Fukushima Research Conference on radiocesium migrations in terrestrial environments and in ecosystems（3rd International Cesium Workshop)(4th 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）

Tadafumi Niizato

（新里　忠史）

Environmental Research Group　Fukushima Environmental Safety Center
Japan Atomic Energy Agency (JAEA)

国立研究開発法人
日本原子力研究開発機構
Japan Atomic Energy Agency

福島県環境創造センター
Fukushima Prefectural Centre for Environmental Creation

Outline

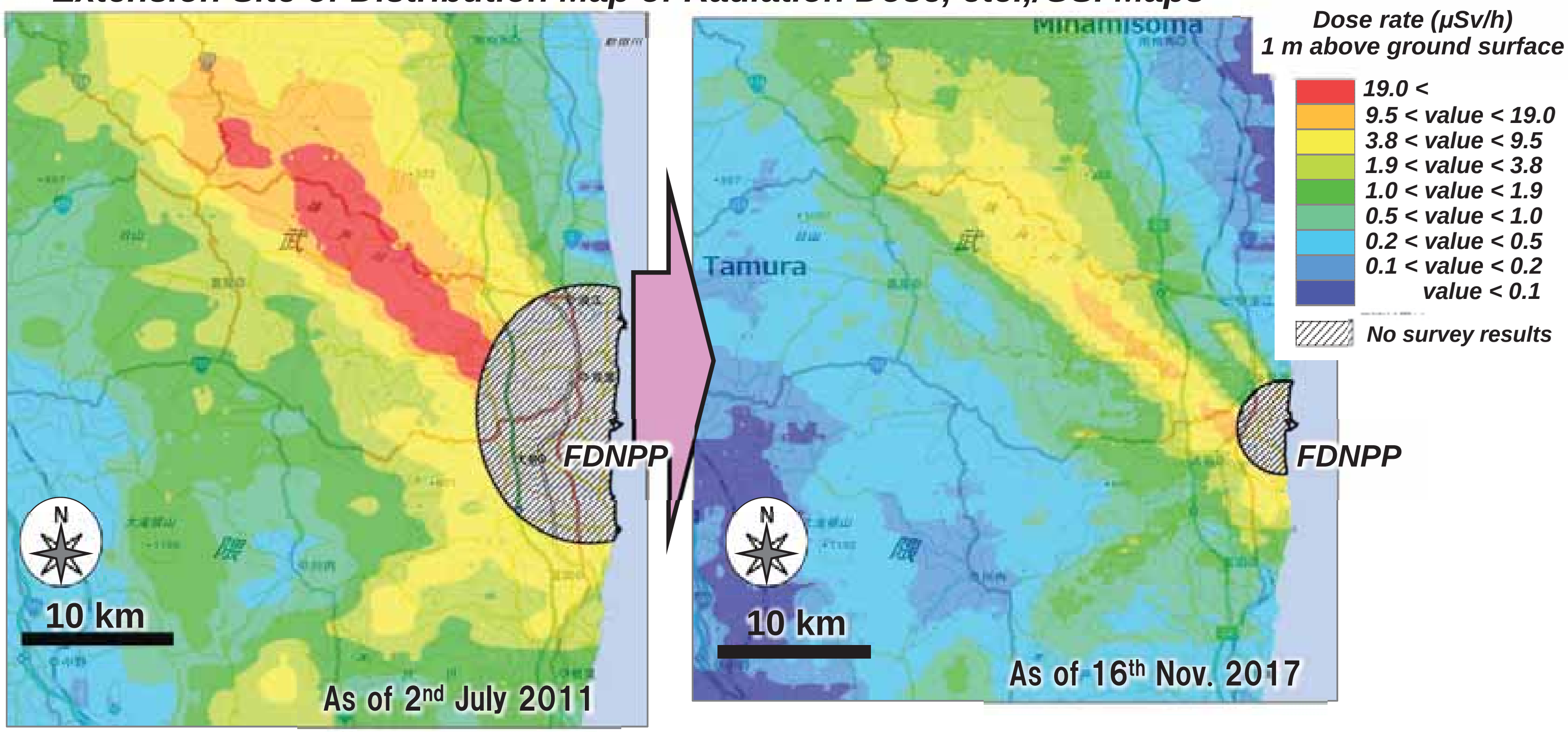
- **Introduction**
  - *Physical geography, land use, radiation dose*
  - *Transport processes*
- **Methods**
  - *Monitoring at experimental plots, tree sampling*
- **Results**
  - *Radiocesium distribution, Input/output budgets*

- **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)

The source of the map;

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

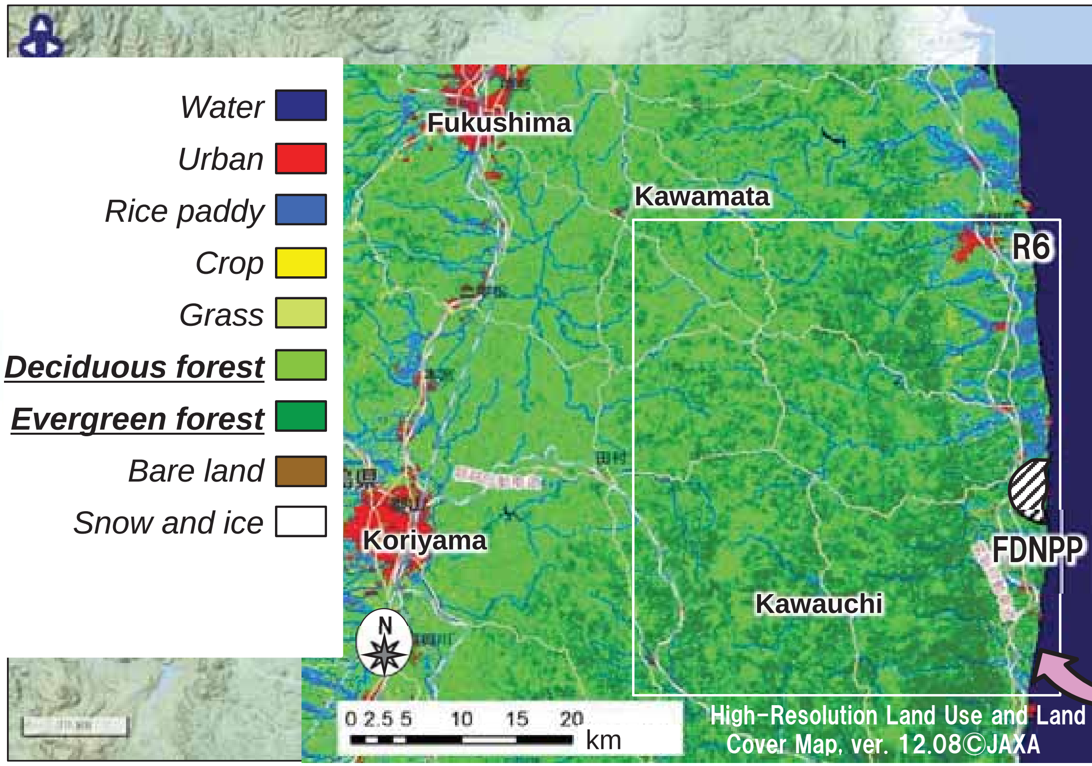


- ***Generally, radiation dose decreases with time***
- ***The area with higher radiation dose still exists***

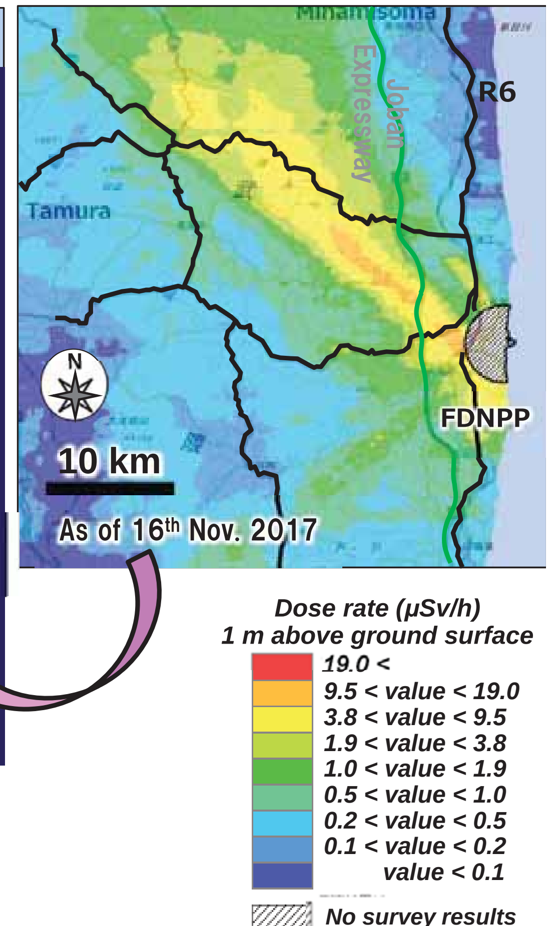
https://ramap.jmc.or.jp/map/eng

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

https://ramap.jmc.or.jp/map/eng

Comparison; physical geography

	Forested land [1),2)]	Mean average temperature [3),4)]	Annual precipitation [3),4)]	Topography
Belarus (Minsk)	42 %	Jan. -4.3 ℃ July 19.5 ℃	690 mm	Lowland Wetland (Pripet Marshes)
Ukraine (Kiev)	16 %	Jan. -4.4 ℃ July 18.5 ℃	608 mm	North; Wetland (Pripet Marches) East & west; Hilly land

Fukushima	<whole Fukushima> 68 %	<Fukushima City> Jan. 1.6 ℃ July 23.6 ℃	1,166 mm Dec. 42 mm Sept. 160 mm	Inland lowland
	<kawamata>[5)] 80 %	Jan. 0.5 ℃ July 23.6 ℃	1,368 mm Dec. 42 mm Sept. 211 mm	Mountainous & Hilly land
	<Kawauchi> 90 %	Jan. -0.7 ℃ July 26.0 ℃	1,465 mm Dec. 50 mm Sept. 227 mm	Mountainous & Hilly land

1) FAOSTAT (2010)
2) The 88th 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

Input

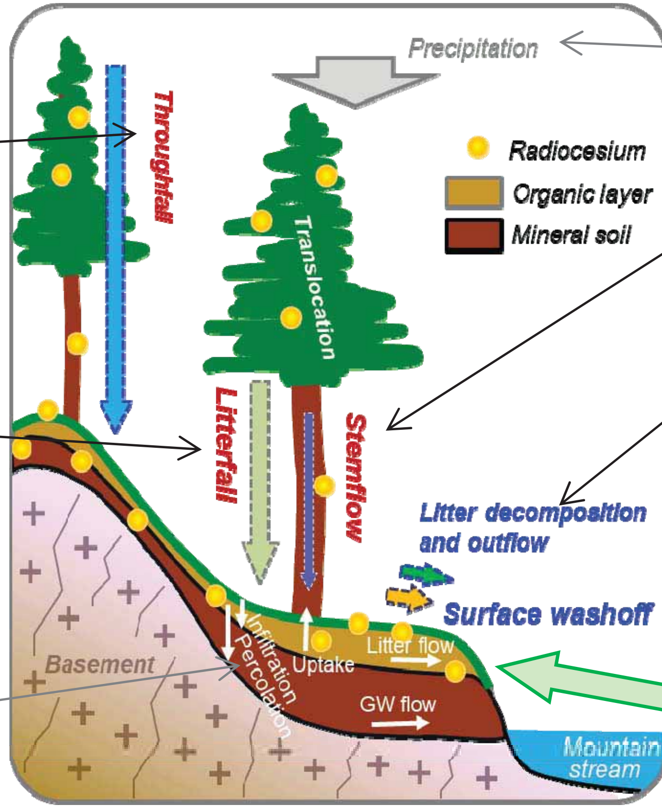
➢ Throughfall

Input

➢ Litterfall

output

➢ Migration in soil layer



➢ Precipitation Input

➢ Stemflow Input

➢ Surface runoff Output

➢ Soil loss Output

Forest floor

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.*

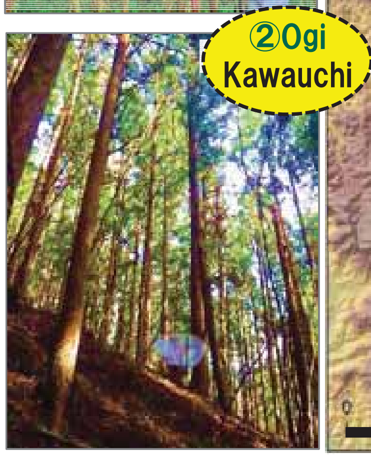
Forest investigation

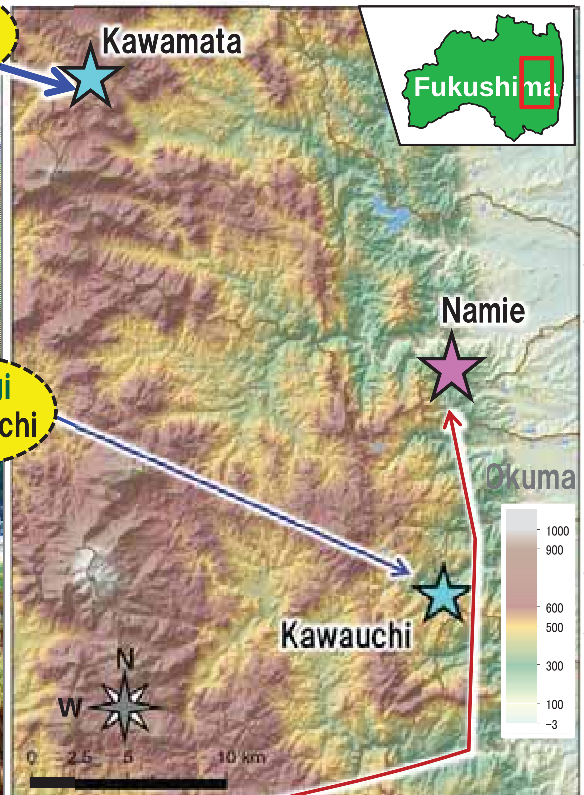
Yamakiya, Kawamata
(mixed forest; Red pine, Konara oak)

Ogi, Kawauchi
(evergreen coniferous forest, Japanese cedar)

Ide, Namie
(evergreen coniferous forest, Japanese cedar)

Outline of study areas





Forest type	Topography	Soil type

① Yamakiya, Kawamata

- ✓ **Mixed forest of deciduous and evergreen trees** (Japanese red pine and konara oak)
- ✓ **Gentle to steep** slope facing the south
- ✓ Brown forest soil (sandy loam to loam)
- ✓ **Decontaminated** and undisturbed plots

Experimental plots (gentle & steep slope)

② Ogi, Kawauchi

- ✓ **Evergreen forest (Japanese cedar)**
- ✓ Ogi A; **Intermediate to steep** slope facing the north
- ✓ Ogi B; **Steep** slope facing south
- ✓ Brown forest soil (silt loam to loam)
- ✓ Undisturbed plot

Experimental plots (steep slope)

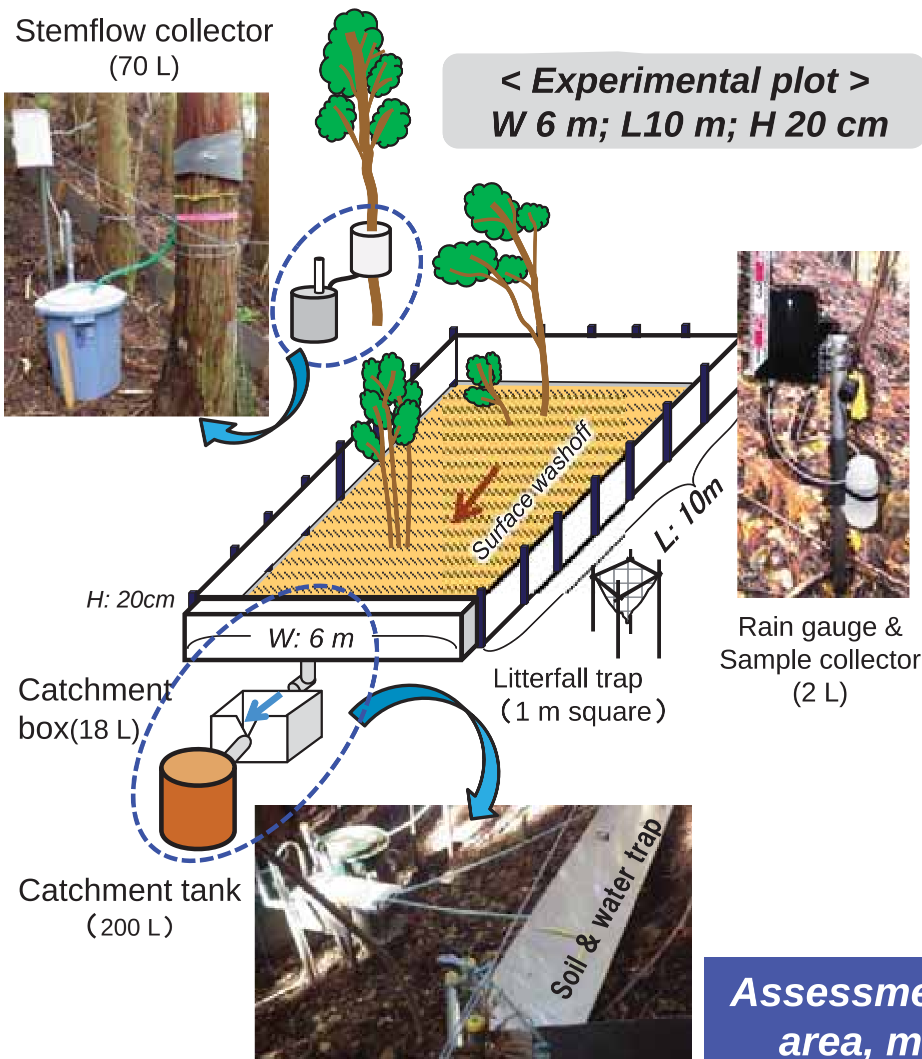
③ Ide, Namie

- ✓ **Evergreen forest (Japanese cedar)**
- ✓ **Steep** slope facing the south
- ✓ Brown forest soil **(sand)**
- ✓ Undisturbed and **disturbed plots by wildfire**

Experimental plots (steep slope)

- ✓ **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



Canopy to forest floor (radioCs input)

- ❑ **Throughfall**
  - ✓ Amount of throughfall (mm=L/m^2)
  - ✓ radioCs activity (Bq/L)
- ❑ **Stemflow**
  - ✓ Stemflow volume (L/m^2)
  - ✓ radioCs activity (Bq/L)
- ❑ **Litterfall**
  - ✓ Weight of litterfall (kg/m^2)
  - ✓ 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^2)
  - ✓ Surface runoff volume(L/m^2)
  - ✓ 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^{-2} period^{-1}]

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

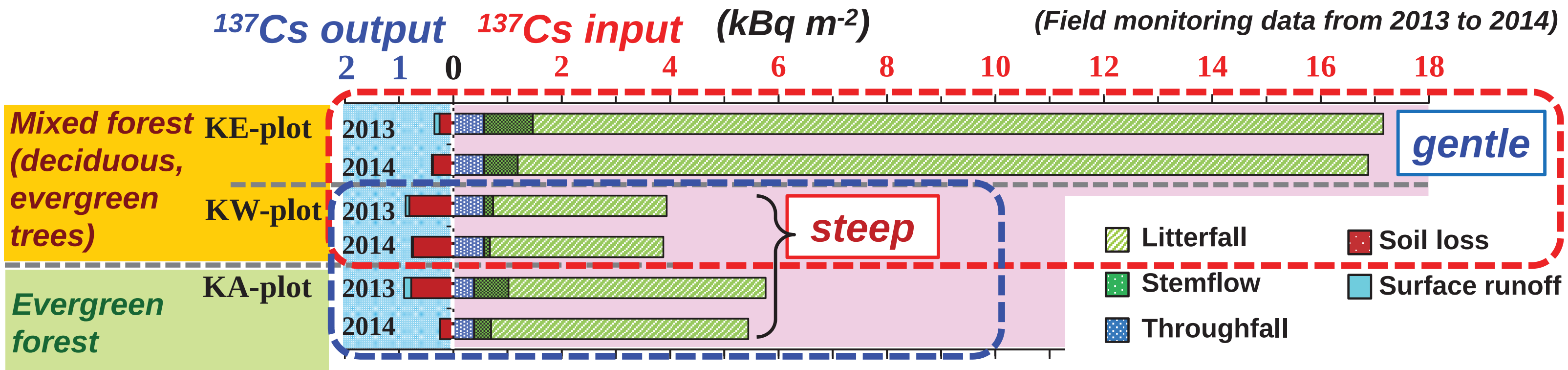
❑ Mass movement

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

❑ Nivation

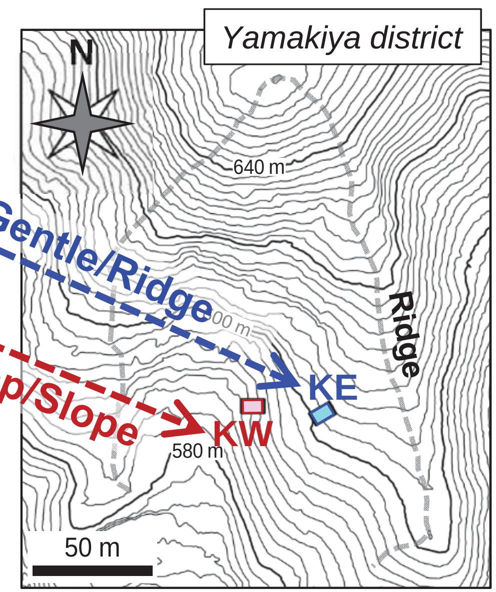
- ✓ Erosion by melting water
- ✓ Shaving and grooving of top soil
- ✓ Snow season and snow melt season
- ✓ Limited area in the forest

^{137}Cs input/output budgets, forest floor basis



(※)6th Airborne monitoring results

Plot	Year	Inventory※ (^{137}Cs (kBq m^{-2}))	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



Contour interval 2m

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

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

^{137}Cs output in the various forest floor conditions

Decontaminated plot

2 m × 4 m, **dips 27° west**

Cover rate approx.30%

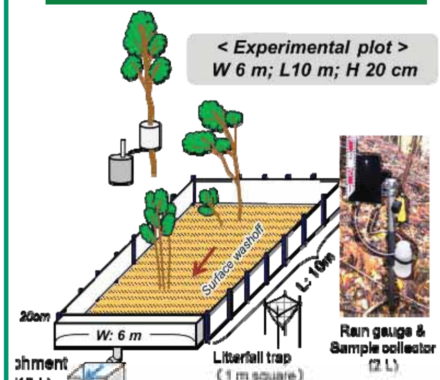
Disturbed plot by wildfire

Monitoring period
19th June – 20th Nov., 2017

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

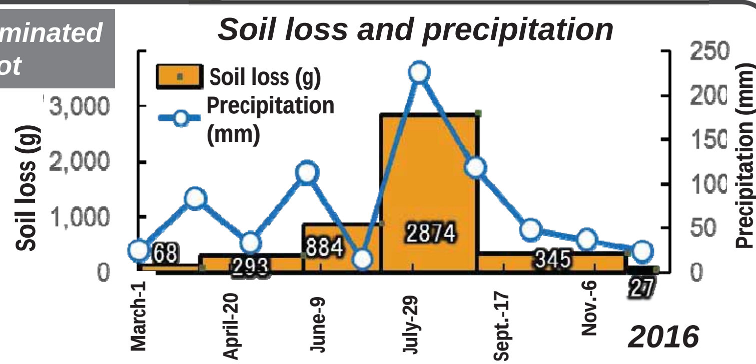
Plot	^{137}Cs Inventory [Bq/m^2]	^{137}Cs output [Bq/m^2]	Output rate
Disturbed	416k	10,377	2.49 %
Undisturbed	547k	798	0.15 %

Undisturbed plot



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

Decontaminated plot



Soil loss and ^{137}Cs output

Soil loss	^{137}Cs output ①	^{137}Cs inventory②	^{137}Cs output rate (①÷②)
0.56 kg/m^2	10.5 kBq/m^2	1,600 kBq/m^2	0.66 %

Undisturbed plot	Yamakiya (mixed forest; steeply inclined)			
Year	2013	2014	2015	2016
^{137}Cs outflow via soil loss [Bq/m^2](April – Nov.)	810	1,721	180	308
^{137}Cs inventory around the plot [kBq/m^2]	1,600 (Nov. 2013)			
^{137}Cs outflow rate [%](April – Nov.)	0.05	0.11	0.01	0.02%

^{137}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

⓪ **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

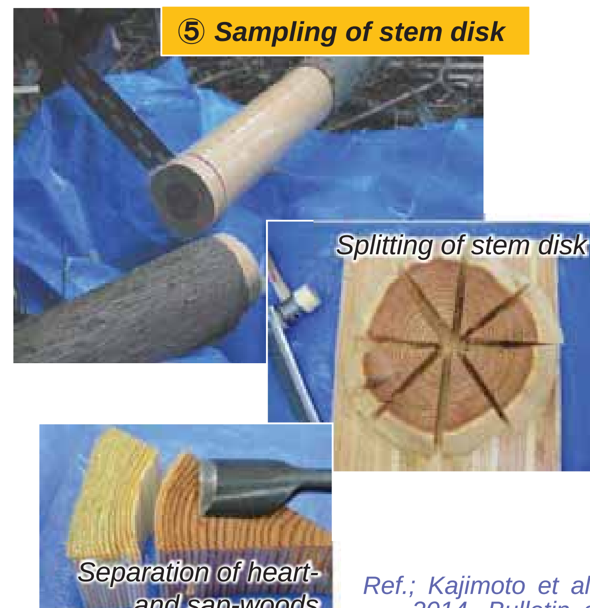
⑤ **Stem disk** → splitting, separation of heart- and sap-woods, weighting, bulk density, pretreatment, Ge analysis

② Cutting

③ Needle and twig sampling

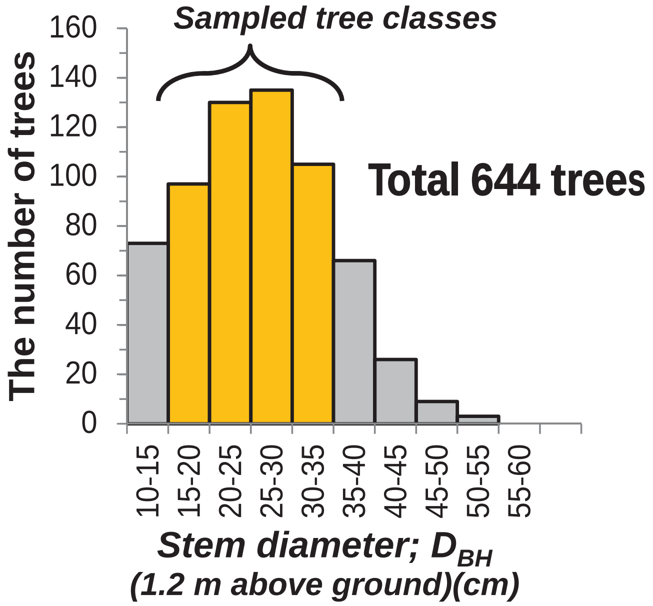
④ Bark sampling

⑤ Sampling of stem disk

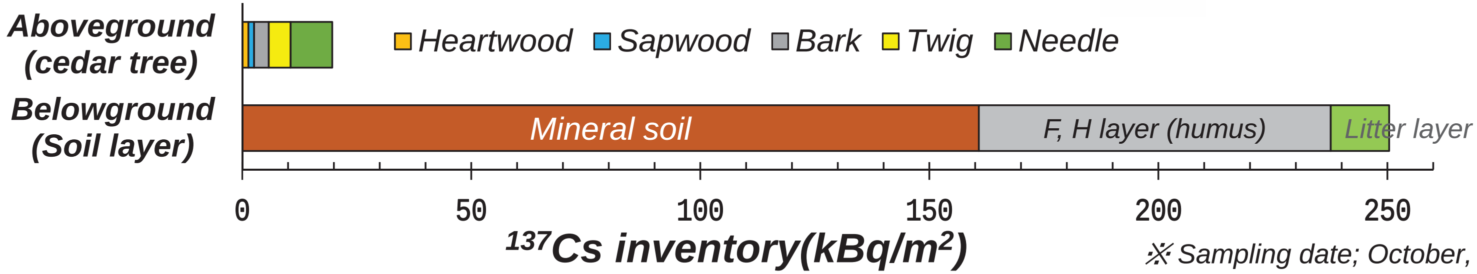


Ref.; Kajimoto et al., 2014, Bulletin of FFPRI, vol.13, no.3, 113-136.

Aboveground and belowground ^{137}Cs inventories



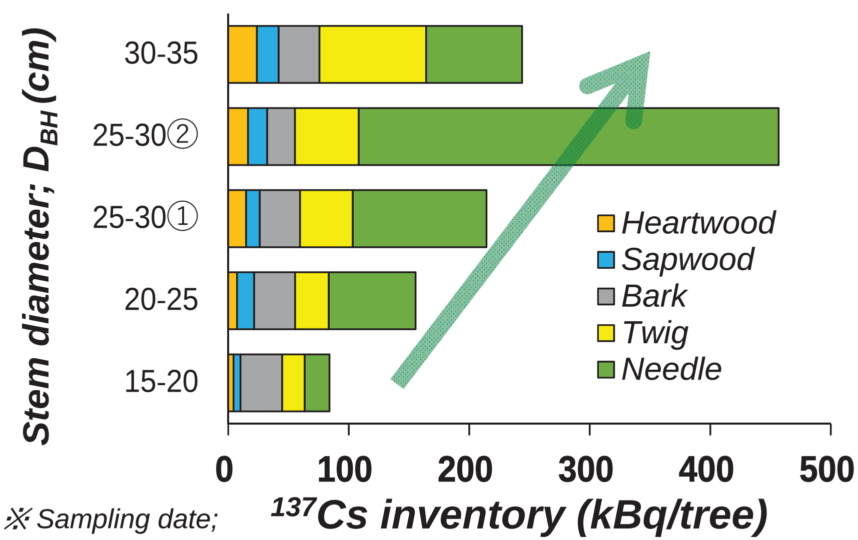
- ***Aboveground (cedar tree)*** **<<** ***Belowground (litter & soil layer)***
- ***radioCs mostly exists in the belowground***
  (approx. 90 % of the ^{137}Cs inventory in the forest)



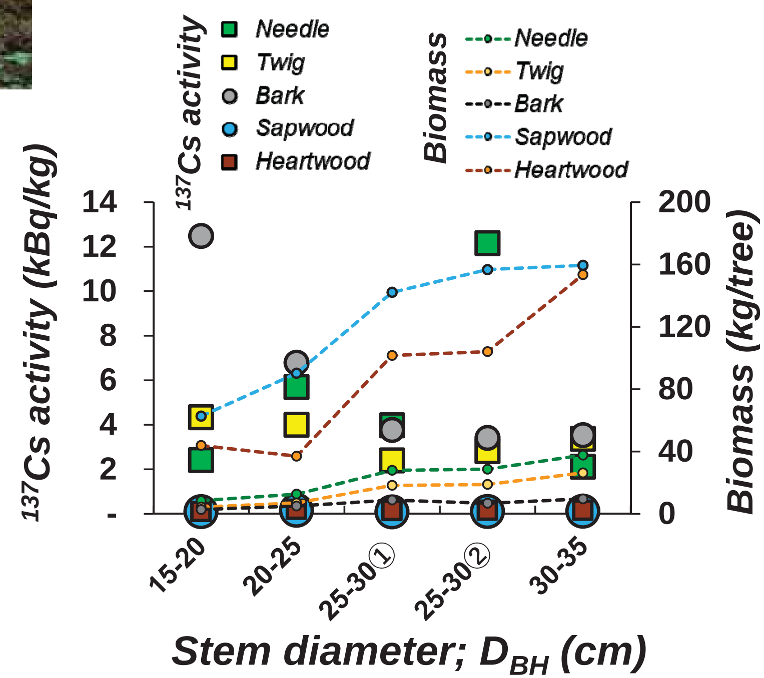
※ Sampling date; October, 2015

^{137}Cs partitioning of aboveground tree system

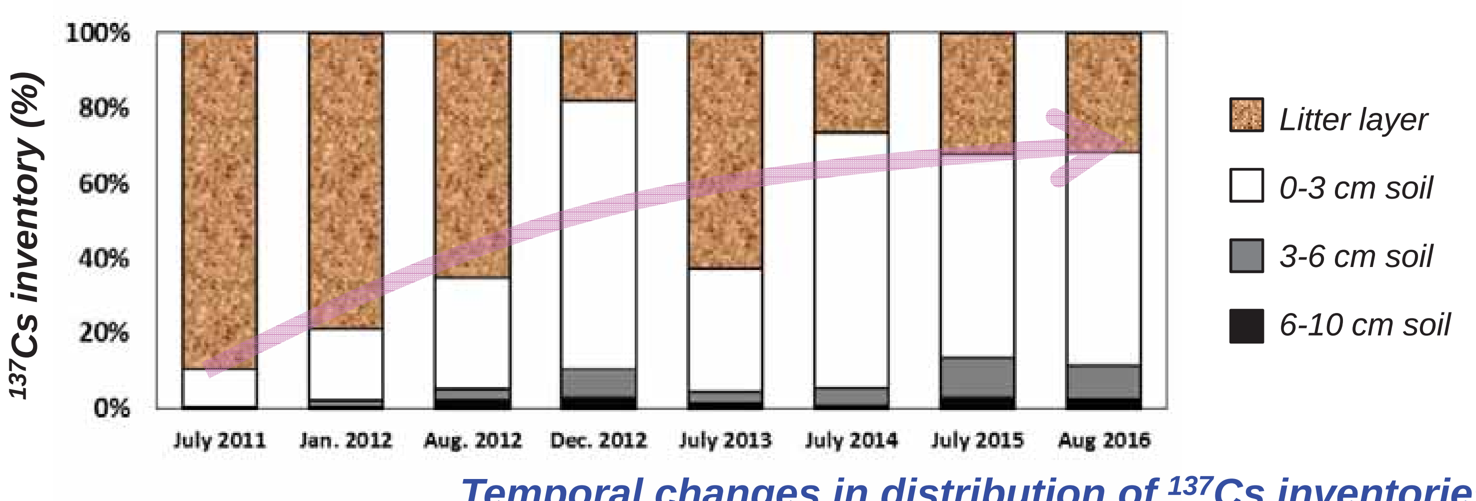
- ^{137}Cs inventory;
  - ✓ **Needle** ≥ Twig ≥ Bark > wood
- ^{137}Cs activity;
  - ✓ **Bark** ≥ Needle ≥ Twig > wood
- ^{137}Cs inventories are increasing with the tree biomass, though needle and bark show large variations in ^{137}Cs activity.



※ Sampling date; October, 2015



Distributions of ^{137}Cs inventories in the litter & soil layers

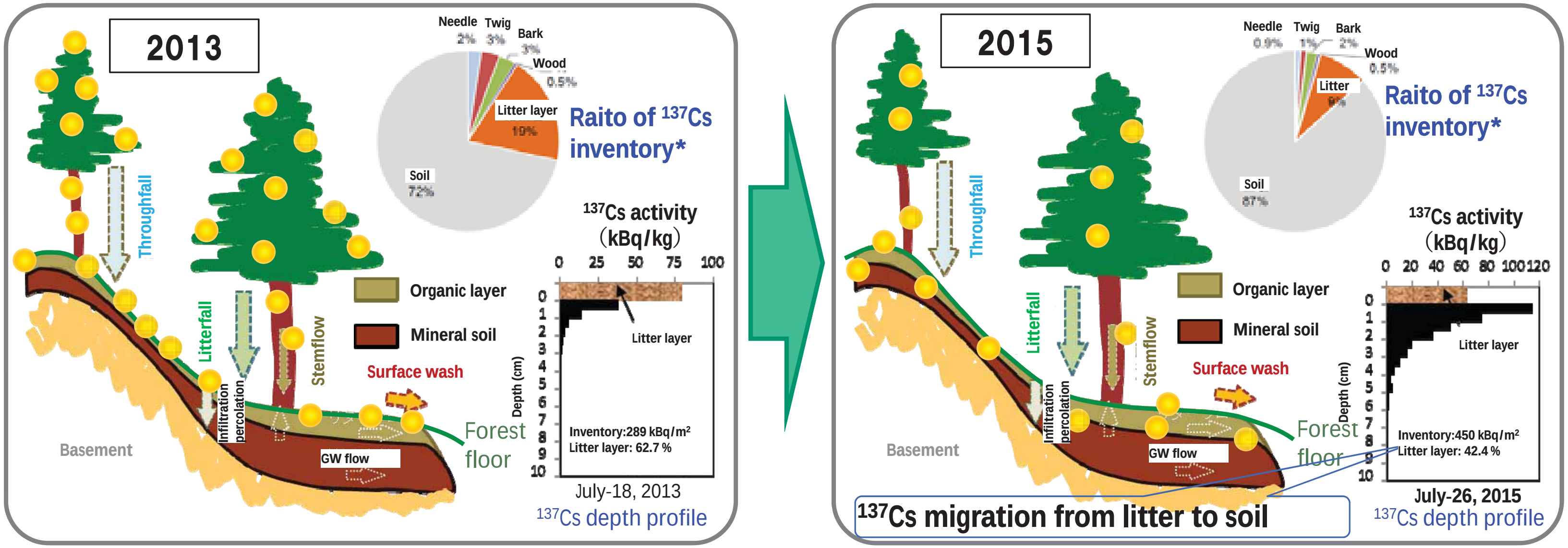


Temporal changes in distribution of ^{137}Cs inventories in litter and soil layers (young cedar forest)

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

- *^{137}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}Cs inventories in the past 5 years.*
- *These indicate that...*
  - ✓ *^{137}Cs transfer from soil surface to the lower layer hardly occur.*
  - ✓ *^{137}Cs remains the litter and soil layers less than 3 cm depth for a long time.*

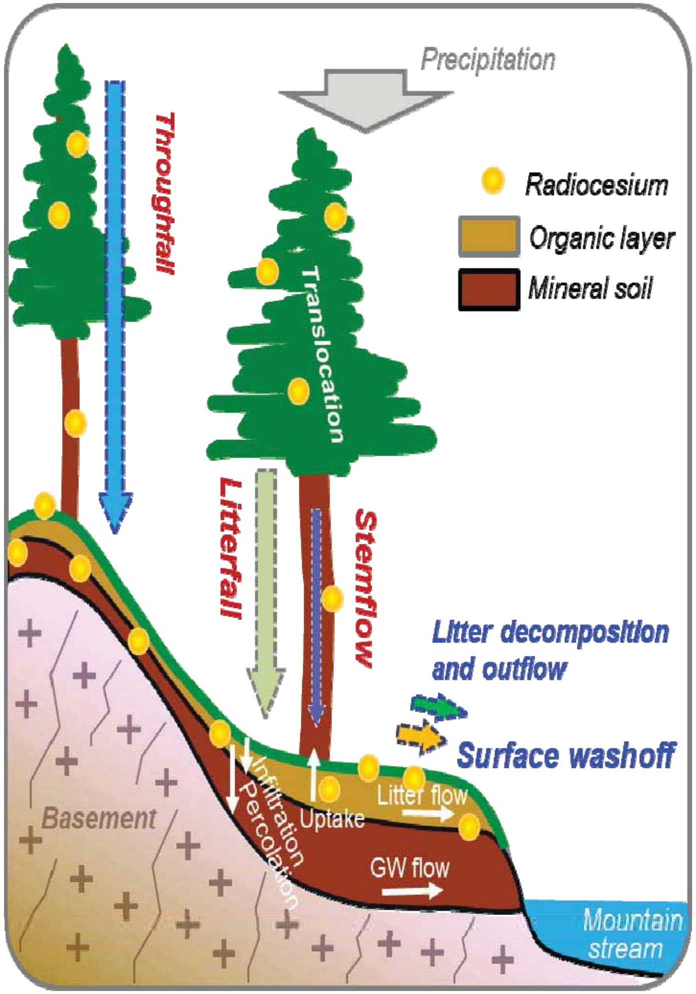
Summary; Temporal and spatial changes of radioCs distribution in the forested ecosystem



*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.

