Journal of Environmental Radioactivity 282 (2025) 107622

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Improved soil-to-plant transfer factors for ^{99}Tc and ^{79}Se in natural and agricultural ecosystems

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ARTICLE INFO

Handling editor: Dr S.C. Sheppard

Keywords:
Soil-to-Plant Transfer Factors
Technetium
Radioselenium
Taxonomic Effects

ABSTRACT

Geological Disposal Facilities (GDFs) are the preferred option for the disposal of high- and medium-level nuclear waste but environmental assessments for GDFs are complex. Models of transfer into the biosphere for radioisotopes that occur in nuclear waste rely on estimated Transfer Factors (TFs) that often have high levels of uncertainty and only exist for a few species. Here, using two key radioisotopes found in nuclear waste, we show that taxonomic analyses and phylogenetically based trait prediction (PTP) can be used to both reduce uncertainty in current estimates of soil-to-plant TF and to predict them for the many species with no measurements. We grew 61 species of plants selected to provide a phylogenetically informed sample, measured their uptake of ^{99}Tc and ^{75}Se, and reconstructed their possible evolutionary relationships using gene sequence information. The uptake of Tc and Se isotopes by plants was correlated, and for Tc was more similar within plant groups than between them and included significant taxonomic and phylogenetic influences. We use these findings to suggest improved soil-to-plant Transfer Factors (TFs) for ^{99}Tc and ^{79}Se. We suggest that the approaches we used might be useful for a range of radionuclides, in both improving current estimates of TF and for predicting TFs to plants and, perhaps, to other biota. Such improvements might be useful not only for environmental assessments of nuclear waste disposal but also the environmental regulation of a nuclear industry being expanded in several nations to help meet targets for reducing global CO_2 emissions.

1. Introduction

The spent fuel from nuclear power generation produces one of humanity's most complex waste management challenges. In Sweden, where implementation of the nuclear waste disposal plan is relatively advanced, the task is regarded as 'one of Sweden's largest environmental protection projects' (https://www.skb.com/, accessed 31/05/24). Geological Disposal Facilities (GDFs) are generally the preferred permanent storage option for nuclear waste but, as yet, there are none that are fully operational. To approve the siting and operation of GDFs, the organisations that construct and regulate them need to be able predict the possible environmental behaviour of radionuclides they contain. Here we report research directly relevant to a GDF in the United Kingdom, but the results also have general implications for modelling the transfer of radioisotopes.

^{99}Tc and ^{79}Se are important radioisotopes in nuclear waste and have been released into the environment, especially from nuclear waste processing facilities. Most of the ^{99}Tc and ^{79}Se that has been produced is, however, in nuclear waste destined for permanent storage facilities. In fact, after the decay of short-lived radionuclides over about the first 1000 years, ^{99}Tc and ^{79}Se contribute very significantly to the long-term total radioactivity in much high-level nuclear waste (IAEA, 1992). ^{99}Tc and ^{79}Se are of particular interest in the environment and in nuclear waste because, in contrast to most other isotopes in nuclear waste, under many environmental conditions they can form mobile anionic chemical species (Bennett and Willey, 2003; Tolu et al., 2014). Both have redox-sensitive environmental chemistries and under oxidizing conditions readily form TcO_4^- and SeO_4^{2-}, which are biologically available (Antanaitis et al., 2008) and readily taken up by organisms, including crops and plants (Bennett and Willey, 2003; Bitterli et al., 2010).

Soil bioavailability is a key factor in the soil to plant transfer of radionuclides but predicting it using chemical extraction of a radionuclide from a soil is not necessarily a good predictor of transfer to plants - for almost any element some plant species take up very little of the element even when it can be extracted chemically and some plant species can actively increase availability (often by root biological activities that change redox potential for example (Konnerup et al., 2017)). In addition, in many instances the transfer of ^{99}Tc or ^{79}Se into many food chains

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https://doi.org/10.1016/j.jenvrad.2025.107622
Received 31 May 2024; Received in revised form 20 January 2025; Accepted 20 January 2025
Available online 31 January 2025

depends significantly on activity in above ground biomass, which can be determined not just by root uptake but also by the root to shoot transfer factor, which for Tc and Se can be both significant and different between crop or plant species (Henner et al., 2013; Sun et al., 2010).

The published compilations of soil-to-plant transfer factors (TFs) of the IAEA can be used for useful general predictions of the transfer of some important radionuclides to the main temperate agricultural crops (IAEA, 2010) and other plants (IAEA, 2014). There is, however, significant uncertainty associated with these TFs. There is even more uncertainty for other crops and species not included in the databases, particularly: a) many important tropical crops, b) minor crops in both temperature and tropical zones (that can contribute significantly to doses in certain groups), and c) the very many wild plant species that might be contaminated by a leak from a GDF and for which there are increasingly stringent legislative requirements for protection (ICRP, 2014). It is impractical to measure TFs for all possible soil-plant-radionuclide combinations, so to reduce uncertainty in all estimates and to be able to predict radionuclide transfer to a wide range of crops and species, biological traits associated with radionuclide transfer have been sought. Species differences in soil-plant TFs have been analysed with respect to plant growth forms (e.g. 'grasses/herbs/shrubs'), growth strategies (e.g. 'competitor/ruderal/stress tolerator') (Willey and Wilkins, 2008), growth rates (Willey et al., 2010), and, in common with analyses of other ecologically significant species traits, taxonomies (Broadley et al., 1999; White et al., 2012; Willey, 2010). Here we build upon the use of taxonomic patterns and report data for flowering plants (angiosperms) based on a phylogenetically informed experimental design and use it to test the influence of phylogeny (evolutionary relationship) on plant uptake of ^{99}Tc and ^{79}Se. We focussed on angiosperms because most crop plants are in this plant group, and they also dominate the vegetation of many natural ecosystems. We chose to test taxonomy and phylogeny because the many recent studies of evolutionary relationships between species mean that, in general, there is now a consensus on relationships for many groups of organisms, including plants, which have found increasing use for predicting differences in a range of biological traits. We conclude that taxonomy and phylogeny can be used to reduce uncertainty in current estimates of TF of ^{99}Tc and ^{79}Se by plants and to predict TFs for these radionuclides when there are currently no measured values, and that the methods we used might be useful for predicting the transfer of other radioisotopes to plants and other organisms.

