Cs and Sr transfers in Chernobyl Pilot Site soils (Chernobyl Exclusion Zone)

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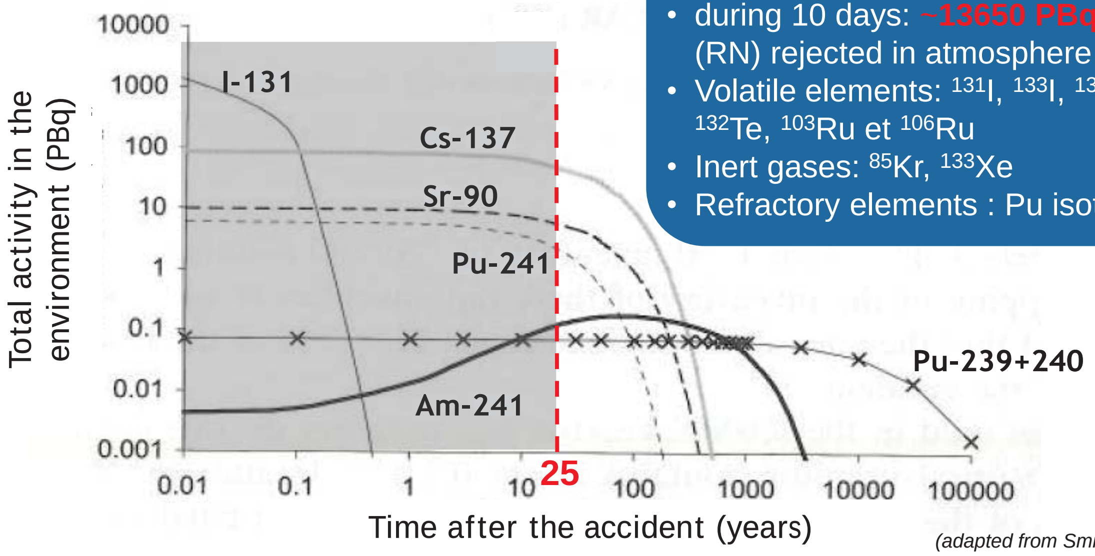
French Institute for Radioprotection and Nuclear Safety (IRSN)

Caesium Workshop -Session 3

- Context
- Site characterization
- Modelling and system understanding
- Summary & Perspectives for EPIC
- Transfer to F-TRACE?

