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

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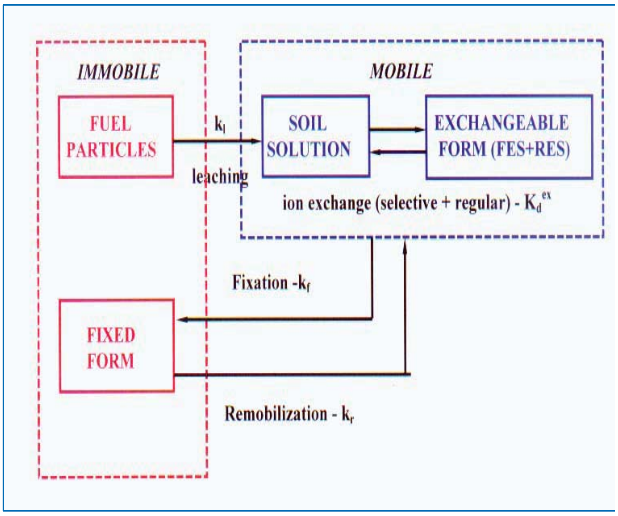
Outline

- Radiocesium solid-liquid distribution in soil-water system;
- Radiocesium 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

Speciation

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).

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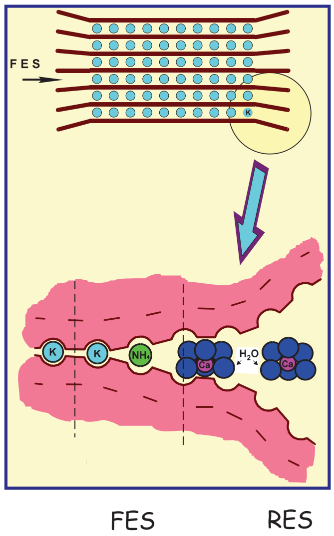
Selective sorption and fixation of radiocesium

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



$$FES-M+{}^{137}Cs^{+} \xleftrightarrow{K_c^{FES}(Cs/M)} FES-{}^{137}Cs+M^{+}$$

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

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

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

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

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Radiocesium solid-liquid distribution at Fukushima

- 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.

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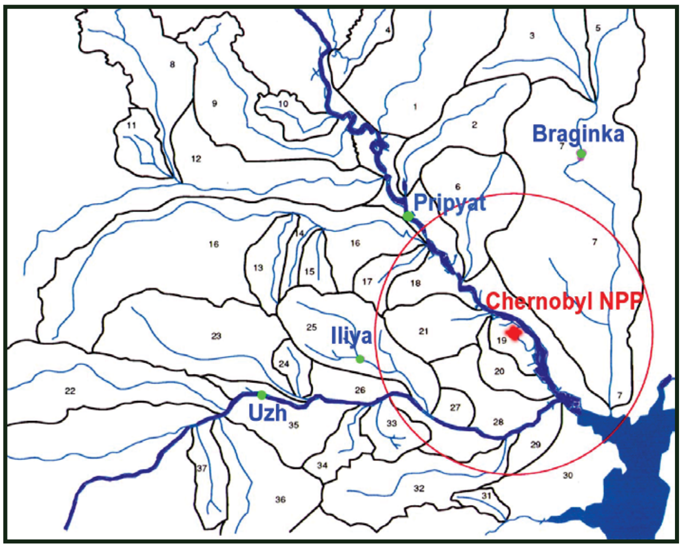
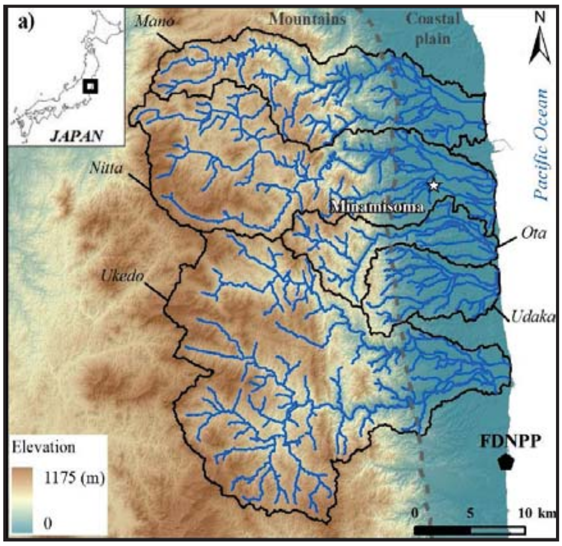
River watersheds in Fukushima and Chernobyl