- ***^{137}Cs output** << **^{137}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}Cs inventory in the forest)*
- *^{137}Cs proportion in the litter layer remarkably decrease.*
- *^{137}Cs remains the litter and soil layers less than 3 cm depth for a long time.*

Key issues; Dynamics of radioCs in the forested land



☐ Outflow from the forested ecosystem
→Projection of radioCs outflow

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

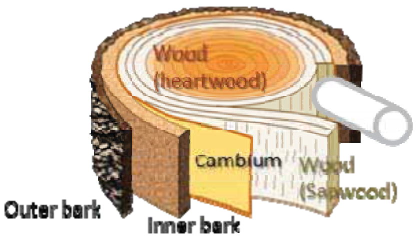
☐ Cycling in the forested ecosystem
→Projection of radioCs concentration in the forest products
→ Projection of radioCs distribution

- ✓ ***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

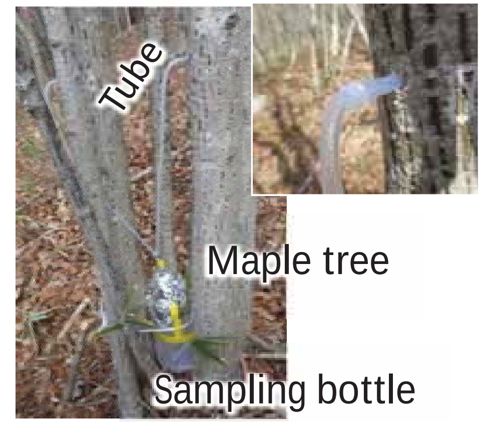


- ***Dissolved radioCs transferred through xylem sap***

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

Radiocesium concentration in xylem sap (Bq/L) (<0.45μm)	
^{134}Cs	5.78±0.32
^{137}Cs	30.6 ±0.10

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



Ge analysis①

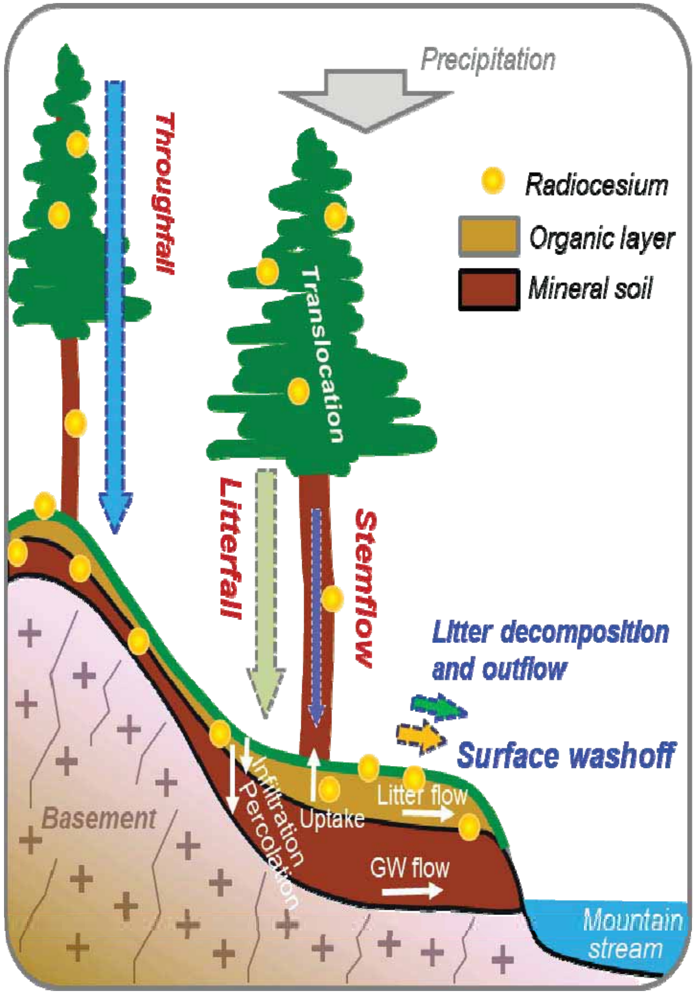
Static culture
65 days 25 ℃
incubation

Ge analysis②

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

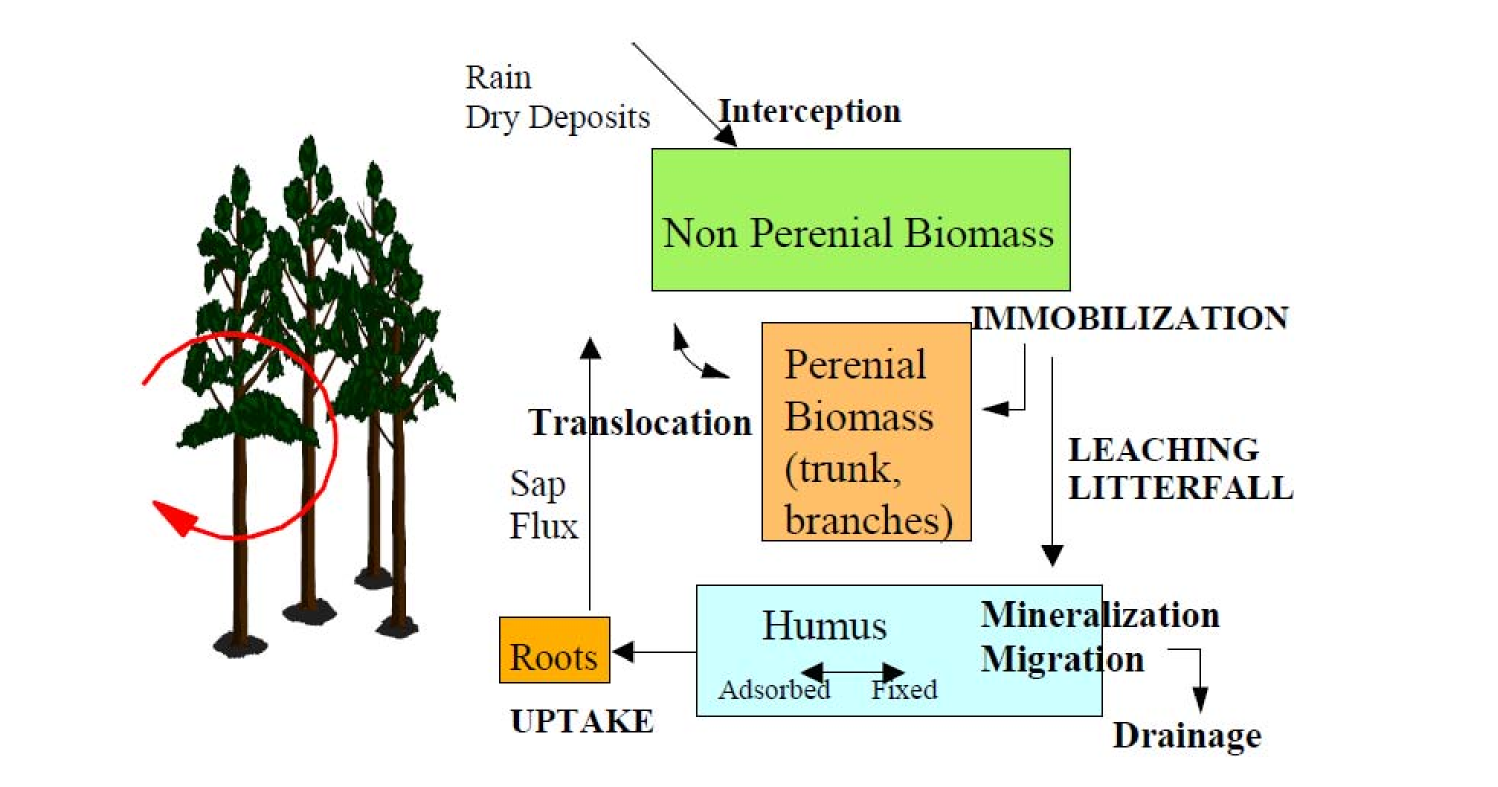
- ✓ *Temporal and spatial changes of radioCs distribution and air dose rate*

Dynamics of radiocaesium in the forest ecosystem

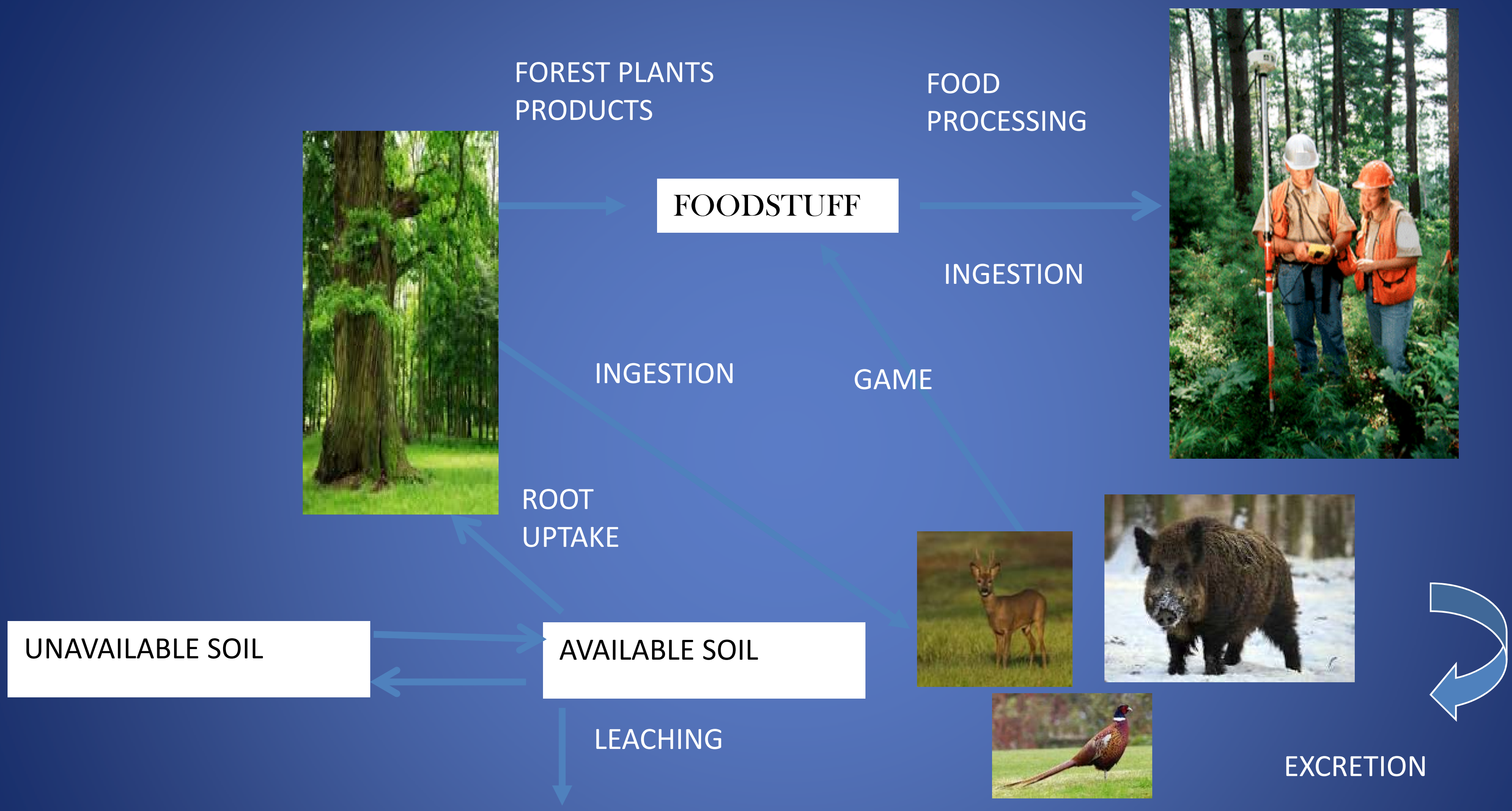
S. Fesenko

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:

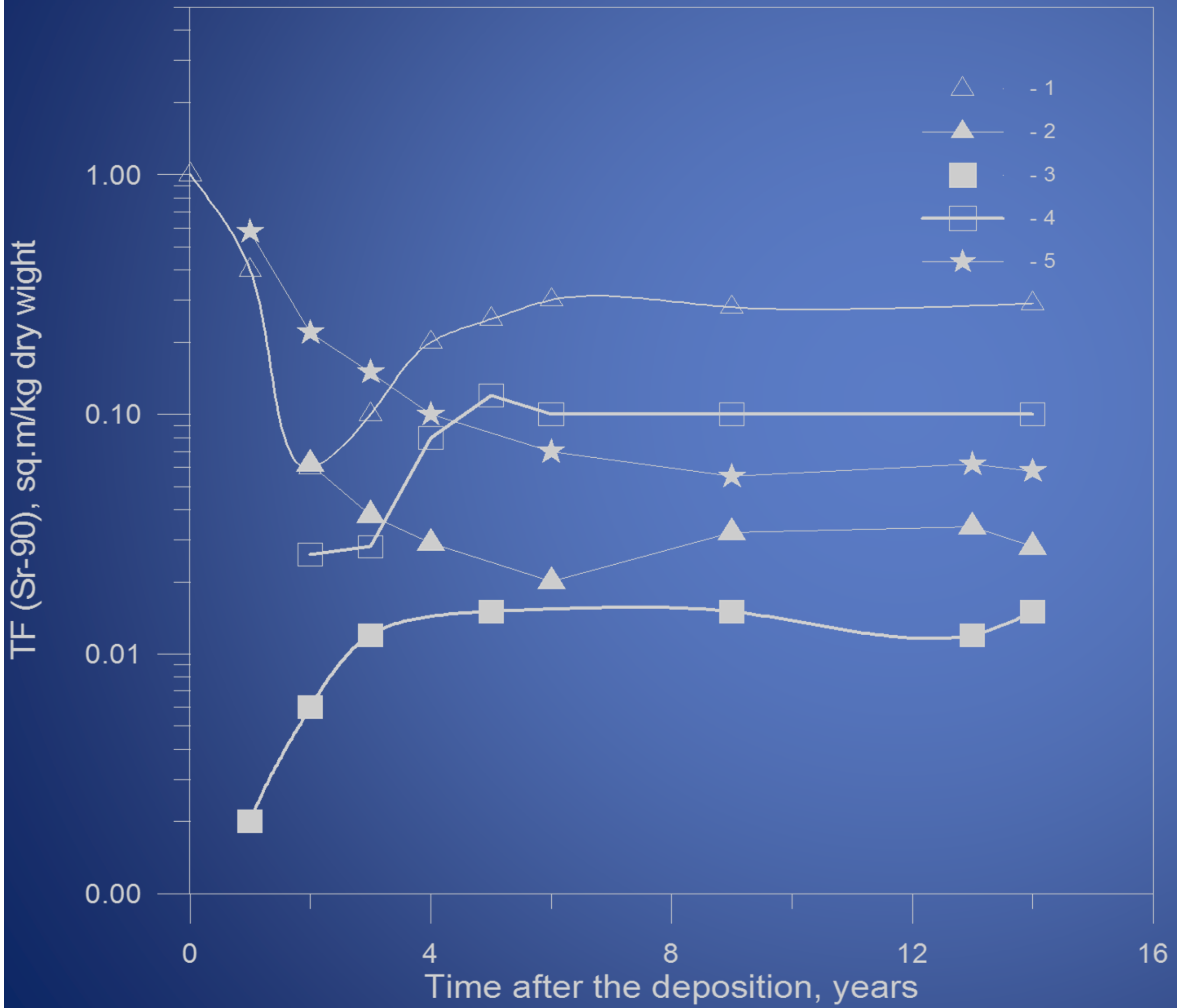
- *High spatial variability of radionuclide levels in many forest components.*
- *The contamination level of a specific forest compartments is being changed due to multiple factors and processes interacting in a very complex way.*

Factors governing variations in ^{137}Cs concentrations in forest plants

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

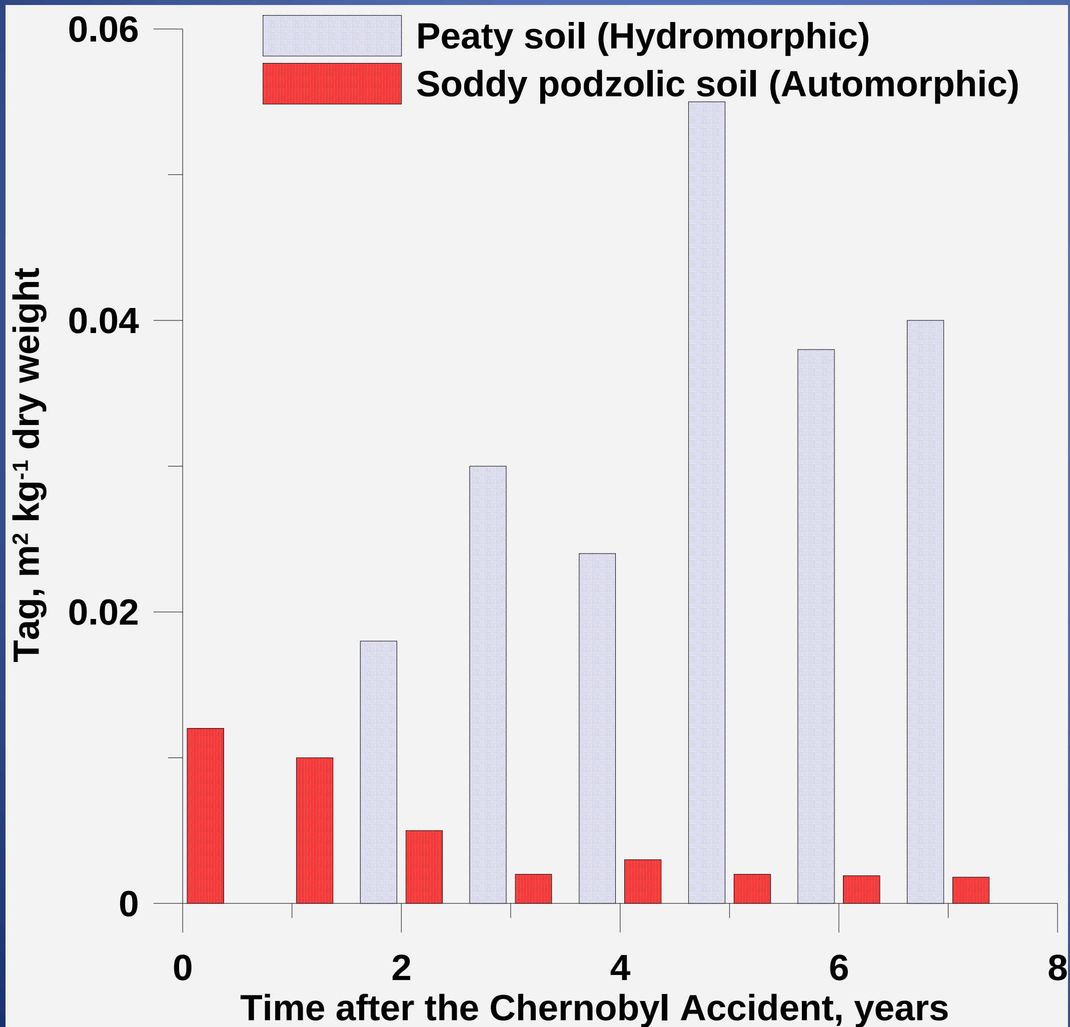
- *Features of forest soil:*
  - *Depth of soil genetic horizons*
  - *Soil/litter properties*
  - *Cs distribution in the top soil*
- *Specific features of plants:*
  - *distribution of fine roots within the top soil;*
  - *ability to accumulate Cs;*
  - *biomass of the plant compartments*

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

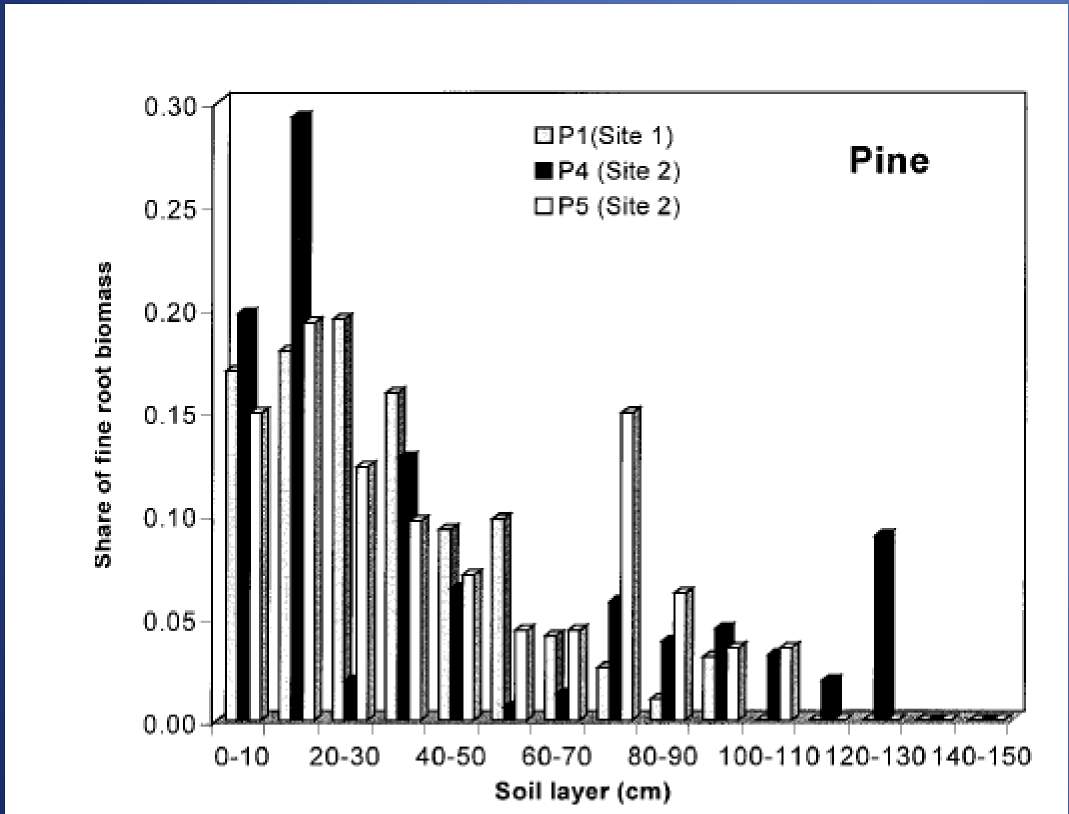


- 1 - leaves
- 2 - branches
- 3 - wood
- 4 – internal bark
- 5 – external bark

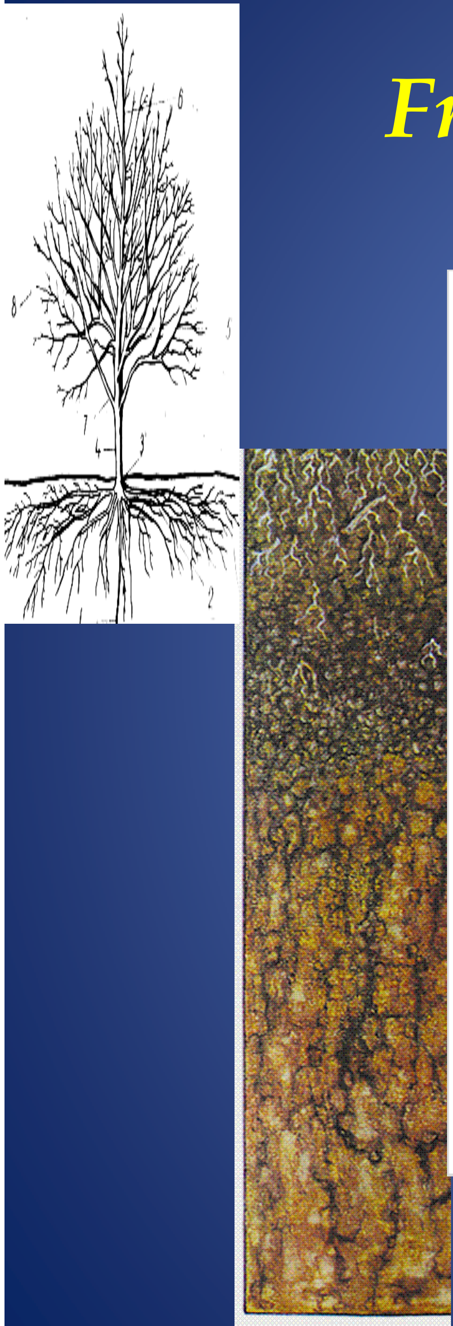
Transfer of ^{137}Cs to pine wood on soils with contrasting properties in areas affected by the Chernobyl accident

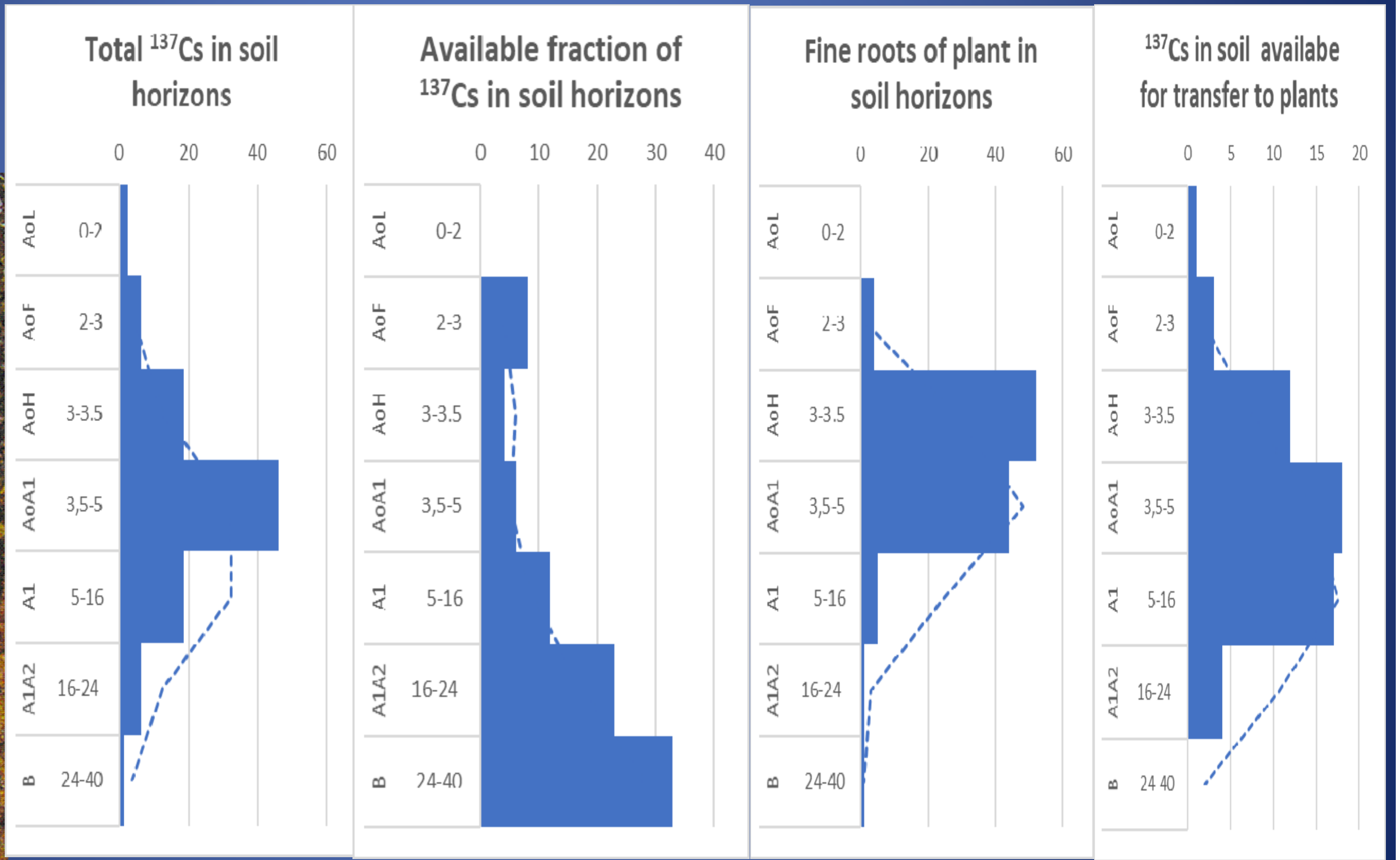


Distribution of fine roots in soil profile

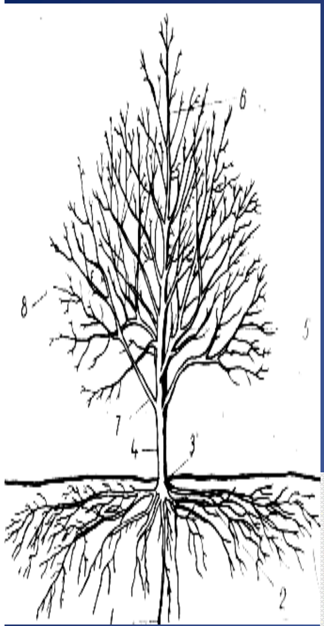


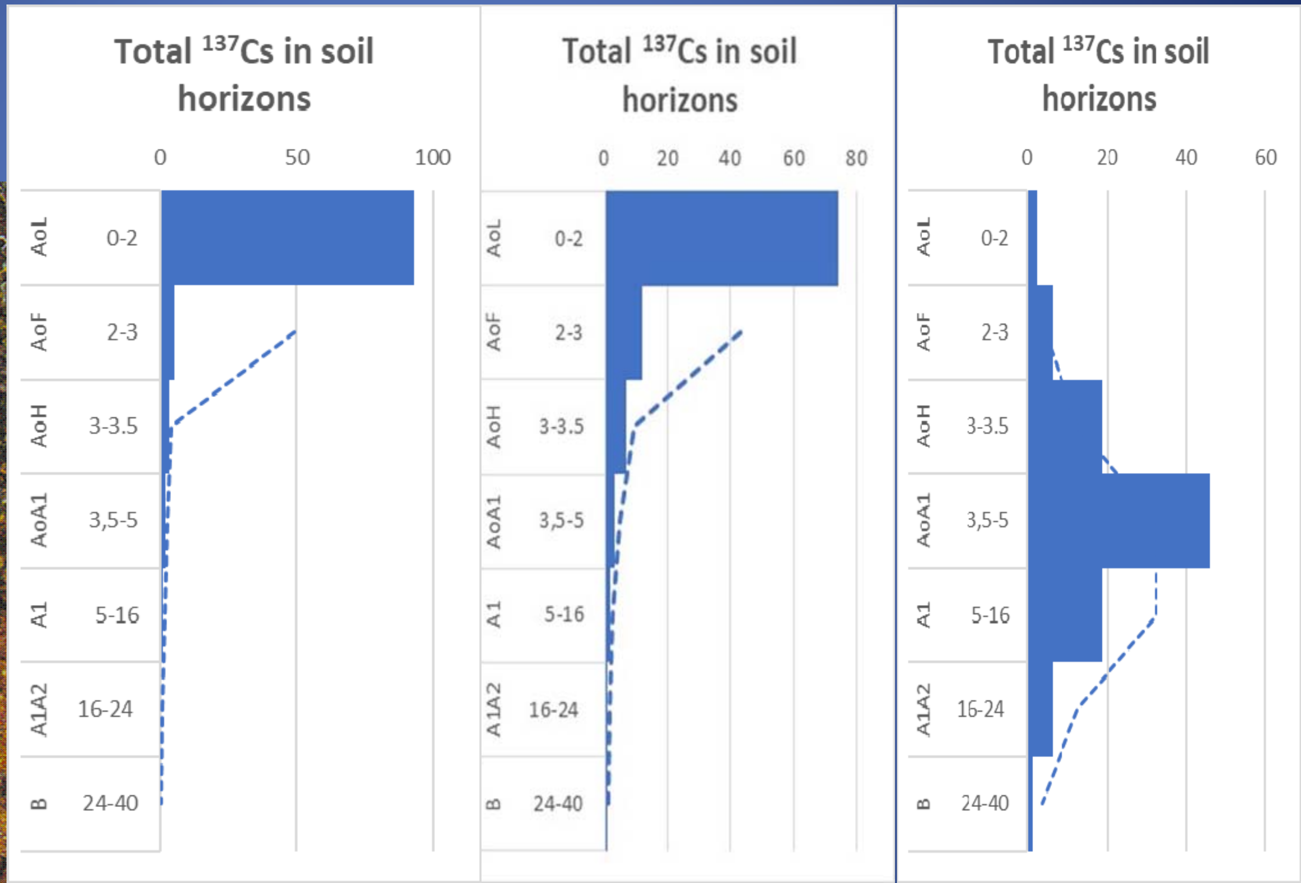
Fraction of Cs available for root uptake





Dynamics of Cs-137 distribution in soil profile





1987 1991 1999

Time dependent amount of ^{137}Cs that is available for transfer to plants

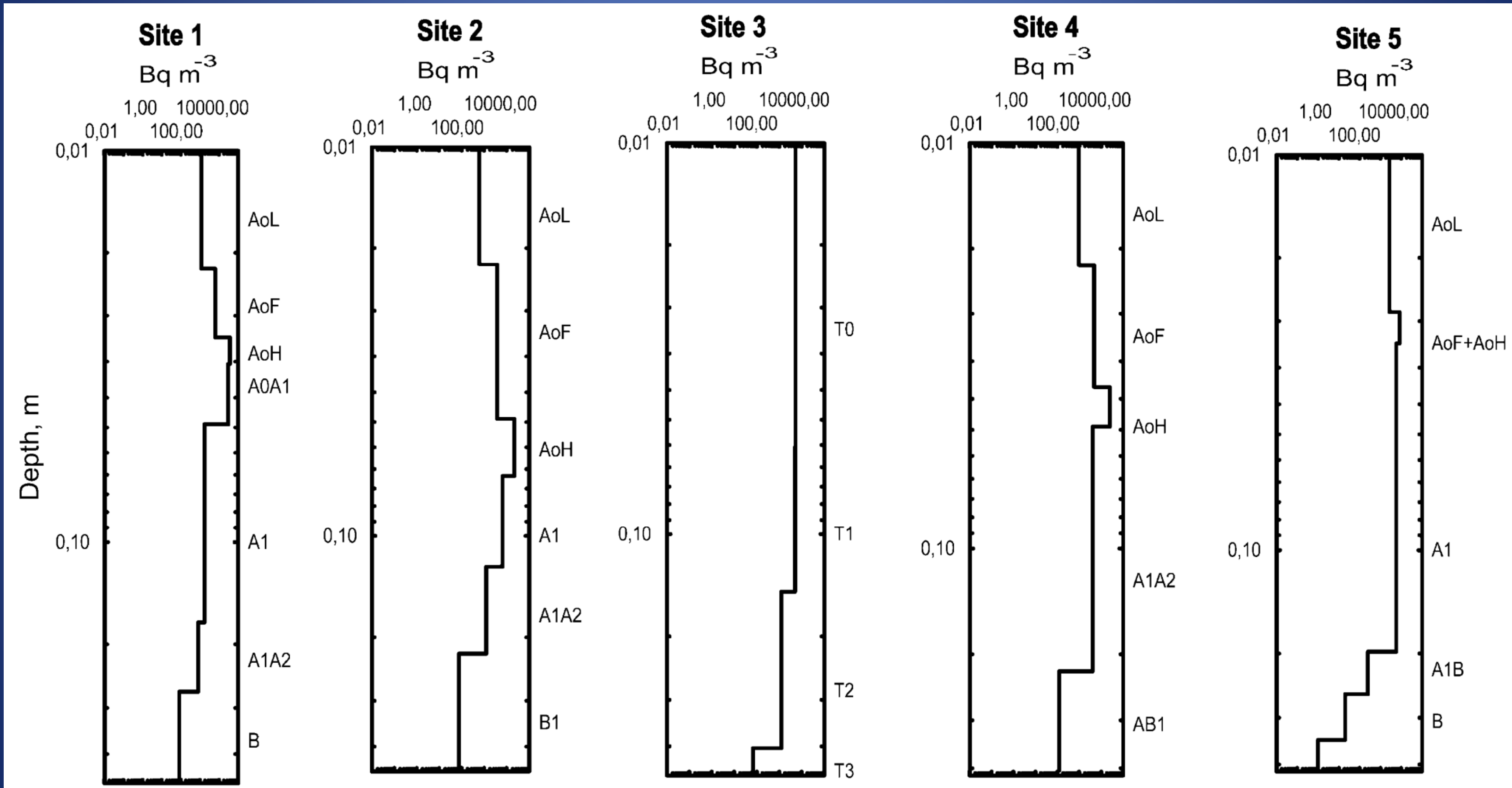
$$BF^{J}(t) = \sum_{i}^{N} \delta_i(t) * q_i(t) * k_i^j$$

- q_i *is the fraction of the total* ^{137}Cs *activity in the soil horizon i;*
- $\delta(t)$ *is the fraction of exchangeable (or available)* ^{137}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 ^{137}Cs extracted by reagents (%)				
			H_2O	$AcNH_4$	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}Cs in soil profile

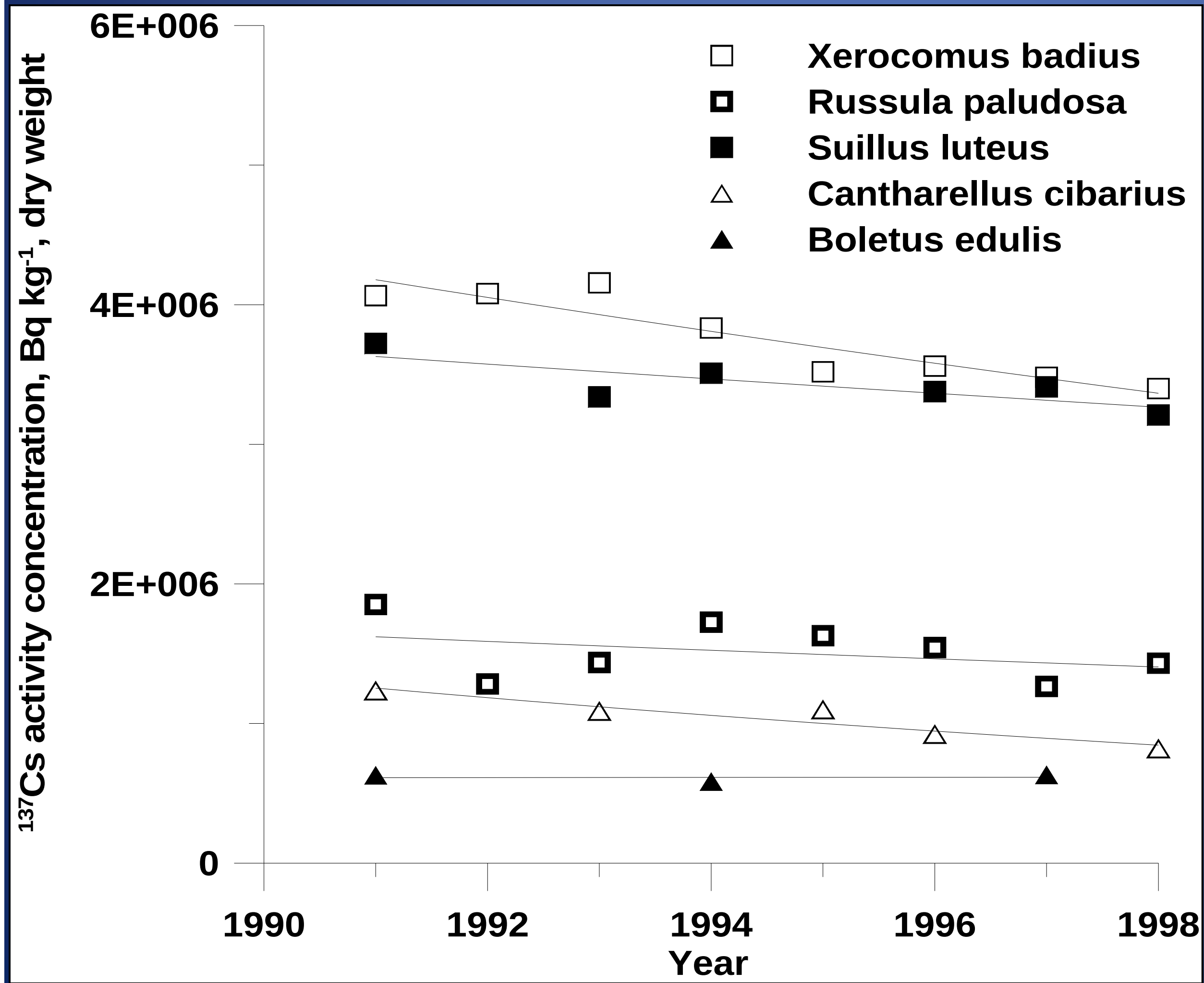


Environmental factors governing the extent of tree contamination by radiocaesium

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*

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

Dynamics of ^{137}Cs activity concentrations in selected mushroom species.

