

Distributions of alkali and alkaline earth metals in several agricultural plants

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Abstract. Several agricultural plants were collected from an experimental field and the concentrations of alkali and alkaline earth metals were determined in different plant components. The concentrations of Cs in older leaves of rice and cabbage plants were higher than those in younger ones. The K concentrations in rice leaf blades decreased with withering, and these were similar in cabbage leaves. The distribution pattern of K concentrations in the plant components was similar to that of Rb, however that of Cs was different. The distributions of Ca, Ba and Sr concentrations in the rice leaf blades followed a relatively similar pattern, whereas that of Mg was different. The concentrations of alkaline earth metals in older leaves of a cabbage plant were higher than those in younger ones. The percentage distributions of Cs and Sr in non-edible plant components were 93 and 99% in rice plants, and 77 and 91% in cabbage leaves, respectively. These results showed that the non-edible plant components were important for understanding the transfer of the alkali and alkaline earth metals and radionuclides in the soil-plant system. These elements could serve as natural analogues to predict the distributions of ^{137}Cs and ^{90}Sr in different plant components.

1. INTRODUCTION

The first commercial nuclear fuel reprocessing plant in Japan is under construction and related nuclear cycle facilities are already operational in Rokkasho-mura, Aomori. In order to assess more precise and realistic estimation of radiation dose to the public around the nuclear facilities, the distribution and transfer of radionuclides in site-specific need to be taken into account. The distributions of radionuclides in plant components provide useful information for understanding the transfer of radionuclides in agricultural fields. Most of the non-edible plant components are returned to the soil as fertilizer where they may be recycled through the soil-plant system. In Japan several plant components, for example rice bran and straw, are also mixed with livestock feed. However, information on the distributions of these nuclides in plants is still limited.

The fate of radionuclides in the environment follows the behavior of stable elements. The particle-size distribution of radioactive ^{129}I aerosols, derived from nuclear facilities, is similar to that of stable I aerosols in the atmospheric environment [1]. The soil-to-plant (mushroom) transfer factor of radioactive ^{137}Cs is well correlated with that of stable Cs [2-4]. The profiles for weapon fallout ^{137}Cs and Cs in the forest soil organic layer are also similar [4]. Similar extraction profiles among sequential extraction fractions in the soils were found for fallout ^{137}Cs and Cs as well as ^{210}Pb and Pb [5]. Consequently, the behavior of the stable element in the environment may be regarded as a useful analogue in predicting the long-term change of the radionuclide.

Cesium-137 and ^{90}Sr are important radionuclides for the assessment of radiation exposure to the public because of their high fission yields, relatively long half-lives, high transferability, and wide distribution in the environment. For a better understanding of transfer processes of the radionuclides, the distributions of alkali (Na, K, Rb and Cs) and alkaline earth (Mg, Ca, Sr and Ba) metals in plant components were determined in the present study.

2. MATERIALS AND METHODS

2.1 Sample collection

Rice, cabbage and potato plants were collected from an experimental field, which was cultivated in a

manner similar to typical agricultural fields in Aomori, Japan. The rice plants were planted in the paddy soil in the middle of May, and harvested in the middle of October. The periods from planting to harvest time of cabbage and potato plants ranged from the beginning of May to the middle of August and from the beginning of June to the middle of October, respectively. The surface soils were also collected from each sampling area.

2.2 Sample preparation

Entire rice plant samples at harvest time were washed and then separated into different components including rice grains, leaves (leaf blade), stems (leaf sheath and culm) and roots. Rice grain samples were threshed and then polished to 90% of the total weight of brown rice. Hull, rice bran and polished rice samples were analyzed separately. The samples were dried at 50°C for 1 week and then pulverized with a stainless steel cutter blender. Polished rice and rice bran samples were ashed at a temperature lower than 450°C to avoid losses of alkali metals and then these samples were used to analyze Cs.

An entire cabbage plant sample at harvest time was carefully cleaned with water to eliminate extraneous soils. In order to determine the distributions of stable elements in leaf components, the leaf parts were cut every 3-5 leaves from the outer (older) to inner (younger) leaves. They were freeze-dried and then pulverized in a stainless steel cutter blender.

The potato plant samples were also cleaned in a similar manner as the previous samples, and then separated into different components such as leaves, petioles, stems and tubers. They were freeze-dried and then pulverized in a stainless steel cutter blender. The soil samples were dried at 50°C and then passed through a 2 mm sieve.

