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Mitigation of Radioactive Contamination from Farmland Environment and Agricultural Products

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Abstract: Radioactive Cs contamination of vast area including crop land with a focus on TEPCO's Fukushima Daiichi Nuclear Power Plant accident in March 2011, still ask farmers to reduce the uptake of radioactive materials to the crops in the farmland even today. Several removal technologies from the paddy and upland soils, volume reduction of waste materials, and potassium fertilizer application to reduce the uptake of radioactive cesium (Cs) to the edible part of plant will be presented. In the case of food supply, as the concentration of radioactive substances in food less than standard value (100 Bq/kg) is required, sufficient removal of radioactive Cs from the field and reduction of transfer factor from soil to plant by maintaining potassium levels in soil throughout the growth stage, successively reduced the number of food samples with excess levels of radioactive Cs in food products. Furthermore, the food processing steps also played an important role to remove the radioactive Cs from the food (such as wheat, soybean, etc.). On the other hand, attention should be paid to reduction of radiation exposure of farm workers during their labor in the fields, by removing or shielding the radioactive Cs from the environment. Several agricultural machineries have been modified to adapt in contaminated areas. As the contaminated soil is limited to the very thin but fertile top soil layer, removal of contaminated soil and top-dressing of non-contaminated but low fertile soil sometimes reduce the fertility of the field and also induce erosion, so we also attempt to develop methods to compensate the productivity after the decontamination of the field by applying forage grasses to increase the soil fertility and reduce the possible erosion of topsoil from the field.

Key words: radioactive cesium, nuclear power plant accident, decontamination of radioactive Cs from field, mitigation of radioactive Cs to plant

1. Introduction

A large area of Eastern Japan was contaminated by radioactive Cesium (^{134}Cs and ^{137}Cs) because of the accident of Fukushima Daiichi Nuclear Power Station (FDNPP) after the attack of Great East Japan Earthquake and subsequent Tsunami occurred in 11th March 2011 (Fig. 1). Not only Fukushima prefecture, those adjacent prefectures were also suffered by radioactive Cs to some extent.

Table 1 indicates the top 12 prefectures of agricultural working population and agricultural production in 2010 among 47 prefectures in Japan. It indicates that those prefectures played important roles in Japanese agricultural activity. As shown in Fig. 1, the contamination by radioactive Cs was mainly spread to the southern direction of Fukushima prefecture. This pattern is also observed in the report of contaminated agricultural products in 2011 (Fig. 2). In 2011, tea production was severely damaged as shown in Fig. 2. And the contamination was reported from as far as Shizuoka prefecture, which is about 400 km from the FDNPP. It is important to consider the situation of vegetation at the time of the radioactive Cs scattering to the air. Though early March is just before the spring in these areas, while as tea plant is evergreen plant species, the canopy was covered with leaves. Thus it is considered that the radioactive Cs was attached to the existing leaves on the top of the canopy. Distribution of radioactive Cs was investigated by dividing plant canopy into several parts as shown in Fig. 3, and it is

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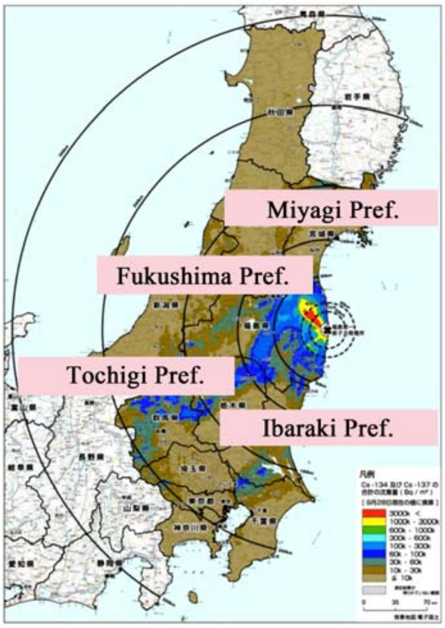

Fig. 1 Airborne monitoring survey of soil surface radioactive Cs (sum of ^{134}Cs and ^{137}Cs, October 12, 2011) [1].

