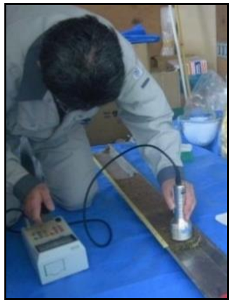
MRS2017_Scientific Basis for Nuclear Waste Management XLI,
29 October-3 November, 2017, Sydney, AUSTRALIA（ID:75）

Apparent Diffusion Coefficient Consistent with Sorption Derived Based on Changes in Depth Profiles of Radiocaesium in Soil Contaminated by the Fukushima Nuclear Accident and Future Prediction of Depth Profile







Haruo SATO [1] & Masato YOSHII [1]

1 Graduate School of Natural Science & Technology,
Okayama University, JAPAN

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Introduction

Introduction (1/2)

- The accident at the TEPCO Fukushima Daiichi NPP occurred following the Great East Japan Earthquake in March 2011, and led to the release of volatile radionuclides (RNs), which were deposited on the environment (soils, forests, residential land, etc.) in the Fukushima and the surrounding prefectures.

- Because radiocaesium (Cs-134 & 137) are now the main contributors to radiation dose rate after the decay of short-lived I-131, behaviour of Cs in the environment is important for future prediction.

- The authors conducted twice field investigations on the depth profiles of RNs in soil in the environment of Fukushima after about 3 months (1st investigation) and about 1 year (2nd investigation) following the 1F-NPP accident and reported the apparent diffusion coefficients (D_a) of RNs (Cs-134 & 137, Te-129m, Ag-110m) obtained from the depth profiles and the distribution coefficients (K_D) of RNs (Cs-137 & I-131) obtained from batch experiments so far [e.g., Sato et al., 2013, 2014; Sato, 2015].

Introduction (2/2)

- Consequently, almost all D_a-values of radiocaesium (Cs-134 & 137) were approximately of order 10^{-11}-10^{-10} m^2/s and the batch K_D-values of Cs-137 ranged 2,080-61,000 ml/g in both investigations.
- However, large discrepancies were found between D_a-values & K_D-values.

  This cause was considered to be due to that depth profiles obtained in both field investigations included the effect of dispersion by rainwater flow near the surface layer of soil which was unsaturated. This indicates that depth profiles in the deeper part of soil which was almost saturated were formed by mainly diffusion process.
- Therefore, in this study, the D_a-values of radiocaesium consistent with sorption (K_D) were derived based on change in the depth profile.

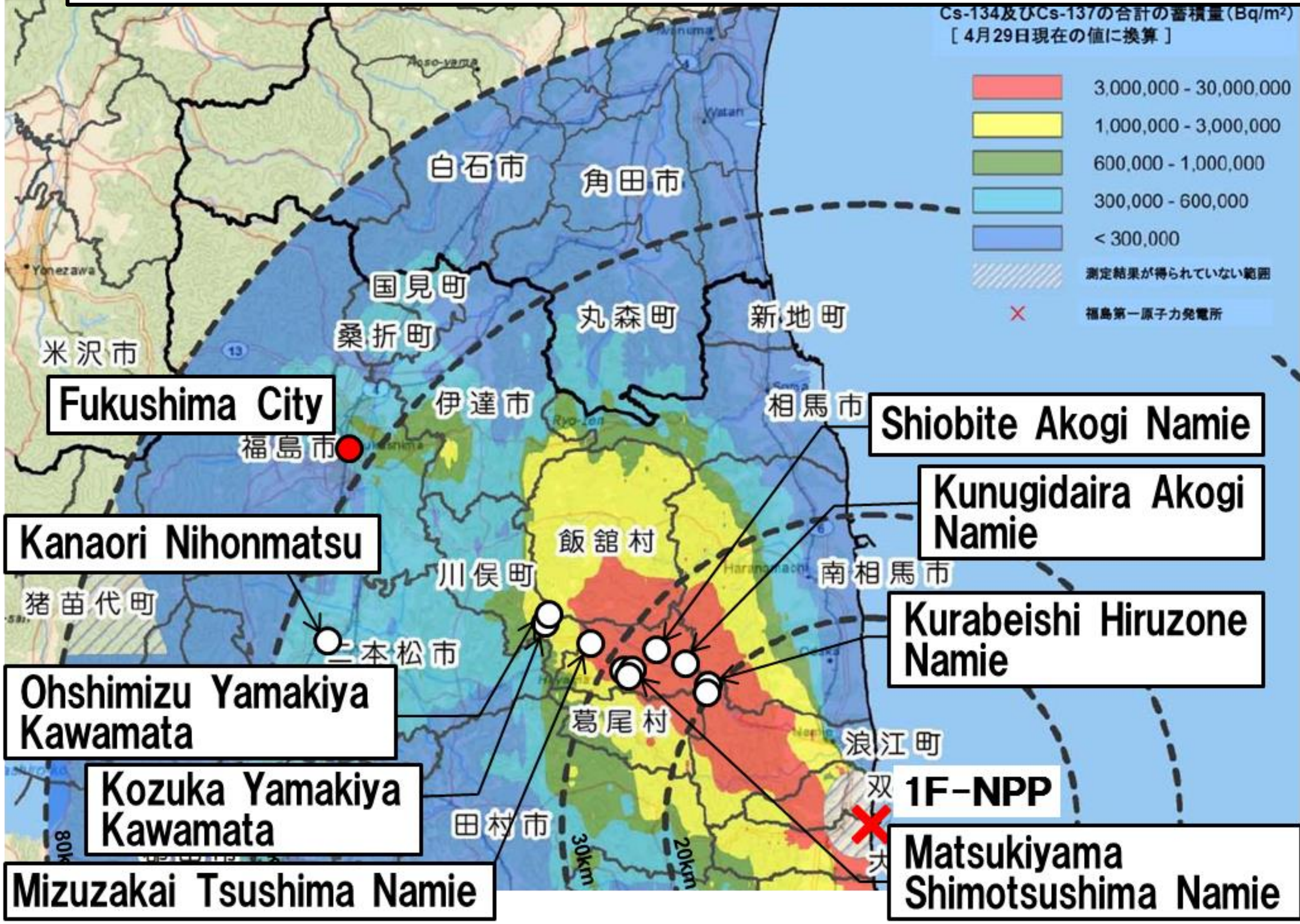
  Furthermore, simulation of the depth profiles of radiocaesium was conducted up to 30 years later after the 1F-NPP accident.

