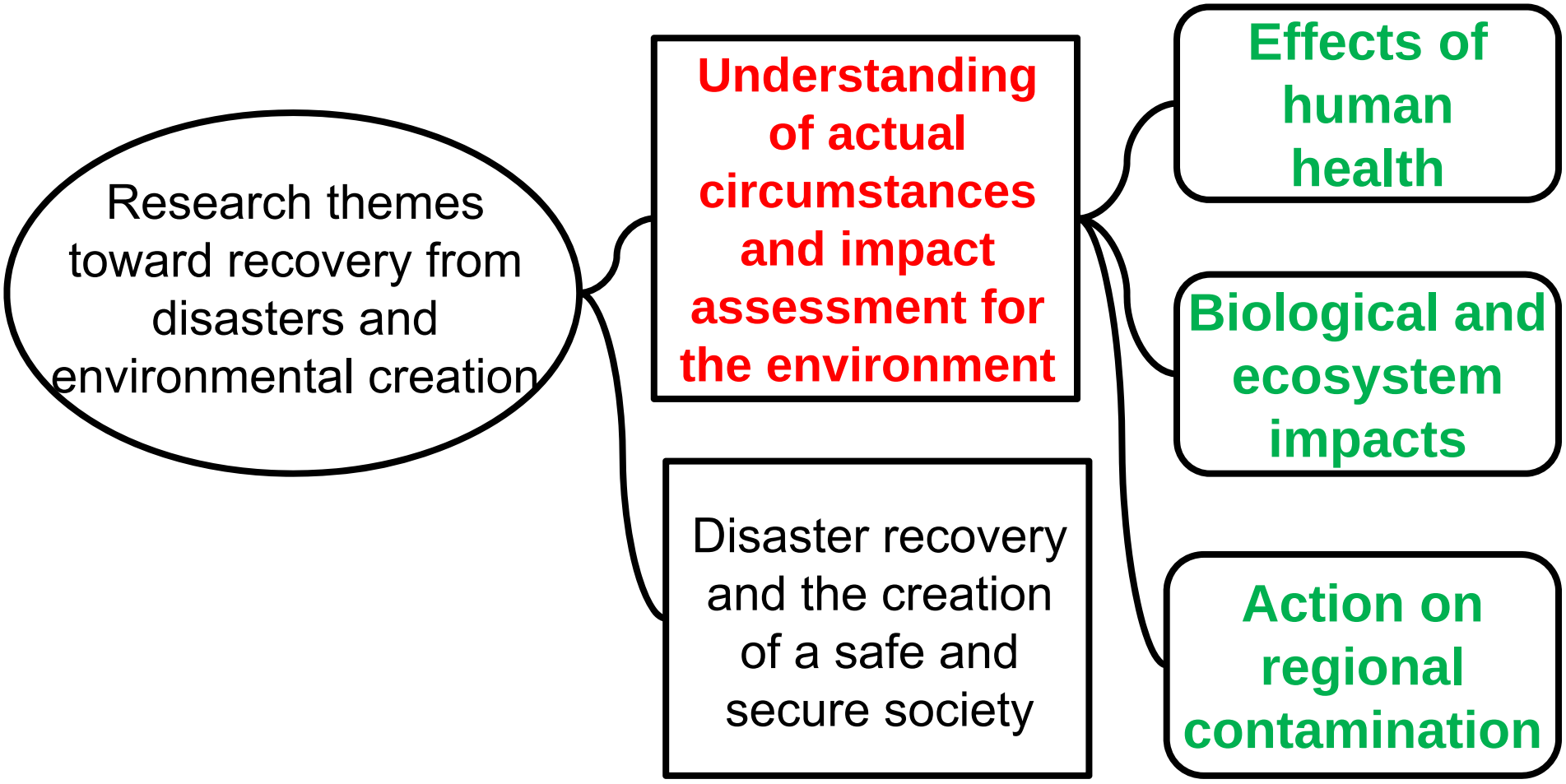
07, June, 2013

Behavior of radionuclides in the environment

- Introduction to National Institute for Environmental Study's research project -

Principal Investigator	**Toshimasa Ohara**
Lead Investigators	
Multimedia modelling	Noriyuki Suzuki
Field measurement	Seiji Hayashi
Ecosystem effect	Masanori Tamaoki
Human exposure	Shoji F. Nakayama
Analytical	Yasuyuki Shibata, Atsushi Tanaka
Science coordinator	Motoyuki Mizuochi

Positioning of **our project**
in the “Panorama of Research on Disaster Environment” overviewed by NIES



Source: NIES, An Overview of Research on Disaster Environment Towards Recovery and Restoration from Disasters and Environmental Creation (April 2012)

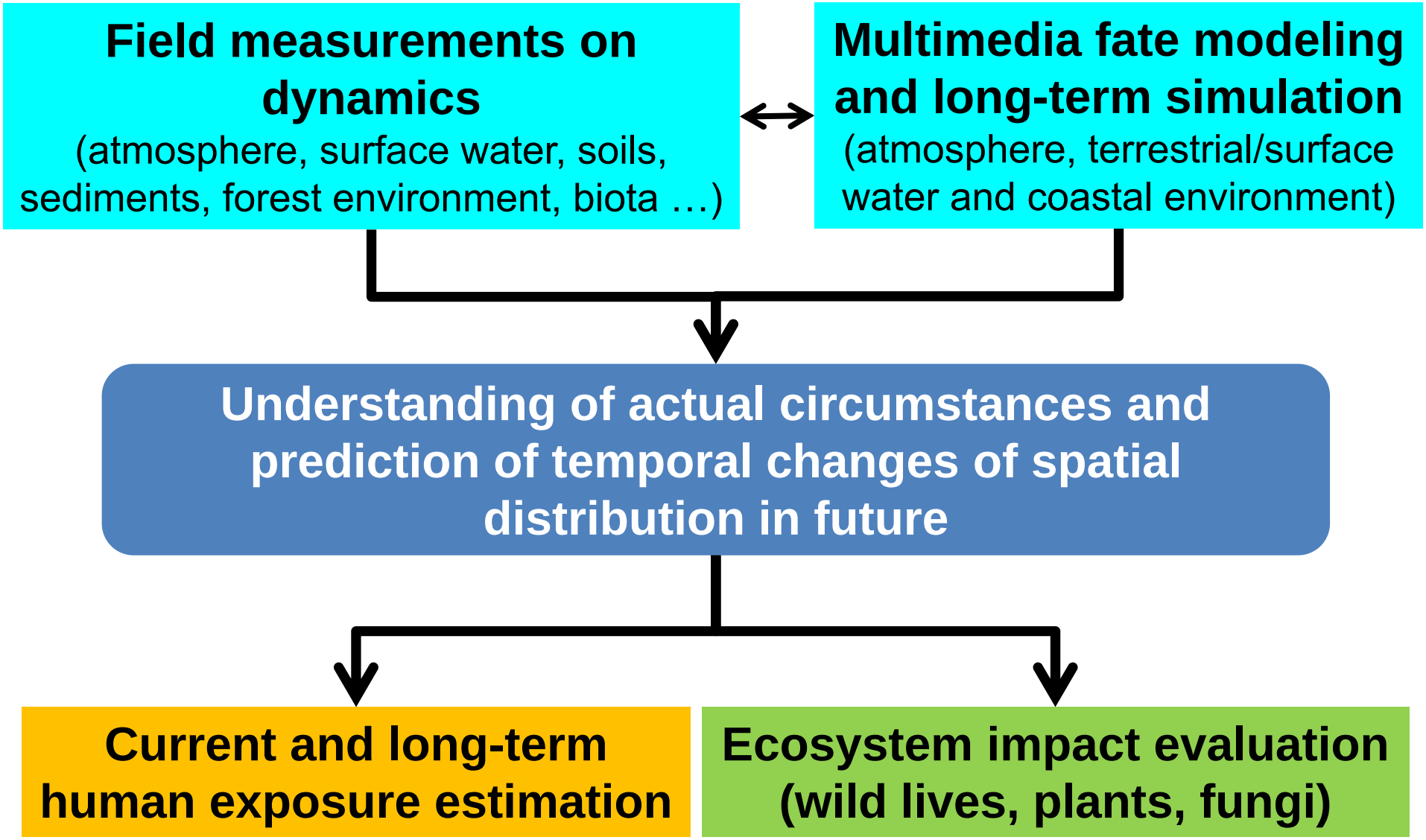
Introduction

- ✓ Due to nuclear accident at the Fukushima Daiichi nuclear power plant (FDNPP), enormous amounts of radionuclides were emitted into the atmosphere and the ocean.
- ✓ Radioactive materials affect human health through the contamination of air, water, soil, and food.

- ✓ **Aims of our project**

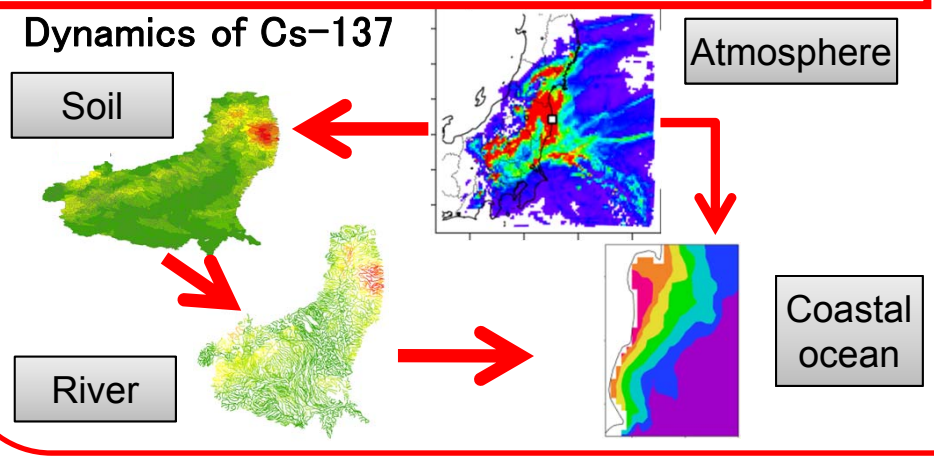
  - Better understand the fate and transport of radionuclides released from FDNPP
  - Predict long-term distribution of radioactive nuclides in the terrestrial and aquatic environment as well as in living organisms
  - Develop a tool set for evaluating the effectiveness of decontamination
  - Build a model to estimate long-term human exposure to radiation
  - Contribute to better understanding of the impacts on human, wildlife and ecosystem health

Project framework



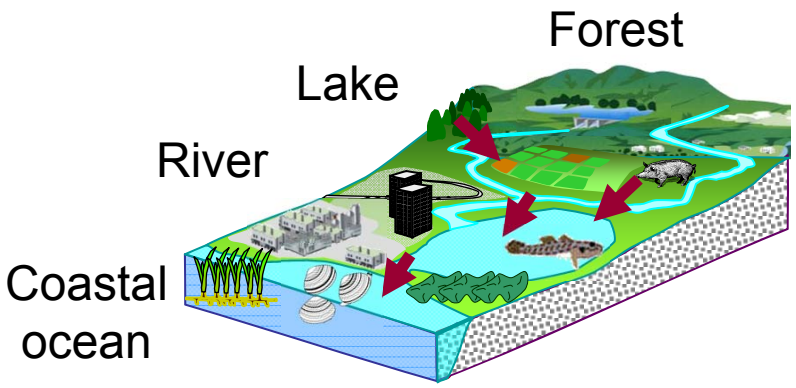
Outline of study on "Behavior of radionuclides in the environment"

Multimedia Modeling



- ✓Developing a multimedia fate model by coupling models for atmospheric, oceanic, and terrestrial environments.
- ✓Analyzing and predicting the dynamics of Cs-137 in the air, soil, river/lake, and coastal ocean.

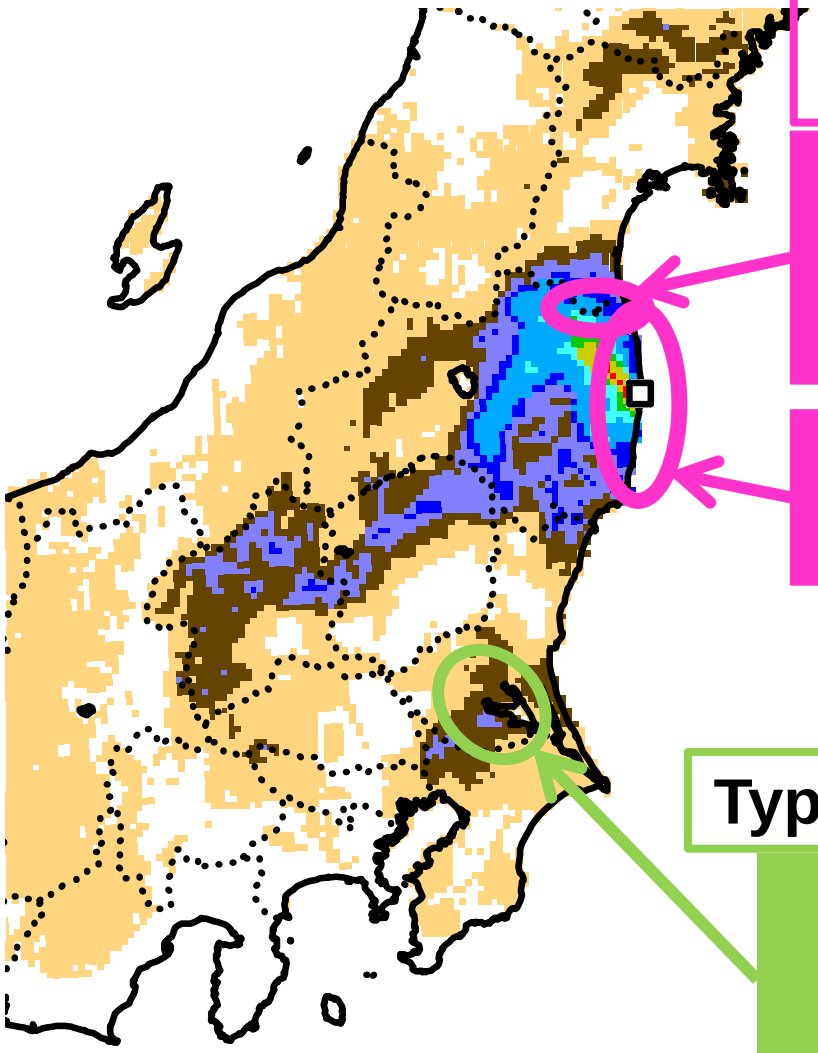
Field Measurements



Field survey in the eastern area of Fukushima prefecture and Mt. Tsukuba/Lake Kasumigaura

- ✓ Monitoring radionuclides in the atmosphere.
- ✓ Measuring radioactive Cs in forest, lake, river, and coastal ocean.
- ✓ Improving the analytical method of I-129, radioactive Sr, and Cs in the water.