2.3 Sample analysis

The dried plant (500 mg) or ashed plant (50 mg) samples were put in a Teflon™ PFA pressure decomposition vessel with about 10 ml of mixed acid ($\text{HNO}_3 + \text{HClO}_4 + \text{HF}$), and decomposed with a microwave digester (SPEX, CDS7000) for 2-5 h. After digestion, the samples were evaporated to dryness. Then, the residues were dissolved in 3% HNO_3 . Aluminum, alkali (excluding K) and alkaline earth metals were determined by inductively coupled plasma-mass spectrometry (VG, PQΩ: ICP-MS) and K was determined by atomic absorption spectrometry (HITACHI, Z-8200). Indium was used as an internal standard to compensate for changes in analytical signals during the operation of ICP-MS. A standard reference material (IAEA, H-10 Hay) was used for validation of the analytical procedure [6].

3. RESULTS AND DISCUSSION

3.1 Concentrations of alkali and alkaline earth metals in surface soils

The concentrations of Al, alkali and alkaline earth metals in the soils are indicated in Table 1. There is no difference among the concentrations of each element collected from the three fields.

3.2 Concentrations of alkali and alkaline earth metals in plant components

It is well known that the major elements are unevenly distributed in a plant component. A typical example is the relative distribution of K and Ca in a rice grain (Figure 1), which was mapped using X-ray

Table 1: Soil concentrations of alkali and alkaline earth metals in an experimental field

Cultivated plant	Alkali metal					Alkaline earth metal			
	Al	Na	K	Rb	Cs	Mg	Ca	Sr	Ba
	mg kg ⁻¹								
Rice	95000	12000	7900	39	3.4	8000	18000	160	250
Cabbage	90000	13000	7500	40	3.3	8000	18000	170	240
Potato	99000	12000	7200	38	3.3	8800	18000	150	220

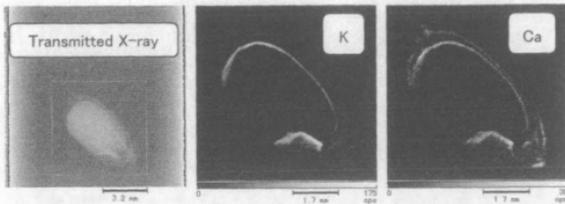


Figure 1: Image of transmitted X-ray and mapping of fluorescent X-ray intensities of K and Ca in rice using X-ray analytical microscopy.

analytical microscopy (HORIBA, XGT-2000W). The image clearly showed that the K and Ca content in rice bran was higher than that in the polished rice and hull components.

The concentrations of alkali and alkaline earth metals were determined at the different leaf blade positions of rice plants, ranging from the youngest to the oldest leaf blades. The surface of rice plant samples, such as leaves, stems and roots, was often contaminated by soil particles, even after washing with water. In order to correct the concentrations of the elements in the samples it was assumed that all Al in the samples originated from the soil particles. The concentrations of Cs in leaf blade positions increased from younger to older ones, whereas those of Na, K and Rb decreased (Figure 2). The Cs/K ratio at each leaf blade position significantly changed, with a 30 fold difference between first and fifth position leaves. These findings suggest that K in rice plants was more translocatable than Cs, which was dependent upon age, and their translocation rates were different. Broadley et al. [7] have found that the dominant inward rectifying K^+ channel in plant roots does not contribute significantly to Cs^+ uptake and voltage-insensitive cation channels mediate most Cs^+ influx. The concentrations of Ca, Sr and Ba were higher for both older and younger leaves rather than middle ones, whereas that of Mg had a relatively constant value independent of the positions. In addition, the relative concentrations of the elements in rice bran, hull, straw and root are shown in Figure 3. The values of K and Rb were higher in both rice bran and straw, and their distributions were similar to each other. However, that of Na was highest in the root. The relative concentrations of Ca and Sr were rich in the straw and roots of rice plants, but their distributions were different from those Mg and Ba. Each element had different distribution characteristics.