Overview of the Field Investigation

Investigation Location

Field Investigation

Depth profile investigation of RNs in soil conducted twice at 11 locations in the city of Nihonmatsu & the towns of Kawamata & Namie, which are located between 20km and 60km from 1F-NPP



1st: ca. 3 months following accident

2nd: ca. 1 year following accident

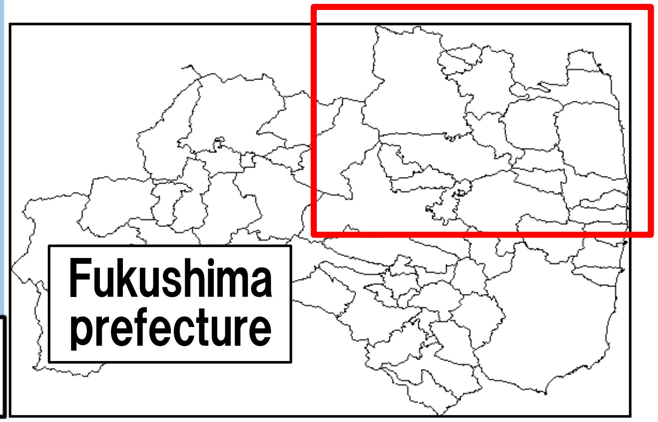
Investigation: 11 locations

(10 locations overlapped)

Kanaori Nihonmatsu

Kawamata(Kuchibuto river basin)

Namie (Ukedo river basin)



Sample Extraction in the Field

Drilling by handheld soil sampler (15cm in inner diameter)

An example of soil core (1/3 of the rod opens)

Sampling for lab. exps. (physical property, mineral analysis, sorption-desorption exps., etc.)

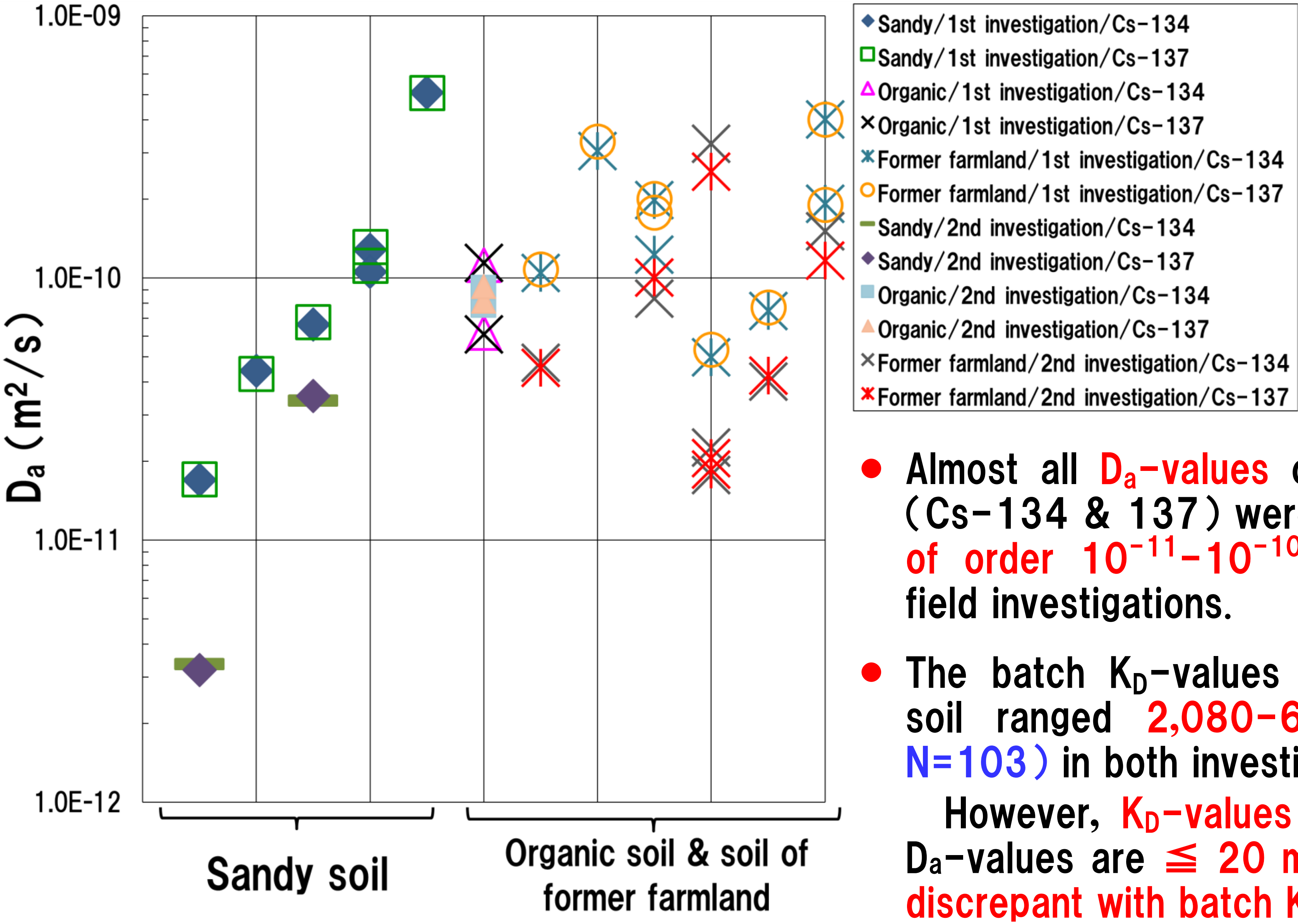
Hydraulic conductivity measurement by water head

Soil samples for lab. exps. (physical property, mineral analysis, sorption-desorption exps., etc.)

