Fit-for-purpose modelling of radiocaesium soil-to-plant transfer for nuclear emergencies: a review

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Keywords:

Nuclear emergency, Chernobyl, Fukushima, radioactive caesium, soil-to-plant transfer model, uncertainty

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Abstract

Numerous radioecological models have been developed to predict radionuclides transfer from contaminated soils to the food chain, which is an essential step in preparing and responding to nuclear emergencies. However, the lessons learned from the application of the existing models to predict soil-to-plant transfer of radiocaesium (RCs) following the Fukushima accident in 2011 renewed interest in RCs transfer modelling. To help guide and prioritise further research in relation to modelling RCs transfer in terrestrial environments, we critically reviewed existing models focusing on transfer to food crops and animal fodders.

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To facilitate the review, we categorised existing RCs soil-to-plant transfer models into empirical, semi-mechanistic and mechanistic, though several models cross the boundaries between these categories. The empirical approach predicts RCs transfer to plants based on total RCs concentration in soil and a transfer factor. The semi-empirical approach takes into account the influence of soil characteristics such as clay and exchangeable potassium content on RCs transfer, and -in contrast to the empirical approach- predicts transfer to plants based on bioavailable rather than total RCs. The mechanistic approach further considers the physical and chemical processes that control distribution and uptake of RCs in soil-plant systems including transport in the root zone and root absorption kinetics.

The empirical approach is simple and requires few inputs, but it is often associated with considerably uncertainty due to the variability in these inputs. The mechanistic approach is instrumental in understanding RCs transfer in soil-plant systems and to identify influential soil and plant parameters; however, it is too complex and data-intensive to be useful for emergency preparedness and response purposes.

We propose that the semi-mechanistic approach is scientifically sound and practical, hence more fit-for-purpose compared with the empirical and mechanistic approaches. We recommend further work to extend the applicability of the semi-mechanistic approach to a wide range of plants and soils.

1. Introduction

Nuclear accidents have released substantial amounts of radioactive caesium (RCs) into the environment. According to Steinhauser et al. (2014), between 157 PBq and 181 PBq (1 PBq = 10^{15} Bq) of RCs were released following the Chernobyl accident in 1986, and between 20 PBq and 83 PBq following the Fukushima accident in 2011.

With a physical half-life of 30 years (^{137}Cs isotope) and biogeochemical behaviour that mimics that of potassium (K), an essential plant nutrient, RCs can contaminate the food chain for a long time (Merz et al., 2013; Beresford et al., 2016). It enters plants through foliage, roots or both (RCs has no known biological role in plant growth, it is taken up by plants because of its chemical similarity to K (White and Broadley, 2000)). Interception and absorption of RCs by foliage contaminate crops and pasture for weeks or months (until harvest), whereas root uptake from contaminated soils results in lasting contamination (for decades), which may call for long-term countermeasures and remediation of the contaminated land (Wright et al., 2003). Often, these countermeasures need to be planned and implemented long before measurements from the field become available. Therefore, reliable prediction of RCs transfer is vital for effective planning of these long-term measures (Raskob et al., 2018).

Several models have been developed to predict soil-to-plant transfer (or simply transfer) of radionuclides including RCs (Whicker and Kirchner, 1987; Kirk and Staunton, 1989; Müller and Pröhl, 1993; Absalom et al., 1999, 2001; Wright et al., 2003; Keum et al., 2007; Casadesus et al., 2008). These models differ in their conceptualisation of the soil-plant system, mathematical structure and data requirements. However, the lessons learned from the application of the existing models to predict soil-to-plant transfer of radiocaesium (RCs) following the Fukushima accident in 2011 renewed interest in RCs transfer modelling (Hinton et al., 2013). Areas of interest include development of transfer models that are applicable to a wide range of environmental conditions -in contrast to existing models, which are applicable to European conditions as we shall demonstrate in this paper- and proper quantification and reduction uncertainty. For instance, the European project CONFIDENCE (COping with uNcertainties For Improved modelling and DEcision making in Nuclear emergenCiEs) focuses on reducing uncertainties in the release and the post-release phases of a nuclear emergency (Raskob et al., 2018). Addressing the long-term behaviour of radionuclides within the environment, CONFIDENCE aims to understand and reduce uncertainties associated with prediction of radionuclide transfer to human foodstuffs.

To help guide and prioritise further studies in relation to RCs transfer modelling, we critically review existing modelling approaches focussing on transfer to food crops and animal fodders. We did not review transfer models for forests since they have already been reviewed by others (e.g. Myttenaere et al., 1993; Schell et al., 1994; Riesen et al., 1999; Goor and Avila, 2003; Shaw et al., 2005; Diener et al., 2017)). This paper summarises these approaches (with example applications), discusses their strengths and limitations and concludes by presenting recommendations for further research to improve the practicality and applicability of RCs transfer models in emergency preparedness and response contexts. To structure this review, we broadly group existing RCs transfer models into empirical, semi-mechanistic or mechanistic (admittedly, many of the existing transfer models cross the boundaries between these approaches).

