Development of Technologies for Removal of Radioactive Materials from Agricultural Soil in JAPAN

October 16, 2011

Ministry of Agriculture, Forestry and Fisheries
Purpose and Goals of the project

• We started the research project in Iitate Village and Kawamata Town from late May.

• When we started the project, the goal of decontamination was not established as Japanese government.

• Therefore, we planed the field trials with immediate goal to decrease the level of radioactive Cs in agricultural land to less than 5,000 Bq/kg, the level banning rice planting.

※ We have to contribute to long term goal of overall decontamination, 1 mSv/year. It is established goal.

• We clarify the suitable methods to remove radioactive materials according to the level of radioactive Cs.
Relative frequency of translation factor of radioactive cesium from soil to rice grain observed in Japan (1959-2001, n=564)

Translation factors in the other reports

- *1 ほぼ試験（国内）、内田ら, 2008
- *2 ほぼ試験（台湾）、J.-J. Wangら, 1998
- *3 ポット試験（国内）、津村ら, 1984
- *4 ポット試験（ハングラデシュ）、M.M.Rahmanら, 2005
- *5 ポット試験（ハングラデシュ）、A.S.Mollahら, 1998
Distribution Map of Radioactive Substances Concentration in Agricultural Land Soil in Fukushima Prefecture (Reference)

Legend

Research Point

Research point Surroundings (※1)

Radioactive cesium concentration in agricultural soil (Bq/kg) (※2)

- 25000<
- 10000—25000
- 5000—10000
- 1000—5000
- <1000
- Evacuation zone

※1: Radioactive cesium concentration in agricultural soil within zone surrounding research points were estimated based on air dose ratio data provided by the Ministry of Education, Culture, Sports, Science and Technology and/or Fukushima prefecture.

※2: In agricultural lands, radioactive cesium concentration in soil was measured using soil collected from 15 cm deep in paddy fields and maximum 30 cm deep in upland fields, taking into account the soil mixing by plowing or depth of plant roots.
Institutes Participated

◎ National Institutions
  • National Agriculture and Food Research Organization (NARO)
  • National Institute for Agro-Environmental Sciences (NIAES)
  • National Institute of Advanced Industrial Science and Technology (AIST),
  • National Institute for Materials Science (NIMS),
  • Japan Atomic Energy Agency (JAEA)

◎ Prefectural Agricultural Experiment Stations
  • Fukushima Agricultural Technology Centre
  • Other Prefectural Agricultural Experiment Stations

◎ Universities

◎ Private enterprises
  (ISEKI & CO., LTD., Dainichiseika Color & Chemicals Mfg. Co., Ltd. etc.)

  • 7 national institutions, 11 universities, 6 prefectural agricultural experiment stations, 1 incorporated foundation and 3 private enterprises have cooperated.
Outline of Demonstration Experiment for Development of Decontamination Technology of Agricultural Soil —

- Removal of topsoil using soil hardener
  Itamizawa, area: 10 a
  Radioactive cesium concentration: 9,100 Bq/kg

- Phytoremediation by high absorbing plants
  (Sunflower, Amaranthus)
  Nimaibashi, area: 0.5 a
  Radioactive cesium concentration: 7,700 Bq/kg

- Decontamination by paddling with water
  Itamizawa, area: 4.2 a
  Radioactive cesium concentration: 15,300 Bq/kg

- Scarping test of grass pasture
  Ittoi, area: 8 a
  Radioactive cesium concentration: 10,400 Bq/kg

- Sunflower combustion test
  Numadaira Clear center

- Topsoil removal
  (Paddy field)
  Ittoi, area: 8 a
  Radioactive cesium concentration: 13,600 Bq/kg

- Operation tests in other locations
  Plowing in (Upside-down)
  (Paddy field)
  Motomiya City, area: 28 a
  Radioactive cesium concentration: 4,100 Bq/kg

Radioactive cesium concentration is the sum of cesium134 and cesium137 (Bq/kg, dry soil)
Basic knowledge on agricultural soil contamination

Table 1: Extractive test of radioactive cesium ($^{134}\text{Cs} + ^{137}\text{Cs}$) from agricultural soil in Fukushima Prefecture

<table>
<thead>
<tr>
<th>Land category</th>
<th>Water extraction</th>
<th>Ammonium acetate extraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Paddy field soil</td>
<td>ND</td>
<td>2.3%</td>
</tr>
<tr>
<td>in Fukushima</td>
<td>ND</td>
<td>5.3%</td>
</tr>
</tbody>
</table>

Note) Detection limit (0.4 Bq/L)

Figure 1: Radioactive cesium concentration of paddy field soil in Itamizawa, Iitate village.

