# Soil to plant transfer of radionuclides: predicting the fate of multiple radioisotopes in plants

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Neil J. Willey

Centre for Research in Bioscience, University of the West of England, Bristol BS16 1QY, UK

ABSTRACT

Predicting soil-to-plant transfer of radionuclides is restricted by the range of species for which concentration ratios (CRs) have been measured. Here the radioecological utility of meta-analyses of phylogenetic effects on alkali earth metals will be explored for applications such as 'gap-filling' of CRs, the identification of sentinel biomonitor plants and the selection of taxa for phytoremediation of radionuclide contaminated soils. REML modelling of extensive CR/concentration datasets shows that the concentrations in plants of Ca, Mg and Sr are significantly influenced by phylogeny. Phylogenetic effects of these elements are shown here to be similar. Ratios of Ca/Mg and Ca/Sr are known to be quite stable in plants so, assuming that Sr/Ra ratios are stable, phylogenetic effects and estimated mean CRs are used to predict Ra CRs for groups of plants with few measured data. Overall, there are well quantified plant variables that could contribute significantly to improving predictions of the fate radioisotopes in the soil-plant system.

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## 1. Introduction

Predicting the soil-to-plant transfer of radionuclides is important to many assessments of radionuclide fate in terrestrial ecosystems. Soil availability of radionuclides contributes significantly to the magnitude of their transfer to plants but there are many plant factors, from molecular to ecological scale, that also help to determine transfer. For most radionuclides these plant factors are, in general, less well understood than the factors controlling availability from soil. Understanding the influence of plant factors on soil-to-plant transfer of radionuclides might help the development of techniques that can predict radionuclide transfer to species for which there are no measured transfer factors at present (Willey, 2010). It might also help clarify the effects of soil and other environmental variables on transfer by providing an explanation for a significant proportion of the variation in soil-to-plant transfer of radionuclides.

Previous research has shown that there are phylogenetic influences on the transfer of radionuclides from soils to plants (e.g. Willey et al., 2010; Willey et al., 2005). Phylogenetics studies the evolutionary history of groups of organisms. The rapid increase in both molecular data and the power of computational techniques has enabled new phylogenies to be described for many groups of organisms including plants. These phylogenies are very useful in comparative biological studies, i.e. in attempts to gain insight about fundamental biological characteristics by comparing them in different taxonomic groups. A molecular-based plant phylogeny for use in comparative biology was first proposed by Soltis et al. (1999). This has continued to be updated by the Angiosperm Phylogeny Group who have constructed one of the most widely accepted phylogenies of flowering plants (Bremer et al., 2003, 2009). These new phylogenies, and taxonomies derived from them, now provide a framework for many comparative analyses in plant biology. One such analysis is that of element concentrations in plants, including elements with radioisotopes.

Previous investigations have suggested that there are phylogenetic effects in plant uptake from soil of several heavy metals (Broadley et al., 2001), numerous elements (Watanabe et al., 2007) and many radionuclides including those of Cs (Broadley et al., 1999; Willey et al., 2005), Cl (Willey and Fawcett, 2005), Sr (Willey and Fawcett, 2006a), Ru (Willey and Fawcett, 2006b), S (Willey and Wilkins, 2006), Co (Willey and Wilkins, 2008) and Tc (Willey et al., 2010). Analyses of phylogenetic and taxonomic effects depend on large datasets of element concentrations in plants. Particularly for radionuclides there are few datasets that provide comparative species data, i.e. data from species studied under the same conditions, for more than 5 or 6 species. In all the aforementioned analyses of taxonomic effects residual maximum likelihood (REML) modelling was used to construct datasets of

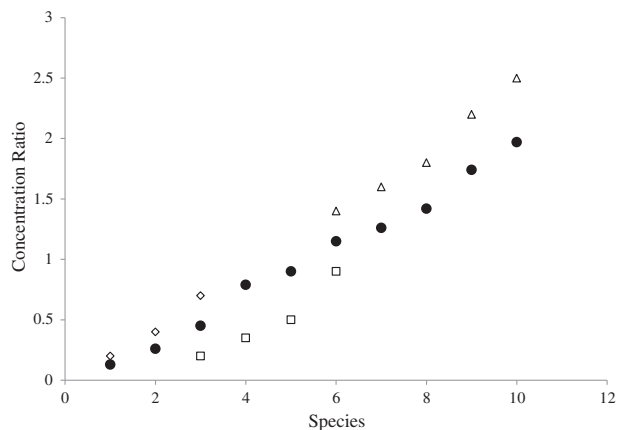E-mail address: Neil.Willey@uwe.ac.uk.