After Konoplev et al., 1988

After Evrard et al., 2012

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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_l and K_s:

$$K_l = \frac{\int_O^T C_W(t)V(t)dt}{\int_S C_S\,dS} = \frac{\overline{C_W}}{\overline{\overline{C_S}}}\frac{V}{S},$$

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

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

$$K_l^* = \frac{K_l}{h} = \frac{K_l S}{V} = \frac{\overline{C_W}}{\overline{\overline{C_S}}},$$

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

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

After Konoplev et al., 1988

Fukushima Prefecture

After Yoshimura et al., 2014

Radiocesium wash-off from runoff plots

Location	Date	Land use	^{137}Cs, kBq/m^2	N_l, mm^{-1}	N_s, m^2/g	K_d, L/kg	References
Chernobyl artificial raining							
Dovlyady	07.1986	Grassland	1310∓170	31×10^{-6}	10×10^{-5}	$(16∓9)\times10^{3}$	Konoplev, 1998
Benevka	10.1986	Grassland	1540∓280	29×10^{-6}	$2,6\times10^{-5}$	$(17∓10)\times10^{3}$	Konoplev, 1998
Chernobyl snowmelt							
Dovlyady	03.1987	Grassland	1310∓170	$0,9\times10^{-6}$	$1,7\times10^{-5}$	$(13∓1,3)\times10^{3}$	Konoplev, 1998
Benevka	03.1987	Grassland	1540∓280	$1,9\times10^{-6}$	$5,2\times10^{-5}$	$(15∓14)\times10^{3}$	Konoplev, 1998
Kopachi	04.1987	Pine Forest	1650∓450	57×10^{-6}	32×10^{-5}	$(17∓3,4)\times10^{3}$	Konoplev, 1998
Korogod	04.1987	Farmland	246∓28	$7,2\times10^{-6}$	$2,0\times10^{-5}$	$(3,9∓1,8)\times10^{3}$	Konoplev, 1998
Fukushima long-term runoff plots							
Kawamata	2011-2013	Grassland	560	-	$2,1\times10^{-5}$	-	Wakiyama et al., 2014
Kawamata	2011-2013	Cedar Forest	440	-	14×10^{-5}	-	Wakiyama et al., 2014
Kawamata	2011-2013	Farmland uncultivated	410	-	$5,3\times10^{-5}$	-	Wakiyama et al., 2014
Kawamata	2012-2013	Farmland cultivated	410	-	$0,97\times10^{-5}$	-	Wakiyama et al., 2014

Radiocesium wash-off from river catchments

River name	^{137}Cs deposition, kBq/m^2	N_l, mm^{-1}	N_s, m^2/g	K_d, L/g
Fukushima, December 2012, Yoshimura et al., 2014				
Ukedo river	2500	1.3×10^{-7}	2.2×10^{-5}	500
Niida river	900	0.8×10^{-7}	1.5×10^{-5}	200
Mano river	400	0.8×10^{-7}	1.6×10^{-5}	200
Abukuma river	120	1.2×10^{-7}	2.8×10^{-5}	600
Chernobyl, January 1988, Konoplev et al., 2002				
Sakhan river	2300	30×10^{-7}	-	-
Iput river	275	50×10^{-7}	-	-
Sozh river	250	50×10^{-7}	-	-
Dneper river	39	76×10^{-7}	17×10^{-5}	15
Pripyat river	31	140×10^{-7}	21×10^{-5}	32