2. Materials & methods

A hierarchical species-sampling regime was designed for three major clades of the angiosperm phylogeny (Fig. 1). The clades were chosen to represent major groups of flowering plants and the selection of species was focused on the flora of the British Isles. In total, 61 quick-growing herbaceous species (Supplementary Information 1) were grown in 5 replicate 12 cm pots with saucers in Levington's F2S compost with supplementary lighting and heating. They were watered on demand to field capacity. Levington's F2S, which is for early growth-stage seedlings, is *Sphagnum* peat-based with added 'lime' (primarily dolomite) and ammonium sulphate (about 3%). The pH is *c.*6.2 and P = *c.*50 mg/L, K = *c.*50 mg/L and nitrate = *c.*10 mg/L. When tested against a range of other composts, peat-based composts such as Levington's F2S provide more reliable and uniform growth of herbaceous plants such *Arabidopsis* for experimental purposes (Drake et al., 2018). Plants were grown in randomised blocks in batches until they were in the exponential phase of their growth (i.e. before they flowered). They were then radiolabelled by dispensing 25 mL of solution labelled with 107 kBq ^{99}Tc/L and 95 kBq ^{75}Se/L evenly onto the surface of the compost. This solution soaked the compost and excess solution was caught by saucers and then reabsorbed. The 1.5 kg of substrate in each pot resulted in mean activity concentrations of 17.8 kBq ^{99}Tc/kg and 15.8 kBq ^{75}Se/kg. During the addition of radiolabel plants were shielded to avoid external contamination. In each batch, after 48 h, all above-ground shoot material was harvested from 1 cm above the compost surface (to avoid plant contamination), dried and about 0.2 g digested in 5 mL nitric acid and 5 mL H_2O_2 until the reaction ceased. When cooled, 5 mL of digest was counted for γ-emissions from ^{75}Se and 100 μL added to 3 mL scintillant to count β-emissions from ^{99}Tc. In each batch, counts of ^{75}Se γ-emissions were decay-corrected to labelling date. After correcting for background counts in each batch, and using counting efficiencies of 0.9 for ^{99}Tc and 0.45 for ^{75}Se (determined by repeat measures of a standard prepared from stock), activity concentrations in CPM/kg were calculated for each sample (Supplementary Information 2). Activity concentration data were analysed using the 'R' platform.

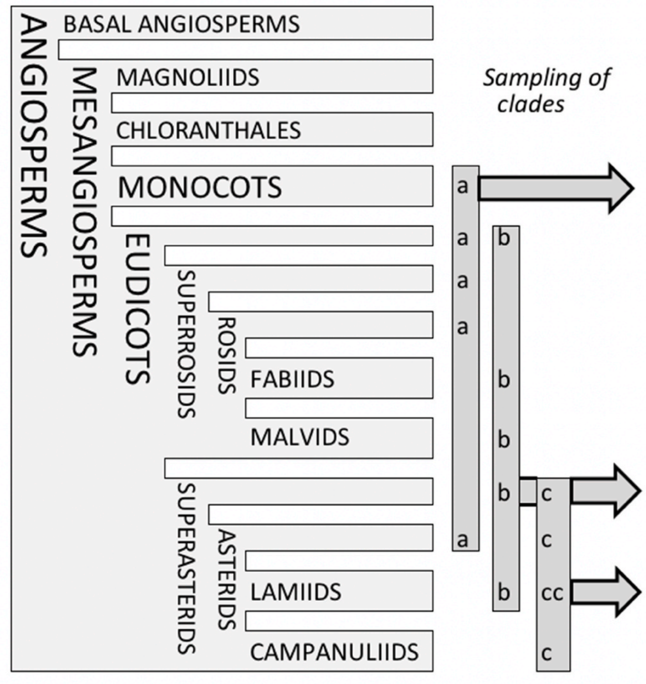

Fig. 1. Clade-based species-sampling regime from the Angiosperm Phylogeny Group IV phylogeny. For each set of species (a, b or c) one key species plus four others from other clades was chosen. In addition to these species, 4 more were chosen within the 'superorder', Order, Family and Genus of the key species on the clade to provide a clade-based set of samples with a hierarchy of relatedness (bold arrows). Two species on the Monocot clade ('a', focused on *Festuca gigantea*) did not grow well leaving the set of samples with 19 species but the Eudicot ('b', focused on *Silene nutans*) and Superasterid ('c', focused on *Salvia pratensis*) sample sets were complete with 21 species each to give 61 species in total. For full list of species see Supplementary Information 1.

The Geneious R8.1 software (https://www.geneious.com) was used to locate gene sequence information for species sampled and then to reconstruct a phylogeny for them. Sequences were found for 55 species for the *rbcL* gene. After multiple sequence alignment the Geneious Tree Builder was then used to build a phylogenetic tree which was exported as a *newick* file. The tree was not dated, and bootstrapping was not used to assess support for the branches. The *newick* file and the R 'phylosignal' library (designed to detect phylogenetic signals across species with determined phylogenetic relationships (Keck et al., 2016)) were used for analyses in the 'R' platform.

