

*Fukushima Research Conference on radiocesium migrations in terrestrial environments and in ecosystems (3rd International Cesium Workshop)(4<sup>th</sup> March, 2018, Fukushima)*

福島リサーチカンファレンス(国際セシウムワークショップ)

「放射性セシウムの陸域環境や生態系における移行」(2018.3.4, 福島)

# ***Distribution and Input/output budgets of radiocesium in the mountainous forest of Fukushima***

**(山地の森林域における放射性セシウムの分布と流入/流出量)**

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Fukushima Prefectural Centre for Environmental Creation

## ■ **Introduction**

- *Physical geography, land use, radiation dose*
- *Transport processes*

## ■ **Methods**

- *Monitoring at experimental plots, tree sampling*

## ■ **Results**

- *Radiocesium distribution, Input/output budgets*

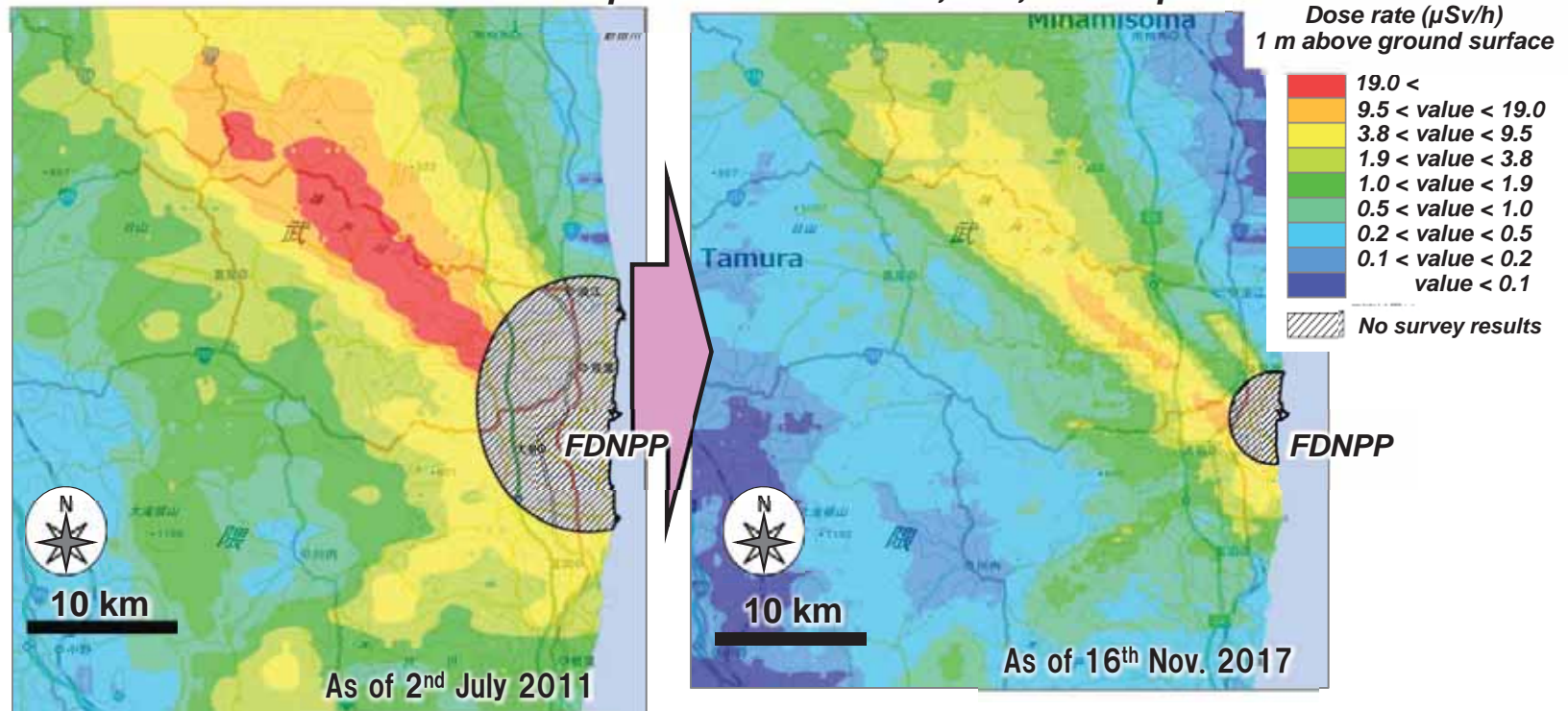
## ■ **Conclusions & key issues**

- *Studies in progress (bioavailability of radiocesium)*
  - ✓ *Sources of dissolved radiocesium (decomposition of litter?)*
  - ✓ *Migration in dissolved fraction (tree system, root uptake)*
  - ✓ *Long term ecological research (interaction between components of forested ecosystem)*

# Distribution map of radiation dose (airborne monitoring) <sup>3</sup>

The source of the map;

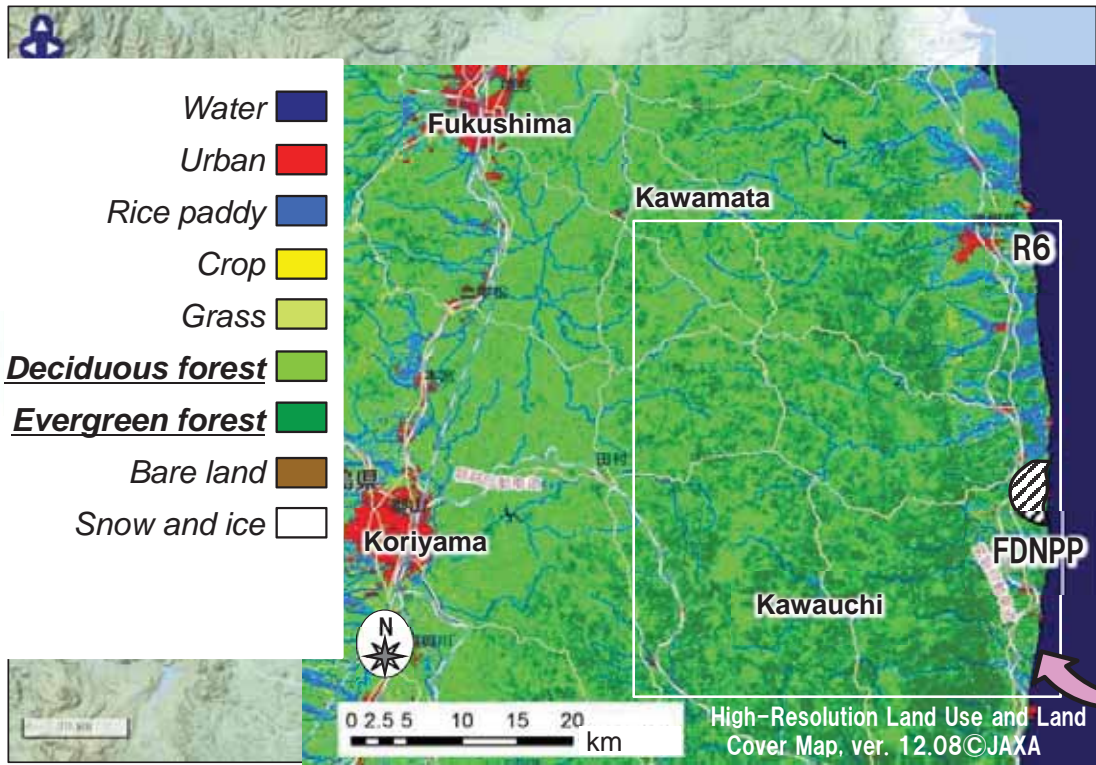
Extension Site of Distribution Map of Radiation Dose, etc./GSI Maps



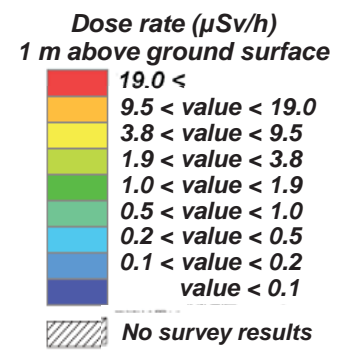
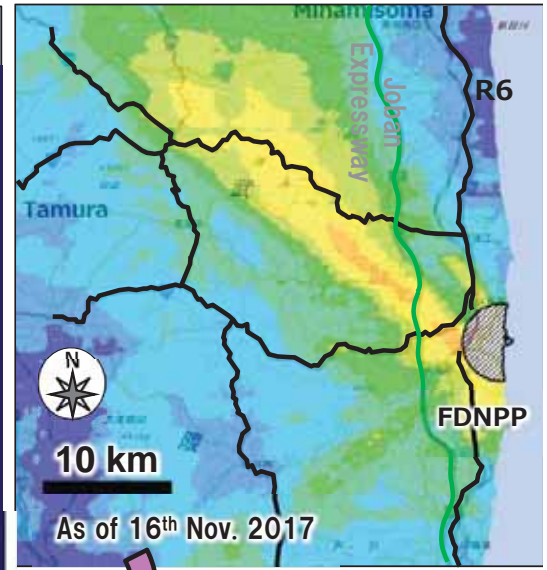
- **Generally, radiation dose decreases with time**
- **The area with higher radiation dose still exists**

# Topography, land use, and radiation dose

## Land use



## Radiation dose



- **Generally, radiation dose decreases with time**
- **The area with higher radiation dose still exists**  
→ **Mountainous forest covers most of the area**

# Comparison; physical geography

	<i>Forested land</i> <sup>1),2)</sup>	<i>Mean average temperature</i> <sup>3),4)</sup>	<i>Annual precipitation</i> <sup>3),4)</sup>	<i>Topography</i>
<b>Belarus (Minsk)</b>	<b>42 %</b>	Jan. -4.3 °C July 19.5 °C	<b>690 mm</b>	<b>Lowland</b> Wetland (Pripyet Marshes)
<b>Ukraine (Kiev)</b>	<b>16 %</b>	Jan. -4.4 °C July 18.5 °C	<b>608 mm</b>	North; <b>Wetland</b> (Pripyet Marshes) East & west; <b>Hilly land</b>
<b>Fukushima</b>	<whole Fukushima> <b>68 %</b>	<Fukushima City> Jan. 1.6 °C July 23.6 °C	<b>1,166 mm</b> Dec. 42 mm Sept. 160 mm	Inland lowland
	<kawamata> <sup>5)</sup> <b>80 %</b>	Jan. 0.5 °C July 23.6 °C	<b>1,368 mm</b> Dec. 42 mm Sept. 211 mm	<b>Mountainous &amp; Hilly land</b>
	<Kawauchi> <b>90 %</b>	Jan. -0.7 °C July 26.0 °C	<b>1,465 mm</b> Dec. 50 mm Sept. 227 mm	<b>Mountainous &amp; Hilly land</b>

1) FAOSTAT (2010)

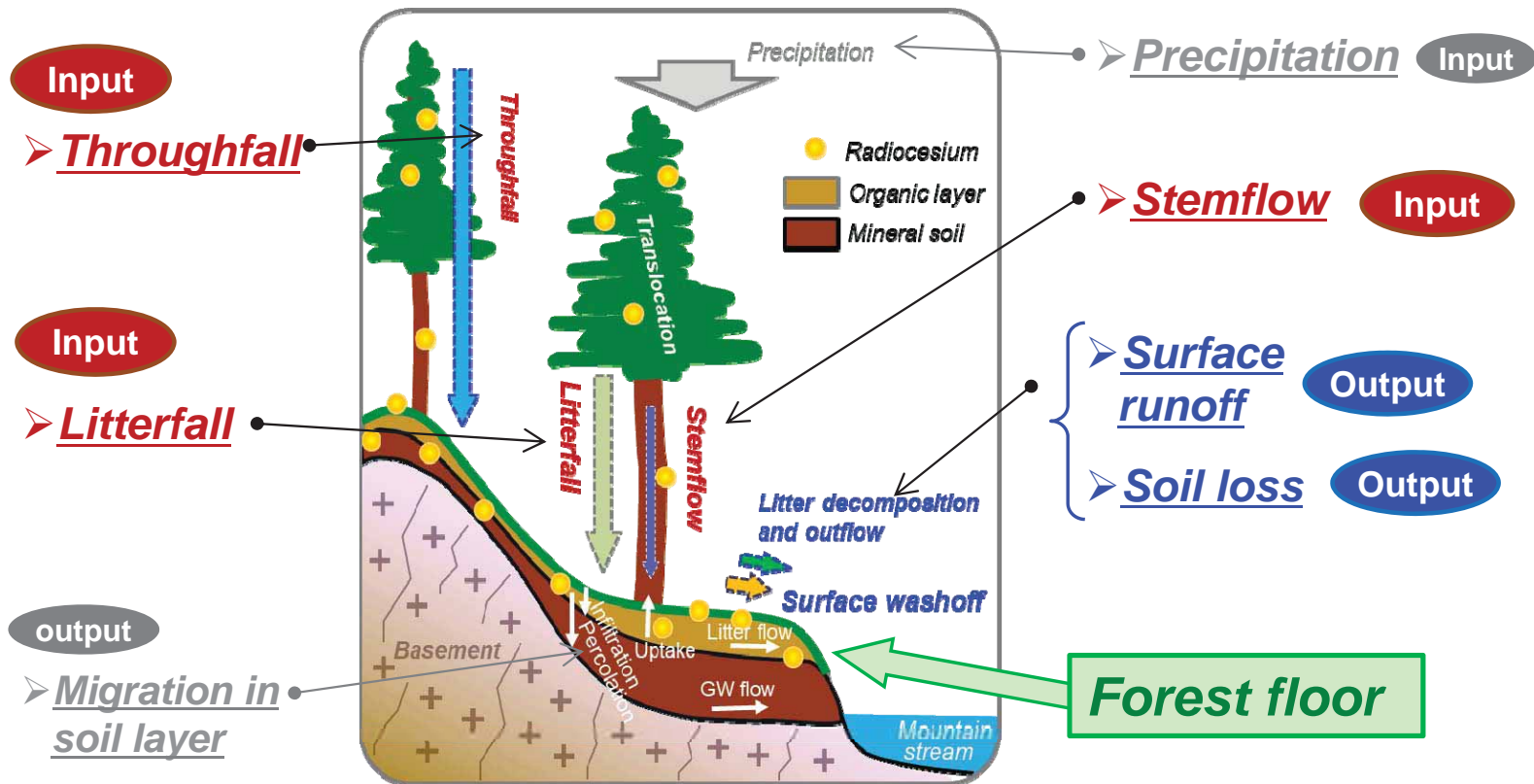
2) The 88<sup>th</sup> statistical yearbook of Ministry of Agriculture, Forestry and Fisheries (2012-2013)

3) Chronological Scientific Tables (2014)

4) Japan Meteorological Agency (2014) <past 30 years (1981-2010)>

5) Kawamata Machi official web page <<http://www.town.kawamata.lg.jp/>> (2014)

# Dynamics of radiocesium in the forested land



## Objectives

*Input and output budgets* of radiocesium within a forested area **with the forest floor of particular emphasis** to clarify whether the forest floor behaves as a **sink (input > output)** or a **source (output > input)** of the contamination.

# Forest investigation



**Yamakiya, Kawamata**  
(**mixed forest;**  
Red pine, Konara oak)

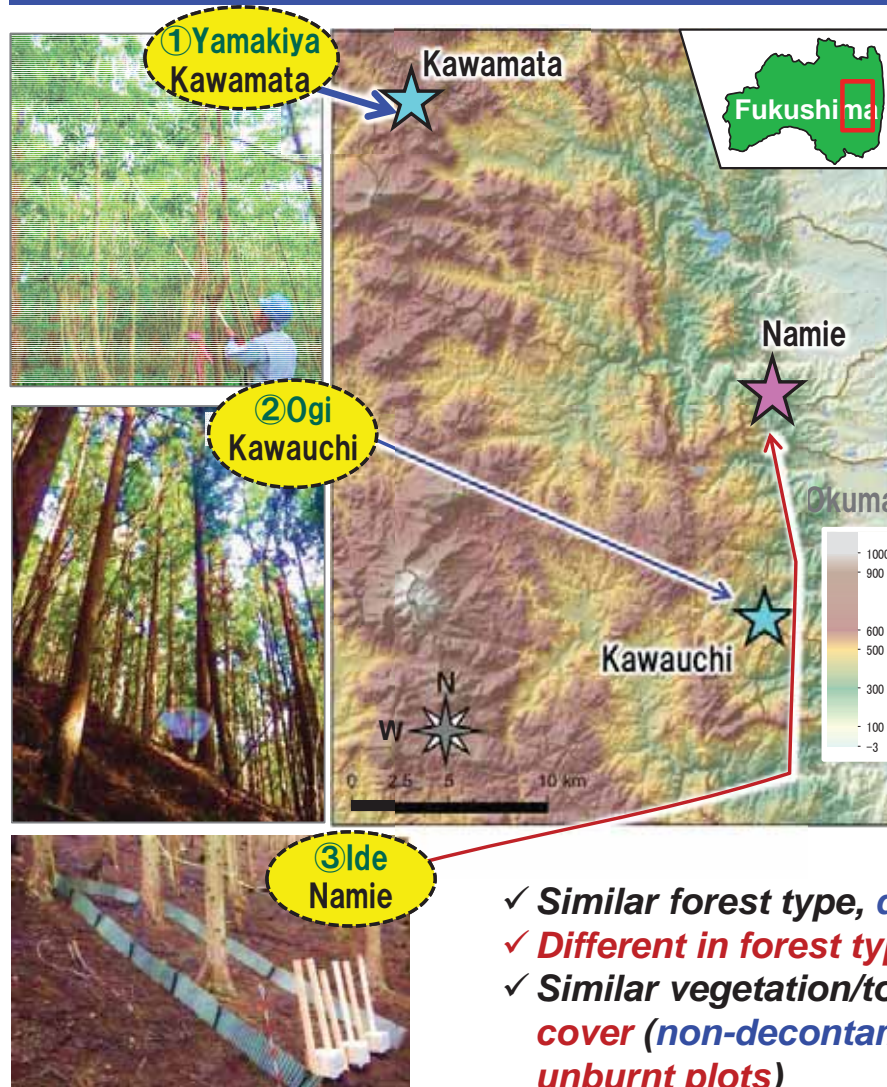


**Ogi, Kawauchi**  
(**evergreen coniferous**  
forest, Japanese cedar)



**Ide, Namie**  
(**evergreen coniferous**  
forest, Japanese cedar)

# Outline of study areas



Forest type	Topography	Soil type
<b>① Yamakiya, Kawamata</b>		
✓ <b>Mixed forest</b> of <b>deciduous</b> and <b>evergreen</b> trees (Japanese red pine and konara oak)	✓ <b>Gentle to steep</b> slope facing the south	Experimental plots ( <b>gentle &amp; steep</b> slope)
✓ Brown forest soil (sandy loam to loam)	✓ <b>Decontaminated</b> and <b>undisturbed</b> plots	
<b>② Ogi, Kawauchi</b>		
✓ <b>Evergreen forest</b> (Japanese cedar)	✓ Ogi A; <b>Intermediate to steep</b> slope facing the north	Experimental plots ( <b>steep</b> slope)
✓ Ogi B; <b>Steep</b> slope facing south	✓ Brown forest soil (silt loam to loam)	
	✓ <b>Undisturbed</b> plot	
<b>③ Ide, Namie</b>		
✓ <b>Evergreen forest</b> (Japanese cedar)	✓ <b>Steep</b> slope facing the south	Experimental plots ( <b>steep</b> slope)
✓ Brown forest soil ( <b>sand</b> )	✓ <b>Undisturbed</b> and <b>disturbed</b> plots by wildfire	

- ✓ **Similar forest type, different in topography** (Yamakiya)
- ✓ **Different in forest type, similar topography** (Ogi - Yamakiya)
- ✓ **Similar vegetation/topography, but different in forest floor cover** (**non-decontaminated vs. decontaminated, burnt vs. unburnt plots**)

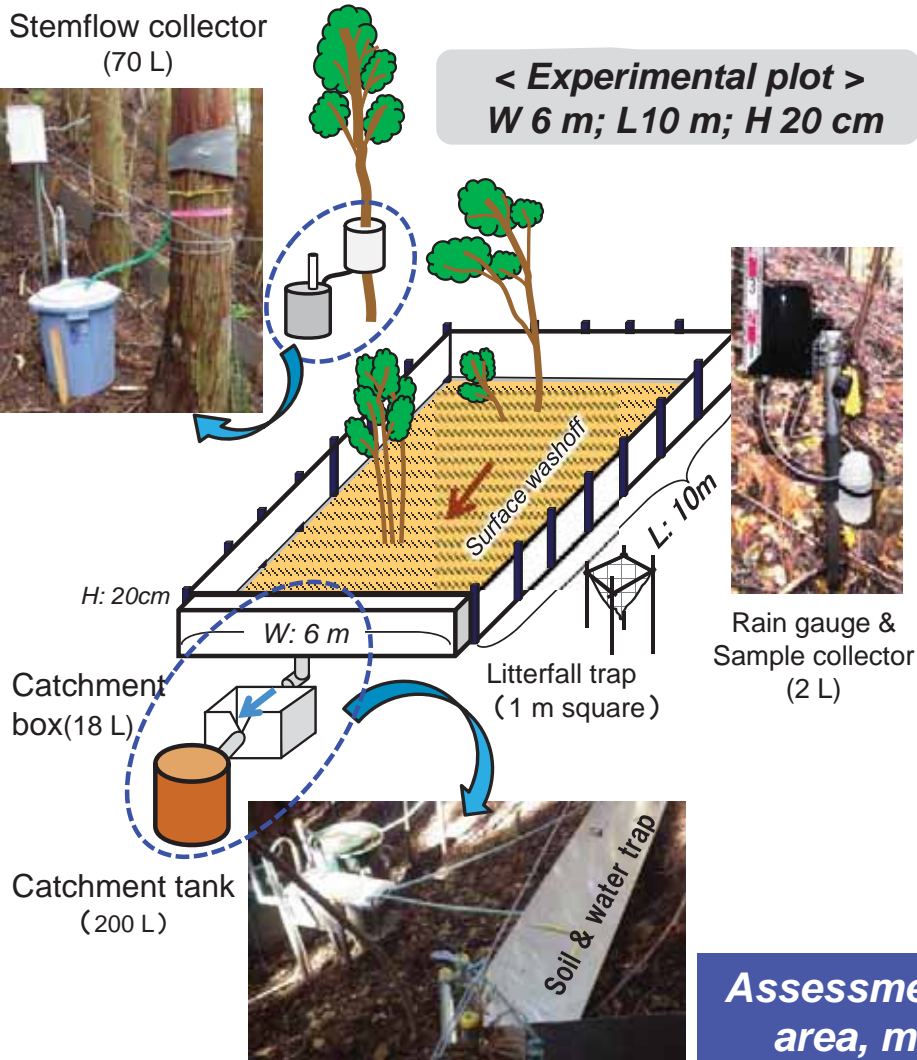


# Monitoring items in the experimental plots

Stemflow collector  
(70 L)



< Experimental plot >  
W 6 m; L 10 m; H 20 cm



Canopy to forest floor  
(radioCs input)

## Throughfall

- ✓ Amount of throughfall (mm=L/m<sup>2</sup>)
- ✓ radioCs activity (Bq/L)

## Stemflow

- ✓ Stemflow volume (L/m<sup>2</sup>)
- ✓ radioCs activity (Bq/L)

## Litterfall

- ✓ Weight of litterfall (kg/m<sup>2</sup>)
- ✓ radioCs activity (Bq/kg)

Forest floor to  
outside of the forest  
(radioCs output)

## Surface washoff (soil loss, surface runoff)

- ✓ Weight of particulate matter (kg/m<sup>2</sup>)
- ✓ Surface runoff volume (L/m<sup>2</sup>)
- ✓ radioCs activities of particulate matter (Bq/kg) and surface runoff water (Bq/L)

Assessment of radioCs input & output in unit area, monitoring period [Bq m<sup>-2</sup> period<sup>-1</sup>]

# Transport processes on forest floor

## Transport & sedimentation processes

### ☐ Raindrop erosion, surface wash

- ✓ Rainfall run-off (overland flow, downslope flow)
- ✓ Raindrop erosion
- ✓ Stem flow
- ✓ Litter flow (run-off over the litter layer, esp. broad-leaved forest)
- ✓ Rainy season (July to October)
- ✓ **Whole forested area**

### ☐ Frost action

- ✓ Freezing and thawing action (frost heave, frost creep)
- ✓ Downslope transport of thawing soil
- ✓ Winter season (December, January to February)
- ✓ **South-facing forested area, thin litter layer**

### ☐ Mass movement

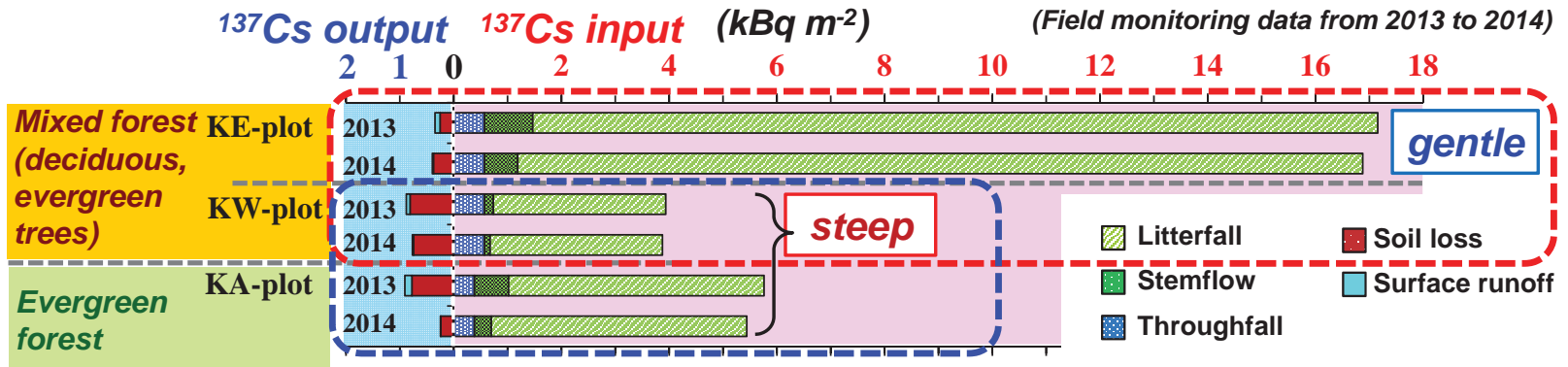
- ✓ Small-scale mudslide
- ✓ Very steep forest slope
- ✓ Mainly, rainy season and snowmelt season (late February to March)
- ✓ **Small area in the forest**

### ☐ Nivation

- ✓ Erosion by melting water
- ✓ Shaving and grooving of top soil
- ✓ Snow season and snow melt season
- ✓ **Limited area in the forest**

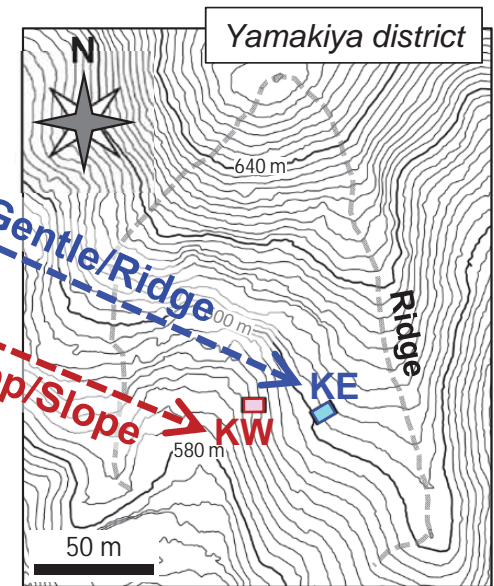


# $^{137}\text{Cs}$ input/output budgets, forest floor basis



(※)6<sup>th</sup> Airborne monitoring results

Plot	Year	$^{137}\text{Cs}$ (kBq m <sup>-2</sup> )			
		Inventory*	Output (rate, %)	Input (rate, %)	Input/output
KE-plot	2013	497.0	0.3 (0.07)	17.2 (3.45)	50
	2014		0.4 (0.08)	16.9 (3.40)	43
KW-plot	2013		0.9 (0.18)	3.9 (0.79)	4
	2014		0.8 (0.15)	3.9 (0.78)	5
KA-plot	2013	487.0	0.9 (0.19)	5.8 (1.18)	6
	2014		0.2 (0.05)	5.4 (1.12)	22



The topographic data were created based on the results of an airborne laser survey conducted by the Geospatial Information Authority of Japan.

- $^{137}\text{Cs}$  output  $\ll$   $^{137}\text{Cs}$  input  
 The forest floor behaves as a sink
- Topographic difference  $>$  Forest type difference

# <sup>137</sup>Cs output in the various forest floor conditions

**Decontaminated plot**

2 m × 4 m, dips 27° west  
**Cover rate approx. 30%**




March, 2016

**Disturbed plot by wildfire**

Monitoring period  
 19<sup>th</sup> June – 20<sup>th</sup> Nov., 2017

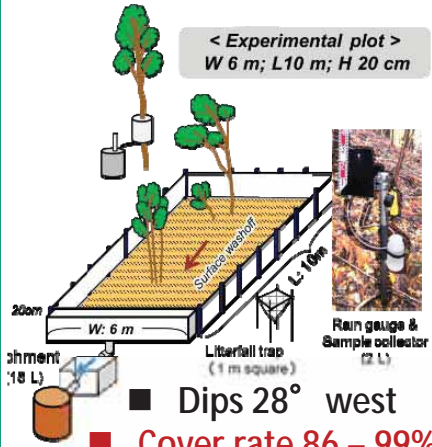
■ Dips approx. 30° south  
 ■ **Cover rate**  
 ✓ 10 – 18% (disturbed)  
 ✓ 90 – 92% (undisturbed)



Plot	<sup>137</sup> Cs Inventory [Bq/m <sup>2</sup> ]	<sup>137</sup> Cs output [Bq/m <sup>2</sup> ]	Output rate
Disturbed	416k	10,377	2.49 %
Undisturbed	547k	798	0.15 %

**Undisturbed plot**

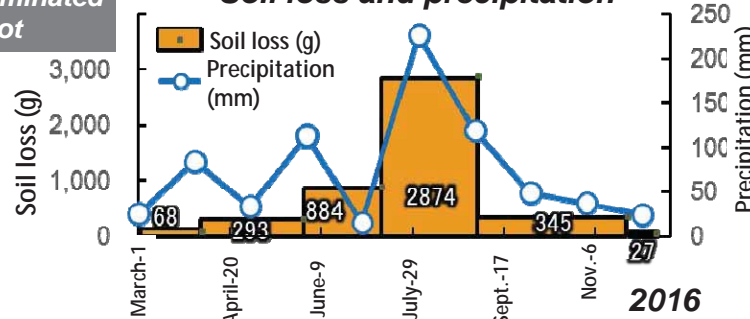
< Experimental plot >  
 W 6 m; L 10 m; H 20 cm



■ Dips 28° west  
 ■ **Cover rate 86 – 99%**

**Decontaminated plot**

**Soil loss and precipitation**



Date	Soil loss (g)	Precipitation (mm)
March-1	68	~50
April-20	293	~80
June-9	884	~180
July-29	2874	~220
Sept.-17	~500	~100
Nov.-6	345	~50
Nov-27	27	~20

**Soil loss and <sup>137</sup>Cs output**

Soil loss	<sup>137</sup> Cs output ①	<sup>137</sup> Cs inventory②	<sup>137</sup> Cs output rate (①÷②)
0.56 kg/m <sup>2</sup>	10.5 kBq/m <sup>2</sup>	1,600 kBq/m <sup>2</sup>	0.66 %

**Undisturbed plot**

**Yamakiya**  
 (mixed forest; steeply inclined)

Year	2013	2014	2015	2016
<sup>137</sup> Cs outflow via soil loss [Bq/m <sup>2</sup> ](April – Nov. )	810	1,721	180	308
<sup>137</sup> Cs inventory around the plot [kBq/m <sup>2</sup> ]	1,600 (Nov. 2013)			
<sup>137</sup> Cs outflow rate [%](April – Nov.)	0.05	0.11	0.01	0.02%

**<sup>137</sup>Cs output in the decontaminated plot (ca.0.6%per year) far exceed that in the undisturbed plot (0.02% per year)**

# Tree sampling (Japanese cedar)

① Selection of tree and covering



② Cutting



① Every tree measurement (the number of trees,  $D_{BH}$ )



① Selection and covering



② Tree cutting



③ Needle and twig → weighting, pretreatment, Ge analysis

④ Bark → size measurement, weighting, pretreatment, Ge analysis

⑤ Stem disk → splitting, separation of heart- and sap-woods, weighting, bulk density, pretreatment, Ge analysis

③ Needle and twig sampling



⑤ Sampling of stem disk



Splitting of stem disk



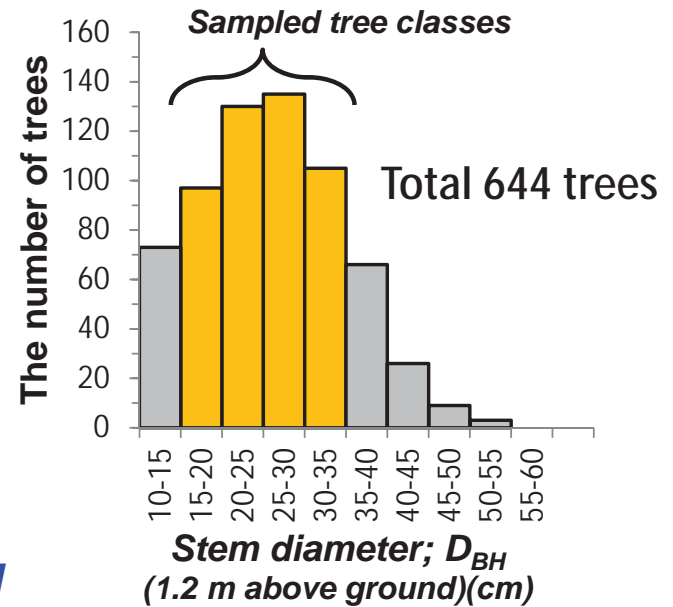
④ Bark sampling



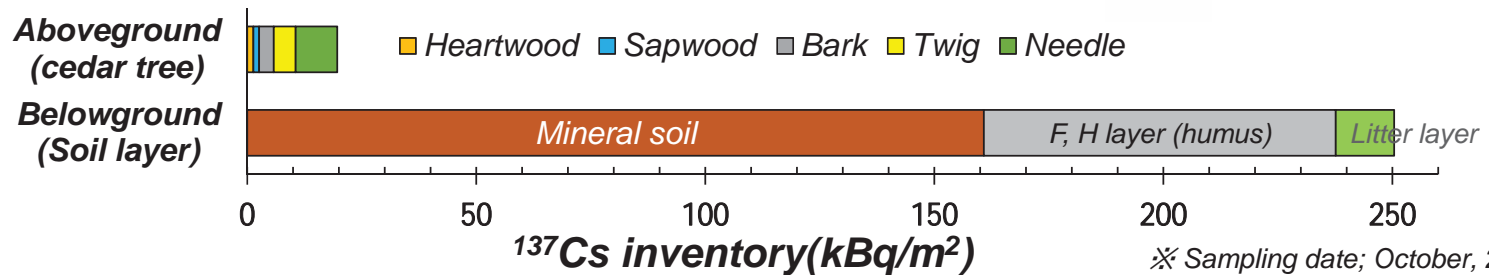
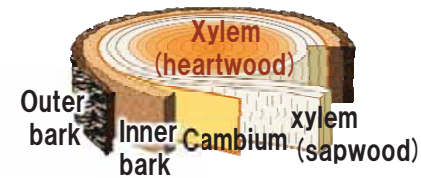
Separation of heart- and sap-woods

Ref.; Kajimoto et al., 2014, Bulletin of FFPRI, vol.13, no.3, 113-136.

# Aboveground and belowground $^{137}\text{Cs}$ inventories <sup>14</sup>



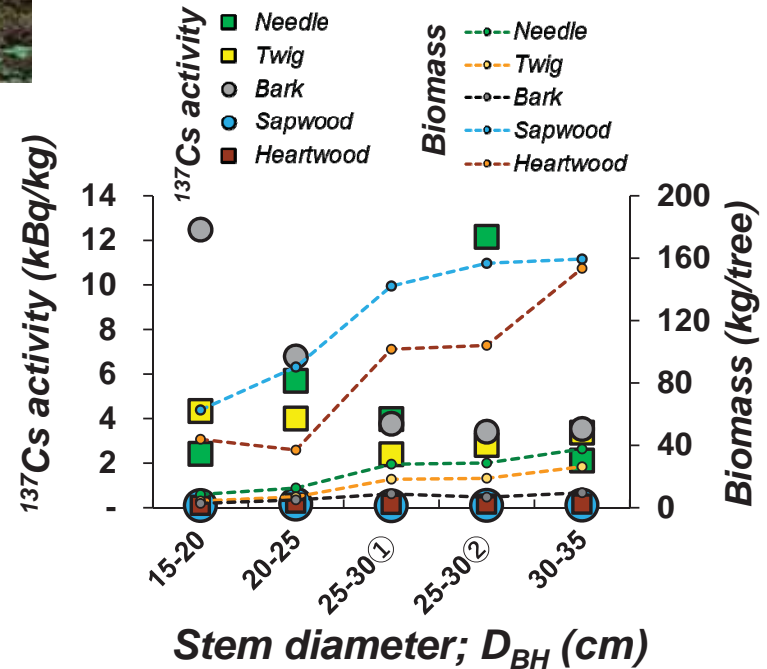
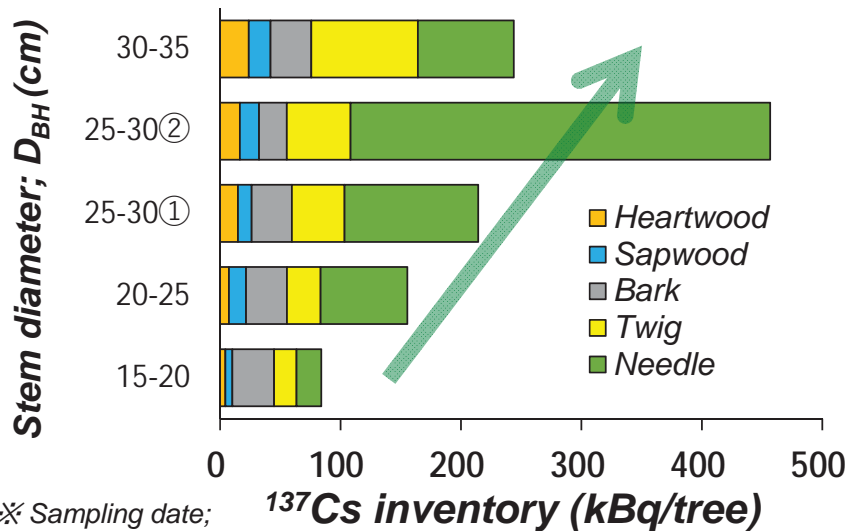
- **Aboveground (cedar tree) << Belowground (litter & soil layer)**
- **radioCs mostly exists in the belowground (approx. 90 % of the  $^{137}\text{Cs}$  inventory in the forest)**



# $^{137}\text{Cs}$ partitioning of aboveground tree system

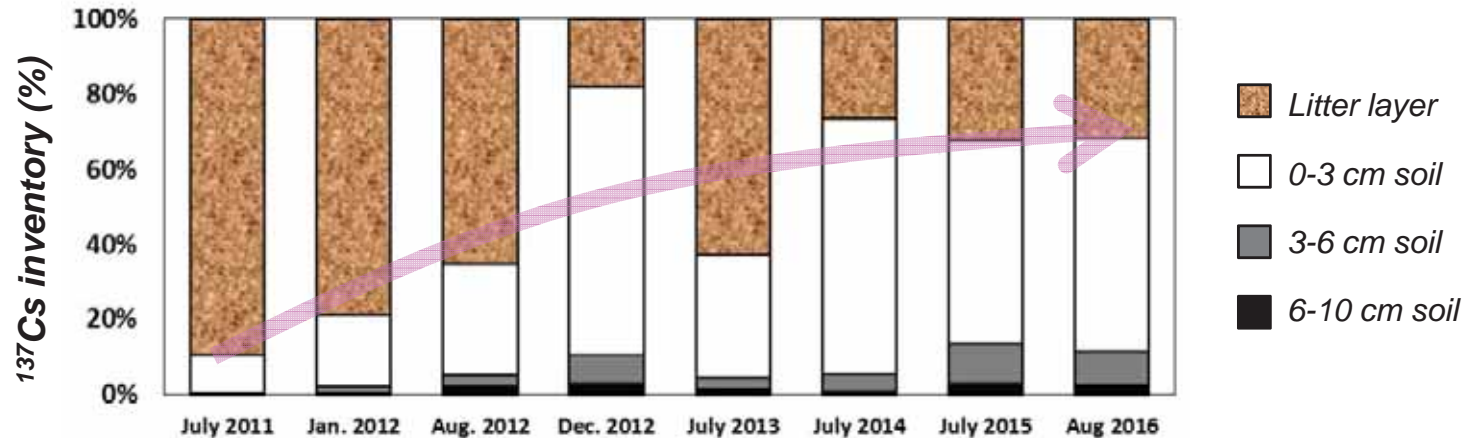


- $^{137}\text{Cs}$  inventory;
  - ✓ **Needle**  $\geq$  Twig  $\geq$  Bark  $>$  wood
- $^{137}\text{Cs}$  activity;
  - ✓ **Bark**  $\geq$  Needle  $\geq$  Twig  $>$  wood
- $^{137}\text{Cs}$  inventories are increasing with the tree biomass, though needle and bark show large variations in  $^{137}\text{Cs}$  activity.



