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Confidence
Coping with uncertainty for improved modelling and decision making in nuclear emergencies

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D9.15 — An evaluation of process-based models and their application in food chain assessments

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Abstract

Vegetation interception and the time of year will largely determine the contamination of foodstuffs immediately following an accidental release. During the transition phase, uptake of radionuclides by vegetation from soil will increase in importance and root uptake will dominate during the long-term rehabilitation phase. Predictions made using radioecological models will be used to make long-term decisions, e.g., with regard to remediation strategies.

Models must be sufficiently robust and fit for purpose with uncertainties reduced where practicable. Most radioecological models use empirical transfer factors to estimate soil-to-plant transfer of radionuclides and these do not credibly cope with variation in root uptake caused by variation in soil properties. Consequently, process-based soil-to-plant models were developed to predict radionuclide (predominantly radiocaesium) transfer based upon relatively readily available soil properties.

The model originally developed by Absalom et al. (1999) and further improved by Absalom et al. (2001) and Tarsitano et al. (2011) is a typical example of a process-based transfer model. It has been applied to predict radiocaesium transfer under a range of environmental conditions with varying degrees of success.

The objective of the CONFIDENCE project's Work Package 3 (WP3) was to improve the capabilities of radioecological models used to predict activity concentrations in foodstuffs and to better characterise, and where possible, reduce uncertainties. The focus of this deliverable is to consider the use of process based models for post-accident predictions. We begin with assessing the applicability of the 'Absalom model' to a range of European soil and plant types that were not included in its initial parameterization. We also demonstrate how the model can be incorporated into the Food Chain and Dose Module for Terrestrial Pathways (FDMT), which is part of the European decision support system, JRODOS. To date, most consideration in the development of process-based transfer models has been focused on radiocaesium. In this deliverable we develop process-based soil-plant transfer models for radiostrontium. Finally, the deliverable reports on end-users views of process-based models.

Our assessment of the Absalom model shows that it is a useful tool for predicting radiocaesium transfer to the human food chain. Its predictions for grass and radish (edible root) were mostly within an order of magnitude of the measurements for most of the study soils. We recommend expanding the model database by considering more soils (with different mineralogies) and plant types in its parameterisation. The Absalom model has successfully been incorporated into the FDMT.

We have successfully established two process-based models to predict strontium concentrations in a range of crops using relatively few soil parameters and the calcium concentration in crops as inputs. To support these methodologies we have produced a collation of calcium concentrations in crops consumed by humans and farm animals (Chaplow et al., submitted). The approach removes the need for empirical concentration ratios and is able to make predictions for crop types for which no radioecological data exist.

Whilst the approaches developed for strontium produced predictions that compared well with measured data, and better than predictions using the commonly used concentration ratio approach, they require further testing against a wider range of soil types and crops.

A weakness of the strontium approaches developed is that they can only be used to make equilibrium predictions. However, they would be sufficient to aid the identification of longer-term 'at risk areas' in the event of an accidental release. The models could be used to estimate parameters to replace existing concentration ratios in models such as FDMT, which would enable their application in dynamic predictions. However, it would be preferable for future studies to consider trying to parameterise dynamic processes in soils within these process-based approaches.

CONTENTS

1 Introduction

Following an accidental release of radioactive substances into the environment, food production systems (e.g. pasture and agricultural lands) may become contaminated for a long time. Therefore, reliable prediction of radionuclide transfer to the food chain from contaminated lands is essential for effective protection of the public from exposure to harmful radiation through consumption of contaminated foodstuffs.

The Chernobyl accident highlighted that some areas may be more 'sensitive' or 'vulnerable' (e.g. have comparatively high transfers to foodstuffs or contribute relatively high fluxes of radionuclides to the public via contaminated foodstuffs) to radiological contamination than other areas. Vulnerability depends upon, for instance, soil type, farming practices and local consumption habits (e.g. Desmet and Myttenaere, 1988; Howard, 2000; Howard et al., 2002; Wright et al., 2002). Because of such factors, predictive models should be able to cope with spatial and temporal variation in radionuclide transfer.

Commonly used models to predict radionuclide activity concentrations in human foodstuffs tend to use empirical soil-to-plant transfer factors (also known as soil-plant concentration ratios) to describe the transfer of radionuclides from soil to crops (e.g. Brown and Simmonds, 1995; Brown et al., 2018). Such models cannot easily cope with variation in root uptake caused by variation in soil properties. In the 1990's – 2000's, semi-mechanistic, or process-based models were developed that could be implemented spatially and predict radiocaesium (RCs) transfer based upon relatively readily available soil properties (e.g. pH, soil organic matter content (OM), clay content, exchangeable potassium) (e.g. Absalom et al., 2001; Gillett et al., 2001; Wright et al., 2003). As well as being able to make predictions based upon local soil properties, the model described by Absalom et al. (2001) could also be used to predict the impact of K-fertilisation as a remediation measure. Consequently, the model was included within a decision support system (Cox et al., 2005).

Following the Fukushima accident, there was interest in applying the model, as presented by Absalom et al. (2001), to predict RCs transfer for impacted areas in Japan. However, the model tended to over-predict RCs retention in Japanese soils and hence underestimate the likely uptake by crops (Uematsu et al., 2015, 2016; Almahayni et al., 2019).

To date, most consideration in the development of process-based soil-to-plant transfer models has been focussed on RCs. Of the other radionuclides, which might be released following a nuclear accident, ^{90}Sr is the one that may be of long-term concern. For instance, ^{90}Sr is present in soils within the Chernobyl exclusion zone at concentrations approaching those of ^{137}Cs (Kashparov et al., 2001). In areas contaminated by releases from the Mayak facility (Russian Urals), ^{90}Sr is a major contributor to dose (Akleyev et al., 2017; Tolstykh et al., 2017).

The Strategic Research Agenda for radioecology (Hinton et al., 2013) prioritised further work on process-based models, stating that: '*By making the models more process-based, we expect (i) a significant reduction in model uncertainty; (ii) a better quantification of environmental variability; (iii) identification of the most influential parameters; and (iv) improved modelling tools capable of predicting radionuclide exposure to humans and wildlife under a variety of conditions, thereby enhancing the robustness of both human and wildlife assessments of exposure to ionising radiation*'. However, whilst offering an approach to reduce the uncertainties associated with empirical, ratio-based models, process-based models have not been adopted for application in emergency planning/management.

In this report, we will:

i) revisit process-based soil-plant models for RCs assessing their applicability to a range of European soil types and crops;
ii) develop process-based soil-to-plant transfer model for radiostrontium;
iii) demonstrate how process-based models can be incorporated into decision support systems;
iv) report on end-users views of process-based models.

2 Radiocaesium soil-to-plant transfer

2.1 Review of modelling approaches

Several RCs soil-to-plant transfer models have been developed and published over the years. Within CONFIDENCE WP3, Almahayni et al. (2019) reviewed these models and assessed their fitness for the purpose of emergency preparedness and response. Specifically, the ability of existing soil-to-plant transfer models to aid decision makers regarding identification and targeting areas vulnerable to RCs deposition and implementation of long-term countermeasures. Existing RCs transfer models belong to the following broad approaches: empirical, semi-mechanistic and mechanistic.

2.1.1 The empirical approach

Empirical transfer models are simple equilibrium concentration ratios (CR), or transfer factors (TF), that relate RCs concentration in plant biomass to RCs concentration in soil. This empirical approach is practical since it only requires RCs concentration in soil as an input and a TF value. Transfer factors have been determined for a wide range of soils and plant species. For instance, the International Atomic Energy Agency (IAEA) maintains and periodically updates a large compendium of TF values (e.g. IAEA, 2009, 2010). Where possible, transfer factors in the IAEA compendium are grouped according to soil texture (i.e. sand, loam and clay) and plant types (e.g. grass, leafy vegetables, root crops, etc.) with the aim of facilitating the selection of the most appropriate value.

Although simple and practical, the empirical approach predictions for a given soil-plant may vary by up to four orders of magnitude (Figure 1). The TF predicts plant uptake of RCs from soil based on total activity concentration in soil, which does not represent the `bioavailable` pool of RCs in soil. A proportion of the total RCs in soil is strongly fixed within soil minerals (e.g. certain types of clays) and is unavailable for plant uptake. Furthermore, the empirical approach does not consider soil pH, OM content or exchangeable potassium (typically influenced by local agricultural practices). These soil parameters strongly affect RCs bioavailability. Other parameters which may affect RCs transfer to plants include chemical speciation of RCs (form deposited or used in a controlled study), time between measurement and contamination, plant cultivar and agricultural practices. Additionally, most existing TF data are appropriate for temperate regions, whereas those appropriate for tropical and arid regions are limited.

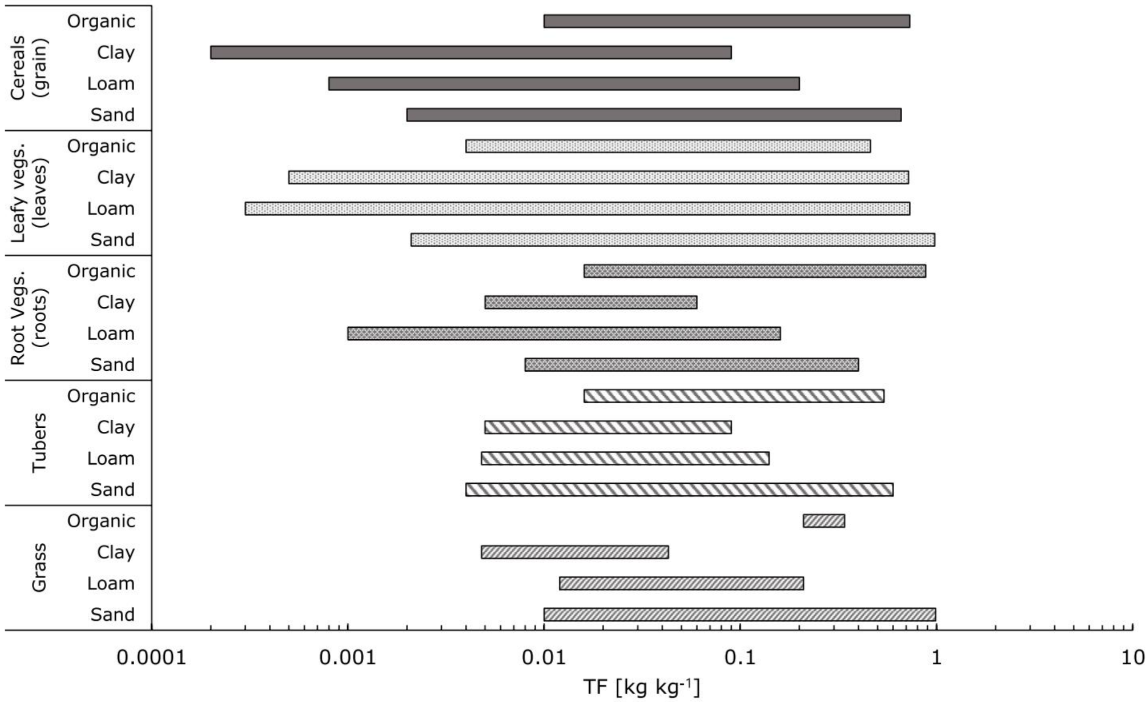


Figure 1 Variation in RCs TF for common soils and plant types (IAEA, 2010). Note logarithmic scale on x-axis.

2.1.2 The semi-mechanistic approach

Unlike the empirical approach, the semi-mechanistic approach accounts for influential soil parameters such as pH, OM content, clay content and exchangeable potassium. The model of Absalom et al. (1999, 2001) and its revised version (Tarsitano et al., 2011) are typical examples of the semi-mechanistic approach. They predict RCs transfer based on RCs deposition and soil pH, organic matter, clay content and exchangeable potassium.

Because the 'Absalom' model includes soil parameters and processes, it is suitable for predicting RCs transfer from soils with differing physical and chemical characteristics. It is also capable of simulating the effect of agricultural practices such as potassium fertilisation and liming. The semi-mechanistic approach of the Absalom model is applicable to any area if appropriate soil parameters are available. The model is also suitable for predicting long-term transfer since it considers time-dependent variation in RCs availability due to fixation to soil minerals. The model has been applied in various contexts (i.e. in different regions and for different foodstuffs) (e.g. Gillett et al., 2001; Beresford et al., 2002; Wright et al., 2002; Cox et al., 2005; Keum et al., 2007; Uematsu et al., 2016).

In Almahayni et al. (2019) we evaluated the Absalom model in terms of mechanistic basis and practicality. We concluded that the model was more mechanistic than the transfer factor approach since it relates RCs uptake to soil physical and chemical characteristics, which in turn are estimated from basic soil parameters (e.g. pH, clay content and OM content). Recently, the model has been extended to include wheat and barley, and restructured following an assessment of parameter redundancy (Tarsitano et al., 2011). This led to the development of a slightly modified version of the original model.

The Absalom model still has some limitations. Because the model was parameterised for European soils and grasses, it predicts RCs transfer under these conditions better than when it is used to predict RCs transfer for different crops (e.g. rice, potatoes, radish, etc.) and/or non-European (e.g. Japanese) soils (e.g. Keum et al., 2004; Rahman and Voigt, 2004; Rahman et al., 2005). For instance, the Absalom model uses clay content to predict RIP (Radiocaesium Interception Potential), which is a quantitative measure of soil intrinsic capacity and selectivity for sorbing RCs, and consequently RCs transfer to plants. This simplification however is not valid when applied to soils with different mineralogies from those included in the initial model calibration (i.e. common European soils). For example, the model overestimated the RIP of Fukushima soils by up to a factor of 16 (by a factor of 3 on average, N=51); the RIP per unit clay in the Fukushima soils was on average threefold lower than that in common Europeans soils (Uematsu et al., 2015). An empirical model with soil OM content and CEC (Cation Exchange Capacity) of the Fukushima soils data predicted RIP in the Fukushima soils more realistically than the Absalom model. Moreover, the model predictions of the TF for grass and Japanese soils differed (by less than two orders of magnitude) depending on whether RIP and dissolved potassium concentration were estimated by the model, or were measured values used as inputs (Uematsu et al., 2016). Recently, RCs transfer to brown rice from 50 Japanese soils was successfully predicted when using a version of the model that used measured soil RIP as an input (Sadao, 2019).

2.1.3 The mechanistic approach

Mechanistic RCs transfer models are largely based on nutrient transport and uptake in soil-plant systems. They often couple dispersion to rate-controlled process such as mineral dissolution-precipitation reactions and enzyme kinetics. These models are predominantly used for research purposes to test hypotheses and to identify important processes and sensitive parameters.

The structure of mechanistic transfer models is complex in comparison with empirical and semi-mechanistic models. Mechanistic models require many parameters that are not readily available or easy to measure (e.g. rate constants and root structure). Consequently, these models are seldom used to predict RCs transfer to plants in the context of nuclear emergency. Additionally, many mechanistic RCs transfer models do not account for the influence of exchangeable potassium on RCs transfer. Potassium has been shown to compete with RCs for plant uptake, especially in soils with low exchangeable potassium (Zhu and Smolders, 2000). This has led to using potassium fertilisers as a countermeasure to reduced RCs uptake from contaminated lands (Salt and Rafferty, 2001). Failure to account for potassium-RCs competition for plant uptake may produce unrealistic (e.g. high) uptake predictions.

2.1.4 Summary and recommendations

The advantages and disadvantages of the main RCs transfer modelling approaches are summarised in

Table 1 Advantages and disadvantages of existing RCs transfer modelling approaches as predictive tools in nuclear emergencies.

. In conclusion, the semi-mechanistic Absalom model is practical, robust and fit for predicting RCs transfer to plants in a nuclear emergency context. However, the model should be parameterised for soils and plants that were not included in its initial parameterisation. To optimise resources, parameterisation could focus on regions around major nuclear power plants to build a model database that is sufficiently representative of soils and plants not only in the temperate environment, but also in tropical, arctic and arid environments.

Table 1 Advantages and disadvantages of existing RCs transfer modelling approaches as predictive tools in nuclear emergencies.

Modelling approach	Advantages	Disadvantages
Empirical **(e.g. transfer factor)**	• Simple • Practical	• Does not account for key soil processes and parameters • Large variability • Lack of data for tropical and arid regions
Semi-mechanistic **(e.g. Absalom model)**	• Practical • Accounts for key soil parameters and processes • Accounts for time-dependent variation in RCs • Practical • Robust • Required soil parameters are often readily available.	• Predictions of RIP and dissolved potassium concentration may not be poor for soils that are outside the calibration range • Works best for European soils and grass (data for other soils and plant types are limited)
Mechanistic **(e.g. nutrient transport and uptake)**	• Instrumental research tool • Test hypotheses and sensitivity of processes and parameters	• Model structure is complex • Requires many non-readily available parameters • Some do not account for key processes and parameters

2.2 Transfer experiments and models

The review of existing RCs soil-to-plant transfer models (Almahayni et al., 2019) identified the semi-mechanistic Absalom model as a practical and robust predictive tool that can be used to plan and prepare countermeasures following nuclear emergencies. To further test the Absalom model and to identify soil parameters that influence RCs transfer to plants, SCK•CEN conducted controlled RCs transfer experiments using European soils (including some from more arid areas) and plants that had not been included in the Absalom model parameterisation.

In the following sections, the experimental setup and the results of these transfer experiments are presented and discussed.

2.2.1 Soil characterisation

Work package 3 partners (SCK•CEN, CEH, DSA and University of Extremadura) provided the soils that were used in the transfer experiments (Table 2). The soils represented temperate (Belgium and UK), Mediterranean (Spain) and boreal (Norway) European regions and were collected from arable land (N=5) and pastures (agricultural and semi-natural) (N=15); arable soils were underrepresented in the study soils.

Most soils had already been characterised in terms of pH, OM content (estimated by loss on ignition) and particle size distribution (sand, silt and clay content); where this information was not already available, analyses were conducted to obtain it. Soil CEC and RCs interception potential (RIP) were determined on subsamples of the soils using standard laboratory procedures (CEC was determined using silver thiourea complex cation, and soil RIP was determined according to Wauters et al (1996)).

Table 2 Soils used in the RCs soil-to-plant transfer experiments.

Country	N	Land use	Soil label
Belgium	6	Pasture	C, F, I, J, M, N
UK	6	Pasture, arable	Brimstone, Bromyard, Chiltern, Corney, North Wales, Spark Bridge
Spain	6	Pasture, arable	Bazagona, Casar de Caceres, Monfrague, Retortillo, Torreorgaz, Valero
Norway	2	Pasture	R13_14, R_15*

*The R_15 soil was included in the transfer experiments but not in subsequent analyses since no information on its clay content was provided.

2.2.2 Experimental setup

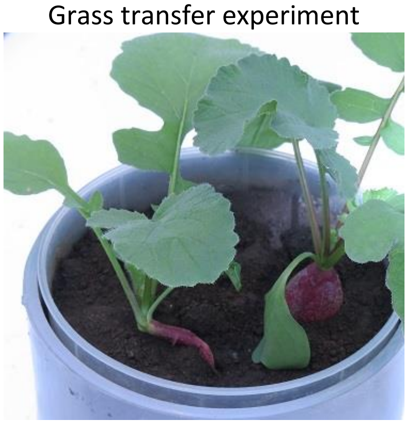
For the soil-to-plant transfer experiments, moist soils (at field capacity; see Table 2) were sieved to 2-mm and contaminated with approximately 400 Bq g^{-1} dw ^{137}Cs. The soils were then fertilised with the recommended amount of nitrogen, phosphorous and potassium, (NPK) fertiliser, homogenised and left to equilibrate for 3 weeks. The soils were subsequently sown with ryegrass seeds (density of 0.65 g pot^{-1}). Water lost by evaporation and root uptake was replenished regularly using demineralised water.