3. Results & discussion

The International Atomic Energy Agency (IAEA) defines soil-to-plant transfer factors (TFs) for any given element as 'the ratio of the dry weight concentration in the plants to the dry weight concentration in the specified soil layer' (IAEA, 2010). In quantifying TFs, account must be taken of, for example, equilibrium times and soil depth. The IAEA's compilations of TFs for a variety of crops and plants (IAEA, 2010, 2014), that are used around the world for predicting the contamination of crops and ecosystems, suggest that for 'grasses and herbs' geometric mean (GM) TFs of 10 for Tc (with GM Standard deviation = 2 and min-max 0.008–20) and 0.44 for Se (GMSD = 3.6, min-max = 0.009–12) (IAEA, 2014: Table 5). These can be adjusted for different soil conditions so that for ^{99}Tc, for example, Health Protection England has recommended TFs of 5 for oxidizing soils and 0.5 for reducing soils (Ewers et al., 2011). Different plant species have, however, long been known to have different uptake capacities for both Tc (Bennett and Willey, 2003; Willey et al., 2010) and Se (Kikkert and Berkelaar, 2013; White et al., 2007; Funes-Collado et al., 2013; Bitterli et al., 2010), with potentially significant impacts on the level of contamination of terrestrial food chains with ^{99}Tc and ^{79}Se but, in contrast to soil factors, there is currently no method to take account of plant factors in predicting TFs for Tc and Se. In order to help take account of plant factors in the prediction of TFs for ^{99}Tc and ^{79}Se in grasses and herbs, we report here results generated using statistical techniques previously used to detect taxonomic and phylogenetic signals in radionuclide transfer (Willey, 2014) and phylogenetic signals for biomonitoring and toxicology (Laras et al., 2014). We chose species from across the angiosperm phylogeny that are representative of plants that grow under the current and possible future conditions of the British Isles so that the predictions are of particular relevance to the UK's proposed GDF. Overall, the research focused on testing the hypothesis that there were significant taxonomic and phylogenetic influences on the uptake of ^{99}Tc and ^{79}Se by plants. It shows that the identification of such influences has the potential to improve the prediction of radionuclide TFs for groups of organisms and to can enable the prediction of TFs for species when no empirical data is available for them.

^{99}Tc and ^{79}Se are isotopes of long-term importance in nuclear waste and, together with isotopes of I and Cl, are amongst the most likely to break through to the surface from a repository (Piqué et al., 2013). ^{99}Tc is a long-lived relatively low energy β-emitter ($\lambda = 2.11 \times 10^5$ y; 263 keV emission energy), as is ^{79}Se ($\lambda = 2.95 \times 10^5$ y; 155 keV emission energy). Radiation doses to living systems from ^{99}Tc and ^{79}Se occur, therefore, primarily via internal exposure - so the primary environmental concern is their entry into food chains. Tc has no stable isotopes and there has been essentially no naturally occurring Tc for most of life's history (primordial ^{99}Tc decayed completely quite early in Earth's history and there have been, perhaps, only minor instances of naturally occurring nuclear fission since) but TcO_4^- in aqueous solution has physico-chemical similarities to several essential nutrient ions, e.g. MoO_4^-, NO_3^-. The most abundant natural stable isotope of selenium is ^{80}Se (abundance about 50% of natural stable Se), with there being almost no natural ^{79}Se. ^{75}Se is an anthropogenic primarily γ-emitting radioisotope (at 136 keV and 265 keV, with $\lambda = 0.327$ y). There is no evidence that plants discriminate significantly during uptake between Se isotopes so here, for experimental convenience, we used ^{75}Se as an analogue of ^{79}Se. Plants have no essential requirement for Tc or Se and no dedicated TcO_4^- or SeO_4^{2-} transporters but protein transporter systems in plants can retrieve the essential nutrient anions MnO_4^-, SO_4^{2-}, PO_4^{2-} and NO_3^- from aqueous solutions in significant amounts. It is not yet known which transport systems control the entry of TcO_4^- to plants but SeO_4^{2-} generally behaves as an SO_4^{2-} analogue in the physiology of plants (White et al., 2007).

The labelling procedure provided easily detectable activity concentrations in plant samples and reliably enabled the determination of the β-emitting ^{99}Tc and the γ-emitting ^{75}Se. In a concentration range up to mM concentrations plant uptake rates of elements can be concentration dependent but the absolute concentrations of ^{99}Tc and ^{75}Se used here, though different are both not only environmentally realistic but extremely low, so concentration-dependent effects are not expected. Mean ^{99}Tc and ^{75}Se activity concentrations of species ranged over approximately two orders of magnitude (269-52,000 Bq ^{99}Tc/kg dry weight and 4269-110,909 Bq ^{75}Se/kg dry weight; Fig. 2) and were essentially log-normally distributed (Supplementary Information 3). Geometric mean activities across all species were 8.51 kBq ^{99}Tc/kg (n = 280) and 37.7 kBq ^{75}Se/kg (n = 280). The log-normal distribution of concentrations is often reported for plant activity concentration data and for this reason geometric means of TFs are provided alongside arithmetic means in IAEA compilations (IAEA, 2010, 2014).

