MINIMIZING THE CONTAMINATION OF AGRICULTURAL ENVIRONMENT TOWARD FOOD SAFETY – WITH PRIMARY FOCUS ON THE FUKUSHIMA NUCLEAR DISASTER

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ABSTRACT

Radioactive materials released during the accident at the Fukushima Daiichi nuclear power station affected farmlands in Fukushima and its neighboring prefectures, contaminating soil and agricultural products. Soon after the accident, high concentrations of radionuclides were detected in cultivated green leafy vegetables and fresh raw milk, caused by the direct deposition of radioactive materials. Shipment of crops from affected areas was restricted. After the early phase of direct contamination, the potential contamination of rice through uptake of radioactive cesium through plant roots from contaminated soil became a concern. Based on the monitoring data since 1959, rice planting was restricted in fields where contamination levels were higher than 5,000 Bq/kg soil. The transfer of radioactive cesium from soil to crops is affected not only by its concentration in soil but also by soil type and soil management practices such as fertilizer application and tillage. In order to estimate the actual contamination conditions for farmland soil, a map showing the concentration distribution of radioactive cesium in farmland soil was created. A long-term problem is soil contamination by radioactive cesium, particularly by 137Cs with a half-life of 30 years. Farming in highly contaminated areas requires active decontamination practices. Depending on the soil conditions, the application of potassium fertilizers and the addition of adsorbents are effective for inhibiting any further uptake of radioactive cesium by plants. While 2011 was an extremely difficult year for agriculture, 2012 will be a year of large-scale recovery; although there is no doubt that a lot of efforts are necessary, which will continue for many years to come.

Keywords: Radiocesium, Fallout, Deposition, Transfer factor, Chernobyl, Fukushima, Decontamination, Food safety

INTRODUCTION

Globally, consumer concerns about the safety of foods are on the rise. Much has been done to reduce hazardous chemicals, such as heavy metals and persistent organic pollutants (POPs), in foods to as low levels as reasonably possible. The accident at the Fukushima Dai-ichi nuclear power plant of Tokyo Electric Power Company (TEPCO) posed another serious problem for food safety.

On March 11, 2011, there was a big earthquake off the eastern coast of Japan. The resulting tsunami devastated the coastline with the tragic loss of 20,000 lives. The Fukushima Dai-ichi nuclear power plant was struck by the tsunami, leading to one of the most severe accidents in the history of nuclear industry, which was categorized as a major accident (level 7) of the International Nuclear and Radiological Event Scale (INES). A large amount of radionuclides were released into the atmosphere and contaminated the environment. An area of 20 km radius from the nuclear power plant and other heavily contaminated area nearby were designated an “Exclusion Zone”, forcing more than 100,000 people to evacuate. Radioactive materials contaminated farmlands in Fukushima and its neighboring prefectures affecting soil and agricultural products, which caused consumer concerns about contamination of foods with radionuclides to reach fever pitch. More than
8,000 ha of farmlands in Fukushima prefecture have been prevented from being cultivated due to heavy soil contamination with radionuclides in 2011. Some agricultural products exceeded the provisional regulation level of radionuclides in foods were banned for shipment from Fukushima and neighboring prefectures. Thus the agricultural sector was hit hard by this accident.

This paper seeks to review the impact of the recent accident to agriculture and discuss future prospects, one year on from the accident.

Artificial radionuclides in the environment

From the experience of the Chernobyl accident, it is known that a variety of different radionuclides are released in various forms during a nuclear power plant accident, most of which have short half-life and/or are volatile. The radionuclides which are most hazardous for the environment and agriculture are iodine-131(\(^{131}\)I), cesium-134(\(^{134}\)Cs), cesium-137(\(^{137}\)Cs), and strontium-90(\(^{90}\)Sr) (Table 1). Immediately after the accident, \(^{131}\)I is the most serious concern since it is released and deposited in large quantities. Once \(^{131}\)I is deposited, it disappears quickly due to its short half-life (8 days). After that, radioactive cesium remains as the major contaminant – \(^{134}\)Cs disappears early (half-life 2 years), while \(^{137}\)Cs remains for a long time because of its long half-life (30 years). \(^{90}\)Sr also exert a serious influence on agriculture and human health. However, the concentration of \(^{90}\)Sr in contaminated soil relative to that of radiocesium from the Fukushima accident is much lower than that from Chernobyl accident or global fallout. It is, therefore, considered that \(^{90}\)Sr is not a contaminant of major concern for the Fukushima accident.

