Radiocaesium mobility in the «soil-water» system – underlying processes. Looking at Fukushima from a Chernobyl perspective

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Outline

- Radiocaesium solid-liquid distribution in soil-water system;
- Radiocaesium wash-off and river transport;
- River flood plain processes: accumulation and loss;
- Irrigation ponds – critical water bodies?
- Monitored natural attenuation as a remediation option;
- Main messages
Mobility and bioavailability of radiocesium are determined by ratio of its chemical forms in fallout and site-specific environmental characteristics determining rates of leaching, fixation/remobilization as well as sorption-desorption of mobile fraction (its solid-liquid distribution).

Concept of exchangeable $K_d$

- The existing methods for estimating the distribution coefficient have a common drawback which is the absence of a clear division of radionuclide speciation based on their ability to exchange with the liquid phase.
- This problem can be resolved by using the concept of exchangeable form of the radionuclide (Konoplev and Bulgakov, 1999).
Selective sorption and fixation of radiocesium

High retention of radiocesium in soils is caused by two main processes: selective reversible sorption on illitic clay minerals and fixation

\[
FES - M + ^{137}Cs^+ \leftrightarrow K_c^{FES} (Cs / M) \rightarrow FES - ^{137}Cs + M^+
\]

\[
RIP^{ex}(M) = K_d^{ex}(Cs) \times m_M = K_c^{FES}(Cs / M) \times [FES]
\]

\[
K_d^{ex}(^{137}Cs) = \frac{RIP^{ex}(K)}{([K^+]_W + K_c (N / K)[NH_4^+]_W)}
\]

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Radiocesium \(K_d(L/g)\) in rivers. Chernobyl-Fukushima comparison

<table>
<thead>
<tr>
<th>River</th>
<th>Period</th>
<th>Min</th>
<th>Max</th>
<th>Median</th>
<th>Geom Mean</th>
<th>Arithm Mean</th>
<th>St Dev</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Chernobyl</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dneper</td>
<td>1987-1999 DB &quot;RUNOFF&quot;</td>
<td>4,1</td>
<td>165</td>
<td>29</td>
<td>28,3</td>
<td>38,7</td>
<td>29,6</td>
</tr>
<tr>
<td>Pripyat Belaya Soroka</td>
<td>1988-1999 DB &quot;RUNOFF&quot;</td>
<td>1,7</td>
<td>42,8</td>
<td>8,1</td>
<td>9,6</td>
<td>15,9</td>
<td>16,4</td>
</tr>
<tr>
<td>Pripyat Chernobyl</td>
<td>1986-1999 DB &quot;RUNOFF&quot;</td>
<td>2,6</td>
<td>176</td>
<td>25,2</td>
<td>24</td>
<td>34</td>
<td>30</td>
</tr>
<tr>
<td>Uzh Cherevach</td>
<td>1986-1990 DB &quot;RUNOFF&quot;</td>
<td>2,5</td>
<td>168</td>
<td>29</td>
<td>28,3</td>
<td>45,5</td>
<td>42</td>
</tr>
<tr>
<td><strong>Fukushima</strong></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Abukuma</td>
<td>2012-2013 Nanba, 2014</td>
<td>56,6</td>
<td>1660</td>
<td>564</td>
<td>561</td>
<td>665</td>
<td>352</td>
</tr>
<tr>
<td>Hiso River</td>
<td>2011 Ueda et al., 2013</td>
<td>92,4</td>
<td>955</td>
<td>139</td>
<td>179</td>
<td>230</td>
<td>226</td>
</tr>
<tr>
<td>Wariki River</td>
<td>2011 Ueda et al., 2013</td>
<td>252</td>
<td>923</td>
<td>367</td>
<td>439</td>
<td>484</td>
<td>229</td>
</tr>
</tbody>
</table>

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$K_d$ values in the surface waters of the Fukushima area are much higher those for the Chernobyl area (by 1-2 orders of magnitude).

This points to a high radiocesium binding capacity of sediments and soils in the Fukushima area and, as a result, a significant part of radiocesium on the area contaminated after the Fukushima accident are transported on suspended material with surface runoff and river flow.