Targeted area of field study in our project



Cs-137 deposition map by airborne monitoring

Typical high contamination area around FDNPP

Udagawa river
＊Typical watershed in Fukushima
(Forest, lake, river, creek**)**

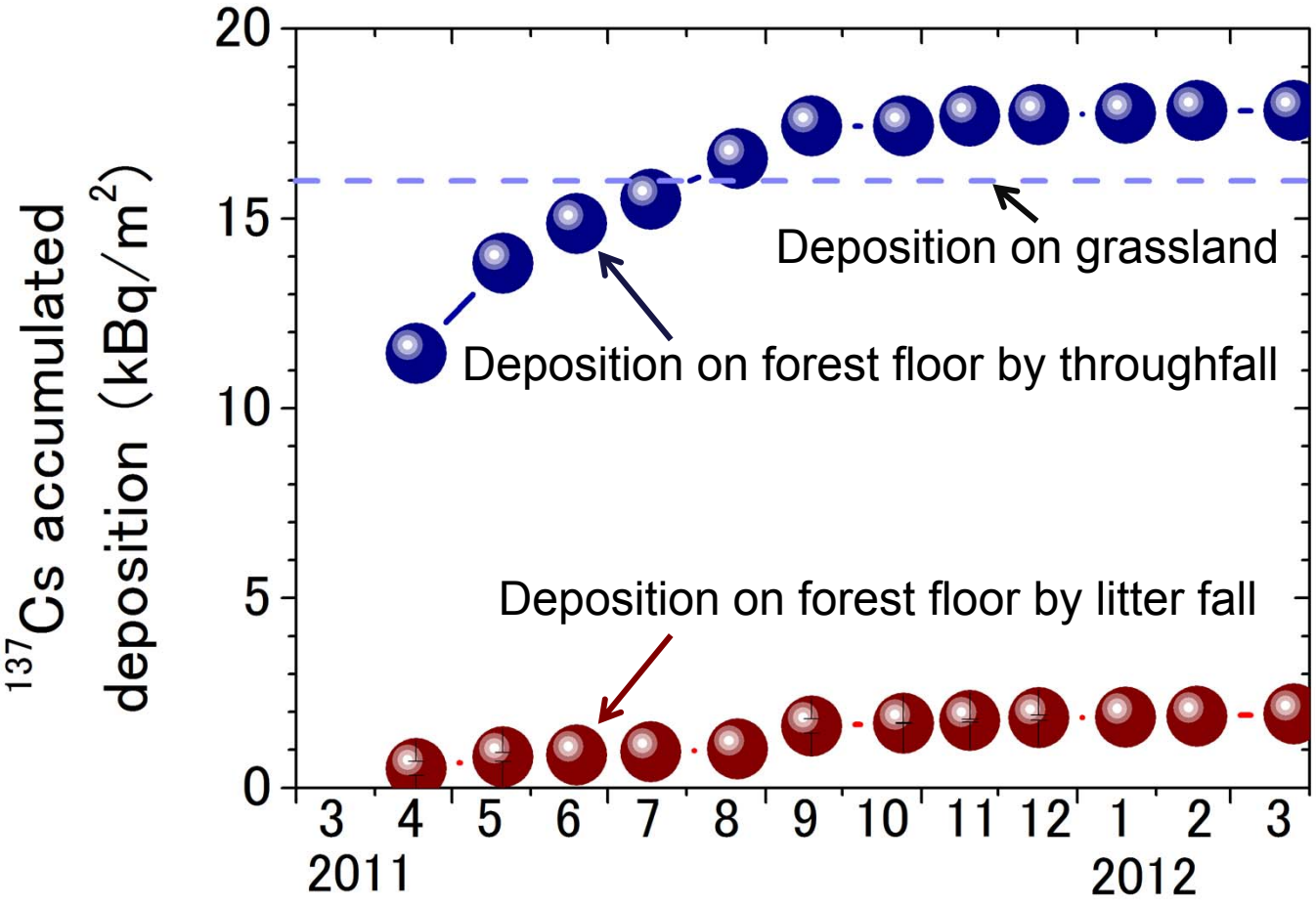
FDNPP surrounding area
<Biological and ecosystem impacts>

Typical moderate contamination area

Mt. Tsukuba, Lake Kasumigaura
* Typical watershed in Kanto area
(Forest, river, lake)

^{137}Cs dynamics in forest of Mt. Tsukuba

Temporal redistribution of ^{137}Cs from the canopy of a cedar forest in Mt. Tsukuba to the forest floor beneath

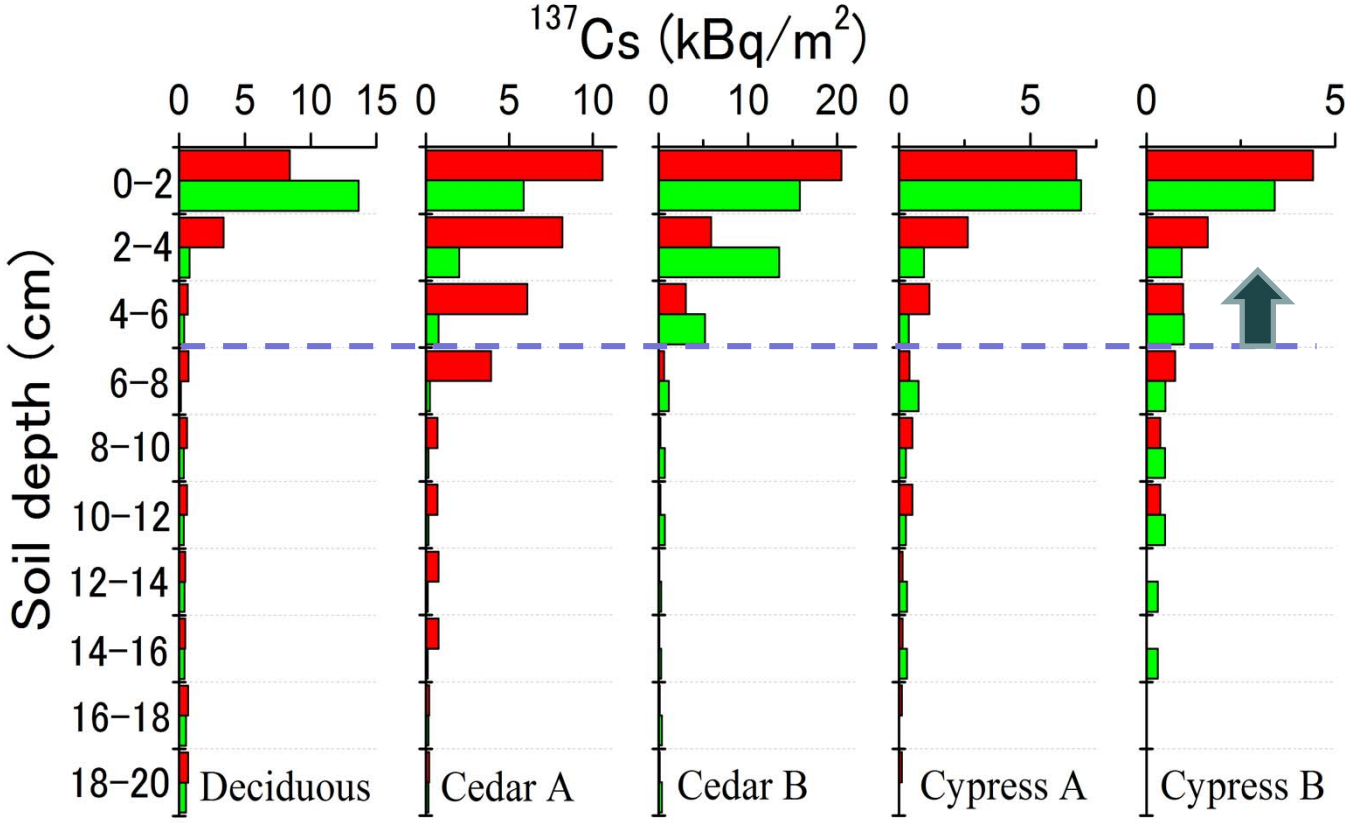


Initial deposition amount on forest canopy
→ Influencing factor on dynamics of radiocaesium in forest for some time after the accident at FDNPP

^{137}Cs accumulation in forest soil (Mt. Tsukuba)

- Accumulation into surface soil including litter layer & little downward migration
- Increase of ^{137}Cs accumulation in surface soil by mechanistic decontamination (through fall and litter fall) of contaminated forest canopy

^{137}Cs profiles in forest soils of main tree species in Mt. Tsukuba



47days after

1 year after

70 – 95% of deposited ^{137}Cs was accumulated within 6cm depth

^{137}Cs accumulation

After 47days,

13.8 ± 12.2 kBq/m^2

Increasing!

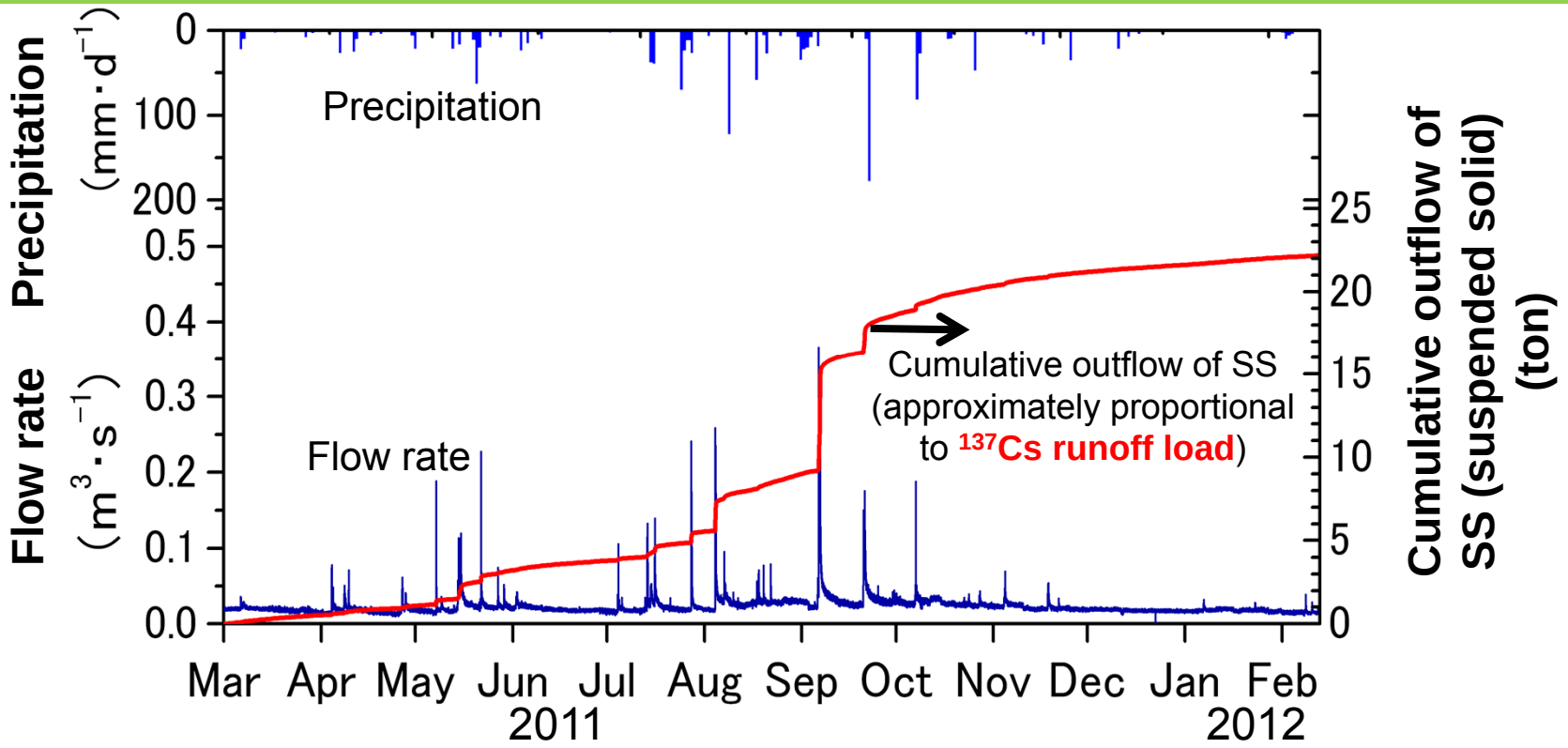
After 1year,

16.8 ± 9.7 kBq/m^2

Runoff characteristics of radiocesium from a forest catchment (Mt. Tsukuba)

