Importance of Understanding Clay-Cs Association for Reduction, Storage and Disposal of Waste from Decontamination Activities in Fukushima

http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_03.pdf

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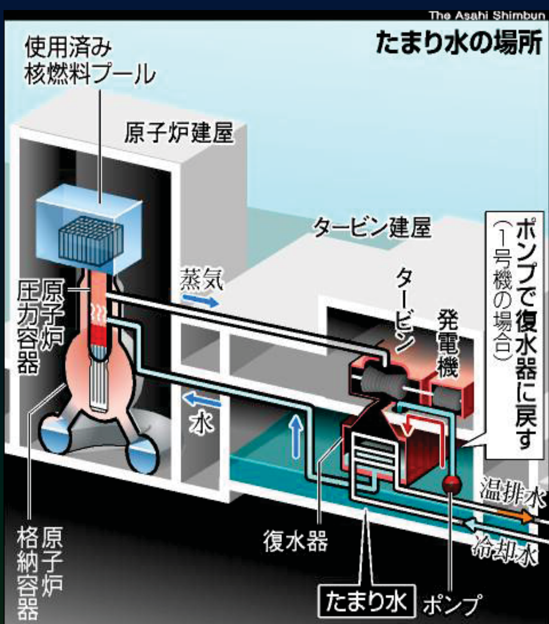
Total Dose and Destination of Radioactive Cesium

Cs−134+Cs137：1380PBq

Land
8-16PBq
Sea
18-26PBq
(2-5%)

Fuel
800−880PBq
(60%)



Water
480PBq
(35%)

→ Adsorbent

P：10^{15}

東電公表のたまり水のデータおよび公開ワークショップ「福島第一原子力発電所事故による環境放出と拡散プロセスの再構築」で紹介されたデータを基に算出

Scientists Showed the Evidence of Clay-Cs Strong Association

~1986: above-ground nuclear test

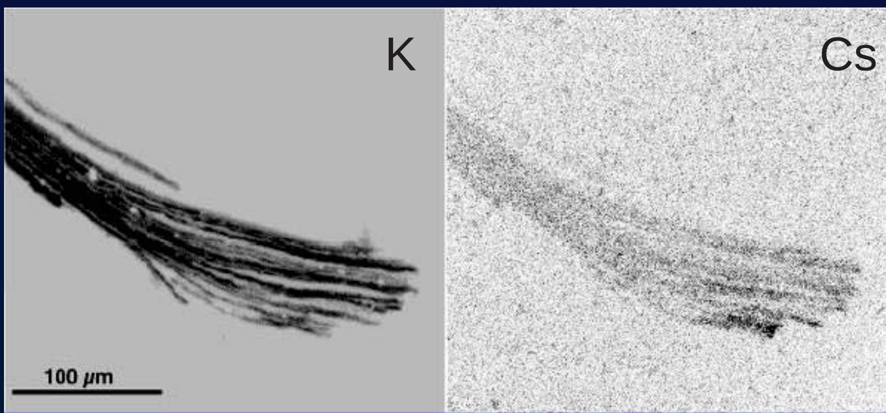
~2000: Chernobyl NPP accident, Hanford's leak

Radiocesium retention in soils is due to the presence of a small number of highly selective sites of clay minerals.

2000~:

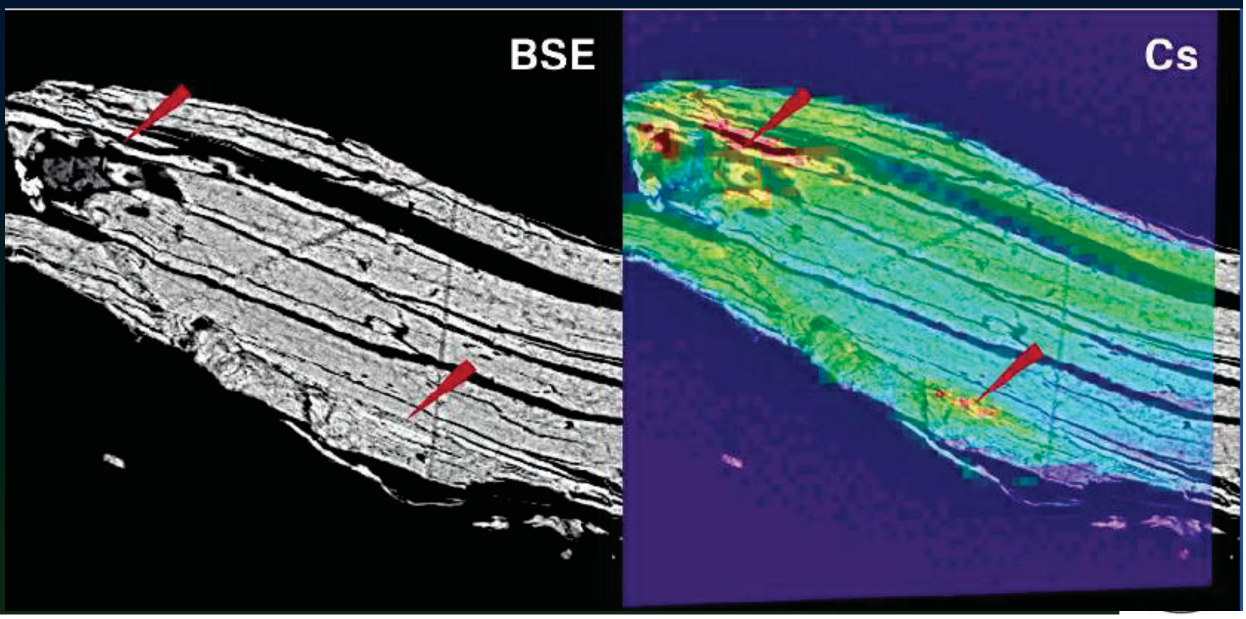
EXAFS study (Bostick et al., 2002): The inner-sphere complexes of Cs may have occurred within the interlayer or at frayed edge sites and were less extractable than the outer-sphere complexed Cs.

Microscopic study (McKinley et al., 2004):

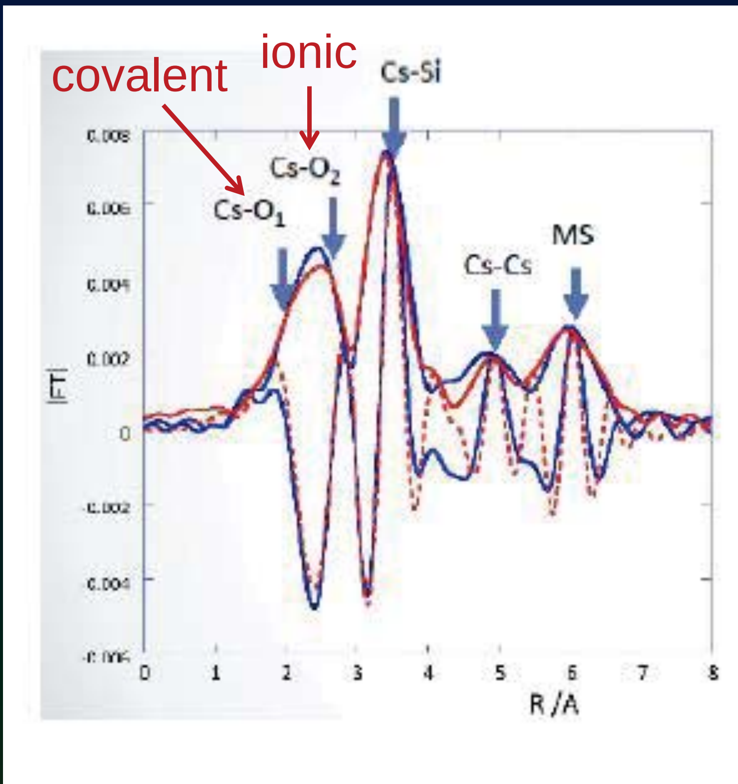


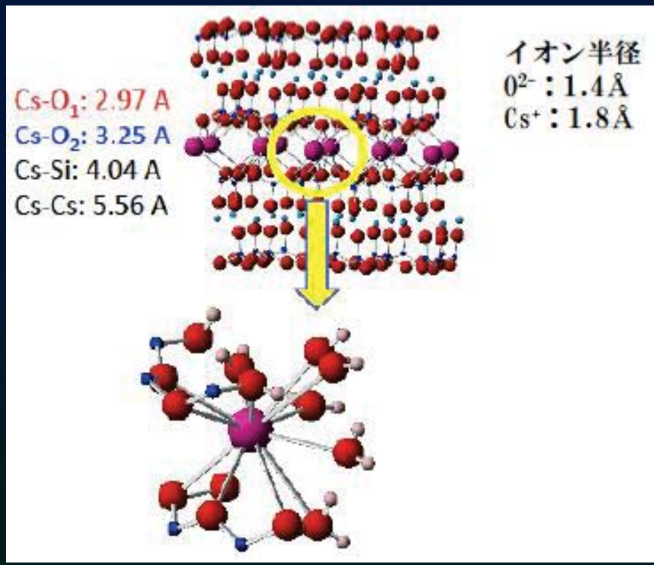
(McKinley et al., 2004)

Frayed Edge!