River watersheds in Fukushima and Chernobyl

After Konoplev et al., 1988

After Evrard et al., 2012
Radionuclide wash-off from catchment - secondary contamination of rivers and lakes by surface run-off

- Wash-off coefficient $K_w$ is the portion of radionuclide deposition on the catchment washed-off with surface runoff to water bodies. Total wash-off coefficient is composed from “solid” and “liquid” wash-off coefficients $K_i$ and $K_s$:

$$K_i = \frac{\int_0^T C_w(t) V(t) dt}{\int_S C_S dS} = \frac{C_w V}{C_S S},$$

$$K_s = \frac{\int_0^T C_a(t) m dt}{\int_S C_S dS} = \frac{C_a M}{C_S S},$$

- Normalization of wash-off coefficients on runoff characteristics allows to predict radionuclide entering to water bodies in dissolved and adsorbed state.

$$K_i^* = \frac{K_i}{h} = \frac{K_i S}{V} = \frac{C_w}{C_S},$$

$$K_s^* = \frac{K_s}{g} = \frac{K_s S}{M} = \frac{C_a}{C_S},$$

where $h$ - runoff depth, mm; $g$ - mass of eroded material from the catchment.

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Experiments on runoff plots

Chernobyl 30-km zone

Fukushima Prefecture

After Konoplev et al., 1988

After Yoshimura et al., 2014
## Radiocesium wash-off from runoff plots

<table>
<thead>
<tr>
<th>Location</th>
<th>Date</th>
<th>Land use</th>
<th>$^{137}$Cs, kBq/ m²</th>
<th>$N_r$, mm⁻¹</th>
<th>$N_s$, m²/g</th>
<th>$K_{dr}$, L/ kg</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Chernobyl artificial raining</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Dovlyady</td>
<td>07.1986</td>
<td>Grassland</td>
<td>1310±170</td>
<td>$31 \times 10^5$</td>
<td>$10 \times 10^4$</td>
<td>$(16\pm9)\times 10^3$</td>
<td>Konoplev, 1998</td>
</tr>
<tr>
<td>Benevka</td>
<td>10.1986</td>
<td>Grassland</td>
<td>1540±280</td>
<td>$29 \times 10^4$</td>
<td>$2,6 \times 10^3$</td>
<td>$(174\pm10)\times 10^3$</td>
<td>Konoplev, 1998</td>
</tr>
<tr>
<td><strong>Chernobyl snowmelt</strong></td>
<td></td>
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</tr>
<tr>
<td>Dovlyady</td>
<td>03.1987</td>
<td>Grassland</td>
<td>1310±170</td>
<td>$0,9 \times 10^4$</td>
<td>$1,7 \times 10^3$</td>
<td>$(13\pm1,3)\times 10^3$</td>
<td>Konoplev, 1998</td>
</tr>
<tr>
<td>Benevka</td>
<td>03.1987</td>
<td>Grassland</td>
<td>1540±280</td>
<td>$1,9 \times 10^4$</td>
<td>$5,2 \times 10^3$</td>
<td>$(15\pm14)\times 10^3$</td>
<td>Konoplev, 1998</td>
</tr>
<tr>
<td>Kopachi</td>
<td>04.1987</td>
<td>Pine Forest</td>
<td>1650±450</td>
<td>$57 \times 10^4$</td>
<td>$32 \times 10^3$</td>
<td>$(173\pm3,4)\times 10^3$</td>
<td>Konoplev, 1998</td>
</tr>
<tr>
<td>Korogod</td>
<td>04.1987</td>
<td>Farmland</td>
<td>246±28</td>
<td>$7,2 \times 10^4$</td>
<td>$2,0 \times 10^3$</td>
<td>$(3,9\pm1,8)\times 10^3$</td>
<td>Konoplev, 1998</td>
</tr>
<tr>
<td><strong>Fukushima long-term runoff plots</strong></td>
<td></td>
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</tr>
<tr>
<td>Kawamata</td>
<td>2011-2013</td>
<td>Grassland</td>
<td>560</td>
<td>-</td>
<td>$2,1 \times 10^3$</td>
<td>-</td>
<td>Wakiyama et al., 2014</td>
</tr>
<tr>
<td>Kawamata</td>
<td>2011-2013</td>
<td>Cedar Forest</td>
<td>440</td>
<td>-</td>
<td>$14 \times 10^5$</td>
<td>-</td>
<td>Wakiyama et al., 2014</td>
</tr>
<tr>
<td>Kawamata</td>
<td>2011-2013</td>
<td>Farmland uncultivated</td>
<td>410</td>
<td>-</td>
<td>$5,3 \times 10^3$</td>
<td>-</td>
<td>Wakiyama et al., 2014</td>
</tr>
<tr>
<td>Kawamata</td>
<td>2012-2013</td>
<td>Farmland cultivated</td>
<td>410</td>
<td>-</td>
<td>$0,97 \times 10^3$</td>
<td>-</td>
<td>Wakiyama et al., 2014</td>
</tr>
</tbody>
</table>

