

## ARTICLE

# Time-Dependent Transfer of $^{54}\text{Mn}$ , $^{60}\text{Co}$ , $^{85}\text{Sr}$ and $^{137}\text{Cs}$ from a Sandy Soil to Soybean Plants

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Greenhouse experiments were performed to investigate the dependence of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  transfer from sandy soil to soybean plants on the growth stage when a radioactive deposition occurs. A solution containing  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  was applied onto the soil surfaces in the lysimeters at six different times – 2 d before sowing and 13, 40, 61, 82 and 96 d after sowing. Soil-to-plant transfer was quantified with a transfer factor ( $\text{m}^2 \text{kg}^{-1}$ -dry) specified for the deposition time. The transfer factor values of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  for the seeds were in the range of  $1.5 \times 10^{-3}$ – $1.0 \times 10^{-2}$ ,  $4.7 \times 10^{-4}$ – $3.2 \times 10^{-3}$ ,  $5.7 \times 10^{-4}$ – $1.0 \times 10^{-2}$  and  $3.0 \times 10^{-5}$ – $2.7 \times 10^{-4}$ , respectively, for different deposition times. The corresponding values for the leaves were  $6.4 \times 10^{-3}$ – $3.2 \times 10^{-2}$ ,  $4.3 \times 10^{-4}$ – $2.0 \times 10^{-3}$ ,  $5.1 \times 10^{-3}$ – $5.3 \times 10^{-2}$  and  $9.2 \times 10^{-5}$ – $1.9 \times 10^{-4}$ , respectively. The values for the seeds were on the whole highest following the middle-growth-stage deposition. After the pre-sowing deposition, the transfer factor values of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  for the seeds decreased annually so those in the fourth year were 53%, 75% and 34% of those in the first year, respectively. The present results may be useful for predicting the radionuclide concentrations in soybean plants due to their root uptake following an acute soil-deposition during the vegetation period, and for validating a relevant model.

**KEYWORDS:** *time, radionuclide, deposition, soybean, transfer factor, year*

## I. Introduction

Radionuclides can be released and deposited onto soils at any time of the year. The physiological activities of plants, the developmental stages of their organs and the availabilities of radionuclides for root uptake, change with time.<sup>1,2)</sup> In the case of an acute release during the vegetation period, therefore, soil-to-plant transfer of radionuclides may greatly depend on the time of their deposition.<sup>1,2)</sup> Accordingly, deposition time-dependent values of a soil-to-plant transfer factor would be useful for estimating the root uptake from an acute vegetation-period deposition.<sup>2,3)</sup>

On the other hand, a great portion of some radionuclides including  $^{137}\text{Cs}$  would remain in the root-zone soil for many years after their depositions.<sup>4-7)</sup> This means that a radioactive deposition onto soil can lead to a root uptake lasting for many consecutive years. For a more realistic assessment of the radiological impact of a soil contamination, therefore, it is necessary to understand a long-term soil-to-plant transfer of radionuclides.

Soybean is one of the staple-food crops for some Asian people, such as Koreans and Japanese. However, little is known about either the dependence of the soil-to-soybean transfer on the time of deposition or its dependence on the number of years after a deposition. In this study, therefore, greenhouse experiments were carried out to investigate both types of time-dependence of soil-to-soybean transfer of four important artificial radionuclides –  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  (substitute for  $^{90}\text{Sr}$ ) and  $^{137}\text{Cs}$ .

## II. Materials and Methods

Soybean plants were grown in lysimeters placed in a

greenhouse. The lysimeters, 0.6 m wide, 0.6 m long and 1.2 m high, were filled with soil from an abandoned field. **Table 1** summarizes physicochemical properties of the topsoil. Each lysimeter was supplied with chemical fertilizers for 1.5 g nitrogen, 1.1 g phosphorus and 1.8 g potassium, 54 g slaked lime and 360 g compost 1 d before sowing. A cultivar of soybean was sown on May 26 and several sprouts were thinned out to make a planting density of 12 plants per lysimeter.

A radioactive solution containing  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  (approximately 50 kBq  $\text{ml}^{-1}$  for each, carrier-free) was applied onto the soil surface at a dose of 64 ml per lysimeter at six different times – 2 d before sowing and 13, 40, 61, 82 and 96 d after sowing. A uniform contamination was made by dropping 1 ml of the solution to each of 64 points evenly distributed on the soil surface of each lysimeter. Following the pre-sowing application, the topsoil was mixed with the applied solution as a result of a simulated plowing. In farmlands, plowing is a normal practice done before sowing.

