Dynamic Analysis of Radioactive Cesium in Decontaminated Paddy Fields

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ABSTRACT
Radioactive contamination resulting from the Tokyo Electric Power Company Fukushima Daiichi Nuclear Power Station (FDNPS) disaster caused by the Great East Japan earthquake and tsunami on March 11, 2011, has affected large areas of land in Fukushima, Japan. At present, agriculture has been limited to some areas, and there are concerns that radioactive substances carried by rainwater might contaminate water bodies such as ponds and rivers. In order to resume safe agricultural activities, evaluating the dynamics of radioactive substances in agricultural water is essential. In this study, we measured the concentration and analyzed the impact of radioactive cesium in irrigation water on rice in five districts having limited residential population. Further, we analyzed the radioactive cesium balance and soil samples from decontaminated paddy fields. The main findings of the study are (1) radioactive cesium in agricultural water was mainly in suspended form at the experimental locations within the 40 km zone; (2) most of the radioactive cesium in the irrigation water was accumulated in the paddy field, but radioactive cesium in irrigation water had limited impact on brown rice cultivation; and (3) continual monitoring of areas with radiation levels higher than those recorded in this study is recommended.

Keywords: decontamination, irrigation management, radioactive substances

INTRODUCTION
A vast geographical area in Fukushima, Japan, was contaminated with large amounts of radioactive substances following the Tokyo Electric Power Company Fukushima Daiichi Nuclear Power Station (FDNPS) disaster caused by the Great East Japan earthquake and tsunami on March 11, 2011 (Chino et al., 2011). Various techniques such as surface soil removal, turning and plowing, and rotary tilling of soil were developed to decontaminate paddy fields (Naka et al., 2012). Nonetheless, the risk of recontamination by radioactive substances still exists because of the inflowing irrigation water (Kubota et al., 2013).

The distribution and behavior of radioactive materials released from the 1986 Chernobyl Nuclear Power Plant accident have been investigated extensively at farmland, grassland, and forest sites (IAEA, 2006). Unlike Chernobyl, which is a continental inland region with 30-year average precipitation of 600 mm, Fukushima has a 30-year precipitation of about 1,200 mm. Further, 6 km area in the west of the FDNPS in the Abukuma highland at an altitude of 500 – 700 m has been completely destroyed (JMA, 2015).
There are many paddy fields in Fukushima. Following the FDNPS disaster, agriculture has been limited to some areas, and there are concerns that water bodies such as ponds and rivers, which are resources of agricultural water might become contaminated by radioactive substances that are carried by rainwater (Shin et al., 2013; Kubota et al., 2014). For ensuring that agricultural activities are resumed in the affected areas, evaluating the dynamics of radioactive substances in irrigation water and reducing the inflow of these substances from the environment into paddy fields or canal systems are essential.

Particulate and dissolved forms of radioactive cesium deposited on land are known to gradually migrate via rainfall into water bodies, such as mountain streams and rivers (Yasutaka et al., 2013). Radioactive cesium in agricultural water exists in the form of dissolved ions and in particulate, suspended form, which is adsorbed by or fixed onto suspended organic and inorganic particles (MAFF, 2014a). Crops do not directly absorb radioactive cesium in the suspended form but can readily absorb it in the dissolved form that is present in the water used to irrigate paddy fields (MAFF, 2014b).

However, it has been suggested that the contamination of brown rice mainly occurs from fallout deposited on the organic matter covering the paddy fields and not via irrigation water (Shiozawa, 2012). This means that the impact of radioactive cesium in irrigation water flowing into paddy fields is not clear. Therefore, investigation of the impact of radioactive contamination in areas that are considered unsafe is necessary before agricultural activities can be resumed.

This study measured the concentrations of each form of radioactive cesium in agricultural water from five districts with limited residential population, that are being considered for resuming cultivation and compared the concentrations with the surveyed air radiation dose of each district. We analyzed the balance, including of the in- and outflow of radioactive cesium in agricultural water from Aug to Sep and determined its concentration in the soil of a paddy field (about 467 m²) located in district A.