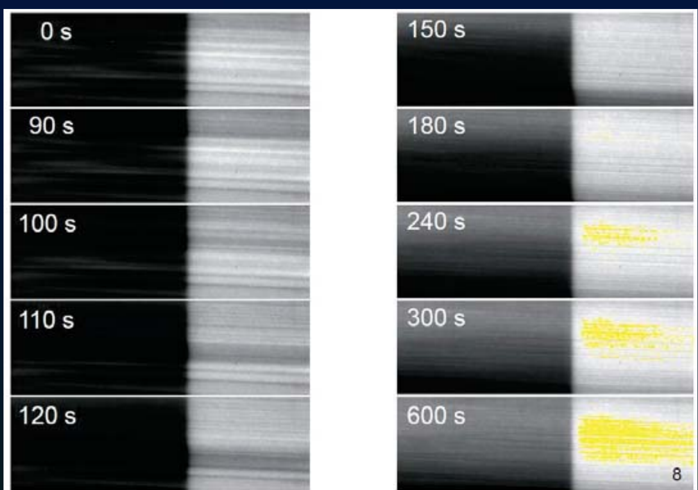
XAS Studies (by JAEA)

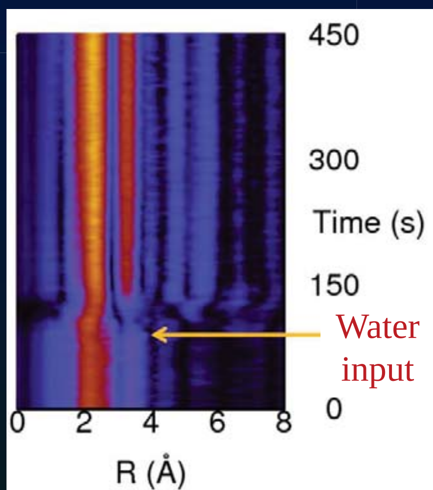


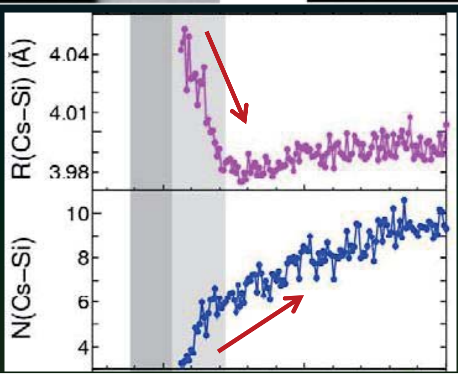


Before and after chemical treatment

Dispersive (time-splitting) XAFS (DXAFS by JAEA)



