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REVIEW

Models for Assessment on the Uptake of Radio-cesium by Rice Using Soil Parameters Calibrated for Paddy Fields in Fukushima Prefecture

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Abstract

Two recently developed models of radio-cesium (RCs) transfer from soil to rice grains that were parameterized and ready-to-use for paddy fields in Fukushima were reviewed to establish model-based countermeasures for food safety management in Fukushima. A probability density function model of the transfer factor from soil to rice grains based on soil exchangeable potassium (K) was compared with that of a rice grain RCs concentration model on the basis of the concentration of soil exchangeable RCs and soil exchangeable K. The relevance between these two models was demonstrated using soil exchangeable RCs as a quantitative index of the availability of soil RCs and observed data. The results indicate that the probability density of the transfer factor is affected by soil exchangeable RCs and that the use of a log-normal distribution in the model is justified by the observed distribution pattern of soil exchangeable RCs. This finding improves the accountability of the differences in the modeled probability densities for different regions and helps design countermeasures using the two models for different purposes, such as precautions for K fertilization or conservative risk assessment of shipping restrictions.

Discipline: Agricultural Environment
Additional key words: bioavailability, risk management, soil exchangeable potassium, soil exchangeable radio-cesium, transfer factor

Introduction

The uptake of radio-cesium (RCs) from soil by rice plants in paddy fields can be effectively suppressed by the application of potassium-containing fertilizers, such as potassium chloride (Fujimura et al. 2013, Saito et al. 2014, Kato et al. 2015). This countermeasure has been adopted in government subsidies for the rehabilitation of agriculture (Fukushima Prefectural Government 2011) and has been widely applied in areas affected by the accident at the Tokyo Electric Power Company, Fukushima Daiichi Nuclear Power Plant, in March 2011. It supported farmers to continue cultivation, as well as to restart cultivations in evacuated areas after soil decontamination (e.g., surface soil removal plus dressing with clean soils).

After successful social implementation, withdrawal of the countermeasure has been proceeding in areas where evacuation orders have not been applied. Consequently, to estimate an appropriate level of soil exchangeable potassium (K) corresponding to different levels of soil contamination and availability of RCs, major focus in research has shifted to optimizing this very effective yet cost-intensive countermeasure. Increasing importance has been placed on the prediction of RCs transfer to rice grains and appropriate levels of soil exchangeable K using models based on soil parameters.

For applications of RCs transfer models, models that are suitable for the purpose and sufficiently robust should be selected. To help establish a comprehensive countermeasure utilizing models and soil data

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inventories, this study focuses on two recently developed models (Yamamura et al. 2018, Yagasaki et al. 2019a) that have been calibrated for rice paddy fields in Fukushima. They have a function that considers the effect of soil exchangeable K on RCs transfer, and thus can be integrated with the ongoing K fertilization countermeasure. The first part of this study presents a short review of the major development history of RCs transfer models to highlight the features and strengths of the two models. Unlike the comprehensive reviews of numerous models by Yasuda (1995) and Almahayni et al. (2019), this study intends to narrow the focus on applications of the two ready-to-use models under the situations in Fukushima, thus presenting a short review only for the comparison and in-depth understanding of the two models. The second part of this study provides an in-depth look at the two models to discuss their effectiveness as well as the development of an integrated approach.