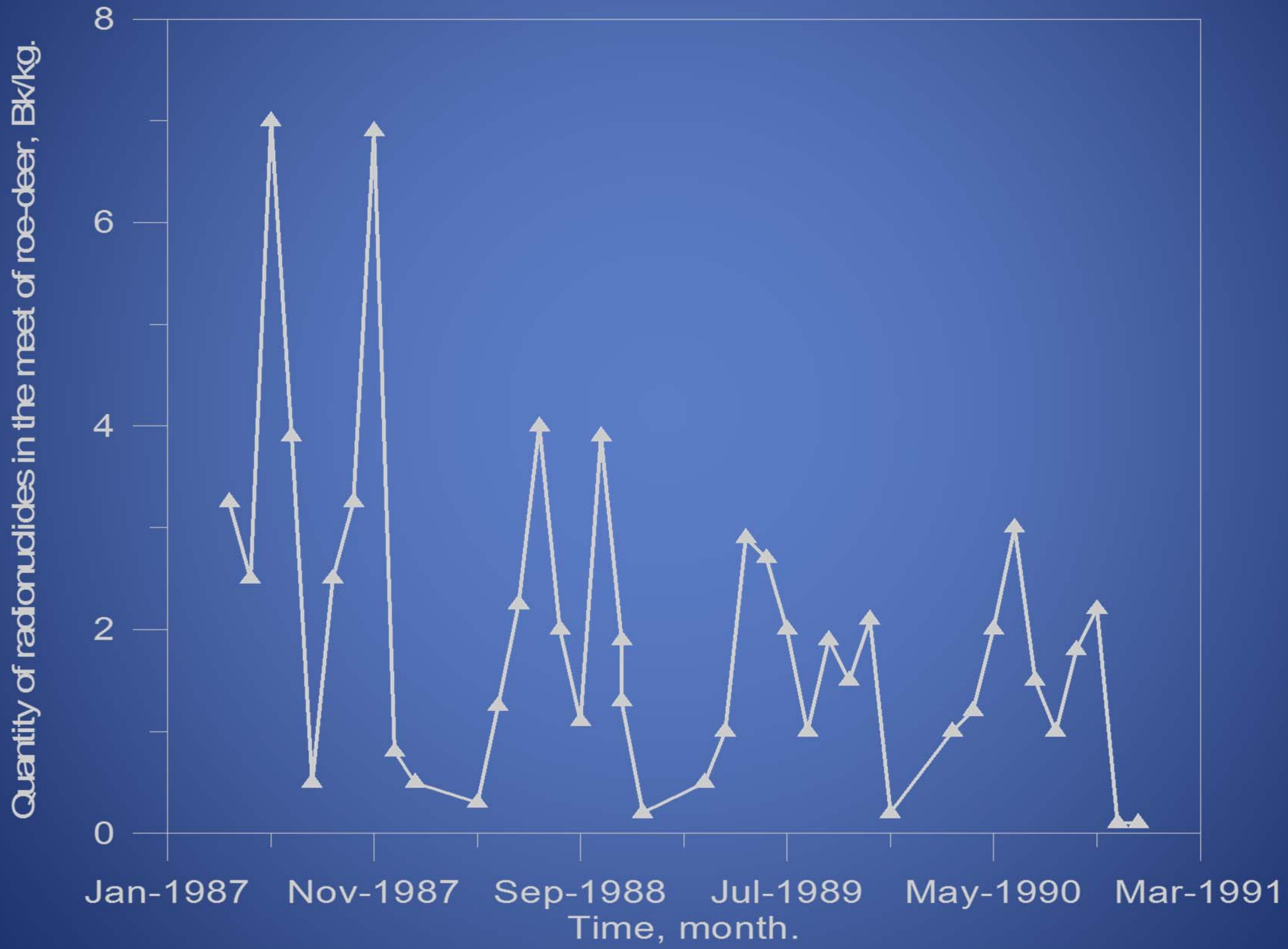


Cs-137 soil deposition at the site in 1986 - 555 kBq m^{-2}

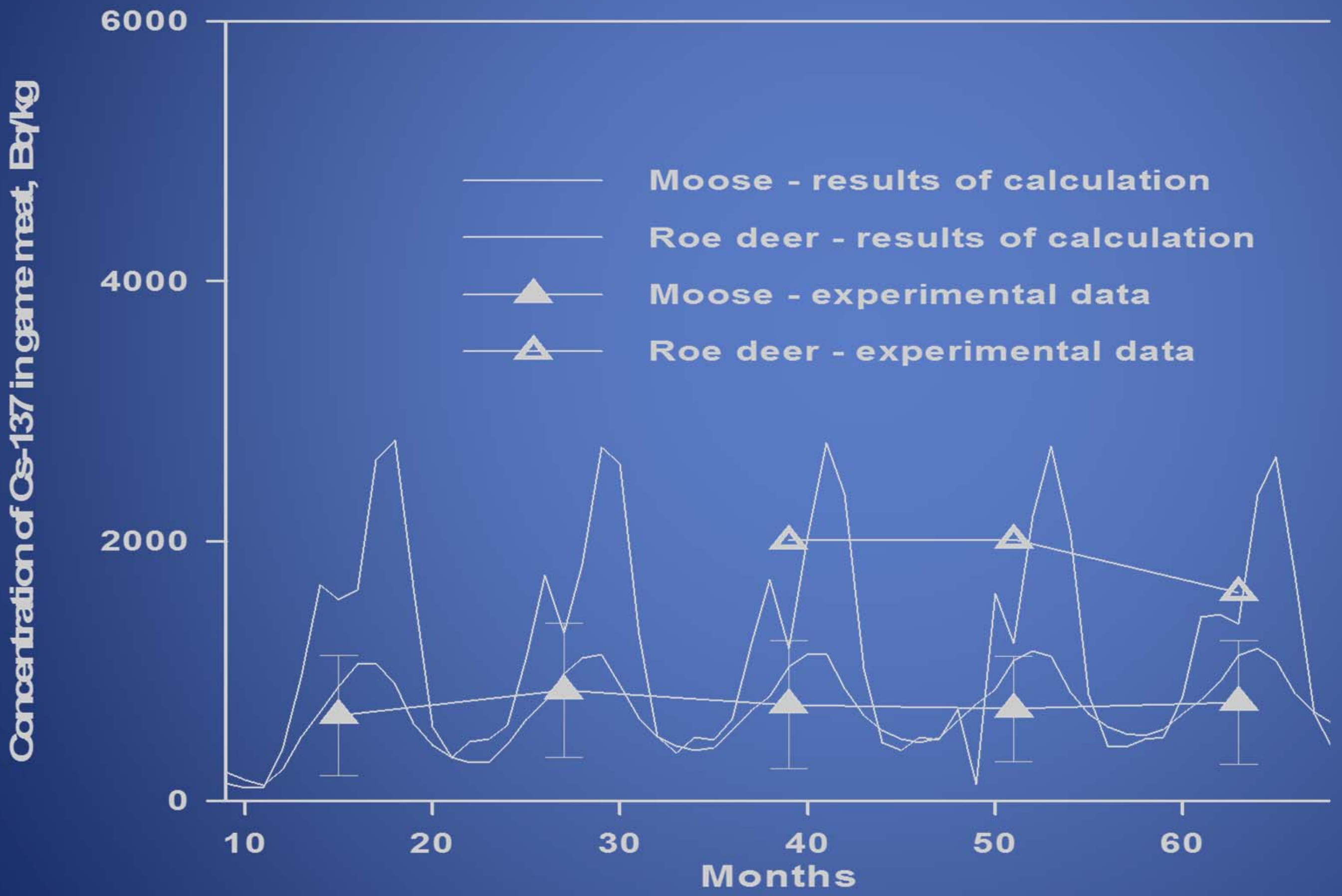
Factors governing variations in ^{137}Cs concentrations in wild animals

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

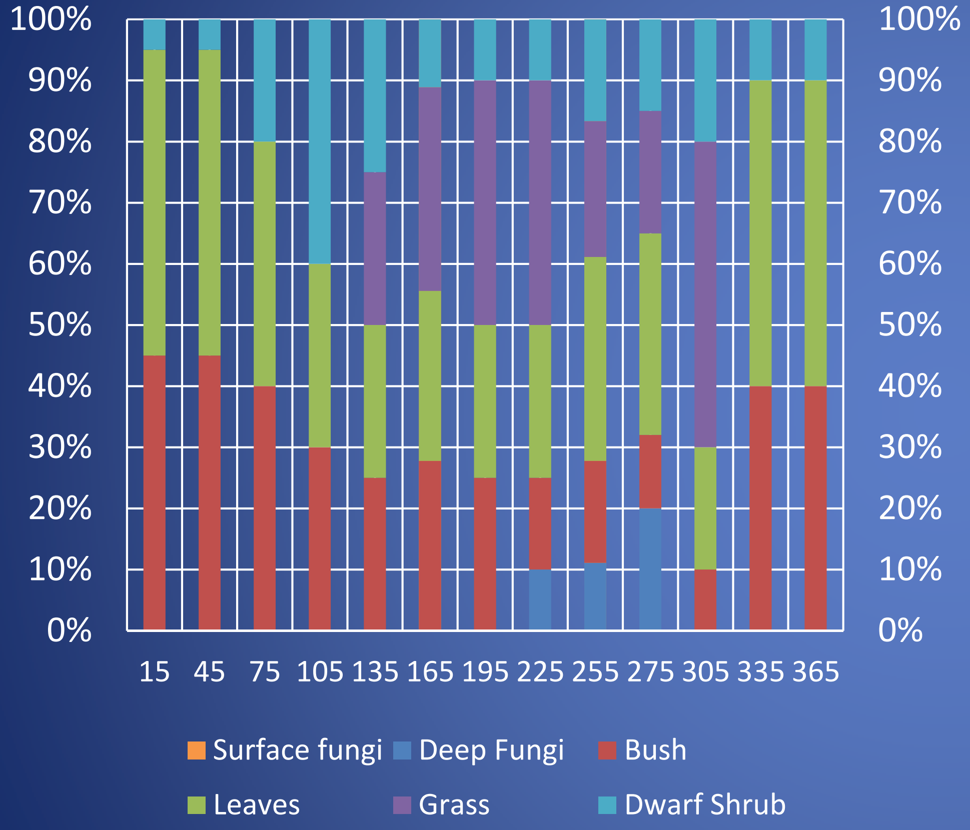
Dynamics of ^{137}Cs content in meat of roe deer in Southern Germany



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

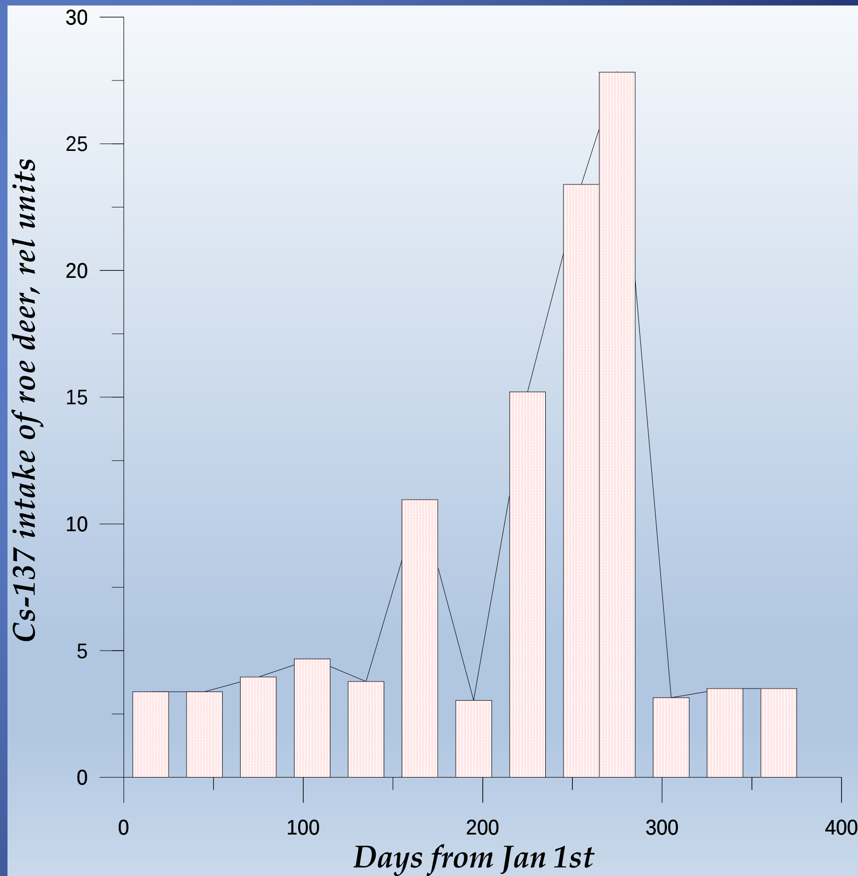


Intake by roe deer: seasonal variations



Fractions of individual feeds

Intake with feeds



Half lives of ^{137}Cs in wild animal

Animal	*Ecological half life, year*	*Biological half life. d*
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}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

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}Cs by forest trees following the Chernobyl accident

ELSEVIER
The Science of the Total Environment 269 (2001) 87–103
the Science of the Total Environment
www.elsevier.com/locate/scitotenv

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

S.V. Fesenko[a,*], N.V. Soukhova[a,c], N.I. Sanzharova[a], R. Avila[b],
S.I. Spiridonov[a], D. Klein[c], P.-M. Badot[c]
...on, 249020,
...du Pays de

MODELLING OF ^{137}Cs BEHAVIOUR IN FOREST GAME FOOD CHAINS

S. FESENKO, S. SPIRIDONOV
Russian Institute of Agricultural Radiology and Agroecology (RIARAE), 249020, Obninsk, Russia
R. AVILA

ELSEVIER
Journal of Environmental Radioactivity 65 (2003) 19–28
JOURNAL OF ENVIRONMENTAL RADIOACTIVITY
www.elsevier.com/locate/jenvrad

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

Radiat Environ Biophys (2000) 39:291–300

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

Available online at www.sciencedirect.com
SCIENCE DIRECT
ELSEVIER
Journal of Environmental Radioactivity 82 (2005) 143–166
JOURNAL OF ENVIRONMENTAL RADIOACTIVITY
www.elsevier.com/locate/jenvrad

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

S.V. Fesenko[a,b,*], G. Voigt[a], S.I. Spiridonov[a], I.A. Gontarenko[a]
[a] *International Atomic Energy Agency, Agency's Laboratories Seibersdorf, 1400 Vienna, Austria*
[b] *Russian Institute of Agricultural Radiology and Radioecology, 249020 Obninsk, 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)

Contents

- **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**

Introduction

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

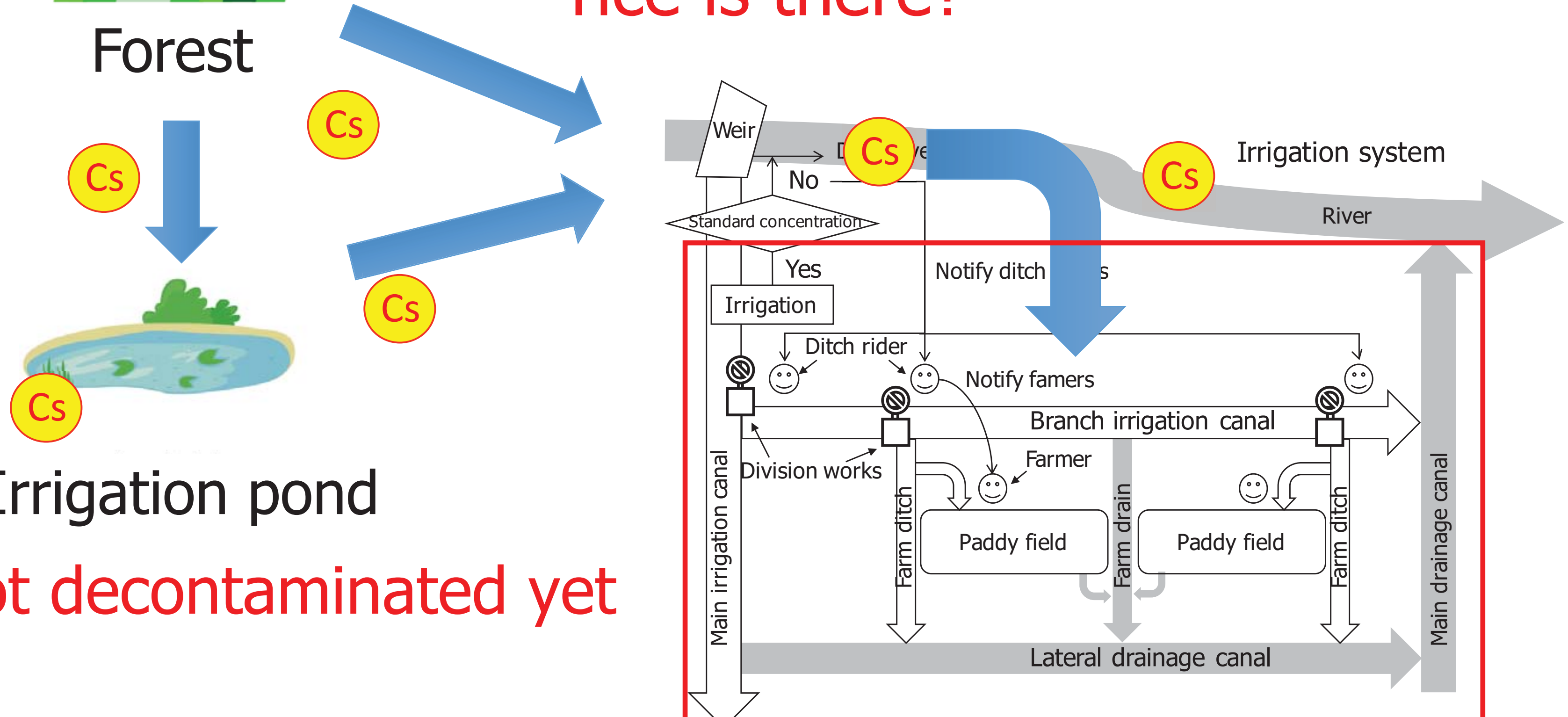
Introduction

Background of research

(1) Is it contaminated again?

(2) How much the impact on brown rice is there?

Forest



Not decontaminated yet

Decontamination

Introduction

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

Introduction

NARO

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: ^{131}I)	Drinking water	300
	Milk, Dairy products 1)	
	Vegetables (Except root vegetables and tubers)	2,000
	Fishery products	
Radioactive cesium 2) (^{134}Cs plus ^{137}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.

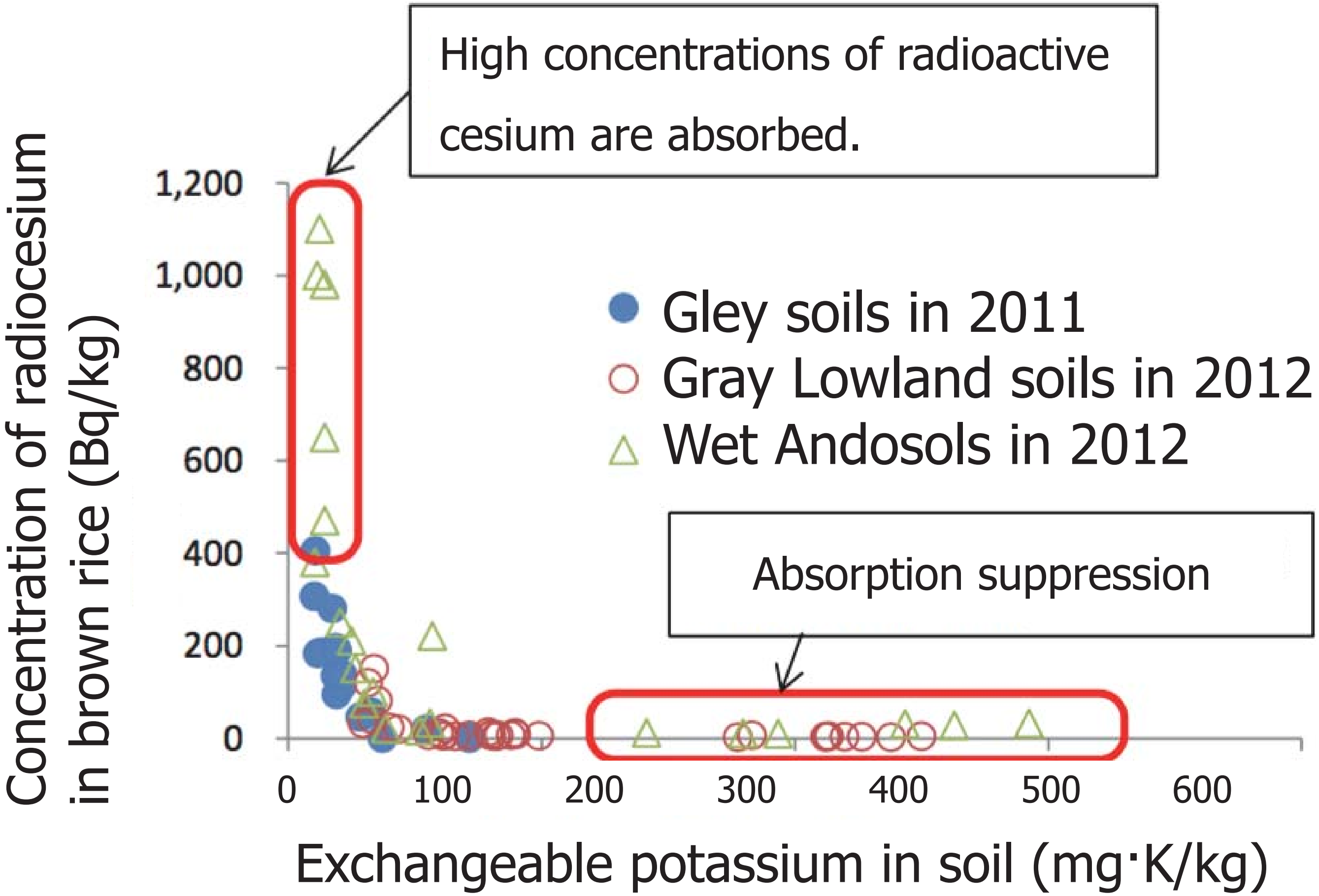
Operational intervention level : < **1 mSv/ year**

MHLW: http://www.mhlw.go.jp/shinsai_jouhou/dl/shokuhin.pdf
http://www.mhlw.go.jp/english/topics/2011eq/dl/new_standard.pdf

Standard limits for radiocesium in Japan

Introduction

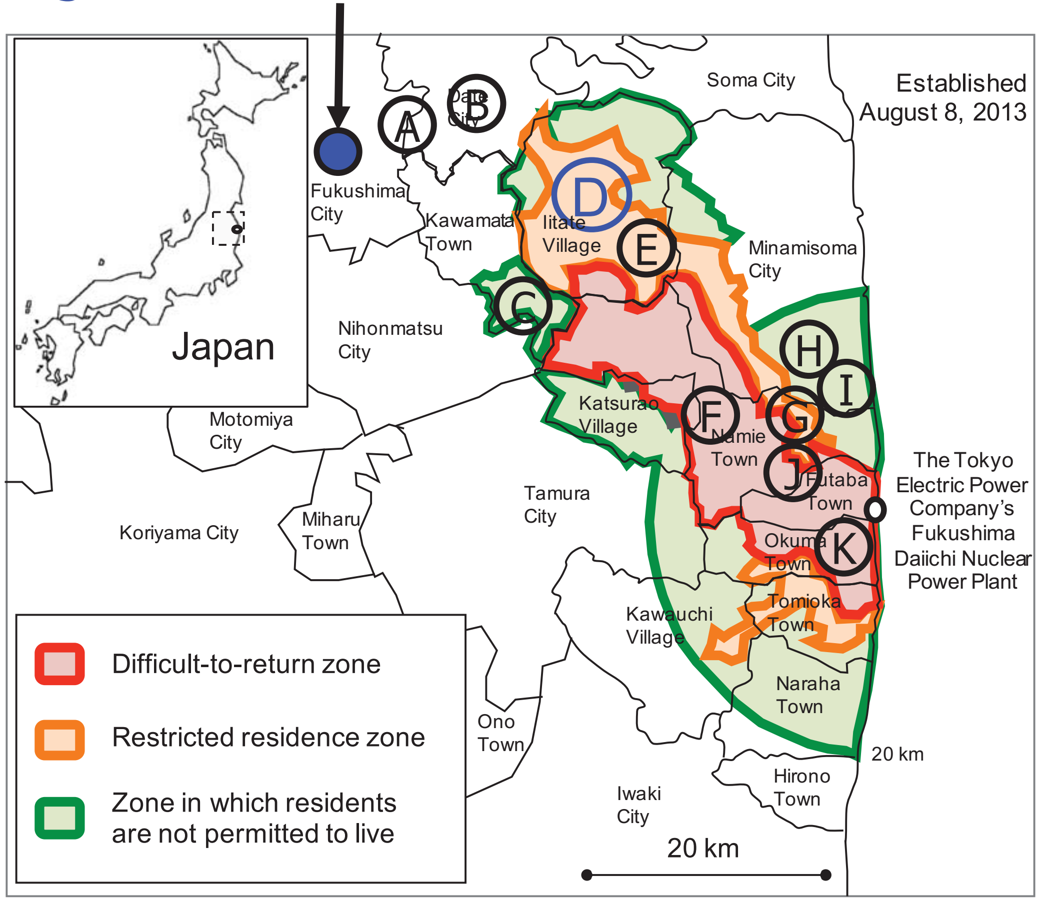
Relationship between radiocesium in rice and exchangeable potassium in soil



Materials and methods

Study area and water sampling sites

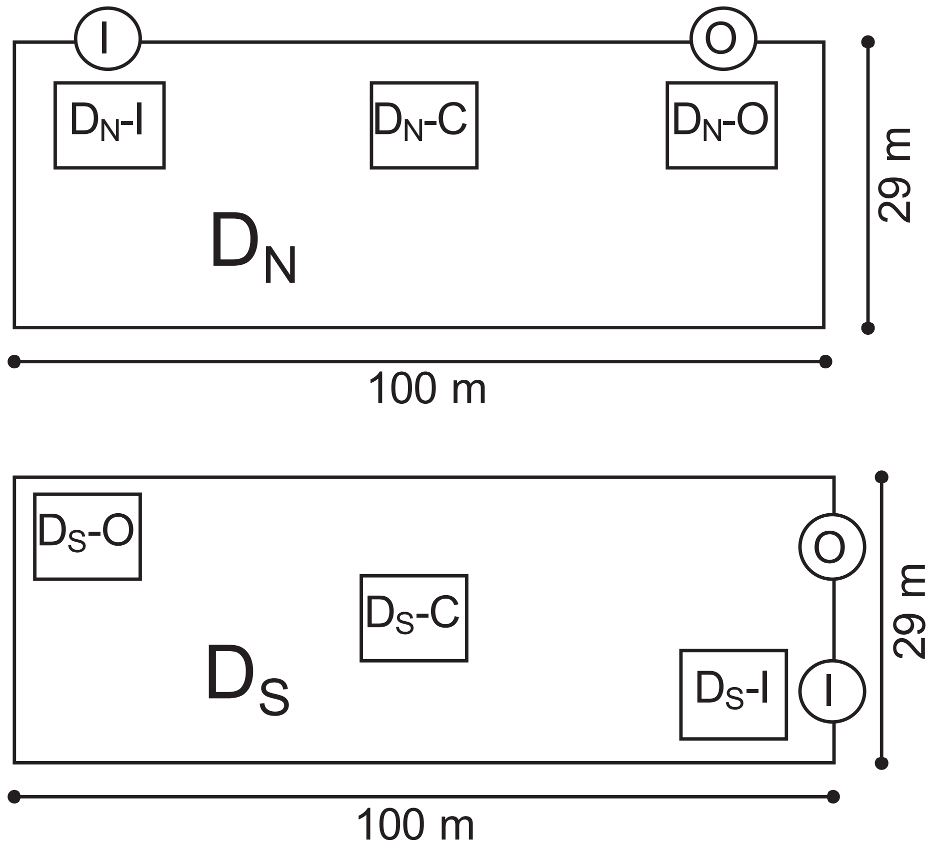
Agricultural Radiation Research Center, NARO/TARC



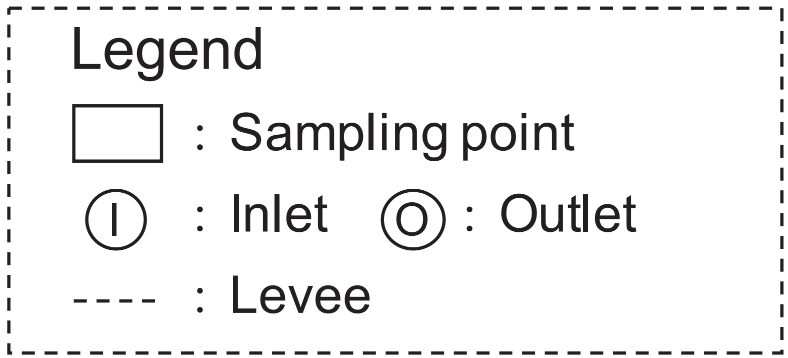
- 40 km northwest of FDNPP
- Decontamination of topsoil removal
- Cultivation trials in two paddy fields
- potassium level to 274 (mg·K)/kg soil