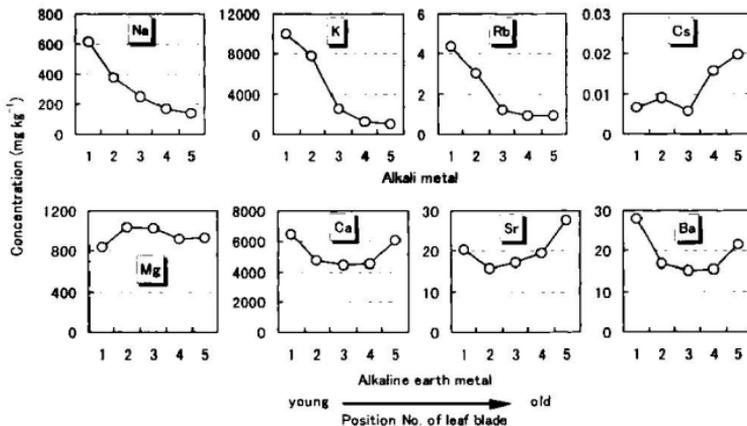


Figure 2: Concentrations of alkali and alkaline earth metals in different leaf blade positions of rice plants.

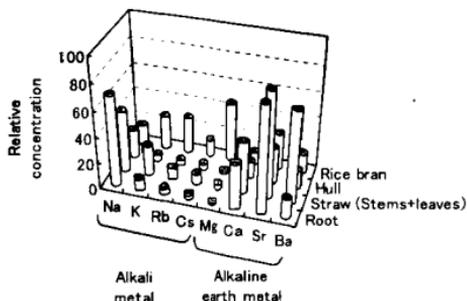


Figure 3: Relative concentrations of alkali and alkaline earth metals in rice plant components. Concentrations of elements in rice plant components were normalized with that in polished rice.

The distributions of the elemental concentrations from outer (older) to inner (younger) leaves in cabbage plants at harvesting time are indicated in Figure 4. The concentrations of the elements were divided into two groups according to their different patterns in the leaf positions as follows: (1) the concentrations of the element were higher in outer leaves than in inner ones (Na, Cs and alkaline earth metals), and (2) the concentrations of the elements had a relatively constant value independent of leaf positions (K and Rb). Because the inner leaves are tightly compressed in globularness while the outer leaves are opened, the amount of transpiration of the outer leaves of a cabbage plant are higher than those in the inner leaves. Therefore, the concentrations of most elements were higher in the outer leaves than in the inner ones. On the contrary, the mobile elements (K and Rb) in cabbage leaves had relatively constant concentrations. A similar observation for ^{137}Cs and ^{85}Sr obtained by radiotracer experiments was reported [8]. The concentrations of K and Cs in outer and inner leaves were within a factor of two, although the concentration ratio of Cs/K in outer leaves varied compared to inner leaves. The concentration ratios for Sr/Ca in cabbage leaf positions showed a relatively constant value, even if the

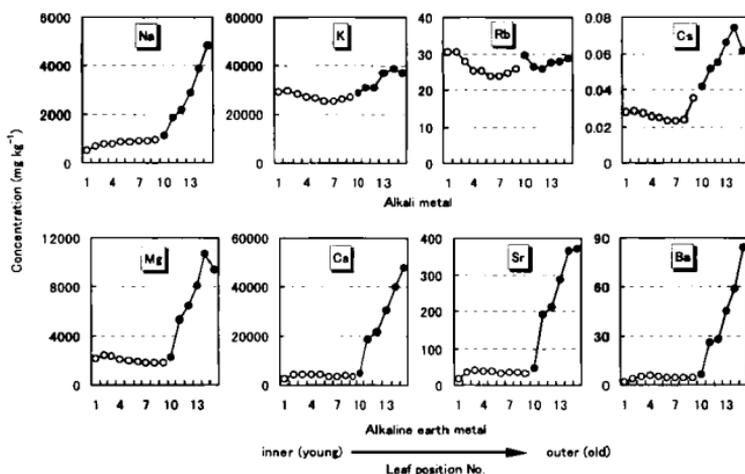


Figure 4: Concentrations of alkali and alkaline earth metals in different leaf positions of a cabbage plant. Leaf positions of Nos. 1-9 show edible parts (inner leaves, ○) and those of Nos. 10-15 show non-edible parts (outer leaves, ●).