Development in the functions of radio-cesium transfer models

Among numerous models, empirical models based on the soil-to-plant transfer factor are the simplest, assuming that the activity concentration of RCs in crop or crop parts is linearly proportional to that in soil. This approach is adopted not only in the so-called equilibrium model (Yasuda 1995) but is also used in dynamic models, such as PATHWAY (Kirchner et al. 1983). Although a database on the transfer factor was compiled (IAEA 2009), the results showed large variability even for the same crop in the same soil groups. Ehlken and Kirchner (2002) argued that large variability is due to the processes influencing plant uptake of radionuclides, such as competing ions and bioavailability of radionuclides in soils. Absalom et al. (1999) also developed a model by considering the effect of K ions as competing ions as well as the availability of RCs in soils. This model and its successor refined models (Absalom et al. 2001, Tarsitano et al. 2011) are based on the concentration of RCs and K in soil solution as the primary driving factors for RCs transfer. The model estimates these concentrations using the distribution coefficient (K_d) between the soil solution and adsorbed phase. The coefficient of RCs is estimated by the radio-cesium interception potential (RIP) (Cremers et al. 1988), as well as by total RCs and exchangeable K in soil. As the RIP is cost-intensive to measure, the models adopt a strategy to reduce the input data requirement by estimating RIP on the basis of the soil clay content and soil solution K concentration. The latter is further estimated with the soil exchangeable K and cation exchange capacity. However, for Japanese soils, the model was found to overestimate RIP, likely due to the lower content of weathered micas in the clay fraction (Uematsu et al. 2015). Thus, this strategy can be effective only when estimations are successful. Almahayni et al. (2019) concluded that extending model parameter tuning for soils and plants that were not included in its initial parameter tuning is encouraged. For example, the model overestimated RCs transfer to grass in areas affected by the Fukushima accident, even when measured RIP was used as input (Uematsu et al. 2016). Overall, the inclusion of the effect of soil K and availability of soil RCs has been a major path in the evolution of the models, which is associated with an increasing necessity for the calibration of multiple parameters.

Alternative modeling approach to consider the availability of soil radio-cesium

After the Fukushima accident, several studies have adopted an alternative approach to consider the availability of soil RCs by using soil exchangeable RCs determined via soil extraction with 1 mol L^{-1} ammonium acetate solution (Koyama 2005, Kondo et al. 2015). This approach directly assesses the soil RCs availability and is in contrast to those using distribution coefficients and total soil RCs. Both approaches consider the concentration of RCs and K in soil solution as two controlling factors for RCs transfer but greatly differ in input data requirements. The soil exchangeable RCs data were included in a multipoint survey conducted by the government in Fukushima after the accident. Using this dataset, Yagasaki et al. (2019a) developed a model and explained 68% of the variance in the observed concentration of ^{137}Cs in rice grains, as follows:

$$\log A_{rice} = a \log A_{ex} + b \log K_{ex} + c \qquad \text{(Eq. 1)}$$

where A_{rice}, A_{ex}, and K_{ex} denote the concentration of rice grain RCs (Bq kg^{-1}), soil exchangeable RCs (Bq kg^{-1}), and soil exchangeable K (mg K kg^{-1}), respectively (Table 1). The importance of the two model variables was found to be the same. The practical application of this model for risk management can also use a 95% upper prediction interval. Additionally, the application of an inverse model can be adopted to estimate the required levels of soil exchangeable K to lower the intervention level of rice grain RCs concentrations at different levels of soil exchangeable RCs (Yagasaki et al. 2019b).

This model considers the logarithm of the rice grain ^{137}Cs concentration. Thus, model parameter tuning is affected by samples in a relatively low concentration

