# TRANSFER FACTOR OF RADIUM-226 FROM SOIL TO RICE

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## ABSTRACT

In the environment, Ra occurs naturally and is distributed widely in environmental materials, such as rock, soil, water and plants; however, its concentration is extremely low. Thus, soil-to-plant transfer factor (TF) data obtained under natural conditions are limited. In this study, we determine the concentrations of Ra-226 in brown rice and associated paddy soils collected from 11 locations throughout Japan to obtain TFs. The radioactivity was measured with a Ge detector system for soil samples and a liquid scintillation counting system for brown rice after radiochemical separation. The recoveries of Ba, used as a chemical yield tracer for Ra, were more than 97% for all the samples. The concentrations of Ra-226 in paddy soil samples ranged from 20 to 65 Bq/kg–dry soil, and those in the brown rice samples ranged from 8 to 65 mBq/kg-dry except for two samples (<DL). Using the data, TFs were calculated and the average TF of Ra-226 from the paddy soil to the rice (brown rice) was 6.8E-4. This value was close to the TFs for maize reported in the IAEA Technical Reports Series No. 364.

## INTRODUCTION

Radium-226 and 228 are in the U and Th decay series, respectively, and these naturally occurring radium isotopes are widely distributed in environmental materials, such as in rock, soil and plant materials. Ra-226, an alpha emitter with a half-life of about 1600 y, is of special interest because it is one of the important radionuclides for the assessment of radioactive waste disposal. Once Ra is taken into the human body by ingestion of food and water or inhalation, it can distribute on bone so it has a long biological half-life; exposure to Ra can cause cancers and other body disorders. Therefore its long-term management is required and understanding of Ra behavior in the environment is important.

For a precise long-term radiological assessment of radionuclides, it is necessary to obtain the local transfer parameters and their variations which are used in mathematical models. Among the parameters used in these models, the soil-to-crop transfer factor (TF) is a key parameter that directly affects the internal dose assessment for the ingestion pathway [1]. With respect to the long-term behavior, measurement of Ra-226 concentration in compartments of the environment gives us useful information for this purpose. However, because Ra-226 concentrations in many environmental materials are extremely low, it is difficult to measure Ra-226 without chemical separations and concentrations from sample matrices, especially for plant samples. Subsequently, soil-to-plant TF data are limited.

In this study, in order to obtain TFs of Ra under natural environmental conditions, we determine the concentrations of Ra-226 in rice grains and associated paddy soils collected throughout Japan using radiation-counting methods. We focused on rice because the consumption of cereals is very high in Japan and other Asian countries.

## MATERIALS AND METHODS

### Samples

The paddy soils were collected from 11 locations throughout Japan, and the rice plants grown on these soils were also collected in the harvesting season. Rice varieties grown traditionally in Japan are classified as short-grain types. Radium-226 concentrations were measured for the soil samples, which were air-dried and sieved with a 2-mm sieve, and for rice samples, which were husked to obtain brown rice (with bran) and they were incinerated at 450˚C for 24 h. For comparison, concentrations of four stable elements, Ca, Sr, Ba and Cs were measured by inductively coupled plasma mass spectrometry (ICP-MS) using a part of each soil and brown rice sample. We have reported method details elsewhere [2].

### Chemical Separation

A chemical separation for Ra-226 was carried out using about 15 g of each incinerated rice sample. Use of 5 g of incinerated rice has been recommended for the standard method used in Japan [3]. However, to obtain a lower level for the detection limit (DL), we tripled the sample amount in the present study. The radiochemical separation steps were the same as used in the standard method [3]. Since there is no stable Ra isotope, the chemical yield of Ra was obtained using barium (Ba), which is just above Ra in the periodic table. This selection was based on their similar chemical behaviors. To check the accuracy of the method, we used two reference materials, IAEA-315 (marine sediment) and IAEA-368 (Pacific Ocean sediment). There is no standard reference material available for plant samples.

### Radioactivity Measurement

After the radiochemical separation from the rice samples, the samples were kept for two weeks to reach activity equilibrium with Rn-222, then, Ra-226 was removed. The activity of Rn-222 was measured with a liquid scintillation counter (Aloka, LB-2 or LB-5) and from the counts, Ra-226 was calculated.

