ELSEVIER
Available online at www.sciencedirect.com
SCIENCE DIRECT
Journal of Environmental Radioactivity 86 (2006) 227–240
JOURNAL OF ENVIRONMENTAL RADIOACTIVITY
www.elsevier.com/locate/jenvrad

Inter-taxa differences in root uptake of $^{103/106}$Ru by plants

N.J. Willey*, K. Fawcett

Centre for Research in Plant Science, Faculty of Applied Sciences, University of the West of England, Frenchay, Bristol BS16 1QY, UK

Received 9 May 2005; received in revised form 31 August 2005; accepted 5 September 2005
Available online 25 October 2005

Abstract

Ruthenium-106 is of potential radioecological importance but soil-to-plant Transfer Factors for it are available only for few plant species. A Residual Maximum Likelihood (REML) procedure was used to construct a database of relative $^{103/106}$Ru concentrations in 114 species of flowering plants including 106 species from experiments and 12 species from the literature (with 4 species in both). An Analysis of Variance (ANOVA), coded using a recent phylogeny for flowering plants, was used to identify a significant phylogenetic effect on relative mean $^{103/106}$Ru concentrations in flowering plants. There were differences of 2465-fold in the concentration to which plant species took up $^{103/106}$Ru. Thirty-nine percent of the variance in inter-species differences could be ascribed to the taxonomic level of Order or above. Plants in the Orders Geraniales and Asterales had notably high uptake of $^{103/106}$Ru compared to other plant groups. Plants on the Commelinoid monocot clades, and especially the Poaceae, had notably low uptake of $^{103/106}$Ru. These data demonstrate that plant species are not independent units for $^{103/106}$Ru concentrations but are linked through phylogeny. It is concluded that models of soil-to-plant transfer of $^{103/106}$Ru should assume that; neither soil variables alone affect transfer nor plant species are independent units, and taking account of plant phylogeny might aid predictions of soil-to-plant transfer of $^{103/106}$Ru, especially for species for which Transfer Factors are not available.

Keywords: Ruthenium; Soil-to-plant transfer; Phylogeny

* Corresponding author.
E-mail address: neil.willey@uwe.ac.uk (N.J. Willey).

0265-931X/$ - see front matter
doi:10.1016/j.jenvrad.2005.09.002

1. Introduction

Ruthenium-106 is a fission product of radioecological importance but there have been relatively few comparisons of species differences in its uptake by plants from soil. Differences between plant species can affect soil-to-plant transfer of other radioecologically important isotopes such as ^{137}Cs and ^{90}Sr (Nisbet and Woodman, 2000) and might affect soil-to-plant transfer of ^{106}Ru. Here we report a database of relative $^{103/106}Ru$ uptake by 114 plant species, by collation of data we generated for 105 species with data in 5 previous studies, and analyse it using techniques established to provide a phylogenetic perspective on inter-species differences in element concentrations.

Ruthenium-103 and ^{106}Ru are γ-emitters produced in significant quantities by nuclear fission. ^{103}Ru has a relatively short half-life (39 d) but ^{106}Ru has a longer half-life (368 d) and is considered a potentially significant long-term radioecological hazard in the ecosystems it contaminates. ^{106}Ru was a significant component of nuclear-weapons testing fall-out (Walton, 1963; Ritchie et al., 1970) and it was one of the common radionuclides deposited in the Chernobyl 30 km zone (Lux et al., 1995; Krouglov et al., 1998) contributing significantly to external doses to humans (Andersson and Roed, 1994). Despite being deposited primarily in fuel particles which settled close to the Chernobyl reactor (Krouglov et al., 1998), ^{106}Ru was detected in significant quantities in Chernobyl fall-out in, for example, Sweden (Kresten and Chyssler, 1989), Italy (Adamo et al., 2004) and Turkey (Polar and Bayülgen, 1991). ^{106}Ru is also a contributor to effluents from Cap de la Hague (Salbu et al., 2003) and has been a focus of attention in modelling potential accidents with Pressurised Water Reactors (Renaud et al., 1999). Given the potential radioecological importance of ^{106}Ru it is important to understand its ecosystem transfer processes, such as that from soil-to-plant.

Many studies, including some of the first radioecological studies performed, have shown that, in general, ^{106}Ru is less available to plants from soil than ^{90}Sr but more available than ^{137}Cs (Nishita et al., 1956, 1961; Bunzl et al., 1984). This is reflected in soil K_d values and soil-to-plant Transfer Factors (TFs) with a mean of 100 and 0.1, respectively (Sheppard, 1985). However, K_d values for ^{106}Ru in organic soils can be very large (Sheppard and Thibault, 1990) and binding to mobile organic fractions can increase its mobility (Polar and Bayülgen, 1991). Uptake of ^{106}Ru by plants is generally greater from soils of high pH and with high base status, for example from the black soils of the Indian Subcontinent (D'Souza and Mistry, 1980). Overall, therefore, ^{106}Ru is considered quite available to plants in many soils. Interestingly, however, much knowledge of ^{106}Ru transfer to plants has been gained using Cl or nitrosyl forms as experimental contaminants but the deposition in the Chernobyl 30 km zone has proved relatively immobile and unavailable due to its deposition in fuel particles, probably as metallic impurities (Krouglov et al., 1998).