Table 1 List of agricultural working population and production (JPY) in top 12 prefectures in Japan (2010) [2].

Rank	**Prefecture**	**Production (billion JPY)**
1	Hokkaido	995
2	Ibaraki	431
3	Chiba	405
4	Kagoshima	401
5	Kumamoto	307
6	Aichi	296
7	Miyazaki	296
8	Aomori	275
9	Niigata	256
10	Tochigi	255
11	Fukushima	233
12	Iwate	229
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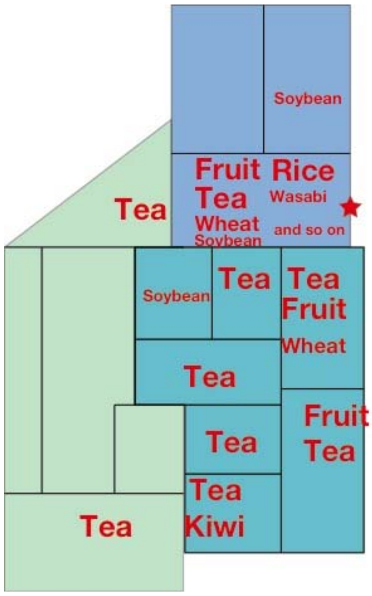

Fig. 2 Agricultural products contamination by radioactive Cs reported in 2011 (Sum of ^{134}Cs and ^{137}Cs were higher than 100 Bq/kg FW) [3].

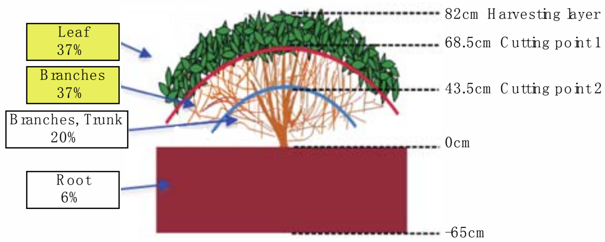

Fig. 3 Distribution of radioactive Cs in the tea plan [4].

confirmed that tea canopy trapped the radioactive Cs from the air. Furthermore, as there is a possibility that newly dressed radioactive Cs to the soil was absorbed by plant, then transported to the newly born tea leaves, stable Cs isotope was applied to soil or existing leaves, then the transportation to the newly born leaves was investigated (Fig. 4) [5]. It was demonstrated that the translocation from soil was negligible but those stable Cs applied to the existing leaves translocated to newly born leaves. Based on these observations, cutting off the leaves and branches at the top canopy was adopted.

This countermeasure was effectively worked to reduce the contamination of tea plant, and further report of exceeding the limit value was not found after 2012. In the case of fruit trees, as most of them were deciduous trees, there was no green leaves at the timing of the accident. Thus, direct attachment of radionuclides occurred on the surface of branch and trunk, and within the bark of tree. To remove the radioactive Cs from the fruit tree (e.g., Pair, peach, grape, persimmon, etc.), bark scraping by hand or high pressure washer was performed. This method effectively decreased the surface dose rate of tree [6].

On the other hand, paddy fields and most of upland fields were not cultivated yet in early March, and most of the fall out was accumulated on the top of soil surface. Thus the decontamination of soil was required.

It is important to divide the strategy to decontaminate radioactive Cs from the soil from the viewpoint of agricultural products contamination and reduction of air dose level (Fig. 4). The purpose of the former target is to protect internal exposure by food intake of the consumers, and for this purpose the target level is put as 100 Bq/kg after 2012. And in this case, the condition of plow layer is focused and the detection is mainly operated by using Ge semiconductor detector. The latter target is to reduce external exposure of local residents, especially of farmers. For this purpose the target level is put not to exceed 1 mSv/year by the additional exposure by fall out. In this case, radioactive compounds located in the top soil layer is focused, thus surface soil layer is most important, and the detection was carried out mainly by survey meter.