## Radiocesium wash-off from river catchments

<table>
<thead>
<tr>
<th>River name</th>
<th>$^{137}$Cs deposition, kBq/ m²</th>
<th>$N_r$, mm⁻¹</th>
<th>$N_s$, m²/g</th>
<th>$K_{dr}$, L/ g</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fukushima, December 2012, Yoshimura et al., 2014</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ukedo river</td>
<td>2500</td>
<td>$1,3 \times 10^{-7}$</td>
<td>$2,2 \times 10^{-5}$</td>
<td>500</td>
</tr>
<tr>
<td>Niida river</td>
<td>900</td>
<td>$0,8 \times 10^{-7}$</td>
<td>$1,5 \times 10^{-5}$</td>
<td>200</td>
</tr>
<tr>
<td>Mano river</td>
<td>400</td>
<td>$0,8 \times 10^{-7}$</td>
<td>$1,6 \times 10^{-5}$</td>
<td>200</td>
</tr>
<tr>
<td>Abukuma river</td>
<td>120</td>
<td>$1,2 \times 10^{-7}$</td>
<td>$2,8 \times 10^{-5}$</td>
<td>600</td>
</tr>
<tr>
<td><strong>Chernobyl, January 1988, Konoplev et al., 2002</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sakhan river</td>
<td>2300</td>
<td>$30 \times 10^{-7}$</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Iput river</td>
<td>275</td>
<td>$50 \times 10^{-7}$</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Sozh river</td>
<td>250</td>
<td>$50 \times 10^{-7}$</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Dneper river</td>
<td>39</td>
<td>$76 \times 10^{-7}$</td>
<td>$17 \times 10^{-5}$</td>
<td>15</td>
</tr>
<tr>
<td>Pripyat river</td>
<td>31</td>
<td>$140 \times 10^{-7}$</td>
<td>$21 \times 10^{-5}$</td>
<td>32</td>
</tr>
</tbody>
</table>
Radiocesium wash-off from the catchments of Fukushima

- Normalized solid wash-off coefficients for Fukushima area very well corresponded to those of Chernobyl;
- Normalized liquid wash-off coefficients \( N_l(\text{mm}^{-1}) \) in the Fukushima NPP area are about an order of magnitude lower than those obtained in the Chernobyl area.
- It was found that the total wash-off coefficient of radiocesium from farmlands in Fukushima area can be high and reaches up to several \( \% \) per year (Yoshimura et al., 2014);
- Generally, high precipitation in the region and steep slopes promote higher wash-off of radiocesium as compared to the Chernobyl case.

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Map of annual precipitation, Japan

Annual precipitation in contaminated area is 1000-1600 mm, 2-3 times higher than in the Chernobyl zone (500-600 mm)!
Locations of monitoring points on Niida river floodplain

Niida river
Hiso river
Iitoi river

Grassmat
Suspended sediment time integrated sampler

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Map of radiocesium contamination of the Niida River catchment

Monitoring sites
1 - sub-catchment number

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Radiocesium vertical profiles in soils of Niida river flood plain

**SN-11**
- No accumulation

**SN-21**
- $S = 0.5 \text{ cm/a}$

**SN-31**
- $S = 3.3 \text{ cm/a}$

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**Irrigation ponds - critical water bodies?**
- **Suzuuchi** - irrigation cascade pond with own catchment
- **Kashiramori** - recreation pond with individual catchment
- **Sakashita rsv.** - general-purpose reservoir
- **Inkyozaka** - filling pond
- **Funasava** - town cascade pond in irrigation system
- **FDNPP**

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## Total $^{137}\text{Cs}$ activity concentrations in water bodies of Okuma town (March 2014)

<table>
<thead>
<tr>
<th>Location</th>
<th>Distance from FDNPP, km</th>
<th>Dose rate, μSv/h</th>
<th>Deposition $^{137}\text{Cs}$, kBq/m²</th>
<th>$^{137}\text{Cs}$, Bq/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>Suzuuchi</td>
<td>3.75</td>
<td>24</td>
<td>5840</td>
<td>59.4±0.9</td>
</tr>
<tr>
<td>Funasawa</td>
<td>3.50</td>
<td>3.25</td>
<td>-</td>
<td>9.3±0.2</td>
</tr>
<tr>
<td>Inkyozaka</td>
<td>0.24</td>
<td>11</td>
<td>4250</td>
<td>3.78±0.06</td>
</tr>
<tr>
<td>Kashiramori</td>
<td>7</td>
<td>3.3</td>
<td>865</td>
<td>0.747±0.003</td>
</tr>
<tr>
<td>Sakashita</td>
<td>8</td>
<td>3.0</td>
<td>672</td>
<td>0.125±0.013</td>
</tr>
</tbody>
</table>