- Obtain core samples of about 40-50cm depth by core sampler.
- Obtain samples for lab. exps. (soil physical property, mineral analysis, sorption exps., RN analysis, etc.).
- Obtain samples for concentration profile (2cm pitch up to 20cm depth, 4cm pitch up to 40cm depth, 5cm pitch in the part deeper than 40cm).

An Example of the Investigation Results

D_a-values of Cs for each type of soil calculated from depth profiles



- Almost all D_a-values of radiocaesium (Cs-134 & 137) were approximately of order 10^{-11}–10^{-10} m^2/s in both field investigations.
- The batch K_D-values of Cs-137 on soil ranged 2,080-61,000 (ml/g, N=103) in both investigations.

  However, K_D-values expected from D_a-values are ≦ 20 ml/g and largely discrepant with batch K_D-values.

Derivation of D_a consistent with K_D based on Change in Depth Profile

Normal Analysis of D_a

Diffusion eq. & analytical solution for one-dimensional non-steady state of a planar source consisting of a limited amount of substance

$$\frac{\partial C}{\partial t} = D_a \frac{\partial C}{\partial x^2} - \lambda C = \left\{ \frac{n_p D_p}{n_p + \rho_{th}(1 - n_p)K_D} \right\} \frac{\partial C}{\partial x^2} - \lambda C$$

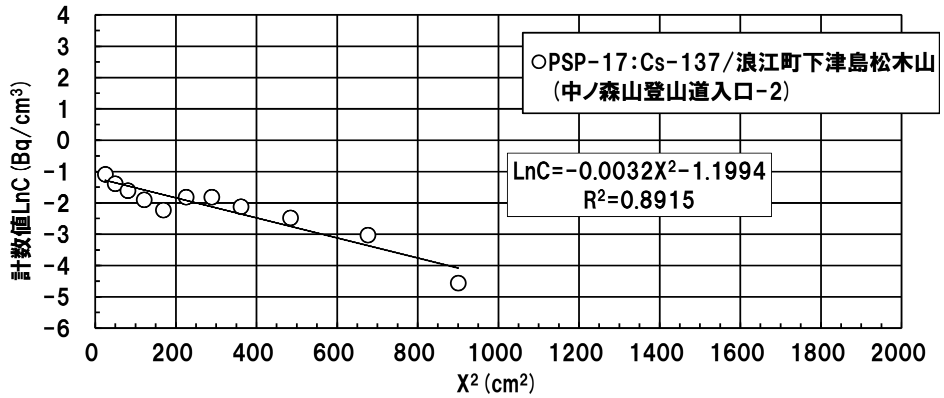
Initial condition $C(t, x) = 0, t = 0, \quad M = \int_0^{+\infty} C\, dx$

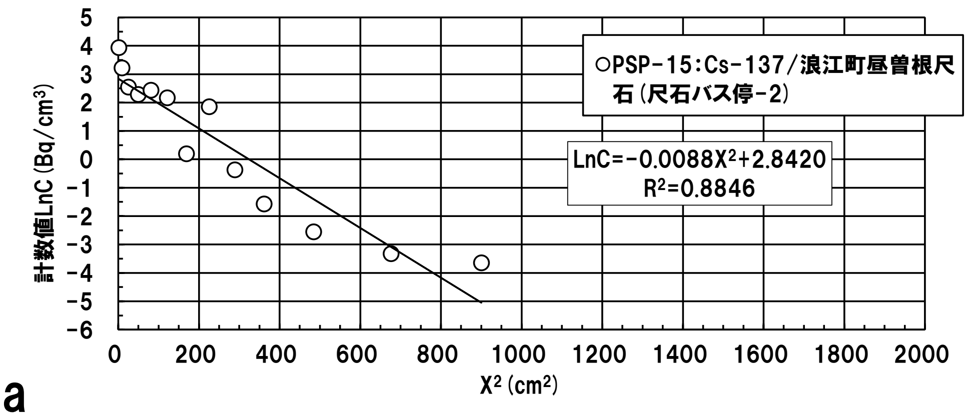
Boundary condition $C(t, x) = 0, t > 0, x = +\infty$

$$C = \frac{M}{\sqrt{\pi D_a t}} exp\left(-\frac{x^2}{4D_a t} - \lambda t\right)$$

$$\ln C = \ln\left(\frac{M}{\sqrt{\pi D_a t}}\right) - \frac{x^2}{4D_a t} - \lambda t$$

C: concentration   t: time   x: depth (distance)
D_p: diffusivity in porewater n_p: porosity
ρ_{th}: solid density K_D: distribution coefficient
λ: decay constant
M: total amount of diffusing substance per unit area





Analysis of D_a from Change in Depth Profile

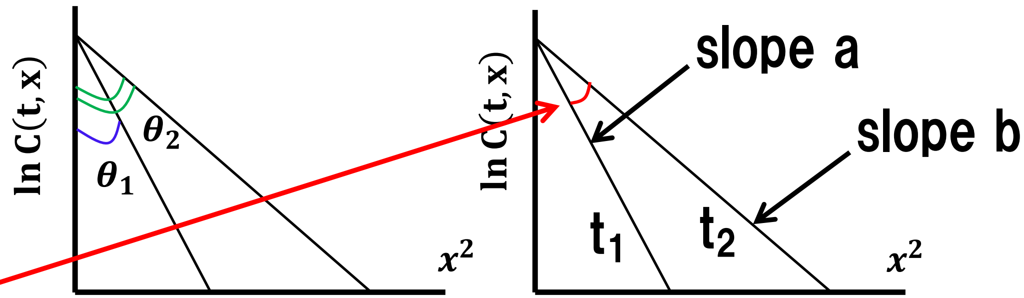
Logarithm of analytical solution

$$\ln C = \ln\left(\frac{M}{\sqrt{\pi D_a t}}\right) - \frac{x^2}{4D_a t} - \lambda t$$