To investigate and develop countermeasures on these topics, NARO put actual proof trials just after the event in 2011. The trials were carried out in Iitate village and adjacent Yamakiya district in Kawamata town (Fig. 5). The trial includes top soil stripping, paddling and suspension removal, phytoremediation, and so on. Of course not all the trials finished successfully, we have concluded that the role of phytoremediation to remove radioactive Cs from soil is very limited (ca. 0.5% per one cultivation cycle), so we do not recommend phytoremediation for the effective method for decontamination of soil.

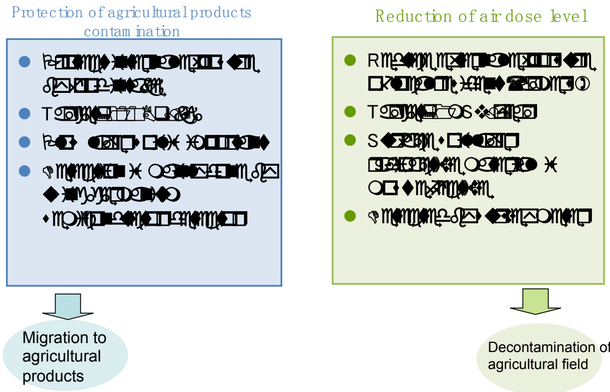

Fig. 4 Scheme of radioactive Cs removal from the soil.

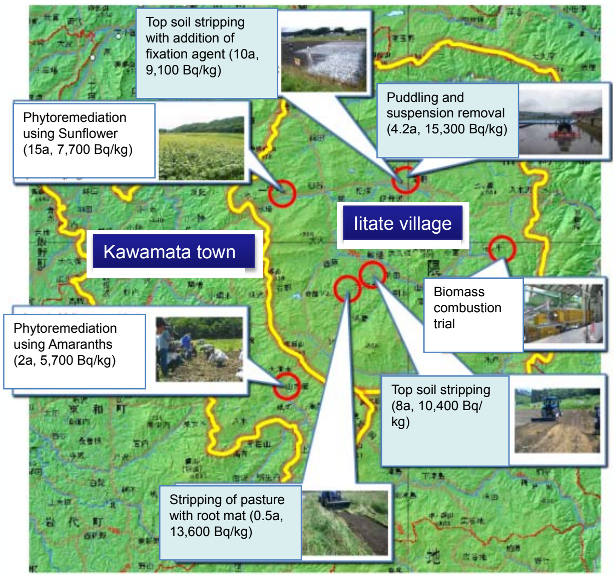

Fig. 5 Actual proof trials in 2011.

It was concluded that physical removal of radioactive Cs from soil is most effective. However, we need to consider the decontamination technique based on two types of field condition, before tillage and after tillage (Fig. 6). If the soil was not disturbed by tillage, as radioactive cesium is limited in the top soil layer, most effective method is to remove top soil by stripping. This method is very effective but the problem is how to manage the disposal of soil after stripping. It is also possible to remove the top soil layer to the lower layer by inversion tillage. This technique also effectively to decrease the surface doses rate by the inhibition of radiation transmission by soil itself. And this technique is applied to those field whose surface radioactivity is not so high (that is, lower than 5,000 Bq/kg), and there is sufficient suitable deep soil layer also. Furthermore, if the soil contamination level is low, it is also possible to apply deep ploughing to dilute the radioactive materials in the soil. On the other hand, if the soil was cultivated before decontamination, top soil stripping is not applicable because huge amount of fertile soil is required to be removed. So, in the case of paddy field, paddling and suspension removal technique has been developed. This technique is based on the observation that most of the radioactive Cs is attached to the small soil particle mainly to the clay particle (Fig. 7) [7].