This paper does not review all the physical and biogeochemical processes that determine the fate of

RCs in terrestrial environments. This task has been achieved by other authors over the years (e.g. Bunzl et al., 2000; White and Broadley, 2000; Ehlken and Kirchner, 2002; Staunton et al., 2002, 2003; Vinichuk et al., 2004; Koarashi et al., 2012; Mishra et al., 2016; Burger and Lichtscheidl, 2018). Here we present as much of this topic as needed to understand the conceptual framework of the models discussed.

2. Bioavailability of RCs in soil-plant systems

Sorption on soil constituents controls RCs mobility and transfer to plants. RCs is predominantly present as a free hydrated cation in soil solution (RCs^{+}) with little or no tendency to form soluble complexes (Zhu and Smolders, 2000). In mineral soils, RCs is selectively retained on the frayed edge sites (FES) of weathered clay minerals such as illites and micas and is progressively incorporated into the clay structure (this ageing effect reduces RCs mobility and its availability to plants), whereas in organic soils, sorption on less selective sites in soil organic matter (SOM) predominates (Sawhney, 1972; Cremers et al., 1988; Absalom et al., 1995; Dumat et al., 2000; Staunton et al., 2002; Koarashi et al., 2012; Fuller et al., 2015). Consequently, RCs is generally more mobile and plant-available in organic soils of pasturelands than in mineral soils of agricultural lands (Van Bergeijk et al., 1992; Sanchez et al., 1999; Fesenko et al., 2002; Kruyts and Delvaux, 2002; Beresford et al., 2007).

Soil solution composition, particularly soluble K, profoundly affects RCs transfer to plants. On the one hand, K enhances RCs mobility in soil through competition for sorption sites (e.g. Absalom et al., 1999), on the other, K reduces RCs transfer to plants through competition for plant uptake. For instance, in a solution culture experiment with spring wheat, the concentration factor of RCs the ratio between RCs activity concentration in the plant and that in solution) was 42 times lower in the 250 μM K treatment than in the 50 μM treatment (Smolders et al., 1996). In a pot experiment with ryegrass grown on thirty grassland soils, the concentration factor decreased by 100 times as the soluble K in soil solution increased from 0.07 mM to 1 mM (Smolders et al., 1997).

It has been suggested that root uptake of Cs is via two predominant pathways on plant cell roots: the K transporter and K channel pathways (White and Broadley, 2000). Zhu and Smolders (2000) suggest that K transporters have a low degree of discrimination against Cs, whereas, K channels strongly discriminate against Cs, although K channels are suggested to be responsible for mediating most of the Cs uptake (White and Broadley, 2000).

Soil micro-organisms can play a role in the sorption of RCs in organic soils (Dighton et al., 1991; Parekh et al., 2008). Mycorrhizal fungi, which occur in symbiotic association with many plant roots, mediate the transport of mineral elements from soils to plants. Consequently, it might be anticipated that they play a role in RCs transfer to plants. However, current evidence suggests that mychorrizal fungi do not contribute significantly to the uptake of RCs by plants (Joner et al., 2004; de Boulois et al., 2008).

3. Modelling RCs transfer from soil

3.1. The empirical approach

The soil-to-plant transfer factor (TF) is exemplary of this approach. It predicts RCs activity concentration in plants, or in specific plant organs, at harvest based on total RCs activity in the rooting depth (Eq. A.1). Transfer factors have been compiled in databases with international recommendations of values for a range of soil-crop combinations (Nisbet and Woodman, 2000; Frissel et al., 2002; IAEA, 2010). When sufficiently large datasets are available, TF values have been categorised according to soil texture and organic matter content (IAEA, 2010).

The TF model assumes that RCs is uniformly distributed over the rooting depth. The IAEA (2010) recommends -for prediction purposes- to standardise the rooting depth for food crops (20 cm) and grasses (10 cm). In (semi-)natural environments the aggregated (TF_{agg}) and weighted (TF_{w}) transfer factors are more applicable since the root and RCs distributions vary considerably with depth (Almahayni (2014) showed that a standard TF would fail to predict any soil-to-plant transfer under such conditions when the release is directly to the subsoil). The TF_{agg} (Eq. A.2) predicts RCs transfer from total RCs deposition, effectively making no presumptions regarding RCs distribution within the root zone (Wright et al., 2003; Keum et al., 2007; IAEA, 2010). The TF_{w} (Eq. A.3) predicts the transfer from the soil weighted mean activity concentration, which combines into a single parameter RCs activity concentrations and the fractional abundance of plant roots in individual soil layers (Wadey et al., 2001; Shaw et al., 2004; Wheater et al., 2007).