Species differences in ^{106}Ru uptake by plants have been reported (Nishita et al., 1961; Handl, 1988) but compared to other radioecologically significant isotopes there is a paucity of comparisons of concentrations to which plants take up ^{106}Ru and all such have been confined to inter-species comparisons. Recently, molecular descriptions of the evolutionary relationships (phylogeny) of many groups of organisms have been useful to analyse differences in phenotypes between taxa at many levels of the taxonomic hierarchy. New phylogenies of flowering plants have been published specifically to aid such comparisons (e.g. Soltis et al., 1999). Treating relative elemental concentration as a phenotype and mapping it to the flowering plant phylogeny have revealed significant phylogenetic effects on the relative concentration in plants of ^{137}Cs (Broadley et al., 1999; Willey et al., 2005), Cu, Zn, Ni, Cd and Pb, (Broadley et al., 2001), Ca (Broadley et al., 2003) and Mg, K, N, and P (Broadley et al., 2004). These studies reveal that, with the exception of N and P, at least some of the inter-species differences in relative concentration can be ascribed to taxonomic levels higher than the species. This shows that, for concentrations of these elements, species have a tendency to behave as groups rather than each species behaving independently. Such phylogenetic effects not only have to be accounted for when predicting soil-to-plant transfer but might also be used as a framework for making general predictions of relative concentrations in plants. Given the paucity of TF data for $^{103/106}$Ru, a knowledge of relative concentrations for $^{103/106}$Ru might be useful to radioecologists because, for a given substrate availability, they might be used to make predictions of uptake for substrate/plant species combinations for which TFs are not available. Here, using the method established in previous studies, we report a database of relative mean $^{103/106}$Ru concentrations in plants and analyse it using a recent flowering plant phylogeny. We conclude that there is a significant phylogenetic effect on $^{103/106}$Ru uptake by plants and discuss its significance.

2. Materials and methods

2.1. Data for $^{103/106}$Ru uptake by plants

Studies in the literature were selected if they contained measurements of significant concentrations of $^{103/106}$Ru in above-ground green shoots in plants in two or more species after identical exposure in the same contaminated soil. Studies in which foliar contamination had occurred, or in which $^{103/106}$Ru activities were very low, were excluded and different experimental treatments were used as separate 'studies'. Both ^{103}Ru and ^{106}Ru data were included because there is no evidence to suggest discrimination between them during uptake by plants from soil. This provided 9 'studies' ('studies' 1–9 Table 1) from 5 sources (Bell et al., 1988; Coughtrey and Jones, 1985; Douka and Xenoulis, 1991; Handley and Babcock, 1972; Wirth et al., 1996) and included data on 12 species. One hundred and six species were chosen for experiments to complement those in the literature and provide a spread across the angiosperm phylogeny. Four species in literature data sets (*Lolium perenne*, *Lycopersicon esculentum*, *Fragaria vesca*, *Brassica oleracea*) were included in experiments. Species selection was biased in favour of herbaceous plants and crops, tree and aquatic species being more problematic to grow and expose to $^{103/106}$Ru.

Experiments with a number of radiolabelling regimes based on those previously used (Bell et al., 1988; Coughtrey and Jones, 1985; Douka and Xenoulis, 1991; Handley and Babcock, 1972; Wirth et al., 1996) showed that the procedure below, which includes $CaCl_2$ and Na_2EDTA to enhance Ru availability, produced high enough ^{103}Ru concentrations to be reliably measured in plant material. Five replicate pots of each species were grown in approximately 90 g of peat-based Levington's F2 compost (Fison's, Ipswich, UK) for approximately 7 weeks. Plants were grown in a randomised block in a greenhouse with 16 h day and 8 h nights at c. 24 °C and 16 °C, respectively. Plants were labelled with ^{103}Ru in the exponential phase of their growth and before they flowered, hence some taxa were slightly younger or older than 7 weeks. During radiolabelling plants were placed in randomised blocks with 350 $\mu Em^{-1} s^{-1}$ light for 16 h day and 8 h night. For radiolabelling 50 mL of 200 μM $CaCl_2$ and Na_2EDTA were added with 3700 kBq $^{103}RuCl_2\ L^{-1}$ to give 41 kBq g^{-1} substrate. The 50 mL of radiolabelled solution saturated the substrate and the excess solution was caught in saucers below the pots and in all cases was reabsorbed into the substrate during radiolabelling, so a homogenous distribution of ^{103}Ru in the substrate was assumed. Plant shoots were harvested 96 h after ^{103}Ru application, 1 cm above the substrate. Radiolabelling took place in 7 events in 14 blocks, each of which was treated as a separate study ('studies' 10–23 in Table 1). ^{103}Ru activity concentrations were measured in dried plant samples by γ-counting in an LKB Wallac 'Compugamma 1282' (NaI(Tl) detector) with appropriate blanks and background corrections.

Table 1
Relative mean Ru concentrations in 114 species of angiosperm classified according to the phylogeny of Soltis et al. (1999)

Class	Subclass	Group	Super-order	Order	Scientific name	Common name	Mean activity (±SE) (Bq/g)	Relative mean concentration	Study (*n*)
Magnoliids	Magnolidae	"	"	Magnoliales	*Annona cherimoia*	Cherimoya	1920.1 ± 17.6	4.042	22,23 (10)
					Annona squamosa	Custard apple	1404.7 ± 52.8	4.467	23 (5)
				Laurales	*Chimonanthus praecox*	Wintersweet	829.5 ± 903	0.149	11 (5)
				Piperales	*Peperomia hederafolia*	Ivy peperomia	312.5 ± 116.7	2.019	14 (5)
					Peperomia rotundifolia	Round-leaved peperomia	265.1 ± 114.0	1.708	14 (5)
					Houttynia cordata	Houttynia	997.7 ± 343.7	1.086	10 (5)
	Monocots	Commelinoids	"	Arecales	*Areca lutescens*	Areca palm	128.5 ± 37.5	−1.637	10 (5)
				Commelinales	*Commelina coelestis*	Blue spiderwort	121.2 ± 25.3	−1.263	10 (5)
				Poales	*Cyperus zumila*	Umbrella plant	916.6 ± 293.2	0.956	10 (5)
					Carex pendula	Pendulous sedge	165.1 ± 11.7	2.686	13 (5)
					Carex comans	Bronze sedge	150.5 ± 85.7	3.225	13 (5)
					Carex stricta	Sedge	98.1 ± 5.5	−0.629	13 (5)
					Hordeum vulgare	Winter barley	182.3 ± 43	3.515	4 (8)
					Lolium perenne	Rye grass	3450,573 ± 1150,64[a]	0.43	3,10 (12)
					Sorghum vulgare	Northern sugar cane	26.2 ± 2.5	−1.333	15 (5)
					Triticum aestivum	Wheat	7.2 ± 2.9	−1.692	15 (5)
					Triticum durum	Durum wheat	4.1 ± 1.7	−2.194	15 (5)
				Zingeberales	*Canna indica*	Canna lily	14.3 ± 4.4	0.294	19 (5)
					Maranta species	Maranta	50.6 ± 11.8	3.758	23 (5)
					Roscoea scillifolia	Roscoea	48.8 ± 15.2	3.376	23 (5)
					Zingiber officinale	Ginger	93.1 ± 8.6	1.305	14 (5)
		Non-Commelinoids	"	Alismatales	*Arisaema wallichianum*	Jack-in-the-pulpit	275.8 ± 98.9	4.922	23 (5)
					Arisaema tortuosum	Jack-in-the-pulpit	89.8 ± 43.6	3.927	23 (5)
					Scindapsis aureus	Devil's ivy	26.1 ± 7.7	0.983	16 (5)
					Philodendron hastatum	Elephant's ear philodendron	10.3 ± 4.0	−0.229	21 (5)
				Liliales	*Lilium formosanum*	Lily	136.8 ± 34.7	4.341	22 (5)