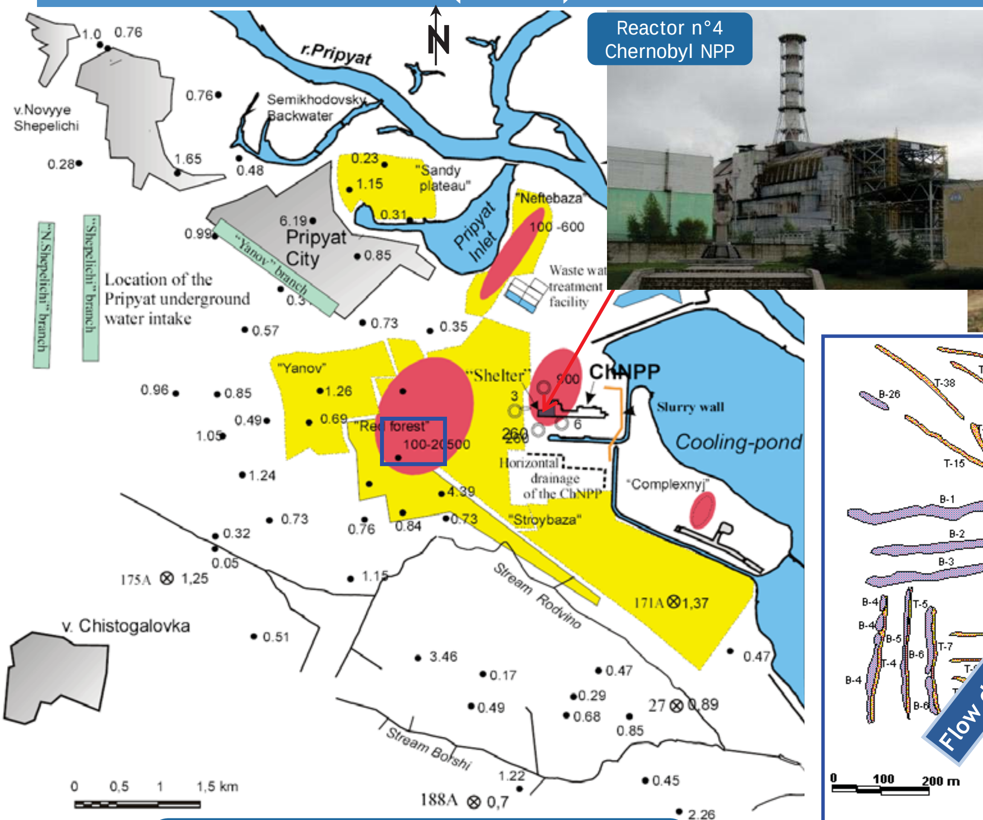
THE CHERNOBYL NPP ACCIDENT

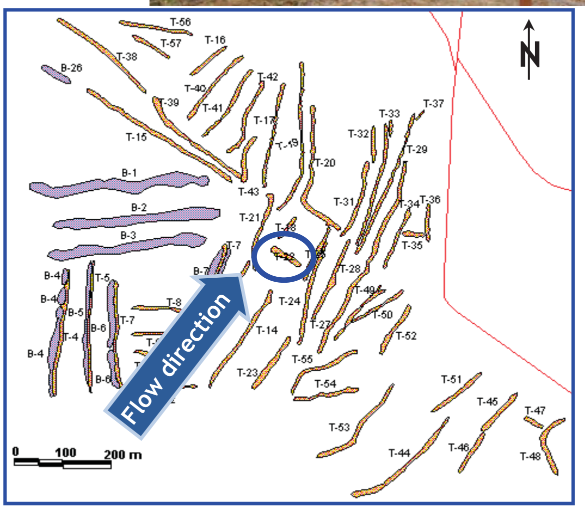
- **04/26/1986: Explosion of Chernobyl NPP's reactor n°4**
- during 10 days: **~13650 PBq** of radionuclides (RN) rejected in atmosphere (till 10 km high)
- Volatile elements: ^{131}I, ^{133}I, ^{137}Cs, ^{90}Sr, ^{129}Te, ^{132}Te, ^{103}Ru et ^{106}Ru
- Inert gases: ^{85}Kr, ^{133}Xe
- Refractory elements : Pu isotopes



(adapted from Smith & Beresford, 2005).

THE PILOT SITE (EPIC)

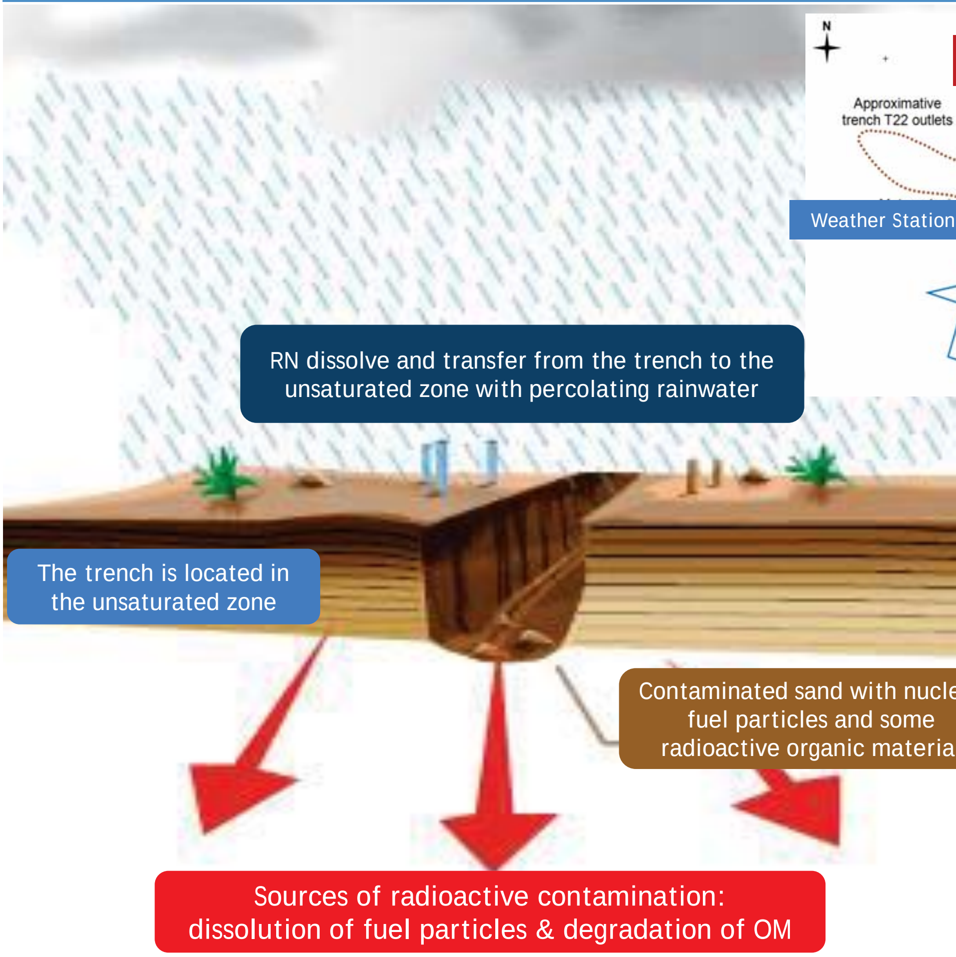


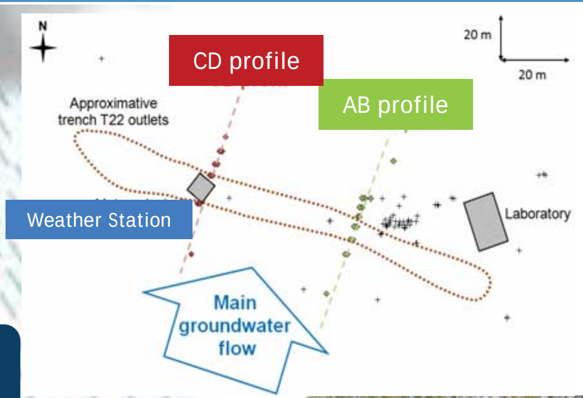


Map indicating the location of different temporary waste deposit zones in Chernobyl exclusion zone and Sr-90 contaminated underground waters *(Antropov et al., 2001)*

Map indicating the location of trenches and mounds in the « Red Forest » zone. T=trench, B=mound *(Antropov et al., 2001)*

