Behavior of radioactive cesium accumulation in water bodies in contaminated area by FDNPP accident

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High Cs concentration in the bottom sediment are detected in water bodies in Fukushima and the surroundings area.

Cs moves from originally contaminated areas to cleaner downstream water bodies.

Accumulated bottom areas become a long-term source of contamination.

Concern over increase in the risk of exposure through the activities such as water use, fishery and recreation.

Background
Purpose

- Understanding migration properties of radioactive cesium in river basin scale
- Understanding actual condition of cesium accumulation into inflowing water bodies
- Evaluating the trend of sediment contamination in water bodies by flows & stocks analysis
Study area

- Lake Kasumigaura Basin (2,157km²) as a mildly-contaminated area
  - Shallow eutrophied lake with long retention time (200 days).
  - Still blocking shipment for some fishes

- Uda River Basin (106.3km²) as a heavily-contaminated area
  - Highly contaminated mountainous upper region compared to lowland region
  - Dam (Matugabou dam) controlling water flow in the basin
  - Large coastal lagoon (Matsukawaura Lagoon) as an accumulation place

$^{137}$Cs deposition from an airborne monitoring survey on Nov. 11th, 2011
Survey against 7 main inflow rivers

- Measuring flow rates, turbidities and SS conc. to calculate SS flux after the accident.
- Sampling river waters during rain runoff events
- Measuring $^{137}$Cs activities associated with SS and dissolved

Sediment survey:
- Temporal change analysis by stationary sampled core at three points
- Measuring $^{137}$Cs accumulation at 68 points by core sampling on Dec., 2012 and Oct., 2013

Runoff survey:
- Observatory of flow rate and turbidity
- Sediment core sampling point

$^{137}$Cs accumulation in basin (Bq·m⁻²)

- <2,500
- 2,500 – 5,000
- 5,000 – 10,000
- 10,000 – 20,000
- 20,000 – 30,000
- 30,000 – 40,000
- 40,000 – 50,000
- 50,000 <


**137Cs flows & stocks survey in Uda River Basin**

**Matugabou Dam**
(Lake Udagawa)

**Discharge & turbidity survey point**

**Runoff survey**
- 137Cs runoff from forested area
- 137Cs outflow flux from the upstream dam
- 137Cs flux and runoff ratio from the whole basin

**Sediment survey**
- 137Cs accumulation amount and profile in the bottoms of upstream dam
- Longitudinal variation of 137Cs activity in the river-bed sediment
- Spatial distribution and total deposition of 137Cs in the lagoon
Behavior of radioactive Cs in the Lake Kasumigaura Basin
Relationship between $^{137}$Cs activity associated with SS and SS concentration

$^{137}$Cs activity in SS (Bq/g-SS) vs. SS (mg/L)

- Sakura R.
- Seimei R.
- Koise R.
- Ono R.
- Sonobe R.
- Tomoe R.
- Hokota R.

Relationship between $^{137}$Cs activity associated with SS and deposition amount in catchment

$^{137}$Cs activity in SS (Bq/g-SS) vs. $^{137}$Cs deposition amount (kBq/m²)

- Sakura R.
- Seimei R.
- Ono R.
- Sonobe R.
- Tomoe R.
- Hokota R.
- Kose R.

$^{137}$Cs activity associated with SS
→ depending on not conc. of SS but initial deposition amount in catchment
**137Cs runoff from main inflow rivers’ catchments of Lake Kasumigaura**

Estimated runoff volume of 137Cs associated with suspended solid from main inflow rivers’ catchments for two years after FDNPP accident

<table>
<thead>
<tr>
<th>River name</th>
<th>Sakura</th>
<th>Koisé</th>
<th>Ono</th>
<th>Seimei</th>
<th>Sonobe</th>
<th>Tomoe</th>
<th>Hokota</th>
</tr>
</thead>
<tbody>
<tr>
<td>Land use (%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Forest</td>
<td>34.6</td>
<td>48.6</td>
<td>18.2</td>
<td>22.7</td>
<td>14.2</td>
<td>21.6</td>
<td>24.2</td>
</tr>
<tr>
<td>Farmland</td>
<td>16.9</td>
<td>18.7</td>
<td>32.8</td>
<td>29.1</td>
<td>41.0</td>
<td>46.9</td>
<td>53.9</td>
</tr>
<tr>
<td>Paddy</td>
<td>28.1</td>
<td>21.6</td>
<td>14.0</td>
<td>20.0</td>
<td>13.7</td>
<td>15.9</td>
<td>11.3</td>
</tr>
<tr>
<td>Urban</td>
<td>8.8</td>
<td>11.1</td>
<td>15.2</td>
<td>9.8</td>
<td>10.2</td>
<td>5.1</td>
<td>3.2</td>
</tr>
<tr>
<td>SS specific runoff volume (kg/m²)</td>
<td>73.8</td>
<td>61.6</td>
<td>32.2</td>
<td>58.3</td>
<td>57.2</td>
<td>47.3</td>
<td>36.3</td>
</tr>
<tr>
<td>137Cs specific runoff volume (kBq/m²)</td>
<td>0.017</td>
<td>0.046</td>
<td>0.080</td>
<td>0.276</td>
<td>0.061</td>
<td>0.041</td>
<td>0.042</td>
</tr>
<tr>
<td>137Cs deposition amount (kBq/m²)</td>
<td>5.00</td>
<td>12.5</td>
<td>26.7</td>
<td>33.5</td>
<td>11.7</td>
<td>11.7</td>
<td>17.4</td>
</tr>
<tr>
<td>137Cs runoff ratio (%)</td>
<td>0.34</td>
<td>0.37</td>
<td>0.30</td>
<td>0.83</td>
<td>0.52</td>
<td>0.35</td>
<td>0.24</td>
</tr>
</tbody>
</table>