※ Sampling date; October, 2015

# Distributions of $^{137}\text{Cs}$ inventories in the litter & soil layers <sup>16</sup>



## Temporal changes in distribution of $^{137}\text{Cs}$ inventories in litter and soil layers (young cedar forest)

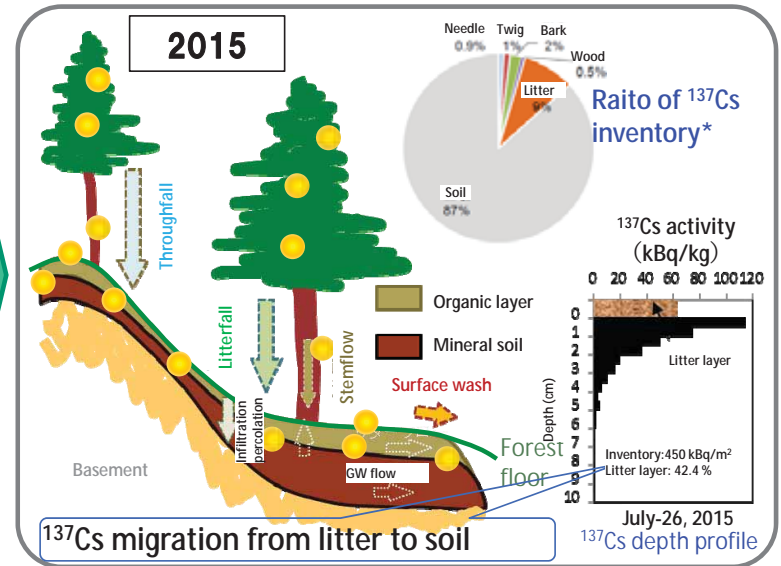
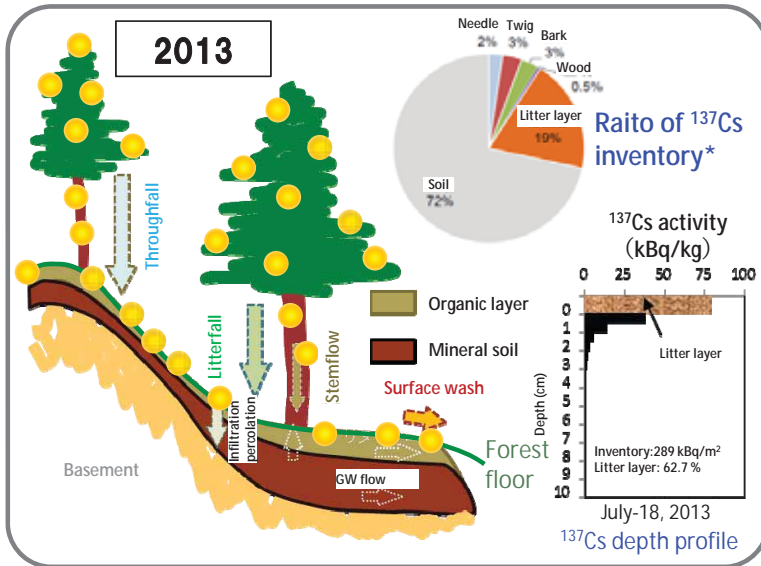


Soil sampling by scraper plate  
(sampling interval: 0.5 cm, 1 cm)

- $^{137}\text{Cs}$  proportion in the litter layer remarkably decrease from approx. 90 % in July 2011 to approx. 30% in August 2016.
- The soil layers deeper than 3 cm *show little variation of  $^{137}\text{Cs}$  inventories* in the past 5 years.
- These indicate that...
  - ✓  $^{137}\text{Cs}$  transfer from soil surface to the lower layer *hardly occur.*
  - ✓  $^{137}\text{Cs}$  *remains* the litter and soil layers *less than 3 cm depth* for a long time.



# Summary; Temporal and spatial changes of radioCs distribution in the forested ecosystem



●  **$^{137}\text{Cs}$  output** <<  **$^{137}\text{Cs}$  input**

✓ the forest floor behaves as a sink, except for the lower cover rate  
(decontaminated and disturbed by wildfire)  
→ **Forest floor cover is key parameter**

● **Aboveground (cedar tree)** << **Belowground (litter & soil layer)**

● radioCs mostly exists in the belowground

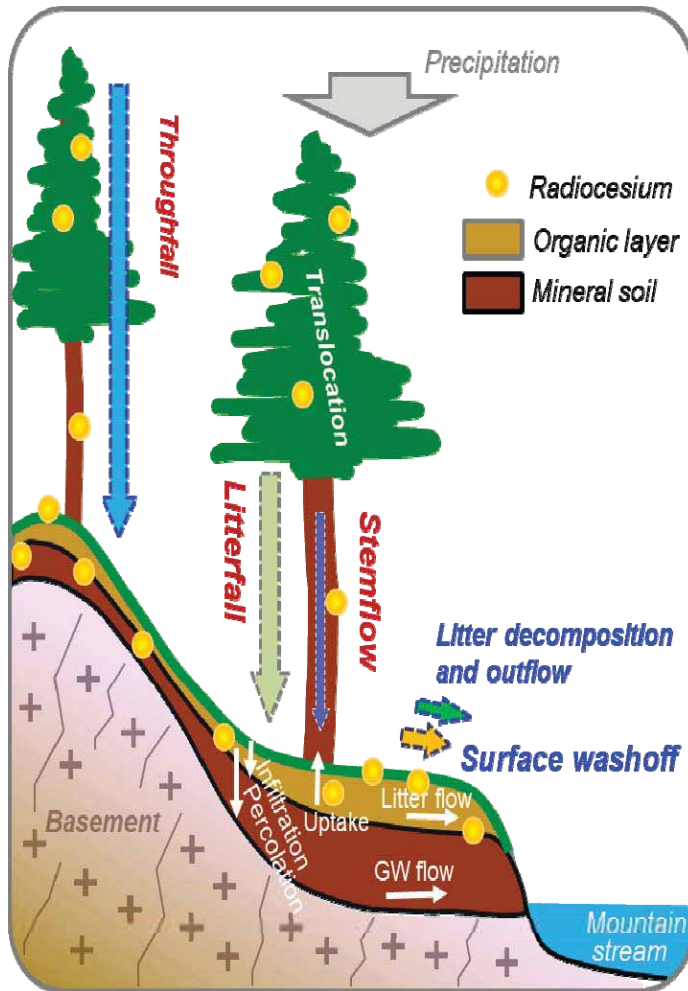
(approx. 90 % of the  $^{137}\text{Cs}$  inventory in the forest)

●  **$^{137}\text{Cs}$  proportion in the litter layer remarkably decrease.**

●  **$^{137}\text{Cs}$  remains the litter and soil layers less than 3 cm depth for a long time.**

\*The investigation results for the present state of radioactive substances distribution in the forest (H27 report) (Forestry Agency, 2016) Niizato et al. (2016), J. Environ. Radiact., vol.161, 11-21.

## Key issues; Dynamics of radioCs in the forested land



### □ Outflow from the forested ecosystem

→ Projection of radioCs outflow

- ✓ Soil loss/outflow of particulate-bound radioCs
- ✓ Surface washoff /outflow of dissolved radioCs

### □ Cycling in the forested ecosystem

→ Projection of radioCs concentration in the forest products

→ Projection of radioCs distribution

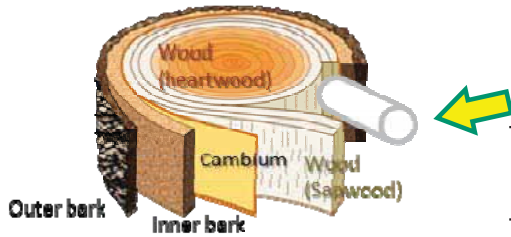
- ✓ Throughfall, Litterfall, stemflow/transport from canopy to forest floor, then into soil layer
- ✓ Litter decomposition/elution of dissolved radioCs and its migration into soil layer
- ✓ Tree and wild plant radioCs uptake/recycling from soil rhizosphere/layer
- ✓ (re)Translocation in plant system

### □ External exposure in the forested land

→ Assessment of external exposure in forested land

- ✓ Temporal and spatial changes of radioCs distribution and air dose rate

# Dissolved radioCs; transfer, elution



Drill a hole to the wood by a drill, we collected the xylem sap

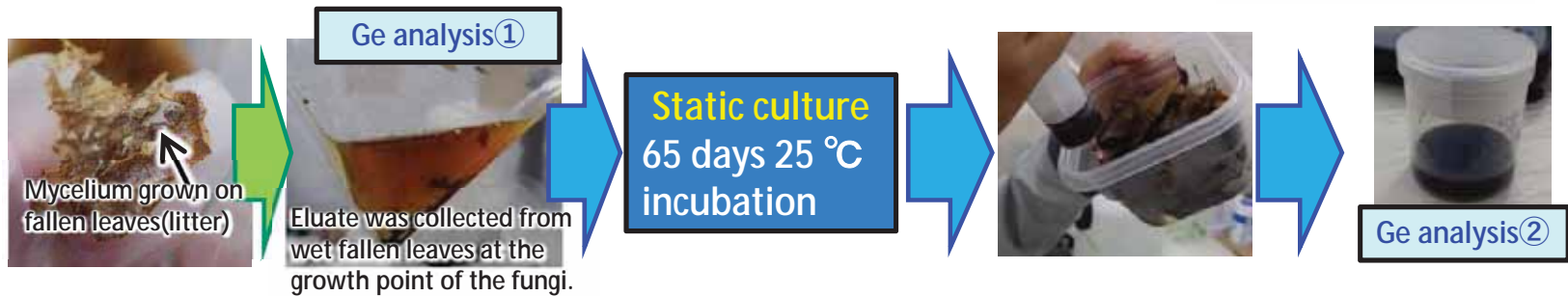
Radiocesium concentration in xylem sap (Bq/L) ( $<0.45\mu\text{m}$ )

$^{134}\text{Cs}$	$5.78 \pm 0.32$
$^{137}\text{Cs}$	$30.6 \pm 0.10$

※Sampling March 2 - April 5, 2016  
About 5 liters of sap collected



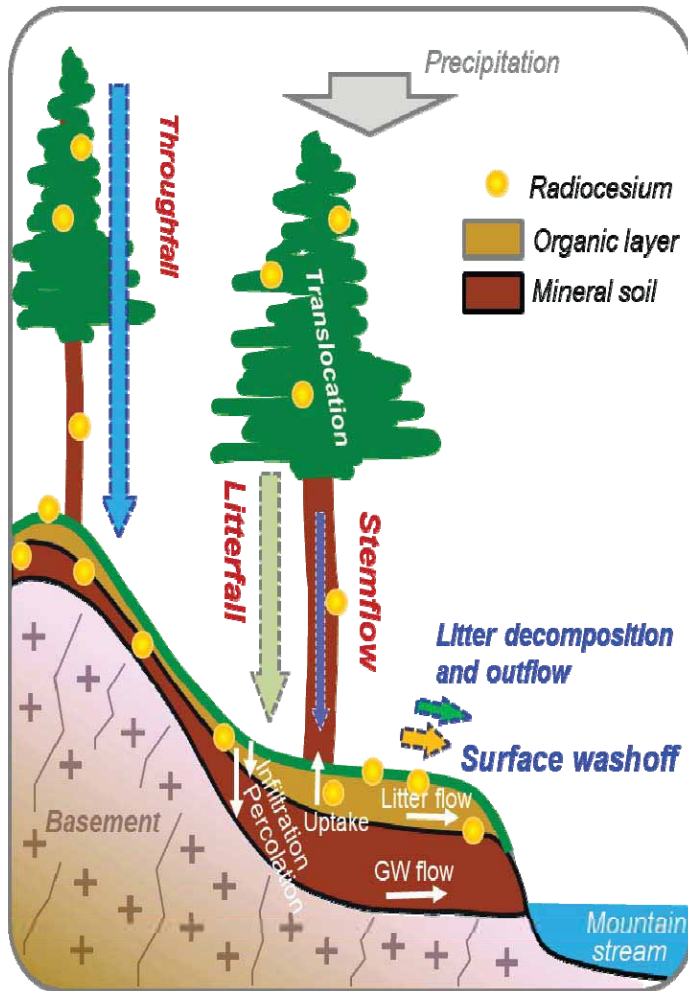
- Dissolved radioCs transferred through xylem sap



		Sampling date	$^{137}\text{Cs}$ (Bq/kg, Bq/L)	$^{137}\text{Cs}$ ratio (Eluate/Litter)
① After sampling	Litter	Sept. 15, 2016	$(8.43 \pm 0.07) \times 10^3$	$7.5 \times 10^2$ ↓
	Eluate		$(1.12 \pm 0.09) \times 10$	
② After incubation	Litter	Nov. 19, 2016	$(9.76 \pm 0.09) \times 10^3$	$1.1 \times 10^2$ ↓
	Eluate		$(9.05 \pm 0.26) \times 10$	

- radioCs activities of eluate; “after sampling “ < “after incubation”
- There is a possibility that litter decomposition by fungi accelerated elution of dissolved radioCs from litter

## Key issues; Dynamics of radioCs in the forested land



### □ Outflow from the forested ecosystem → Projection of radioCs outflow

- ✓ **Soil loss**/outflow of particulate-bound radioCs
- ✓ **Surface washoff** /outflow of dissolved radioCs

### □ Cycling in the forested ecosystem → Projection of radioCs concentration in the forest products → Projection of radioCs distribution

- ✓ **Throughfall, Litterfall, stemflow**/transport from canopy to forest floor, then into soil layer
- ✓ **Litter decomposition**/elution of **dissolved radioCs** and its migration into soil layer
- ✓ **Tree and wild plant radioCs uptake**/recycling from soil rhizosphere/layer
- ✓ **(re)Translocation** in plant system

### □ External exposure in the forested land → Assessment of external exposure in forested land

- ✓ **Temporal and spatial changes of radioCs distribution and air dose rate**

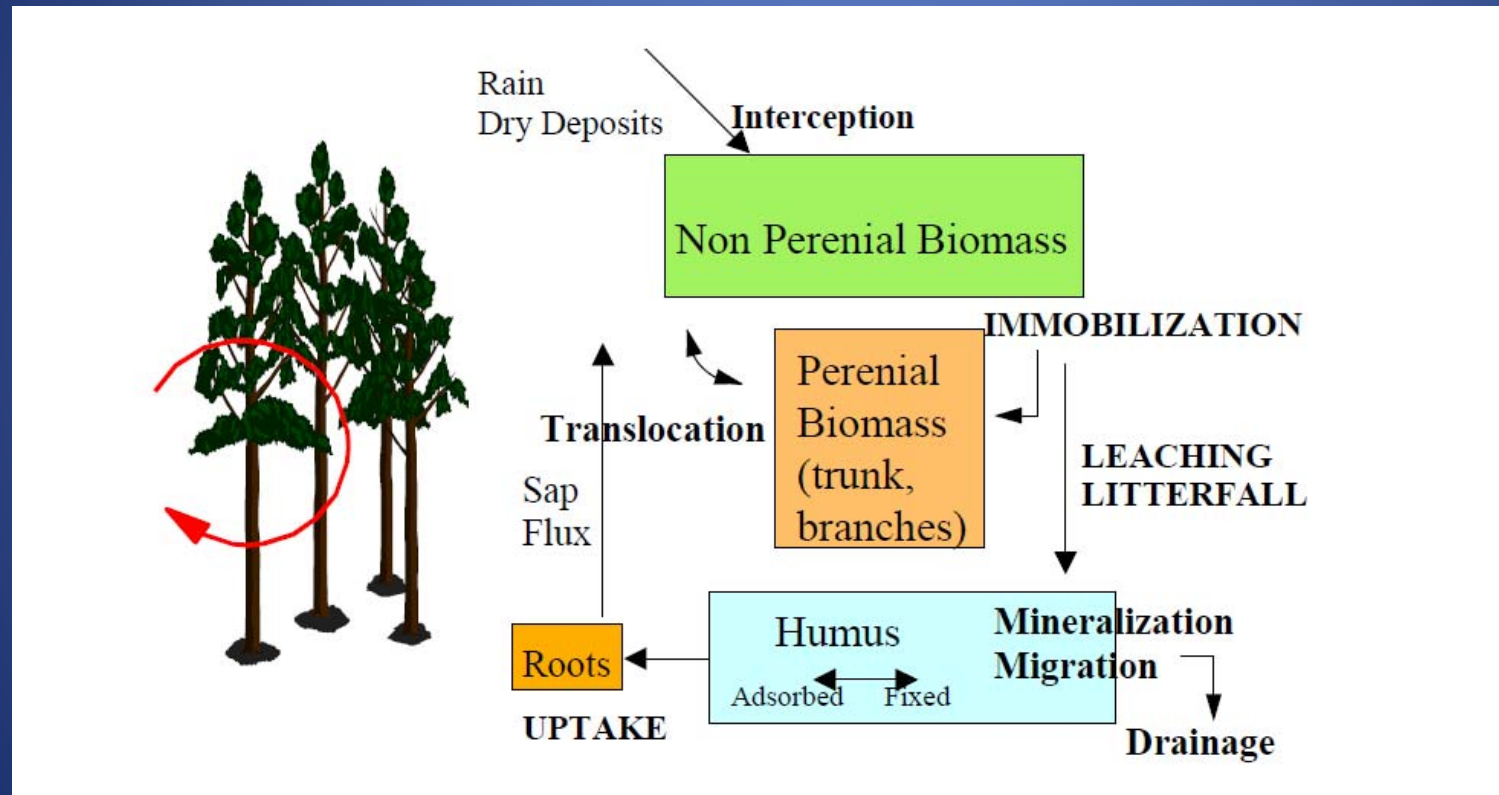


# *Dynamics of radiocaesium in the forest ecosystem*

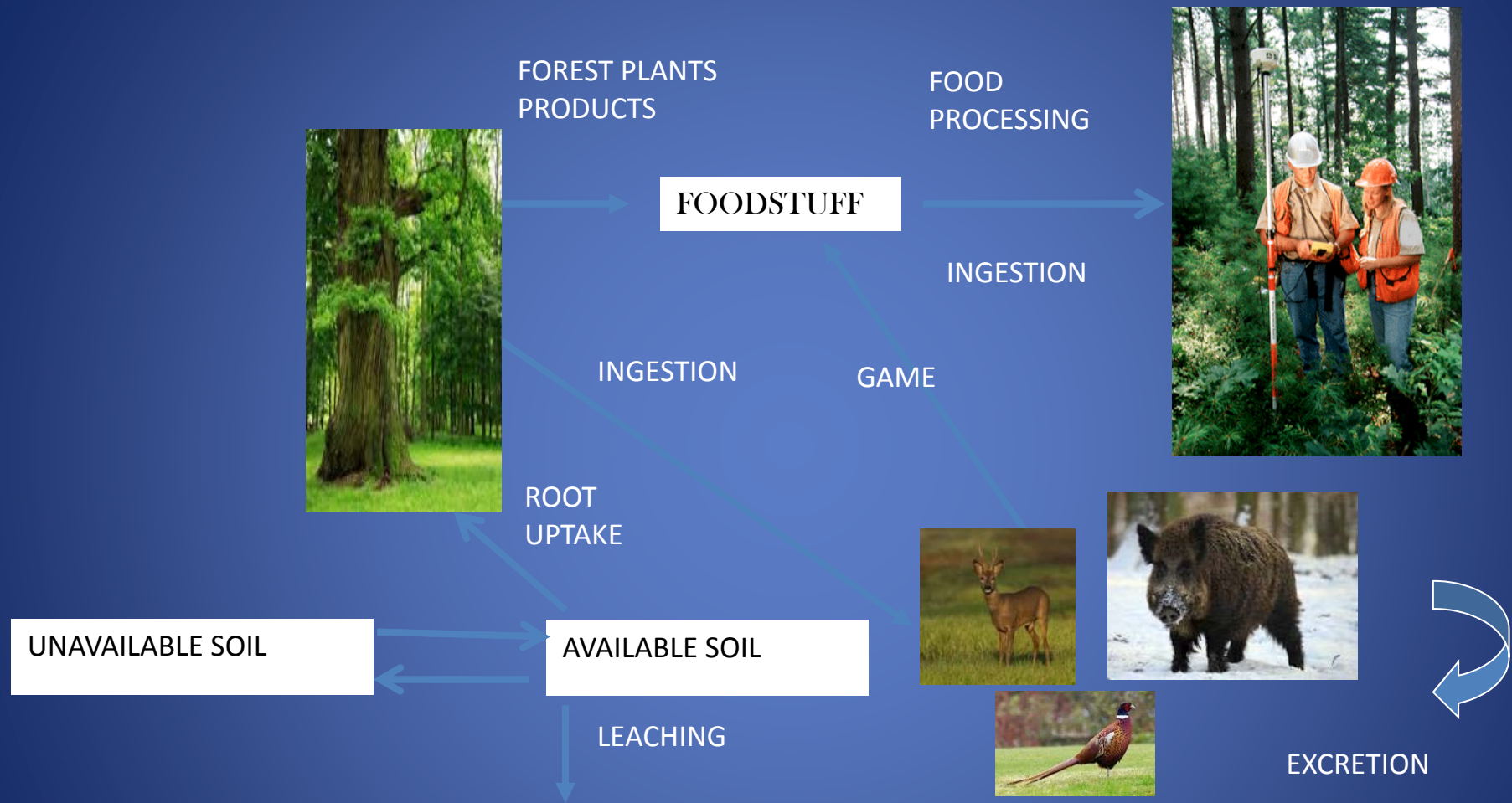
*S. Fesenko*

*Russian Research Institute of  
Radiology and Agroecology*

# Biogeochemical cycling (IAEA, 2002)



# Environmental pathways and processes





## *Main radioecological features of forest ecosystems*

- High typological diversity.*
- Comprehensive structure of trophic chains.*
- Multi-floor structure of vegetation cover.*
- Considerable accumulation of radionuclides in biomass.*
- Manifested heterogeneity properties of soil profile.*

### *As a result:*

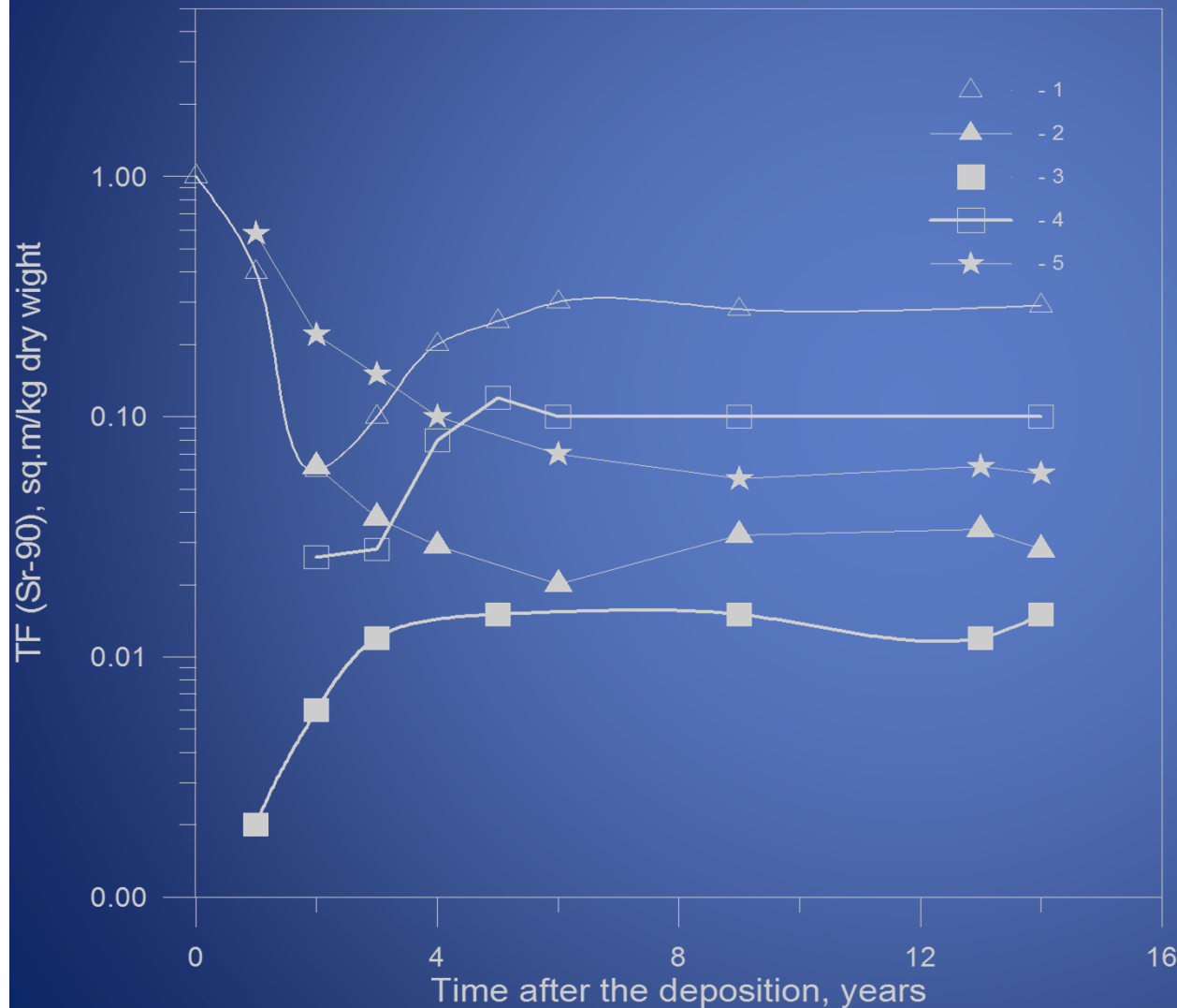
- High spatial variability of radionuclide levels in many forest components.*
- The contamination level of a specific forest compartments is being changed due to multiple factors and processes interacting in a very complex way.*

## *Factors governing variations in $^{137}\text{Cs}$ concentrations in forest plants*

*Available for transfer  $^{137}\text{Cs}$  in soil is a major source for time dependent radionuclide transfer to plants.*

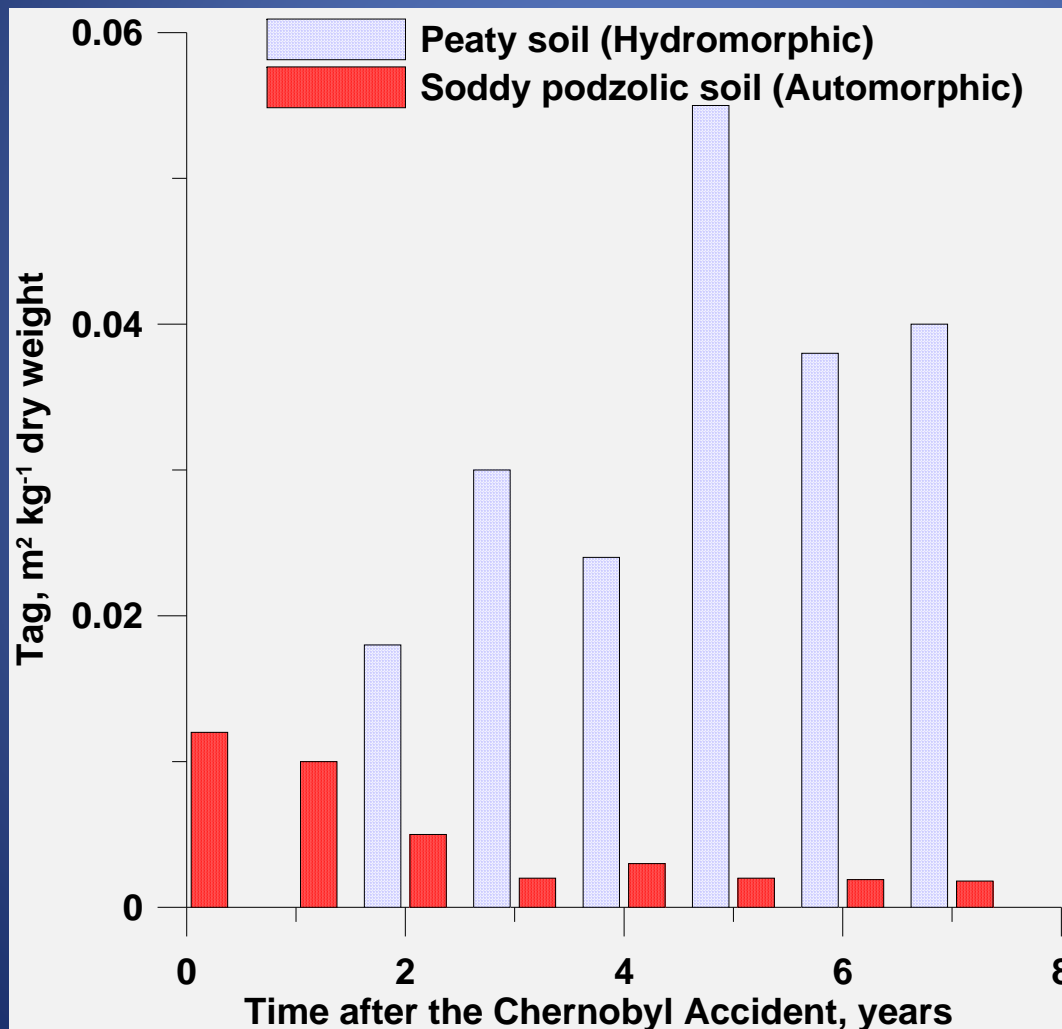
- *Features of forest soil:*
  - *Depth of soil genetic horizons*
  - *Soil/litter properties*
  - *Cs distribution in the top soil*
- *Specific features of plants:*
  - *distribution of fine roots within the top soil;*
  - *ability to accumulate Cs;*
  - *biomass of the plant compartments*

# Typical dynamics of $^{90}\text{Sr}$ activity concentrations in different components of birch forest after the Kyshtym accident

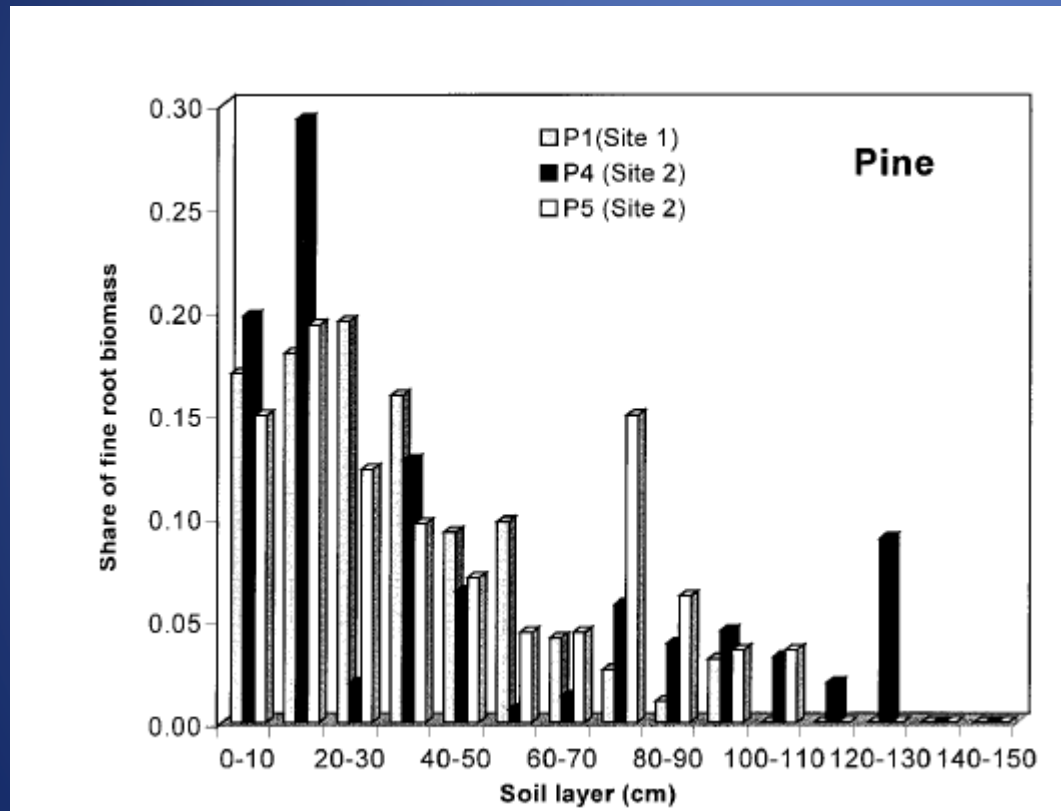


- 1 - leaves
- 2 - branches
- 3 - wood
- 4 - internal bark
- 5 - external bark

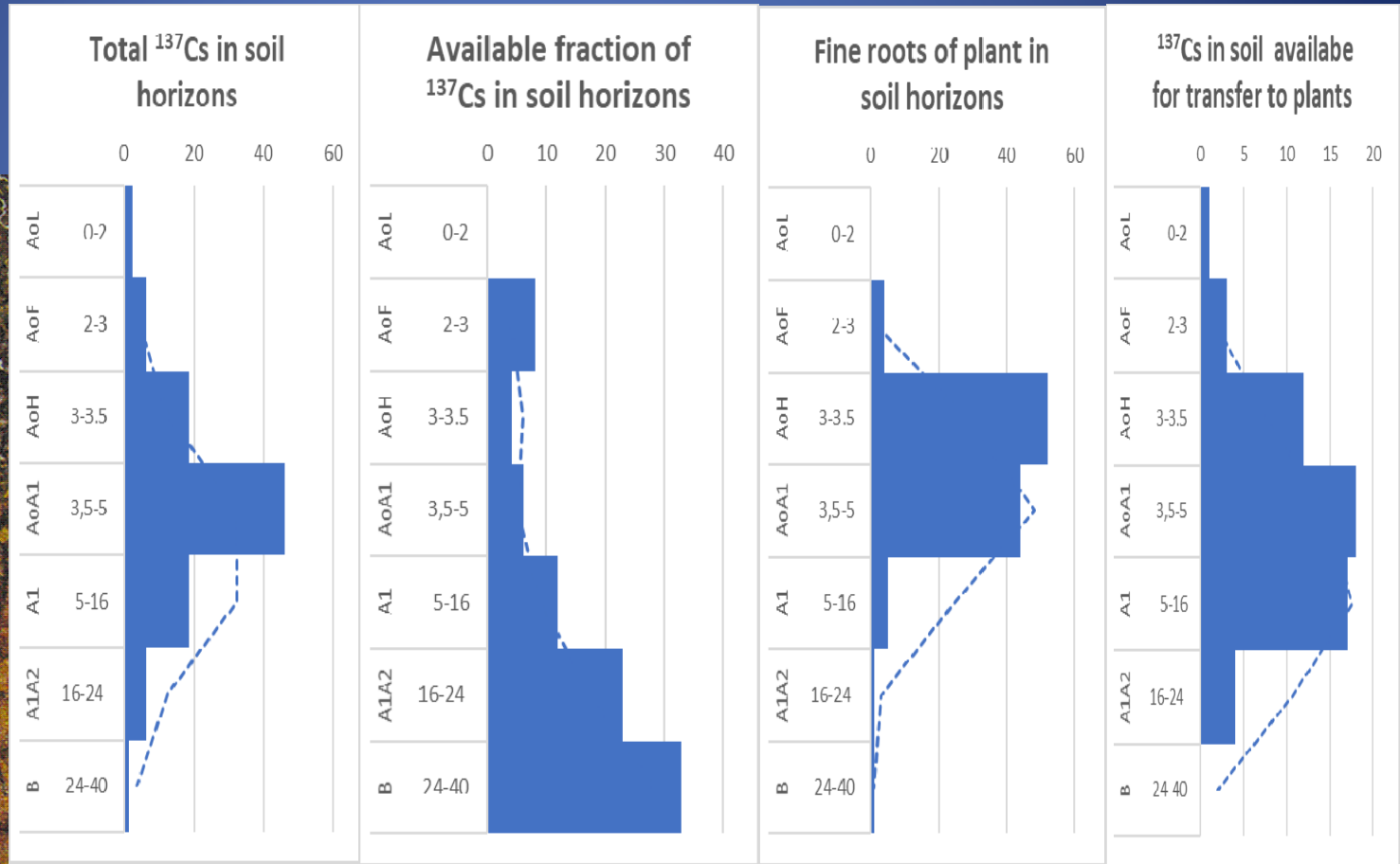
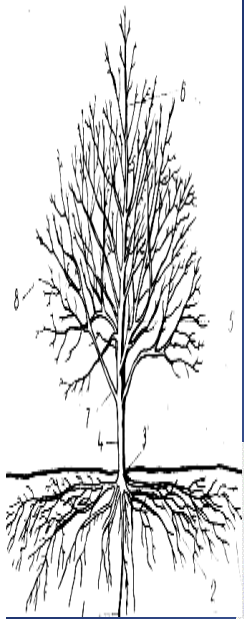
*Transfer of  $^{137}\text{Cs}$  to pine wood on soils with contrasting properties in areas affected by the Chernobyl accident*



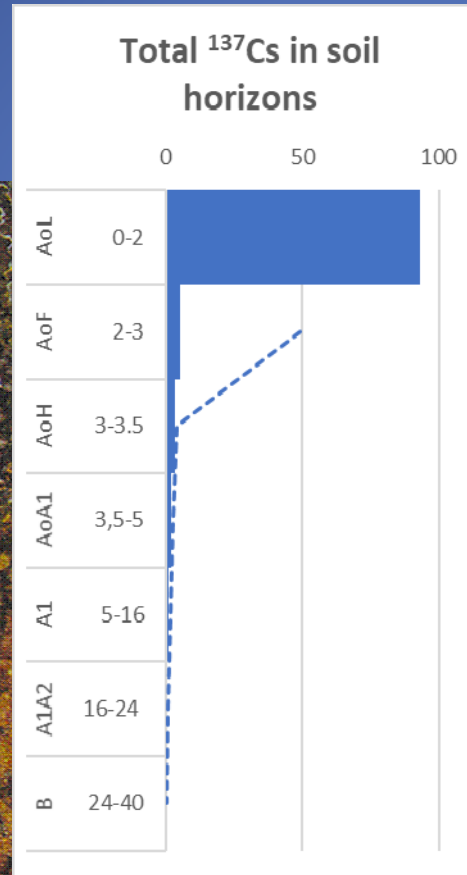
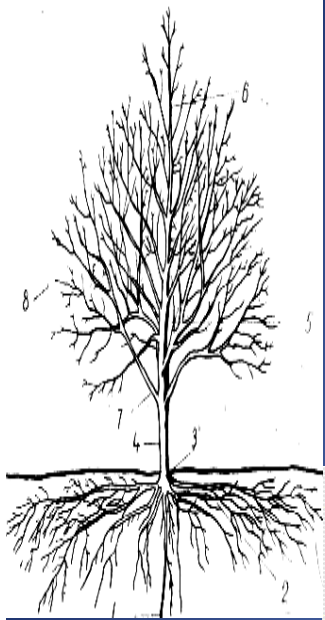
# *Distribution of fine roots in soil profile*



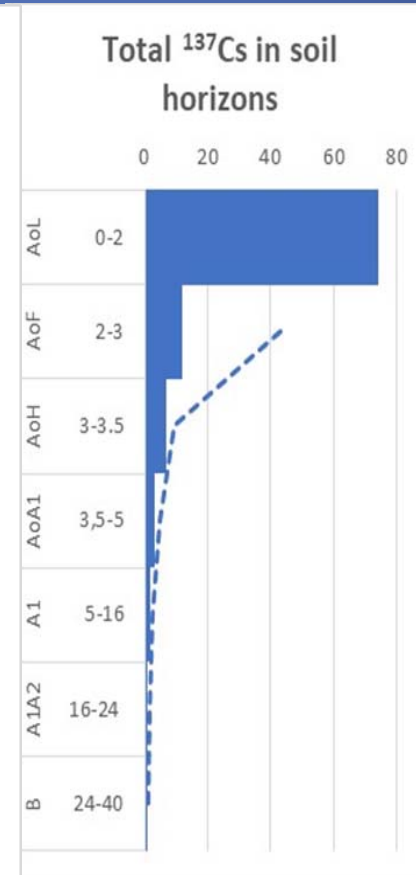
# Fraction of Cs available for root uptake



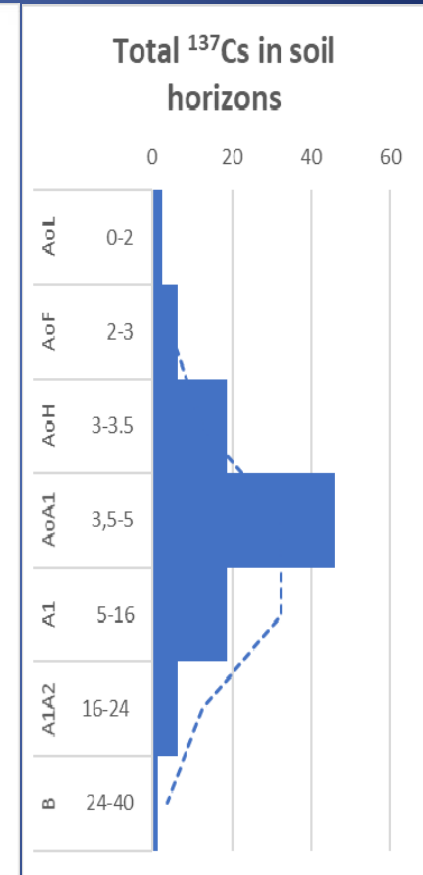
# Dynamics of Cs-137 distribution in soil profile



1987



1991



1999

## *Time dependent amount of $^{137}\text{Cs}$ that is available for transfer to plants*

$$BF^J(t) = \sum_i^N \delta_i(t) * q_i(t) * k_i^j$$

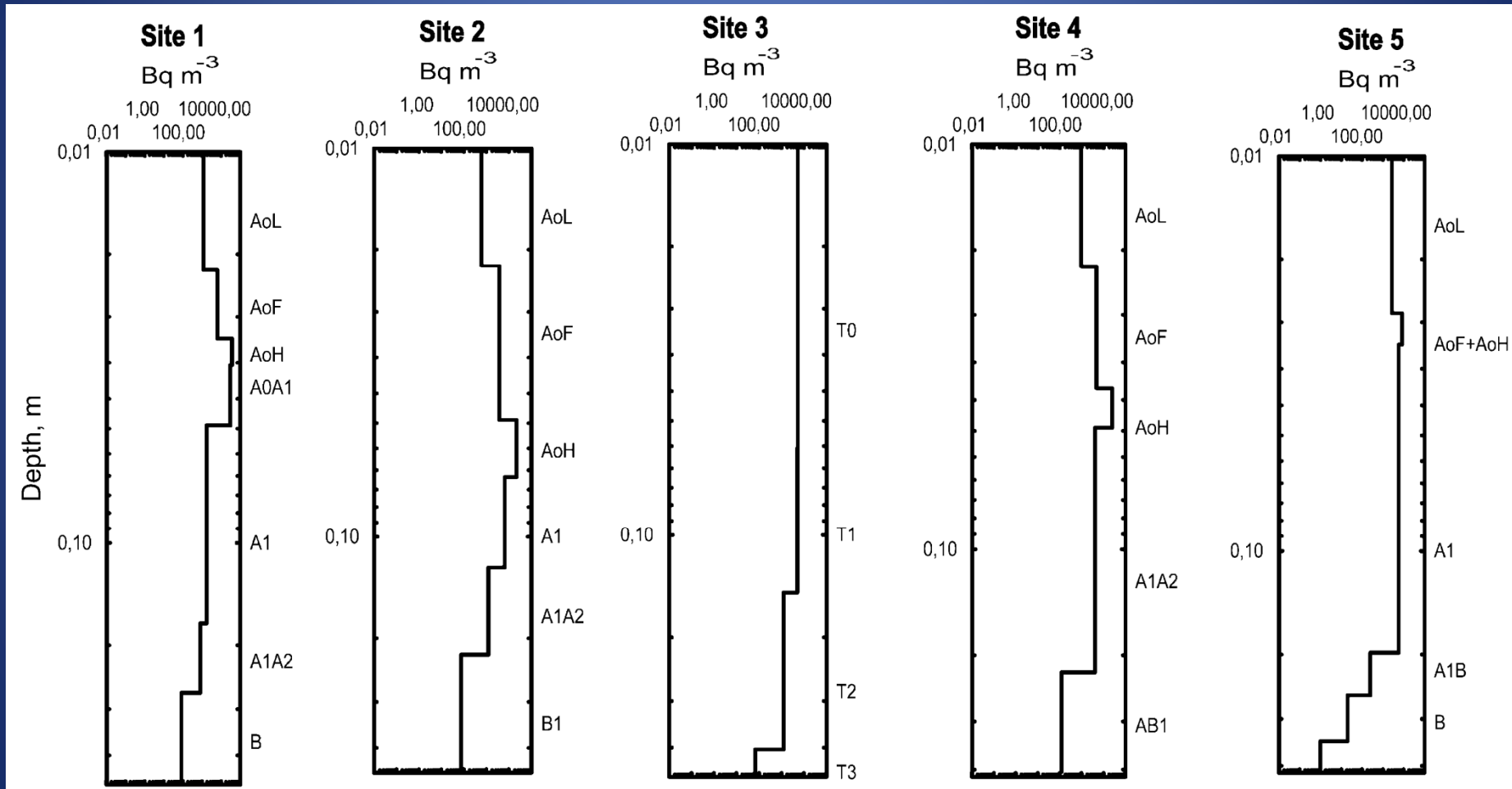
- $q_i$  is the fraction of the total  $^{137}\text{Cs}$  activity in the soil horizon  $i$ ;
- $\delta(t)$  is the fraction of exchangeable (or available)  $^{137}\text{Cs}$  in the  $i$ -th horizon;
- $k_i$  is the fraction of roots biomass of  $j$ -th species of plants in the  $i$ -th horizon;
- $N$  is the number of genetic litter-soil horizons of concern.