3.2. The semi-mechanistic approach

The semi-mechanistic approach factorises the effect of soil characteristics on RCs transfer. The influential soil characteristics are typically selected based on their mechanistic relevance to the transfer process, how much of the observed variation in the transfer they explain or, preferably, both (Rigol et al., 2008; Yamamura et al., 2018).

The transfer model of Absalom et al. (1999) is an example of the semi-mechanistic approach (henceforth we refer to the model as 'Absalom1999'). It predicts RCs transfer based on the activity concentration of soluble RCs instead of total RCs in soil. The model also accounts for the competition between K and RCs as well as the effect of RCs ageing on its availability to plants (Appendix B). The main inputs of the model are RCs deposition, soil clay and exchangeable K. Parameterised for ryegrass and mineral soils, the model explained the variation in RCs activity concentration observed in wheat grains (92%; $P < 0.001$), barley grains (87%; $P < 0.001$), potato tubers (81%; $P < 0.001$) and cabbage (59%; $P < 0.001$). To extend the applicability of Absalom1999 to organic soils, Absalom et al. (2001) included non-specific sorption of RCs on

SOM and its competition with NH_4 for sorption sites (consequently, Absalom2001 requires three additional inputs: soil pH, SOM and soluble NH_4 concentration). Parameterised for grass and soils with a broad range of SOM (3.4% to 97%), the model explained 52% ($P < 0.001$) of the variation in RCs transfer to barley from mineral and organic soils, but it underestimated the transfer from soils either high in clay ($>$ 30%) or in OM ($>$ 50%) content (in three cases the deviation between model and observation was a factor of 3 greater than the residual standard deviation). Tarsitano et al. (2011) refined Absalom2001 by removing redundant parameters and re-parameterising the model using a wider range of plants (barley and wheat in addition to grass) and soils (mineral and organic). The revised version was structurally simpler yet performed better than the original Absalom2001 version.

The Absalom model has been used for different purposes and in different geographical contexts. In Europe, the model has been applied to assess contamination of crops and fodder grass with Chernobyl-derived RCs (van der Perk et al., 2000; Gillett et al., 2001; van der Perk et al., 2001; Beresford et al., 2002) and to optimise countermeasure strategies for contaminated lands (Cox et al., 2005). In Asia, the model has been applied to predict RCs transfer under (sub)tropical and flooded conditions (typical of paddy rice fields) (Rahman and Voigt, 2004; Rahman et al., 2005; Keum et al., 2007). Recently, Uematsu et al. (2016) used the Absalom model to predict RCs transfer from Fukushima-contaminated soils in Japan (mainly Andosols and Gleysols).

3.3. The mechanistic approach

Mechanistic RCs transfer models take a similar approach to traditional nutrient models, which couple a standard advection-dispersion equation to supply and demand relationships and Michaelis−Menten absorption kinetics in soil-plant systems to simulate nutrient redistribution and uptake by plants (e.g. Barber and Cushman, 1981; Oates and Barber, 1987).

Kirk and Staunton (1989) applied the mechanistic approach to predict the fate of RCs in grasslands. Their model takes into account leaching, instantaneous and time-dependent sorption and uptake by a root system that is uniformly distributed over depth. The root absorbing power parameter, which quantifies plant demand for RCs, can be adjusted to account for the effect of soluble K concentration on RCs uptake. The model predictions were not validated due to insufficient data.

Darrah and Staunton (2000) introduced new features to the Barber-Cushman model including a finite life-time of the roots, variable roots density with depth, RCs cycling within the plant and a feedback loop to soil. Kirk and Staunton and Darrah and Staunton presented no validation of the model against experimental or field data.

Roca−Jove and Vallejo−Calzada (2000) applied the Barber-Cushman model to predict RCs transfer from loamy and loamy sandy soils to pea plants in pot experiments. The model explained 88% (N=11) of the variation in RCs concentration in plant leaves and stems (the statistical significance of the correlation between model predictions and the measurements could not be established because the predictions and the measurements were not fully independent).

The BioRUR model of Casadesus et al. (2008) predicts RCs uptake based on K uptake, the ratio between K and RCs concentration in soil solution and a selectivity coefficient to account for the transport mechanism discrimination between K and RCs. Parameterised using data for sunflower grown in hydroponics, the

BioRUR overestimated RCs transfer from soil to barely by a maximum of two orders of magnitude. However, when the model accounted for K depletion in the rhizosphere, the model overestimated RCs transfer by just one order of magnitude.

4. Discussion

Each of the models presented earlier has its own advantages and disadvantages. The empirical TF model is simple and easy to use, but its predictions could be associated with considerable uncertainty due to the variability in TF data even for nominally the same plant-soil combination (Figure 1). This variability has been attributed to different sources including methodological approaches and experimental conditions such as the form of Cs used in the transfer experiment, the equilibration period and growth conditions (readers interested in a discussion of these factors should consult other publications such as IAEA (2010)).