THE PILOT SITE (EPIC)





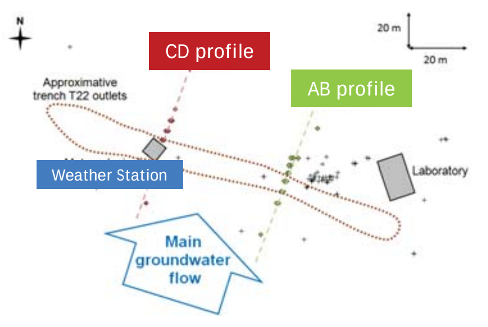
THE PILOT SITE (EPIC)

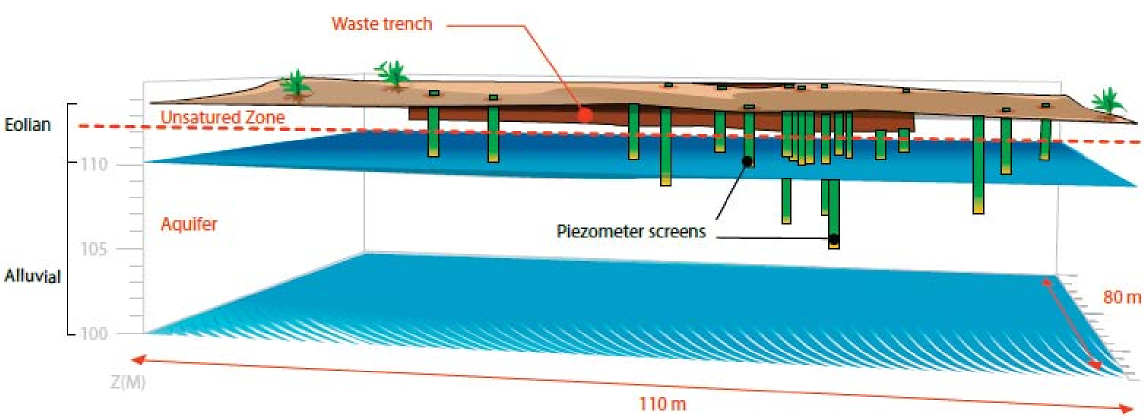
Site instrumentation (UZ):

- Suction pressure,
- moisture content and temperature captors
- Soil water samplers

Site instrumentation (SZ & background):

- Piezometers
- Weather station
- Laboratory





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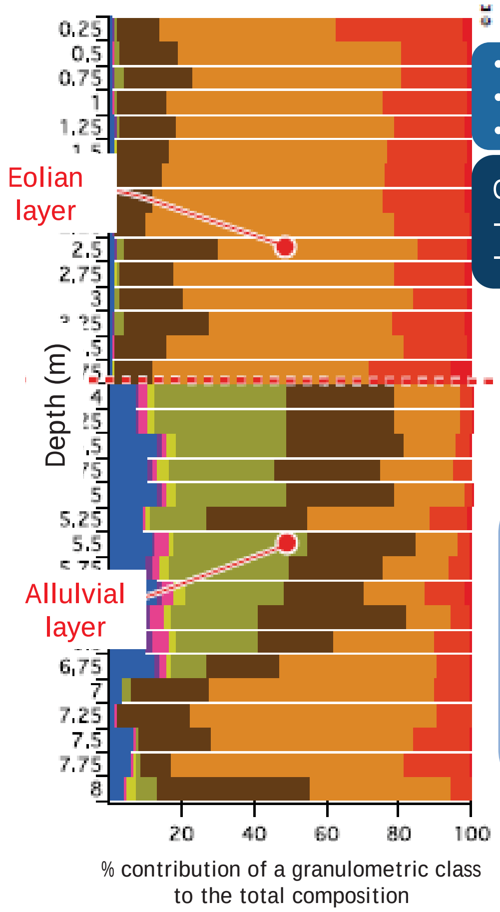
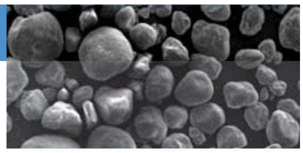


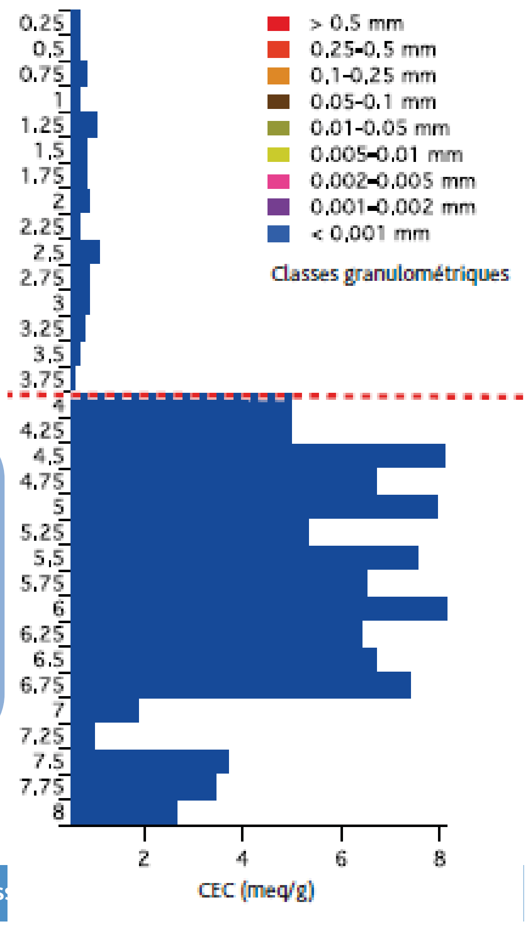
- Quartz (98-99%)
- Heavy minerals (1-2%)
- Organic carbon (<0.3%)