137Cs slightly run off in catchment scale
Change in spatial distribution of $^{137}$Cs accumulation in sediment of the Lake Kasumigaura

Estimated spatial distributed map of $^{137}$Cs accumulation in sediment by spline function using activities of sediment cores at 68 points

December, 2012

October, 2013

- Higher activities in the western side $\rightarrow$ Effect of initial direct deposition
- Locally high activities at some river mouths $\rightarrow$ Effect of inflow from the river
- Change in $^{137}$Cs accumulation $\rightarrow$ Slightly increase?, Promotion of vertical mixing

18 kBq/m$^2$ (to 15cm depth)

15 kBq/m$^2$ (to 15cm depth)

19 kBq/m$^2$ (to 25cm depth)

Cs-$^{137}$accumulation (kBq $\cdot$ m$^{-2}$)

- <2.5
- 2.5 - 5.0
- 5.0 - 10
- 10 - 20
- 30 - 40
- 40 - 50
- 50 <
Analysis of stocks and flows of $^{137}\text{Cs}$ in the Kasumigaura Basin in 21 months after the accident

- Deposition over the basin’s entire land area
  - $14\text{kBq/m}^2 (19\text{TBq})$

- Direct deposition on the lake surface
  - $\approx 10\text{kBq/m}^2$

- Inflow of dissolved $^{137}\text{Cs}$ from land
  - $0.12\text{kBq/m}^2 (0.17\text{TBq})$
  - [Deposition $\times$ annual runoff ratio (0.5%) $\times 21/12$]

- Accumulation in the sediment
  - $18\text{kBq/m}^2 (2.9\text{TBq})$

- Accumulation in the sediment per unit area derived from land
  - $1.0\text{kBq/m}^2$

- Outflow from lake
  - Unknown but small

- Small contribution of $^{137}\text{Cs}$ associated with SS both in the past and future?
- Large effect of direct deposition on the lake surface and the runoff of radioactive Cs from impervious soon after deposition on the accumulation in the sediment
Temporal change of dissolved $^{137}$Cs in the Lake Kasumigaura

Increase of Cs concentration in summer → effect of $\text{NH}_4^+$ production in sediment?
Effect of sediment environment on production of dissolved Cs

Relation of NH$_4^+$ concentration in the lake bottom water to distribution condition of $^{137}$Cs between sediment surface layer and lake water

- Marked growth and decomposition of algae
- Generation of reduction condition and NH$_4^+$ in lake sediment
- Promotion of Cs solubilization from solid phase

Concern about the effect on Cs transition to aquatic ecosystem and agricultural food
Same phenomena possibly occurring in eutrophied lakes and ponds in Fukushima prefecture.
Behavior of radioactive Cs in the Uda River Basin
\[^{137}\text{Cs}}\text{ runoff from forested area in the Uda River Basin}

- Cs associated with SS is main component of runoff (Kd > 10^5)
- Slight decrease in Cs concentration from 2012 to 2013
- Very limited runoff of Cs even in highly contaminated region

**Relationship between \[^{137}\text{Cs}}\text{ activity associated with SS and SS concentration**

**Runoff volume and ratio of \[^{137}\text{Cs}}\text{ associated with SS**

<table>
<thead>
<tr>
<th></th>
<th>Catchment No.1</th>
<th>Catchment No.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>[^{137}\text{Cs}}\text{ deposition (kBq/m^2)}]</td>
<td>170</td>
<td>230</td>
</tr>
<tr>
<td>Conc. of [^{137}\text{Cs}}\text{ associated with SS (kBq/kg)}]</td>
<td>11±4.6</td>
<td>18±6.3</td>
</tr>
<tr>
<td>Runoff volume of [^{137}\text{Cs}}\text{ associated with SS (kBq/m^2)}]</td>
<td>0.21</td>
<td>0.27</td>
</tr>
<tr>
<td>[^{137}\text{Cs}}\text{ runoff ratio (%)}]</td>
<td>0.12</td>
<td>0.12</td>
</tr>
</tbody>
</table>
Accumulation of $^{137}$Cs in sediment of dam lake

