Chapter 4
Absorption of Radioceasium in Soybean

Naoto Nihei and Shoichiro Hamamoto

Abstract Radioactive materials, primarily radiocesium (^{134}Cs + ^{137}Cs), were released into the environment by the Fukushima Daiichi Nuclear Power Plant accident in March 2011. The percentage of soybean plants that had a concentration of radiocesium over 100 Bq/kg was higher than that of other crops. To examine the reason why the concentration of radiocesium in soybeans was high, its concentration and distribution in seeds were analyzed and compared to rice.

Potassium fertilization is one of the most effective countermeasures to reduce the radiocesium uptake by soybean and nitrogen fertilizer promotes soybean growth. To use potassium and nitrogen fertilizers safely and efficiently, applied potassium behavior in soil and the effect of nitrogen fertilizer on radiocesium absorption in soybean were studied.

Keywords Nitrogen · Seed · Soybean · Potassium · Radiocesium

4.1 Introduction

The Great East Japan Earthquake occurred on March 11, 2011 and it was immediately followed by the nuclear accident at the Fukushima Daiichi Nuclear Power Plant, Tokyo Electric Power Company. Radiocaesium, the dominant nuclide released, was deposited on agricultural lands in Fukushima and its neighboring prefectures, which contaminated the soil and agricultural products.

To revitalize agriculture in the affected regions, the authorities in Fukushima Prefecture have been promoting countermeasures for reducing radiocaesium (RCs) uptake by plants and the remediation of polluted agricultural land. Some of these remediation techniques include the application of potassium (K) fertilizer, plowing

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to bury the topsoil, and stripping the topsoil. Emergency environmental radiation monitoring of agricultural products (hereafter, referred to as “monitoring inspections”) have been conducted by the Nuclear Emergency Response Headquarters to assess the effectiveness of the countermeasures to keep food safe (Nihei 2016). Approximately 500 food items were monitored, which produced 100,000 data points by the end of March 2016. The monitoring inspections indicated that the percentage of soybean plants with a radiocaesium content of greater than 100 Bq kg^{-1} (fresh weight), was higher compared to other cereal crops (Fig. 4.1); 100 Bq kg^{-1} is the maximum allowable limit of radiocaesium in general foods. Because the cultivation area of soybean plants in Fukushima Prefecture is the second largest after rice, the analysis of RCs uptake by soybean plants is particularly important.

To cultivate soybean after the accident, farmers were recommended to apply potassium fertilizer until the exchangeable potassium (Ex-K; extracted with 1 mol L^{-1} ammonium acetate) is greater than 25 mg K_2O 100 g^{-1} or higher. This recommendation was made because it is known that potassium fertilization is effective for reducing radiocaesium concentration in agricultural crops. However, Ex-K did not increase in the soil for some soybean fields in Fukushima Prefecture after the application of K, resulting in relatively higher RCs concentration in those seeds. Moreover nitrogen fertilizer has a large effect on crop growth, but few studies have examined how nitrogen contributes to the absorption of RCs in soybean.

In this chapter, the reasons why RCs concentration in soybean is higher compared to other crops, potassium behavior in the soil with the low effectiveness of potassium application, and the effect of nitrogen fertilization on RCs absorption in soybean will be discussed.

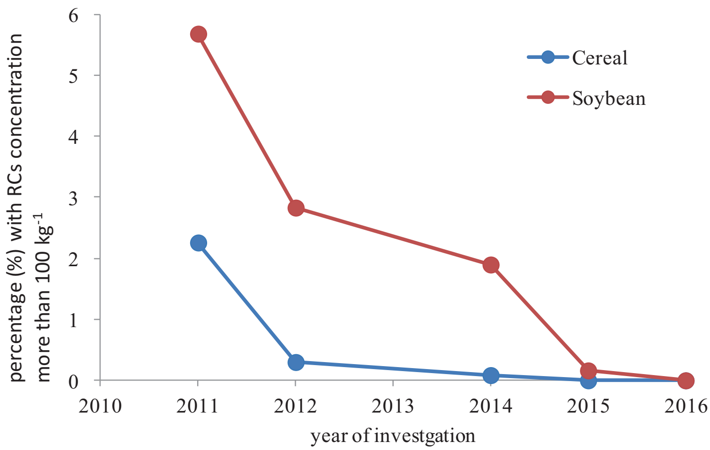

Fig. 4.1 The percentage of soybean plants and cereals with a radiocaesium content exceeding 100 Bq kg^{-1} in the monitoring inspections carried out in Fukushima prefecture

4.2 The Concentration Distribution of Cs in Soybean Seeds

Even though there are some reports of Cs uptake by soybean plants, it is not clear why the concentration of Cs in soybean seed is higher than those in other crops such as rice.

