Integrated watershed approach in support of environmental impact assessment including remediation

AMORAD French project [2013-2019]

Olivier Evrard, P. Laceby, S. Huon, O. Cerdan, I. Lefèvre, H. Jaegler, F. Pointurier, H. Lepage, C. Chartin, Y. Onda, S. Hayashi, O. Cerdan, V. Landemaine, R. Vandromme, L. Garcia-Sanchez *and others*

ALLIANCE – NERIS workshop, Munich

March 28, 2018

IRSN
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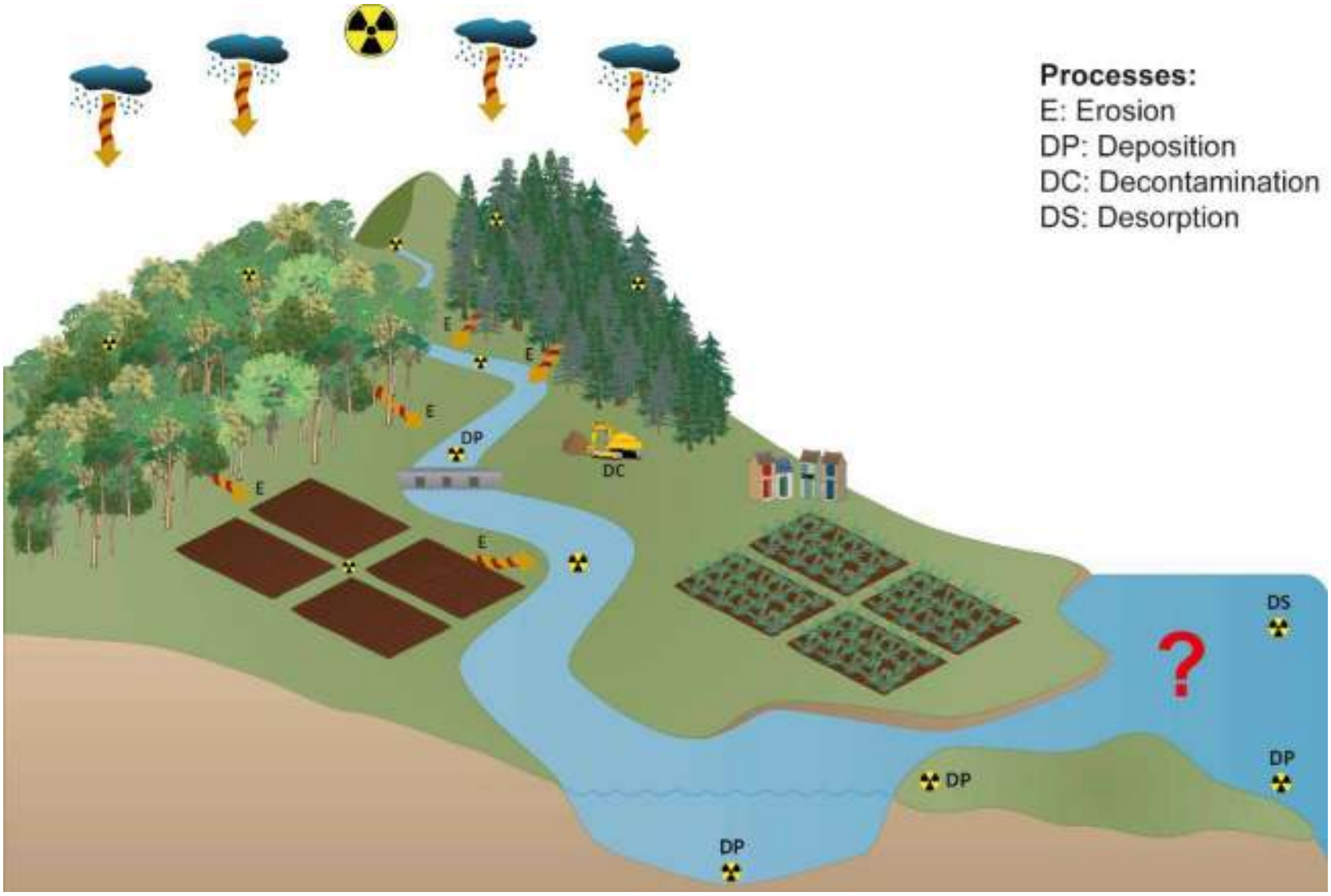
université PARIS-SACLAY
iEES
brgm
TOFU
National Institute for Environmental Studies, Japan

7 years ago….

(Source: The Telegraph, 2011)

(Source: Emily Welenc's Blog)


Processes:
E: Erosion
DP: Deposition
DC: Decontamination
DS: Desorption
E
E
E
E
E
DP
DC
DS
?
DP
DP
DP

Diversity of radionuclides released by FDNPP

Table 1
Radionuclides detected in environmental samples after the FDNPP accident.

Radionuclide	Half-life (d)	Half-life (y)	Reference
^{129m}Te	0.05	–	Tazoe et al. (2012)
^{132}I	0.10	–	Shozugawa et al. (2012)
^{135}Xe	0.38	–	Steinhauser (2014)
^{91}Sr	0.40	–	Shozugawa et al. (2012)
^{140}La	1.70	–	Shozugawa et al. (2012); Tagami et al. (2011)
^{133m}Xe	2.20	–	Steinhauser (2014)
^{239}Np	2.35	–	Shozugawa et al. (2012)
^{132}Te	3.30	–	Shozugawa et al. (2012)
^{133}Xe	5.25	–	Steinhauser (2014)
^{131}I	8.00	–	Shozugawa et al. (2012); Tagami et al. (2011); Tazoe et al. (2012)
^{131m}Xe	11.90	–	Steinhauser (2014)
^{140}Ba	12.80	–	Shozugawa et al. (2012)
^{136}Cs	13.20	–	Tagami et al. (2011)
^{129m}Te	33.60	–	Tagami et al. (2011); Tazoe et al. (2012)
^{95}Nb	35.00	–	Shozugawa et al. (2012); Tagami et al. (2011)
^{103}Ru	39.40	–	Steinhauser (2014)
^{59}Fe	44.60	–	Shozugawa et al. (2012)
^{89}Sr	50.53	–	Steinhauser (2014)
^{91}Y	58.50	–	Shozugawa et al. (2012)
^{95}Zr	64.00	–	Shozugawa et al. (2012)
^{35}S	87.37	–	Steinhauser (2014)
^{110m}Ag	252.00	–	Shozugawa et al. (2012); Tagami et al. (2011); Tazoe et al. (2012)
^{134}Cs	–	2.06	Tagami et al. (2011); Tazoe et al. (2012)
^{85}Kr	–	10.75	Steinhauser (2014)
^{3}H	–	12.32	Kakiuchi et al. (2012); Steinhauser (2014)
^{241}Pu	–	14.33	Steinhauser (2014)
^{137}Cs	–	30.20	Shozugawa et al. (2012); Tagami et al. (2011); Tazoe et al. (2012)
^{238}Pu	–	87.70	Steinhauser (2014)
^{241}Am	–	432.60	Steinhauser (2014)
^{14}C	–	5730.00	Steinhauser (2014)
^{240}Pu	–	6561.00	Steinhauser (2014)
^{239}Pu	–	24110.00	Shozugawa et al. (2012); Steinhauser (2014)
^{99}Tc	–	2.10E+05	Steinhauser (2014)
^{36}Cl	–	3.00E+05	Steinhauser (2014)
^{129}I	–	1.57E+07	Steinhauser (2014)
^{236}U	–	2.34E+07	Steinhauser (2014)
^{235}U	–	7.04E+08	Steinhauser (2014)
^{238}U	–	4.47E+09	Steinhauser (2014)

Short-lived

Long-lived

(Evrard, Laceby et al., JER 2015)

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Short-lived

Long-lived

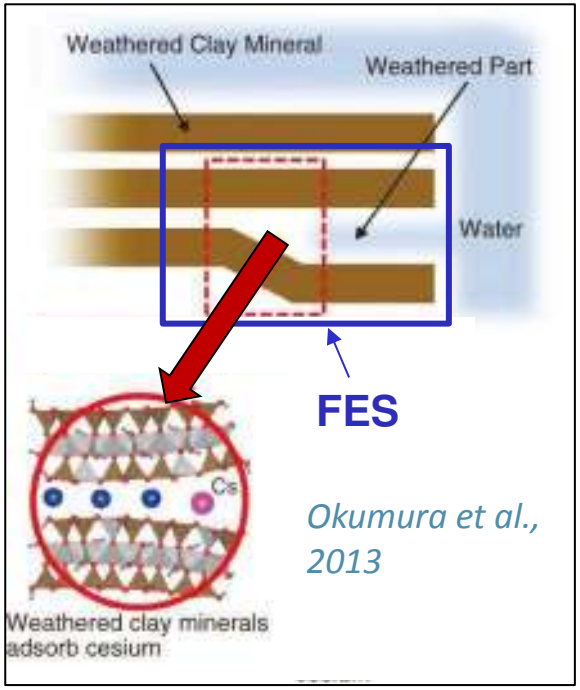
Cesium-134 ($t_{1/2}$: 2y) Unique FDNPP Accident Tracer

Cesium-137 ($t_{1/2}$: 30y) Long-term Human Health Concern

(Evrard et al., JER 2015)

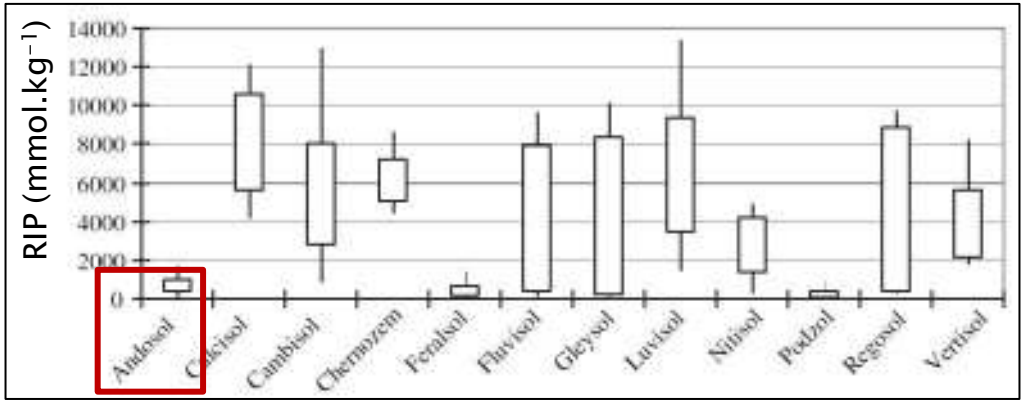
Behaviour of radiocesium in Japanese Andisols

^{137}Cs strongly fixed on Frayed Edge Sites (FES)