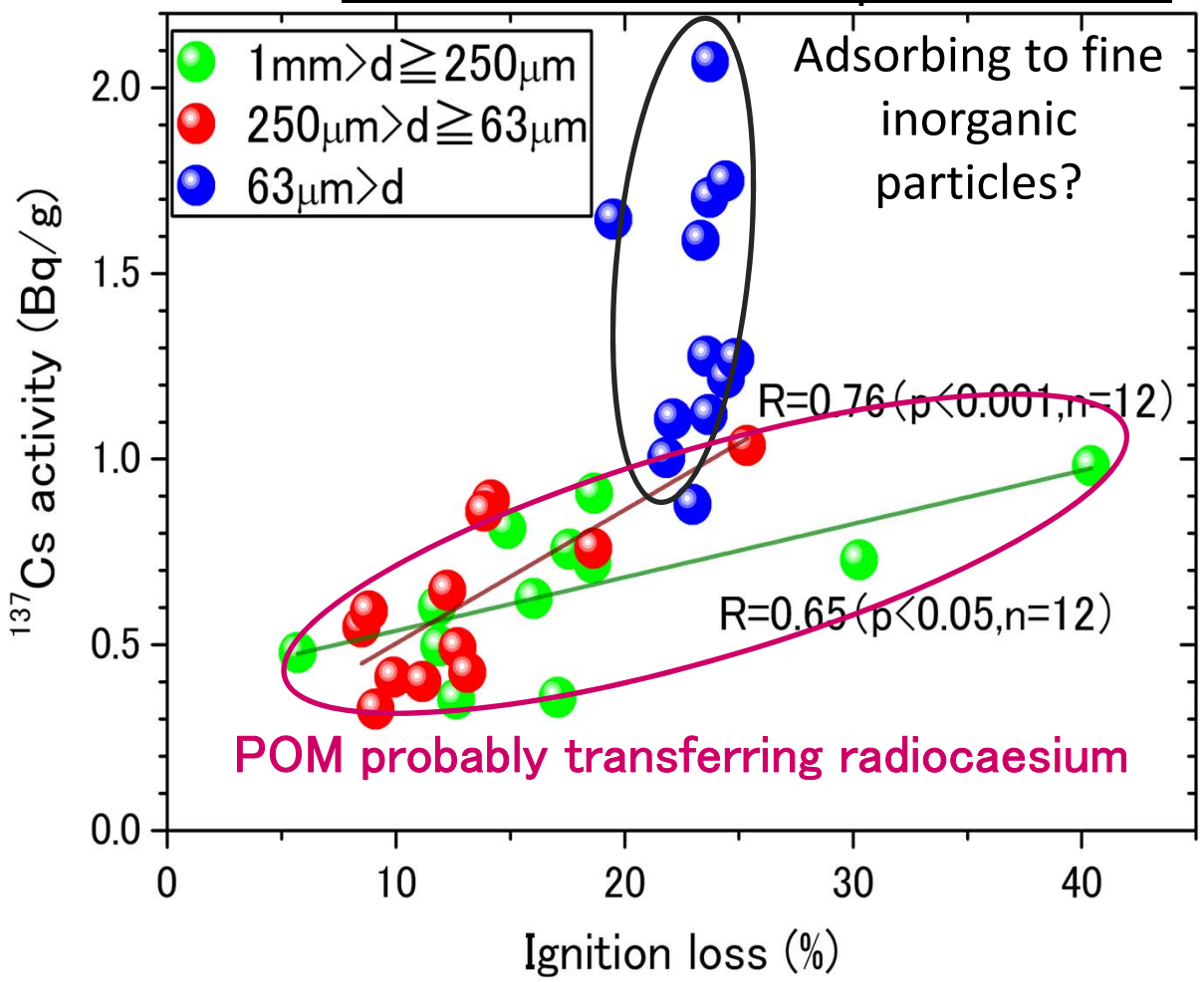
- Continuous hydrological observations started immediately after the accident and stream water sampling during the rain events at a forested catchment
- Estimated ^{137}Cs annual runoff load was 0.04 kBq/m^2 for one year, corresponding to only 0.3% of the total amount deposition in the catchment.
- Very little contribution of forested area as a source of ^{137}Cs at present

^{137}Cs runoff = [^{137}Cs concentration in SS (900 Bq/kg-SS)] x (runoff volume of SS)



Contribution of POM to ^{137}Cs runoff from forest catchment in Mt. Tsukuba

Relationship between ^{137}Cs activity and organic matter content of sieve classified suspended solid

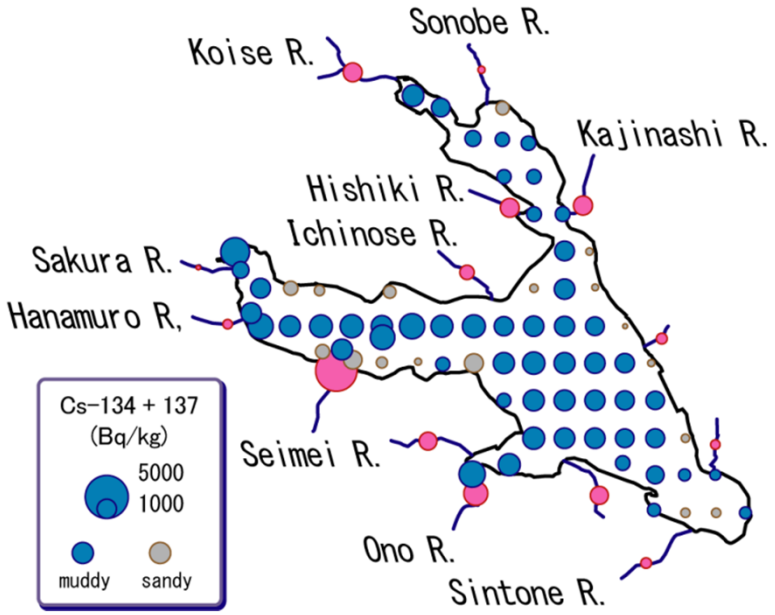


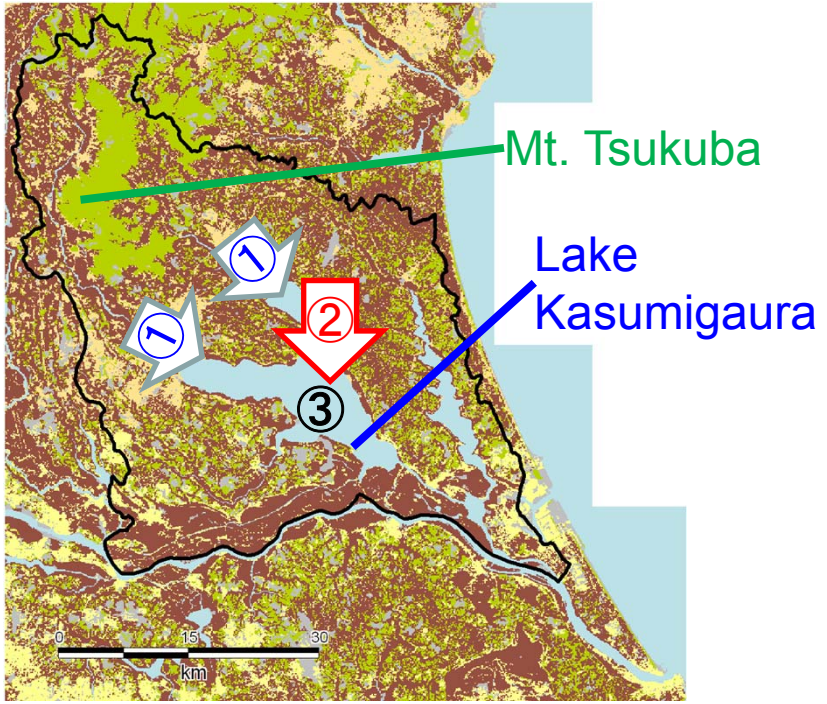
- POM : Particulate organic matter
- A slight amount of fraction over 1mm →below the detection limit of ^{137}Cs
- Fraction over 63μm covering 15～48% of total ^{137}Cs activity

Transfer and the bioconcentration of radiocaesium into aquatic organisms through food web should be concerned

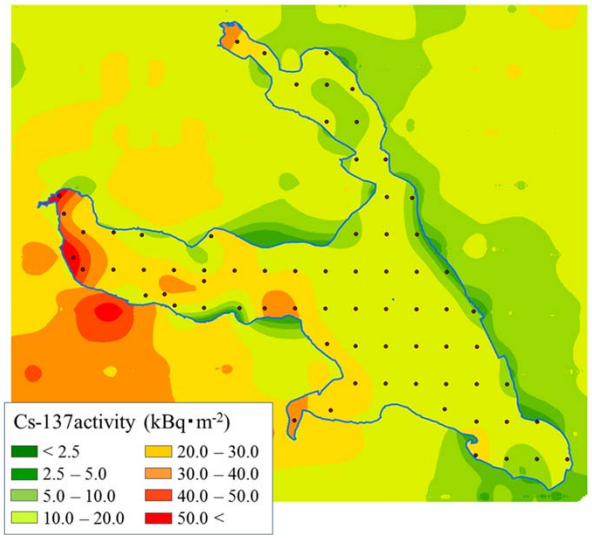
Stock and flow of Cs-137 (Lake Kasumigaura)

Horizontal profiles of activity in sediment





Spatial distributed accumulation

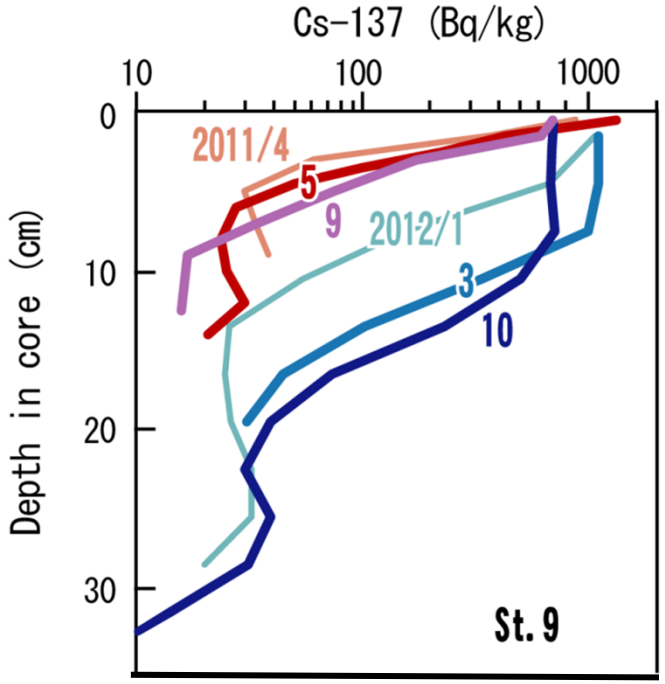


Budget of Cs-137 in Lake

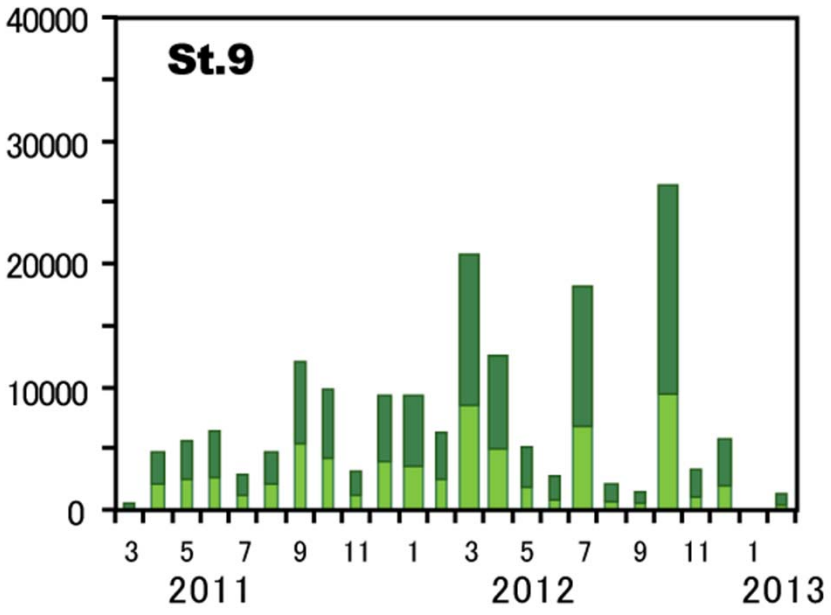
① Inflow via river 0.2 TBq
(runoff ratio from entire land to lake = 0.5 %)
② Deposition from air to lake 2.7 TBq
(= ③ - ①)
③ Stock into the lake sediment 2.9 TBq
(estimation based on the measurements)

Temporal variation of Cs-137 (Lake Kasumigaura)

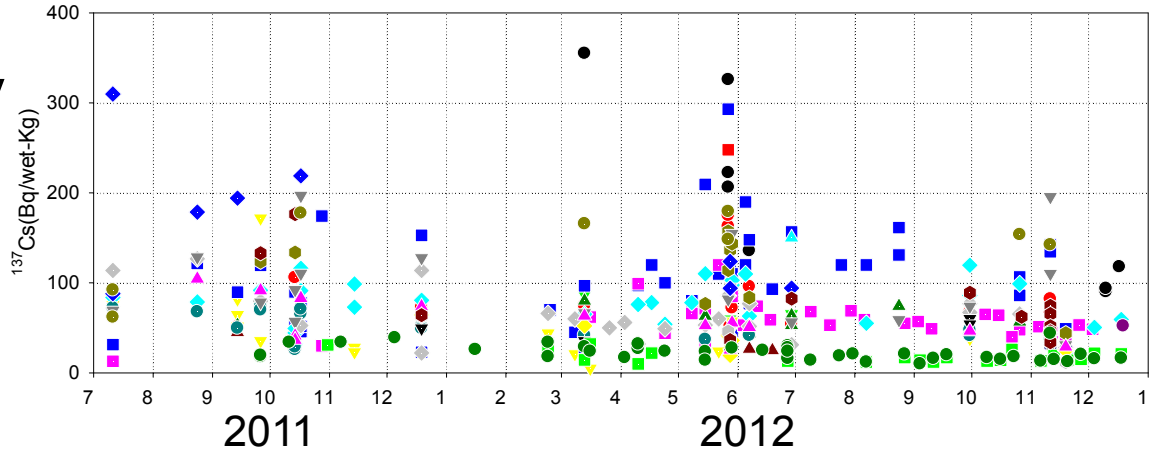
Vertical profiles of activity in sediment



Monthly variation of activity in sediment at St. 9



Time variation of activity in fishes (Color: kinds of fish)



Observation in Udagawa River catchment

Transfer

Sedimentation in river

Sedimentation & fluid in lagoon & coast

Deposition & runoff in forest

Sedimentation in Lake

Runoff

Matsukawaura Lagoon

Coast of Fukushima Pref.

