

Predicting Soil to Plant Transfer of Radiocesium Using Soil Characteristics

J. P. ABSALOM,*[†] S. D. YOUNG,[†]
N. M. J. CROUT,[†] A. F. NISBET,[‡]
R. F. M. WOODMAN,[‡]
E. SMOLDERS,[§] AND A. G. GILLETT[†]

School of Biological Sciences, Sutton Bonington Campus,
University of Nottingham, Loughborough,
Leicestershire LE12 5RD, U.K.

National Radiological Protection Board, Chilton, Didcot,
Oxfordshire OX11 0RQ, U.K.

Laboratory of Soil Fertility and Soil Biology, Faculty of
Applied Biological and Agricultural Sciences, K. U. Leuven,
K. Mercierlaan, 92, B 3001 Heverlee, Belgium

A model is presented that dynamically estimates the radiocesium activity in herbage from readily available soil parameters. Three key properties underlying the bioavailability of radiocesium in soils were estimated in the model: the labile radiocesium distribution coefficient (k_{dl}), the solution K^+ concentration ($[m_K]$), and the radiocesium concentration factor (CF, $Bq\ kg^{-1}\ plant/Bq\ dm^{-3}\ soil\ solution$). These were determined as functions of the soil clay content and exchangeable K status. The effect of time on radiocesium fixation was described by two first-order decay equations. The model was initially parametrized using radiocesium uptake data from a ryegrass pot trial. Without further parameterization, the model was then tested for a wide range of soil and crop combinations using a database of published and unpublished information from a variety of sources and covering contamination time periods of 0.5–11 years. Model predictions of activity concentrations in crops were in generally good agreement with observed values.

Introduction

In May 1986, radiocesium (^{137}Cs , ^{134}Cs) from the Chernobyl accident was deposited over large areas of Europe, Belarus, Ukraine, and Russia. At the time of deposition, it was widely thought that the radiocesium would be immobilized rapidly in the soil following adsorption by clay minerals. However, radiocesium remained bioavailable in many areas. Belarus, Ukraine, and Russia received the greatest deposition with Belarus, for example, losing an estimated 20% of its cultivatable land due to contamination (1). Large areas of Western Europe were also affected, especially Scandinavia, Greece, and Northern Britain. In the U.K., for example, sheep grazing such contaminated land still contain activity concentrations of radiocesium that exceed the U.K. and EC limits of 1000 $Bq\ kg^{-1}$ fresh weight. Restrictions on the movement, sale,

and slaughter of sheep are in place 12 years after the Chernobyl accident with more than 350 farms affected in Wales. Radiocesium contamination of agricultural products in the areas of Belarus, Ukraine, and Russia also remains high. Failure to predict this long-term availability of radiocesium was partly due to the differences between the organic, acidic soils with a low clay and nutrient (K) status which received most of the U.K. deposition and the low-land clay-rich mineral soils on which most previous Cs studies had been conducted (2). Illitic clay is the principal adsorptive surface for radiocesium in soil, while potassium is the major competitor for plant and soil sorption sites. Thus, these two soil properties have a large influence on the bioavailability of radiocesium in soil.

Established models which consider radiocesium uptake by plants, such as ECOSYS (3) and PATHWAY (4), do not incorporate the effects of soil properties on radiocesium bioavailability but instead describe radiocesium uptake from a generic soil. However, radiocesium bioavailability has been shown to be strongly influenced by soil properties such as K status and clay content (5, 6), both of which vary greatly in soils. In this paper, a semi-mechanistic model is presented to estimate the activity concentrations of radiocesium in plants from soil characteristics that are routinely measured or readily estimated.

Model Description

In the model, radiocesium uptake by plants is determined from a concentration factor (CF) defined as the ratio of radiocesium activity in the plant (Cs_{plant}) to that in soil solution (Cs_{sol}). The activity concentration of radiocesium in solution is estimated from a variable distribution coefficient, k_{dl} , which is related to the clay content of the soil (% clay) and the concentration of K^+ in the soil solution ($[m_K]$). The latter property is not commonly recorded in soil databases but may be estimated from more readily available soil data: exchangeable K and clay content. Time-dependent fixation of radiocesium is described as a two-component first-order kinetic process. Figure 1 shows the relationship between conceptual pools of ^{137}Cs and the equations that relate them; the model equations are described below.

Activity Concentration of Radiocesium in Plants. Plant activity concentration, Cs_{plant} ($Bq\ kg^{-1}$), is calculated from the product of CF ($dm^3\ kg^{-1}$) and Cs_{sol} ($Bq\ dm^{-3}$)

$$Cs_{plant} = (CF)Cs_{sol} \quad (1)$$

The use of eq 1 requires the prediction of the activity concentration of radiocesium in soil solution and the value of CF. Smolders et al. (5) showed that the CF for radiocesium uptake by ryegrass may be related to $[m_K]$

$$\log(CF) = -(k_2 \log(\min([m_K], k_{lim}) + k_1)) \quad (2)$$

where k_1 and k_2 are empirical constants and k_{lim} is the K^+ concentration above which CF has a constant minimum value (4).

