陸域環境における放射性セシウムの挙動と存在形態

Behavior of Radiocesium in a Terrestrial Environment and Its Physicochemical fractions

Institute of Environmental radioactivity: Hirofumi Tsukada
塚田　祥文

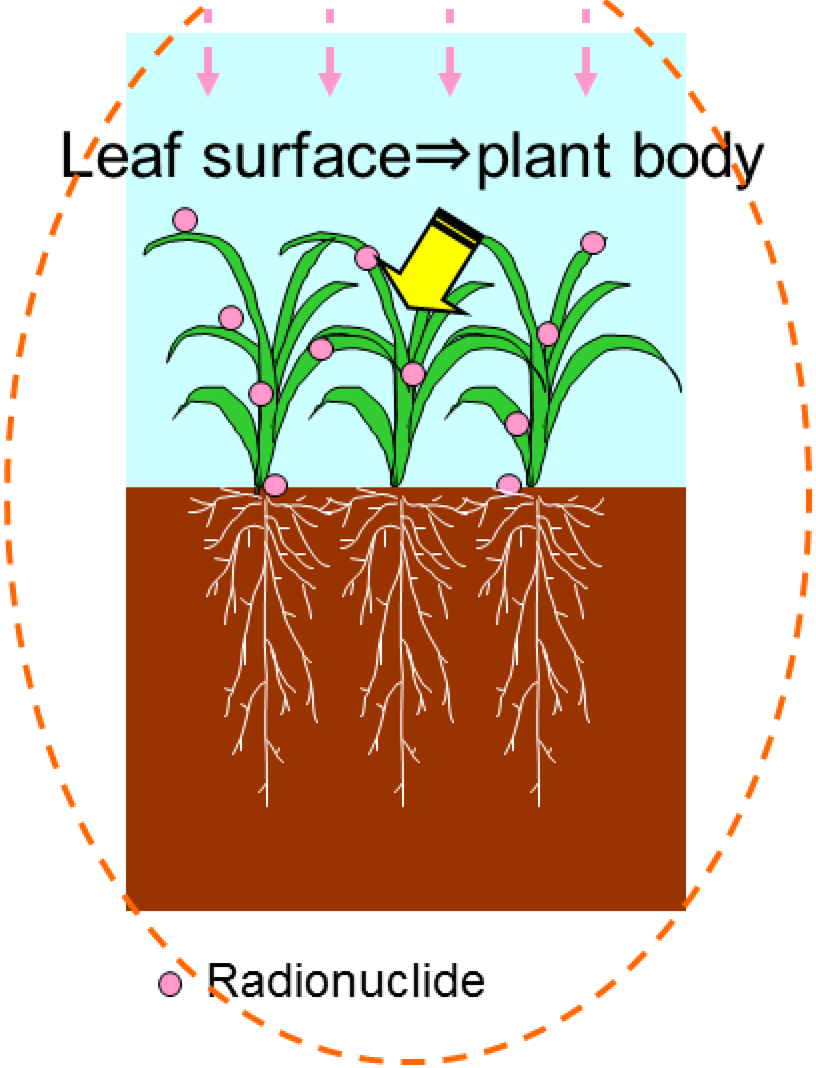
Why information on the physicochemical fractions of radionuclides is needed?

⇩

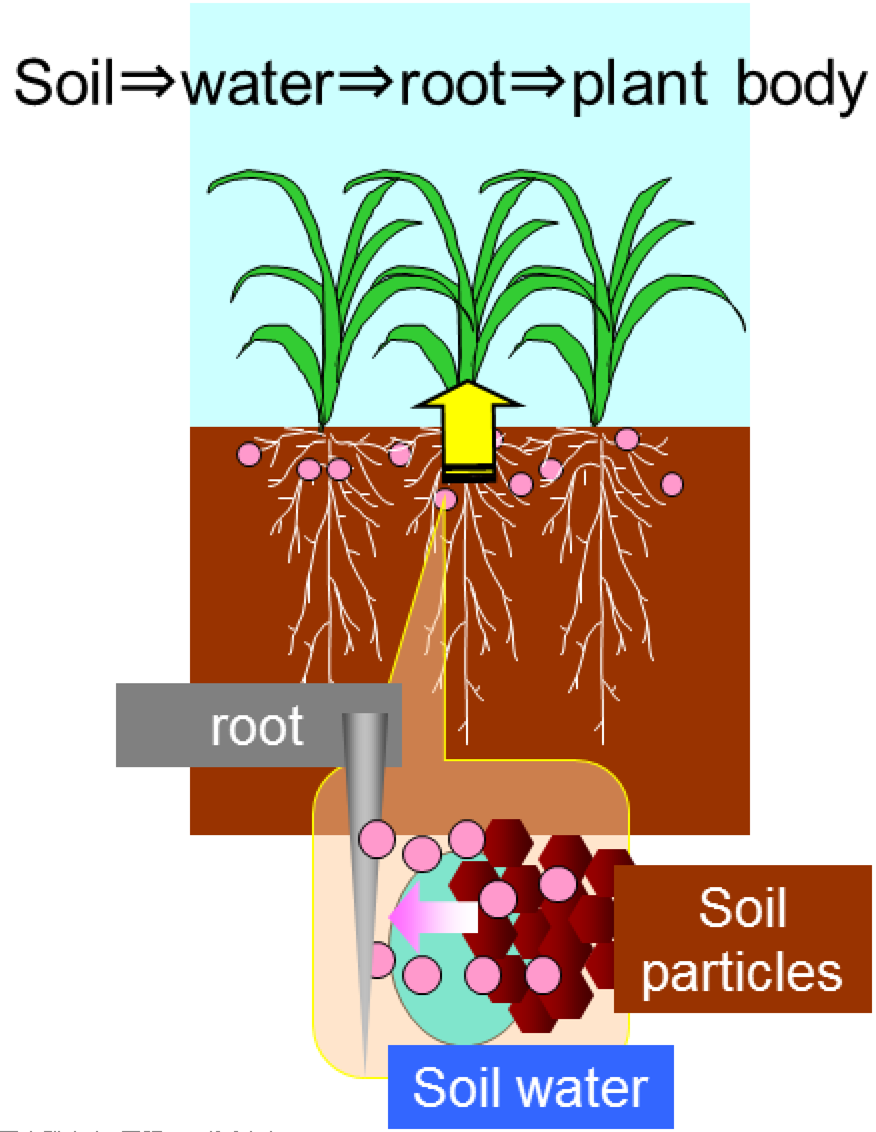
Because the migration of radionuclides in the environment was depended on their physicochemical forms

Transfer processes of radionuclides in plants

Leaf-to-plant (foliar uptake)

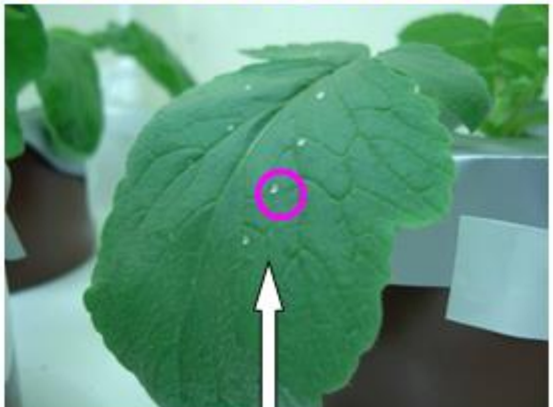


Soil-to-plant (**root uptake**)

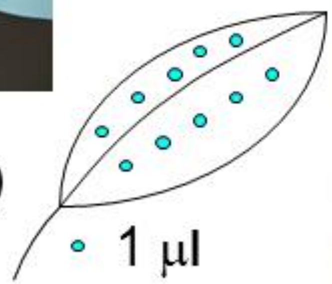


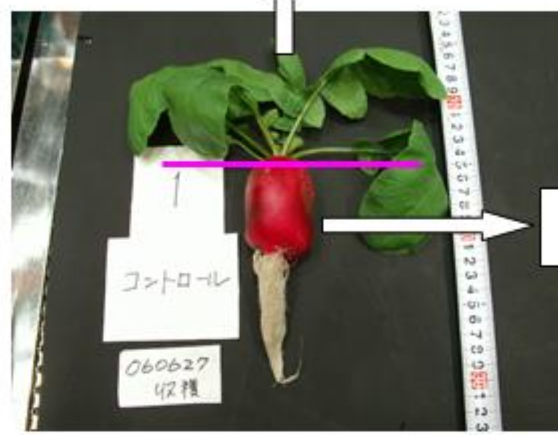
Experiment of foliar uptake

Cs solution was applied to the surface of radish leaves ⇒ Radish was harvested 4 days after the application



Solution drop 1μl (Cs)





Wash with pure water → Resultant water

Leaf parts → Wash with pure water

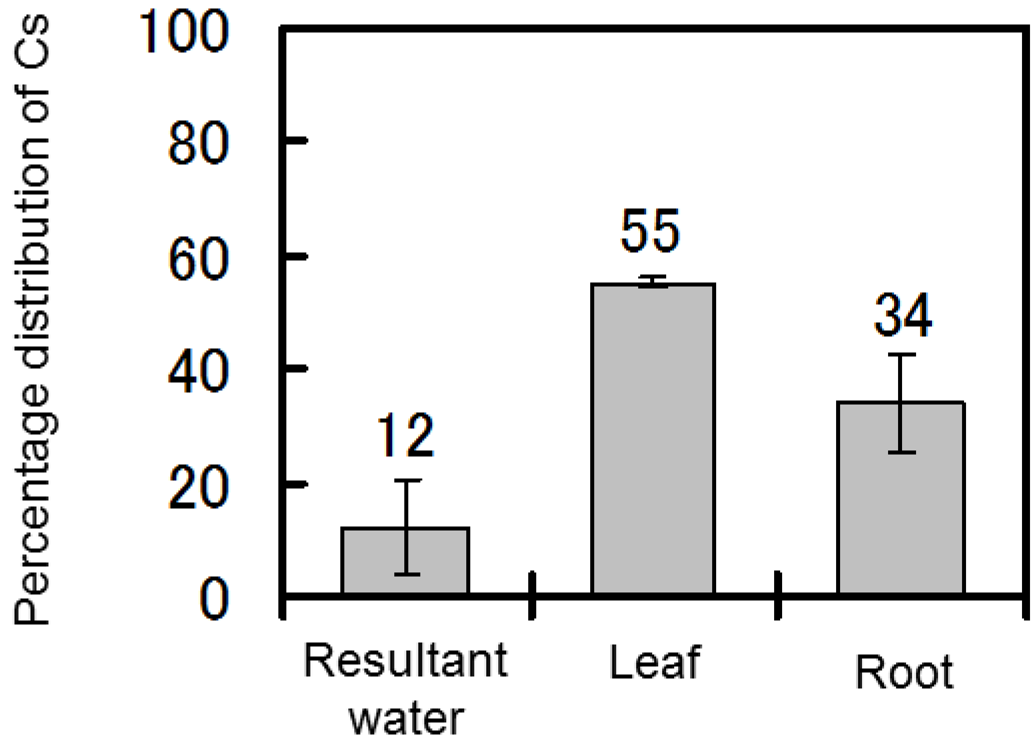
Leaf parts → Freeze-dried and then decomposed with mixed acid

Root part → Freeze-dried and then decomposed with mixed acid

Freeze-dried and then decomposed with mixed acid → Analysis of Cs with ICP-MS

Resultant water → Analysis of Cs with ICP-MS

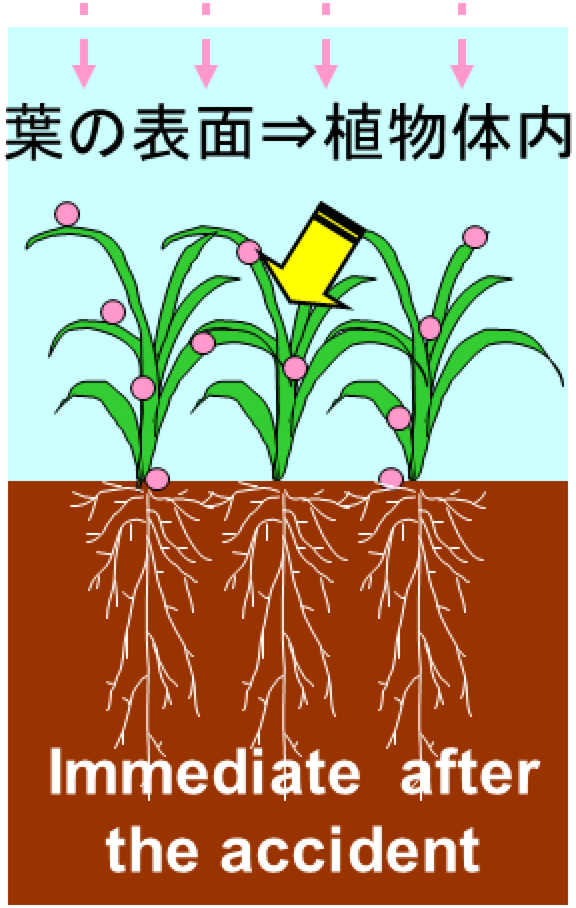
Distribution of transported Cs from leaf surface among various parts of the radish



Translocation of Cs by foliar uptake was easily moved to leaf body and root parts.