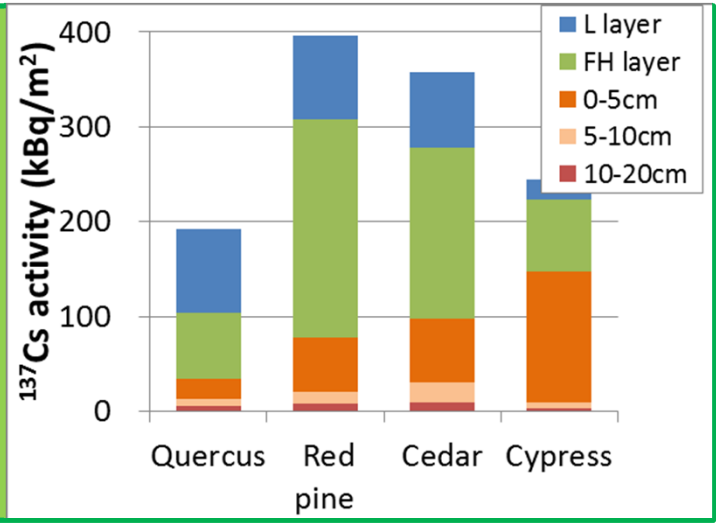
Tamano Pond

Lake Udagwa

Transfer

Deposition in forest soil (Sep. 2012)

Ave. deposition 330kBq/m²

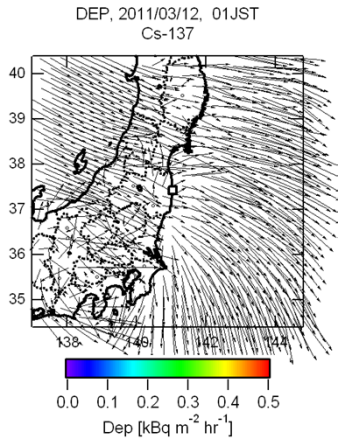


Runoff from forest area (Jul. 2012 to Feb. 2013)

- ^{137}Cs runoff amount per unit area
  0.04 ~ 0.07 kBq/m²
- Runoff ratio to deposition
  0.02 ~ 0.03%

Outline of multimedia fate modeling

To establish simulation model to estimate the long-term (up to several tenth years) fate of radioactive substances, combining existing atmospheric, multimedia and ocean fate models

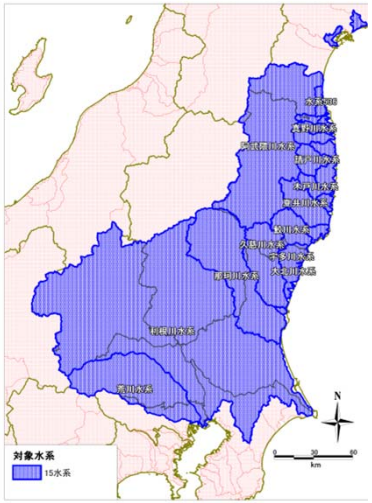


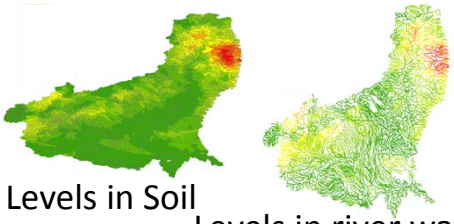
Atmospheric transport/deposition model

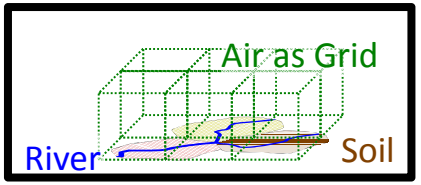
Deposition

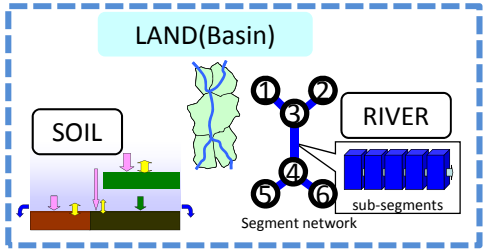
Deposition

Terrestrial multimedia model









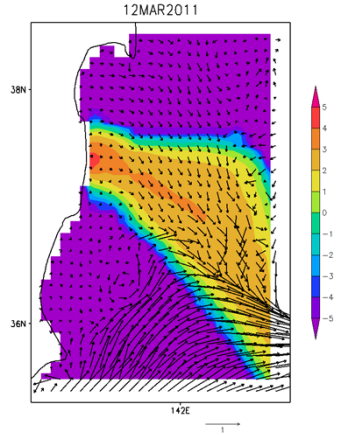
Outflow

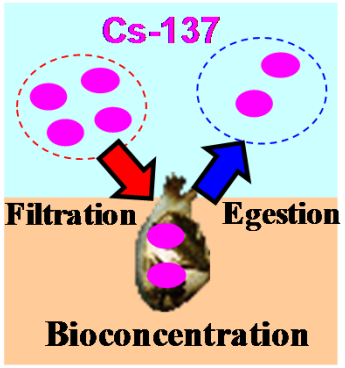
Coastal Ocean model

Fluid dynamics model

Transport model in water & sediment

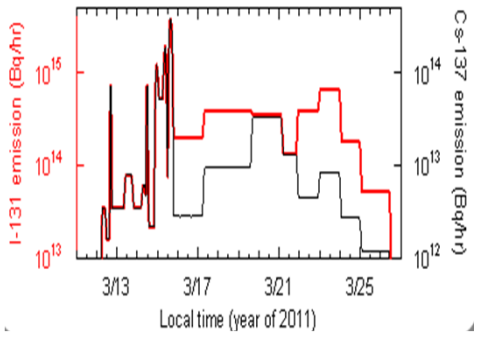
Ecological model



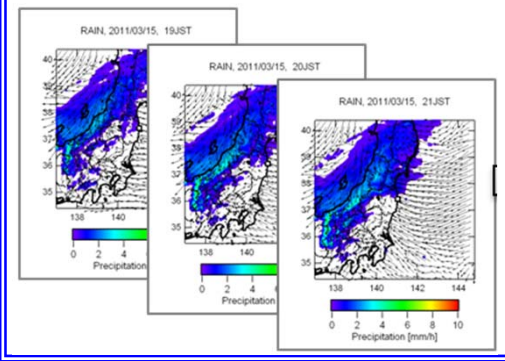


Atmospheric model

Emissions

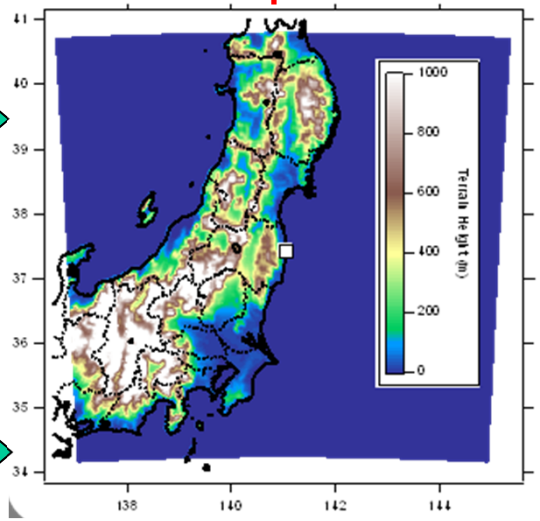


Regional Meteorological model



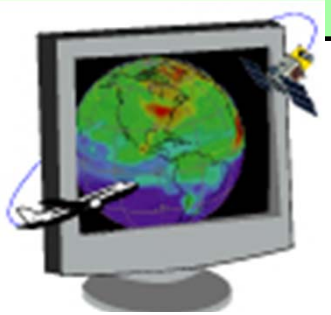
Wind, Temperature, Humidity, Solar radiation, rainfall etc.

Chemical Transport Model for air pollutants

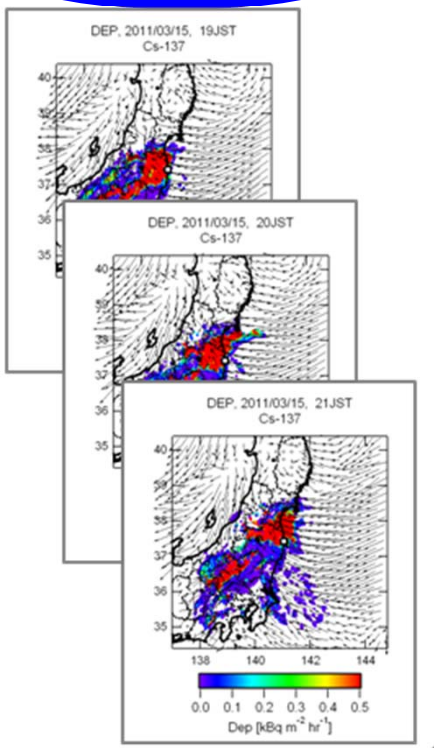


Physical processes

- **Transport**
- **Emission**
- **Deposition**
- **Radioactive decay**



3D distribution of air concentration and deposition



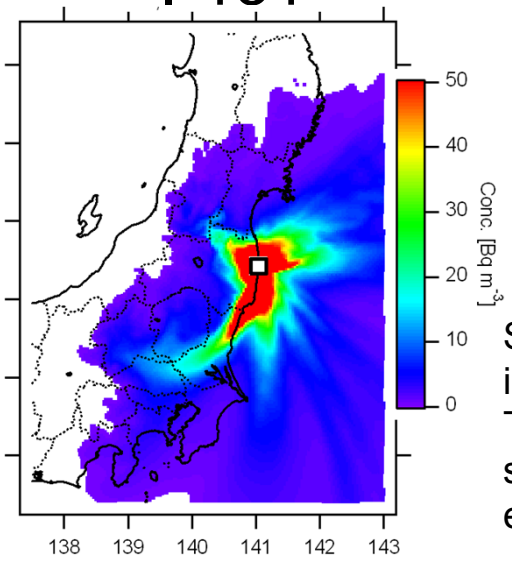
Verification

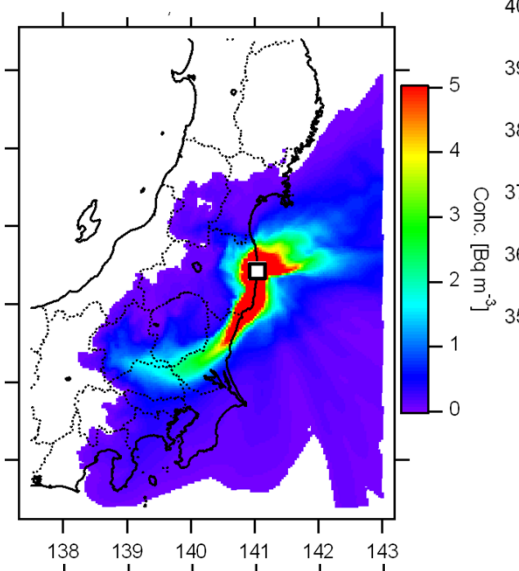
Spatial distribution of I-131 and Cs-137 (surface concentration and deposition)

I-131

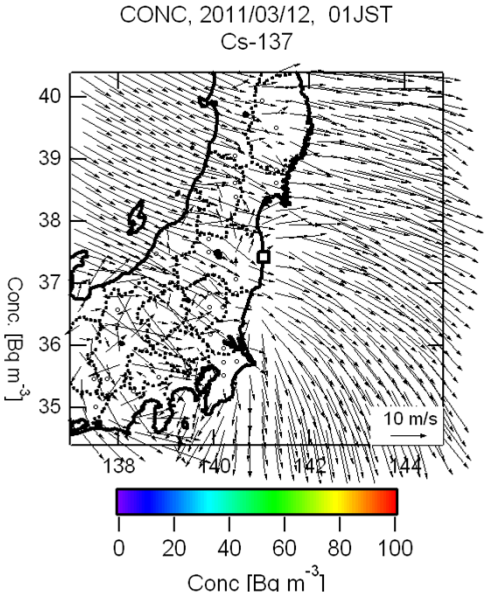
Cs-137

Surface conc. (2 months mean)

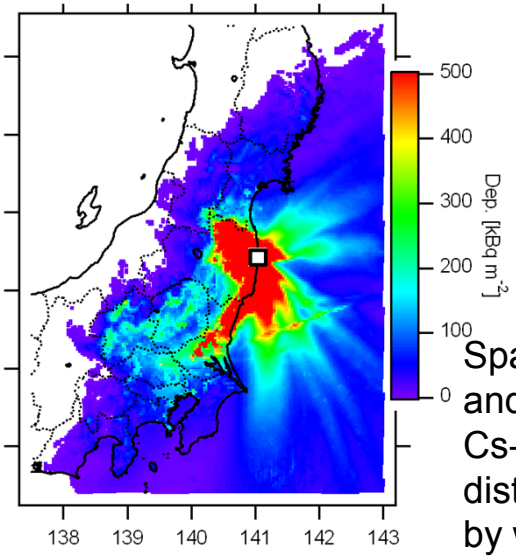


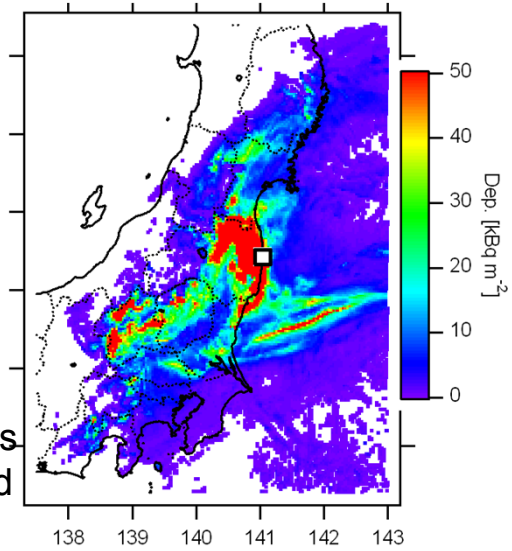


Spatial distribution of I-131 is similar to Cs-137. The elevated areas are simulated around the emission source.