The assumption that RCs transfer could be solely predicted form total RCs can hardly be justified since plant-available RCs does not correlate to total RCs in many soils (Ehlken and Kirchner, 2002; Tamponnet et al., 2008). As discussed in section 2, the amount of plant-available RCs in soil is largely influenced by

RCs sorption, ageing and competition with soluble K, which are not accounted for by the TF approach. Additionally, despite the wealth of TF data available for temperate conditions, data for tropical, subtropical and arid environments are still limited. Transfer of RCs under these conditions may differ substantially. For instance, soil sorption capacity in (sub)tropical regions is lower due to faster mineral weathering (Velasco et al., 2008), and hence the greater RCs transfer in these regions (Wasserman et al., 2002a,b).

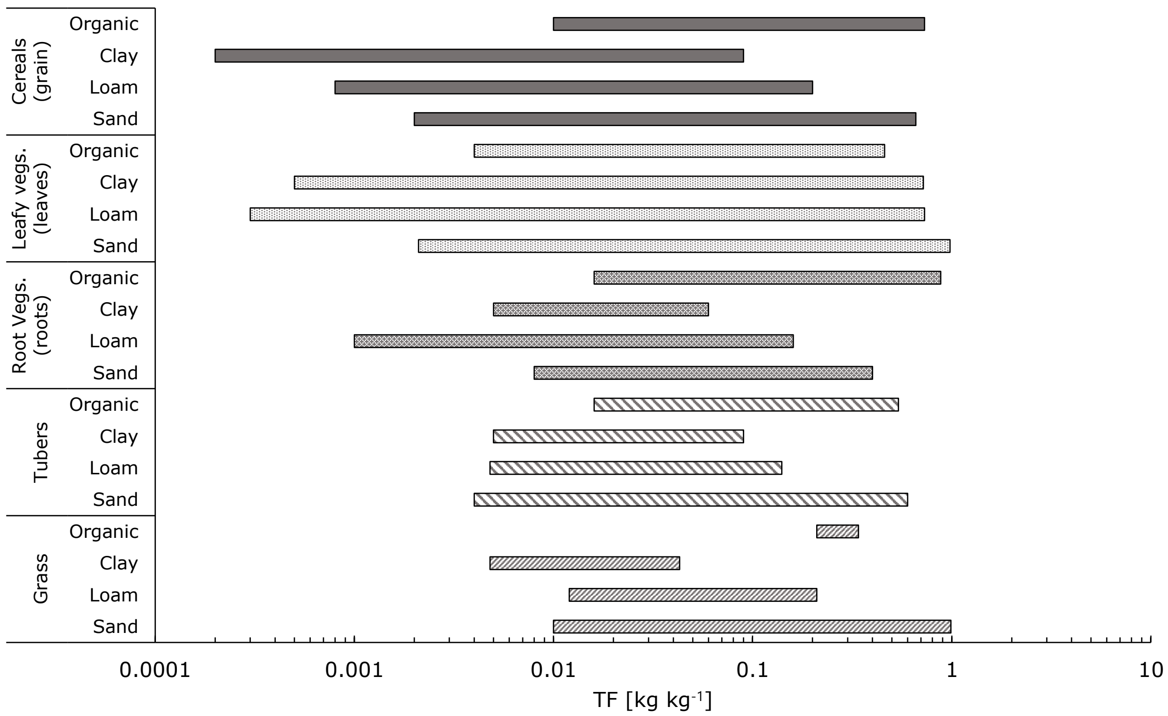

Figure 1: Variability in TF data reported in IAEA (2010) for typical food crops and grasses.

Unlike the TF model, the semi-mechanistic approach of the Absalom model relates RCs transfer to soluble RCs, which should correspond to plant-available RCs better than total RCs in soil. And because the Absalom model accounts for RCs ageing and competition with K, the model should predict RCs transfer without being unnecessarily conservative. The model inputs should be readily available from existing soil databases (e.g. Hengl et al., 2017). This makes it a practical tool to use in the context of emergency preparedness and response to identify areas vulnerable to RCs deposition and to assess the effectiveness of countermeasures that involve application of K-fertilisers.

The results from these applications suggest that the Absalom model works better for certain conditions. For instance, the model reproduced the downward trend in RCs activity in fodder grass over the ten-year simulation period in the study of van der Perk et al. (2001), and estimated reasonably well the TF value for grass (4.8×10^{-2}) compared to the measured value (5.3×10^{-2}) in the studie of Rahman et al. Predictions of RCs transfer to grass from Japanese Andosols, Gleysols, Cambisols, Acrisols and Histosols were within the range of the measurements (Uematsu et al., 2016). However, the model systematically overestimated RCs transfer to species such as potato (van der Perk et al., 2001) and rice (Rahman et al., 2005); its predictions of raddish TF correlated poorly with the measurements (Rahman and Voigt, 2004). The Absalom model tended to underestimate RCs mobility in the Japanese soils especially in the low range (RIP $<$ 1000 mmol kg^{-1}; Uematsu et al. (2016); the model-predicted RIP for these soils was up to 10 times greater than the measured value.