Okumura et al., 2013

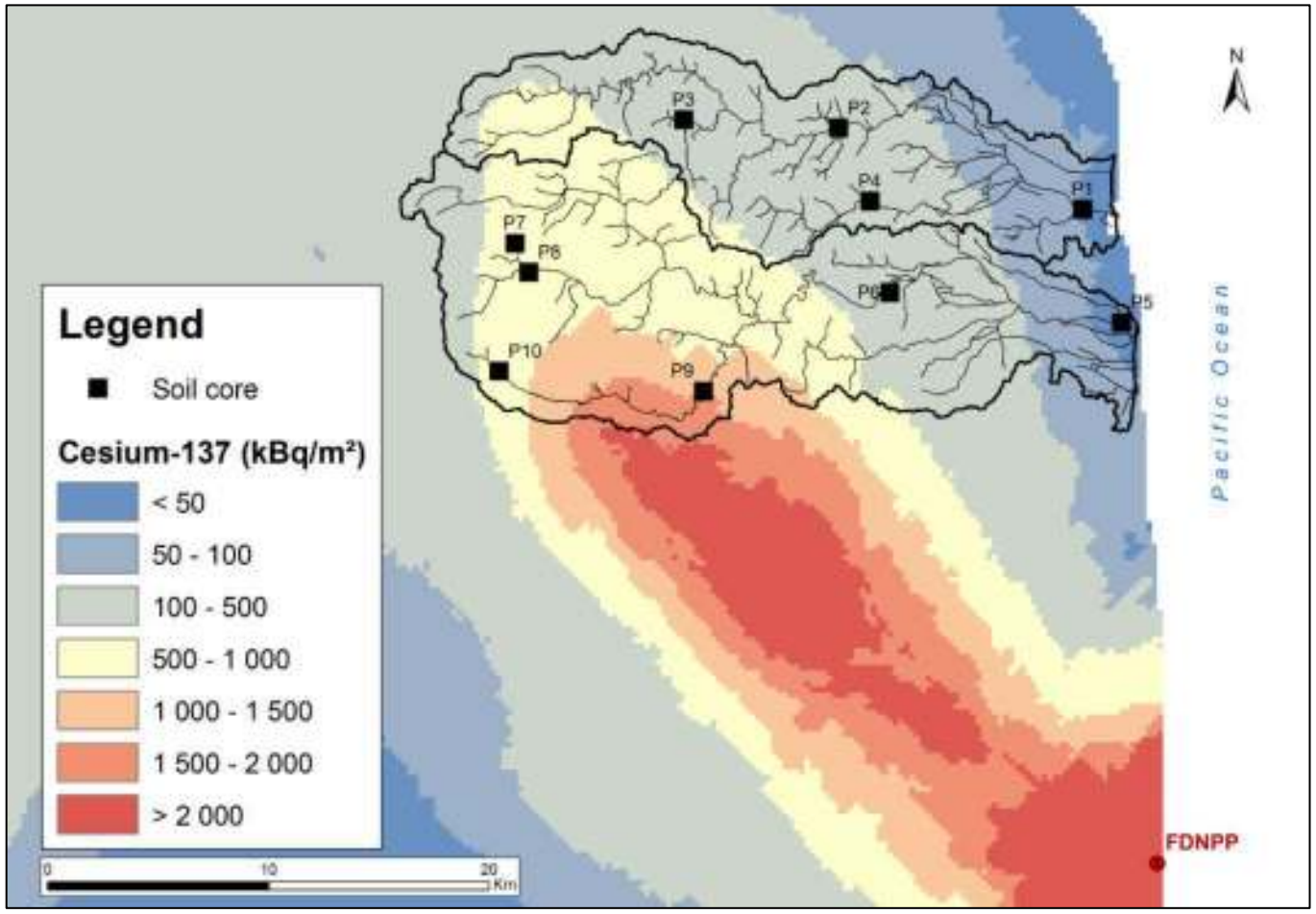
Andisol characterized by low FES



Vandebroek et al., 2012

Did ^{137}Cs remain in the uppermost centimetres and could it be transported by erosion ?

Behaviour of radiocesium in Japanese Andisols



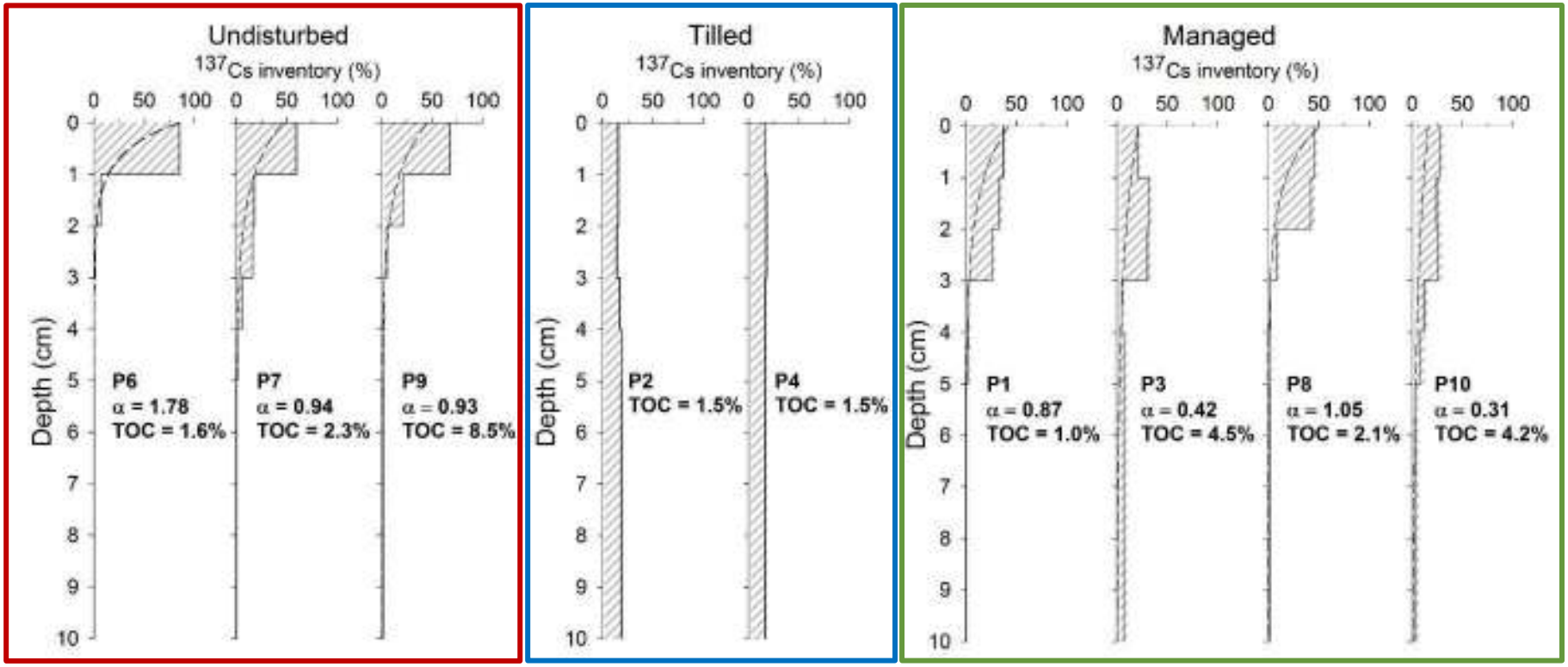
Focus on paddy fields

(highly connected to the rivers)

cm
0-1
1-2
2-3
3-4
4-5
5 - 10

Lepage et al., 2015 (JER)

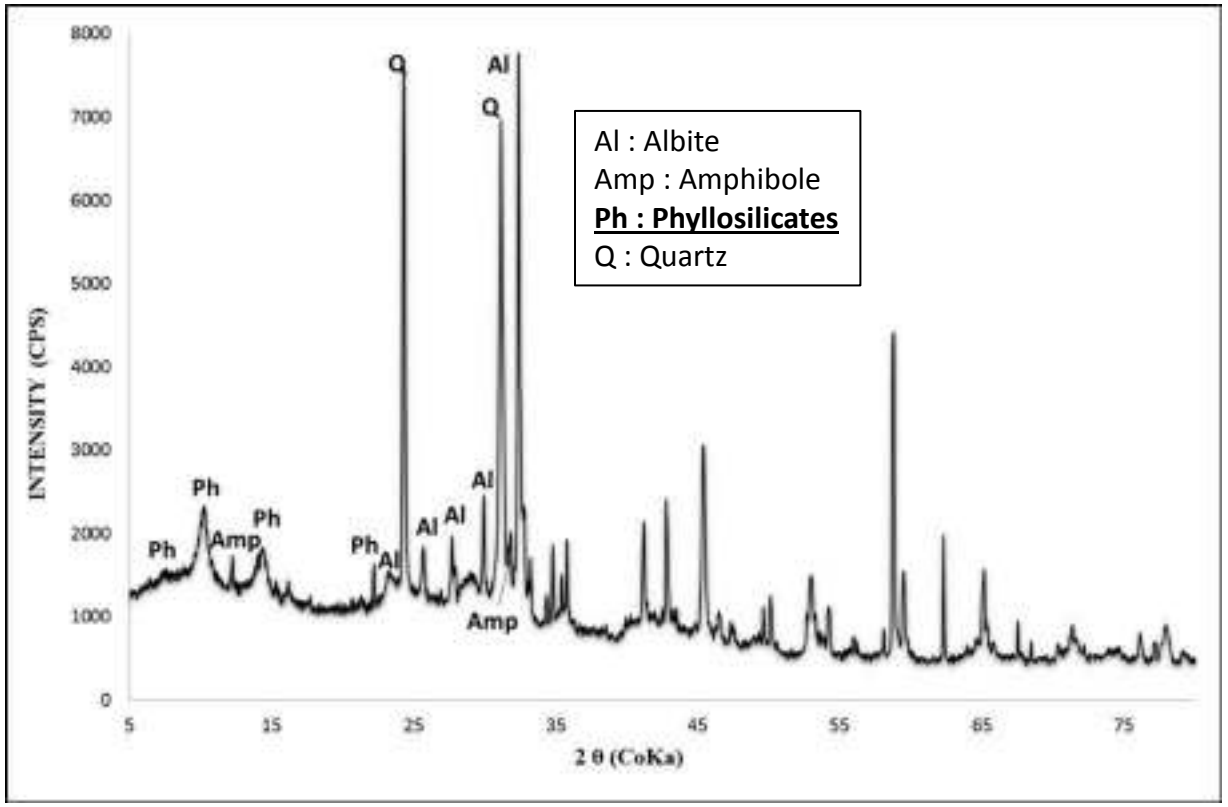
Behaviour of radiocesium in Japanese Andisols



- Major part of ^{137}Cs remains in the uppermost 5 cm even 30 months after the accident.
  - Natural migration of radiocesium is very low in paddy field soils in the studied catchments

Lepage et al., 2015 (JER)

Why does radiocesium strongly bind to Japanese Andisols?



XRD analysis:

- Presence of **phyllosilicates** (chlorite, kaolinite, illite) known to strongly fix radiocesium *(Nakao et al., 2012)*

Sources of these minerals:

- Eroded in the upstream mountains and transported to the paddy field by river *(Nemoto et al., 2013)*
- Aeolian deposition of dust from China (*Eguchi et al., 2013)*

Radiocesium

- **^{134}Cs and ^{137}Cs** are considered the most serious risk to the current and long term health of the local population (Kitamura et al., 2014; Saito et al., 2015)
- Radiocesium is strongly and rapidly fixated to fine soil particles (He and Walling, 1996; Sawhiney, 1972)
- Majority of cesium is transported with the **particulate** fraction (see table – Mean K(d) ~5 x 10^5)

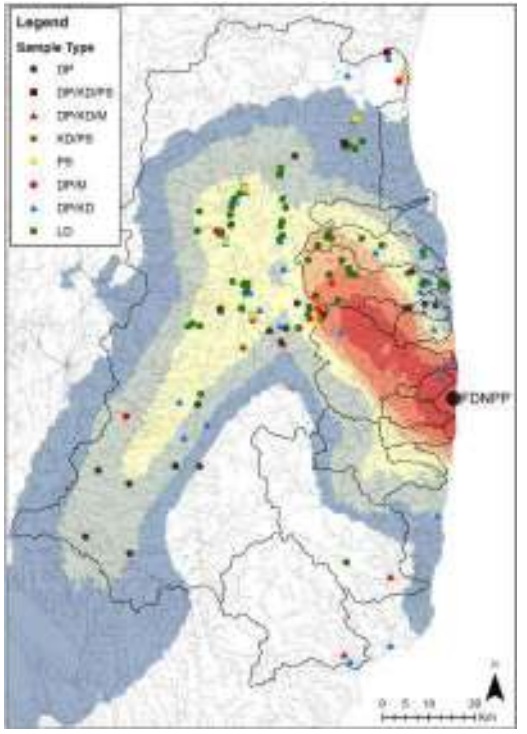


Table 3
Summary of research examining radiocesium in particulate/dissolved and K_d.