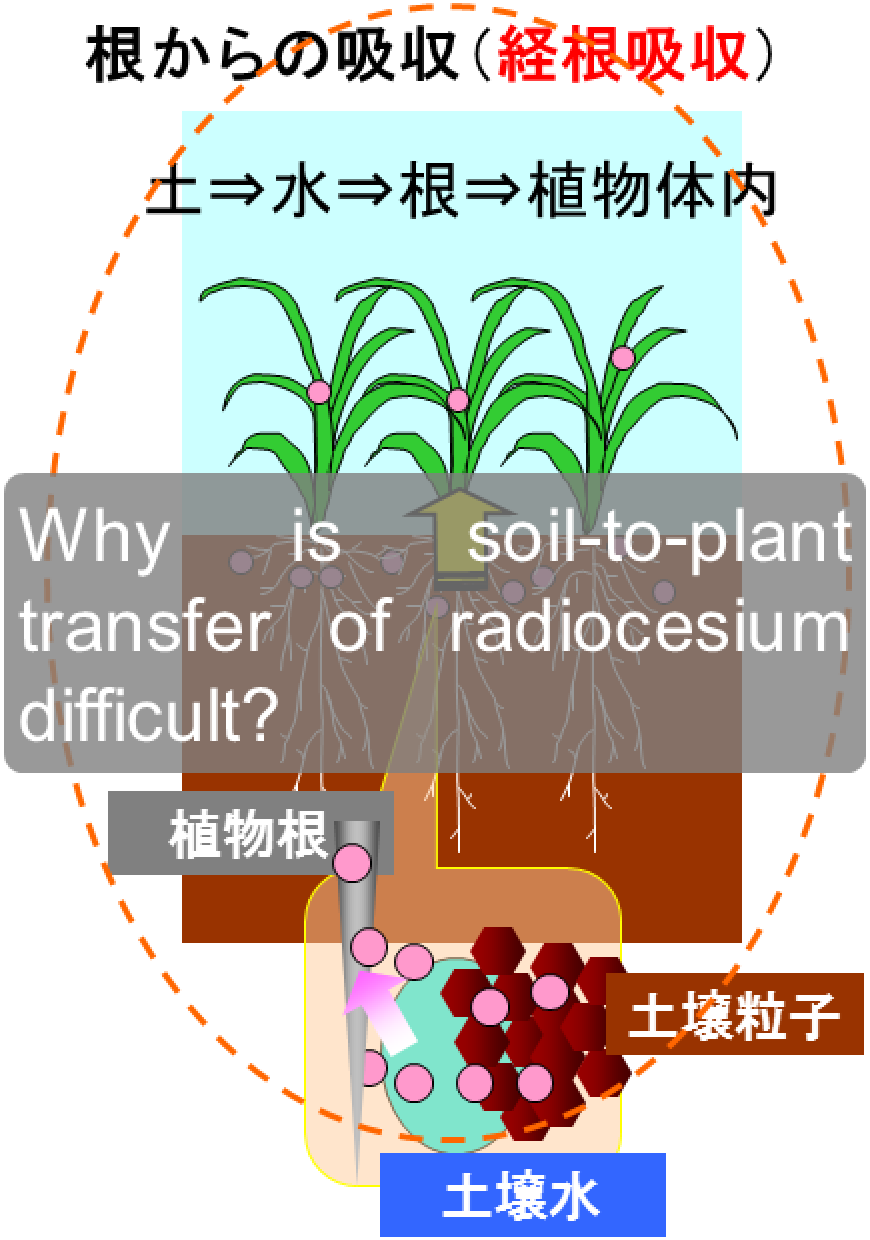
植物へ吸収される放射性核種の移行経路

葉からの吸収（葉面吸収）



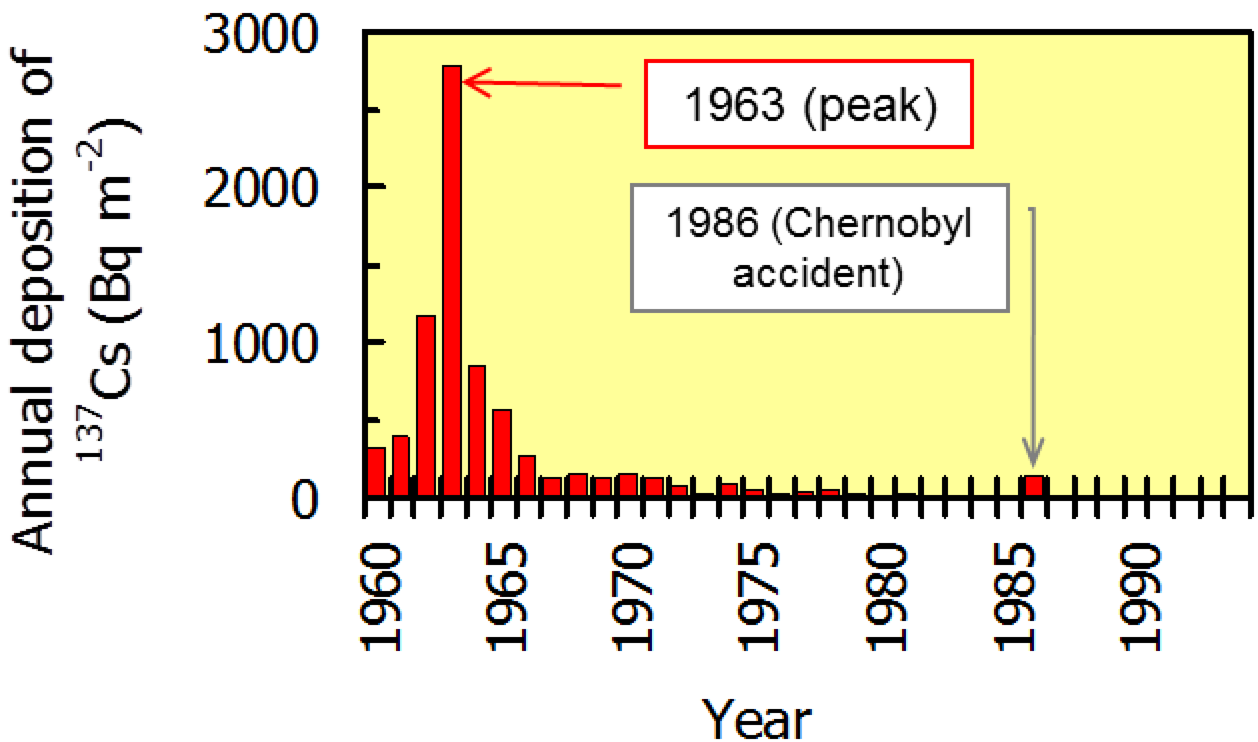
○ 放射性核種

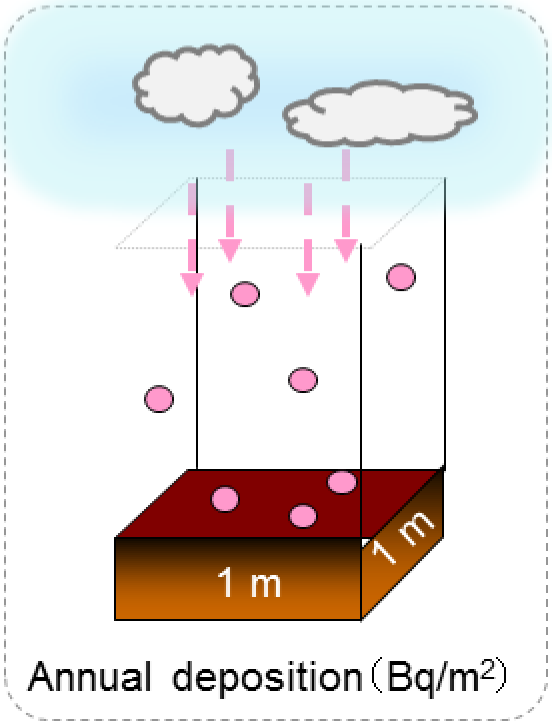
根からの吸収（経根吸収）



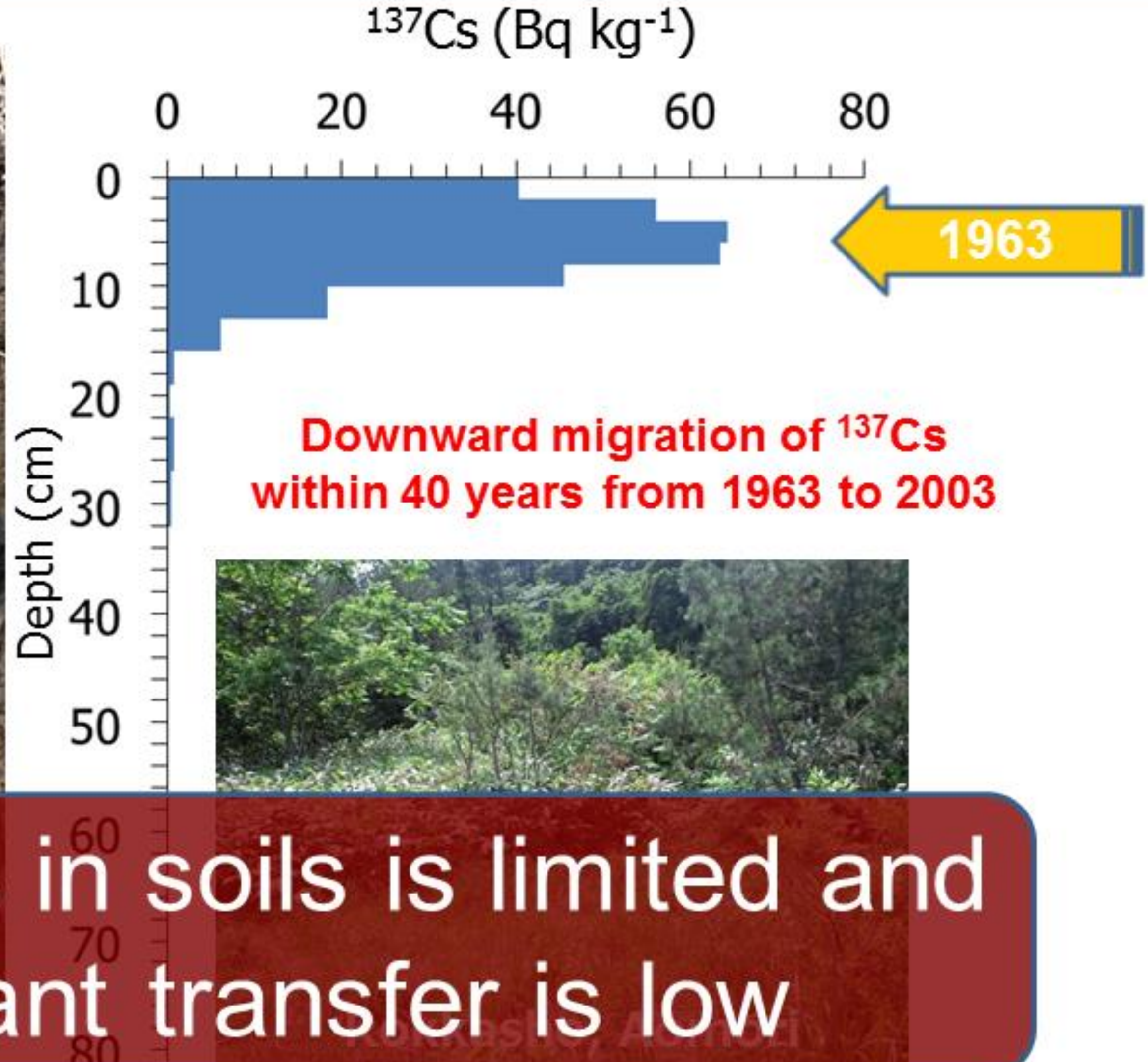
Global fallout of ^{137}Cs in Aomori, Japan from 1960 to 1994 (before the TEPCO's Fukushima Daiichi Nuclear Power Station Accident)