Soil Solution K^+ Concentration ($[m_K]$). Work by Nemeth et al. (7) suggests that a linear relationship exists between the concentration of potassium in soil solution ($[m_K]$, $mol\ dm^{-3}$) and the relative occupancy of the inorganic cation-exchange capacity (CEC_{inorg} , $cmol_c\ kg^{-1}$) by potassium (% K_x).

In the absence of clay mineralogical information, the value of CEC_{inorg} may be estimated by multiplying the % clay content by a representative charge density of 50 $cmol_c\ kg^{-1}$ (7). To

* Corresponding author. Phone: (44 115) 9516317 (outside U.K.), (0115) 9516317 (within U.K.); fax: (44 115) 9516261 (outside U.K.), (0115) 9516261 (within U.K.); e-mail: jon.absalom@nottingham.ac.uk

[†] University of Nottingham.

[‡] National Radiological Protection Board.

[§] K. U. Leuven.

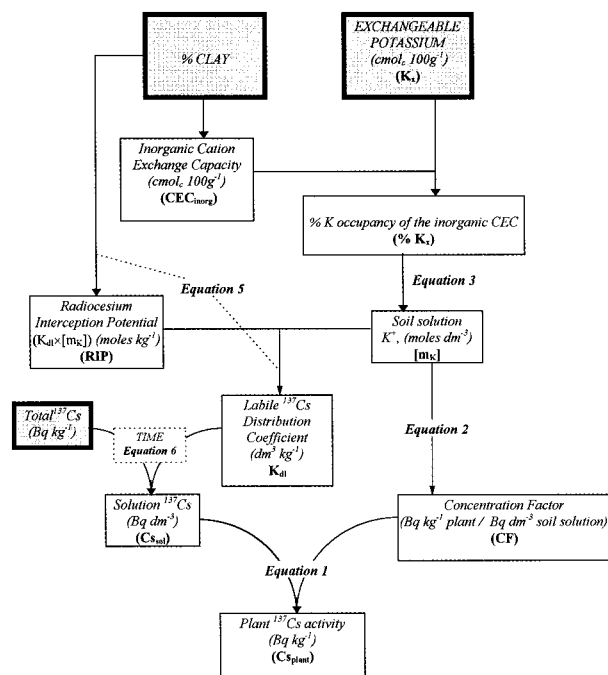


FIGURE 1. Relationship between conceptual pools of radiocesium. Shaded boxes indicate model input data. Equations describing the relationship between pools are referenced to those given in the text.

test this relationship between sorbed and solution K, published data from several sources were reanalyzed for a wide range of soil types ($r^2 = 0.837$, Figure 2). However, further application of this approach to data provided for 27 Belgian soils (Smolders, per communication) yielded mixed results:

only around 60% of the Belgian data conformed to the relationship shown in Figure 2. Lack of agreement was shown by soils with exceptionally large solution K concentrations: these were mainly coarse-textured soils with a low buffer capacity. Altman and Dittmer (8) conducted a survey of $[m_K]$ in 155 soils from 40 literature sources. The distribution of their results is overlain in Figure 2: the *non-conforming* Belgian data belong to the upper 15% of this distribution. It appears, therefore, that the simple linear relationship between $[m_K]$ and $\% K_x$ only breaks down at exceptionally large K^+ concentrations (approximately $>0.002 \text{ mol dm}^{-3}$, Figure 2) where *non-specific* adsorption sites on humus or expanded clays may control the K^+ equilibria.

To further illustrate this, a Gapon relationship between $\% K_x$ and $[m_K]$, representing wholly nonspecific adsorption, is also shown in Figure 2 (represented by a Gapon exchange coefficient, $K_G = 1.0$) (9). The large $[m_K]$ values found for some coarse-textured soils lie between the Nemeth and Gapon relationships, consistent with a transitional position between specific and nonspecific adsorption. Thus, the Nemeth relationship may be valid for soils, generally, if it is assumed that potassium is *specifically adsorbed* on the *inorganic* fraction. On this basis, a relationship between $[m_K]$ and both exchangeable K (K_{exch} , $\text{cmol}_c \text{ kg}^{-1}$) and soil inorganic CEC (a function of % clay) was derived

$$[m_K] = (k_3(\% K_x)) + k_4 \quad (3)$$

where k_3 and k_4 are coefficients derived by linear regression (Figure 2).