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## Radiocesium vertical profiles in soils of Okuma town

### Suzuuchi SPS-02

- $^{137}\text{Cs}$, Bq/g
- $^{134}\text{Cs}$, Bq/g

### Inkyozaka SPI-02

- $^{137}\text{Cs}$, Bq/g
- $^{134}\text{Cs}$, Bq/g

### Sakashita

- $^{137}\text{Cs}$, Bq/g
- $^{134}\text{Cs}$, Bq/g

### Kashiramori

- $^{137}\text{Cs}$, Bq/g
- $^{134}\text{Cs}$, Bq/g

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Simplified diffusion model of radiocesium vertical migration

\[ Q(x) = \frac{Q_0}{\sqrt{\pi Dt}} \times e^{-\frac{x^2}{4Dt}} \]

- \( Q(x) \) – radiocesium content on depth \( x \) (Bq/m\(^2\)cm);
- \( Q_0 \) – radiocesium inventory in soil (Bq/m\(^2\));
- \( x \) – depth (cm);
- \( t \) – time after deposition;
- \( D \) – effective diffusion coefficient (cm\(^2\)/year)

Effective diffusion of radiocesium in soils of Okuma town

Suzuuchi soil profiles described by diffusion equation

\[ D_{eff} = 2.48 \text{ cm}^2/\text{year} \]
Effective diffusion of radiocesium in soils of Okuma town

<table>
<thead>
<tr>
<th>Location</th>
<th>Deposition, kBq/ m²</th>
<th>Soil type</th>
<th>$D_{\text{eff}}, \text{cm}^2/\text{year}$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fukushima</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Suzuuchi</td>
<td>5840</td>
<td>Sandy Fluvisols</td>
<td>2.48</td>
</tr>
<tr>
<td>Inkyozaka</td>
<td>4250</td>
<td>Sandy Fluvisols</td>
<td>2.24</td>
</tr>
<tr>
<td>Kashiramori</td>
<td>865</td>
<td>Sandy-loam Cambisols</td>
<td>9.27</td>
</tr>
<tr>
<td>Sakashita</td>
<td>672</td>
<td>Loam Inceptisols</td>
<td>5.0</td>
</tr>
<tr>
<td><strong>Chernobyl</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Benevka</td>
<td>1500</td>
<td>Alluvial sandy-loam</td>
<td>0.5</td>
</tr>
<tr>
<td>Chernobyl</td>
<td>750</td>
<td>Cultivated sandy-loam</td>
<td>0.6</td>
</tr>
</tbody>
</table>

- $D_{\text{eff}}$ for Fukushima soils are higher than for Chernobyl soils (Konoplev et al., 1990) – result of higher precipitation;
- Similarly to Chernobyl case migration rate in forest soil is essentially higher than in grassland and farmland.

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Monitored natural attenuation - an option for water remediation

- Natural attenuation, by definition, constitutes the least invasive approach to environmental remediation;
- Reliance on natural attenuation requires detailed characterization of the site and adequate monitoring owing to the evolution of natural systems with time;
- The purpose of MNA is to take advantage of natural processes that reduce the flux of a contaminant reaching any given receptor.
Conclusions

- Normalized solid wash-off coefficients for Fukushima area very well corresponded to those of Chernobyl;
- Normalized liquid wash-off coefficients $N_l$ ($\text{mm}^{-1}$) in the Fukushima NPP area are about an order of magnitude lower those obtained in the Chernobyl area.
- Generally, high precipitation in the region and steep slopes promote higher wash-off of radiocesium as compared to the Chernobyl case;

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Conclusions

- Irrigation ponds can be considered as a critical water bodies in terms of radiocesium mobility and bioavailability and should be surveyed before water application;
- $D_{\text{eff}}$ for Fukushima soils are higher than for Chernobyl soils;
- Similarly to Chernobyl case migration rate in forest soil is essentially higher than in grassland.

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Main messages

- For remediation strategy development and decision making the long-term wide scale (catchment-river-lake) monitoring program should be in place.
- Long-term monitoring program should include not only measurements of radionuclide activity concentrations (solution, solid etc.) but also their speciation and environmental characteristics determining radionuclide behavior such as RIP, competing ions concentrations etc.
- Information on radionuclide chemical forms, their transformation in other words mobility and bioavailability should be taken into account when rehabilitation and decontamination strategies are developed on local or regional scale.

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THANK YOU VERY MUCH FOR YOUR ATTENTION!

QUESTIONS?

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