To consider the impact of the Fukushima nuclear plant accident on the environment and on agriculture, estimates of baseline radioactive nuclides deposition in agricultural environment before the accident are necessary.

Prior to the Fukushima accident, farmlands in Japan have been subjected to radionuclides deposition. After the first atmospheric nuclear weapon tests in 1945, a large number of tests were conducted by USA, UK, and USSR, which were discontinued by the end of 1962. After the signing of the test ban treaty by these three countries in 1963, only a small number of such tests have been conducted by China and France, with the last explosion occurring in 1980. A total of 598 explosions took place up to 1980. The total explosive yield has been estimated to amount to 427.9 Mt, 57% of which was concentrated in a 16-month period from September 1961. Thermonuclear tests were frequently conducted at high altitude, with sufficient energy to inject most of the debris into the stratosphere, spreading the radionuclides all over the world. (Wright et al., 1999).

As over 90% of test explosions have been carried out in the northern hemisphere, most radioactive deposition has occurred there, since there is little mixing between the two hemispheres. Contamination of soil with radiocesium (\(^{137}\)Cs) from global fallout is still present all over the world and poses a problem of food contamination in some areas.

Concerns about contamination of foods with radionuclides became serious among consumers in the 1950s, which triggered the monitoring of artificial radionuclides in the environment. In Japan, activity concentrations of radioactive cesium and strontium in soil and cereals have been monitored at 17 monitoring sites throughout Japan since 1959. Changes in \(^{137}\)Cs activity concentration of paddy soil, brown rice and white rice have been monitored over time (Komamura et al., 2005).

Average values of the monitoring sites are shown in Fig. 1. The highest average values were observed in 1963 and 1964, where \(^{137}\)Cs concentration of soil and brown rice are 42.7 Bq/kg

<table>
<thead>
<tr>
<th>Physical half life</th>
<th>Amount released at the chernobyl accident (PBq)</th>
</tr>
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<tbody>
<tr>
<td>Iodine-131((^{131})I)</td>
<td>8.04 d</td>
</tr>
<tr>
<td>Cesium-134((^{134})Cs)</td>
<td>2.06 a</td>
</tr>
<tr>
<td>Cesium-137((^{137})Cs)</td>
<td>30.00 a</td>
</tr>
<tr>
<td>Strontium-90((^{90})Sr)</td>
<td>29.12 a</td>
</tr>
</tbody>
</table>

1PBq=1000TBq=1×10\(^{15}\)Bq
and 11.5 Bq/kg, respectively. The average value of $^{137}$Cs concentration has decreased consistently, and comes to one-tenth and one-hundredth of the peak, in soil and brown rice, respectively. Although the concentration of $^{90}$Sr is one-tenth of that of $^{137}$Cs, the fluctuation in concentration of $^{90}$Sr showed a similar tendency to that of $^{137}$Cs. 

These results can be used as background to estimate the deposition of radionuclides from the Fukushima nuclear power plant.

**Fukushima nuclear power plant accident and release of radionuclides**

The Fukushima Dai-ichi nuclear power station has 6 reactors. On March 11, 2011, 3 of them (reactors 1, 2 and 3) were in operation and the other 3 (reactors 4, 5, and 6) were undergoing routine maintenance and were not operating.

At 2:46 pm, the magnitude 9.0 earthquake rocked Fukushima Dai-ichi nuclear power station, nuclear reactors 1, 2, and 3 were automatically shut down by the tremor. At 3:27 and 3:35 pm, a 14-meter tsunami, unleashed by the earthquake, came over the seawall designed to protect the plant from a tsunami of 5.7 meters, flooding the facility and disabling the backup diesel generators, and washed away their fuel tanks. This resulted in the total loss of cooling system of the nuclear reactors. Meltdown, a nuclear reactor accident that results in core damage from overheating, was considered to have occurred during the day.