**MATERIALS AND METHODS**

**Study area**

The government of Japan established four zones in Fukushima after the FDNPS disaster on April 1, 2013 (Reconstruction Agency, 2013). The study area included decontaminated paddy fields and agricultural water in district A (difficult-to-return zone); districts B and C (restricted residence zone) in Iitate, Fukushima; and districts D and E (deliberately evacuated zone) in Kawamata, Fukushima, Japan. The paddy fields were located about 40 km away from the FDNPS (Fig. 1). District A receives irrigation water from a pond, whereas the other districts receive water from rivers (Table 1). Rice was cultivated on a trial basis at each location in 2013.
Fig. 1 - Sampling locations in Fukushima, Japan.

Table 1 - The air radiation dose [μSv/h] and number of water samples.

<table>
<thead>
<tr>
<th>Survey date of air radiation dose</th>
<th>District</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Difficult-to-return zone</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Restricted residence zone</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Deliberately evacuated zone</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water source</td>
<td>Pond</td>
<td>3.62</td>
<td>1.32</td>
<td>1.66</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>River</td>
<td></td>
<td></td>
<td></td>
<td>0.83</td>
<td>0.69</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>4.08</td>
<td>1.38</td>
<td>1.44</td>
<td>0.78</td>
<td>0.66</td>
</tr>
</tbody>
</table>

Number of samples in 2013 (unit: pieces)

<table>
<thead>
<tr>
<th>Monitoring period</th>
<th>Jun 10 to Oct 7</th>
<th>Jul 17 to Oct 8</th>
<th>Jul 5 to Sep 11</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raw water</td>
<td>20 (5)*1</td>
<td>6</td>
<td>4</td>
</tr>
<tr>
<td>Dissolved form</td>
<td>7 (2)*1</td>
<td>3</td>
<td>3</td>
</tr>
</tbody>
</table>

*1 This value is number of samples from outlet.
**Survey and analysis method**

We measured air radiation dose by a survey meter (TCS-172; Hitachi Aloka Medical, Ltd. Japan) and periodically sampled agricultural water from the five districts in 2013. Table 1 shows the air radiation dose and the number of samples from each district. For analysis, 20 L water samples were passed through a 0.45 μm filter over a few days, and the filtrate was collected into beakers; the filtrate was then analyzed using the evaporative concentration method by placing the beakers on a hotplate at 180°C in a draft chamber until the volume reduced to 2 L. Temperature of the sample was about 80°C. Since the concentration of radioactive cesium in the dissolved form was low in many cases, the total volume of the filtrate as well as some volume of raw water was analyzed using the evaporative concentration method as a pre-treatment; this was considered as the dissolved form of radioactive cesium. Radioactive substances in the water sample, including all forms of radioactive cesium ($^{134}$Cs and $^{137}$Cs), raw water, and concentrated filtrate for dissolved form were analyzed using germanium semiconductor radiation detectors (GC4020-7500SL or GC2520-7500SL; Canberra Co. Ltd., USA) and a 2 L marinelli vessel (Fig. 2).

The in- and outflow of radioactive cesium in a paddy field of district A were quantified by measuring the inlet and outlet flows, respectively, as well as the water level in the paddy field by using a weir (Fig. 3). Soil core samples (5 cm in diameter) were collected at 2.5 cm depth from 46 sampling points from the paddy field at district A after rice was harvested. The samples were dried at 105°C, and the radioactive cesium concentrations of different fractions were measured by sieving the soil samples through a 2-mm screen. Radioactive substances in the soil samples, including all forms of radioactive cesium ($^{134}$Cs and $^{137}$Cs), were analyzed using the germanium semiconductor radiation detectors and a U8 vessel (Fig. 2).