Group	Superorder	Order	Scientific name	Common name	Mean activity (±SE) (Bq/g)	Relative mean concentration	Study (*n*)
			Nemophila menziesii	Californian bluebell	702.2 ± 161.8	3.887	17 (5)
			Lycopersicon esculentum	Tomato	33,22,102.8 ± NA, NA,24	3.278	5,6,15 (1,1,7)
			Nicotiana glauca	Yellow tree tobacco	615.8 ± 239.6	2.557	12,13,15,16 (20)
			Nicotiana sylvestris	Tobacco 'Only the Lonely'	1130.5 ± 220.8	1.93	12 (5)
			Solanum sisymbrifolium	Solanum	617.9 ± 76.1	3.592	17 (5)
	Euasterid 2	Apiales	*Angelica hispanica*	Angelica	35.6 ± 14.2	2.578	21 (5)
			Apium graveolens	Celery	2271.0 ± 812.7	2.429	12 (5)
			Coriandrum sativum	Coriander	242.7 ± 36.0	2.634	18 (5)
			Daucus carota	Carrot	99.6 ± 33.6	4.124	4 (13)
			Hedera helix	Ivy	55.7 ± 8.2	0.572	14 (5)
			Pittosporum species	Pittosporum	27.8 ± 6.9	2.821	22 (5)
		Asterales	*Centaurea species*	Cornflower	393.4 ± 95.4	3.646	16 (5)
			Helianthus annuus	Sunflower	1366.6 ± 135.5	3.595	14 (5)
			Lactuca sativa	Lettuce	47.9 ± 10.0	2.912	15 (5)
			Tithonia rotundifolia	Mexican sunflower	377.2 ± 128.9	3.596	15,17 (10)
Rosids	Basal	Saxifragales	*Liquidambar styraciflua*	Sweet gum	14.4 ± 3.7	1.045	13 (5)
			Heuchera micrantha	Alum-root	140.4 ± 55.3	−2.624	20 (5)
			Heuchera sanguinea	Heuchera	36.5 ± 7.7	2.727	21 (5)
			Bergenia purpurascens	Bergenia	84.5 ± 16.6	3.886	22,23 (10)
			Bergenia cordifolia	Bergenia	103.1 ± 23.9	4.1	22 (5)
		Geraniales	*Geranium pyrenaicum*	Pyrenian cranesbill	594.5 ± 158.8	4.097	16 (5)
			Geranium sylvaticum	Wood cranesbill	77.9 ± 9.4	3.573	21 (5)
			Pelargonium alchemilloid	Ladys Mantle-leaved pelargoniu	363.2 ± 86.1	3.596	16 (5)
		Myrtales	*Callistemon subdulatus*	Tonghi bottle-brush	12.2 ± 4.8	0.697	13 (5)
			Clarkia bottea	Clarkia	661.6 ± 140.3	3.788	19 (5)
			Oenothera hookeri	Giant Yellow evening primrose	481.9 ± 94.8	1.07	12 (5)

(*continued on next page*)

Table 1 (*continued*)

Class	Subclass	Group	Super-order	Order	Scientific name	Common name	Mean activity (±SE) (Bq/g)	Relative mean concentra-tion	Study (*n*)
				Asparagales	*Allium ameloprasum*	Leek	326.9 ± 82.6	0.659	12 (5)
					Allium cepa	Onion	424.5 ± 396.4	−0.588	12 (5)
					Allium schoenoprasum	Chives	414.9 ± 143.3	0.823	12 (5)
					Allium tuberosum	Garlic chives	139.7 ± 49.4	3.134	13 (5)
					Asparagus officinalis	Asparagus	443.0 ± 148.4	−0.753	11 (5)
					Tigridia pavonia	Peacock flower	175.1 ± 29.4	3.068	20 (5)
					Crocosmia masonorum	Montbretia	8.1 ± 4.5	−2.114	21 (5)
Eudicots	Basal	"	"	Proteales	*Grevillea robusta*	Silk oak	337.3 ± 258.6	3.25	21 (5)
					Platanus orientalis	Oriental plane	145.8 ± 25.9	4.121	21 (5)
				Ranunculales	*Papaver pilosum*	Hairy poppy	237.6 ± 103.8	3.078	20 (5)
					Papaver somniferum	Opium poppy	443.9 ± 229.7	2.875	18 (5)
					Putsatilla vulgaris	Pasque flower	72.3 ± 12.4	3.557	21,22 (10)
		Caryophyllids	"	Caryophyllales	*Beta vulgaris*	Beet	100.6 ± 24.6	3.518	15 (5)
					Dianthus seguiri	Pink	570.2 ± 112.4	1.277	12 (5)
					Dianthus superbus	Superb pink	322.8 ± 144.6	3.65	13 (5)
					Dianthus gratinopoulis	Cheddar pink	1994.0 ± 449.1	0.932	11 (5)
					Gypsophila elegans	Baby's tears	1057.3 ± 287.1	4.113	19 (5)
					Gypsophila paniculata	Baby's tears	635.6 ± 134.0	3.732	19 (5)
					Silene chalcedonia	Campion	252.6 ± 27.0	2.757	18,19 (10)
					Rheum tataricum	Rhubarb	1484.9 ± 170.8	1.612	10 (5)
					Rumex acetosa	Sorrel	1533.6 ± 369.6	0.67	11 (5)
					Rumex sanguineus	Bloodwort	330.6 ± 114.1	2.791	18 (5)
	Core Eudicots	Asterids	Basal	Ericales	*Vaccinium myrtillis*	Bilberry	50 ± NA[b]	0.354	8 (1)
					Camellia sinensis	Camellia (Tea)	207.9 ± 39.9	1.875	14 (4)
			Euasterid 1	Lamiales	*Mentha piperata*	Peppermint	112.6 ± 34.5	2.972	20,21 (10)
					Mentha spicata	Spearmint	958.6 ± 173.6	1.785	12 (5)
					Salvia officinalis	Sage	3930.7 ± 1535.3	2.25	10 (5)