Labile ^{137}Cs Distribution Coefficient, k_{dl} . The role of organic matter in the adsorption of Cs has been extensively studied, and it has generally been shown that, in all but highly organic soils ($>80\%$ soil organic matter), humus is responsible for a negligible amount of Cs sorption (6, 10, 11). Rather, Cs

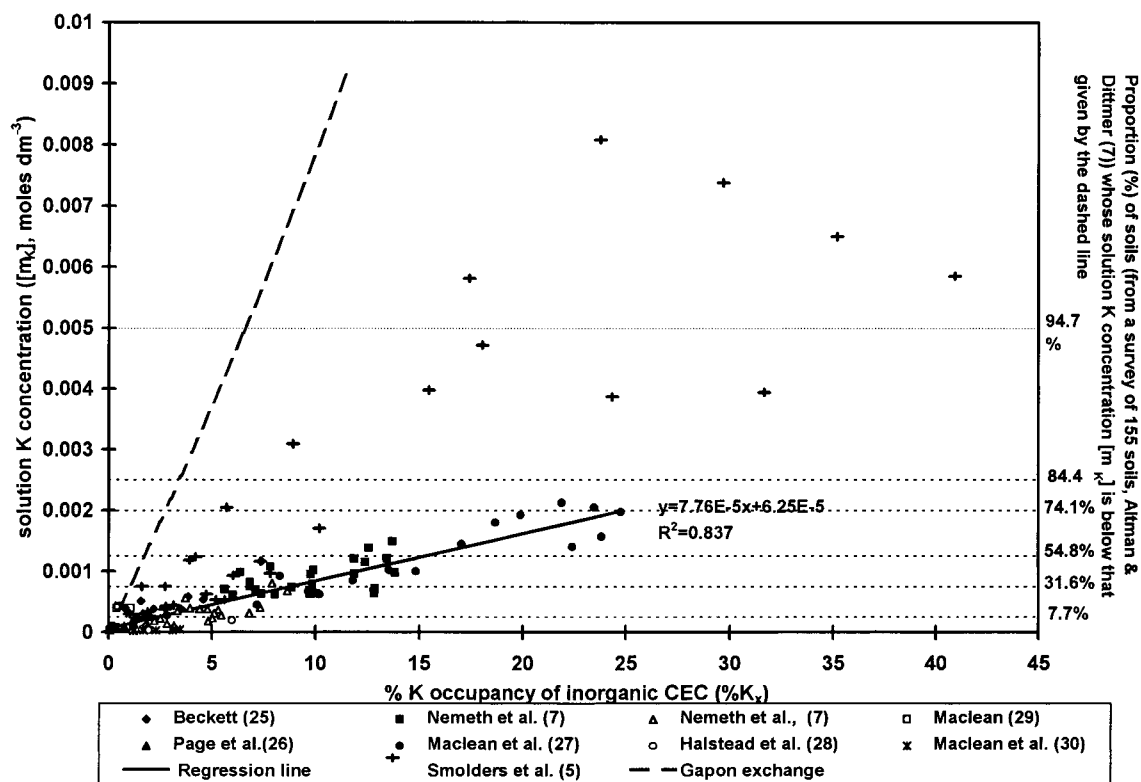


FIGURE 2. Relationship between solution K^+ ($[m_K]$, mol dm^{-3}) and $\% K$ saturation of the inorganic CEC ($\% K_x$). The solid line is a regression fit to eight literature studies; the data of Smolders et al. (1997) (+) are not included in the derivation of eq 3. The broken line is a Gapon relationship assuming $K_G = 1.0$ (Bolt, 1982), $m_{ca} = 0.01 \text{ M}$, and $\% Ca_x = 100 - \% K_x$. Horizontal dotted lines denote the frequency distribution of $[m_K]$ values from a survey of 40 literature sources (8).

TABLE 1. Description of Published Data Used To Test the Value of P_{fast} (0.81, Equation 6)

study	soil type	crop	duration, years	no. of observns	r^2 ^a
(19) Sanzharova et al. (1996)	sandy/loamy	meadow grass	7	8	0.94
	loam/clay	meadow grass	7	8	0.82
	peat/gley	meadow grass	7	8	0.88
(18) Haak and Lonsjo (1996)	sandy	natural pasture	18	18	0.85
	clay	grazing pasture	18	18	0.69

^a r^2 values represent the fit of equation 6 to the observed data in these two studies.

is strongly adsorbed at *specific* sites present in the clay mineral fraction, in particular the weathered edges of illitic and micaceous tactoids. These minerals, even when present in small amounts, are adequate to sorb the trace quantities of radiocesium resulting from deposition (6, 11). The only significant competitors for adsorption at these selective sites are potassium and ammonium. As nitrate N is likely to dominate the inorganic nitrogen pool of aerobic agricultural soils, the role of NH_4^+ may be ignored in most cases. Therefore, the solid-solution equilibrium of radiocesium in soil may be described by a radiocesium interception potential (RIP) (12). The RIP of a soil is the product of its specific $Cs^+ \leftrightarrow K^+$ exchange constant and the content of Cs/K-specific sites which are assumed to be occupied almost exclusively by K^+ . Thus, RIP may be defined in terms of just two variables, k_{dl} and $[m_K]$,

$$RIP = k_{dl}[m_K] \quad (4)$$

Data presented by Smolders et al. (5) indicated that a relationship exists between RIP and clay content. Thus, eq 4 was rearranged and k_{dl} then empirically related to $[m_K]$ and % clay by

$$k_{dl} = \frac{k_5 + (k_6[\% \text{ clay}]^2)}{[m_K]^{n_1}} \quad (5)$$

where k_5 , k_6 , and n_1 are empirical constants.