Author(s)	Catchment(s)	Sampling period	Radionuclide(s)	Particulate fraction of transported ^{137}Cs (%)	Average K (d)
Nagao et al. (2013)	Same/Natsui	July 2011–Dec. 2011	^{134}Cs & ^{137}Cs	99 flood conditions 33 base flow	2.5×10^5
Nagao et al. (2014)	Niida	May 2011–Nov. 2012	^{134}Cs & ^{137}Cs	47 (before Sept. 2011) 84 (after Sept. 2011)	N/A
Sakaguchi et al. (2015)	Abukuma	June 2011–Dec. 2012	^{137}Cs	80	8.8×10^5
Shinomiya et al. (2014)	Experimental Catchment (Abukuma)	June 2012	^{134}Cs & ^{137}Cs	92–97 flood conditions 12–96 base flow	N/A
Tsuji et al. (2014)	Abukuma, Ota	Sept. 2012–May 2013	^{134}Cs & ^{137}Cs	64	N/A
Ueda et al. (2013)	Niida	July 2011–Nov. 2011	^{134}Cs & ^{137}Cs	>90 flood conditions 40 base flow	3.5×10^5
Yamashiki et al. (2014)	Abukuma	June 2011–May 2012	^{134}Cs & ^{137}Cs	82–93	N/A
Yoshikawa et al. (2014)	Abukuma	April 2012–Sept. 2012	^{137}Cs	60–88 flood conditions 69–95 base flow	1.2×10^5 base 10.6×10^5 event
Yoshimura et al. (2015b)	Abukuma, Mano, Same, Fujiwara, Niida, Ota, Odaka, Asami, Ukedo,	Dec. 2012	^{134}Cs & ^{137}Cs	58–100[a]	3.6×10^5

[a] Excluding an estuary site.

(Evrard, Laceby et al., JER 2015)

Climate

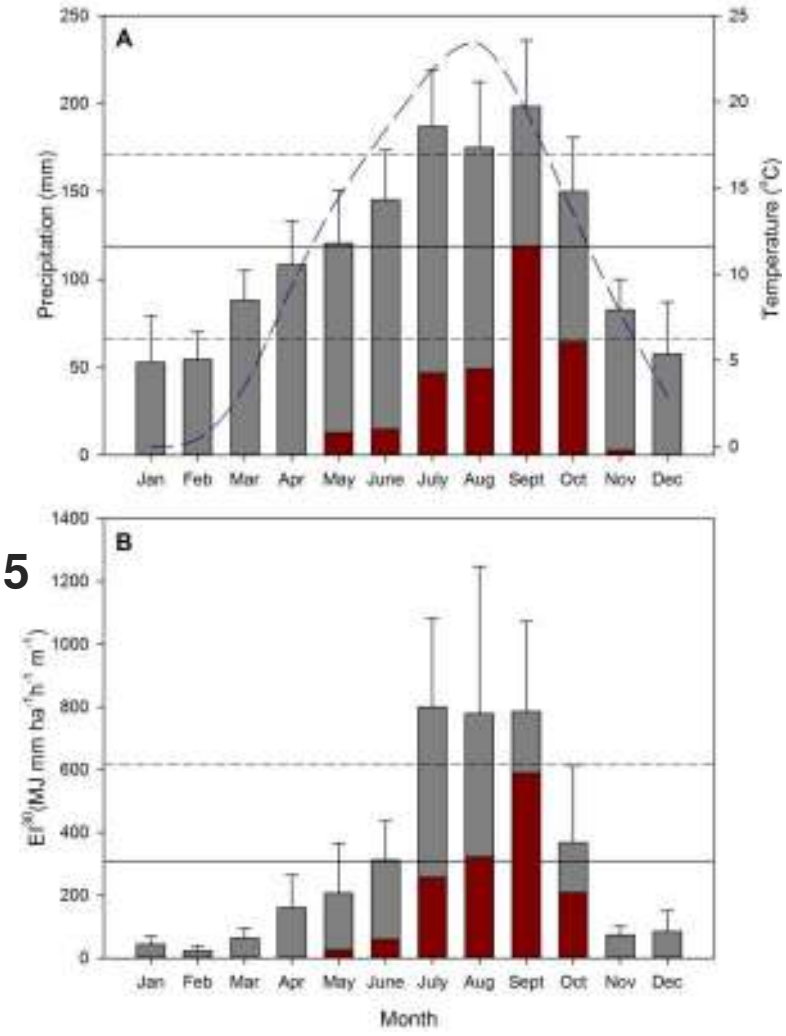
Seasonal Variability

Total precipitation:

60% between June and October

Rainfall erosivity:

82% between June and October



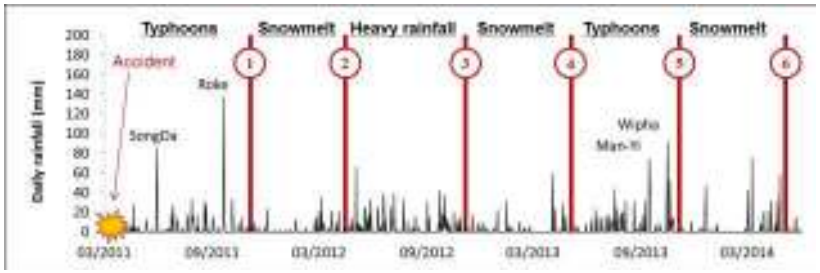
17 typhoons/tropical storms between 2011-2015

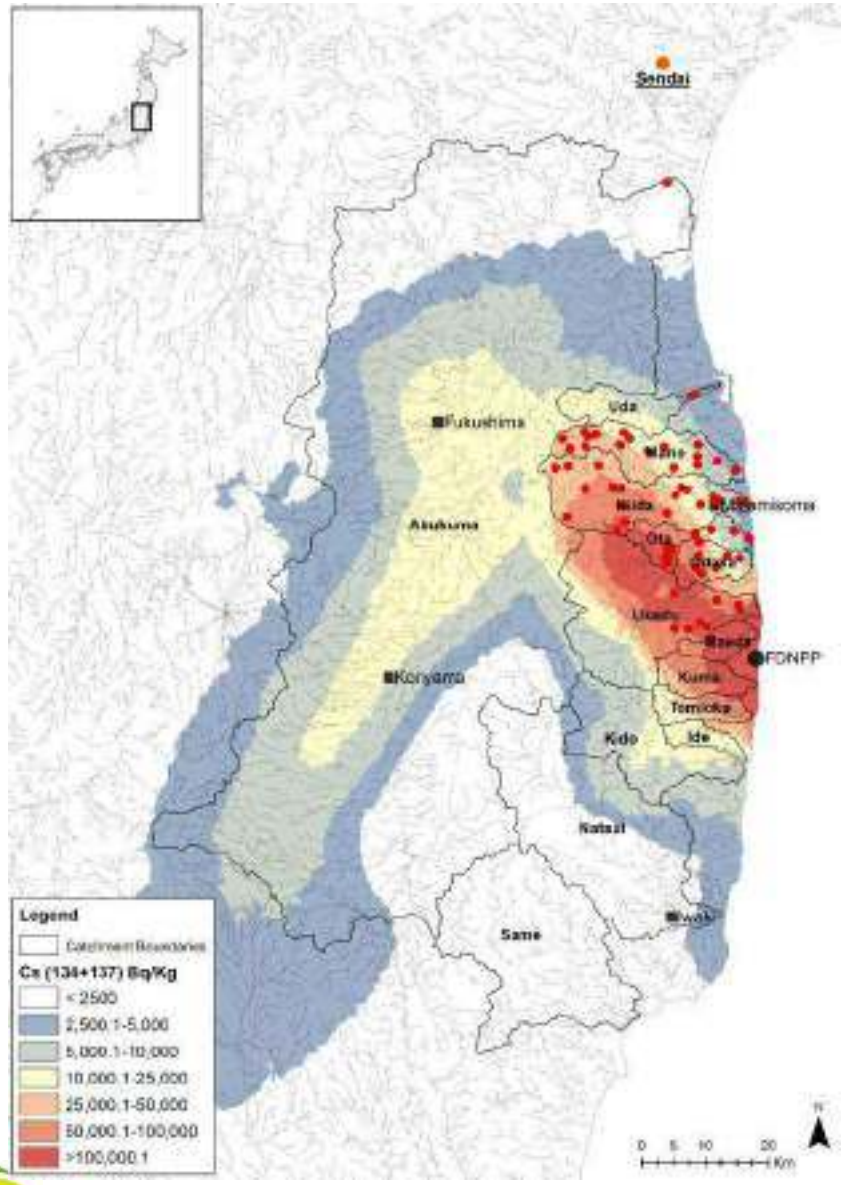
- ***Largest:***
  - **Roke** (2011)
    Precip - 231 mm (SD 48, Max 339)
    R - **1695** (SD 1066, Max 5089)MJ mm ha h
  - **Etau** (2015)
    Precip - 240 mm (SD 125, Max **573**)
    R - **1581** (SD 1477, Max **7887**) MJ mm ha h

(Laceby et al., 2016 HESS)