**Fig. 1.** An illustration of the REML modelling technique. Three different datasets of concentration ratios (open symbols) have, overall, different means because of differences in the environment that the plants grew in. If there are species in common between datasets, perhaps because they overlap or because datasets can be produced that link them, the REML procedure can minimise differences due to, for example soil type, and make a prediction (closed symbols) for the relative CRs for all the species.

estimated relative concentrations in many species (Fig. 1). The iterative REML procedure can minimize the influence of one factor on variation in measured values in order to reveal the influence of another factor. In the procedure these are termed the 'random' and 'fixed' factors respectively. In the aforementioned analyses the random factor was 'study' and the fixed factor was' species'. Classifying data by 'study' included all the soil and environmental variables that might have contributed to determining the element concentration in plants. REML modelling across several datasets requires that they have values for the fixed factor in common. In most instances data for plant concentrations or activities was generated to overlap with species data from the literature. REML analyses of these datasets were used to minimize the influence of soil and environmental variables and thus provide estimates of relative values in many species. Full details of the analysis are provided in Willey (2010).

Data analysed for phylogenetic effects in element concentrations in plants has focused on concentrations in green shoots. Although there are many plants with significant woody parts and the element concentrations in roots also contribute to ecosystem flows of elements, green shoots often have the most dynamic role in the flows of nutrients and elemental contaminants in ecosystems. Many radionuclides and elements have been shown separately to have low uptake by monocotyledonous plants as compared to eudicot plants. For some elements there are significant differences between the commelinid and non-commelinid monocot groups (Broadley et al., 2003). For some elements there are significant differences between the Asterid and Rosid groups but for several elements with important radionuclides including Cs, Co, Sr and Tc, plants on the Caryophyllid clade have the highest relative activities (Willey et al., 2005; Willey and Fawcett, 2006a, b; Willey and Wilkins, 2008). Although a proportion of the difference between concentrations of elements in plants is related to phylogeny, its contribution to these differences is not the same for different elements. For alkaline earth metals like Ca and Mg the proportion of the differences between species related to phylogeny is in excess of 60% (Broadley et al., 2003), whereas for some metals less than 20% is related to phylogeny (Broadley et al., 2001). It is also possible that there is an interaction between environmental variables and phylogenetically constrained differences, perhaps most obviously with differences in the concentrations of elements that plants are exposed to. This was recently tested in herbage from the

Park Grass Experiment at Rothamsted UK in which hay meadows have been exposed to different fertiliser regimes for well over a century. For those species that inhabited all of the great variety of meadows produced by differing fertiliser regimes, principle components analysis for their concentrations of 17 elements showed that plant family explained a significant proportion of differences (White et al., 2012). Plants ionomes, i.e. their elemental concentrations, are an increasingly important area of study for understanding the flows of nutrients in agricultural and natural ecosystems. The evidence that plant ionomes are, at least in significant part, constrained phylogenetically is increasing, suggesting that they have predictable biodiversity (Willey, 2012).

Fundamental understanding of the biodiversity of plant ionomes might be very useful for predicting the fate of radionuclides. For the vast majority of plant species there are no measurements of concentration ratios (CRs). Databases of CRs for major crop plants are extensive and thus provide, for major soil types at least, quite robust estimates of the partitioning of important radionuclides between soil and plant. Data is, however, much less extensive for the many minor crop plants that are often of great significance to critical groups. For almost all wild plants, which have to be demonstrably protected from the effects of radioactive contamination in an increasing number of nations, CRs are non-existent. Methods that can predict CRs in any plant might, therefore, make a significant contribution to radiological protection. In addition, species selection can be very important in environmental monitoring programmes and in phytoremediation schemes — both of which might be guided by the phylogenetic constraints on transfer. Further, phylogenetic constraints are really a reminder that there is no *a priori* reason why species should be thought of as the origin of difference in CR — the species is a reproductive unit and there is no particular reason to think that fundamental ionomic differences vary just at the species level. Here I report an analysis for some alkali earth metals, including Sr, which reveals they all have quite similar phylogenetic constraints on their transfer. I suggest that for alkali earth metals at least general predictions of the fate radionuclides in the soil-plant system for a wide variety of plant species can be made using recent plant taxonomies and stoichiometric ratios.

## 2. Methods

Datasets were derived from previous publications that had used REML modelling to compile estimates of relative concentrations of Ca, Mg and Sr (Broadley et al., 2003, 2004; Willey and Fawcett, 2006a). Before REML modelling raw concentrations or CRs were $\log_e$ transformed. This provided data that was normally distributed for modelling in the original analyses. For analyses reported here the REML-estimated relative species means of $\log_e$ transformed data were selected because they were approximately normally distributed rather than the exponents of the transformed means which, although they convert data back to scales relevant to concentrations, are approximately $\log_e$-normally distributed. Data for all the species reported in Broadley et al. (2003, 2004) for Ca and Mg, and for Sr in Willey and Fawcett (2006a) were included in analysis.

The estimated relative mean concentrations of Ca, Mg and Sr were standardized to give a mean of zero and a standard error of 1. Running means for $n = 10$ were then calculated. Transformed means were then plotted in the order of the taxonomies used in the original publications that were based on Angiosperm Phylogeny Group I phylogeny (Soltis et al., 1999). The data for Ca, Mg and Sr each included a large dataset generated under a single set of conditions in the greenhouse plus a compilation of numerous literature reports of CRs. The datasets generated in the greenhouse were used to link literature studies and provide phylogenetic spread. In total for Ca $n = 206$ species, Mg $n = 117$, and for Sr $n = 155$. There was