For the soil samples, about 60 g of each pretreated soil sample were charged and sealed into a plastic vessel. The sample was kept at room temperature for more than 30 days to reach an equilibrium condition with its progeny nuclides. Then, the concentration of Pb-214 was measured with a Ge detector system (Seiko EG&G Ortec) from which Ra-226 could be calculated.

## RESULTS AND DISCUSSION

### Radium-226 Concentrations in Brown Rice and Associated Paddy Soils in Japan

The concentrations of Ra-226 in the rice samples and the soils are listed in Table I. Radium-226 concentrations in the soils ranged from 20 to 65 Bq/kg-dry with an average of 35 Bq/kg-dry. It is difficult to compare the concentration differences among soil types and places because only 11 samples were measured in the present study. More data are needed for detailed discussions. For brown rice samples, the concentrations ranged from <0.007 to 0.065 Bq/kg-dry; these values were much lower than those in the soils. Since Ra-226 concentrations are extremely low in plant samples in the natural environment, we strongly recommend radiochemistry procedures be done to separate and concentrate Ra-226 from the plant matrix.

**Table I. Concentrations of Ra-226 in brown rice and associated paddy soils collected in Japan, 2002.**

| Sampling sites | Soil type | Radium-226 (Bq/kg-dry) | |
|---|---|---|---|
| | | Soil | Brown rice |
| Hokkaido | Gray lowland soils | 34.7±1.0 | <0.010 |
| Iwate | Wet andosols | 24.9±1.1 | 0.012±0.002 |
| Fukushima | Wet andosols | 33.7±0.9 | 0.015±0.002 |
| Miyagi | Gray lowland soils | 19.9±0.9 | <0.007 |
| Chiba | Gley lowland soils | 30.9±0.9 | 0.008±0.002 |
| Ibaraki | Brown lowland soils | 24.0±0.9 | 0.008±0.002 |
| Toyama | Gray upland soils | 40.8±1.0 | 0.065±0.003 |
| Hyogo | Gray lowland soils | 31.9±0.9 | 0.025±0.003 |
| Mie | Gray lowland soils | 46.0±1.0 | 0.030±0.002 |
| Hiroshima | Gley lowland soil | 64.9±1.1 | 0.055±0.003 |
| Fukuoka | Gray lowland soil | 37.1±0.9 | 0.027±0.003 |

±: Counting error (1 s.d.)

In the radiochemistry procedures for Ra-226 measurement, Ba was used as a yield tracer of Ra, and the recovery throughout the separation procedure were more than 97% for all the samples. The DL was lowered since a 15-g ash sample was used; that is, the DL obtained was about 0.7 Bq/kg-ash. The ash-to-dry weight ratios ranged from 0.011 to 0.016 so that the minimum DL for dried brown rice was 0.007 Bq/kg-dry. If 5-g ash samples were used as recommended in the standard method [3], the DL would be 2 Bq/kg-ash. Unfortunately, even though we lowered the DL, Ra-226 concentrations for two brown rice samples could not be measured in the present study. Further study is needed to measure ultra trace levels of Ra-226 in plant samples.

**Transfer Factors of Ra-226, Ca, Sr, Ba, and Cs**

Using the obtained data for Ra-226 in the brown rice and soil samples, we calculated the TFs. The TF can be quantified as:

$$TF = \frac{\text{Concentration of the element in the edible parts at harvest (Bq/kg-dry or g/kg-dry)}}{\text{Concentration of the element in the soil (Bq/kg-dry or g/kg-dry)}}$$

The TFs of Ra-226 (TF-Ra) ranged from 0.0003 to 0.0016 with an average of 0.00068. This value was close to the TFs for maize (0.00024-0.006 with an average of 0.0012) reported in the IAEA TRS 364 [1]. Only two TF data are available for brown rice collected in Japan [4], and their average TF was 0.0047, which is one order of magnitude higher than the present results. The sample number of the previous report should be responsible for this difference.

Since the TFs of Ra are limited, we measured TFs of Ca, Sr and Ba which are in the same alkaline earth metal group. By comparison we could clarify the distribution similarity between Ra and these elements. We also obtained TFs of stable Cs; Cs-137, a long-lived radionuclide, should show similar behavior to stable Cs if the two isotopes have reached an equilibrium condition. Radioactive Cs-137, like Ra-226, is an important radionulide for long-term dose assessment and we also compared their TFs.