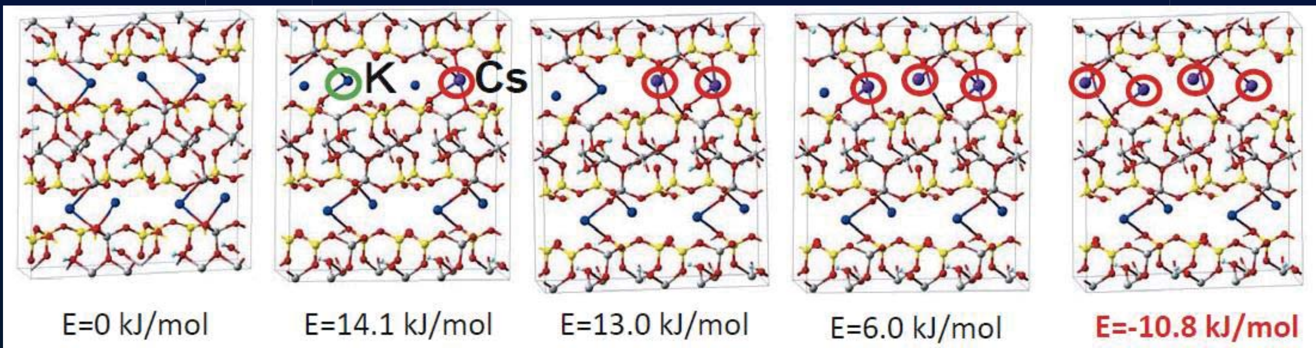


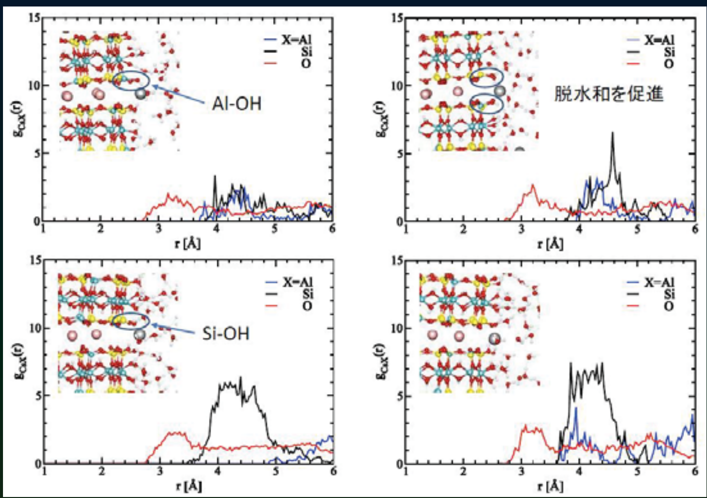


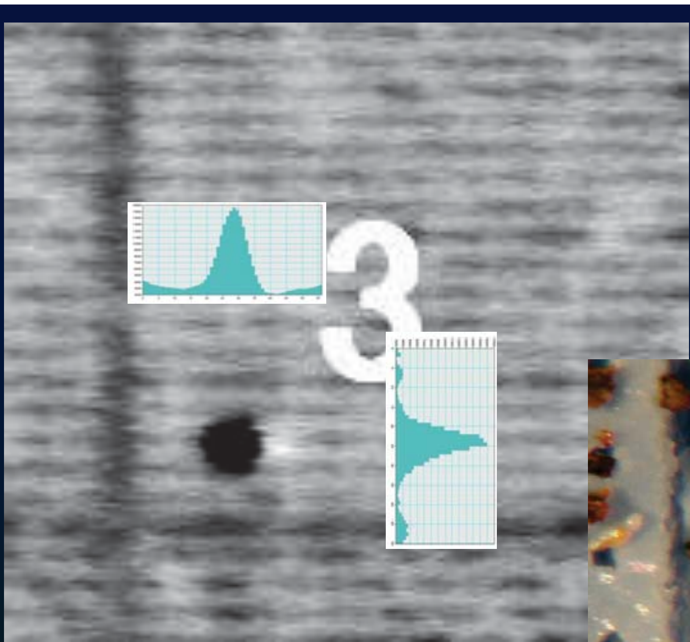
Interlayer collapse

Cs uptake into interlayer

Ab initio MD by JAEA

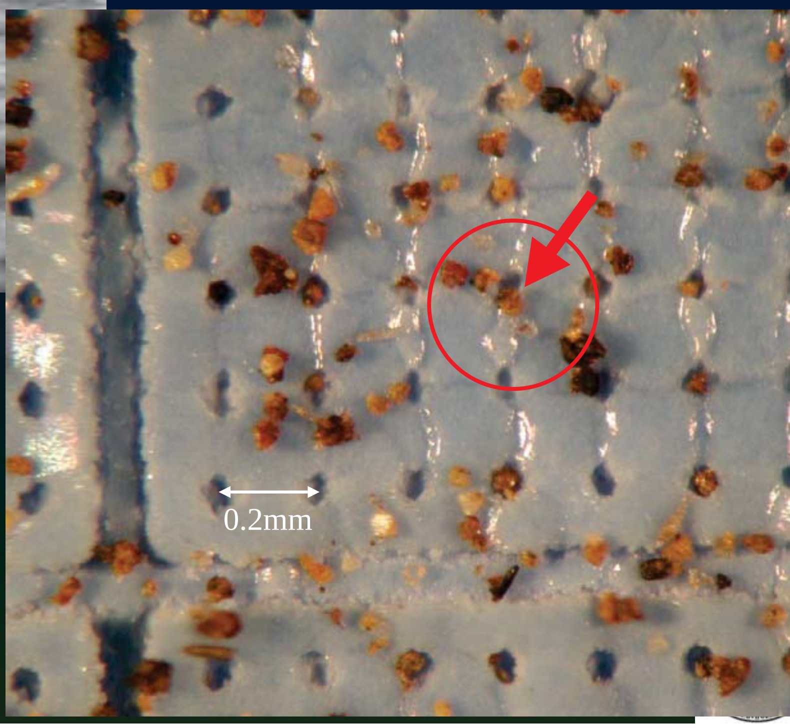




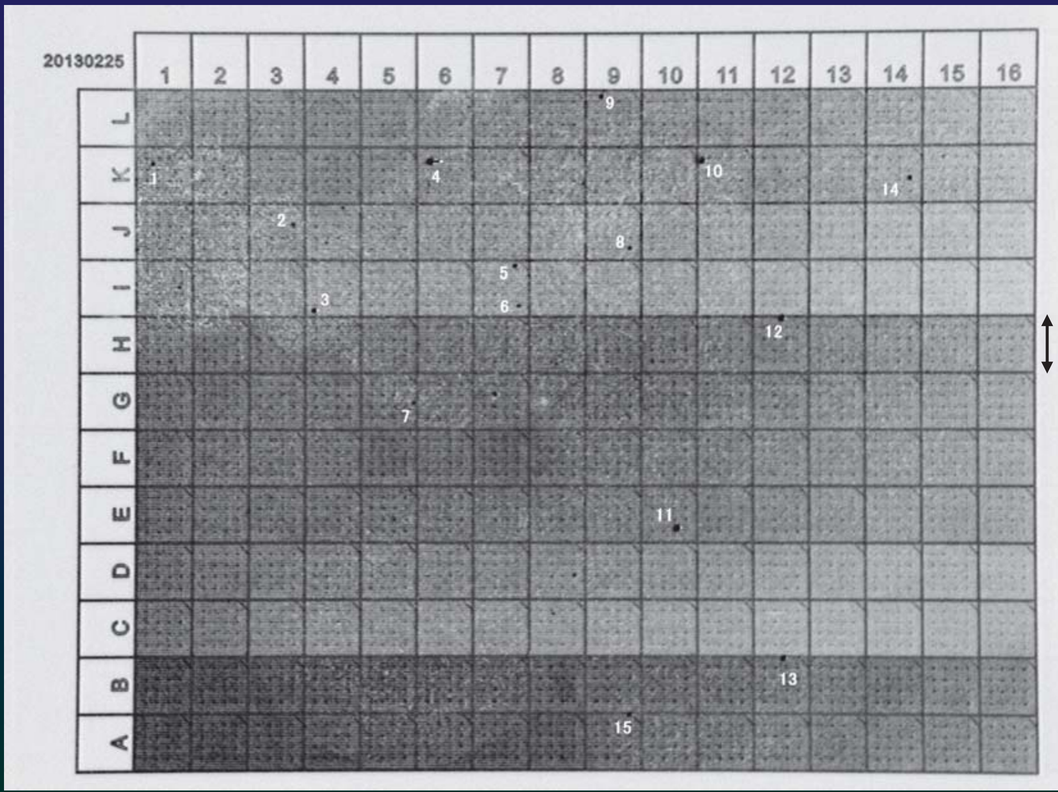


IP(Imaging plate) image

Finding of particles by IP
By Prof. Kogure of UT



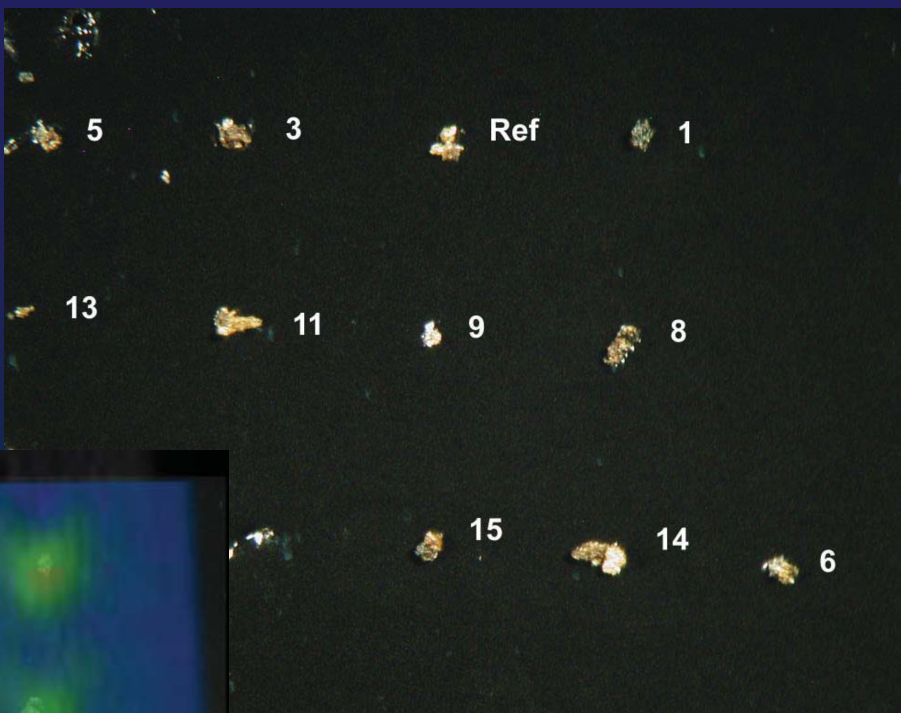
Microscopic image

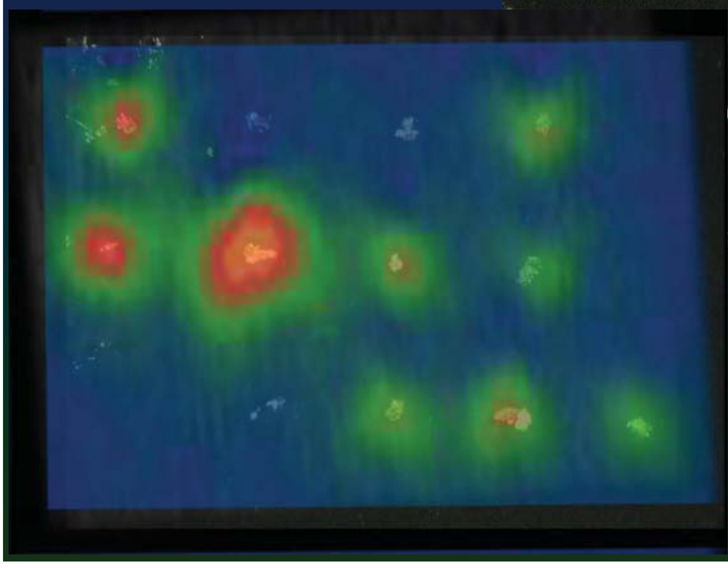


IP image

5mm

Around 20,000 particles were put on this plate.
However, only 15 radioactively contaminated particles were observed in this case.