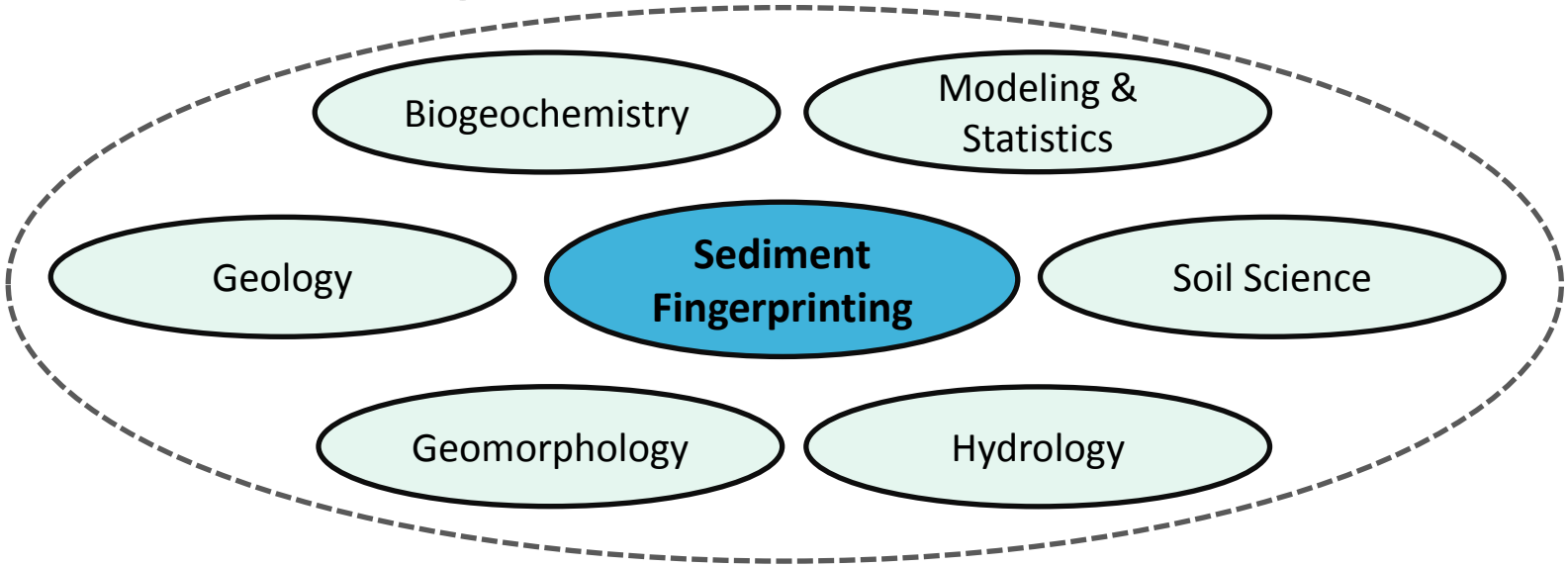
Fieldwork

SURVEYS EVERY 6 MONTHS



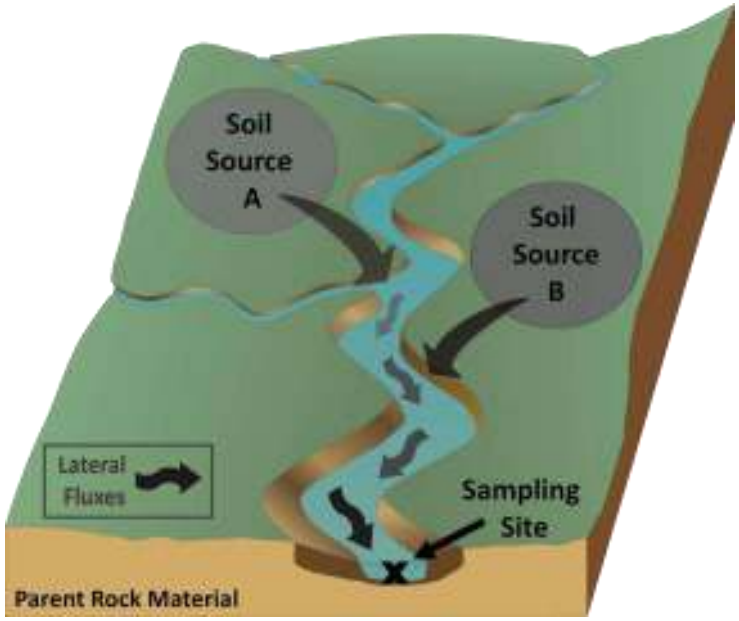


Sediment Tracing



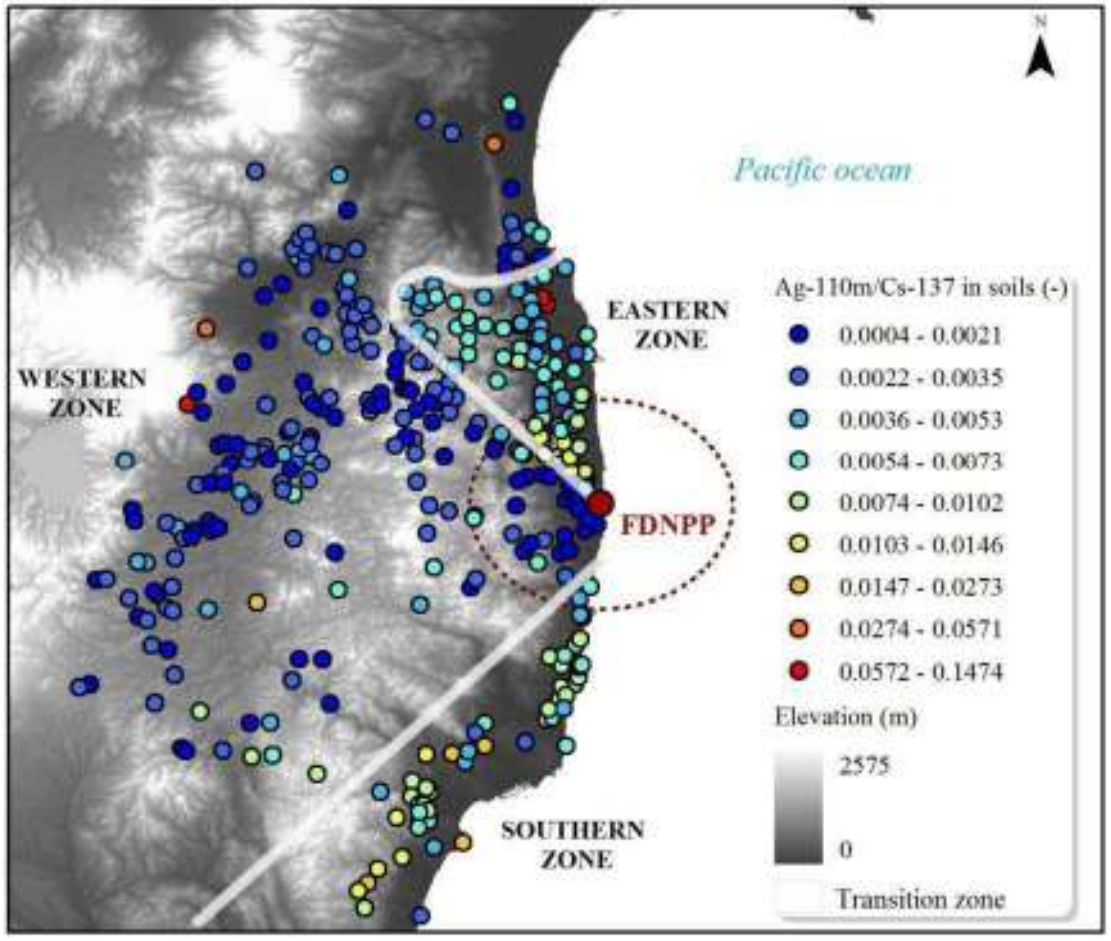
Three main approaches:

1. **Spatial pattern of radionuclide deposition**
2. Soil types (geochemistry)
3. Land use sources (organic matter)



Spatial pattern of ^{110m}Ag deposition

Homogeneous ^{134}Cs:^{137}Cs ratio (~ 1) in soils across the area

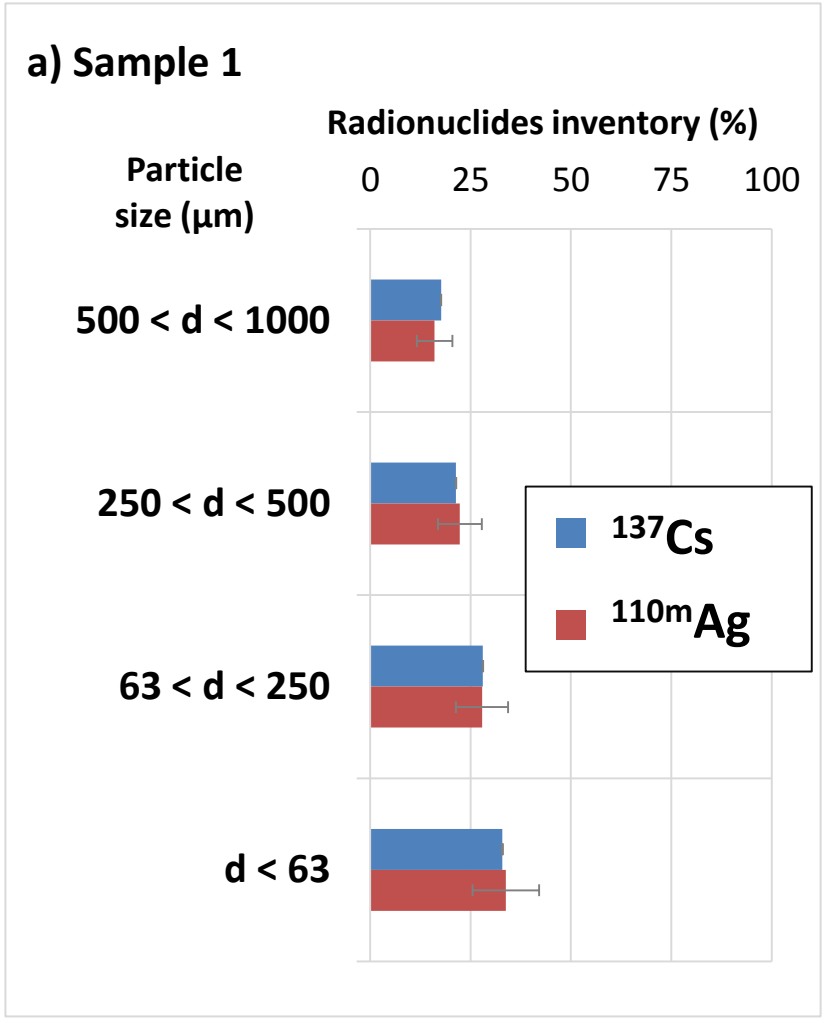


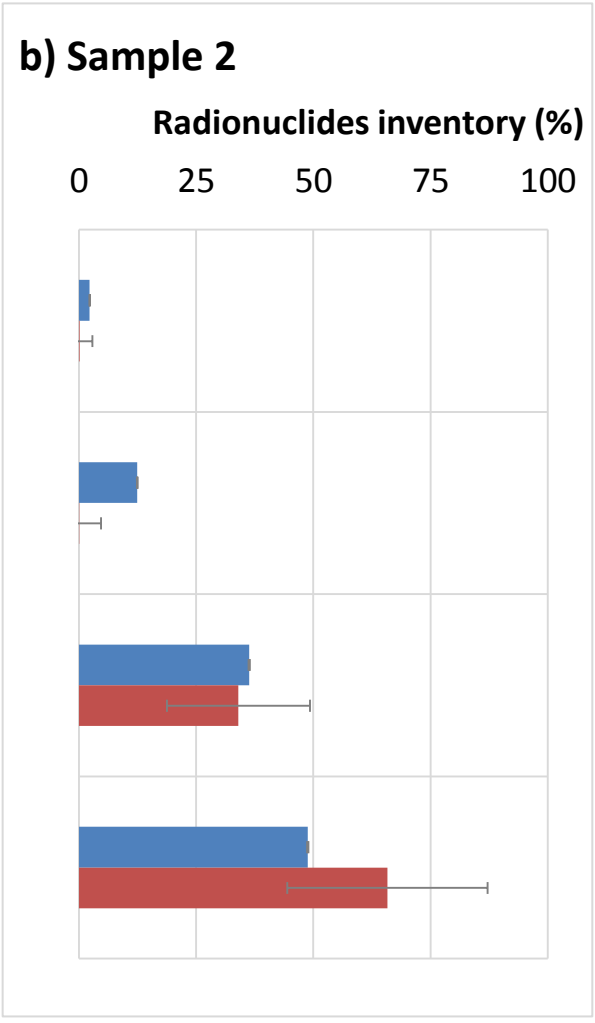
Large spatial variations of ^{110m}Ag :^{137}Cs ratio

Chartin et al. (2013) *Anthropocene*.

Spatial pattern of ^{110m}Ag deposition

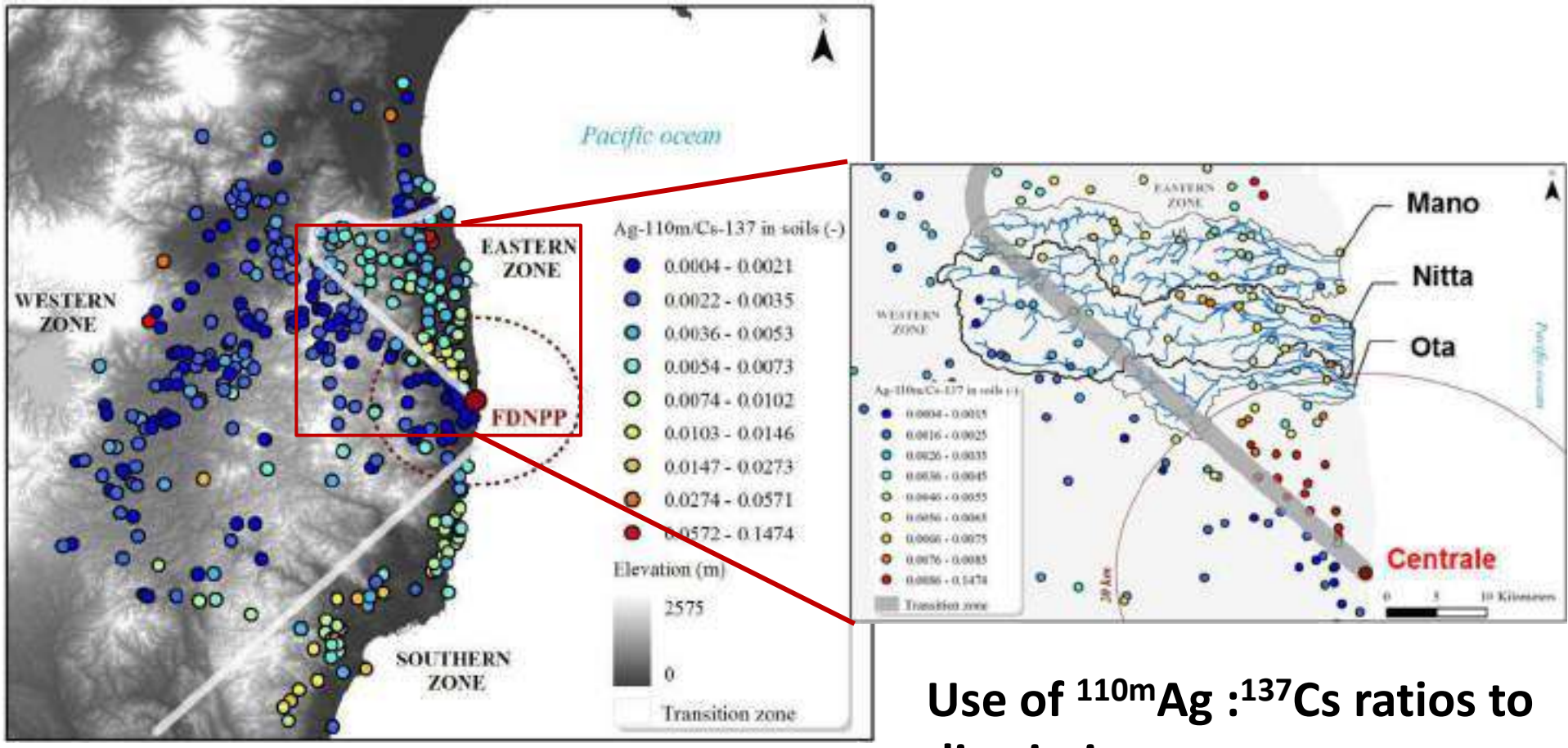
Affinity for the finest particle fractions