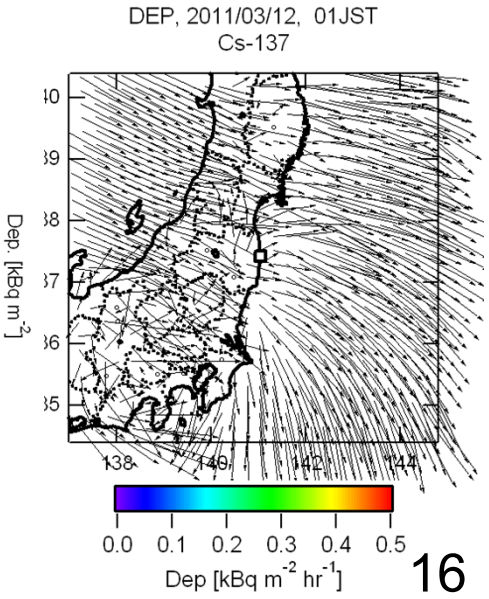


Deposition (2 months total)

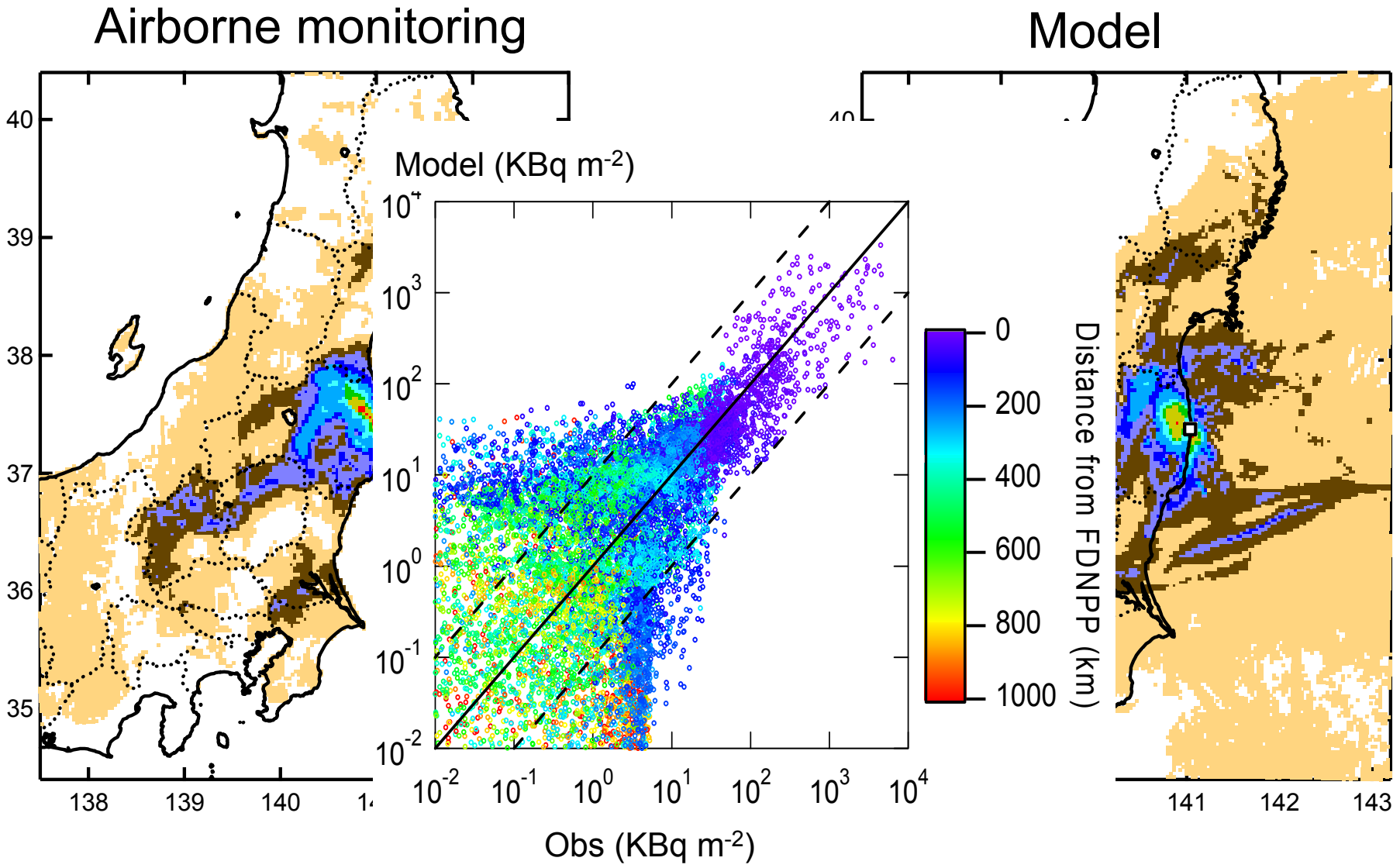




Spatial distribution of I-131 and Cs-137 is quite different. Cs-137 shows inhomogeneous distribution (hot spots) caused by wet deposition of particles.



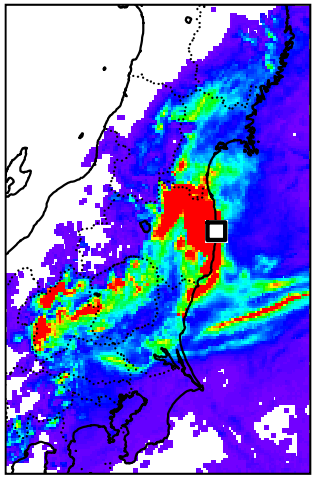
Observed and modeled Cs-137 deposition map



Bias = -14 %; Error = 78 %; r = 0.67

Terrestrial multimedia model

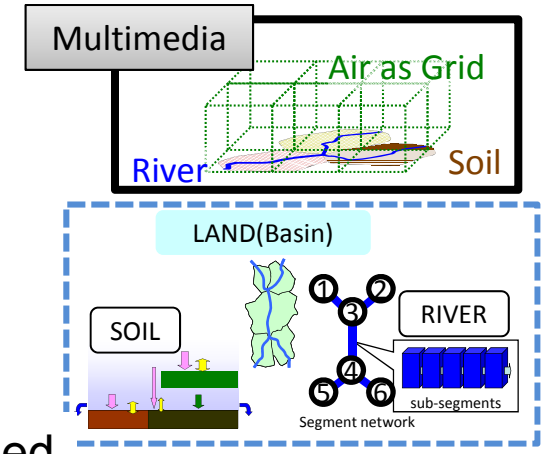
Atmospheric model

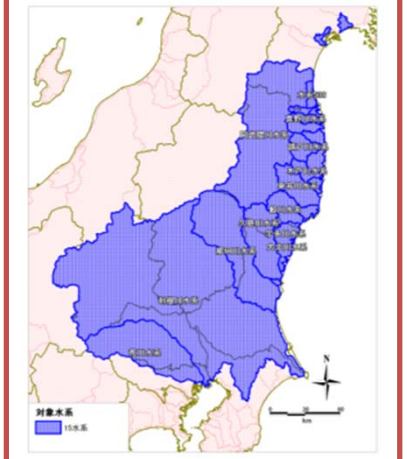


Cumulative deposition of Cs-137 from atmosphere to surface soil

Time varied gridded deposition data

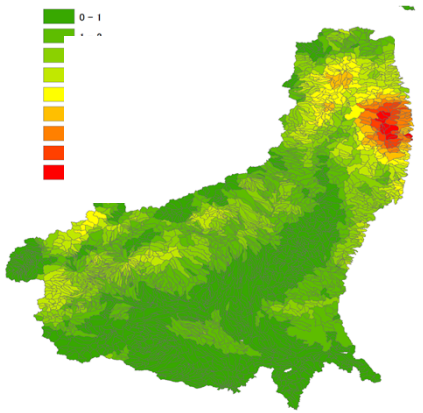
Terrestrial model





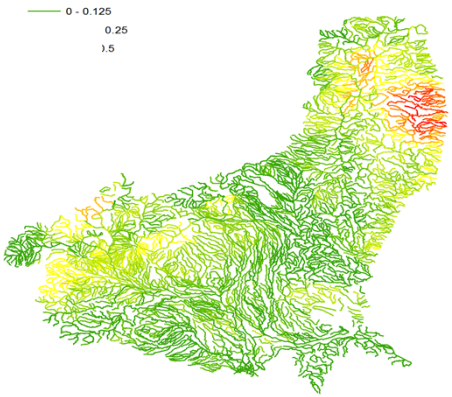
Model domain

15 river watersheds around Fukushima prefecture



Cs-137 in soil

Level of Cs-137 in soil at March 23, 2011



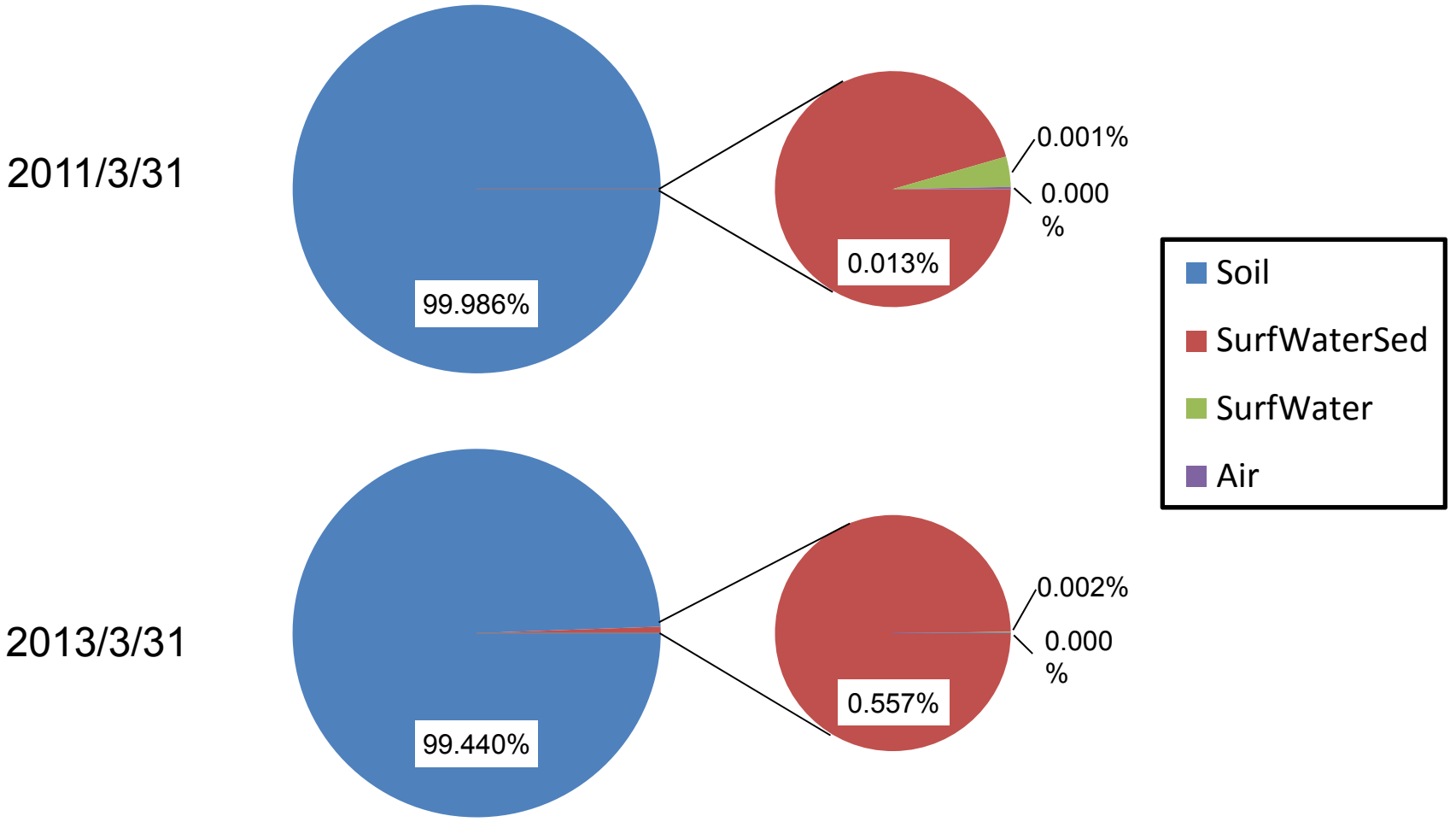
Cs-137 in river water

Level of Cs-137 in river water at March 23, 2011 (Soluble+Particulate, ordinary flow rate)