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Efforts of JAEA and Issues on Decontamination

23rd April, 2012

- Decontamination of building and road : applicable by existing methods
- Decontamination of farm and forest lands is extremely difficult
- The following two technologies should be developed
- Reduction of the wastes
  - Incineration
  - Washing or leaching
  - (Wet classification)

  To develop the above technologies, understanding of clay-Cs association is definitely needed.
- Highly developed monitoring technolo

http://www.jaero.or.jp/data/02topic/fukushima/interview/nakayama_t.html

OFFICE OF ENVIRONMENTAL MANAGEMENT

EPA United States Environmental Protection Agency

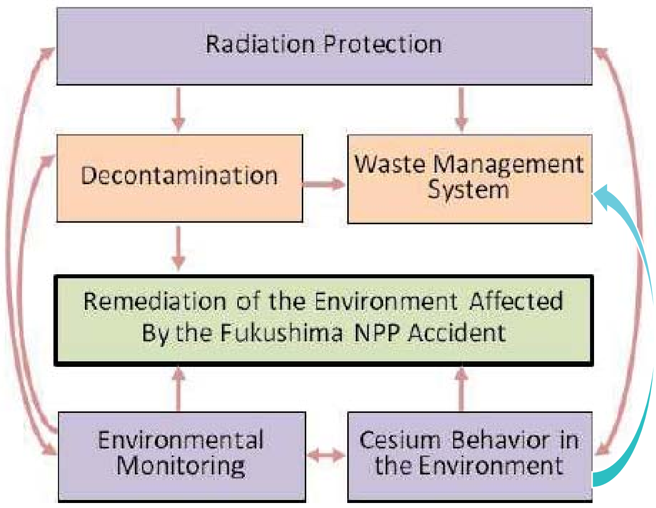
ESF Mission Overview

- **US State Department's Embassy Science Fellowship Program was used to provide expert support to Japan's Ministry of the Environment (MOE) in its decontamination efforts in areas outside of the Daiichi nuclear plant site.**
- **Overall intent was to draw upon US DOE and US EPA remediation experience to:**
  - Share methods and lessons learned
  - Offer suggestions for enhancing Japan's off-site decontamination efforts, and
  - Identify areas for future collaboration
- **Assignment duration:** February – March 2013
- **Approach:**
  - Worked closely with MOE's Decontamination Team
  - Reviewed extensive set of program documents, guidelines, procedures, methods, status, etc.
  - Met with staff from related agencies: Japan Atomic Energy Agency, National Institute for Environmental Studies, Nuclear Regulation Authority, etc.
  - Visited Fukushima Prefecture and municipal government decontamination offices
  - Visited decontamination and waste storage sites in evacuated and non-evacuated areas
  - Met with decontamination contractors carrying out full-scale remediation and demonstration projects

http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_03.pdf

Systems Perspective for Fukushima Offsite Remediation

- **System diagram shows key program elements for offsite remediation**
- **Framework was used by ESFs to organize all aspects of review including formulation of observations and recommendations**
- **Connections between system elements need to be developed and maintained to enhance overall remediation effectiveness.**
- **One cross-cutting consideration, public involvement, was identified as a having a significant role in the success of many of these program elements.**



Program Elements for an Environmental Remediation System for a Populated Region Contaminated by Cesium

Lee et al. (2013)

http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_03.pdf

Be Systematic for Intelligent Decontamination and Waste Management!

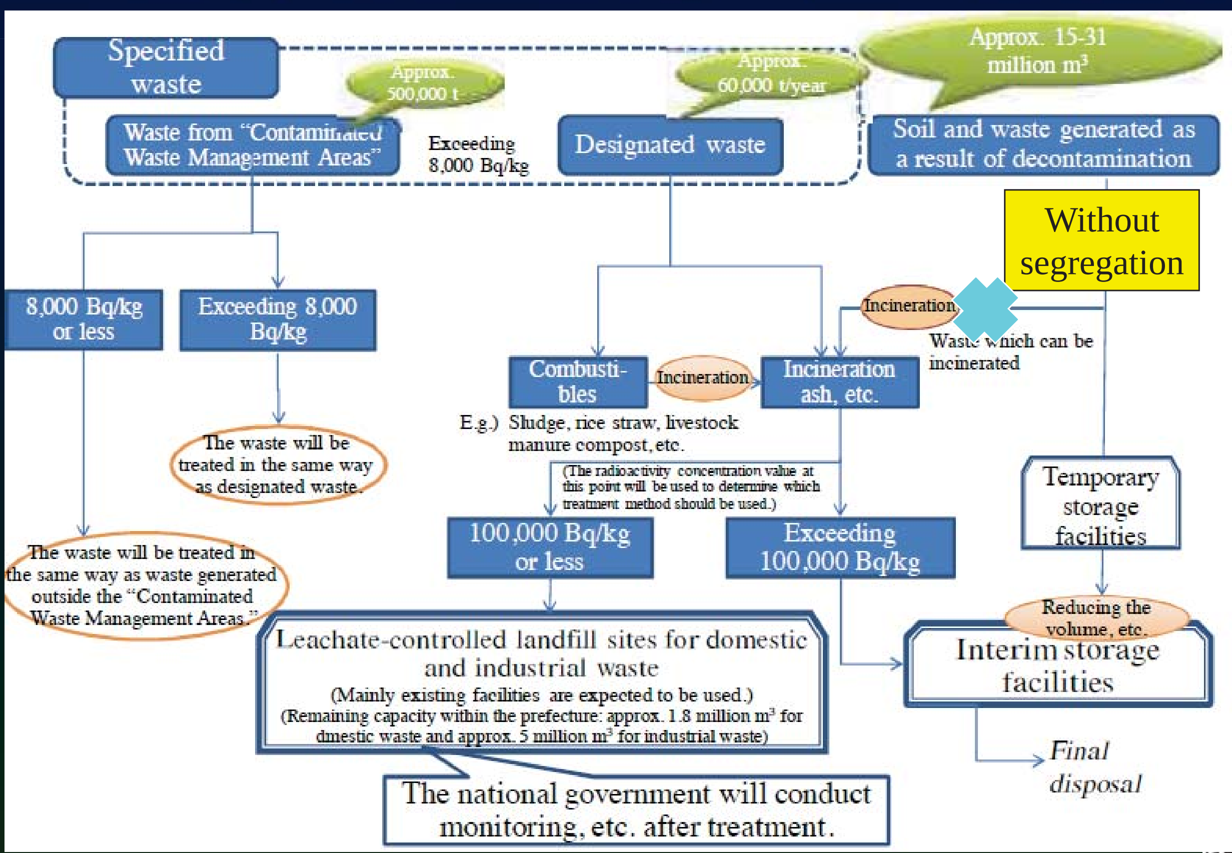
✘Reductionism（要素還元主義）

- Persons in charge of decontamination are always thinking "effectiveness of their methods in the decontamination"
- Persons in charge of decontamination do not care the waste produced by their decontamination activities.

●Systematic approach

- So far, there does not appear to be a clearly defined process for evaluating options for full scale decontamination or for making the decision of whether and how to proceed for the contaminated areas.
- Should conduct a systematic analysis of the existing performance data to identify potential factors or practices that could improve effectiveness of future decontamination efforts.

Flow Diagram for the Treatment of Specified Waste and Waste Generated as a Result of Decontamination