Plants were harvested on September 20, 117 d after sowing. Harvested plants were divided into four parts of seeds, shells, stems and leaves, and air-dried in the greenhouse for about one month. Dried plant samples were put into plastic containers for the measurements of radionuclide concentrations by gamma-spectrometry using a properly-calibrated HPGe detector (EG&G ORTEC).

**Table 1.** Physicochemical properties of the topsoil

pH (1:5)	OM (%)	CEC	EC ( $\text{cmol kg}^{-1}$ )		Sand (%)	Silt (%)	Clay (%)
			K	Ca			
5.2	0.91	4.2	0.12	1.84	79.0	17.0	4.0

OM: Organic matter, CEC: Cation exchange capacity ( $\text{cmol kg}^{-1}$ ), EC: Exchangeable cation.

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After the radionuclide concentrations in the different parts of the plants were measured, soil-to-plant transfer factors (TF,  $\text{m}^2 \text{kg}^{-1}\text{-dry}$ ) were determined as the ratio of the plant concentration ( $\text{Bq kg}^{-1}\text{-dry}$ ) at harvest to the soil-deposition density ( $\text{Bq m}^{-2}$ ) in the beginning. More details of the experimental can be found elsewhere.<sup>2,3)</sup>

### III. Results and Discussions

#### 1. Deposition-Time Dependent Transfer Factor

Figures 1 and 2 show the variations of soil-to-soybean TF values of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  with the times of their deposition during the vegetation period. Fig. 1 is about the reproductive parts (seeds and shells) and Fig. 2 is about the vegetative parts (stems and leaves).

In general, the variations with the deposition times were by a factor of up to 10. The TF values of the radionuclides for the seeds and the shells were on the whole highest at a middle-growth-stage deposition. It was generally true of the stems if marked rises for  $^{54}\text{Mn}$  and  $^{60}\text{Co}$  at the latest deposition are disregarded. In the leaves, only  $^{54}\text{Mn}$  had the highest value at a middle-growth-stage deposition. The TF value of  $^{85}\text{Sr}$  for the leaves decreased as the deposition time got closer to harvest, whereas that of  $^{137}\text{Cs}$  was rather constant regardless of the deposition times. Rise in the TF value at the latest deposition was most significant in the stems, whereas there was no significant rise in the seeds. This can be attributed to the fact that translocation of the absorbed radionuclides to the seeds decreases near maturity.

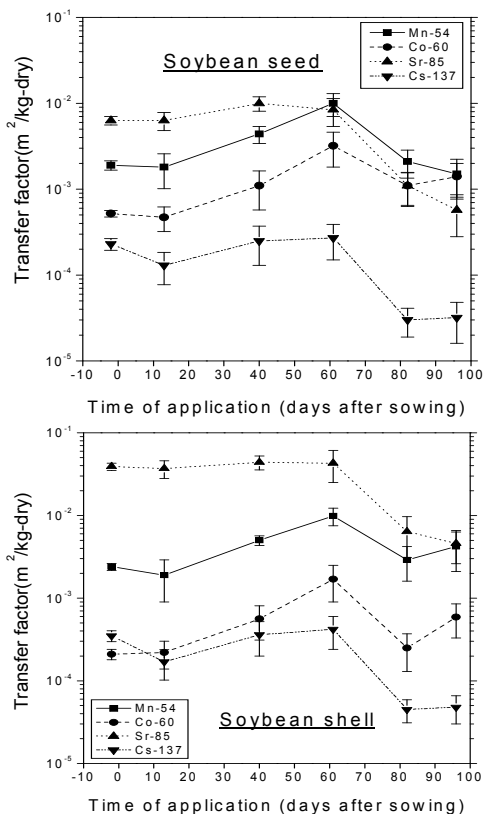


Fig. 1. Soil-to-plant transfer factors of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  for the reproductive parts of soybean plants varying with the times of radioactive deposition.