Derived from Atmospheric nuclear weapons tests
Mainly during 1950s and 1960s



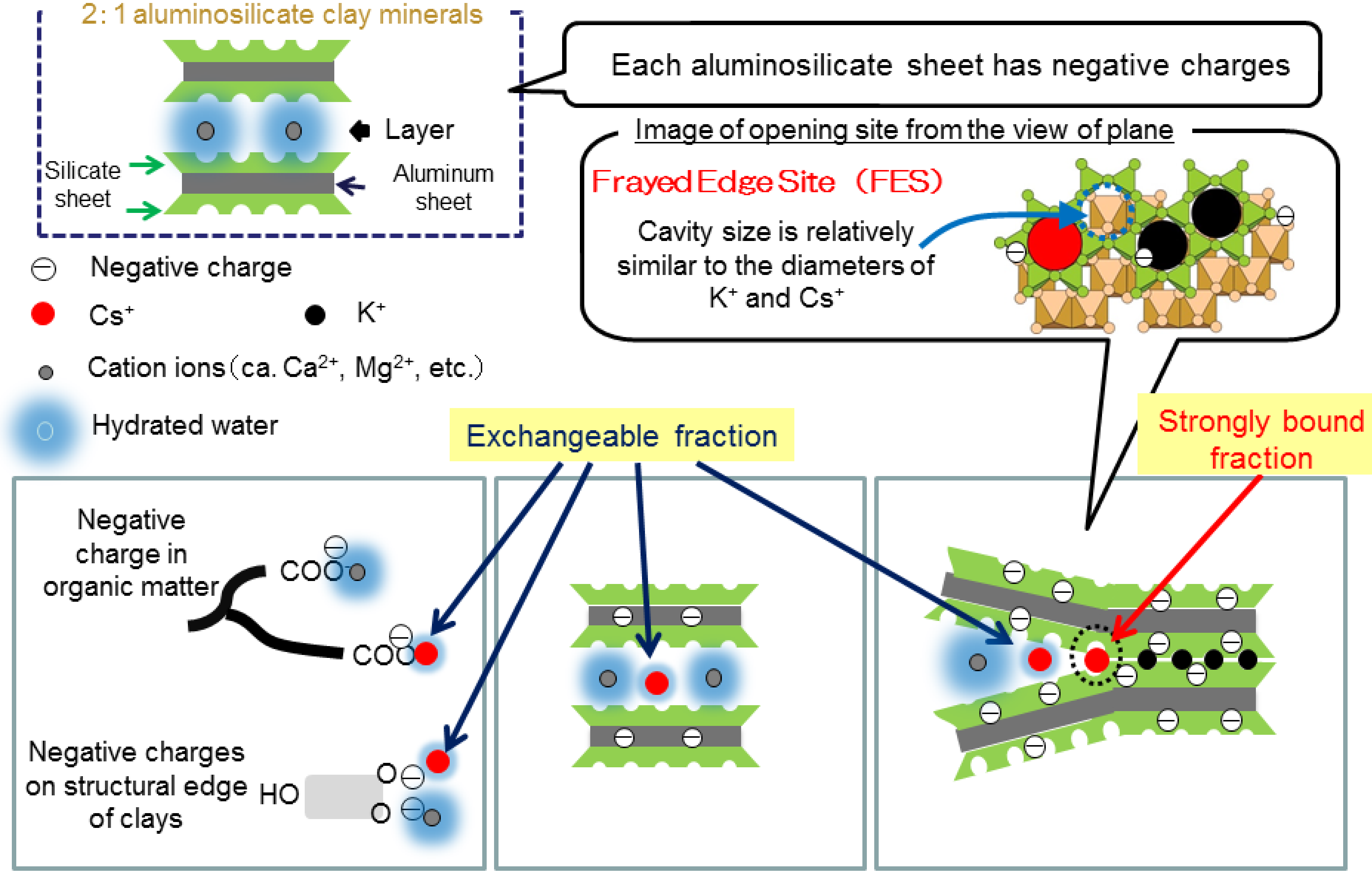


Vertical distribution of ^{137}Cs in uncultivated soil collected in 2003 (before the accident)

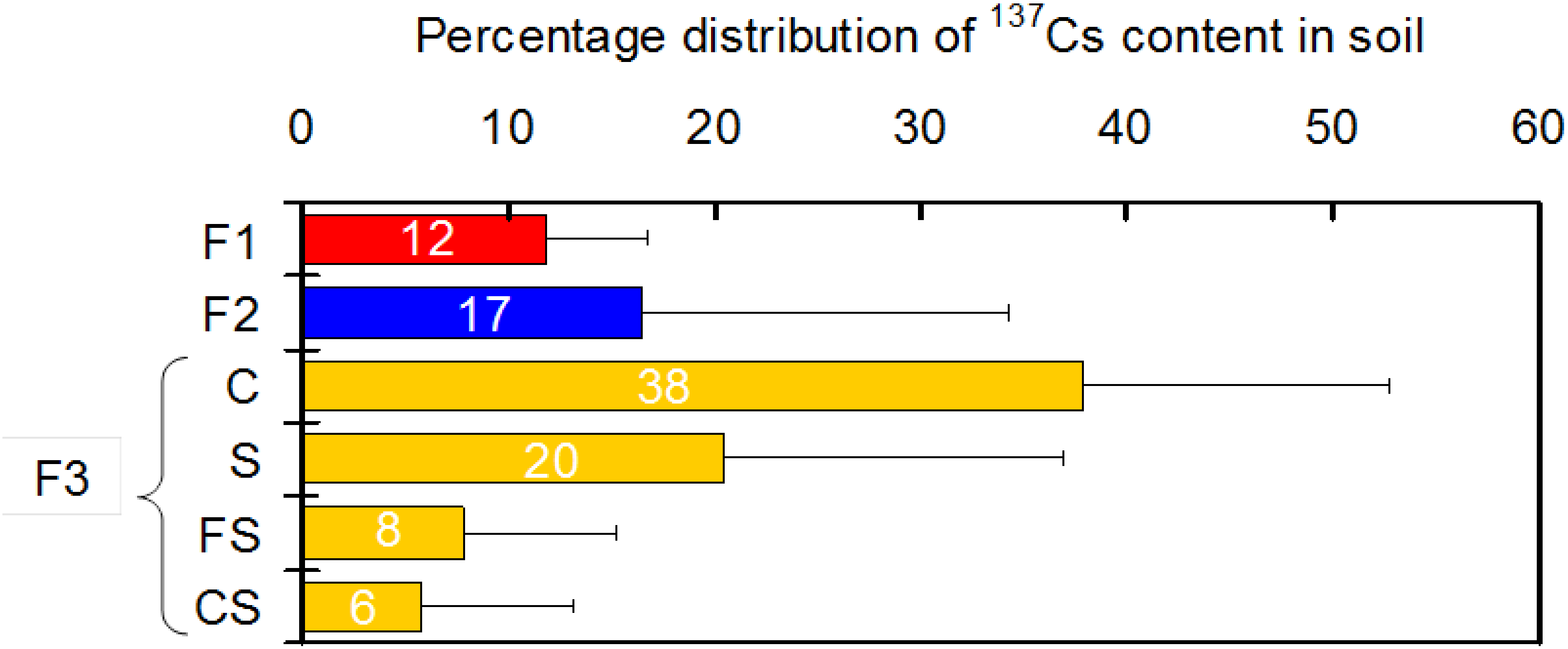


Mobility of Cs in soils is limited and soil-to-plant transfer is low

Radiocesium in soil



Percentage distribution of ^{137}Cs in physicochemical fractions in soil (n=11)



F1: Exchangeable fraction

F2: Organic-bound material fraction

F3: Strongly bound fraction (residue)

C (Clay, <0.002 mm), S (Silt, 0.002-0.02 mm),

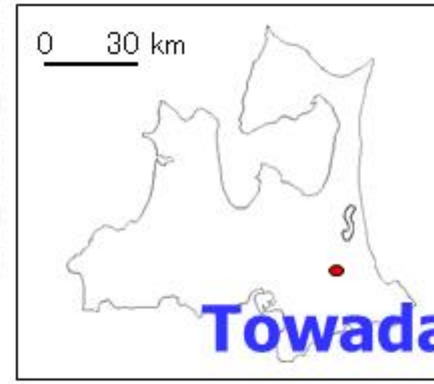
FS (Fine sand, 0.02-0.2 mm), CS (Coarse sand, 0.2-2 mm)

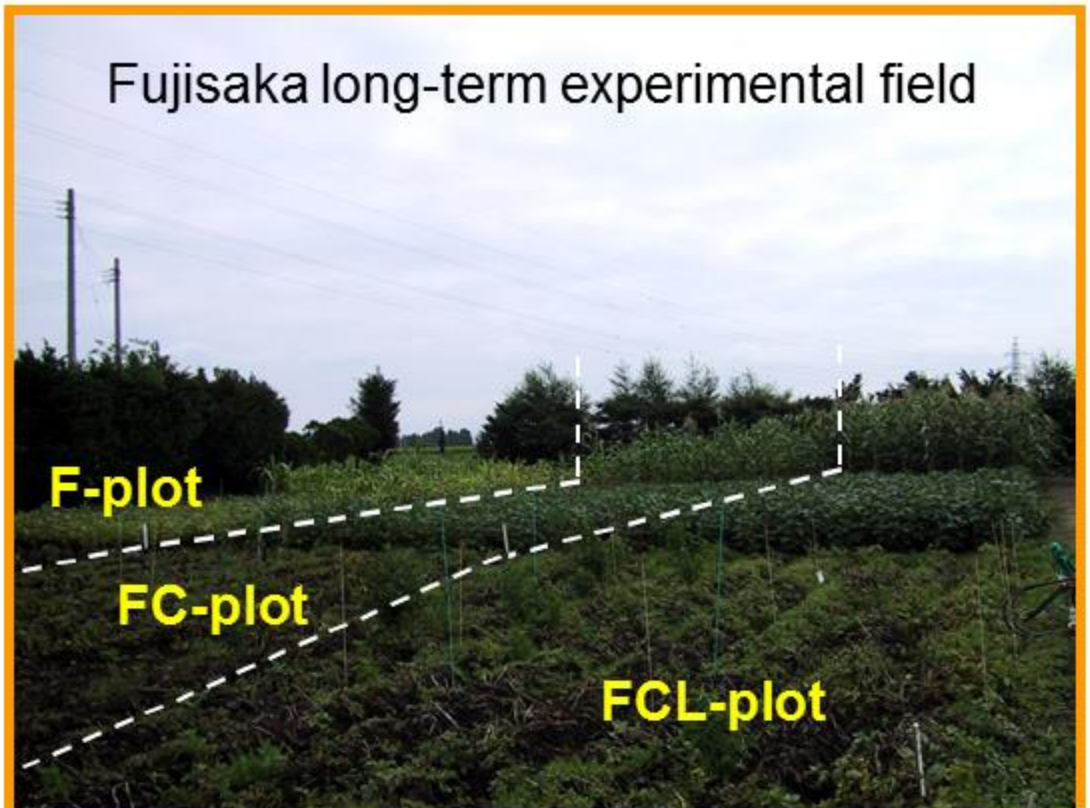
Long-term experimental field in Towada, Aomori (Established in 1940: sampling at August, 2001)