Assuming t_1 & t_2 ($t_2 > t_1$) as elapsed time after the deposition of radiocaesium, slopes a & b given by a plot of lnC versus x^2 can be given by the following Eqs. as θ_1 & θ_2 for angle to vertical axis (lnC), respectively

$$a = -\frac{1}{4D_{a1}t_1} = \frac{1}{\tan\theta_1}$$

$$b = -\frac{1}{4D_{a2}t_2} = \frac{1}{\tan\theta_2}$$



The change quantity of slope against time change from t_1 to t_2 ($t_2 > t_1$) is given by the following relation

$$\frac{1}{\tan(\theta_2 - \theta_1)} = \frac{1}{4D_a(t_2 - t_1)}$$

$$D_a = -\frac{\tan\left\{\arctan\left(\frac{1}{b}\right) - \arctan\left(\frac{1}{a}\right)\right\}}{4(t_2 - t_1)} = -\frac{\tan\{\arctan(-4D_{a2}t_2) - \arctan(-4D_{a1}t_1)\}}{4(t_2 - t_1)}$$

D_{a1} & D_{a2}: apparent diffusion coefficient for elapsed times t_1 & t_2

Calculation of K_D from D_a

Relation between D_a & K_D for typical porous media

$$D_a = \frac{n_p D_p}{n_p + \rho_d K_D} = \frac{n_p D_p}{n_p + \rho_{th}(1 - n_p)K_D} = \frac{n_p D^o}{n_p + \rho_{th}(1 - n_p)K_D}\left(\frac{\delta}{\tau^2}\right)$$

$$K_D = \frac{n_p(D^o - 3D_a)}{3\rho_{th}(1 - n_p)D_a}$$

D^o: diffusion coefficient in free water
δ: constrictivity (in the case of no interaction of ions with pore wall, $\delta = 1$)
τ^2: tortuosity

Analytical condition of K_D

Parameter	Input information & value
n_p (porosity)	Measured data of soil sampled at each investigation location
ρ_{th} (solid density)	
δ (constrictivity)	When pore size is large enough compared to diffusion species (ions), approximately $\delta = 1$
τ^2 (tortuosity)	For uniform and typical porous media, geometrically $\tau^2 = 3$
D^o (m^2/s)	D^o for Cs^+ ions: $D^o = 2.1 \times 10^{-9}$ (m^2/s, 25℃)

Calculated Results of D_a & K_D

Calculations of D_a & K_D for each location based on data obtained in twice investigations

$$D_a = -\frac{\tan\left\{\arctan\left(\frac{1}{b}\right) - \arctan\left(\frac{1}{a}\right)\right\}}{4(t_2 - t_1)} = -\frac{\tan\{\arctan(-4D_{a2}t_2) - \arctan(-4D_{a1}t_1)\}}{4(t_2 - t_1)}$$

$$K_D = \frac{n_p(D^o - 3D_a)}{3\rho_{th}(1 - n_p)D_a}$$

Investigation location	D_a (m^2/s)		K_D (ml/g)	
	Cs-134	Cs-137	Cs-134	Cs-137
Kanaori Nihonmatsu C.	2.76E-14	2.72E-14	9.66E+3	9.82E+3
Kurabeishi Hiruzone Namie T.	1.98E-14	1.97E-14	1.98E+4	1.99E+4
	4.53E-14	4.64E-14	8.67E+3	8.46E+3
Oshimizu Yamakiya Kawamata T.	1.58E-14	1.36E-14	1.32E+4	1.54E+4
Mizuzakai Tsushima Namie T.	6.70E-15	8.38E-15	9.61E+4	7.69E+4
	1.75E-14	1.06E-14	3.68E+4	3.04E+4
Matsukiyama Shimo-tsushima Namie T.	2.94E-14	2.07E-14	2.14E+4	1.67E+4
Shiobite Akogi Namie T.	2.37E-14	2.35E-14	1.65E+4	1.67E+4
Kunugidaira Akogi Namie T.	1.22E-14	1.06E-14	2.04E+4	2.37E+4

$K_D = 10^3 \sim 10^5$ ml/g

Consistent

Batch K_D-values

K_D=2,080～61,000 (ml/g)

(N=103)

Simulation of Change in Depth Profile of Cs in Soil

Numerical Analysis

Governing eq. (D const.)

$$\frac{\partial C}{\partial t} = D\frac{\partial C}{\partial x^2} - \lambda C$$

Initial condition $C(t,x) = 0, t = 0, x > 0$

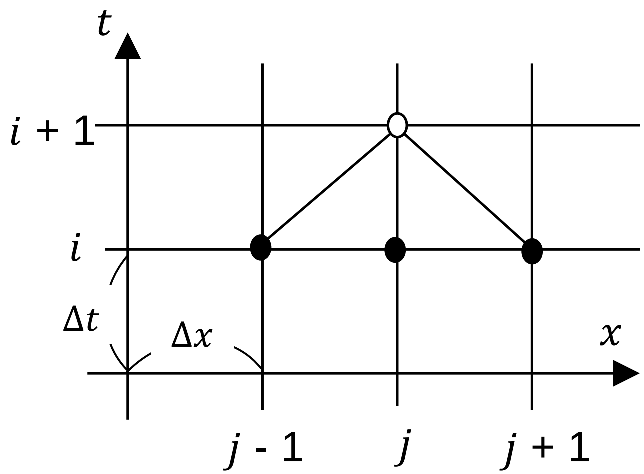
$$M = \int_0^{+\infty} C\, dx$$

$D = D_{a2}$: $0 \le t \le 1y$
$D = D_a$: $1y < t \le 30y$

} 2 steps of analyses considered change in D_a

Boundary condition $C(t,x) = 0, t > 0, x = +\infty$

Discretization by difference method



$$\frac{C_j^{i+1} - C_j^i}{\Delta t} = D\frac{C_{j+1}^i - 2C_j^i + C_{j-1}^i}{(\Delta x)^2} - \lambda C_j^i$$