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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 (mm^{-1}) in the Fukushima NPP area are about an order of magnitude lower 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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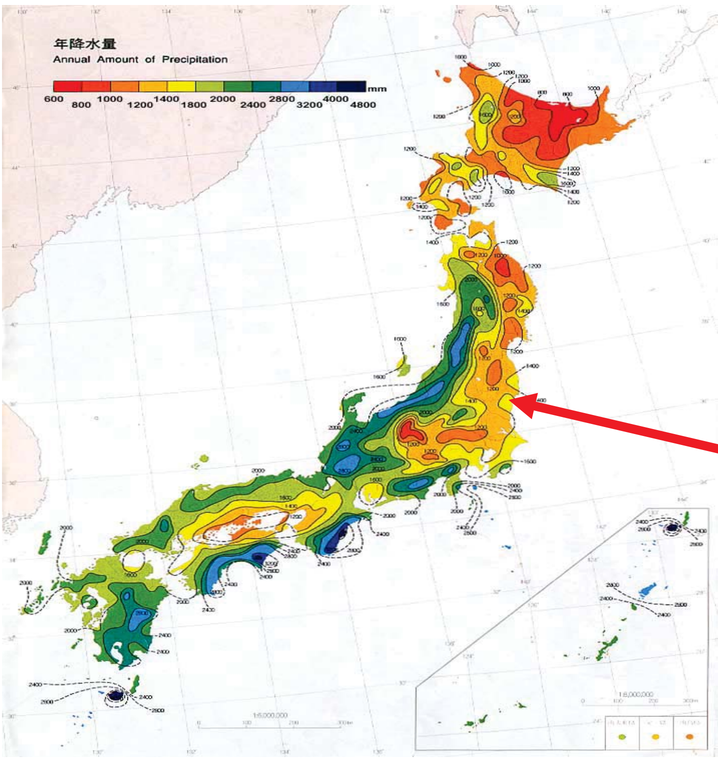