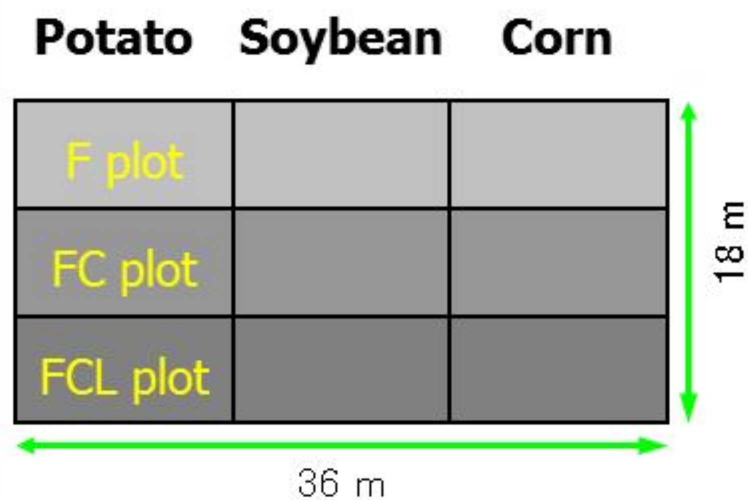
F−plot (**Chemical fertilizers**, N:P:K = 12:15:10 g m^{-2} y^{-1})

FC−plot (Chemical fertilizers ＋**Compost**, 1100 g m^{-2} y^{-1})

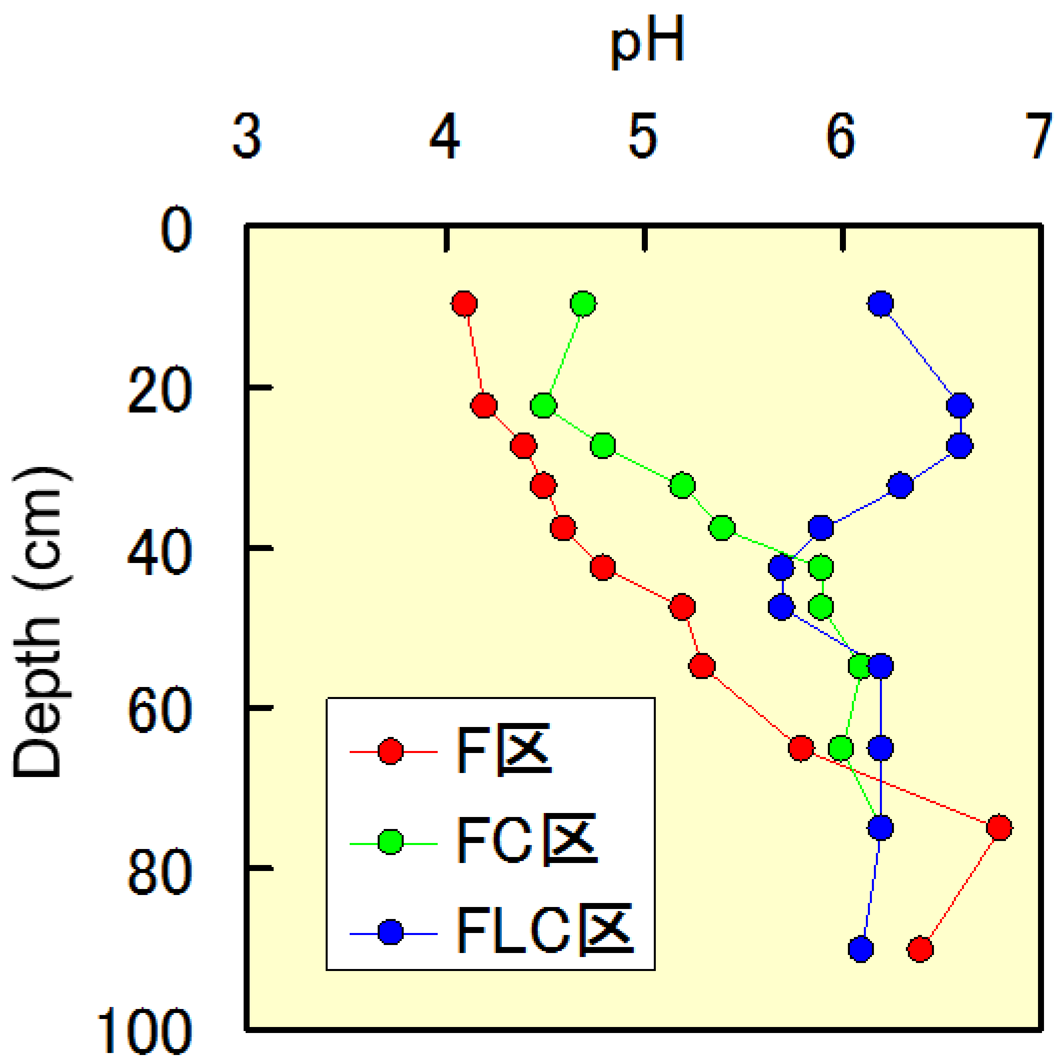
FCL−plot (Chemical fertilizers ＋Compost ＋**Lime**, 75 g m^{-2} y^{-1})



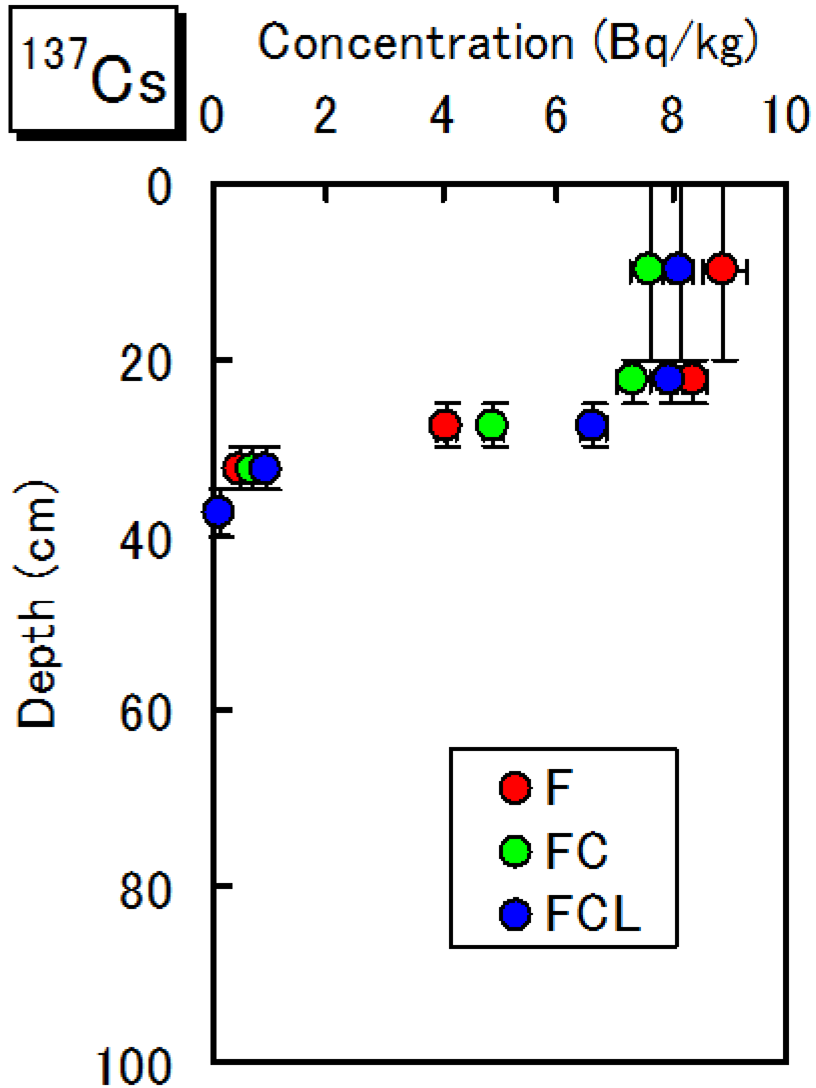




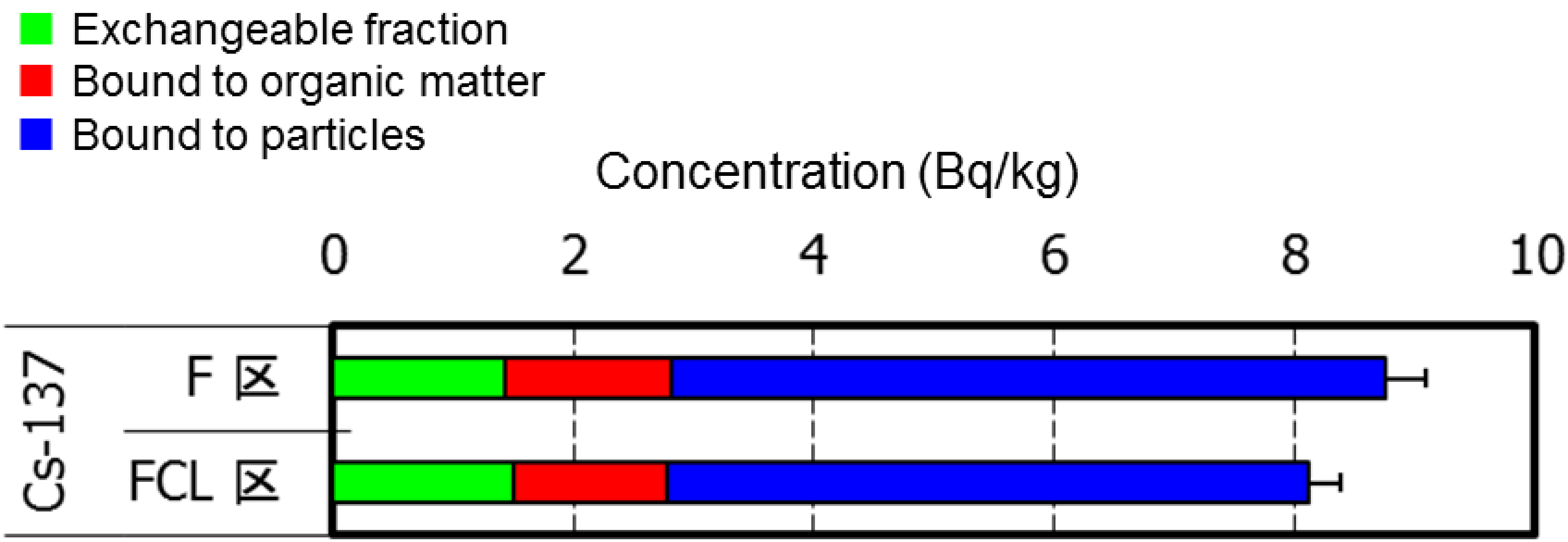
Vertical profile of pH in water extract from soil in long-term experimental field