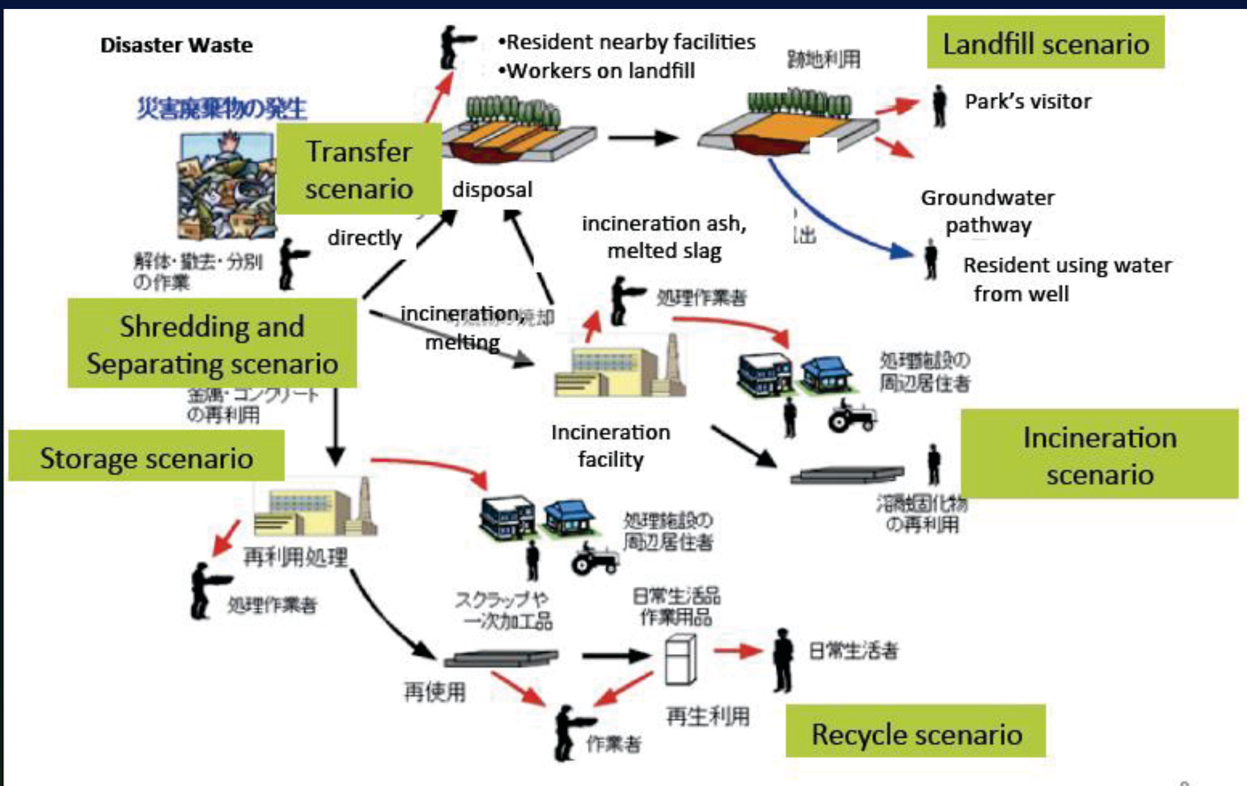
http://josen.env.go.jp/en/roadmap/pdf/chart1_5.pdf

Issues in Waste Generated as a Result of Decontamination

MOE did not decided the followings

- how to treat the wastes according to their doses
- how to treat the wastes for reducing their volumes
- how to reuse the soil generated after volume reduction processes
- how to reduce amount of wastes from the site of occurrence

Outline of Exposure Scenario Considered Waste Management



Safety of cross-jurisdictional waste treatment (June 19, 2011)

http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_01.pdf

Results of the Assessment

Scenario		Target for Evaluation	Radioactivity concentration resulting in an exposure dose of 1mSv/y
Storage	Waste loading and unloading work	Workers (1000h/y)	12,000 Bq/kg
	Those living around storage sites	Public (Outside in 20% resident time)	100,000 Bq/kg *A certain distance from a storage
Transfer	Waste transfer work	Workers (1000h/y)	10,000 Bq/kg
	Those living around transfer routes	Public (450 h/y)	160,000 Bq/kg
Incineration	Incinerator repair work	Workers (900h/y)	30,000 Bq/kg
	Those living around incineration facilities	Public (Outside in 205 resident time)	5,500,000 Bq/kg
Landfill	Incineration ash landfill operations	Workers (1000h/y)	10,000 Bq/kg
	Dewatered sludge, etc. landfill operations	Workers (1000h/y)	8,000 Bq/kg
	Those living nearby the final disposal site	Public (Outside in 20% resident time)	100,000 Bq/kg *A certain distance from a landfill
	Use of the landfill site as a park	Public (200h/y)	170,000 Bq/kg *exposure dose of 10 μSv/y
	Ingestion of crops grown with underground water	Public	46,000 Bq/kg *exposure dose of 10 μSv/y

Operational period

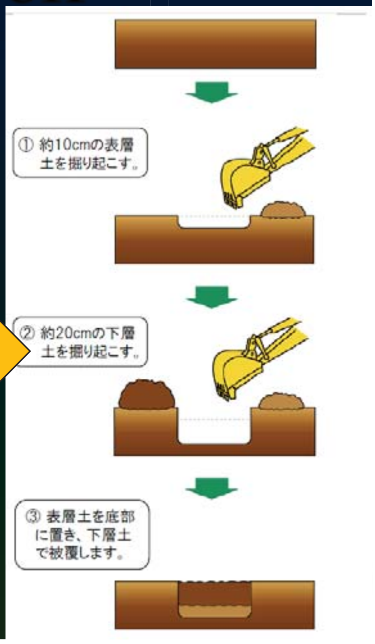
Post-closure period

http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_01.pdf

The Most Effective Way to Reduce Amount of the Contaminated Soil

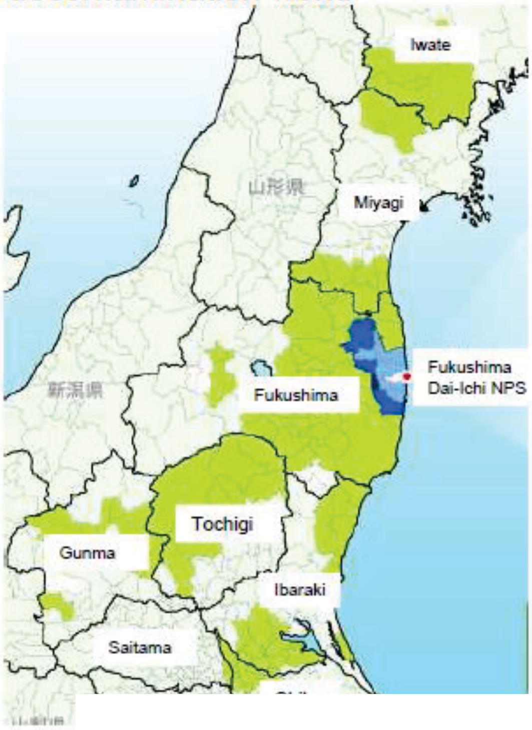
- from removing to turning over the soil
- reduce the amount of soil peeled off
- storage at the sites

MOE recommended
the above method
in their guideline
for radiation protection



Intensive Contamination Survey Area

100 municipalities, designated as Intensive Contamination Survey Area, shall implement monitoring survey and formulate the decontamination implementation plan (the plan) which stipulates area, method and contractors to implement decontamination work.



○As of the end of March 2013, the plans have been formulated in 94 municipalities.

○As decontamination target covers large area including public facilities, residential houses, roads, farmland and forest, municipalities shall clarify the objects and priorities, in consideration with the protection of public health.

⇒Decontamination work is implemented based on the Plan developed by each municipality.

http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_01.pdf

Intensive Contamination Survey Area

Implementation of decontamination is in progress along the Plans. Especially in space related to children and in public facilities, the works are getting close to the end. Continuous implementation of decontamination work is indispensable for another few years.

In Fukushima pref. (As of the end of Jan.,2013)	Ordering Ratio	Implementation Ratio
Public facility, etc.	more than 90%	approx. 75%
Residence	approx. 80%	approx. 60%
Road	approx. 75%	approx. 60%
Farmland & meadow	approx. 80%	approx. 60%
Forest(living area)	nearly 20%	less than 10%

Outside Fukushima pref. (As of the end of Dec.., 2012)	Ordering Ratio	Implementation Ratio
School, nursery school	almost ordered	more than 80%
Park, sports facility	approx. 80%	approx. 60%
Residence	approx. 40%	approx. 20%
Public facility, etc.	approx. 70%	approx. 70%
Road	approx. 60%	approx. 60%
Farmland & meadow	approx. 70%	approx. 30%
Forest(living area)	Partially ordered	Partially implemented

Note: Decontamination plans are as of the end of FY2012. The numbers could be changed according to adjustment by the municipalities.

※Please refer to the Information Site on Decontamination for individual municipality status. http://josen.env.go.jp/zone/index.html

http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_01.pdf

Background and objectives of Workshop

….. Local dose rates generally decrease as decontamination proceeds, but reservoirs of radiocaesium remain in un-remediated environments (particularly forests) and stored waste, which must be assured not to cause a risk of re-contamination. ……

What should be done for the most intractable decontamination challenges such as forest land decontamination?