Map of annual precipitation, Japan



Annual precipitation in contaminated area is 1000-1600 mm, 2-3 times higher than in the Chernobyl zone (500-600 mm)!

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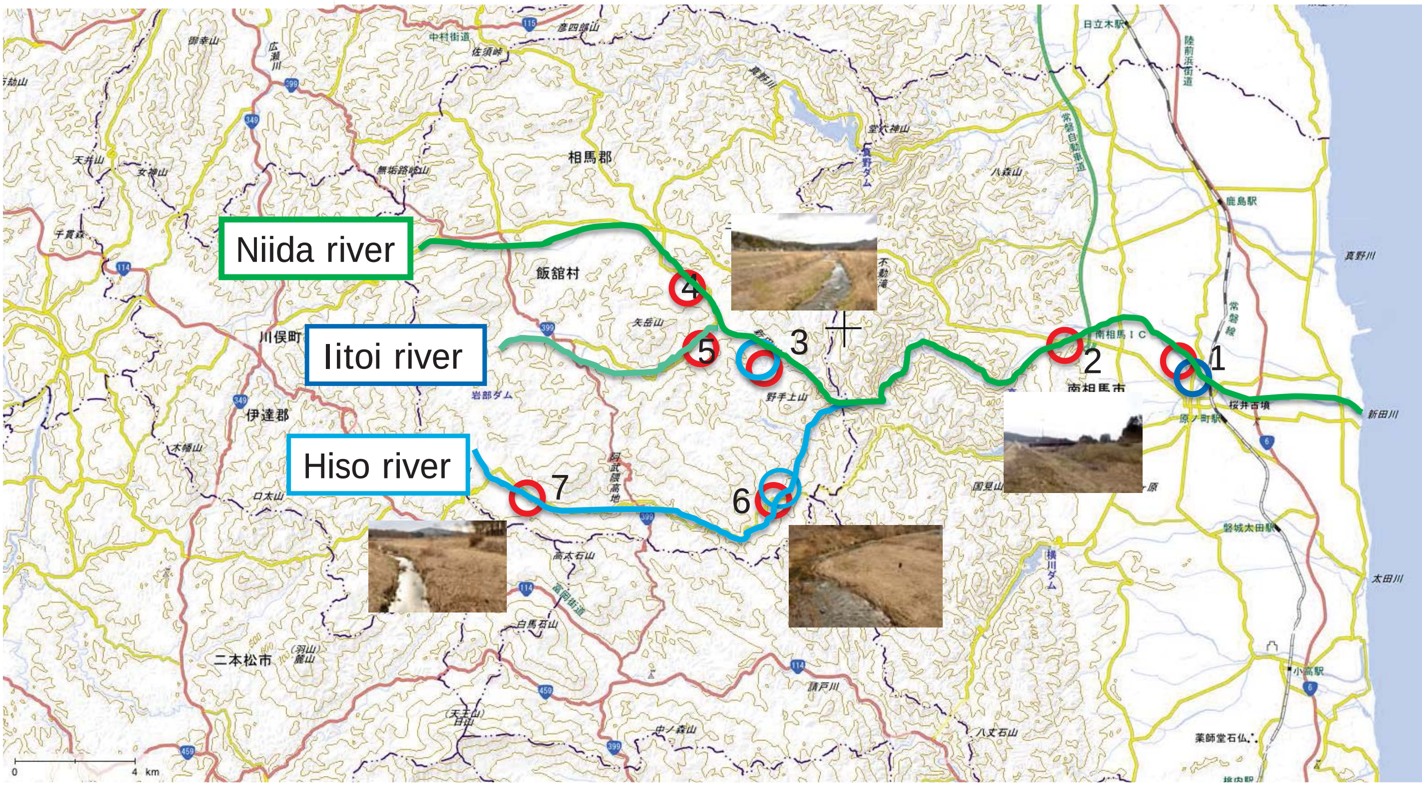
Locations of monitoring points on Niida river floodplain