## *Cs-137 distribution in the soil profile*

Soil horizon	Depth of the layer (cm)	Percentage of the total activity (%)	Percentage of <sup>137</sup> Cs extracted by reagents (%)				
			H <sub>2</sub> O	AcNH <sub>4</sub>	1 N HCl	3 N HCl	Rest
Site 1							
AoL	0–2	2.4	7.5	8.0	6.8	7.9	69.9
AoF	2–3	6.4	0.26	3.9	4.0	10.2	81.6
AoH	3–3.5	18.7	0.08	3.2	4.3	11.6	80.9
AoA1	3.5–5	46.2	0.02	1.1	3.0	9.0	86.8
A1	5–16	18.8	0.17	11.1	7.8	14.9	66.0
A1A2	16–24	6.3	0.28	18.5	8.5	14.1	58.6
B	24–40	1.3	4.4	29.4	15.1	16.9	34.3
Site 2							
AoL	0–2	1.2	8.2	17.0	11.8	18.3	44.7
AoF	2–5	11.3	1.1	8.6	5.7	12.9	71.7
AoH	5–7	43.4	0.28	6.7	5.0	12.3	75.7
A1	7–12	32.9	0.04	3.1	1.4	2.1	93.3
A1A2	12–20	9.8	0.22	3.2	1.4	1.8	93.3
B1	20–40	1.5	2.3	16.1	5.1	12.2	64.4

# Distribution of available $^{137}\text{Cs}$ in soil profile



# *Environmental factors governing the extent of tree contamination by radiocaesium*

## *Influencing factors*

## *Examples of hierarchy for trees*

*Soil type*

*peat-gley > peat-podzolic > soddy-podzolic > podzolized chernozems*

*Moisture regime*

*central depression > terrace basement > terrace slope > slope upper part > watershed top*

*Stand composition*

*Monospecific coniferous stand > mixed coniferous-deciduous forest*

*Stand age*

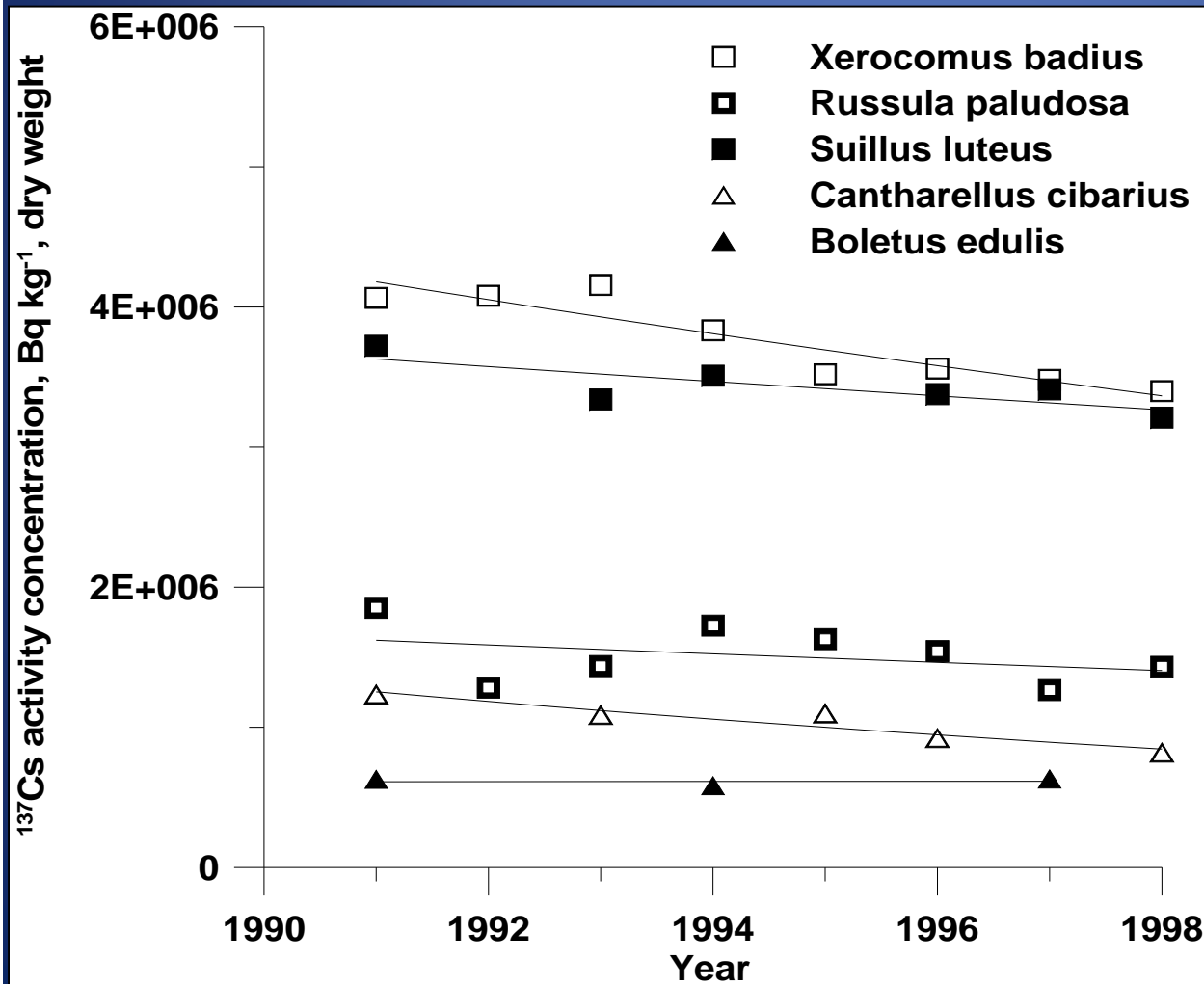
*0-30 > 30-60 > 60-90 > +90*

*Tree species*

*aspen > oak > birch > pine > lime > spruce*

<i>Mushroom species</i>	<i>Edibility and life mode of mushrooms</i>	<i>Caesium transfer factor (<math>m^2.kg^{-1}</math> dry weight)</i>		
		<i>GM<sup>(1)</sup></i>	<i>Min.</i>	<i>Max</i>
<i>Agaricus arvensis</i>	<i>Edible. Humus saprophytic</i>	$5 \times 10^{-3}$	$6 \times 10^{-4}$	$1 \times 10^{-2}$
<i>Amanita vaginata</i>	<i>Not edible. Symbiotic</i>	5	-	
<i>Boletus edulis</i>	<i>Edible. Symbiotic</i>	$9 \times 10^{-2}$	$4 \times 10^{-3}$	1.4
<i>Cantharellus cibarius</i>	<i>Edible. Symbiotic</i>	$2 \times 10^{-1}$	$1.5 \times 10^{-2}$	$7 \times 10^{-1}$
<i>Clitocybe nebularis</i>	<i>Not edible. Litter saprophytic</i>	$2 \times 10^{-1}$	-	
<i>Collybia butyracea</i>	<i>Not edible. Litter saprophytic</i>	-	$1 \times 10^{-1}$	$2 \times 10^{-1}$
<i>Coprinus comatus</i>	<i>Edible. Saprophytic</i>	$5 \times 10^{-3}$	$4 \times 10^{-4}$	$1.5 \times 10^{-2}$
<i>Lactarius sp.</i>	<i>Symbiotic</i>	3.9	$5 \times 10^{-1}$	9
<i>Suillus elegans or S. grevillei</i>	<i>Edible. Symbiotic</i>	$4 \times 10^{-1}$	$7 \times 10^{-2}$	$9 \times 10^{-1}$
<i>Tylopilus felleus</i>	<i>Not edible. Symbiotic</i>	2.5	$8 \times 10^{-1}$	8
<i>Xerocomus badius</i>	<i>Edible. Symbiotic</i>	1.3	$2 \times 10^{-3}$	7

# Dynamics of $^{137}\text{Cs}$ activity concentrations in selected mushroom species.

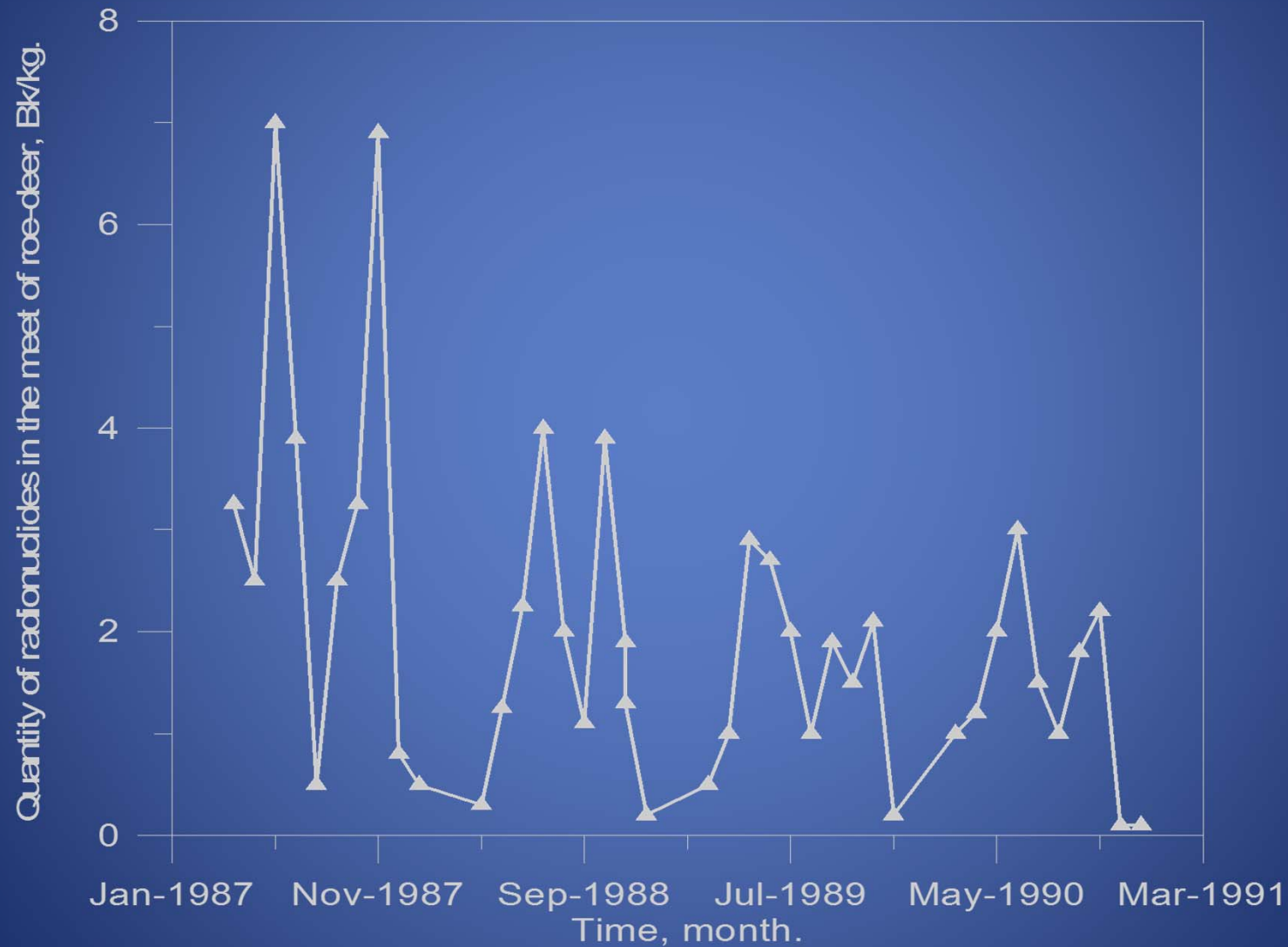


*Cs-137 soil deposition at the site in 1986 - 555 kBq m<sup>-2</sup>*

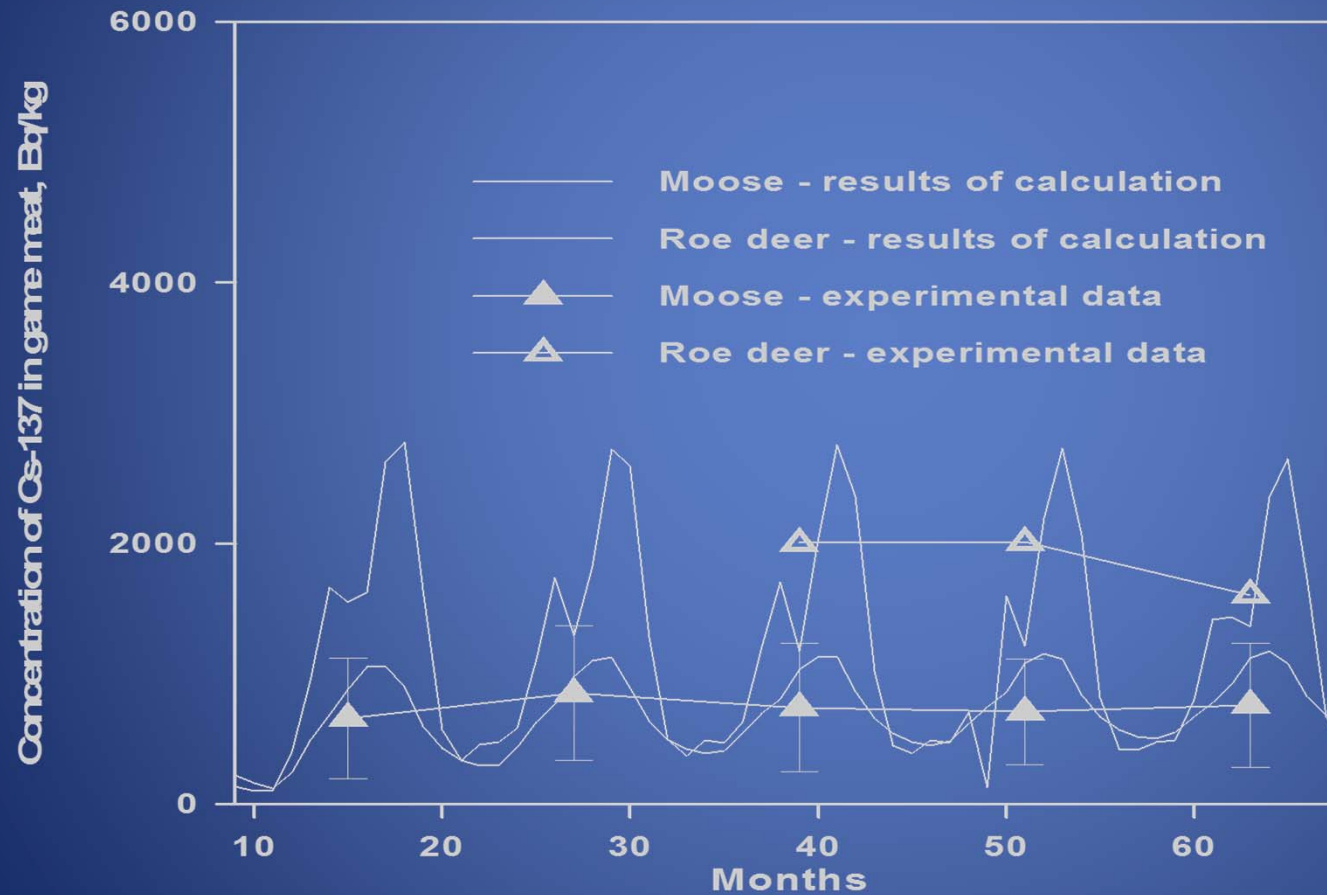
## *Factors governing variations in $^{137}\text{Cs}$ concentrations in wild animals*

- *Feeding ration of game (time dependent);*
- *Concentrations of  $^{137}\text{Cs}$  in feeds (time dependent)*
- *Specific features in metabolism: biological half lives;*
- *Availability of feeds?*

# *Dynamics of $^{137}\text{Cs}$ content in meat of roe deer in Southern Germany*

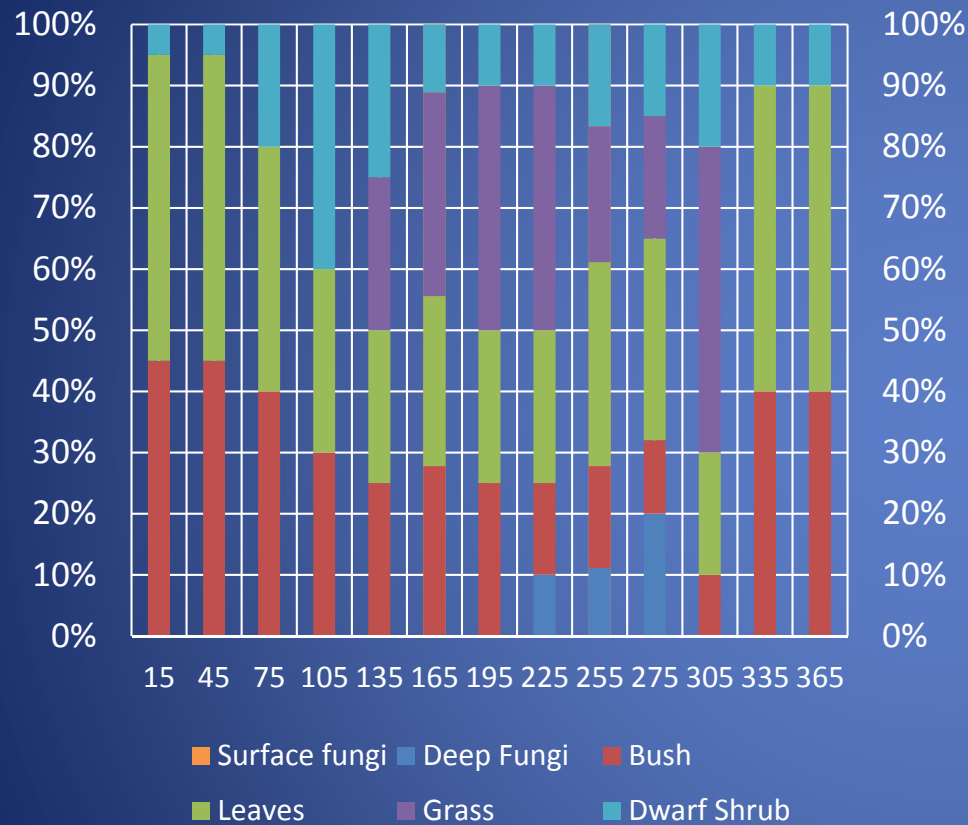


# *Dynamics of $^{137}\text{Cs}$ concentrations in muscles of roe deer and moose in Harbo area (Central Sweden)*



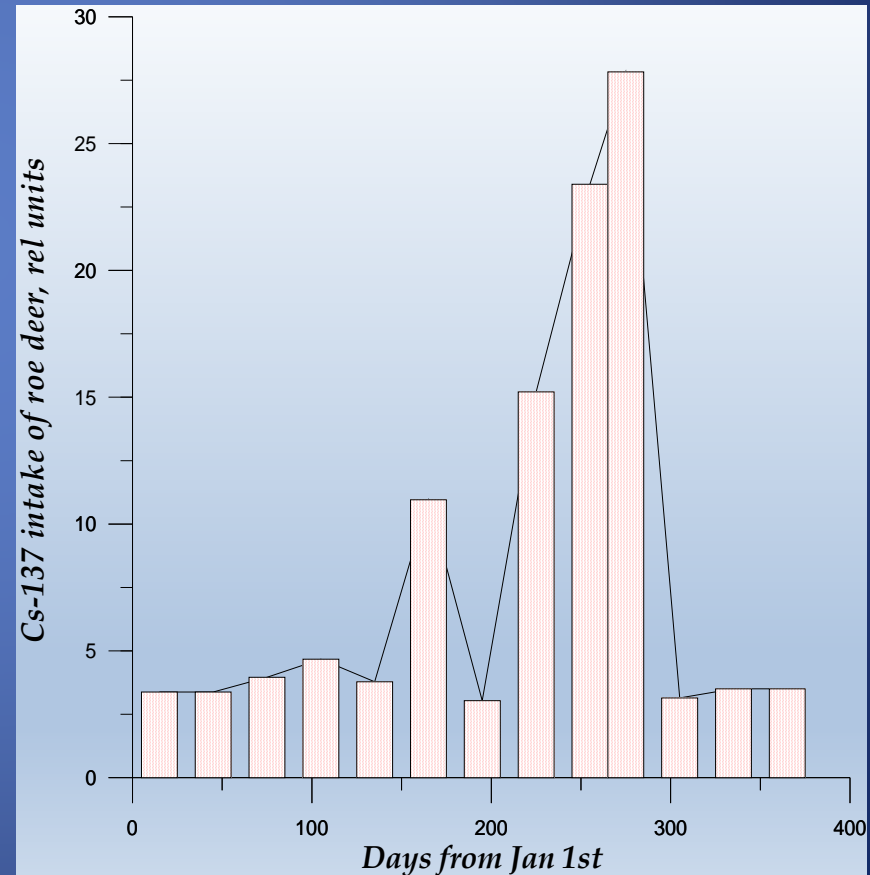


# Intake by roe deer: seasonal variations



*Fractions of individual feeds*

*Intake with feeds*



## *Half lives of $^{137}\text{Cs}$ in wild animal*

<i>Animal</i>	<i>Ecological half life, year</i>	<i>Biological half life. d</i>
Roe deer	6-7	10 -25
Rein deer	9.3-17.4	20-40
Chamois	7.7	9-15
Wild boar	> 10, no decrease	12-40

## *Concentrations of $^{137}\text{Cs}$ in muscles*

*wild boar > roe deer > chamois > deer*

# Outputs of the model oriented forest monitoring program can be found in:

Radiat Environ Biophys (2001) 40:105–113

© Springer-Verlag 2001

## ORIGINAL PAPER

S.V. Fesenko · N.V. Soukhova · N.I. Sanzharova  
R. Avila · S.I. Spiridonov · D. Klein · E. Lucot  
P.-M. Badot

### Identification of processes governing long-term accumulation of $^{137}\text{Cs}$ by forest trees following the Chernobyl accident



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Journal of Environmental Radioactivity 65 (2003) 19–28

www.elsevier.com/locate/jenvrad

JOURNAL OF  
ENVIRONMENTAL  
RADIOACTIVITY

### $^{137}\text{Cs}$ distribution among annual rings of different tree species contaminated after the Chernobyl accident



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Journal of Environmental Radioactivity 82 (2005) 143–166

www.elsevier.com/locate/jenvrad

JOURNAL OF  
ENVIRONMENTAL  
RADIOACTIVITY

### Decision making framework for application of forest countermeasures in the long term after the Chernobyl accident

S.V. Fesenko<sup>a,b,\*</sup>, G. Voigt<sup>a</sup>, S.I. Spiridonov<sup>a</sup>,  
I.A. Gontarenko<sup>a</sup>

<sup>a</sup>International Atomic Energy Agency, Agency's Laboratories Seibersdorf, 1400 Vienna, Austria

<sup>b</sup>Russian Institute of Agricultural Radiology and Radioecology, 249020 Obninsk, Russia



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The Science of the Total Environment 269 (2001) 87–103

the Science of the  
Total Environment

An International Journal of the Scientific Community  
for the Environment and its Relationship with Man

www.elsevier.com/locate/scitotenv

### $^{137}\text{Cs}$ availability for soil to understory transfer in different types of forest ecosystems

S.V. Fesenko<sup>a,\*</sup>, N.V. Soukhova<sup>a,c</sup>, N.I. Sanzharova<sup>a</sup>, R. Avila<sup>b</sup>,  
S.I. Spiridonov<sup>a</sup>, D. Klein<sup>c</sup>, P.-M. Badot<sup>c</sup>

on 249020,

du Pays de

### MODELLING OF $^{137}\text{Cs}$ BEHAVIOUR IN FOREST GAME FOOD CHAINS

S. FESENKO, S. SPIRIDONOV

Russian Institute of Agricultural Radiology and Agroecology (RIARAE),  
249020, Obninsk, Russia

R. AVILA

Radiat Environ Biophys (2000) 39:291–300

© Springer-Verlag 2000

## ORIGINAL PAPER

S.V. Fesenko · G. Voigt · S.I. Spiridonov  
N.I. Sanzharova · I.A. Gontarenko · M. Belli  
U. Sansone

### Analysis of the contribution of forest pathways to the radiation exposure of different population groups in the Bryansk region of Russia



# Behavior of radiocesium in irrigation water and its effect on brown rice

**Moono Shin Ph.D.**

Senior Researcher

Rice Production Group

Agricultural Radiation Research Center

Tohoku Agricultural Research Center NARO (NARO/TARC)

- **Introduction**
  - Background of research
  - Purpose of research
  - Standard limits for radiocesium in Japan
  - Relationship between radiocesium in brown rice and exchangeable potassium in soil
  
- **Materials and methods**
  - study area and sampling sites
  - Survey methods
  - Radiocesium analyses
  
- **Results and Discussion**
  
- **Conclusions**

## *Background of research*



On March 11, 2011, the Great East Japan earthquake and tsunami caused an accident at the Tokyo Electric Power Company's Fukushima Daiichi Nuclear Power Plant (FDNPP).

- Removal (Decontamination)
- Inflow prevention
- Diffusion prevention

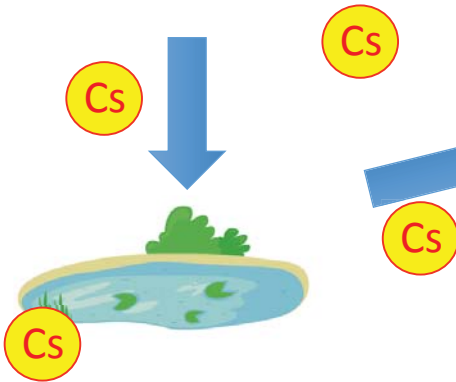
# Introduction

## Background of research

- (1) Is it contaminated again?
- (2) How much the impact on brown rice is there?

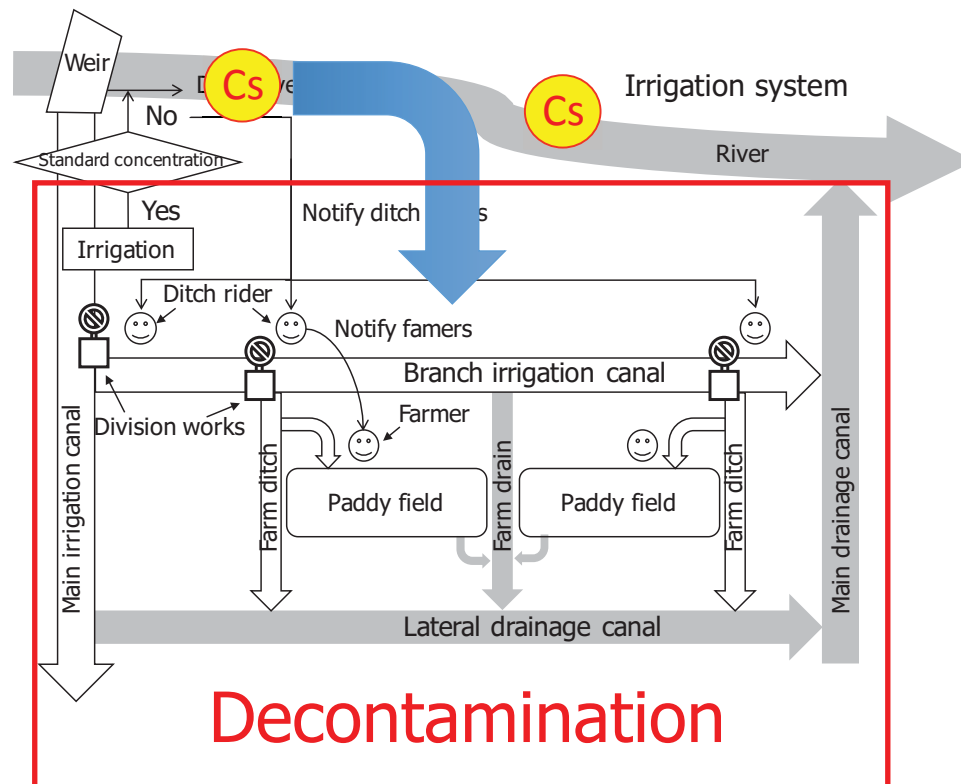


Forest



Irrigation pond

Not decontaminated yet





## *Purpose of research*

We conducted trials in decontaminated paddy fields with restricted cropping.

- Quantify the different forms of radiocesium in the irrigation water
- Investigate the dynamics of radiocesium in decontaminated paddy fields
- Quantify the mass balance of radiocesium in the paddy fields by analyzing the water balance
- Clarify the effect of the radiocesium in irrigation water on the radiocesium concentration in brown rice

## Standard limits for radiocesium in Japan

Provisional regulation values  
for radioactive iodine and cesium  
Date of enforcement: March 17, 2011

Nuclide	Category	Limit
Radioactive iodine (Representative radio-nuclides among mixed radionuclides: <sup>131</sup> I)	Drinking water	300
	Milk, Dairy products <sup>1)</sup>	
	Vegetables (Except root vegetables and tubers)	2,000
Fishery products		
Radioactive cesium <sup>2)</sup> ( <sup>134</sup> Cs plus <sup>137</sup> Cs)	Drinking water	200
	Milk, Dairy products	
	Vegetables	500
	Grains	
	Meat, Eggs, Fish, etc.	

- 1) Provide guidance so that materials exceeding 100 Bq/kg are not used in milk supplied for use in powdered baby formula or for direct drinking.  
2) These values take into account the contribution of radioactive strontium.

Operational intervention level : < 5 mSv/ year

Standard limits for radiocesium  
Date of enforcement  
:April 1, 2012

Category	Limit
Drinking water	10
Milk	50
Infant foods	50
General foods	100

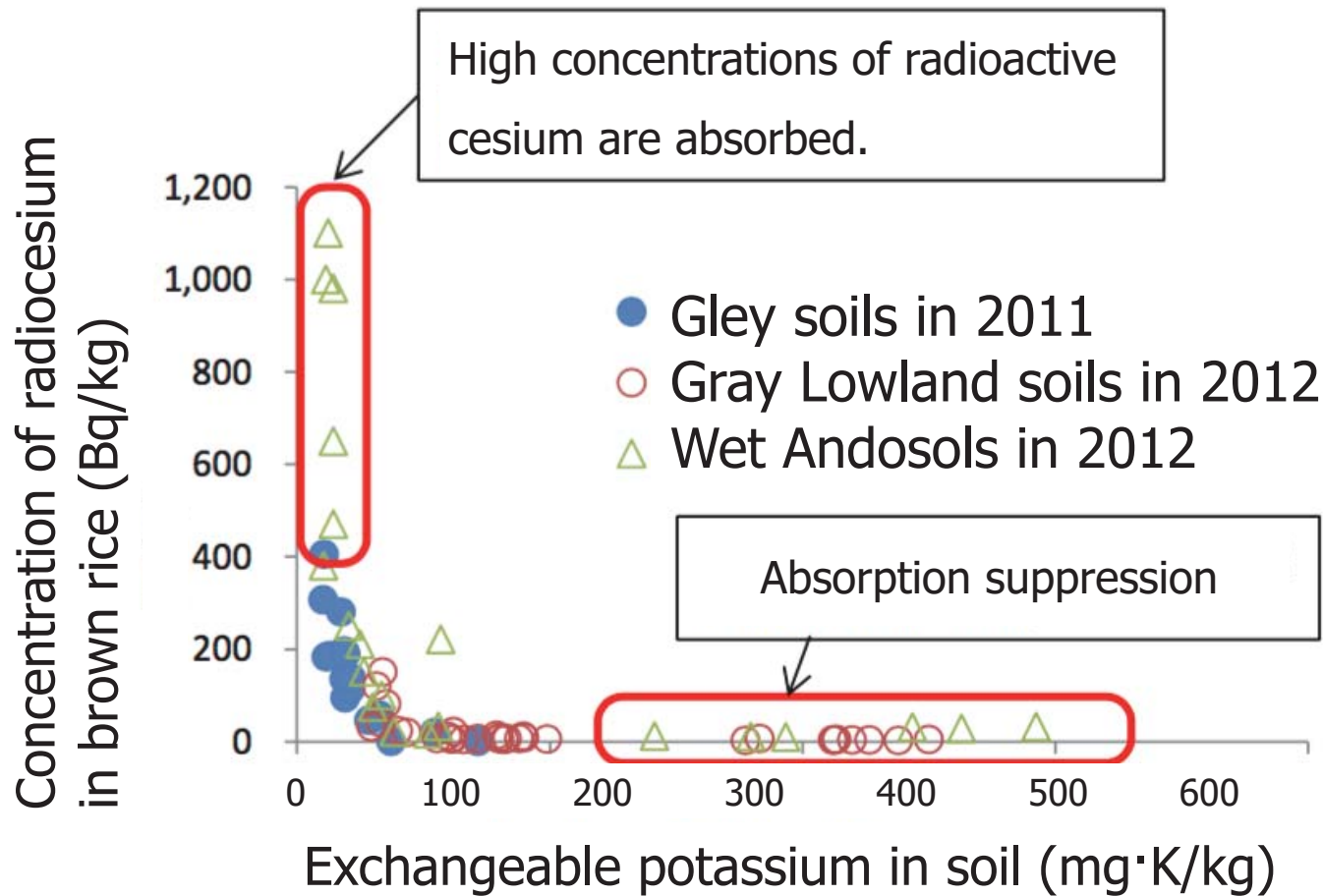
3) These limits take into account the contribution of Sr-90, Pu, and Ru-106.