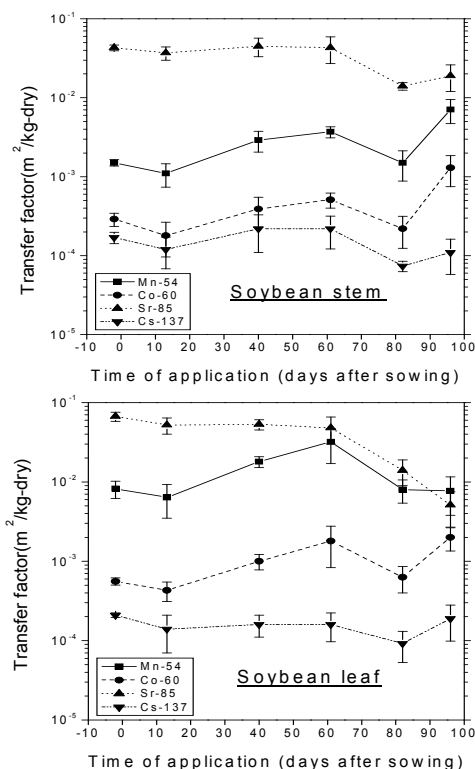


Fig. 2. Soil-to-plant transfer factors of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  for the vegetative parts of soybean plants varying with the times of radioactive deposition.

The TF values from the pre-sowing deposition were on the whole higher than those from the earliest post-sowing deposition by a factor of up to 2. This may indicate that the soil mixing following the pre-sowing deposition increased the root uptake of the radionuclides. Such an effect was most significant for  $^{137}\text{Cs}$  and least significant for  $^{85}\text{Sr}$ .

According to generally-adopted methods<sup>8,9)</sup> of estimating the soil-to-plant transfer of radionuclides deposited acutely during the vegetation period, a higher transfer would be predicted from an earlier deposition.<sup>2,3)</sup> A great part of the present results do not agree with this estimation. Therefore, the use of such empirical data as presented here may be helpful for a more realistic estimation.

In most cases, the TF value was highest for  $^{85}\text{Sr}$ , followed by  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  in turn. The TF of  $^{85}\text{Sr}$  was 1-2 orders of a magnitude higher than that of  $^{137}\text{Cs}$ . Similar trends have been reported for various types of plants.<sup>2,3,10,11)</sup> Differences between  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  TF values were smaller in the seeds than in the other parts. This may reflect that the mobility of radiocesium in the plant body is much higher than that of radiostromium.<sup>12-14)</sup> The difference in the mobility between  $^{137}\text{Cs}$  and  $^{85}\text{Sr}$  is clearly demonstrated by Fig. 3 which shows that  $^{137}\text{Cs}$  was distributed in the seeds by about 3.5 times higher percentage than  $^{85}\text{Sr}$  did.

For  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$  and  $^{85}\text{Sr}$ , the TF values for different plant parts decreased on the whole in the order of leaf > shell > seed > stem, seed > leaf > shell > stem and leaf  $\geq$  stem > shell > seed, respectively. In an experiment for kidney bean by D'Souza and Mistry,<sup>15)</sup> the  $^{58}\text{Co}$  concentration was also

higher in the seeds than in the other parts. The order for  $^{85}\text{Sr}$  reflects again its low mobility within the plant body. At the latest deposition, however, the orders for  $^{54}\text{Mn}$  and  $^{60}\text{Co}$  changed into leaf  $\geq$  stem  $>$  shell  $>$  seed and leaf  $>$  seed  $\geq$  stem  $>$  shell, respectively, because of the above-mentioned rises in the TF values. The decreasing order for the TF value of  $^{137}\text{Cs}$  was shell  $>$  seed  $>$  leaf  $\geq$  stem for the earliest four depositions, but leaf  $>$  stem  $>$  shell  $>$  seed for the other two depositions. This may support that root uptake until a middle-growth stage and subsequent translocation contribute effectively to the transfer of  $^{137}\text{Cs}$  from soil to the seeds.

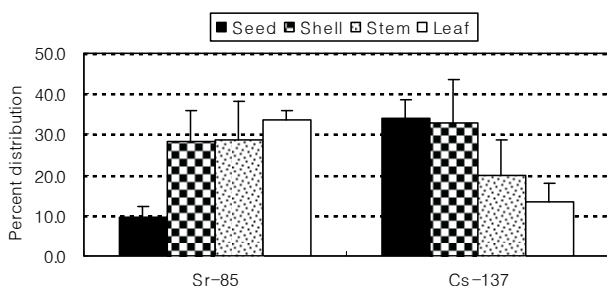


Fig. 3. Distributions of  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  among different parts of mature soybean plants.

