

INVESTIGATION OF ADSORPTION BEHAVIOUR OF FISSION PRODUCTS IN SOILS USING MULTITRACER TECHNIQUE

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The adsorption mechanism of fission products in soils has been investigated by using multi-tracer technique. The multitracer method used in this work contained fission products as carrier free isotopes which were prepared by neutron irradiation of ^{235}U at Kyoto University Research Reactor. After the soil sample was mixed into the multitracer solution for two weeks, the solution was separated from soil sample. The gamma-ray spectrometry was carried out for the separated solution and the original multitracer solution which has not been mixed with soils. Distribution coefficients of fission products for soil samples were estimated by measured photoelectric peaks. In this work, distribution coefficients of ten elements for thirty typical Japanese soils were observed.

1 Introduction

Electric power generation depends on nuclear power largely, and nuclear power plants and nuclear fuel reprocessing plants are adjacent to the civic life in Japan. In the case of nuclear accident in such plants, fission products and/or nuclear fuel material may be released into the environment. These radioactive materials are transferred into the air and deposited onto the ground surface by radioactive fallout. The fallout isotopes may be adsorbed on surface soils and/or migrate in the ground waters, rivers, and oceans to diffuse. A part of diffuse isotopes would be taken in plants, animals, and human in a food chain, and cause internal exposure. To estimate the influence of released radioisotopes, it is important to investigate the behaviour of the fallout isotopes in soils. One of the basic parameters to study their behaviour in soils is distribution coefficients for soils.

The distribution coefficients of some elements for typical soils have been reported [1], however, there are very little data for fission products. And the adsorption behavior of the fallout isotopes should be observed at trace level concentration because a possible difference may exist between the adsorption behaviour of trace and macro amount of elements.

Multitracer technique is one of the useful methods to investigate the behaviour of multiple elements at trace levels [2,3]. We have previously developed the preparation method of a multitracer using the thermal neutron-induced fission reaction of ^{235}U [4,5,6]. The fission products can be produced as carrier-free isotopes in this method. The distribution coefficients of fission products were determined for thirty kinds of typical Japanese soils using the fission multitracer in this study.

2 Experimental

The multitracer prepared at KUR (Kyoto University Reactor) was used to determine the distribution coefficients of fission products for soil samples. The multitracer was prepared by thermal neutron-induced fission reaction of ^{235}U , thus producing several radioactive isotopes (fission products) simultaneously [4,5,6]. Soil sample was mixed with the multitracer solution and had been contacted with the solution for fourteen days. After the contact with the soil, the solution was separated from soil sample. The radioactivity of the solution was measured to determine the amount of adsorbed fission products onto the soil. Thirty kinds of Japanese soils sampled at agricultural field in Japan were used in this work. The details of sample soils are listed in Table 1.

Table 1. Properties of the soil samples used in this work

| No. | Prefecture | Use | Soil type |
|-----|------------|--------------|--------------------|
| 1 | Hokkaido | rice paddies | grey lowland soil |
| 2 | Hokkaido | field | Kuroboku soil |
| 3 | Hokkaido | field | grey lowland soil |
| 4 | Akita | rice paddies | lowland paddy soil |
| 5 | Akita | field | Kuroboku soil |
| 6 | Fukushima | field | brown forest soil |
| 7 | Tochigi | rice paddies | wet Kuroboku soil |
| 8 | Tochigi | field | gray lowland soil |
| 9 | Chiba | field | grey lowland soil |
| 10 | Chiba | field | Kuroboku soil |
| 11 | Nagano | rice paddies | brown lowland soil |
| 12 | Nagano | field | Kuroboku soil |
| 13 | Shizuoka | rice paddies | grey lowland soil |
| 14 | Shizuoka | field | regosol |
| 15 | Niigata | rice paddies | grey lowland soil |
| 16 | Fukui | rice paddies | grey lowland soil |
| 17 | Fukui | field | gray lowland soil |
| 18 | Gifu | rice paddies | gray lowland soil |
| 19 | Gifu | field | brown lowland soil |
| 20 | Aichi | rice paddies | grey lowland soil |
| 21 | Aichi | field | yellow soil |
| 22 | Shiga | rice paddies | gray lowland soil |
| 23 | Hyogo | field | gray lowland soil |
| 24 | Tottori | rice paddies | gray lowland soil |
| 25 | Tottori | field | gray lowland soil |
| 26 | Kagawa | rice paddies | lowland paddy soil |
| 27 | Kagawa | field | lowland paddy soil |
| 28 | Saga | field | lowland paddy soil |
| 29 | Saga | field | lowland paddy soil |
| 30 | Kagoshima | rice paddies | lowland paddy soil |

The multitracer used in this work was prepared by the following method. A powder of $^{nat}\text{UO}_2$ (10-20 mg) and RbCl (10-20 mg) in same weight was mixed together and ground well. The mixed powder was pressed to form a pellet. The pellet of UO_2/RbCl was wrapped with an aluminum foil and sealed in a quartz tube in vacuo. The quartz tube containing the pellet was inserted in a polyethylene capsule. The capsule was irradiated by neutron flux of $2.8 \times 10^{13} \text{ n/cm}^2/\text{s}$ using pneumatic transferring system (Pn-2) at KUR for 30-40 min. The irradiated pellet was dissolved in few millilitres of 0.1 M HCl . The UO_2 powder which is not dissolved in dilute acid was removed from the solution containing fission products by filtration. A few hundred milliliters of distilled water was added to the separated solution to adjust the concentration of radioactivities of fission products. And a few drops of aqueous ammonium were added to the solution to adjust the pH at 5.0 to 5.5 that is typical pH range of the surrounding water of Japanese soil.