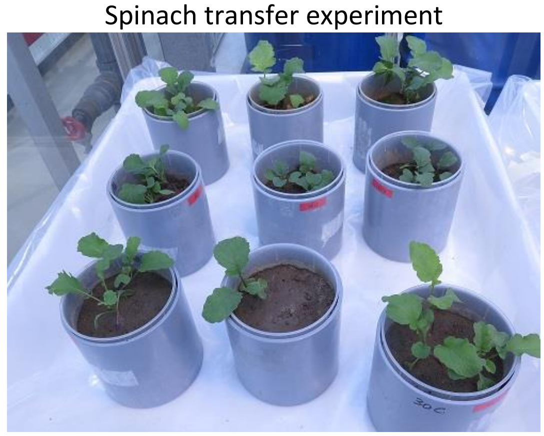
Grass was harvested after 23 days. The plants were cut at 2 cm above soil surface and washed to avoid soil contamination. Plant biomass was oven-dried (70°C), ashed and dissolved in hydrochloric acid ready for analysis. To prepare the soils for the next experiment, plant roots were removed from soils; the soils were then re-fertilised and equilibrated for 5 days.

The same procedure as described for grass was repeated for spinach (leaves) and radish (edible root). The growth period for these two plants was 35 days.

Soil collection | RIP measurements: soil equilibration

Grass transfer experiment | Spinach transfer experiment





Radish transfer experiment

Figure 2 The setup of the RCs soil-to-plant transfer experiments.

2.2.3 Results and discussion

The study soils (N=20) covered a wide range of physical and chemical characteristics (Figure 3). Soil pH ranged between 5 (Corney) and 8 (Brimstone); OM content varied between 4% (Bromyard) and 60% (Corney); clay content was between 1% (Bazagona and R13_14) and 40% (Bromyard) and the range in RIP was between 8 $cmol_c$/kg (R13_14) and 1049 $cmol_c$/kg (M).

The characteristics of the study soils varied between regions within the same country and between land management practices. For instance, the coefficient of variation in OM content, clay content and RIP between the British soils amounted to 107%, 70%, 101% respectively. The arable soils had greater clay contents, CEC and RIP than the pasture soils (Figure 4 and 5). Particularly, the greater RIP of the arable soils suggests that RCs transfer to crops grown in these soils would be lower than that from crops grown in the pasture soils.

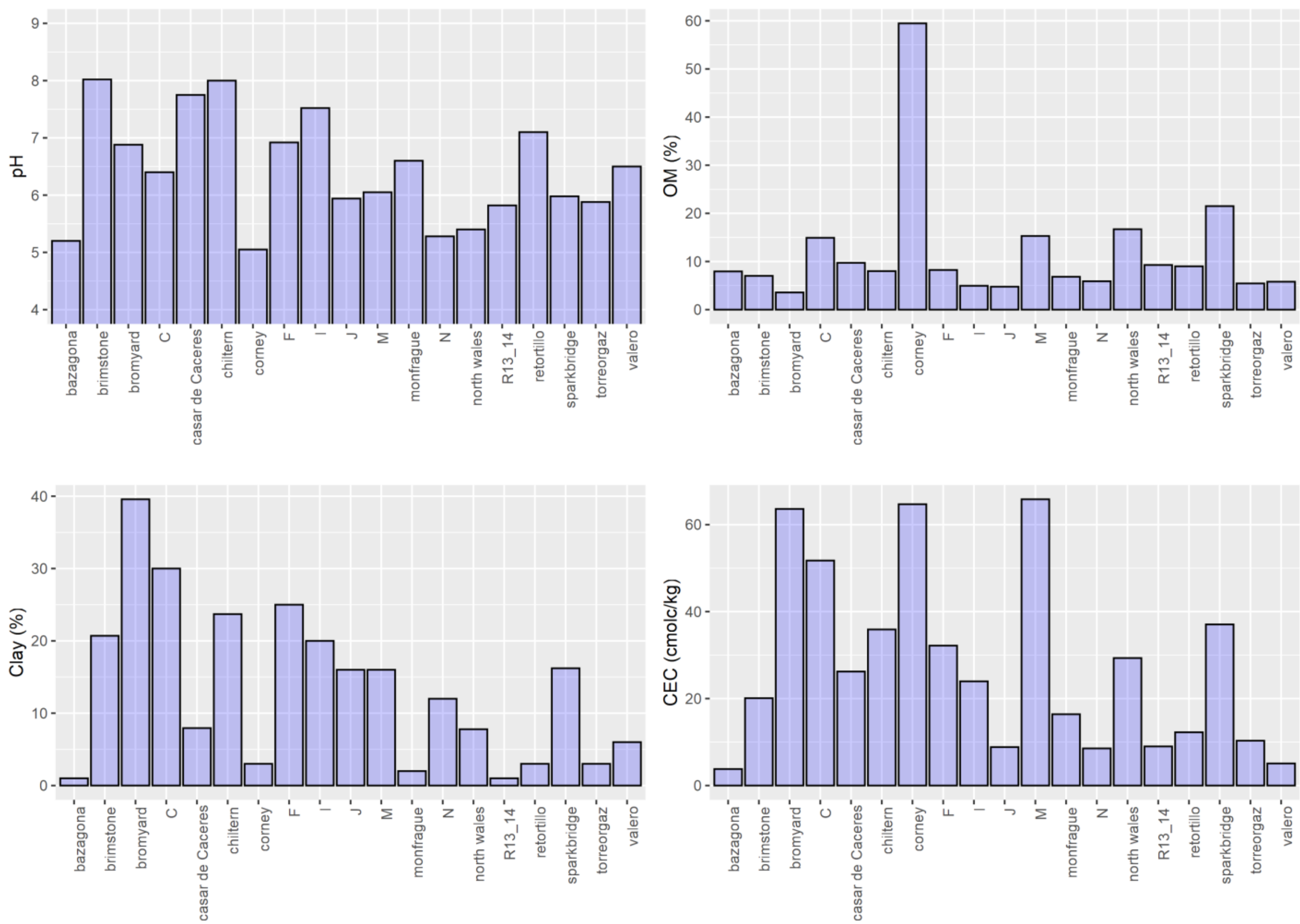


Figure 3 Physical and chemical characteristics of the study soils

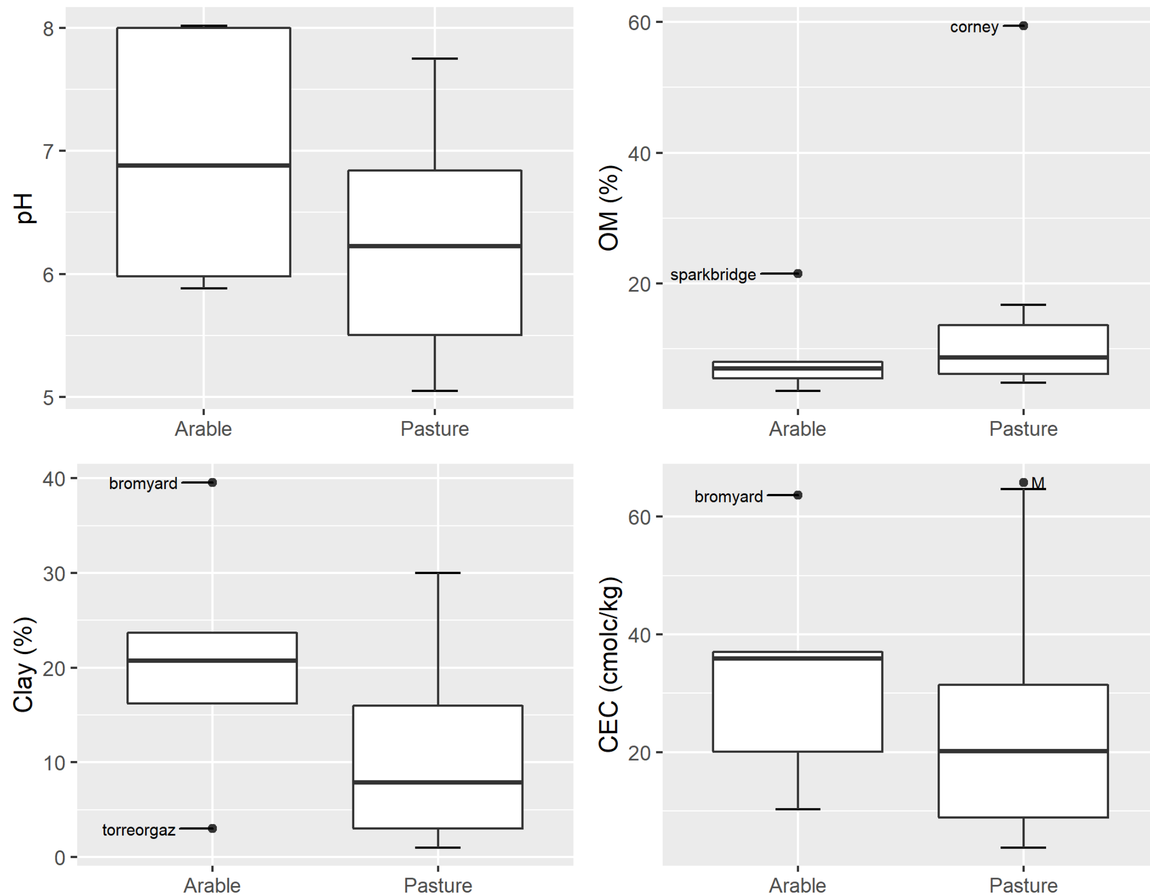


Figure 4 Range of the physical and chemical characteristics of the study soils grouped by land use. In the box and whisker plots: the thick horizontal line is the median, the lower and upper edges of the box are the 25th and 75th percentiles and the extending lines from both ends of the box are the minimum and maximum.

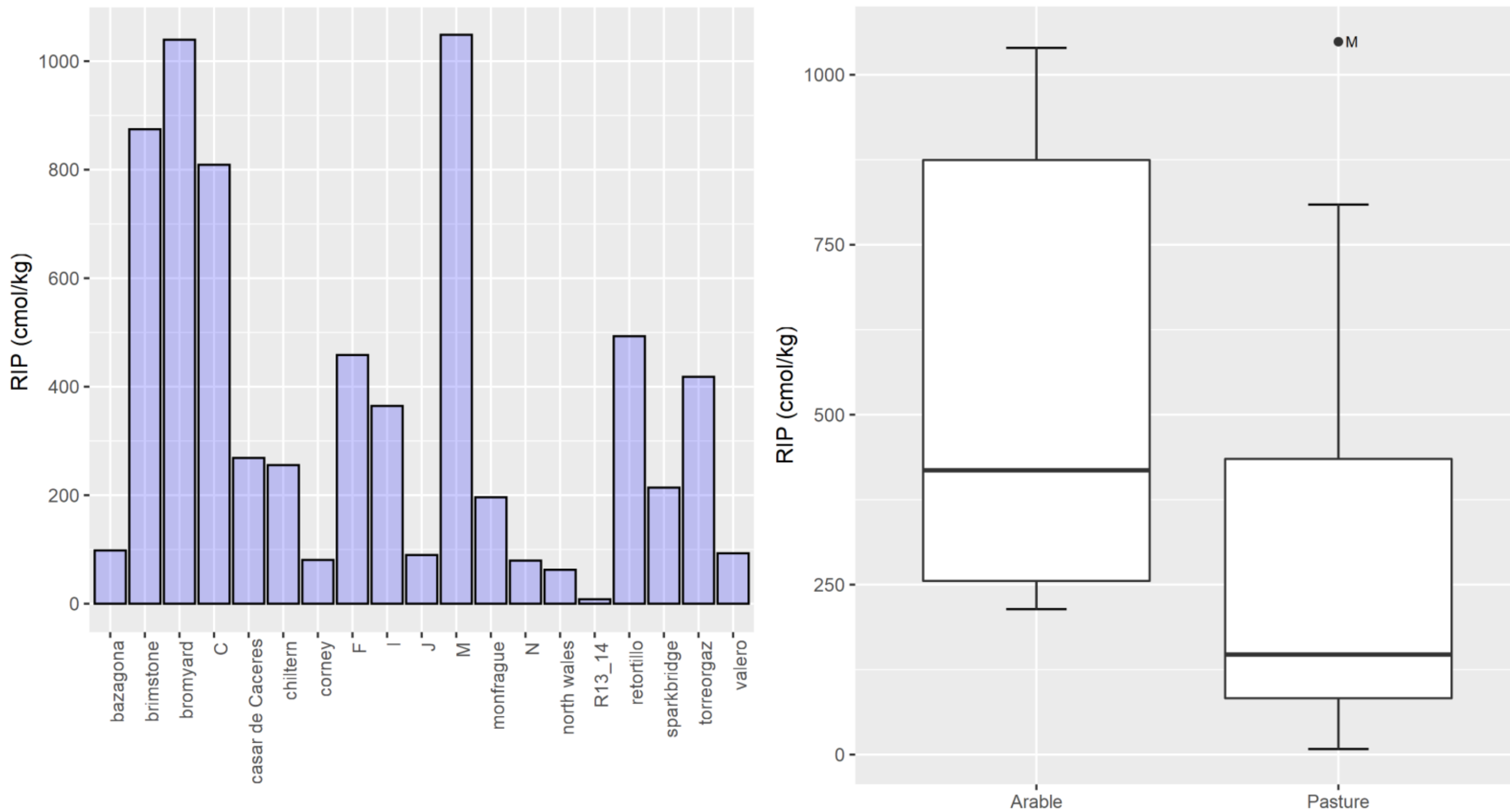


Figure 5 RIP of the study soils and its distribution according to land use; the thick horizontal line on the box whisker plot is the median, the lower and upper edges of the box are the 25th and 75th percentiles and the extending lines from both ends of the box are the minimum and maximum (excluding outliers: full circles).

2.2.3.1 Estimating RIP from basic soil parameters

It has been suggested that RIP strongly influences soil capacity to retain RCs and hence RCs availability to plants (Cremers et al., 1988; Delvaux et al., 2000; Waegeneers et al., 2005; Uematsu et al., 2016). Additionally, RIP is a key parameter in the semi-mechanistic Absalom model, which can either be estimated from OM and clay content or used directly as an input. However, soil RIP values are neither readily available or are they routinely measured.

We investigated whether the RIP in our dataset could be estimated from pH, clay content, OM content and CEC. Overall, the RIP correlated significantly to pH and clay content (Table 3). Stepwise regression with RIP as the dependent variable and these parameters as the independent variables retained the clay content as the only significant predictor (Table 4).

However, RIP and CEC correlated well in the Belgian soils as shown in Figure 6. Further analysis of the relationship between the two parameters in these soils revealed a strong and significant linear association (Table 5). We found no such correlation in the British soils despite having RIP and CEC ranges comparable to those of the Belgian soils, suggesting that other factors (not measured in our study) may have influenced the RIP-CEC dependence.

Table 3 Kendall's rank correlation coefficients and related P values between RIP and soil parameters in the study soils (N=19).

Soil parameter	Correlation coefficient	P values
pH	0.39	0.02
OM (%)	0.05	0.8
Clay (%)	0.43	0.01
CEC ($cmol_c$/kg)	0.32	0.06

Table 4: Results of the linear regression of RIP against pH, clay (%), OM (%) and CEC ($cmol_c$/kg) of the study soils (N=19). R^2 is the adjusted coefficient of determination of the model (RIP = $\beta_0 + \beta_1 \times clay$) and related P value in parenthesis.

Coefficient	Value	Std. error	P value	R^2
$\boldsymbol{\beta_0}$	943	961	0.3	
$\boldsymbol{\beta_1}$	203	56	<0.01	0.40 (<0.01)

Table 5: Results of the linear regression of RIP against CEC ($cmol_c$/kg) of the Belgian soils only (N=6). R^2 is the adjusted coefficient of determination of the model (RIP = $\beta_0 + \beta_1 \times CEC$) and related P value in parenthesis.

Coefficient	Value	Std. error	P value	R^2
$\boldsymbol{\beta_0}$	-598	116	<0.01	1 (<0.01)
$\boldsymbol{\beta_1}$	168	3	<0.01	

We also tested the empirical equations of Absalom et al. (2001) that estimate soil RIP mainly from clay content, OM content and pH. We compared RIP values estimated for our soils (Table 2) based on the relationships with RIP measurements given in Absalom et al. (2001). Whilst the two sets of RIP values correlated significantly, the estimates were significantly lower than the measurements (Table 6), suggesting that the empirical equations of Absalom et al. (2001) overestimated RCs mobility (RCs sorption in soil is proportional to RIP), and possibly its uptake, in the test soils.

Table 6: Results of the linear regression of measured RIP (cmol/kg) against Absalom-predicted RIP (cmol/kg) in the study soils (N=19). R^2 is the adjusted coefficient of determination for the model (RIP (measured) = $\beta_0 + \beta_1 \times RIP\ (Absalom)$) and related P value in parenthesis.

Coefficient	Value	Std. error	P value	R^2
$\boldsymbol{\beta_0}$	114	92	0.2	0.40 (<0.01)
$\boldsymbol{\beta_1}$	37	10	<0.01	

In our study, the RIP of the M soil was 12 times that of the J soil despite having identical clay contents (16%) (Figure 6). We observed similar behaviour in the British soils; the RIP of the Brimstone soil was three times that of the Chiltern soil despite their comparable clay content. Similarly, the average RIP of the Belgian and British soils was twice the average RIP of the Spanish soils despite having 5 times the amount of clay content. These results support findings from previous studies regarding the likely influence of clay mineralogy on soil RIP (Vandebroek et al., 2012; Uematsu et al., 2015; Almahayni et al., 2019).

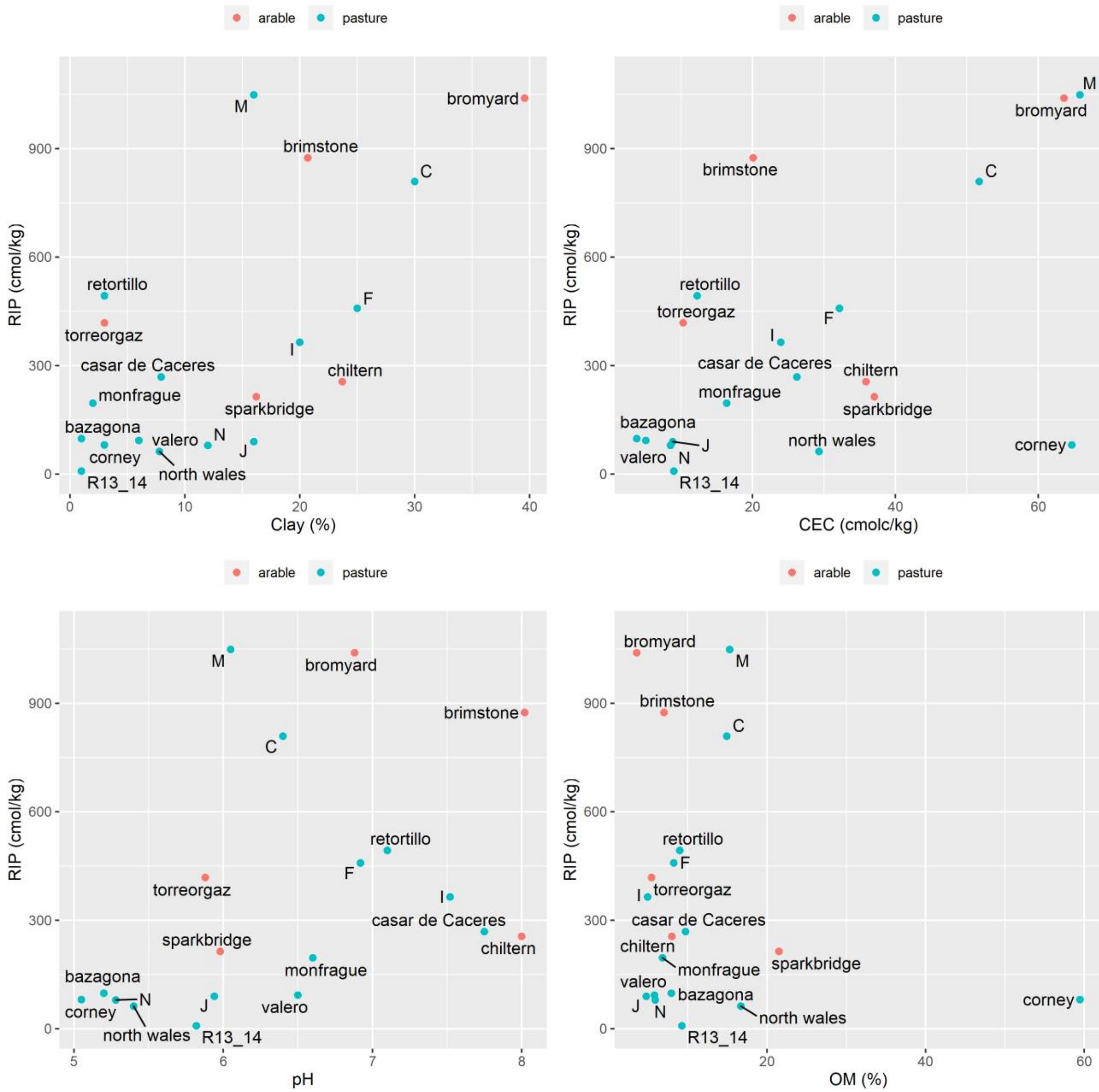


Figure 6 Soil RIP as a function of soil characteristics.