Operational intervention level : < 1 mSv/ year

MHLW: [http://www.mhlw.go.jp/shinsai\\_jouhou/dl/shokuhin.pdf](http://www.mhlw.go.jp/shinsai_jouhou/dl/shokuhin.pdf)  
[http://www.mhlw.go.jp/english/topics/2011eq/dl/new\\_standard.pdf](http://www.mhlw.go.jp/english/topics/2011eq/dl/new_standard.pdf)

# Introduction

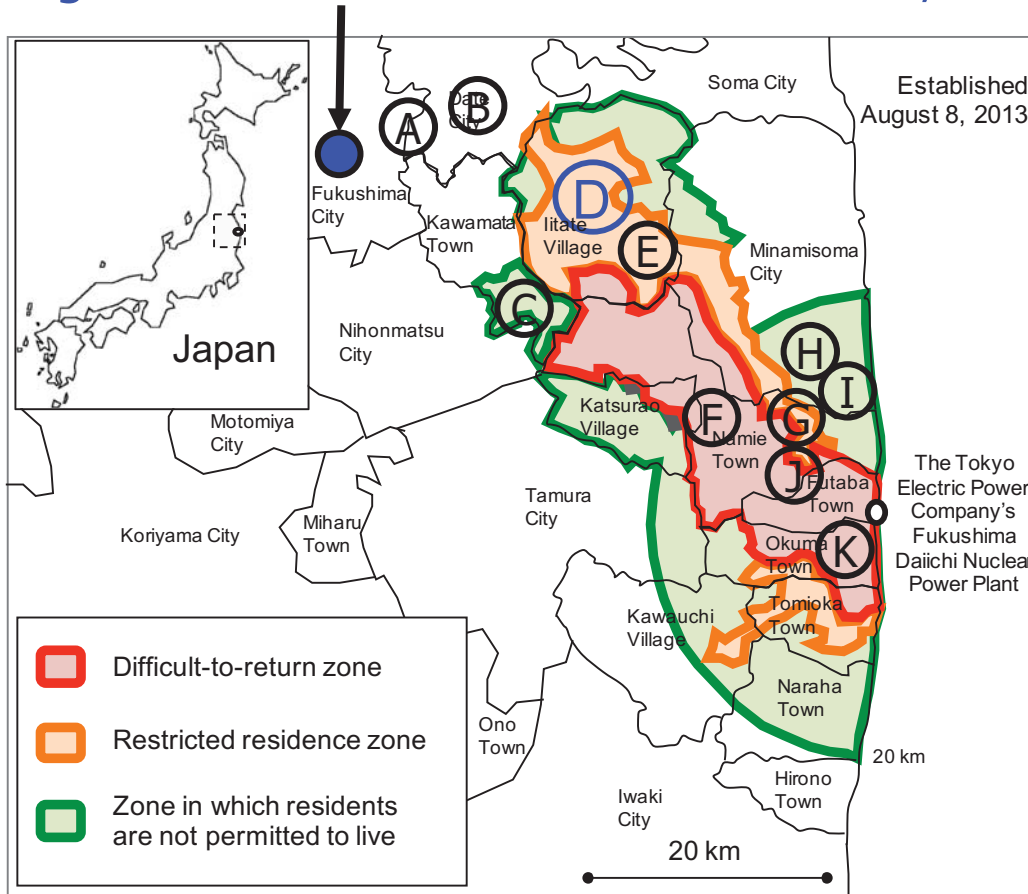
## *Relationship between radiocesium in rice and exchangeable potassium in soil*



# Materials and methods

## *Study area and water sampling sites*

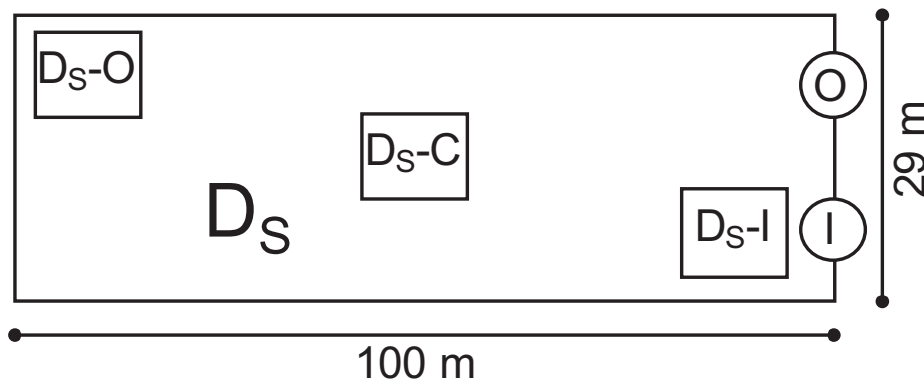
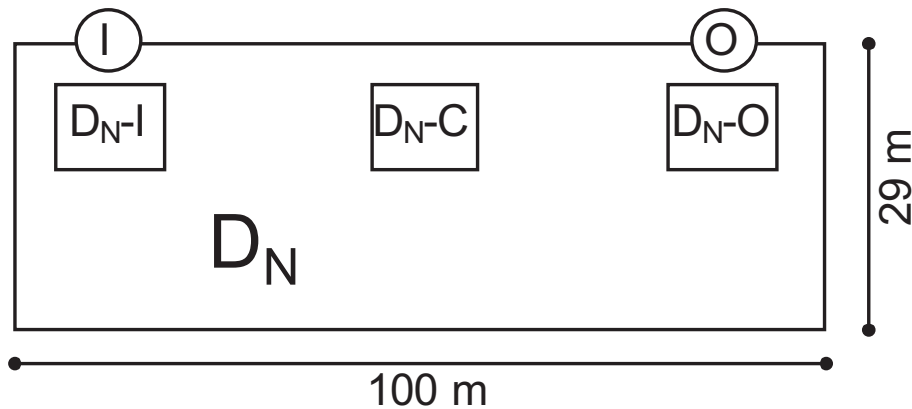
Agricultural Radiation Research Center, NARO/TARC



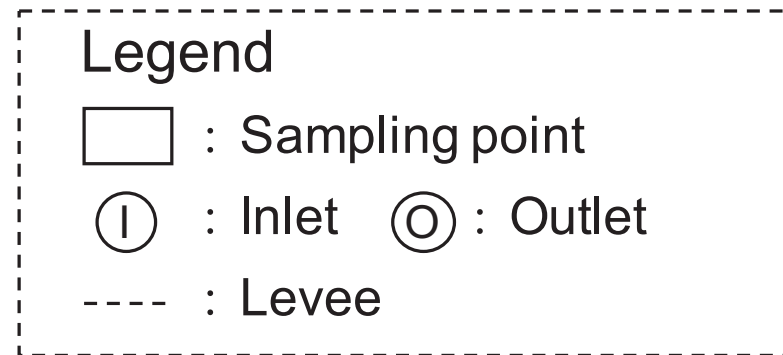
- 40 km northwest of FDNPP
- Decontamination of topsoil removal
- Cultivation trials in two paddy fields
- potassium level to 274 (mg·K)/kg soil

# Materials and methods

## Survey methods

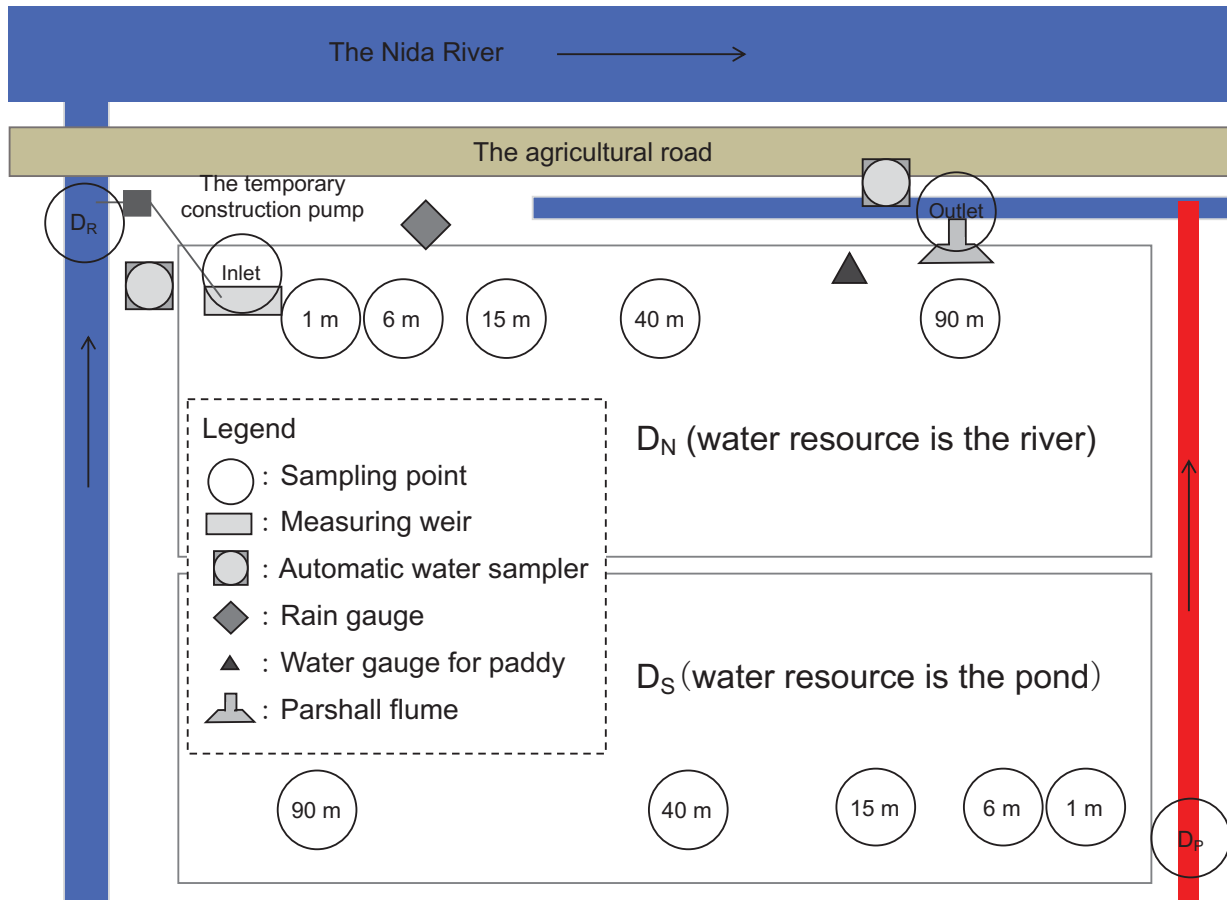


- Irrigation water
- Soil
- Brown rice



Soil and brown rice quadrat sampling sites

## Survey methods



- Irrigation water
- Ponding water
- Drainage water

Water sampling points and irrigation equipment at site D

# Materials and methods

## Survey methods



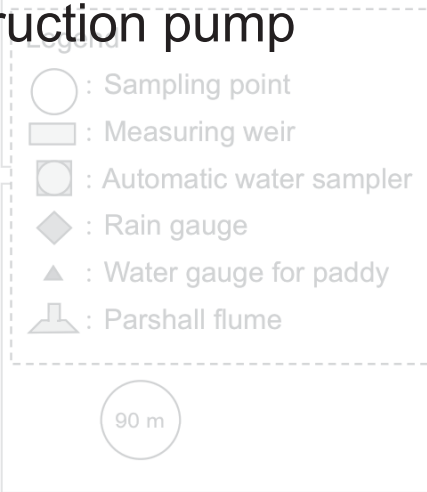
The temporary construction pump



Automatic water sampler



Parshall flume



Water gauge for paddy



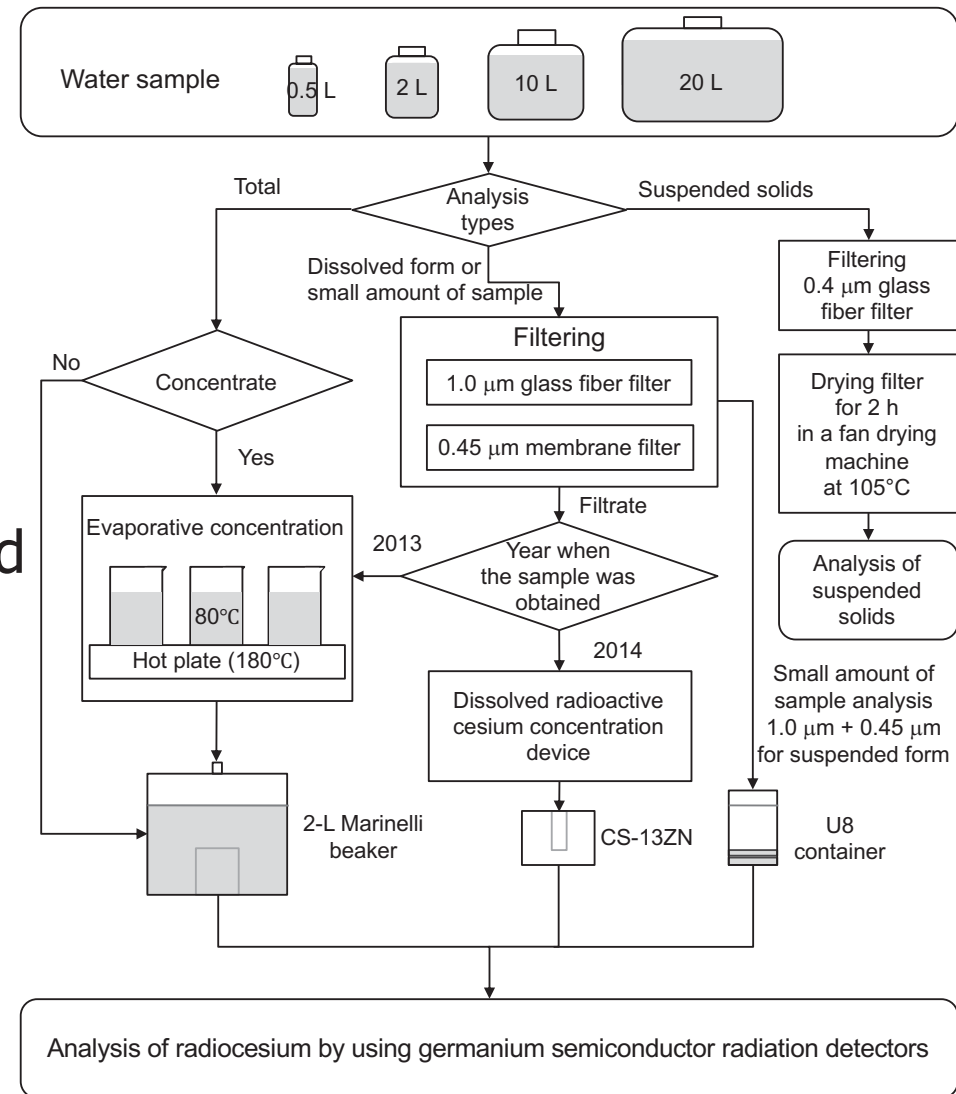
Rain gauge

Water sampling points and irrigation equipment at site D

# Materials and methods

## *Radiocesium analyses*

- Analysis water sample
  - Total radiocesium
  - Dissolved radiocesium
- Pre-concentration
  - Evaporative concentration
  - Nonwoven fabric impregnated with Prussian blue
  - Raddisk
- Analyses  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ 
  - GC4020-7500SL, Canberra
- Analyses of suspended solids and exchangeable potassium



Procedures for laboratory analysis



# Materials and methods

## *Radiocesium analyses*



Evaporative concentration



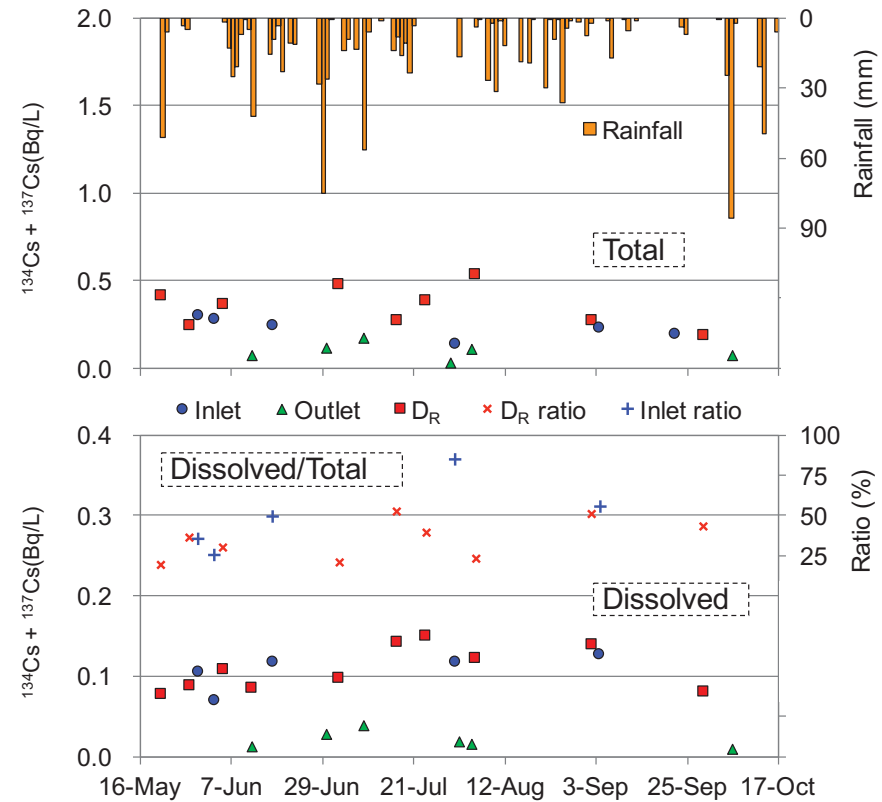
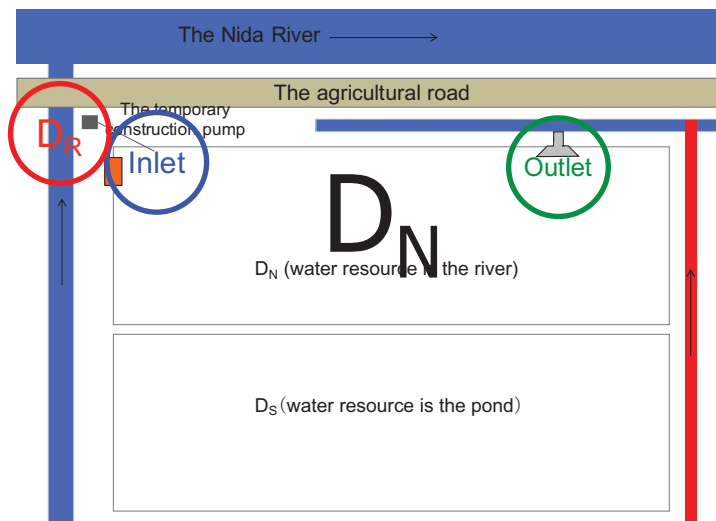
Nonwoven fabric impregnated  
With Prussian blue



Raddisk

# Results and discussion

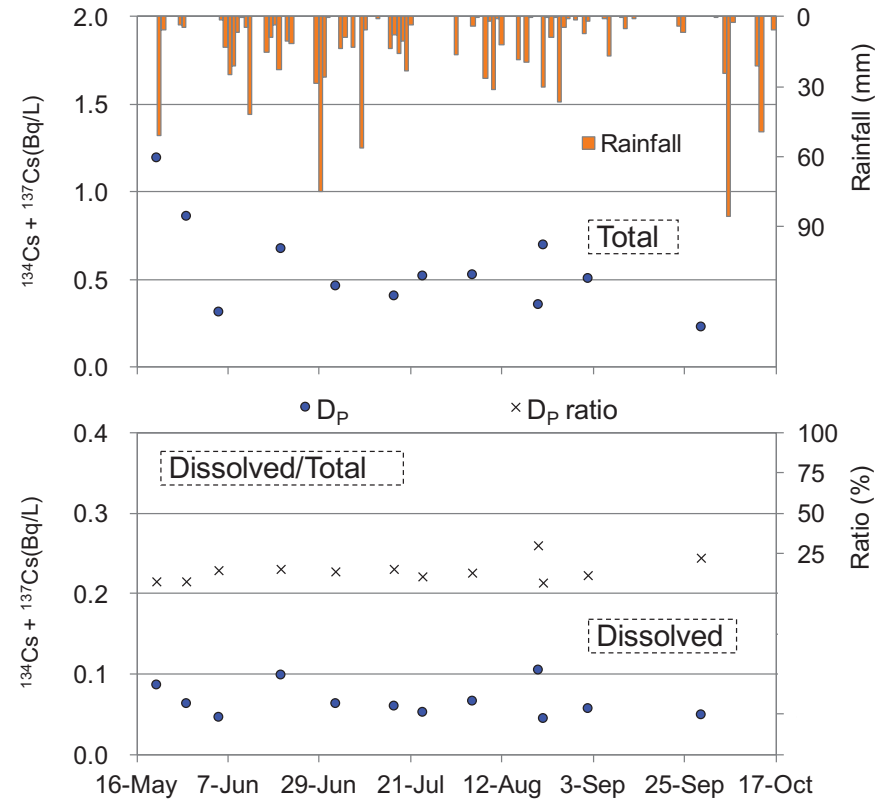
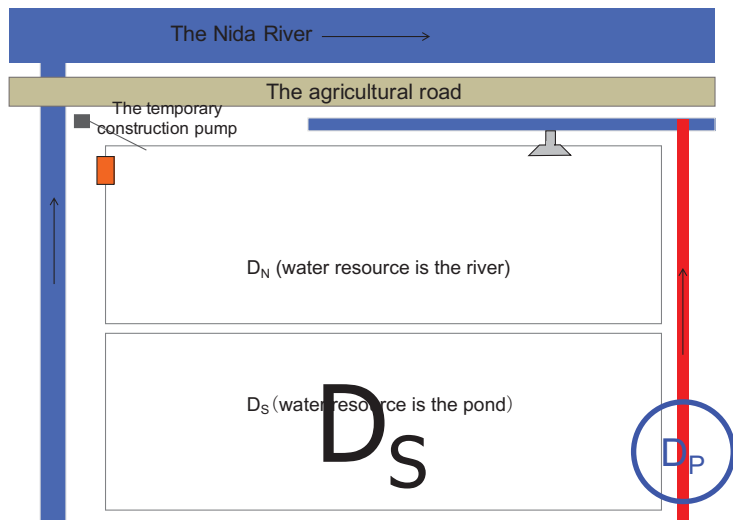
## *Temporal changes in the radiocesium concentration in irrigation water*



Concentrations of radiocesium in  $D_R$  (the irrigation water from the river at site D), inlet and outlet at site  $D_N$  (the northern part of site D) during the irrigation period in 2014

# Results and discussion

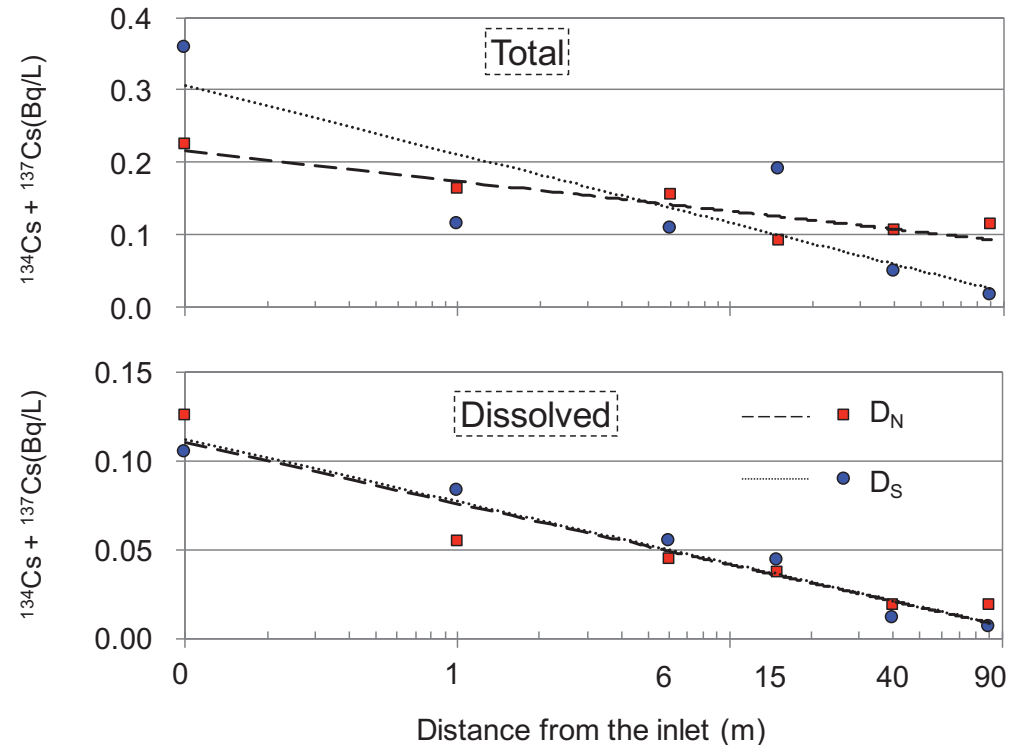
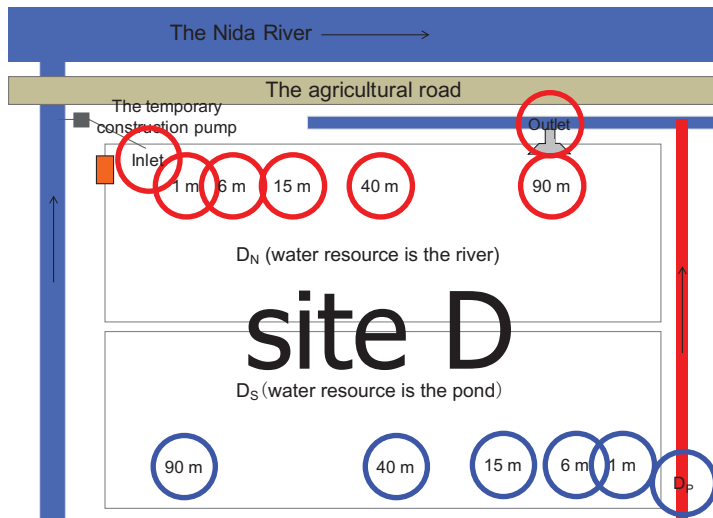
## *Temporal changes in the radiocesium concentration in irrigation water*



Concentrations of radiocesium in  $D_P$  (the irrigation water from the pond at site D) at site  $D_S$  (the southern part of site D) during the irrigation period in 2014

# Results and discussion

## *Spatial dynamics and form changes of radiocesium in irrigation water*



Concentrations of radiocesium in the irrigation water in 2014 at various distances from the water inlet at sites  $D_N$  and  $D_S$  (the northern and southern parts of site D, respectively)

# Results and discussion



## *Radiocesium concentration in soil and brown rice*

	$D_N$				$D_S$			
	Ex-K (mg·K)/kg	Soil Bq/kg	Rice Bq/kg	Transfer Factor (TF)	Ex-K (mg·K)/kg	Soil Bq/kg	Rice Bq/kg	Transfer Factor (TF)
Inlet	560	810	1.64	0.0020	245	377	1.75	0.0046
Center	449	1087	1.65	0.0015	240	390	2.67	0.0068
Outlet	415	898	1.32	0.0015	322	501	1.51	0.0030
Average	475	932	1.54	0.0017	269	423	1.98	0.0047

Amount of exchangeable potassium contents in the soil, and concentrations of radiocesium (Bq/kg) in the soil and brown rice after cultivation trials in 2014

# Results and discussion



## *Quantification of the radiocesium balance in the paddy fields*

Site	$D_N$	$D_S$
Area (m <sup>2</sup> )	2900	2900
(1) Average (Bq/L) Total (Dissolved)	(a) 0.288 (0.108)	0.562 (0.064)
(2) Inflow [Bq/(m <sup>2</sup> ·year)] Total (Dissolved)	(b) 95.3 (37.2)	(b) ÷ (a) × (1) 186.0 (22.0)
(3) Outflow [Bq/(m <sup>2</sup> ·year)] Total (Dissolved)	(c) 14.7 (3.2)	(c) ÷ (b) × (2) 28.7 (1.9)
Balance [Bq/(m <sup>2</sup> ·year)] (2)-(3) Total (Dissolved)	80.6 (34.0)	157.3 (20.1)

Estimated volume of radiocesium balance in irrigation water in 2014

In the present study, we quantitatively analyzed the balance of radiocesium in irrigation water and monitored its dynamics two decontaminated paddy field sites within 40 km of FDNPP.

## Our key findings were as follows

- Most of the radiocesium in the irrigation water flowing into the paddy fields accumulated near the inlet.
- The total and dissolved amount of radiocesium in the irrigation water flowing into the paddy fields were generally  $<200$  Bq/m<sup>2</sup>/year and  $<40$  Bq/m<sup>2</sup>/year, respectively.
- The impact on brown rice from radiocesium in the irrigation water was limited. In 2014, the concentration of radiocesium in brown rice was generally  $<3.0$  Bq/kg at the investigated paddy fields.

# Acknowledgments

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This study presents part of the results (2013–2014) from the project “Development of Decontamination Technologies for Radioactive Substances in Agricultural Land” commissioned by the Ministry of Agriculture, Forestry and Fisheries. We gratefully acknowledge the help of many people during our research.

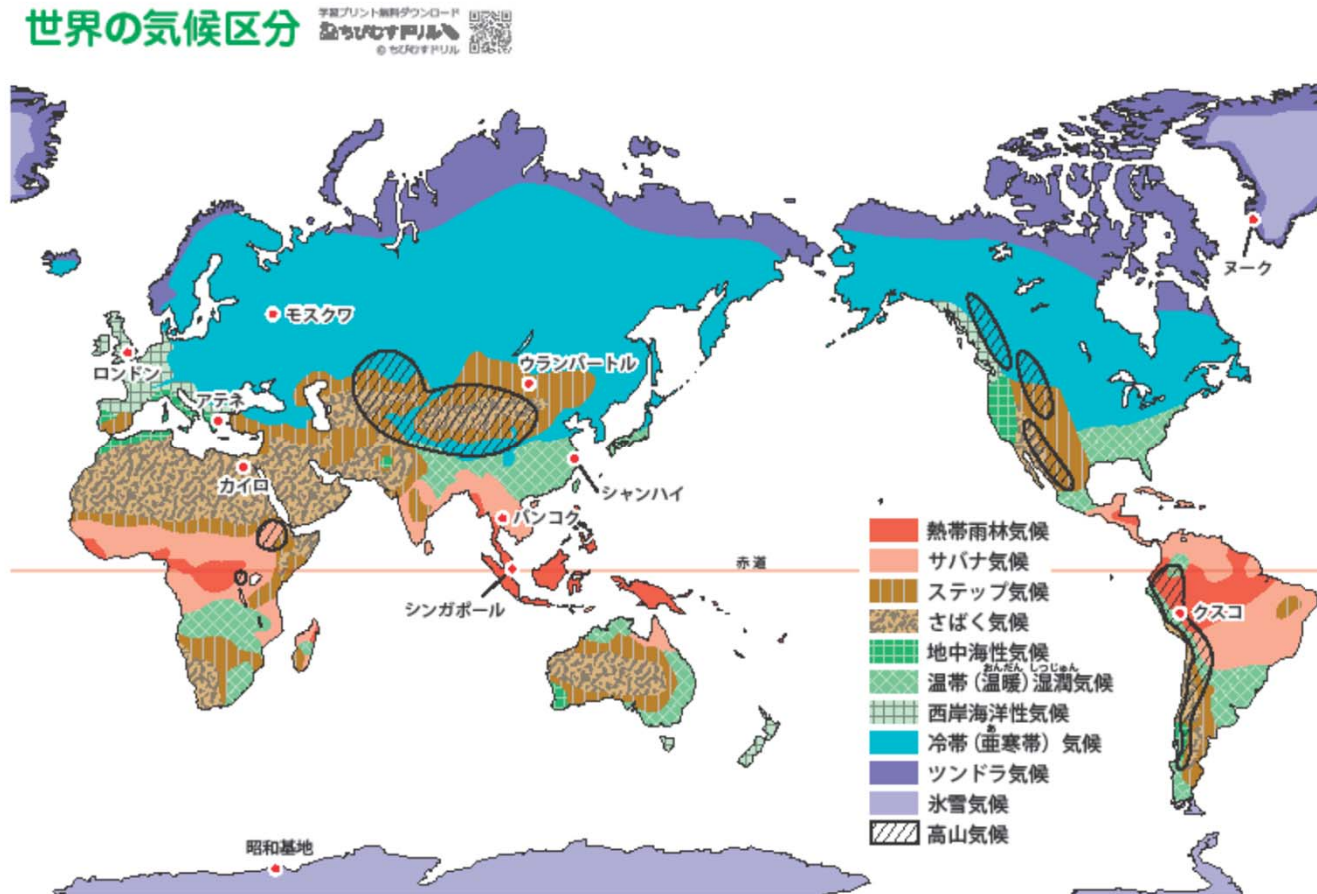
**MAFF**

Ministry of Agriculture, Forestry and Fisheries





Do we now know enough about radiocaesium in terrestrial ecosystems?



B Howard

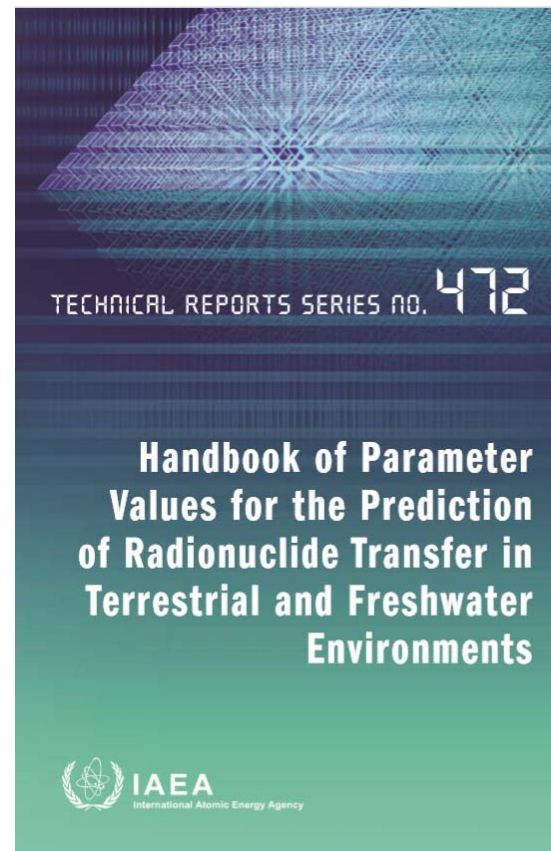
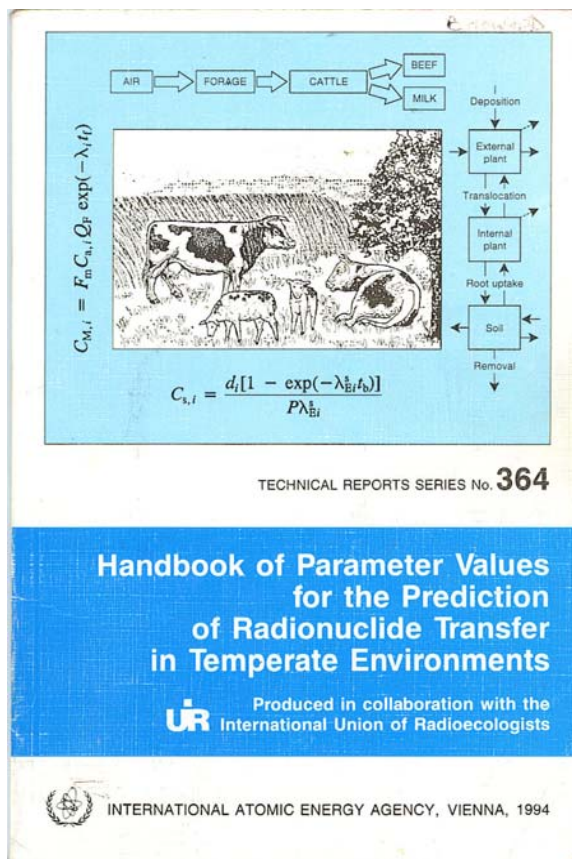
University of Nottingham, UK



## Why is radiocaesium important?

- Component of atmospheric weapons tests fallout
- Planned releases from nuclear facilities
- Component of releases after accidents
- Environmentally mobile
- Significant external doses
- Long-lived contributor to effective dose for humans and also important for other organisms

# IAEA publications compiling data



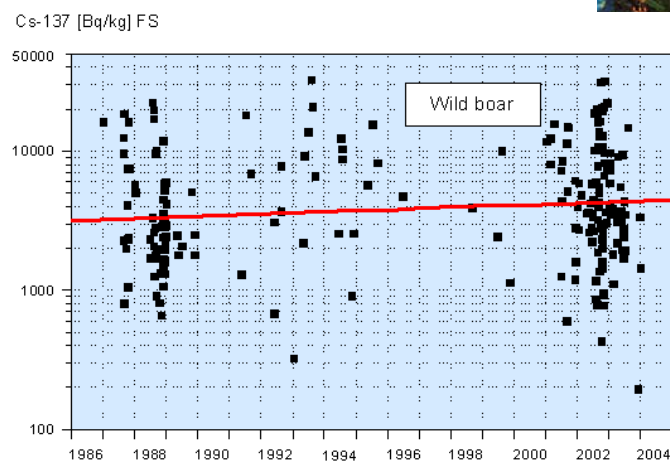
*Analysis of radioecological data in IAEA Technical Reports Series publications to identify key radionuclides and associated parameter values for human and wildlife assessments*

*Report of Working Group 4 of the Uncertainties and Variability in Data and Modelling for Radiological Impact Assessments (MODARIA) Programme*

# Post-Chernobyl Cs knowledge

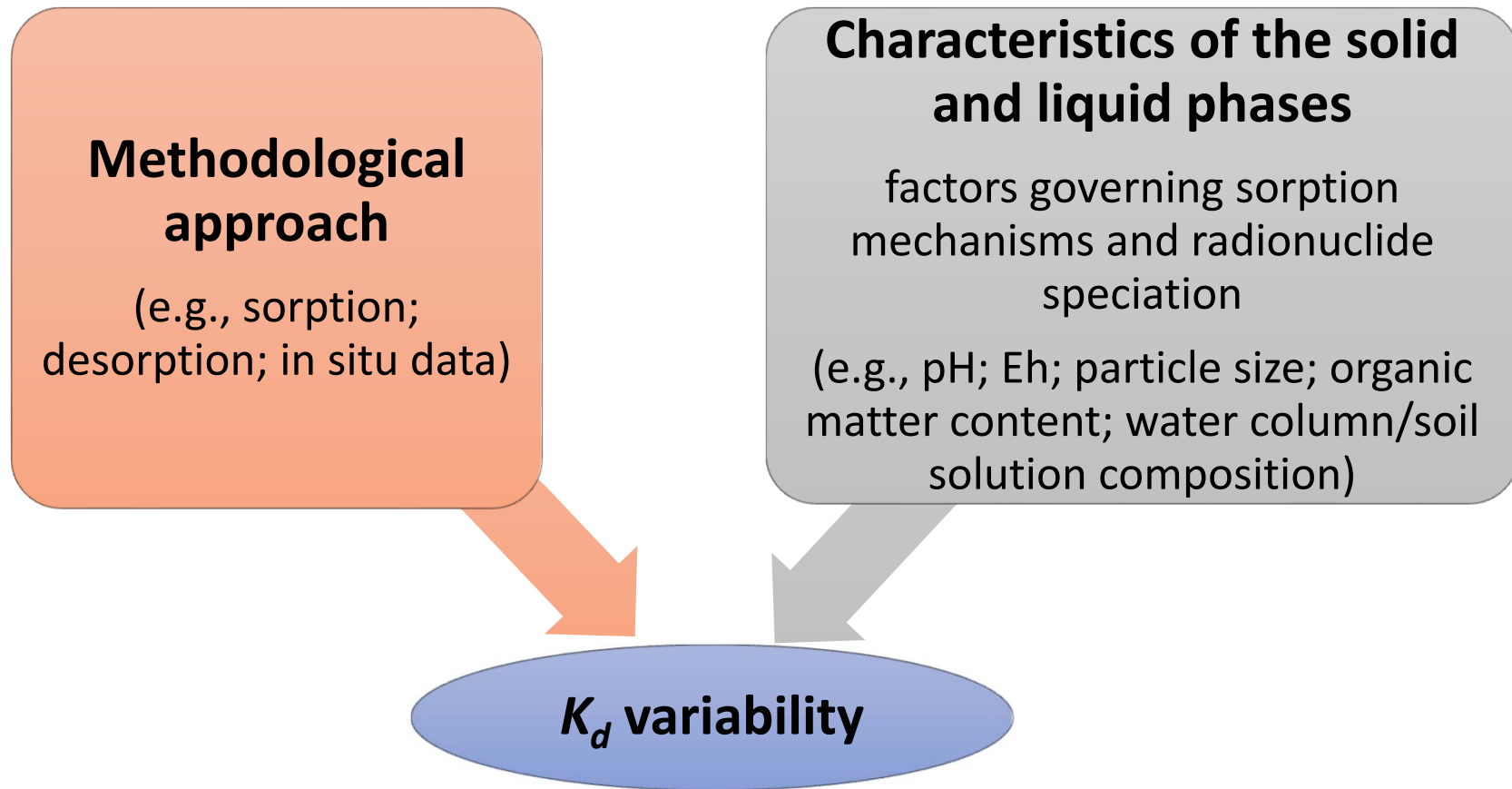
Importance of :

- radiocaesium density deposition initially
- soil type in short, medium and longer term
- Production systems in affected area (eg. animal production)
- Free food collected from forests and other semi-natural systems
- Long effective half-lives



# Approach to $K_d$ in MODARIA

$$K_d = C_{solid} / C_{liquid}$$



(Vidal et al MODARIA report in press)

# Radiocaesium $K_d$ data in TRS 472/TECDOC 1616

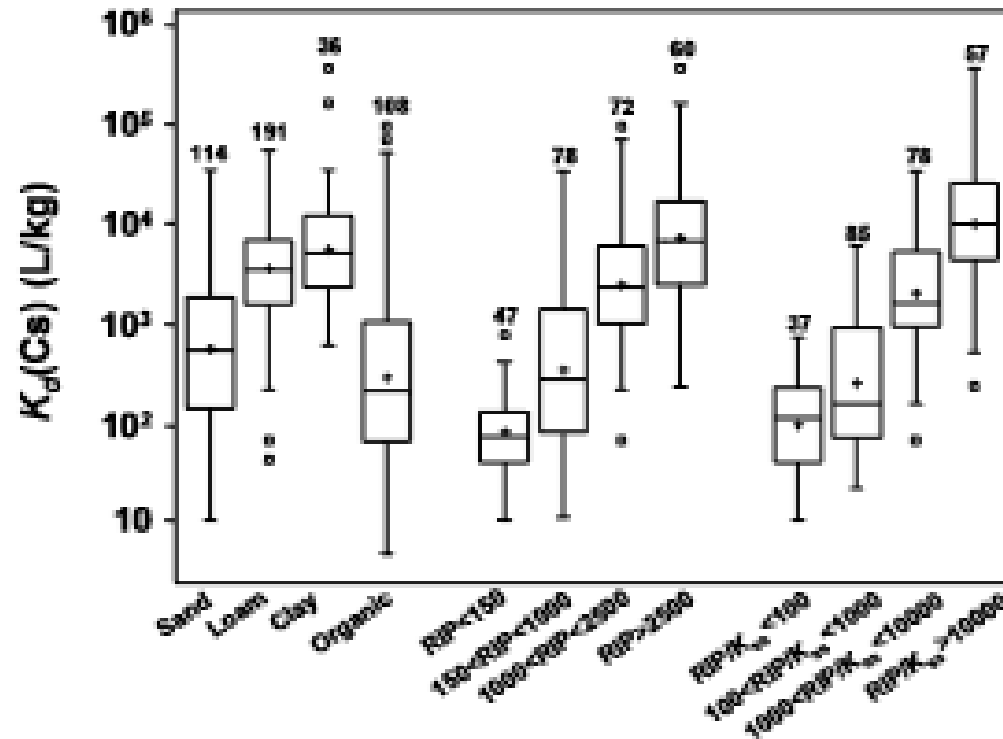
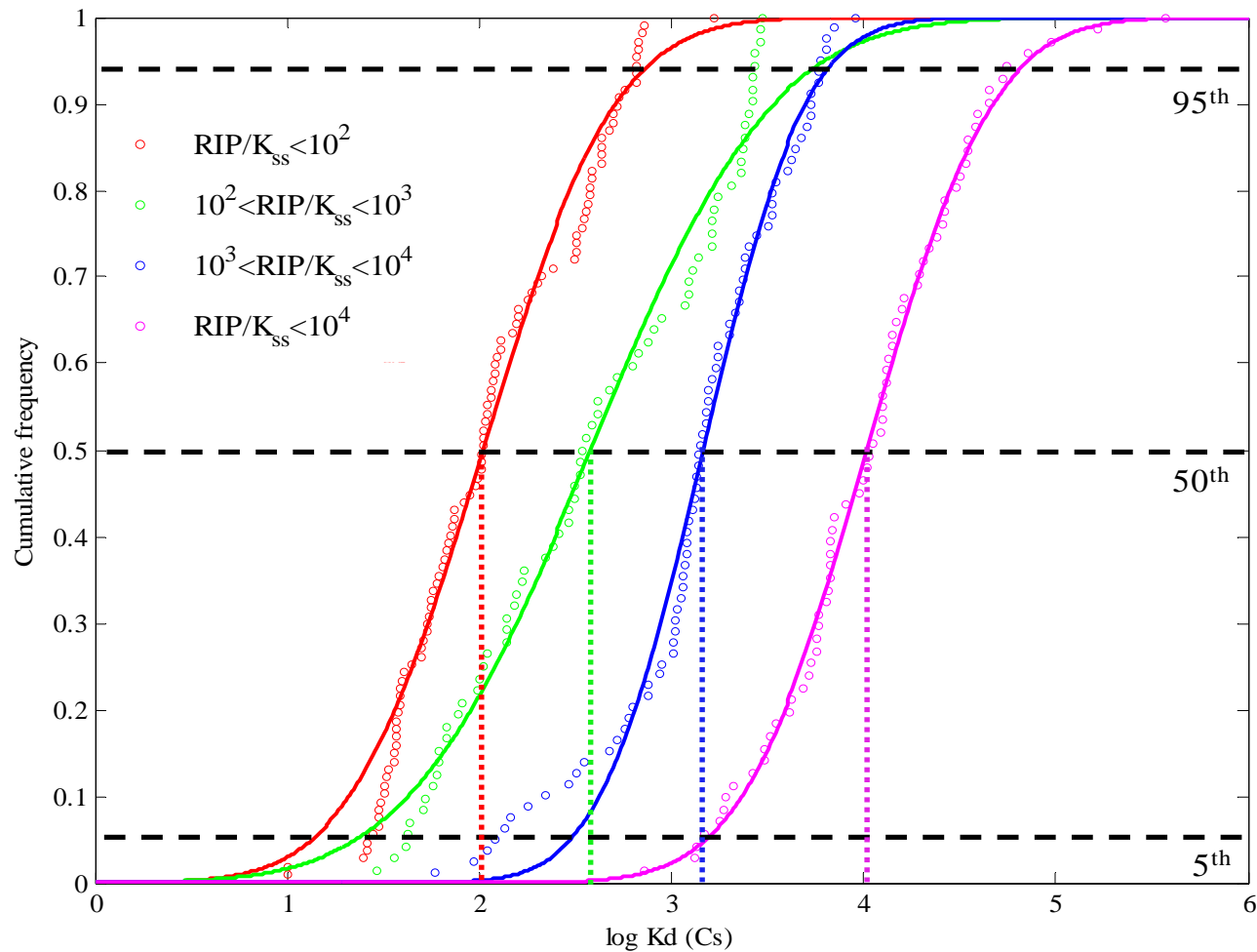


FIG. 2. Box-and-whisker plots of  $K_d(\text{Cs})$  for soils grouped according to the texture/OM and cofactor criteria. The box encloses the middle 50% of the distribution of values, and the median is represented as a horizontal line inside the box. Vertical lines extend to the limits of the 1.5 interquartile ranges. Other symbols represent GM (+) and points at >1.5 interquartile ranges ( $\square$ ).