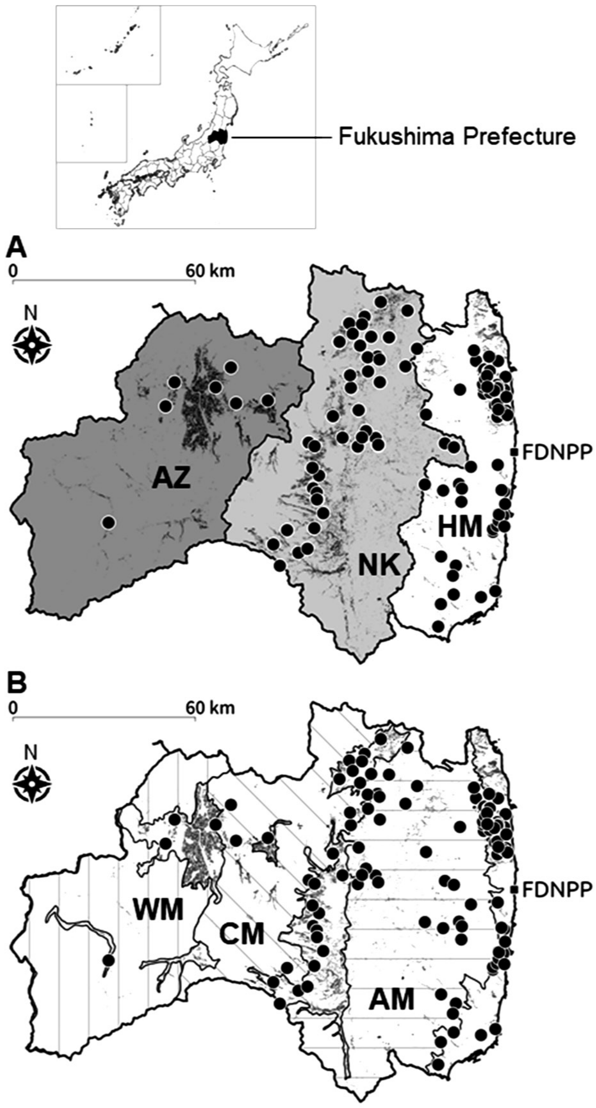
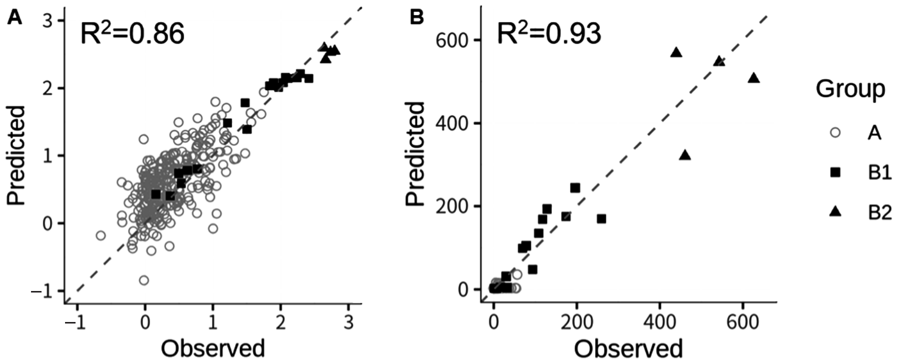


Fig. 1. Location of soil sampling points of the multipoint survey and field experiment data superimposed on districts and topographic classification in Fukushima Prefecture

Open circle: soil sampling points
Black filled area: agricultural fields [1]
A: Districts [2]
HM: Hamadori (Eastern coastal)
NK: Nakadori (Central)
AZ: Aizu
B: Topographic classification [3]
AM: Abukuma mountains
CM: Central mountains
WM: Western mountains
[1] *Hitsu*-polygon data, Ministry of Agriculture, Forestry and Fisheries (MAFF), Japan
[2] Digital National Land Information, Ministry of Land, Infrastructure, Transport and Tourism (MLIT), Japan
[3] Fundamental Land Classification Survey, MLIT, Japan

range of 10^{-1}-10^{1} Bq kg^{-1}, which is not very important when considering risk management. Using results from multipoint survey and field experiments (Fig. 1), described in detail by Yamamura et al. (2018) and Yagasaki et al. (2019a), respectively, this study proposes two alternative expressions for this model–a weighted linear regression model (Fig. 2A) and a nonlinear model (Fig. 2B). The influence of samples in a relatively low concentration range, which ranges from 0.1 to 10 Bq kg^{-1}, as discussed earlier, can be less in these two models



$\log A_{rice} = a \log A_{ex} + b \log K_{ex} + c$ $\quad$ $A_{rice} = A_{ex}{}^{\alpha} + K_{ex}{}^{\beta} + \gamma$

$a = 0.76,\ b = -1.71,\ c = 3.02$ $\quad$ $\alpha = 2.84,\ \beta, = 3.09,\ \gamma = 2.80$

A_{rice} : concentration of ^{137}Cs in rice grains (brown rice), in Bq kg^{-1}
A_{ex} : concentration of soil exchangeable ^{137}Cs in Bq kg^{-1}
K_{ex} : concentration of soil exchangeable K, in mg K kg^{-1}
$a, b, c, \alpha, \beta, \gamma$: regression coefficients