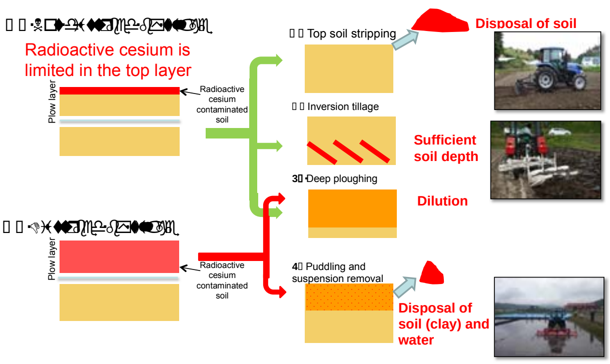

Fig. 6 Decontamination of radioactive Cs from soil.

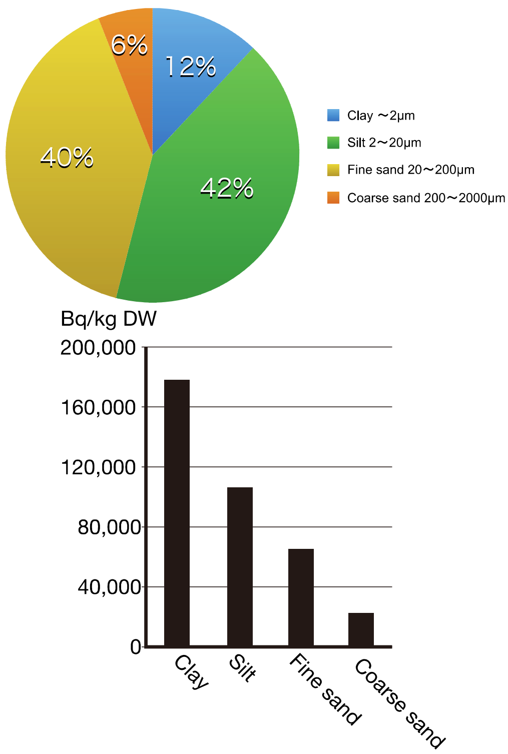

Fig. 7 Distribution of radioactive Cs in clay, silt, fine sand and coarse sand (upper) and concentration of radioactive Cs in each fraction (lower).

Fig. 8 Paddling and suspension removal on small scale paddy field. (National Institute for Agro-Environmental Sciences, NARO Tohoku Agricultural Research Center).

Paddling and suspension technique is widely used paddy rice cultivation technique to prepare the soil suitable for transplanting of rice seedling. Small soil particle containing clay fraction can be separated by applying this technique, radioactivity of the soil decreased effectively (Fig. 8) [8].

Radioactive Cs distribution is not uniform and it is required to be monitored precisely. If the contamination level is high and required to treat not to disperse the soil during the decontamination, soil hardener technique is also proposed to fix the top soil (Fig. 9). Which make visible check of the target area and decrease the risk of soil particle intake by inhalation who work there. Though the proposed technology and corresponding contamination level is indicated, it should be decided based on each field condition and farmer's request.

In 2011, though those areas which were suffered radioactive Cs fall out severely, were prohibited to cultivate rice plant, rice cultivation was possible outside the prohibited area but some brown rice showed higher contamination level. To make clear the reason why high radioactive Cs concentration of brown rice occurred, produced rice and corresponding field soil data were collected and analyzed. At first the radioactive Cs concentration was plotted between soil and brown rice (Fig. 10). The relationship is not clear, and it is required to include another parameter to explain the result. It is reported that Cs uptake is mainly carried out by the activity of K transporter and the ratio of Cs to K concentration in the soil solution is important [10]. Thus K addition effectively decrease Cs uptake and transfer from soil to plant [11-13]. When exchangeable K and transfer factor (TF) of rice in the corresponding field was plotted, there was a clear negative relationship (Fig. 11) [14]. Which confirms that Cs uptake is also regulated by K transporter mediated pathway, thus the level of K in the soil is important. By using this result, if the soil radioactive cesium concentration is 5, 000 Bq/kg, and the TF is 0.01 indicates that the radioactive Cs content in the rice would be 50 Bq/kg. So, it is proposed to keep the soil ex. K level to 25 mg K_2O/100 g throughout the growth. Thus, sufficient amount of potassium fertilizer, mostly KCl, was dressed before the normal fertilizer application. And this guideline was introduced for the rice production in 2012 and still in use.