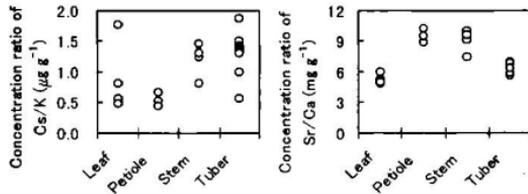


Figure 5: Concentration ratios of Cs/K and Sr/Ca in potato plant components. The samples were collected on June 1, July 18 and 30, and August 14, 1996.

concentrations of Ca and Sr in outer leaves were one order of magnitude higher than those in inner leaves. These findings suggest that the translocation rates of both Ca and Sr were similar within the cabbage leaves, while those of K and Cs were different.

The concentration ratios for Cs/K and Sr/Ca in potato plant components collected on 4 different dates were plotted in Figure 5. The concentrations of K and Cs in the plant components were within one order of magnitude for each element, although the Cs/K ratios varied. The concentrations of Ca and Sr in the tuber were two orders of magnitude lower than in leaf, petiole and stem parts. The Sr/Ca ratio in each component had an individual value.

3.3 Percentage distribution of alkali and alkaline earth metals in plant components

The percentage distributions of dry weight, alkali and alkaline earth metals were indicated in Figure 6. This study assumed 4% as root percentage to total rice plant [9], because the entire root system could not be completely collected from the soil. Percentage distribution of dry weight in the edible portion (polished rice is commonly eaten) was 34% at harvest time and the weight percentage of non-edible components was 66%, 50% of which was straw. The distributions of dry weight components in a rice plant described by Myttenaere et al. [10] were comparable to those found in this study. The distribution of Cs in polished rice is 7% because of the lower concentration of Cs in polished rice, though its weight is 34%. Straw contained 73%, followed by rice bran (10%), hull (7%) and root (3%) in non-edible components. These findings suggest that more than 90% of Cs uptake by rice plants is utilized in the soil-plant system and/or transferred to the feed-livestock pathway. The distribution of Cs in rice plant components in the present study was in agreement with the results obtained by a tracer experiment [10]. The reported value by Wang et al. [11] was different from the present one. For their sampling periods in

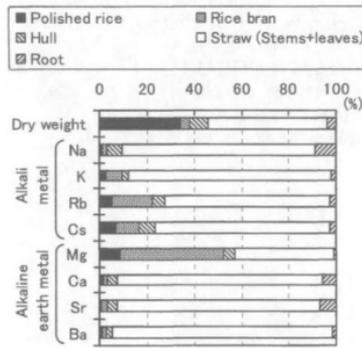


Figure 6: Distribution of elements in rice plant components.

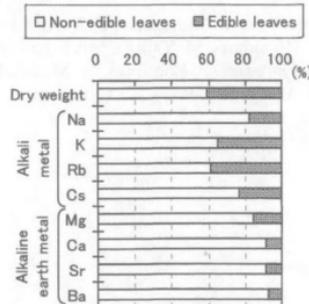


Figure 7: Distribution of elements in non-edible and edible cabbage leaves.

1982, the fallout of ^{137}Cs following the 26th Chinese atmospheric nuclear weapon test in 1980 had caused higher concentrations even during 1982, and then the contamination was not only from root uptake but also by the direct deposition of ^{137}Cs on the rice plant. The distributions of Na, K and Rb in the non-edible parts were 99, 98 and 95%, respectively. The distributions of Ca, Sr and Ba had relatively similar values and those in the non-edible parts were 99%. That of Mg was extremely rich in rice bran (44%) because the Mg concentration in rice bran was 46 times higher than that in polished rice.

The distributions of the dry weight, alkali and alkaline earth metal contents in edible and non-edible cabbage leaves are shown in Figure 7. The percentage distributions of dry weight contents in edible and non-edible leaves of a cabbage plant at harvest time were 41 and 59%, respectively. However, each element had an individual value. The percentage distribution of Sr (first group of elements) in non-edible leaves was 91% and those of the first group elements were rich in the non-edible portion. While those of K and Rb (the second group of elements) in non-edible leaves were 65 and 61% respectively, which were relatively similar values to their dry weight distribution. These findings suggest that the distribution of Cs in plants is different from that of other alkali metals such as Na, K and Rb, and the distribution of Sr is similar to that of Ca.

The distributions of stable elements in agricultural fields may be regarded as a useful analogue in predicting the long-term changes of radionuclides. Consequently, studies utilizing the transfer of elements could be regarded as a convenient substitute to help describe the long-term behavior of radionuclides due to human activities in the environment. In order to have a better understanding of the long-term fate of radionuclides in the environment, more studies on the transfer and distribution of stable isotopes and related elements in the total environment will be required.

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