Table 2: Measurement of radioactive cesium ($^{134}\text{Cs} + ^{137}\text{Cs}$) concentration in water system around the paddy field in Iitate village experimental site.

<table>
<thead>
<tr>
<th>Water category</th>
<th>Concentration of radioactive substances</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water in farm pond</td>
<td>ND</td>
</tr>
<tr>
<td>Water in Nitta river</td>
<td>ND</td>
</tr>
<tr>
<td>Ground water</td>
<td>ND</td>
</tr>
<tr>
<td>Drain water</td>
<td>ND</td>
</tr>
</tbody>
</table>

Table 3: Result of measurement of radioactive cesium concentration per soil particle from surface soil (0-2.5 cm, Itamizawa), untreated sample

<table>
<thead>
<tr>
<th>Particle size</th>
<th>Percent (%)</th>
<th>Bq/kg</th>
<th>Bq/Total</th>
<th>Bq ratio (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clay</td>
<td>4.8</td>
<td>174,300</td>
<td>8,400</td>
<td>13.0</td>
</tr>
<tr>
<td>Silt</td>
<td>29.6</td>
<td>103,300</td>
<td>30,600</td>
<td>46.4</td>
</tr>
<tr>
<td>Fine sand</td>
<td>45.2</td>
<td>48,000</td>
<td>21,700</td>
<td>32.9</td>
</tr>
<tr>
<td>Coarse sand</td>
<td>20.4</td>
<td>25,900</td>
<td>5,280</td>
<td>8.0</td>
</tr>
</tbody>
</table>

Note) Radioactivity concentration of surface layer (0 to 2.5 cm) in total is 65,923 Bq/kg. Soil was sampled in June, 2011

Note) Detection limit (4～7 Bq/L)
Topsoil removal

- Pulverization by tractor equipped with a vertical harrow
  - Concentration of radioactive cesium within soil
    - 10,370 Bq/kg → 2,600 Bq/kg (reduction :75%)

- Scraping by tractor equipped with a rear blade instead
  - Air dose rate on the topsoil surface
    - 7.1 μSv/h → 3.4 μSv/h

- Disposal/ sandbagging by tractor equipped with a back hoe
  - Volume of disposed soil approximately 40t/10a
  - Time required for removal approximately 55~70min./10a

This method is applicable in agricultural land regardless of dose of radioactivity and land category.

Volume of disposed soil is larger.
Topsoil removal using soil hardener

- Spray of solution by dissolving magnesium hardener in water

- Hardener solution permeates and topsoil hardens (7 to 10 fine weather days)

- Concentration of radioactive cesium within soil
  9,090 Bq/kg → 1,671 Bq/kg (reduction: 82%)

- Air dose rate on the topsoil surface
  7.8 μSv/h → 3.6 μSv/h

- Volume of disposed soil approximately 30t/10a
  (less than topsoil removal without soil hardener)

- Removal of topsoil

  - Hardening of soil prevents scattering of soil dusts, so it is effective for safety of operator.
  - Topsoil was colored white, so it facilitates visual checking omission of removing.
  - Cost of hardener is approximately 170,000 yen/10a.
  - No harmful effect to soil after decontamination.
  - The same result is expected when polyion or molecular polymer is used.
Removal of turf and grasspasture

- Concentration of radioactive cesium in soil
  13,630 Bq/kg → 327 Bq/kg
  (reduction : 97%; scrape thickness 3cm)

- Volume of soil to dispose (including glass)
  approximately 40t/10a

- Operation time
  250 minute / 10a (approximately)

Decontamination efficiency is high, it is possible combining weeding and decontamination as one task.

This method is applicable in a turf, grasspasture and weed where root matt is well grown.
Removal of soil after paddling with water (Paddy field)

Paddle soil
(surface layer soil)

- Radioactive cesium concentration (dry soil)
  15,254 Bq/kg → 9,689 Bq/kg
  (reduction: 36%)

- Surface dose rate
  7.6 μSv/h → 6.5 μSv/h

- Volume of soil to dispose
  about 1.2～1.5t/10a

- Concentration of radioactive cesium in top layer clear water is below detection limit

  Reduction of concentration of radioactive cesium within soil differed from types of soil and in a range between 30% and 70%.

  Desirable results are not expected where clay content in the soil is small.

  The advantage of this method can reduce the soil wastes after decontamination.
Inverting Plowing

**Plow with a jointer (Plowing depth 30 cm, tow by a tractor)**

**Modified 2 layered plow (Plowing depth 45 cm, tow by a tractor)**

**Two layered plow (Plowing depth 60 cm, tow by a D6 bulldozer)**

Topsoil was buried in the layer of 25 to 40 cm deep.

Topsoil was buried in the layer of 40 to 60 cm deep.

