Uptake of Radioactive Strontium and Cesium in Rice Plants

(1) Accumulation of Sr and Cs in Rice Grains through Roots

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ABSTRACT

Water-cultured rice plants were exposed to $^{89}\text{Sr}$ or $^{137}\text{Cs}$ through roots for five days at their various growth stages and continued to grow until harvest. The harvested grains were radiochemically analysed and the concentration factors were calculated.

The maximum uptake of $^{137}\text{Cs}$ in the grains was found at the booting stage, while that of $^{89}\text{Sr}$ was at the flowering stage. The Cs uptake was 400 times higher at the booting stage, and 30 times higher at the flowering stage than those with Sr.

The growth stage dependency of the uptake of Sr and Cs was the most important factor for a selective enrichment of Cs in rice grains.

The specific affinity of Cs to cell sap and that of Sr for membrane substances of rice grains probably caused a selective redistribution inside the plant body.

INTRODUCTION

From standpoint of the environmental contamination with fission products released from nuclear detonation or industrial accidents in nuclear reactor operation, radio-contamination of rice with Sr and Cs is very important problem. The reasons are that rice is a staple food in Japan, and that both radioactive Sr and Cs have long lives and a high availability to crops.

During a national radio-contamination survey for $^{90}\text{Sr}$ and $^{137}\text{Cs}$ in soils and rice, which has been conducted in the National Institute of Agricultural Sciences* in co-operation with the Institute of Public Health, it has been pointed out that the

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content of $^{137}$Cs in polished rice is 10 times or more higher than that of $^{90}$Sr\(^\text{(1-4)}\). For instance, 50 pCi of $^{137}$Cs per kg and 4 pCi of $^{90}$Sr per kg were observed for the polished rice samples collected in 1962, in spite of the ratio of $^{137}$Cs to $^{90}$Sr being reported as 2.5:1 in the original fall-out itself\(^\text{(5)}\).

The mechanisms, governing such selectivity for Cs and Sr in the course of migration of these elements from fall-out to polished rice, should be studied as one of the most important problems of the contamination in food chain.

It is well known that there are four pathways in grain contamination: (a) absorption from soil through root, (b) plant-base absorption, (c) foliar absorption, and (d) floral absorption\(^\text{(6,7)}\). The former is an "indirect contamination" and the latter three are "direct contamination".

In this paper, root absorption of $^{89}$Sr and $^{137}$Cs by water-cultured rice plant was dealt with. For the sake of comparison, $^{32}$P was also applied to the plants, because phosphorus is one of the indispensable elements for plants and its behaviour has been well studied.

\section*{MATERIALS AND METHODS}

Forty-four days-old rice seedlings were transferred to Wagner pot (1/5000 are) on June 24, 1963, and were grown on water-culture. The composition of the nutrient solution is shown in Table 1. Tap water containing 33 ppm of Ca was used in this water-culture. The culture solution was adjusted to pH 5.5 with HCl or NaOH, and was renewed every 5 days.

The plants were treated with radioisotopes in a green house as schematically shown in Fig. 1. At each stage of the plant growth, $^{32}$P was singly applied into a pot, whereas $^{89}$Sr and $^{137}$Cs were simultaneously added to a pot, with nutrient sol-

\begin{table}[h]
\centering
\caption{Composition of the nutrient solution}
\begin{tabular}{|c|c|c|c|c|c|c|}
\hline
Elements & N & P & K & Mg & Fe & Mn \\
\hline
Concentration & 20 ppm N & 10 ppm P$_2$O$_5$ & 20 ppm K$_2$O & 10 ppm MgO & 3 ppm Fe$_2$O$_3$ & 0.5 ppm MnO \\
Chemicals & NH$_3$NO$_3$ & KH$_2$PO$_4$ & KH$_2$PO$_4$ & K$_2$SO$_4$ & MgCl$_2$·6 H$_2$O & FeCl$_3$·6 H$_2$O·3 H$_2$O & MnCl$_2$·4 H$_2$O \\
\hline
\end{tabular}
\end{table}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Experimental design of isotope treatments}
\end{figure}
ution in all cases. After isotope treatments the plants were once removed from pot, and the roots were rinsed three times with tap water. And then the plants were transferred to another pot to be continued to grow under ordinary water-culture conditions until harvest.