The calculated TFs for Ca, Sr, Ba and Cs had ranges of 0.003-0.036, 0.0018-0.006, 0.0006-0.006 and 0.0004-0.0042, respectively. Each result is plotted against TF-Ra in Figure 1. TF-Ca, TF-Sr and TF-Cs did not show any correlations with TF-Ra. It is well known that Ca is an essential element for plants, but Ra is not, so this difference was reasonable. Concerning Cs, the element belongs to the alkaline metal group, therefore, it is natural that Cs did not show any correlation with Ra. Interestingly, only TF-Ba was similar to the TFs of Ra giving a relatively high positive correlation ($r=0.65$, $p \leq 0.05$). Since the ionic radii of Ra (1.43Å) and Ba (1.35Å) are close, their transfers from soil-to-rice grain had the same trend.

In the present study, we measured TFs of Ra in brown rice samples; however, it is also interesting to collect TFs for other crops, e.g., wheat, green vegetables and so on. We are collecting these data and the findings will be discussed in the near future.

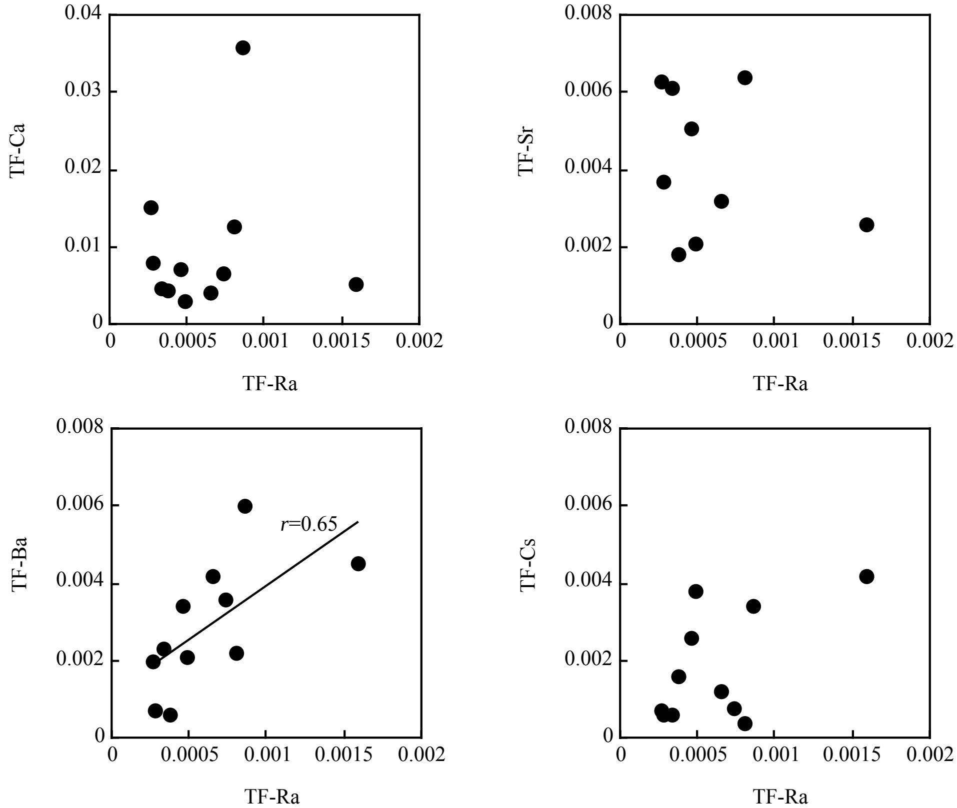


**Fig. 1. Relationships between TF-Ra and TFs-Ca, Sr, Ba and Cs from soil to brown rice collected in Japan.**

## CONCLUSION

Concentrations of Ra-226 in brown rice and associated paddy soils collected from 11 locations throughout Japan were measured to obtain TFs. The recoveries of Ba, used as a chemical yield tracer for Ra, were more than 97% for all the brown rice samples. The Ra-226 concentrations in the soils ranged from 20 to 65 Bq/kg-dry and those in brown rice samples ranged from <0.007 to 0.065 Bq/kg-dry. The TFs of Ra-226 ranged from 0.0003 to 0.0016 with an average of 0.00068, which was close to the TFs for maize (0.00024-0.006 with an average of 0.0012) reported in the IAEA TRS 364 [1]. Among the same alkaline earth metal group members, TF-Ba was similar to the TFs of Ra which we obtained.

## ACKNOWLEDGEMENTS

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