Materials and methods

Survey methods



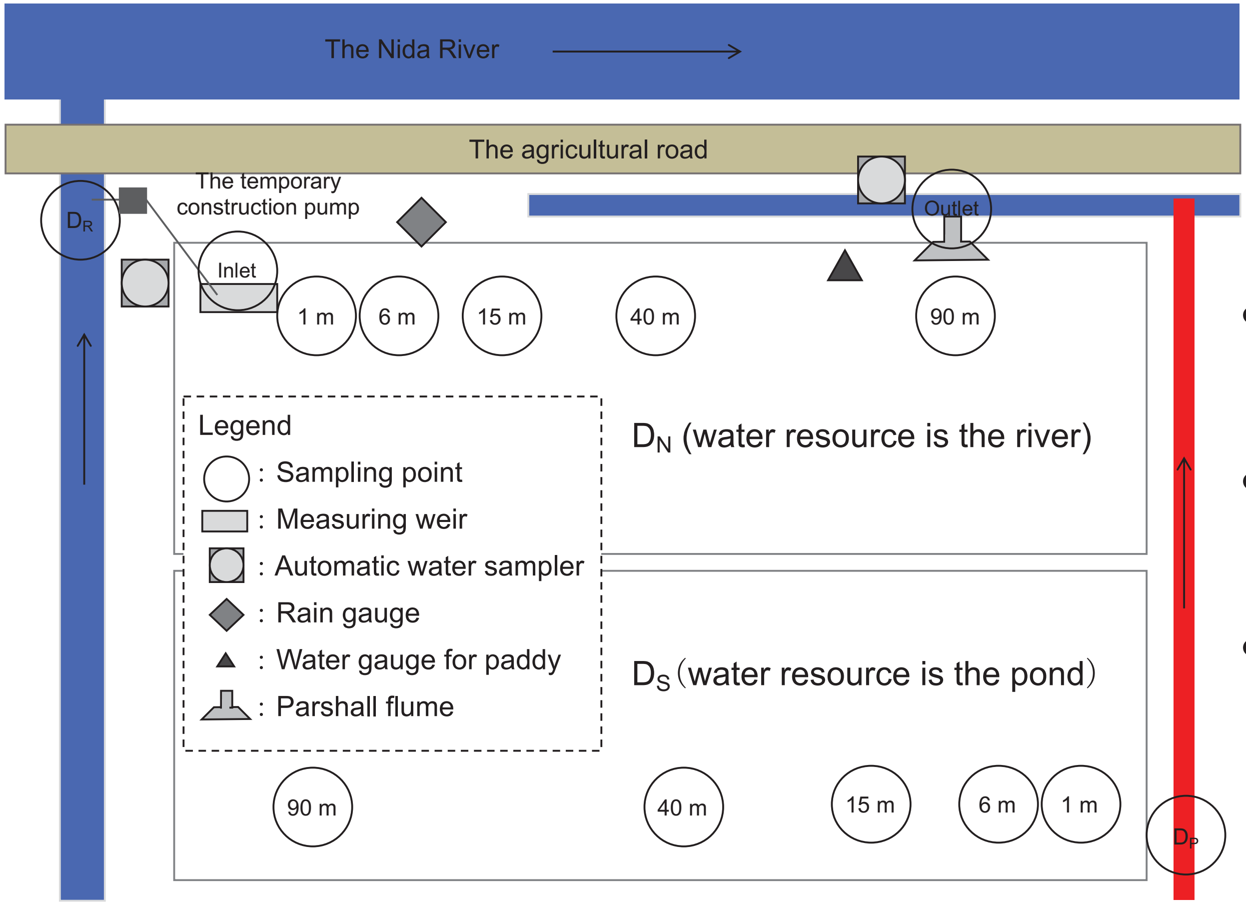
- Irrigation water
- Soil
- Brown rice



Soil and brown rice quadrat sampling sites

Materials and methods

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

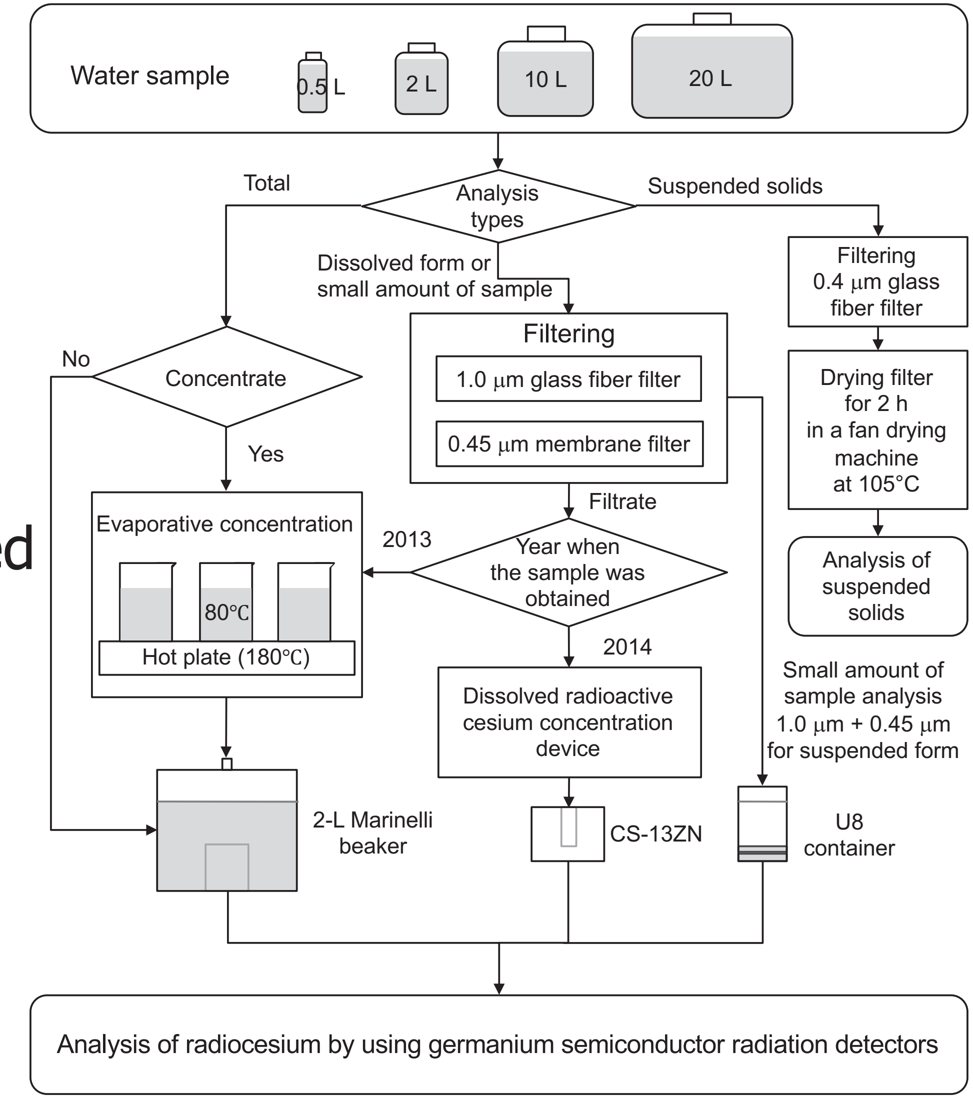
Rain gauge

Water sampling points and irrigation equipment at site D

Materials and methods

Radiocesium analyses

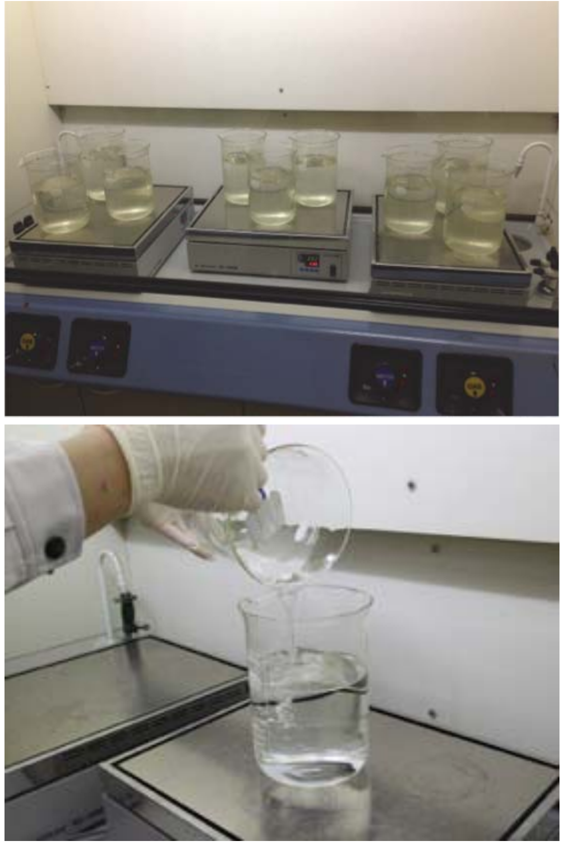
- Analysis water sample
  - Total radiocesium
  - Dissolved radiocesium
- Pre-concentration
  - Evaporative concentration
  - Nonwoven fabric impregnated with Prussian blue
  - Raddisk
- Analyses ^{134}Cs and ^{137}Cs
  - GC4020-7500SL, Canberra
- Analyses of suspended solids and exchangeable potassium



Procedures for laboratory analysis

Materials and methods

Radiocesium analyses



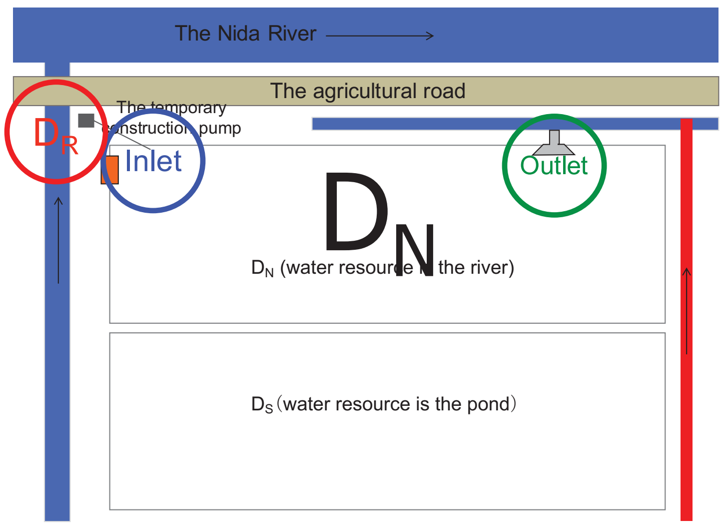
Evaporative concentration

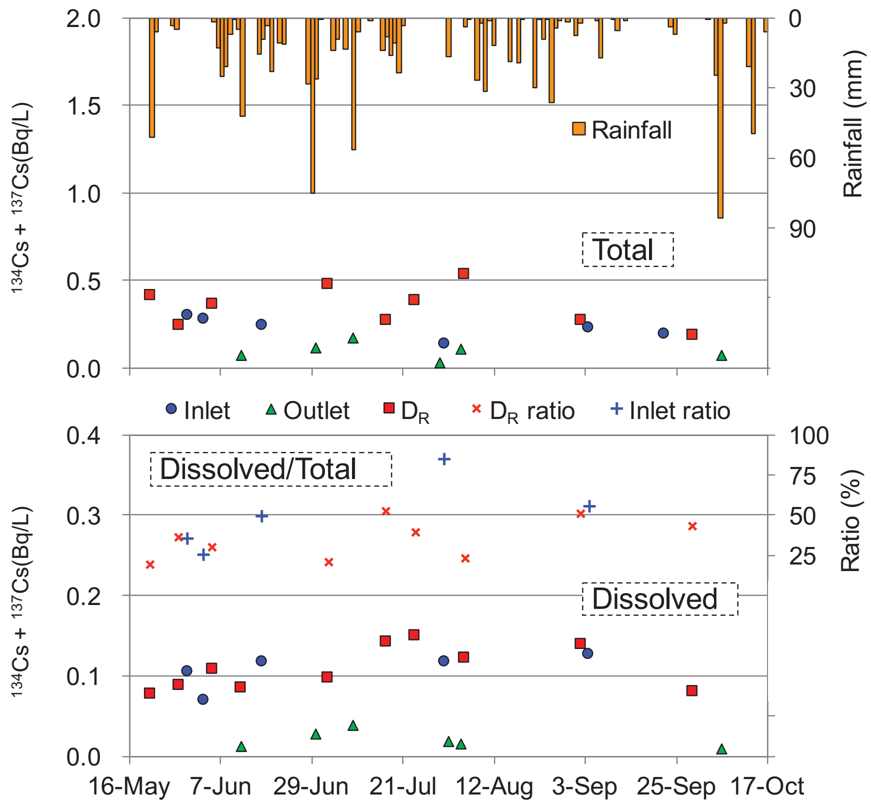
Nonwoven fabric impregnated
With Prussian blue

Raddisk

Results and discussion

Temporal changes in the radiocesium concentration in irrigation water

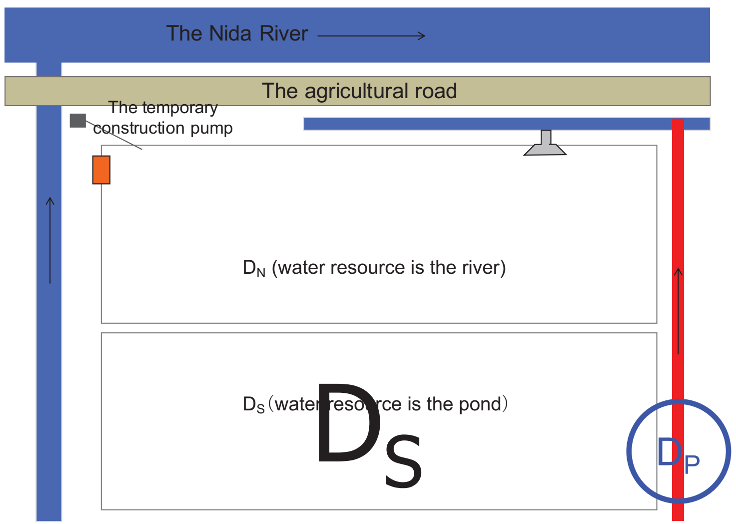


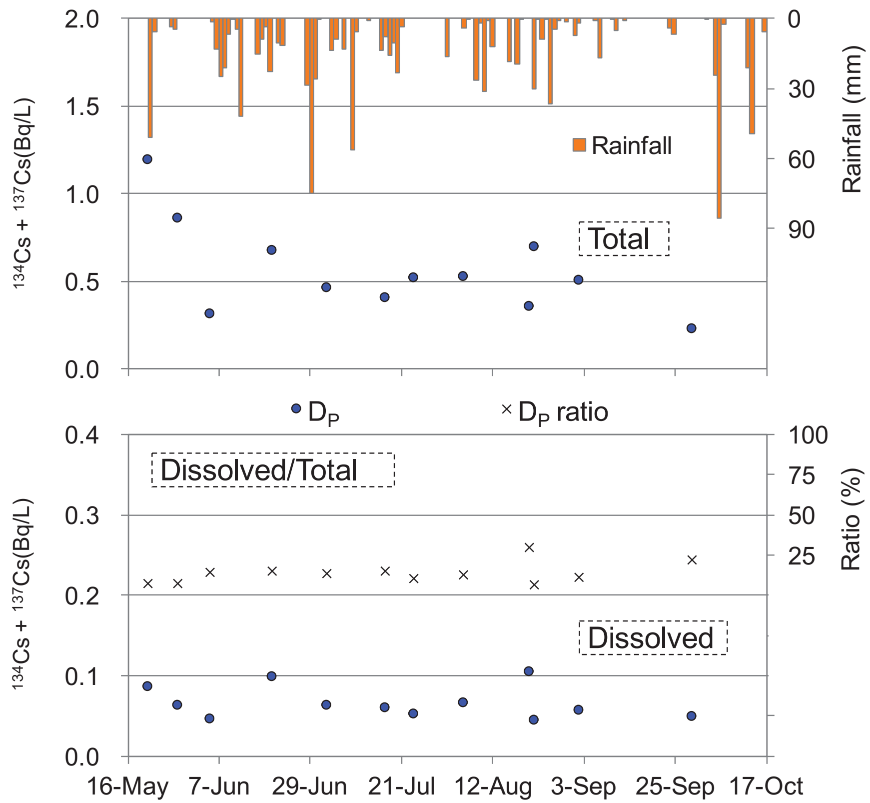


Concentrations of radiocesium in D_R (the irrigation water from the river at site D), inlet and outlet at site D_N (the northern part of site D) during the irrigation period in 2014

Results and discussion

Temporal changes in the radiocesium concentration in irrigation water

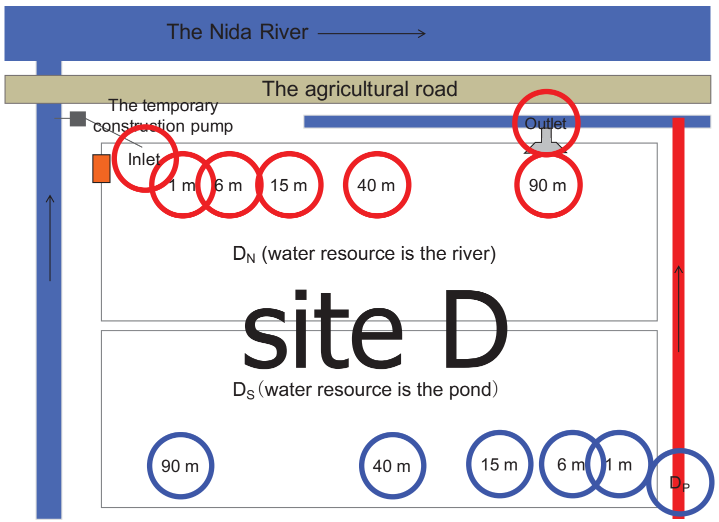


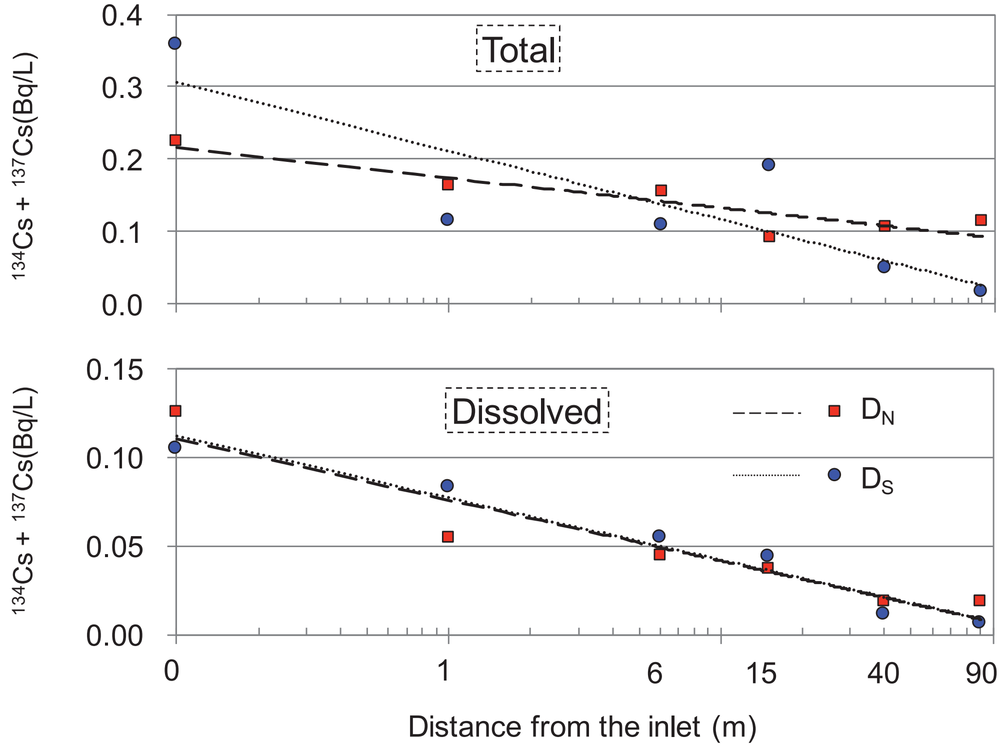


Concentrations of radiocesium in D_P (the irrigation water from the pond at site D) at site D_S (the southern part of site D) during the irrigation period in 2014

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 (m²)	**2900**	**2900**
(1) Average (Bq/L) Total (Dissolved)	(a) 0.288 (0.108)	0.562 (0.064)
(2) Inflow [Bq/(m2·year)] Total (Dissolved)	(b) 95.3 (37.2)	(b) ÷ (a) × (1) 186.0 (22.0)
(3) Outflow [Bq/(m2·year)] Total (Dissolved)	(c) 14.7 (3.2)	(c) ÷ (b) × (2) 28.7 (1.9)
Balance [Bq/(m2·year)] (2)-(3) Total (Dissolved)	80.6 (34.0)	157.3 (20.1)

Estimated volume of radiocesium balance in irrigation water in 2014

Conclusions

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

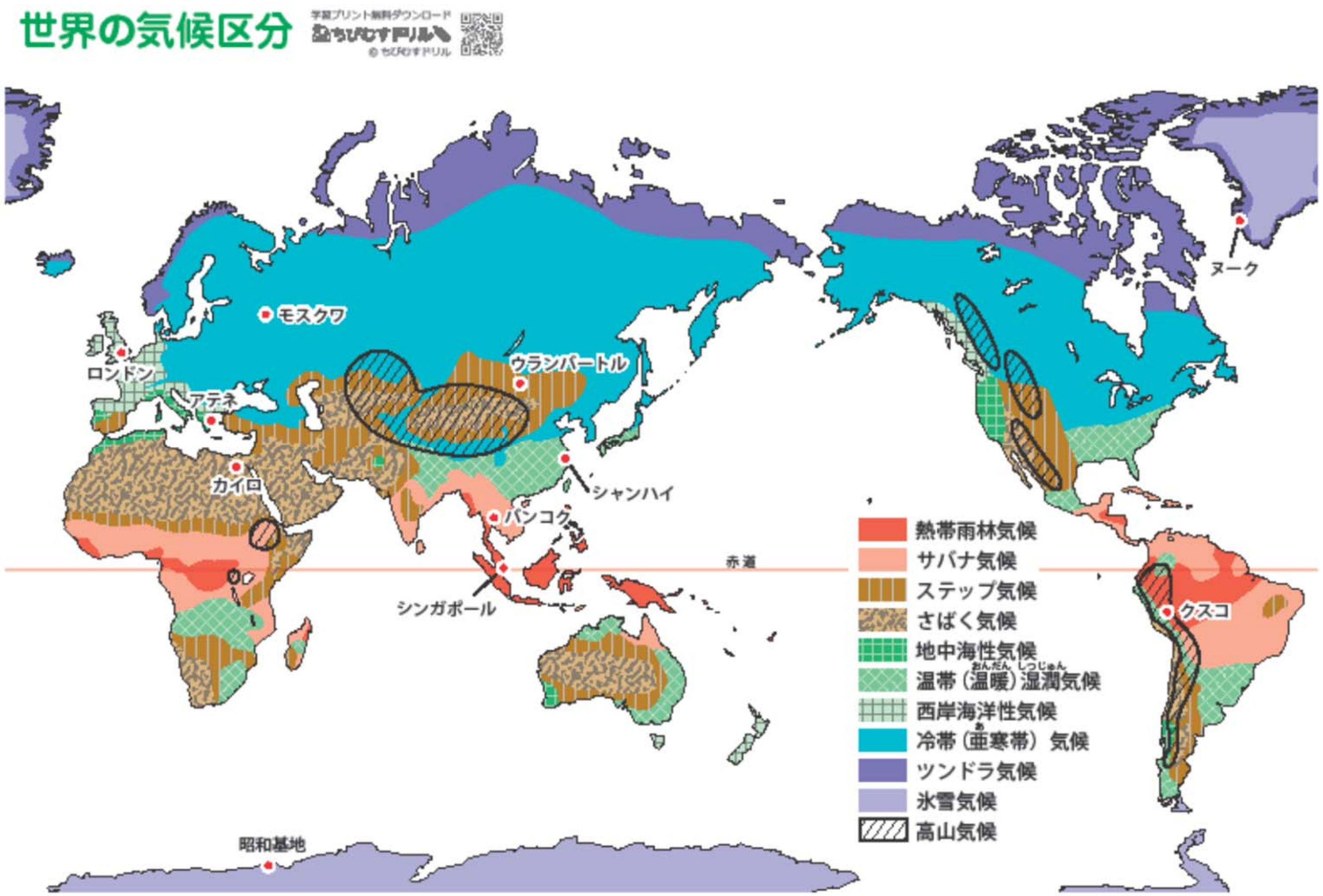
- Most of the radiocesium in the irrigation water flowing into the paddy fields accumulated near the inlet.
- The total and dissolved amount of radiocesium in the irrigation water flowing into the paddy fields were generally <200 Bq/m^2/year and <40 Bq/m^2/year, respectively.
- The impact on brown rice 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

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.

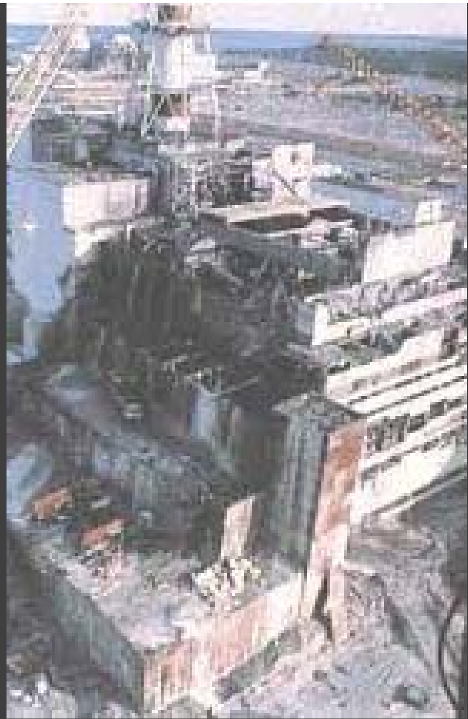
MAFF
Ministry of Agriculture, Forestry and Fisheries

Do we now know enough about radiocaesium in terrestrial ecosystems?



B Howard

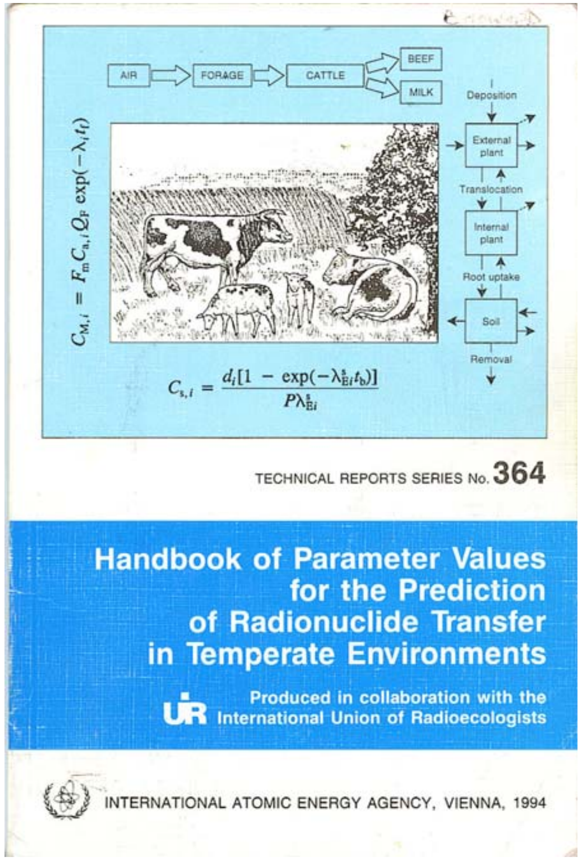
University of Nottingham, UK

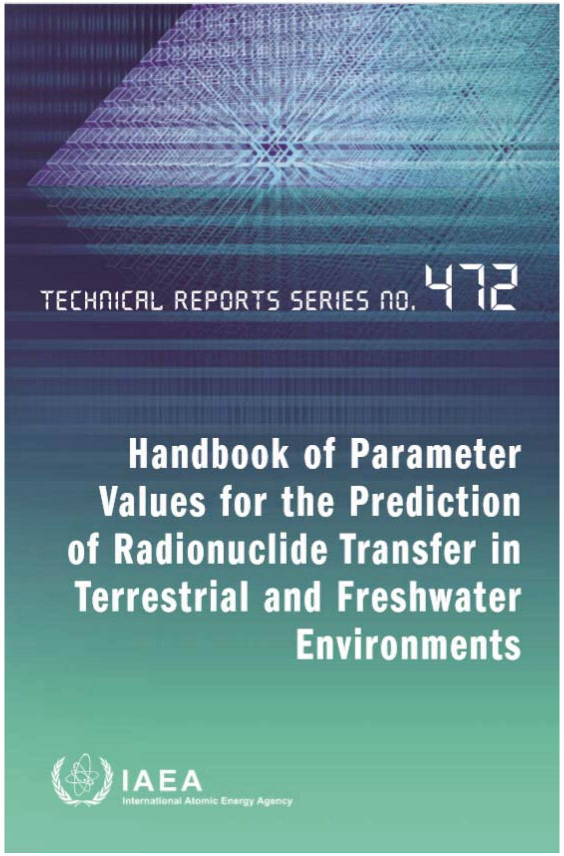


Why is radiocaesium important?

- 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





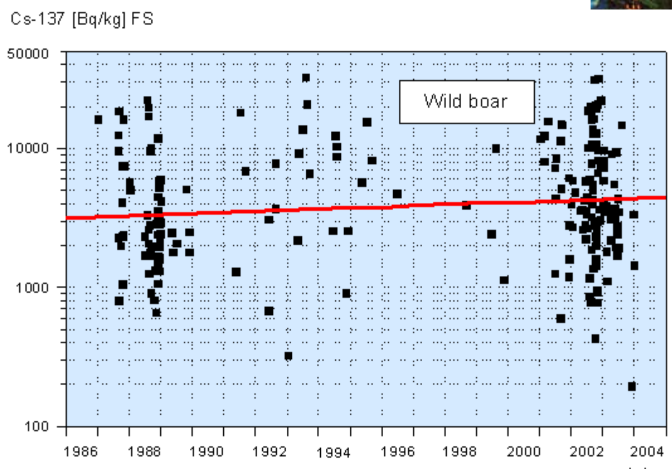
Analysis of radioecological data in IAEA Technical Reports Series publications to identify key radionuclides and associated parameter values for human and wildlife assessments

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

Post-Chernobyl Cs knowledge

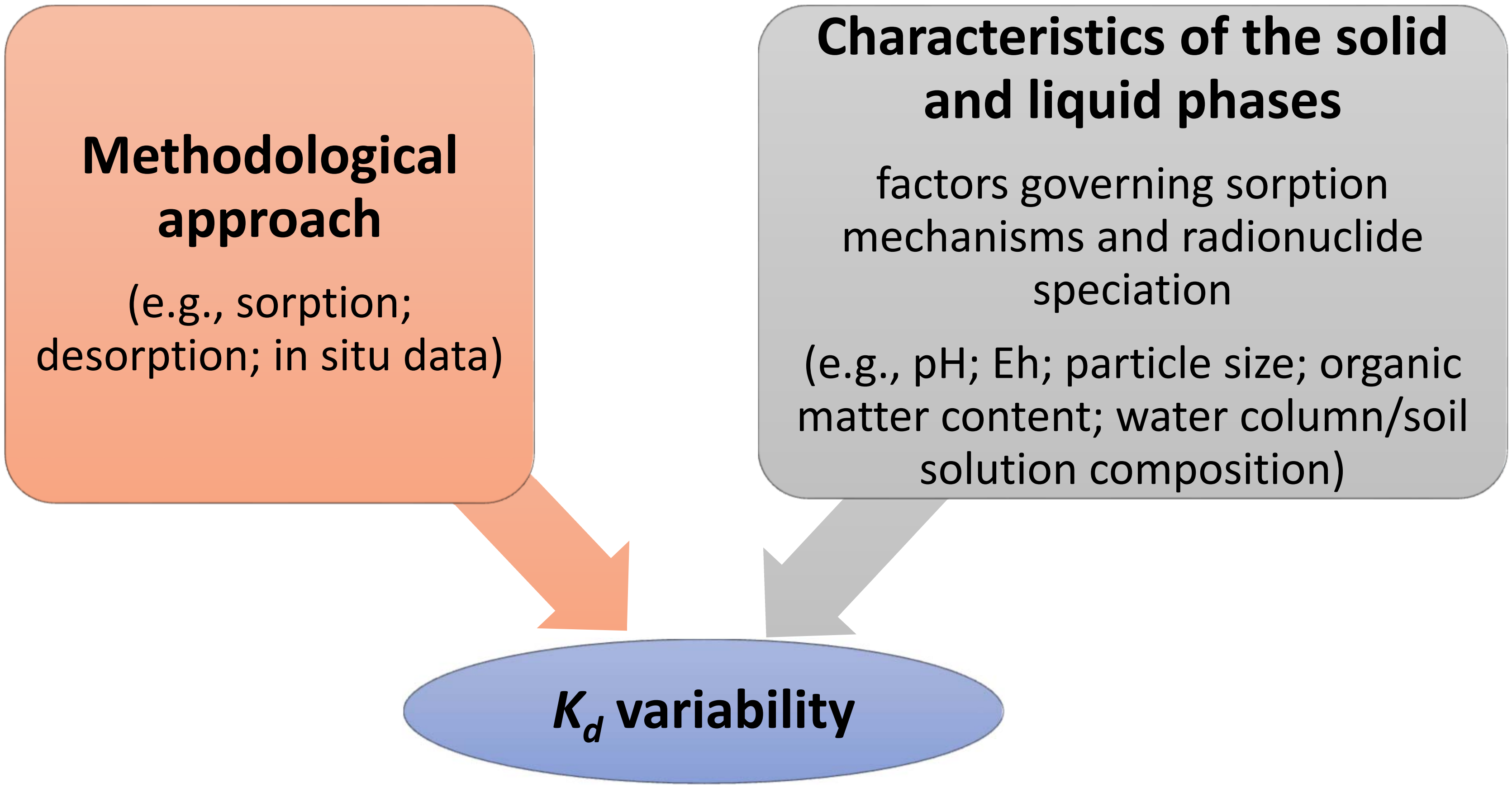
Importance of :

- 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

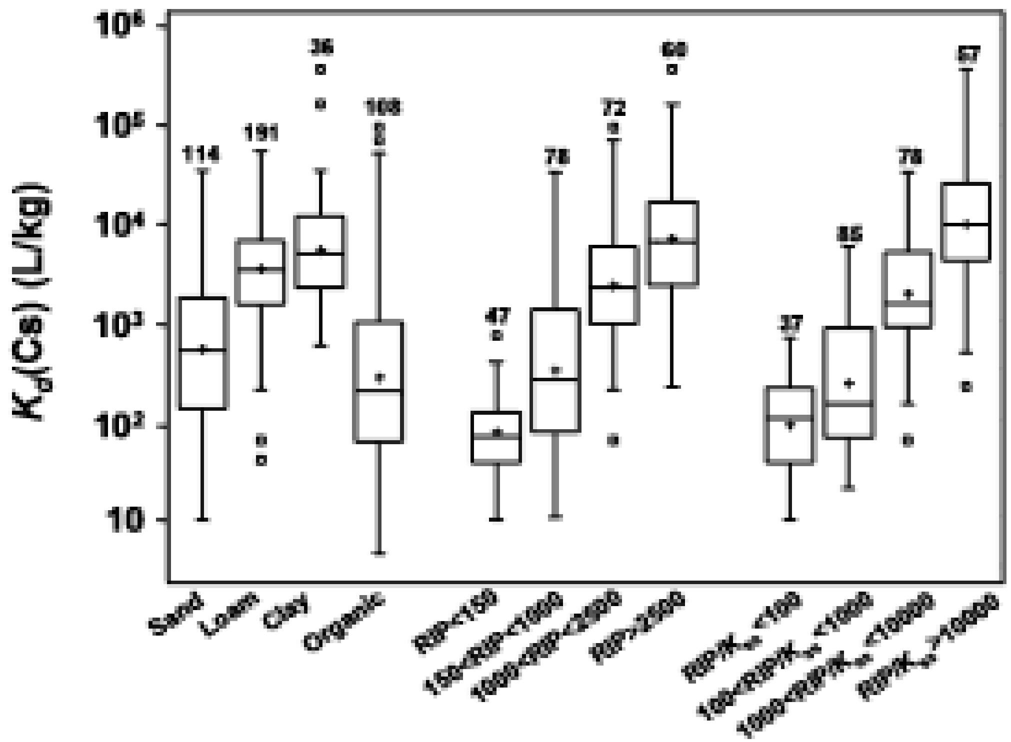
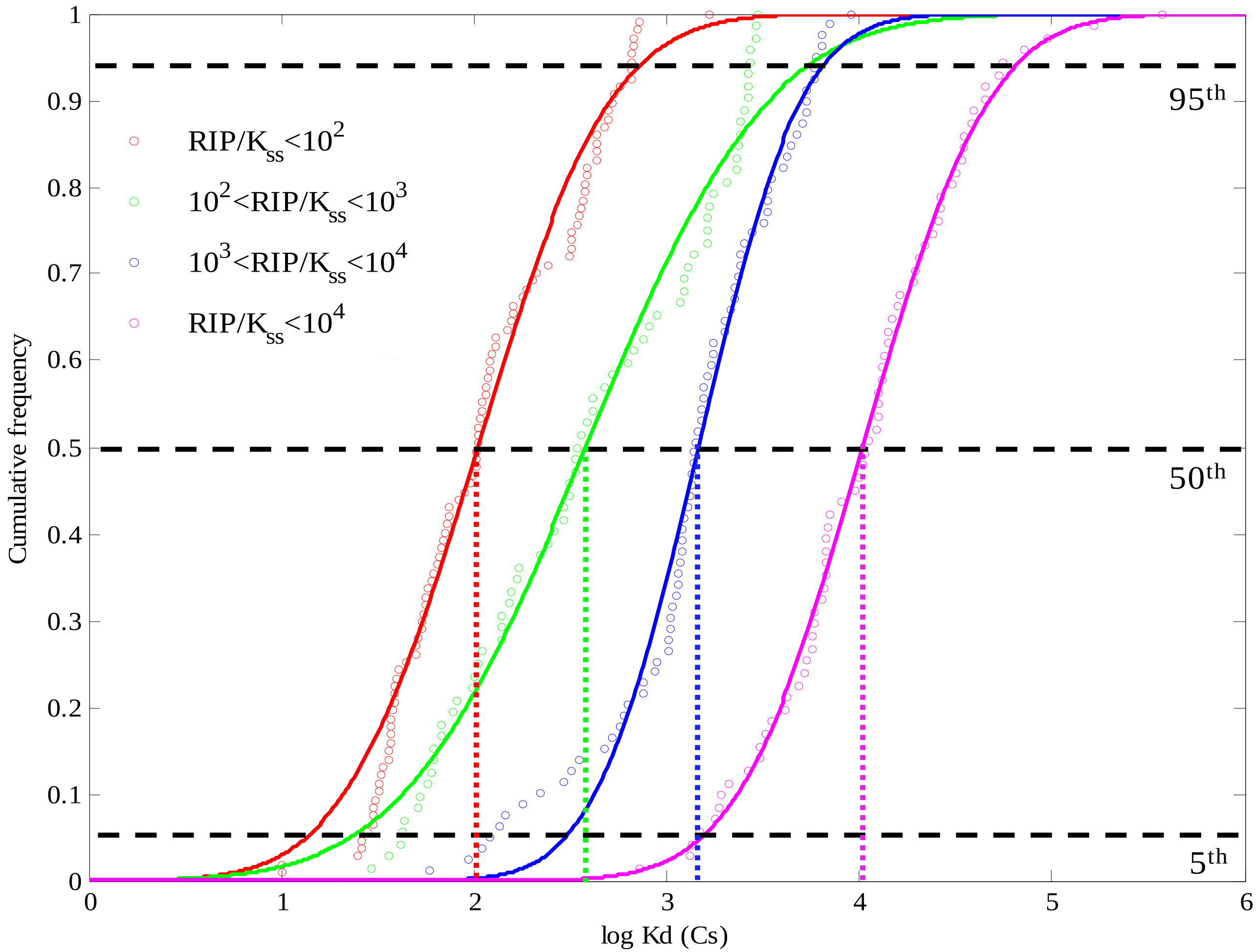