Fig. 2. Prediction of rice grain ^{137}Cs concentrations for data from a multipoint survey in Fukushima during 2012-2015 based on concentrations of soil exchangeable ^{137}Cs and K by using a multiple linear regression model modified from Yagasaki et al. (2019a) and an alternative nonlinear model

A (left): A multiple linear regression model, including logarithms with base 10 of rice grain ^{137}Cs concentration, soil exchangeable ^{137}Cs, and K concentrations, respectively, with weighting by $(\log A_{rice})^2$

B (right): A nonlinear regression model, taking exponentials of soil exchangeable ^{137}Cs and K concentrations

Group: Surveys and field experiments conducted in rice paddy fields in Fukushima Prefecture in 2012-2015 (A: Multipoint survey by MAFF, Japan. B1 and B2: Field experiment sites at the Fukushima Agricultural Technology Centre). Units for rice grain ^{137}Cs concentration, soil exchangeable ^{137}Cs, and K are Bq kg^{-1}, Bq kg^{-1}, and mg K kg^{-1}, respectively. The same dataset containing 308 observations was used for both models, A and B, and was obtained from surveys and field experiments conducted in Fukushima Prefecture in 2012-2015.

(Fig. 2A, Fig. 2B). Prediction by the nonlinear exponential model explained 93% of the variance in observation (Fig. 2B) and showed improved prediction for samples over 100 Bq kg^{-1} (Fig. 2B), which were underestimated when using the original model.

Model of the probability density of radio-cesium transfer for conservative risk assessment

The model by Yamamura et al. (2018) estimates the probability density of the transfer factor at given levels of soil exchangeable K, and not the individual transfer factor under various soil conditions. Compared with simple models based on the transfer factor, this model greatly reduces uncertainty in prediction by considering the effect of soil exchangeable K, but also maintains high usability with low data requirements by setting its purpose on risk assessment for a population of rice grains exceeding the intervention level. Additionally, the application of an inverse model can be adopted to estimate the required levels of soil exchangeable K at given total soil RCs to lower the intervention level in the case of high possible transfer in a population (e.g., region).

The model expresses the probability density of the transfer factor using a log-normal distribution function with empirical parameters, including the shape parameter and scaling factor (Table 1). The scaling factor is a function of soil exchangeable K with two empirical parameters b_0 and b_1 that account for the effect of regions and years and that of soil exchangeable K, respectively (Table 1). The following sections show in-depth model visualization and data analysis using the results from the multipoint survey and field experiments described earlier.

The cross-sections of the modeled probability of transfer factor at three different concentrations of soil

Table 1. Two models of radio-cesium transfer to rice grains with different soil parameters

	Model of probability density function for TF	Model of rice grain RCs concentration
Reference	Yamamura et al. (2018)	Yagasaki et al. (2019a, b)
Model description	A model of probability density function of TF, $L(x)$, based on a log-normal distribution with 1 soil variable, 2(3) model parameters α (b_0, b_1), and σ, obtained by maximum likelihood estimation; $L(x\mid\log_e(\alpha),\sigma)=\frac{1}{x\cdot\sqrt{2\pi\sigma^2}}\cdot\exp\left\{\frac{-(log_e(x/\alpha))^2}{2\sigma^2}\right\}$ where, L : probability density function of TF x : TF σ : shape parameter $\log_e(\alpha)$: scaling factor (a function of exK) $\log_e(\alpha) = b_0 + b_1 \log_e(K)$ where; b_0 : on effect of districts and years, with a set of discrete values. b_1 : on effect of soil exK. K : soil exK concentration (mg K kg^{-1}).	A model of rice grain (brown rice) RCs concentration in Bq kg^{-1}, $A(R, K)$, based on multiple linear regression with 2 soil variables including an interaction term; $\log_e A = a\cdot\log_e R + b\cdot\log_e K + c + d\cdot\log_e R\cdot\log_e K$ $A(R, K) = \exp(a\cdot\log_e R + b\cdot\log_e K + c + \mathrm{d}\cdot\log_e R\cdot\log_e K)$ where, A : rice grain RCs concentration (Bq kg^{-1}) R : soil exRCs concentration (Bq kg^{-1}) K : soil exK concentration (mg K kg^{-1}) a, b, c, d : coefficients