Vertical profile of ^{137}Cs concentrations in a long-term experimental field

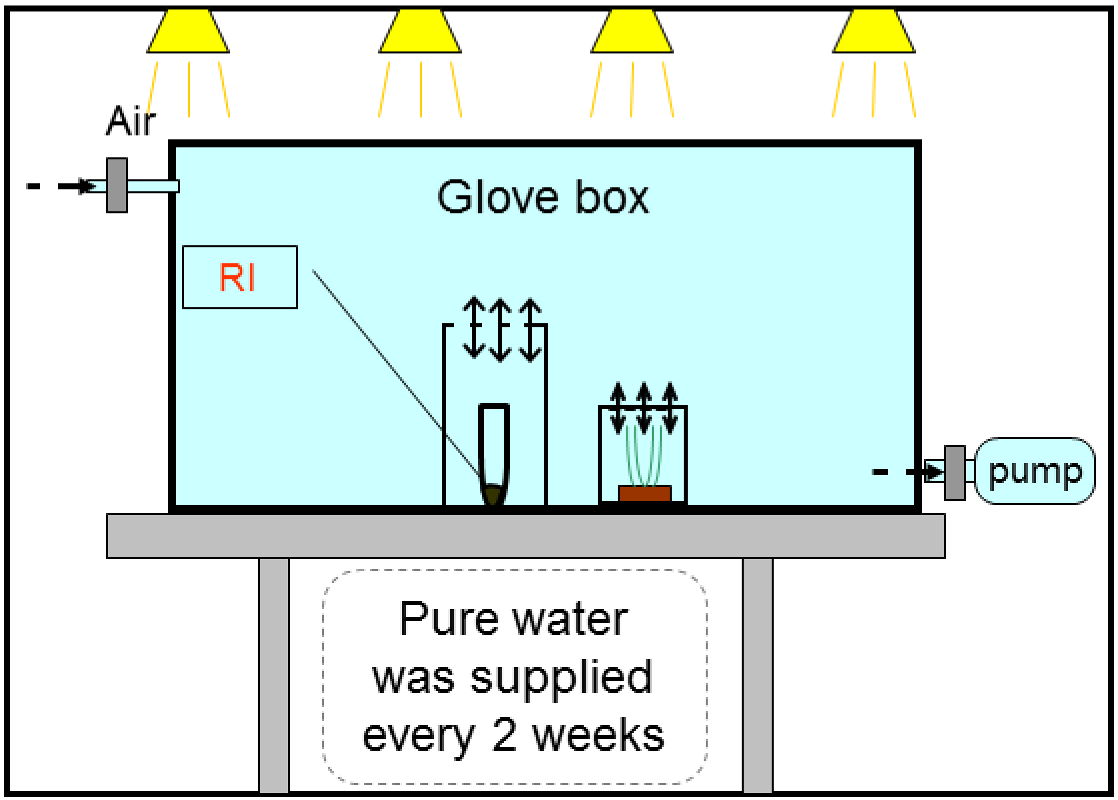


Physicochemical forms of ^{137}Cs in surface soil collected from a long-term experimental field

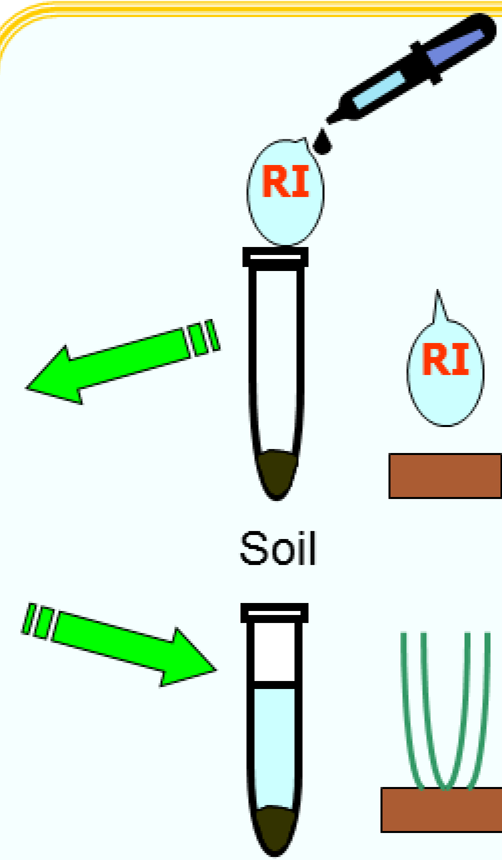


F区, pH 4.1; FCL区, pH 6.2

Experimental design of water extractability, exchangeable fraction and soil-to-plant transfer of ^{137}Cs with the time of elapsed

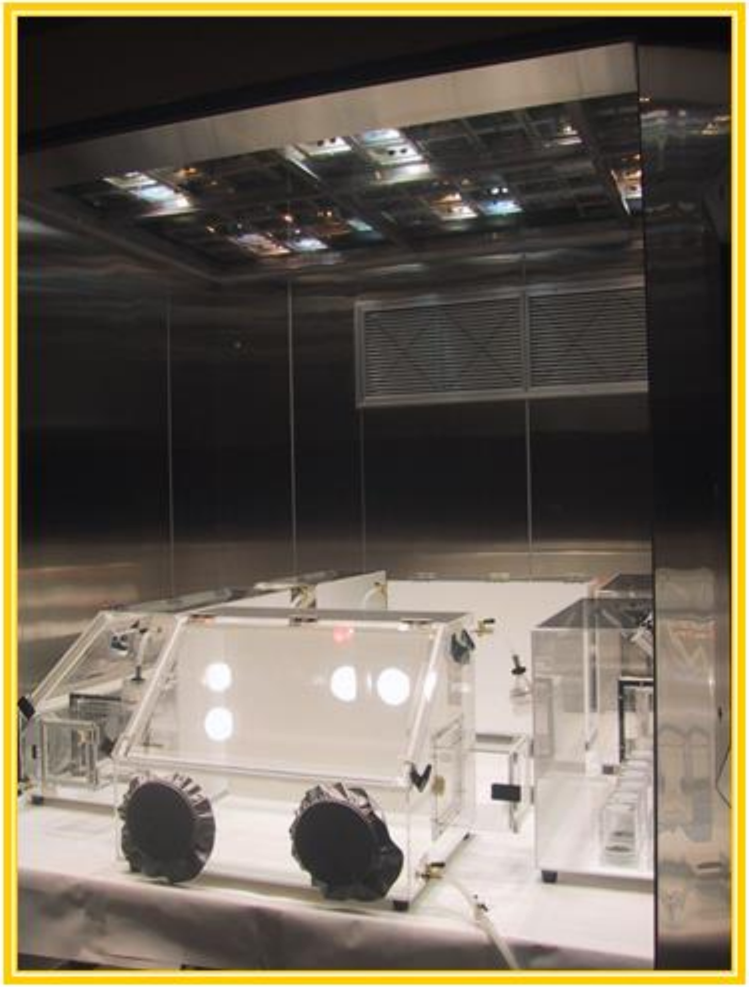


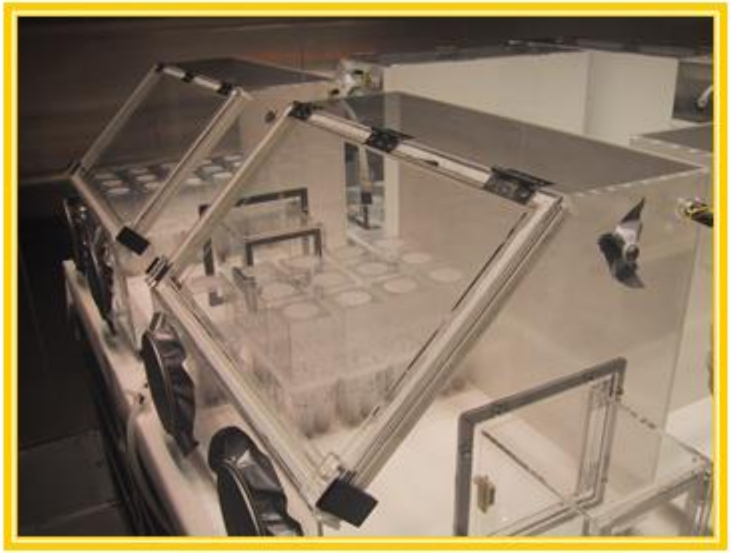
Artificial chamber: room temperature, 17℃; humidity, 60%; light intensity, 30000 lx; daylight hour, 12 h