$$\therefore C_j^{i+1} = C_j^i + \frac{\Delta t}{(\Delta x)^2} D\left(C_{j+1}^i - 2C_j^i + C_{j-1}^i\right) - \lambda C_j^i \Delta t$$

$$C_j^i = C\left(t_i, x_j\right),\quad t_i = i\Delta t,\quad x_j = j\Delta x,\quad i = 0,1,2,3,\cdots,\quad j = 0,1,2,3,\cdots$$

Initial condition $C_0^0 = C_o,\quad C_j^0 = 0,\quad j = 1,2,3,\cdots$

Boundary condition $C_0^i = C_o - \lambda C_o i\Delta t - \sum_{j=1}^{n} C_j^i,\ i = 1,2,3,\cdots$

Condition for preventing numerical dispersion

$$\frac{\Delta t}{(\Delta x)^2} D + \frac{\lambda \Delta t}{2} \le \frac{1}{2}$$

Analytical Condition & Input Data

Analytical condition

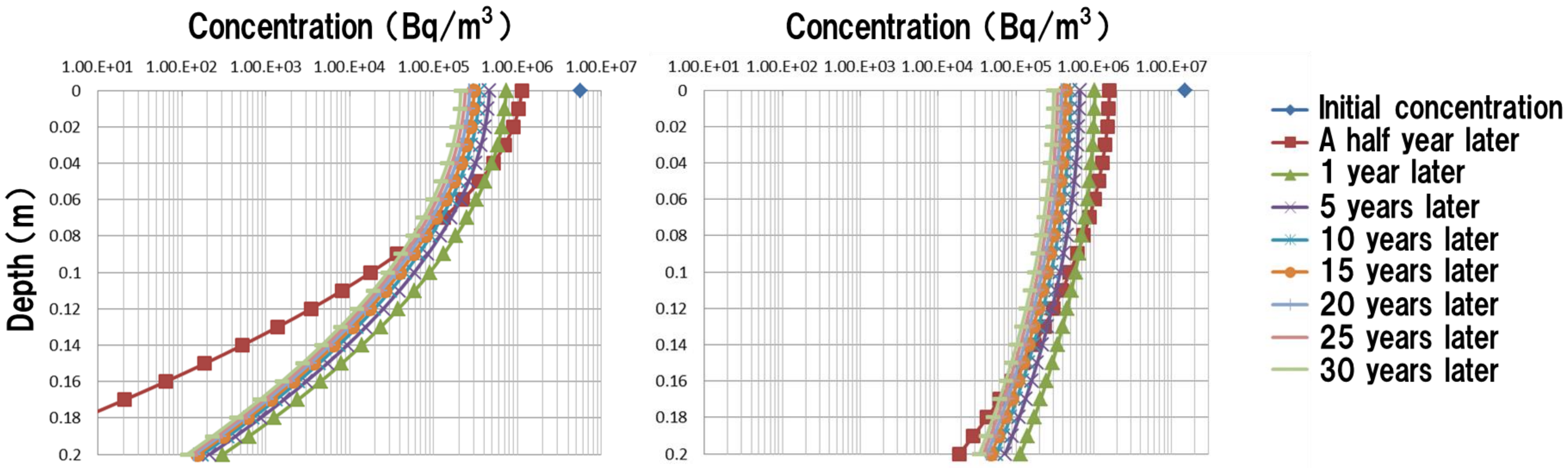
Analytical location: 9 locations
Δt (time step): 86,400sec (=1d)
Δx (distance step): 0.01m (=1cm)
$D=D_{a2}$ ($t \leqq 1y$): D_{a2} data for each location (calculated from depth profile after 1y)
$D=D_a$ ($1y < t \leqq 30y$): data for each location

Input data to each location for simulation of depth profile

Investigation location	D_{a2} (m^2/s) ($t \leqq 1y$)		D_a (m^2/s) ($t > 1y$)		Inventory M (Bq/m^2)	
	Cs-134	Cs-137	Cs-134	Cs-137	Cs-134	Cs-137
Kanaori Nihonmatsu C.	3.41E-11	3.51E-11	2.76E-14	2.72E-14	9.34E+5	9.47E+5
Kurabeishi Hiruzone Namie T.	7.85E-11	8.17E-11	1.98E-14	1.97E-14	1.29E+7	1.26E+7
	7.85E-11	8.17E-11	4.53E-14	4.64E-14	8.36E+6	8.28E+6
Oshimizu Yamakiya Kawamata T.	4.75E-11	4.54E-11	1.58E-14	1.36E-14	6.19E+5	6.01E+5
Mizuzakai Tsushima Namie T.	8.32E-11	1.01E-10	6.70E-15	8.38E-15	6.97E+5	7.11E+5
	8.32E-11	1.01E-10	1.75E-14	1.06E-14	4.03E+5	4.15E+5
Matsukiyama Shimotsushima Namie T.	2.26E-11	2.05E-11	2.94E-14	2.07E-14	2.80E+6	2.77E+6
Shiobite Akogi Namie T.	4.03E-11	4.23E-11	2.37E-14	2.35E-14	2.78E+5	2.78E+5
Kunugidaira Akogi Namie T.	1.51E-10	1.16E-10	1.22E-14	1.06E-14	7.49E+6	7.45E+6

Simulated Results of Depth Profile

Examples of the simulated results of change in depth profile (Cs-134+137)



Change in depth profile of radiocaesium (Cs-134+137) (left: Shiobite Akogi Namie Town & right: Kunugidaira Akogi Namie Town)

- Depth profiles in soil gradually extended with time, but little change in the formation of the depth profiles is found for 30yrs (this is due to that D_a of Cs is very slow).
- This is consistent with the fact that much percentage of radiocaesium still exists near the surface layer of soil, even now it passed over 6 years after the accident.