Completely matured rice grains (rough rice, 34 g per pot in average) were dried at room temperature for about 10 days and were then husked. The brown rice was polished at the 90% milling rate. Two grams of the polished rice and brown rice were ashed respectively, in an electric muffle furnace by raising the temperature gradually up to 450°C and kept at the same temperature for 3 hrs.

$^{89}$Sr was determined on the basis of the method of Kodaira$^{8}$ and $^{137}$Cs analysis was made by the method recommended in the text book$^{9}$ (refer to another paper too$^{10}$). The procedure is briefly outlined in the appendix. According to this procedure, neither cross contamination possibly arising from $^{89}$Sr and $^{137}$Cs nor interference by the constituents of rice grain was observed, as examined respectively in the previous paper$^{10}$ and in the appendix of this paper. $^{32}$P was precipitated as ammonium phosphomolybdate by usual analytical method. $\beta$-radioactivities of $^{32}$P and $^{137}$Cs were measured by a conventional GM method, whereas for $^{89}$Sr a 4 $\pi$ gas flow low background counter was used.

RESULTS

The data were expressed in terms of concentration factor (C. F.) in rice grain as defined as follows:

$$C.\text{ F.} = \frac{\text{Radioactivity (cpm) of the contaminant}}{\text{Radioactivity (cpm) per 1 g of the initial nutrient solution}} \times \frac{\text{accumulated into 1 g of rice grain}}{\text{per 1 g of the}}$$

Table 2. Concentration factors (C. F.) for $^{32}$P in rice grain

| Treatments  | P C. F. | B C. F. | F C. F. | M C. F. |
|-------------|---------|---------|---------|---------|
| Brown rice  | 73 24   | 194 31  | 204 32  | 137 27  |
| Polished rice| 43 14   | 114 18  | 120 19  | 100 16  |
| Bran        | 340 110 | 910 144 | 880 137 | 830 130 |

Table 3. Concentration factors (C. F.) for $^{137}$Cs in rice grain

| Treatments  | P C. F. | B C. F. | F C. F. | M C. F. |
|-------------|---------|---------|---------|---------|
| Brown rice  | 1,649 5.8 | 2,478 9.9 | 1,604 5.8 | 365 1.32 |
| Polished rice| 727 2.6  | 1,069 3.9 | 770 2.8  | 155 0.56 |
| Bran        | 9,950 36 | 15,160 55 | 9,110 33 | 2,250 8.1 |

Table 2. Concentration factors (C. F.) for $^{32}$P in rice grain

Table 3. Concentration factors (C. F.) for $^{137}$Cs in rice grain

note: Isotope solution; 3.1 cpm/ml for treatment P, 6.3 cpm/ml for treatments B, F & M.
* See Fig. 1.

Table 2. Concentration factors (C. F.) for $^{32}$P in rice grain

Table 3. Concentration factors (C. F.) for $^{137}$Cs in rice grain

note: Isotope solution; 277 cpm/ml.
* See Fig. 1.
For $^{32}$P, as shown in Table 2 and Fig. 2, the concentration factors in both brown rice and polished rice were fairly constant, and kept higher levels than those for $^{137}$Cs and $^{89}$Sr, throughout all growth stages. This fact indicates a very high mobility in plant body for phosphorus.
For $^{137}$Cs, as shown in Table 3 and Fig. 2, the concentration factors obtained from the treatments P, B and F, except for the M, showed almost the similar values, although there was a gentle peak at the treatment B, and kept extremely high contamination levels in comparison with $^{89}$Sr.

For $^{89}$Sr, as shown in Table 4 and Fig. 2, the concentration factors showed a sharp peak at the treatment F.