# MODARIA: CDFs of $K_d$ (Cs) for soils grouped according to $RIP/K_{ss}$ criterion





# SUMMARY OF $K_d$ (Cs) DATA FOR DIFFERENT CONTAMINATION SCENARIOS AND SOIL TYPES

Which information is available?	$K_d$ (Cs) group	GM (L kg <sup>-1</sup> )	GSD	5 <sup>th</sup> (L kg <sup>-1</sup> )	95 <sup>th</sup> (L kg <sup>-1</sup> )	
None	Overall	2600	8.6	$7.6 \times 10^1$	$9.2 \times 10^4$	
Elapsed time since contamination	Short-term	1500	5.6	$8.8 \times 10^1$	$2.6 \times 10^4$	
	Long-term	25000	2.9	$4.2 \times 10^3$	$1.4 \times 10^5$	
Elapsed time since contamination; OM%	Short-term	Organic (OM $\geq$ 50%)	71	2.8	$1.3 \times 10^1$	$3.8 \times 10^2$
		Mineral (OM < 50%)	2600	3.8	$2.9 \times 10^2$	$2.3 \times 10^4$
	Long-term	Organic (OM $\geq$ 90%)*	250	2.7	$4.5 \times 10^1$	$1.2 \times 10^3$
		Mineral (OM < 90%)	27000	2.7	$5.2 \times 10^3$	$1.4 \times 10^5$
Elapsed time since contamination; OM%; soil texture	Short-term	Clay+Loam <sup>a</sup>	3700	3.1	$5.7 \times 10^2$	$2.4 \times 10^4$
		Sand <sup>b</sup>	1200	5.1	$8.6 \times 10^1$	$1.7 \times 10^4$
	Long-term	Clay+Loam <sup>c</sup>	32000	2.4	$7.6 \times 10^3$	$1.4 \times 10^5$
		Sand <sup>d</sup>	18000	2.5	$4.0 \times 10^3$	$8.2 \times 10^4$
Radiocaesium Interception Potential (RIP)**; potassium in soil solution ( $K_{ss}$ )	Short-term	$RIP/K_{ss} < 10^2$	100	3.5	$1.3 \times 10^1$	$8.1 \times 10^2$
		$10^2 \leq RIP/K_{ss} < 10^3$	380	5.5	$2.3 \times 10^1$	$6.3 \times 10^3$
		$10^3 \leq RIP/K_{ss} < 10^4$	1500	2.6	$3.0 \times 10^2$	$7.1 \times 10^3$
		$RIP/K_{ss} \geq 10^4$	10000	2.6	$1.5 \times 10^3$	$7.2 \times 10^4$



## Consider pre-2011

Limited data for:

- andosols
- Rice, fruit, non-European foods
- Contaminated mountainous catchments
- Areas with frequent typhoons

Six years on.....

Enormous effort to:

- Enhance the number of scientists working in the field
- Collect data on radiocaesium behaviour in Japanese terrestrial and aquatic systems
- Provide data on gov web sites and social media
- Support affected people with information centres / interactive fora
- Estimate historical, current and future doses

## Data improvements:

- Radiocaesium uptake from site specific soils
- Uptake by rice and behaviour in irrigated paddy fields
- Lateral transport from catchments
- Impact of typhoons
- Huge extension in data for different types of foodstuffs
- Identification of bark to fruit pathway
- Significant advances in mechanisms governing RCs behaviour
- Time dependent data from early after deposition
- Development of site specific remediation





**WG 4 :**

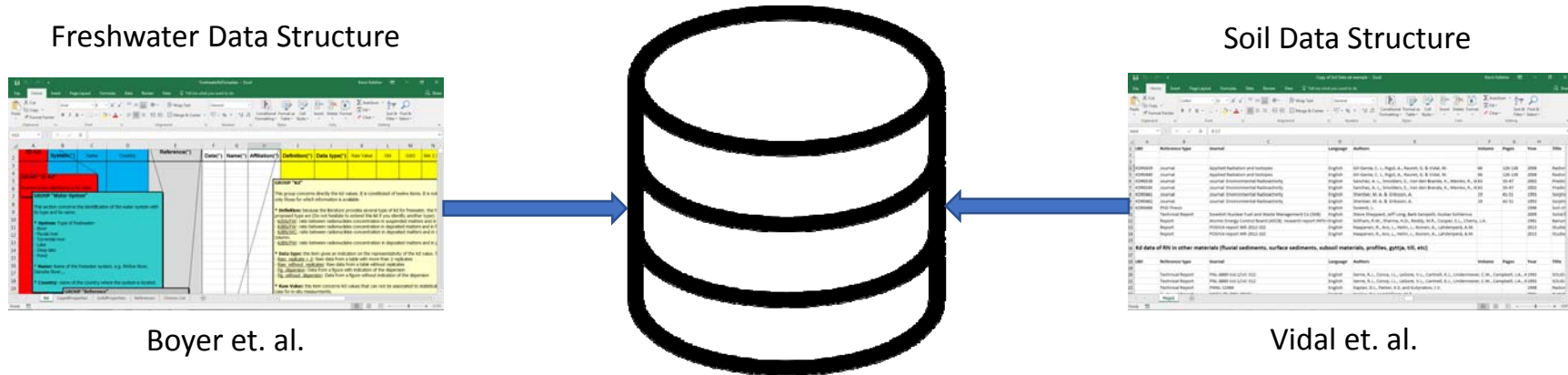
**Working Group 4 –  
Transfer processes and  
data for radiological  
impact assessment**

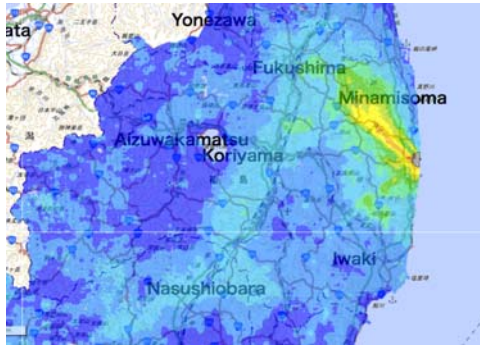
**MODARIA II activities**



# $K_d$ sub group: Development of Global $K_d$ database prototype

MS Access database, based primarily on commonalities between current structure for Freshwater and Soil  $K_d$  datasets.

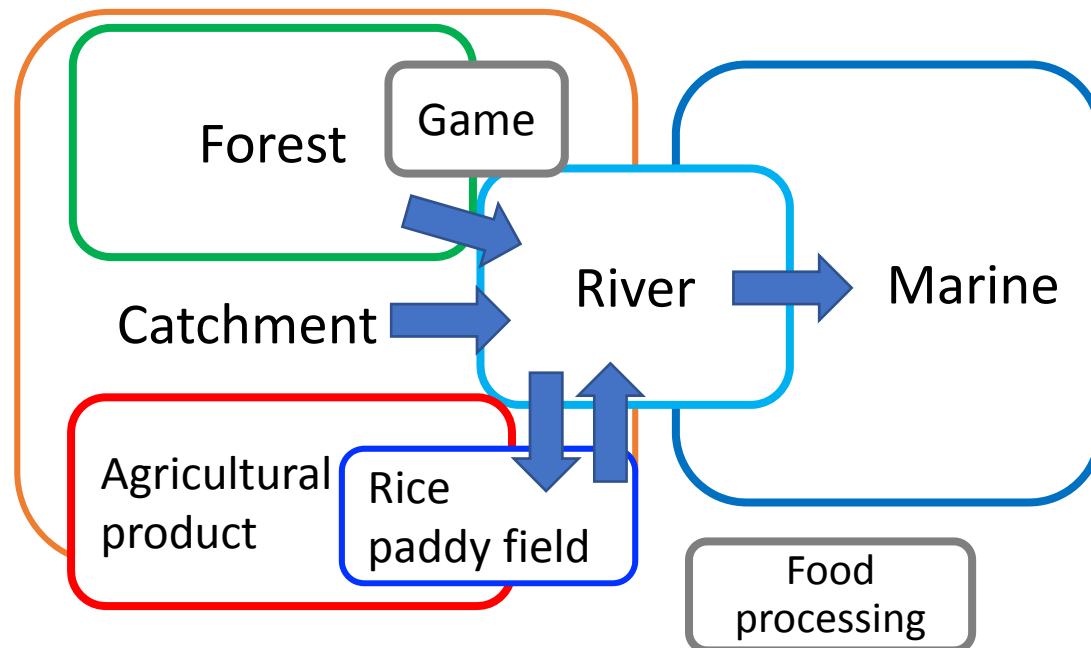




## WG4 sub-group

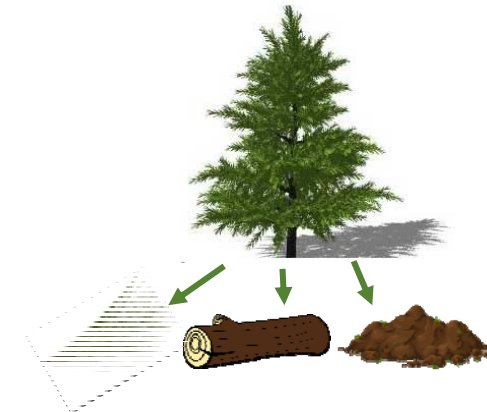
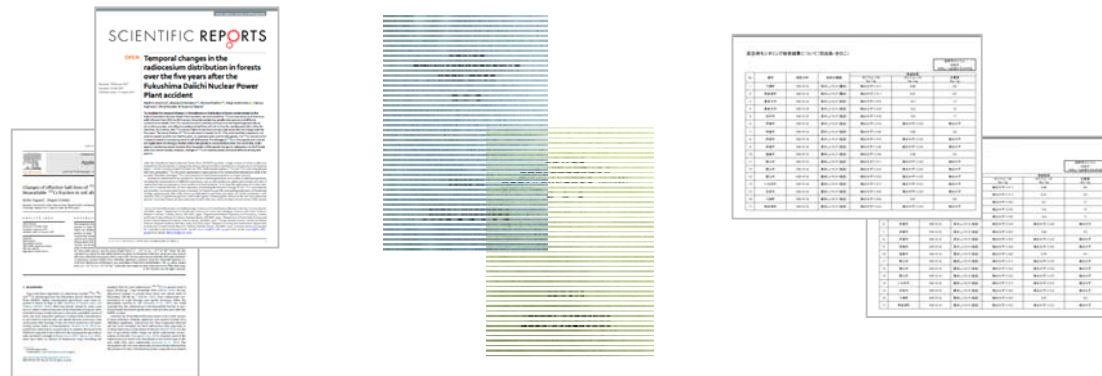
Collation of environmental transfer parameters after the Fukushima accident

**(Fukushima parameters)** Leader: K. Tagami (QST)



# Forest data

—Data compilation ongoing (Hashimoto et al )



Journal papers

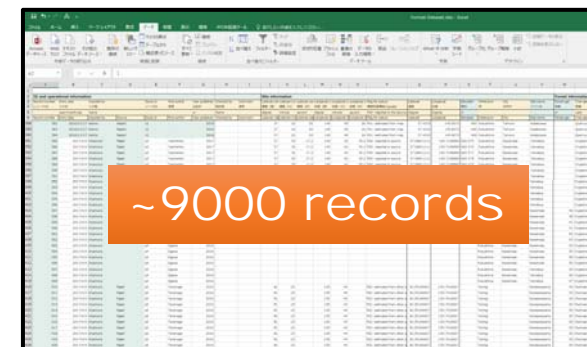
Reports by governments

On the Web

• **Concentration and Inventory** data of

- Trees (each organ, e.g. leaf, wood ...)
- Organic layers (litter layers)
- Mineral soils
- Mushrooms
- Small wild animals (e.g. earthworms, insects etc., NOT large mammals)

## DATABASE



Information on time, location, and forest characteristics etc.



## TRS 472 (2010) vs Fukushima database

	Parameters	RI / stable	
472 Terrestrial & Freshwater (equilibrium)	$a$ , m <sup>2</sup> /kg	RI	Interception coefficient
	$F_{tr}$ , -	RI	Translocation ratio
	$K_s$ , m <sup>-1</sup>	RI	Resuspension factor
	$K_d$ , L/kg	RI	Distribution coefficient in soil
	$F_v$ , -	RI	Concentration ratio from soil to plant
	$F_m$ , $F_f$ , d/kg or d/L	RI+S	Feed transfer coefficient, animal products
	$T_{ag}$ , m <sup>2</sup> /kg	RI	Aggregated transfer factor, semi-natural ecosystem
	$T_{eff}$ , t	RI	Effective half-lives (limited)
	CR	RI	Concentration ratio, water-biota
	$F_r$	RI	Food processing retention factor
Fukushima (accidental)	$K_{d(a)}$ , L/kg	RI	Agricultural soil and River
	$F_v$ , $T_{eff}$	RI	Rice and other Crops
	$T_{ag}$ , m <sup>2</sup> /kg	RI	Wild animals, plants, mushrooms
	$T_{eff}$ , t	RI	River (water, sediment, biota), Forest (tree, soil, litter), Typhoon (heavy rain/storm) event
	CR	RI	Water-Freshwater biota
	$F_r$	RI, Stable	Wild edible plants, etc.
	$a$ , $F_m$ , $F_f$ , $K_s$ ,	RI	Limited data available

Some  
| Challenges  
for the  
future

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Reservoir of  
radiocaesium in  
forested catchments

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High proportion of  
forest in SDA 1, 2 and  
3

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Aim to repopulate up  
to the NPP/repository  
site



2018. 3. 4 JAEA Workshop for studies of environmental dynamics of radiocesium released by NPP accident

Behavior of dissolved radiocesium in river water in a high-dose-rate forested watershed in Fukushima Prefecture

Hideki TSUJI, Shoko Ito, Seiji HAYASHI  
National Institute for Environmental Studies, Japan



# Outline

1. Background
2. Study site, materials and methods
3. Dissolved  $^{137}\text{Cs}$  concentration in a forest river water under baseflow and storm runoff conditions
4. The source of dissolved  $^{137}\text{Cs}$  in river water
5. Conclusion

# Background

A lot of radiocesium has been released from the Fukushima Daiichi Nuclear Power plant, and river water was contaminated

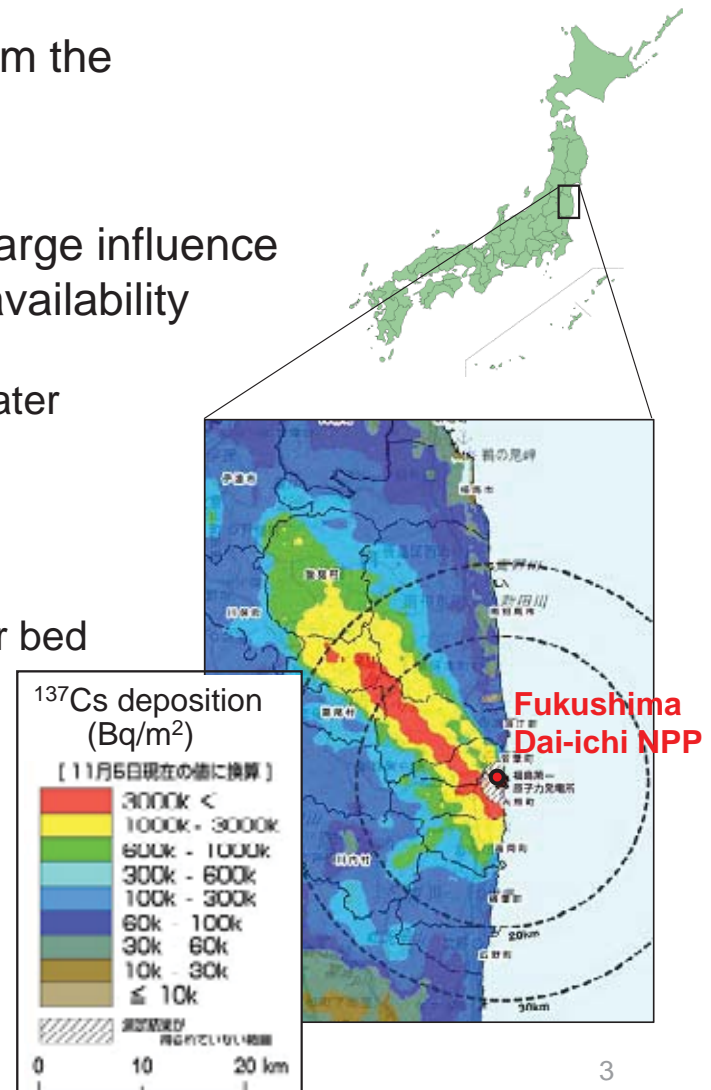
The dissolved form of radiocesium has a large influence on aquatic ecosystem due to the high bioavailability

Dissolved  $^{137}\text{Cs}$  concentration in forest river water increased at runoff event (Shinomiya et al., 2014 ; Iwagami et al., 2017)



Indicated the elution of  $^{137}\text{Cs}$  from litter on river bed (Sakai et al., 2016)

Comprehensive behavior of dissolved radiocesium in forested watershed has not been fully understood

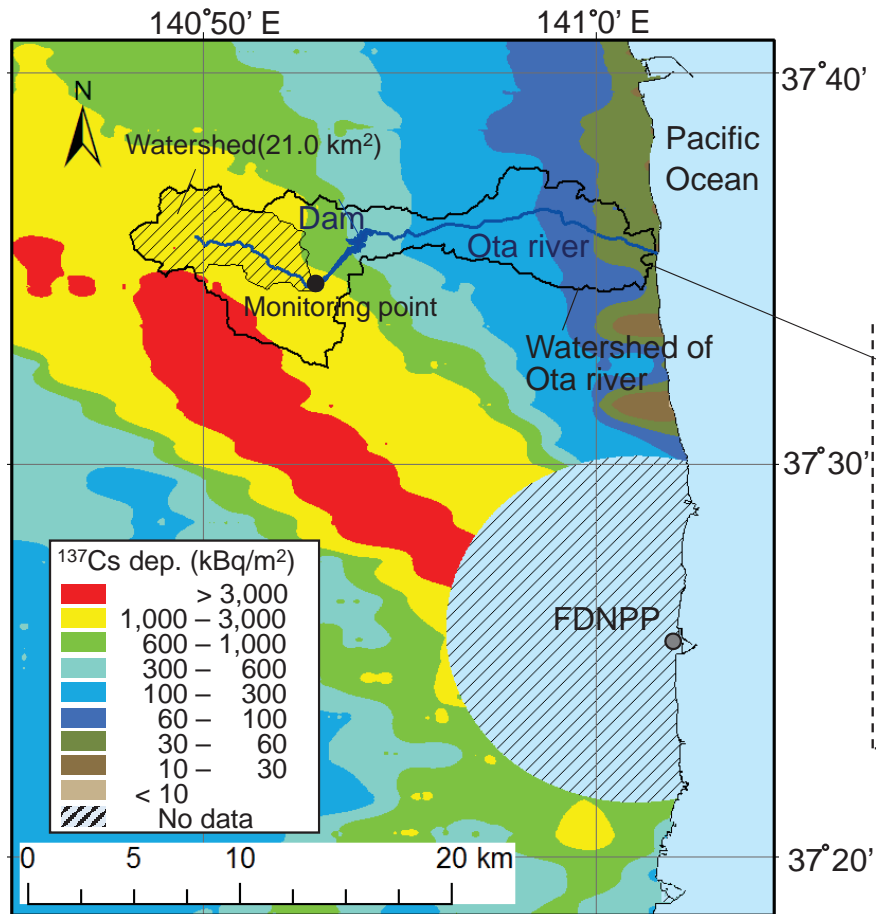


# Purpose of this study

Investigate the behavior of dissolved  $^{137}\text{Cs}$  in a high-dose-rate forested watershed

# Study site

## Ota river watershed

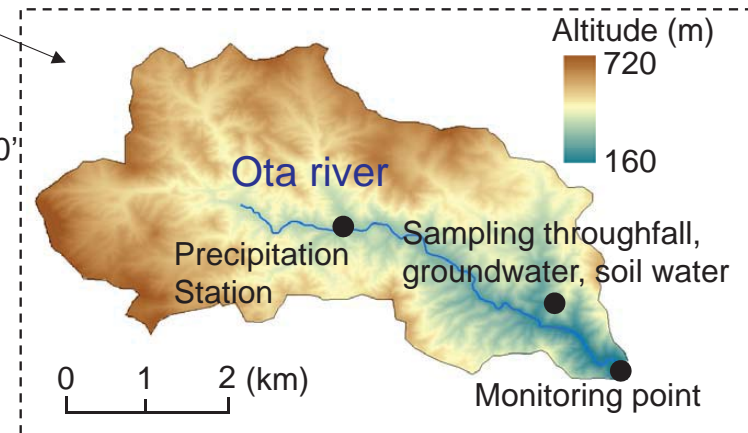


Land Use :

99.2% of forest area

Averaged <sup>137</sup>Cs deposition :

1.7 MBq/m<sup>2</sup>

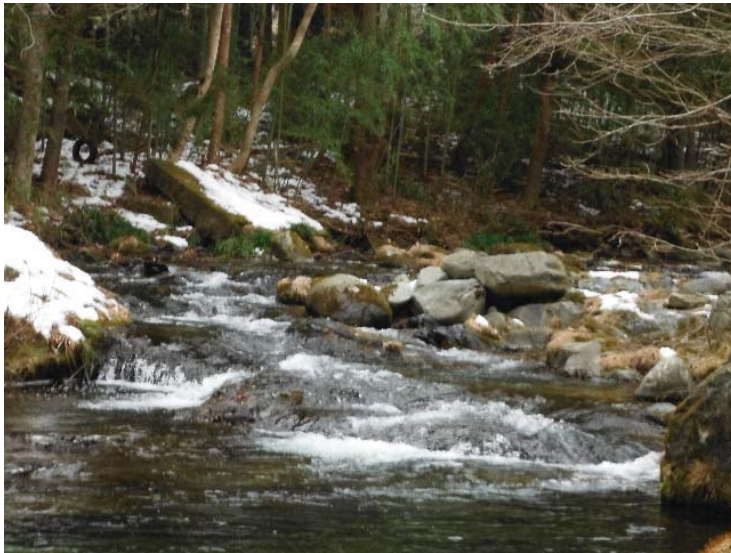




# Sampling of water

## Base flow condition

1. river water  
once-twice a month  
(Jan. 2014 – Nov. 2016)
2. soil water at 20-50 cm depth
3. groundwater



## Storm runoff condition

1. river water  
sampled at intervals of 30-60 min  
including the peak runoff
2. surface water flowing in the dry valley
3. rain water and throughfall water



# Measurement

## 1. dissolved $^{137}\text{Cs}$ [Bq/L]

- river water  
concentrated dissolved  $^{137}\text{Cs}$  in 20-40 L water  
by the cartridge filters method
- rain water, throughfall water, groundwater, soil water:  
concentrated dissolved  $^{137}\text{Cs}$  by evaporation

Radioactivity in sample was detected by coaxial high-purity germanium detectors (GC2518, Canberra)

\* $^{137}\text{Cs}$  in suspended solid in river water was also measured

## 2. coexisting ion concentration ( $\text{K}^+$ , $\text{NH}_4^+$ ...)

by ion chromatography (ICS-1600)

## 3. dissolved organic carbon concentration (DOC)

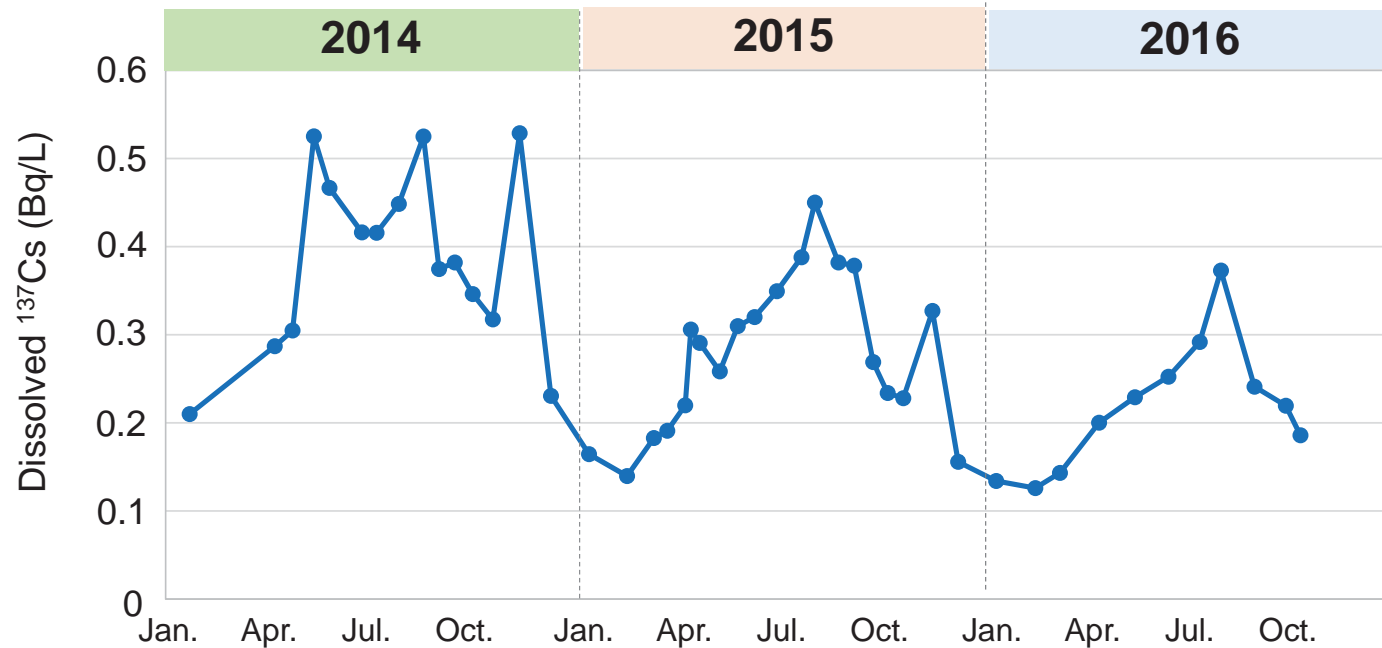
by TOC analyzer (TOC-L)



Cartridge filters device  
(Tsuji et al., 2014)  
(Yasutaka et al., 2015)

# Results

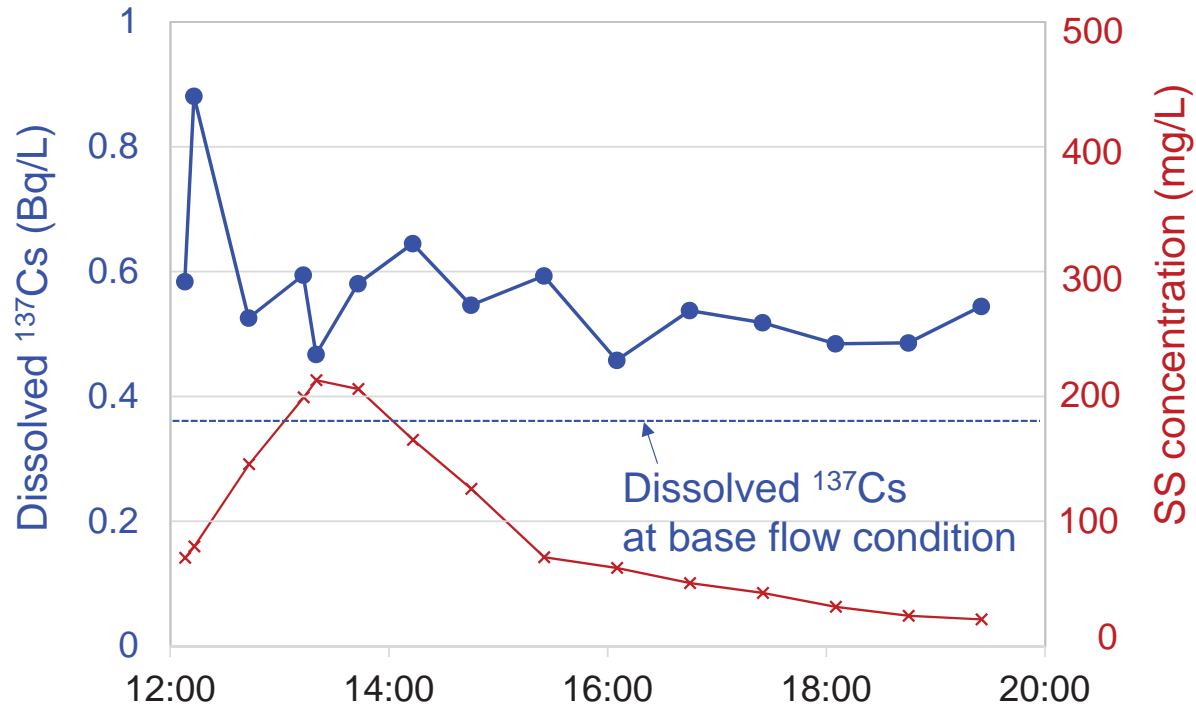
# Dissolved $^{137}\text{Cs}$ concentration under base flow condition



- Dissolved  $^{137}\text{Cs}$  concentration was 0.1-0.5 Bq/L
- Seasonal fluctuation was observed (higher in summer season)

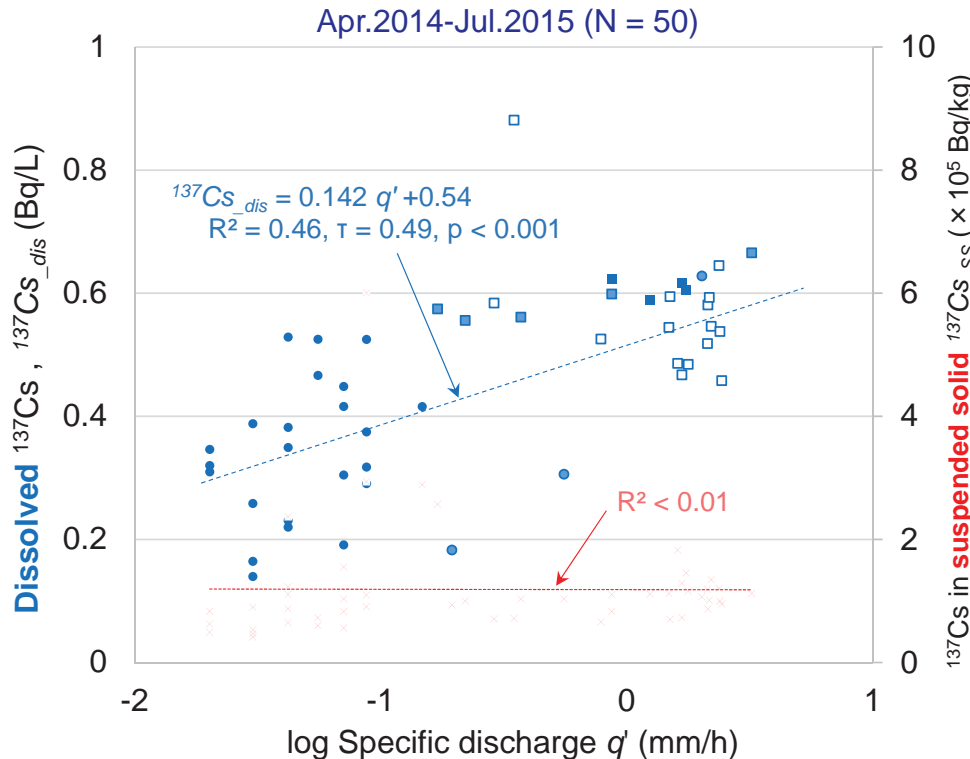
# Dissolved $^{137}\text{Cs}$ concentration under storm runoff condition

Runoff event on Jul.16, 2015



Dissolved  $^{137}\text{Cs}$  concentrations were higher than those at base flow condition

# Comparison of the magnitude of runoff and dissolved $^{137}\text{Cs}$ concentration



Dissolved  $^{137}\text{Cs}$  (● base flow, ■ Oct. 6, 2014, ▣ Oct. 14, 2014, □ Jul. 16, 2015, ● Others)  
 $^{137}\text{Cs}$  in suspended solid ×

The higher the flow rate of the river, the higher the dissolved  $^{137}\text{Cs}$  conc. However,  $^{137}\text{Cs}$  conc. in SS did not increase.

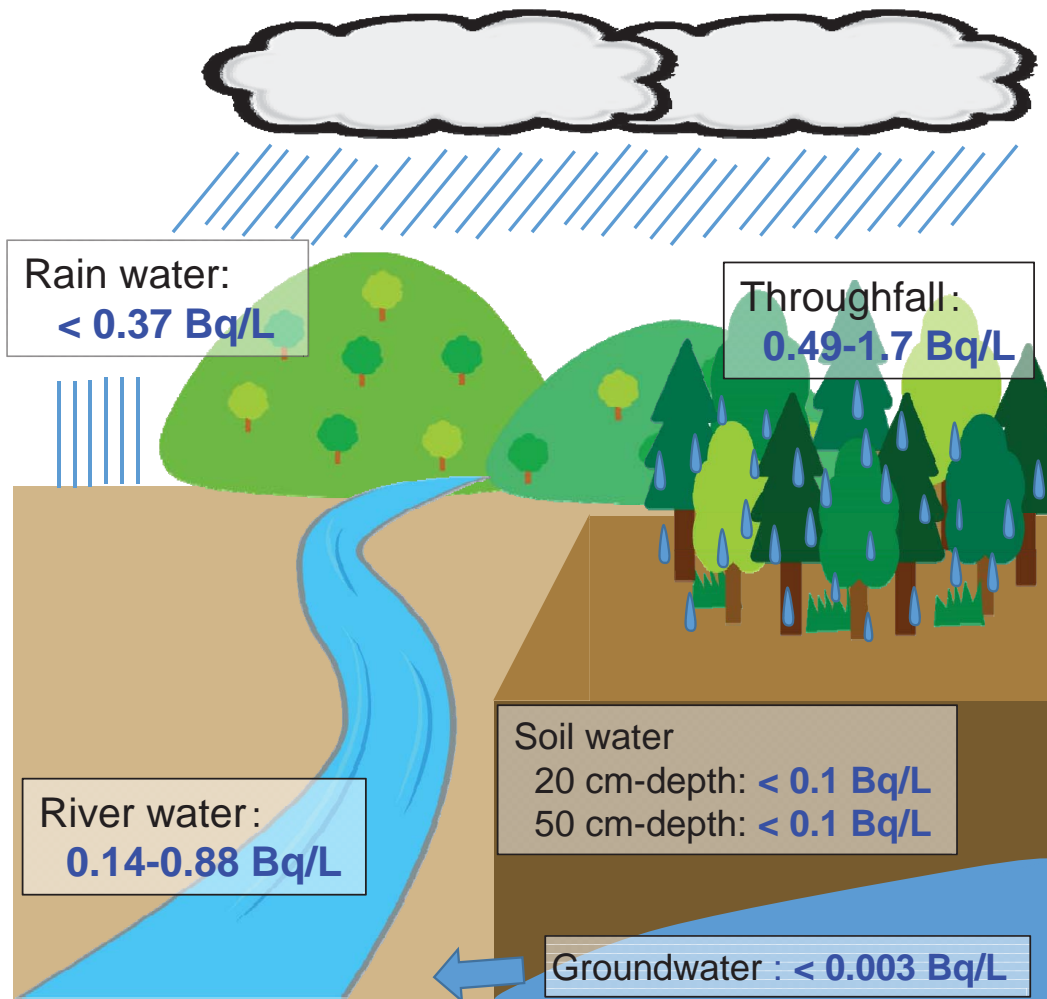


Cannot explain the increase of dissolved  $^{137}\text{Cs}$  conc. by solid-liquid distribution equilibrium with suspended matter in water



Where does the dissolved  $^{137}\text{Cs}$  generate in the forest?