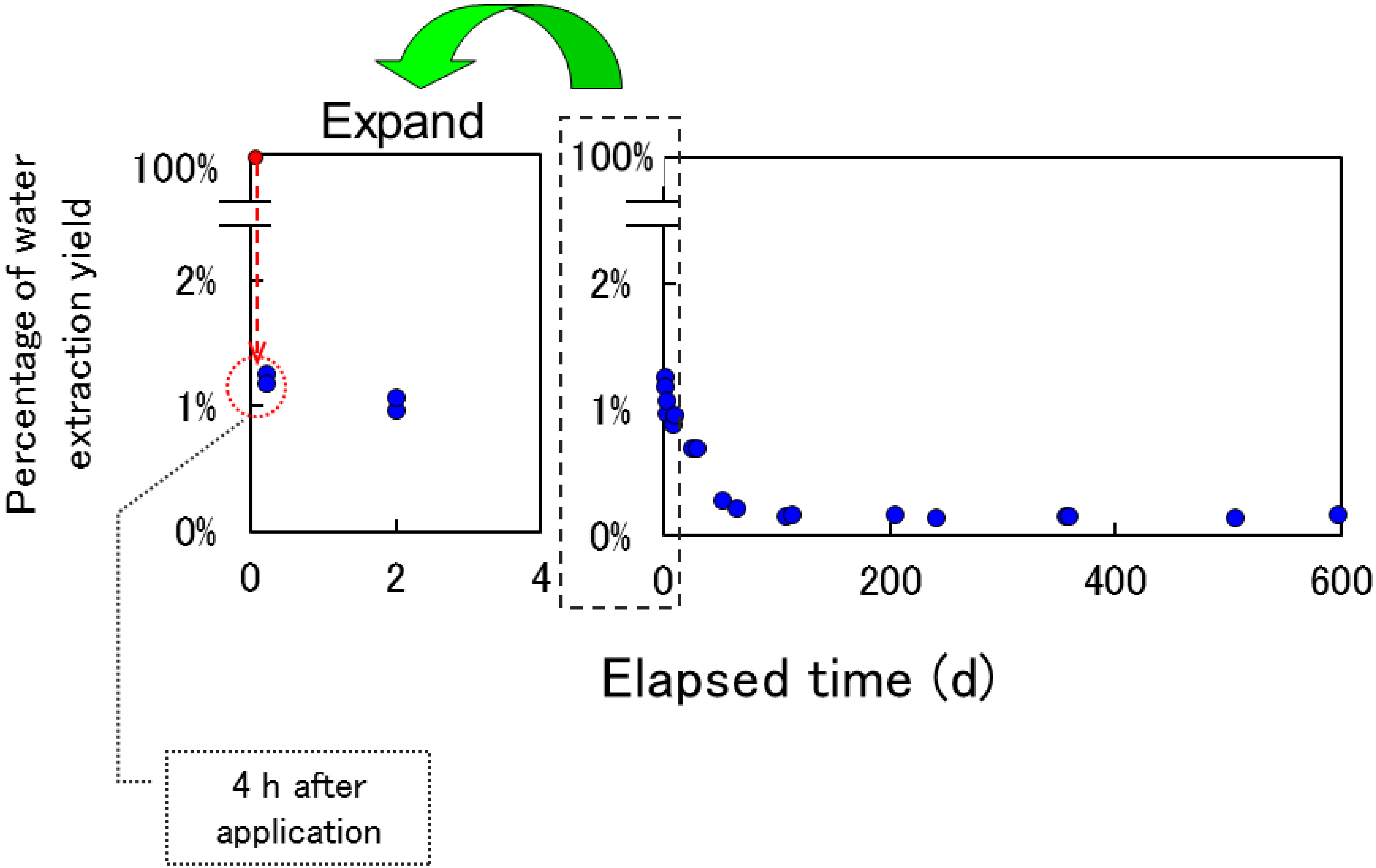
Extraction and cultivation of pasture

Photos in experiment

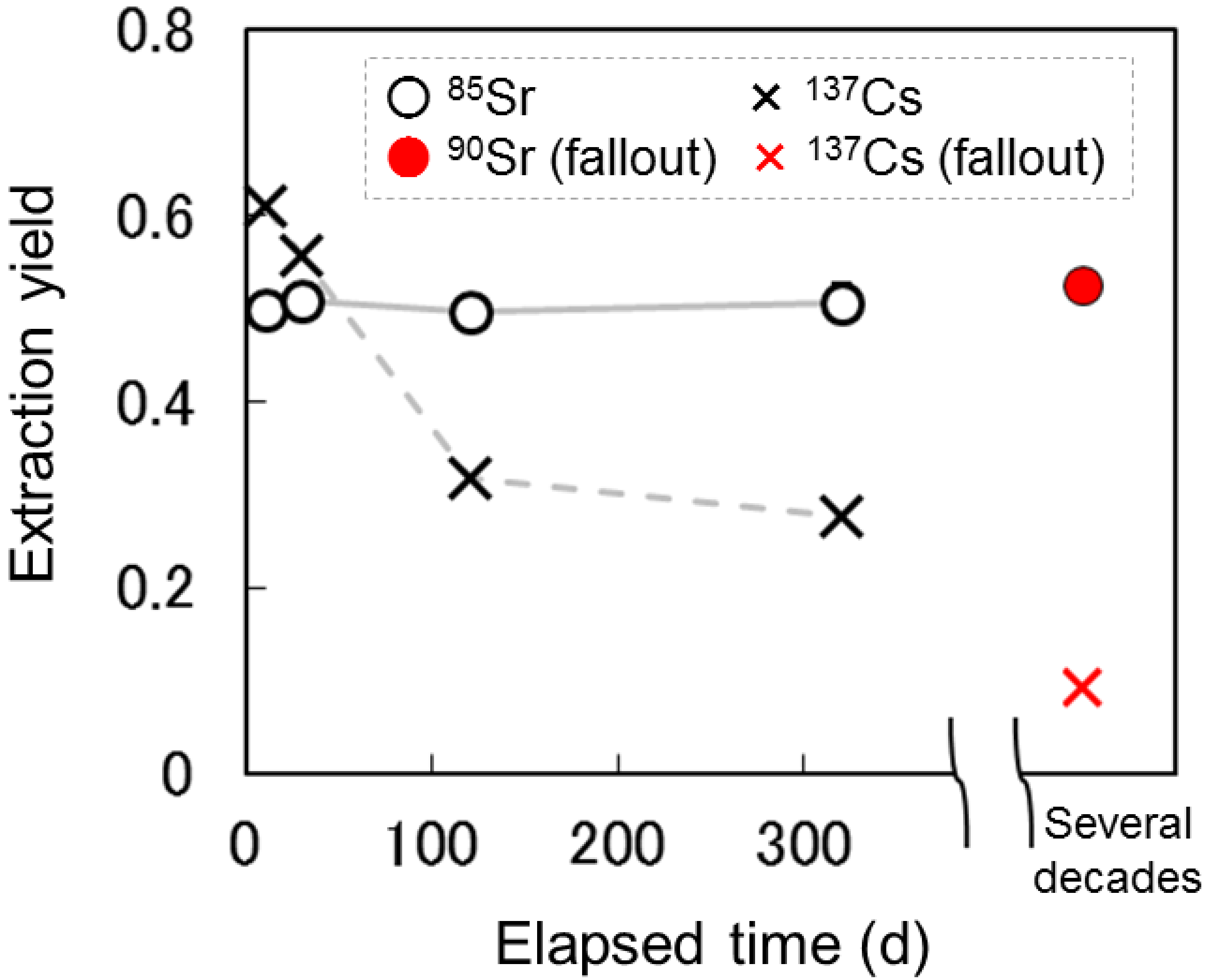




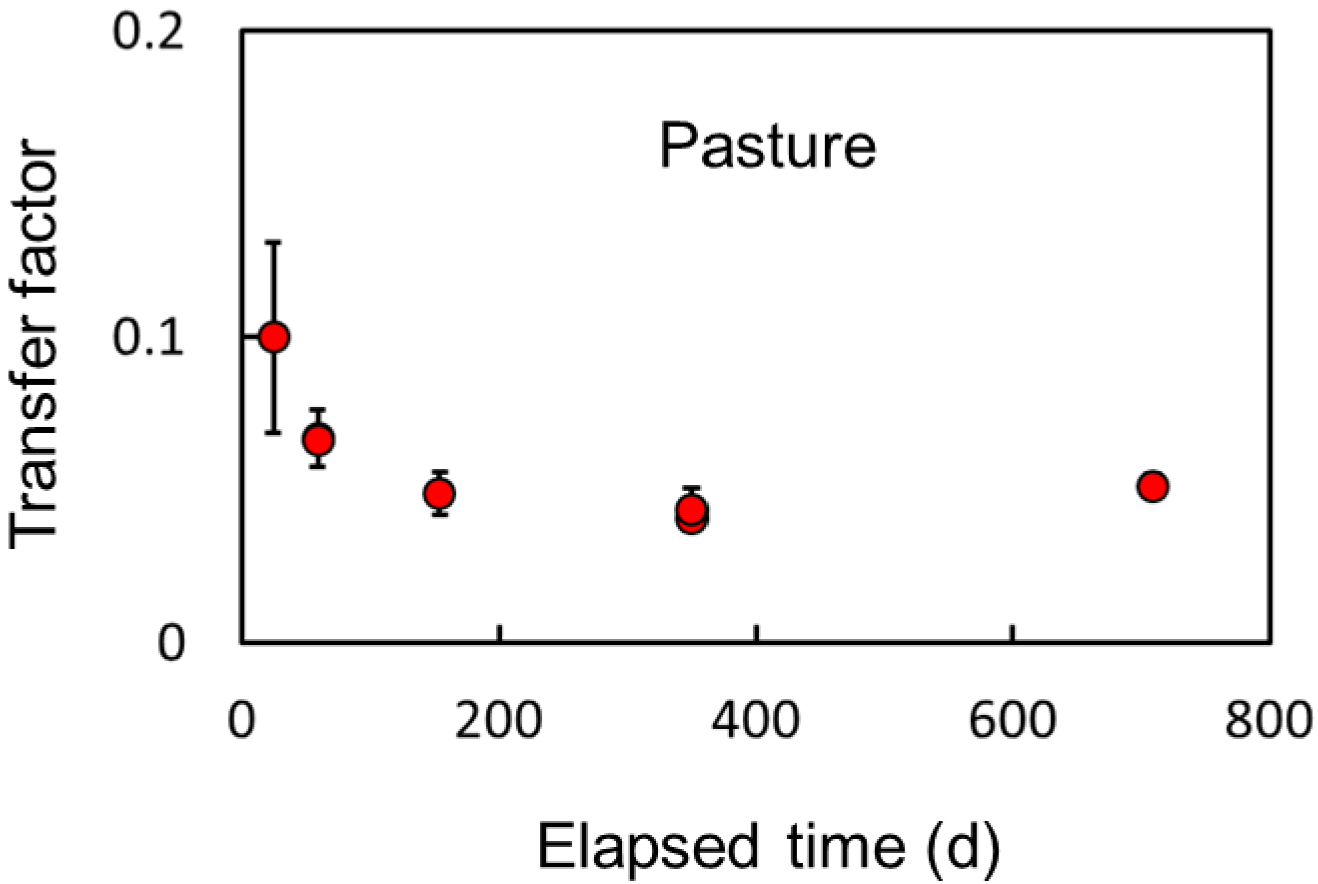
Temporal extraction yield of ^{137}Cs with water in soil



Extraction yields of added ^{85}Sr and ^{137}Cs with 1M ammonium acetate solution with time elapsed, and those of ^{90}Sr and ^{137}Cs derived from global fallout

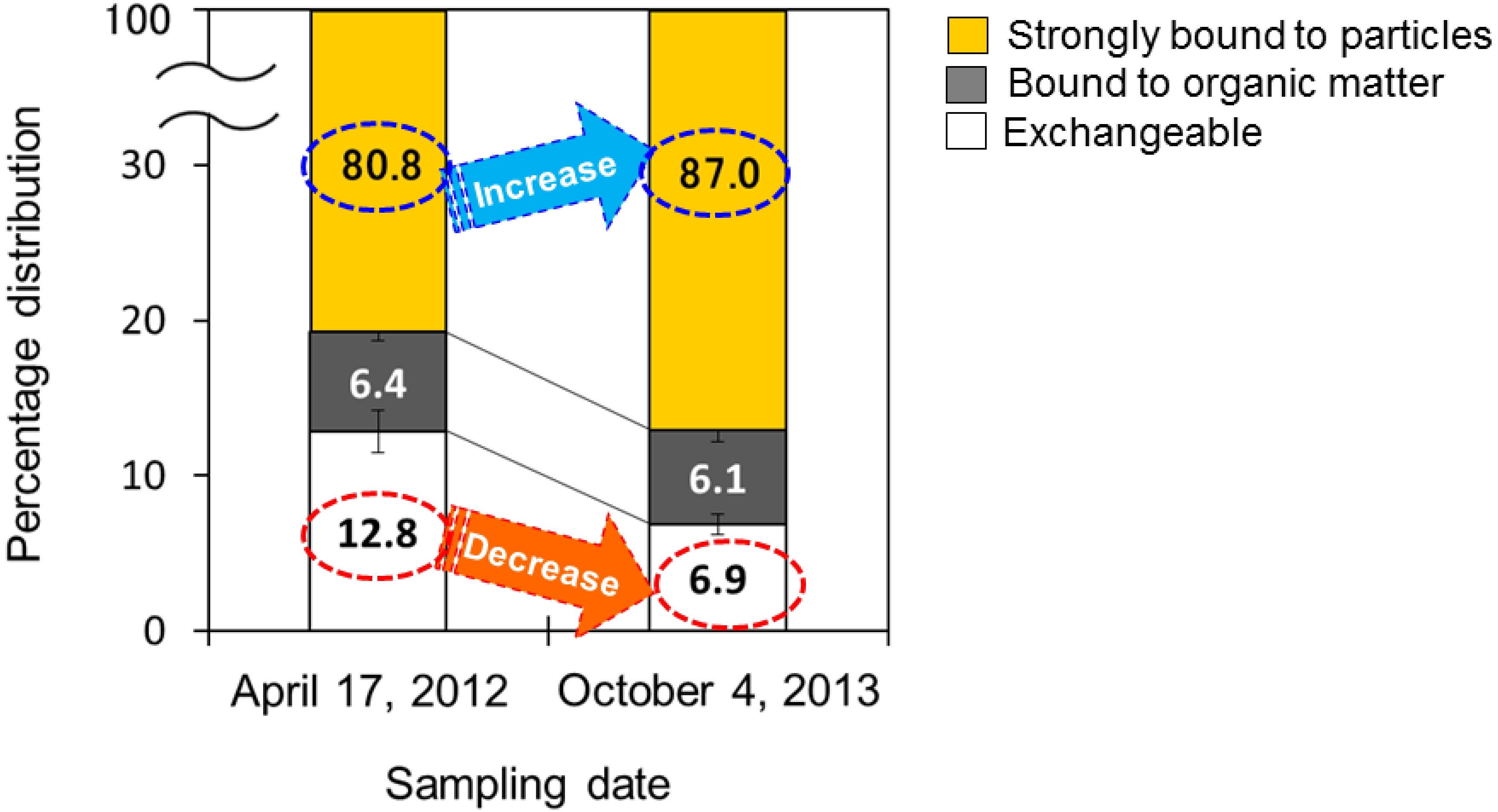


Temporal change of soil-to-plant transfer factor



$$\text{Transfer factor} = \frac{\text{Concentration of } ^{137}\text{Cs in plant}}{\text{Concentration of } ^{137}\text{Cs in soil}}$$