DISCUSSION

1. Similarities of Sr to Ca and Cs to K

The problem on the similarity and the discrimination of Cs vs. K in soils and plant body does not seem to be so clear, in spite of many efforts\(^5\)\(^6\)\(^11\)\(^-\)\(^18\) compared with the relationship between Sr and Ca. Tensho et al\(^17\) discussed on the selectivity of Cs vs. K, and obtained 0.03 as an observed ratio for a system of rice plant and soil under a flooded condition. Among the indispensable nutrients, although the discrimination is significant, K is still only one element which has a relatively high similarity to Cs in the behaviour inside the plant body.

Numerous studies on the relationship between Sr and Ca in soils and plants have been reported\(^5\)\(^6\)\(^13\)\(^19\)\(^-\)\(^24\). In most of these studies, a very high similarity of Sr to Ca has been revealed.

2. Growth stage dependency of the uptake of Sr and Cs

Generally in plant body, the most metabolically active parts are young organs at each growth stage. Therefore the leaves developed in early growth stage accumulate various ions very actively. Meanwhile when they become old, other organs emerge and develop to take the place of the most active parts. Consequently, with the progress of plant growth, the ions accumulated previously in the leaves in early period are redistributed more or less to newly developed organs, finally to grains. Whereas the ions absorbed in late period can be translocated rather directly to grains.

According to Takahashi\(^25\), K is mainly accumulated by rice plant in early growth stage, in contrast with the accumulation of Ca which shows a maximum just before flowering stage.

In this experiment as shown in Fig. 2, the maximum accumulation of Cs in the grains was resulted from the treatment B (booting stage), while that of Sr was from the treatment F (flowering stage). These patterns of ion uptake were in good accordance with those which was expected from Takahashi's pointing out, taking account of the similarities of Cs to K and Sr to Ca.

It seemed that the plant absorbed a large quantity of Cs in early period and then redistributed it into rice grain, but could not absorb it so much in late period due to the decrease in its absorption power. On the contrary the same plant seemed to absorb only a small amount of Sr in early period because of its poor absorption power; in addition, the redistribution might be difficult by the cause of the affinity of Sr for membrane substances as discussed later. But in late period
the plant seemed to absorb a considerably large amount of Sr together with Ca, and then translocated it into rice grain rather directly, not necessary through the redistribution function.

To compare the degree of accumulation of Cs and Sr in rice grains, the ratio of the concentration factors for these elements was calculated and shown in Table 5. The ratio of 400:1 for brown rice was resulted from the treatment B. The ratio of 30:1 was resulted from the treatment F, by which a maximum accumulation of Sr took place. This experimental fact can be a strong evidence for the explanation of the predominance of $^{137}$Cs to $^{90}$Sr which has been observed in results of the routine work of radio-contamination survey.

3. Distribution of Sr and Cs in rice grain

According to Ozaki\textsuperscript{26}, polished rice contains 0.13\% of P, 0.12\% of K and 0.014\% of Ca. At present time the value of Ca should be written as 0.006\% according to a recent work\textsuperscript{27}. In any way the content of K is about 10 to 20 times higher than that of Ca in polished rice.

If discriminating function of Cs vs. K and Sr vs. Ca is neglected, in other words, if Cs and Sr behave quite equally to K and Ca respectively, ten to twenty-fold accumulation of Cs against Sr can be expected. This idea also can be one of the interpretations for the predominance of Cs to Sr in polished rice.

On the other hand, as shown in Table 6, the concentration factors were compared in respect of brown rice and polished rice. The ratios were fairly specific to each element (2:1 for Cs and 3:1 for Sr), regardless with the period of treatment.

| Treatments* | P   | B   | F   | M   | Average |
|-------------|-----|-----|-----|-----|---------|
| C. F. in brown rice | Cs  | 2.2:1 | 2.3:1 | 2.1:1 | 2.3:1 | 2.2:1 |
| C. F. in polished rice | Sr  | 3.3:1 | 3.1:1 | 2.7:1 | 2.9:1 | 3.0:1 |

* See Fig. 1.

| Treatments* | P   | B   | F   | M   | Average |
|-------------|-----|-----|-----|-----|---------|
| cpm in bran | Cs  | 60:40 | 61:39 | 67:33 | 62:38 | 60:40 |
| cpm in polished rice | Sr  | 73:27 | 71:29 | 68:32 | 69:31 | 70:30 |