Group	Superorder	Order	Scientific name	Common name	Mean activity (±SE) (Bq/g)	Relative mean concentration	Study (*n*)
			Oenothera tetragona	Evening primrose' Sundrops'	279.9 ± 97.4	0.429	12 (5)
	Eurosid 1	Malpighiales	*Hypericum olympicum*	Dwarf St. John's Wort	90.1 ± 13.4	3.165	13 (5)
			Passiflora caerulescens	Passion flower	53.3 ± 5.6	3.538	22 (5)
		Rosales	*Humulus japonicus*	Japanese hop	81.1 ± 27.1	3.24	15 (5)
			Elaeagnus multiflora	Elaeagnus	25.7 ± 1.7	1.701	13 (5)
			Morus alba	White mulberry	312.7 ± 86.3	0.612	12 (5)
			Maclura pomifera	Osage orange	53.8 ± 13.2	2.23	13 (5)
			Fragaria vesca	Strawberry	20,80,50,2243 ± NA,529[b]	1.585	7,8, 9,10,11,12 (17)
			Rubus idaeus	Blackberry	20 ± NA[b]	0.242	9 (1)
			Rubus saxitilus	Blackberry	20,20 ± NA[b]	0.105	7,8 (1,1)
			Pilea cadierei	Pilea	430.9 ± 98.5	2.548	14 (5)
		Fabales	*Lupinus angustifolius*	Lupin	8212.5 ± 2778.4	3.162	10 (5)
			Medicago lupulina	Black Medik	173.9 ± 79.1	3.269	13 (5)
			Medicago sativa	Lucerne	533 ± 36[a]	−0.009	3 (1)
			Phaseolus vulgaris	Bean	52,11 ± NA	3.197	5,6 (1)
			Pisum sativum	Pea	45.8 ± 14.5	0.42	12 (2)
			Trifolium pratense	Red clover	595.5 ± 68.2	3.539	17,18 (10)
			Trifolium repens	White clover	507.1 ± 119.5	3.713	19,20 (10)
			Trifolium arvense	Hare's foot clover	744.0 ± 260.6	1.347	12 (5)
		Curcurbitales	*Curcurbita maxima*	Pumpkin 'Blue Hubbard'	2693.8 ± 470.6	4.437	14 (5)
			Curcurbita pepo	Pumpkin	3311.5 ± 530.1	4.647	14 (5)
	Eurosid 2	Brassicales	*Alyssum montanum*	Alyssum	1720.2 ± 392.5	0.729	11 (5)
			Alyssum saxatile	Alyssum	80.5 ± 16.4	1.682	19 (5)
			Alyssum petraeum	Alyssum	2467.0 ± 655.3	2.596	12 (5)
			Brassica oleracea	Cabbage	6.9,22.1,316.1 ± 4.1,4,49.8	3.139	1,2,4,17,20 (4,12,14)
			Tropaeolum perigrinum	Canary creeper	131.7 ± 39.3	3.76	15 (5)

(*continued on next page*)

Table 1 (*continued*)

Class	Subclass	Group	Super-order	Order	Scientific name	Common name	Mean activity (±SE) (Bq/g)	Relative mean concentration	Study (*n*)
					Antirrhinum X	Snapdragon	446.1 ± 123.7	3.117	18 (5)
					Digitalis ambigua	Large Yellow foxglove	1853.8 ± 540.9	2.398	12 (5)
					Digitalis purpurea	Wild foxglove	1469.0 ± 228.6	2.231	12 (5)
				Solanales	*Ipomoea purpurea*	Purple morning glory	694.6 ± 136.6	3.867	14,15, 16 (15)

Studies 1–2: Bell et al., 1988, seasons 1 + 2, sandy loam soil UK, plants grown to maturity; study 3: Douka and Xenoulis, 1991; mean of harvests 2–4, clay soil pH 8 Greece with 339 Bq kg^{-1} ^{106}Ru, mean of shoots. Study 4: Coughtrey and Jones, 1985, brown sand Freckenham series, 0.29 μCi ^{103}Ru/5 kg pot, mean of shoots. Studies 5–6 Handley and Babcock, 1972, hydroponics Hoagland's solution, 38.4 μCi/4 L, mean of three plants, new growth and old growth. Studies 7–9: Wirth et al., 1996, collected at 3 sites in Bavaria in 1992, TForg. Studies 10–23 experiments for this paper. n = Number of replicate measurements. Mean activities in plants from studies carried out for this paper also listed.