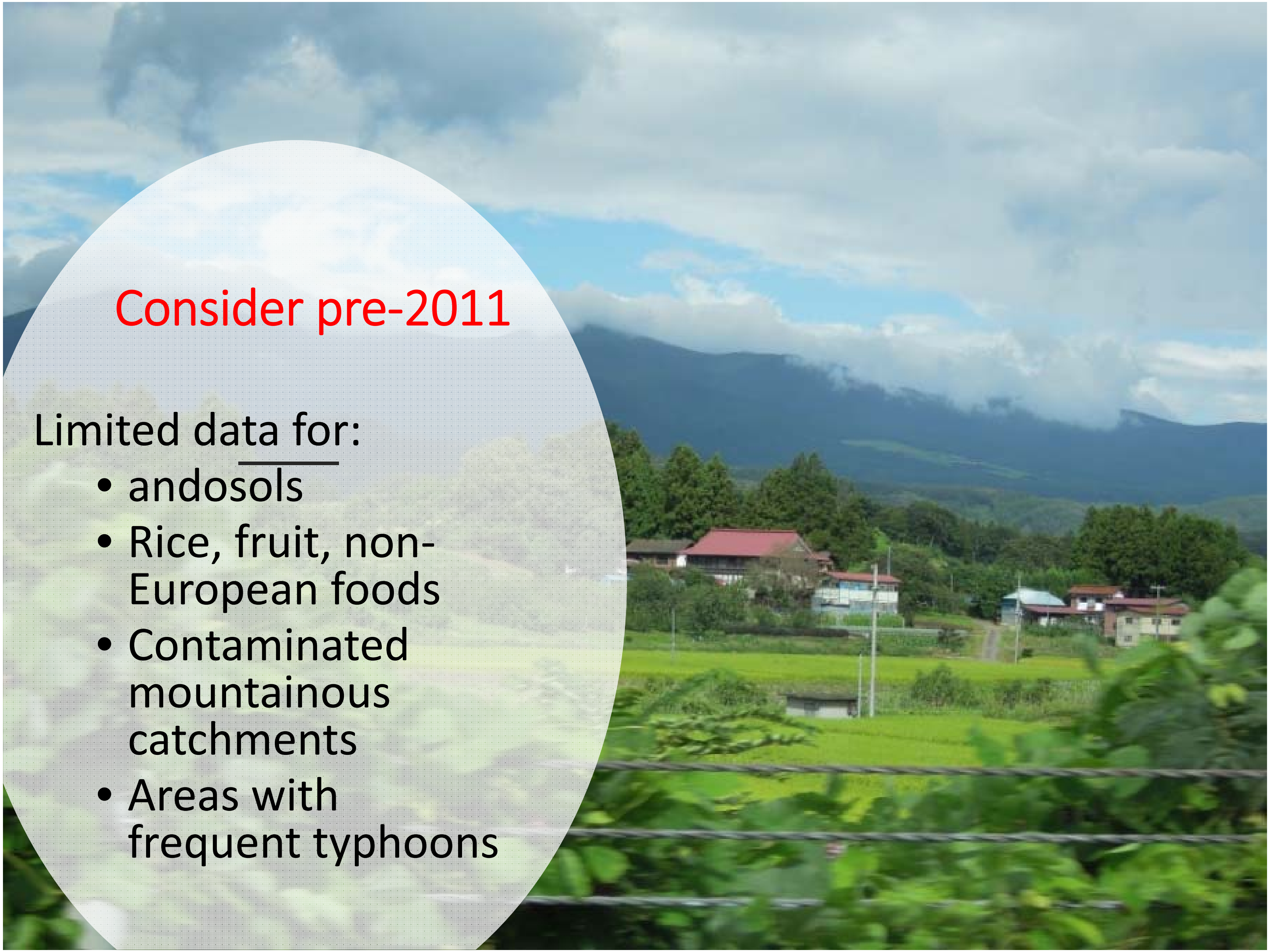
FIG. 2. Box-and-whisker plots of K_d (Cs) for soils grouped according to the texture/OM and cofactor criteria. The box encloses the middle 50% of the distribution of values, and the median is represented as a horizontal line inside the box. Vertical lines extend to the limits of the 1.5 interquartile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

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


Consider pre-2011
Limited data for:
• andosols
• Rice, fruit, non-European foods
• Contaminated mountainous catchments
• Areas with frequent typhoons

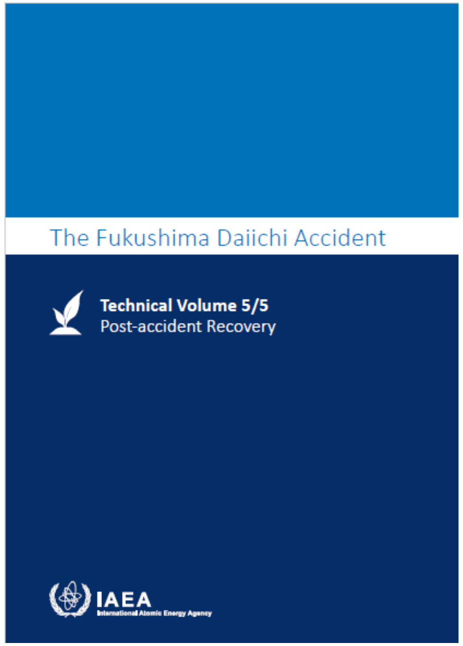
Six years on……

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 :

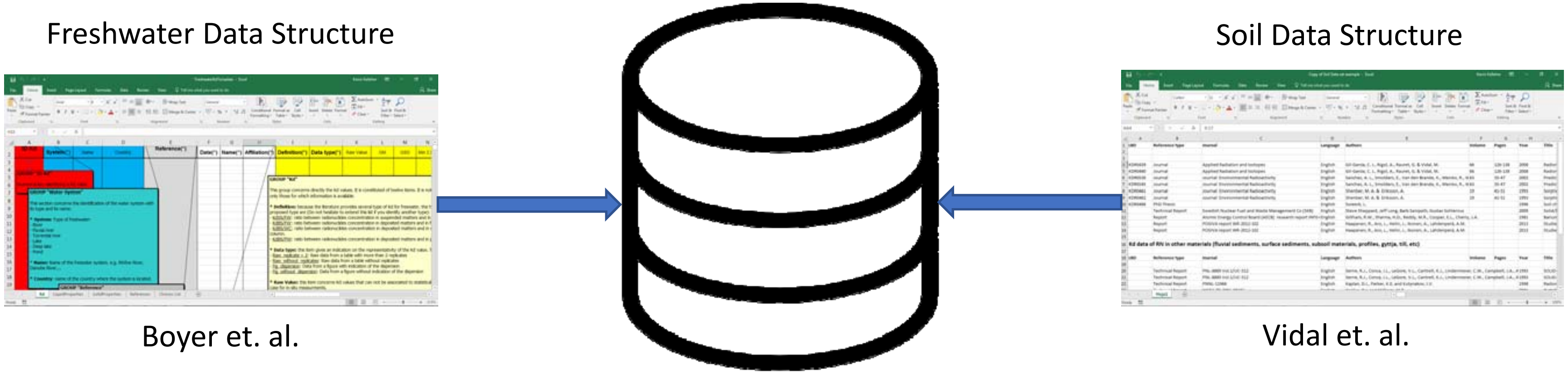
Working Group 4 – Transfer processes and data for radiological impact assessment

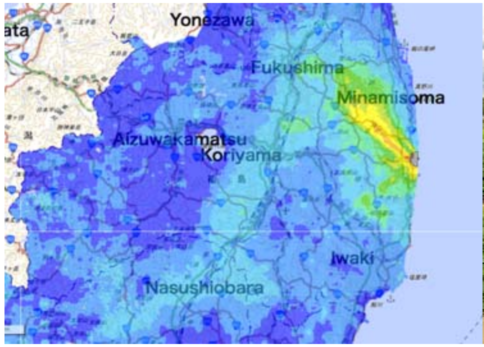
MODARIA II activities

IAEA
International Atomic Energy Agency

K_d sub group: Development of Global K_d database prototype

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



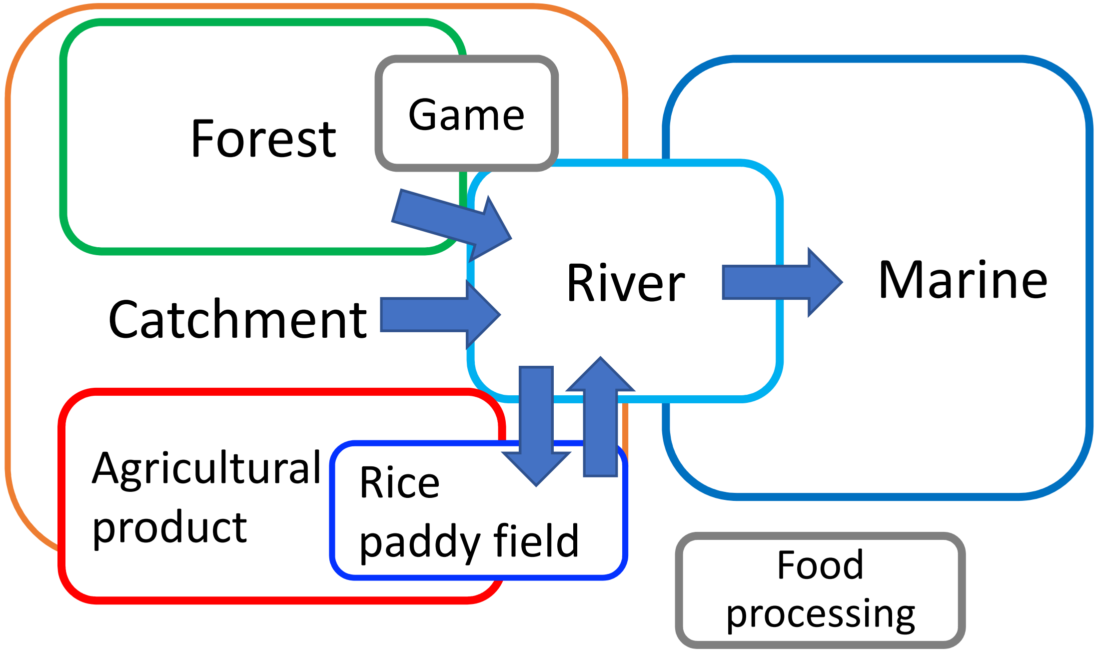




WG4 sub-group

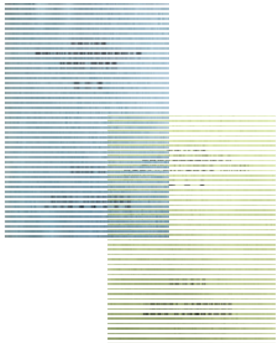
Collation of environmental transfer parameters after the Fukushima accident

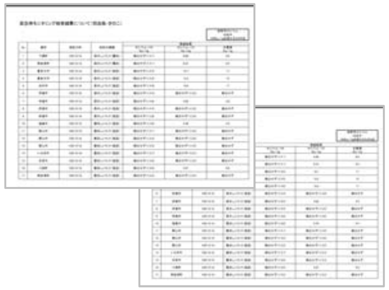
(Fukushima parameters) Leader: K. Tagami (QST)

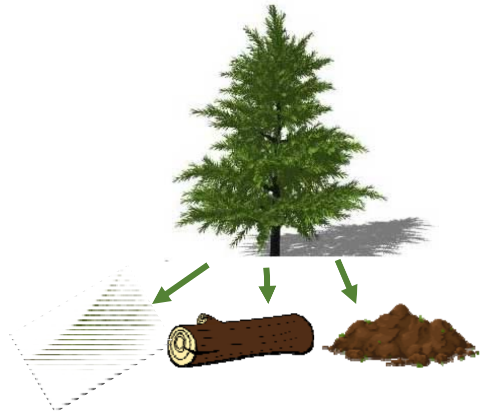


Forest data

—Data compilation ongoing (Hashimoto et al)



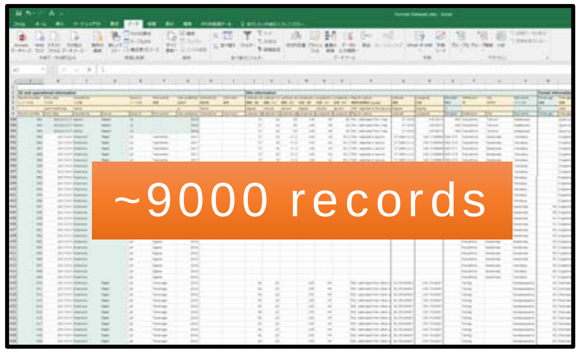




Journal papers **Reports by governments** **On the Web**

- **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

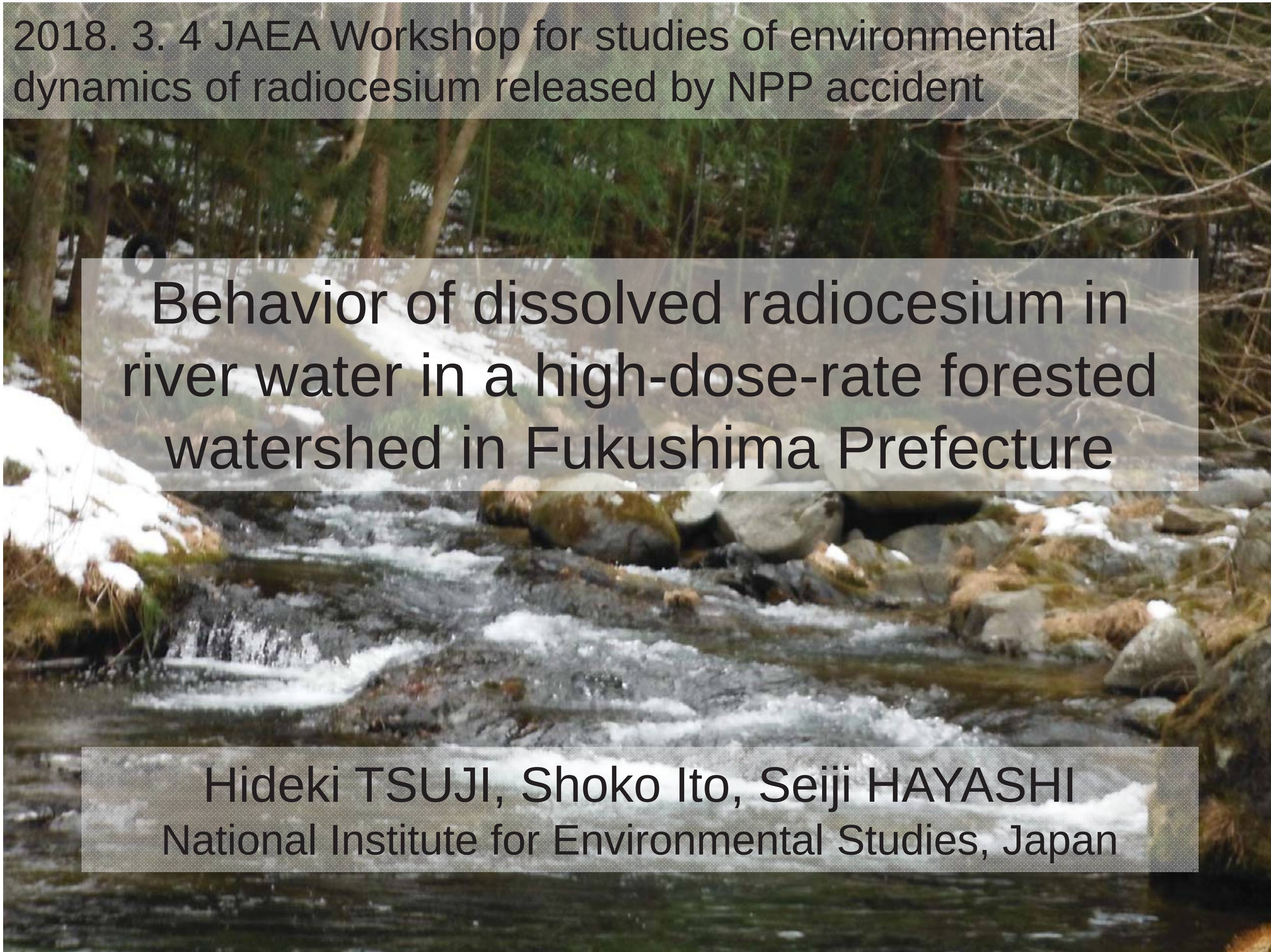
	Parameters	RI / stable	
472 Terrestrial & Freshwater (equilibrium)	a, m^2/kg	RI	Interception coefficient
	F_{tr}, -	RI	Translocation ratio
	K_s, m^{-1}	RI	Resuspension factor
	K_d, L/kg	RI	Distribution coefficient in soil
	F_v, -	RI	Concentration ratio from soil to plant
	F_m, F_f, d/kg or d/L	RI+S	Feed transfer coefficient, animal products
	T_{ag}, m^2/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^2/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

Reservoir of radiocaesium in forested catchments

High proportion of forest in SDA 1, 2 and 3

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}Cs concentration in a forest river water under baseflow and storm runoff conditions

4. The source of dissolved ^{137}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

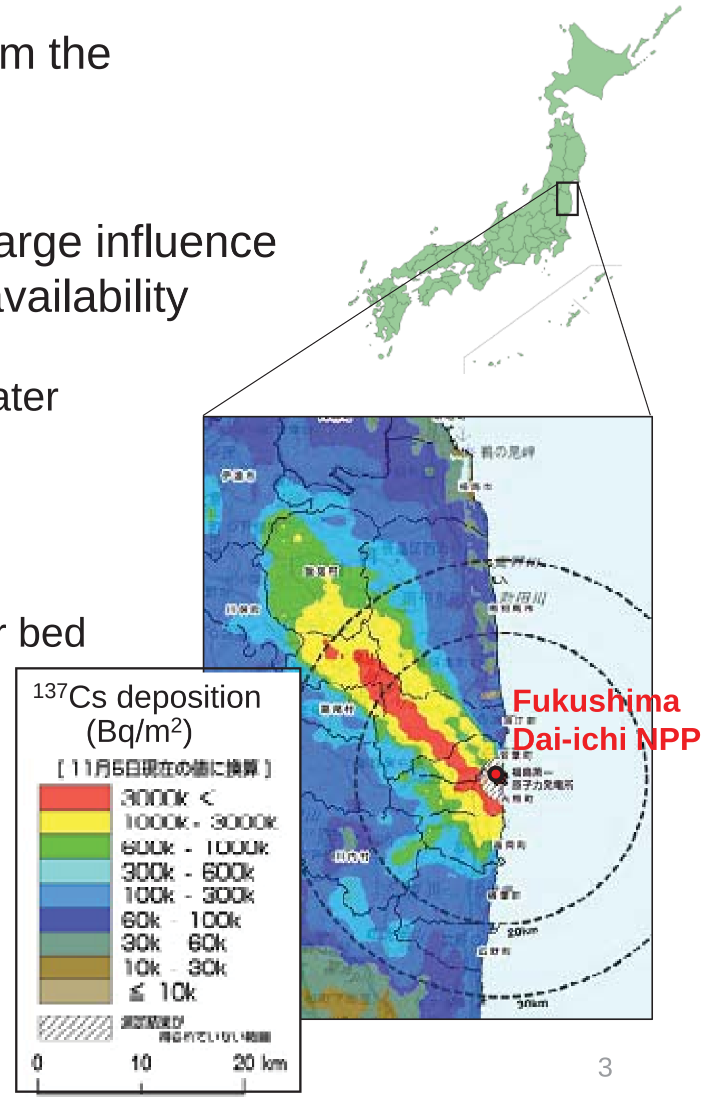
The dissolved form of radiocesium has a large influence on aquatic ecosystem due to the high bioavailability

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

⇧

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

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

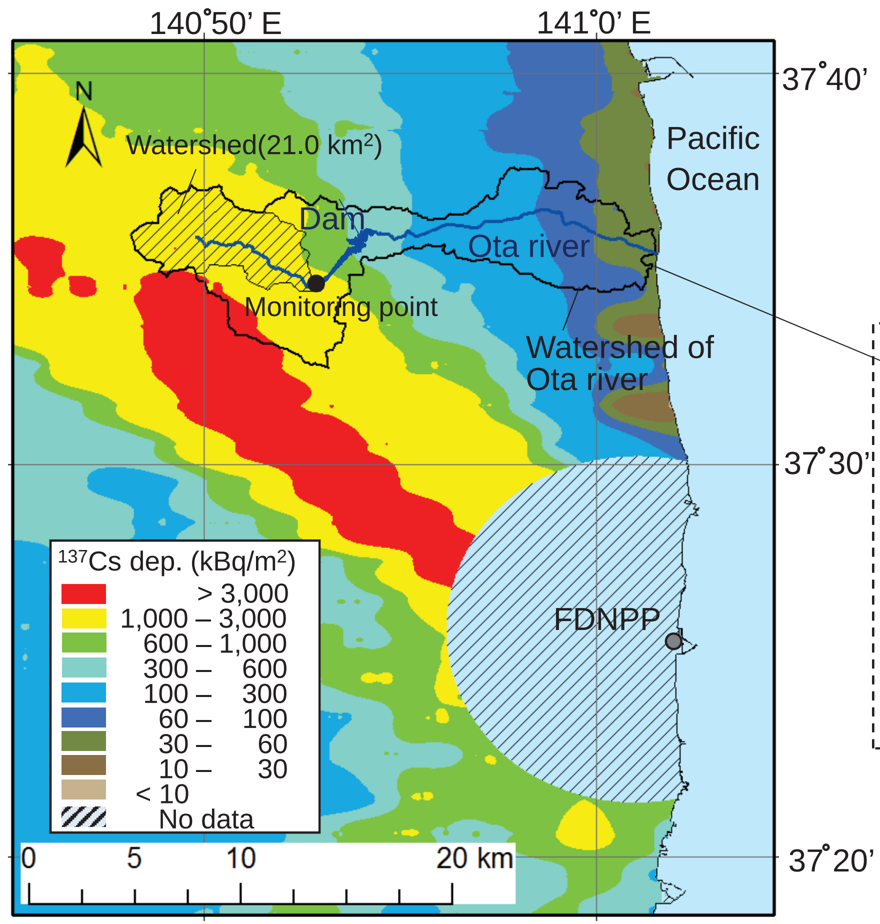


Purpose of this study

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

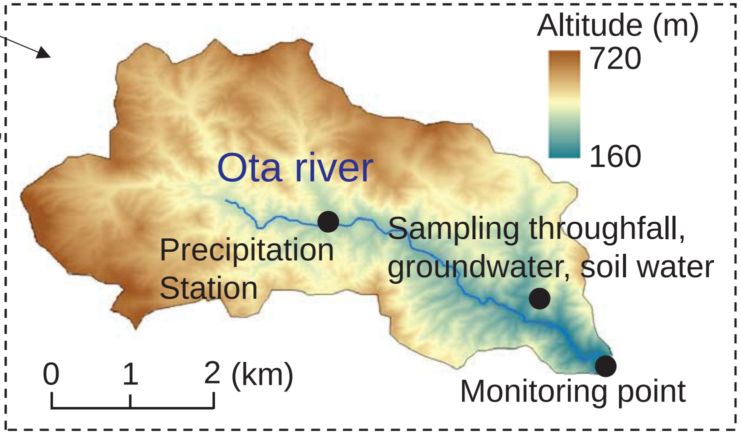
Study site

Ota river watershed



Land Use：
99.2% of forest area

Averaged ^{137}Cs deposition：
1.7 MBq/m²



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}Cs [Bq/L]

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

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

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

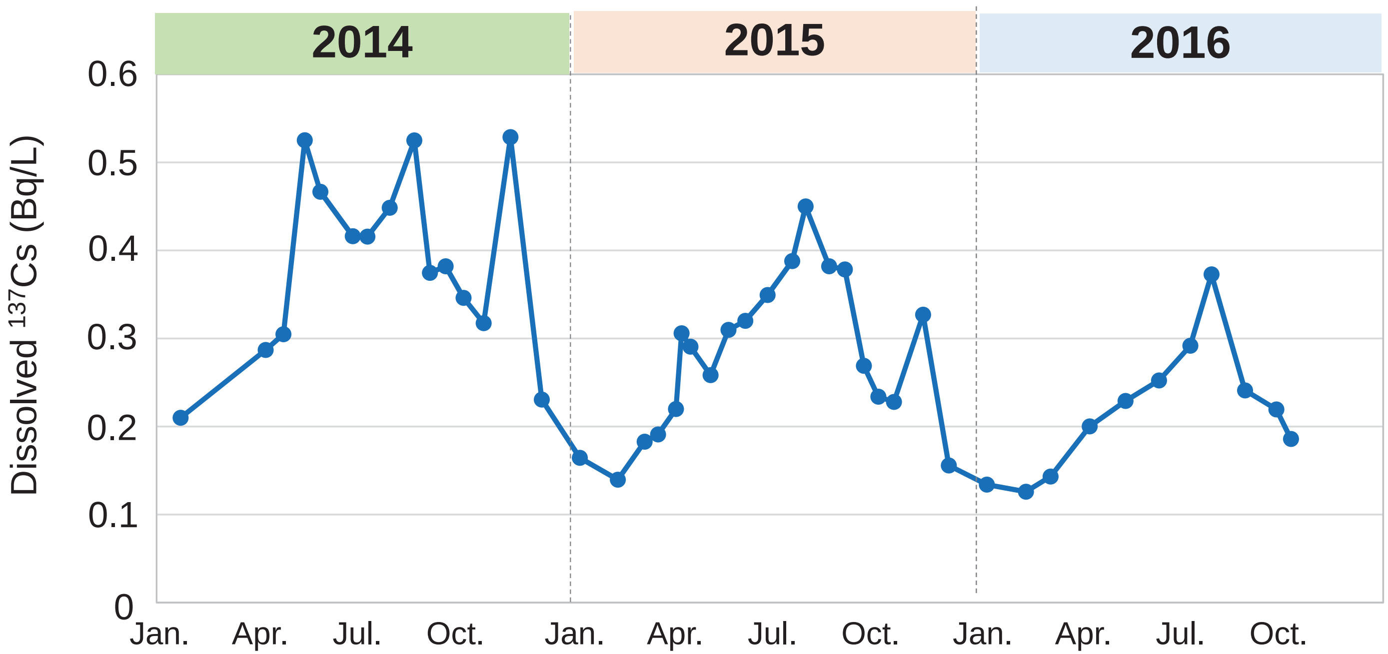
2. coexisting ion concentration (K^+, NH_4^+…)
   by ion chromatography (ICS-1600)

3. dissolved organic carbon concentration (DOC)
   by TOC analyzer (TOC-L)

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

Results

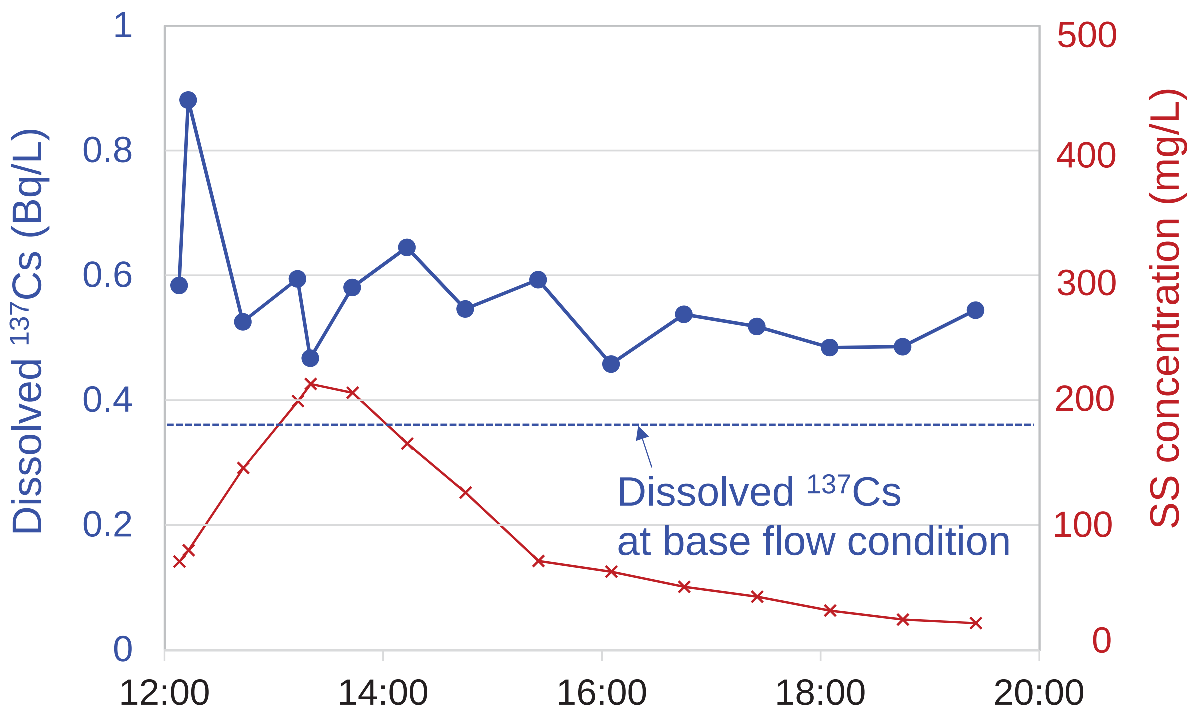
Dissolved ^{137}Cs concentration **under base flow condition**



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

Dissolved ^{137}Cs concentration **under storm runoff condition**

Runoff event on Jul.16, 2015



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

Comparison of the magnitude of runoff and dissolved ^{137}Cs concentration

AGU PUBLICATIONS JGR

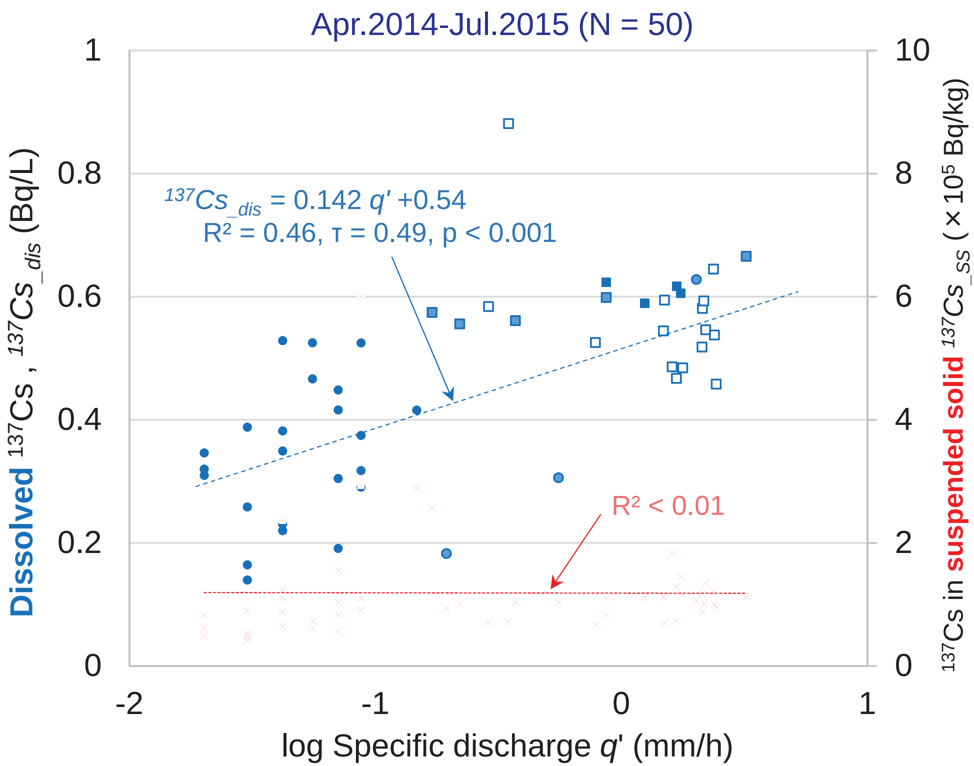
Journal of Geophysical Research: Biogeosciences

RESEARCH ARTICLE
10.1002/2016JG003428

Key Points:
• Dissolved 137Cs concentration in

Behavior of dissolved radiocesium in river water in a forested watershed in Fukushima Prefecture

H. Tsuji[1], T. Nishikiori[1,2], T. Yasutaka[3], M. Watanabe[1], S. Ito[1], and S. Hayashi[1]



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

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

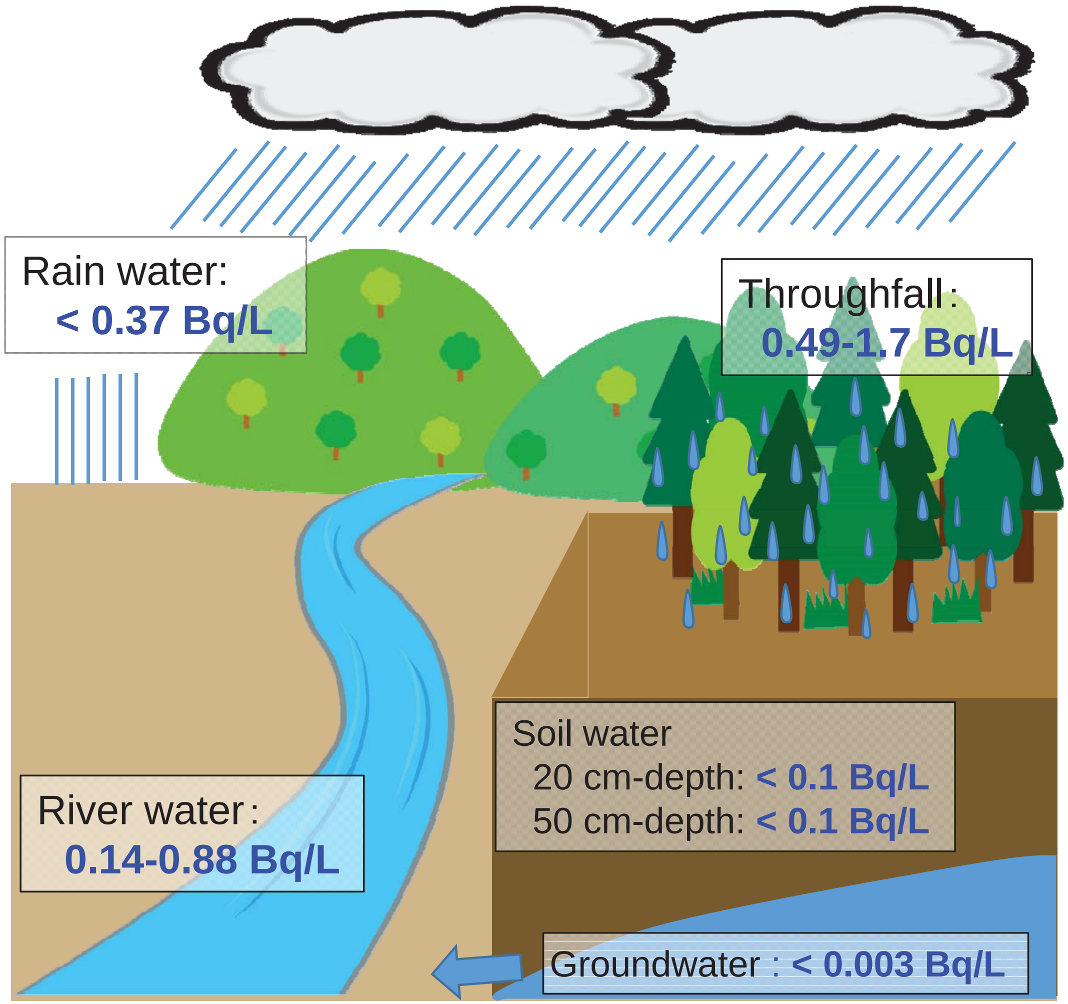
⇩

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

⇩

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

Dissolved ^{137}Cs concentration in the influent water of the river water



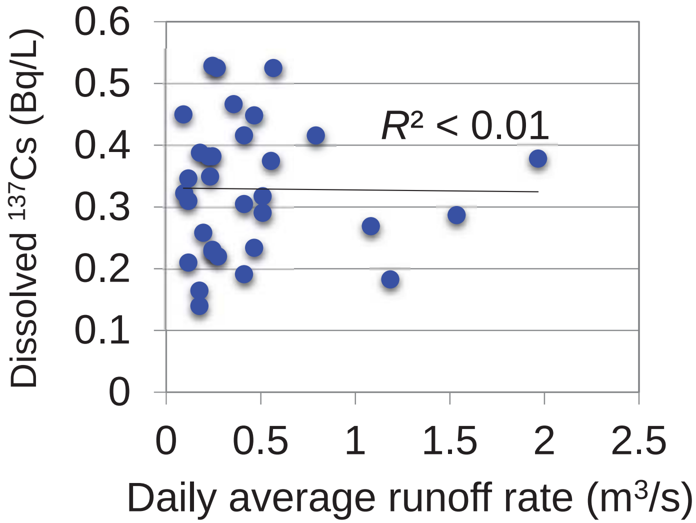
Dissolved ^{137}Cs in throughfall water was higher than river water, but it will little contribute the increase of dis.^{137}Cs in river water, because it is adsorbed by mineral particles during the infiltration process

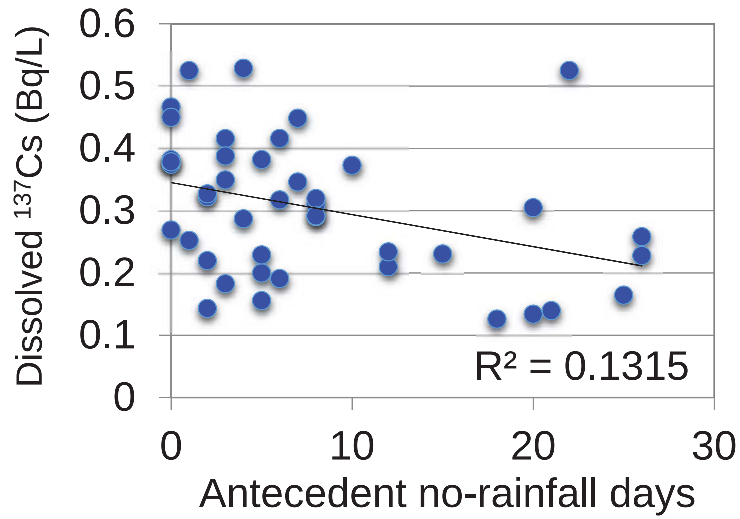
(Nakanishi et al., 2014)