Activity concentrations of ^{99}Tc and ^{75}Se were correlated across the 61 species (Pearson's $r = 0.635$, $t = 13.696$, $df = 278$, $P < 0.001$, Fig. 2 inset, Supplementary Information 4). Correlations in activity concentrations between isotopes of elements with similar chemical properties in the environment have been reported before, for example ^{90}Sr/Ca (Burger and Lichtscheidl, 2019) and ^{137}/^{134}Cs and K, but the dataset reported here is, for the uptake of Tc and Se, the most wide-ranging yet reported in terms of species. The correlation between ^{90}Sr/Ca correlation has been very useful for predicting the contamination of crops and wild plant species but is stronger than that reported here for ^{99}Tc and ^{75}Se, which is more redolent of the looser ^{137}/^{134}Cs and K correlation. Selenium is not an essential nutrient for plants, but it is for animals - so its behaviour in the soil-plant system, especially in agricultural systems, is quite well known. The data reported here, depending on the strength of the correlation, might be useful for understanding the behaviour of not only ^{79}Se but also ^{99}Tc. This is important because the available data for ^{99}Tc are sparse compared to those for stable Se. Further investigations of the strength of the correlation between ^{99}Tc and ^{75}Se might, therefore, be radioecologically useful.

In the experiments reported here final activity concentrations of ^{99}Tc were, overall, less than those of ^{75}Se. The TF for soil-to-plant transfer of radioisotopes is best applied to instances in which soil and plant concentrations are at equilibrium (as far as it occurs) and is best understood as, overall, a representative value for transfer. The experimental protocol reported here was not intended to generate equilibrium conditions or values for TF databases but if the TF calculation is applied then the activity concentrations of ^{99}Tc and ^{75}Se produces 'TFs' of 0.46 for ^{99}Tc and 2.37 for ^{75}Se. The mean % removed of total radioisotope applied was 0.03% for ^{99}Tc and 2.08% for ^{75}Se – so for plant uptake the very small change in activity concentration of the substrate is unlikely to have produced significant concentration-dependent effects. Thus, the substrate and exposure time used here is one in which transfer of Tc is, in contrast to what happens in some other substrates or exposure times, less than that of Se. The concentration of sulphate in the compost may be associated with high uptake of ^{75}Se relative to ^{99}Tc because it may have prevented SeO_4^{2-} adsorption to exchange sites. Alternatively, the occurrence of sulphate, Tc, and Se together may have resulted in higher relative uptake of Se (Se, could, for example, be better at competing for sulphate transporters in the root plasma-membrane than is Tc in the compost used here under these conditions). Overall, the activity concentrations in plants are unlikely, given the short radiolabelling periods, to have reached an equilibrium concentration so discussions of absolute activity concentrations are of limited value - the focus of investigations reported here was a comparison of the relative values produced in different plant species under a particular set of conditions following acute exposure.

Acute exposure of roots to radioactive contaminants is an important potential route of soil-to-plant transfer, especially for radioisotopes of elements such as Tc and Se that often have low K_d values (distribution coefficient between soil and solution) meaning that they are soluble, mobile, and bioavailable in the regolith. Radioisotopes with higher K_d values in the soil-plant system (such as ^{137}Cs and ^{90}Sr which are important contaminants around the accident sites at Khyshtym, Chernobyl and Fukushima) and that have arrived in the soil via aerial contamination, sometimes as nuclear fuel particles, tend to remain in the upper layers of soil for extended periods of time (Beresford et al., 2016) and are more likely to produce chronic exposure. Isotopes such as ^{99}Tc and ^{79}Se which can form soluble ions have availabilities that are much more profoundly affected by transient hydrological and climatic regimes

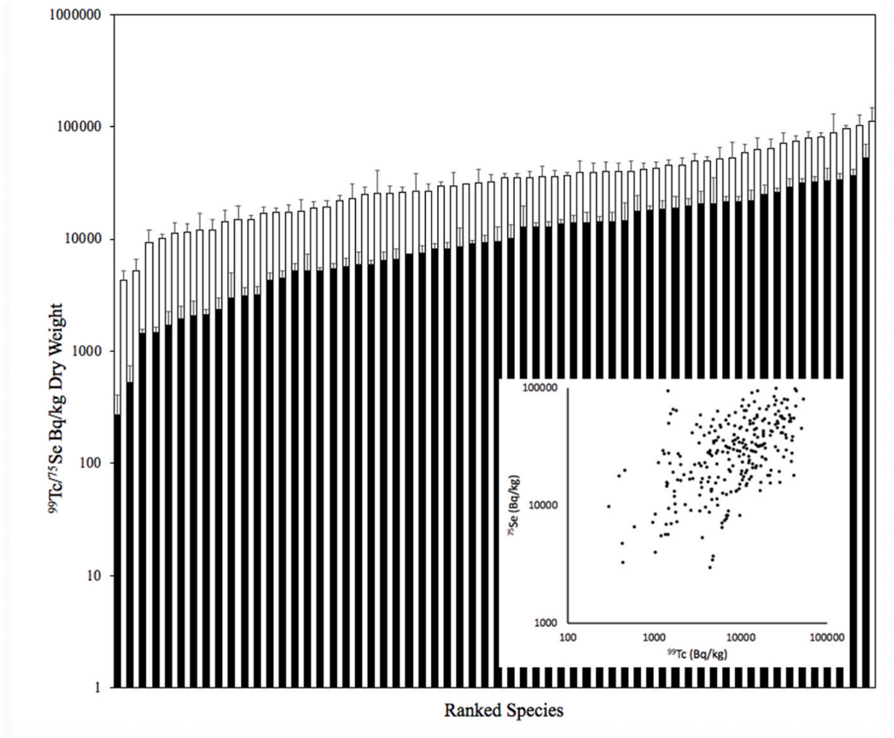

Fig. 2. Mean ± SE ($n = 5$) activity concentrations of ^{99}Tc and ^{75}Se (each ranked separately) for 61 species of plants selected from the flora of Britain and grown in pots under controlled conditions. Empty bars = Se, filled bars = Tc. Inset shows relationship between ^{99}Tc and ^{75}Se concentrations in the 61 species ($r = 0.635$, $P \leq 2.2 \times 10^{-16}$).

and by biological activities – all of which can produce short episodes of exposure. This is especially the case if the contamination route is not down from the surface but up from below. The potential importance of this exposure type has been noted for many years in studies of the highly soluble ^{36}Cl in buried radioactive waste (Smith et al., 2014). Thus, acute exposure of plants through the soil is useful for understanding the possible behaviour of the mobile nuclear waste components ^{99}Tc and ^{79}Se. Further, in the experiments reported here we provided acute exposures to herbaceous plants in the exponential phase of growth. It is during this phase of growth that the uptake of nutrient elements is most significant (Chen et al., 2012), so although the values reported cannot be used directly as TFs, they are likely to relate to the relative differences between species in uptake from a contamination scenario of importance to understanding the transfer of radioisotopes from nuclear waste.