On March 12, there was a massive explosion in the outer structure of unit 1. The concrete building surrounding the steel reactor vessel collapsed as a result of the explosion. However, no damage is believed to have been caused to the reactor itself. An explosion occurred subsequently at unit 3 and 4, on March 14, and 15, respectively.

The total release of $^{137}$Cs is estimated to be 13 PBq (13x10$^{15}$ Bq) or 15% of the total from Chernobyl. For a few days after the accident, the direction of the wind around the nuclear power plant was from west to east, carrying radioactive materials to the ocean. From 15 to 16 March, the wind shifted; northeasterly, northerly, and southeasterly winds transported radioactive materials to inland areas. Furthermore, rainfall occurred in the north-west area of the nuclear power plant, resulting in a heavy ground deposition of radionuclides in the area.

From 20 to 23 March, a transient cyclone passed over Japan. Rain fell in a large area including Tokyo and surrounding regions, causing effective deposition of radioactive materials in a large area. As a result, farmland and agricultural products were contaminated.

Most of the land surface deposition occurred two weeks after the accident. The factors that influenced the deposition of radionuclides were the amount of radionuclides released, the direction of the wind, and the levels of precipitation.

![Graph showing dynamics of $^{137}$Cs and $^{90}$Sr in soil and brown rice in Japan](image)

**Fig. 1.** Dynamics of $^{137}$Cs (left) and $^{90}$Sr (right) activity concentration of paddy field soil and rice in Japan
Map of contamination levels of agricultural land

As was revealed by the Chernobyl nuclear plant accident, contamination levels do not always decline in a gradual linear way with geographical distance from the accident site, and the levels in some spots are found to be high (hot spot). It is usually necessary to get information about the actual contamination conditions of farmland soil in order to develop and implement countermeasures to reduce adverse effects of contaminated radionuclides. The concentration of radioactive cesium in soil was measured at 580 locations in Fukushima and neighboring prefectures (3,500 locations were added later) to create a map showing the concentration distribution of radioactive cesium in farmland soil (Fig. 2).

Since correlations were found between radiocesium activity concentration of soil and air dose rate of 1 meter above ground, the data of gamma radiation dose in air in the area were also used to create a map. Correlation between radiocesium concentration of soil and air dose rate varied depending on soil type (andosol or non-andosol) and type of agricultural land use (paddy field or upland field).

It was observed that contamination was greatly affected by wind direction and rainfall immediately after the accident and was distributed unevenly, not in a concentric fashion. The highest radiocesium contamination areas, exceeding 25,000 Bq/kg soil, were distributed in the 20 km radius area (exclusion zone) and in the northwest area from the power plant (extended exclusion zone). The actual distribution pattern of hot spots is complicated. In the middle part of Fukushima prefecture, the levels of contamination in most of the agricultural land are higher than that of the surrounding area (exceeding 1,000 Bq/kg soil).

From these results, the area of farmland with radiocesium exceeding 5,000 Bq/kg soil and between 1,000 and 5,000 Bq/kg soil is estimated to be more than 8,000 ha and 53,000 ha, respectively.

Contamination of agricultural products by direct fallout and deposition

Soon after the Fukushima nuclear plant accident, contamination of agricultural products with
radionuclides became a serious public concern. In response, the government set provisional guideline levels for radionuclides in foods on March 17. The values of the provisional guideline are comparable to those of EU and USA (Table 2).

Germanium semiconductor detectors of γ-ray spectra are used for the accurate measurement of concentrations of radioactive cesium and strontium. Since the semiconductor detectors were available in the National Institute for Agro-Environmental Sciences (NIAES), we implemented monitoring of radionuclides concentration of soil and foods in Fukushima and neighboring prefectures. High concentrations of radionuclides (such as $^{131}$I, $^{134}$Cs, and $^{137}$Cs), which exceeded the provisional regulation values for the radionuclides were immediately detected particularly in cultivated leafy vegetables (such as spinach), flowerhead brassicas (such as broccoli), and fresh raw milk in the area. High concentrations of radioiodine ($^{131}$I) (10 times higher than that of radiocesium) were found. Food from areas where contaminated foods exceeding the provisional regulation level were found were not shipped or exported from those areas. These served to increase consumer concern about food safety.