Lepage et al., 2014 (JER)

Spatial pattern of ^{110m}Ag deposition

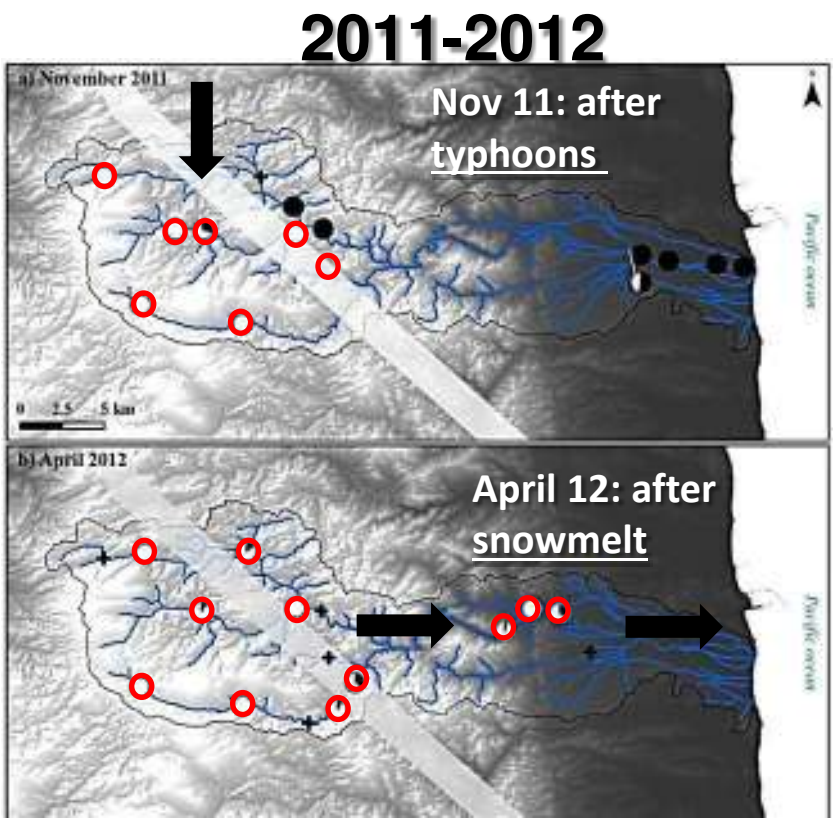


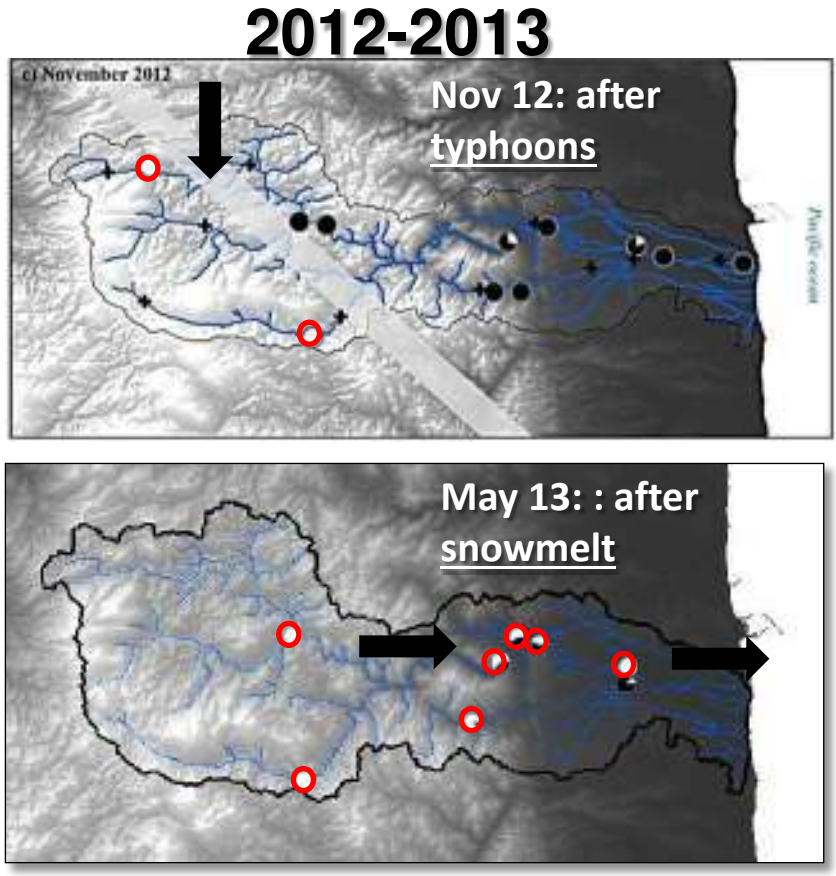
(Chartin et al., 2013)

Use of ^{110m}Ag :^{137}Cs ratios to discriminate upstream vs. downstream soils

Spatial pattern of ^{110m}Ag deposition

Seasonal cycle of dispersion



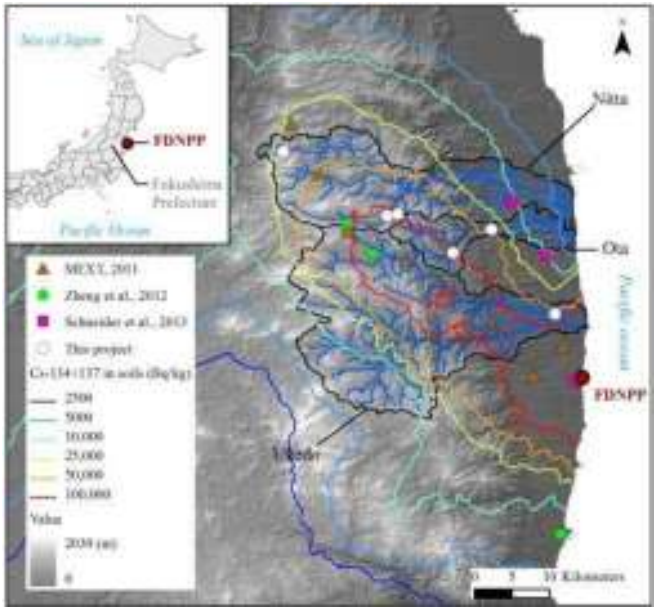


Proportion (%) of upstream soil

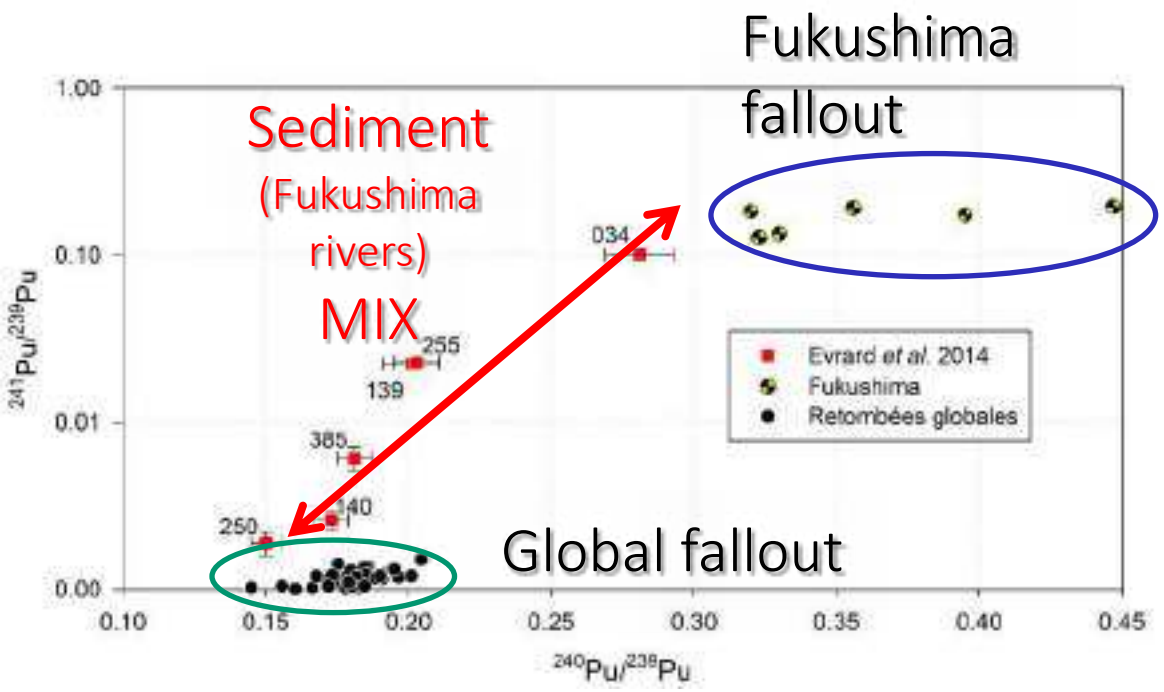
Proportion (%) of downstream soil

Rapid decay of ^{110m}Ag (half-life of 250 d) and low initial fallout levels

What about longer-lived radioisotopes ?



- Analysis of Pu isotopes by mass spectrometry (^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu).
- Detection of Pu **ultra-traces** in flood sediment deposits along Fukushima rivers.

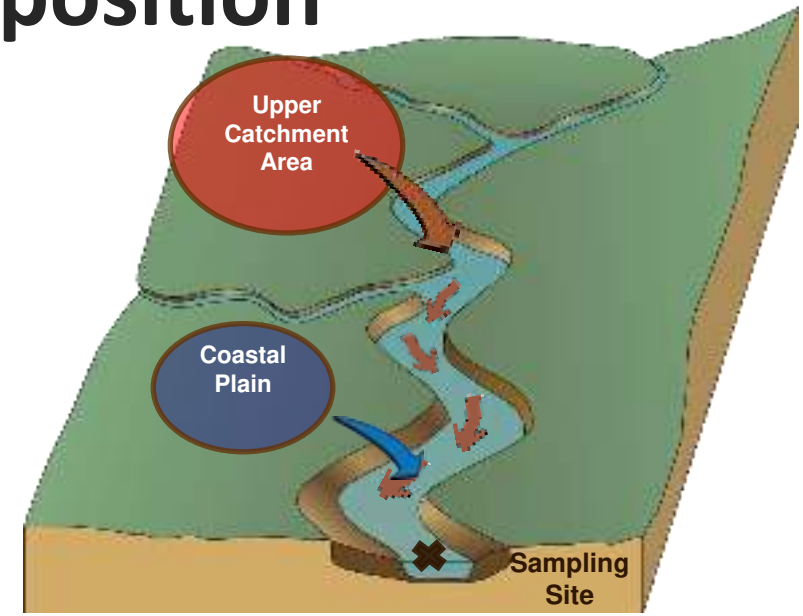


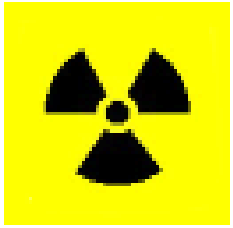
Evrard et al., *ES&T* (2014)