Forest land should be managed periodically for Fukushima recovery

- Plantation（植栽）
- Clearance of under bush（下草刈り）
- Improvement cutting（除伐）
- Tree thinning（間伐）

Research program for Acceleration of Forest Reclamation at Fukushima by Prefectural Government

- To check effects of ordinal forest management procedure on air dose rate at forest before and after the management

Forest Management Procedure

Opening of working road

Cutting

Cross Cutting

Collection

Storage

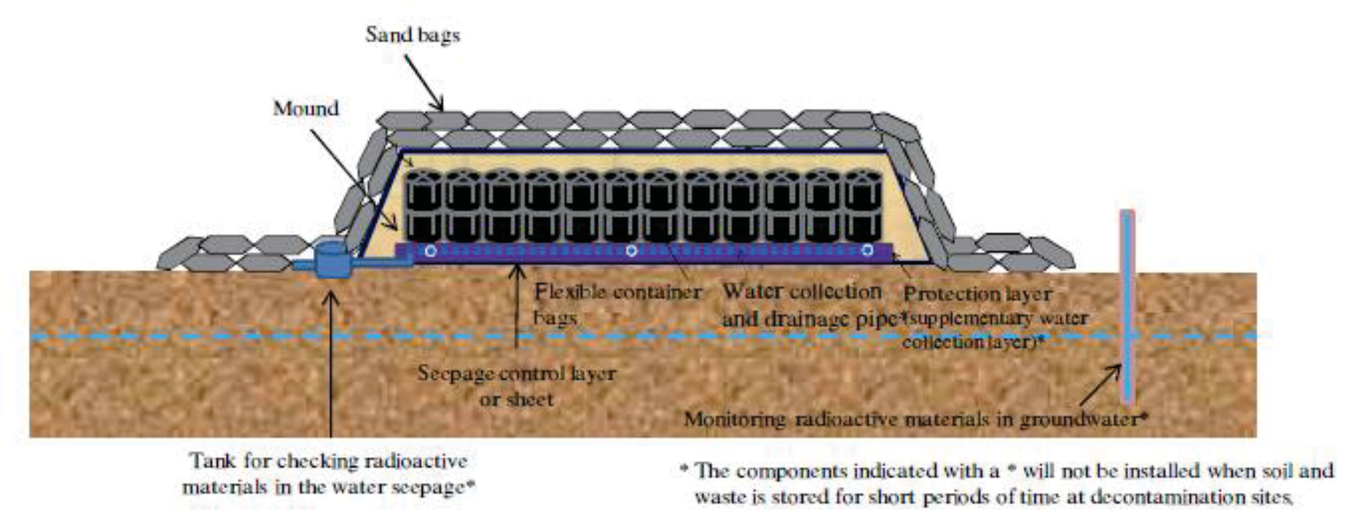
Collection of leaves and branches

Temporal Storage Site

The Research Program Teaches us…

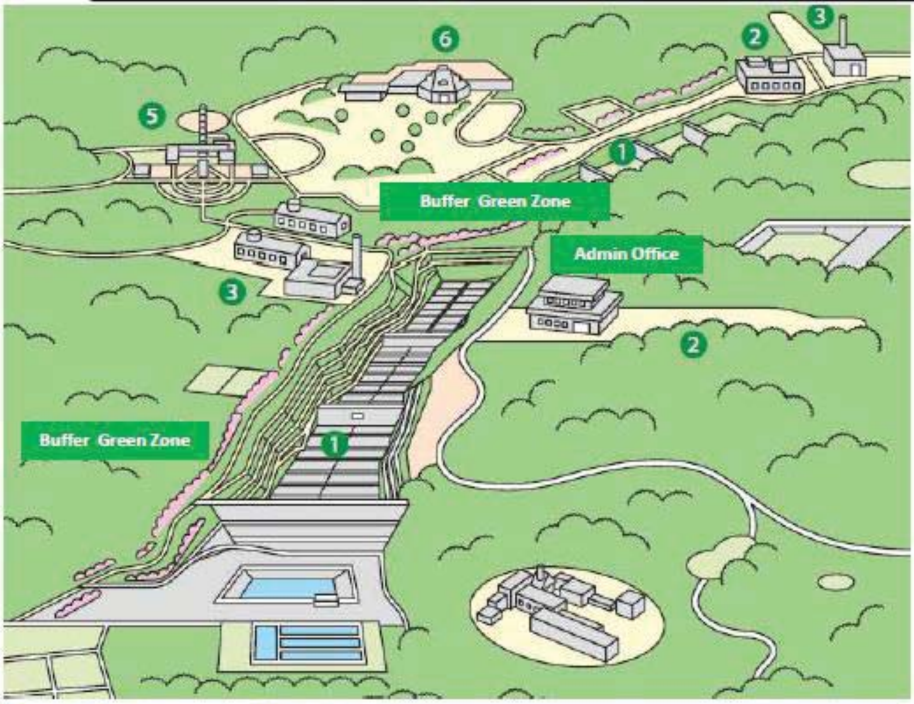
- Forest management is necessary for recovery and re-population of Fukushima regardless of necessity in decontamination
- Forest management is also necessary for prevention of soil erosion
- Approximately 10-20% reduction in air dose rate would be expected by ordinal forest management if we collect leaves and branches after the management
- Forest management for broad-leaf tree is more effective for the reduction of air dose rate. We can not expect an effective reduction of dose rate at forest management operation for Japanese cedar.
- We have to care soil erosion during the forest management procedure

Temporal Storage Sites



Present State of Temporal Storage site at Fukushima（Photo by Dr. Hatta of JIRCAS）

For Secure Interim Storage Facility



①Storage Facility

②Emplacement & Segregation Facility

③Volume Reduction Facility

④24hour monitoring Equipment(placed in several points, not specifically indicated)

⑤R & D Facility

⑥Public information Center

Scale of the whole facility (estimation)
Total storage volume ranges between 15-28 million m³, which is 12-23 times big as a baseball stadium(approx. 1.24million m³)

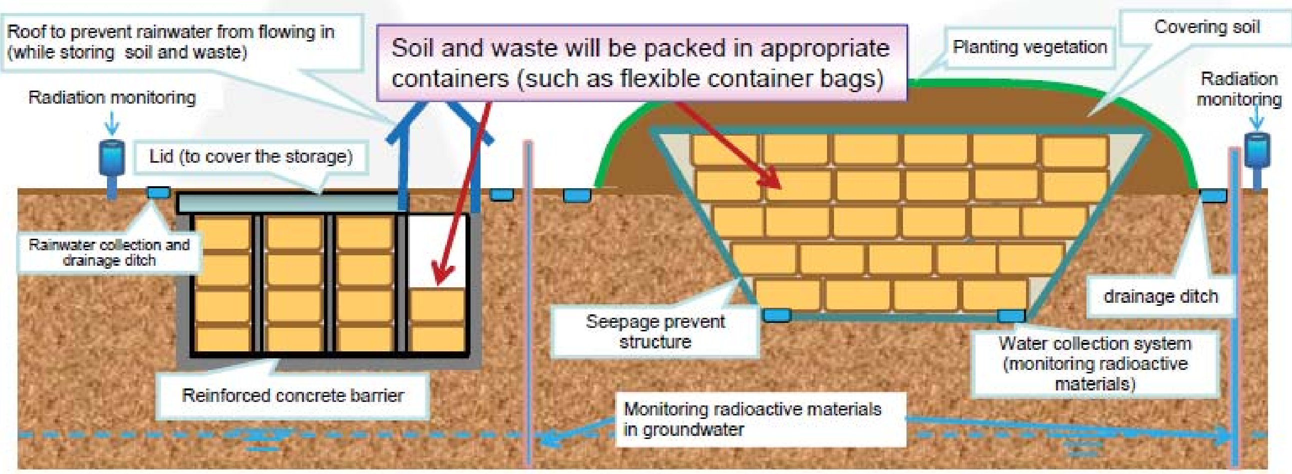
http://josen.env.go.jp/en/documents/pdf/workshop_july_17-18_2013_01.pdf

For Secure Interim Storage Facility

- Several types of storage facilities may be installed according to the characteristics of stored soil and waste.
  - ・Level of contamination
  - ・Leachate traits under various environmental scenario.