Although the absolute values in Fig. 2 extend the species for which we have data for ^{99}Tc and ^{75}Se, further analyses are necessary if they are to be useful in situations other than those of the experiments reported here. We therefore focused on analysing taxonomic and phylogenetic influences in producing differences in the values. For Tc there are significant differences in the coefficients of variation (CVs) at different levels of the taxonomic hierarchy overall, and for all individual sample sets (Table 1). The M-SLRT test (command 'mslr' in R 'cvequality' library (Marwick and Krishnamoorthy, 2019) was used because it is robust to different sample numbers in each category. For each sample set we planted 5 replicates of 5 species at 5 different taxonomic levels. As shown in Supplementary Information 2, a few replicates did not grow sufficiently well for radiolabelling, so no sample set was an exactly balanced 125 samples - hence the use of the 'mslr' test. This sampling regime was designed to provide the activity concentration values for 5 species at each of 5 levels of the taxonomic hierarchy. For example, for sample set 'a', which was focused on monocotyledonous clades, uptake measurements were made for 5 species within the genus *Festuca*, then for 5 genera (including *Festuca*) within the family Poaceae, then for 5 families within the order Poales, and so on.

Overall, sample set 'a' included species that were the most taxonomically distant of the three sample sets (its hierarchical sampling included species from across both monocots and eudicots and then focused down a 'monocot' clade, Fig. 1) and had the most significant difference between CVs at the different taxonomic levels (Table 1, Supplementary Information 5). These results, though from a small proportion of all species, indicate that uptake of Tc by 5 different species is likely to vary more between 5 distantly related species than between 5 closely related species. This indicates that there are taxonomic, and potentially phylogenetic, influences in ^{99}Tc uptake across the species - if the trait (^{99}Tc activity concentration) was determined entirely on a species-by-species basis and each species acted independently for this trait, no differences in CVs would be expected at the different taxonomic levels. For ^{75}Se there were no statistically significant differences between CVs at different levels of the taxonomic hierarchy, either in sample sets or overall (Table 1). Previous analyses of the uptake of other radionuclides by plants have reported differences in uptake between taxonomic groups. In particular, differences in uptake between

Table 1
Table of Results from MLSR test for Differences in Coefficients of Variation of Tc and Se Activities Between Different Levels of the Taxonomic Hierarchy.

	Sample Set 'a' (*n=113, levels=5*)		Sample Set 'b' (*n=121, levels=5*)		Sample Set 'c' (*n=111, levels=5*)		All Samples (*n=345, levels=5*)	
	Test Stat	P value	Test Stat	P value	Test Stat	P value	Test Stat	P value
Tc	31.85	*2.50E-06*	10.09	*0.027*	14.48	*0.0059*	21.34	*0.00027*
Se	7.55	*0.11*	3.16	*0.53*	6.32	*0.18*	9.26	*0.055*

Monocots and Eudicots have been noted for ^{99}Tc (Willey et al., 2010), together with differences between particular Orders and Families of plants. Taxonomic differences in the accumulation of metals by plants have long been known (e.g. Cappa and Pilon-Smits, 2014). Ultimately, these taxonomic differences are most useful if they can be ascribed statistically to a known phylogeny. This requires techniques targeted to find phylogenetic influences in data from a sample of taxa.

A phylogenetic influence can be manifest when closely related taxa have more similar values of a trait than do distantly related taxa (Münkemüller et al., 2012). For many ecological studies, it is important to take account of phylogenetic influences in traits because they reveal that taxa are not behaving independently, but phylogenetic signals are also increasingly being used in phylogenetically-based trait prediction (PTP). For example, for microbes, in which there is often an analogous situation to that of radionuclide uptake by plants (i.e. quantitative data for a trait is only available for a small proportion of all taxa) PTP has been suggested as a basis for predicting a range of traits (Goberna and Verdú, 2016). Phylogenetic analyses of activity concentrations of ^{99}Tc and ^{75}Se in the plant species revealed a significant phylogenetic signal in activity concentrations for ^{99}Tc (Blomberg's $K = 0.319$ ($P = 0.009$), Fig. 3) but not for ^{75}Se. The phylogeny used for analyses here provided an estimate of the evolutionary distances between the species based on sequences from a single gene. Other genes, or consensus phylogenies based on multiple genes, are likely to provide improved estimates of the relationships between the species, but the data reported here provide, for the first time, a set of plant concentration values for ^{99}Tc and ^{75}Se for species together with their estimated phylogeny and suggests that there is a phylogenetic signal in ^{99}Tc uptake. The data and analyses presented here are, we suggest, the best evidence yet that there might be phylogenetic influences on plant uptake of some radionuclides. The ^{99}Tc and ^{75}Se activities in Fig. 2 show a general positive relationship but the hierarchical distribution of variance (Table 1) is stronger for ^{99}Tc so the signal for ^{75}Se, in a data set with considerable variance, has not met the statistical threshold for a phylogenetic signal. Overall, it seems that both soil and plant factors can affect the absolute value of TFs for ^{99}Tc and ^{75}Se and that taxonomy and phylogeny can be used as a proxy to predict some of the influence of plant factors on the transfer of ^{99}Tc.