# Dissolved $^{137}\text{Cs}$ concentration in the influent water of the river water

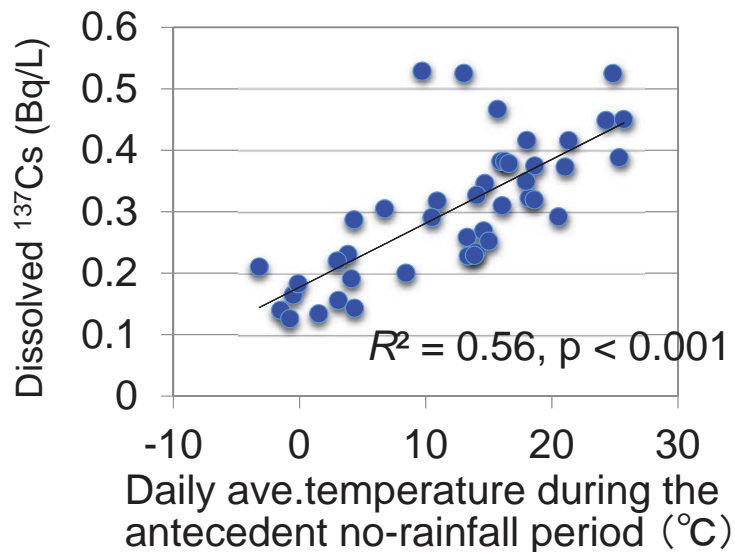
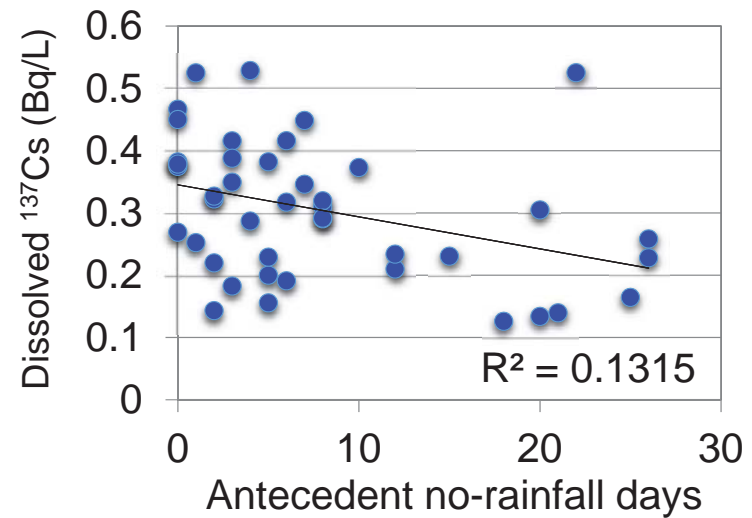
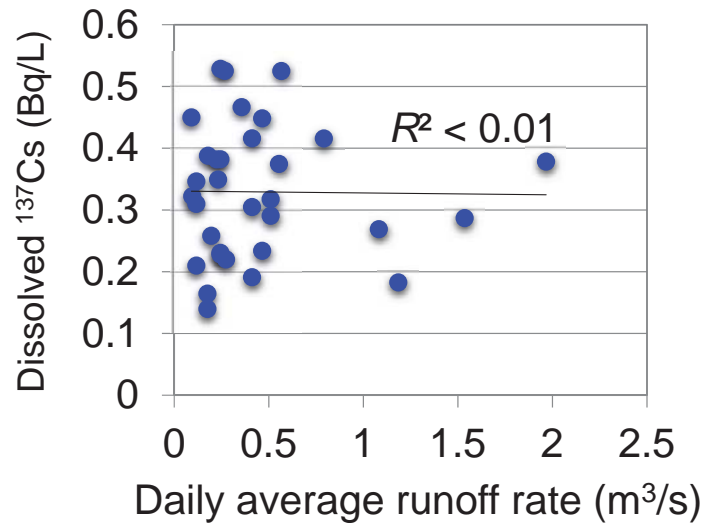


Dissolved  $^{137}\text{Cs}$  in throughfall water was higher than river water, but it will little contribute the increase of dis. $^{137}\text{Cs}$  in river water, because it is adsorbed by mineral particles during the infiltration process (Nakanishi et al., 2014)



Dissolved  $^{137}\text{Cs}$  was generated around the surface flow area?

## Relationship of dissolved $^{137}\text{Cs}$ and environmental factor at base flow condition

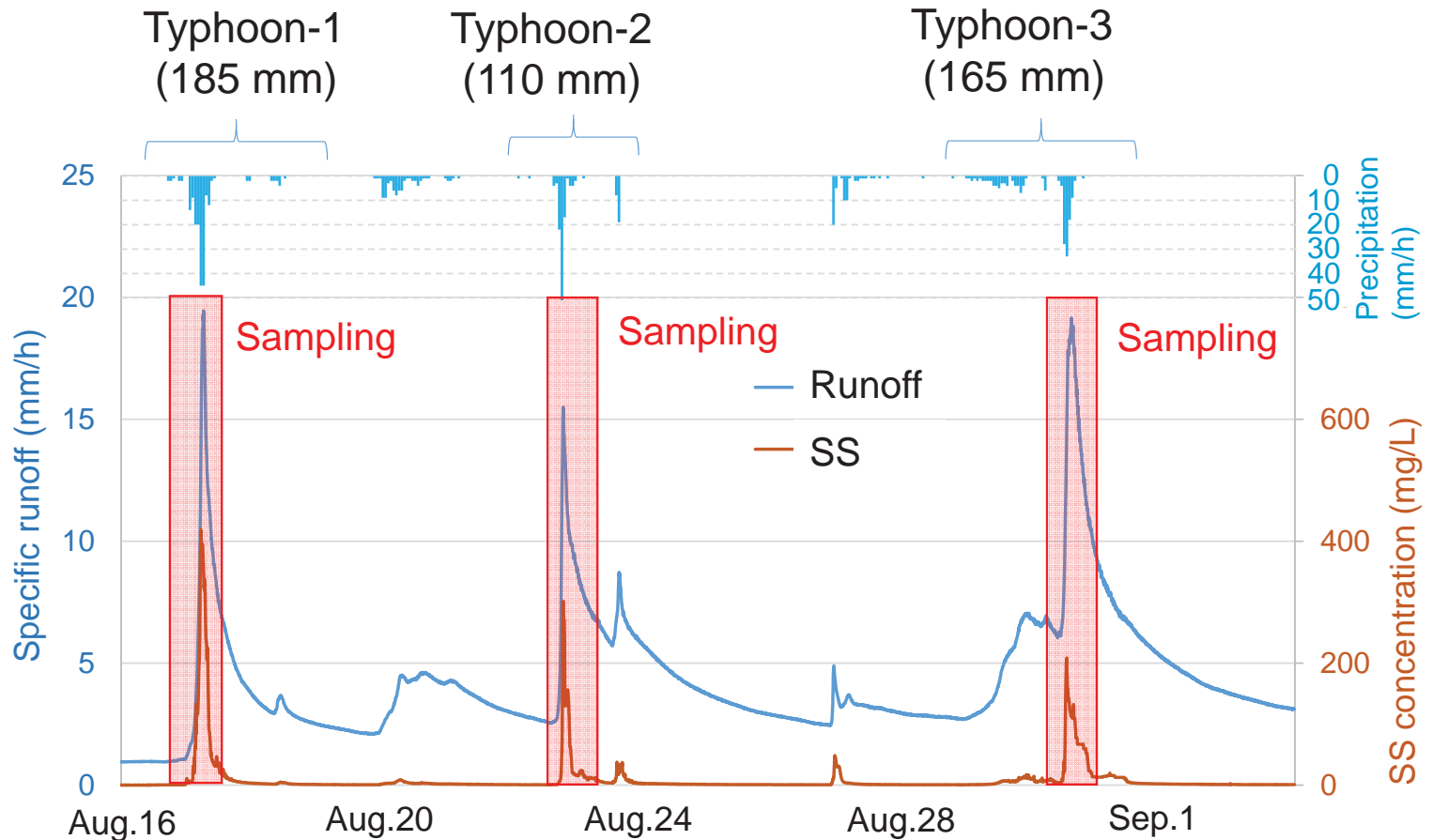


Suggesting that dissolved  $^{137}\text{Cs}$  was supplied by the decomposition of organic matter by microorganisms

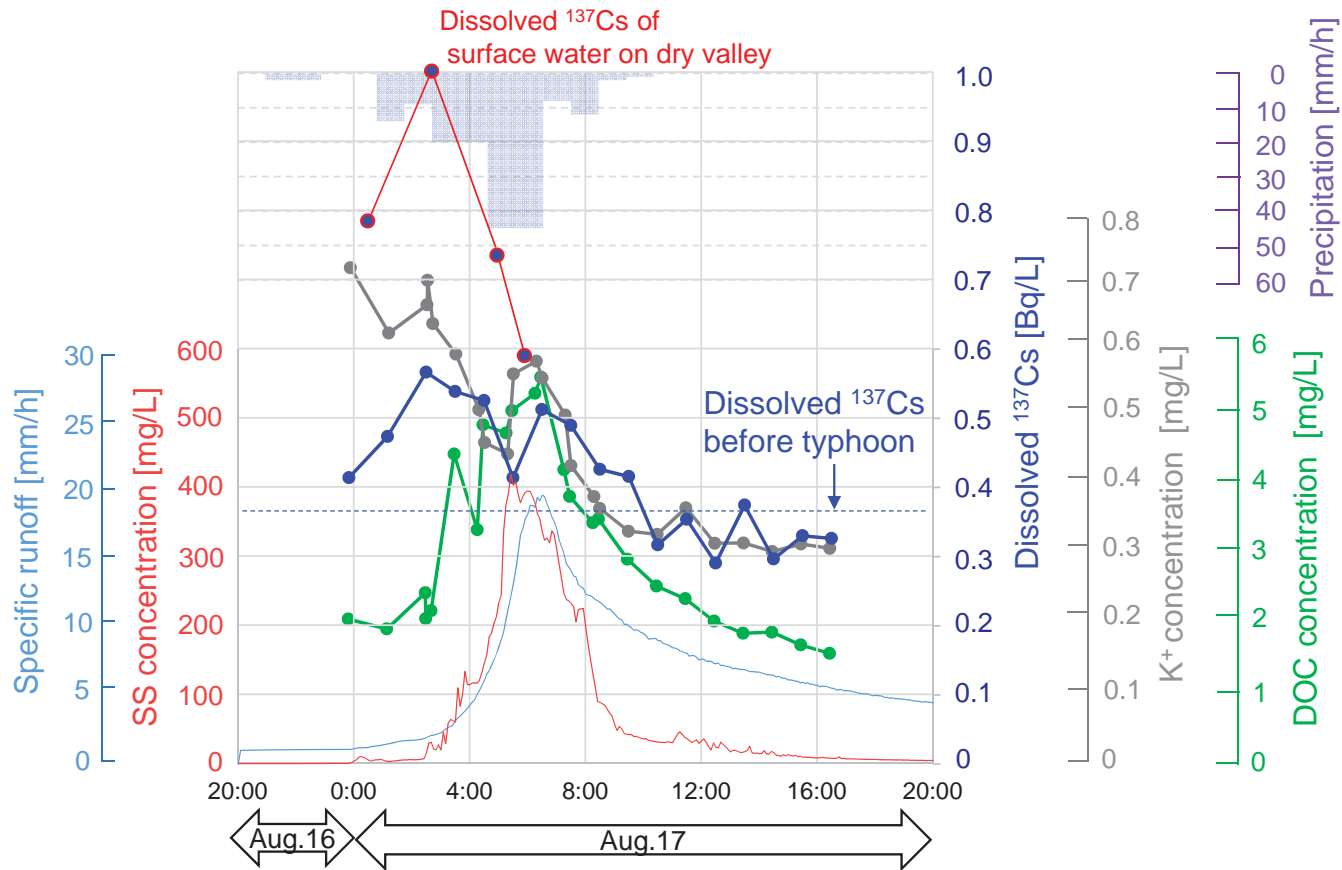


# A series of storm runoff events in Aug.-Sep.2016

Total precipitation : 640 mm



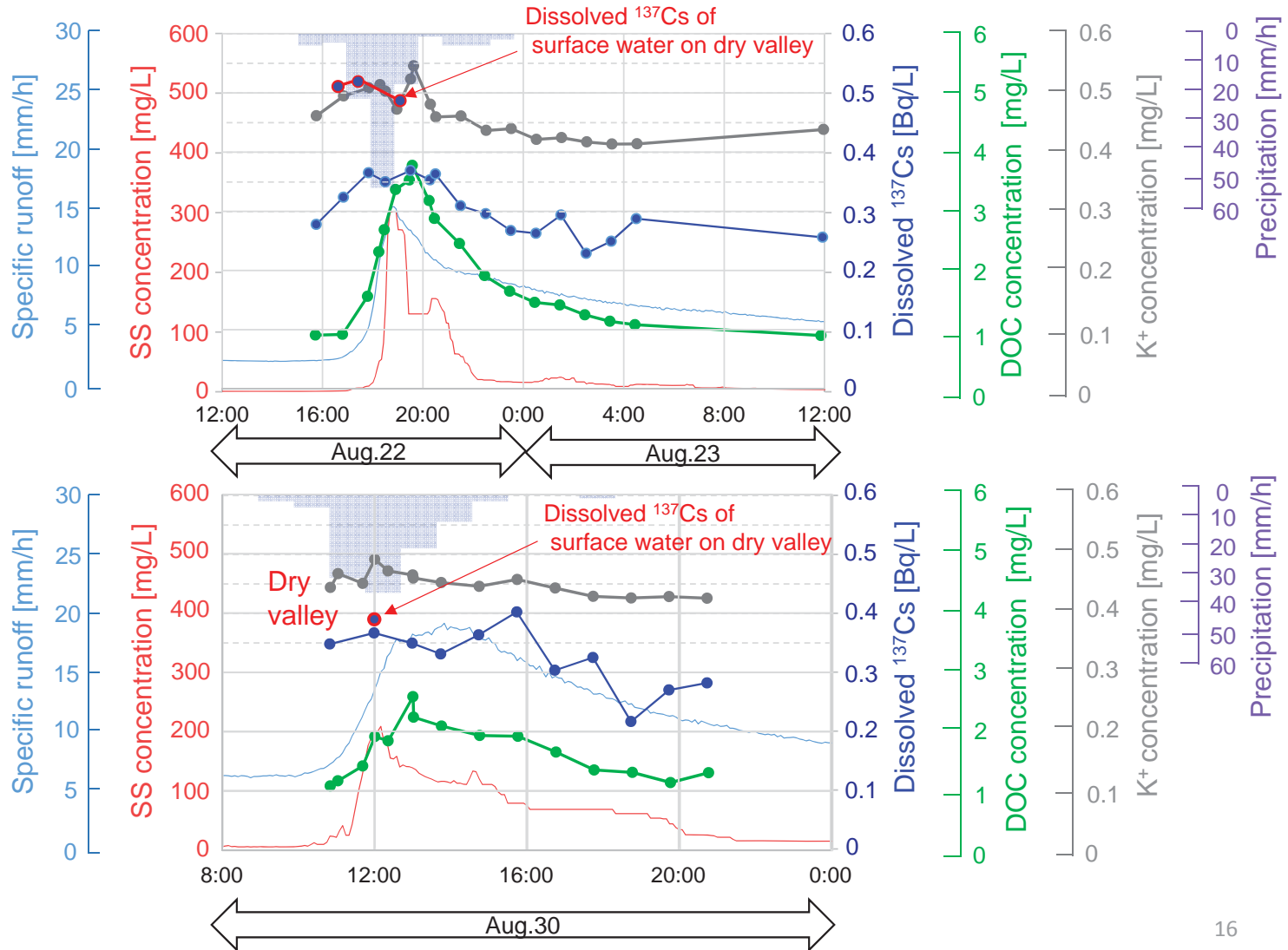
# Dissolved $^{137}\text{Cs}$ and coexisting solute conc. at the first runoff event



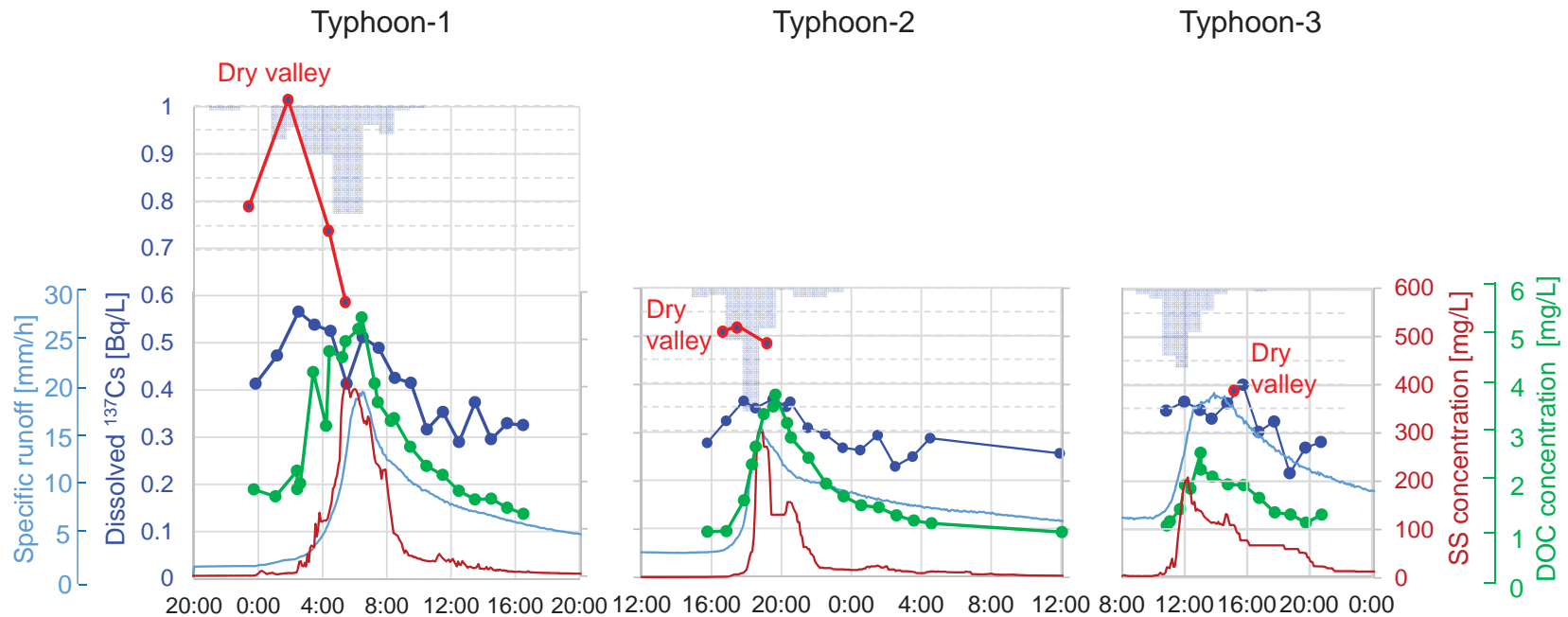
- Dissolved  $^{137}\text{Cs}$  before the runoff peak was higher than those at base flow condition
- Dissolved  $^{137}\text{Cs}$  in dry valley was higher than those of river water

⇒ Dissolved  $^{137}\text{Cs}$  generated in the dry valley was one of the factors to increase dissolved  $^{137}\text{Cs}$  in river water at storm runoff event

# Dissolved $^{137}\text{Cs}$ and coexisting solute at the second and third runoff event



# Time change of dissolved $^{137}\text{Cs}$ conc. during the three runoff events

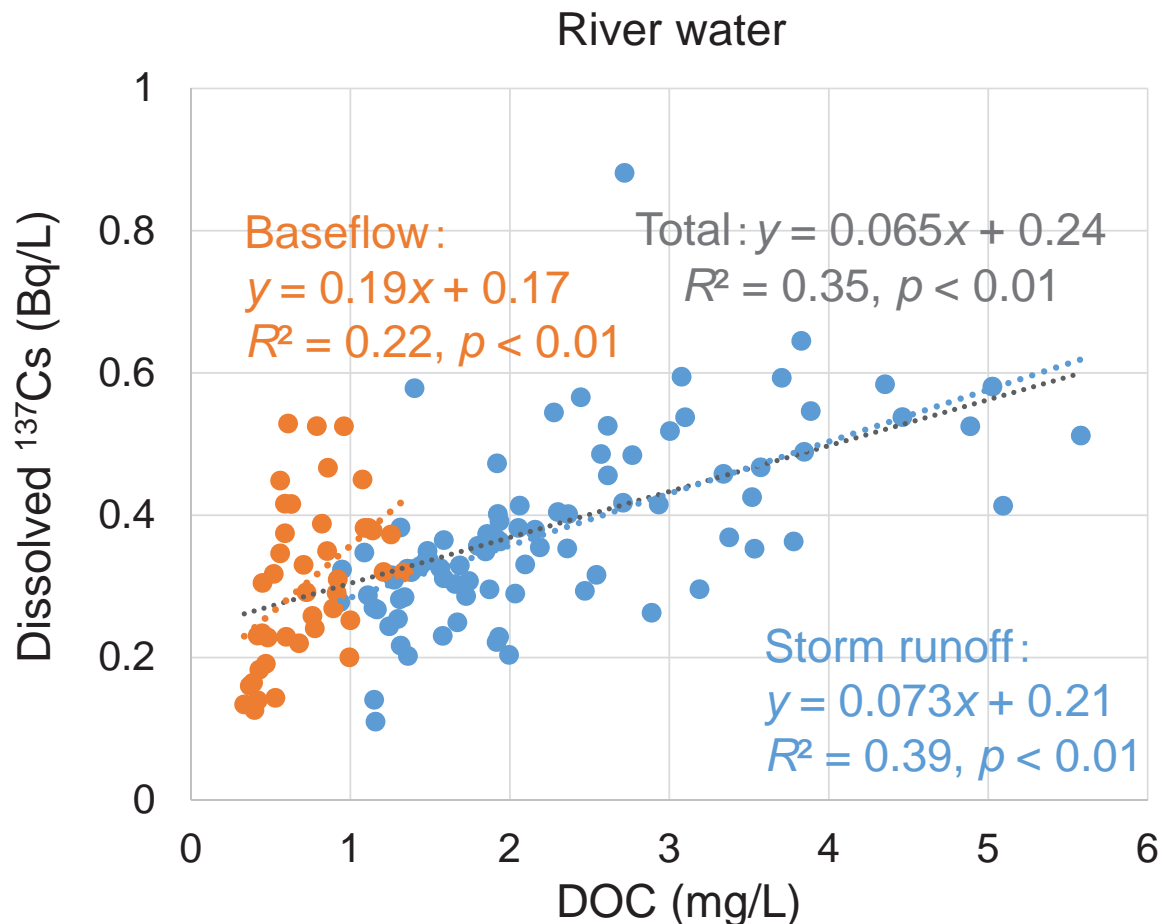


- The peak of dissolved  $^{137}\text{Cs}$  conc. in each runoff event gradually decreased
- The peak of DOC concentration also decreased



Because considerable amount of  $^{137}\text{Cs}$  (within the litter layer) with solubilization potential was washed out at the first typhoon event, the solubilizable  $^{137}\text{Cs}$  stock may have dropped at the later runoff event.

# Relationship of dissolved $^{137}\text{Cs}$ and DOC concentration



Surface soil water

{ DOC :  $> 10$  mg/L  
dis.  $^{137}\text{Cs}$  :  $< 0.1$  Bq/L

groundwater

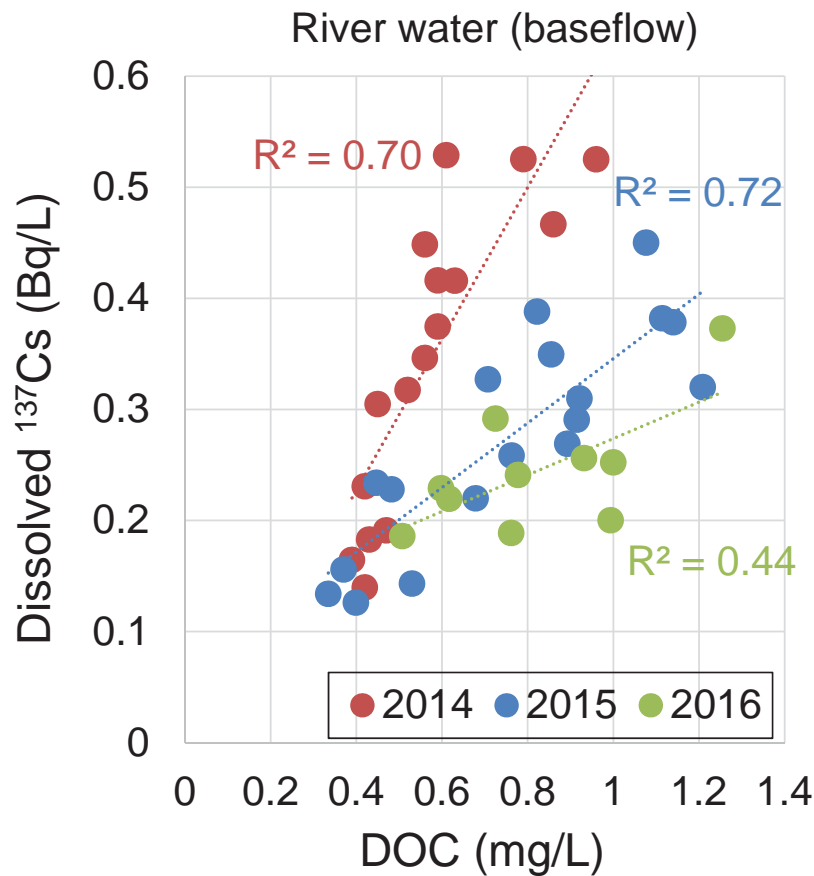
{ DOC :  $< 0.5$  mg/L  
dis.  $^{137}\text{Cs}$  :  $< 0.01$  Bq/L

Significant positive relationship was observed between dissolved  $^{137}\text{Cs}$  and DOC concentration

The coefficient of the regression line between DOC and  $^{137}\text{Cs}_{\text{dis}}$  under storm runoff condition was significantly lower than that under baseflow condition.

→ **Surface soil water** may be the main source of DOC at storm runoff event

# Time change of the dependence of dis.<sup>137</sup>Cs conc. to DOC conc.



Dissolved <sup>137</sup>Cs concentration estimation equation using “DOC conc.” and “elapsed time” as explanatory variables

$$\text{Dissolved } ^{137}\text{Cs [Bq/L]} \sim a \cdot \text{DOC} + b \cdot \frac{\text{DOC} \times \text{time}}{\text{interaction}} + c$$

Time dependence of DOC

= the significance of coefficient “*b*”



$$b = -0.178 \pm 0.024, p < 0.001$$

The dependence of dissolved <sup>137</sup>Cs conc. on DOC conc. significantly decreased.

→ Strongly suggest the contribution of solute from litter decreased with time

# The process of increasing dis.<sup>137</sup>Cs in river water at runoff event

- Initial runoff:  
Elution of <sup>137</sup>Cs from riparian litter along with the rising water level



Also the source of <sup>137</sup>Cs at baseflow

- Around the runoff peak:  
Elution of <sup>137</sup>Cs from litter on the dry valley when occurring the return flow



Expansion of surface flow network in the process of runoff



# Conclusion

$^{137}\text{Cs}$  eluted from litter by microbial decomposition was an influential source of dissolved  $^{137}\text{Cs}$  in river water

brings the following phenomenon :

- dissolved  $^{137}\text{Cs}$  concentration was higher in summer than winter under base flow conditions
- dissolved  $^{137}\text{Cs}$  concentration was higher under storm runoff conditions than base flow conditions

Main source of dissolved  $^{137}\text{Cs}$

Under baseflow condition or the initial phase of storm runoff :  
elution of  $^{137}\text{Cs}$  from riparian litter along with the rising water level

Around the runoff peak of storm runoff :  
elution of  $^{137}\text{Cs}$  from litter on the dry valley when occurring the return flow





The 3<sup>rd</sup> International Cesium Conference

# Transport of radiocesium into aquatic biota and its biological effects

放射性セシウムの水生生物相への移行と生物影響

**Tomisato Miura**

Hirosaki university

Graduate School of Health Sciences

*Ukedo River, Namie, Fukushima*



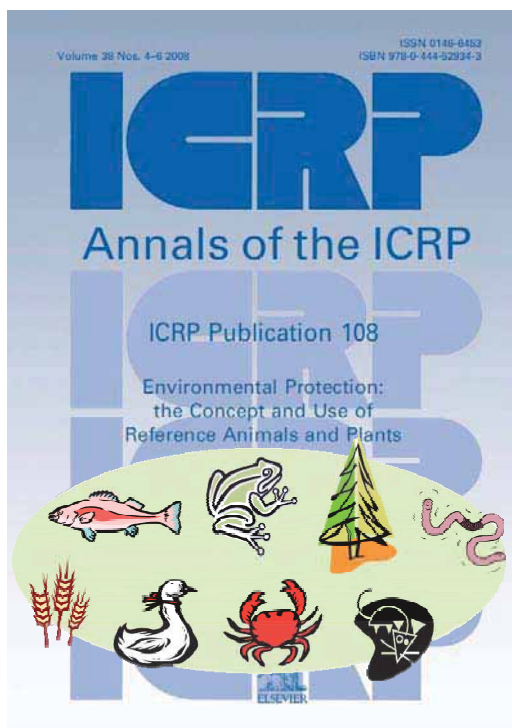
Hirosaki University, Chromosome Research Group

Tomisato Miura

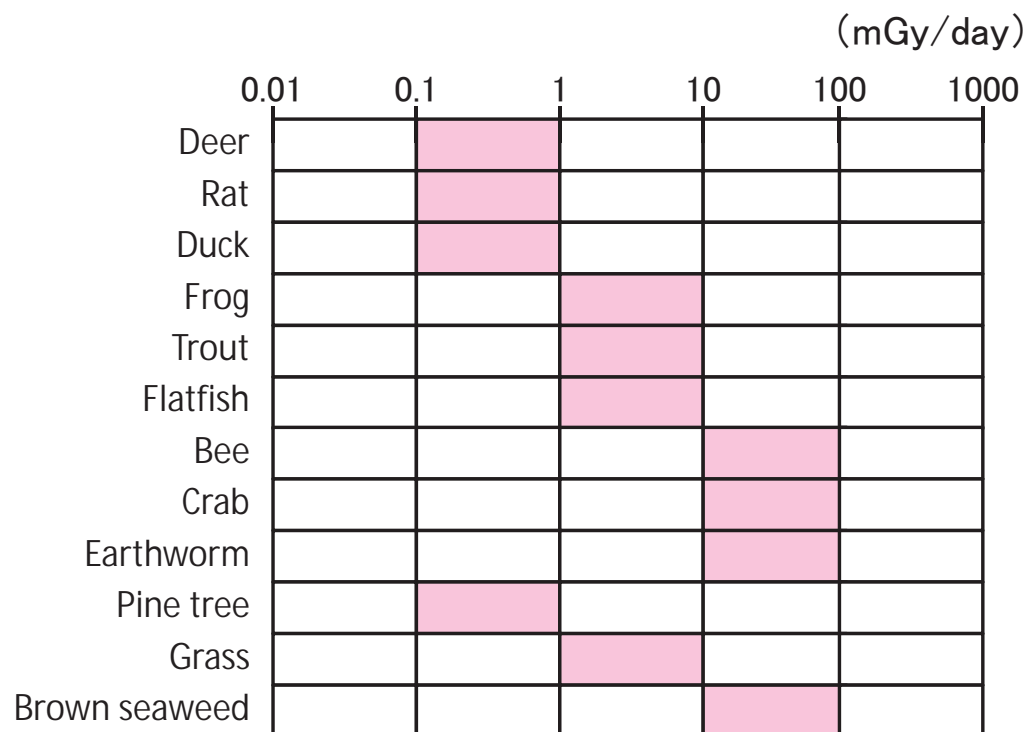


# Derived Consideration Reference Levels: DCRL

Each DCRL constitutes a band of dose rates for each RAP within which there is likely to be some chance of the occurrence of deleterious effects (ICRP Committee 5).



ICRP Publication 108. Ann. ICRP 38 (4-6)



ICRP Publication 108: Ann. ICRP 38, (2008) より改変





# Animal researches in Hirosaki University

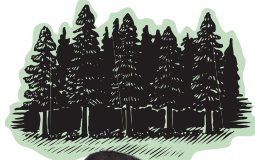
- Dynamics analysis of radioactive materials
- External and internal exposure doses
- Biological effects (chromosome aberrations)
- Environmental evaluation

## Land area



Japanese monkey

### forest



Field mice

### urban area



Cat



Raccoon

## Hydrosphere



Trout



Aquatic biota



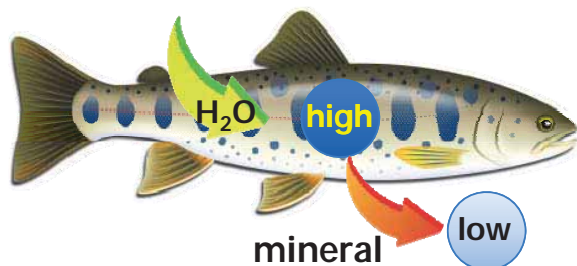


# Difference in salt discharge mechanism between freshwater fish and saltwater fish

## Migration and homeostasis due to concentration gradient

### Freshwater

(Hypotonic, low mineral conc.)



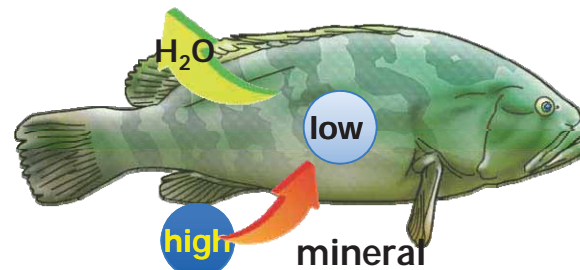
Leakage of K and Cs from the body



Do not discharge K or Cs

### Saltwater

(Hypertonic, high mineral conc.)



Inflow of K and Cs from outside the body



Proactive discharge of K and Cs

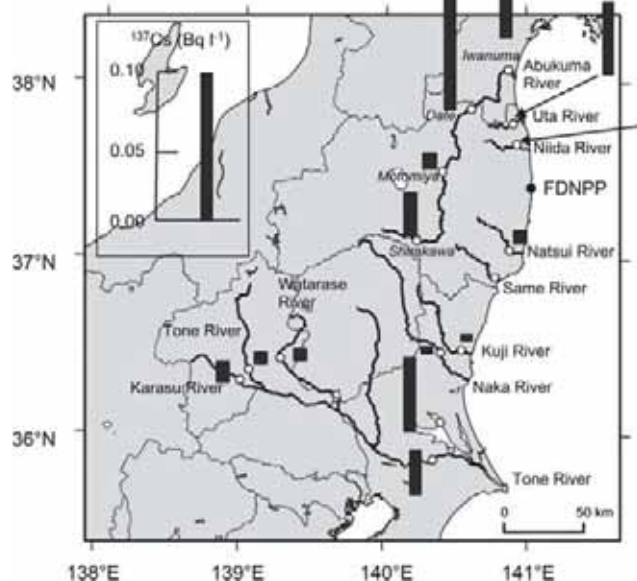
**Biological half-life of radioactive cesium is long in freshwater fish**



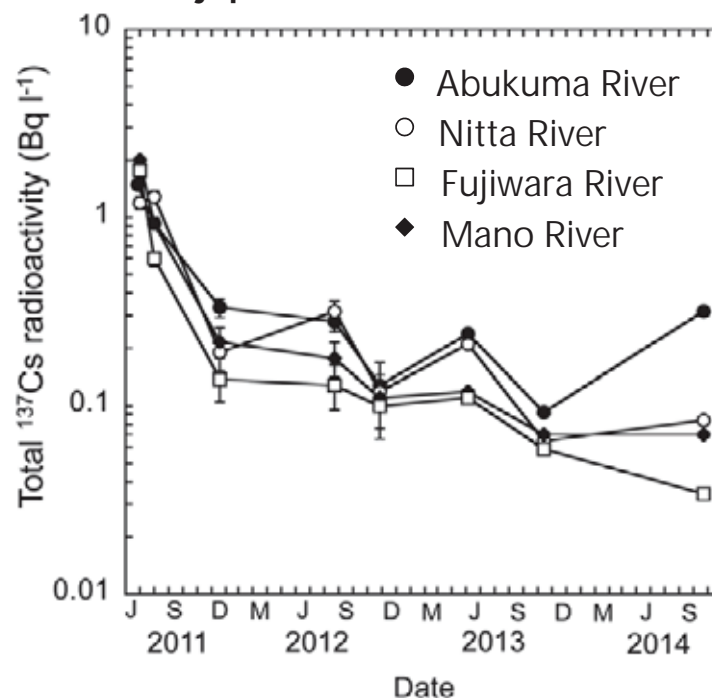


# Radioactive cesium in rivers after FDNPP accident

August 24-26, 2012



Changes in Cs-137 Concentration in rivers, Fukushima, Japan

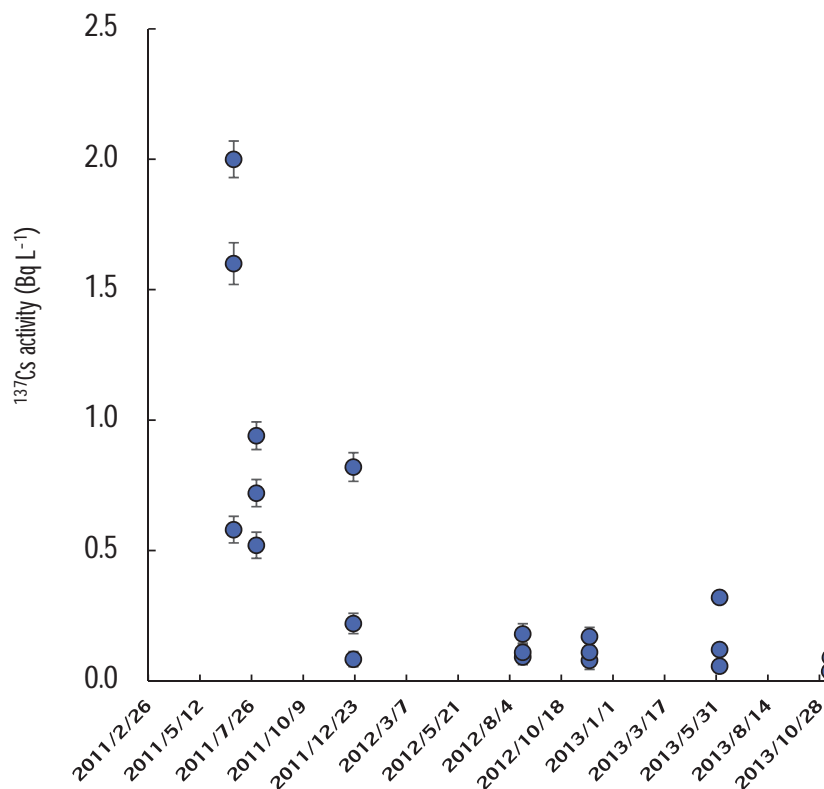


長尾誠也, 地球化学49, 217-226 (2015) より引用



# Radioactive cesium concentration in the rivers

## Mano River



## Ukedo River

Field	Date	Radioactive cesium concentration (Bq/L)	
		Cs-134	Cs-137
Murohara Bridge	2015. 5. 19	<0.85	<0.88
	2015. 6. 11	<0.61	<0.69
	2015. 7. 15	<0.79	<0.63
	2015. 8. 14	<0.86	<0.80
	2015. 9. 30	<0.62	<0.88
	2015. 10. 19	<0.65	<0.84
	2015. 11. 7	<0.64	<0.75
	2015. 12. 16	<0.48	<0.99
	2016. 1. 9	<0.63	<0.80
	2016. 2. 2	<0.85	<0.92
Ukedo Bridge	2015. 5. 19	<0.82	<0.80
	2015. 6. 11	<0.63	<0.80
	2015. 7. 9	<0.61	<0.69
	2015. 8. 8	<0.81	<0.70
	2015. 9. 30	<0.67	<0.69
	2015. 10. 19	<0.61	<0.63
	2015. 11. 10	<0.79	<0.88
	2015. 12. 15	<0.75	<0.96
	2016. 1. 9	<0.86	<0.92
	2016. 2. 6	<0.47	<0.63

Radioactive Material Monitoring Surveys of the Water Environment (Ministry of the Environment)