2.2.3.2 Soil-to-plant transfer factors

Radiocaesium transfer from the contaminated soils (see section 2.2.2) varied between soils and plant species (Figure 7). The TF varied between soils by up to three orders of magnitude. The TF was on average lower for the arable soils than for the pasture soils, indicating that arable soils retained RCs more effectively, thus reducing the potential for RCs plant uptake compared to their pasture counterparts (Figure 8). Indeed, these soils had on average a greater RIP (median of 418.2 cmol/kg) than the pasture soils (median of 196.2 cmol/kg). These differences may explain the lower RCs transfer from the arable soils.

We acknowledge that the difference in uptake from the arable and pasture soils may be due to our sampling strategy. The study soils were not purposefully selected to compare transfer between arable to pasture soils but to represent a range of environmental conditions.

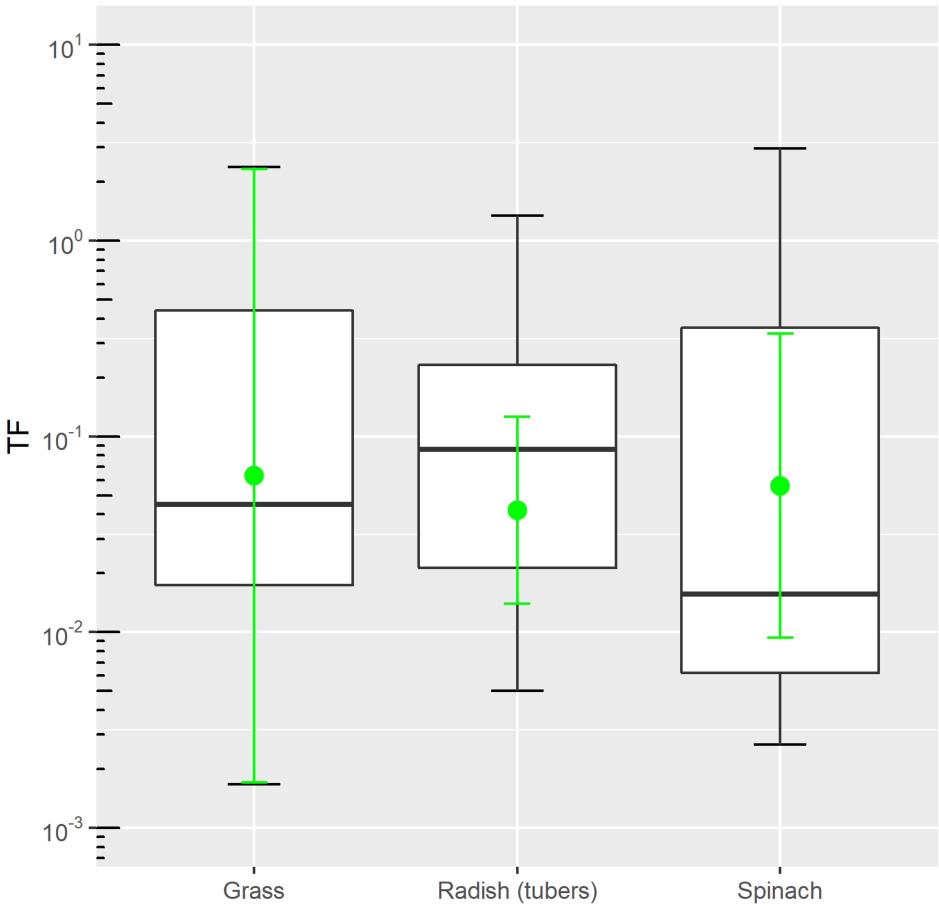


Figure 7 Variation in RCs soil-to-plant transfer factor over all soils. Green circles are the geometric mean (GM) of the TF reported in the IAEA (2010) for grass, root crops and leafy vegetables.

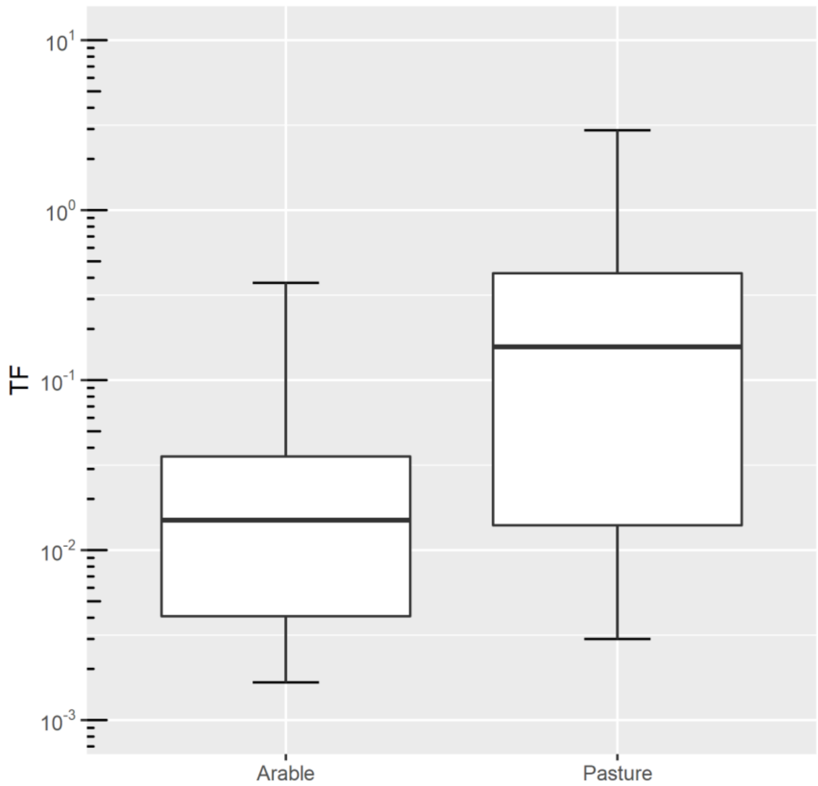


Figure 8 Variation in RCs soil-to-plant transfer factor, across all study crop types, between the main land uses in our study; the thick horizontal line is the median, the lower and upper edges of the box are the 25th and 75th percentiles and the extending lines from both ends of the box are the minimum and maximum.

The TF data we obtained for our study soils compare reasonably (same order of magnitude) to those reported in the IAEA (2010) compendium for similar plant types (Table 7). The GSD values in Table 7 highlight the large uncertainty in TF values.

Table 7: Geometric mean (GM) and geometric standard deviation (GSD) of TF measured in our experiments and those reported in IAEA (2010) for similar plant species.

	Confidence WP3			**IAEA**			
	N	**GM**	**GSD**		**N**	**GM**	**GSD**
Grass	20	7.8×10^{-2}	9	**Grass**	64	6.3×10^{-2}	37
Radish	20	7.8×10^{-2}	5	**Root crops (roots)**	81	4.2×10^{-2}	3
Spinach	20	4.2×10^{-2}	11	**Leafy veg.**	290	6.0×10^{-2}	6

2.2.3.3 Predicting Transfer Factors from basic soil parameters

Predicting TFs from basic soil parameters would have advantages for predicting RCs transfer to the food chain. Predicting TFs from soil parameters would allow us to bridge the data gap in existing TF compendia, which generally lack data for specific soil categories, plant types or both. The approach could utilise soil data in existing soil databases allowing spatially-distributed prediction of RCs transfer to pasture and food crops and for the construction of soil vulnerability maps (as has been previously demonstrated by Wright et al. (2003) who applied a model parametrised using soil OM content to upland areas of England).

We investigated the potential to predict RCs TF for grass, radish edible root and spinach from pH, OM content, clay content, CEC and RIP of the study soils. Despite the apparent association between the TF and some of these parameters (clay content, CEC and RIP in the radish dataset as shown in Figure 9 to Figure 13), the overall association between the TF and the aforementioned soil parameters was not statistically significant (Table 8), indicating that these parameters, on their own, could not predict the TF reliably.

Table 8: Results of the stepwise regression analyses of the TF against pH, OM content (%), clay content (%) and RIP (cmol/kg) of the study soils (N=19). R^2 is the adjusted coefficient of determination of the model ($TF = \beta_0 + \beta_1 \times pH + \beta_2 \times clay + \beta_3 \times OM + \beta_4 \times RIP$) and related P value in parenthesis.

Soil parameter	Coefficient	Std. error	P value	R^2
Grass				
$\boldsymbol{\beta_0}$	3.05	1.3	0.04	0.26 (0.07)
$\boldsymbol{\beta_1}$	-3.13E-1	2.0E-1	0.04	
$\boldsymbol{\beta_3}$	-2.05E-2	1.4E-2	0.2	
$\boldsymbol{\beta_4}$	-9.10E-4	5.1E-4	0.1	
Radish (edible roots)				
$\boldsymbol{\beta_0}$	4E-1	1.2E-1	<0.01	0.12 (0.09)
$\boldsymbol{\beta_4}$	-4.7E-4	2.6E-4	0.09	
Spinach				
$\boldsymbol{\beta_0}$	3.3	1.3	0.02	0.18 (0.08)
$\boldsymbol{\beta_1}$	-0.42	0.2	0.04	
$\boldsymbol{\beta_3}$	-0.02	0.01	0.1	

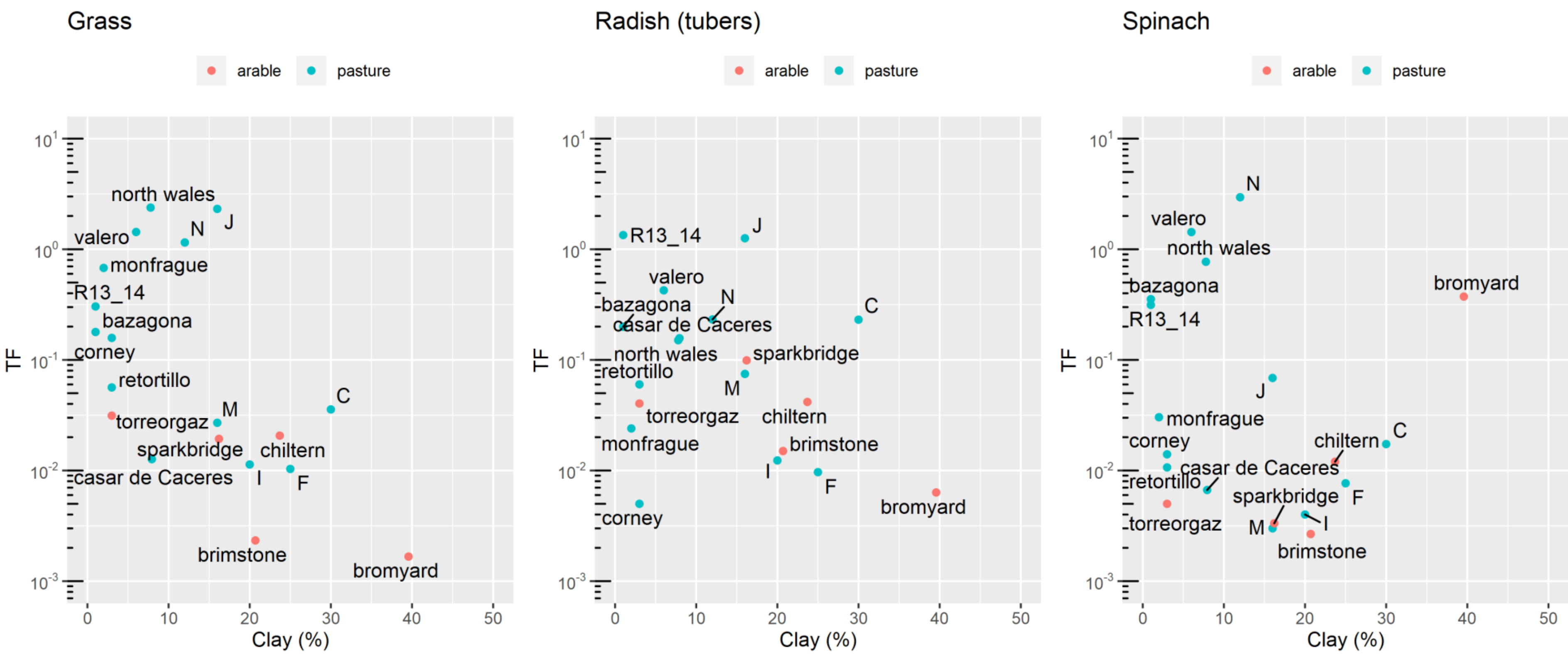


Figure 9 Measured TF for grass, radish edible root and spinach as a function of clay content of the study soils.

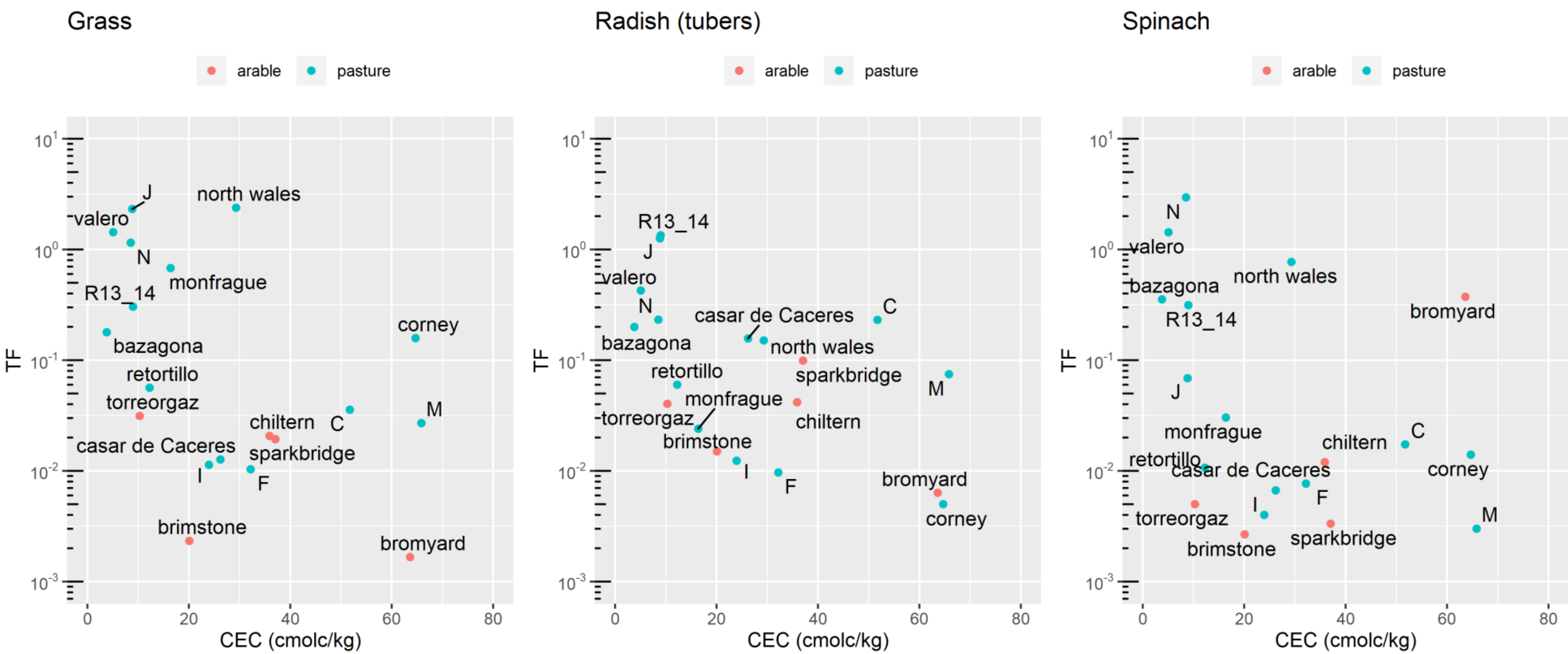


Figure 10 Measured TF for grass, radish edible root and spinach as a function of CEC of the study soils.

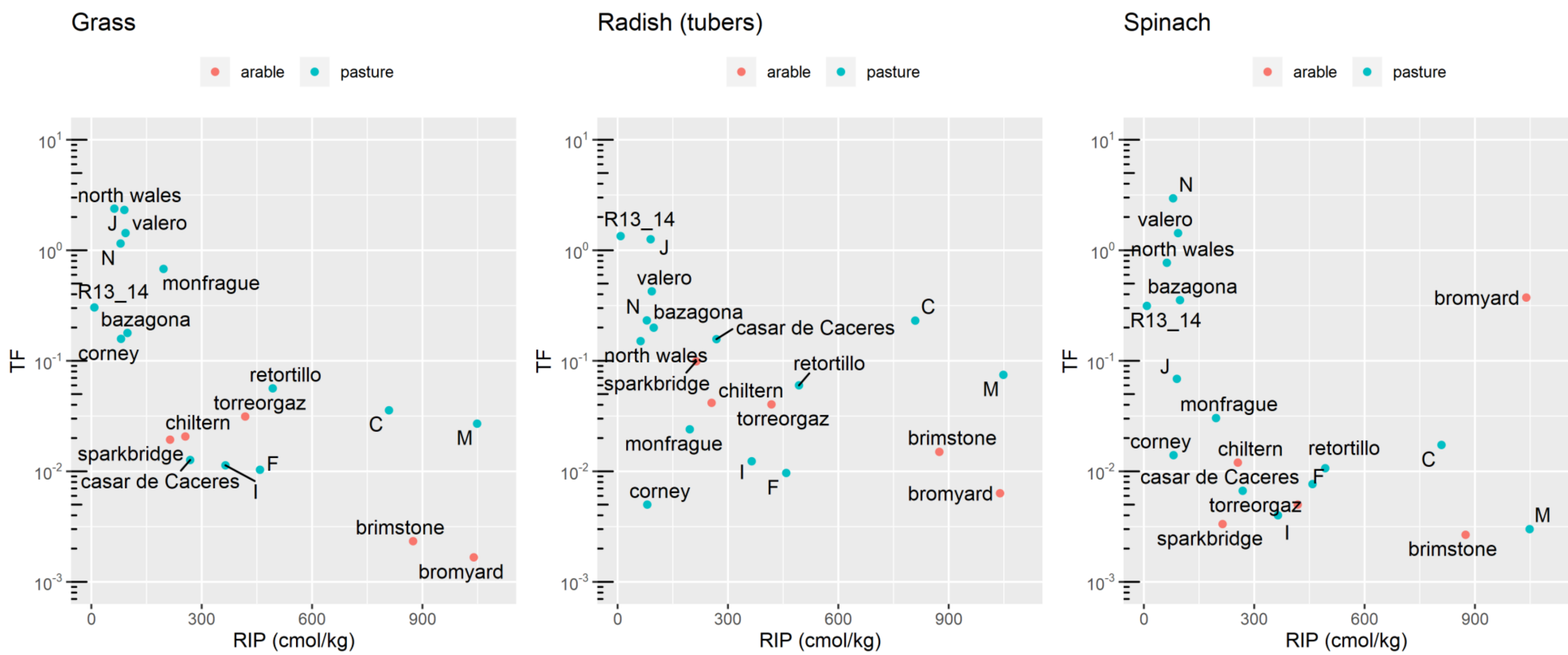


Figure 11 Measured TF for grass, radish edible root and spinach as a function of soil RIP.