The IAEA publishes a Technical Report Series (TRS) that includes compilations of soil-to-plant TFs. TRS 479 includes TFs for wildlife in natural terrestrial systems and TRS 472 includes TFs for agricultural systems (IAEA, 2010, 2014). These compilations, based on the best available data, are used by regulators worldwide as a guide to soil-to-plant transfer of radionuclides. For Tc and Se, the compiled mean TFs are based on the relatively few available data points and have some uncertainty, and for many agricultural and almost all wild plant species there are no data. The analyses reported here for Tc and Se transfer provide an opportunity to improve the prediction of transfer for ^{99}Tc and ^{75}Se by reducing uncertainty and filling gaps in compilations of TF.

The analysis of CVs at different levels of the taxonomic hierarchy (Table 1) suggests that there are some systematic differences at high levels, i.e. between major categories, for ^{99}Tc. This is potentially useful because the major categories into which almost all plant species are classified are widely agreed. In general, the geometric mean, if there are sufficient data for it to be calculated, is probably the more useful of the two mean values (arithmetic and geometric) provided for TFs in TRS 479 because in large data sets TFs tend to be log-normally distributed - as is the case with the data reported here. Using the TRS 479 geometric means for 'grasses and herbs' (10 and 0.44 for Tc and Se respectively) the activity concentrations in plants reported here can be converted into a set of predicted TFs for 61 species with geomeans of 10 and 0.44 (; Supplementary Information 5). If these TFs for Tc are categorized into major groups, they have statistically significantly different geometric means and geometric standard deviations that are smaller than those listed in TRS 479. Thus, we suggest that the recommended TF for soil-to-plant transfer of Tc could be split into four separate geometric means depending on the taxonomic position of a species (Fig. 4a). This provides improved estimates of TF for species in these groups and a method for filling gaps for species whose TF has not yet been measured – a value based on taxonomic group will generally provide a better estimate than the current single average in the IAEA compilations. If the same data conversion process is followed for Se it is, despite the lack of taxonomic effects, useful because it provides the basis (based on a purposefully wide sample) for reducing the geometric standard deviation around the geometric mean (Fig. 4b).

For prediction in agricultural systems (TRS 472) crop plants are divided up into groups and the foodstuffs they produce into plant compartments. The compiled TFs for Tc for these compartments, where they exist, are reproduced (from table 18 in TRS 472) in Table 2, together with new values and SDs converted to TFs using the values for maize leaves (because it is based on the highest number of samples), and the effects identified above. It is suggested that to estimate TFs for stems and shoots, using the same values for cereals and grasses (forage) as for maize, and the lower SD shown on the table for all the aforementioned monocots, is appropriate until more detailed measurements become available. For rice caution is necessary because the anaerobic conditions it is often grown in reduce Tc TFs considerably. For leafy vegetables, leguminous fodder, pasture and the leaves of herbs and other crops the differences between Rosids and other eudicots is used to suggest, for the first time, TFs for transfer of Tc to particular foodstuffs. There are no values for estimating the transfer of radioactive Se in TRS 472 but the overall difference in Tc and Se TFs in TRS 479 for a variety of plants, and the correlation between Tc and Se revealed in Fig. 2, suggests a basis for making them.

The case for authorising the construction of a geological disposal facility for nuclear waste necessitates predicting the pathways radioisotopes might take in the environment if they ever reached it. At present, the IAEA TRS publications provide a widely accepted foundation for such predictions. However, for many radionuclides they provide data based on a small proportion of all species and often with significant uncertainty. The research reported here provides some direct suggestions for reducing uncertainty and for filling data gaps for the ^{99}Tc and ^{79}Se that will dominate much nuclear waste in the medium term. It is also an approach that might be useful to pursue for other radionuclides and organisms. In addition to the control exerted by soil factors on the availability of many radionuclides, a number of plant factors influence soil-to-plant transfer. Of these plant factors the one for which most information is likely to be available without requiring measurement is their taxonomy and phylogeny, especially at higher taxonomic levels. Thus, treating TF as a trait suggests that taxonomically and phylogenetically informed trait prediction could be useful in studies of radioisotopes in the environment. Further, it is frequently noted in radioecology that understanding the behaviour of radioisotopes can be very revealing about the behaviour of stable isotopes. In the data reported here the phylogenetic signal is stronger for Tc than for Se, and Tc is a less close chemical analogue of a plant nutrient than Se is. Previous studies of taxonomic effects in element concentrations in plants have also noted that phylogenetic effects can be stronger for contaminant elements with no nutrient analogues than for those which do behave as nutrient analogues. Further work might include an assessment of whether contaminant elements without nutrient analogues, whose entry into plants is probably dependent on accidental plant characteristics, differ more in the concentrations to which they accumulate than contaminant elements with nutrient analogues, which may be subject to the homeostatic concentration controlling mechanisms evolved for their nutrient analogues.