Conclusions

- Derived D_a consistent with sorption (K_D) from change in depth profile.
- Simulated change in the depth profiles of radiocaesium (Cs-134 & 137) up to 30 years later after the 1F-NPP accident.
- Moreover, simulated change in air dose rate based on the simulated results of the depth profiles of radiocaesium. The details will be reported in next time.

Thank you for your attention !

Acknowledgements

- The field investigations were conducted as part of the contract work of the Ministry of Education, Culture, Sports, Science and Technology (MEXT).
- The authors would like to thank many scientists and staff of Japan Atomic Energy Agency (JAEA) for conducting the field work, laboratory experiments and documentation together.

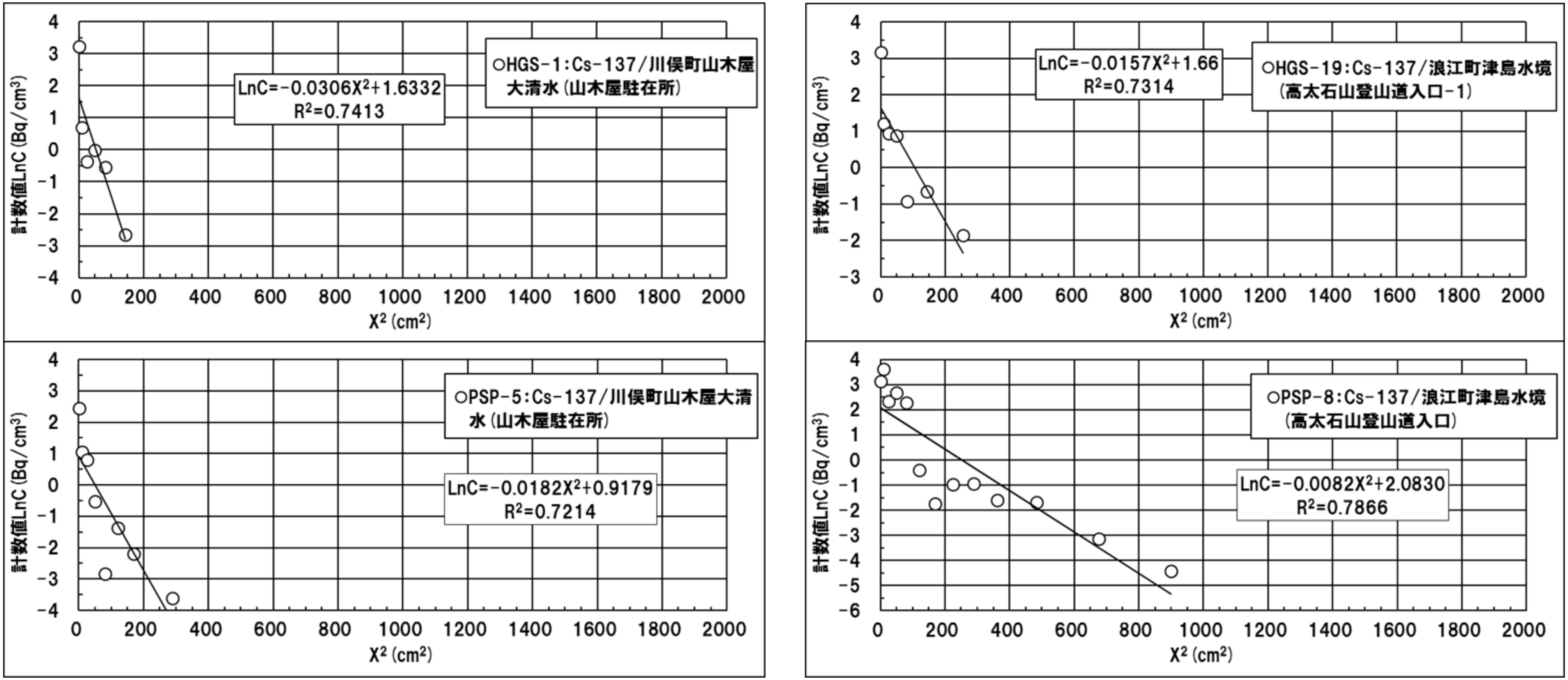
APPENDIX

Investigation Outline

Investigation item	1st investigation	2nd investigation
Investigation (sampling) date	June 7-19 (June 10-16), 2011	Feb. 23-Mar. (Feb. 27-Mar. 8) 10, 2012
Investigation location	11 locations Nihonmatsu C., Kawamata T., Namie T.	11 locations Nihonmatsu C., Kawamata T., Namie T.
Depth distribution	259 samples (16 slices: 11 locations) analysis: Ge semiconductor detector	180 samples (12 cores: 11 locations) analysis: Ge semiconductor detector
Extraction (drilling) method	Geoslicer (wide & handheld): 29 slices wide-sized: 1 slice at 1 location 1.1m wide×1m deep×2cm thick handheld: 28 slices at 11 locations 10cm wide×1m deep×2cm thick	Core sampler (handheld): 19 cores 15cm inner diameter×50cm deep
Soil observation/description	29 slices (all slices)	19 cores (all cores)
Soil physical property	24 samples (11 locations) 2 depth intervals/slice density, porosity, water content, etc.	24 samples (11 locations) 2 depth intervals/core density, porosity, water content, etc.
Classification by elutriation		12 samples (3 locations): 3 cores 3 types of soils (sandy, clayey, organic) clay (<2μm), silt (2-20μm), sand (>20μm)
Mineral analysis (XRD)		12 samples (3 locations): 3 cores 3 types of soils (sandy, clayey, organic) oriented (clay), un-oriented (all samples)
Chemical property (CEC & AEC)		12 samples (3 locations): 3 cores 3 types of soils (sandy, clayey, organic) elutriated samples (clay, silt, sand)
Sorption-desorption exp. (^{137}Cs & ^{131}I)	24 samples (11 locations)(only sorption) 2 depth intervals/slice sorption: ionized water	12 samples (3 locations): 3 cores 3 types of soils & elutriated samples sorption: ionized water, desorption: 0.33M-KCl