Media distribution of ^{137}Cs in the simulation domain

- Most part of ^{137}Cs exists in soil compartment
  - More than 99% in soil after 2 years

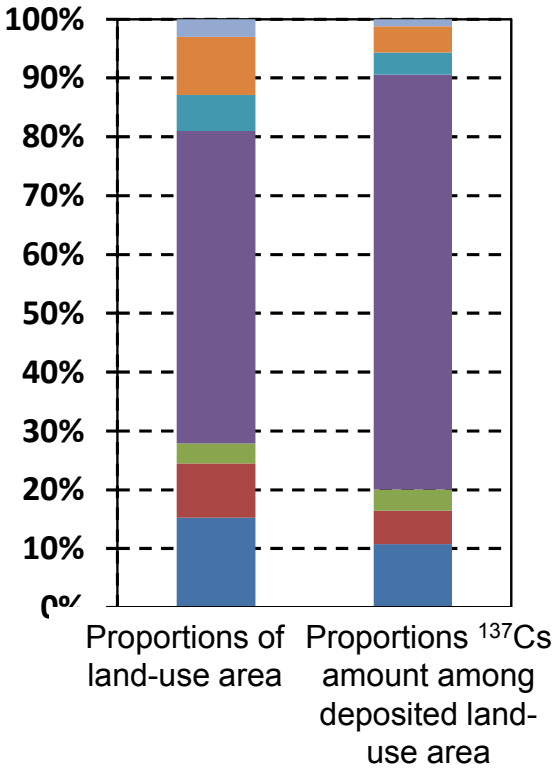
- Second largest part exists in surface water sediment

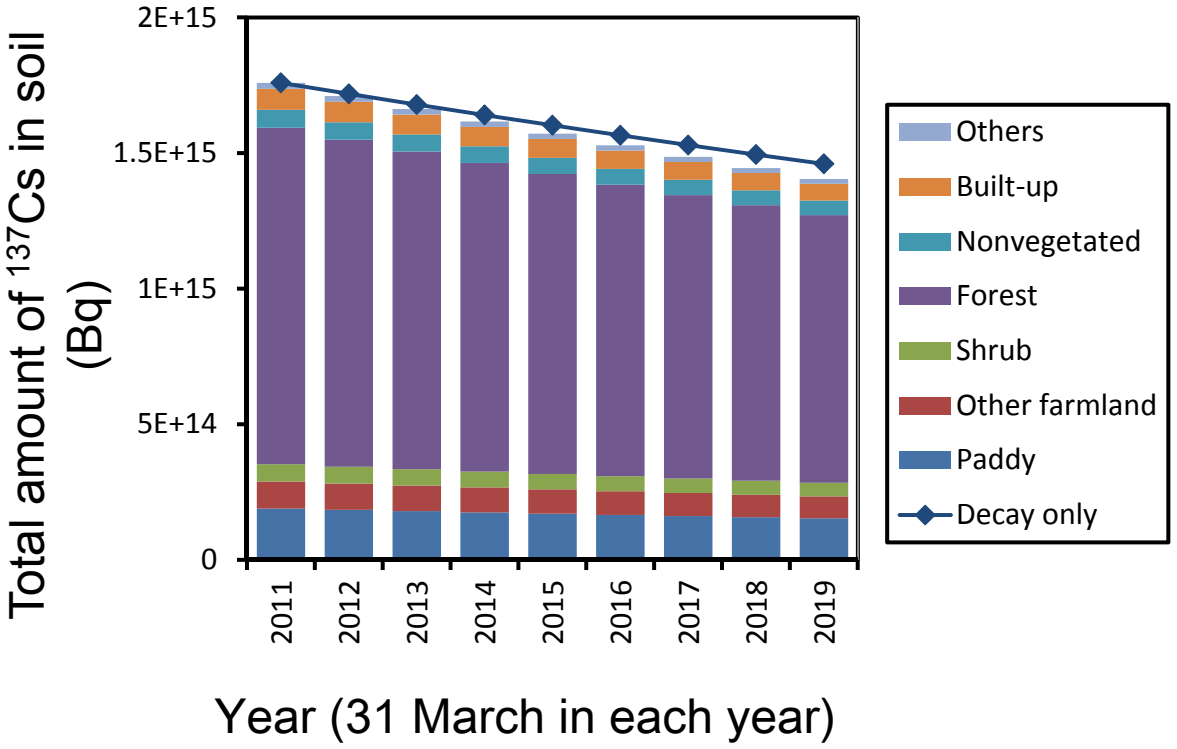


Simulated trend of ^{137}Cs in soil

- Most part of ^{137}Cs were mainly deposited to forest area
  - Contaminated plume passed above forest area

- Decreasing trend of ^{137}Cs in soil
  - Simulated to slightly faster than radioactive decay, by runoff processes



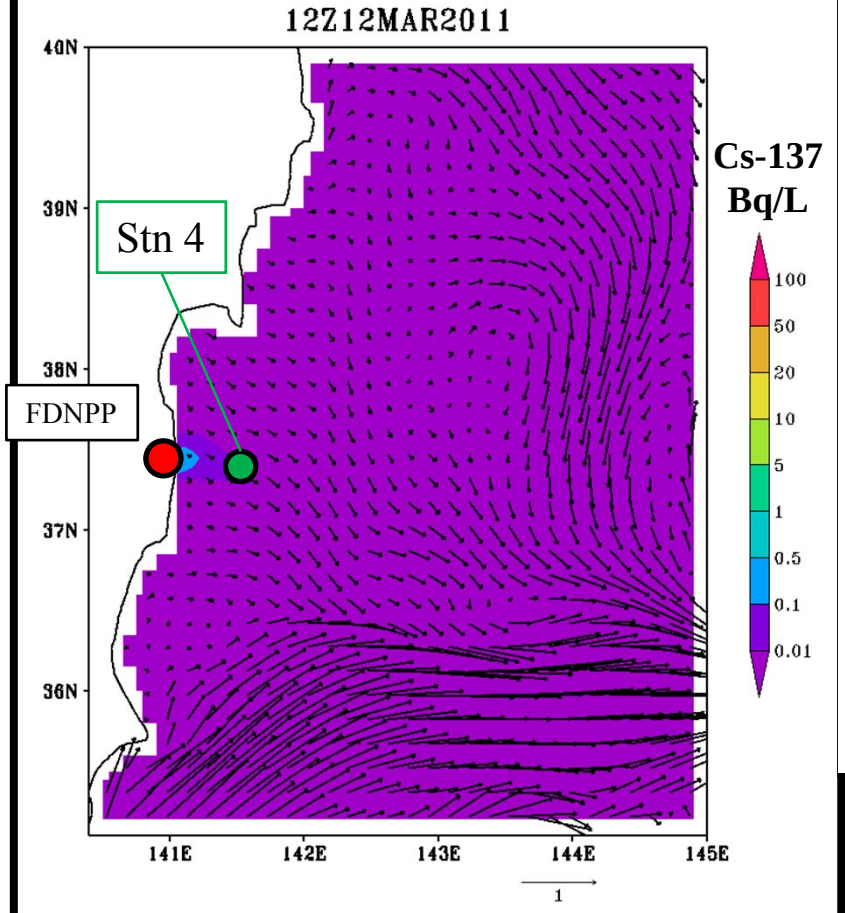


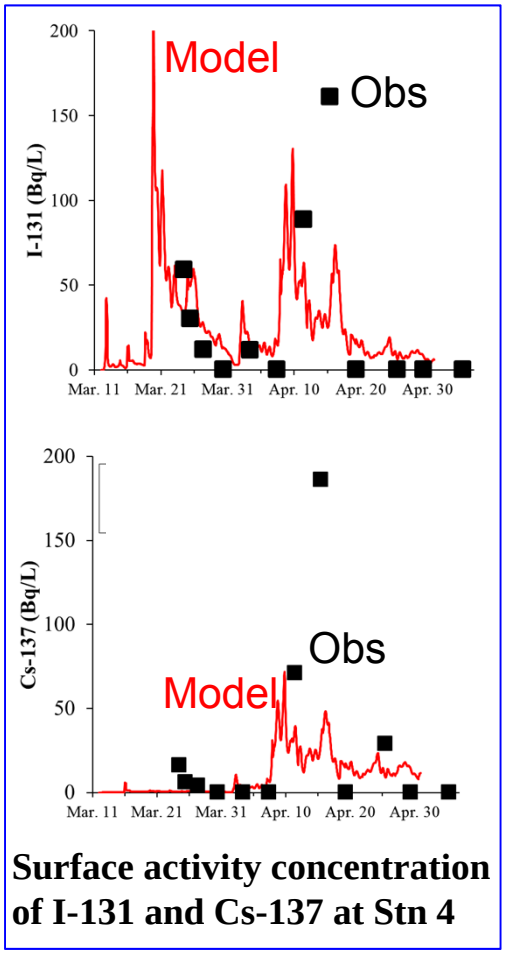
Coastal Ocean Model (Dispersion and Sedimentation)

Deposition (Atmospheric model)

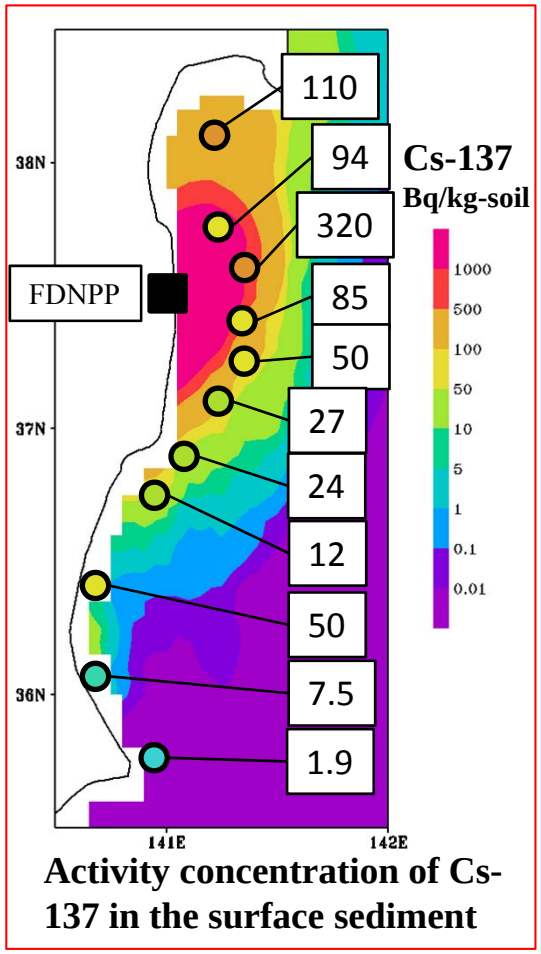
Loading (River basin model)

Radionuclides Dispersion Model





Surface activity concentration of I-131 and Cs-137 at Stn 4



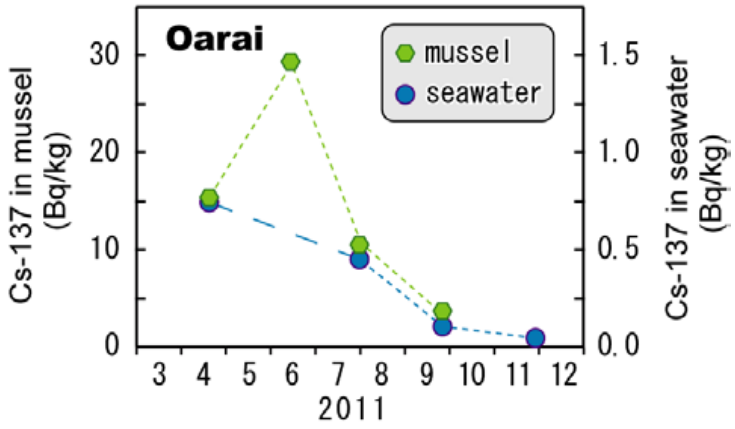
Activity concentration of Cs-137 in the surface sediment

The model reproduced the observed distribution of radionuclides in the sea and the sediment.