We analyzed radioactive cesium in the lower limit of quantification of the radiation detectors that soil, raw water and dissolved form were 10 Bq/kg, 0.1 Bq/L and 0.01 Bq/L respectively. For all the samples, we measured to an accuracy of 10% of the relative standard deviation, or more precisely; we applied attenuation compensation using the day of sampling for the samples.
RESULTS AND DISCUSSION

Analysis of radioactive cesium concentration in irrigation water

The concentrations of radioactive cesium in raw water and in the dissolved form in agricultural water were measured (Fig. 4). The average concentration in raw water was 0.5 – 2.0 Bq/L and in the dissolved form was 0.01 – 0.07 Bq/L. The standard
Radioactive cesium concentration permitted in agricultural water has not yet been defined in Japan; however, the standard limits of radioactive cesium in drinking water is 10 Bq/kg for Japan (MHLW, 2012). Therefore, the concentrations found in the samples were not considered to be high.

In this study, the relative amount of dissolved form of radioactive cesium to raw water was lower in the pond than in the river water; this was possibly because of the absorption of dissolved radioactive cesium by soil particles that flowed into the pond. The ratio of radioactive cesium concentration in the dissolved form to that in raw water was 2 – 14%, suggesting that radioactive cesium in environmental water was mainly derived from suspended solids (FFPRI, 2012; Shin et al., 2015).

In the districts where the air radiation dose was high, the concentration of total radioactive cesium in the irrigation water was also high. However, in districts B and C—where the air radiation dose was low—the concentration of radioactive cesium in the dissolved form was higher than that in district A, where the concentration of total radioactive cesium and air radiation dose were both elevated (Figs. 5 and 6). This means that there is a possibility to be affected dissolved radioactive cesium of agricultural water in the air radiation dose even in low areas.

![Graph showing concentration of radioactive cesium in irrigation water of the study districts and its ratio.]

Fig. 4 - Concentration of radioactive cesium in irrigation water of the study districts and its ratio.
Changes in the concentration of radioactive cesium in agricultural water

The concentration of radioactive cesium in irrigation water was high in district A (5.6 Bq/L) during mid-September (Fig. 7); this was likely due to a decrease in the water level and disturbance of the farm pond for easy water management during this period. The total radioactive cesium concentration in agricultural water varied considerably; however, within a given district, there was little change in the concentration of radioactive cesium in the dissolved form. Fluctuations in the concentration of the suspended form were mainly attributed to rainfall events.
In general, the concentration of radioactive cesium in the outlets of drainage water was lower than that in the inlets of irrigation water. This finding suggests that radioactive cesium from irrigation water might have accumulated in paddy fields.

**Quantitative analysis of radioactive cesium**

We calculated the volume of intake from the inlet by using a water gauge, and converted the level of water during the relevant period by dividing the volume with the size of paddy field. Further, we calculated the transfer of radioactive cesium from value to multiply the level of water by concentration of radioactive cesium. The inflow and outflow of radioactive cesium on the basis of water balance from Aug 3 to Sep 25 in a paddy field in district A is shown in Table 2. The water balance analyzed in the two months because there was missing the data from being behind in the equipment installation.

The amount of inflow of radioactive cesium by irrigation water was lesser than the deposition amount because of the scattering of radioactive cesium of 3,000 kBq/m² or more during the disaster from results of the third airborne monitoring by MEXT (MEXT, 2011). Therefore, the effect of radioactive cesium in irrigation water on rice cultivation in this paddy field was considerable less during the irrigation period in 2013.
Table 2 - Estimated value of radioactive cesium balance in district A from Aug to Sep.