As the model includes the dynamic process of Cs^+ fixation (see below), the value of k_{dl} describes the constant distribution between the *adsorbed labile* and *solution* activity concentrations of radiocesium. This contrasts with the normal definition of a soil k_d value, which simply expresses a total Cs solid:solution ratio that would increase with time as Cs^+ fixation progresses.

Effect of Time on the Bioavailability of Radiocesium.

The bioavailability of radiocesium decreases with time (13, 14, 6). The two major processes responsible for this decline are likely to be leaching and fixation of radiocesium in the soil. As radiocesium is strongly sorbed in the soil (large k_d values), the effect of leaching is probably small. For example, apparent migration rates in German soils have been reported at between 0.8 and 2 cm year⁻¹ (15). Similarly, Golikov et al. (16) found the average vertical migration rate of radiocesium in a range of soils to be 0.61 cm year⁻¹. Thus, radiocesium is present in the soil rooting zone for decades after deposition, and leaching would appear to contribute little to the progressive reduction in bioavailability which has been observed.

Fixation is thought to occur as radiocesium slowly migrates into the collapsed interlayers of illitic and micaceous minerals (6, 11), a process that has been described by diffusion kinetics (17). The characteristics of the main Cs^+ adsorption sites and the process of fixation are probably similar for all soils containing illitic and micaceous minerals. Even highly organic soils have been shown to contain significant quantities of illite (18). Therefore, the proportional rate of decrease in the

TABLE 2. Values of the Model Parameters Used in Equations 2, 3, 5, and 6

model param	value
k_{fast} , days ⁻¹	0.0019
k_{slow} , days ⁻¹	0.00019
P_{fast}	0.814
k_{lim} , mol dm ⁻³	0.0024
k_1	5.23
k_2	2.42
k_3	7.65×10^{-5}
k_4	6.25×10^{-5}
k_5	2.38
k_6	0.27
n_1	0.676

bioavailability of radiocesium may be fairly constant over a range of soils, regardless of adsorption capacity. Studies of contaminated agricultural products have indicated that the initial rate of decline in radiocesium activity may be described by an effective ecological half-life of around 1 year (19), while, concurrently, long-term reduction rates may be modeled using a 10-year half-life (20).

Within the model, soil Cs is divided into two pools with relative magnitudes P_{fast} and $1 - P_{fast}$ subject to rapid and slow fixation rates, respectively. These are combined to calculate a dynamic factor (D , ranging from 0.0 to 1.0, eq 6) that is used to modify modeled labile and, hence, solution activity concentrations (from k_{dl} , eq 5) through time:

$$D = P_{fast}e^{-k_{fast}t} + (1 - P_{fast})e^{-k_{slow}t} \quad (6)$$

where k_{fast} and k_{slow} are the fixation rate constants (1.9×10^{-3} and 1.9×10^{-4} day⁻¹, respectively) and P_{fast} is the proportion of $Cs^0_{bioavailable}$ subject to fixation at the rate $k_{fast} = 0.81$.

Values of P_{fast} were estimated by fitting to literature study data given in Table 1, which describe temporal changes in transfer factors (Bq kg⁻¹ plant / Bq kg⁻¹ soil). The data used to determine P_{fast} came from two studies of several soil types over periods of between 8 and 17 years (21, 22) (Table 1). Equation 6 was fitted to each dataset, and in each case, significant fits to the observed data were found with r^2 values varying between 0.69 ($n = 18$) and 0.92 ($n = 8$) (Table 1).

Model Parameterization. The empirical constants in the model equations were derived by simultaneous fitting of eqs 2 and 5 to observed data (5) using a Marquardt nonlinear regression technique (23). The data for the ryegrass pot experiment conducted with a range of Belgian soils (5) included measured values of $[m_K]$, k_{dl} , and Cs_{plant} . Observed values of k_{dl} and $[m_K]$ were measured prior to the pot experiment ($k_{dl} = k_d$ at $t = 0$). Equation 6 (Figure 1) was implemented using a time period equivalent to that of soil ¹³⁷Cs exposure ($t = 60$ days). Table 2 shows the fitted values of the model parameters, while the relationship between the measured and modeled Cs_{plant} , $[m_K]$, and k_{dl} are shown in Figure 3 (a, b, and c, respectively).