Grassmat

Suspended sediment time integrated sampler

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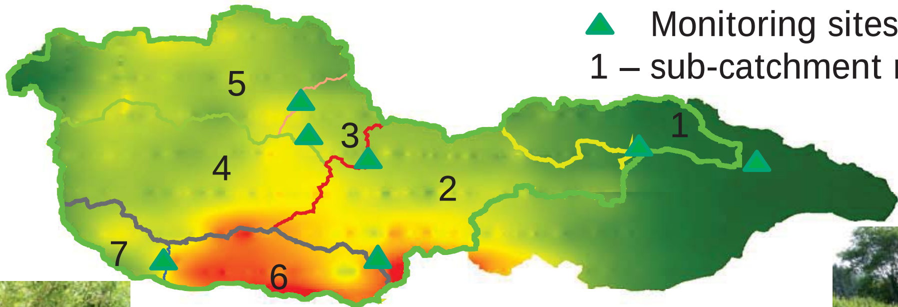
Map of radiocesium contamination of the Niida River catchment



Monitoring sites
1 – sub-catchment number

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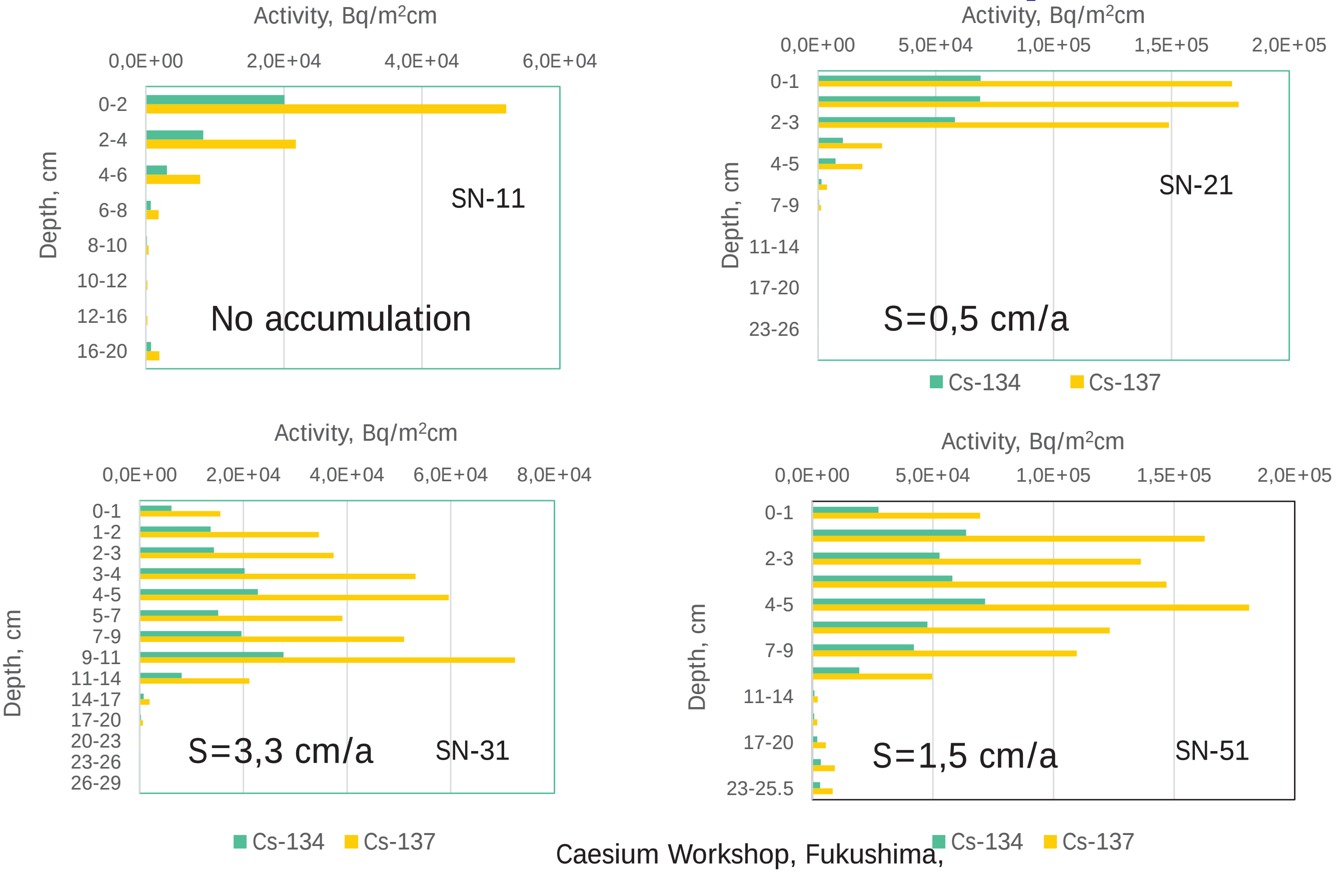
Radiocesium vertical profiles in soils of Niida river flood plain