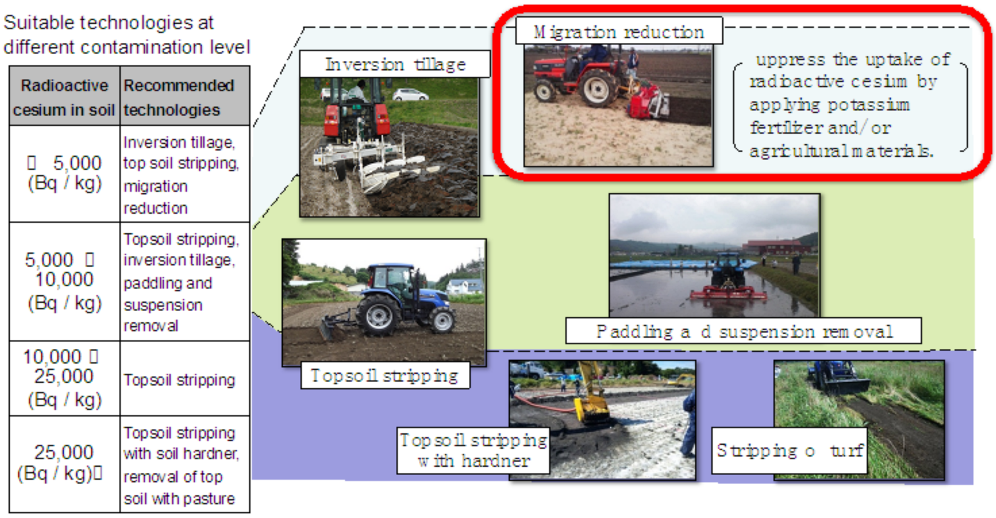

Fig. 9 Scheme of proposed decontamination of radioactive Cs from agricultural field.

Based on these decontamination and mitigation efforts, the prohibition area for rice cultivation decreased year by year (Fig. 12). In 2011, the area was about 8,500ha, and in 2014 it was about 2,100 ha.

The relationship between soil Exchangeable K and TF is also observed in field crops such as soybean and buckwheat (Fig. 13). Thus the Cs uptake from soil is dependent on the K status of the soil regardless of paddy or upland condition.

Beside decontamination and mitigation of radioactive Cs, it is also important to address another aspect how to regulate the radioactive Cs contamination in the food. In Table 2, food processing factor using soybean is demonstrated. Food processing factor is designated as Food processing factor = Radioactive Cs content after processing (Bq/kg fresh weight)/Radioactive Cs content before processing (Bq/kg fresh weight). To make up the food stuff, we

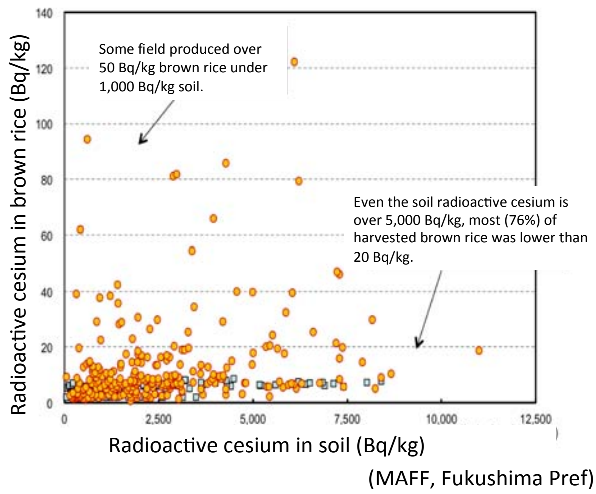

Fig. 10 Relationship between radioactive Cs concentration of soil and brown rice grown in a same field [9].