[a] Expressed as Bq kg^{-1}.

[b] Represented as TF 10^4.

2.2. Statistical analyses

Data were obtained, from the literature and experiments, for 114 species across 23 'studies'. Due to species selection, and replication between blocks, every data set had at least one species in common with another data set. Some species were represented multiple times in the data sets whilst others were present only once. Residual Maximum Likelihood (REML) analysis was used to produce a database of relative mean $^{103/106}Ru$ concentrations in the 114 plant species. Studies were used as 'blocks' and species as 'treatments' in the REML analysis which was run on the statistical software package Genstat for Windows 5th Ed release 4.2 (VSN International, Oxford, UK) (Thompson and Welham, 2001) using the programme of Broadley et al. (1999, 2001, 2003, 2004). Defining blocks and treatments in this way takes account of the absolute differences in concentrations related to experimental conditions (studies) to reveal relative mean concentrations for the treatments (species). REML analyses, which here included $\log_e$-transformation of raw values, can produce relative mean concentrations that are both positive and negative (Thompson and Welham, 2001). An ANOVA of REML-transformed values, coded using the ordinal phylogeny of Soltis et al. (1999) was then performed. The ordinal phylogeny of Soltis et al. (1999) was used because it was published specifically for such analyses and to enable direct comparison of Ru results with previous analyses for other elements that used this phylogeny. The categories 'Class', 'Subclass', 'Group' and 'Superorder' (Table 1) were used nominally above the level of the Order because the relationship between the Linnaean hierarchy they are derived from and higher taxonomic groups on recent phylogenies is unresolved. Normality tests used a Kolmogorov–Smirnov test in SigmaStat 3.0 for Windows.

3. Results and discussion

The relative mean $^{103/106}Ru$ concentrations in 114 plant species, together with absolute values from each experimental study, are shown in Table 1. The REML procedure accounts for variance in absolute concentrations associated with different experimental conditions ('studies') in the input data in order to estimate relative mean concentrations for plant species across

Group	Superorder	Order	Scientific name	Common name	Mean activity (±SE) (Bq/g)	Relative mean concentration	Study (*n*)
		Malvales	*Cistus palhinhae*	St. Vincent Cistus	68.8 ± 26.1	2.361	13 (5)
			Althaea rosea	Hollyhock	5054.1 ± 701.7	1.93	11 (5)
			Malva sylvestris	Common mallow	6570.0 ± 421.1	5.191	16 (5)
		Sapindales	*Pistachia chinensis*	Chinese pistachio	36.4 ± 6.8	0.712	18 (5)
			Ruta graveolens	Rue	3041.6 ± 1667.9	1.046	11 (5)

all studies. There were significant effects of block in the analysis confirming that values for all these species could not be compared without taking it into account. The values in Table 1 cannot, therefore, be regarded as concentration ratios or TFs for plant species under a given set of conditions but rather they are predicted relative mean concentrations across a variety of conditions, i.e. which species tend to have, relative to each other, higher or lower concentrations. There are, however, a number of factors that might interact under a particular set of conditions to produce relative concentrations somewhat different to those in Table 1. First, the length of exposure to $^{103/106}$Ru in almost all the data sets collated was acute. The relationship between concentrations produced in plants after acute and chronic exposure to $^{103/106}$Ru from the soil is little known. For other radionuclides, such as ^{90}Sr and ^{137}Cs, there is evidence that much uptake takes place during the exponential phase of growth (Weaver et al., 1981), as is the case with many mineral nutrients (Marschner, 1995). As majority of the species in Table 1 were

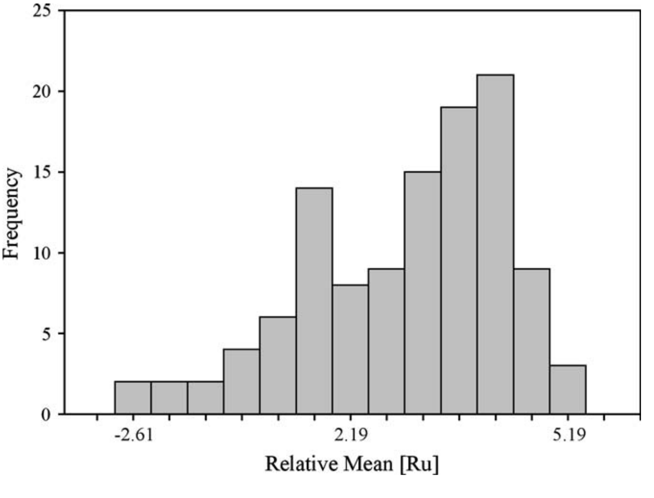


Fig. 1. The frequency distribution of relative mean Ru concentrations in 114 species of angiosperm (Kolmogorov–Smirnov distribution 0.11, $P < 0.001$).