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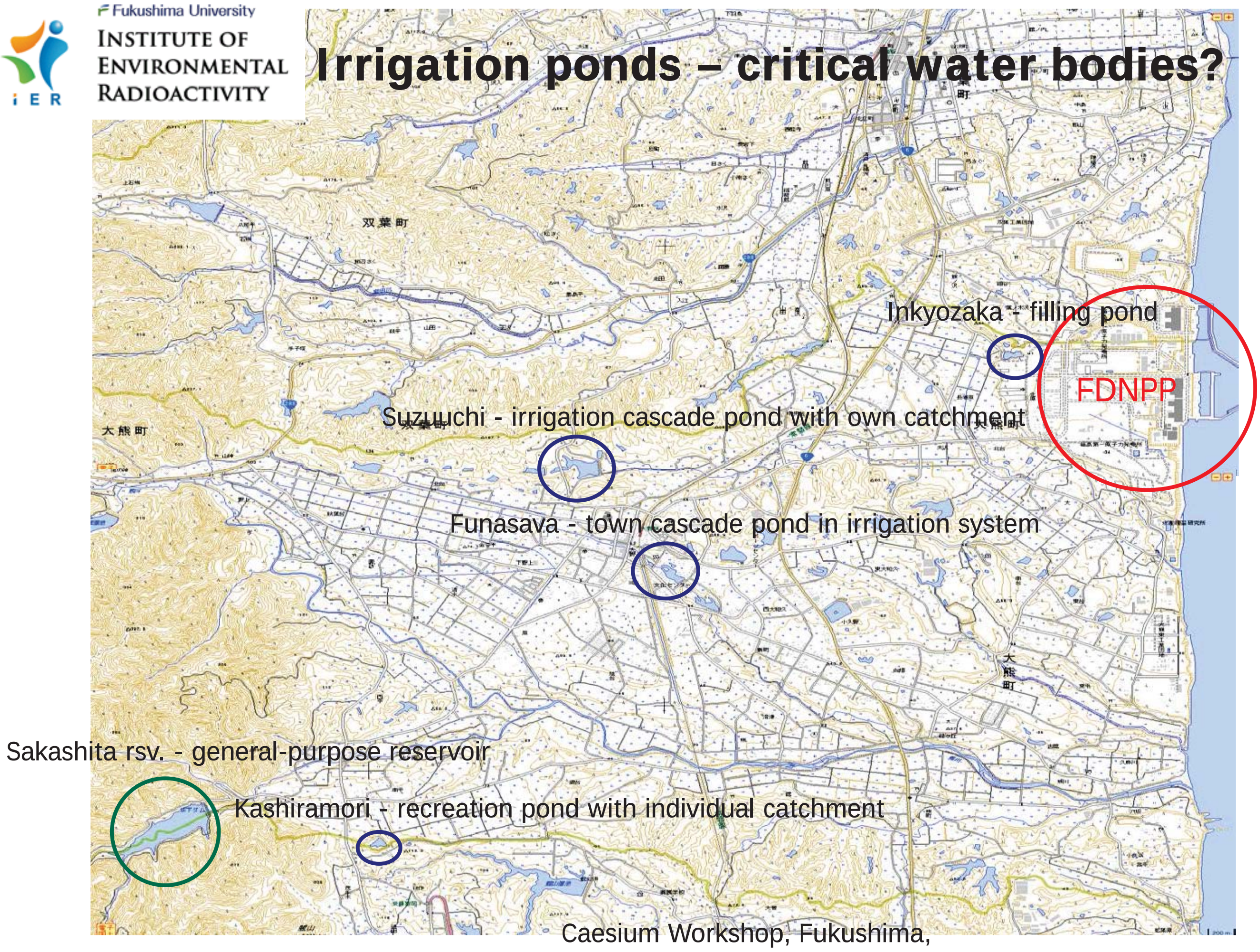
Irrigation ponds – critical water bodies?



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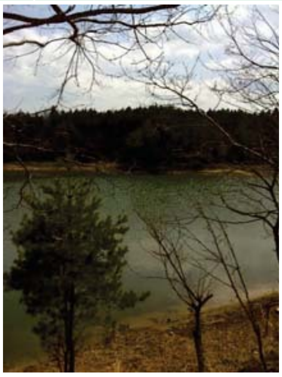
Total ^{137}Cs activity concentrations in water bodies of Okuma town (March 2014)

Location	Distance from FDNPP, km	Dose rate, uSv/h	Deposition ^{137}Cs, kBq/m^2	^{137}Cs, Bq/L
Suzuuchi	3,75	24	5840	59,4∓0,9
Funasawa	3,50	3,25	-	9,3∓0,2
Inkyozaka	0,24	11	4250	3,78∓0,06
Kashiramori	7	3,3	865	0,747∓0,003
Sakashita	8	3,0	672	0,125∓0,013