### Depth distribution of radioactive cesium after plowing in (30 cm)

- **Plowing in (30 cm)**
  - Surface dose rate: $0.7 \text{ \mu Sv/h} \rightarrow 0.3 \text{ \mu Sv/h}$
  - Soil to dispose: no
  - Operation time: 30 minutes / 10a

Before operation, risk assessment of ground water contamination and assessment of soil are essential.
Phytoremediation by high absorbing plants

- Concentration of radioactive cesium at stem and leaf
  52 Bq/kg (fresh weight)
  $\rightarrow$ approximately 520 Bq/m$^2$

- Concentration of radioactive cesium of soil
  7,715 Bq/kg $\rightarrow$ 1,068,000 Bq/m$^2$

- Absorb from soil approximately 1/2000

Sunflower is not practical for decontamination technology as quite long time to decontamination.

Sunflower is one of the crops to absorb radioactive cesium. (Institute for Environmental sciences. [http://www.ies.or.jp/japanese/research/seikaH21pdf/3000_H21.pdf])

Measure quantity of absorption substantially in upland field of Iitate village (radioactive cesium with soil : 7,700 Bq/kg)

Burn test of plant remains of sunflower including radioactive cesium
Scheme for applying the methodology for decontamination of radioactive cesium from agricultural field

For the moment, it is purposed to reduce the level of agricultural land of 5,000 Bq/kg or above to less than that (estimated, paddy field: 6,300 ha, upland field: 2,000 ha).

Note) ● denotes method producing disposal soil while ○ denotes method without disposal.

<table>
<thead>
<tr>
<th>Radioactive cesium concentration within soil</th>
<th>Upland field</th>
<th>Paddy field</th>
</tr>
</thead>
<tbody>
<tr>
<td>To 5,000 Bq/kg</td>
<td>From the point of view to reduce uptake of radioactive cesium by crops as much as possible, as well as reducing air dose rate, ○ plowing or ○ cultivation technology to reduce uptake is applied as appropriate.</td>
<td></td>
</tr>
<tr>
<td><strong>5,000 Bq/kg to 10,000 Bq/kg</strong></td>
<td>In low case (value shall be studied) ● Topsoil removal ○ Plowing</td>
<td>In high case (value shall be studied) ● Topsoil removal ○ Plowing</td>
</tr>
<tr>
<td><strong>10,000 Bq/kg to 25,000 Bq/kg</strong></td>
<td>● Topsoil removal</td>
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</tr>
<tr>
<td><strong>25,000 Bq/kg to</strong></td>
<td>● Topsoil removal Removal thickness of 5 cm or deeper. Provided that study is required for operational technology in high dose conditions. (For example, use of soil hardener for antiscattering of soil dust)</td>
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<table>
<thead>
<tr>
<th>Ground water level</th>
<th>Soil condition assessment / Ground water level</th>
</tr>
</thead>
<tbody>
<tr>
<td>5,000 Bq/kg to 10,000 Bq/kg</td>
<td>Lowland soil ● Topsoil removal ○ Removal of soil after paddling with water ○ Plowing (Plow layer may disturb)</td>
</tr>
<tr>
<td><strong>10,000 Bq/kg to 25,000 Bq/kg</strong></td>
<td>Other than lowland soil ● Topsoil removal ○ Removal of soil after paddling with water (Less effective when compared with lowland soil) ○ Plowing (Plow layer may disturb) (Only applicable when ground water level is low)</td>
</tr>
<tr>
<td><strong>25,000 Bq/kg to</strong></td>
<td>● Topsoil removal Removal thickness of 5 cm or deeper. Provided that study is required for operational technology in high dose conditions. (For example, use of soil hardener for antiscattering of soil dust)</td>
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Treatment of disposed soil
(temporary custody by portable concrete containers)

Trial concrete containers
• External dimension: 1.5 m by 1.5 m by 1.5 m
• Wall thickness: 15 cm
• Capacity: 1.6m$^3$
• Weight: 4.2 t

Flexible container bag with soil inside a container

Surface dose rate
2.9 $\mu$Sv/h $\rightarrow$ 0.29 $\mu$Sv/h (reduction: 91%)

During operation tests, radioactivity intercepting portable concrete containers were used for temporary custody and found to be competent.

Technologies for desorbing and/or removing of radioactive cesium from disposed soil shall be continuously researched.
Remaining Research Subjects

Researches shall be continuously conducted, to develop technologies to;

• Isolation of radioactive cesium from disposed soil.
• Remove radioactive materials from farmland neighboring facilities such as dike and drain.
• Reduce concentration of radioactive cesium in agricultural products by improving cultivation methods.
• Safer methods of removing radioactive materials.
• Recovery of soil fertility after removal of top soil or inverting plowing.