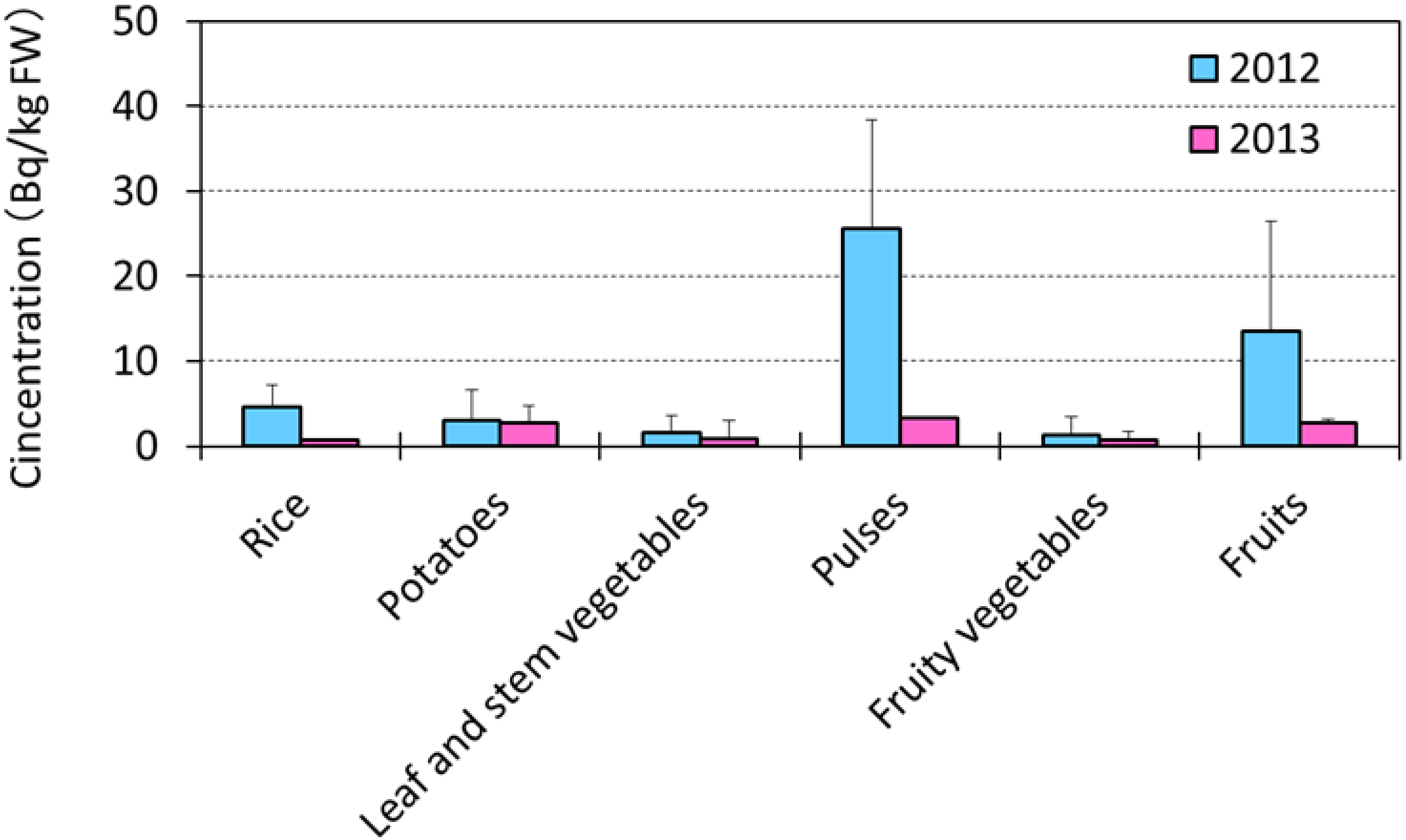
Change of physicochemical fractions of ^{137}Cs in a soil collected from Oguni, Date, Fukushima



Concentration of radiocesium in brown rice collected throughout Fukushima Prefecture in 2011 and 2012

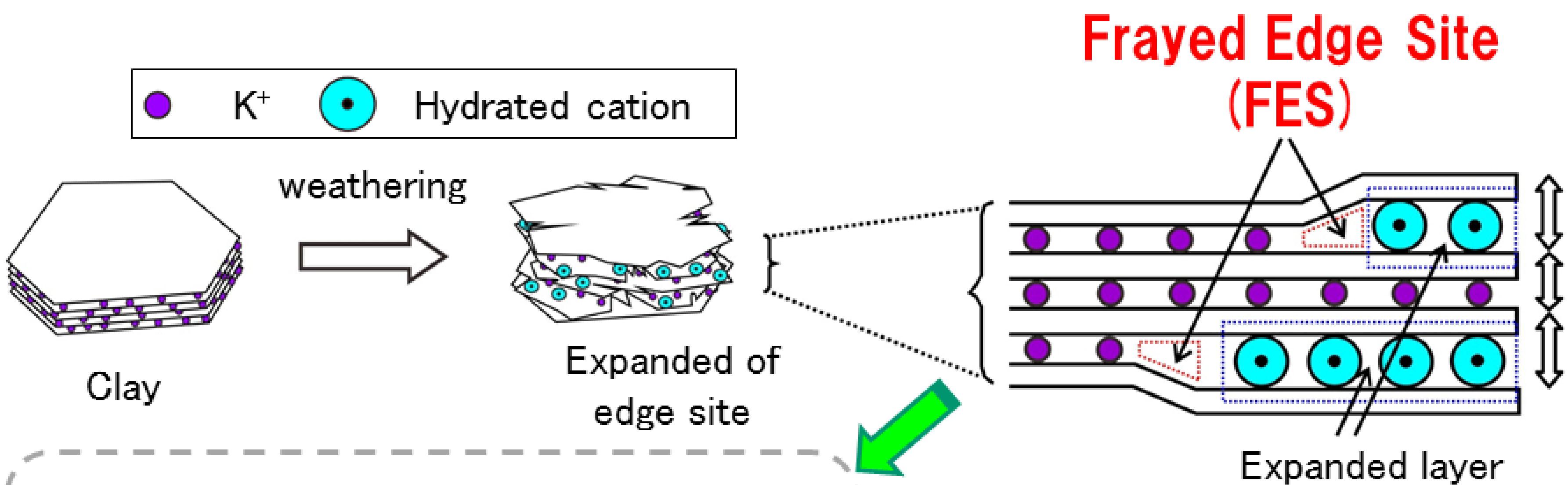
It was not any brown rice which was over the New Standard Limit (100 Bq/kg) collected throughout Fukushima Prefecture in 2014

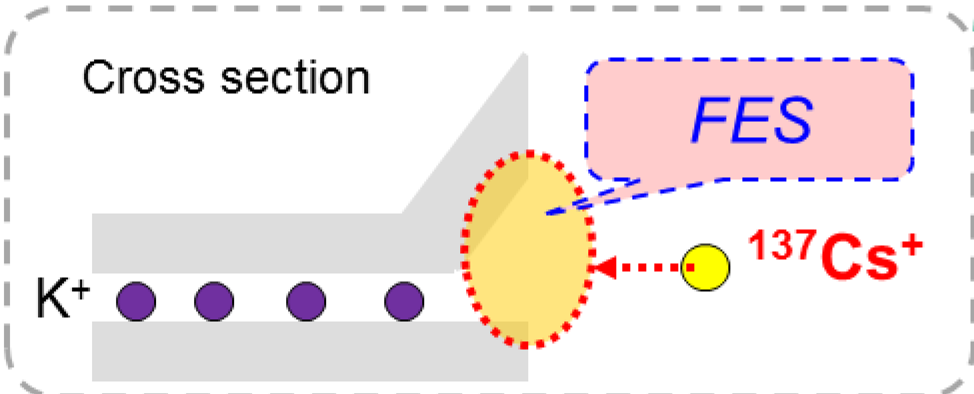
Concentration of radiocesium in agricultural products limited to production within the Fukushima Prefecture



Year	Number	Average	Min―Max	^{40}K（Average）
2012	36	7.2 → 1/4	＜0.2 ― 40	130
2013	42	2.0	＜0.1 ― 14	110

Interception of radioactive Cs in Soil





Selectivity of cation in FES

$Cs^+ >> NH_4^+ > K^+$

1000 : 5 : 1

After radioactive Cs adsorbed in the FES,

✔ Strongly bound to soil

✔ Difficulties of dissolved and transfer to plant

Radiocesium Interception Potential (RIP)

It is very difficult to determine the capacity of FES directly. Therefore Radiocesium Interception Potential (RIP) instead of FES was measured.

What is RIP?

- ✔ Indicator of vulnerability for radiocesium adsorption in soil
- ✔ Negative correlation with soil-to-plant transfer factor
- ✔ Wide range and different with soil types

放射性セシウム捕捉ポテンシャル
(Radiocaesium Interception Potential, RIP)

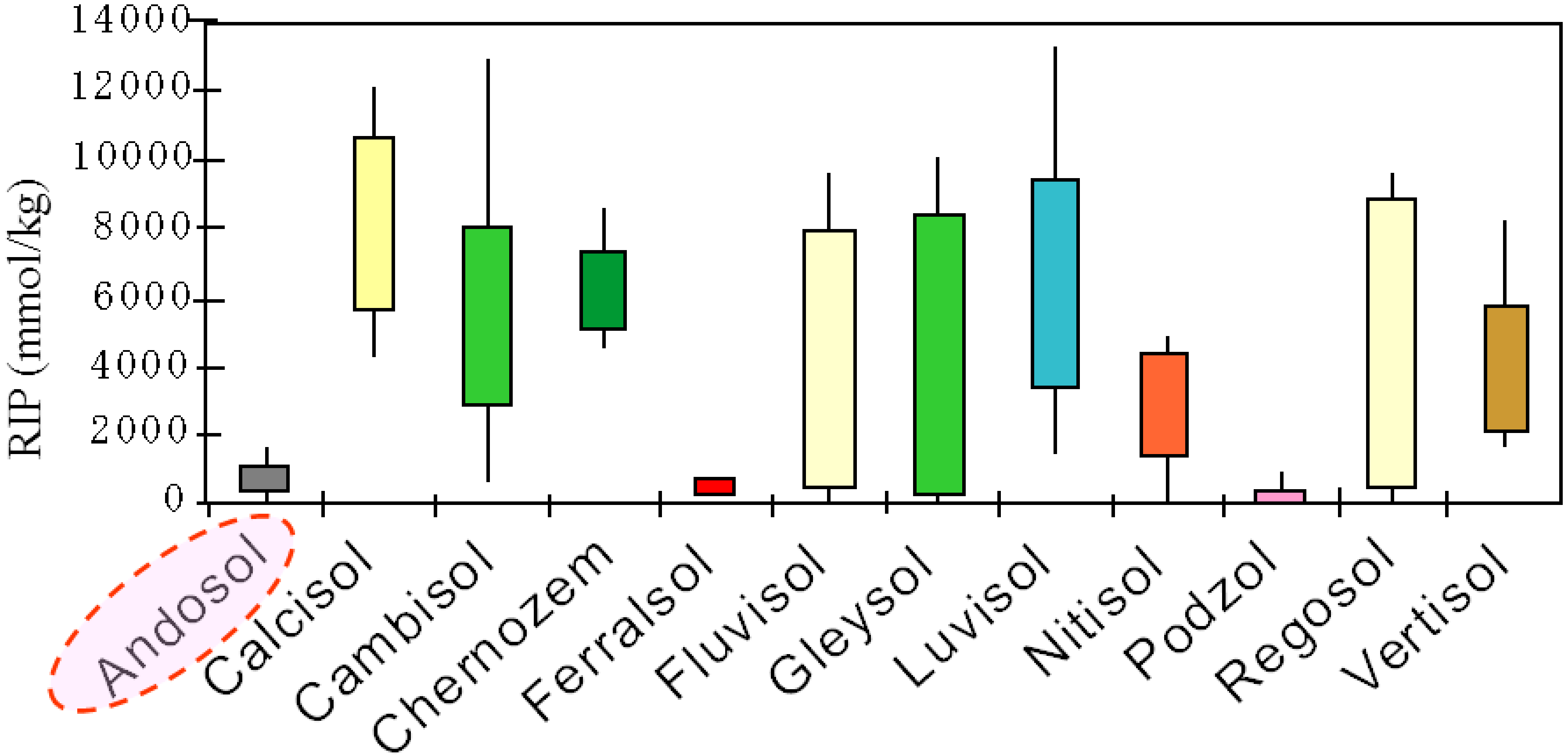
RIP（mmol/kg） ＝ $K_c^{EFS}(C_S\text{-}K) \times [FES]$

- $K_c^{EFS}(C_S\text{-}K)$: Selectivity coefficient of trace Cs to K of the FES
- [FES]: Amount of FES

The FES was saturated with K^+ and other exchangeable cation sites are saturated with Ca^{2+} under 0.1 mol/L $CaCl_2$+ 0.5 mmol/L KCl solution.