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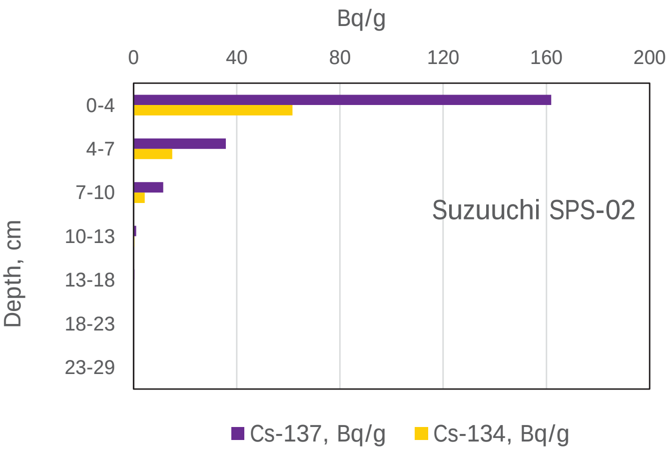
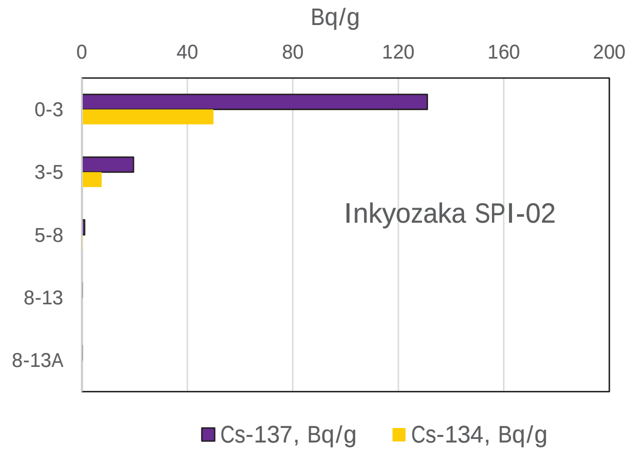
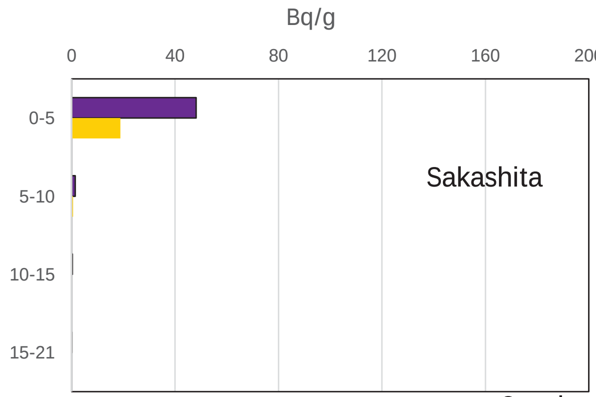
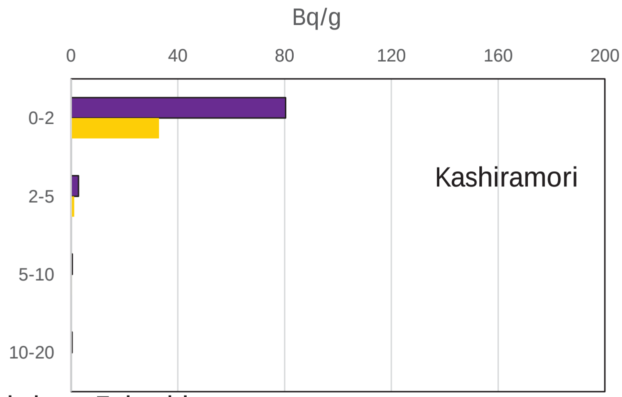
Radiocesium vertical profiles in soils of Okuma town