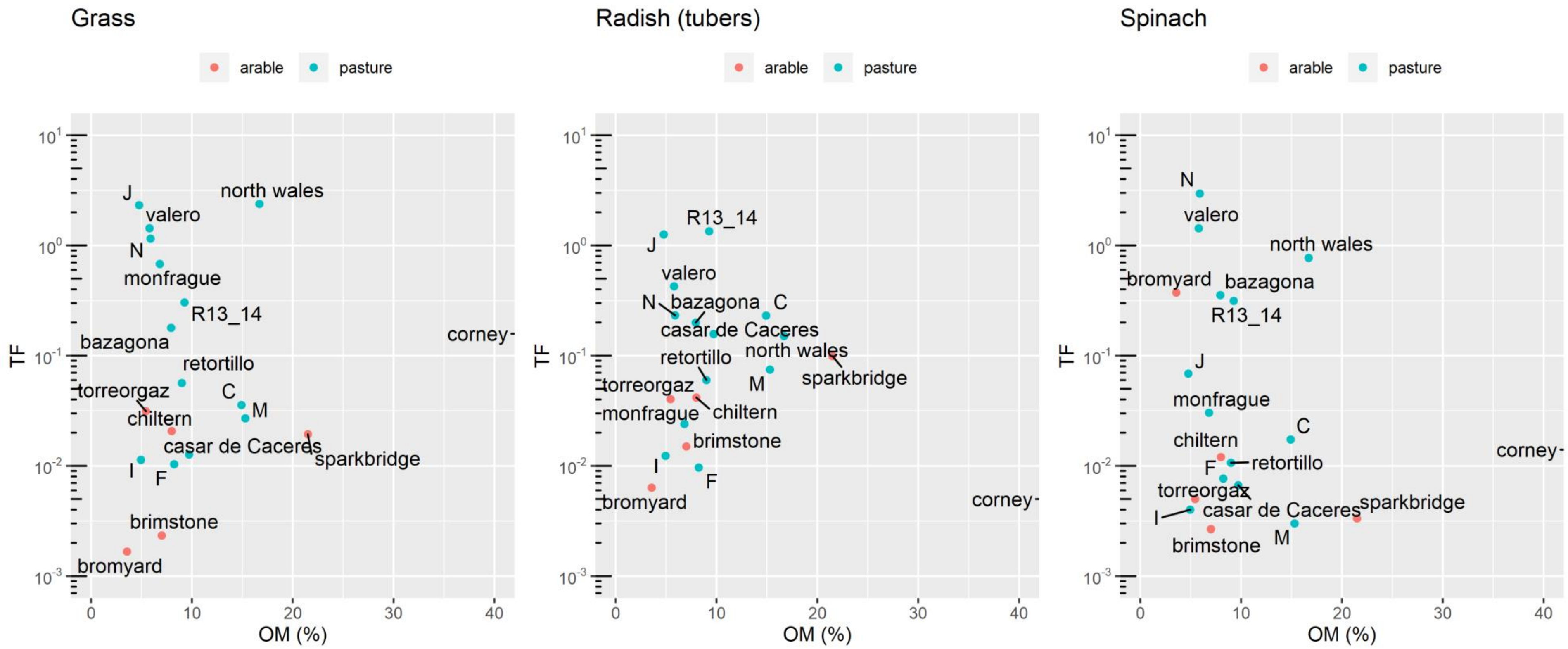


Figure 12 Measured TF for grass, radish edible root and spinach as a function of soil OM content (estimated by loss on ignition).

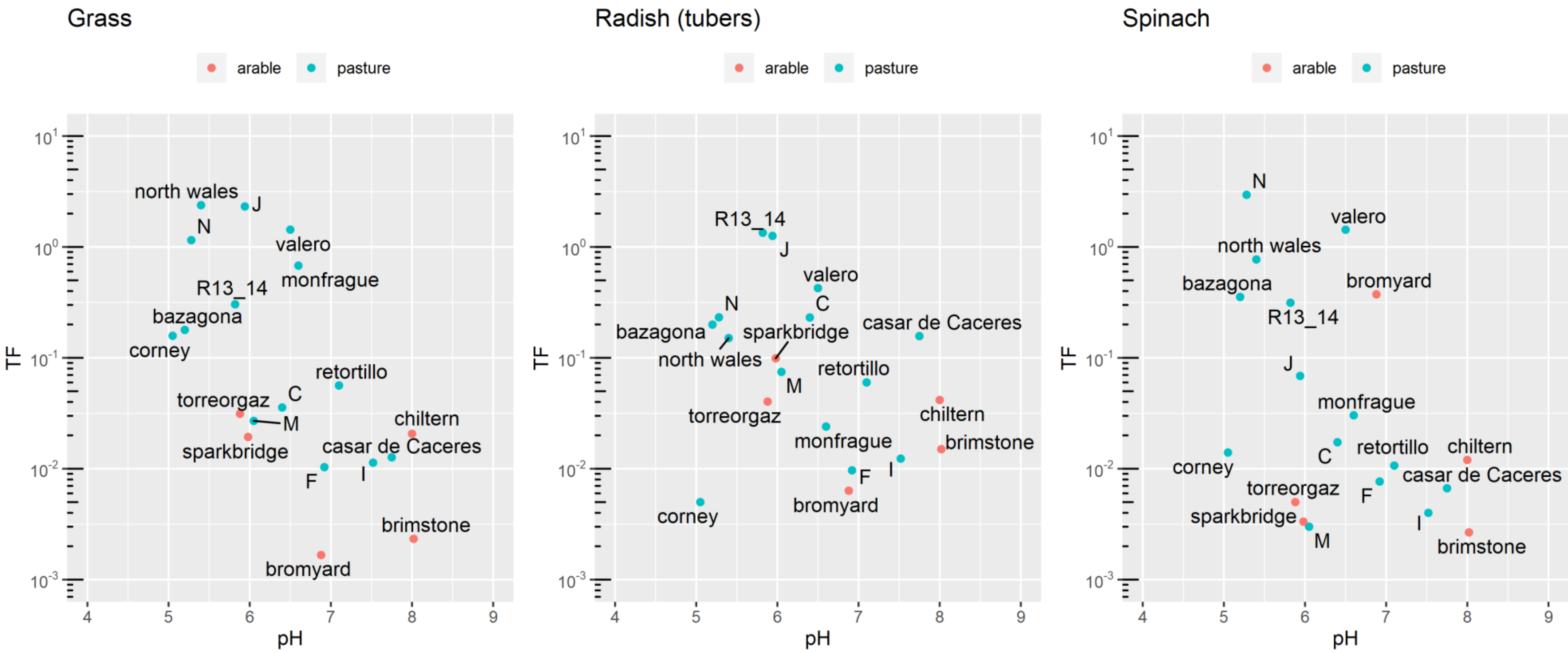


Figure 13 Measured TF for grass, radish edible root and spinach as a function of soil pH.

2.2.3.4 Predicting Transfer Factors using the Absalom model

As we demonstrated in Section 2.2.3.3, the TF did not correlate significantly to any of the basic soil parameters we measured during our experiments.

We have applied the Absalom et al. (2001) model to predict RCs TF from our soils to grass, radish edible root and spinach using as inputs RCs concentration in soil, pH, OM content, clay content and exchangeable potassium. The model was applied with default parameter values for grass (see Almahayni et al., 2019 for full description of the model and its parameters). Consequently, model predictions presented in this report represent RCs transfer to grass from our study soils, and variation in those predictions reflect variations in the model inputs rather than variations in uptake between plants. Predictions for grass are compared to measured values form our studies for grass, radish and spinach.

The Absalom model predicted RCs TF for grass reasonably well for most soils with predictions being within an order of magnitude of the measurements (the 1:1 line in Figure 14). Predictions for a few soils (N and J from Belgium, North Wales from UK and Valero from Spain) were within two orders of magnitude of the measured values. The variation of two orders of magnitude in model predictions across all soils is similar to the overall range for grass TF values as reported in IAEA (2010) for a wider range of soils.

The Absalom model reproduced RCs concentration in grass biomass for most soils (Figure 15); predictions in grass biomass correlated significantly with our measurements (Table 9). However, estimates of RCs concentration in grass biomass based on an average TF (GM in IAEA (2010)) and soil contamination, did not correlate with our measurements, and we were unable to reproduce the variation in the RCs concentrations across our study soils (Figure 15).

The results of the Absalom model validation are encouraging and increase confidence in the model. The model predictions of the TF correlated significantly with our TF measurements for grass and radish edible root (Table 9), indicating relevance of the Absalom predictions to the actual data (predictions were mostly within an order of magnitude of our measurements as shown in Figure 14). However, the Absalom model TF predictions correlated poorly with our TF measurements for spinach. These results suggest that the model—even with the default parameters—is useful for estimating TF for some species other than grass (e.g. edible root). Making predictions for leafy vegetables appears to require further model calibration based on a sufficiently large datasets.

Table 9 Kendall's rank correlation coefficients and related P values (in parenthesis) between Absalom predictions and the measurements of grass, radish (edible root) and spinach TF.

	Grass TF (measured)	Radish TF (measured)	Spinach TF (measured)
Grass TF (Absalom)	0.49 (< 0.01)	0.25 (>0.1	0.18 (>0.3)

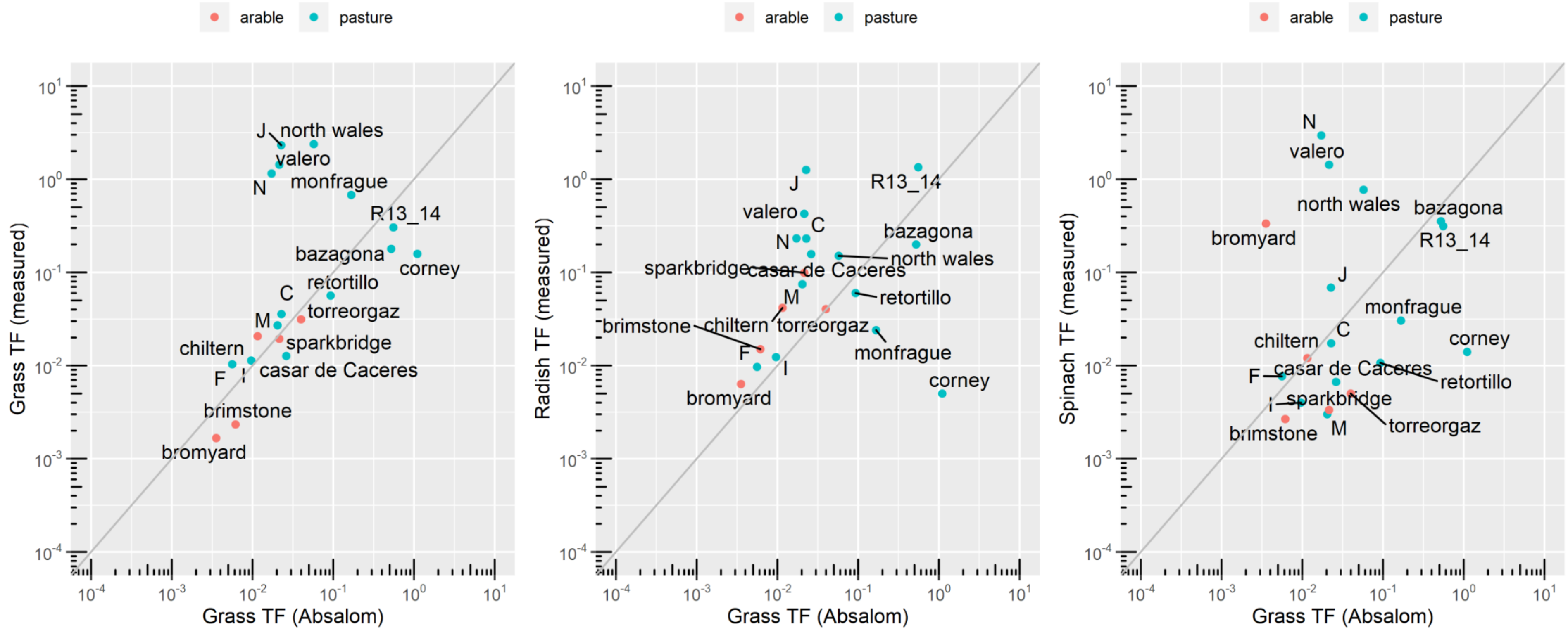


Figure 14 Measured versus Absalom-predicted TF. The diagonal line represents the 1:1 line, which defines the perfect match between the predictions and the measurements.

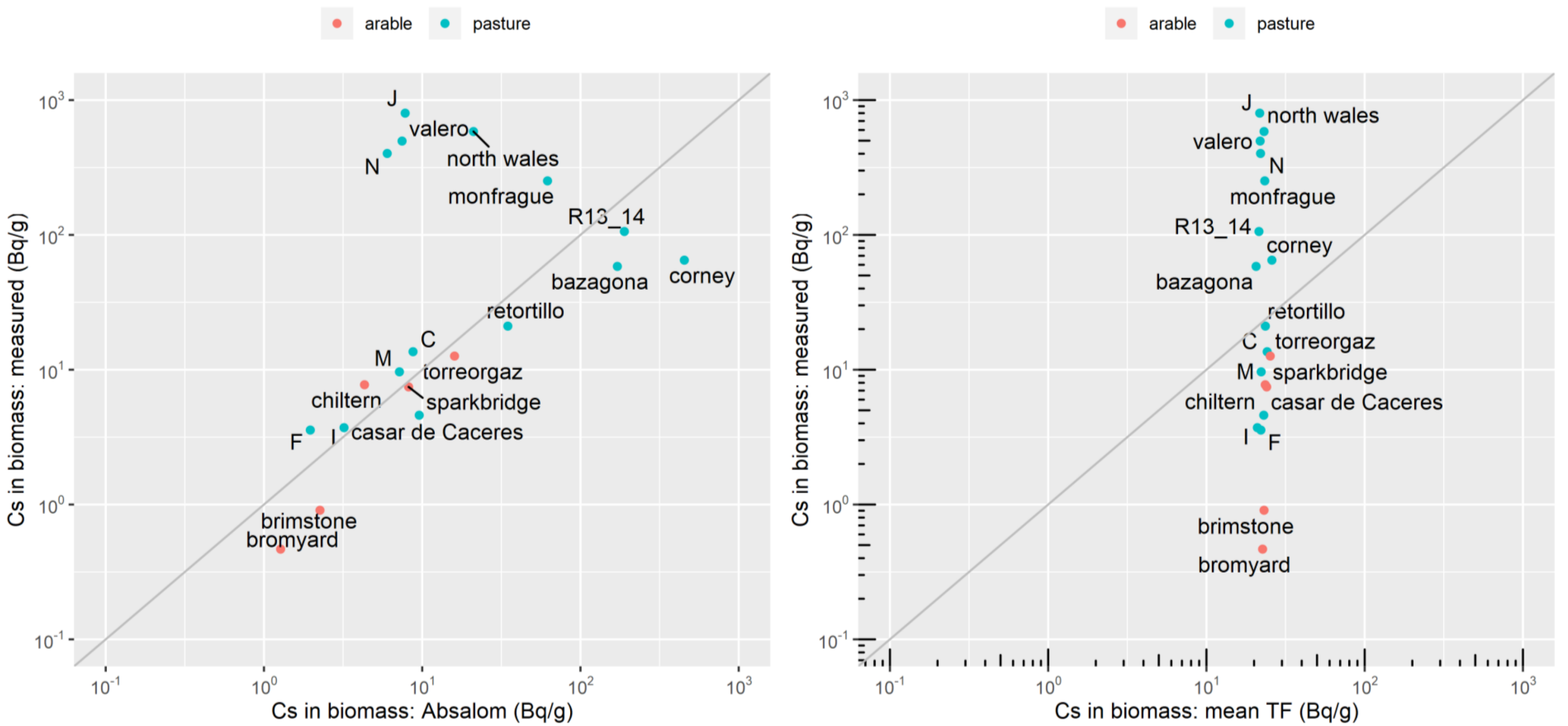


Figure 15 Measured versus predicted (with Absalom and TF) RCs concentration in dry grass biomass across all soils. The Absalom model reproduced the measured concentration and its variation across study soils, whereas the TF-based predictions (using IAEA (2010) GM TF for grass) were poor.

2.2.4 Summary and recommendations

We determined RCs transfer to grass, radish edible root and spinach from 20 European arable and pasture soils with varying pH, OM content, clay content, CEC and RIP. The median TF, a measure of RCs transfer to plants, increased in the following order: spinach < grass < radish edible root, and the values determined in our study were generally comparable to those reported in the IAEA (2010) compendium. Our TF values varied by up to three orders of magnitude across each soil—plant combination. This variability is in line with the variability in published TF data for similar plant categories.

We attempted to derive an empirical equation that predicts soil RIP from readily available soil parameters to alleviate the need to measure RIP or rely on model predictions, which requires special procedures and involves using radioactive isotopes. The RIP correlated significantly to clay content of the study soils, but the correlation was not sufficiently strong to derive a useful predictive equation. The RIP correlated significantly to the CEC of the Belgian soils, whereas no such correlation existed in the British soils, for which RIP and CEC values were comparable to their Belgian counterparts. This calls for further investigation of the correlation between RIP and CEC.

We also tested the ability of the Absalom model to estimate the RIP of the study soils from their clay content, OM content and pH. The model significantly and systematically underestimated the RIP of the study soils suggesting a need to revise how it predicts soil RIP (e.g. consider not only the clay content but also clay mineralogy).

We also attempted to derive an empirical equation that predicts TF from readily available soil parameters (available from existing soil maps and/or databases), which would enable spatial predictions of RCs transfer to food chains. However, the TFs in our dataset did not correlate significantly to any particular soil parameter or set of parameters. Additionally, the size of our dataset (19 soils) is relatively small; a larger soil collection might potentially reveal statistically meaningful correlations between TF and soil parameters.

The Absalom (2001) model is a useful tool for predicting RCs transfer to the food chain. The model predictions of the TF for grass and radish edible root were mostly within an order of magnitude of the measurements for most of the study soils. This is encouraging considering that we applied the model with the default parameter values (the experimental data were too few to allow proper calibration of the model). We recommend expanding the model database by considering more soils (with different mineralogies) and plant types in its parameterisation.

3 Implementation of the Absalom model into FDMT

FDMT (Food Chain and Dose Module for Terrestrial Pathways) is a model developed to simulate transfer of radionuclides along the human food chain and to calculate activity concentrations in food and feedstuff. It is the food chain transfer module of the European JRodos and ARGOS decision support systems (Brown et al., 2018)

FDMT is largely based upon the earlier dynamic model ECOSYS-87 (Müller and Prohl, 1993) that was originally implemented within Microsoft EXCEL™. Much of the developmental work including the numerical specification of many of the parameters used in ECOSYS-87 (and therefore FDMT) was completed in the 1980s and hence many of the later, large numbers of radioecology studies prompted by the 1986 Chernobyl accident were not considered. This latter shortcoming has now been addressed, to a degree, within CONFIDENCE (Brown et al., 2018) and elsewhere (Staudt, 2016; Thørring et al., 2016) through an extensive updating process.

As described in Brown et al. (2018), there are numerous limitations associated with the FDMT model as incorporated within the decision support systems noted above. Lack of flexibility with regards to modifying model components and the assessor being restricted to only run simulations deterministically, have been identified as the main limitations. Overcoming these restrictions has provided the rationale for extracting the model and implementing it within a probabilistic-enabled modelling platform called ECOLEGO. In so doing, more simulation options were introduced, further enabling an exploration of the factors that introduce variability within model predictions, e.g. region specific parameters such as growing seasons and dietary habits. ECOLEGO is a platform for creating dynamic models and performing deterministic or probabilistic simulations (Avila et al., 2005); http://ecolego.facilia.se/ecolego/show/HomePage).

Transferring FDMT to the ECOLEGO platform provided us with flexibility to modify and develop the existing models, by either including new processes or adding sub-models. For the present work, the default soil model of FDMT has been replaced by the 'Absalom' soil-to-plant transfer model, which predicts ^{137}Cs in soil solution and selected vegetation with time (as described above), originally developed for grass. The mathematical specification and description of Absalom et al. (2001) model as presented in Appendix 1 of Tarsitano et al. (2011) is the version that has been implemented within ECOLEGO platform (after fixing some errors in Tarsitano et al. in consultation with the originating authors).

The Absalom model has the advantage that soil gravimetric clay content (g/g), gravimetric organic content (g/g), pH and exchangeable potassium ($cmol_c$/kg) can be considered specifically as inputs. To date within CONFIDENCE, effort has been placed on defining these parameters for Ukrainian podzoluvisols for which empirical data exist that could be used to evaluate model performance. Lind et al. (2019) introduced and discussed these data in relation to the development of models that account for 'hot particles' in soils.