Spatial distribution of Cs accumulation
Behavior of inflow Cs associated with SS highly depending on flow property in the lake
Behavior in highly accumulated area
Current inflow sediment shielding initial Cs deposition layer

Matsugabou Dam (Lake Udagawa)

- 335kBq/m² (Oct., 2013)
- 485kBq/m² (Nov., 2012)
- 434kBq/m² (Nov., 2012)
- 703kBq/m² (Oct., 2013)

Direct deposition & initial inflow?
Typhoon Typhoon

Discharge control for reservation of irrigation water and flood prevention

\[ \text{retention ratio to inflow } ^{137}\text{Cs to dam lake} = \left( \frac{^{137}\text{Cs outflow} \times \text{SS outflow} \times ^{137}\text{Cs conc.}}{^{137}\text{Cs inflow} \times \text{(catchment deposition} \times \text{runoff ratio)}} \right) \]

\[ = \left( \frac{31 \times 10^3 \text{(kg)} \times 30,000 \text{(Bq/kg)}}{300,000 \text{(Bq/m}^2) \times 25.6 \times 10^6 \text{(m}^2) \times 0.0011} \right) = 0.9 \]

Discharge control for reservation of irrigation water and flood prevention

Surface layer intake

most inflow SS and Cs accumulating to bottom sediment

Role of dam in prevention of the proliferation of Cs to downstream area was confirmed
$^{137}$Cs runoff from the entire Uda River Basin

$^{137}$Cs associated with suspended solid (SS)

<table>
<thead>
<tr>
<th>SS (mg/L)</th>
<th>$^{137}$Cs activity (Bq/g-SS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>100</td>
<td>2</td>
</tr>
<tr>
<td>200</td>
<td>4</td>
</tr>
<tr>
<td>300</td>
<td>6</td>
</tr>
<tr>
<td>400</td>
<td>8</td>
</tr>
<tr>
<td>500</td>
<td>10</td>
</tr>
<tr>
<td>600</td>
<td>12</td>
</tr>
</tbody>
</table>

- **2012**
- **2013**

Observed river flow rate and SS load (Jul. 2012 to Jan. 2014)

$^{137}$Cs Runoff condition from the entire basin

- Total runoff amount associated with SS: 0.35kBq/m$^2$ (34GBq)
- $^{137}$Cs runoff ratio: 0.17\% (Jul. 2012 to Jan. 2014)

$^{137}$Cs runoff is currently limited from the whole basin as well as forested area.
Spatial distribution of rCs concentration profile in the bottom sediment of Matsukawaura Lagoon

134Cs + 137Cs (Bq/kg-sediment)

Sediment core sampling points

West
Mid-west
Mid-East
South-west
South-east

North

Sediment depth (cm)

West
North
Mid-west
Mid-East
South-west
South-east

Koizumi R.
Uda R.
Ume R.

Spatial distribution of 134Cs and 137Cs concentration profile in the bottom sediment of Matsukawaura Lagoon.
Relationship between sediment property and rCs concentration in the Matsukawaura Lagoon

Relationship between radioactive Cs concentration and organic and muddy contents in the surface layer (0-5 cm) at all sampling points.

\[ ^{134}\text{Cs} + ^{137}\text{Cs} (\text{Bq/kg DW}) \]

\[ R = 0.90 \]

\[ R = 0.81 \]

\[ \text{Ignition Loss, Muddy Content Rate (\%)} \]

\[ \text{MCR: ratio of fraction < 63\mu m to total content of sediment} \]

Distribution property of accumulated Cs

→ Multiple contribution by inflow amount and flow property in the lagoon.

Detailed survey and utilization of numerical simulation model are currently underway.
Spatial distribution of $^{137}$Cs accumulation on the bottom sediment of Matsukawaura Lagoon

Total accumulation of $^{137}$Cs to 20cm depth on July, 2013: 220GBq (29kBq/m$^2$)

Inflow amount of $^{137}$Cs associated with SS from the catchment (170km$^2$)

Total deposition on the catchment × annual runoff ratio × elapsed years after the accident

$= 68$GBq $\rightarrow 9.2$kBq/m$^2$ on the bottom sediment

Sediment inflow probably has the role in Cs accumulation in the lagoon.

- Reflecting the difference of contamination level between upstream and downstream area
- Outflow from the lagoon to coastal sea should be considered for more precise estimation.
Summary of behavior of radioactive cesium accumulation in water bodies

- Direct deposition and initial inflow from land strongly contributed to current Cs accumulation condition in water bodies.

- Further accumulation into water bodies in a cleaner region with highly contaminated upper region should be considered.

- Role of dam in the prevention of proliferation of Cs to downstream area was confirmed, but production of bioavailable Cs from accumulated Cs should be examined.
Thank you for your attention!