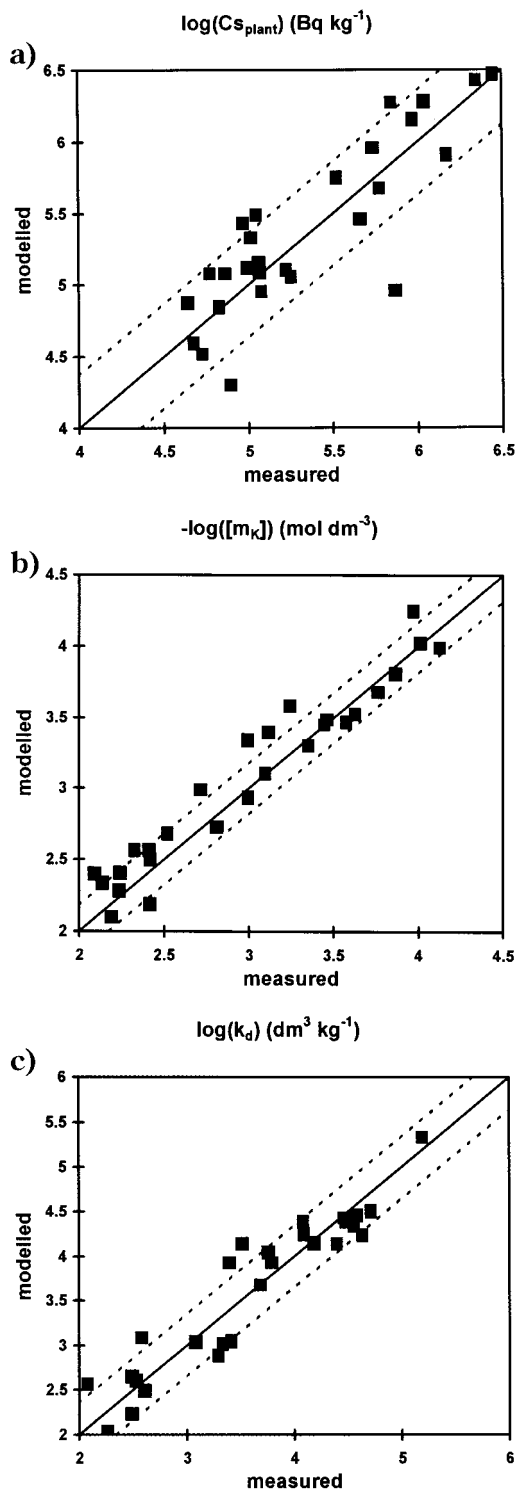


FIGURE 3. Observed versus predicted output from eqs 1–3 and 5. RSD = residual standard deviation; broken lines indicate 1 RSD on either side of the 1:1 relationship (solid line). (a) Observed versus predicted $\log(Cs_{\text{plant}})$. $r^2 = 0.58$, $n = 27$. (b) Observed versus predicted $-\log([m_k])$. $r^2 = 0.92$, $n = 27$. (c) Observed versus predicted $\log(k_d)$. $r^2 = 0.81$, $n = 27$.

Model Validation and Extension to Other Crop Types

A large number of studies of radiocesium contamination of a variety of crops have recently been collated from the literature and personal communications (24). The resulting database contains soil-to-plant transfer factors for a wide range of soil and crop types and includes information on the

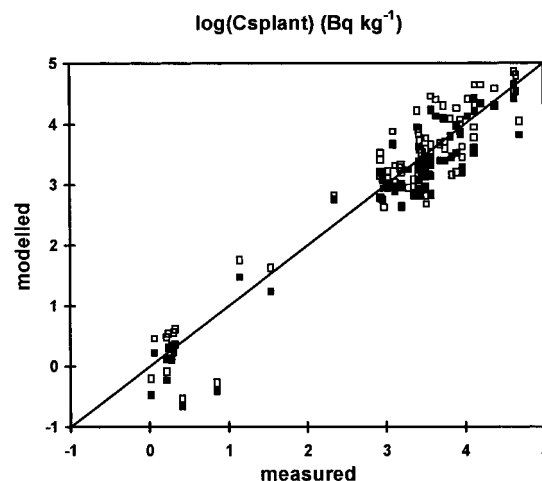


FIGURE 4. Measured versus modeled ^{137}Cs plant activity (Bq kg^{-1} , log values) for wheat (inedible fraction). Results using uptake parameters optimized for both ryegrass (■) and wheat (□) are given (Table 2); the 1:1 relationship is shown as a solid line.