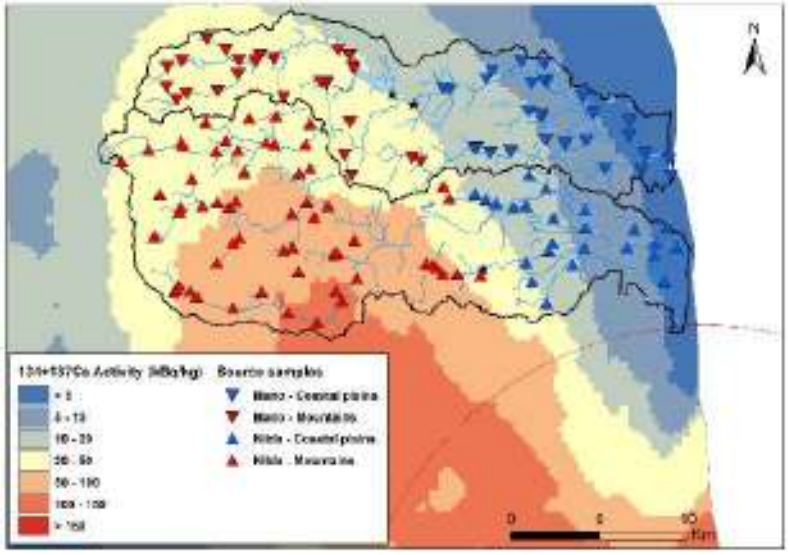
Spatial pattern of ^{137}Cs deposition

Two major sources characterised by contrasting contamination levels in ^{137}Cs:

- Coastal plain
- Upper catchment region

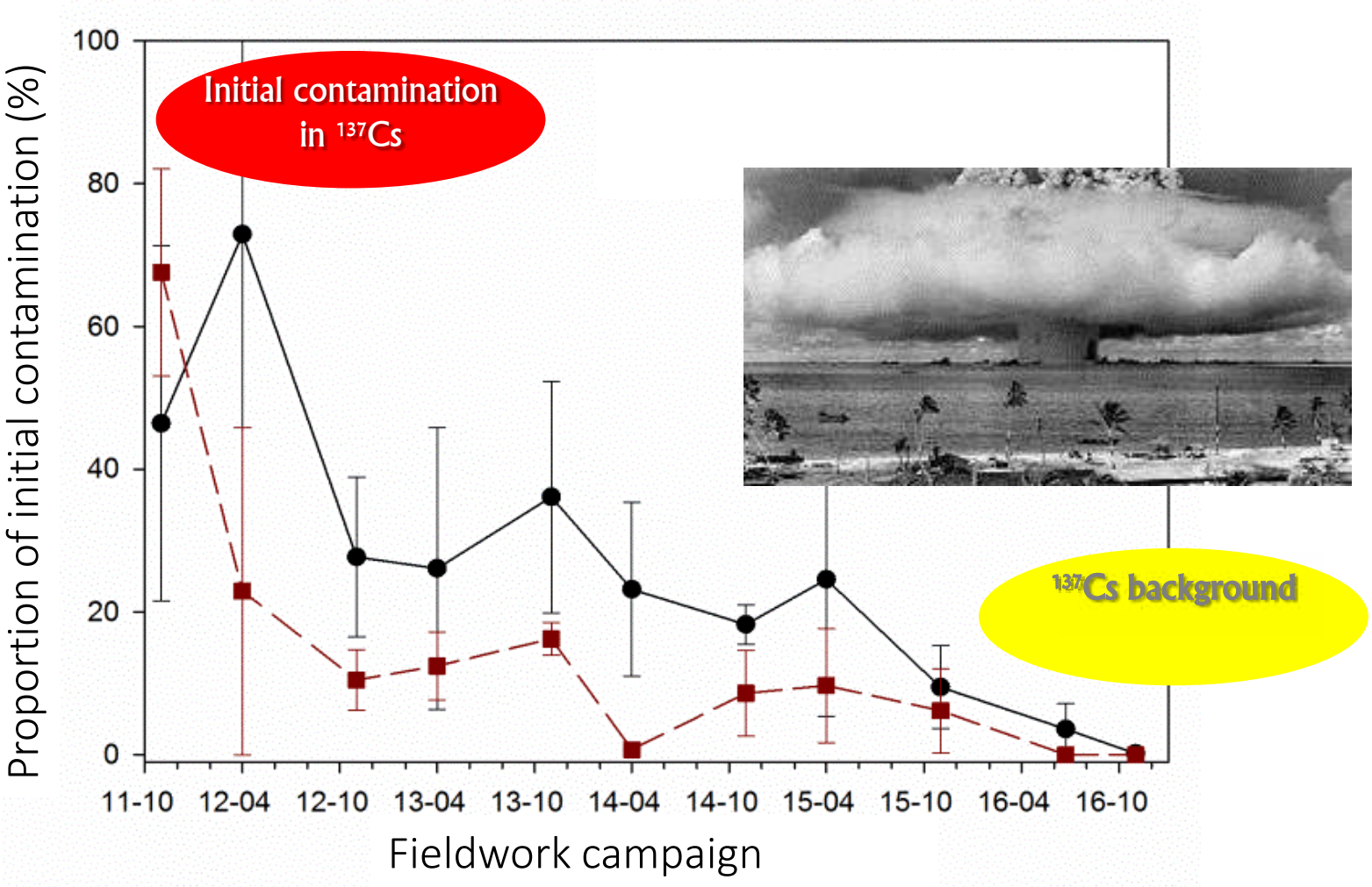






(Lepage et al., 2016 Anthropocene)

Strong decrease of the initial contamination from 2011 to 2017



After Evrard et al., *Sc. Rep*. (2016)

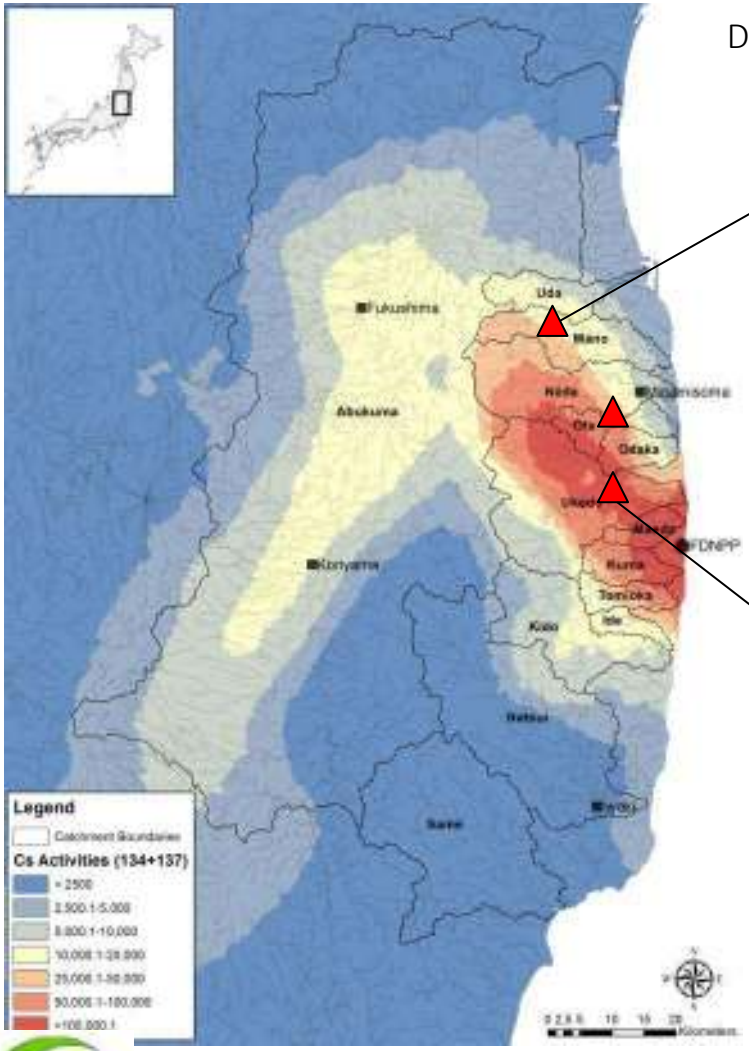
How can we explain this decrease?

Where is the residual contamination stored today?

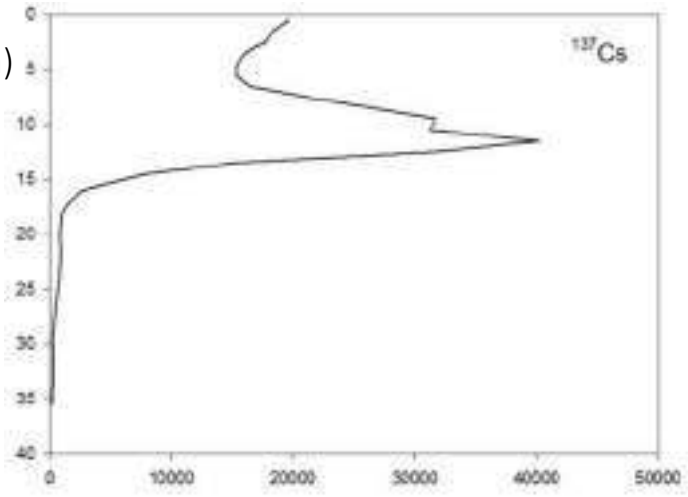
Forests

Dams/reservoirs

Today, the main sources of contamination are forests and reservoirs



Depth (cm)



^{137}Cs (Bq/kg)

Mano Dam

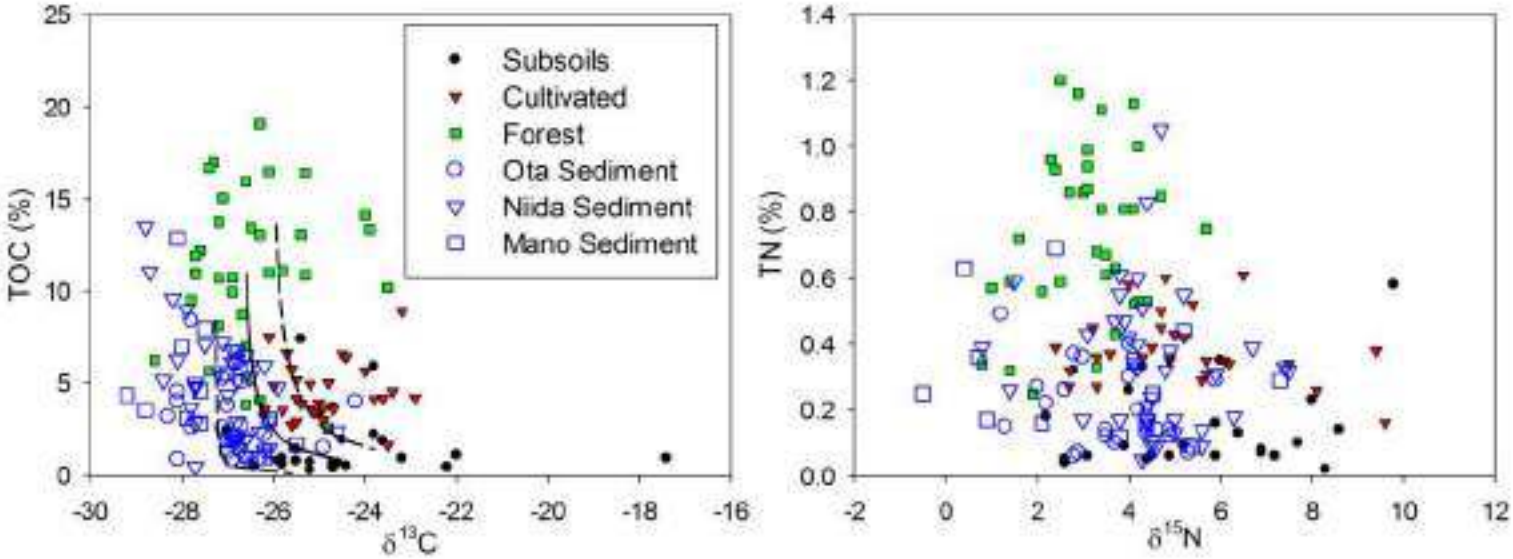
Ogaki Dam

National Institute for Environmental Studies

独立行政法人
日本原子力研究開発機構
Japan Atomic Energy Agency

Focus on the Mano Dam Reservoir

Where does the sediment accumulated in the reservoir come from?