Contamination of agricultural products with radionuclides is brought about either through fallout and direct deposition to plants or through uptake by plant roots from contaminated soil (Fig. 3). The contaminations of foods observed soon after the accident, mentioned above, were caused by the fallout and direct deposition of radionuclides.

The contamination of vegetables by radionuclides with concentrations exceeding provisional regulation values has gradually decreased over time and has not been reported since June 2011. This is due to the short physical half-life of $^{131}$I deposited and a reduction in radioactive emissions from the damaged nuclear power station. In addition milk contamination (exceeding regulation level of $^{131}$I) has not been reported since July 2011. Thus the effect of direct deposition seems to have been diminished.

However in May 2011, tea leaves, bamboo shoots, and tree fruits such as yuzu (Chinese lemon), Japanese apricot, and Japanese persimmon, were found with radioactive cesium levels exceeding the provisional regulation level. Since fruits are major agricultural products of Fukushima prefecture, the impact on agriculture in the area was enormous. This contamination has been caused by the direct surface deposition of radionuclides on leaves and stems, which were absorbed and subsequently transferred to edible part of the plants.

Contamination of beef with radiocesium happened unexpectedly. In some area of Tohoku district, rice straw is cut down and spread on drained paddy fields after harvest. Radiocesium, released by the accident, was deposited on the rice

Table 2. Provisional guideline levels (Bq/kg) for radionuclides in foods*

<table>
<thead>
<tr>
<th></th>
<th>Japan</th>
<th>EU</th>
<th>USA</th>
</tr>
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<tbody>
<tr>
<td><strong>Iodine $^{131}$I</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Drinking water</td>
<td>300</td>
<td>500 (infant)</td>
<td>170</td>
</tr>
<tr>
<td>Milk, dietary products</td>
<td>100 (infant)</td>
<td>150 (infant)</td>
<td></td>
</tr>
<tr>
<td>Vegetables(exc. root vege)</td>
<td>2000</td>
<td>2000</td>
<td></td>
</tr>
<tr>
<td><strong>Cesium $^{134}$Cs</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Drinking water</td>
<td>200</td>
<td>10*</td>
<td>1200</td>
</tr>
<tr>
<td>Milk, dietary products</td>
<td>50*</td>
<td>400 (infant)</td>
<td></td>
</tr>
<tr>
<td>Vegetables</td>
<td>500</td>
<td>100*</td>
<td></td>
</tr>
<tr>
<td>Crops</td>
<td>1250</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Meat, egg, fish</td>
<td></td>
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</table>

*Levels of new guide line applied from April 1, 2012
straw that had been laid on the paddy field from the previous fall. In April 2011, after the accident, farmers collected the straw and fed them to cows without knowing that the straw was contaminated. The beef of cows fed with these contaminated straw was found to have very high levels of radiocesium, by the radiological survey of foods. By that time, some of the contaminated beef had already been distributed to outlets.

These contaminations accelerated the concern of consumers since they could not be forecasted beforehand.

**Contamination by uptake through plant roots from soil**

While contamination of agricultural products by direct deposition of radionuclides after the accident had reduced, the potential contamination of foods by uptake of radioactive cesium from soil through plant roots became a concern. In particular, the possibility of rice contamination, a staple food of Japan, was worrying.

To predict the radionuclide transfer from soil to plant, soil-to-plant transfer factor (TF) is useful (Uchida et al, 2009).

\[
TF = \frac{\text{plant concentration (Bq/kg)}}{\text{soil concentration (Bq/kg)}}
\]

TF for rice was estimated to be 0.1 using the long-term monitoring data collected from paddy fields throughout Japan since 1959, which consists of more than 400 datasets. Since the provisional guideline level of radiocesium content for brown rice is 500 Bq/kg, rice planting was restricted in fields where contamination levels were equal to, or higher than 5,000 Bq/kg, which will assure the production of safe rice with radiocesium concentration below 500 Bq/kg.