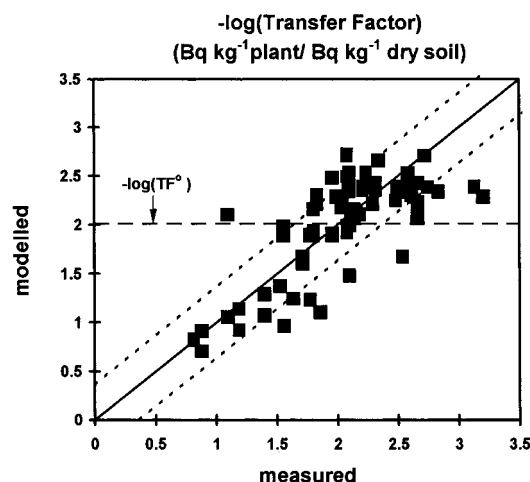


FIGURE 5. Measured versus modeled radiocesium transfer factor for wheat (grain). The model was fitted to measured transfer factor data. Broken lines indicate 1 RSD on either side of the 1:1 relationship (solid line) ($r^2 = 0.71$, $n = 78$). A single transfer factor model (TF^0) was also fitted to measured TF data and is indicated by a horizontal dashed line.

age and level of contamination, exchangeable soil K status, and % clay content. The data, which were derived from pot, lysimeter, and field investigations (summarized in Table 3), thus provide the required inputs to the model (Figure 1) and enable comparisons to be made between the predicted and observed activity concentrations of radiocesium in plants. All these verification data were screened for systematic differences resulting from experimental design, resulting in the exclusion of pot trial data in the case of wheat straw (2 data points excluded from a total of 80—Table 3). No other data points were excluded.

Two comparisons between the measured and modeled activity concentrations of radiocesium in wheat are shown in Figure 4. The first illustrates the relationship between the measured and modeled vegetation activity concentrations using model parameters estimated for ryegrass (Table 2), and applied to wheat straw ($r^2 = 0.867$, Table 3). In the second case, parameters k_1 , k_2 , and k_{lim} have been re-estimated for wheat straw (all other parameters remain unchanged), and closer agreement was achieved ($r^2 = 0.919$, Table 3). Equation 2 describes the dependence of radiocesium uptake on $[m_k]$, and it is reasonable to assume that this response varies

TABLE 3. Results of Applying the Dynamic Radiocesium Model to Various Crops^a

crop	<i>r</i> ²		model parameters			observations				
	grass, Table 1	crop-specific	<i>k</i> ₁	<i>k</i> ₂	<i>k</i> _{lim} , mol dm ⁻³	total	pot	lysimeter	field	time period, years
wheat (straw)	0.89 ^b	0.92 ^{b,d}	6.86 ± 0.25	2.93 ± 0.25	0.0014 ± 0.00046	78	0	67	11	0.5–3.5
wheat (grain)	0.75 ^b	0.91 ^{b,d}	7.22 ± 0.25	2.91 ± 0.52	0.0014 ± 0.00044	80	2	67	11	0.5–3.5
barley (straw)	0.79 ^b	0.87 ^{b,d}	1.73 ± 0.2	1.46 ± 0.49	0.022 ± 0.0012	47	4	8	35	0.5–8.5
barley (grain)	0.82 ^b	0.89 ^{b,d}	3.76 ± 0.16	1.94 ± 0.4	0.0018 ± 0.00094	77	4	8	65	0.5–10
potato (tubers)	0.62 ^b	0.81 ^{b,d}	3.73 ± 0.14	2.15 ± 0.37	0.0056 ± 0.0	80	0	14	66	0.5–11
potato (inedible fraction)	–0.429 ^c	0.69 ^{b,d}	0.85 ± 0.21	1.43 ± 1.79	0.0056 ± 0.00	23	0	14	9	0.5–11
cabbage	0.087 ^c	0.59 ^{b,d}	5.04 ± 0.63	2.65 ± 0.48	0.0056 ± 0.0	44	0	9	35	0.5–10

^a Values of *r*² represent the fit between the observed and modeled plant ¹³⁷Cs activity (log [Bq kg⁻¹]) using values of *k*₁, *k*₂, and *k*_{lim} (eq 2) derived for ryegrass (grass, Table 1) or optimized for individual crop types (crop-specific). Parameters used in eqs 3 and 5 to derive [*m*_k] and *k*_{dl} and the description of progressive radiocesium fixation (eq 7, Table 2) were independent of crop type (± values represent 1 estimated standard error). ^b *P* < 0.001. ^c Not significant. ^d In each case, the *r*² values for crop-specific parameters were significantly (*P* < 0.001) greater than the *r*² for ryegrass.

between plant species, giving unique values for the parameters *k*₁, *k*₂, and *k*_{lim} (eq 2). Therefore, crop-specific parameters were determined by fitting eq 2 to the observed data for selected crop types in the database. The parameters used in eqs 3 and 5, to predict solution K and *k*_{dl}, respectively, remained unchanged for this analysis, as they describe soil properties and are thus considered independent of crop species. The estimated values of *k*₁, *k*₂, and *k*_{lim} for each crop are given in Table 3. The model fit given by parameters derived for ryegrass were compared to those found using species-dependent values of *k*₁, *k*₂, and *k*_{lim}. For all crop types, a significant improvement in fit was observed (*P* < 0.001, Table 3). The crop data showing the greatest improvement in model fit, relative to simulations using the ryegrass parameters, were cabbage and potato. In terms of taxonomy and morphology, these two crops are the least similar to ryegrass of those tested.