(Cremers et al., 1988; Wauters et al., 1996)

RIP value of soil in world wide



(Vandebroek et al., 2012, Journal of Environmental Radioactivity)

Calculation of RIP value on applying zeolite in soil

Soil

Amount of surface soil per 10 a (depth 0–20 cm)

⇒ 140,000 kg/10 a (bulk density: 0.7 g/mL); RIP value, 1000 mmol/kg

Applying zeolite

Amount of application: 200 kg/10a (content in soil: 0.14% w/w)

RIP value in 4 kinds of zeolite: ①1,430 (min), 18,300, 48,600, ②56,500 (max) mmol/kg

	Increment of RIP in the application of zeolite	⇒	RIP value in soil after the application
Applied zeolite①:	2.0 mmol/kg	⇒	1002 mmol/kg (0.2%増加)
Applied zeolite②:	81 mmol/kg	⇒	1081 mmol/kg (8 %増加)

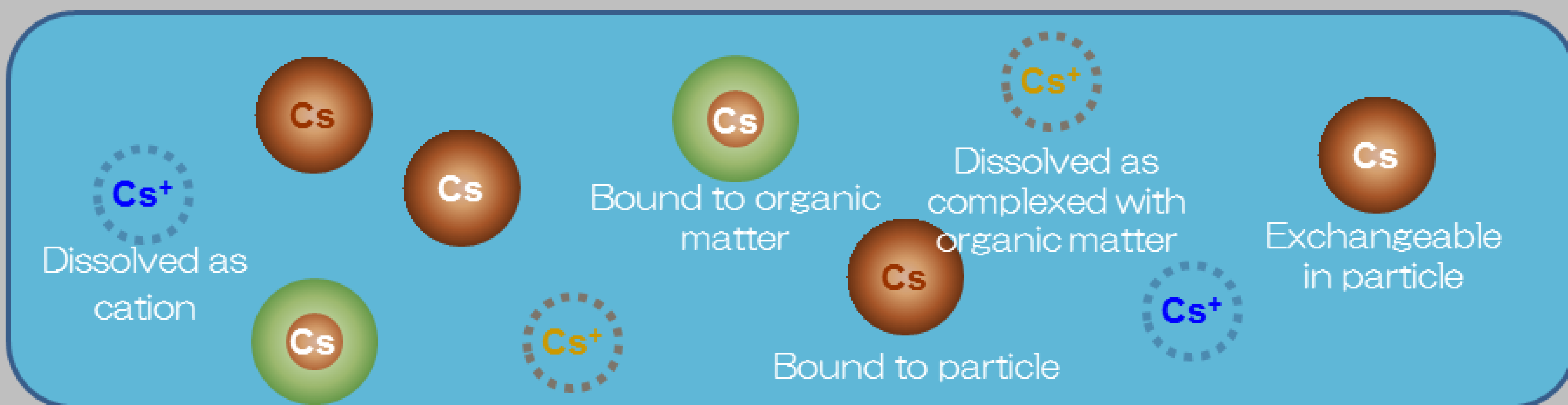
It is possible to calculate the effectiveness of applying amendment. It need to measure the RIP in amendments before applying.

Radiocesium in inland water

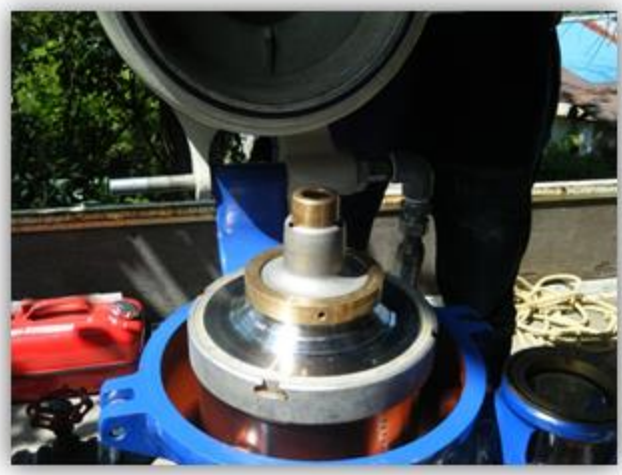
✔ Why is the physicochemical forms in inland water important?

⇒ Bioavailability of radiocesium is different from its physicochemical forms such as dissolved, suspended fractions.

Physicochemical forms of radiocesium in inland water



Sampling of suspended substances with continuously centrifugal method



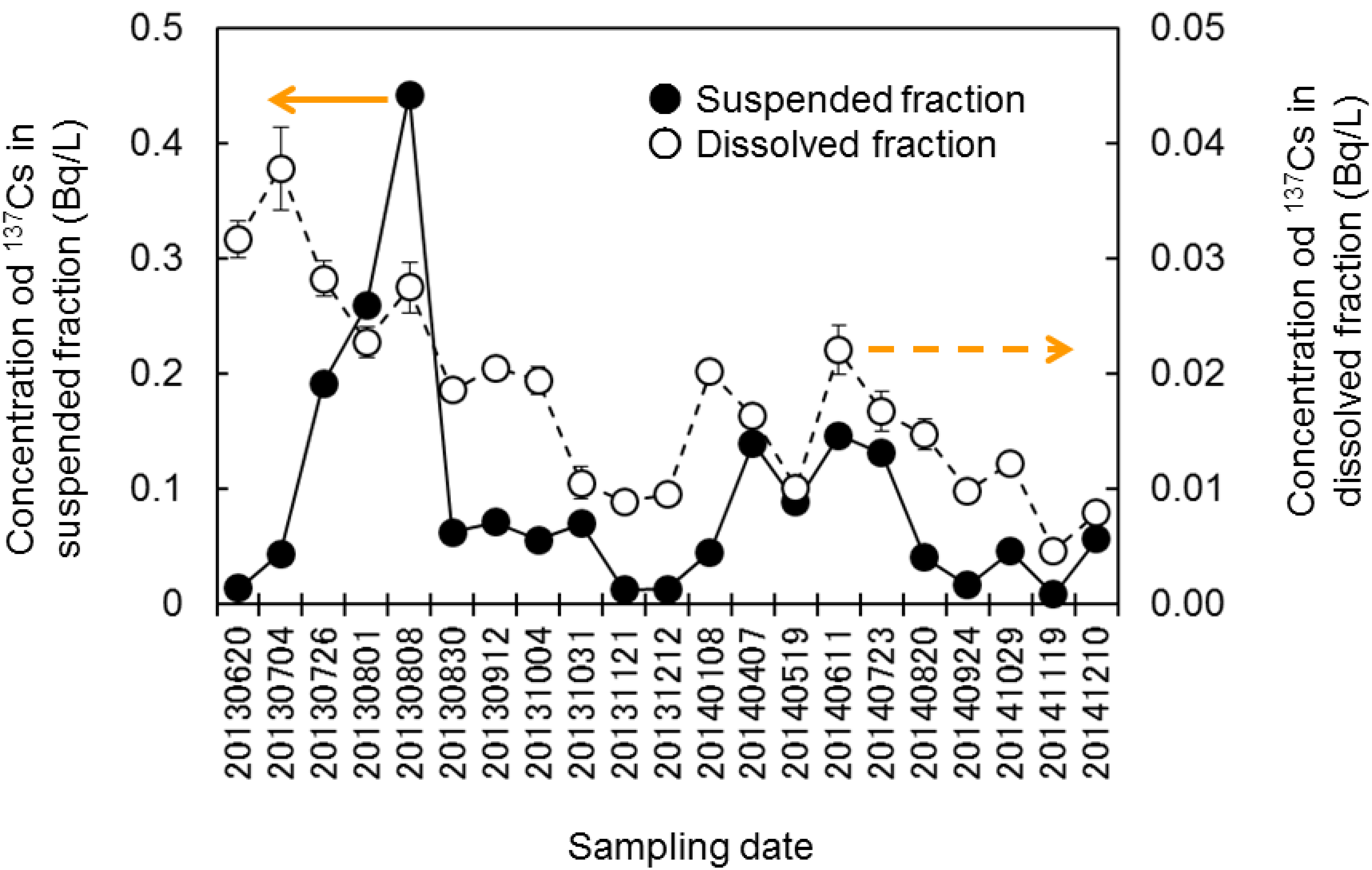
Collecting time: 4-6 h

Flow rate: 12.5 L/min

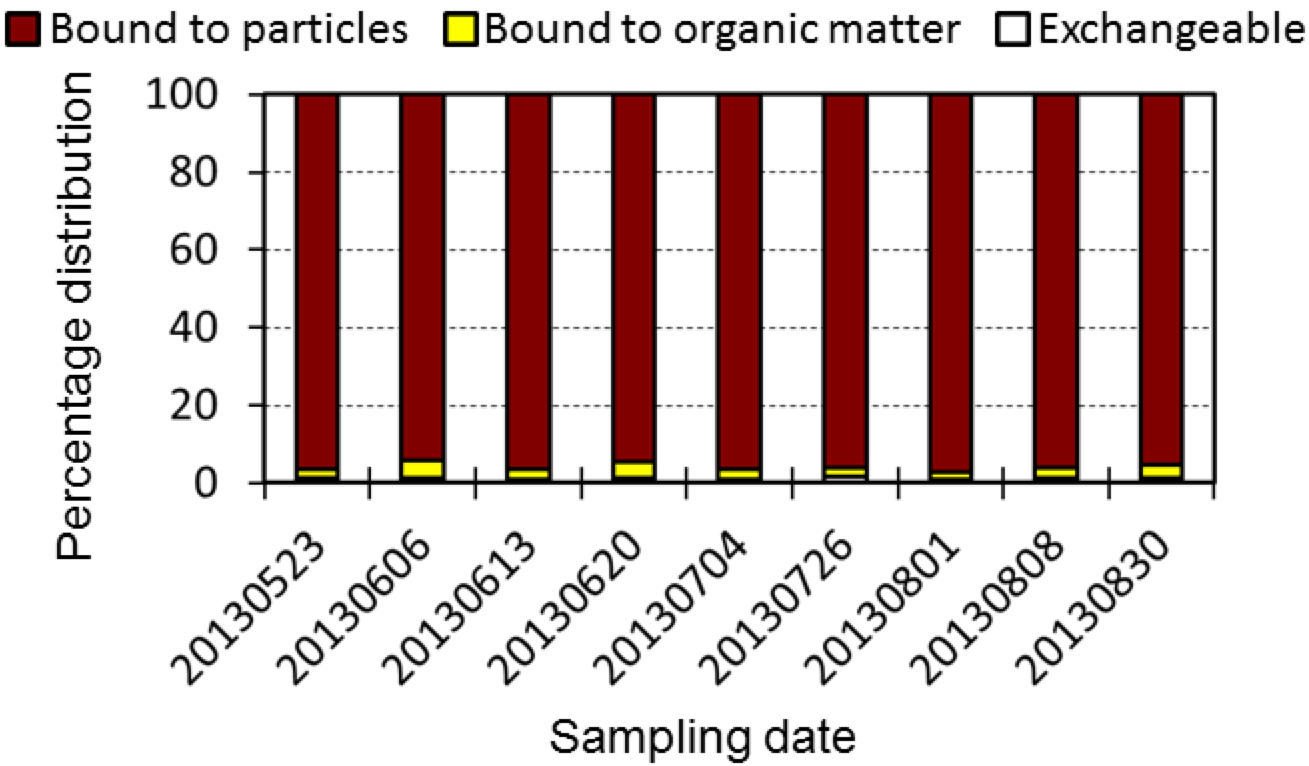
Total flow amount: 2000-4000 L

Collected suspended matter: **2-20 g**

Temporal change of ^{137}Cs concentration in dissolved and suspended fractions collected in Oguni river, Date



Physicochemical fractions of ^{137}Cs in suspended matter collected in Oguni river, Date



存在割合(%)	平均値	最小値		最大値
□ Exchangeable	1.2	0.90	—	1.5
□ Bound to organic matter	2.9	1.8	—	4.5
■ Bound to particles	96	94	—	97

→ Difficult to dissolve in water and to transfer to plant

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Thank you very much for your attention!