<table>
<thead>
<tr>
<th>Volume of intake</th>
<th>Level of water</th>
<th>Concentration of radioactive cesium</th>
<th>Transfer of radioactive cesium</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a) [m³]</td>
<td>(b) = (a) / 467 m²</td>
<td>(c) [Bq/L]</td>
<td>(d) = (b) × (c) [kBq/m²]</td>
</tr>
</tbody>
</table>

Inflow

<table>
<thead>
<tr>
<th>Period</th>
<th>Volume</th>
<th>Level</th>
<th>Concentration</th>
<th>Transfer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aug 3 to Aug 13</td>
<td>73.4</td>
<td>157</td>
<td>2.02</td>
<td>0.32</td>
</tr>
<tr>
<td>Aug 13 to Aug 19</td>
<td>122.0</td>
<td>261</td>
<td>1.94</td>
<td>0.51</td>
</tr>
<tr>
<td>Aug 19 to Aug 30</td>
<td>60.4</td>
<td>129</td>
<td>1.79</td>
<td>0.23</td>
</tr>
<tr>
<td>Aug 30 to Sep 25</td>
<td>34.8</td>
<td>75</td>
<td>1.43</td>
<td>0.11</td>
</tr>
<tr>
<td>(Total)</td>
<td>622</td>
<td></td>
<td>1.16</td>
<td></td>
</tr>
</tbody>
</table>

Outflow

<table>
<thead>
<tr>
<th>Period</th>
<th>Volume</th>
<th>Level</th>
<th>Concentration</th>
<th>Transfer</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Total)</td>
<td>173</td>
<td>1.32</td>
<td></td>
<td>0.23</td>
</tr>
</tbody>
</table>

Balance

<table>
<thead>
<tr>
<th>Period</th>
<th>Volume</th>
<th>Level</th>
<th>Concentration</th>
<th>Transfer</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Total)</td>
<td></td>
<td></td>
<td>0.93</td>
<td></td>
</tr>
</tbody>
</table>

*1 This value is the average radioactive cesium concentration in the outlet of raw water shown in Fig. 7 during the irrigation period in 2013.

Analysis of radioactive cesium concentration in the soil

The concentration of radioactive cesium in the soil core samples was high for only one sample that was obtained from a paddy field from immediately below the inlet; there were no major differences among the cesium concentrations recorded for the samples collected from other points (Fig. 8).

The effect of radioactive cesium present in the irrigation water could not be determined because of the large variation in its concentration in the paddy fields. Nonetheless, the concentration of radioactive cesium was lower in the irrigation water than in the soil after the paddy field was decontaminated.
CONCLUSIONS
This study analyzed the concentration of radioactive cesium in agricultural water and soil in paddy fields; these experiments were conducted for determining whether the paddy fields could be used for cultivation after decontamination, as well as the impact of such contamination on rice. Our study yielded several key findings.

1) District A had the highest concentration of cesium in raw water from irrigation water, whereas district C had the highest concentration of radioactive cesium in the dissolved form. At the experimental locations within the 40 km zone, radioactive cesium in the irrigation water was mainly in suspended form.

2) The concentration of radioactive cesium in raw water varied remarkably by location, but the concentration of the dissolved form was similar across the sampled areas.

3) Most of the radioactive cesium in the irrigation water flowing into the paddy field did not drain out, and was accumulated in the paddy field, but the impact of radioactive cesium in irrigation water on brown rice was minimal in the five districts where rice cultivation was tested.

4) The amount of inflow of radioactive cesium by irrigation water was lesser than the amount deposited by scattering of radioactive cesium at the time of the disaster.

This study was conducted in areas that have limited residential population; therefore the water management practice might have been different from that used in conventional farming. Since radioactive cesium in irrigation water had likely accumulated in paddy fields, continuous monitoring would be required in areas where radiation level was higher than the values measured in this study.
ACKNOWLEDGMENTS
This study was performed out as a research agenda of the “Movement monitoring of radioactive cesium in decontamination paddy” project commissioned by the Ministry of Agriculture, Forestry, and Fisheries Research. The authors express their gratitude to the Ministry of Agriculture, Forestry, and Fisheries.

REFERENCES


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