Example of facilities for radioactive waste which can generate leachate

Example of facilities for radioactive waste which does not generate leachate



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Leaching Test of Radioactive Cs from the Contaminated Soils

Sample	Sampling term	Land use	Cs-134 (Bq/kg乾)	Cs-137 (Bq/kg乾)	Cs合計 (Bq/kg乾)	Leachate by water: 溶出液 Cs-134 (Bq/L)	Leachate by water: 溶出液 Cs-137 (Bq/L)
農地土壌−1	平成24年12月	褐色森林土(畑)	2,889	5,132	8,021	ND	ND
農地土壌−2	〃	黒ボク土(畑)	6,932	12,294	19,225	ND	ND
農地土壌−3	〃	多湿黒ボク土(水田)	19,235	33,834	53,069	ND	ND
農地土壌−4	〃	灰色低地土(水田)	50,166	87,949	138,115	ND	ND
農地土壌−5	〃	褐色森林土(樹園地)	59,525	104,762	164,287	ND	ND
宅地土壌−1	平成23年12月	礫まじり砂質細粒土	683	1,311	1,994	ND	ND
宅地土壌−2	〃	砂礫質細粒土	1,348	2,416	3,764	ND	ND
宅地土壌−3	〃	礫まじり細粒土	2,592	4,615	7,207	ND	ND
宅地土壌−4	〃	細粒土	3,365	6,134	9,500	ND	ND
宅地土壌−5	〃	礫まじり砂質細粒土	4,028	7,359	11,387	ND	ND
宅地土壌−6	平成24年4月	砂礫質細粒土	4,018	7,596	11,614	ND	ND

Effects of Co-existing Cations on Leaching of Cs

Leaching %

試験溶液: 共存イオン	共存イオン濃度 [mol/L]	共存イオン濃度 [mg/L]	放射性Cs(134+137) 溶出濃度 [Bq/L]	溶出率 [%]	備考
NH_4^+	1×10^{-4}	1.8	52	0.6	作付水田土壌溶液中の濃度 2×10^{-4}～6×10^{-4} [mol/L] (3.6～10.8[mg/L])
	1×10^{-3}	18	−	−	
	1×10^{-2}	180	303	3.7	
	1×10^{-1}	1800	723	8.7	
	1	18000	987	9.9	
K^+	1×10^{-3}	39	38	0.4	作付水田土壌溶液中の濃度 6×10^{-5}～2×10^{-4} [mol/L] (2.3～7.8 [mg/L])
	1×10^{-2}	391	83	1.0	
	1×10^{-1}	3910	486	6.0	
	1	39100	789	9.8	
Na^+	5×10^{-3}	115	38	0.4	
	5×10^{-2}	1150	54	0.7	
	5×10^{-1}	11500	210	2.6	海水濃度相当

Effects of NH_4^+ ion on Leaching of Cs

NH_4^+濃度		Sample 1		Sample 2		Sample 3		Sample 4	
		11,614[Bq/kg]		53,069[Bq/kg]		138,115[Bq/kg]		164,287[Bq/kg]	
[mol/L]	[mg/L]	溶出濃度 [Bq/L]	溶出率[%]	溶出濃度 [Bq/L]	溶出率[%]	溶出濃度 [Bq/L]	溶出率[%]	溶出濃度 [Bq/L]	溶出率[%]
1×10^{-4}	1.8	ND	<3.0	30	0.9	52	0.6	36	0.3
1×10^{-3}	18	ND	<3.1	41	1.2	-	-	80	0.7
1×10^{-2}	180	ND	<3.2	81	2.6	303	3.7	174	1.5
1×10^{-1}	1800	44	4.7	208	7.1	723	8.7	667	5.7
1	18000	90	11.3	340	10.0	987	9.9	1278	9.2

Effects of pH, Solidification Agent (SA), Humic Acid and Temperature ion on Leaching of Cs

酸・アルカリ，固化剤の影響（農地土壌－4／138,115Bq/kg）

	試験溶液 項目	試験溶液 範囲	放射性Cs(134+137) 溶出濃度 [Bq/L]
pH	初期pH	4	ND
		7	ND
		12	ND
SA MgO	固化剤（MgO）	1 wt%	ND
		4 wt%	ND
		10 wt%	ND
SA CaO	固化剤（CaO）	1 wt%	ND
		3 wt%	ND
		5 wt%	ND

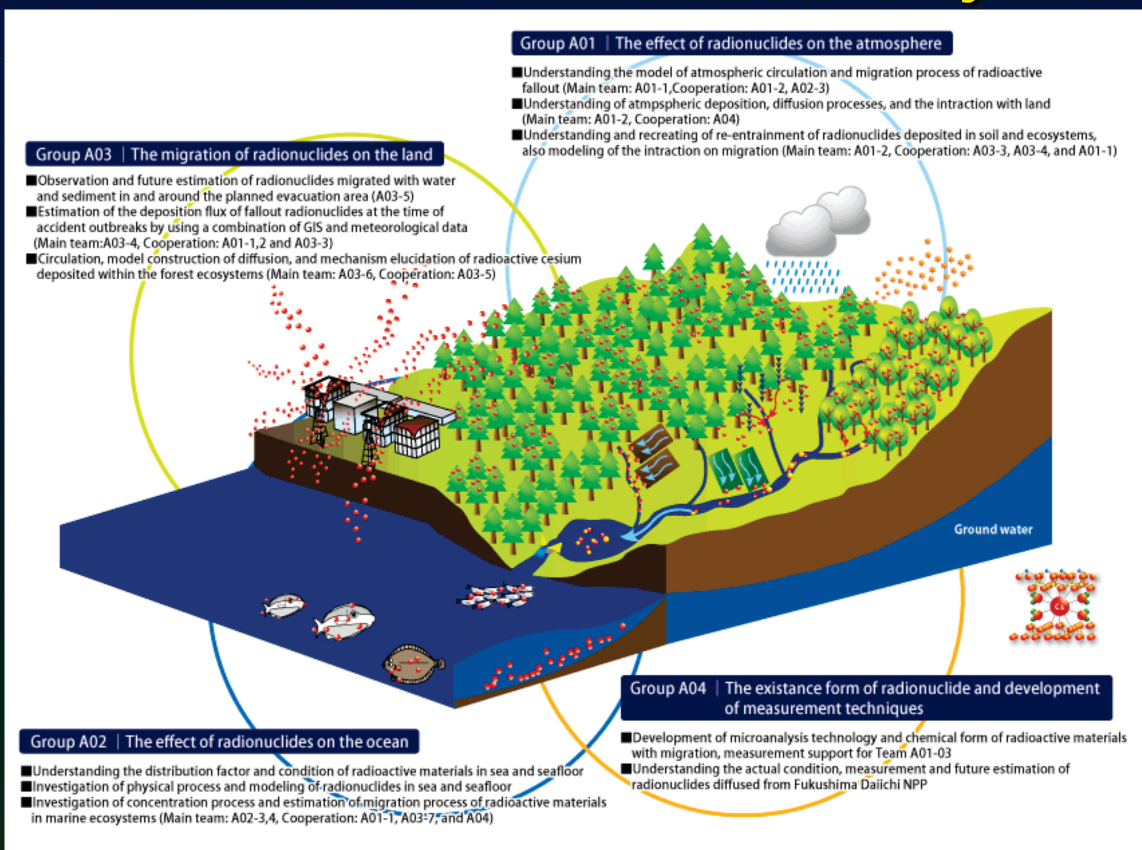
フミン酸，温度の影響（農地土壌－4／138,115Bq/kg）

	試験溶液 項目	試験溶液 範囲	放射性Cs(134+137) 溶出濃度 [Bq/L]
Humic Acid	フミン酸	10 ppm	ND
		50 ppm	ND
		100 ppm	ND
Temp.	温度	10 ℃	ND
		25 ℃	ND
		60 ℃	41