Most of the applications we reviewed used the Absalom model without adjustment (calibration) of its parameters, which might partially explain why the model predicted transfer to grass comparatively better than to other species. Specifically, Eq. B.7, which describes the effect of soluble K concentration on RCs uptake, have been derived for grass, and its parameters $b_{1,2}$ have been consistently identified amongst the most sensitive parameters of the model (Absalom et al., 2001; Tarsitano et al., 2011). Indeed, our calculations (not reported here) showed that increasing the default value of b_1 (1.57) by 25% (to 1.95) increased the concentration factor by 1 (at 1 mM of soluble K) to 2 (at 100 μM of soluble K) orders of magnitude, whereas increasing the default b_2 value (2.57) by the same percentage (to 3.21) decreased the concentration factor by a maximum of 1 order of magnitude (the 100 μM to 1 mM range of soluble K concentration is experimentally relevant). Differences of 25%, or more, in the default $b_{1,2}$ values are likely to be observed between plant species as we illustrate in Figure 2. Therefore, it is reasonable to assume that the Absalom model requires adjustment before applying to non-grassy vegetation (Tarsitano et al. (2011).

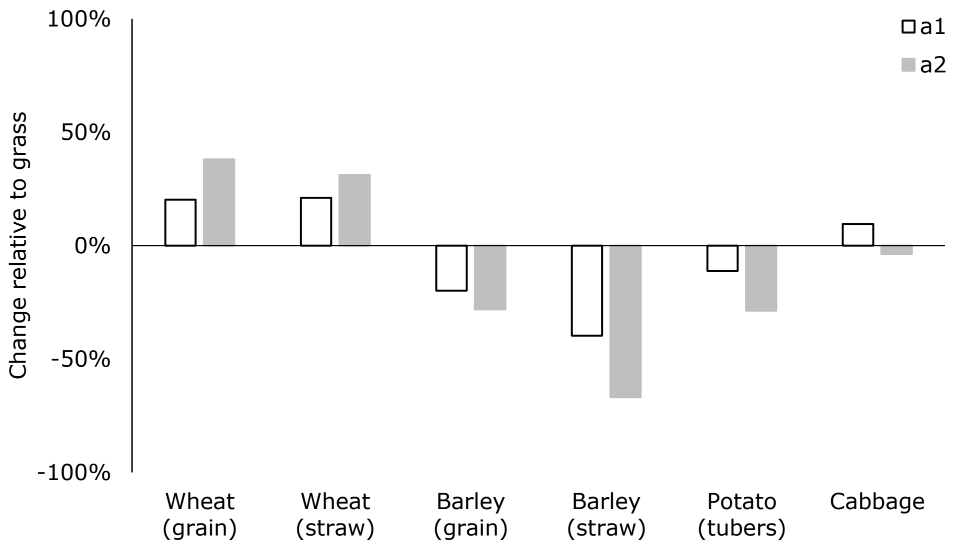

Figure 2: Between-plants variation in $a_{1,2}$ (Eq. B.3), which have the same function as $b_{1,2}$, based on Absalom et al. (1999) data. The bars represent the percentage change in the default values of a_1 (2.42) and a_2 (5.23) across different species.

Overestimation of the RIP of the Japanese soils could possibly have been due to mineralogical differences between these soils and the European soils that were used in the initial parameterisation of the Absalom model. The Japanese soils have on average three times lower RIP-per-gram-of-clay value than the European soils, possibly due to minor content of micaceous clay compared to amorphous minerals (Uematsu et al., 2015).

Mechanistic models are instrumental in fundamental RCs transfer research to better understand processes, identify sensitive parameters and hence prioritise experiments. For instance, Casadesus et al. demonstrated the sensitivity of RCs transfer to K depletion in the rhizosphere using different configurations of the BioRUR model. Achieving this task would have been less feasible using non-mechanistic models.

However, these advantages of the mechanistic models are often diminished by the complexity and the very high requirements for specific data (e.g. sorption rates and detailed description of root architecture) that are rarely available or readily measurable. Furthermore, some of the so-called mechanistic models (e.g. Darrah and Staunton, 2000; Roca−Jove and Vallejo−Calzada, 2000) do not, interestingly, seem to consider the effect of K on RCs transfer, which raises questions regarding how mechanistic these models really are. Consequently, mechanistic transfer models are seldom validated or applied to field conditions in emergency preparedness and response.

5. The way forward

In conclusion, we propose the semi-mechanistic approach of the Absalom model is a more fit-for-purpose approach for application in emergency preparedness and response context than the empirical and mechanistic approaches. It combines a sound scientific basis and practicality (using relatively available parameters).