Soil pH was one of the few parameters reported explicitly in the datasets for the Ukrainian soils. In the process of model parameterisation in Lind et al. (2019) an indicative value of soil acidity, with pH 6.61, was selected from the available datasets for relevant Ukrainian soils (for use in the implementation of the Absalom model). Ivanov and Khomutinin (2015) provides information on exchangeable potassium, expressed as K_2O (mg/kg), from which representative values for exchangeable potassium $[K]_{exch.}$ can be derived for podzoluvisols in nearby contaminated areas of Ukraine. Finally, since data for site specific clay and organic matter data were not available, indicative values for podzoluvisols (sampled in Poland) were used from Vandebroek et al. (2012). The parameter defined in Tarsitano et al. (2011) as 'a_1' was taken as that relating specifically to wheat (this particular crop was selected because we had a reasonable coverage with empirical data against which our prediction could be compared). These indicative input parameters are provided in Table 10, all other parameters have been left as defaults.

Table 10 Parameters used in the Absalom model set-up.

Clay content (%)	Organic matter content (%)	$[K]_{exch}$ $cmol_c/kg$	pH	a_{1wheat} $log_{10}(L/kg)$
9.1	1.22	0.1	6.61	3.45

3.1 Solid-liquid distribution coefficient (K_ds)

Solid-liquid distribution coefficients (K_ds) for ^{137}Cs based on empirical collations (IAEA, 2010) can be compared with values that have been calculated from the Absalom et al. (2001) model.

Table 11 ^{137}Cs K_ds based on collated empirical data (IAEA 2010) and from applied models. GM: geometric mean, GSD: geometric standard deviation.

Soil Type	K_d L/kg			
	GM	GSD	Min	Max
All soils*	1.20E+03	7	4.3	3.80E+05
Sand*	5.30E+02	5.8	9.6	3.50E+04
Loam + clay*	3.70E+02	3.6	39.0	3.80E+05
Organic*	2.70E+02	6.8	4.3	9.50E+04
			Best Estimate	
Podzoluvisol - Absalom			1.31E+04	

*IAEA TRS-472 (IAEA, 2010)

The Absalom model estimates of ^{137}Cs K_d fall at the higher end of the empirical range for all soil types (in Table 11) suggesting that most of the ^{137}Cs activity in soil would be predicted to be associated with the solid phase. This result is consistent with field observations that show a significant fraction of RCs being associated with strongly bound phases in soils sampled from the Ukrainian part of the Chernobyl exclusion zone, several years after the accident (Oughton et al., 1992). Consequently, the Absalom model's prediction of K_d seems reasonable.

3.2 Modelled radionuclide activity concentrations in wheat

A comparison of the results of FDMT (as implemented into ECOLEGO, see Brown et al. (2018)) and two variants of Absalom model outputs with respect to ^{137}Cs (Bq/kg) activity concentrations in wheat following a deposition event is shown in Figure 16. Soil conditions do not affect the FDMT model output for activity concentrations in wheat, because a generic soil-to-plant concentration ratio is applied by default, for all soil types.

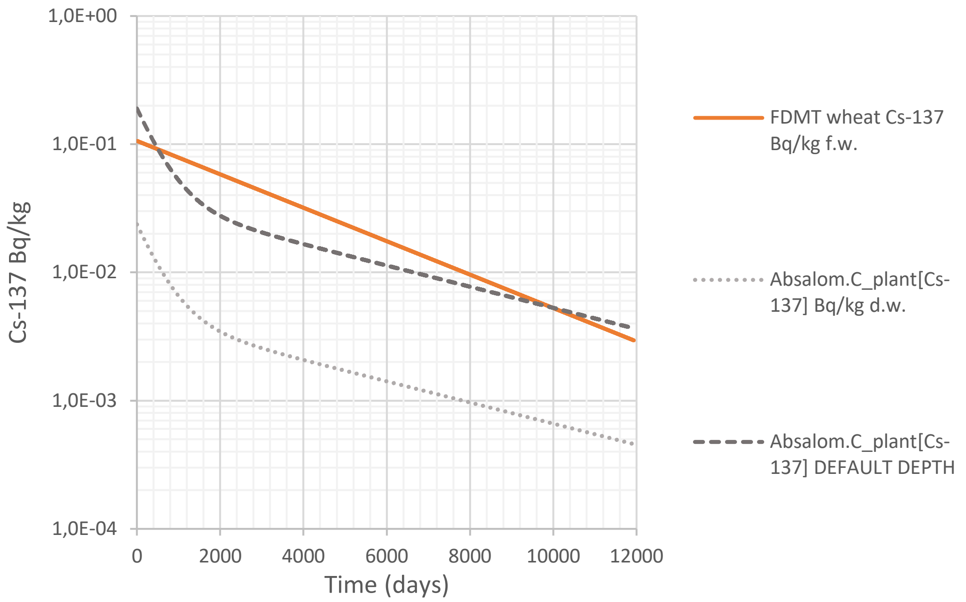


Figure 16 ^{137}Cs activity concentrations (Bq/kg) in wheat simulated using 2 models: Absalom (grey dotted line) and FDMT (solid orange line); Deposition = 1 kBq/m^2 ^{137}Cs. The dark grey, short-dashed line shows the Absalom model output using the models default depth (over which ^{137}Cs is assumed to be distributed) of 2.5 cm.

The application of two variants of the Absalom model requires some elaboration. Initially, for the derivation of activity concentration in soil, a depth of 0.2 m (in line with the default specified in a new soil model developed for particles, as described in Lind et al. (2019)) was used for the Absalom model. This approach required substantial adjustment of the deltaZ (m) parameter (defining the depth over which ^{137}Cs is assumed to be distributed) in the Absalom model from the default of 0.025 m. Both FDMT and the Absalom models predict a continual decline in the levels of ^{137}Cs in wheat following the deposition event. Whereas one variant of the Absalom model (using 20 cm rooting depth) yields values substantially below FDMT, using the variant of Absalom with default depth (2.5 cm) provides results with a high degree of similarity with FDMT. Reverting to the default deltaZ (m) value, therefore, yields results from the Absalom model that are much more in line with values generated using the FDMT model simply reflecting the observation that for the 20 cm model run we are essentially diluting the activity in soil by a level approaching a factor of 10. The Absalom model returns the most elevated ^{137}Cs activity concentration in wheat of the model predictions for the first 500 or so days using this default configuration.

Note further that the Absalom model returns activity concentrations on a dry mass basis, whereas the FDMT models make predictions for fresh mass. No attempt has been made to adjust for this. The dry matter content of wheat grain is relatively high with a value of 88% being provided in IAEA (2010) and hence accounting for a fresh to dry matter conversion for wheat grain would make little difference to the model estimates.

3.3 Comparison with validation data

Simulations have been run using ^{137}Cs depositions based on measured ^{137}Cs activity concentrations in soils, from locations in Ukraine, that were decay-corrected to the date of deposition (26th April 1986). The comparison of empirical data versus FDMT model predictions is given in Figure 17. The comparisons were made for the period 2011 to 2018 for which an empirical dataset on soil and selected crop ^{137}Cs activity concentrations was available, as described by Lind et al. (2019). ^{137}Cs activity concentrations in grain from the FDMT and Absalom models (using a distribution depth, deltaZ (m), of

0.2 m as opposed to a default of 0.025 m) result in values that are lower than empirical determinations by approximately a factor of 10 and 100 respectively. Using the Absalom model with the default deltaZ (m) parameter yielded results similar to those of FDMT, i.e. decrease to a value of approximately a factor of 10 below measured values. The substantial underprediction of the FDMT and Absalom models may be counterintuitive, in the sense that part of the contamination in the studied areas is associated with particles that might be expected to restrict the availability of ^{137}Cs for plant uptake. However, a detailed analysis of the system using a bespoke 'hot particle' model has provided us with insights into the dynamics of transfer (Lind et al., 2019). Although ^{137}Cs bioavailability may be diminished within the first years after deposition, at later stages, once particles have weathered many years after deposition, transfer to crops may actually become augmented above predictions based upon 'commonly-applied' models that do not account for particle behaviour. Further details are provided in Lind et al. (2019). The predictive efficacy of the FDMT model is low. In addition to the aforementioned substantial under-prediction by the model, there is only a weak correlation between predicted and observed ^{137}Cs activity concentrations in crops.

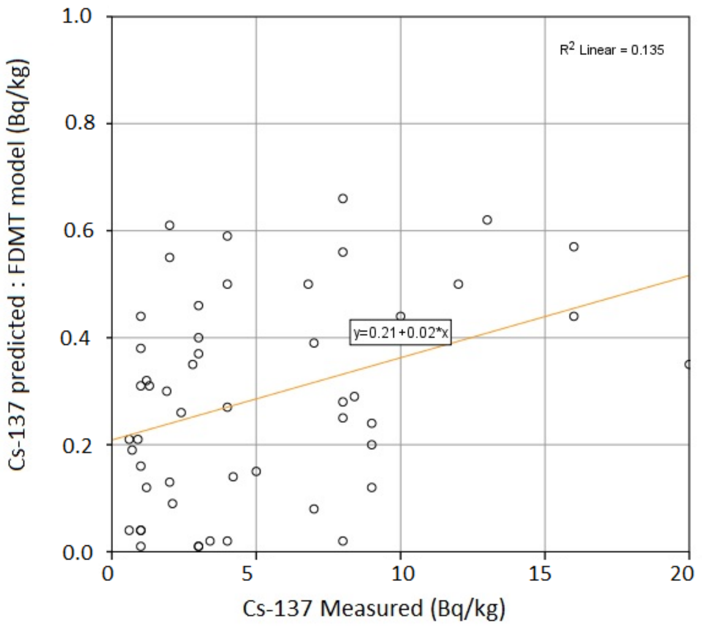


Figure 17 Activity concentrations of ^{137}Cs (Bq/kg) in crops (wheat, Rye, barley); x-axis = measured (grain data 2011-2018) and y-axis = predicted using FDMT for the corresponding year.

3.4 Summary and recommendations

Having implemented FDMT into the ECOLEGO platform, we have access to a flexible tool for incorporating and exploring new or modified model components and parameters. This flexibility has been demonstrated here by replacing FDMT's default soil model with a process-based model. In this way, the impact of various processes and factors can be modelled. The next step is to incorporate and study the influence of other process-based models (which have been developed elsewhere in the CONFIDENCE project) upon the FDMT's outputs. This, combined with the new possibility of being able to conduct sensitivity analysis, can be used to guide future research related to the transfer of radionuclides through the human food chain and also allow us to identify the focus of further model developments in this field.

4 Development of process-based soil plant models for radiostrontium

Although previous studies have derived relationships between soil properties and (i) K_d for Sr and/or (ii) the transfer of Sr to specific plant species (e.g. Camps et al., 2004; IAEA, 2006), to our knowledge there has been no development of more generic process-based approaches as has been attempted for Cs. In this chapter, we describe our development of two process-based models to predict the soil-plant transfer of (radio)strontium. The first approach involves derivation of a novel and relatively simple (in terms of required inputs) methodology, whereas the second approach adapted a well-established, chemical speciation model (Tipping, 1994; Tipping et al., 2011) to predict radiostrontium concentrations in plants. Before discussing model development, we will describe plant growth studies conducted to provide data with which we could test the predictions of the developed models.

4.1 Plant growth studies

Studies were conducted using six different soils selected with a range of characteristics anticipated to influence the transfer of radiostrontium to plants (Table 12). Three of these soils were obtained from England and one from Spain; the 'Spark Bridge' (England) and 'Casar de Cáceres' (Spain) soils were also used in the radiocaesium studies described in Chapter 2. Two further soils were obtained from a UK commercial supplier (these were sourced from southeast England); the commercially supplied soils were unsterilized.

A variety of different crops were selected: radish (*Raphanus sativus*); lettuce (*Lactuca sativa*); grass (*Agrostis capillaris*); chard (*Beta vulgaris subsp. vulgaris*); courgette (*Cucurbita pepo var. cylindrical*); strawberry (*Fragaria × ananassa*) and potato (*Solanum tuberosum*). The radish (variety 'Sparkler'), lettuce (cultivar 'Clarion'), grass, chard (cultivar 'White Silver 2') and courgette (cultivar 'Midnight') were grown in the study soils from seed. Potatoes (cultivar 'Charlotte') were obtained as 'seed potatoes' and 'bare root' strawberry (cultivar 'Romina') plants were obtained from a commercial grower. A variety of pot sizes (1.8 L to 12 L) were used depending upon plant size. All plants were grown in the soils from England, including the two obtained from a commercial supplier, at CEH Lancaster during 2018. After starting in a greenhouse, potatoes, courgettes and chard were moved outside; all other plants remained in the greenhouse for the course of the study. All plants were grown to maturity with the edible portion, including radish leaves, being collected (leaves and 'runners' from strawberry plants were also retained). All plants were grown in Spain (Extremadura) in the Casar de Cáceres soil in 2018 with the exception of strawberries; the selected variety could not be obtained in Spain and for reasons of plant health legislation, it was not possible to export the plants from the UK to Spain. In Spain, all plants were germinated and grown to maturity outside. At the end of the study pore waters were collected using rhizon samplers (https://en.eijkelkamp.com/products/ground-water-samplers/rhizon-soil-moisture-samplers.html).

Because strawberries could not be grown in Spain in 2018, some of the Spanish study soil was exported to the UK where studies were conducted in 2019. For comparative purposes, strawberries and grass were grown in 2019 in both the Casar de Cáceres and Spark Bridge soils. As the radish harvest had been generally poor in 2018, radish was grown again in all six study soils.

4.1.1 Sample analyses

Following weighing and washing in de-ionised water, plant samples were stored frozen prior to being freeze-dried and then finely ground (<2 mm). Potatoes were peeled after washing and prior to being frozen (the peel was retained for analyses but results are not discussed here). Soil samples were dried at 20°C (soils sent from Spain to the UK for analyses were first dried at 60°C) and then finely ground using an agate ball mill.

In preparation for ICMPS/ICPOES analysis, the finely ground soil and plant samples were digested using aqua regia (3:1 mixture of concentrated HNO_3 and HCl 'Aristar' grade acids) at 175°C for 12 minutes using a microwave oven (CEM, MarsXpress). A second sub-sample of the soils was also digested using a $BaCl_2$ extraction method (see Lofts et al., 2001). Filtered and acidified pore waters were analysed directly. All samples were then diluted 100-fold prior to analyses. Analysis of the digests and pore waters was conducted by ICP-MS (Perkin Elmer, NeXION 300D) (for Cs and Sr) or ICP-OES (Perkin Elmer, Optima 7300 DV) (for Ca, K and Mg). ICP-MS and ICP-OES quantitative measurements were made using external calibration after spiking the digests with 10 µg l^{-1} Ga, In and Re as internal standards to compensate for instrumental drift and matrix effects. ICP-MS and ICP-OES instrument limits of detection (LOD) were calculated by using the mean and three standard deviation measurements for the digestion reagent blank. Total method LODs for each element were then calculated to take account the sample mass and the dilution arising from the digestion procedure.

A Shimadzu TOC-L analyser, equipped with a TNM-L module, was used to measure NPOC (non-purgeable organic carbon) in pore waters. Before analysis, samples were acidified with 1M HCl and then purged with Zero grade air for 6 minutes to remove any inorganic carbon. The sample was then injected into the analyser and the remaining carbon, as NPOC, measured by combustion at 720°C, with a catalyst, which converted all carbon to carbon dioxide. The carbon dioxide was measured by an infrared detector.

To determine the percentages of clay, silt and sand in the study soils, three 0.5 g replicate sub-samples of each soil type were treated with H_2O_2 to remove soil organic matter. The remaining sample was then put into a 5% Calgon solution and left on an orbital shaker overnight. A Beckman Coulter LS13 320 laser diffraction particle size analyser was then used to determine particle size in the resultant suspension. Soil textural classifications (according to the Soil Survey of England and Wales) were attributed to the study soils using the results of these measurements and the calculator available on http://www.landis.org.uk/services/tools.cfm.

Soil pH was determined on samples of fresh soil using the method of Allen (1989). Soil loss on ignition (LOI) was determined on replicates of each soil type (Allen, 1989).

4.1.2 Results

Table 13 presents summarised data for the dry matter (DM) concentrations of Ca, Mg and Sr in study soils and Table 12 presents Ca and Sr values for plants (pore water results are shown in Figure 22 below). It can be seen from Table 12 and Table 13 that the study soils encompassed a range of soil properties.

Table 12 Properties of soils used in plant growth studies. Values shown are arithmetic mean ± standard deviation

Soil Descriptor	**Land use/ source**	**Textural classification[4]**	**pH [N]**	**% LOI [N]**	**% Sand[*]**	**% Silt[*]**	**% Clay[*]**
Spark Bridge[1]	Horticultural	Sandy silt loam	6.2±0.3 [14]	21.0±1.5 [14]	39.8±0.4	44.0±0.4	16.2±0.1
Clay loam	Arable	Clay loam - Sandy silt loam[5]	7.5±0.4 [11]	4.9±0.3 [9]	49.4±4.9	33.0±3.1	17.6±1.8
Heath	Commercial supplier[2]	Loamy sand	4.9±0.4 [11]	13.5±1.1 [9]	84.0±5.2	13.3±4.3	2.7±1.0
Loamy sand	Arable	Loamy sand	7.0±0.3 [11]	3.4±0.1 [9]	78.9±1.1	13.0±0.8	8.1±0.3
Top	Commercial supplier[2]	Sandy loam	7.3±0.2 [11]	3.5±0.3 [9]	60.3±1.4	31.1±1.1	8.6±0.3
Casar de Cáceres[3]	Pasture (semi-natural)	Sandy loam	7.0±0.6 [9]	5.5±2.2 [9]	52.7±1.3	39.4±1.0	7.9±0.3

[1]Referred to as 'Allotment' in dataset associated with the with the plant growth study (Barnett et al., submitted); [2]Available from Bailey's of Norfolk Ltd. https://www.norfolktopsoil.co.uk/;[3]Referred to as 'Spanish' in dataset associated with the plant growth study (Barnett et al. submitted); n/a data not available; [4]according to http://www.landis.org.uk/services/tools.cfm; [5]the individual samples for this site were classified as either 'clay loam' or 'sandy silt loam'; [*]All % Clay, % Sand and % Silt measurements are based on three measurements.

Table 13 Extractable concentrations of soils used in plant growth studies as determined by aqua regia and $BaCl_2$ extractions. Values shown are arithmetic mean ± standard deviation.

Soil Descriptor	Ca aqua regia extractable concentration (mg kg^{-1} DM) [N]	Ca $BaCl_2$ extractable concentration (mg kg^{-1} DM) [N]	Mg aqua regia extractable concentration (mg kg^{-1} DM) [N]	Mg $BaCl_2$ extractable concentration (mg kg^{-1} DM) [N]	Sr aqua regia extractable concentration (mg kg^{-1} DM) [N]	Sr $BaCl_2$ extractable concentration (mg kg^{-1} DM) [N]
Spark Bridge[1]	13600±920 [22]	8530±486 [22]	6610±450 [22]	467±40.0 [22]	59.3±4.82 [22]	35.1±1.71 [22]
Clay loam	6200±1330 [13]	2750±67.6 [13]	12400±1160 [13]	672±19.6 [13]	13.4±1.77 [13]	9.23±0.35 [13]
Heath[3]	2380±225 [17]	2000±308 [17]	266±61.9 [17]	176±30.7 [17]	7.01±0.90 [17]	7.02±0.74 [17]
Loamy sand	2690±225 [13]	1470±130 [13]	2390±127 [13]	264±19.4 [13]	4.83±0.72 [13]	2.93±0.26 [13]
Top[3]	3990±518 [14]	2460±169 [14]	732±75.4 [14]	56.1±12.2 [14]	9.97±1.19 [14]	6.59±0.58 [14]
Casar de Cáceres[2]	3500±1470 [18]	2190±544 [18]	1560±288 [18]	220±86.9 [18]	22.4±5.75 [18]	12.2±1.66 [18]

[1]Referred to as 'Allotment' in dataset associated with the plant growth study (Barnett et al., submitted); [2]Referred to as 'Spanish' in dataset associated with the plant growth study (Barnett et al. submitted); [3]Obtained from Bailey's of Norfolk Ltd. https://www.norfolktopsoil.co.uk/.