further process the material such as by adding water, boiling, fermentation, etc. And final food radioactivity can be evaluated by using food processing factor as shown in Table 2. By using food processing factor, it

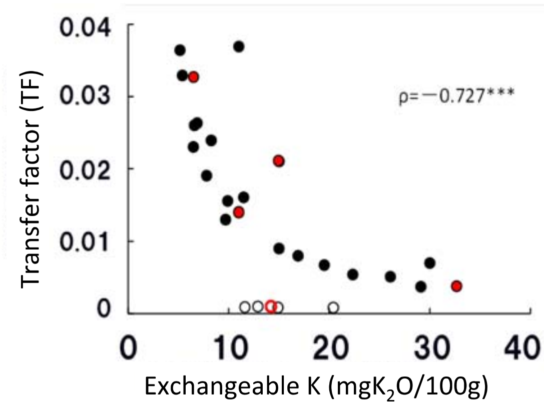

Fig. 11 Relationship between soil exchangeable K and transfer factor. Transfer factor (TF) = radioactive Cs in brown rice/radioactive Cs in soil.

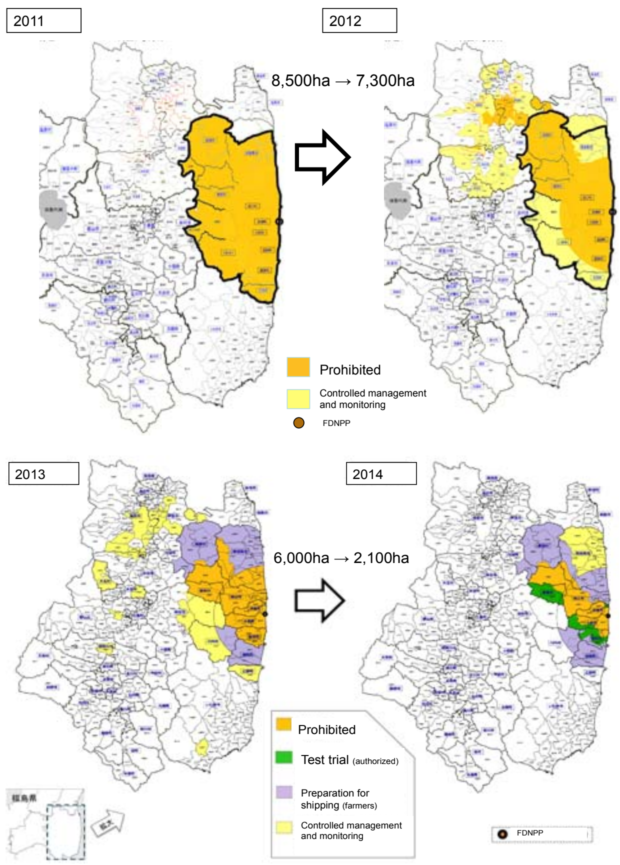

Fig. 12 Successive change of the restricted area for rice production [15, 16].

is clearly shown that the level of radioactive Cs decreased by the most of food processing. And these results make the food manufacturer consider safer to use the agricultural products even under the limit value.