The soil sample (2 g) and the multitracer solution (20 mL) were mixed in a polyethylene tube and shaken once a day for fourteen days. The contacting time of fourteen days was determined by our previous work, in which it was found that fourteen days are enough to saturate the adsorption of the elements onto soils. After fourteen-day contact, the solution and soil were separated by centrifuge roughly and filtrated with cellulose acetate filter with pore size of $45 \mu\text{m}$. The adsorption of trace amount of isotopes to walls of polyethylene tube and filtering membrane was certified to be negligible in our previous work. The final pH of the solutions was measured, and there is no significant change between before and after the contact. In order to measure the activity of remained fission products in the solution, aliquots of the separated solution (10 mL) were measured by a Ge-detector for 40 000 seconds. And the solution which was not contacted with soil (original solution) was also measured to observe the difference of activities between before and after the contact. The experiments were carried out twice for several soil samples to confirm the reproductibility of the experiments; good reproducibility was found.

3 Result and Discussion

The adsorbed fission products were determined by gamma-ray spectrometry. Ten isotopes of ^{86}Rb , ^{95}Zr , ^{95}Nb , ^{103}Ru , ^{131}I , ^{134}Cs , ^{140}Ba , ^{140}La , ^{141}Ce , and ^{147}Nd were identified in the measured spectra. The peak areas assigned to the individual isotopes were calculated using a gamma spectral analyzing computer program. Activities of the tracers were corrected for the nuclear decay of each radioisotope. Distribution coefficients of the tracers were calculated for every soil samples using the determined activities of each isotope via the equation

$$K_d = \frac{C_0 - C_e}{C_e} \frac{V}{W}$$

where K_d is the distribution coefficient of tracers, and C_e and C_0 are the activities of the separated solution and the original solution. V and W are the volume of the multitracer solution and the weight of the soil. The distribution coefficients obtained for all the soil samples are presented in Fig.1. In this figure, squares show averaged K_d values, and vertical bars show the region of K_d values for every isotopes. There are some lower limits of

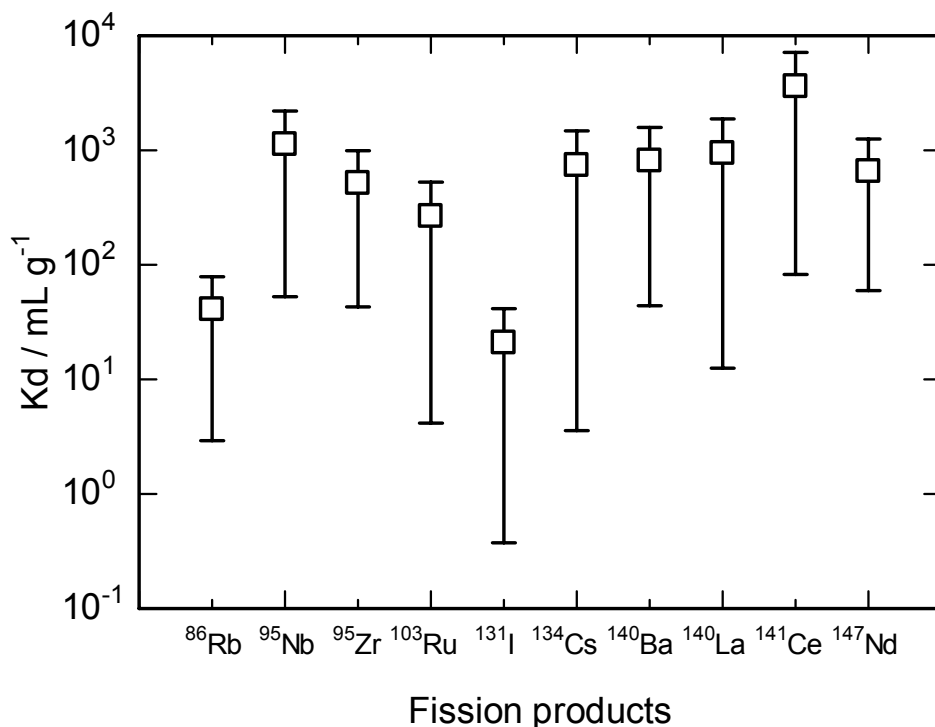


Fig. 1. Obtained distribution coefficients (K_d) for all soil samples. Squares show averaged K_d values, and vertical bars show the region of K_d values for every isotope

K_d values because photo peaks were not found in measured spectra for separated solutions.