Examples of Depth Profiles

Correlations of lnC versus x^2 for Cs-137



Upper: 3 months (t_1), lower: 1 year (t_2) after accident (Yamakiya Kawamata)

Upper: 3 months (t_1), lower: 1 year (t_2) after accident (Mizuzakai Namie)

- Slopes of lnC versus x^2 decrease with increasing time.
- Based on change in slope between times t_1 & t_2, D_a can be calculated.

Calculated Results of D_a from Depth Profile

D_a-values of Cs for each type of soil calculated by normal method

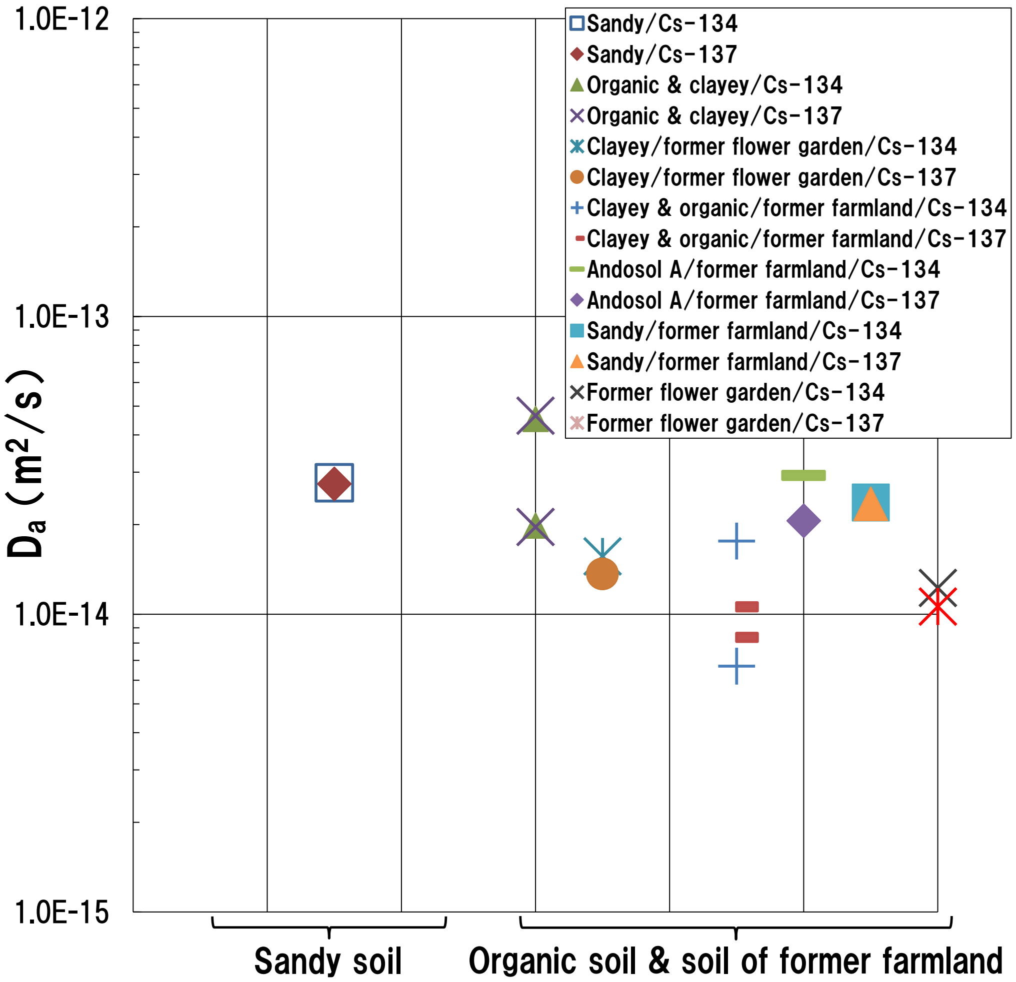
Investigation location	Soil type	D_{a1} (m^2/s) 1st investigation (after 3 months)		D_{a2} (m^2/s) 2nd investigation (after 1 year)	
		Cs-134	Cs-137	Cs-134	Cs-137
Kozuka Yamakiya Kawamata T.	Sandy soil	1.70E-11	1.71E-11	3.38E-12	3.19E-12
		4.43E-11	4.31E-11		
Kanaori Nihonmatsu C.		6.63E-11	6.78E-11	3.41E-11	3.51E-11
Matsukiyama Shimotsushima Namie T.		1.28E-10	1.32E-10		
		1.06E-10	1.12E-10		
Teshichiro Akogi Namie T.		5.09E-10	5.09E-10		
Kurabeishi Hiruzone Namie T.	Organic soil	1.12E-10	1.14E-10	7.85E-11	8.17E-11
		6.17E-11	6.10E-10		
				9.18E-11	9.28E-11
Ohshimizu Yamakiya Kawamata T.	Soil of former farmland (including flower garden)	1.04E-10	1.08E-10	4.75E-11	4.54E-11
Kozuka Yamakiya Kawamata T.		3.05E-10	3.31E-10		
Mizuzakai Tsushima Namie T.		1.98E-10	2.01E-10	8.32E-11	1.01E-10
		1.23E-10	1.78E-10		
Matsukiyama Shimotsushima Namie T.		4.97E-11	5.33E-11	2.26E-11	2.05E-11
				3.25E-10	2.54E+10
				1.77E-11	1.86E-11
Shiobite Akogi Namie T.		7.49E-11	7.71E-11	4.03E-11	4.23E-11
Kunigidaira Akogi Namie T.		1.92E-10	1.91E-10	1.51E-10	1.16E-10
		4.03E-10	4.03E-10		