TF: transfer factor of RCs from soil to rice grain, exRCs: soil exchangeable RCs, exK: soil exchangeable K.

exchangeable K are shown by a contour plot using a set of parameter values they suggested. As the levels of soil exchangeable K decreased, the model predicted an increase in the probability of transfer factor with a range of relatively higher values (Fig. 3A). The plot shows a rise in the contour line and an increase in the 95th percentile of the model-estimated probability (upper edge of the gray-colored zone in Fig. 3B) as soil exchangeable K decreases. Another illustration (Fig. 4) indicated that the model-predicted probability density within a range of soil exchangeable K levels, expressed by a log-normal distribution, mimics these observations, especially the increase in the probability of the transfer factor with a higher value range. For practical model application, it may be desirable to use a safety factor, to be multiplied with the 95th percentile of the prediction, because using the 95th percentile alone could not cover some of the observed variability of the transfer factor that exceeded the predicted 95th percentile (Fig. 4).

Radio-cesium availability as a factor controlling probability density of transfer factor

Here, an assumption is made that the model by Yamamura et al. (2018) considers the soil RCs availability implicitly as a probability density. I investigated whether soil exchangeable RCs could explain changes in the transfer factors at a constant concentration of soil exchangeable K and soil RCs by data stratification (Fig. 5). For demonstration, the data presented earlier and stratified into eight different levels of soil exchangeable K (Fig. 4) were further stratified on the basis of eight different concentration ranges of soil RCs. From this stratified dataset, only subsets with > 15 samples were chosen and used for linear regression analysis of the transfer factor against soil exchangeable RCs. Consequently, only two subsets were obtained, as shown in Figure 5. In both subsets, statistically significant positive increases in the transfer factor were observed with increasing soil

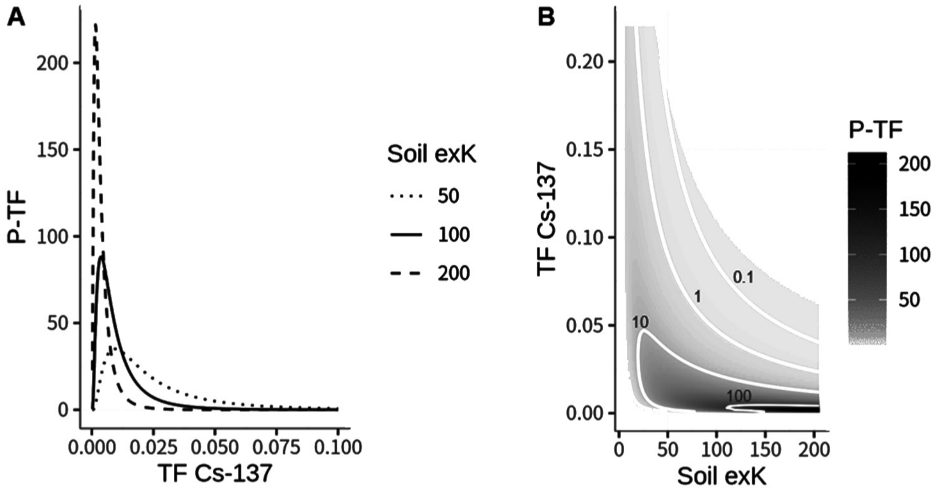


Fig. 3. An illustration of a model of probability density function for transfer factor of Cs-137 from soil to rice grain (brown rice) based on Yamamura et al. (2018)

A (left): Cross-sections of the modeled probability of transfer factor at three different concentrations of soil exchangeable potassium

B (right): Contour plot of the modeled probability of transfer factor. Only probability values greater than the 95th percentile are shown.

TF, transfer factor; **P-TF,** probability of TF; **exK,** soil exchangeable potassium (mg K kg^{-1}). Model parameters for the Hamadori district in 2012 proposed by Yamamura et al. (2018), which have the largest value of model scaling factor compared with other regions and years, were used.