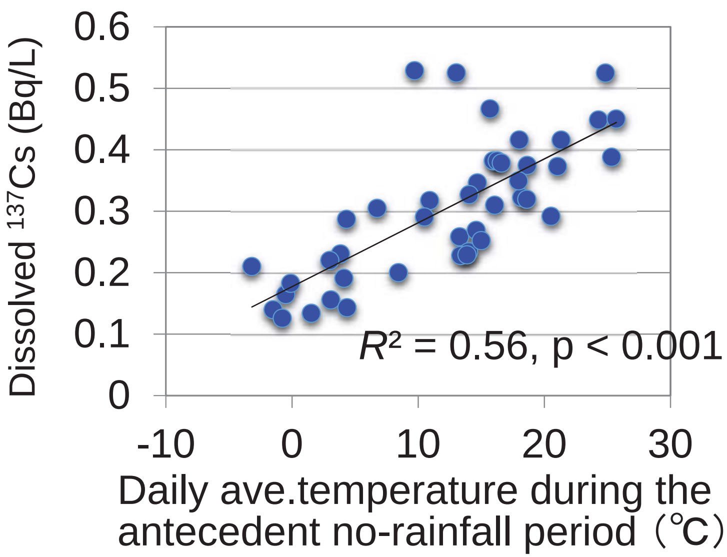
⇩

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

Relationship of dissolved ^{137}Cs and environmental factor at base flow condition



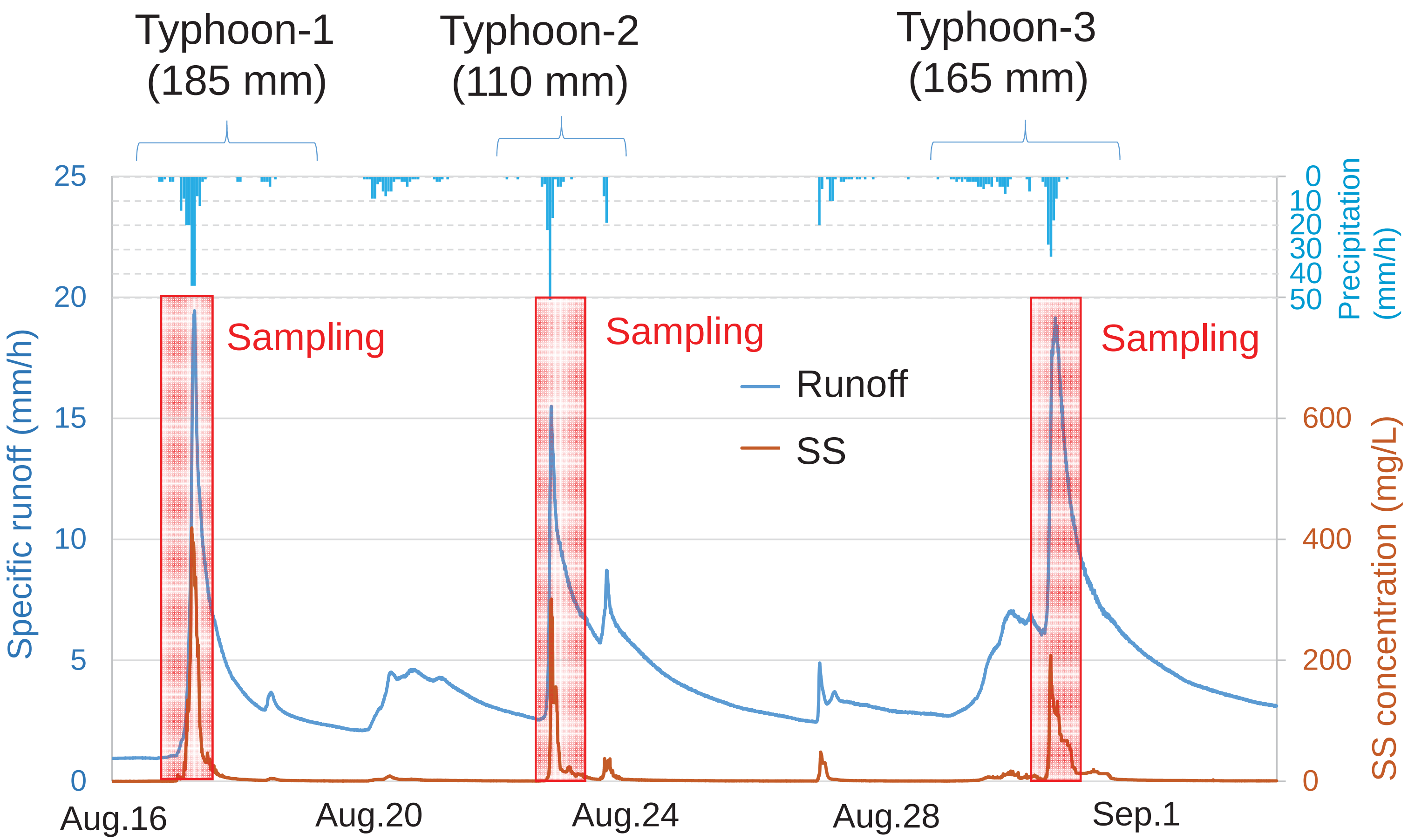




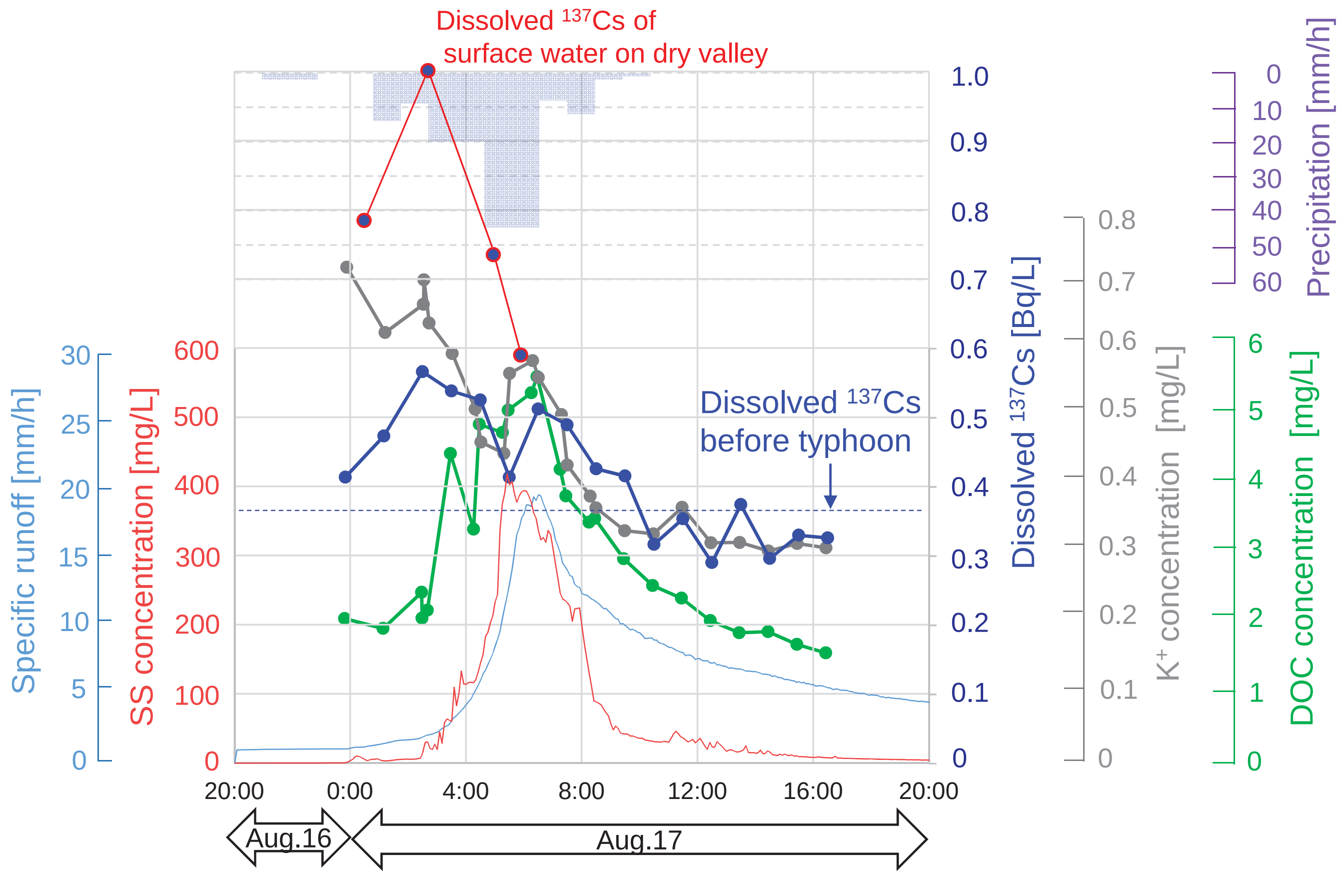
Suggesting that dissolved ^{137}Cs was supplied by the decomposition of organic matter by microorganisms

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

Total precipitation：640 mm



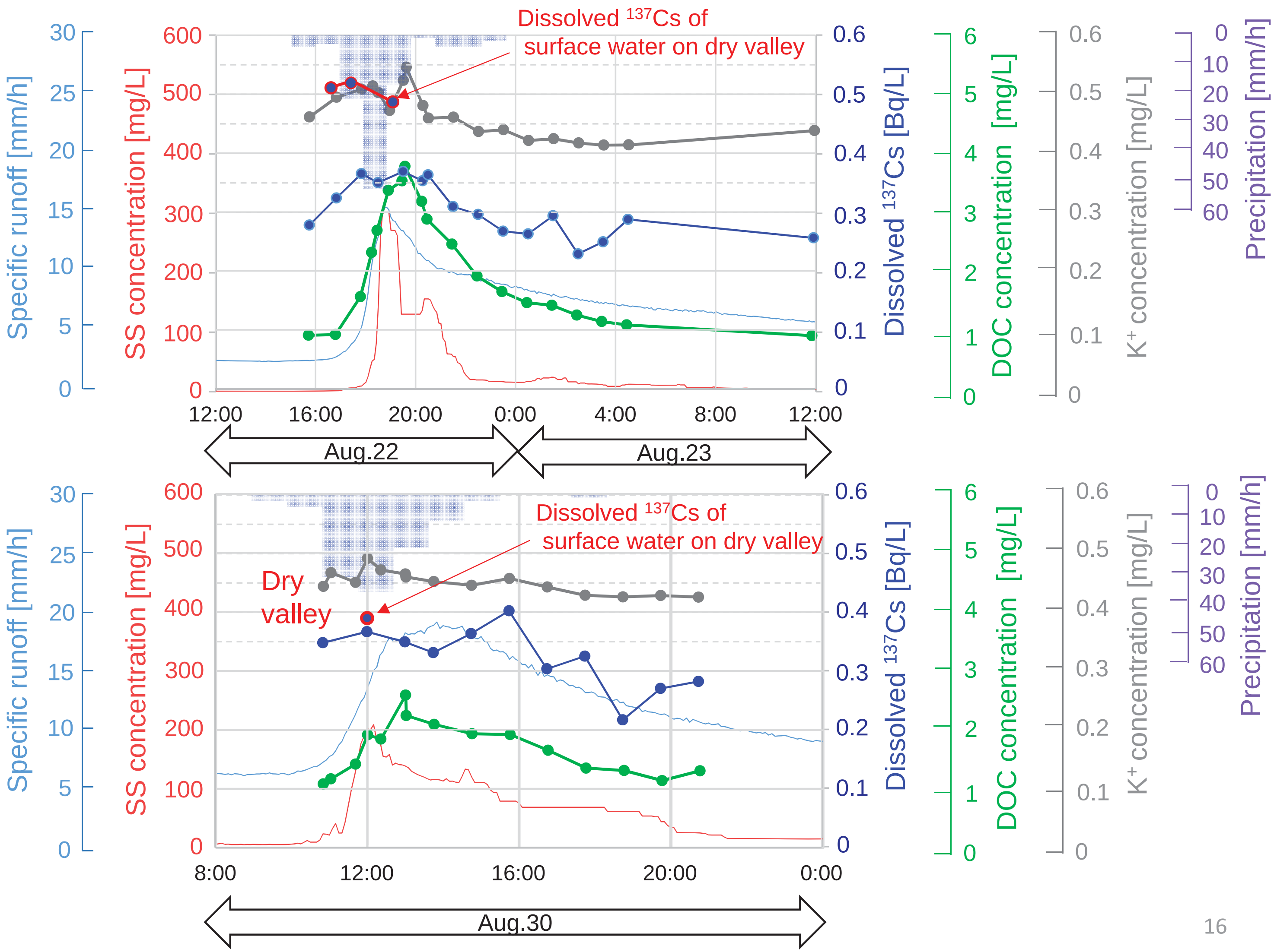
Dissolved ^{137}Cs and coexisting solute conc. at the first runoff event



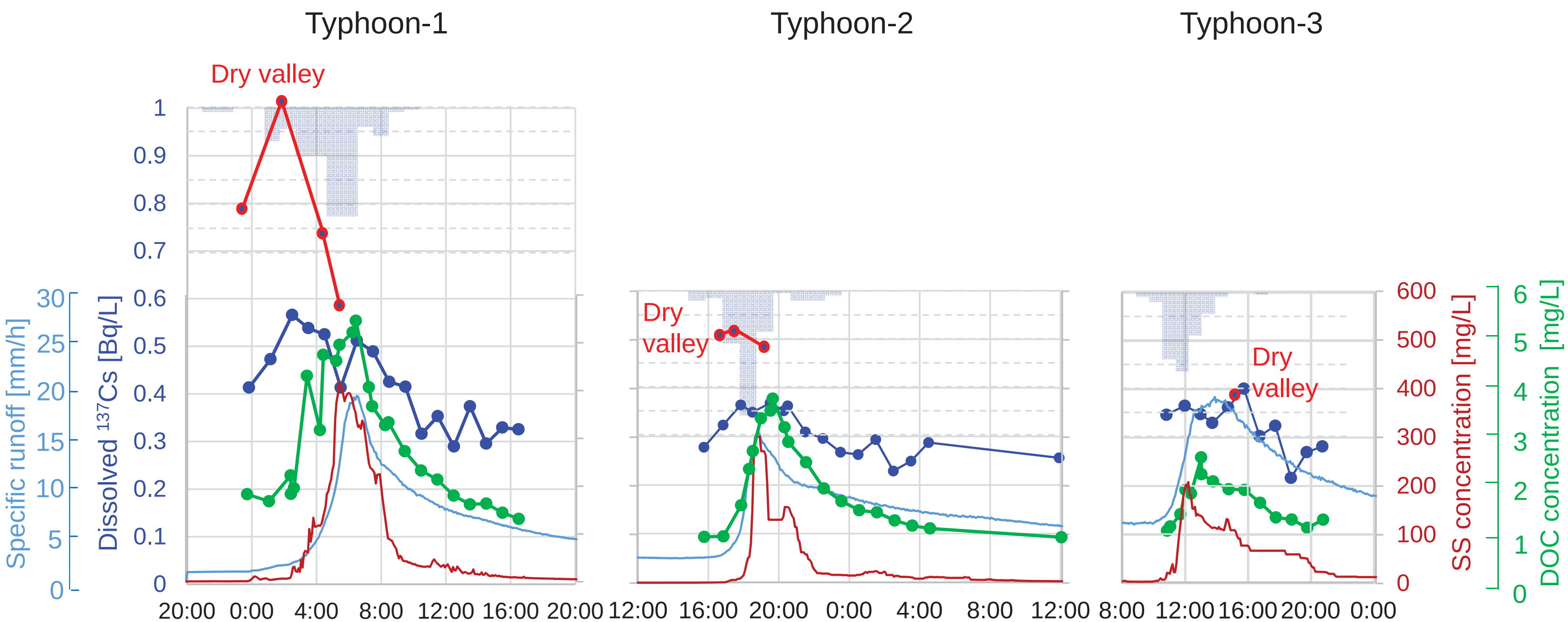
- Dissolved ^{137}Cs before the runoff peak was higher than those at base flow condition
- Dissolved ^{137}Cs in dry valley was higher than those of river water

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

Dissolved ^{137}Cs and coexisting solute at the second and third runoff event



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

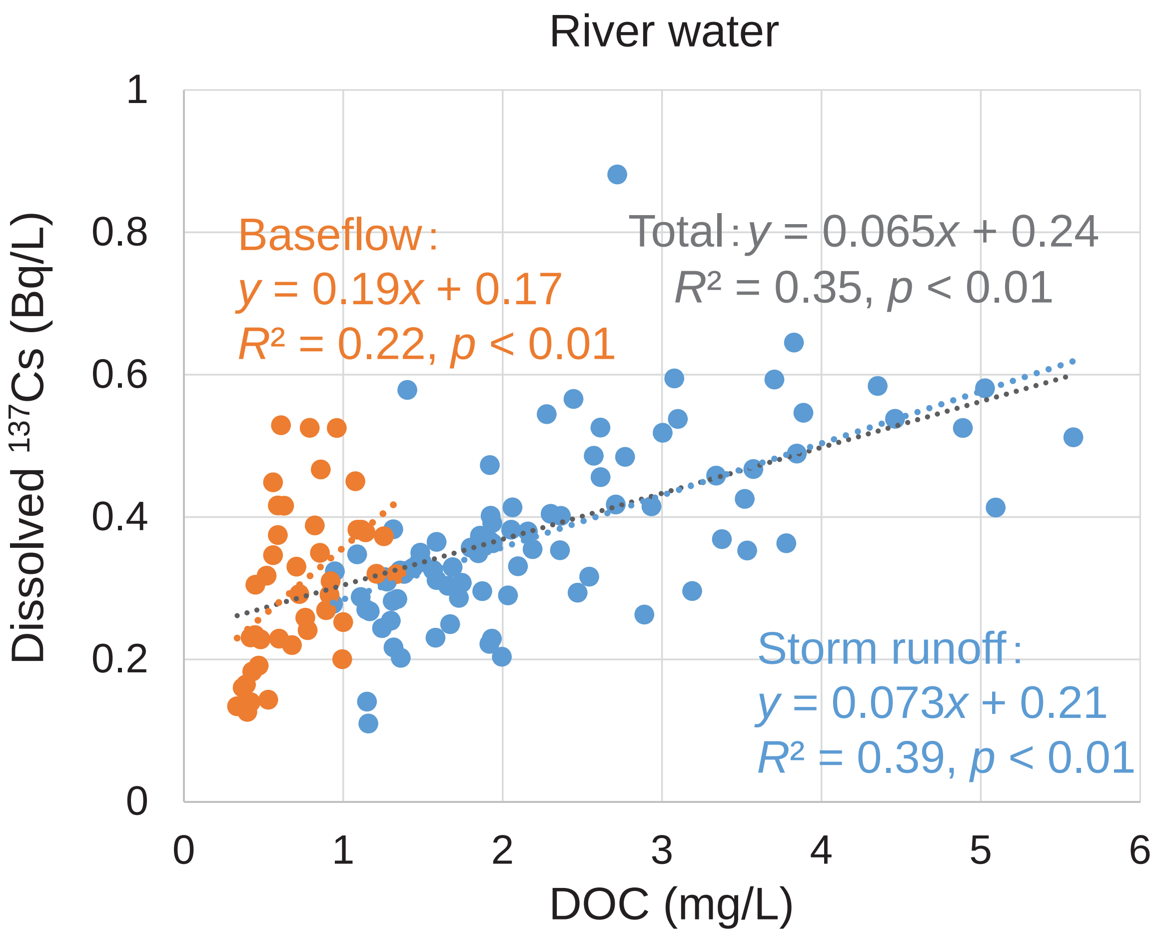


- The peak of dissolved ^{137}Cs conc. in each runoff event gradually decreased
- The peak of DOC concentration also decreased

⇩

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

Relationship of dissolved ^{137}Cs and DOC concentration



Surface soil water

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

groundwater

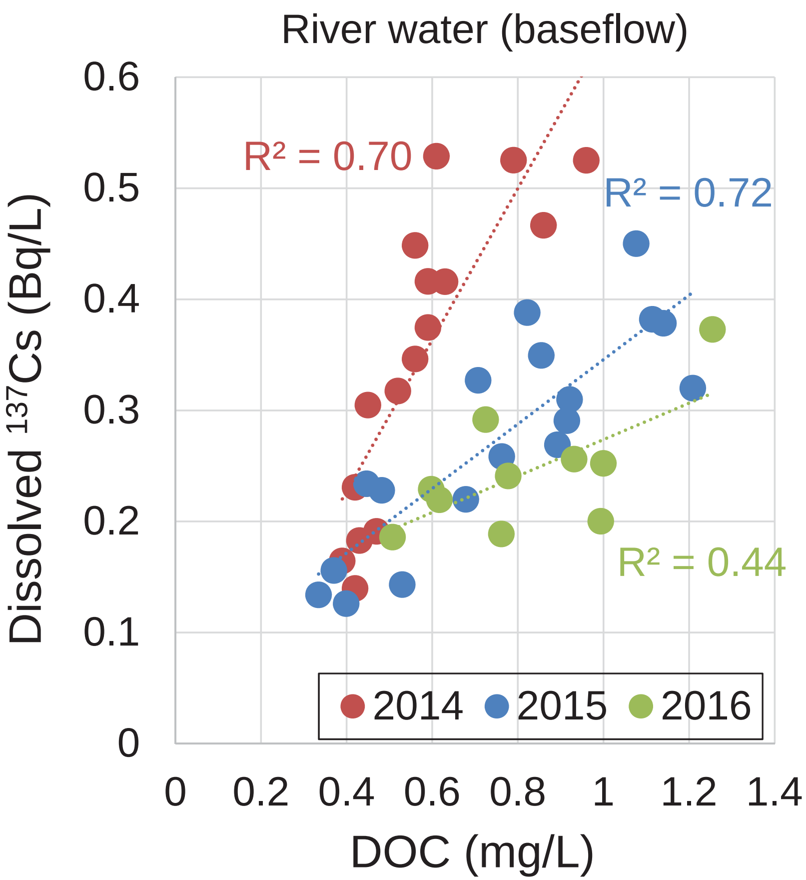
- DOC : ＜0.5 mg/L
- dis.^{137}Cs : ＜0.01 Bq/L

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

The coefficient of the regression line between DOC and $^{137}Cs_{\_dis}$ 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.^{137}Cs conc. to DOC conc.



Dissolved ^{137}Cs concentration estimation equation using "DOC conc." and "elapsed time" as explanatory variables

Dissolved ^{137}Cs [Bq/L]

$\sim a \cdot \mathrm{DOC} + b \cdot (\underline{\mathrm{DOC} \times \mathbf{time}}) + c$

interaction

Time dependence of DOC

= the significance of coefficient "*b*"

↓

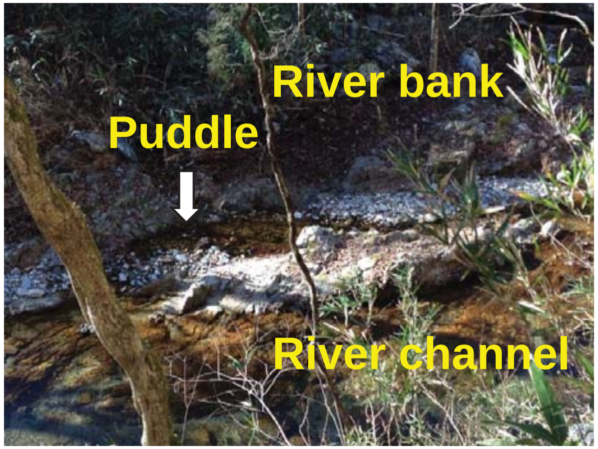
$b = -0.178 \pm 0.024$, $p < 0.001$

The dependence of dissolved ^{137}Cs conc. on DOC conc. significantly decreased.

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

The process of increasing dis.^{137}Cs in river water at runoff event

- Initial runoff:
  Elution of ^{137}Cs from riparian litter along with the rising water level



Also the source of ^{137}Cs at baseflow

- Around the runoff peak:
  Elution of ^{137}Cs from litter on the dry valley when occurring the return flow

Expansion of surface flow network in the process of runoff

Base flow（river）   Increasing runoff process   At the runoff peak

Conclusion

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

brings the following phenomenon :

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

Main source of dissolved ^{137}Cs

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

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

The 3rd 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

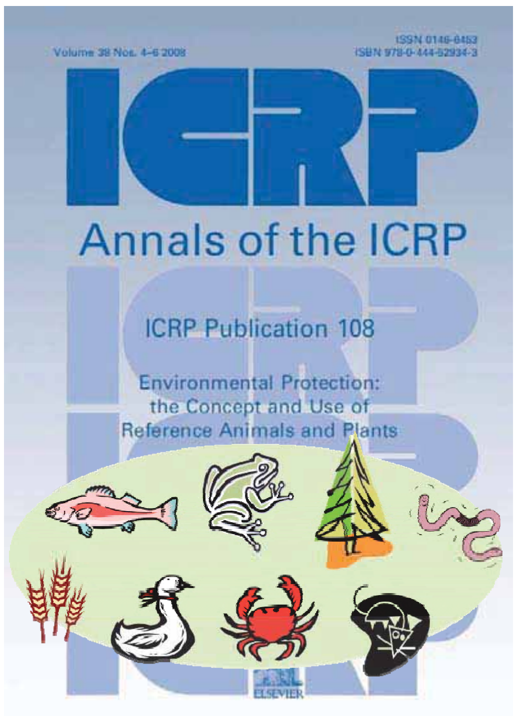
Hirosaki university

Graduate School of Health Sciences

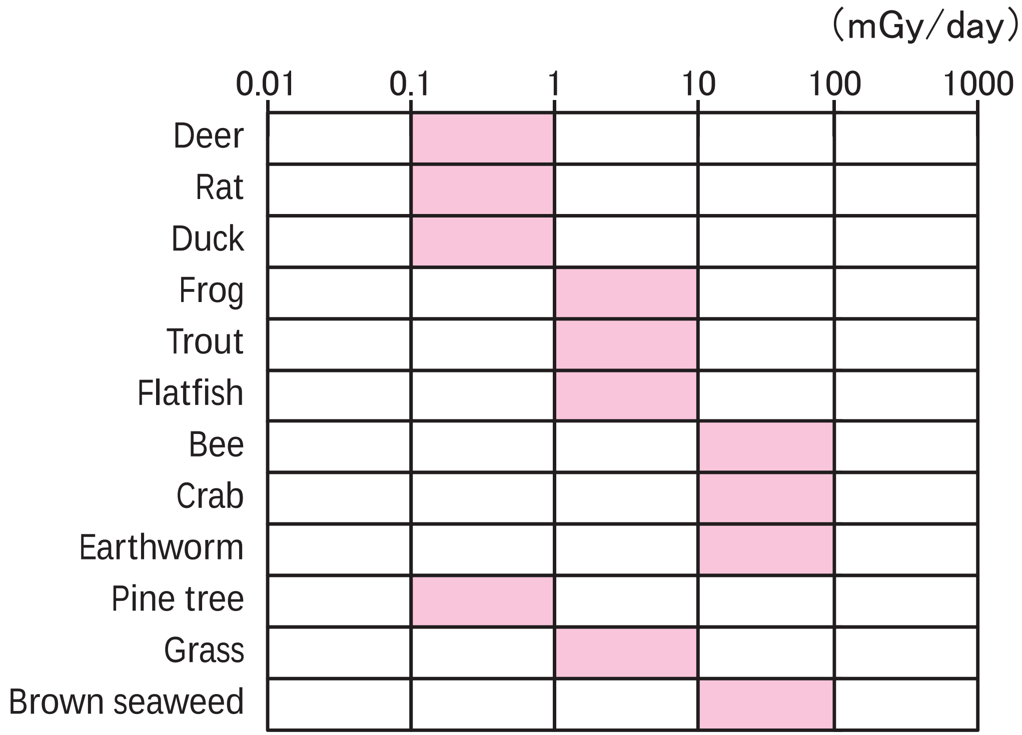
Ukedo River, Namie, Fukushima

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

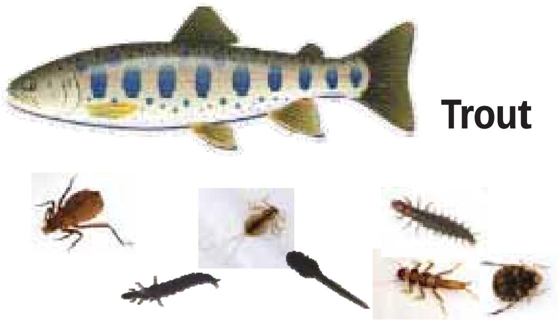
Land area

forest

urban area

Japanese monkey | Field mice | Cat | Raccoon

Hydrosphere



Trout

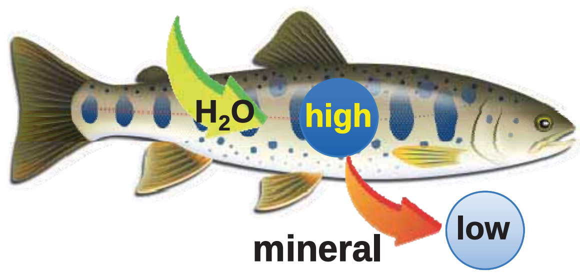
Aquatic biota

Difference in salt discharge mechanism between freshwater fish and saltwater fish

Migration and homeostasis due to concentration gradient

Freshwater

(Hypotonic, low mineral conc.)

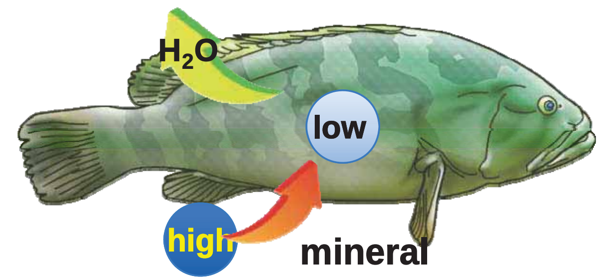


Leakage of K and Cs from the body

Do not discharge K or Cs

Saltwater

(Hypertonic, high mineral conc.)



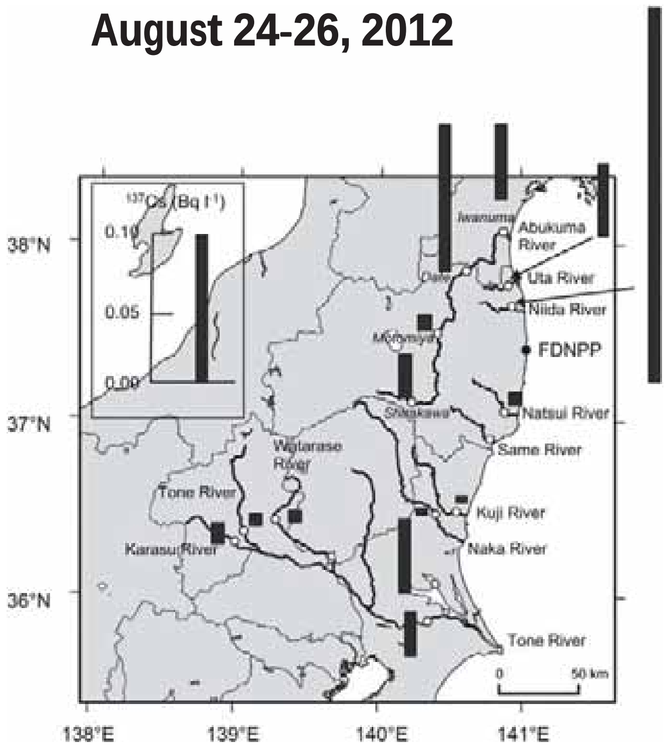
Inflow of K and Cs from outside the body

Proactive discharge of K and Cs

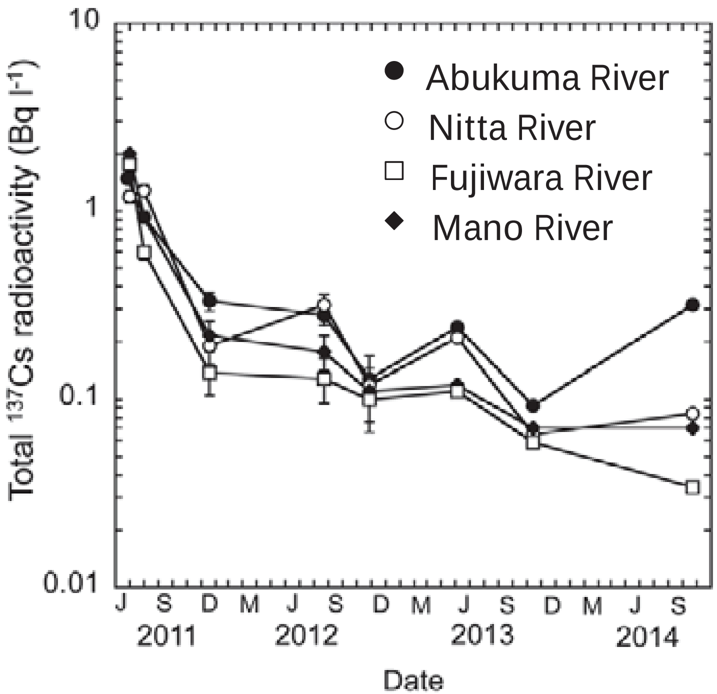
Biological half-life of radioactive cesium is long in freshwater fish

Radioactive cesium in rivers after FDNPP accident

August 24-26, 2012



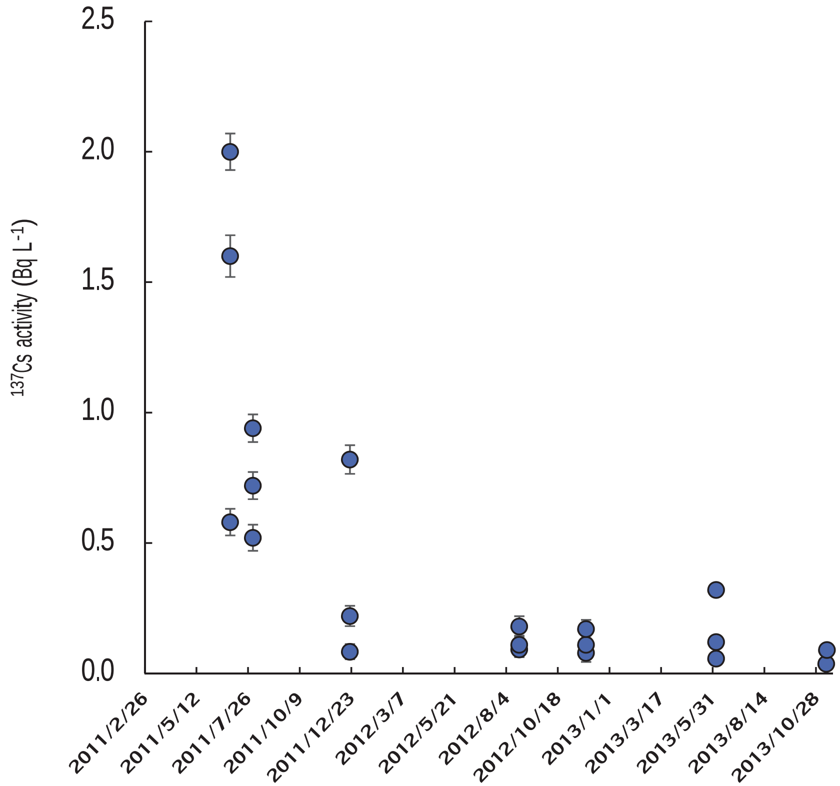
Changes in Cs-137 Concentration in rivers, Fukushima, japan



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

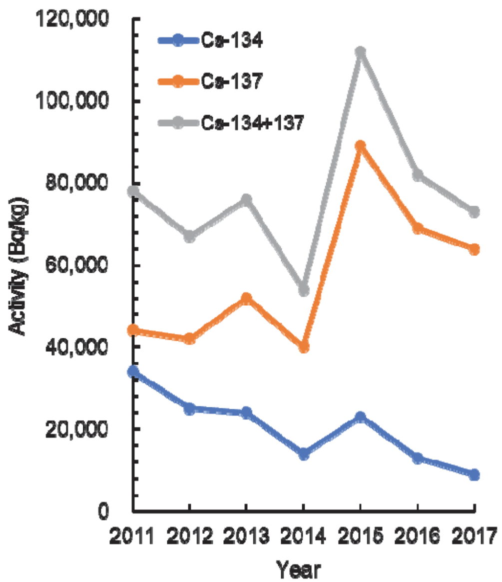
Field	Date	Radioactive cesium concentration (Bq/L)	
		Cs-134	Cs-137
Murohara Bridge	2015. 5. 19	<0.85	<0.88
	2015. 6. 11	<0.61	<0.69
	2015. 7. 15	<0.79	<0.63
	2015. 8. 14	<0.86	<0.80
	2015. 9. 30	<0.62	<0.88
	2015. 10. 19	<0.65	<0.84
	2015. 11. 7	<0.64	<0.75
	2015. 12. 16	<0.48	<0.99
	2016. 1. 9	<0.63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

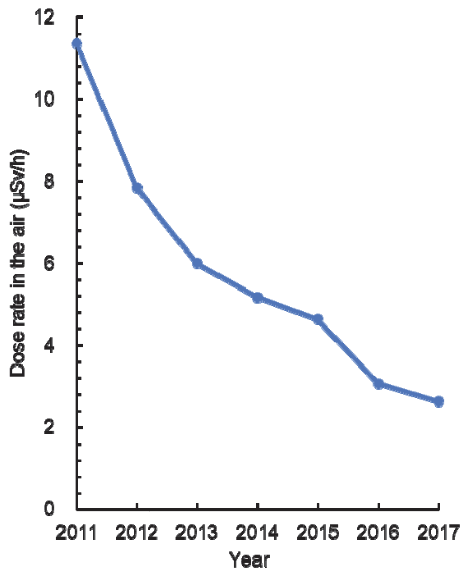
Radioactive material monitoring surveys in Ukedo River