Clay fraction (<1%):
-illite, smectite, chlorite
-feldspaths

quartz (90-94%)
feldspath K-Na (5-9%)
heavy minerals (<0.5%)
hydromica,
montmorillonite,
cristalline calcite,
quartz, amorphic iron oxides (<0.5%)

MEB observations of Pripyat Zaton sands, x 35 (Ardois et al., 2002)





DERIVATION OF Cs AND Sr DATABASE

Site instrumentation (UZ):
- Suction pressure, moisture content and temperature captors
- Soil water samplers

Site instrumentation (SZ & background):
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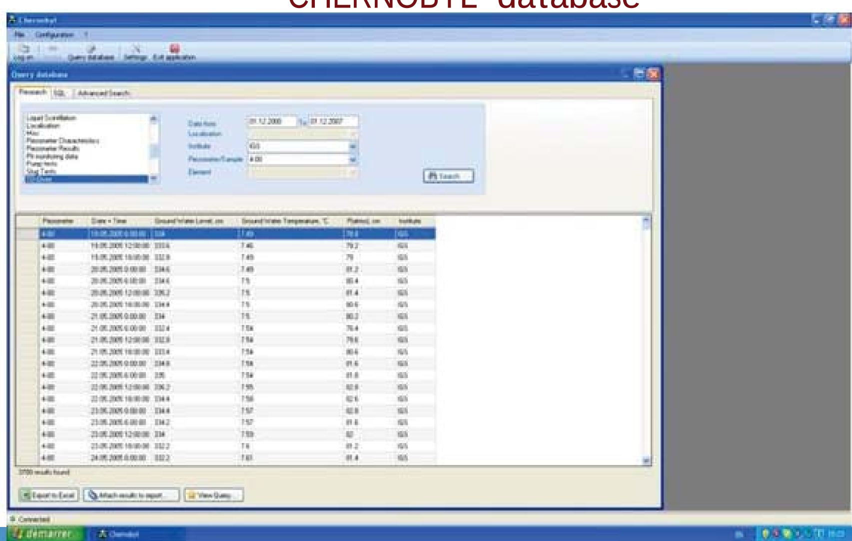
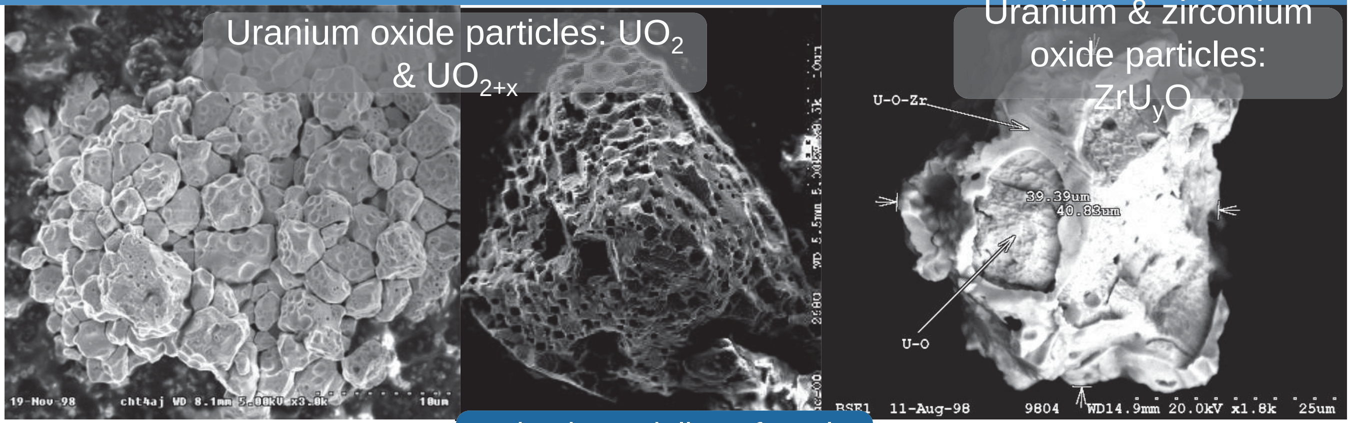
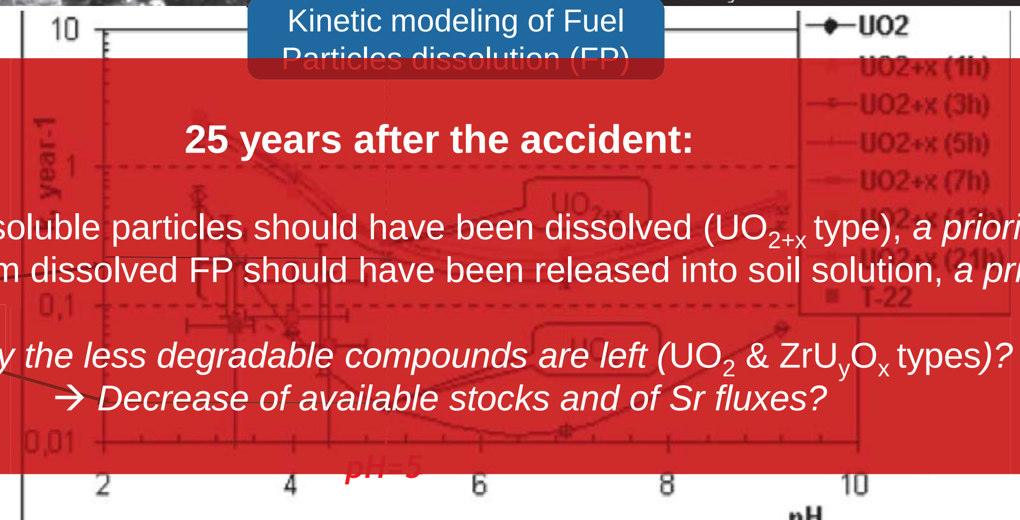
Monitoring data