## 2. Dependence on the Number of Years after Deposition

Fig. 4 shows the annual changes in the TF values of  $^{60}\text{Co}$ ,  $^{54}\text{Mn}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  for the seeds following the pre-sowing deposition. For  $^{85}\text{Sr}$ , TF values beyond the second year could not be acquired because of its relatively short half life.

The TF value of  $^{137}\text{Cs}$  decreased annually so the fourth-year value was lower than the first-year value by a factor of 3. An additional decrease could be expected for the fifth year and beyond. For  $^{54}\text{Mn}$ , an annual decrease occurred only until the third year in which the TF value was lower than the first-year value by a factor of 2. There was no significant annual decrease in the TF value of  $^{60}\text{Co}$  and the fourth-year value was only 25% lower than the first-year value. No decrease was observed for  $^{85}\text{Sr}$ .

Such annual decreases as mentioned above can be mainly attributed to the irreversible fixation of radionuclides by soil matrices like clay minerals.<sup>16-18)</sup> It is well known that radiocesium is readily fixed in soil.<sup>3,16-19)</sup> In lysimeter experiments for several crop species, Noordijk *et al.*<sup>18)</sup> found that the transfer of  $^{137}\text{Cs}$  to the edible parts decreased on an average by a factor of 1.5 in 1-2 years and a factor of 4 in seven years. They found no change in the transfer of  $^{90}\text{Sr}$ .

The present results for  $^{137}\text{Cs}$  and  $^{85}\text{Sr}$  are in general agreement with the findings by Noordijk *et al.*<sup>18)</sup> For  $^{60}\text{Co}$ , however, the observed decrease was much smaller than the decrease found in wild grasses by Boikat *et al.*<sup>16)</sup> The effect of ageing would depend on the soil type and the plant species.<sup>18)</sup> According to Choi *et al.*,<sup>3)</sup> Chinese cabbage and radish showed more significant decreases in the second year than the corresponding present results. On the other hand, downward migration of radionuclides may be another factor for annual decreases in the root uptake.<sup>20,21)</sup>

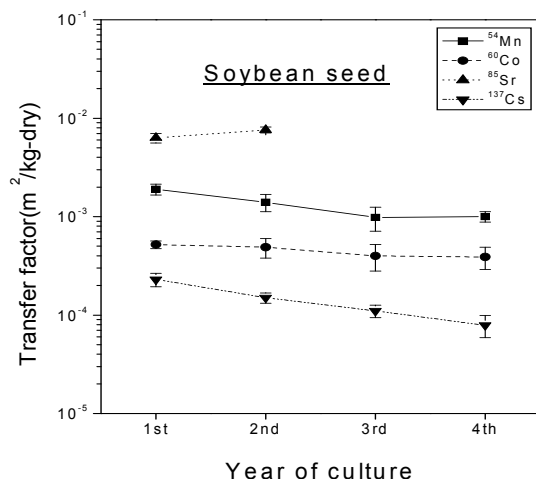


Fig. 4. Annual changes in the TF values of the radionuclides for the soybean seeds after the pre-sowing deposition in the first year

## IV. Conclusion

Two kinds of time-dependence were investigated for the transfer of  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{85}\text{Sr}$  and  $^{137}\text{Cs}$  from a sandy soil to soybean plants. One is the dependence on the time of deposition during the vegetation period, and the other is the dependence on the number of years after deposition.

Soil-to-plant transfers of the radionuclides varied considerably with the times of deposition. In most cases, the observed trends for deposition time-dependent transfer do not agree with the expectation from generally-adopted methods. This supports that the acquired experimental data may be useful for a more realistic prediction of the radionuclide concentrations in soybean following their acute deposition during the vegetation period.

The dependence of the TF value on the number of years after deposition was characterized by its annual decrease except for  $^{85}\text{Sr}$ . Annual decrease in the transfer was most significant for  $^{137}\text{Cs}$ , followed by  $^{54}\text{Mn}$ . In a long-term prediction of the root uptake, such annual decreases should be considered to avoid an unnecessary overestimation.

Present data are acquired from a single experiment and a prediction using them may be accompanied by a high uncertainty. Further experiments need to be carried out to sufficiently cover various environmental conditions to help reduce the uncertainty in predicting the root uptake following a vegetation-period deposition.

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