In the previous work, we could obtain K_d values of fission products for seventeen Japanese soils [7]. A tendency in K_d values has been found in the previous work. 1) The K_d value of ^{86}Rb for all soils is about one hundred times lower than those for most other fission radionuclides. 2) There is a significant difference in K_d values between cesium and rubidium though they both are alkali metal elements. 3) All other transition elements show K_d values of about 10^3 . Rare earth elements show especially high values, close to 10^4 in some cases. 4) Varied values from a few to 10^3 were observed for ruthenium. Similar tendencies are also found in this work.

Correlations between K_d values and characteristics of soils (pH, amount of exchangeable-Ca, exchangeable-K, active-Al, active-Fe, and content of elements) could be investigated in this work because the number of soil samples increases. It is important to evaluate the correlation between K_d values and amount of active-Al because the active-Al is one of the main absorbents in Japanese soils [8]. The active aluminium (active-Al) is aluminium which exists on surface of soil particles as functional group of $\text{Al}(\text{OH})(\text{OH}_2)$ and/or $\text{Al}(\text{OH}_2)_2$ and adsorbs various ions strongly. The correlations between K_d values and active-Al for ^{95}Nb , ^{95}Zr , ^{103}Ru , ^{140}Ba , ^{141}Ce , and ^{147}Nd were shown in Fig. 2. It is

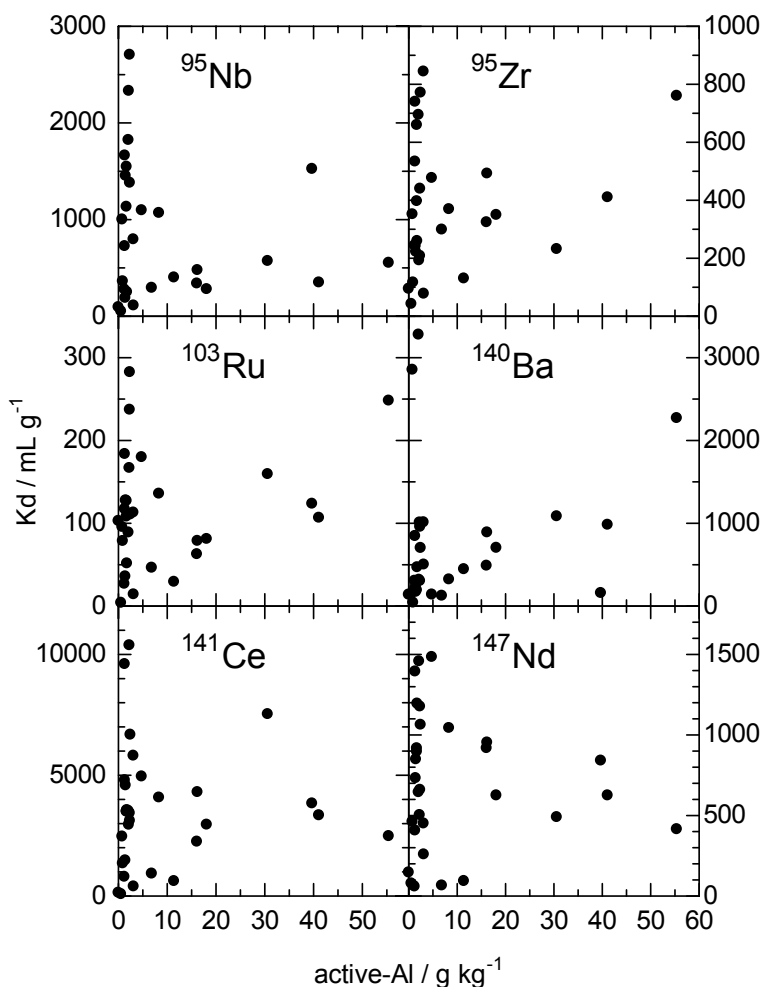


Fig. 2. Correlation between K_d values and the amount of active-Al for ^{95}Nb , ^{95}Zr , ^{103}Ru , ^{140}Ba , ^{141}Ce , and ^{147}Nd .

found that there are two kinds of positive correlations; very strong and weak dependence on the K_d values. There are more complex correlations for other elements. The difference of dependence means that there might be at least two kinds of chemical species which binds in different forms with active-Al and/or some components might disturb the adsorption to active-Al in the case of weak dependence. If difference of chemical species dictates the adsorption to active-Al, the correlation might become more complex. And there exists similar situation (two kinds of relation) in the correlation between K_d values and the amount of active iron, and two components correspond each other. Effect of active iron for absorption is generally considered of less importance than that of active aluminium, however, active iron is also the main absorbent in the soil [10]. Thus, there

might be common critical effect to dictate the contribution of active-Al, existence of some kinds of inhibitor for example.

4 Conclusions

Adsorption behaviour of fission products in thirty kinds of Japanese soils has been observed by using the fission multitracer. K_d values of fission products for soil samples were obtained. There are two components in the correlation between obtained K_d values and the amount of active-Al, that is, very strong and weak dependence on the K_d values. The similar dependence is found in the correlation between K_d values and active-Fe. It is suggested that there might be common inhibitor for the adsorption.

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