CHERNOBYL database



THE SOURCE TERM





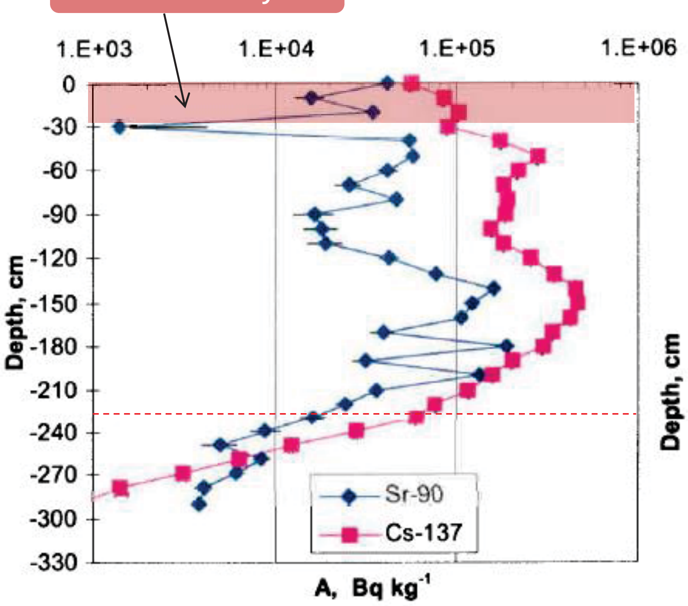
(Van Meir et al., 2009).

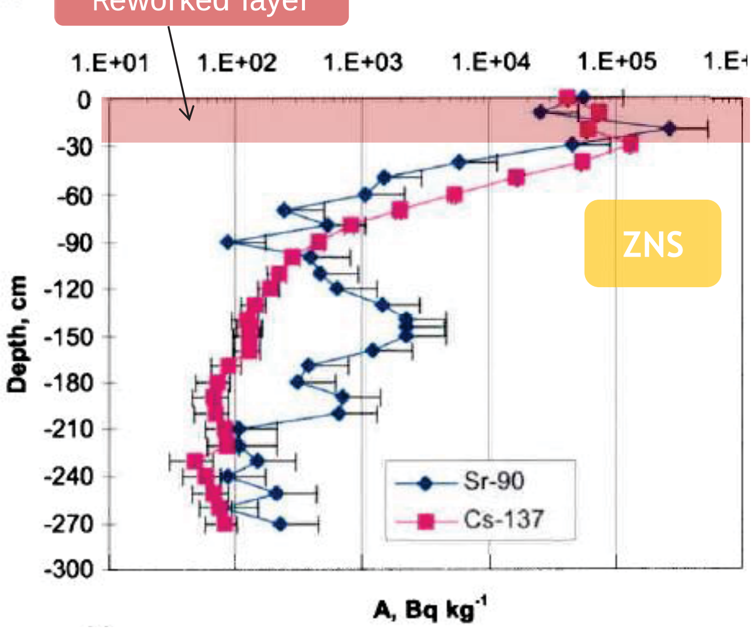
25 years after the accident:

* All the soluble particles should have been dissolved (UO_{2+x} type), *a priori*
* All RN from dissolved FP should have been released into soil solution, *a priori*

→ *Mainly the less degradable compounds are left* (UO_2 & ZrU_yO_x types)*?*
→ *Decrease of available stocks and of Sr fluxes?*