However, the following limitations of the model need to be addressed in order to reduce the uncertainty in its predictions and broaden its applicability. Firstly, the model does not seem to work well for non-grassy species or soils with low micaceous clay content. We encourage further experimental work to parameterise the model for plants and soils that were not included in its initial parameterisation, focussing on regions around likely sources of RCs release (nuclear power plants). The purpose is to build a database of the model key inputs and parameters that are sufficiently representative of those regions to support model application. In addition to targeted transfer experiments, model parameterisation efforts could and should tap the radioecological data collected in the wake of the nuclear accidents. Particularly, the data collected following the Fukushima accident could extend the applicability of the model to none European conditions (e.g. Yoshida and Takahashi, 2012; Tazoe et al., 2012; Mikinori and Takeki, 2012; Ogura et al., 2014; Yamaguchi et al., 2016).

It is worth investigating if phylogenetic relationships, which relate soil-to-plant transfer of a number of radionuclides including RCs to plant phylogeny (or evolutionary history) (Willey, 2010), could be used to expand the applicability of the model to a wider range of crops.

The explicit assumption of the model that clay content is a good estimator of soil RIP is a gross simplification and should be revisited. Additional predictors of soil RIP (e.g. clay mineral composition) should be considered.

We recommend applying systematic model reduction techniques (e.g. Cox et al., 2006; Crout et al., 2009; Tarsitano et al., 2011) to avoid including in the model more mechanisms and parameters than can be supported by existing data (i.e. over-parameterising the model).

Acknowledgements

The work described in this paper was conducted within the CONFIDENCE project which is funded by the European Atomic Energy Community Programme H2020 under grant agreement 662287.

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Appendix A. Mathematical description of the transfer factor (TF) models

Standard TF

$$C_{plant} = \mathrm{TF} \times C_{soil} \tag{A.1}$$

$C_{plant}\left[\mathrm{Bq\ kg^{-1}_{plant\ dw}}\right]$ is the activity concentration in plants, or a specific plant organ, at harvest; $C_{soil}\left[\mathrm{Bq\ kg^{-1}_{soil\ dw}}\right]$ is soil mean activity concentration in the rooting depth.

Aggregated TF

$$C_{plant} = \mathrm{TF}_{agg} \times A_{soil} \tag{A.2}$$

$A_{soil}\left[\mathrm{Bq\ m^{-2}}\right]$ is RCs activity deposition on the ground.

Weighted TF

$$C_{soil} = \sum_{i=1}^{n} C_i f_i \tag{A.3}$$

$C_i \left[\mathrm{Bq\,kg^{-1}_{soil\,dw}}\right]$ is RCs activity concentration and f_i $[-]$ is the fractional abundance of plant roots in the i^{th} soil layer.

Appendix B. Mathematical description of the Absalom models

Absalom et al. (1999) version

$$C_{plant} = \mathrm{CF} \times C_{solution} \tag{B.1}$$

where CF $\left[\mathrm{L\,kg^{-1}}\right]$ is the concentration factor and $C_{solution}$ $\left[\mathrm{Bq\,L^{-1}}\right]$ is RCs activity concentration in soil solution.

The model estimates the CF based on the following empirical relationship (established for ryegrass plants):

$$-\log_{10}\mathrm{CF} = a_1 \log_{10}\left(\mathrm{MIN}\left(\left[\mathrm{K}^+\right], \left[\mathrm{K}^+\right]_{\mathrm{lim}}\right)\right) + a_2 \tag{B.2}$$

where $[\mathrm{K}^+]$ $\left[\mathrm{mol\,L^{-1}}\right]$ is solution K^+ concentration; $[\mathrm{K}^+]_{\mathrm{lim}}$ $\left[\mathrm{mol\,L^{-1}}\right]$ is the value of $[\mathrm{K}^+]$ above which CF is minimum.

$$\left[\mathrm{K}^+\right] = a_3\,\%\mathrm{K_x} + a_4 \tag{B.3}$$

where $\%\mathrm{K_x}$ $[-]$ is the percentage of the exchange sites on soil clay minerals occupied by K. The model assigns a default value of 50 $\left[\mathrm{cmol_c\,kg_{clay}^{-1}}\right]$ for $\mathrm{CEC^{clay}}$, which is multiplied by θ^{clay} $\left[\mathrm{kg_{clay}\,kg_{soil}^{-1}}\right]$ to express $\mathrm{CEC^{clay}}$ relative to soil weight. However, B.3 is not valid for soils where sorption onto non-specific sites in humus controls solution K^+ concentration.

$$C_{solution} = \frac{D(t)}{K_{dl}} \times C_{soil} \tag{B.4}$$

where $D(t)$ $[-]$ is a dynamic factor that modifies $C_{solution}$ to account for ageing of RCs sorbed on the solid phase:

$$D(t) = F_{\text{fast}} \times \exp^{-k_{\text{fast}} t} + \left(1 - F_{\text{fast}}\right) \times \exp^{-k_{\text{slow}} t} \tag{B.5}$$

where F_{fast} $[-]$ is the fraction of soil RCs undergoing fast fixation and $k_{\text{fast, slow}}$ $\left[\text{year}^{-1}\right]$ are the rate constants of the fast and slow fixation processes.