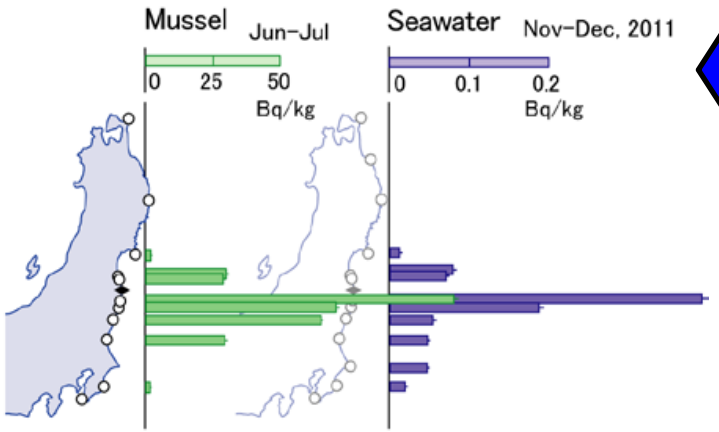
Bioconcentration model

Coastal Ocean Model (Bioconcentration)

Monitoring



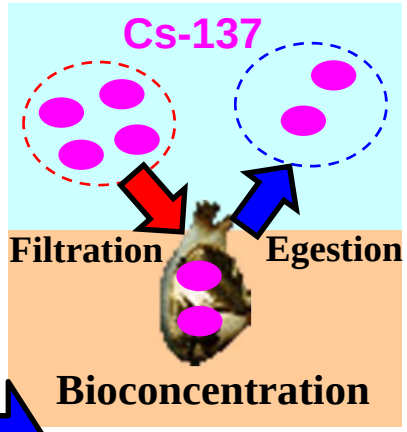
Time series of Cs-137 concentrations in bivalve mussel and in seawater at Oharai coastal region

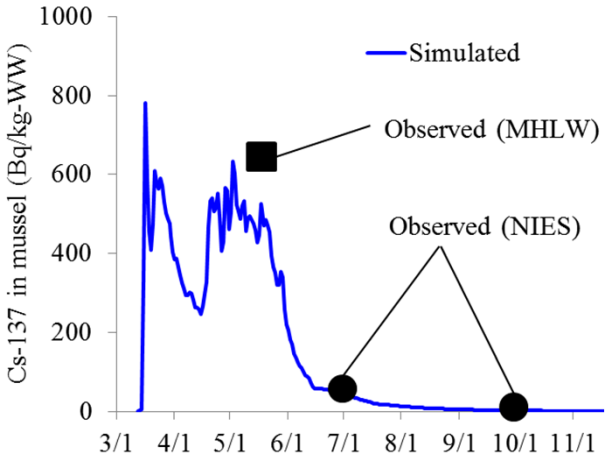


Spatial distribution of Cs-137 concentrations in bivalve mussel and in seawater

Simulated Cs-137 concentration

Bivalve (*M. galloprovinciallis*) Model





Cs-137 in mussel at Soma coastal region

Concentration Factor: 12~ 47 (simulated)
← consistent with the observed factor (12~59)

Biological Half-Life: 26 days (simulated)
← relatively shorter than the observed days for the other bivalves (50～90 days)

Human exposure estimation

Exposure monitoring, case studies

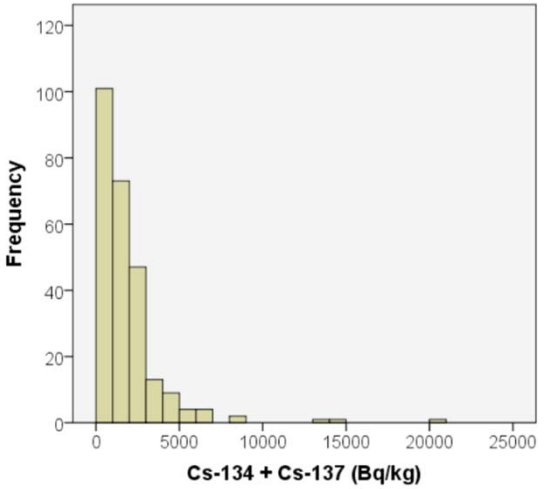
- ✓ External exposure: Outdoor and indoor dose, personal dosimeter monitoring and behavioral record
- ✓ Internal exposure: Total diet, soil and house dust measurement
- ✓ Parameter and validation data generation for exposure models

Exposure modelling

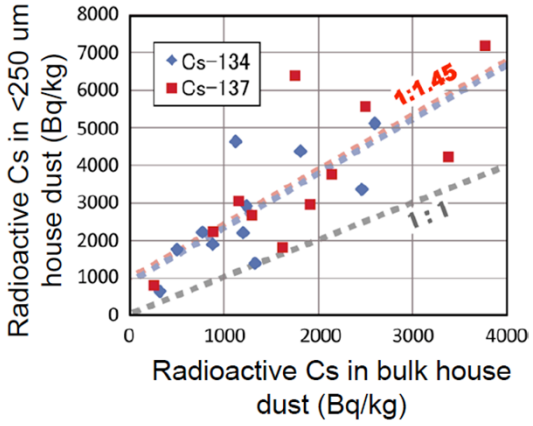
- ✓ Use of monitoring data to construct a long-term exposure model for general population
- ✓ External and internal models
- ✓ Long-term exposure estimation with input from fate models
- ✓ Potential contribution of health effect assessment

Exposure monitoring

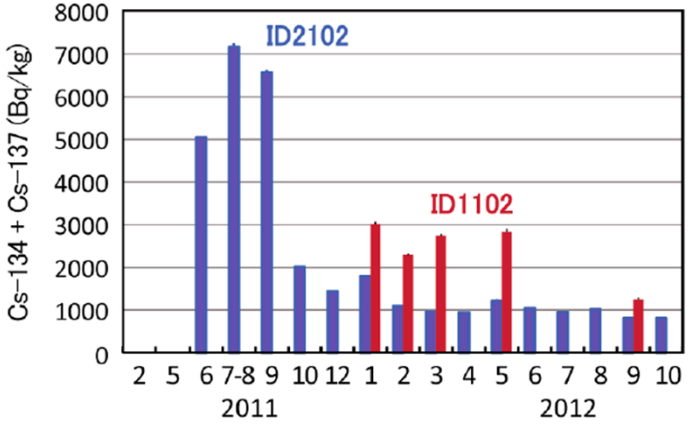
- Vacuum cleaner dust measurement
  - The most radioactive material in the indoor environment
  - Log-normal distribution
  - Decreased in the first several months but reached the plateau after that
  - Distribution in ingestible size (< 250 μm)
  - Provided a model with parameters



Radioactive Cs in bulk house dust (n=254, April, 2012)

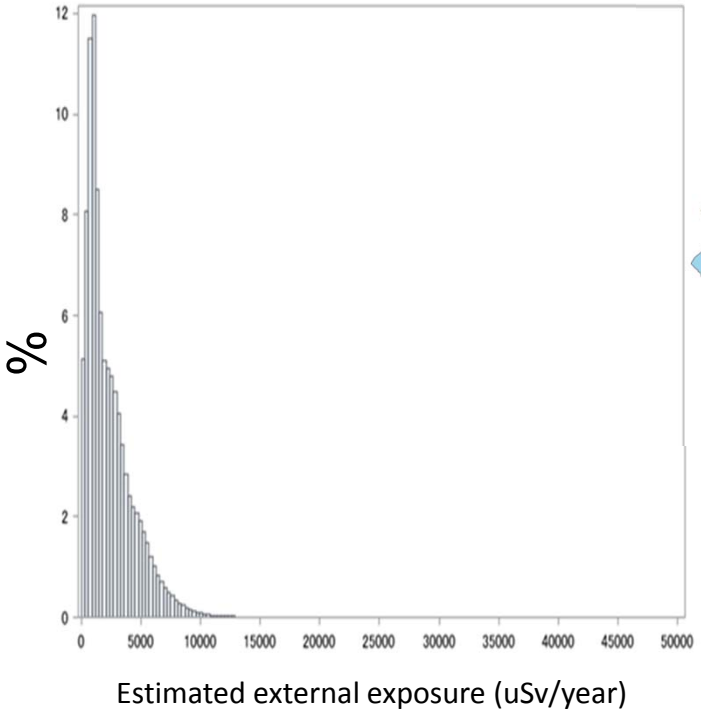


Estimation of the enrichment factor

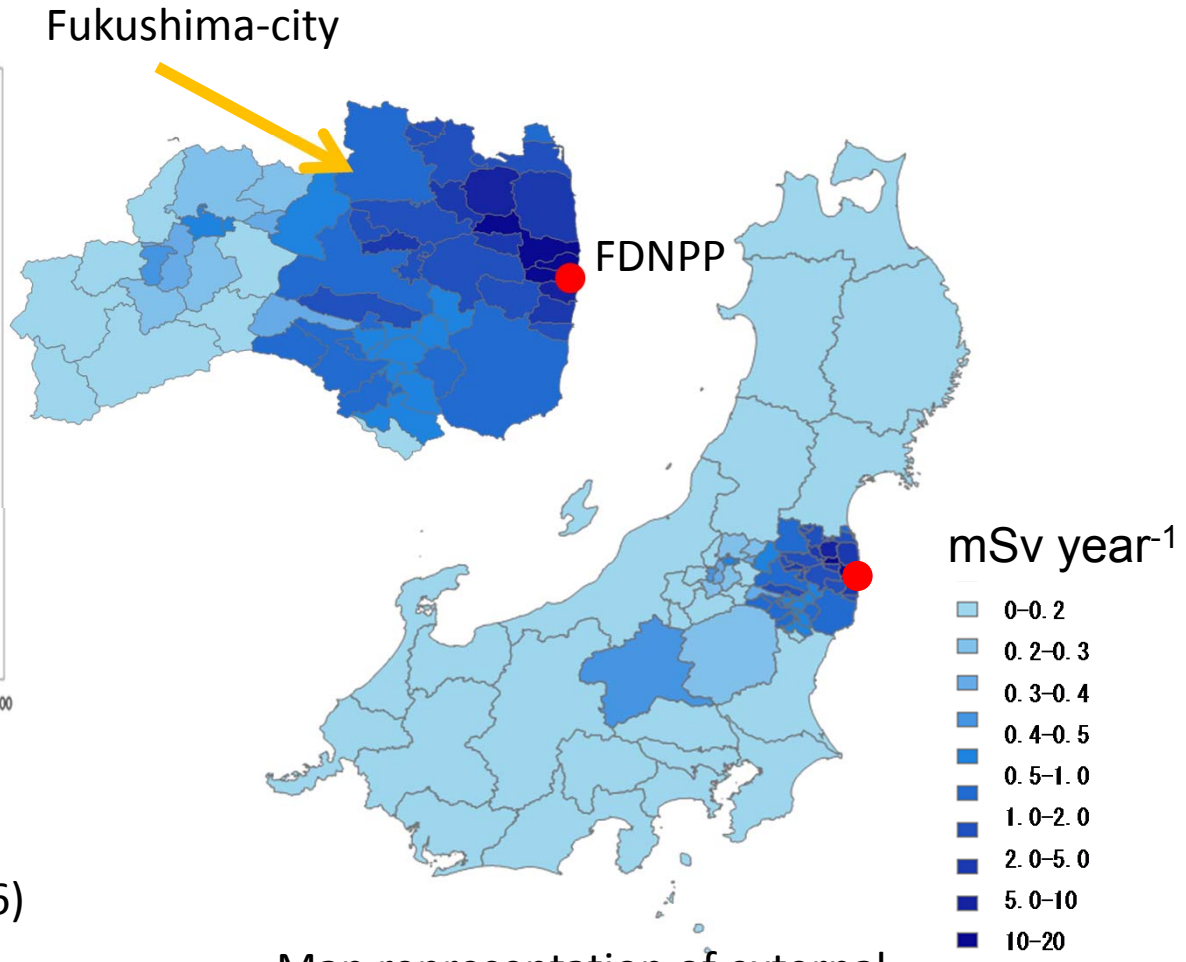


Temporal change of radioactive Cs in house dust

External exposure



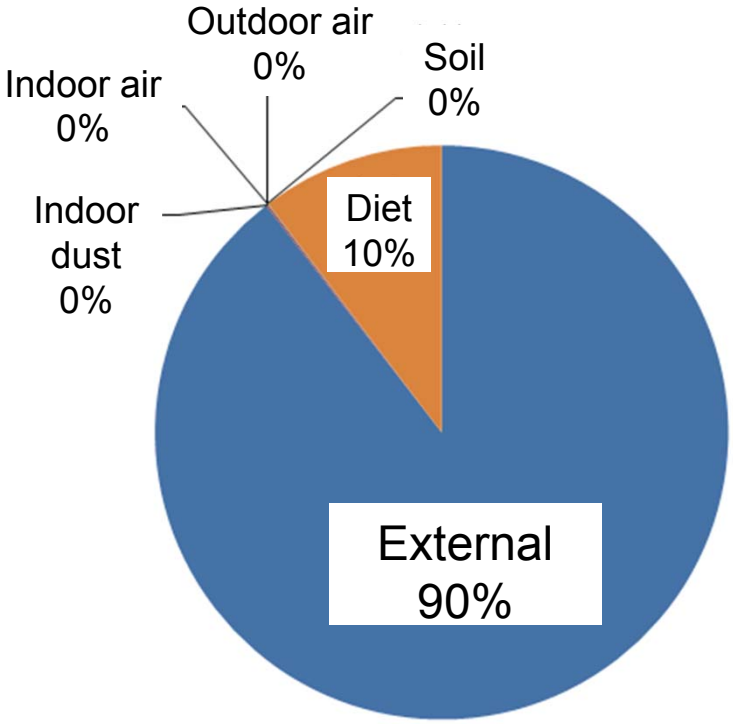
Estimated external exposure level in Fukushima-city (age 1-6) (Cs-134+Cs-137)