The database used in the validation exercise (24) provides information on the radiocesium activity concentrations in crops resulting from radiocesium activities in soil which vary by up to 6 orders of magnitude. This difference in the level of applied radiocesium accounts for a large proportion of the variation in the measured plant activity. This was demonstrated by fitting a simple transfer factor model to the data; i.e.,

$$Cs_{\text{plant}} = (TF^0)Cs_{\text{soil}} \quad (7)$$

where TF⁰ is a constant transfer factor applied to all the data (Bq kg⁻¹ plant dry weight/Bq kg⁻¹ dry soil) and Cs_{soil} is the activity concentration of radiocesium in the soil (Bq kg⁻¹ dry soil). The resulting comparison between measured and modeled log plant activity is significant (*r*² = 0.85, *n* = 78) and compares favorably with the corresponding comparison for the proposed model (*r*² = 0.92, *n* = 78). However, the additional advantage of the model is clear from Figure 5, where a comparison between measured and modeled log TF is shown. The model can describe the observed variation in TF (over 3 orders of magnitude) in contrast to the use of the best fitting single TF value (shown in Figure 5 for comparison). Fitting the model to measured transfer factor data gave parameters that varied little from those optimized to plant activity data (Table 3), e.g., wheat grain values of *k*₁ (7.35), *k*₂ (2.95), and *k*_{lim} (0.0014).

Discussion

A model describing the availability and distribution of radiocesium in soil–plant systems is presented, incorporating the effects of soil K status, clay content, and time. Literature studies suggest that these are the major factors controlling radiocesium bioavailability. The parameterization of the

model, to the results observed for ryegrass, appeared satisfactory (Figure 3). The application of this parameterized model to the observed data for a variety of crops suggested that reasonable fits to the observed data could be achieved for similar crop species (e.g., grass and cereals). For other crops, improvements in the fit between the observed and modeled values were obtained by fitting the parameters that describe radiocesium uptake by plants in response to levels of K in soil solution (eq 2).

This represents a robust test of the model, as the compiled datasets for crop uptake of radiocesium include the results from a variety of soil types and experimental designs, over a considerable range of time periods compared to the Cs fixation half-lives observed in the literature (Table 3).

The model presented may be used to predict temporal changes in activity concentrations in plants if data are available on the exchangeable K status, clay content, and initial level of radiocesium in the soil. In the case of grass, this information could then be used to estimate the radiocesium activity in sheep or cow milk by use of simple transfer factors. Applied in such a way, estimates may be made of the time taken for animal products derived from a particular contaminated site to be reduced to acceptable levels. In the case of other crops (e.g., wheat, potato), the prediction of radiocesium activity in foodstuffs at harvest may be estimated, and decisions may be taken quickly on the most appropriate management strategy for the contaminated produce.

In the first few months following deposition, the activity concentrations of radiocesium in vegetation will be controlled by interception and translocation processes. The model described here is therefore not applicable during this early phase. Other models such as ECOSYS (3) and PATHWAY (4) contain detailed descriptions of the plant contamination processes in the early stages of a contamination event, such as interception, wash-off, and rain splash. However, these models describe plant uptake of radionuclides from the soil as being independent of soil type. For example, PATHWAY (4) uses a single transfer factor value of 0.01 Bq kg⁻¹ plant/Bq kg⁻¹ soil taken from the literature for all crops and soil types. This compares well with the single fitted value of TF⁰ (0.009, Figure 5) estimated here. ECOSYS (3) has crop-specific transfer factor values that are independent of soil type. The value given for cereals is 0.02 Bq kg⁻¹ plant/Bq kg⁻¹ soil. However, Figure 5 indicates that transfer factors for wheat grain may vary by up to 3 orders of magnitude, and the model proposed has the advantage of accounting for a significant proportion of this variation.

The influence of soil K on the adsorption of radiocesium by soils and its uptake by plants is mechanistically accounted for by the model. Consequently, it offers the potential to predict the effect of remediation strategies such as the

application of K fertilizers on a site-specific basis. The semi-mechanistic approach proposed here could be used in conjunction with existing models to improve their capability to predict plant uptake of radiocesium in response to different soil conditions.

It has been noted that pH and organic matter may play a significant role in radiocesium sorption in soils containing high (>80%) organic matter (6, 10, 11). The effects of such soil properties may also be investigated and incorporated to the model given here, increasing its applicability across a wider range of soils. Similarly, a database of soil mineralogical properties would allow the existing model to be constructed more mechanistically.

Acknowledgments

This work was funded by the European Commission DGXII, Contract No. F14PCT950015, as part of the SAVE project (Spatial Analysis of Vulnerable Ecosystems).