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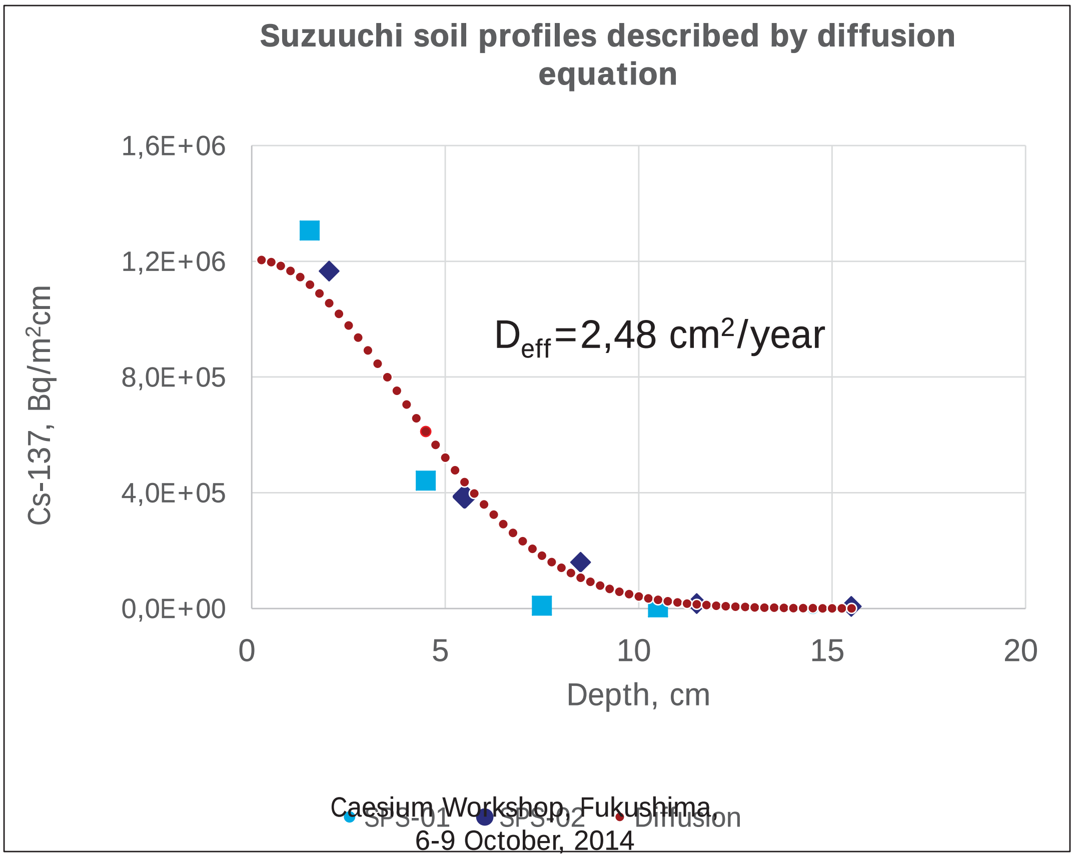
Simplified diffusion model of radiocesium vertical migration

- $$Q(x) = \frac{Q_0}{\sqrt{\pi D t}} \times e^{-\frac{x^2}{4Dt}}$$
- Q(x) – radiocesium content on depth x (Bq/m^2cm);
- 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



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Effective diffusion of radiocesium in soils of Okuma town

Location	Deposition, kBq / m²	Soil type	D_{eff}, cm² / year
Fukushima			
Suzuuchi	5840	Sandy Fluvisols	2,48
Inkyozaka	4250	Sandy Fluvisols	2,24
Kashiramori	865	Sandy-loam Cambisols	9,27
Sakashita	672	Loam Inceptisols	5,0
Chernobyl			
Benevka	1500	Alluvial sandy-loam	0,5
Chernobyl	750	Cultivated sandy-loam	0,6

- D_{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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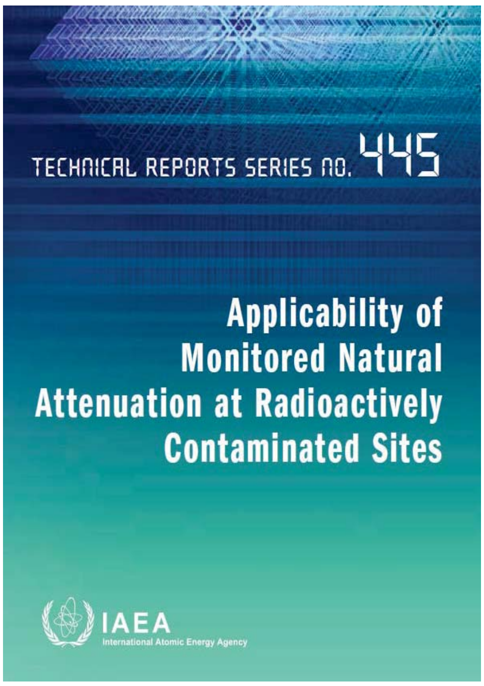
Monitored natural attenuation - an option for water remediation

- 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.



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Conclusions

- Normalized solid wash-off coefficients for Fukushima area very well corresponded to those of Chernobyl;
- Normalized liquid wash-off coefficients N_l (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_{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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