Table 14 Concentrations of Ca and Sr in study plants. Values shown are arithmetic mean ± standard deviation and (range); table continues to page 49.

Crop	Soil descriptor	Ca concentration (mg kg^{-1} DM)	N	Sr concentration (mg kg^{-1} DM)	N
Chard	Spark Bridge[1]	11500±2580 (8680-14400)	5	39.6±7.13 (31.3-49.2)	5
Courgette	Spark Bridge[1]	2840±970 (2680-5220)	5	9.36±2.34 (6.55-12.8)	5
Grass - 2018	Spark Bridge[1]	6580±1400 (4110-7590)	5	32.9±5.83 (22.8-37.3)	5
Grass - 2019	Spark Bridge[1]	8690±448 (7980-9060)	5	41.3±2.25 (38.3-43.7)	5
Lettuce	Spark Bridge[1]	12600±1640 (11300-15400)	5	28.0±3.11 (26.1-33.5)	5
Potato (without peel)	Spark Bridge[1]	253±5.20 (247-259)	5	0.79±0.05 (0.74-0.83)	5
Strawberry fruit - 2018	Spark Bridge[1]	1990±223 (1810-2330)	5	5.44±0.45 (5.14-6.23)	5
Strawberry fruit - 2019	Spark Bridge[1]	1640±199 (1430-1950)	5	4.37±0.90 (3.63-5.78)	5
Strawberry leaf - 2018	Spark Bridge[1]	16100±1920 (13900-19100)	5	47.9±5.34 (43.3-56.8)	5
Strawberry leaf - 2019	Spark Bridge[1]	11900±1180 (10100-13200)	5	38.0±4.44 (32.2-43.2)	5
Radish edible root	Spark Bridge[1]	3940±235 (3650-4200)	5	17.4±1.51 (15.3-18.8)	5
Radish leaf	Spark Bridge[1]	36900±2010 (34500-39400)	5	79.7±4.72 (74.4-84.0)	5
Chard	Clay loam	9960±4240 (6610-17300)	5	19.7±10.0 (14.0-37.5)	5
Courgette	Clay loam	12500	1	30.7	1
Grass	Clay loam	8680±1270 (7170-9880)	5	29.3±4.78 (24.6-35.8)	5

Crop	Soil descriptor	Ca concentration (mg kg^{-1} DM)	N	Sr concentration (mg kg^{-1} DM)	N
Lettuce	Clay loam	24100±2220 (21200-27100)	5	50.3±5.54 (442-57.8)	5
Potato (without peel)	Clay loam	192±21.4 (175-221)	4	0.58±0.06 (0.52-0.65)	4
Strawberry fruit	Clay loam	3280	1	8.19	1
Strawberry leaf	Clay loam	13600	1	36.6	1
Radish edible root	Clay loam	3230±484 (2630-3750)	5	13.9±2.61 (10.6-16.3)	5
Radish leaf	Clay loam	34200±9210 (23800-44400)	5	69.2±20.2 (48.9-92.8)	5
Chard	Heath	20000±4290 (13200-23200)	5	55.1±12.5 (38.1-66.6)	5
Courgette	Heath	2940±2180 (1030-6310)	5	6.72±5.18 (2.08-15.4)	5
Grass - 2018	Heath	4690±214 (4380-4910)	5	26.6±1.54 (24.5-27.9)	5
Lettuce	Heath	29600±2150 (4380-4910)	5	69.1±6.94 (60.5-79.2)	5
Potato (without peel)	Heath	193±8.54 (186-204)	4	0.52±0.024 (0.49-0.55)	4
Strawberry fruit	Heath	1850±497 (1320-2520)	6	4.23±1.07 (2.93-5.59)	6
Strawberry leaf	Heath	15100±6290 (11400-26300)	5	41.0±17.2 (29.6-71.5)	5
Radish edible root	Heath	2490±283 (2090-2750)	5	9.66±1.34 (8.27-11.4)	5
Radish leaf	Heath	18500±2230 (16600-22300)	5	40.3±6.48 (35.7-51.0)	5
Chard	Loamy sand	8760±1880 (7430-11800)	5	10.9±2.70 (8.59-14.8)	5

Crop	Soil descriptor	Ca concentration (mg kg^{-1} DM)	N	Sr concentration (mg kg^{-1} DM)	N
Courgette	Loamy sand	3070±1950 (1850-6490)	5	3.50±2.26 (2.17-7.52)	5
Grass	Loamy sand	8550±464 (7970-9270)	5	24.1±2.23 (21.3-27.3)	5
Lettuce	Loamy sand	17000±3240 (13400-19900)	5	19.4±4.29 (14.5-23.1)	5
Potato (without peel)	Loamy sand	302±62.7 (241-364)	4	0.48±0.01 (0.39-0.59)	4
Strawberry fruit	Loamy sand	2720±1110 (1800-4530)	6	4.76±1.98 (3.12-7.87)	6
Strawberry leaf	Loamy sand	16800±2460 (13800-19000)	5	26.6±5.70 (19.6-31.8)	5
Radish edible root	Loamy sand	4030±266 (3660-4270)	5	11.0±1.23 (9.66-12.4)	5
Radish leaf	Loamy sand	31200±2850 (28200-35600)	5	35.7±4.23 (31.0-41.6)	5
Chard	Top	14800±5050 (10200-21300)	5	55.0±24.8 (32.4-87.7)	5
Courgette	Top	7020±5210 (3330-10700)	2	12.1±9.66 (5.24-18.9)	2
Grass	Top	9410±1040 (7750-10600)	5	29.4±4.29 (23.0-33.7)	5
Lettuce	Top	17200±6440 (9390-26900)	5	26.0±8.58 (15.1-38.6)	5
Potato (without peel)	Top	464±66.5 (420-563)	4	1.05±0.15 (0.94-1.27)	4
Strawberry fruit	Top	3470±641 (2730-4290)	4	7.94±2.34 (6.33-11.4)	4
Strawberry leaf	Top	15600±2540 (12700-19600)	5	34.3±5.65 (26.9-41.8)	5

Crop	Soil descriptor	Ca concentration (mg kg^{-1} DM)	N	Sr concentration (mg kg^{-1} DM)	N
Radish edible root	Top	3854±426 (3120-4160)	5	19.9±2.10 (16.3-21.6)	5
Radish leaf	Top	35400±4050 (28800-39500)	5	95.2±11.8 (76.9-106)	5
Chard	Casar de Cáceres[2]	14800±5050 (10200-21300)	5	55.0±24.8 (32.4-87.7)	5
Courgette	Casar de Cáceres[2]	12000±2000 (10100-14100)	3	36.2±5.21 (30.9-41.3)	3
Grass - 2018	Casar de Cáceres[2]	6770±649 (6220-7580)	5	49.9±9.80 (41.2-65.9)	5
Grass – 2019*	Casar de Cáceres[2]	7290±445 (6930-8030)	5	39.8±4.68 (33.2-45.9)	5
Lettuce	Casar de Cáceres[2]	9820±537 (9440-10200)	2	29.2±1.77 (27.9-30.4)	2
Potato (without peel)	Casar de Cáceres[2]	441±54.6 (384-493)	3	1.41±0.23 (1.19-1.64)	3
Radish edible root*	Casar de Cáceres[2]	3850±426 (3120-4160)	5	19.9±2.10 (16.9-21.6)	5
Radish leaf*	Casar de Cáceres[2]	35400±4050 (28800-39500)	5	95.2±11.8 (76.9-106)	5
Strawberry fruit*	Casar de Cáceres[2]	1430±121 (1290-1620)	5	4.04±0.61 (3.03-4.54)	5
Strawberry leaf*	Casar de Cáceres[2]	12700±696 (12100-13600)	5	48.3±5.64 (42.1-55.4)	5

[1]Referred to as 'Allotment' in dataset associated with plant growth study (Barnett et al., submitted); [2]Referred to as 'Spanish' in dataset associated with plant growth study (Barnett et al., submitted); *Plants grown in UK in Spanish soil, other crops were grown in Spain.

4.2 A 'simple' approach to estimating plant Sr concentrations

The derivation of this methodology starts with two basic equations (see below) that define the soil-to-plant transfer factor (TF) and the observed ratio ($OR_{plant\text{-}soil}$). The $OR_{plant\text{-}soil}$ was proposed in the 1950's as a measure of the discrimination in Ca and Sr transfer between different environmental compartments (e.g. Comar et al., 1957):

$$TF = \frac{Dry\ mass\ activity\ concentration\ in\ plant\ (Bq/kg)}{Dry\ mass\ activity\ concentration\ in\ soil\ (Bq/kg)} \qquad [1]$$

$$OR_{plant-soil} = \frac{Plant\ Sr\ dry\ mass\ activiy\ conc.\ (Bq/kg) \Big/ Plant\ Ca\ dry\ mass\ conc.\ (mg/kg)}{Soil\ Sr\ dry\ mass\ activity\ conc.\ (Bq/kg) \Big/ Soil\ Ca\ dry\ mass\ conc.\ (mg/kg)} \qquad [2]$$

Strontium concentrations (conc.) can be radioisotope activity concentrations (Bq/kg) or stable element concentrations (mg/kg) depending upon the available data. These equations can be rearranged to give the following:

$$OR \times \frac{Plant\ Ca\ dry\ mass\ conc.(mg/kg)}{Soil\ Ca\ dry\ mass\ conc.(mg/kg)} = \frac{Plant\ Sr\ dry\ mass\ activiy\ conc.(Bq/kg)}{Soil\ Sr\ dry\ mass\ activiy\ conc.(Bq/kg)} \qquad [3]$$

Whicker & Schultz (1982) state that $OR_{plant\text{-}soil}$ generally approximates to a value of 1; this is supported by White & Broadley (2003) who suggest that there is no discrimination between Ca^{2+} and Sr^{2+} transport to plant shoots. The data from our studies described in Section 4.1 give a mean $OR_{plant\text{-}soil}$ value of 0.9 further supporting the assumption of an approximate value of 1. Therefore, we can rearrange the above equation to give an expression that predicts the strontium (activity) concentration in plants:

$$Plant\ Sr\ dry\ mass\ activiy\ conc.(Bq/kg) = \frac{(Plant\ Ca\ dry\ mass\ conc.\ (mg/kg)) \times (Soil\ Sr\ dry\ mass\ activiy\ conc.\ (Bq/kg))}{Soil\ Ca\ dry\ mass\ conc.\ (mg/kg)} \qquad [4]$$

Calcium is an essential plant nutrient under homeostatic control (Tang and Luan, 2017). Consequently, for a given crop type it is likely that there will be relatively little variation in plant Ca concentrations. Consequently, if we assume typical Ca concentrations for crops are available, then all that is required to estimate the Sr activity concentration in a crop type at a given site are estimates of the Sr activity concentration and calcium concentration in soil.

4.2.1 Testing the 'simple model'

We could have used the Ca concentrations as determined in the study crops in the experiments described above (i.e. Table 3.3) to test the proposed model. However, to enable a more independent test we have compiled a database of Ca concentrations in crops (including farm animal foodstuffs) from various published and on-line compilations (the resultant database has been published as Chaplow et al. (submitted)). Table 15 presents Ca concentrations from this database for the crops we studied in the above experiments. Note that values in Table 15 may be based on multiple measurements depending upon the source database.

Using the values from Table 15 we can predict Sr concentrations in the study crops using the measured concentrations of Sr and Ca in soils (i.e. from Table 13). Predicted and measured Sr crop concentrations are compared in Figure 18. Figure 18 presents comparisons using the aqua regia soil extraction results; there was no significant difference between measured and predicted plant Sr concentrations using either the aqua regia or $BaCl_2$ extraction results for soils ($P > 0.1$, paired t-test). No crop or soil type was consistently under- or over-predicted. It can be seen from Figure 18 that there is encouraging agreement between the predicted and measured values. The agreement is considerably better than if predictions are made using plant concentration ratio values recommended in IAEA (2010) (Figure 19). There was also consistent under prediction for most crops and soil types using the IAEA concentration ratios, the exception being the Spark Bridge soil for which predictions were generally higher than the measured values. Predictions for potatoes were over-predicted for all soil types using the IAEA concentration ratios.

Table 15 Typical Ca concentrations in study crops from Chaplow et al. (submitted)

Crop	Ca concentration arithmetic mean (range) mg kg^{-1} (DM)	Number of entries
Chard	6990 (-)	1
Courgette	4610 (3000-5420)	3
Grass	5660 (3600-7400)	15
Lettuce	7380 (3570-15300)	9
Potato	373 (320-495)	4
Radish edible root	5755 (4286-6860)	3
Radish leaf	19400 (-)	1
Strawberry	1930 (1840-2020)	2

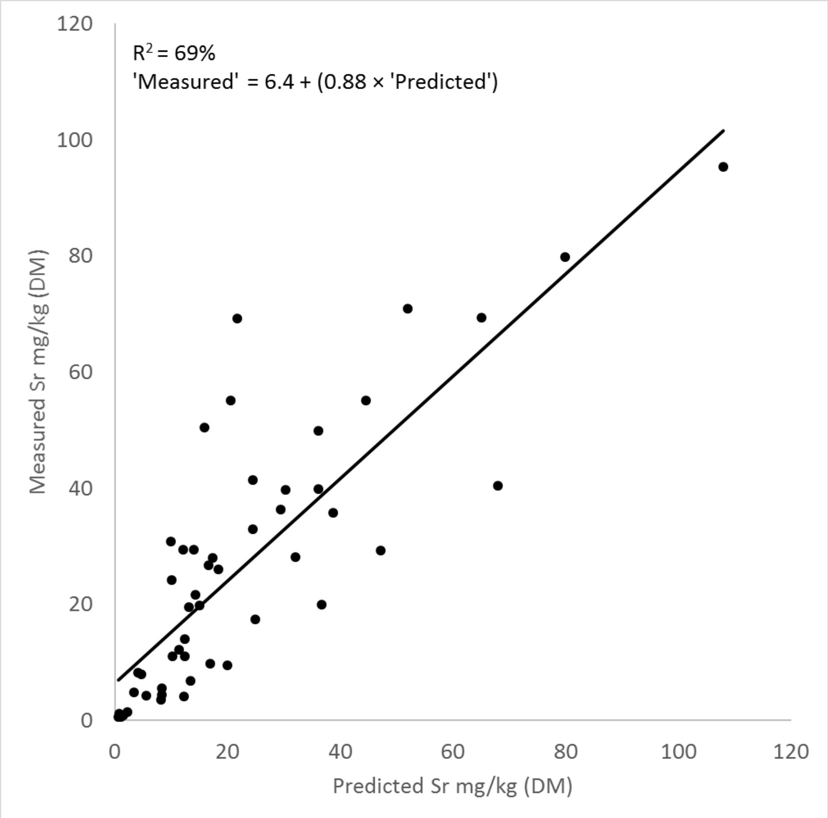


Figure 18 A comparison of measured Sr concentrations in the study crops and predictions using the simple model derived above.

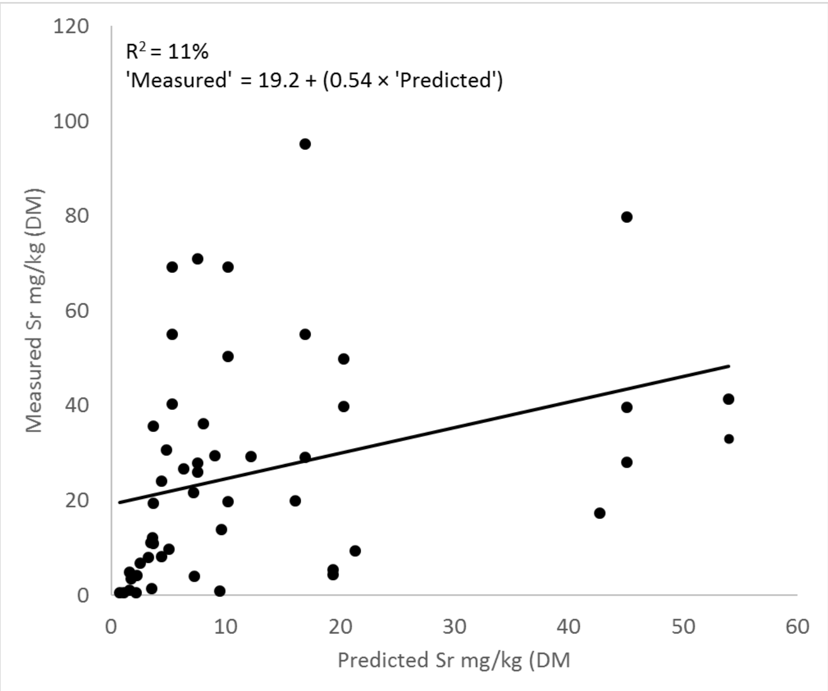


Figure 19 A comparison of measured Sr concentrations in the study crops and predictions using the recommended concentration ratios in IAEA (2010).

4.3 Adapting a chemical speciation model

The Windermere Humic Aqueous Model (WHAM) (Tipping, 1994; Tipping et al., 2011) is a process-based model of chemical equilibrium applicable to soils, waters and sediments. The model comprises sub-models for computation of cation binding to organic matter (represented by humic and fulvic acids), mineral oxides of iron(III), aluminium, manganese and silicon (Lofts and Tipping, 1998), a clay cation exchanger, and to small ligands (e.g. carbonate) in solution. WHAM has been extensively parameterised for the binding of cations, including Sr, to humic and fulvic acids (Tipping et al., 2011) and mineral oxides (Lofts and Tipping, 1998).

To calculate speciation in a soil, WHAM requires as inputs, concentrations of the solute(s) of interest, of the solute(s) that compete with those solute(s) for binding to solid and solution phase ligands, of major ions that contribute to the ionic strength of the system, and of the major binding phases (soil and dissolved organic matter, mineral oxides, clay). In practice, this means that as well as the concentrations of the solute(s) of interest, WHAM requires concentrations of the major ions Na, Mg, K, Ca, Cl, NO_3 and SO_4. To calculate speciation of metals that bind strongly to organic matter and/or mineral oxides, WHAM also requires concentrations of the competing ions Al and Fe(III) in the soil solution, although their concentrations can be estimated *a priori* (Tipping, 2005; Lofts et al., 2008).

In application to the speciation of soils, the solute concentrations required by WHAM are not the total concentrations, because these will include a portion that is not 'geochemically active', i.e. that is not participating in chemical equilibria on a sufficiently short timescale to be contributing to the solubility of the solute. This includes, for example, metals incorporated into the structure of primary and secondary minerals, and precipitates (e.g. $CaCO_{3\,(s)}$). Thus, the required concentrations of 'geochemically active' solutes must be estimated by measurements, which is typically by chemical extraction.

The above discussion shows that WHAM, as a relatively complex, process–based model, requires a large amount of driving data and can require extensive fitting to optimise agreement between measurements and outputs. For the purposes of practical post-accident predictions of ^{90}Sr, the model is over parameterised. Below we describe how we have reduced the model to make it more applicable for use in radiological assessments.

4.3.1 Soil speciation model setup

The WHAM model has already been parameterised for Sr binding to humic and fulvic acids and to mineral oxides. The model is not set up for ion binding to specific clays instead, a generic clay cation exchanger with a specific surface area of 100 $m^2\ g^{-1}$ and a cation exchange capacity of 1 μeq m^{-2} is used as a default. Strontium binding by cation exchange is likely to be an important process controlling its partitioning in soils and so parameterisation of the clay sub-model is a useful first step in model assessment.