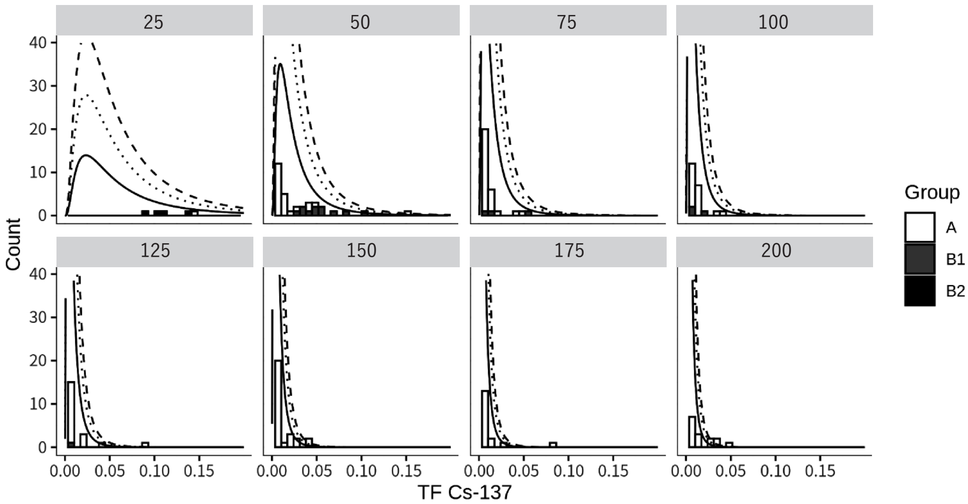


Fig. 4. Cross-section of the probability density function for the transfer factor of Cs-137 from soil to rice grain (brown rice), stratified at eight different levels of soil exchangeable potassium concentrations superimposed on the estimates by the model of Yamamura et al. (2018)

The figures on the top of each plot indicate the selected levels of soil exchangeable potassium concentration in mg K kg^{-1}. The lines in the figure indicate the 95th percentile of the model and its multiples (solid: 95th percentile; dotted and dashed: 95th percentile multiplied by a factor of 2 and 3, respectively).

TF: Transfer factor

Group: Surveys and field experiments conducted in rice paddy fields in Fukushima Prefecture during 2012-2018 (A: Multipoint survey by MAFF, Japan. B1 and B2: Field experiment sites at the Fukushima Agricultural Technology Centre). For details on these surveys and experiments, see the body text. Model parameters for the Hamadori district in 2012 proposed by Yamamura et al. (2018), which have the largest value of model scaling factor compared with other regions and years, were used.

exchangeable RCs ($P < 0.001$) (Fig. 5). Although only two conditions were examined in this study, the result suggests that soil exchangeable RCs are a factor affecting transfer factor variance. Second, the distribution patterns of soil exchangeable RCs at eight stratified levels of soil exchangeable K were investigated using histograms (Fig. 6). The histograms show that the distribution patterns of soil exchangeable RCs resemble a log-normal distribution (Fig. 6). In conclusion, these results suggest that the use of the log-normal distribution for the transfer factor probability density in their stratified levels of soil exchangeable K model is justified by the distribution of RCs availability.

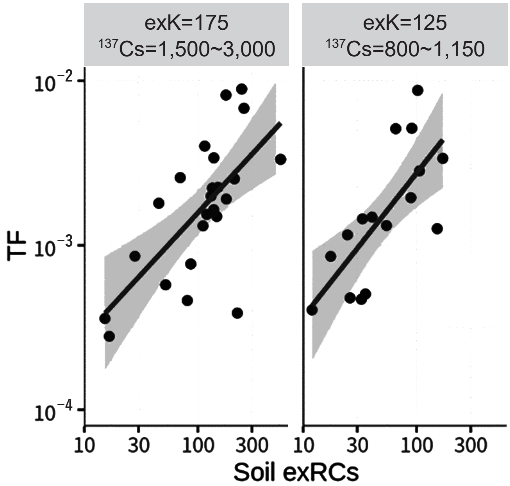


Fig. 5. Relationship between the transfer factor for rice grains and soil exchangeable ^{137}Cs concentration at two different concentration levels of soil exchangeable K and soil RCs