When the concentration distribution of Cs in soybean seeds were analyzed using radioluminography with RCs (Nihei et al. 2017), it was found that Cs was uniformly distributed in the soybean seed, as was potassium, both of which likely accumulated in the cotyledon. The chemical behavior of Cs is expected to be similar to that of K because they are both alkali metal elements and have similar physicochemical properties. Therefore, it is assumed that Cs is also accumulated in the cotyledon like K. In the case of rice grain, the concentration distribution of RCs is localized and rice grains accumulate Cs in the embryo (Sugita et al. 2016), which is only a small part of the rice grain. The different distributions of RCs for rice grain and soybean seed appear to be derived from their seed storage tissues. Soybean seed does not develop its albumen and is therefore called an exalbuminous seed. The cotyledon capacity occupies the largest part of soybean seed. The monitoring inspections measured the edible parts of the crop, i.e., seeds and grains for soybean and rice, respectively. The results suggested that the large capacity of Cs accumulation in soybean seeds is one of the reasons why the concentration of radiocaesium in soybeans was higher than that of rice in the monitoring inspections. In addition, the Cs concentration of each organ and the ratio of absorbed Cs to seeds in mature soybeans were examined. Approximately 40% of absorbed Cs was accumulated in the soybean seeds (Nihei et al. 2018) (Fig. 4.2), while rice grains accumulate only 20% of the entire amount absorbed (Nobori et al. 2016). It is not clear whether the amount of Cs that the soybean plants absorb is larger than that of rice. However, the results from this examination indicates that soybean plants translocate absorbed Cs to its seeds more easily than rice.

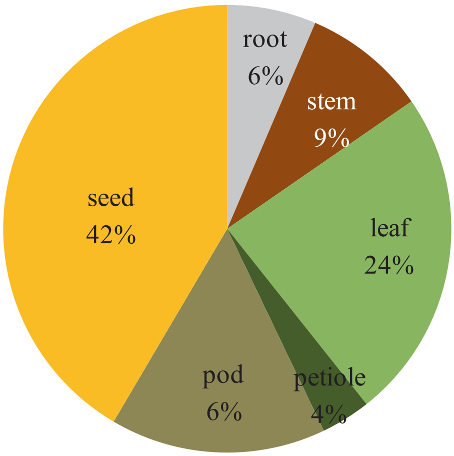

Fig. 4.2 Percentage of ^{137}Cs activity in different organs of mature soybean plants

4.3 Potassium Behavior in the Soil with Low Effectiveness of Potassium Application

It is important to understand the behavior of applied K and the soil characteristics with low Ex-K content to establish efficient techniques to decrease RCs in crops. Therefore, we examined the behavior of K in soil following K fertilizer application (Hamamoto et al. 2018). We tested two types of soil in Fukushima Prefecture (Fig. 4.3). Soil A increases Ex-K with K fertilization (i.e., control treatment), and soil B does not increase Ex-K.

First, a batch experiment was conducted with the two types of soil. After adding KCl (27 mg, 54 mg, 81 mg K 100 g^{-1}) to these soils and culturing for 5 days, Ex-K in soil A increased with the addition of KCl, however, Ex-K in soil B did not increase (Fig. 4.4). There are two reasons for this result: (1) leaching of applied K from the soil and (2) fixation of applied K in the soil. Since this experiment was carried out in a closed system, it was considered that the reason why Ex-K did not increase in soil B was due to the strong adsorption of applied K onto the soil which could not be extracted with ammonium acetate.

Next, a column transport experiment was undertaken to investigate fertilized K behavior in detail (Fig. 4.5). In the repacked soils, the radioisotope tracer, ^{42}K (half-life =12.36 h), was applied to the top 4-cm soil layer, and the soil beneath the top layer was ^{42}K free. Water was applied to the top of the column using a rainfall

Fig. 4.3 Soil A sampling in Fukushima prefecture

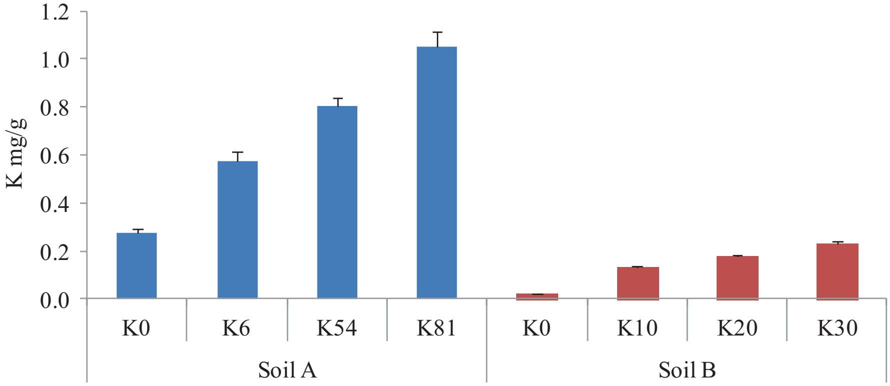

Fig. 4.4 The ratio of exchangeable K to applied K in soils A and B. K0, no K fertilizer. K27, K54, and K81, adding 0.27, 0.54, 0.81 mg K (as KCl) to 1 g soil