@Murohara Bridge in Ukedo River

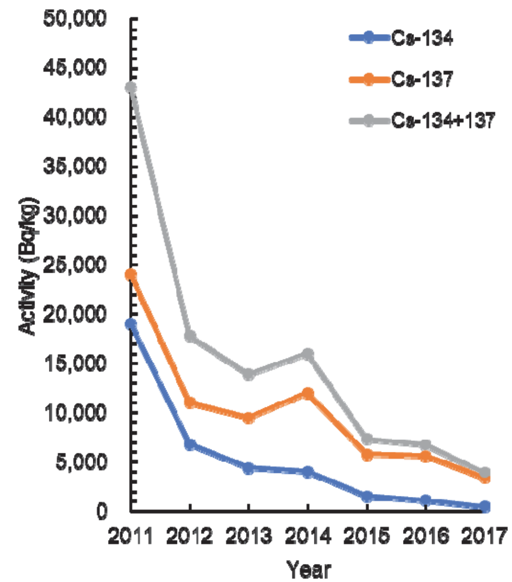
Soil in the left bank



Air dose rate



Bottom sediment



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

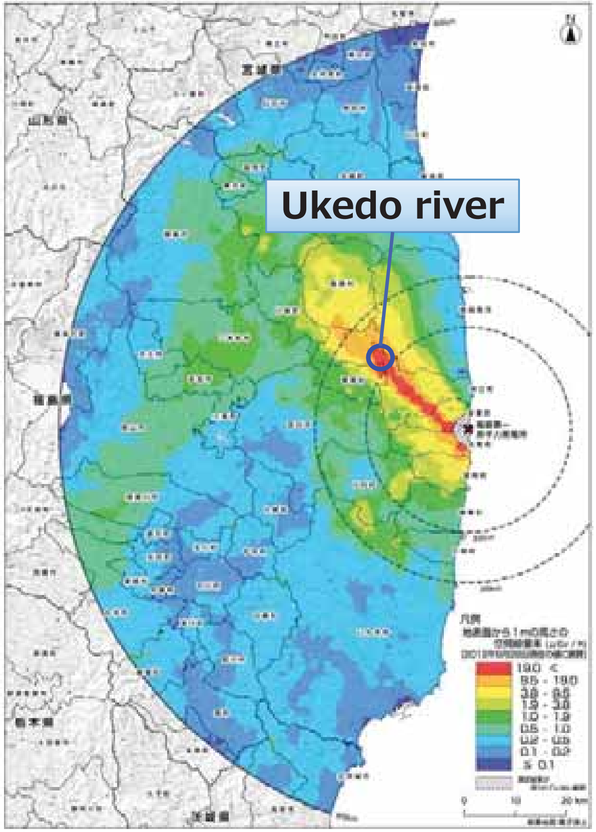
Study of Landlocked masu trout

Landlocked masu trout

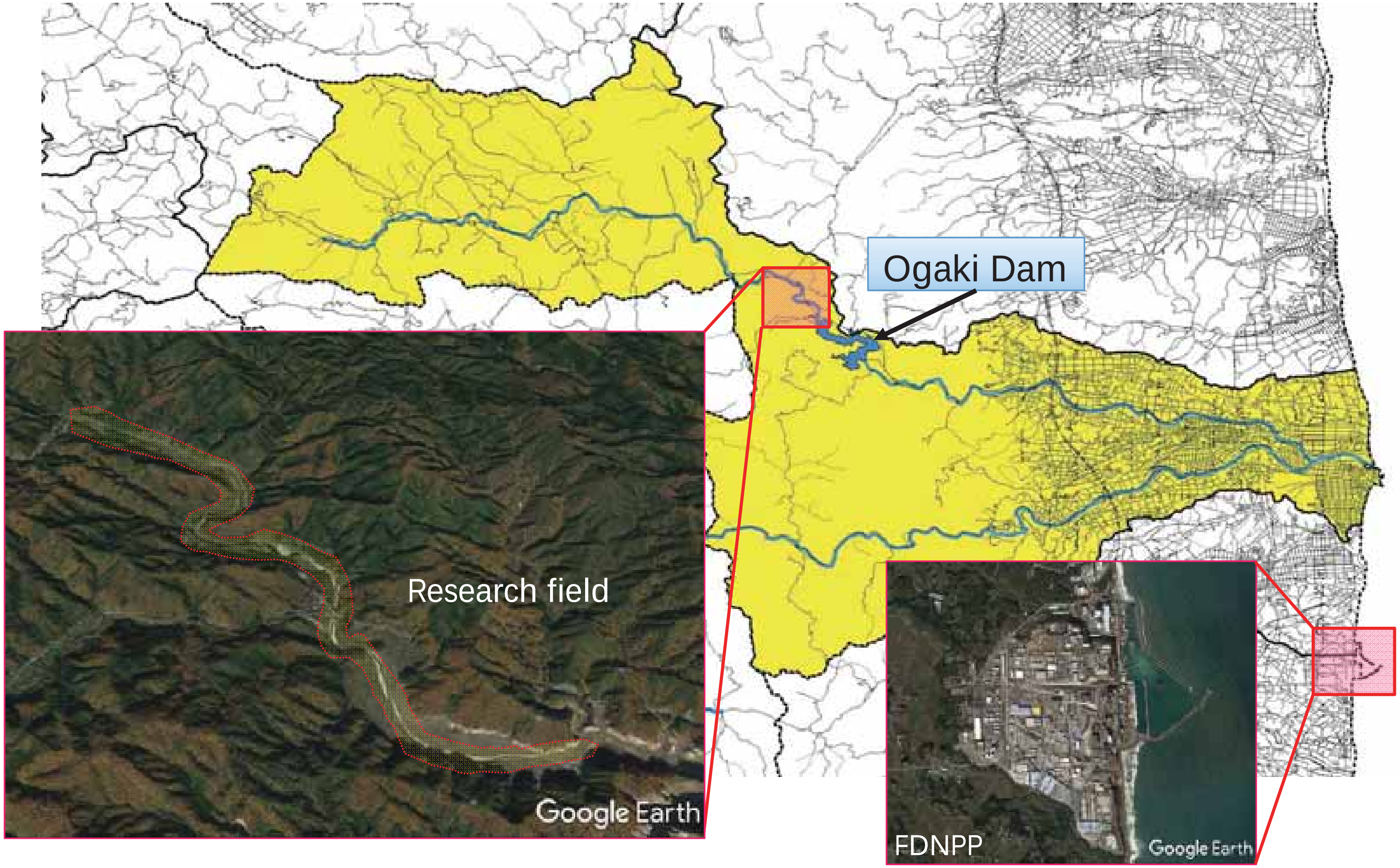
Oncorhynchus masou masou

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

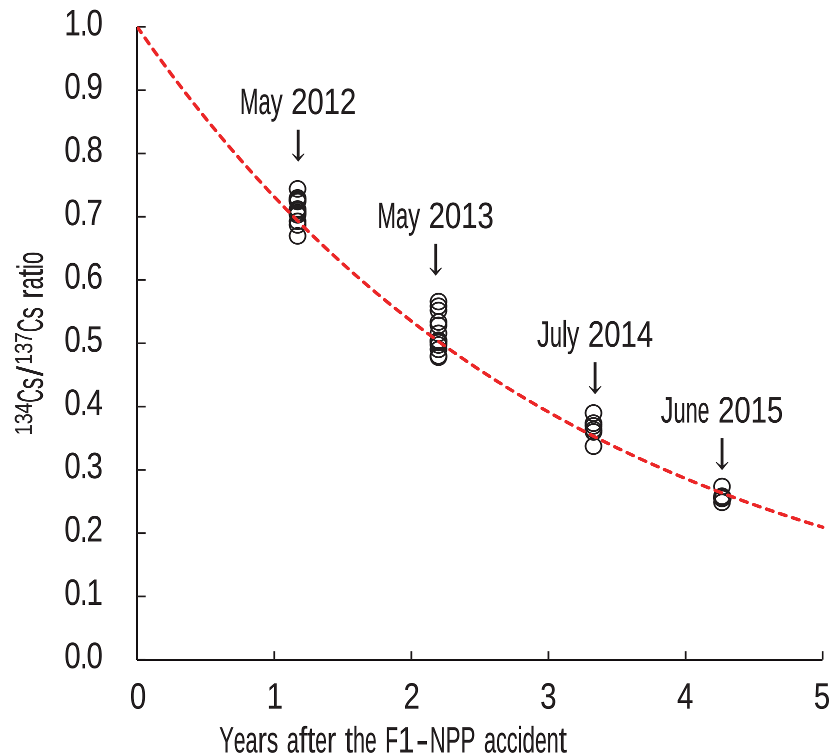
Aircraft monitoring results
(MEXT：June 28, 2012)



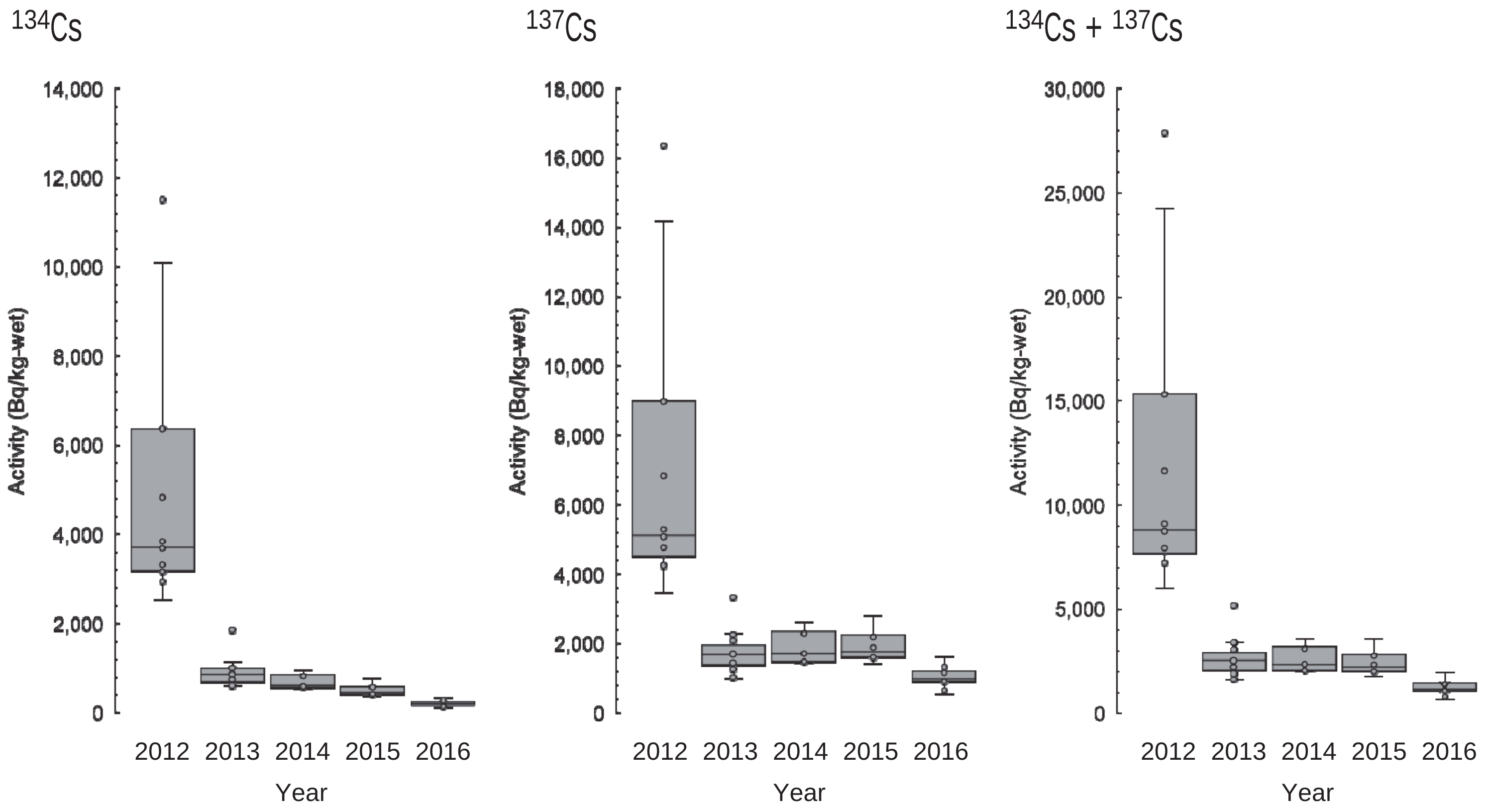
Ukedo River in Namie, Fukushima



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

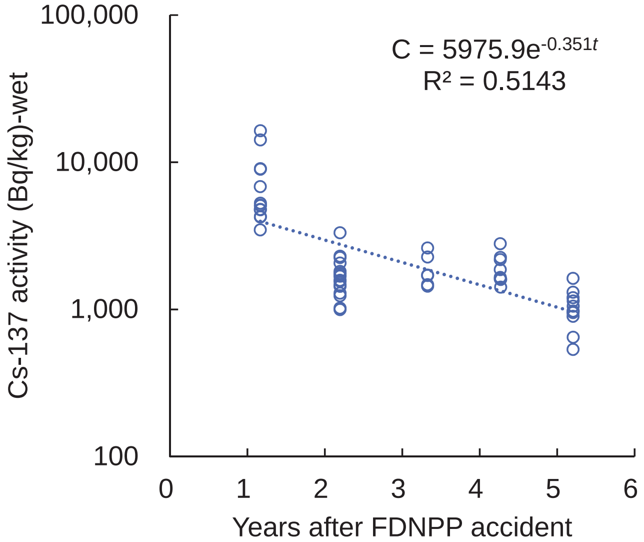


Kinetics of radioactive cesium concentration in muscle of trout captured in Ukedo River



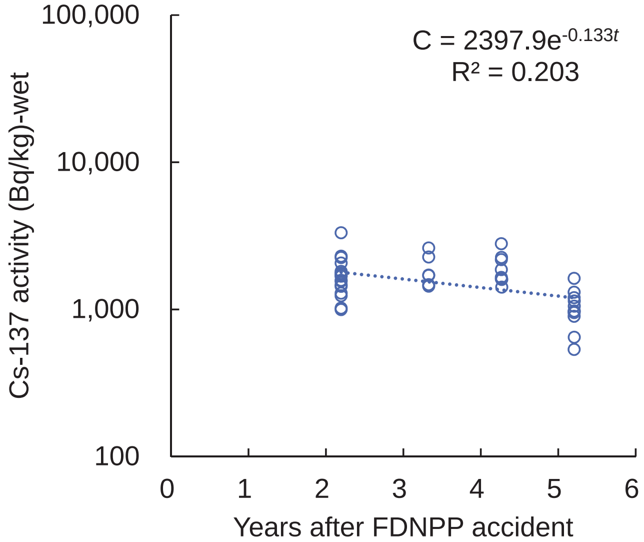
Biological half-life of ^{137}Cs

From 2012 to 2016



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

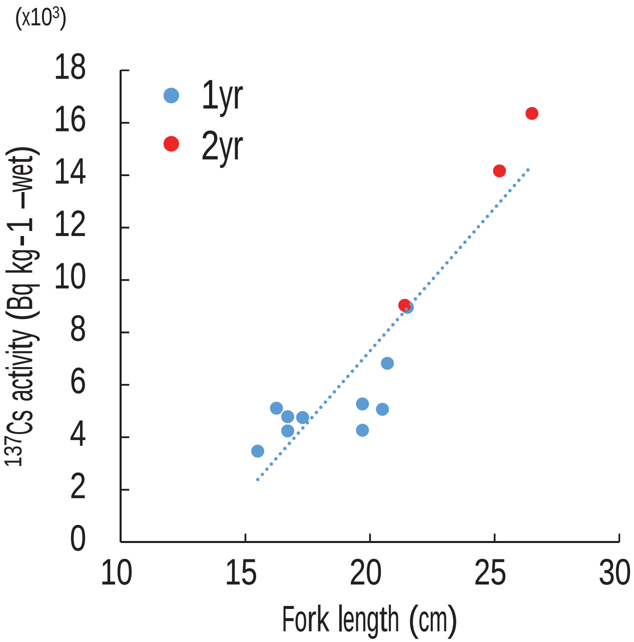
From 2013 to 2016



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

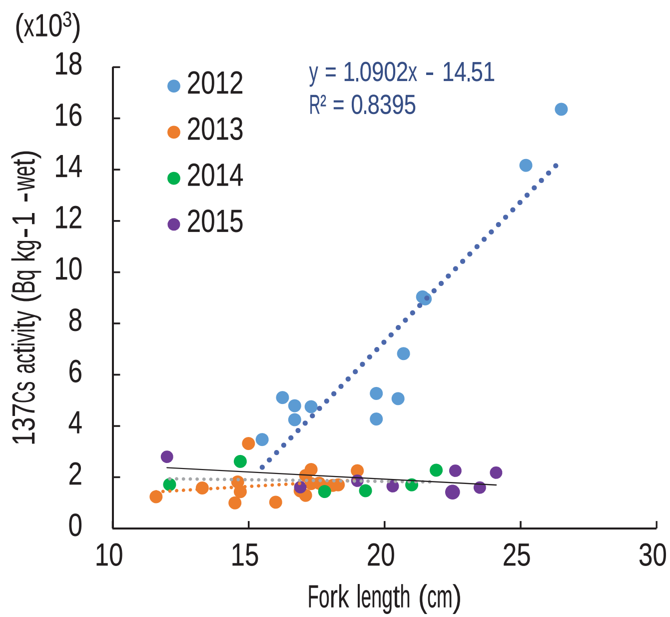
Relationship between age and radioactive cesium concentration in trout captured at the Ukedo River in 2012

Omm13
circuli on the scale: 38
annuli on the scale: 2 (2 year-old)

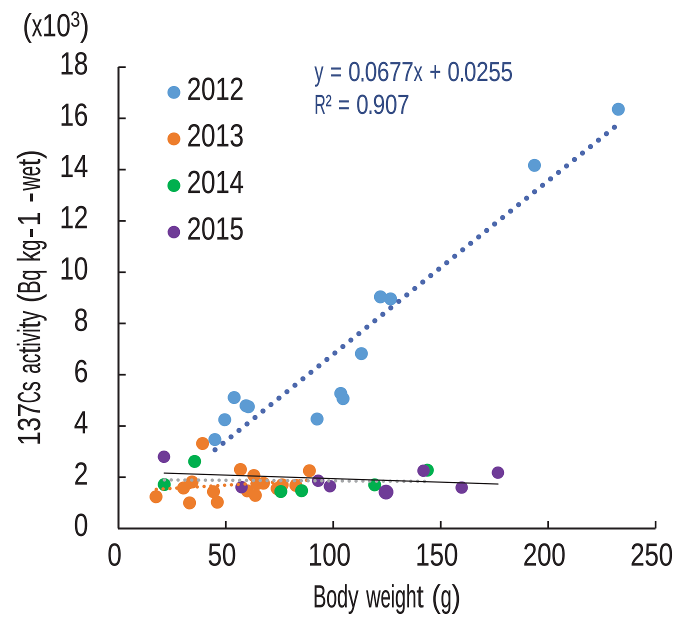


Comparison of radioactive cesium concentration in muscle with body length (fork length) and body weight

Fork length vs. ^{137}Cs



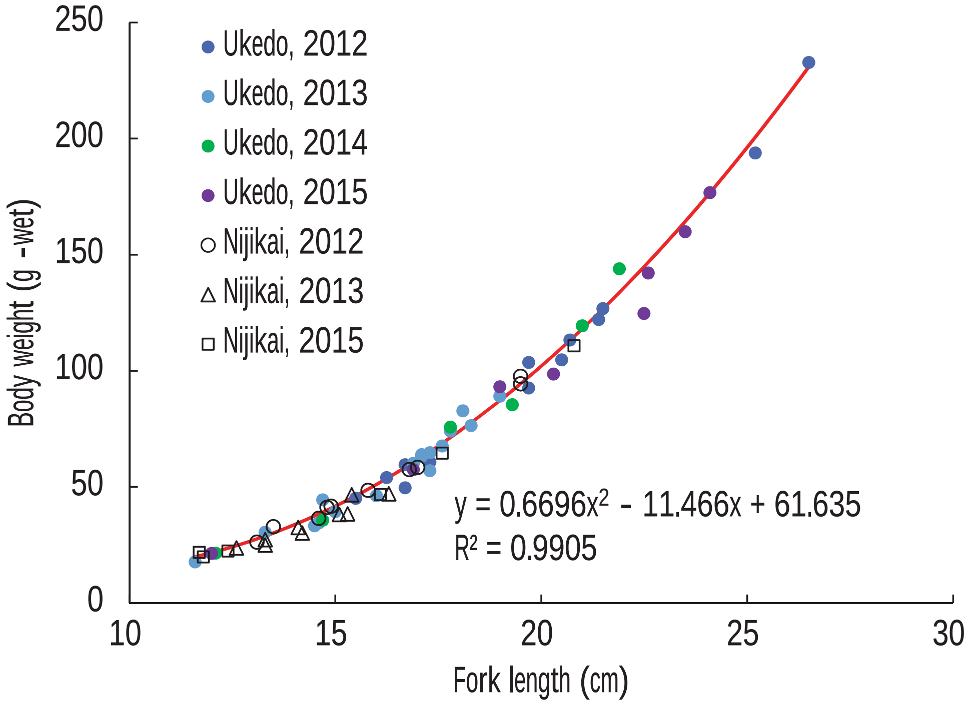
Body weight vs. ^{137}Cs



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

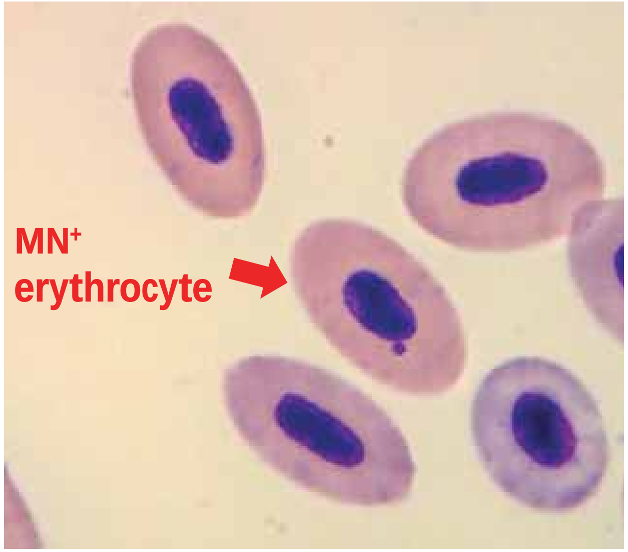
River	Month/Year	n	a	b ± SE	r^2	95% CL of b	Growth pattern
Ukedo	May 2012	13	0.00000760	3.092 ± 0.0991	0.9888	2.873–3.310	Isometric
	May 2013	17	0.00000376	3.233 ± 0.1451	0.9707	2.924–3.543	Isometric
	July 2014	6	0.00000434	3.205 ± 0.1354	0.9929	2.829–3.581	Isometric
	June 2015	8	0.00001429	2.970 ± 0.1073	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

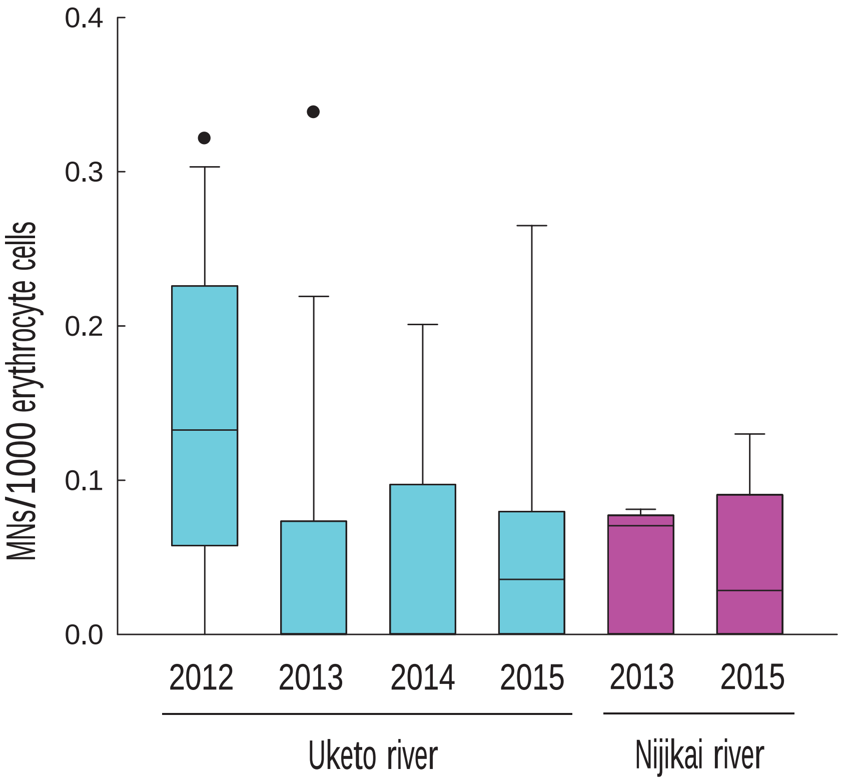
The growth of trout captured in the Ukedo River and Nijikai River during from 2012 to 2015



Kinetics of micronucleus frequency in trout erythrocyte cells

Giemsa-stained blood cell of trout

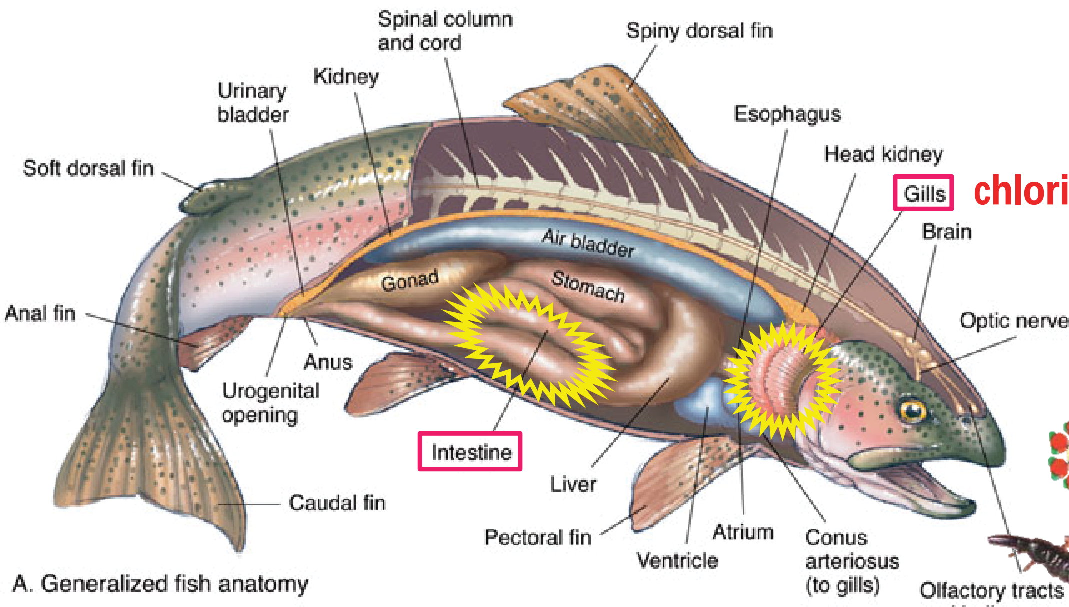




Uptake route of radioactive cesium into the body

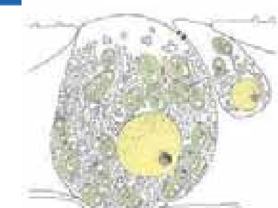
Radioactive cesium absorbed to suspended particles is low bioavailability

Uptake route of radioactive cesium assumed



http://caliskan-ogrenciler.blogcu.com/fish-anatomy-lab11/9752916

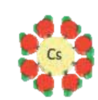
chloride cell



http://proposal.ducr.u-tokyo.ac.jp/ccr_usr/data/image/org/05866/05866_02.jpg

Route 1

Dissolved radioactive cesium



Route 2

Feed (aquatic insect etc.)

Transfer pathway of radioactive cesium


Fallen leaves


Sedimentary organic

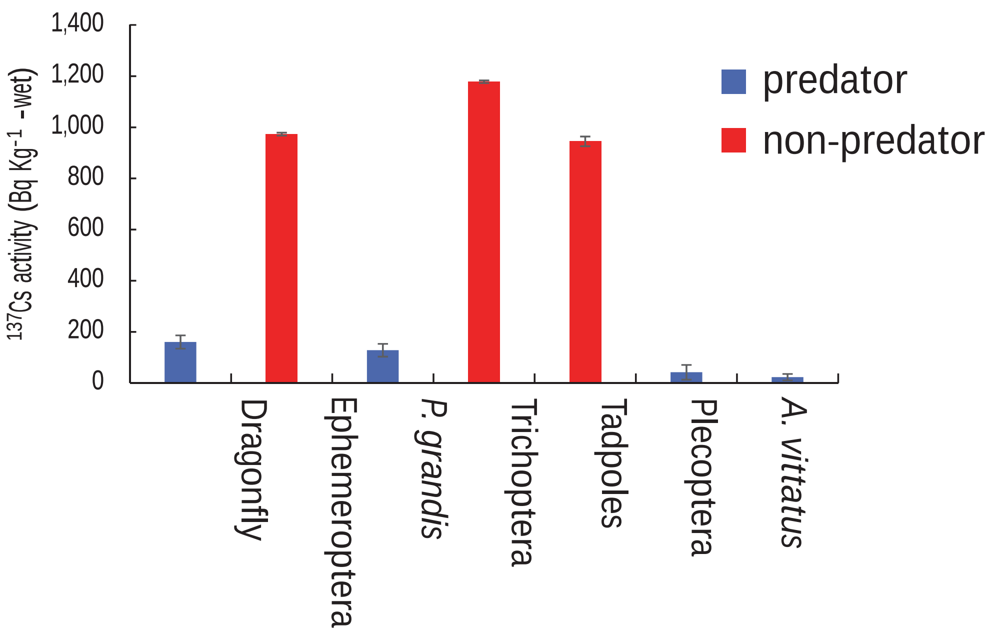

aquatic insect

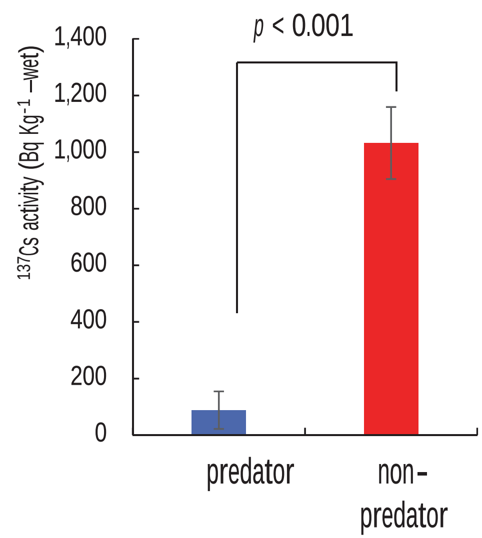

Attached algae


Stomach content

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

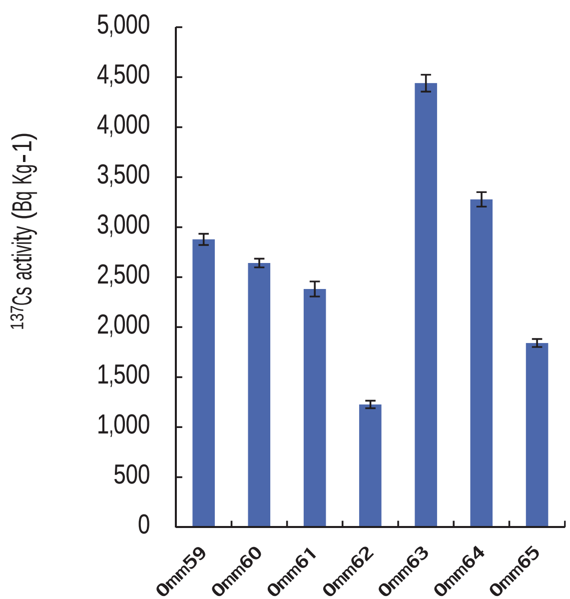
Dragonfly | Ephemeroptera | *Protohermes grandis* | Trichoptera | Tadpoles | Plecoptera | *Aphelocheirus vittatus*



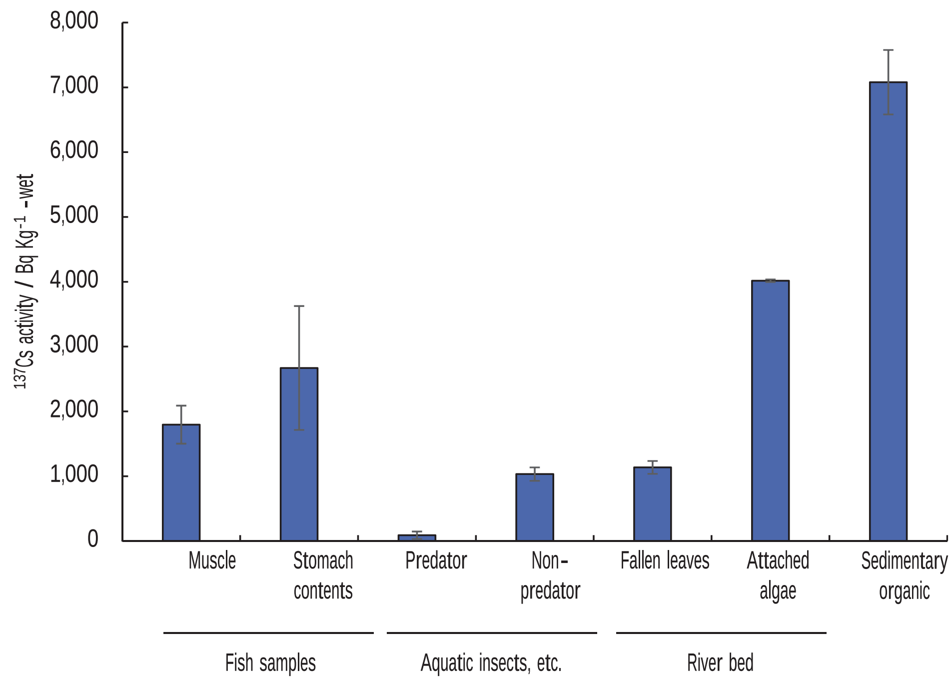


Comparison of radioactive cesium concentration of fish sample and environmental sample in 2015

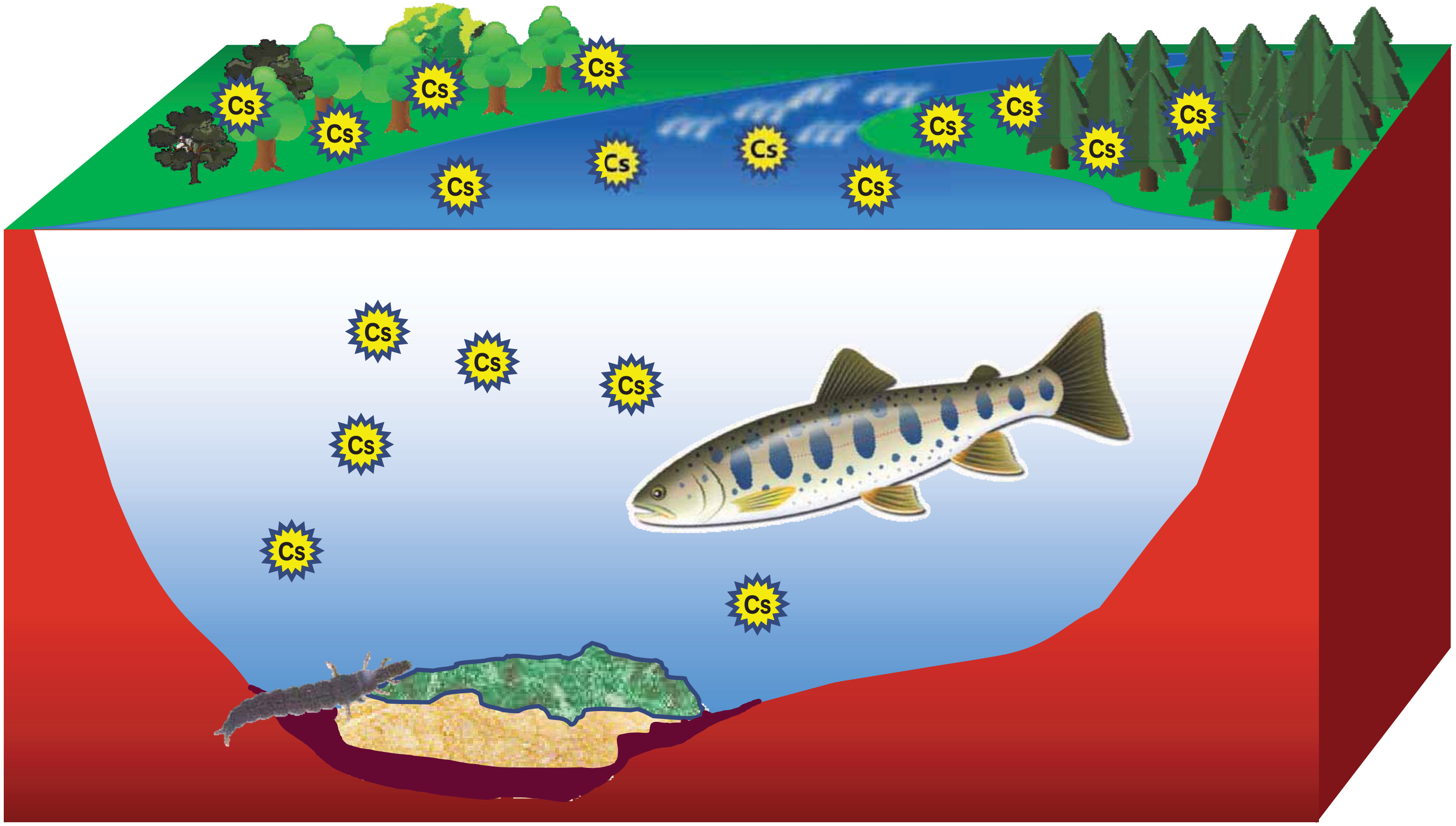
Stomach contents



Comparison of ^{137}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***
  Dr. Kosuke Kasai, Mr. Yohei Fujishima, Mr. Hiroki Ueno, Morikawa Chise
- ***Hirosaki University, Institute of Radiation Emergency medicine***
  Prof. Mitsuaki A. Yoshida, Dr. Kentaro Ariyoshi
  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)