THE SOURCE TERM: Cs and Sr distribution

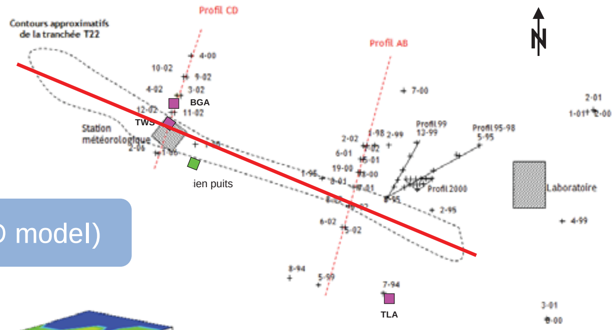




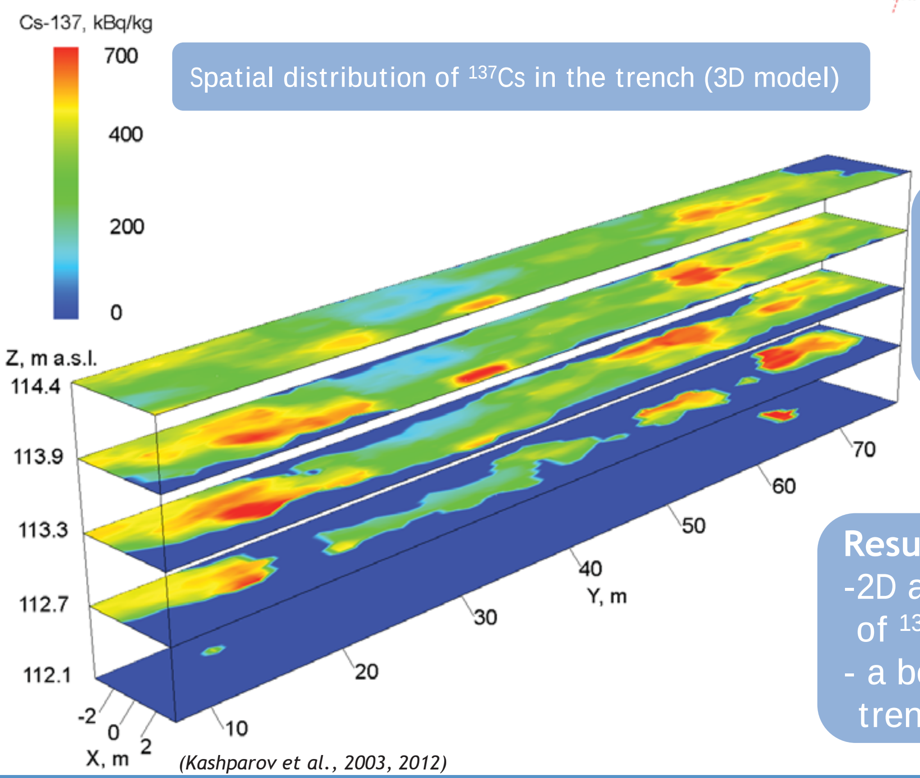
Specific activity in ^{137}Cs & ^{90}Sr in the trench 22 is ~10^5 to 10^6 Bq/kg
(Guillou et al., 2000 ; Kashparov et al., 2004 ; Dewière et al., 2004)

THE SOURCE TERM: Cs and Sr distribution

Layout of the trench–3D model



Spatial distribution of ^{137}Cs in the trench (3D model)

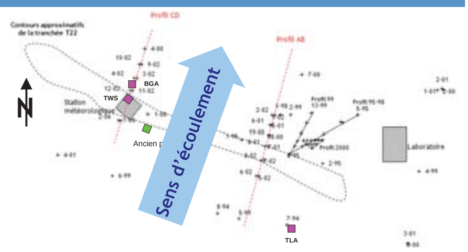


(Kashparov *et al.*, 2003, 2012)

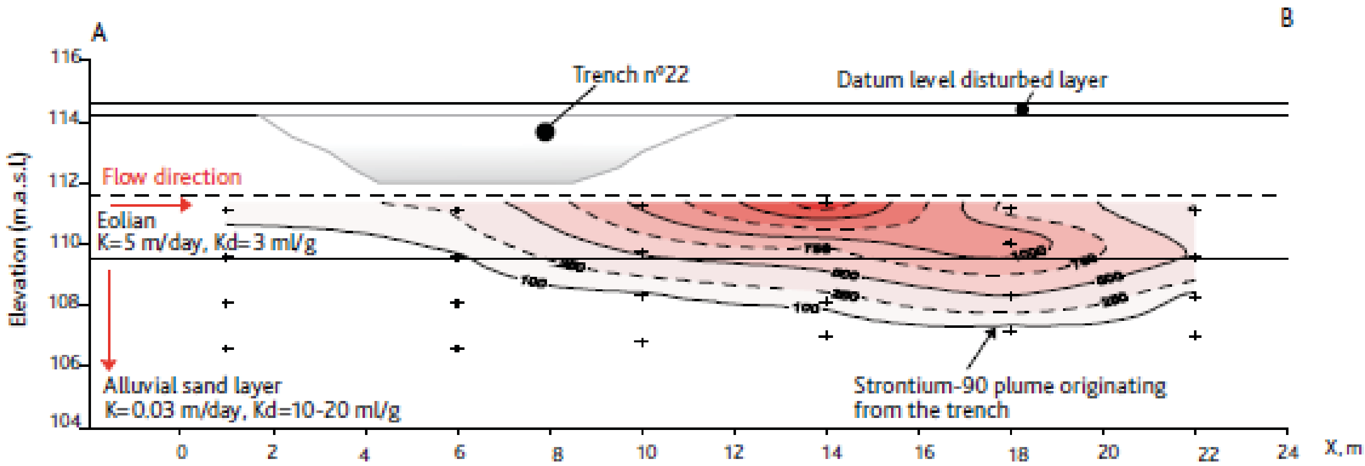
- Integration of ^{137}Cs activity ponctual data
- Interpolation of these data by kriging method

Results :
-2D and 3D spatial distribution of ^{137}Cs in the trench
- a better definition of the trench layout

Parameter	Value
Groundwater level elevation (multi-year mean value), m a.s.l.	111.5
Flow direction in eolian layer	North (±15°)
Horizontal hydraulic head gradient in eolian layer	0.0015
Vertical hydraulic head gradient in alluvial layer	0.03
Infiltration recharge rate, mm/y	300
Hydraulic conductivity of eolian layer (isotropic), m/d	3.6
Hydraulic conductivity of alluvial layer (anisotropic),	
Kx, m/d	0.5
Kz, m/d	0.0275