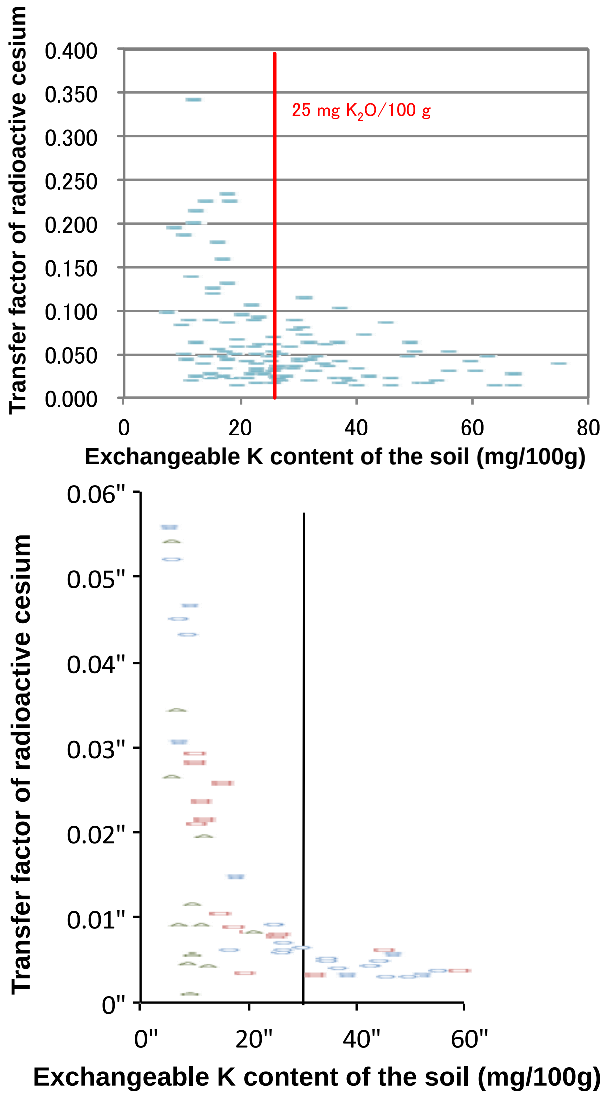

Fig. 13 TF of soybean (upper) [17] and buckwheat (lower) [18].

Table 2 Food processing factor from soybean [19].

Processed food stuff from soybean		Food processing factor
Tofu	Soy pulp	0.18
	Soy milk	0.13
	Tofu	0.12
Natto	Pressurized steam cooked beans	0.40
	Natto	0.40
Boild beans		0.20

Though decontamination of the field by top soil stripping has been progressed to decrease the radioactivity, removal of contaminated soil can not be a simple answer. After top soil removal, soil is covered

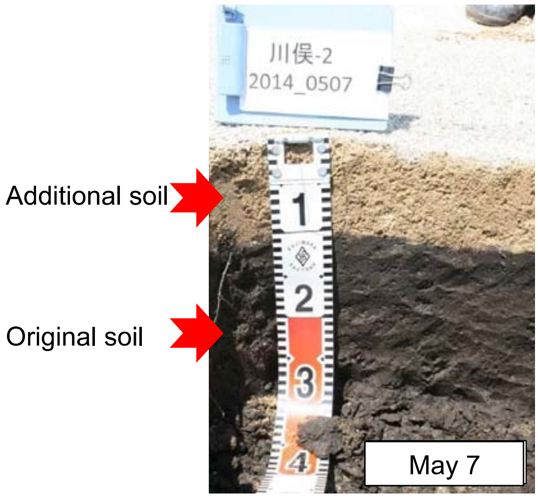

Fig. 14 Additional soil dressed after top soil stripping. (Iitate village, 2014).

with additional soil as shown in Fig. 14. The required amount of additional soil is vast and they were taken from the surrounding mountainous area. The chemical properties of the additional soil is very poor especially in total nitrogen, humus, exchangeable K and CEC (data not shown). Furthermore, decontamination by top soil stripping is preceded the initiation of agriculture, which means that even decontamination is finished, management of the field does not start immediately. Then weed invasion and erosion (especially in the upland field) are becoming another problem to be solved in these areas and cover crop introduction has been proposed and actual trial started.

Though applying decontamination and mitigation seems successfully improved the situation of food contamination and field radiation dose rate, the radioactive Cs contamination still trouble the farmers in the suffered area. To help them, we will further try to exclude the possibility of contamination of radioactive Cs from the environment to the food.

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