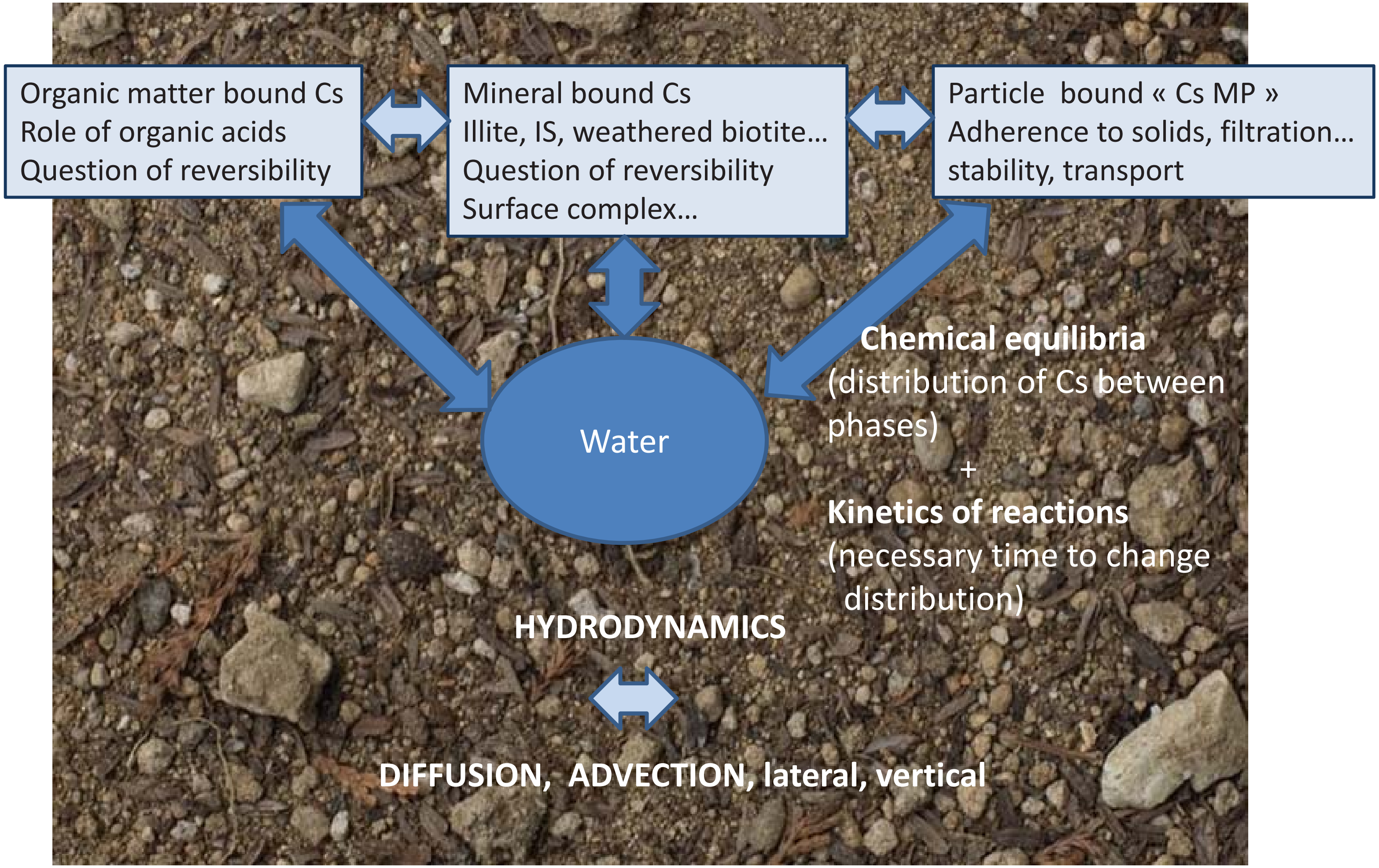
** IAEA Advanced Science Research Center

Interaction Cs-soil

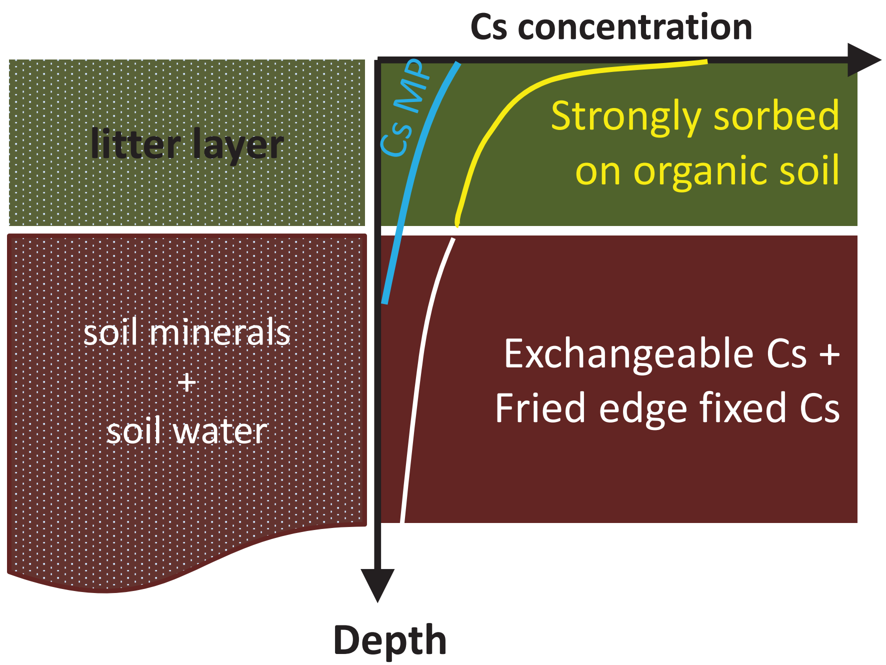
- Sorption of radiocesium [1, 2]:
  - Fixed on "frayed edge" sites FES of mica type minerals :
    - Weak or no desorption ("fixed fraction")
  - Uniform sorption on weathered biotite
  - Sorption of Cs in form of microparticles??
  - Impact of litter/organic matter
- Migration in depth:
  - As a consequence, 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 100 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 environnement depend on its chemical form



Diverse interactions of Cs with soils

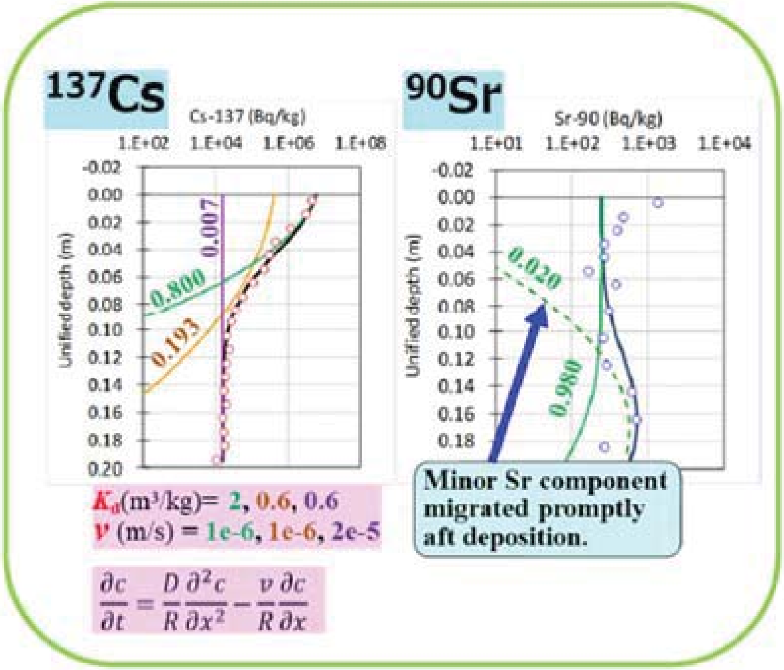


Analytics of stable Cs and Cs 137

- 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**



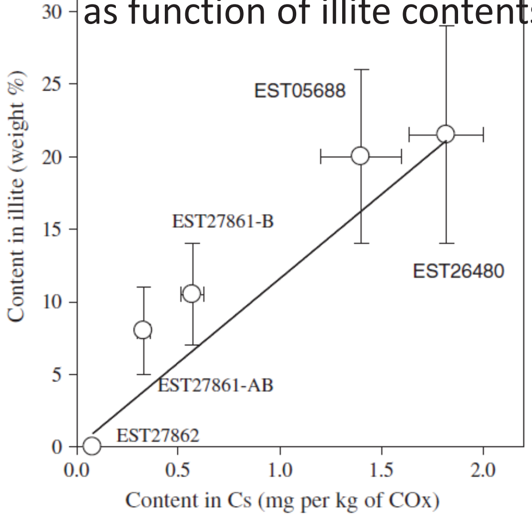
$$\frac{\partial c}{\partial t} = \frac{D}{R}\frac{\partial^2 c}{\partial x^2} - \frac{v}{R}\frac{\partial c}{\partial x}$$

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

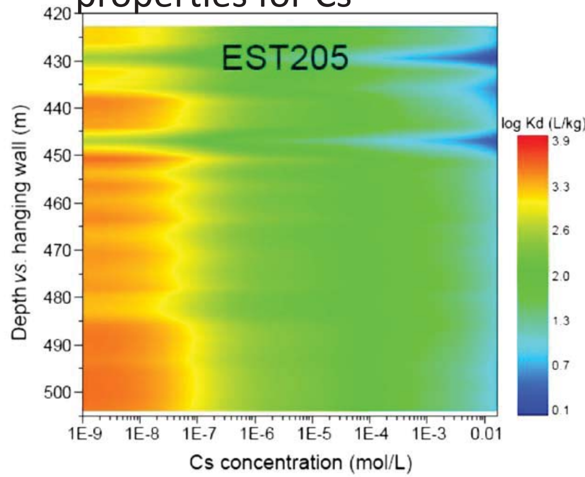
- variation in soil composition
- Organic/mineral distribution
- temporal evolution,
- Fraction of irreversible fixation of Cs etc:

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

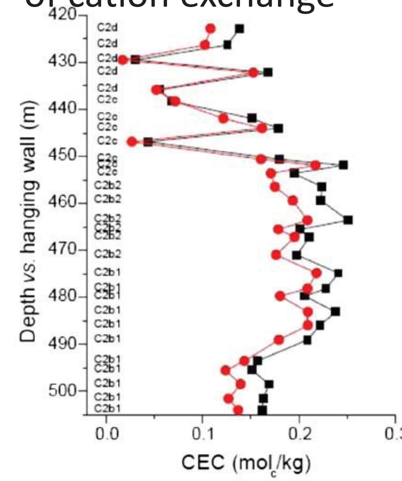
Natural Cs content in clay rock as function of illite contents



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
		NH_4/Na	3.5
Type II	Illite	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	Cs/Na	1.7
		K/Na	1.1
		Ca/Na	0.6
		Mg/Na	0.6
		Sr/Na	0.3

Parameters for weathered biotite missing:
Collaboration U Hokkaido

Chemical Geology 387 (2014) 47–58

Contents lists available at ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

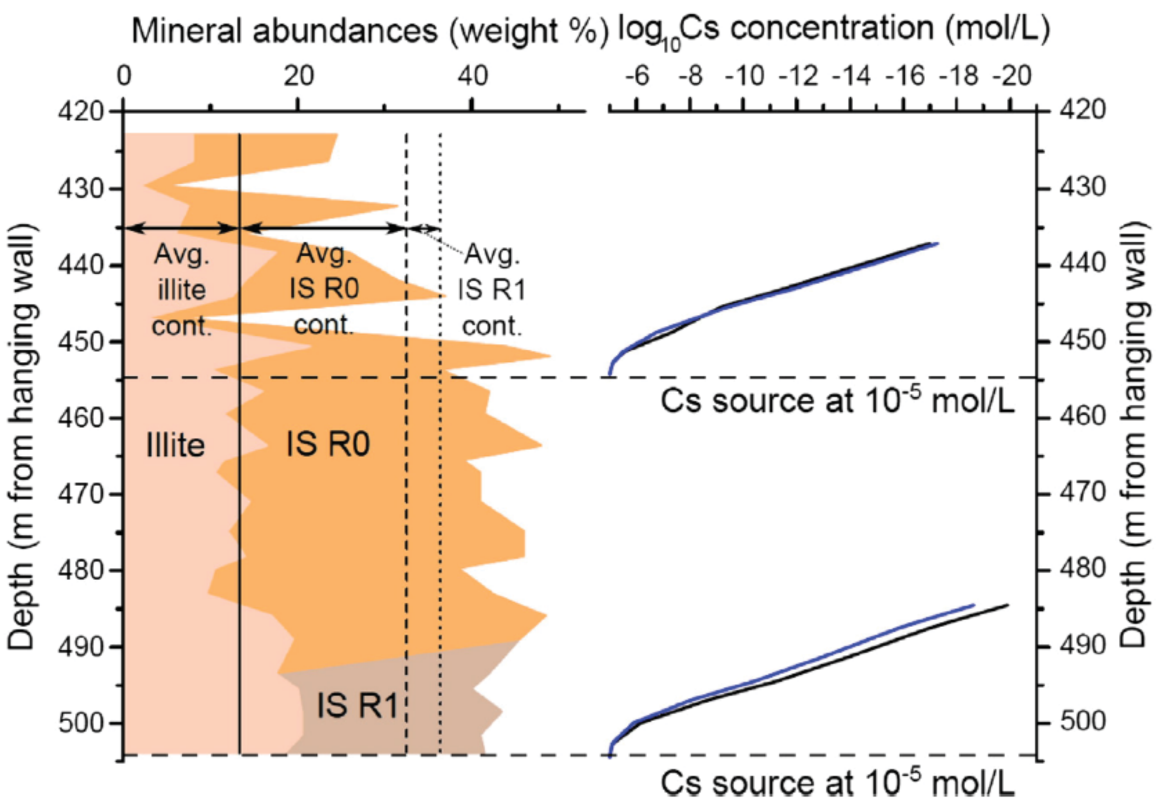
ELSEVIER

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

Z. Chen [a,b], G. Montavon [a,*], S. Ribet [a], Z. Guo [b], J.C. Robinet [c], K. David [a], C. Tournassat [d], B. Grambow [a], C. Landesman [a]

Predicting Cs transport by diffusion, considering mineralogical heterogeneity

Example: Reactive transport calculation with large depth in clayrock

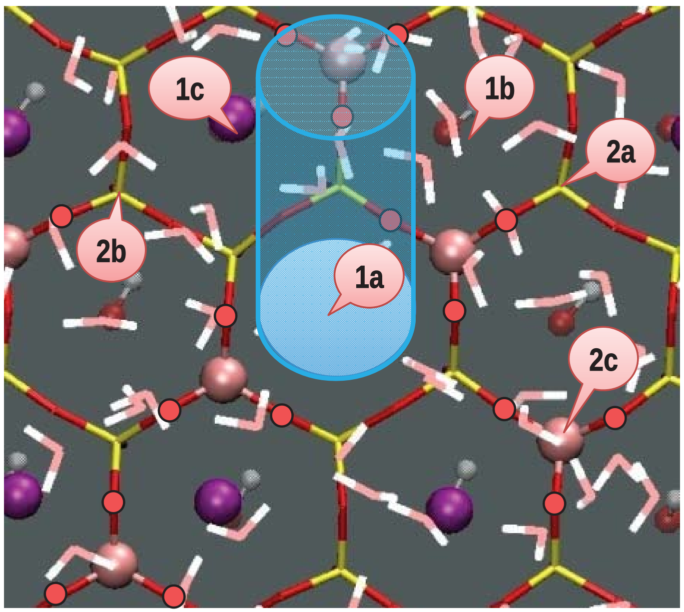


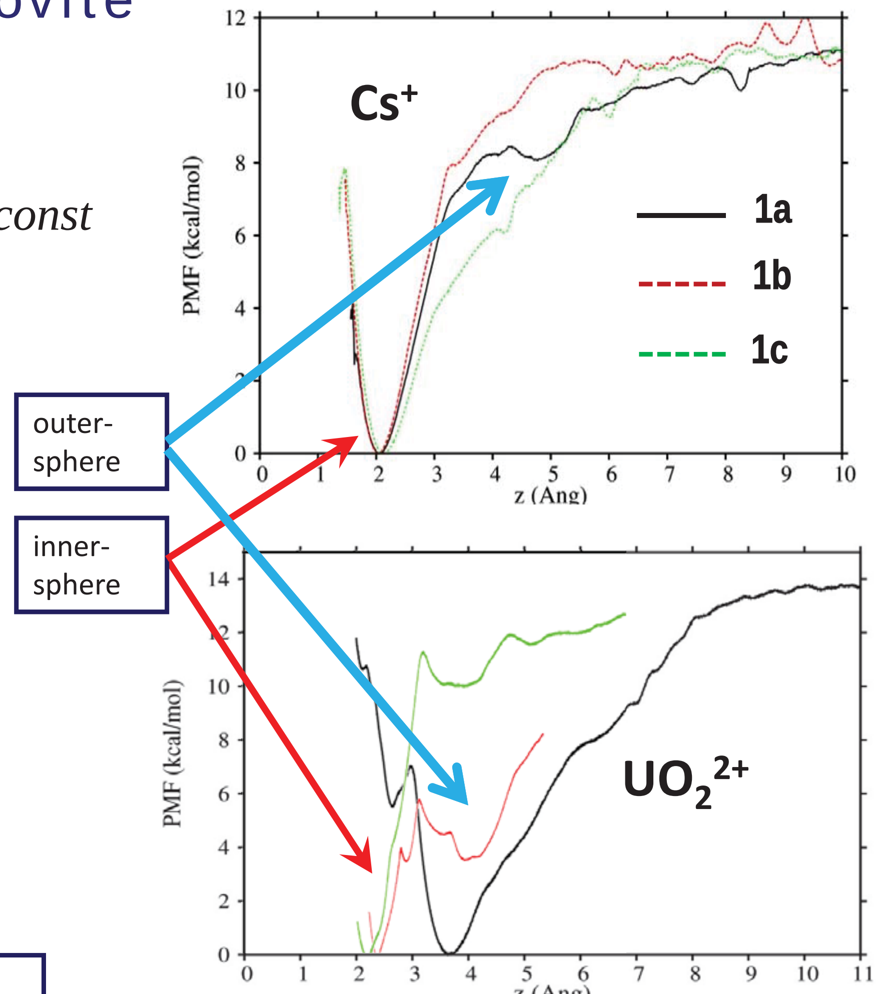
Molecular modelling: Surface Adsorption Sites for Cs^+ and UO_2^{2+} on Muscovite

Potential of Mean Force:

$$\mathrm{PMF} \equiv W(z) = -k_{\mathrm{B}}T \ln\big(\rho(z)\big) + const$$

z - distance from the surface





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

What is the time dependency of desorption of Cs?

The desorption rate depends on soil type

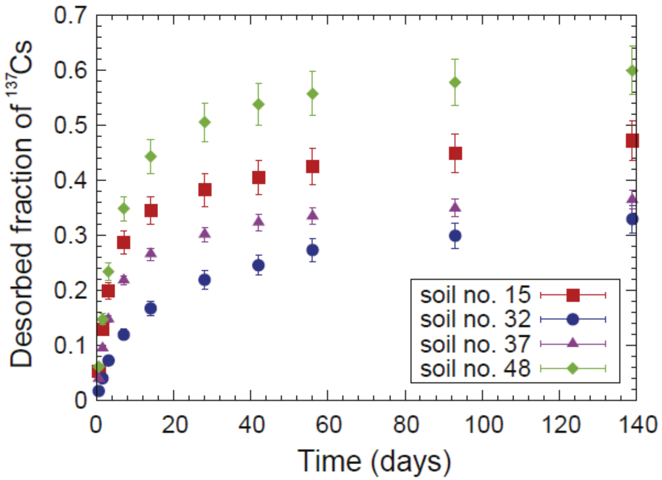


Fig. 1. Fractions of the cumulative desorption of ^{137}Cs from the four different soil samples (soil no. 15, 32, 37, and 48) by 10^{-3} M KCl solution in the presence of cation exchange resin. The amounts of the soils were 5 g.

Behavior can be modeled

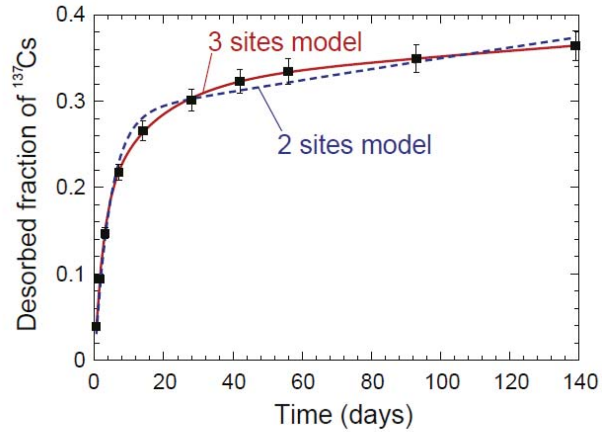


Fig. 6. Fitting results of the three- (continuous line) and two- (dotted line) sites desorption models for the fraction of the cumulative desorption of ^{137}Cs from the soil no. 37 by 10^{-3} M KCl as a function of time shown in Fig. 3.

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.

Journal of Environmental Radioactivity 153 (2016) 134–140

Contents lists available at ScienceDirect

ELSEVIER

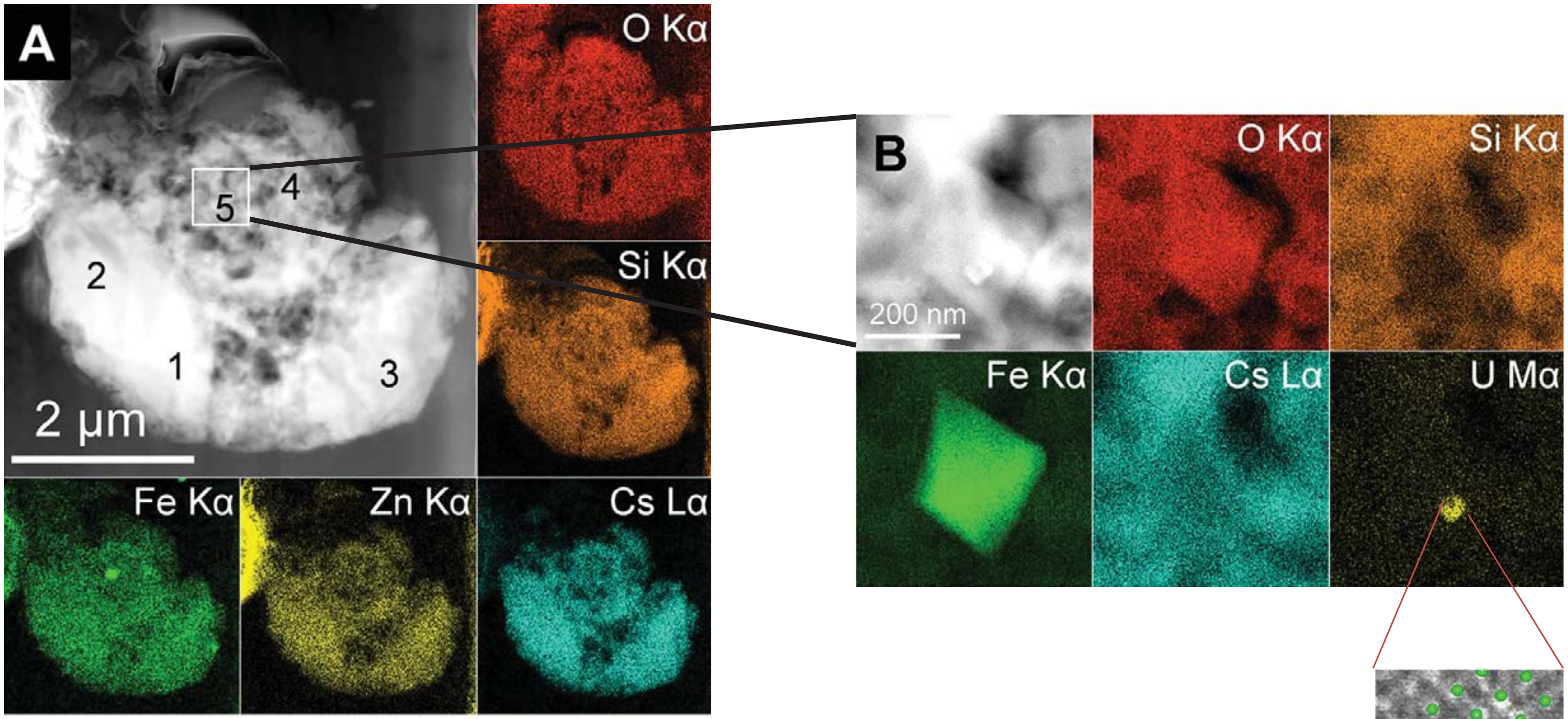
Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

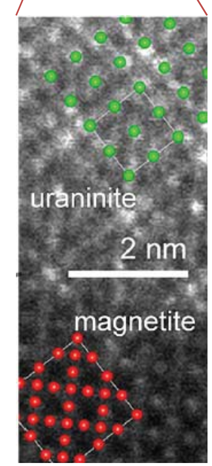
Desorption kinetics of cesium from Fukushima soils

Kento Murota [a], Takumi Saito [b, *], Satoru Tanaka [a, 1]

What about Cs in form of Cs microparticles?

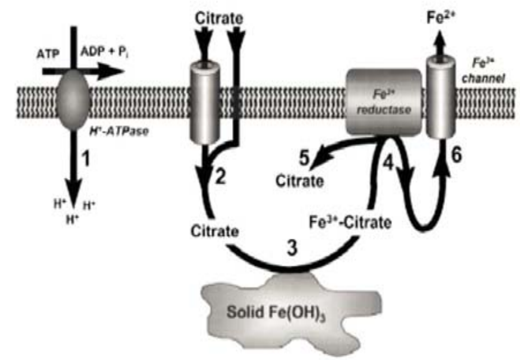


- Which fraction of Cs is in particulate form at which geographical position?
- Does the fraction of Cs in particulate from change with time?
- What is the impact on dose (the residence time of Cs MP in the human body is different)?
- What are transport and sorption properties of the particles ?
- What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



Fabiola GUIDO GARCIA

Organic acids - soil microbes interaction



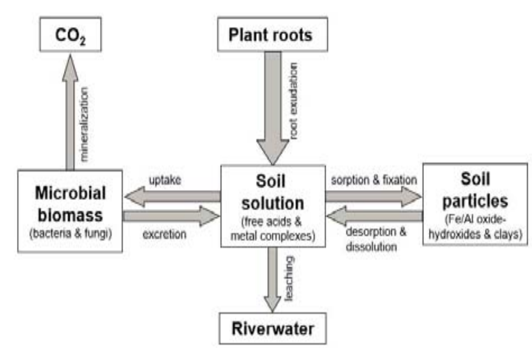
Schematic representation of the role of organic acids in Fe uptake by dicotyledonous plant roots (Jones 1998).

Purpose:

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

Approach:

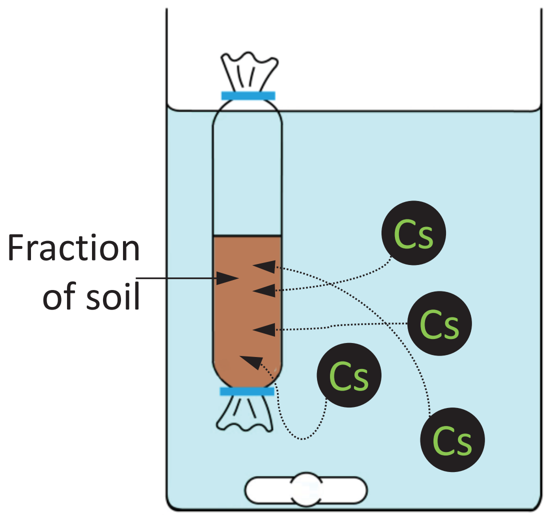
- Activation of microbes by fungi organic acid on production of siderophore.
- Synergy effect of fungi organic acids and microbes siderophores on dissolution of Cs from minerals.



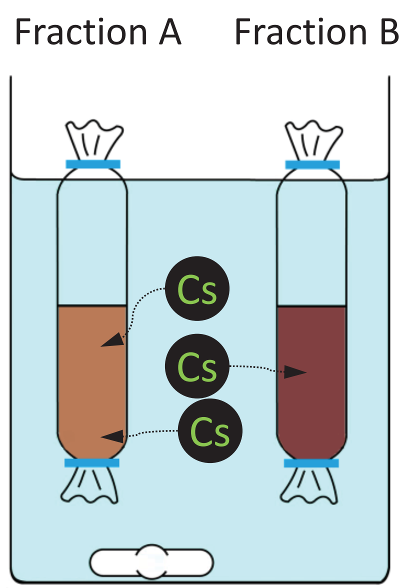
Schematic diagram of major organic acid fluxes and pools in soil (Jones1998).

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

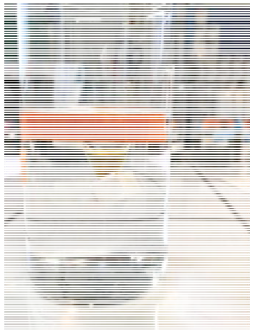
Single system

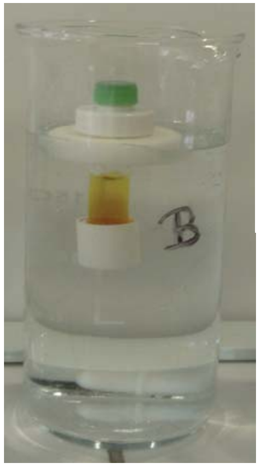


Binary system



Example of dialysis membranes

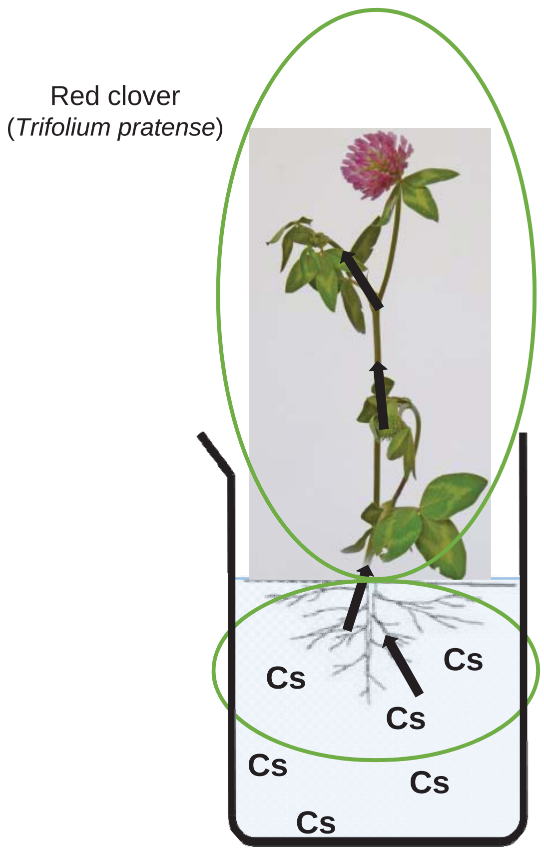




Experiments on phytoextraction of Cs from soil

Phytoextraction of Cs with red clover

Hydroponic system



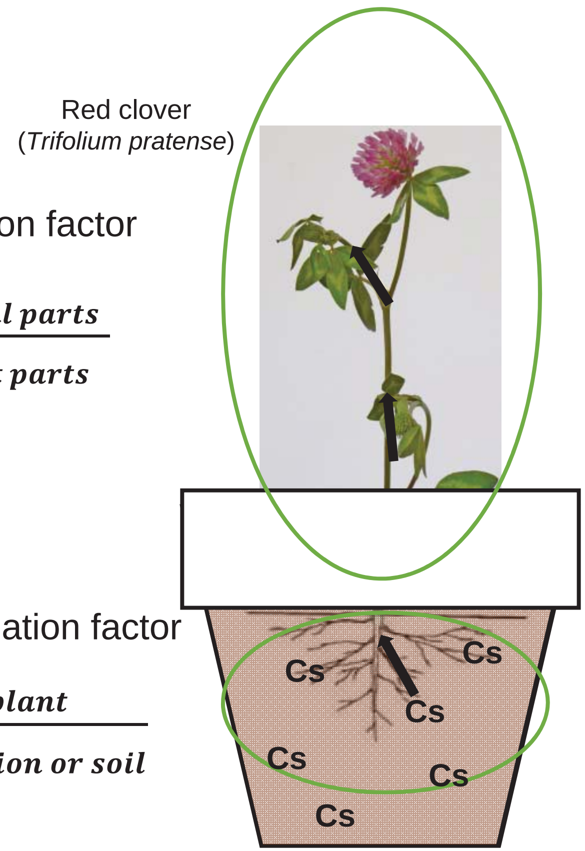
TF : translocation factor

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

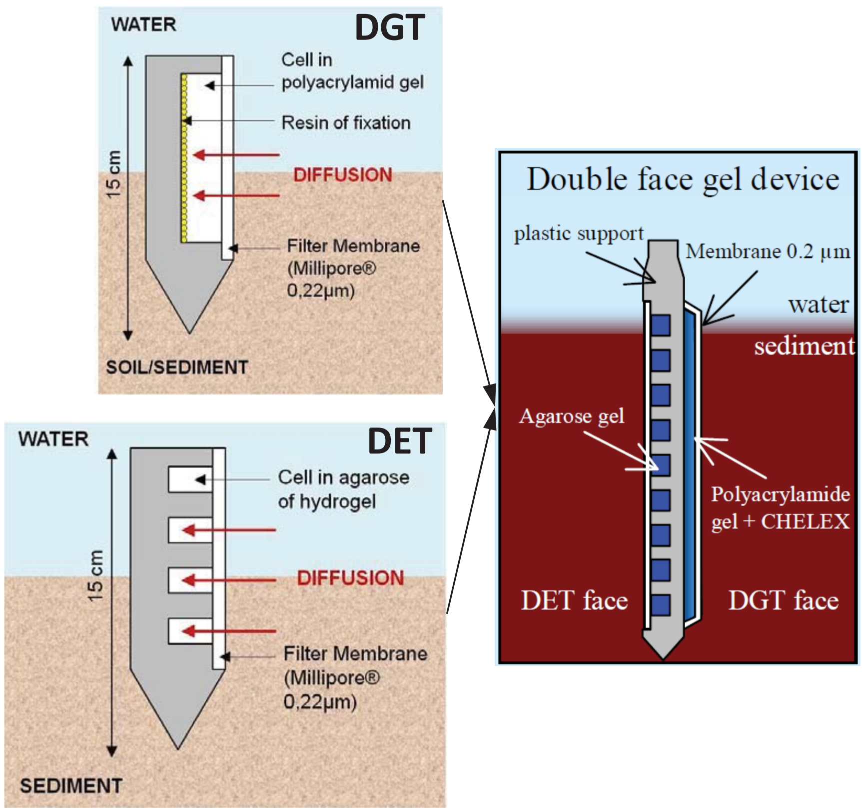
BF : bioaccumulation factor

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

Soil pots



Experiments for monitoring Cs in sediment using DGT and DET

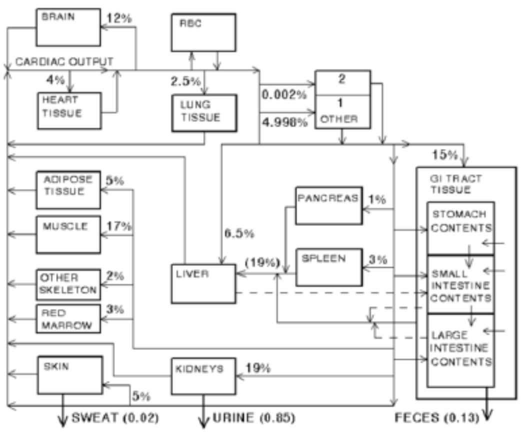


- In situ method
- Implementation time:
  - few hours
- Access to double information
  - DET: measurement of the concentration in interstitial water
  - DGT: measurement of the concentration of labile species

Metzger E. et al., EMEC3: 11-14 december 2002, Geneva

Missing link between assessment of contamination and health

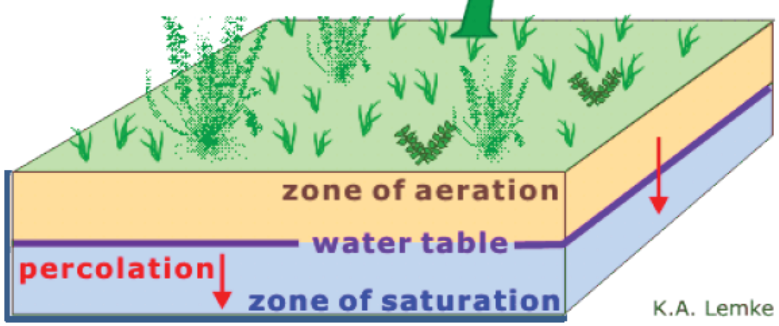
In Fukushima, whole body counters show 10 times lower Cs in humans than expected from equal soil contamination in chernobyl



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

Need for mechanistic molecular understanding

Knowledge in this area is usefull for many scientfic fields



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,