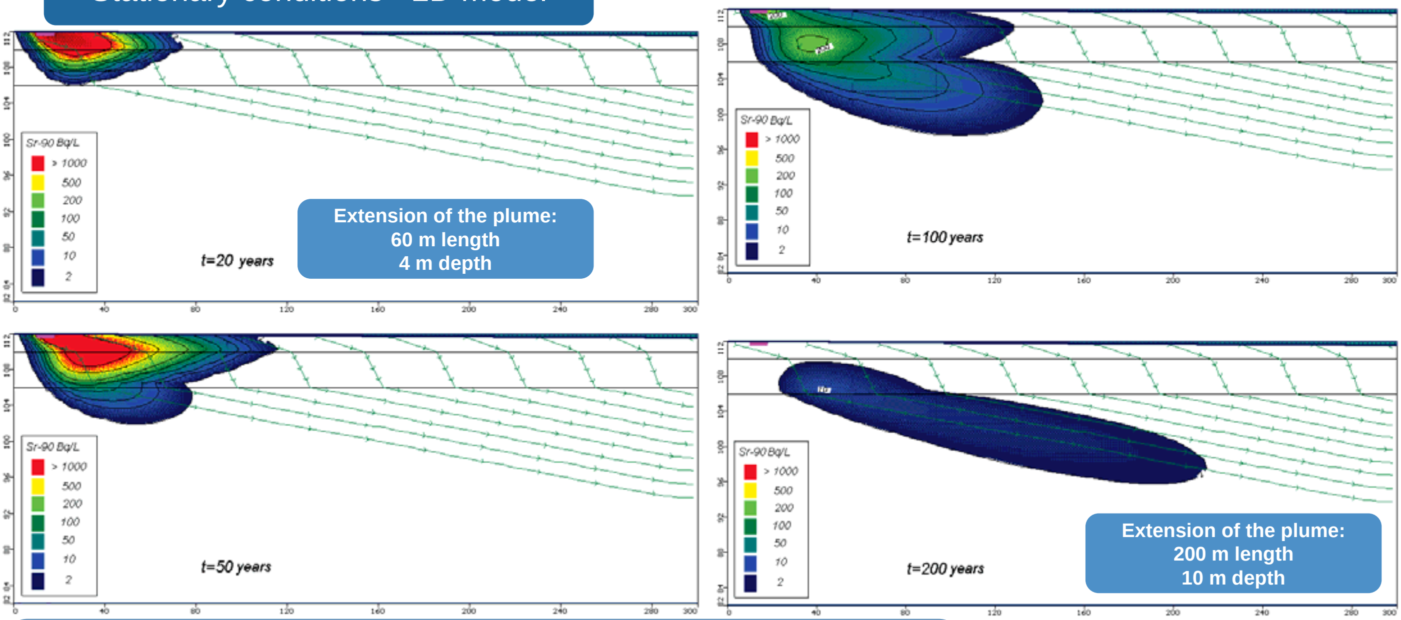
(Bugai *et al.*, 2012)



(Van Meir *et al.*, 2009)

NUMERICAL SIMULATION OF Sr90-PLUME EVOLUTION

Stationary conditions - 2D model



(Bugai et al., 2011)

Global model

- Kinetic model of fuel particles dissolution (dynamic model)
- Stationary model for the saturated zone (plume)

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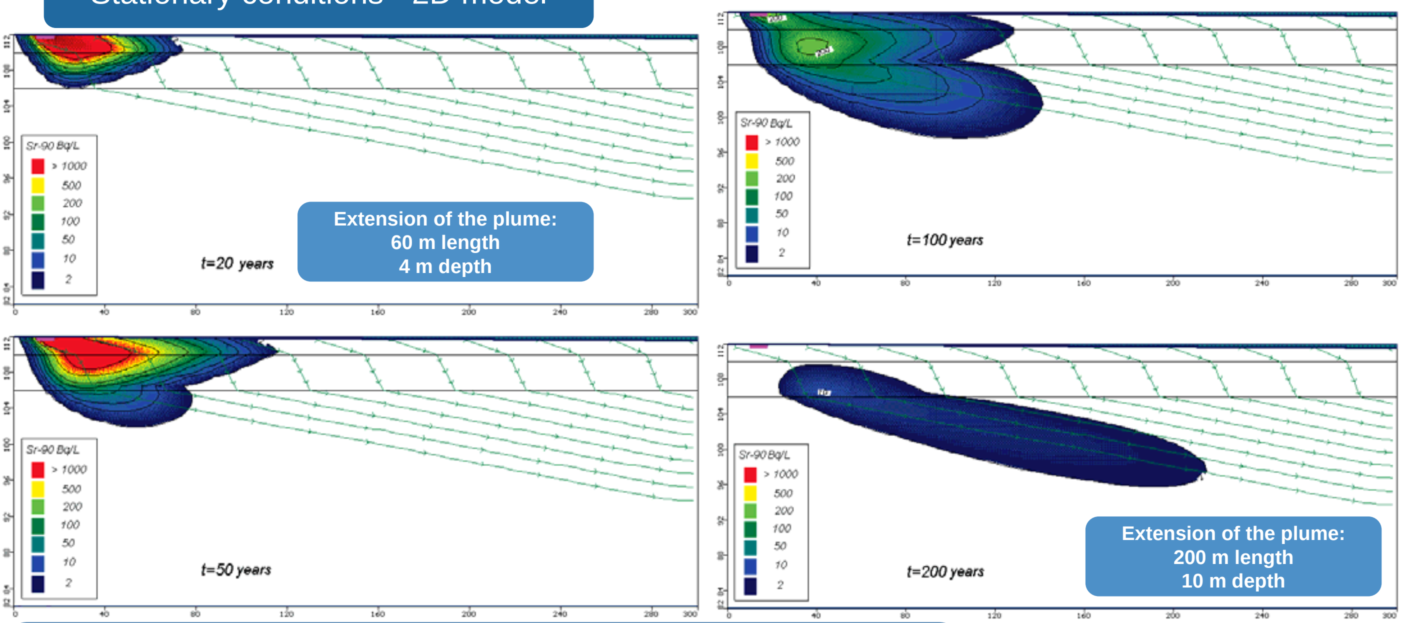