Parameterisation of a clay model comprises selection and/or fitting of the specific surface area (SSA, m^2/g), cation exchange capacity (CEC, eq/m^2) and selectivity coefficients for binding. The Donnan cation exchange model in WHAM assumes that cations accumulate at the clay surface, within a diffuse Donnan layer, to fully neutralise the fixed negative charge. The volume of the Donnan layer, (V_{DL}, dm^3/g clay) is a function of the solution ionic strength. The concentration of a cation within the Donnan layer, $c_{\mathrm{D},i}$ (in units of moles per unit volume of the Donnan layer), is related to its concentration in the solution phase water, $c_{\mathrm{S},i}$, by the expression:

$$c_{\mathrm{D},i} = K_{\mathrm{sel},i} c_{\mathrm{S},i} R^{z_i} \qquad [5]$$

where $K_{\mathrm{sel},i}$ is the selectivity coefficient for cation i, z_i is the cation charge and R is a ratio value (with units of dm^3 solution phase water per dm^3 Donnan layer water). Values of $c_{\mathrm{D},i}$ are found by adjusting R until the total cation charge in the Donnan layer neutralises the fixed charge on the clay:

$$\frac{\sum z_i c_{\mathrm{D},i}}{V_{\mathrm{DL}}} = -\mathrm{CEC} \cdot \mathrm{SSA}\,[\mathrm{CLAY}] \qquad [6]$$

where $[\mathrm{CLAY}]$ is the mass of clay per unit volume of water (g/dm^3). Thus, accumulation of higher charge cations (e.g. Mg^{2+}, Ca^{2+}, Sr^{2+}) is favoured over lower charge cations (e.g. Na^+, K^+, NH_4^+).

While it would be possible to use parameters of an existing clay cation exchanger and adjust the amount of 'active' clay to fit observations of Sr speciation, the current parameterisation does not include selectivity coefficients, and thus predictions will be prone to unknown error. In order to address this, the clay sub-model was re–parameterised based on the work of Missana et al. (2008) and Huertas et al. (2001). Both studies used a bentonite clay, comprising 96% smectite, from Spain.

Missana et al. (2008) studied the pH dependence of the Sr partition coefficient under varying conditions of ionic strength ($NaNO_3$, I = 0.002, 0.01, 0.05 and 0.1 mol/dm^3). These data were suitable for establishing $K_{\mathrm{sel,Sr}}$ relative to Na (i.e. where $K_{\mathrm{sel,Na}} = 1$). Data from pH 3.5 to 8.0 were used for fitting, higher (more alkaline) pH data were rejected as being likely unrealistic soil conditions. The SSA was fixed to 700 m^2/g and the CEC to 1.5 µeq/m^2, both derived from the measurements of the clay properties. Initial fitting showed that the model was not able to reproduce the trend in partition coefficient with changing ionic strength at a given pH by adjusting $K_{\mathrm{sel,Sr}}$ only. A better trend could be achieved by fixing the Donnan layer volume at 0.005 dm^3/g, rather than allowing it to vary with pH. Using this layer volume value, an optimal $K_{\mathrm{sel,Sr}}$ of 6.0 was found. Fitting results are shown in Figure 20.

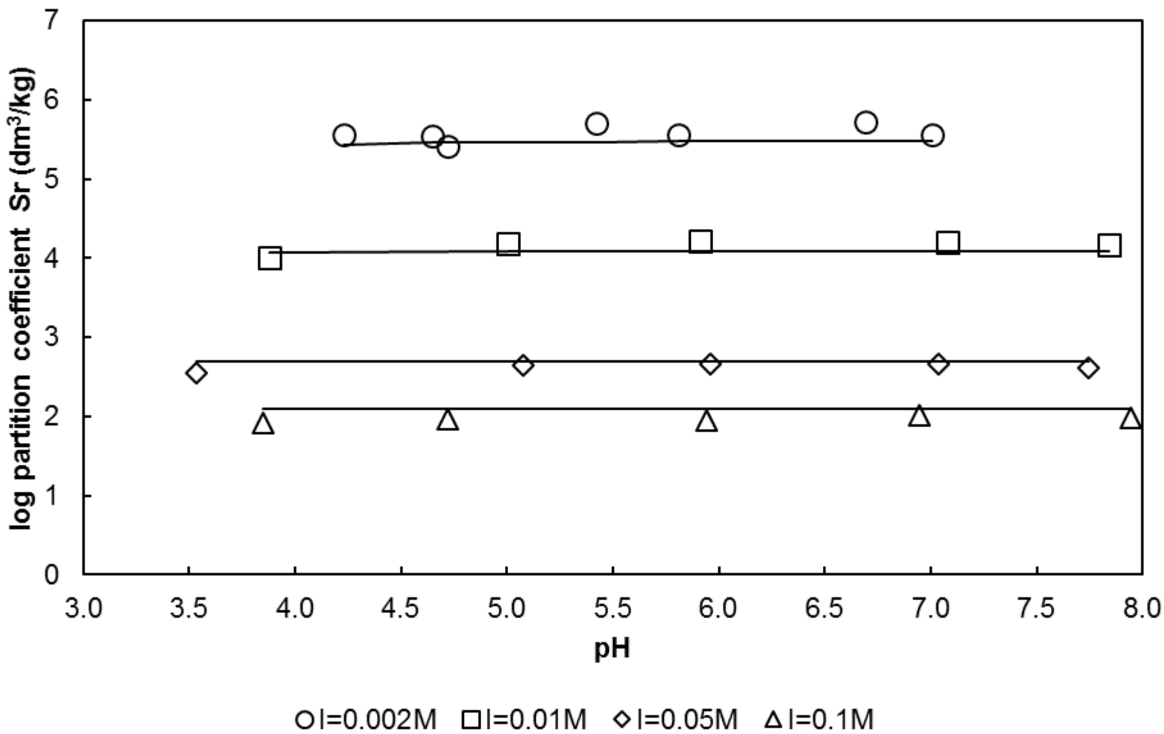


Figure 20 WHAM fits to the Sr-smectite partitioning data of Missana et al. (2008).

Missana et al. (2008) also measured the binding of ranges of concentrations of Sr to the smectite at a constant pH of 6.5 and varying ionic strength. These data were used to check the model predictions for the clay. The results are shown in Figure 21. There is a tendency to underestimate the sorbed Sr, but trends with ionic strength are predicted very well.

Huertas et al. (2001) measured the binding of Mg, K and Ca to the smectite in competition with Na, at a constant pH of 5.6. Optimal selectivity coefficients of 1.8, 6.7 and 2.4 respectively were found by fitting to observed partition coefficients (Figure 22).

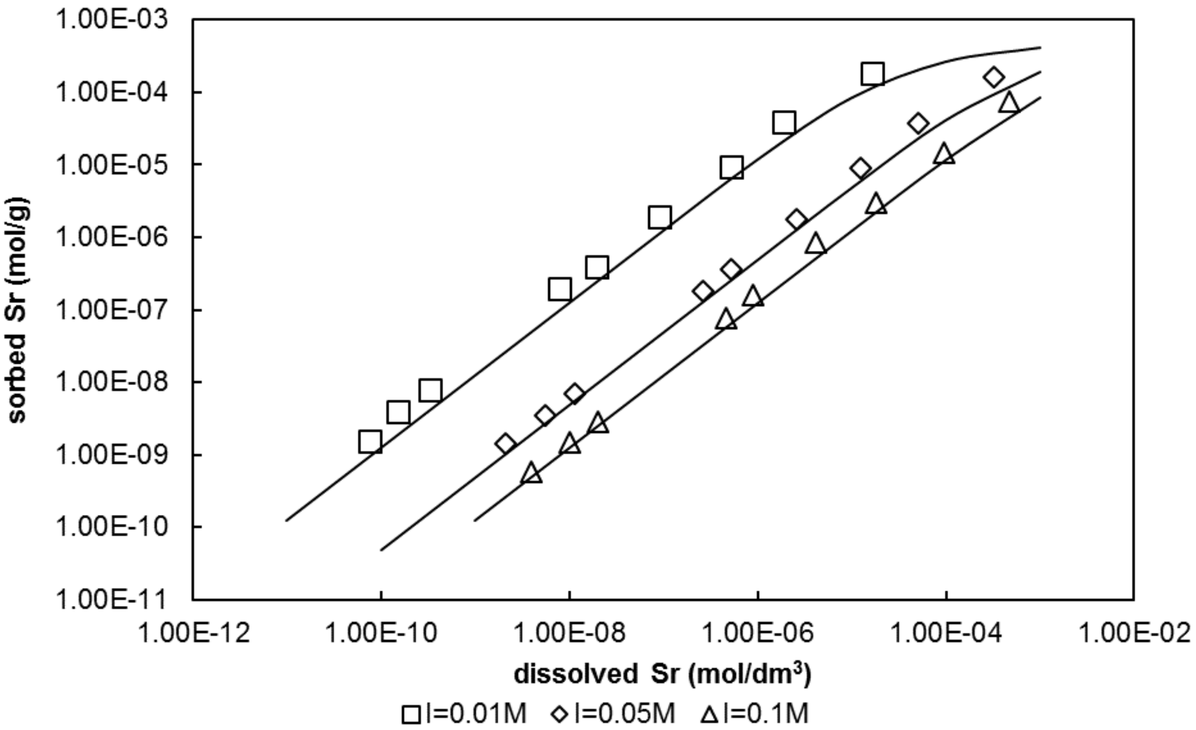


Figure 21 WHAM predictions of the Sr-smectite isotherm data of Missana et al. (2008).

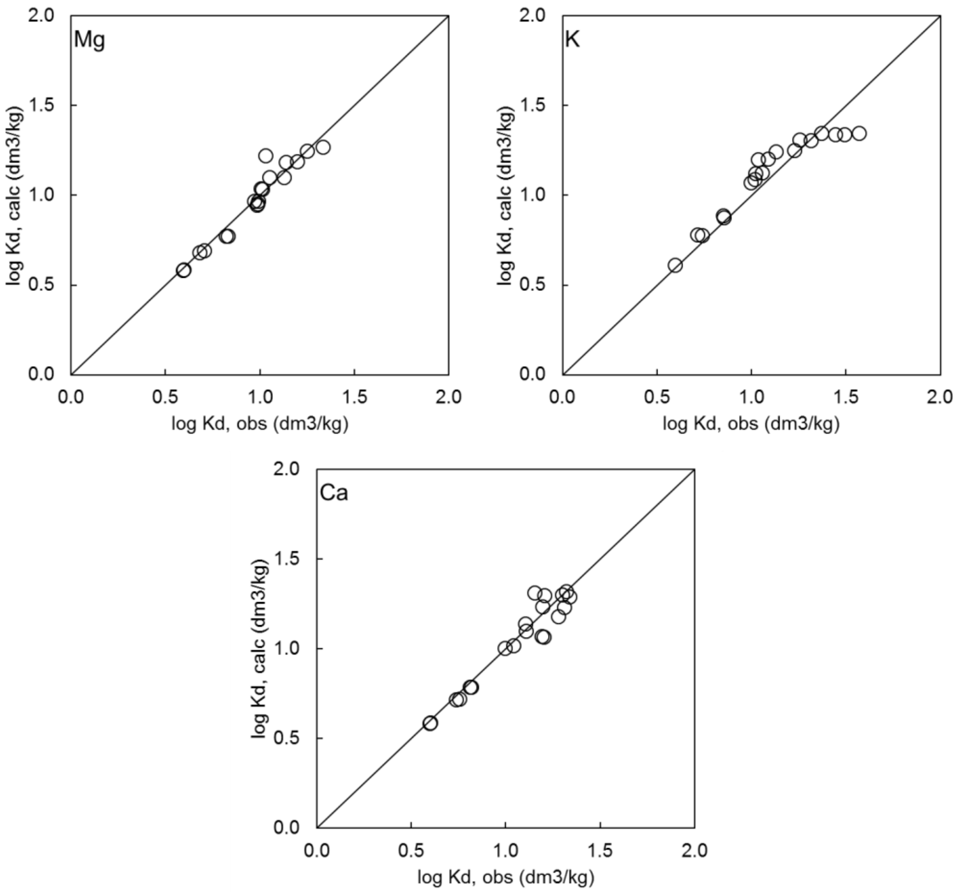


Figure 22 WHAM fitting to the dataset of Huertas et al. (2001): modelled partition coefficients of Mg, K and Ca on smectite in the presence of Na at pH 5.6.

4.3.2 Model sensitivity assessment

The sensitivity of the parameterised model to removal of input variables was assessed using a published set of Sr soil partitioning data (Gil-García et al., 2008). This dataset comprised measurements of the Sr partition coefficient for 30 Spanish soils. The soils were characterised for their pH, organic C concentration and exchangeable major cations (Na, Mg, K, Ca, NH_4) by barium chloride-triethanolamine extraction. This extraction is particularly suitable for estimation of the chemically active pools of major cations in soils, due to the swamping action of the Ba^{2+} cation in removing major cations from exchange sites by competition, without the potential confounding effects of harsher extractants, e.g. dissolution of precipitates.

WHAM was initially applied with the following input variables:

- The measured pH;
- Exchangeable Na, Mg, K, Ca and NH_4;
- Humic and fulvic acid concentrations computed from the measured organic C concentration, assuming them to be present in a ratio of 1:1;
- The carbonate system was simulated by assuming a partial atmospheric pressure of CO_2 of 400ppm;

- Al and Fe were not measured. Their chemistry was simulated using the empirical expressions of Tipping (2005) and (Lofts and Tipping, 2011), respectively, for estimation of the free ion activity in the soil solution, which allows their full speciation to be computed;
- To provide for a realistic ionic strength, the concentration of Cl in each soil was computed by forcing a charge balance (i.e. adjusting the Cl concentration until the ratio of positive to negative charge was unity);
- No concentrations of DOM (dissolved organic matter) in the solution phase of the experiment were measured. The concentration of DOM was estimated by assuming that 0.5% of the SOM (soil organic matter) was in solution. This is an oversimplification but allows for a rough assessment of the influence of DOM on predicted partitioning.

The initial model prediction overestimated the Sr partition coefficient for 29 of the 30 soils. Therefore, to provide a useful baseline for sensitivity testing, the model was optimised to the observed partition coefficients by adjusting the concentrations of clay and SOM to find the best fit to the observed Sr partition coefficients. Clay and SOM concentrations at 40% of the observed values provided the best fit (Figure 23).

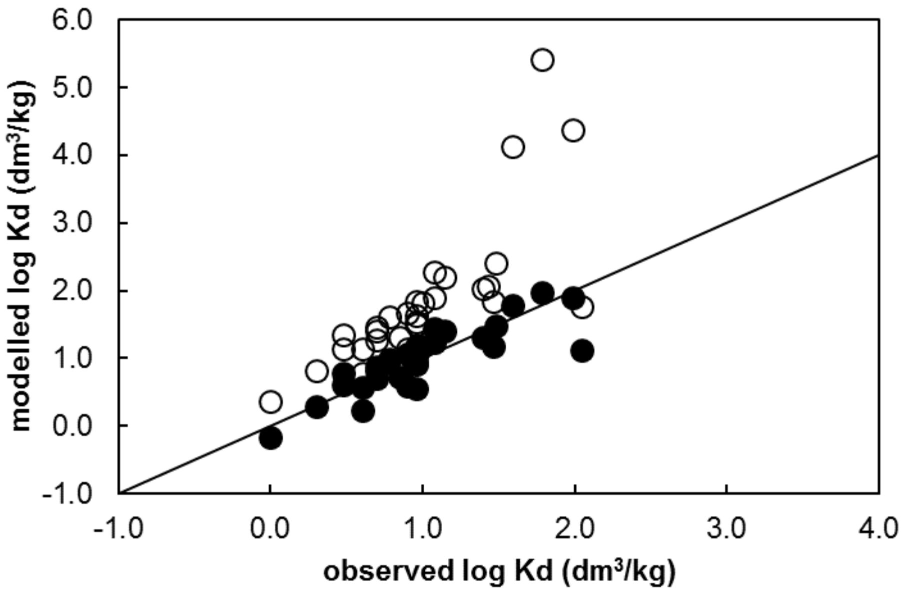


Figure 23 Blind-predicted (open circles) and optimised (closed circles) modelling of Sr partition coefficients in the dataset of Gil–García et al. (2008).

The sensitivity of the model to removal of input variables was assessed by selective removal of one or more variables to construct a series of test files. The results are summarised in Table 16, with the final test (#12) being that with the minimal set of input variables.

Table 16 Summary of model variable removal tests and outcomes. Note that soil pH could not be removed from the model and was included in all tests.

Test number	Variable(s) removed	Sum of squares error in log Kd
	none	0.259
1	Na	0.222
2	K	0.244
3	NH_4	0.259
4	Na, K, NH_4	0.208
5	Mg	0.696
6	Ca	0.402
7	Mg, Ca	2.901
8	SOM	0.366
9	DOM	0.259
10	Clay	0.779
11	CO_2	0.260
12	all except Mg, Ca, SOM, clay	0.227

The outcomes of the sensitivity testing can be summarised as follows:

- Removal of monovalent cations (Na/K/NH_4) has little effect on the goodness of prediction, showing that these cations are not greatly important influencers on Sr speciation and partitioning;
- Removal of divalent cations (Mg/Ca) has a clear effect on the goodness of prediction. In this exercise Mg had the greater effect, likely because it had on average a higher concentration than Ca in the experimental systems;
- Removal of both SOM and clay has a clear effect on the goodness of prediction, suggesting that in these soils both phases are important for binding Sr;
- Removal of either DOM or CO_2 had negligible effect.

The final chosen combination of input variables (exchangeable Mg and Ca, SOM, clay) is clearly able to predict the Sr partition coefficients as well as the full input variable set. It comprises the two main binding phases for Sr and the two main competing ions.

4.3.3 Predicting Sr concentrations in crops

To estimate Sr concentrations in crops we replace soil Sr and Ca concentrations in Equation 4 with those for predicted concentrations of Sr and Ca in soil pore waters.

Predictions of the Ca and Sr concentrations in soil pore waters using the simplified WHAM model were poor (see Figure 24). However, to predict Sr concentrations in vegetation it is in-effect the ratio of Sr:Ca concentrations in pore waters which need to be reliably predicted and not necessarily the absolute pore water concentrations. Substituting pore water concentrations into Equation 4 gives relatively good predictions of Sr concentrations in crops (Figure 25). Unlike for the simple model discussed above, there was some bias in predictions using the modified WHAM model. All predictions for the clay loam soil were under-predicted, as were all of those for grass, chard and lettuce.

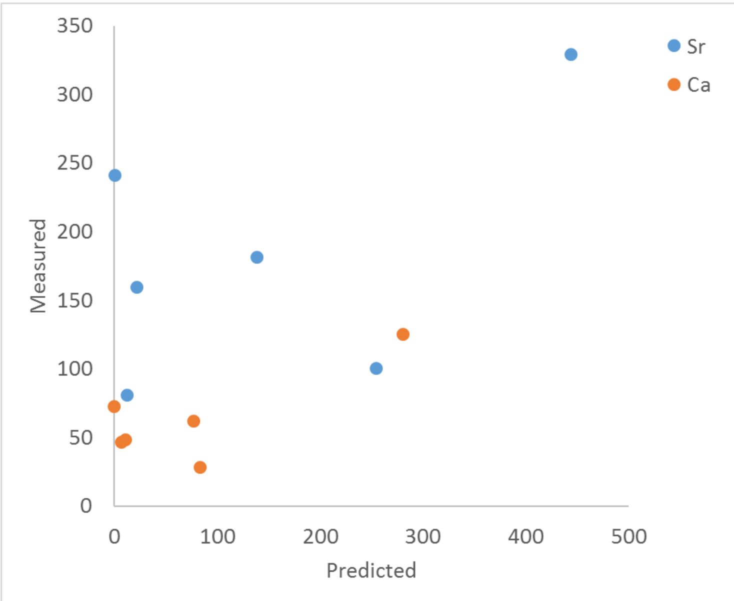


Figure 24 A comparison of measured and predicted pore water concentrations (Ca mg kg^{-1}; Sr µg kg^{-1}).