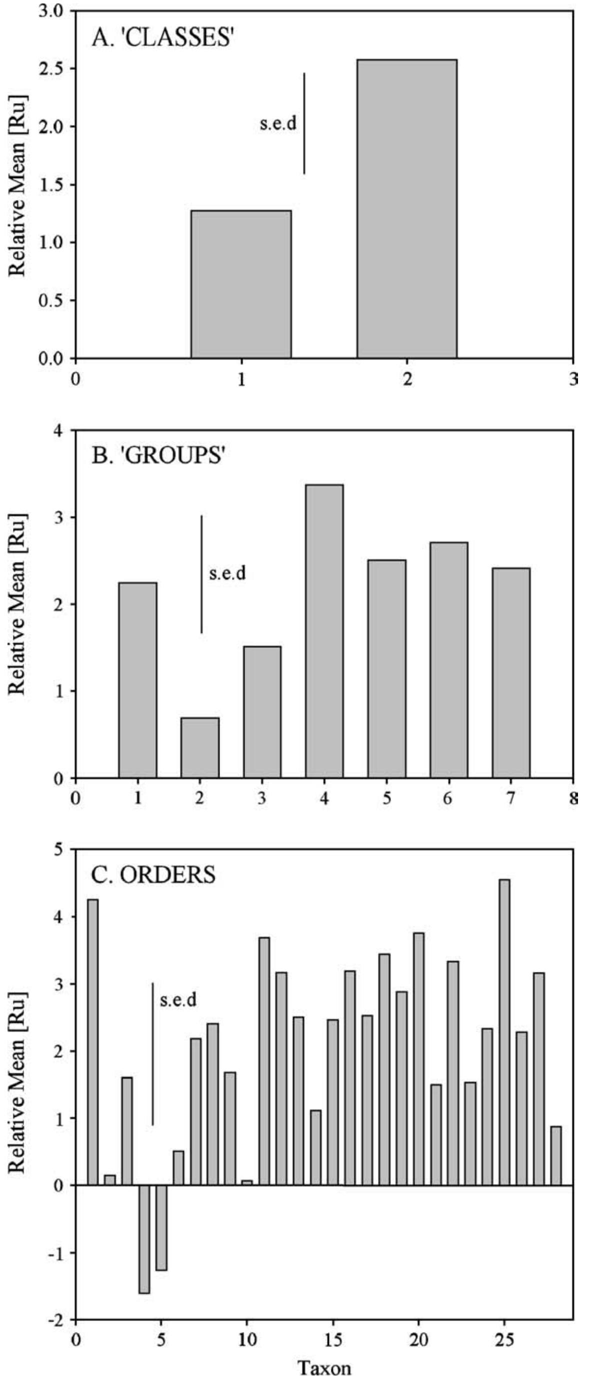
Table 2
Results of ANOVA for relative mean Ru concentrations in 114 angiosperm species classified according to Soltis et al. (1999)

	df	Sum of Squares	% Sum of Squares	Cumulative % Sum of Squares	Mean square	Variance ratio
Class	2	3816	4.2	4.2	1272	4.46
Subclass	3	226	0.2	13.3	113	0.4
Group	4	8153	8.9	13.1	2038	7.15
Superorder	4	1881	2.1	15.4	470	1.65
Order	14	21 775	23.7	39.1	1555	5.45
Family	22	10 168	11.1	50.2	462	1.62
Genus	36	37 697	41.1	91.3	1047	3.67
Residual	28	7987	8.7	100.0	285	
Total	113	91 706				

exposed during their exponential growth phase, it seems likely that the relative mean concentrations in Table 1 will relate to chronic exposures, but it is possible that this relationship is not close. Further, for ^{106}Ru, as for other radioisotopes, the chemical species present in soil can affect its behaviour (Krouglov et al., 1998). It is possible that different compounds of $^{103/106}$Ru might not produce the same relative concentrations as those in Table 1. In fact, it is possible that a number of such edaphic factors might interact with relative $^{103/106}$Ru concentrations because all species cannot grow equally well under different conditions. There is variety in $^{103/106}$Ru compound and experimental conditions used to generate data for Table 1, which therefore provides relative mean concentrations across them, but the full range of exposure conditions might not produce results identical to those in Table 1. Overall, Table 1 does not, therefore, provide definitive relative mean concentrations between plant species. However, as the largest inter-species comparison of uptake of $^{103/106}$Ru by plants is yet to be reported, Table 1 does provide an estimate of the relative mean concentrations for a wide variety of plants and a starting point for analysing them phylogenetically.

Log_e-transformed relative mean $^{103/106}$Ru concentrations ranged from −2.62 to 5.19 across the 114 species in the database (Table 1), indicating that absolute concentrations might differ by more than 2000 fold ($e^{7.81} = 2465$) if all species could be grown simultaneously under the same conditions. In experimental data derived under a single set of conditions (studies 10–23) the lowest $^{103/106}$Ru concentration was 4.1 Bq g^{-1} in *Triticum durum* and the highest concentration was 6570 Bq g^{-1} in *Malva sylvestris*, roughly agreeing with this estimate. $^{103/106}$Ru REML values were not normally distributed but significantly skewed (Fig. 1) and there were no significant outliers that could be removed to produce normality. Overall, these results suggest

Fig. 2. Average relative mean Ru concentration in angiosperm taxa according to Soltis et al. (1999) (s.e.d. = standard error of the difference at 0.05). A: 'Classes' (for ANOVA, $P < 0.001$) 1 = Magnoliids ($n = 33$ species), 2 = Eudicots ($n = 81$). B: 'Groups' (for ANOVA, $P = 0.002$) 1 = Magnoliidae (6), 2 = Commelinoid monocots (15), 3 = non-Commelinoid monocots (12), 4 = Basal Eudicots (5), 5 = Caryophyllids (10), 6 = Asterids (24), 7 = Rosids (42). C: Orders (for ANOVA, $P = 0.004$) 1 = Magnoliales (2), 2 = Laurales (1), 3 = Piperales (3), 4 = Arecales (1), 5 = Commelinales (1), 6 = Poales (9), 7 = Zingerberales (4), 8 = Alismatales (4), 9 = Liliales (5), 10 = Asparagales (3), 11 = Proteales (2), 12 = Rannunculales (3), 13 = Caryophyllales (10), 14 = Ericales (2), 15 = Lamiales (6), 16 = Solanales (6), 17 = Apiales (6), 18 = Asterales (4), 19 = Saxifragales (5), 20 = Geraniales (3), 21 = Myrtales (4), 22 = Malphigiales (2), 23 = Rosales (8), 24 = Fabales (8), 25 = Cucurbitales (2), 26 = Sapindales (5), 27 = Malvales (3), 28 = Brassicales (2).