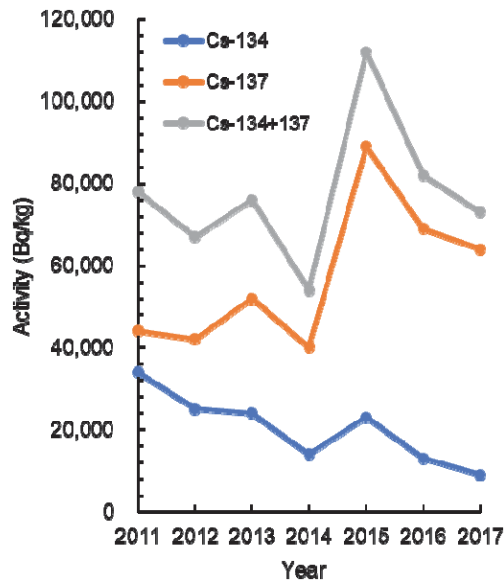




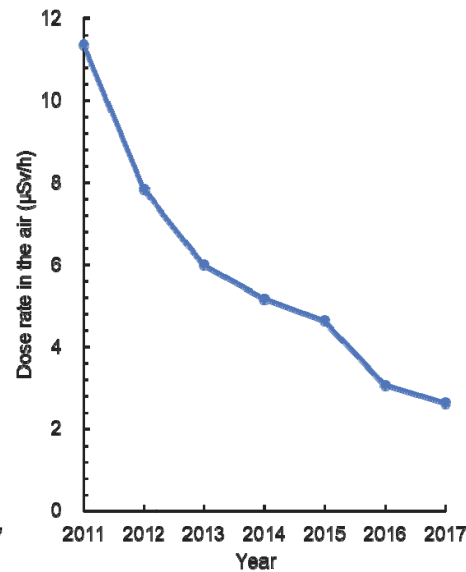
# Radioactive material monitoring surveys in Ukedo River

## @Murohara Bridge in Ukedo River

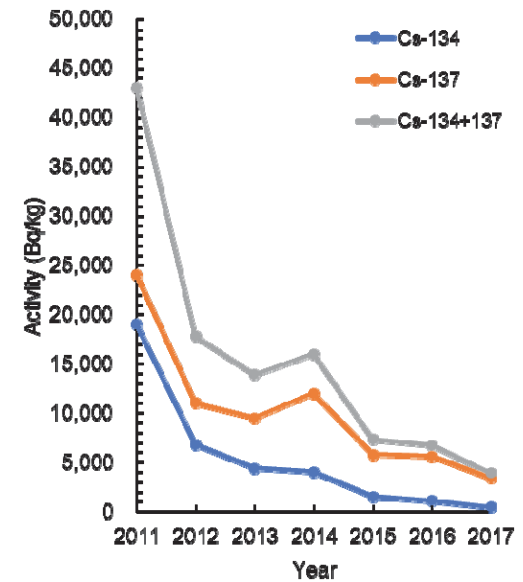
### Soil in the left bank



### Air dose rate



### Bottom sediment



Radioactive Material Monitoring Surveys of the Water Environment (Ministry of the Environment)







# Study of Landlocked masu trout

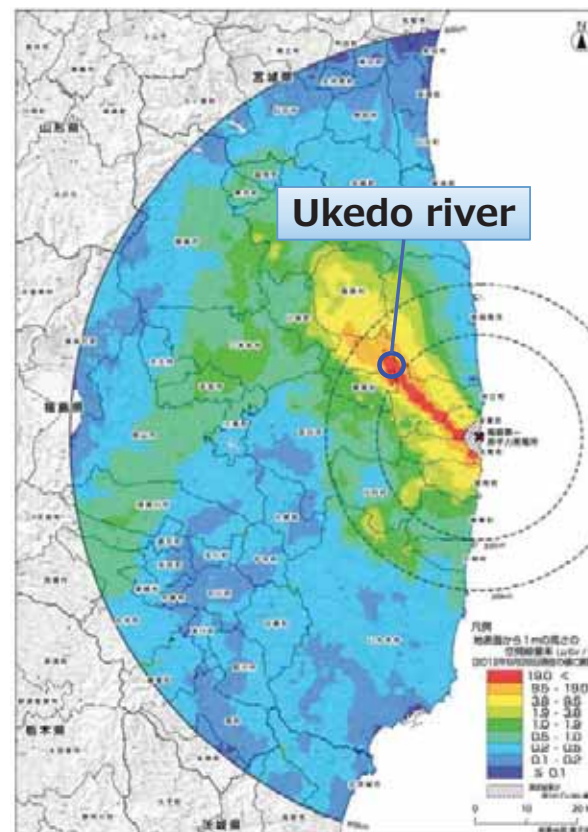
## Landlocked masu trout

*Oncorhynchus masou masou*



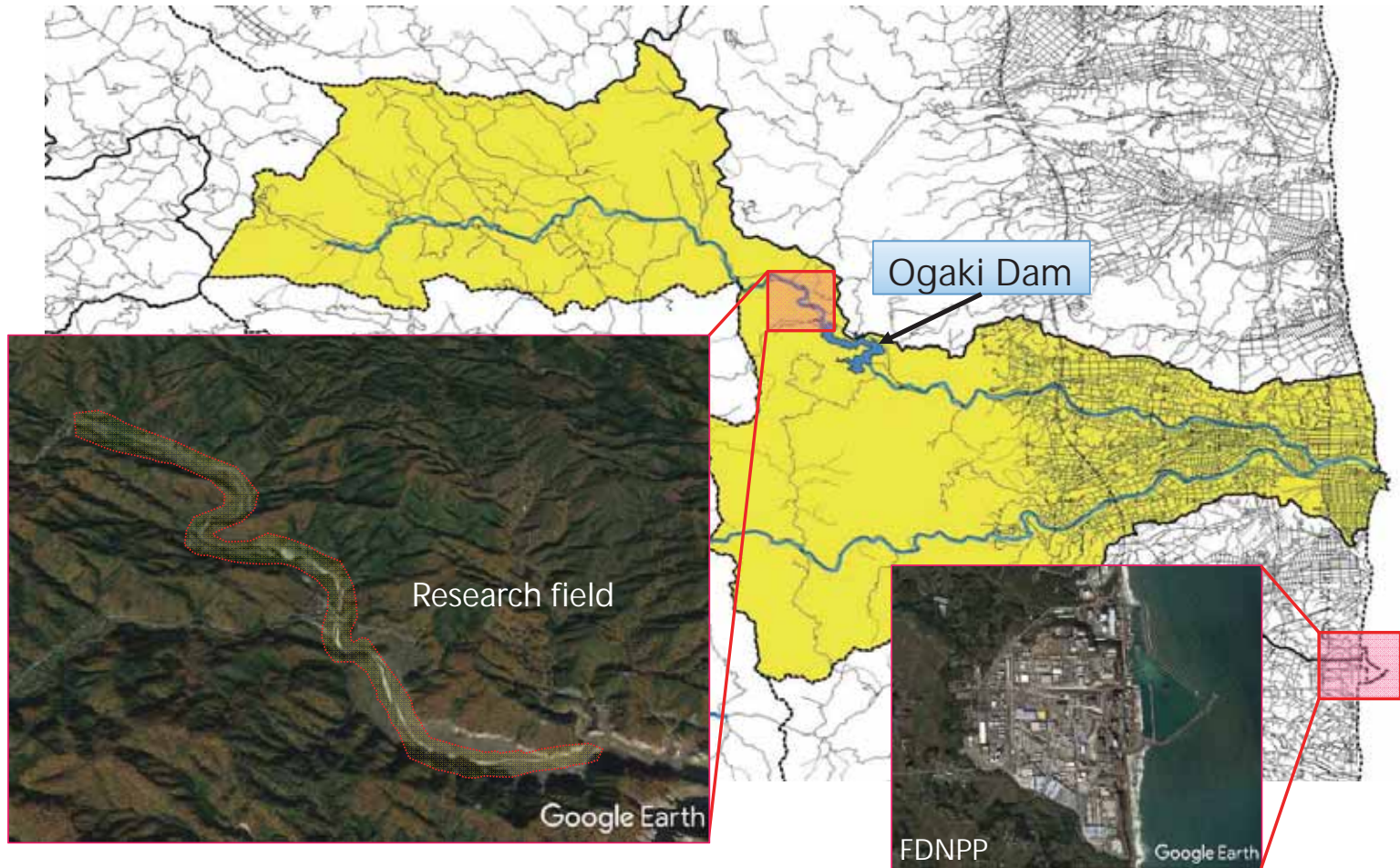
Field	Ukedo River (Namie, Fukushima) Nijikai River (Owani, Aomori)
Capture	Lure fishing
Analysis	Age Body weight and length Chromosome damage (micronucleus) Internal exposure

Aircraft monitoring results  
(MEXT : June 28, 2012)





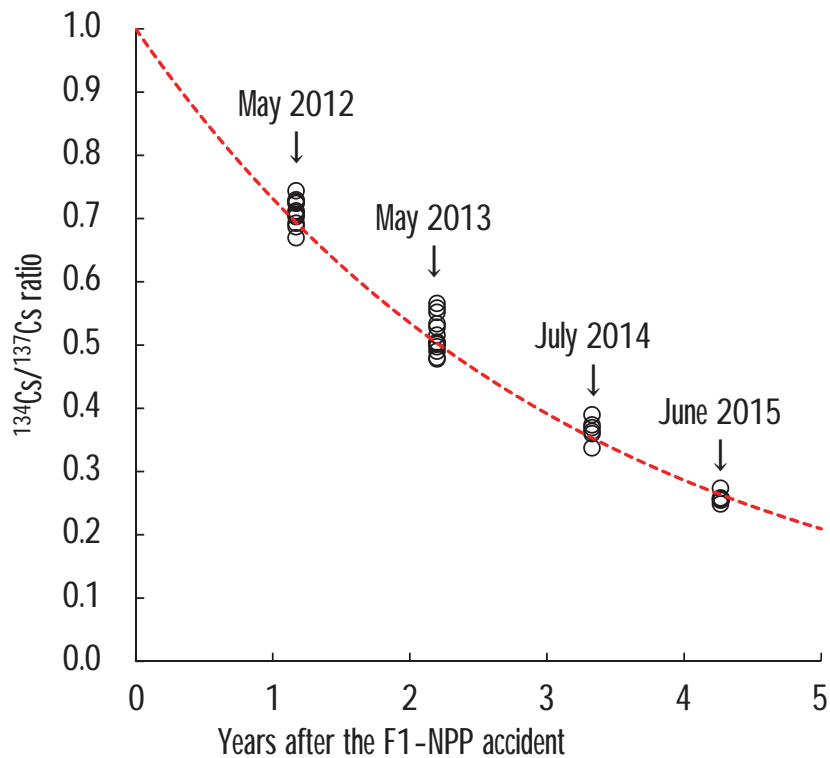
# Ukedo River in Namie, Fukushima







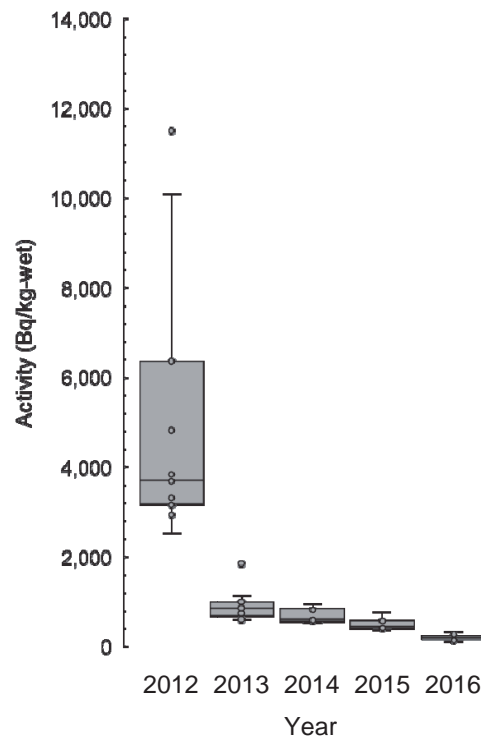
# $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in muscle of trout



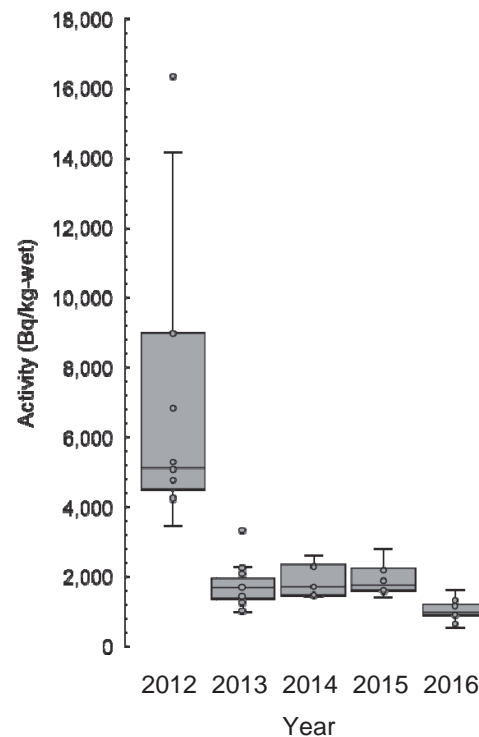


# Kinetics of radioactive cesium concentration in muscle of trout captured in Ukedo River

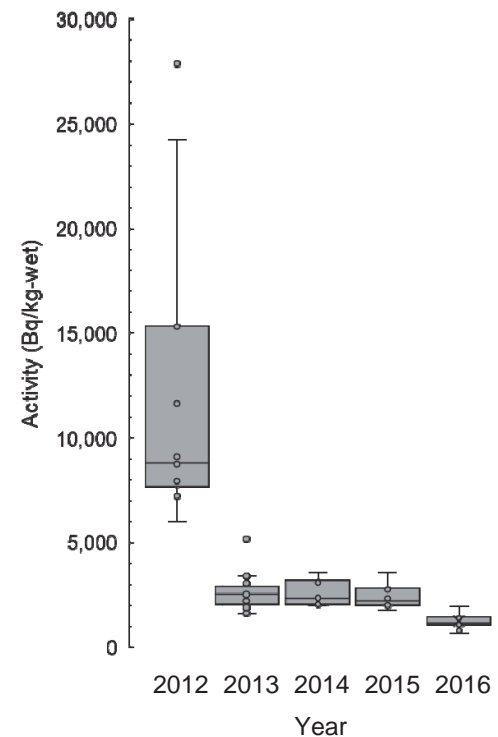
$^{134}\text{Cs}$



$^{137}\text{Cs}$



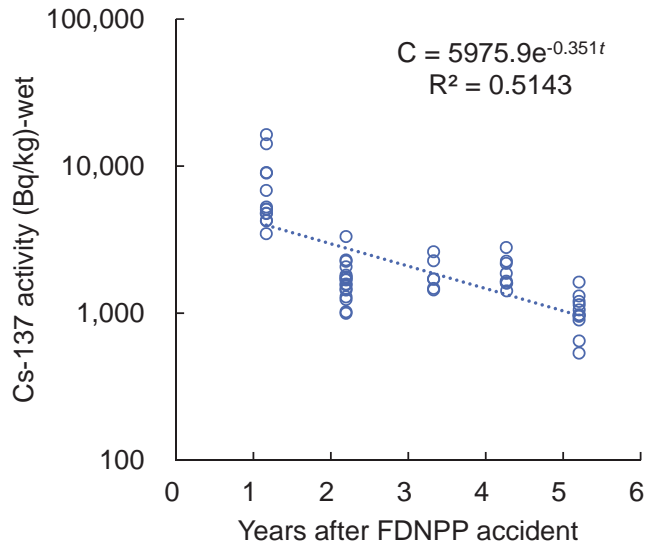
$^{134}\text{Cs} + ^{137}\text{Cs}$





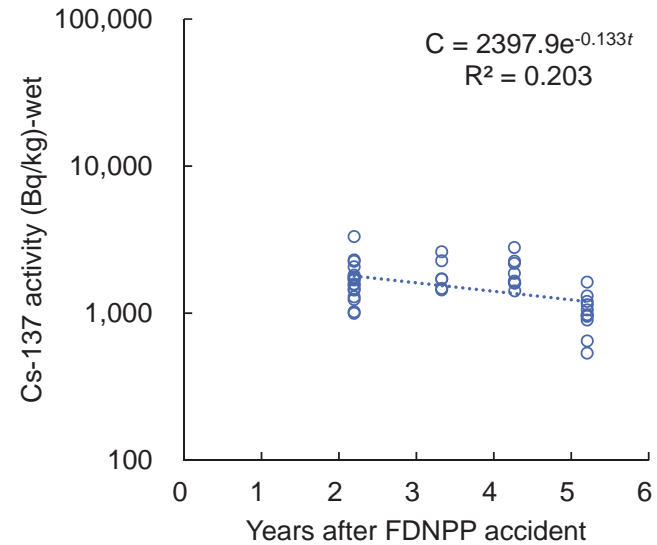
# Biological half-life of $^{137}\text{Cs}$

From 2012 to 2016



Estimated biological half-life of  $^{137}\text{Cs}$ : 1.85 yr

From 2013 to 2016



Estimated biological half-life of  $^{137}\text{Cs}$ : 2.98 yr





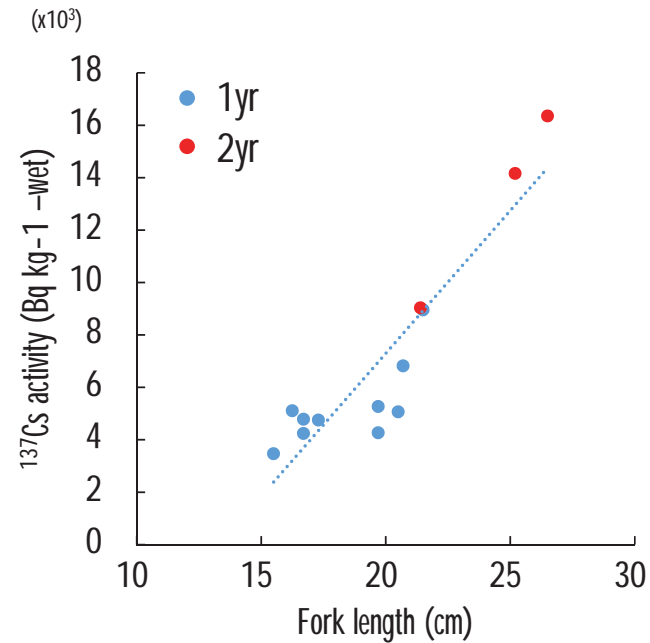
# Relationship between age and radioactive cesium concentration in trout captured at the Ukedo River in 2012



**Omm13**

# circuli on the scale: 38

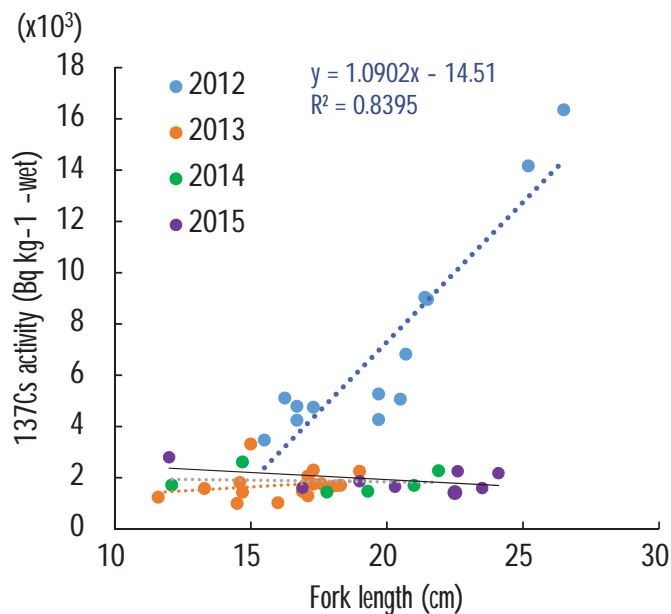
# annuli on the scale: 2 (2 year-old)



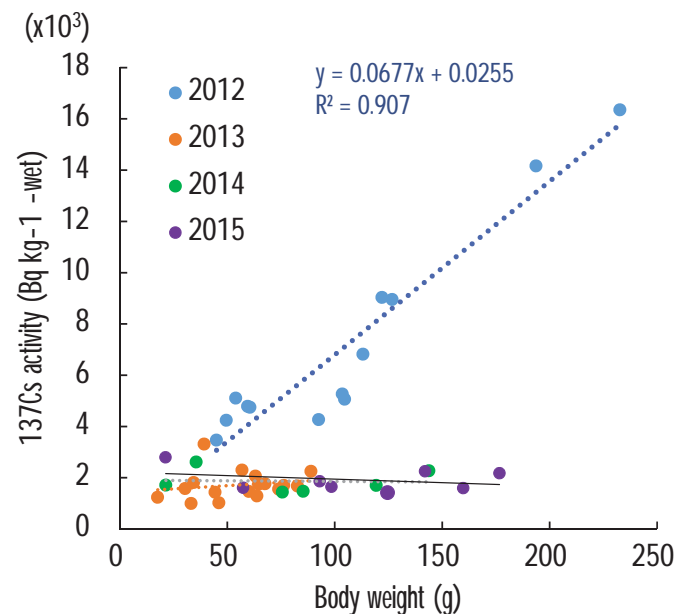


# Comparison of radioactive cesium concentration in muscle with body length (fork length) and body weight

## Fork length vs. $^{137}\text{Cs}$



## Body weight vs. $^{137}\text{Cs}$







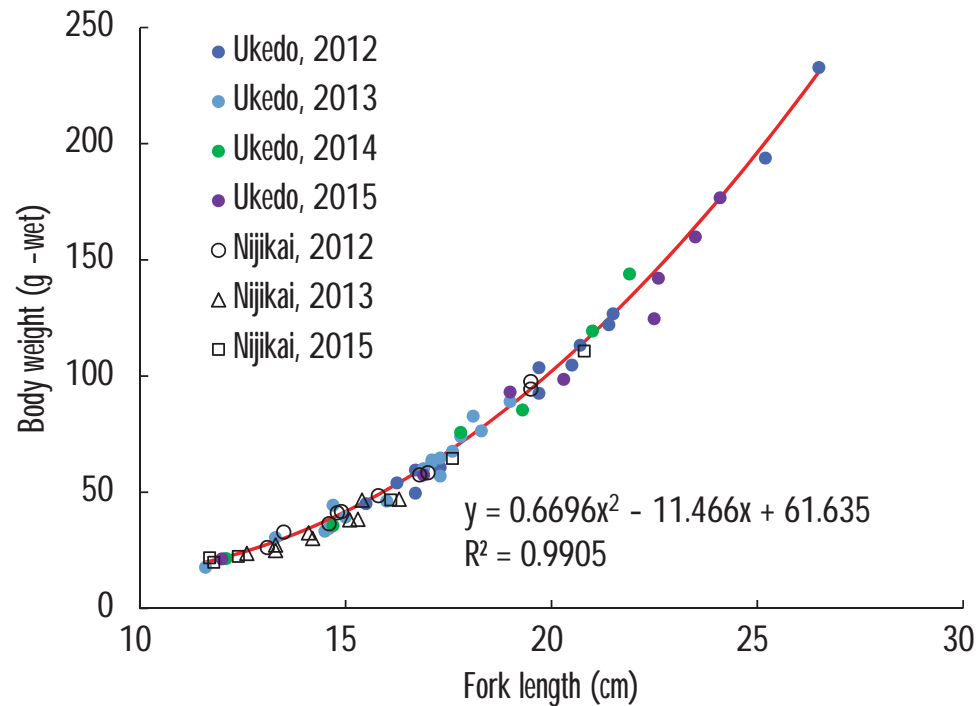
## The relationship between body weight and fork length in trout from Uketo River and Nijikai River

River	Month/Year	n	a	b ± SE	r <sup>2</sup>	95% CL of b	Growth pattern
Ukedo	May 2012	13	0.00000760	3.092 ± 0.0991	0.9888	2.873–3.310	Isometric
	May 2013	17	0.00000376	3.233 ± 0.1451	0.9707	2.924–3.543	Isometric
	July 2014	6	0.00000434	3.205 ± 0.1354	0.9929	2.829–3.581	Isometric
	June 2015	8	0.00001429	2.970 ± 0.1073	0.9922	2.708–3.233	Isometric
	Total	44	0.00000689	3.082 ± 0.0527	0.9879	2.975–3.188	Isometric
Nijikai	May 2012	10	0.00000778	3.092 ± 0.1164	0.9888	2.824–3.361	Isometric
	May 2013	9	0.00001625	2.927 ± 0.2837	0.9383	2.256–3.598	Isometric
	July 2015	6	0.00001877	2.912 ± 0.1279	0.9923	2.557–3.267	Isometric
	Total	25	0.00001347	3.049 ± 0.0950	0.9782	2.852–3.245	Isometric





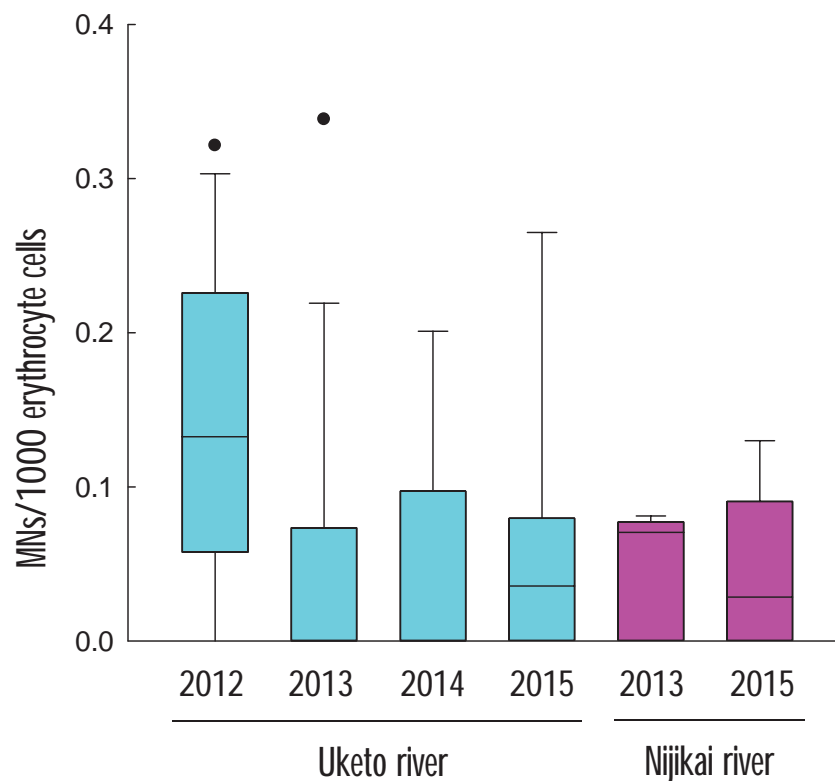
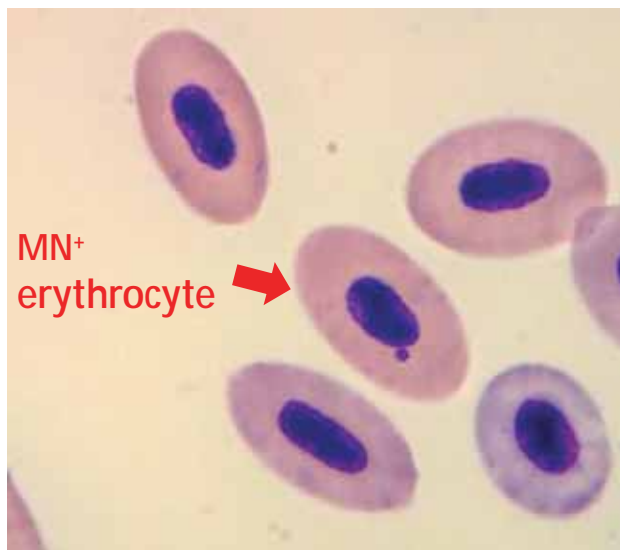
# The growth of trout captured in the Ukedo River and Nijikai River during from 2012 to 2015





# Kinetics of micronucleus frequency in trout erythrocyte cells

Giemsa-stained blood cell of trout



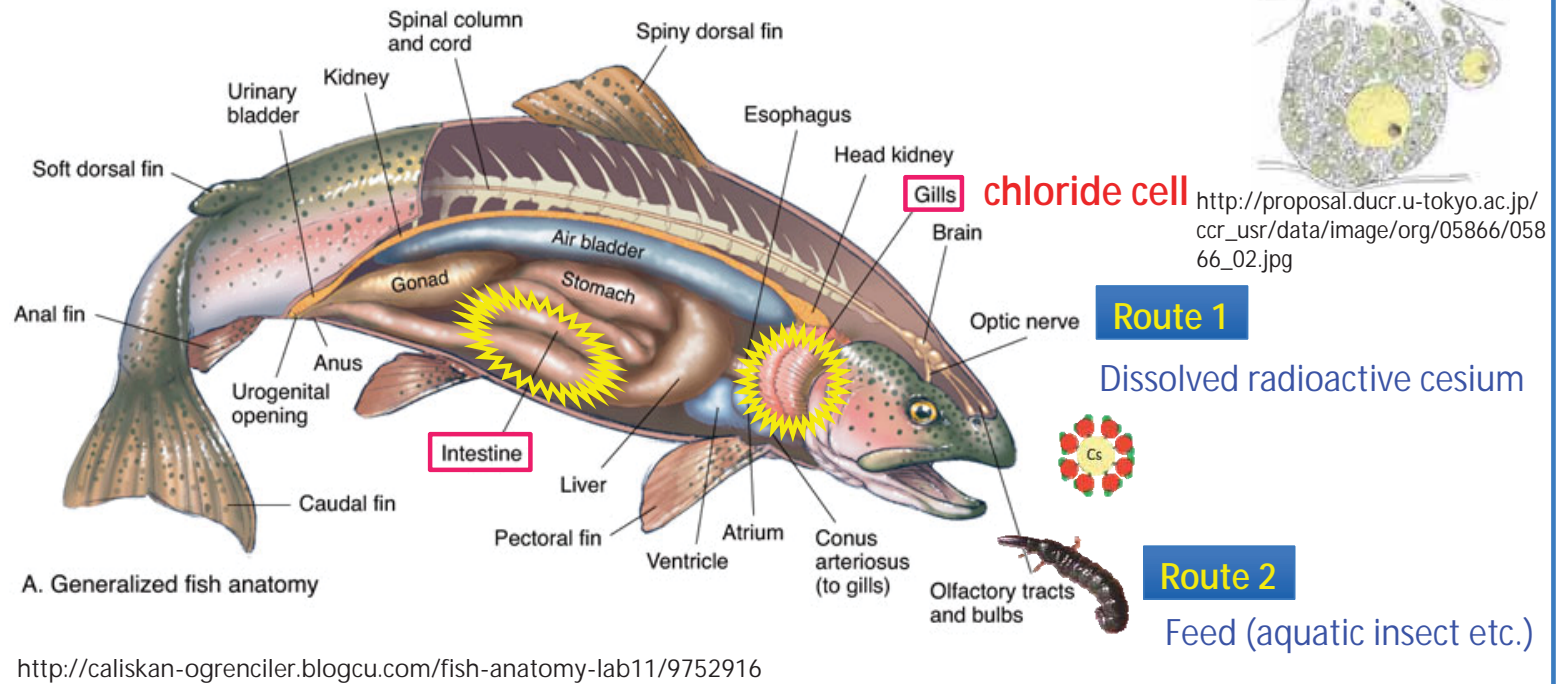


# Uptake route of radioactive cesium into the body

Radioactive cesium absorbed to suspended particles is  
low bioavailability



## Uptake route of radioactive cesium assumed





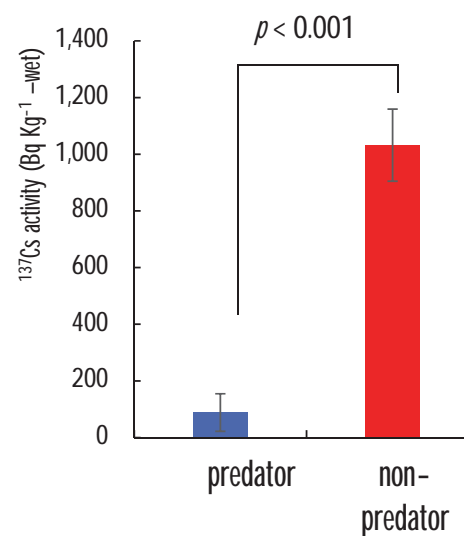
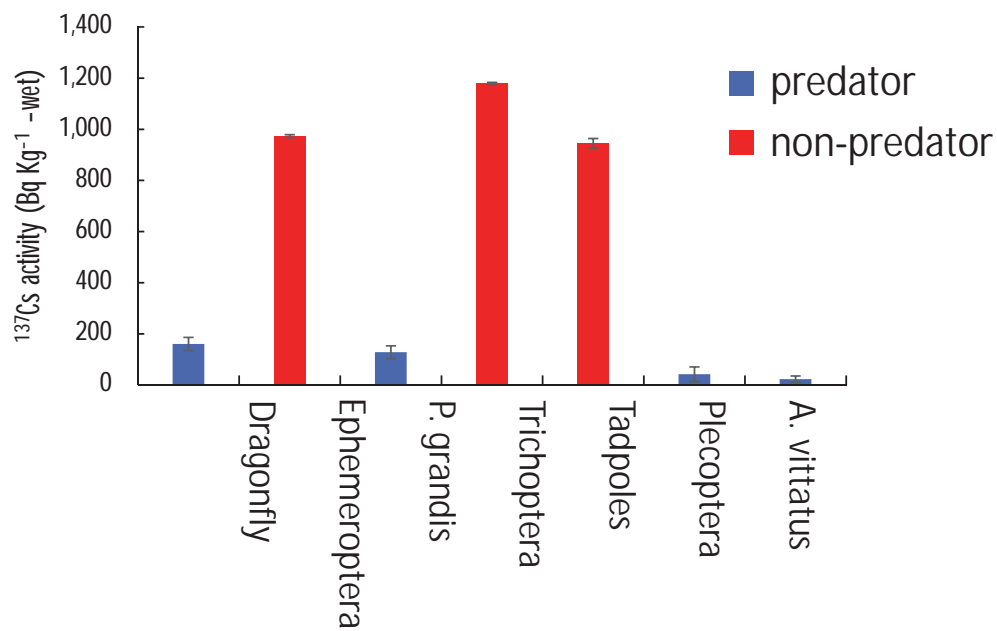
# Transfer pathway of radioactive cesium





# Radioactive cesium concentration in aquatic insects collected at the Ukedo River in 2015

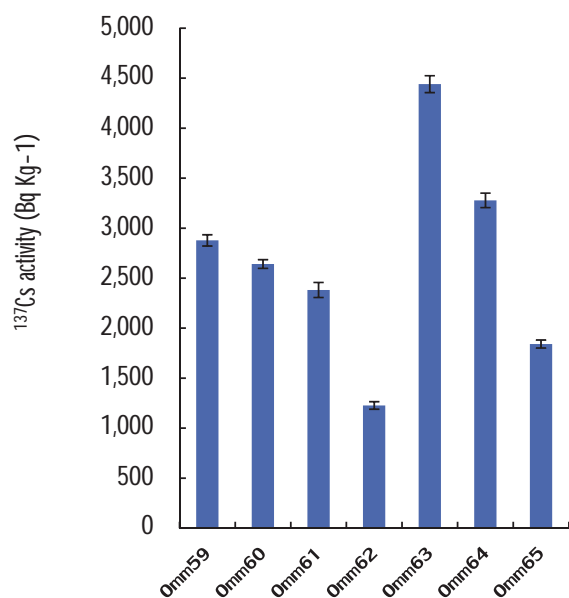
Dragonfly Ephemeroptera *Protohermes grandis* Trichoptera Tadpoles Plecoptera *Aphelocheirus vittatus*



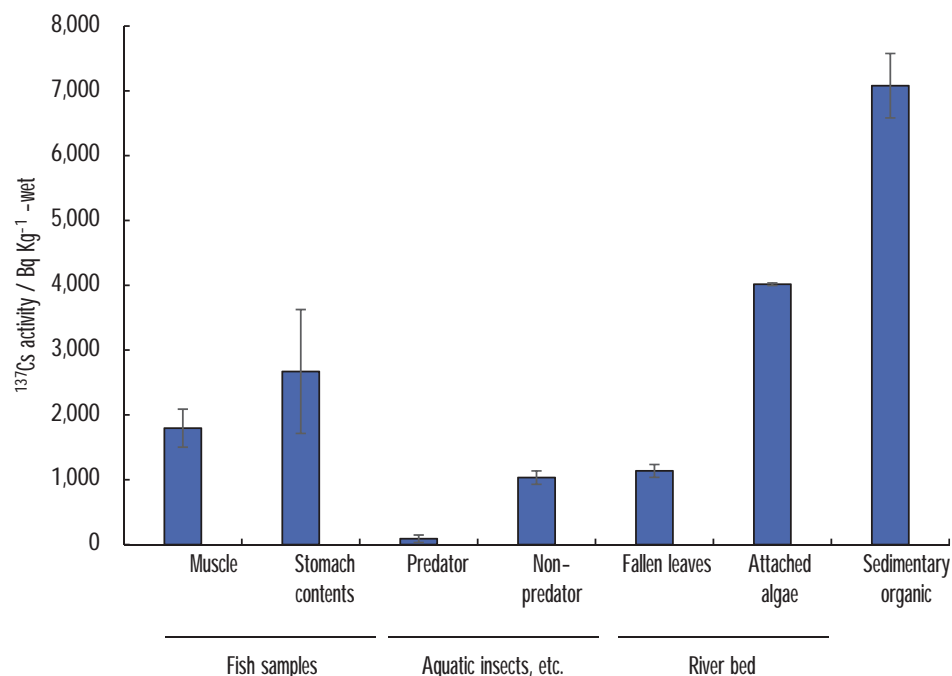


# Comparison of radioactive cesium concentration of fish sample and environmental sample in 2015

## Stomach contents

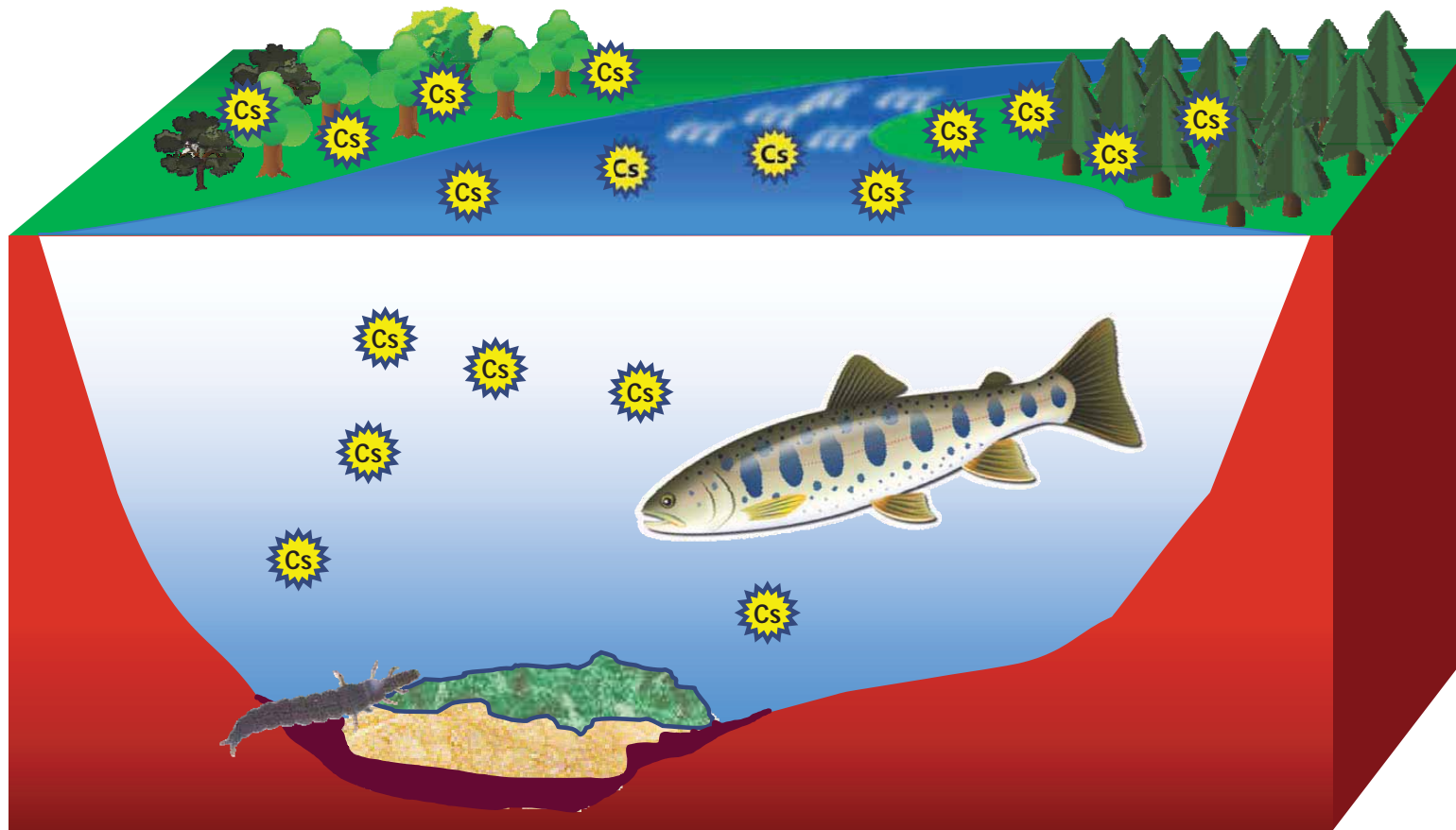


## Comparison of <sup>137</sup>Cs concentration





# Dynamics of radioactive cesium in river







# Conclusion

- Radioactive cesium was accumulated at high concentrations in the muscle tissues of trout in Ukedo River, and its concentration was high even in 2016.
- Size effect was confirmed only in the trout which was captured in the Ukedo River in 2012
- Analysis "Body length-body weight relationship" showed no growth retardation due to radiation.
- Analysis of chromosomal aberrations (micronuclei) showed a high tendency only in individuals captured in the Ukedo River in 2012. However, there was no correlation between body length and radioactive cesium concentration in muscle.
- Radioactive cesium concentration is significantly higher in non-predator type-aquatic insects, which ingest organic deposits and attached algae in the Ukedo River, and it is considered that radioactive cesium accumulates in the body of trout by ingesting these foods.