K_{dl} $\left[\text{L kg}^{-1}\right]$ is the distribution coefficient between exchangeable and dissolved RCs:

$$K_{dl} = \frac{\text{RIP}}{[\text{K}^+]} = \frac{a_5 \left(\theta^{\text{clay}}\right)^2 + a_6}{[\text{K}^+]^n} \tag{B.6}$$

where RIP $\left[\text{mol kg}^{-1}\right]$ is the RCs interception potential, which is estimated from θ^{clay} expressed as a percentage.

Absalom et al. (2001) version

Absalom et al. (2001) found that k_{lim} in Eq. B.2 was not necessary, hence they reduced it to the following equation:

$$-\log_{10} \text{CF} = b_1 \log_{10}\left(\left[\text{K}^+\right]\right) + b_2 \tag{B.7}$$

Absalom et al. (2001) adapted B.3 to account for the competition between K^{+} on one side and calcium (Ca^{+}) and magnesium (Mg^{+}) cations on the other side for sorption sites on soil humus. $[K^{+}]$ is calculated assuming equilibrium conditions between solution K^{+} and that sorbed on soil humus:

$$\left[K^{+}\right] = \frac{K_x^{\text{humus}} \times \sqrt{[Ca^{+} + Mg^{+}]}}{k_G^{\text{humus}} \times \left(CEC^{\text{humus}} - K_x^{\text{humus}}\right)} \tag{B.8}$$

where K_x^{humus} $\left[\text{cmol}_c\,\text{kg}_{\text{humus}}^{-1}\right]$ is exchangeable K^{+} sorbed on soil humus; $[Ca^{+} + Mg^{+}]$ $\left[\text{mol}\,\text{L}^{-1}\right]$ is solution Ca^{+} and Mg^{+} concentration; k_G^{humus} $\left[\text{mol}\,\text{L}^{-1}\right]^{-0.5}$ is the Gapon exchange coefficient for humus and CEC^{humus} $\left[\text{cmol}_c\,\text{kg}_{\text{humus}}^{-1}\right]$ is the cation exchange capacity of soil humus. Absalom et al. (2001) express most of the soil variables in Eq. B.8 in terms of more readily available information:

$$K_x^{\text{humus}} = \frac{K_x^{\text{soil}}}{\dfrac{k_G^{\text{clay}} \times \theta^{\text{clay}} \times CEC^{\text{clay}}}{k_G^{\text{humus}} \times CEC^{\text{humus}}} + \theta^{\text{humus}}} \tag{B.9}$$

$$\log_{10}\left(\left[Ca^{+} + Mg^{+}\right]\right) = b_3\text{pH} - b_4 \tag{B.10}$$

$$CEC^{\text{humus}} = b_5\text{pH} + b_6 \tag{B.11}$$

$$CEC^{\text{soil}} = \theta^{\text{clay}}CEC^{\text{clay}} + \theta^{\text{humus}}CEC^{\text{humus}} \tag{B.12}$$

where K_x^{soil} $\left[\text{cmol}_c\,\text{kg}_{\text{soil}}^{-1}\right]$ is exchangeable K^+ and θ^{humus} $\left[\text{kg}_{\text{humus}}\,\text{kg}_{\text{soil}}^{-1}\right]$ is gravimetric humus content of the soil.

In contrast to the model of Absalom et al. (1999), Absalom et al. (2001) express the labile distribution coefficient of RCs in terms of those of soil clay $\left(K_{dl}^{\text{clay}}\right)$ and humus $\left(K_{dl}^{\text{humus}}\right)$:

$$K_{dl}^{\text{clay}} = \frac{\text{RIP}^{\text{clay}}}{[\text{K}^+] + b_7 \times \left[\text{NH}_4^+\right]} \tag{B.13}$$

$$\log_{10}\text{RIP}^{\text{clay}} = -b_8\left(\frac{\theta^{\text{humus}}}{\theta^{\text{clay}}}\right) + b_9 \tag{B.14}$$

$$K_{dl}^{\text{humus}} = \frac{0.01 \times \text{K}_x^{\text{humus}}}{[\text{K}^+]} \tag{B.15}$$

$$K_{dl} = \theta^{\text{clay}} \times K_{dl}^{\text{clay}} + \theta^{\text{humus}} \times K_{dl}^{\text{humus}} \tag{B.16}$$

where RIP^{clay} $\left[\text{mol}\,\text{kg}_{\text{clay}}^{-1}\right]$ is the RIP of soil clay and $\left[\text{NH}_4^+\right]$ $\left[\text{mol}\,\text{L}^{-1}\right]$ is ammonium concentration in solution.