A. 'CLASSES'
s.e.d
Relative Mean [Ru]
3.0
2.5
2.0
1.5
1.0
0.5
0.0
0
1
2
3
B. 'GROUPS'
s.e.d
Relative Mean [Ru]
4
3
2
1
0
0
1
2
3
4
5
6
7
8
C. ORDERS
s.e.d
Relative Mean [Ru]
5
4
3
2
1
0
-1
-2
0
5
10
15
20
25
Taxon

that there is a significant range of relative mean $^{103/106}$Ru concentrations between plant species and that this range, and its frequency distribution, might usefully be considered in soil-to-plant transfer involving multiple plant species.

There were statistically significant effects of phylogeny on $^{103/106}$Ru concentrations in plants at the level of the 'Class', 'Group', Order and Genus (Table 2). Overall, 39% of the Sums of Squares was associated with the level of Order and above, and Genus was associated with the greatest % Sum of Squares. Between the plants categorized here by 'Class', the Magnoliids ($n = 33$ species) had significantly lower relative mean Ru concentrations than the Eudicots ($n = 81$) (Tables 1 and 2; Fig. 2A). Significant differences at the 'Group' level were marked by relative mean Ru concentrations that were significantly lower in Commelinoid monocots than most other groups (Tables 1 and 2; Fig. 2B). At the Ordinal level the Cucurbitales and Magnoliales had the highest relative mean concentrations but were both only represented by two species (Table 1; Fig. 2C). Of the orders with greater numbers of representatives, the Geraniales ($n = 3$) and Asterales ($n = 4$) had the highest, and the Poales ($n = 9$) the lowest relative mean Ru concentrations. The Apiales, Caryophyllales, Lamiales and Fabales had, despite some high or low values for individual species, relative mean Ru concentrations close to the overall mean (2.19). Relative mean concentrations for these higher taxa do not necessarily ensure that all species within them have low or high values but rather there are significant tendencies to low or high values. In comparison to other studies of ion concentrations in plants down to the Ordinal level, the phylogenetic signal for Ru of 39% is greater than that for P (6.8%) and N (3.3%) (Broadley et al., 2004), Cs (15%) (Willey et al., 2005), Pb (20%), Cr (23%), Cu (24%), and Cd (27%) (Broadley et al., 2001), and Na (23%) (Broadley et al., 2004), but less than that for Zn (44%), Ni (46%) (Broadley et al., 2001), K (49%) (Broadley et al., 2004) and Ca (63%) (Broadley et al., 2003).

The Commelinoid monocots have been noted to have unusually low Ca uptake (Broadley et al., 2003) and the monocots are known to have low uptake of Cs (Broadley and Willey, 1999; Willey et al., 2005), so it seems likely that the relatively low uptake of Ru reported here is part of a pattern of unusual uptake of at least some elements by plants on this clade. Certainly, given the importance of the cereals crops on this clade it is a hypothesis worth further investigation. The few relative mean Ru concentrations for the Cucurbitales and Brassicales in Table 1 suggest that these orders might have relatively high and low uptake of Ru, respectively. There are indications that for other elements these Orders also have characteristic uptake (Broadley et al., 2003, 2004) and we suggest that it might be worthwhile investigating further their uptake of Ru. The Caryophyllales have high relative uptake of Cs (Broadley and Willey, 1999; Willey et al., 2005) but the data reported here suggest that they are not unusual in their Ru uptake.

4. Conclusion

There are significant differences between plant species in the concentration to which they take up acute doses of $^{103/106}$Ru. Clearly, there are soil factors that affect soil availability of $^{103/106}$Ru but the data in Table 1 strongly suggest that, from a given availability, plant uptake will differ significantly between species and needs to be taken into account in understanding soil-to-plant transfer. Fig. 1 suggests that inter-species differences are not normally distributed and that parametric methods might have to be used with care for modelling differences across numerous species. A priori there is no reason why $^{103/106}$Ru concentrations should differ just between species (which is primarily a reproductive unit that can be difficult to define in plants) and the data presented here strongly suggest that radioecologists should consider taxonomic units other than the species when modelling soil-to-plant transfer of $^{103/106}$Ru. Overall, for $^{103/106}$Ru uptake species do not behave independently but are affected by phylogenetic position. This has enabled us to suggest testable hypotheses about which taxonomic units of plants have relatively high and low uptake of Ru and to make general predictions of uptake for taxa in which few TF values exist.

Acknowledgement

We would like to acknowledge the financial support of the UK Food Standards Agency, and to thank Judy Brown for radioanalytical support and Andrew Mead of Warwick HRI for developing the Genstat programme.

References

Adamo, P., Arienzo, M., Pugliese, M., Roca, V., Violante, P., 2004. Accumulation history of radionuclides in the lichen *Stereocaulon vesuvianum* from Mt. Vesuvius (south Italy). Environ. Pollut. 127, 455–461.

Andersson, K.G., Roed, J., 1994. The behaviour of Chernobyl ^{137}Cs, ^{134}Cs and ^{106}Ru in undisturbed soil – implications for external radiation. J. Environ. Radioact. 22, 183–196.

Bell, J.N.B., Minski, M.J., Grogan, H.A., 1988. Plant uptake of radionuclides. Soil Use Manage 4, 76–84.

Broadley, M.R., Willey, N.J., 1999. A comparison of caesium uptake by 30 plant species. Environ. Poll. 97, 11–15.

Broadley, M.R., Willey, N.J., Mead, A., 1999. A method to assess taxonomic variation in shoot caesium concentration among flowering plants. Environ. Pollut. 106, 341–349.

Broadley, M.R., Willey, N.J., Wilkins, J., Baker, A.J.M., Mead, A., White, P.J., 2001. Phylogenetic variation in heavy metal accumulation in angiosperms. New Phytol. 152, 9–27.

Broadley, M.R., Bowen, H.C., Cotterill, H.L., Hammond, J.P., Meacham, M.C., Mead, A., White, P.J., 2003. Variation in the shoot calcium content of angiosperms. J. Exp. Bot. 54, 1–16.