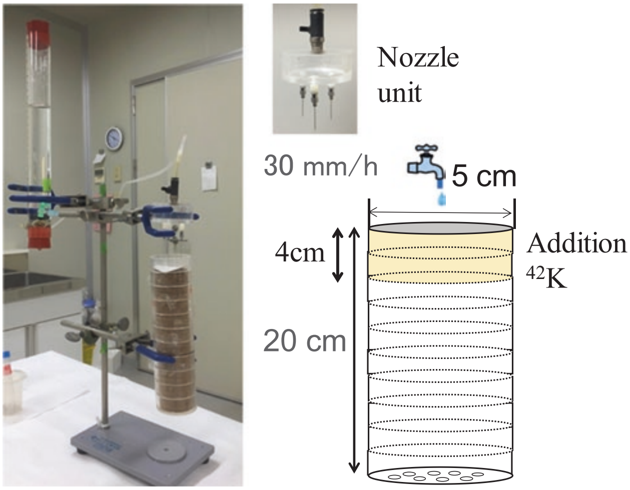

Fig. 4.5 Column transport experiments with soils A and B

simulator connected to a Mariotte's tank. During the experiment, rainfall intensity was maintained at 30 mm/h. After 1 h of 'rainfall', the soil column was horizontally sectioned in 2-cm discs. Samples were taken from each disc, extracted with water (i.e., water-soluble K fraction) and ammonium acetate (i.e., exchangeable K fraction). The ^{42}K activity of each fraction was measured with a semiconductor detector. The ^{42}K obtained by subtracting ^{42}K for ammonium acetate extracts from total ^{42}K was defined as the fixed form. Although 30 mm of water was applied, water content increased almost up to the bottom of the column, however the mobility of the applied ^{42}K was very low in both soils. Only a small quantity of ^{42}K was detected at a soil depth of 4–6 cm after applying ^{42}K to the top 4-cm soil layer in both soils. In soil A, about 75% of ^{42}K retained in each 2-cm disc was the exchangeable form, while in soil B, about 60% of ^{42}K was the fixed form. Again, the findings suggest the soil B can fix a large amount of applied K. Further, from the result of X-ray diffraction (XRD) charts, the clay mineral of soil A was mainly smectite and zeolite, and soil B was mainly vermiculite. In vermiculite such as 2:1 clay mineral, a hollow six-membered ring exists on the tetrahedral silicon sheet facing the layer boundaries. The radius of this void and the potassium ion radius are nearly equal, and the ion is attracted to the six-membered ring. Moreover, the degree of weathering of micaceous minerals (i.e., vermiculitization) may affect the extent of K fixation to layer charge (Sawhney 1970). Since vermiculite, which originates from the weathering of granite, is one of the major clay minerals found in Fukushima, Japan, special attention is needed when K application is used to reduce RCs transfer to crops in such soils.

4.4 The Effect of Nitrogen Fertilization on RCs Absorption in Soybean

Nitrogen (N) has a large effect on crop growth. However, few studies have examined how nitrogen contributes to RCs absorption in soybean. Focusing on this point, we studied the effect of nitrogen fertilizers on RCs absorption in soybean seedlings. The RCs concentration in soybean increased as the amount of nitrogen fertilizer increased. The different forms of nitrogen treatment increased the RCs concentration of soybean in the following order: ammonium sulfate > ammonium nitrate > calcium nitrate. Hence, ammonium-nitrogen increased RCs absorption more than nitrate.

Geometrically, RCs ions are adsorbed and fixed strongly to the clay mineral, and these ions are probably not available for plant uptake. However, because the ionic radius of the ammonium ion is similar to that of the cesium ion, ammonium exchanged and released RCs from the soil. We found that the amount of RCs extracted by the ammonium-fertilizer increased the day after fertilization, and thus, RCs would become available for uptake by soybean plants. In addition, the ammonium and cesium ions are both univalent cations, and ammonium has been found to restrict cesium absorption in hydroponics (Tensyo et al. 1961). However, with the soil was used in the current study, ammonium ions were oxidized to nitrate ions during cultivation. Therefore, we suggest that ammonium fertilizer promotes the activity of RCs in soybean without restricting it.

Soybean cultivation area in Fukushima Prefecture is the second largest after rice cultivation. Therefore, to assist the recovery and revitalization of agriculture in the contaminated regions, it is important to develop agricultural techniques that inhibit Cs accumulation in soybean plants. One technique is the use of K fertilizer, which has been used in contaminated regions. Further studies will be needed to develop more efficient techniques, such as the examination of the mechanism of Cs accumulation in soybean seeds and the improvement of soybean varieties able to alleviate the absorption of radiocaesium.

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