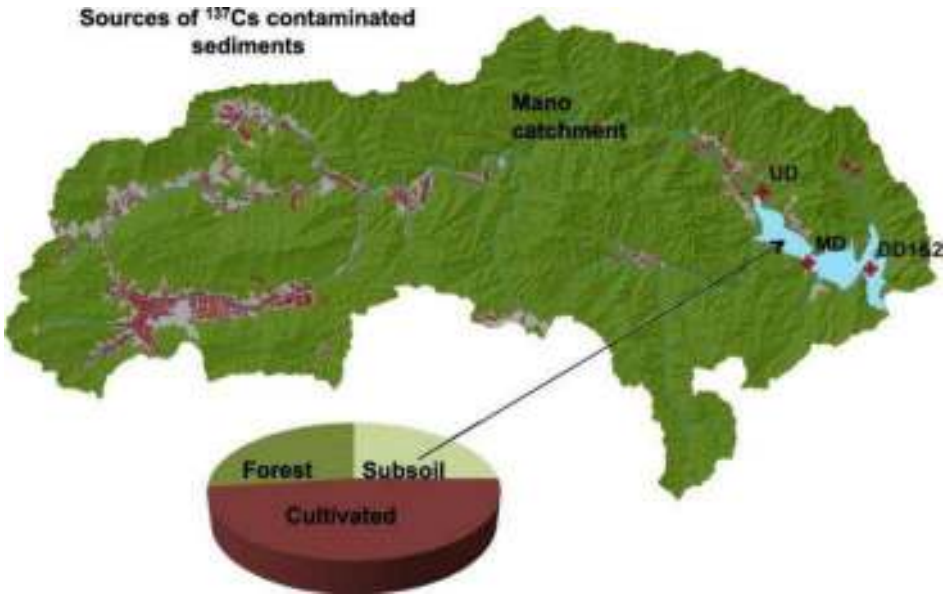
Sources and particulate matter parameters for the <63 µm fraction with artificial concentration dependent mixing lines

Laceby et al. (2017)

Source contributions to Mano Dam sediment

Cultivated sources contributed 48%

(SD 10%)



Forests contributed 27%

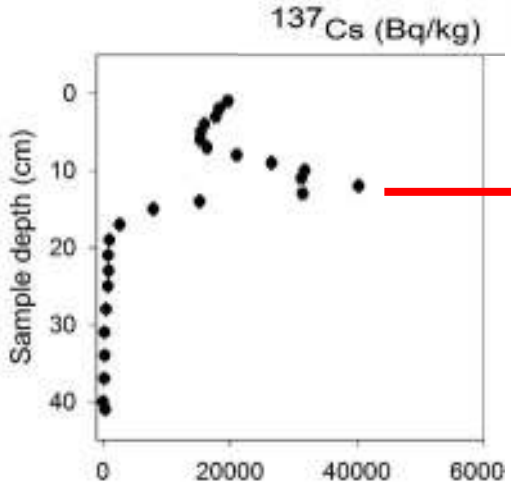
(SD 10%)

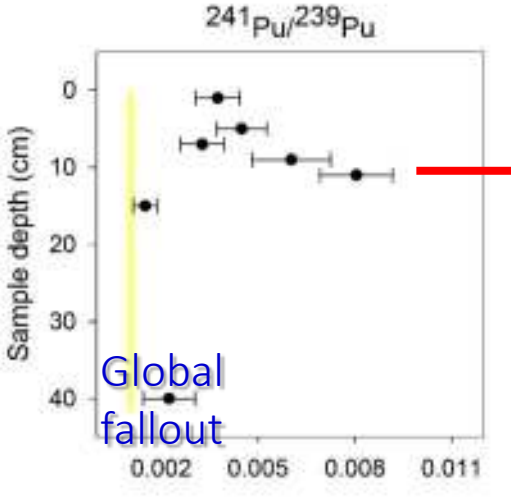
Subsoils contributed 26%

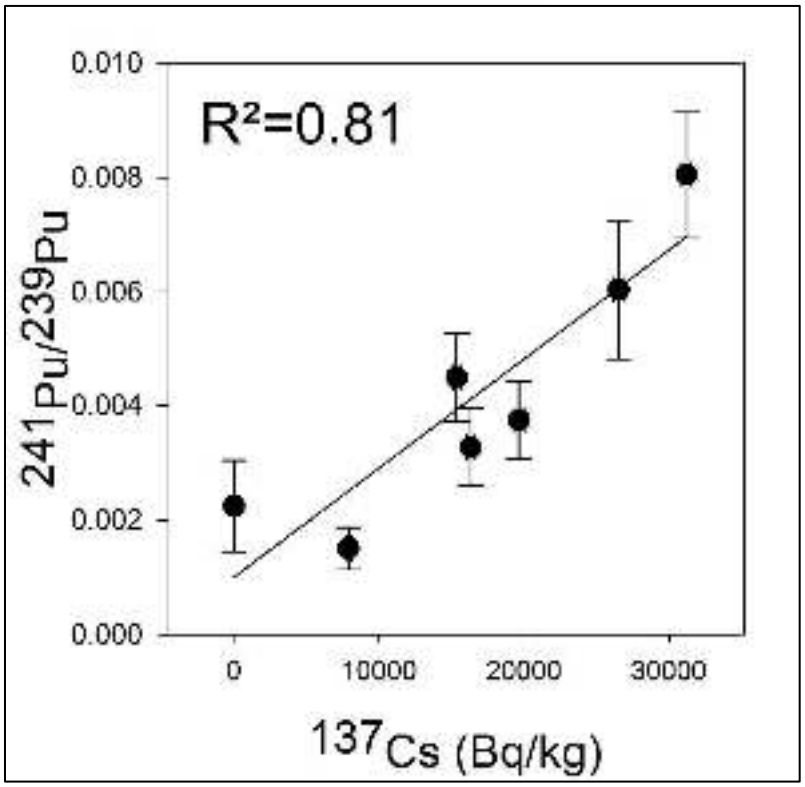
(SD 8%)

(Huon et al, 2018 STOTEN)

Changes of radionuclide contents/signatures with depth in the dam

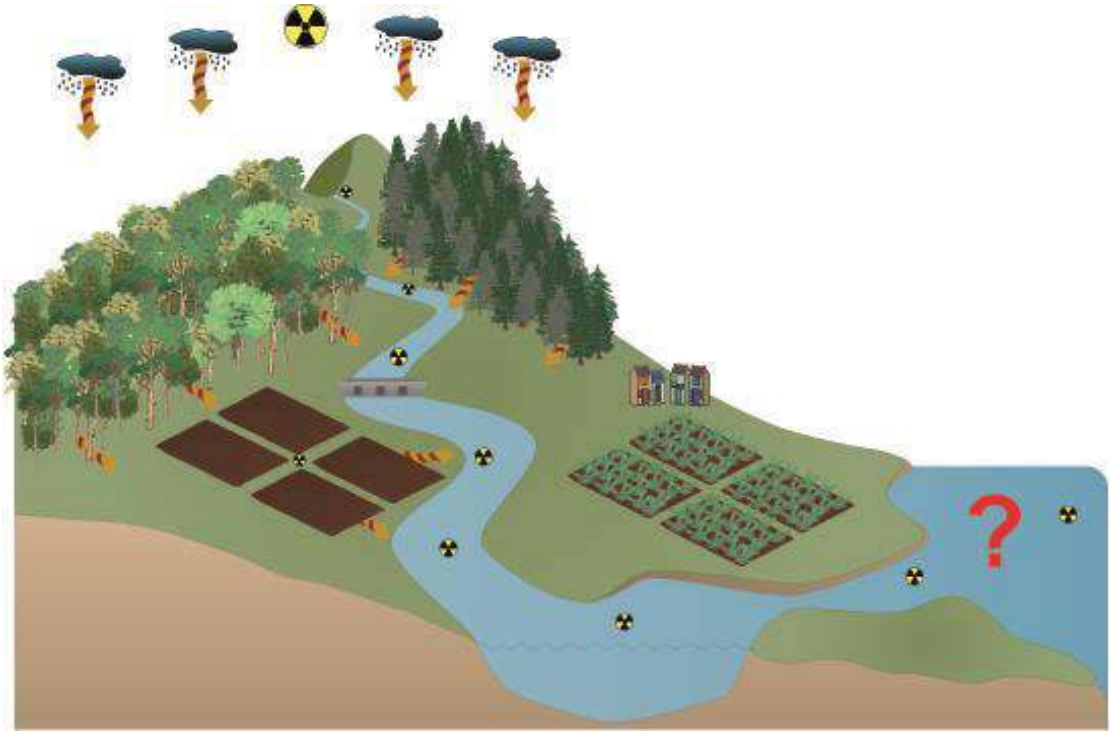




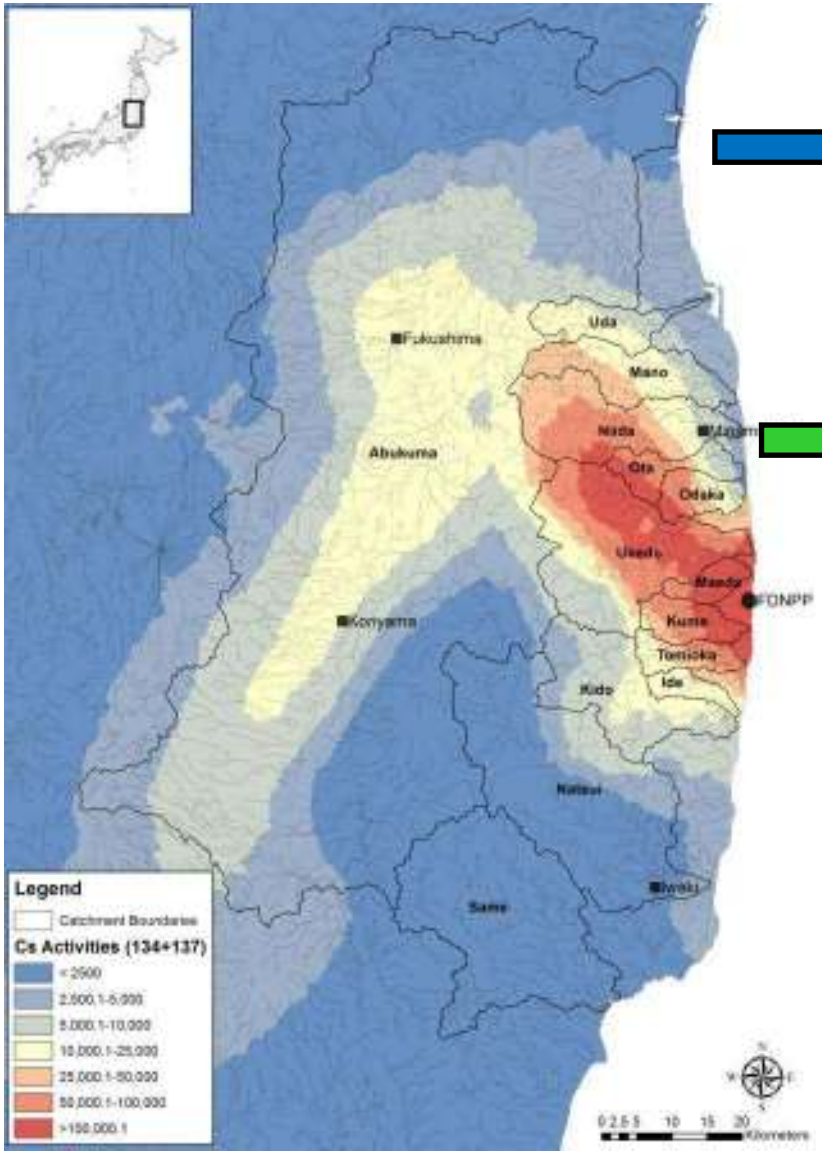


Jaegler et al. (*in preparation*)

WHAT WE KNOW 7 YEARS AFTER THE ACCIDENT



Radiocesium exports to the Pacific Ocean



Abukuma River

3–10 TBq yr^{-1}

(Kitamura et al., 2014; Yamashiki et al., 2014)

Coastal rivers ~ 5 TBq yr^{-1}

(Kitamura et al., 2014)

FDNPP direct releases in March 2011: 12–41 PBq

(Bailly du Bois et al., 2012)