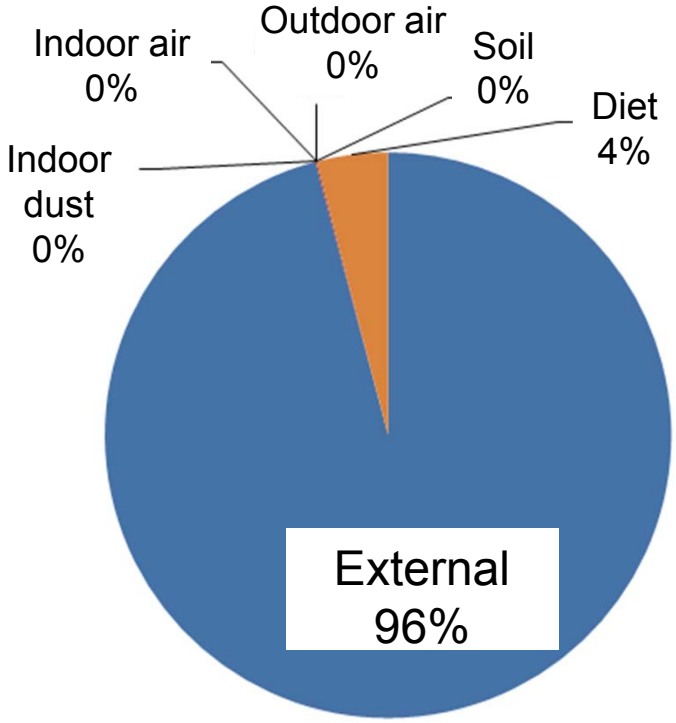
Map representation of external exposure level (age 1-6) (Median Cs-134+Cs-137)

Exposure route

Kashiwa-city



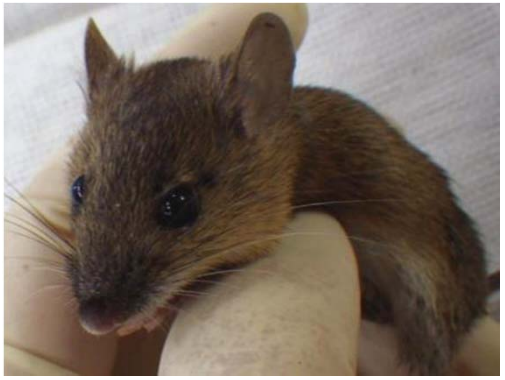
Fukushima-city



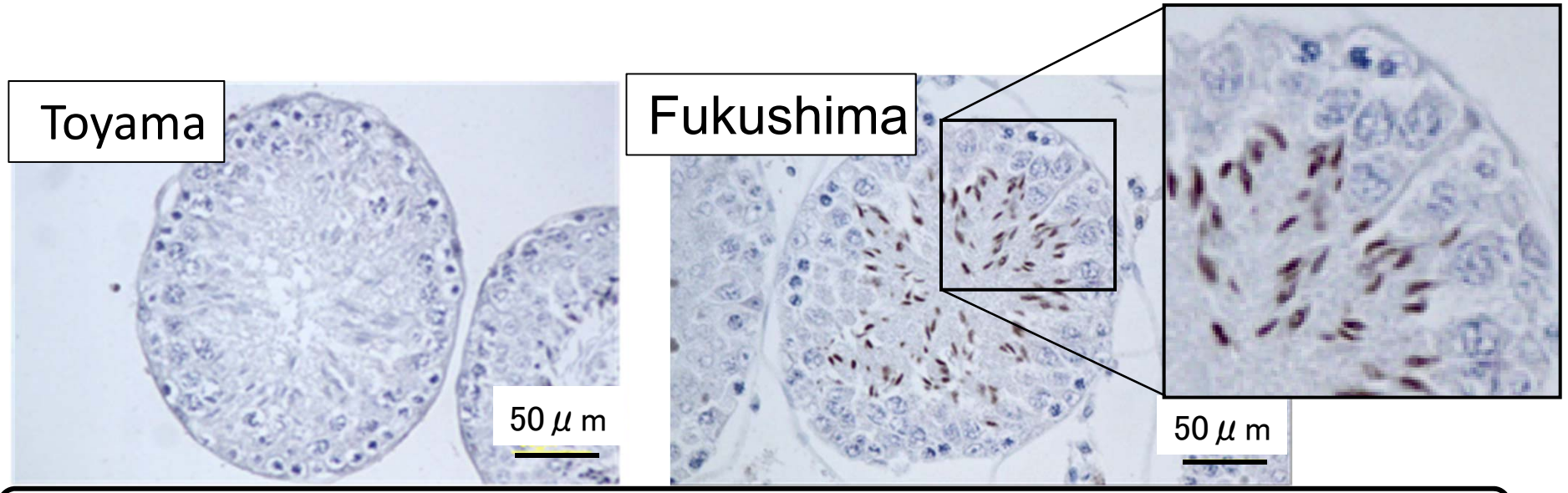
Ecosystem evaluation

(1) Effect for Wild animals

Wild mouse, *Apodemus speciosus*, were captured at high-gamma-dose area in Fukushima and low-gamma-dose area (Aomori and Toyama).



Check DNA oxidization by gamma irradiation at sperm cells using 8-OHdG antibody



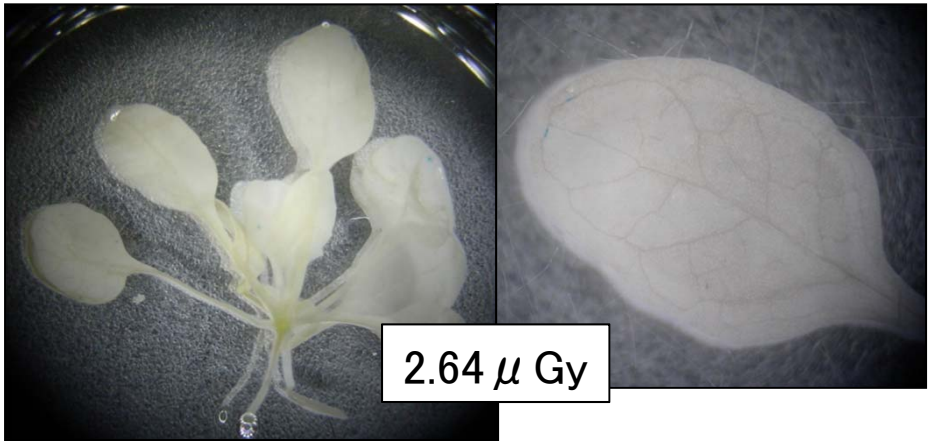
DNA oxidization in sperm cells was remarkable in Fukushima

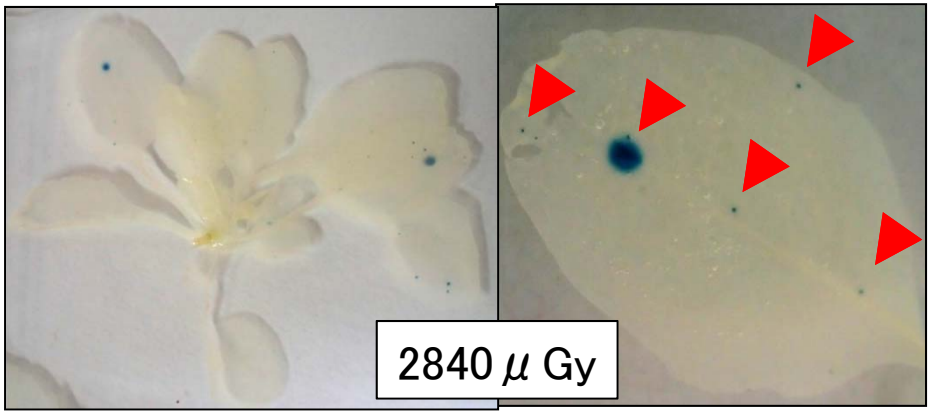
Ecosystem evaluation (to be continued)

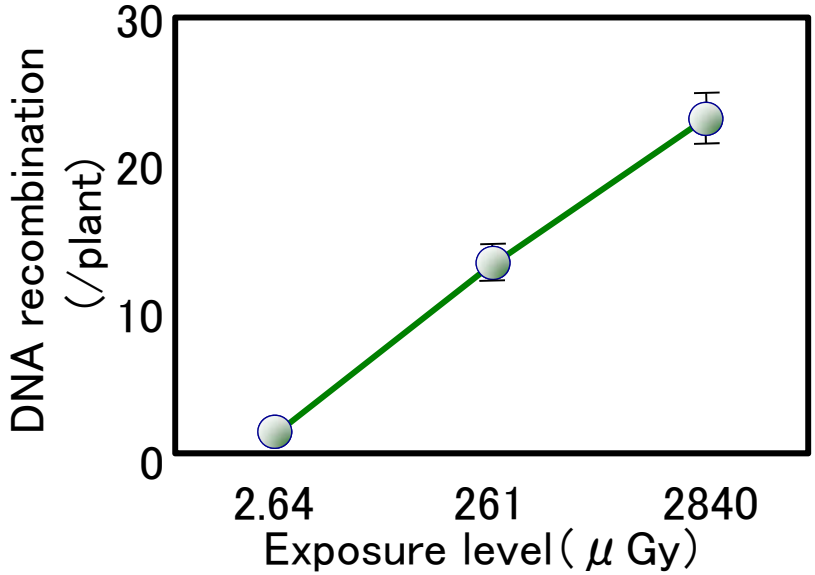
(2) Plants (development of DNA damage monitoring system)

Established transgenic plants that can detect homologous DNA recombination results from DNA damage by gamma-irradiation

Plants were grown on contaminated soil collected from Fukushima, then we estimated frequency of DNA recombination



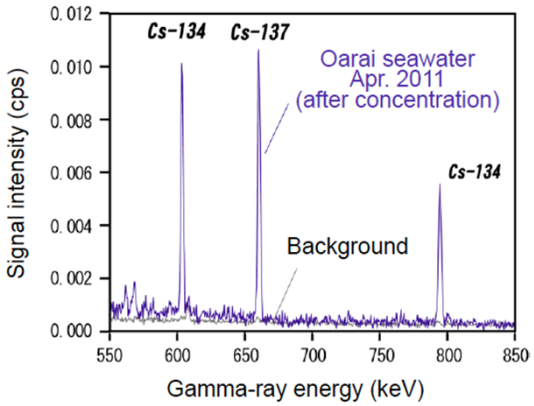


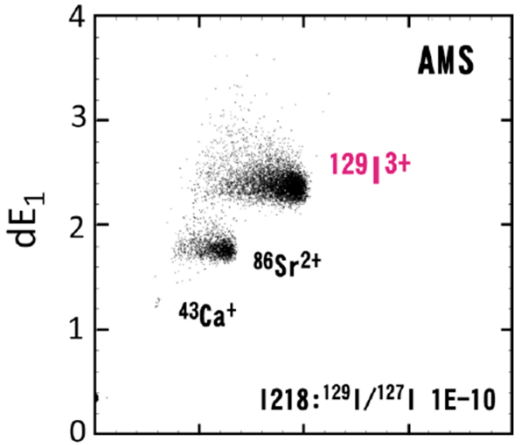


DNA damage by gamma-irradiation can be assessed with the transgenic plant

Analytical method development

- Dissolved radioactive ceasium
  - Rapid and trace measurement of dissolved Cs in water using Cs selective disc
- Radioactive strontium
  - An effective solid phase extraction method developed using crown ether resin
- Iodine 129
  - Ultra trace determination of ^{129}I by an accelerator MS
  - Reconstruction of short-lived ^{131}I distribution
- Imaging plate
  - Disposition in organisms, house dust and wastes





^{129}I Analysis by Accelerator Mass Spectrometry

[Purpose of the study]

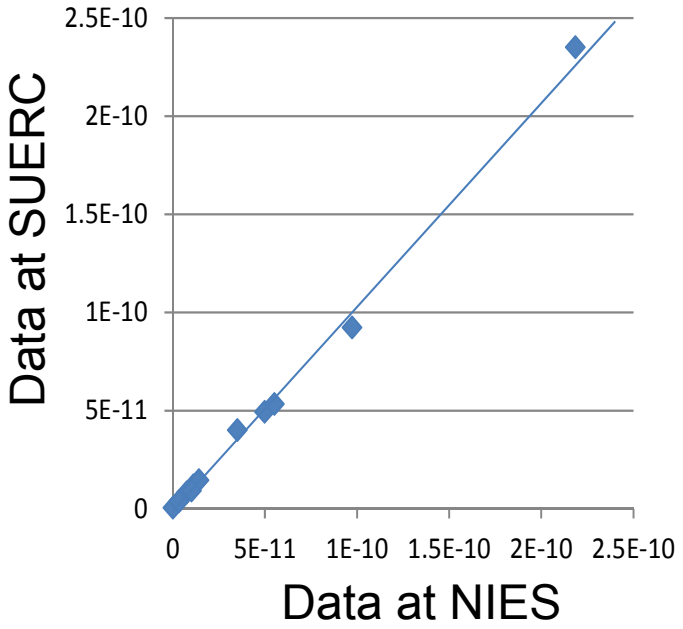
- To reconstruct distribution of short-lived radioactive iodine (^{131}I) by using ^{129}I (half life = 15.7 million y) as surrogate

[Current status and future plan]

- Establish sample preparation and analytical condition
  => compare the data with AMS at Scottish U. Environ. Res. Center
- Analyze air filters to reveal ^{129}I/^{131}I ratios emitted from Fukushima Daiichi NPP

^{129}I/^{131}I: Quartz filters=7.4~9.9
Active carbon f.=7.1~13.3

to analyze more samples for improving reliability of the data and revealing temporal / areal variations



Future work

- Better understanding of the dynamics of radionuclides in environment based on the field measurements
- Long-term monitoring (terrestrial, aqueous, oceanic environment)
- Improve and validate the fate models and simulations
- Calibrate exposure model using more data (Food, drinking water, dust, soils, ...)
- Combining fate models and exposure models for long-term exposure assessment
- Better understanding of the impacts on wildlife and ecosystem health

NIES's plan in Fukushima-ken Environmental Creation Center

Three Research Programs