To account for the limited fixation of RCs^+ in soil humus, Absalom et al. (2001) modified the dynamic factor in B.5 by a factor k_{dr}:

$$k_{dr} = \frac{\theta^{\text{clay}} K_{dl}^{\text{clay}}}{\theta^{\text{clay}} K_{dl}^{\text{clay}} + \theta^{\text{humus}} K_{dl}^{\text{humus}}} \tag{B.17}$$

$$D\left(t\right) = F_{\text{fast}} \times \exp^{-k_{dr} k_{\text{fast}} t} + \left(1 - F_{\text{fast}}\right) \times \exp^{-k_{dr} k_{\text{slow}} t} \tag{B.18}$$

Tarsitano et al. (2011) version

The authors retained Eq. B.7, B.12, B.13 in their revised version of the Absalom model, and they simplified Eq. B.8, B.15 and B.17 to:

$$\left[\text{K}^{+}\right] = \frac{\text{K}_{\text{x}}^{\text{soil}}}{c_1 \theta^{\text{clay}} + c_2 \theta^{\text{humus}} - c_3 \text{K}_{\text{x}}^{\text{soil}}} \tag{B.19}$$

$$K_{dl} = \theta^{\text{clay}} \times K_{dl}^{\text{clay}} + K_{dl}^{\text{min}} \tag{B.20}$$

$$D\left(t\right) = \exp^{-k_{dr} k_{\text{fast}} t} \tag{B.21}$$

where K_{dl}^{min} is the minimum K_{dl} value for soils with negligible RCs sorption on clay minerals (e.g. organic soils).

The reset of the equations in the Absalom2001 version were removed based on their limited added value. Readers interested in the reduction analysis of Tarsitano et al. (2011) may consult the original paper.

Default values of the Absalom model parameters

Table B.1: Default values of the Absalom model parameters

Parameter	**Equation**	**Absalom1999**	**Absalom2001**	**Tarsitano2011**
a_1 (ryegrass)	B.2	2.42		
a_1 (wheat/straw)	B.2	2.93		
a_1 (wheat/grain)	B.2	2.91		
a_1 (barley/straw)	B.2	1.46		
a_1 (barley/grain)	B.2	1.46		
a_1 (potato/tubers)	B.2	2.15		
a_1 (cabbage)	B.2	2.65		
a_2 (ryegrass)	B.2	5.23		
a_2 (wheat/straw)	B.2	6.86		

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Table B.1 – continued from previous page

Parameter	Equation	Absalom1999	Absalom2001	Tarsitano2011
a_2 (wheat/grain)	B.2	7.22		
a_2 (barley/straw)	B.2	1.73		
a_2 (barley/grain)	B.2	3.76		
a_2 (potato/tubers)	B.2	3.73		
a_2 (cabbage)	B.2	5.04		
a_3	B.3	7.65×10^{-5}		
a_4	B.3	6.25×10^{-5}		
a_5	B.6	0.27		
a_6	B.6	2.38		
n	B.6	0.676		

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Table B.1 – continued from previous page

Parameter		Equation	Absalom1999	Absalom2001	Tarsitano2011
$[K^+]_{lim}$	$[mol\,m^{-3}]$	B.2	2.42		
CEC^{clay}	$[cmol_c\,kg^{-1}_{clay}]$	B.3	50	50	50
k_{fast}	$[year^{-1}]$	B.5	0.693	0.693	
k_{slow}	$[year^{-1}]$	B.5	0.0693	0.0693	
F_{fast}		B.5	0.814	0.814	
b_1 (ryegrass)		B.7		1.56	1.64
b_2 (ryegrass)		B.7		2.57	2.49
b_2 (wheat)		B.7			3.45
b_2 (barley)		B.7			3.21
b_3		B.10		0.16	

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Table B.1 – continued from previous page

Parameter		Equation	Absalom1999	Absalom2001	Tarsitano2011
b_4		B.10		3.368	
b_5		B.11		29.72	
b_6		B.11		−34.66	
b_7		B.13		4.167	1.57
b_8		B.14		0.043	0.077
b_9		B.14		1.74	1.65
$k_{\mathrm{G}}^{\mathrm{humus}}$	$[\mathrm{mol\,L^{-1}}]^{-0.5}$	B.8		2.32	
$k_{\mathrm{G}}^{\mathrm{clay}}$	$[\mathrm{mol\,L^{-1}}]^{-0.5}$	B.9		3.18	
c_1		B.19			2451
c_2		B.19			4645

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Table B.1 – continued from previous page

Parameter		Equation	Absalom1999	Absalom2001	Tarsitano2011
c_3		B.19			69.2
K_{dl}^{min}	$[\mathrm{L\,kg^{-1}}]$	B.21			134.8