Projects for Understanding of Cs Behavior at Fukushima: F-TRACE by JAEA

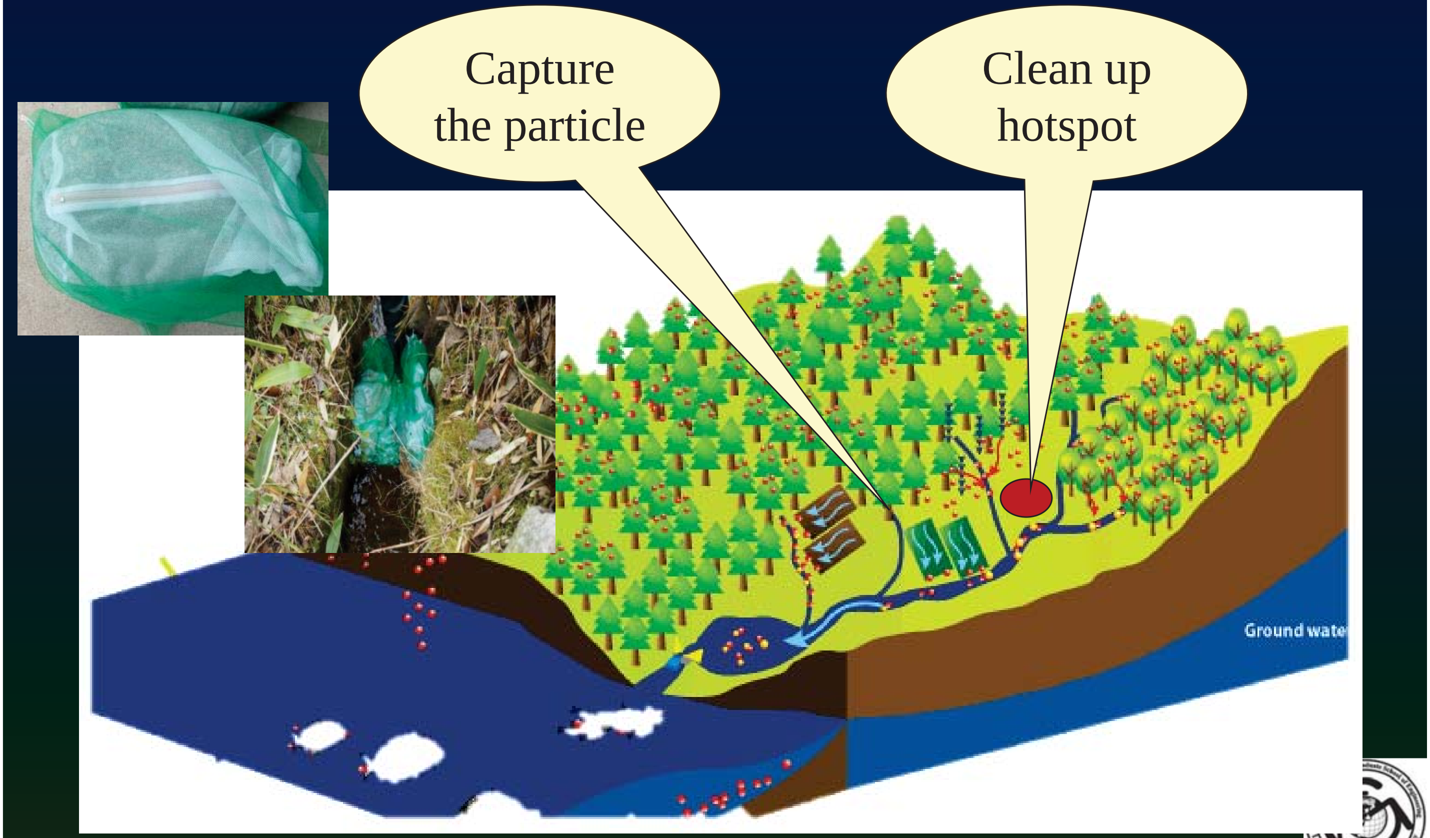
http://fukushima.jaea.go.jp/initiatives/cat01/pdf01/4.pdf

Projects for Understanding of Cs Behavior at Fukushima: ISET-R founded by MEXT

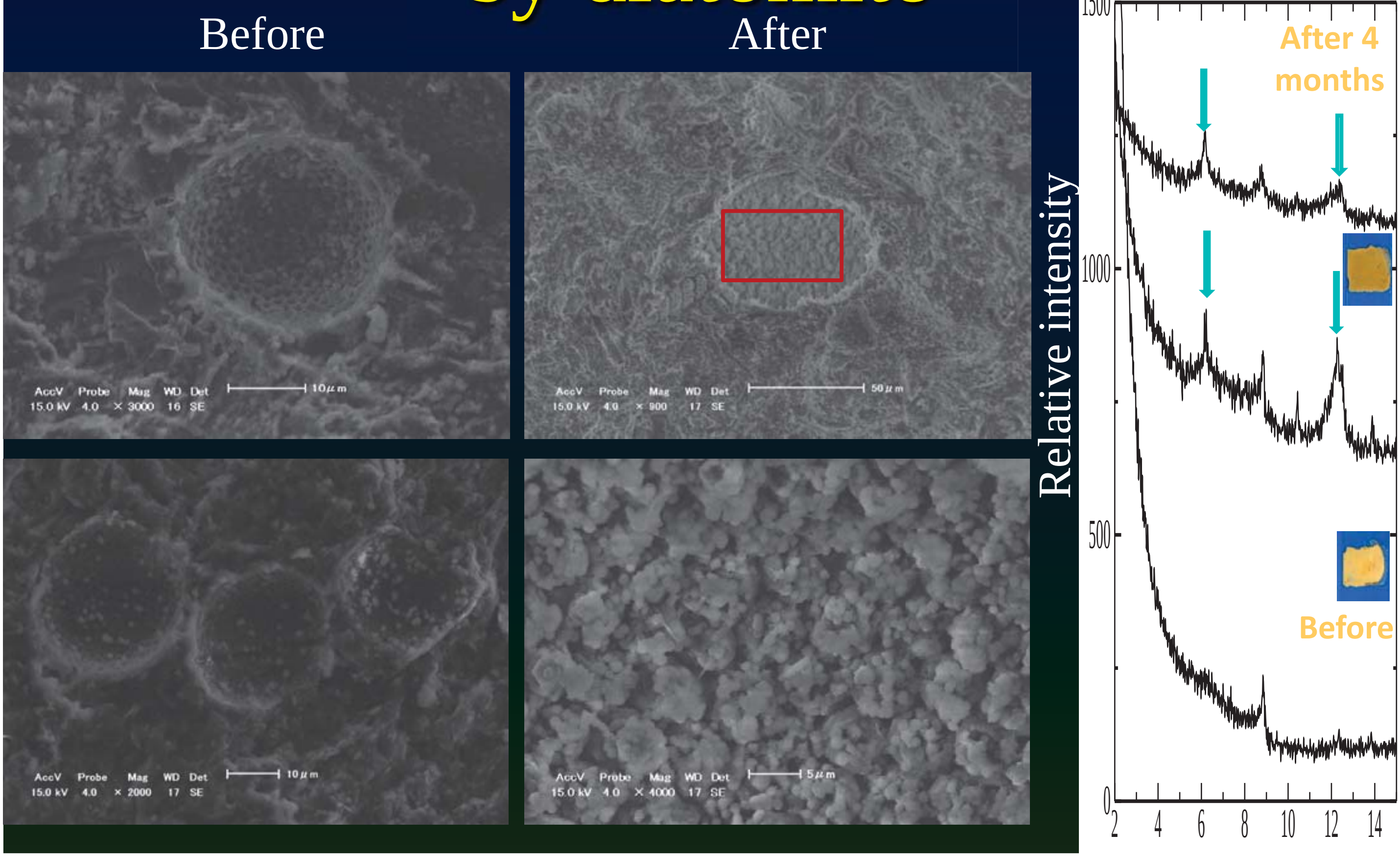


http://isetr.suiri.tsukuba.ac.jp/ISETRen/researchEN.html

Not only active decontamination but also passive decontamination should be considered



Capture the contaminated particles by diatomite



Summary

- Understanding of Clay-Cs association is very important for volume reduction of the waste, reliable storage and disposal of the waste.
- Conduct a systematic analysis of the existing performance data to identify potential factors or practices that could improve effectiveness of future decontamination efforts
- Advanced analyses about clay-Cs association should make answers to questions and recommendations on
  - Why Cs release from clay in the case of coexisting compete cations? Is this always or not?
  - Is Cs released from the clay particles at mixing points between river and sea water? If yes, why?