Concentration of radioactive cesium in brown rice in Fukushima prefecture harvested last fall was low in most cases. However in some cases, the concentration exceeded the provisional regulation values.

Studies have been conducted to elucidate the mechanism that brought about radiocesium contamination of rice. Paddy fields, from which contaminated rice were harvested, are located close to the exclusion zone with a high radiocesium...
concentration in the soil. Furthermore, the transfer of radioactive cesium from soil to crops is also affected by the soil type and by soil management practices used such as fertilizer application and tillage. Potassium is known to be a competitor with cesium for uptake by the plant. Although more than enough fertilizers are generally applied to agricultural land in Japan, some farmers have reduced the application of potassium fertilizer to paddy fields, in the belief that potassium fertilizer might have an adverse effect on the taste of rice. In some cases, highly contaminated rice was harvested from fields with low exchangeable $K_2O$ concentrations of soil. For these fields, application of potassium has been shown to be effective in reducing radiocesium contamination of rice.

**Agriculture countermeasures to reduce the radiological impacts**

Countermeasures consist of protective actions at the emergency stage and remedial actions at the post-emergency stage. They are taken to reduce the level of exposure as much possible. For the Chernobyl accident, a wide range of countermeasures were used, from urgent evacuation of the inhabitants from the area of highest radioactive contamination to long-term monitoring of radionuclides in foodstuffs in many European countries.

**International and national radiological criteria:**

Most of these environmental countermeasures are driven by relevant international and national radiological criteria. The International Commission on Radiation Protection (ICRP) is an international academic society of experts that makes recommendations about radiation protection. The ICRP’s 2007 recommendations state that in emergencies, radiation control standards different from the ones for normal times should be used. Moreover, it is suggested that emergency situations should be classified into two phases: an emergency phase and a rehabilitation phase, and the following protection measures are a guideline (Fig. 4).

1) Normal time: the dose should be limited to 1 mSv or less in a year.
2) Emergency phase: the exposure dose attributable to an accident should not exceed 20-100 mSv.
3) Rehabilitation phase after the accident settles down: the dose should not exceed 1-20 mSv in a year.

In the rehabilitation phase (recovery phase), during which operations such as decontamination will be continued to improve the situation, a reference level in the lower part of the 1-20 mSv/year band is recommended.

A reference level of exposure considered “normal” is appropriate in the long-term. Where
doses are kept below these levels, there should be no excessive health concerns. Similarly, eating foods with radionuclide concentration below internationally acceptable levels should be considered safe.

Based on the epidemiological data of Hiroshima and Nagasaki, the Food Safety Commission of Japan (FSCJ) concluded that more than 100 mSv cumulative dose of radiation during a lifetime could increase health risk to a person. FSCJ also mentioned that even if people are exposed to more than “around 100 mSv” of the extra cumulative exposure, it will not necessarily mean they will have adverse health effects. Based on these studies, together with the recommendations of ICRP, new regulation levels for radionuclides in foods were determined and implemented from April 2012 (Table 2).

Behavior of radiocesium in soil: Radionuclides deposited in soil causes internal and external radiation doses. In order to clean up the contaminated soil, information about behavior and distribution of radiocesium is essential. The radioecological behavior of radiocesium is influenced by the amount and nature of clay minerals present in soil (Sawhney, 1972). Selective sorption and fixation of Cesium ion (Cs+) by 2:1 type clay mineral, such as vermiculite and illite, is known to occur, because the cesium ion fits in the cavity of the silicate tetrahedral sheet of the clay mineral. Thus, radiocesium deposited in soil becomes fixed firmly by clay minerals in a short time, decreasing the soluble and exchangeable form of radiocesium in soil. Hence, in contaminated soil rich in clay minerals, uptake of radiocesium by plants is very low. To the contrary, radiocesium deposited in sandy soil or in organic soil such as peat, is more mobile, and plants grown in these soils are susceptible to contamination (Fig. 5).