Nuclear fission produces waste material with many short-lived radioisotopes but after a few years (the 'cooling period'), in spent nuclear fuel the fission products ^{137}Cs and ^{90}Sr dominate total activity for a few hundred years. Some actinides are, as a result of neutron bombardment in fuel, also present in nuclear waste (e.g. $^{239/240}$Pu, ^{241}Am) and are important components of total activity after a few hundred years. However, in the medium-term ^{99}Tc and ^{79}Se will be amongst the most

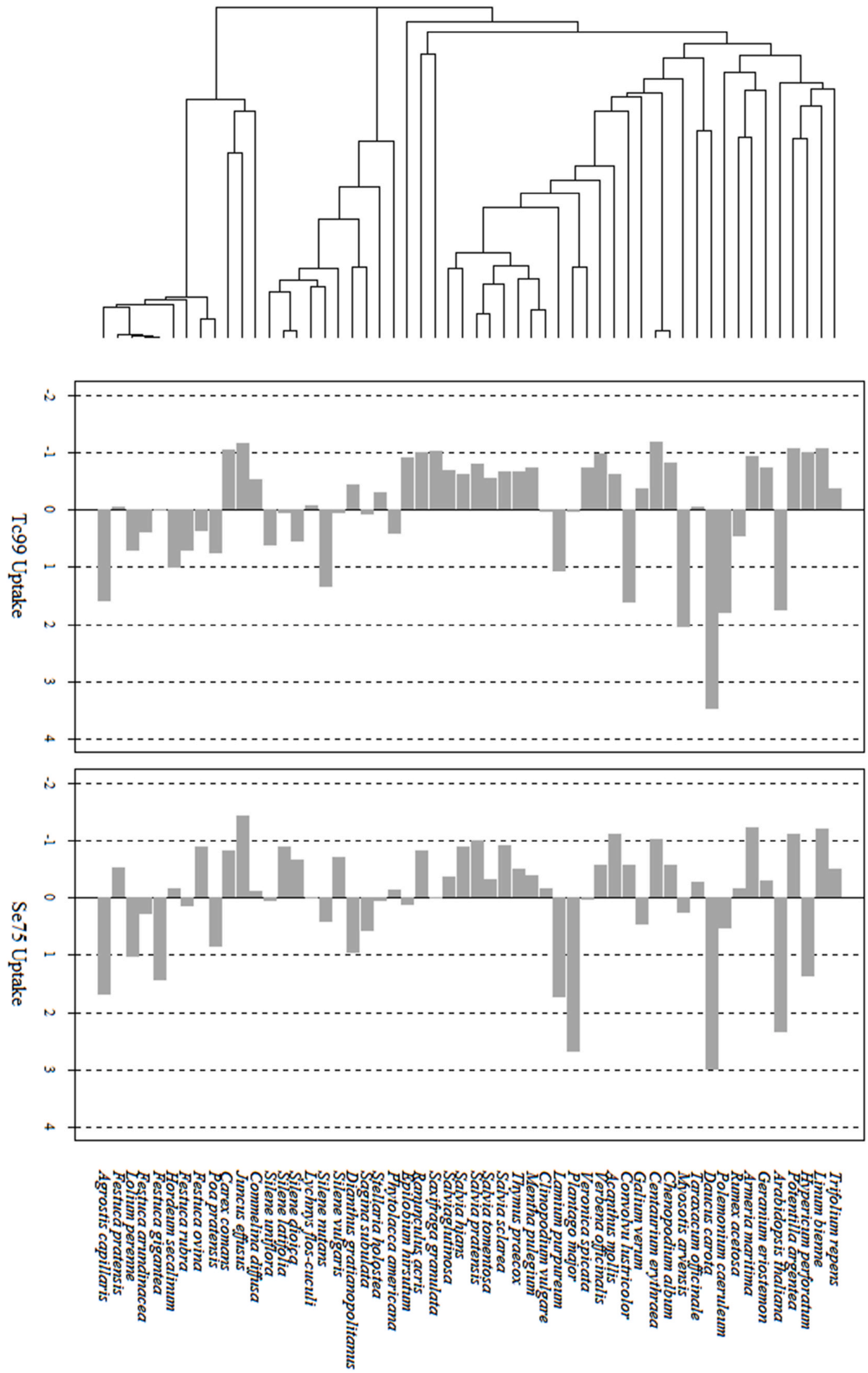

Fig. 3. Phylogenetic analysis for ^{99}Tc and ^{75}Se uptake by 55 plant species. The phylogeny was generated in the Genious software using multiple sequence alignment of *rbc*L gene sequences. Signals were modelled in the 'phylosignal' library on 'R' using a Brownian Model (BM) of evolution. For ^{99}Tc for phylogenetic signal was statistically significant (Blomberg's K = 0.319, P = 0.009).