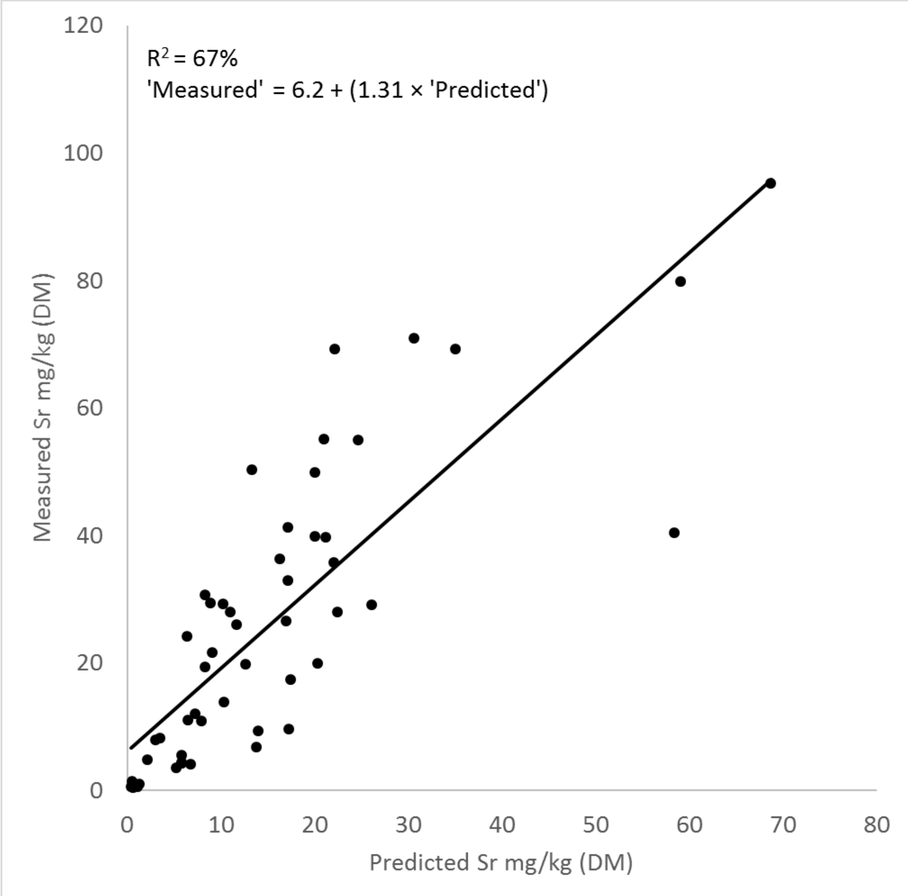


Figure 25 A comparison of measured Sr concentrations in the study crops and predictions using a modified version of the WHAM model.

4.4 Summary and recommendations

We have successfully established two process-based models to predict strontium concentrations in a range of crops using relatively few soil parameters and the calcium concentration in crops as inputs. To support these methodologies we have produced a collation of Ca concentrations in crops consumed by humans and farm animals (Chaplow et al., submitted). The approach removes the need for empirical concentration ratios and is able to make predictions for crop types for which no radioecological data exist.

Whilst the approaches produced predictions that compared well with measured data, and better than predictions using the commonly used concentration ratio approach, they require further testing against a wider range of soil types and crops. It may also be worthwhile investigating if the inclusion of Mg concentrations in plants and soil (or soil solution) improves predictions of root uptake.

A weakness of the approaches is that they can only be used to make equilibrium predictions. However, they would be sufficient to aid the identification of longer-term 'at risk areas' in the event of an accidental release. The models could be used to estimate parameters to replace existing concentration ratios in models such as FDMT (see Brown et al., 2018), which would enable their application in dynamic predictions. However, it would be preferable for future studies to consider trying to parameterise dynamic processes in soils within these process-based approaches. It would also be preferable to improve predictions of pore water concentrations using the modified WHAM model.

5 Discussion

5.1 Overview of progress on process-based models

We have successfully tested the Absalom et al. (2001) model using a wide range of European soils, with different physical and chemical characteristics, and common plant species. The test highlighted the strengths of the model and areas for further improvements. The model reproduced RCs transfer to grass well for most soils; predictions were better than those based on the TF approach. Even without calibration, the model predictions of RCs transfer to radish and spinach from many soils were acceptable considering the uncertainty associated with published TF data. Our tests also suggest that to improve the Absalom model, it should be further calibrated using more soils and plant types than had been considered in its initial parameterisation (see Section 5.3).

We have successfully developed process-based approaches to predict the strontium concentration in crops which need a minimal number of soil parameters and some knowledge of the calcium concentration of crops under consideration (to support this we have published a compendium of Ca concentrations in crops used as human and farm animal foodstuffs (Chaplow et al., submitted)).

We have demonstrated that process-based models can be relatively simply incorporated into the food chain model of an existing decision support system.

5.2 End-user views

Whilst progress was made on the development of process-based models for Cs in the 1990's-2000's such models have not been adopted for application in emergency planning/management. To begin a discussion of process-based models with end-users a workshop was held; the workshop was attended by representatives of industry, regulatory organisations, international organisations and scientists (see Appendix A for a report on this workshop).

Participants in the workshop expressed the following reservations about process-based models:

- Process-based models are too complicated requiring a considerable amount of data to implement them.
- Because of their complexity, process-based models are difficult to communicate to stakeholders, including the public.
- Process-based models have not been sufficiently tested and hence end users are not confident in their use.
- Scientists have not 'made the case' for process-based models.

Conversely, a number of advantages offered by process-based models were also highlighted:

- Process-based models offer an approach to understand/cope with the high degree of variability in empirical plant-soil concentration ratios and provide predictions more relevant to a given site.
- Process-based models (if not too complex) may be easier to explain to the public than a 'black-box' model as they better reflect reality (e.g. a model that bases predictions on easily understandable soil parameters such as percentage clay, organic matter content and/or soil potassium is easier to explain than a 'black-box' model with ratios and rate constants).
- Process-based models may be useful for site-specific assessments of existing exposure scenarios.
- Process-based models may be useful in emergency planning (though site-specific data such as soil properties would be needed).
- Process-based models may help to justify model simplifications.

5.3 Future studies and the way forward

The Absalom RCs transfer model represents a useful basis, but we recommend that the model be tested (and if required adapted) for a wider range of soil types with differing mineralogies and crops. The model predicted RCs transfer to grass from our study soils well (see Section 2). One option that should be considered to expand the range of crops the model can be applied to is to consider using 'phylogenetic' models (e.g. Willey, 2010). In this way predicted RCs activity concentrations in grass from the Absalom model could be used to make predictions for a wider range of crops. Some consideration is required with respect to the default soil depth used in the Absalom model for its application in decision support systems.

The Sr models developed here require further testing and there needs to be consideration with respect to their incorporation into dynamic human food chain models (i.e. how do we parameterise dynamic processes in soils within these process-based approaches).

The adaptation of an established chemical speciation model to predict Sr concentrations in crops is encouraging but predictions of pore water Sr and Ca concentrations were poor; the prediction of Sr concentrations in crops requires a good prediction of the Sr:Ca ratio in pore waters rather than their absolute values. Until we understand why pore water concentrations are poorly predicted we cannot recommend consideration of using such models in the development of process-based soil-plant models for other radionuclides.

There are clearly some issues we need to address before soil-plant process-based models become more widely accepted. To gain wider acceptance, when communicating process-based models to regulators and other stakeholders we need to make it clear that process-based models are not necessarily very complicated and/or resource intensive (e.g. one of the Sr models proposed in section 3 requires only an estimate of soil and crop Ca concentrations). We also acknowledge the need to validate available soil-plant process-based models for a wider range of soil types and crops than have currently been studied. Once this is done, then uptake of process-based models would benefit from some well-designed training provision aimed at different stakeholders with demonstrations of the comparative predictions of process-based and conventional empirical concentration ratio-based models.

Acknowledgements

We would like to thank May Van Hees, Clare Wells and Jacky Chaplow for assistance during the plant growth studies described in Chapters 2 and 3, and Jack Chaplow for compiling the plant Ca concentration database. We acknowledge the CEH Lancaster Analytical Chemistry Group and CEH Bangor Analytical Chemistry Laboratories for sample analyses. Finally, our thanks go to all the attendees of the workshop held in association with this deliverable and to Almudena Real (Ciemat) who was the much appreciated local organiser of the workshop.

Appendix A: Workshop 'Do process-based models have a role in human food chain assessments'

(Madrid 9-10 September 2019)

The aim of the workshop was to discuss process-based soil-to-plant transfer models. Forty stakeholders representing the industry, regulators, scientists and international organisations (see attendance list) attended the workshop and participated in the discussions regarding whether end-users saw benefit in the use and development of process-based soil-to-plant transfer models. In part, this was motivated by the priority given to process-based models by scientists (e.g. Hinton et al., 2014) versus a perceived lack of uptake of previously developed process-based models (i.e. Absalom et al., 2001) by end-users.

To enable discussion, presentations were given on: 'conventional' food chain models (e.g. the FDMT module of JRodos (Müller et al., 2004)) and FARMLAND (Brown and Simmonds, 1995); an overview of soil-to-plant transfer models for Cs; application of process-based soil-to-plant transfer models for Cs post Fukushima in Japan; and CONFIDENCE activities on Cs and Sr process-based soil-to-plant transfer models (as described in Sections 2 and in Sections 3 of this report).

Presentations were followed by facilitated 'breakout' sessions to discuss process-based models and their use. To prompt discussion, the following questions were posed:

- Q1) What is stopping 'you' from using process-based models?
- Q2) Do process-based models have a use in post-accident management?
- Q3) When should process-based models be used/when are they useful?
- Q4)Are we confident that process-based models have been sufficiently parameterised/tested?
- Q5) Are they useful in communicating information?

Below we summarise the discussions 'for and against' process-based models and the main 'take home' messages for CONFIDENCE. These discussions will also help to revise the Strategic Research Agenda for the radioecology (https://radioecology-exchange.org/content/strategic-research-agenda).

The case against process-based models

A number of participants expressed some doubts about process-based models, which can be summarised as:

1) Process-based models are too complicated requiring a considerable amount of data to implement them.
2) Because of their complexity, process-based models are difficult to communicate to stakeholders including the public.
3) Process-based models have not been sufficiently tested and hence end users are not confident in their use.
4) Scientists have not 'made the case' for process-based models.
5) Change to an established system has financial and time implications.

The case for process-based models

Other participants (including regulator/industry end-users) were of the opinion that process-based models could be useful:

1) Process-based models offer an approach to understand/cope with the high degree of variability in empirical plant-soil concentration ratios and provide predictions more relevant to a given site.
2) Process-based models (if not too complex) may be easier to explain to the public than a 'black-box' model as they better reflect reality (e.g. a model that bases predictions on easily understandable soil parameters such as percentage clay, organic matter content and/or soil potassium is easier to explain than a 'black-box' model with ratios and rate constants). A good example of this was given by a regulator who used a process-based model to describe wild boar RCs levels that took into account consumption of deer truffles (Urso et al., 2015).
3) Process-based models may be useful for site-specific assessments of existing exposure scenarios.
4) Process-based models may be useful in emergency planning (though site-specific data such as soil properties would be needed).
5) Process-based models may help to justify model simplifications.

'Take home' messages for CONFIDENCE

There are clearly some issues we need to address before process-based (or other sorts of) soil-to-plant transfer models become more widely accepted. Scientifically, for Cs, although we appear to be able to make relatively good predictions of activity concentrations in grass, predictions for other crops are currently relatively poor (see Section 2). For Sr, CONFIDENCE has made good progress in developing process-based soil-to-plant transfer models (see Section 3). Whilst useful, these models currently can only make equilibrium predictions of Sr activity concentrations in crops. However, this is an improvement on the equilibrium concentration ratio/transfer factor approach. We have to acknowledge the need to validate available process-based soil-to-plant transfer models for a wider range of scenarios (soil types and crops) than have currently been studied. Once (or if) this is done, then uptake of process-based models would benefit from some well-designed training provision aimed at different stakeholders with demonstrations of the comparative predictions of process-based and conventional empirical concentration ratio-based models.

To gain wider acceptance, when communicating process-based models to regulators and other stakeholders, we need to make it clear that process-based models are not necessarily very complicated and/or resource intensive. For instance, the Tarsitano et al. (2011; Figure A.1) implementation of the 'Absalom' model for Cs soil-to-plant transfer requires only soil clay and organic matter contents and exchangeable K. However, the model can be presented in a relatively complicated manner (Figure A.1) which may be off-putting to stakeholders. If described as '*we are using a model which takes into account the clay, organic matter and potassium contents of your soil*' it is likely that stakeholders would appreciate that the model is aiming to make predictions that are more relevant to the assessment area (and will likely reduce uncertainty compared to predictions from conventional empirical models). In the case of Sr, the simplest model proposed by CONFIDENCE requires only calcium concentrations in soil and plants (there are published compilations of Ca concentrations in a wide range of crops openly available (see Section 3). For most European countries many of the soil parameters required by process-based models will likely be available in spatial datasets such that the models could be implemented in geographical information systems to make spatial predictions (e.g. Gillett et al., 2001).

One presentation[1] emphasised that users (regulators, governmental agencies and ministries) need to have confidence in the outputs of models at their disposal. The example was given of the lack of confidence of Japanese authorities to use predictions from the Japanese government's System for Prediction of Environmental Emergency Dose Information (SPEEDI) in the management of the post-Fukushima situation (see Funabashi and Kitazawa, 2012). This further demonstrates the importance of communicating models to end users and of model validation and inter-comparison exercises (e.g. as advocated through programmes such as the IAEA's MODARIA[2]).

During the workshop, it was suggested with some agreement, that when being developed process-based models (or conceptual representations) could consider many processes. However, as they are developed the processes/parameters included in the model should be optimised to those few key parameters that really matter (i.e. through sensitivity testing). The development of models for Sr in CONFIDENCE is an example of such parameter optimisation/model reduction (see Section 3).

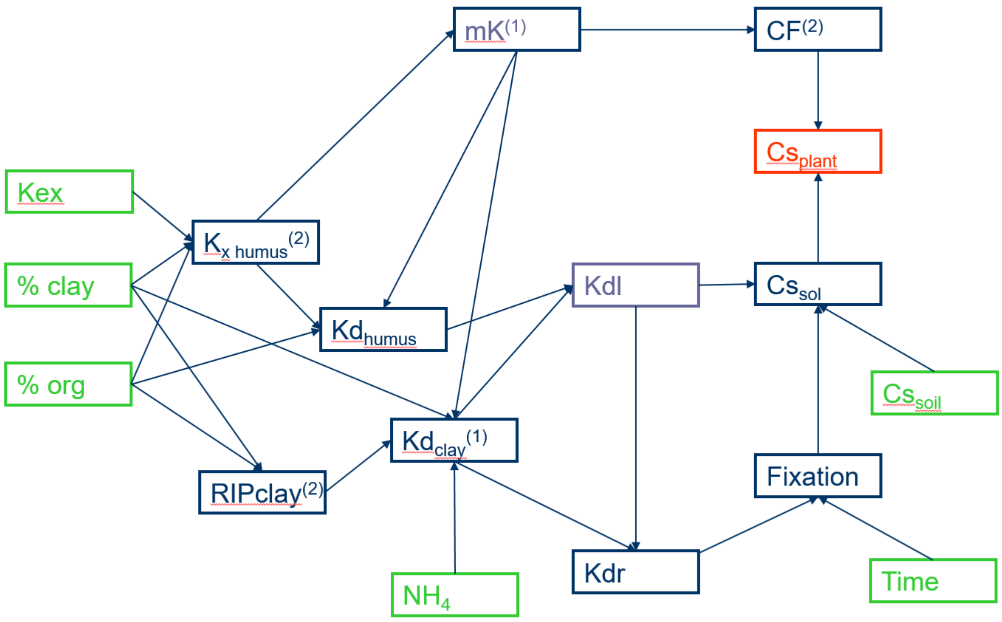


Figure A.1. A diagrammatic representation of the soil-to-plant transfer Cs model as proposed by Tarsitano et al. (2011) (courtesy of Prof. N.M.J. Crout, University of Nottingham).

With respect to post-accident response, the majority of participants agreed that the application of process-based models would become more relevant as time progressed and when more specific questions with regard to contaminated areas had to be answered. A presentation during the workshop on the application of a modification of the 'Absalom' model to optimise potassium fertilisation of rice paddy fields in areas of Japan impacted by the Fukushima accident, was considered a good example of how processed-based models could be used to address specific questions. In the earlier stages after an accidental release, many considered that conventional models (e.g. FDMT, FARMLAND etc.) would be adequate to make predictions of ingestion dose and to identify if countermeasures would be

[1]Presentation available: https://radioecology-exchange.org/sites/default/files/The%20ECOSYS%20FDMT%20model%20Overview%20advantages%2C%20limitations%20and%20suggestions%20for%20further%20development%20Proehl.pdf
[2]https://www-ns.iaea.org/projects/modaria/modaria2.asp?s=8&l=129

required. That said if process-based models were sufficiently validated and spatially implemented, they could also play an early role in identifying areas where food chain issues may persist into the longer-term. The comment (made a number of times) that, conventional models would be sufficient in the short-term but perhaps not optimal in the longer-term, implies that long-term predictions from conventional models should be communicated with care.

The observation that the model complexity may change depending upon need led to the suggestion that it would be useful to have one modelling package from which different components could be selected. The implementation of both FDMT and the 'Absalom' model into the EGOLEGO package within CONFIDENCE (Brown et al., 2018) is a step to meeting this recommendation.

Finally, it was observed that the majority of soil-to-plant transfer process-based modelling development had focussed on radionuclides of longer-term importance following an accidental release from a nuclear facility. It was suggested that process-based models may be of relevance to other scenarios, and hence radionuclides, including long-term assessments of waste disposal facilities (some work beginning to consider repository relevant radionuclides has recently been conducted, e.g. Shaw et al. (2019), RATE (2018)).

Attendance list

Forename	Surname	Organization	Country
Talal	Almahayni	SCK•CEN (Belgian Nuclear Research Centre)	Belgium
Zhanat	Baigazinov	Institute of Radiation Safety and Ecology (IRSE)	Kazakhstan
Catherine	Barnett	Centre for Ecology & Hydrology (CEH)	UK
Nick	Beresford	Centre for Ecology & Hydrology (CEH)	UK
Geert	Biermans	Federal Agency for Nuclear Control (FANC)	Belgium
Penny	Birtle	Magnox Ltd.	UK
Joanne	Brown	International Atomic Energy Agency (IAEA)	Austria
Antonella	Cristina	SCK•CEN (Belgian Nuclear Research Centre)	Belgium
Neil	Crout	Univeristy of Nottingham	UK
Damien	Didier	Institute for Radiological Protection and Nuclear Safety (IRSN)	France
Vanessa	Durand	Institute for Radiological Protection and Nuclear Safety (IRSN)	France
Sergey	Fesenko	State Scientific Center – Research Institute of Atomic Reactors (RIAR)	Russia
Laureline	Février	Institute for Radiological Protection and Nuclear Safety (IRSN)	France
Simon	French	University of Warwick	UK
Blanca	Garcia-Puerta	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Ciemat)	Spain
Javier	Guillén	University of Extremadura	Spain
Ali	Hosseini	Norwegian Radiation and Nuclear Safety Authority (DSA)	Norway
Andra-Rada	Iurian	International Atomic Energy Agency (IAEA)	Austria
Ole Christian	Lind	Centre for Environmental Radioactivity (CERAD)/ Norwegian University of Life Sciences (NMBU)	Norway
Stephen	Lofts	Centre for Ecology & Hydrology (CEH)	UK
Pilar	López Ferrando	The Spanish Nuclear Safety Council (CSN)	Spain
María	López-Ponte	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Ciemat)	Spain
Sergey	Lukashenko	Institute of Radiation Safety and Ecology (IRSE)	Kazakhstan
Milagros	Montero Prieto	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Ciemat)	Spain
Juan Carlos	Mora	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Ciemat)	Spain
Deborah	Oughton	Centre for Environmental Radioactivity (CERAD)/ Norwegian University of Life Sciences (NMBU)	Norway
Danyl	Pérez-Sánchez	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Ciemat)	Spain
Gerhard	Proehl	Consultant	Germany
Almudena	Real	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Ciemat)	Spain
Päivi	Roivainen	Radiation and Nuclear Safety Authority (STUK)	Finland
Eguchi	Sadao	National Agriculture and Food Research Organization (NARO)	Japan
Lindis	Skipperud	Centre for Environmental Radioactivity (CERAD)/ Norwegian University of Life Sciences (NMBU)	Norway
Justin	Smith	Public Health England (PHE)	UK
Martin	Steiner	German Federal Office for Radiation Protection (BfS)	Germany
Agustina	Sterling Carmona	The Spanish Nuclear Safety Council (CSN)	Spain
Simon	Streeter	Food Standards Agency (FSA)	UK
Yifu	Tong	Univeristy of Nottingham	UK
Jose Angel	Vega Vilanova	Asociación Nuclear Ascó-Vandellós II AIE (ANAV)	Spain
Tamara	Yankovich	International Atomic Energy Agency (IAEA)	Austria
Eduardo	Gallego	Technical University of Madrid (UPM)	Spain

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