* See Fig. 1.
This fact may be due to the constancy of chemical composition of rice grain. Brown rice is usually divided into polished rice and bran. The distribution rates of Cs and Sr between these two parts are shown in Table 7. 40% of Cs and 30% of Sr entered polished rice respectively, the remainder contaminated only bran.* This fact may be due to the difference in the constituent materials between polished rice and bran. Namely polished rice is essentially consisted of starch, whereas bran is a sort of degenerated endocarps and endoderms. Cs might behave together with K in cell sap and be incorporated into sugar substances in early period of starch formation. Meanwhile Sr might be caught together with Ca by membrane substances of bran fraction. The affinity of Sr for membrane substances probably gives rise to inhibit the redistribution into other parts in early period of growth, but to accelerate the incorporation into bran in late period.

4. Mechanism of contamination through roots

The pathways of $^{137}$Cs and $^{90}$Sr from soil to rice grain through roots can be outlined in Table 8.

Fall-out materials containing about 2.5 of $^{137}$Cs and 1 of $^{90}$Sr are accumulated into soil (refer to Kodaira’s previous paper). Fixation onto soil takes place especially significantly for $^{137}$Cs. The ratio of $^{137}$Cs to $^{90}$Sr present in soil as an exchangeable form (plant available form) becomes different from that of original fall-out materials. In fact Japanese soils collected in 1964 were observed to contain 147 pCi $^{137}$Cs/kg and 320 pCi $^{90}$Sr/kg, the ratio being 1: 2.*

As pointed out by Tensho et al, if the soil is flooded for the cultivation of lowland rice, a part of $^{137}$Cs can be liberated through a replacement action of NH$_4^+$ ion formed under an anaerobic condition. Also as reported by Nishigaki et al, the physiological activity of rice root is highly dependent to soil conditions and plant growth, for instance the oxidation-reduction level of the soil, and the distribution of underground root system in both longitudinal and horizontal directions.

In addition to these processes mainly induced by soil conditions, the aforementioned plant nutritional processes accelerate very much a selective enrichment of $^{137}$Cs in rice grains.

### Table 8. Outline of the pathways of Cs and Sr from soil to rice plants

| Environments | Soil | Rice plants |
|--------------|------|-------------|
| Conditions   | Fixation | Early stage | Late stage |
| Functions    | N. F.* | Accum.* | Redistr.* | Accum.* | Redistr.* |
| Contaminants | Cs high | low | high | medium | high | medium | high | low | medium | low |
|              | Sr low | high | medium | high | low | medium | low |

*N. F. = Not flooded, F = Flooded, Accum = Accumulation, Redistr = Redistribution.

* In this experiment the weight of bran was 10% of that of brown rice. The extent of contamination per unit weight of bran was very high.

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Appendix

Outline of radiochemical separation of $^{89}$Sr and $^{137}$Cs in plant ash

The ashed samples were extracted with 0.8 N HCl, to which 15 mg of Ca was added as a co-precipitating reagent for $^{89}$Sr, and evaporated to dryness. After removal of silica, 0.1 g of ammonium phosphomolybdate, purified in the authors laboratory, was added to the extract as a collector of $^{137}$Cs under about 0.8 N acidity of HCl, and kept standing overnight. The precipitate was filtered, washed with a diluted acid solution, and finally with ethyl alcohol to prevent creeping. The remaining filtrate was concentrated to a volume of about 50 ml. From this solution, $^{89}$Sr was co-precipitated together with calcium oxalate.

The reliability of this method was examined as seen in the table below.

| Components          | Initial acidity, HCl | 0.36 N | 1.2 N |
|---------------------|----------------------|--------|-------|
| mg in 50 ml of final solution | Ca$^{2+}$ | - | 15 | 15 |
|                     | K$^+$ | - | 50 | 50 |
|                     | Mg$^{2+}$ | - | 20 | 20 |
|                     | PO$_4$$^{3-}$ | - | 50 | 50 |
| Ash obtained from 10 g of brown rice | - | - | - | added |
| Yield of Sr, % ($\leq \pm 4\%$ error) | 100* | 98 | 94 | 96 |

*Taken as 100.