Later leakages

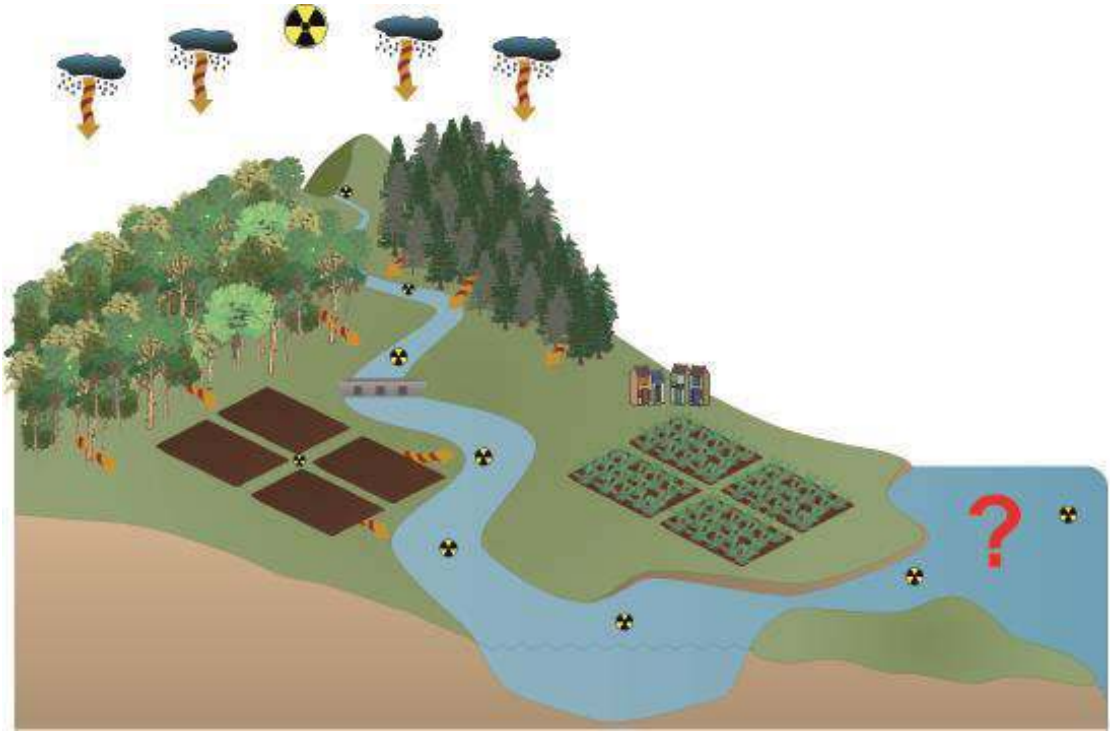
June 2011 – Sept 2012 : 17 TBq

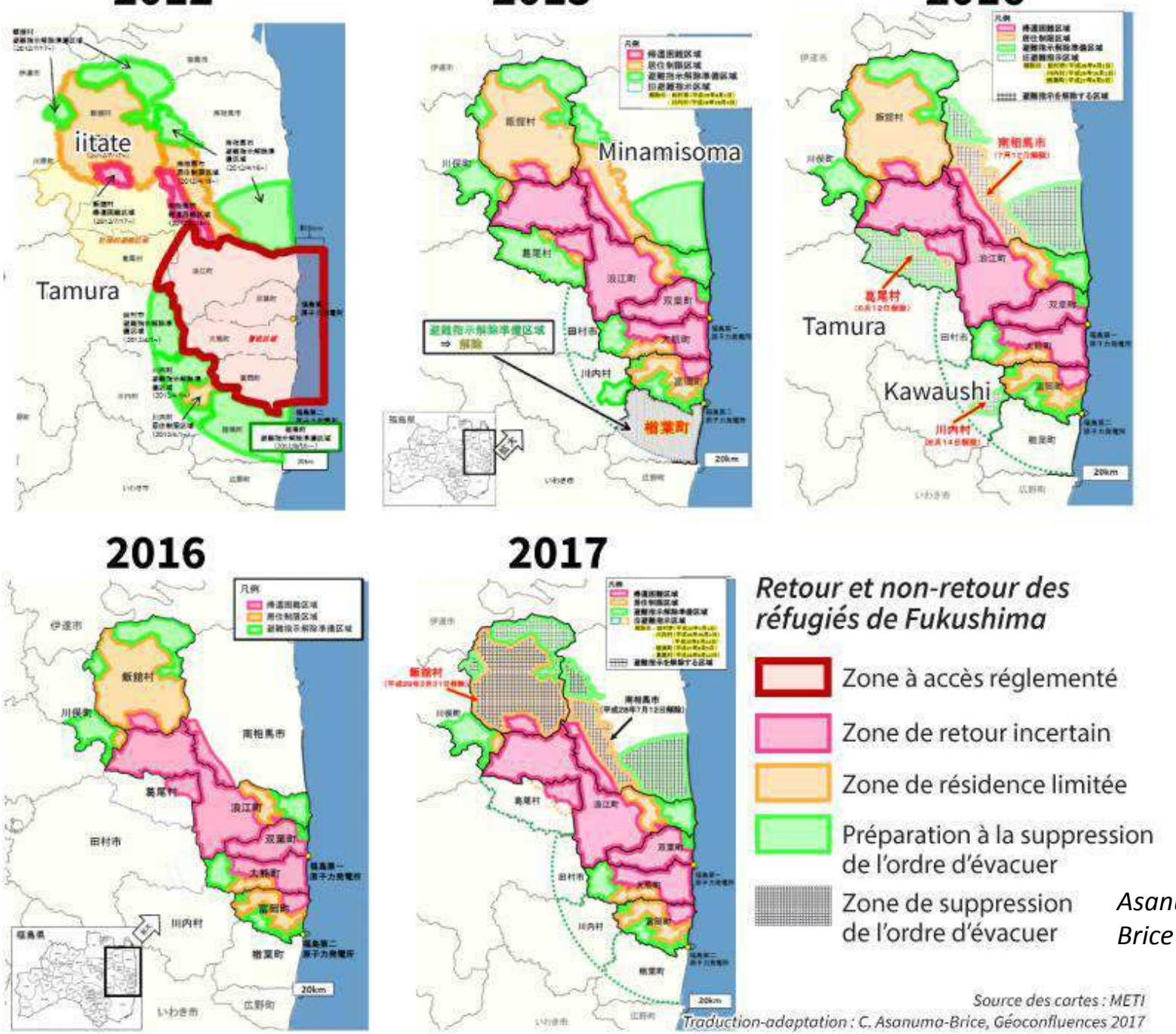
August 2012: 24 TBq

(Yamashiki et al., 2014)

1 TBq = 10^{12} Bq; 1 PBq = 10^{15} Bq

WHAT WE DON'T KNOW 7 YEARS AFTER THE ACCIDENT



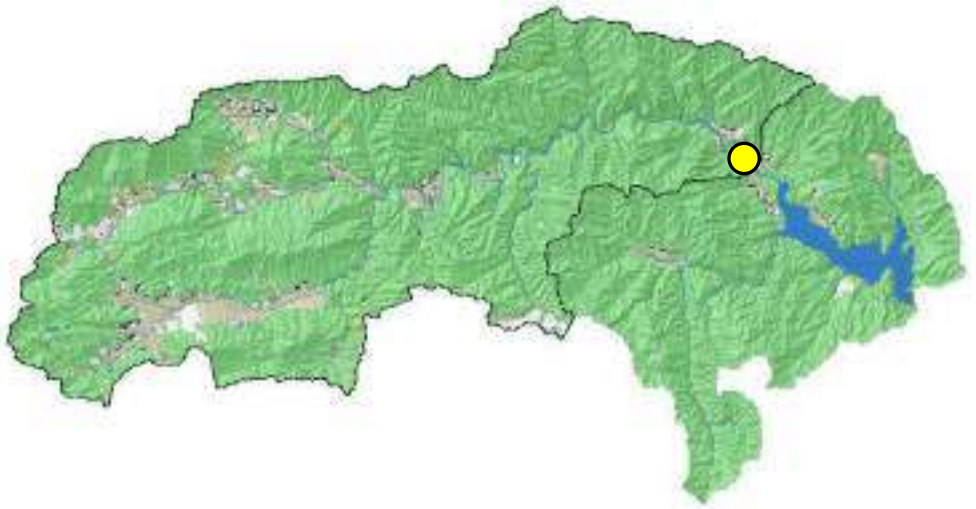

2012
2015
2016
2016
2017
iitate
Tamura
Minamisoma
Tamura
Kawaushi
南相馬市
葛尾村
川内村
楢葉町
飯舘村
川俣町
浪江町
双葉町
大熊町
富岡町
田村市
広野町
いわき市
20km
Retour et non-retour des réfugiés de Fukushima
Zone à accès réglementé
Zone de retour incertain
Zone de résidence limitée
Préparation à la suppression de l'ordre d'évacuer
Zone de suppression de l'ordre d'évacuer
Asanuma-Brice (2017)
Source des cartes : METI
Traduction-adaptation : C. Asanuma-Brice, Géoconfluences 2017

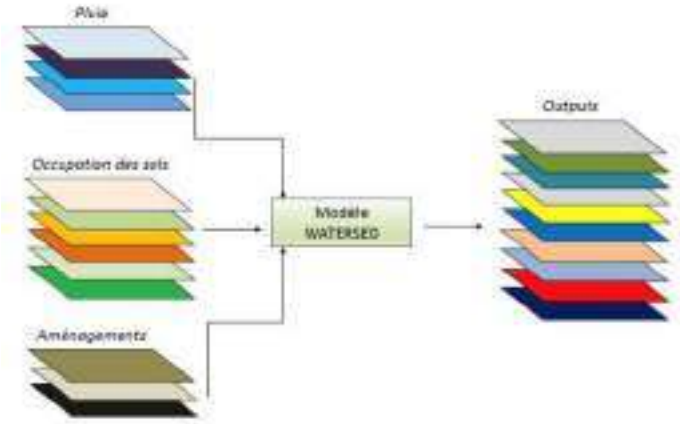
Asanuma-Brice (2017)

?

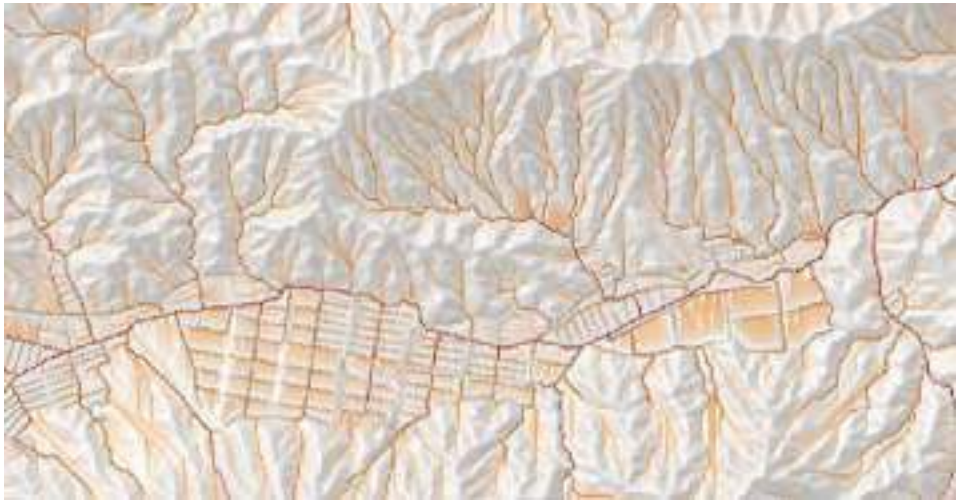
Perspectives

Adapt the **WaterSed** model to the Mano Dam catchment.





*The WaterSed model is the next iteration of the STREAM model (Cerdan et at., 2002)



Develop a **radiocesium** module

Conclusions

- Considerable lab and fieldwork has been conducted in the coastal **river catchments** draining the main **radioactive pollution plume** since 2011;

- The flood events (triggered by typhoons) and human actions have significantly **reduced** the **radiocesium contamination levels** measured in sediment transiting these rivers;

- After the completion of **remediation** works, future research should focus on the impact of the **return of refugees** and their practices on further radionuclide redistribution;

- Improvement of **models** (e.g. ongoing work on WaterSed) and investigation of **forgotten radionuclides** are essential to meet this goal !

Many thanks for your attention!

Believe in the Power of Knowledge. Be Together.

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