Broadley, M.R., Bowen, H.C., Cotterill, H.L., Hammond, J.P., Meacham, M.C., Mead, A., White, P.J., 2004. Phylogenetic variation in the shoot mineral concentration of angiosperms. J. Exp. Bot. 55, 321–336.

Bunzl, K., Bachhüber, H., Schimmack, W., 1984. Distribution co-efficients of ^{137}Cs, ^{85}Sr, ^{141}Ce, ^{103}Ru, ^{131}I and ^{95m}Tc in the various horizons of cultivated soils in Germany. In: Udulft, P., Mekel, B., Prosl, K.M. (Eds.), Proceedings of International Symposium on Recent Investigations in the Zone of Aeration. pp. 567–577.

Coughtrey, P.J., Jones, A., 1985. Experimental Studies on the Dynamics of Radionuclide Transport in Soils and Plants: An Investigation of the Effects of Chemical Form and Time of Administration. Associated Nuclear Services, UK, Report No. 413.

D'Souza, T.J., Mistry, K.B., 1980. Behaviour of gamma-emitting fission products ^{106}Ru, ^{125}Sb, ^{134}Cs and ^{144}Ce deposited on established pastures in tropical environs. J. Nuc. Agric. Biol. 9, 50–53.

Douka, C.E., Xenoulis, A.C., 1991. Radioactive isotope uptake in a grass/legume association. Environ. Pollut. 73, 11–23.

Handl, J., 1988. Transfer from soil to plants of ^{106}Ru as nitrosyl and as chloride. Health Phys. 28, 548–555.

Handley, R., Babcock, K.L., 1972. Translocation of ^{85}Sr, ^{137}Cs and ^{106}Ru in crop plants. Radiat. Bot. 12, 113–119.

Kresten, P., Chyssler, J., 1989. The Chernobyl fallout: surface soil deposition in Sweden. Geol. Foren. Stockh. Forh. 111, 181–185.

Krouglov, S.V., Kurinov, A.A., Alexhakin, R.M., 1998. Chemical fractionation of ^{90}Sr, ^{106}Ru, ^{137}Cs and ^{144}Ce in Chernobyl-contaminated soils: an evolution in the course of time. J. Environ. Radioact. 38, 59–76.

Lux, D., Kammerer, L., Ruhm, W., Wirth, E., 1995. Cycling of Pu, Sr, Cs and other long-living radionuclides in forest ecosystems of the 30 km zone around Chernobyl. Sci. Total Environ. 173, 375–384.

Marschner, H., 1995. The Mineral Nutrition of Plants, second ed. Academic Press, London.

Nisbet, A.F., Woodman, R.F.M., 2000. Soil-to-plant transfer factors fro radiocaesium and radiostrontium in agricultural systems. Health Phys. 78, 279–288.

Nishita, H., Kowalewsky, B.W., Steen, A.J., Larson, K.H., 1956. Fixation and extractability of fission products contaminating various soils and clays: I ^{90}Sr, ^{91}Y, ^{106}Ru, ^{137}Cs, ^{144}Ce. Soil Sci. 81, 317–326.

Nishita, H., Romney, E.M., Larson, K.H., 1961. Uptake of radioactive fission products by crop plants. J. Agric. Food Chem. 9, 101–106.

Polar, E., Bayülgen, N., 1991. Differences in the availabilities of $^{134/137}Cs$ and ^{106}Ru from a Chernobyl-contaminated soil to a water plant, duckweed, and to the terrestrial plants bean and lettuce. J. Environ. Radioact. 13, 251–259.

Renaud, P., Réal, J., Maubert, H., Roussel-Debet, S., 1999. Dynamic modelling of the cesium, strontium and ruthenium transfer to grass and vegetables. Health Phys. 76, 495–501.

Ritchie, J.C., Clebsch, E.E.C., Rudolph, W.K., 1970. Distribution of fallout and natural gamma radionuclides in litter, humus and surface mineral soil layers under natural vegetation in the Great Smoky Mountains, North Carolina–Tennessee. Health Phys. 18, 479–489.

Salbu, B., Skipperud, L., Germain, P., Gueguenîat, P., Strand, P., Lind, O.C., Christensen, G., 2003. Radionuclide speciation in effluent from La Hague reprocessing plant in France. Health Phys. 85, 311–322.

Sheppard, M.I., 1985. Radionuclide partitioning coefficients in soils and plants and their correlation. Health Phys. 49, 106–111.

Sheppard, M.I., Thibault, D.H., 1990. Default soil solid/liquid partition coefficients, K_ds, for four major soil types: a compendium. Health Phys. 59, 471–482.

Soltis, P.S., Soltis, D.E., Chase, M.W., 1999. Angiosperm phylogeny inferred from multiple genes as a research tool for comparative biology. Nature 402, 402–404.

Thompson, R., Welham, S.J., 2001. REML analysis of mixed models. In: Payne, R.W. (Ed.), The Guide to Genstat-Part 2. Statistics. VSN International, Oxford, UK, pp. 413–503.

Walton, A., 1963. The distribution in soils of radioactivity from weapons tests. J. Geophys. Res. 68, 1485–1496.

Weaver, C.M., Harris, N.D., Fox, L.R., 1981. Accumulation of strontium and caesium by kale as a function of plant age. J. Environ. Qual. 10, 95–98.

Willey, N.J., Tang, S., Watt, N., 2005. Predicting inter-taxa differences in plant uptake of $^{134/137}Cs$. J. Environ. Qual. 34, 1478–1489.

Wirth, E., Kammerer, L., Ruehm, W., Steiner, M., Hiersche, L., Krestel, R., Mamikhin, S., Tsvetnova, T., Kuchma, K., 1996. Uptake of radionuclides by understorey vegetation and mushrooms. In: Belli, M., Tikhomirov, F. (Eds.), Behaviour of Radionuclides in Natural and Semi-natural Environments. European Commission, Brussels, pp. 61–79. Experimental Collaboration Project No. 5, Final Report EUR 16531 EN.