TF: transfer factor for ^{137}Cs from soil to rice grains in (Bq kg^{-1}) (Bq kg^{-1})$^{-1}$

Soil exRCs: soil exchangeable ^{137}Cs concentration in Bq kg^{-1}

exK: soil exchangeable K (mg K kg^{-1})

^{137}Cs: Soil ^{137}Cs concentration in Bq kg^{-1}

The values shown above the plots indicate the conditions on the stratified concentration ranges of soil exchangeable K and ^{137}Cs from which data were extracted. The gray solid lines and zones in the plots indicate the regression lines and confidence intervals of the linear regressions, respectively. Note that the vertical and horizontal axes used a logarithmic scale.

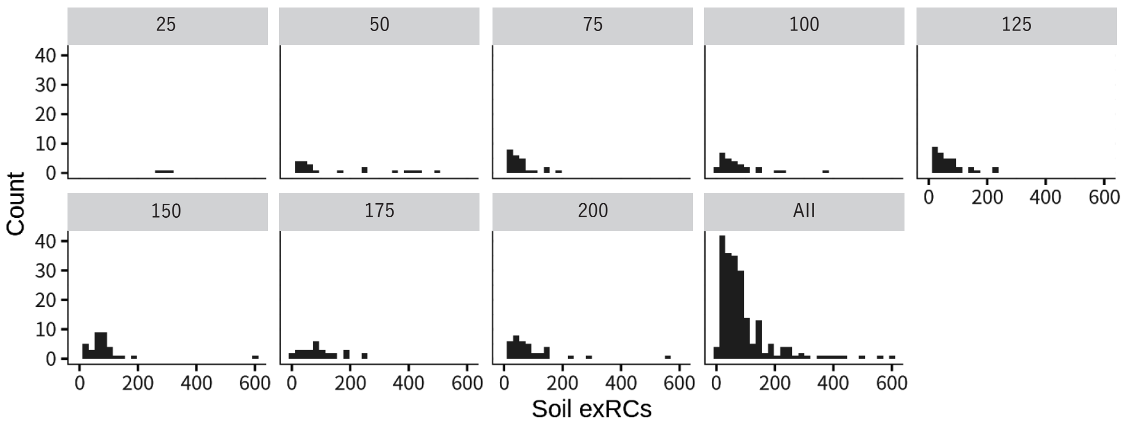


Fig. 6. Histograms of soil exchangeable ^{137}Cs concentration at eight different stratified concentration ranges of soil exchangeable K

Soil exRCs: soil exchangeable ^{137}Cs concentration in Bq kg^{-1}

Values shown above the plots indicate the concentration ranges of soil exchangeable K under the experimental conditions in mg K kg^{-1} from which data were extracted.

Implications for model applications at regional scale for multiple purposes and situations

The relevance between the models demonstrated in the previous section has significant implications for model applications, as described below. The prediction of the highest possible transfer factor by probability density can be regarded as a case of relatively high levels of soil exchangeable RCs in a region. Hence, the model application can be too rigid when applied to cultivation or shipping in subregions with a relatively low proportion of soil exchangeable RCs. Conversely, it can be too conservative when an inverse model is used to predict the required soil exchangeable K level to lower the intervention level. The model using soil exchangeable RCs accounts for the variation in RCs availability, and thus has the advantage of avoiding unnecessary conservative assessment, especially in regions with relatively high soil RCs concentrations and low availability. This model may not be necessary for all regions, especially for those with very low soil RCs concentrations, as data acquisition of soil exchangeable RCs is somewhat cost-intensive, and an alternate model of the probability density of transfer factor multiplied with soil RCs concentration can provide a quick and conservative assessment to ensure safety. Therefore, the study implications can be adapted to the design and accountability of comprehensive model applications for multiple purposes and situations. Further research is needed to compare the outputs of risk assessments by the two models using the same soils and regions. Additionally, it is desirable to elucidate the spatial variations of the soil exchangeable RCs concentration at both local and regional scales by expanding the multipoint survey and by geostatistical model prediction.

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