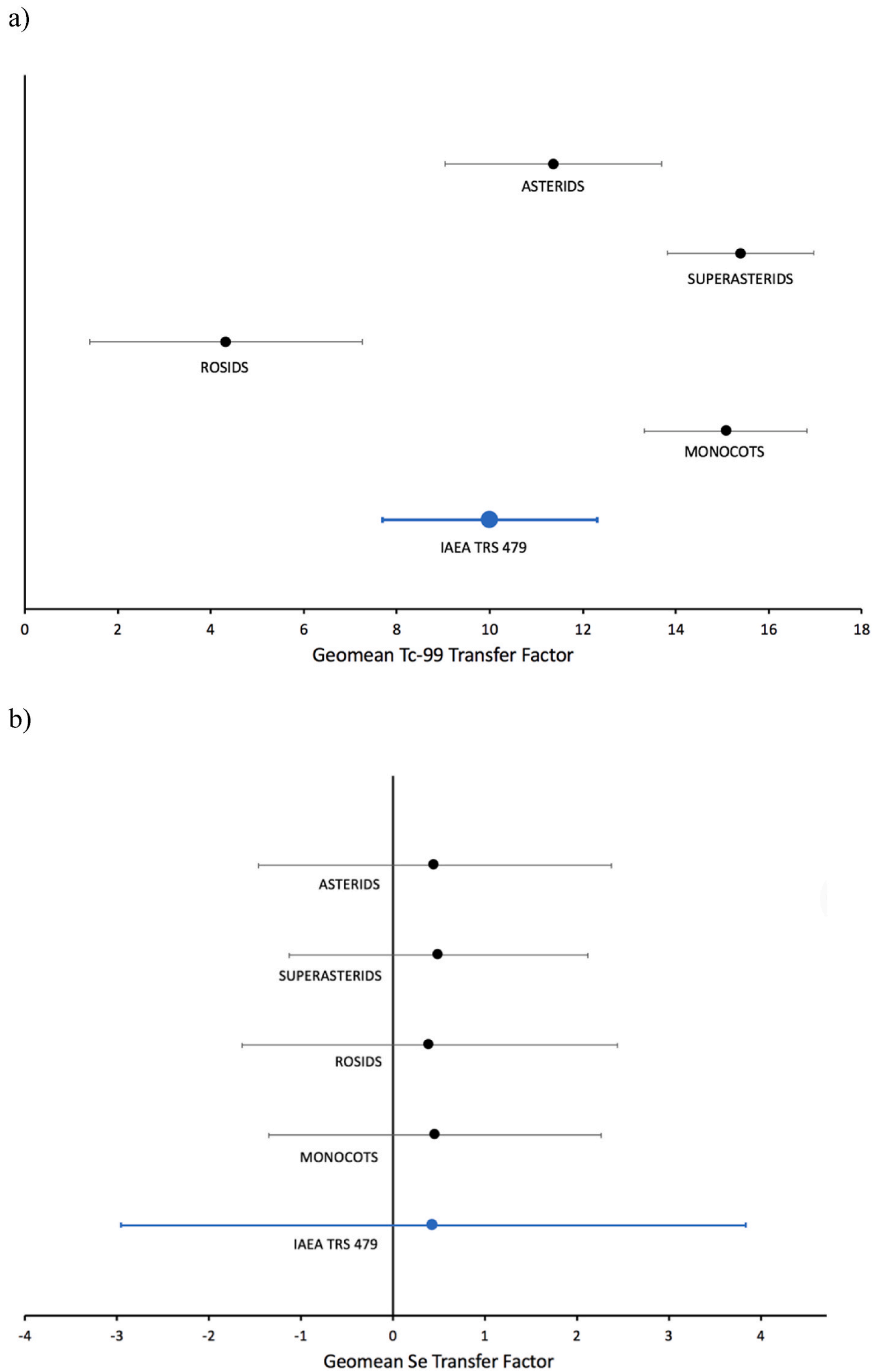

Fig. 4. Suggested improvements in recommended TFs for Tc and Se. Current recommendations are in blue, improved suggestions in black and distinguished by phylogenetic groups. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

important isotopes in nuclear waste – a period in which sustained integrity of GDFs becomes important and possible releases of radionuclides to the environment is a consideration. A global annual power output from nuclear generation of between 2200 and 2600 TWh has been sustained for at least the last 20 years and there are now 416 nuclear reactors generating electricity with 59 currently being built or planned (https://pris.iaea.org/PRIS/home.aspx, accessed 31/5/24). The geological disposal of all the high-level nuclear waste produced during the nuclear age is only just beginning and will be a major undertaking during the next several decades. The results reported here might be useful for environmental assessments for geological disposal facilities, whilst the insights they provide might be of general use in radioecology.

Notes'

The authors declare no competing interests.

Table 2
Suggested Geometric Mean Tc TFs For Crop Groups and Crop Compartments Based on Phylogenetic Effects on Tc Transfer (non-shaded suggested = stay the same, shaded suggested = new).

Crop Group	Crop Compartment	TRS472 TFs Mean TF	TRS472 TFs SD	Suggested TFs Mean TF	Suggested TFs SD	
Cereals	Grains, seeds and pods	1.3		1.3		
	Stems and shoots			6.4	0.44	
Maize	Grains, seeds and pods	3.8	8.2	3.8	8.2	
	Stems and shoots	**6.4**	3.30	6.4	0.45	
Rice	Grains, seeds and pods					
	Stems and shoots					
Leafy vegetables	Leaves	180		180	29.81	Non-Rosids
				58.1	46.40	Rosids
Non-leafy vegetables	Fruits, heads, berries and buds					
Leguminous vegetables	Grains, seeds and pods	4.3	5.2	4.3	5.2	
Root crops	Roots					
Tubers	Tubers					
Fruits	Fruits, heads, berries and buds					
Grasses (cultivars)	Stems and shoots			6.4	0.44	
Leguminous fodder (cultivars)	Stems and shoots			4.4	3.51	
Pasture (mixture)	Stems and shoots	76	3			
Herbs	Leaves			180.0	29.81	Non-Rosids
				58.1	46.40	Rosids
	Grains, seeds and pods					
	Fruits, heads, berries and buds					
Other crops	Grains, seeds and pods					
	Leaves			180.0	29.81	Non-Rosids
				58.1	46.40	Rosids
	Stems and shoots					
	Fruits, heads, berries and buds					
	Roots					
	Tubers					

CRediT authorship contribution statement

Eleni Siasou: Investigation, Methodology, Validation. **Neil Willey:** Conceptualization, Project administration, Supervision, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was carried out as part of the UK Natural Environment Research Council's (NERC) Radioactivity and the Terrestrial Environment (RATE) programme which was part funded by Radioactive Waste Management Ltd and the Environment Agency of England. Grant No NE/L000342/1.

We would like to thank Francois Keck of the Department of Aquatic Sciences, Swedish University of Agricultural Sciences, Uppsala, Sweden for his comments on the manuscript.

We would like to thank Alison Halliday and Daisy Hunt of the School of Life Sciences, University of the West of England, Bristol for support with, respectively, laboratory work, and producing Fig. 3.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2025.107622.

Data availability

The data is available from Environmental Information Data Centre (EIDC) of the UK Natural Environment Research Council (https://doi.org/10.5285/e5122291-b5e2-47de-bf14-db67125f4c14).

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