Literature Cited

- (1) Smith, F. B.; Clark, M. J. Meteorological Office Scientific Paper No. 42, HMSO, London, U.K., 1989.
- (2) Wynne, B. *New Sci.* **1991**, *1573*, 44–46.
- (3) Muller, H.; Pröhl, G. *Health Phys.* **1993**, *64* (3), 232–252.
- (4) Ward Whicker, F.; Kirchner, T. B. *Health Phys.* **1987**, *52*, 171–737.
- (5) Smolders, E.; Van den Brande, K.; Merckx, R. *Environ. Sci. Technol.* **1997**, *31*, 3432–3438.
- (6) Absalom, J. P.; Young, S. D.; Crout, N. M. J. *Eur. J. Soil Sci.* **1995**, *46*, 461–469.
- (7) Nemeth, K.; Mengel, K.; Grimme, H. *Soil Sci.* **1970**, *109*, 179–185.
- (8) Altman, P. L.; Dittmer, D. S. *Environmental Biology*; Federal American Society of Experimental Biology, Federation of American Societies for Experimental Biology: Bethesda, MD, 1966.
- (9) Bolt, G. H. *Soil Chemistry B. Physicochemical Models*; Elsevier Scientific: Amsterdam, 1982.
- (10) Valcke, E.; Cremers, A. *Sci. Total Environ.* **1994**, *157*, 275–283.
- (11) Hird, A. B.; Rimmer, D. L.; Livens, F. R. J. *J. Environ. Radioact.* **1995**, *26*, 103–118.
- (12) Cremers, A.; Elsen, A.; De Preter, P. *Nature* **1988**, *335*, 247–249.
- (13) Livens, F. R.; Baxter, M. S. *J. Environ. Radioact.* **1988**, *7*, 75–86.
- (14) Wauters, J.; Sweek, L.; Valcke, E.; Elsen, A.; Cremers, A. *Sci. Total Environ.* **1994**, *157*, 239–248.
- (15) Beckmann, C.; Faas, C. *Analyst* **1992**, *117*, 525–527.
- (16) Golikov, V. Y.; Balonov, M. I.; Ponomarev, A. V. In *The Chernobyl Papers*; Merwin, S., Balonov, M., Eds.; Vol. I, pp 247–288.
- (17) Bulgakov, A. A., & Konoplev, A. V. *Rad. Protect. Dosim.* **1996**, *64* (12), 11–13.
- (18) Absalom, J. P.; Crout, N. M. J.; Young, S. D. *Environ. Sci. Technol.* **1996**, *30* (9), 2735–2741.
- (19) Smith, J. T.; Fesenko, S. V.; Howard, B. J.; Horrill, A. D.; Sanzharova, N. I.; Alexahin, R. M.; Elder, D. G.; Naylor, C. *Environ. Sci. Technol.* **1999**, *33* (1), 1–208.
- (20) Jacob, P.; Likhtarev, I. Joint Study Project No. 5, Final Report EUR 16541 EN, European Commission, 1996.
- (21) Haak, E.; Lonsjo, H. *Proceedings of the International Symposium on Radioecology 1996—Ten years terrestrial radioecological research following the Chernobyl accident*; Gerzabek, M., Desmet, G., Howard, B. J., Heinrich, G., Schimmack, W., Eds.; Austrian Soil Society: Vienna, 1996; pp 129–136.
- (22) Sanzharova, N.; Belli, M.; Arkhipov, A.; Ivanova, T.; Fesenko, S.; Perepelyatnikov, G.; Tsvetnova, O. In *The Radiological consequences of the Chernobyl accident*; Karaoglou, A., Desmet, G., Kelly, G. N., Menzel, H. G., Eds.; EUR16544 EN; European Commission DG XII: 1996; pp 507–510.
- (23) Press, W. H.; Flannery, B. P.; Teukolsky, S. A.; Vetterling, W. T. *Numerical Recipes*; Cambridge University Press: Cambridge, MA, 1986.
- (24) Woodman, R. F. M.; Nisbet, A. F. Memorandum, NRPB, Chilton, 1998.
- (25) Beckett, P. H. T. *J. Soil Sci.* **1964**, *15* (1), 9–23.
- (26) Page, A. L.; Bingham, F. T.; Ganje, T. J.; Garber, M. J. *Soil Sci. Soc. Am. Proc.* **1963**, *27*, 323–326.
- (27) Maclean, A. J.; Halstead, R. L.; Mack, A. R.; Jasmin, J. J. *Can. J. Soil Sci.* *44*, 66–75.
- (28) Halstead, R. L.; Nielsen, K. F.; Maclean, A. J. *Can. J. Soil Sci.* **1957**, *37*, 61–70.
- (29) Maclean, A. J. *Can. J. Soil Sci.* **1961**, *41*, 196–206.
- (30) Maclean, A. J.; Doyle, J. J.; Hamlyn, F. G. *Can. J. Soil Sci.* **1957**, *37*, 29–33.

Received for review August 26, 1998. Revised manuscript received January 6, 1999. Accepted January 20, 1999.

ES9808853