Remediation of contaminated soil: The fact that radiocesium enters soil by deposition is fixed at the surface of the soil and moves downward to lower layers only gradually, indicates that contaminated soil can be cleaned up by removing the surface layer of the soil.

In 2011, the following three methods were used to test their effectiveness to remediate heavily contaminated fields soils in Fukushima prefecture’s lidate village, located in an extended nuclear evacuation zone (Fig. 6).

1) Top soil removal: The top 2-5 cm soil in which radiocesium is concentrated was removed. Surface soil was crushed by a power hollow, and collected by tractor equipped with an ear blade. This method generates highly contaminated waste soil.

2) Separation of clay fraction: This method is used in paddy fields. Under waterlogged condition, the surface is agitated and suspended. Soil suspension was transferred to “ponds” or large vessels for precipitation, and left there. Sediments were collected as contaminated waste and the supernatant, almost free from contaminants, was discarded. The amount of waste soil generated was less than that by simple top soil removal.

3) Soil turning tillage (skim and burial): Contaminated topsoil and non-contaminated subsurface soil was mixed. Highly contaminated waste soil was not generated. However, soil fertility was
disrupted because the fertile topsoil (plow layer) was buried deeper.

The results of these tests were promising, with a 70 to 95% reduction of radiocesium concentration in soil. The effectiveness varied depending on soil type and soil conditions. Larger-scale experiments will be carried out later this year. However, waste soil, generated with high-level contamination of cesium, will become another problem.

Guideline for remediation techniques, by the Ministry of Agriculture, Forestry, and Fisheries Japan (MAFF) is shown (Table 3). Recommended options depend on the level of soil contamination, and proactive remediation is recommended for soils with radiocesium concentration exceeding 5,000 Bq/kg.

Similar methods were tested for the remediation of radiocesium contaminated soil for Chernobyl.
accident as a pilot scale testing. However, these methods were not used for practical purpose for the Chernobyl accident because of its high cost and impact on soil fertility.

CONCLUSION

Future prospects

The Fukushima nuclear power plant accident occurred suddenly, and exerted a massive influence on various aspects of Japanese life. Although there have been neither acute deaths due to radiation nor evident radiation health damage, many people have concerns about long-term health effects of living in the affected areas. Apart from direct health effects from radiation, social, psychological, and economic effects of the accident have been enormous and will be around for a long time.

As was seen in the Chernobyl accident, the agricultural sector is the area of the economy worst hit by the effects of an accident of this kind. Many incidences of food contamination have been reported during the last year, some of which occurred in unexpected ways. This raised consumer concerns about food safety. Countermeasures have been taken to protect food safety, and knowledge has been accumulated about mechanisms of food contamination. Soil contamination by radioactive cesium, particularly by 137Cs with a half-life of 30 years, represents a long-term problem for Japan. From the experience of the Chernobyl accident, the contamination of agricultural products through root uptake can be expected to decrease over the course of the next few years since the accident. Continued efforts are needed to develop and promote agricultural products that can be produced safely in radionuclide contaminated soils. So far, obligatory radiological monitoring of agricultural products in Fukushima and surrounding regions of the affected area this year has shown that the radiocesium content of agricultural products is below the new guideline level.

Concerted efforts for decontamination of heavily contaminated agricultural land soils area are resulting in significantly improved living and agricultural conditions there. Work is underway within the evacuated area for people to prepare to return to their homes. However, some areas with air dose rates exceeding 50 mSv/h will remain unsettled for a long period. People from there cannot yet contemplate returning home yet.

Radioesium concentration exceeding national guideline levels has been found in forest food products eaten by rural farmers such as edible wild grasses, bamboo shoots, mushrooms, and wild boar. The contamination is due to the persistent recycling of radiocesium, particularly in forest ecosystems. These high levels of radionuclides in forests are expected to persist for a couple of decades.

Considering the long half-life of 137CS, it is important to understand the long-term behavior of radioactive materials by monitoring the contamination due to the accident.

While 2011 was an extremely difficult year for agriculture, 2012 will be a year of large-scale recovery; although there is no doubt that a lot of efforts are necessary, which will continue for many years to come.

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