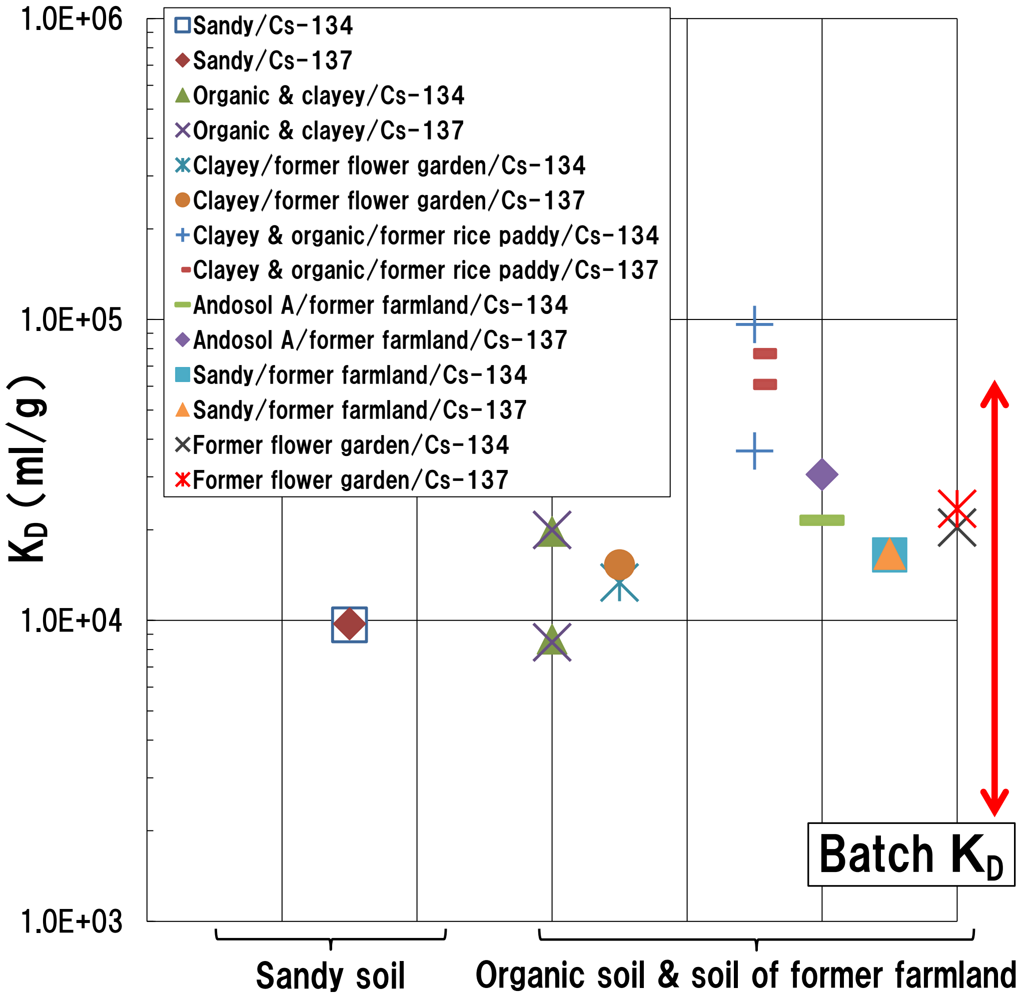
Calculated Results of D_a & K_D

D_a & K_D-values of Cs for each type of soil calculated based on change in depth profile between twice investigations

Investigation location	Soil type	Physical property of soil			D_a (m^2/s)		K_D (ml/g)	
		Porosity (%)	Dry density (Mg/m^3)	Solid density (Mg/m^3)	Cs-134	Cs-137	Cs-134	Cs-137
Kanaori Nihonmatsu C.	Sandy soil	50.9	1.159	2.719	2.76E-14	2.74E-14	9.66E+03	9.74E+03
Kurabeishi Hiruzone Namie T.	Organic soil	59.0	0.969	2.566	1.99E-14	1.96E-14	1.98E+04	2.00E+04
					4.53E-14	4.64E-14	8.66E+03	8.46E+03
Ohshimizu Yamakiya Kawamata T.	Soil of former farmland (including flower garden)	44.9	1.378	2.729	1.56E-14	1.37E-14	1.34E+04	1.53E+04
Mizuzakai Tsushima Namie T.		69.8	0.728	2.511	6.69E-15	8.37E-15	9.63E+04	7.70E+04
					1.76E-14	1.06E-14	3.66E+04	6.08E+04
Matsukiyama Shimotsushima Namie T.		70.2	0.724	2.619	2.93E-14	2.06E-14	2.15E+04	3.05E+04
Shiobite Akogi Namie T.		59.3	1.053	2.604	2.37E-14	2.35E-14	1.65E+04	1.67E+04
Kunugidaira Akogi Namie T.		48.4	1.062	2.631	1.23E-14	1.06E-14	2.04E+04	2.35E+04

Calculated Results of D_a & K_D





- D_a-values are approx. of order 10^{-14} m^2/s and movement by diffusion is very slow.

⇨ Consistent with the fact that almost all Cs still exists near the surface layer of soil.

- Calculated K_D-values are in the range of 10^3–10^5 ml/g, which are consistent with batch K_D-values (K_D=2,080–61,000 ml/g).