# Acknowledgements

## Colleagues

- *Hirosaki University Graduate School of Health Sciences*  
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Prof. Masatoshi Yamada, Dr. Hirofumi Tazoe
- *Hokkaido Pharmaceutical University School of Pharmacy*  
Dr. Akifumi Nakata
- *Fukushima Medical University*  
Dr. Yu Abe
- *Michinoku Fauna Research*  
Dr. Kojun Suzuki
- *National Agriculture and Food Research Organization, Institute of Fruit Tree Science*  
Dr. Toshihiko Shoji

## Supported by

- Namie town
- Takase River/Murohara River Fisheries Cooperative Association
- Izumida River Fisheries Cooperative Association
- Inland Water Fisheries Research Institute,  
Aomori Prefectural Industrial Technology Research Center





# Issues and future prospects of the studies of environmental dynamics of radiocesium

Bernd Grambow\*, \*\*

\* SUBATECH (Institut Mines Telecom Atlantique, CNRS, University of Nantes)

\*\* IAEA Advanced Science Research Center

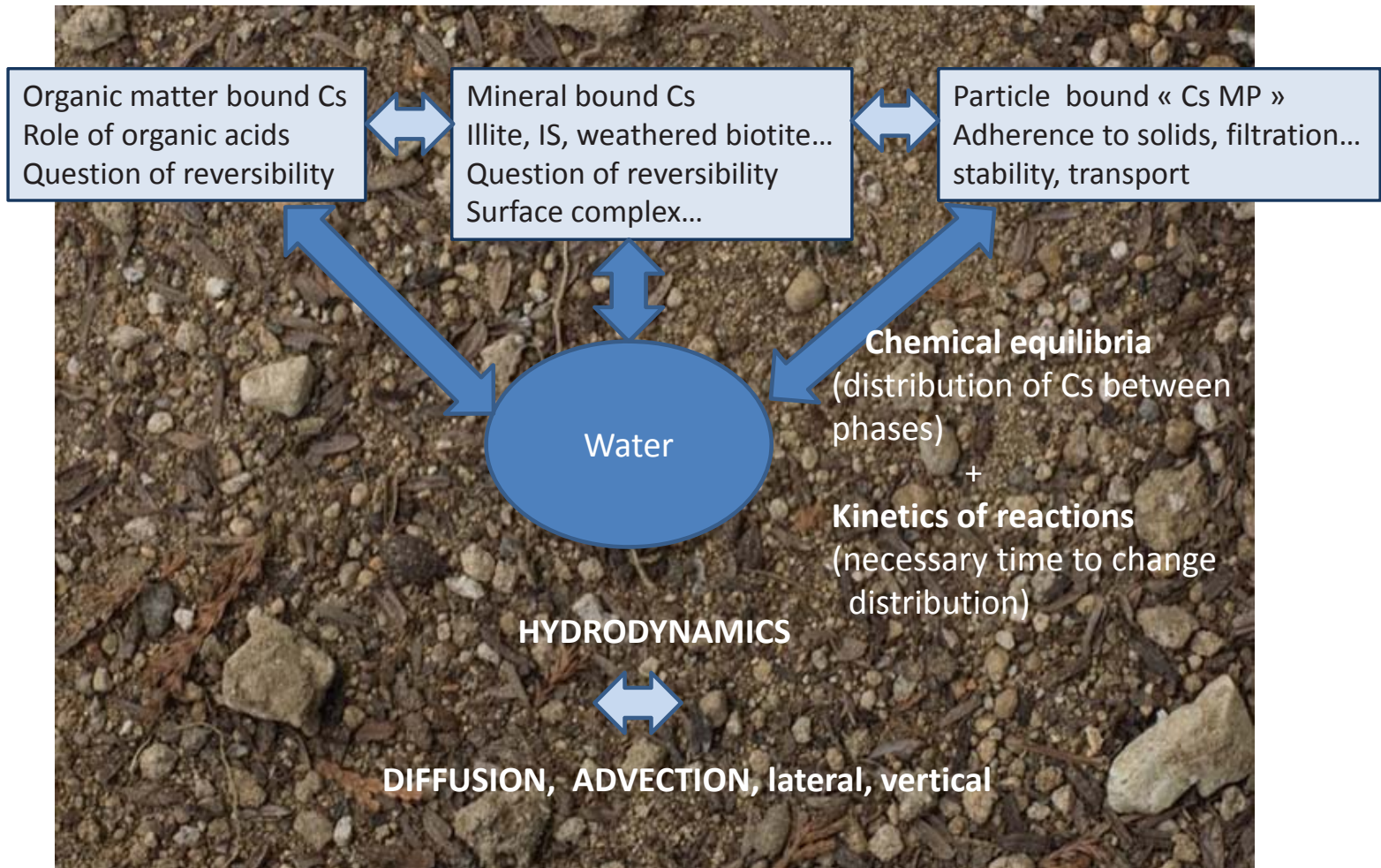
# Interaction Cs-soil

- Sorption of radiocesium [1, 2]:
  - Fixed on “frayed edge” sites FES of mica type minerals :
    - Weak or no desorption (“fixed fraction”)
  - Uniform sorption on weathered biotite
  - Sorption of Cs in form of microparticles??
  - Impact of litter/organic matter
- Migration in depth:
  - As a consequence, low uptake by vegetation
  - Redistribution in soil by erosion, rainfall and sediment transport
- Key questions:
  - In what chemical form is the Cs in the soil?
  - What are the physiochemical processes altering the depth distributions of cesium in soil and its evolution over time?
  - What is the regional variability?

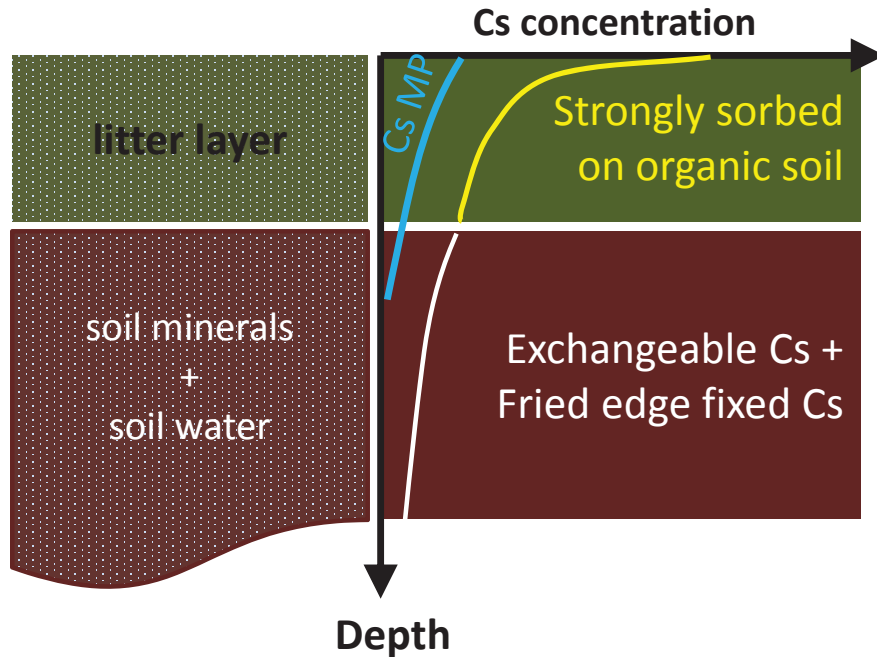
[1] H. Kurikami et al. , J. Env. Radioactivity 171 (2017) 99-109

[2] Chen, Montavon et al. Chemical Geology 387 (2014) 47–58

# The fate of Cs in the environment depend on its chemical form



# Diverse interactions of Cs with soils

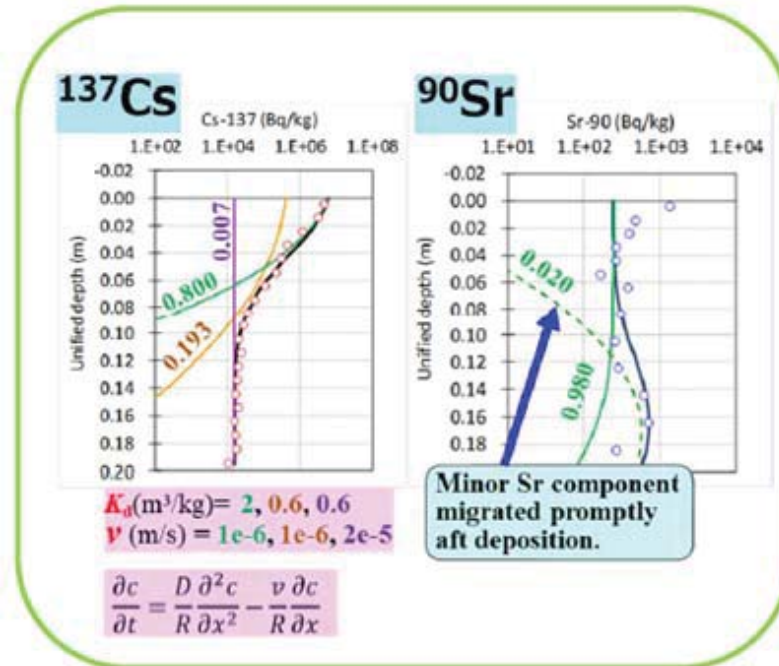


Analytics of stable Cs and Cs 137

- Bound to different soil components and in water
- Desorption tests:
  - exchangeable fractions
  - time constants
- Ratio of Cs in soil water/adsorbed

- Predicting the temporal evolution of the soil profile:
  - model interaction of Cs MP + water, litter and soil minerals
  - predict Cs wash off
  - predict evolution of Cs mobility with time

# Interpretation of the distribution of Cs in soils with depth by chemical models: Results obtained by the University of Kyoto

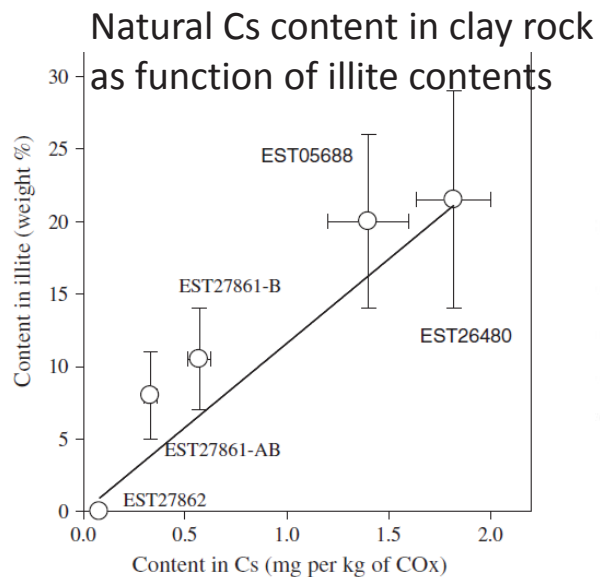


However, soils vary in mineral contents, organic matter, litter cover etc. There is still a long way to go from this interpretation to predictive models: We need to account for

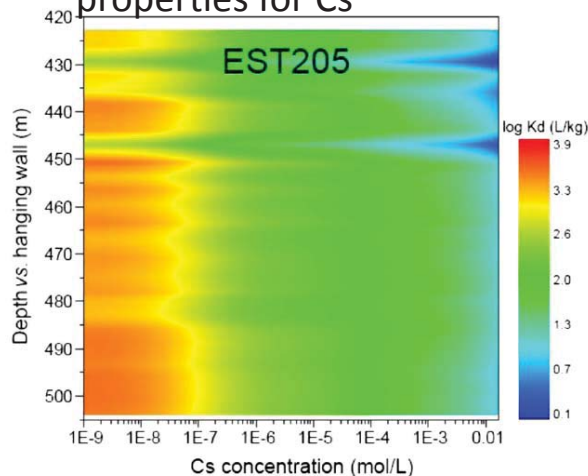
- variation in soil composition
- Organic/mineral distribution
- temporal evolution,
- Fraction of irreversible fixation of Cs etc:



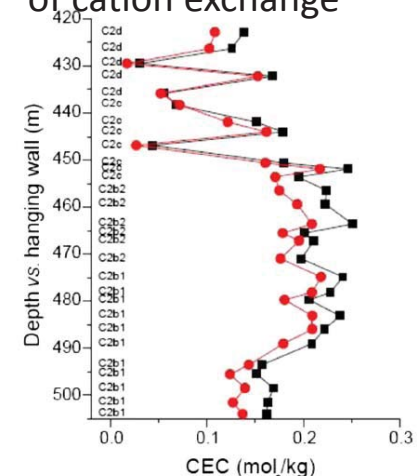
# How taking into account the mineralogical heterogeneity: Example from SUBATECH study on clay rock



predicted retention properties for Cs



measured heterogeneity of cation exchange



## Parameters used to describe Cs sorption

Type of site	Reference clay	Exchange reaction	logK		
"FES"	Illite	Cs/Na	7		
		K/Na	2.4		
Type II	Illite	NH <sub>4</sub> /Na	3.5		
		Cs/Na	3.6		
		K/Na	2.1		
Planar	Illite	Cs/Na	1.6		
		K/Na	1.2		
		Ca/Na	0.7		
		Mg/Na	0.7		
		Sr/Na	0.7		
		Montmorillonite	Montmorillonite	Cs/Na	1.7
				K/Na	1.1
				Ca/Na	0.6
				Mg/Na	0.6
		Parameters for weathered biotite missing:		Sr/Na	0.3
Sr/Na	0.3				


Collaboration U Hokkaido

Chemical Geology 387 (2014) 47–58

Contents lists available at ScienceDirect

**Chemical Geology**

journal homepage: [www.elsevier.com/locate/chemgeo](http://www.elsevier.com/locate/chemgeo)

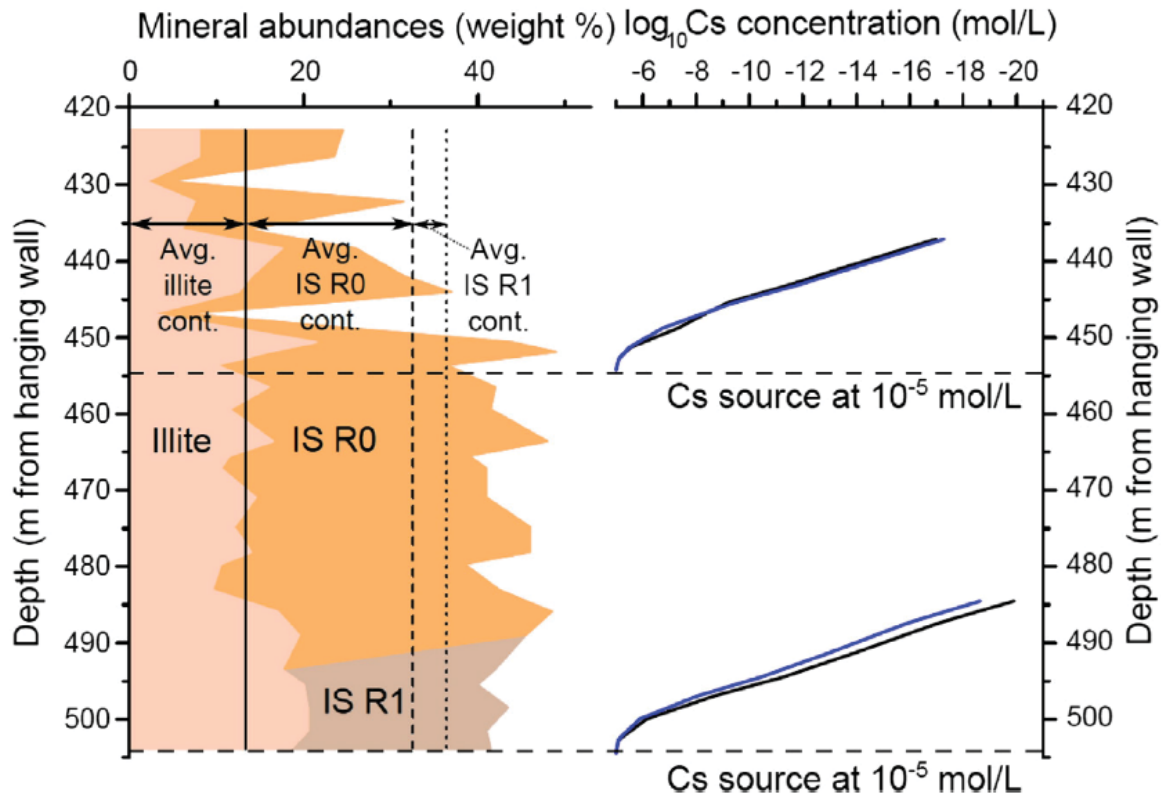


## Key factors to understand *in-situ* behavior of Cs in Callovo–Oxfordian clay-rock (France)

Z. Chen <sup>a,b</sup>, G. Montavon <sup>a,\*</sup>, S. Ribet <sup>a</sup>, Z. Guo <sup>b</sup>, J.C. Robinet <sup>c</sup>, K. David <sup>a</sup>, C. Tourmassat <sup>d</sup>, B. Grambow <sup>a</sup>, C. Landesman <sup>a</sup>

# Predicting Cs transport by diffusion, considering mineralogical heterogeneity

Example: Reactive transport calculation with large depth in clayrock

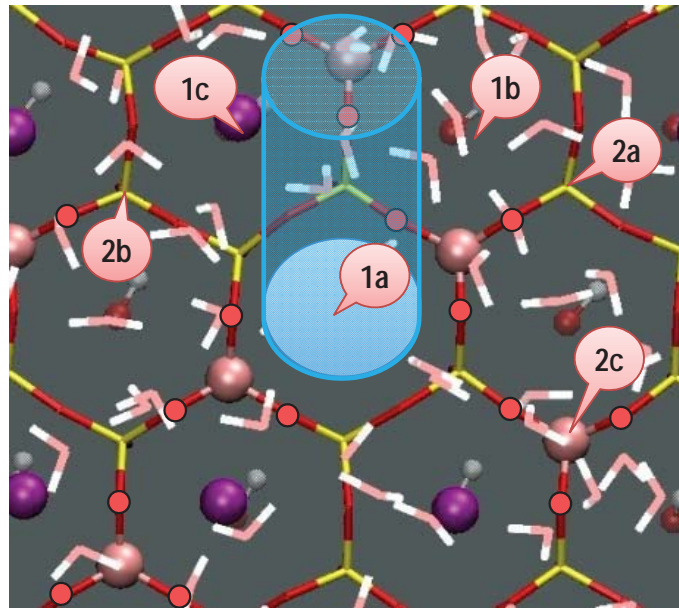


# Molecular modelling: Surface Adsorption Sites for Cs<sup>+</sup> and UO<sub>2</sub><sup>2+</sup> on Muscovite

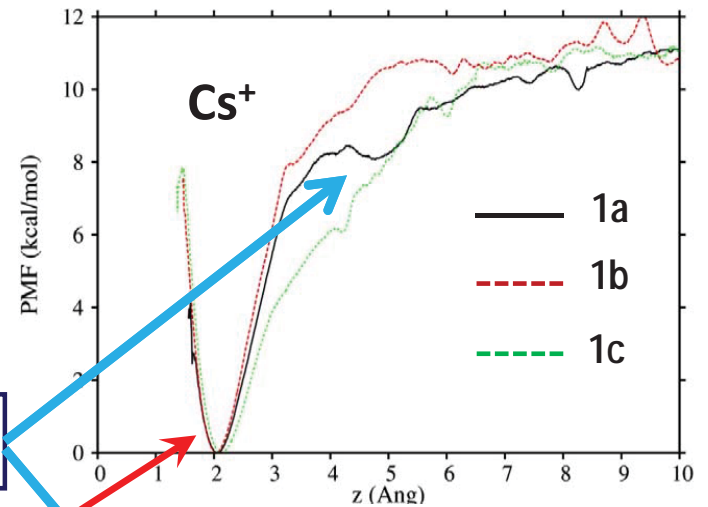
## Potential of Mean Force:

$$\text{PMF} \equiv W(z) = -k_B T \ln(\rho(z)) + \text{const}$$

$z$  - distance from the surface

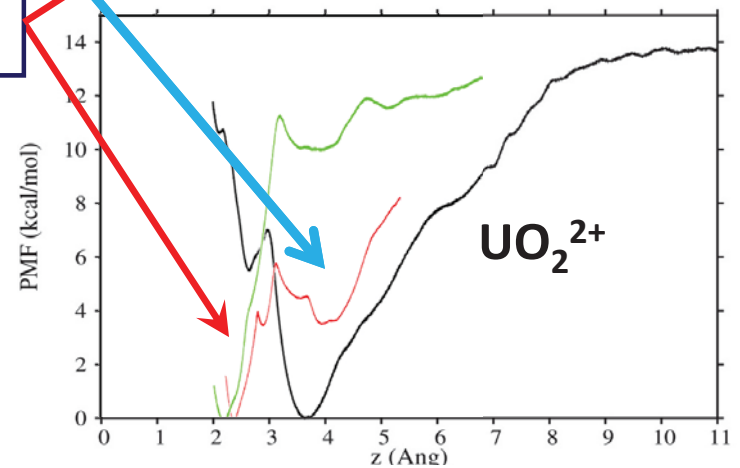


N. Loganathan, A.G.Kalinichev, 2013.



outer-sphere

inner-sphere



# What is the time dependency of desorption of Cs?

The desorption rate depends on soil type

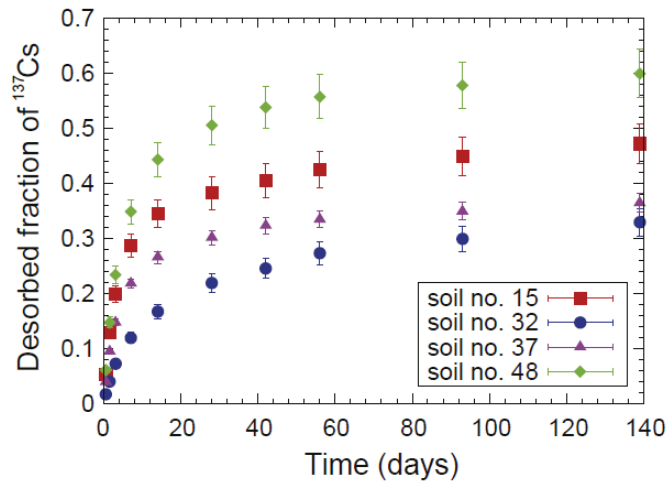


Fig. 1. Fractions of the cumulative desorption of  $^{137}\text{Cs}$  from the four different soil samples (soil no. 15, 32, 37, and 48) by  $10^{-3}$  M KCl solution in the presence of cation exchange resin. The amounts of the soils were 5 g.

Behavior can be modeled

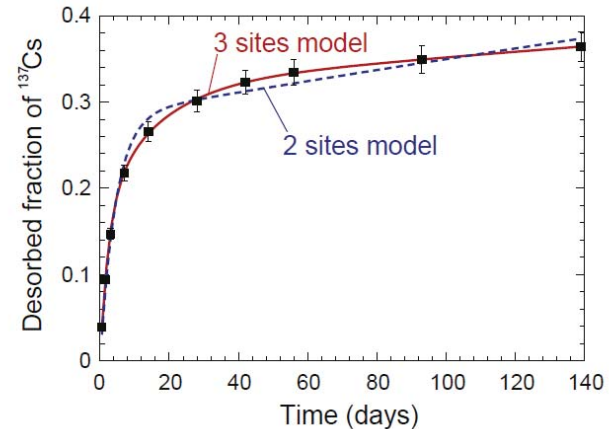


Fig. 6. Fitting results of the three- (continuous line) and two- (dotted line) sites desorption models for the fraction of the cumulative desorption of  $^{137}\text{Cs}$  from the soil no. 37 by  $10^{-3}$  M KCl as a function of time shown in Fig. 3.

Journal of Environmental Radioactivity 153 (2016) 134–140

We have to take into account long-term kinetics of Cs sorption and desorption for assessments of its migration from soils by explicitly considering it in reactive transport modeling.



Contents lists available at [ScienceDirect](https://www.sciencedirect.com)

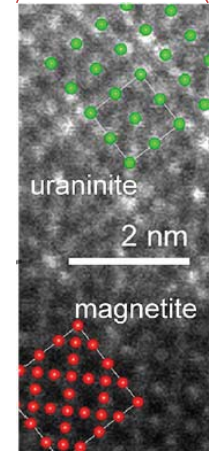
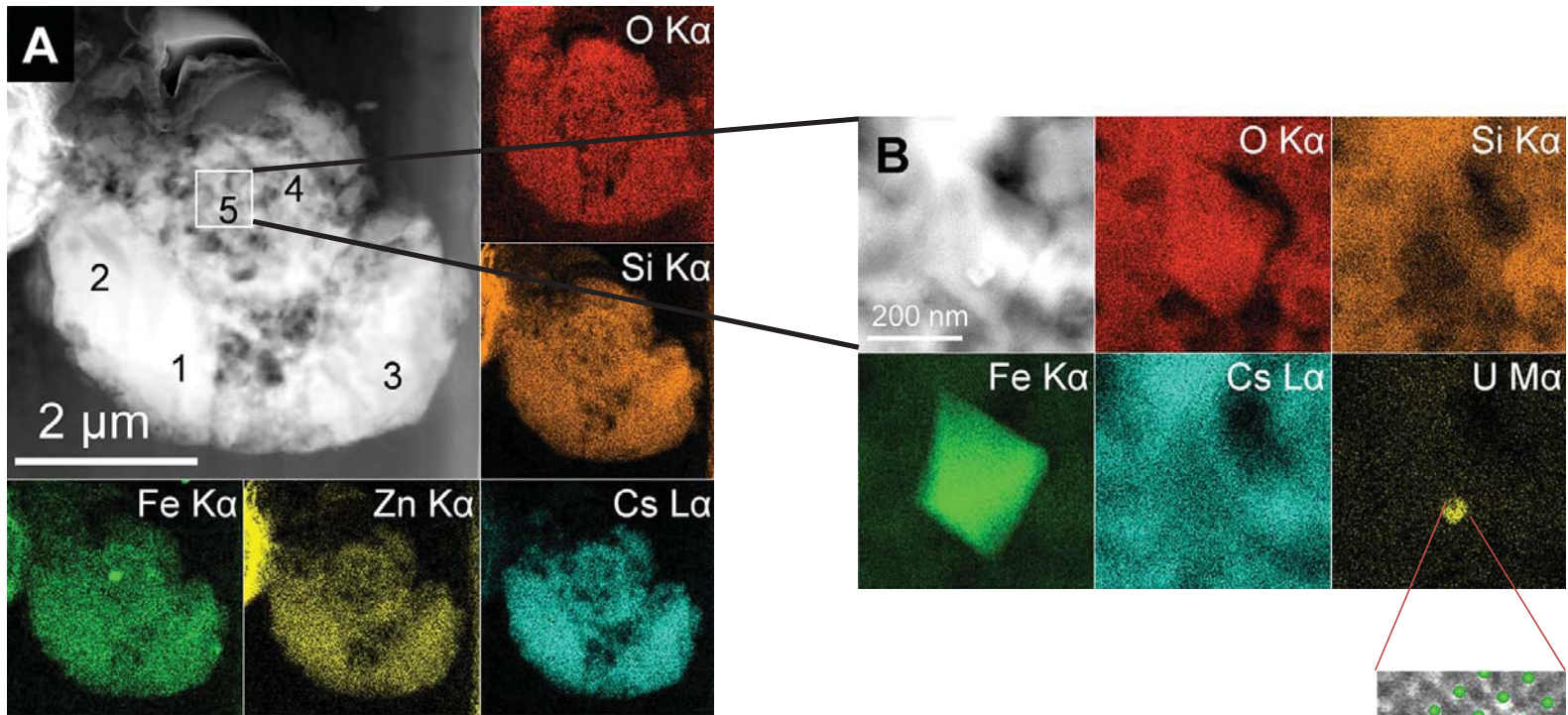
Journal of Environmental Radioactivity

journal homepage: [www.elsevier.com/locate/jenvrad](http://www.elsevier.com/locate/jenvrad)

Desorption kinetics of cesium from Fukushima soils

Kento Murota <sup>a</sup>, Takumi Saito <sup>b,\*</sup>, Satoru Tanaka <sup>a,1</sup>

# What about Cs in form of Cs microparticles?



- Which fraction of Cs is in particulate form at which geographical position?
- Does the fraction of Cs in particulate form change with time?
- What is the impact on dose (the residence time of Cs MP in the human body is different)?
- What are transport and sorption properties of the particles ?
- What is the effect of filtration?

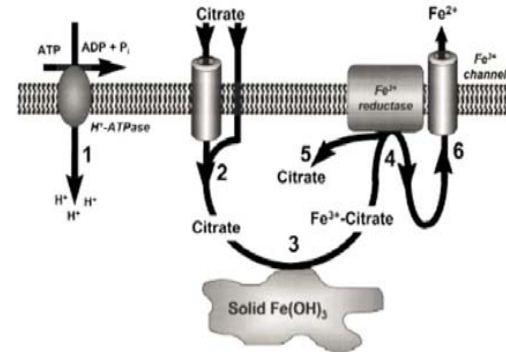
# Organic acids - soil microbes interaction

## Purpose:

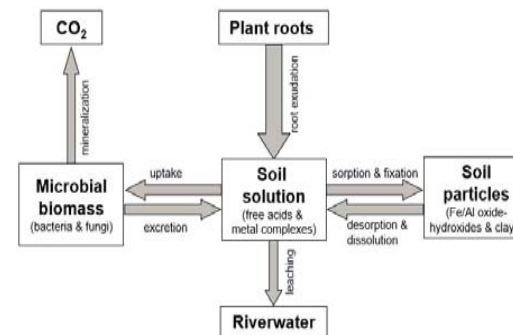
To elucidate if microbial and (plant) endophyte produced siderophores/organic acids play a role in the accumulation of cesium by fungi/mushrooms.

## Approach:

- Activation of microbes by fungi organic acid on production of siderophore.
- Synergy effect of fungi organic acids and microbes siderophores on dissolution of Cs from minerals.



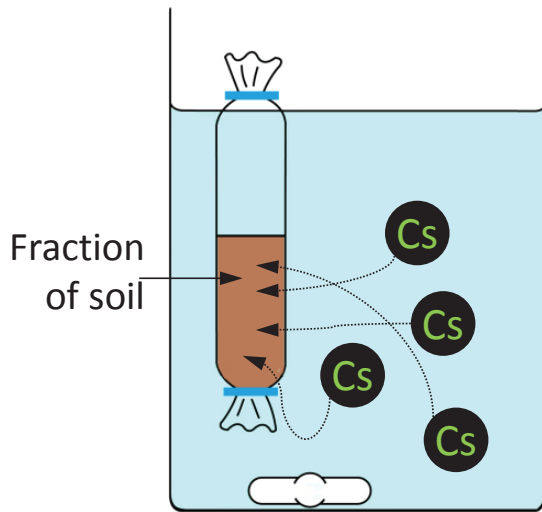
Schematic representation of the role of organic acids in Fe uptake by dicotyledonous plant roots (Jones 1998).



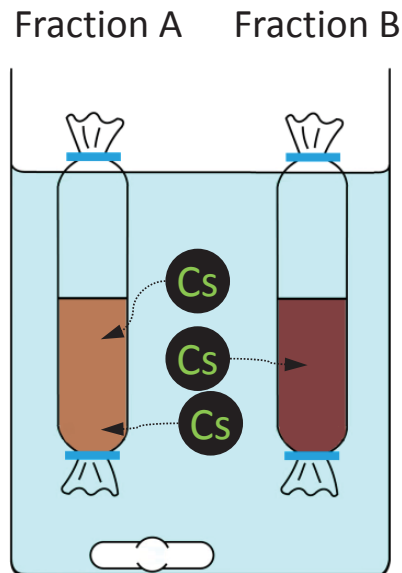
Schematic diagram of major organic acid fluxes and pools in soil (Jones 1998).

# Some experimental methods to study Cs affinity on different mineral fractions of Fukushima soil

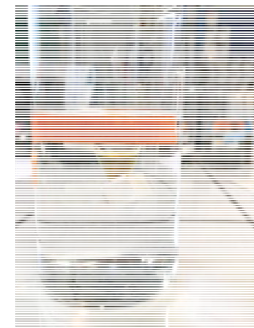
Single system



Binary system



Example of dialysis membranes





# Experiments on phytoextraction of Cs from soil

## Phytoextraction of Cs with red clover

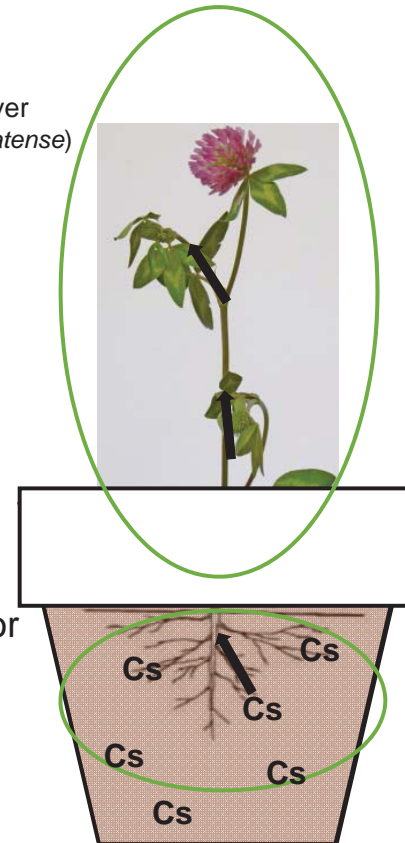
Hydroponic system

Red clover  
(*Trifolium pratense*)



Soil pots

Red clover  
(*Trifolium pratense*)



TF : translocation factor

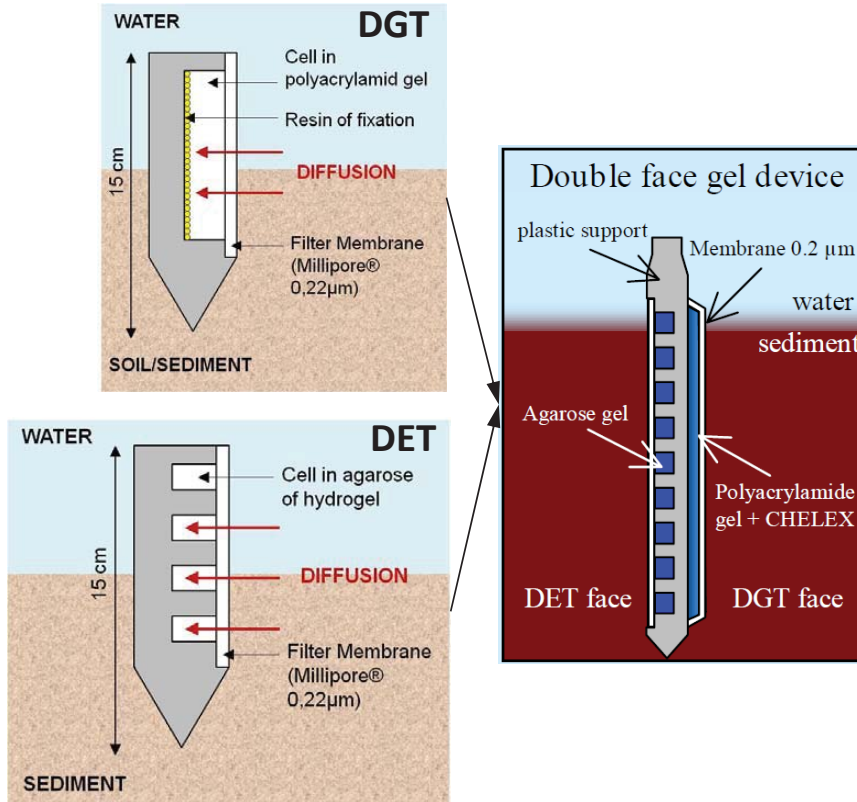
$$TF = \frac{[Cs]_{aerial\ parts}}{[Cs]_{root\ parts}}$$

BF : bioaccumulation factor

$$BF = \frac{[Cs]_{plant}}{[Cs]_{solution\ or\ soil}}$$



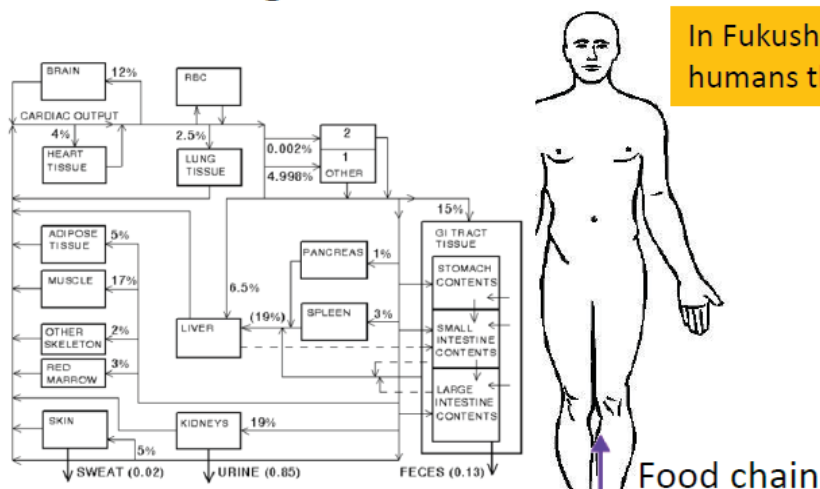
# Experiments for monitoring Cs in sediment using DGT and DET



- In situ method
- Implementation time:
  - few hours
- Access to double information
  - DET: measurement of the concentration in interstitial water
  - DGT: measurement of the concentration of labile species

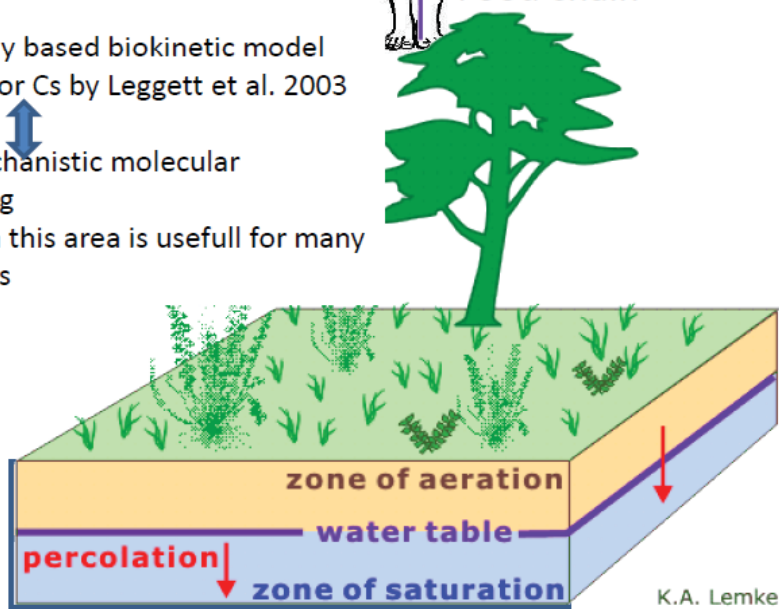
# Missing link between assessment of contamination and health

In Fukushima, whole body counters show 10 times lower Cs in humans than expected from equal soil contamination in chernobyl



Physiologically based biokinetic model developed for Cs by Leggett et al. 2003

Need for mechanistic molecular understanding  
Knowledge in this area is useful for many scientific fields



K.A. Lemke

S.F. Boulyga / International Journal of Mass Spectrometry 307 (2011) 200–210

## Nuclear medicine

*Empirical data: urines, blood...*

*Dynamic models:*

Bio-kinetic/ physiological based pharmacokinetic models (see ICRP100)



Too little interaction missing is a joint large scale studies including epidemiological perspectives

## Agricultural and Environmental radiochemistry

Distribution and fluxes of radionuclides between soils and water and food stuff in the environment

Molecular and large scale geochemical and transport models

Multiscale molecular metabolic flux "Soil/Plant model" i.e. linking K and Cs cycle,