(Bugai et al., 2011)

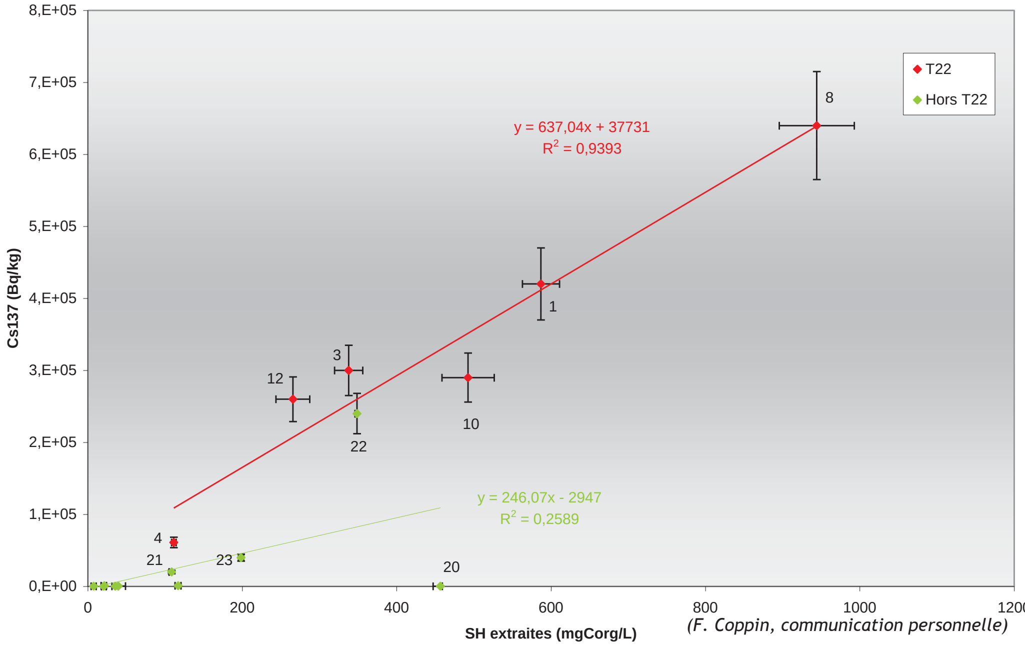
Global model

- Kinetic model of fuel particles dissolution (dynamic model)
- Stationary model for the saturated zone (plume)

Transport model in the groundwater:
- No chemical-physical variations
- No variations of groundwater flow
- No seasonality
- No OM as part of the source term

THE SOURCE TERM: Cs and Sr distribution

What about the Cs plume at T22?



(F. Coppin, communication personnelle)

- migration velocity ^{137}Cs : ~0.16 cm/y (Szenknect, 2003; Guillou et al., 2000)
- Low content in ^{137}Cs measured in the aquifer downgradient T22 : <0,1 Bq/L
- 2011-12: above detection limit concentrations in the groundwater: >3 Bq/L
- ^{137}Cs activities in T22 soil solution: 45-52 Bq/L >> 0,03 Bq/L in Pripyat river
- **Strong affinity of Cs with OM** (cf. fig)
- Cs is used by the plant (competing with nutrients like; Ca, K, etc.)

^{137}Cs is mainly located in the trench and use by the vegetation

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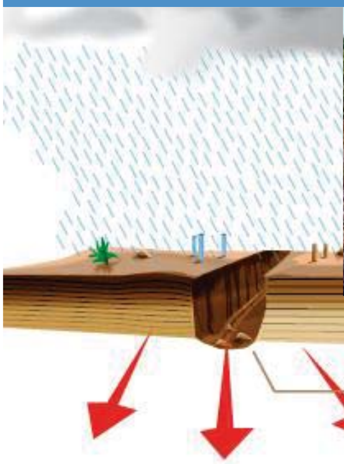
THE SECONDARY SOURCE TERM



- tree-trunks
- forest contaminated soils + liter (OM 3-6%)
- vegetable debris: needles, branches, …
- other sup. plants: herbs, shrubs, …

Kinetic of organic decaying:
- fast decaying of easily degradable compounds (**$T_{1/2}$= 3-4 years**) for fine liter (Pausas, 1997)
- slow decaying of less degradable compounds (**$T_{1/2}$= 7-42 years**) for coarser materials (Currie *et al.*, 2002

25 years after the accident:

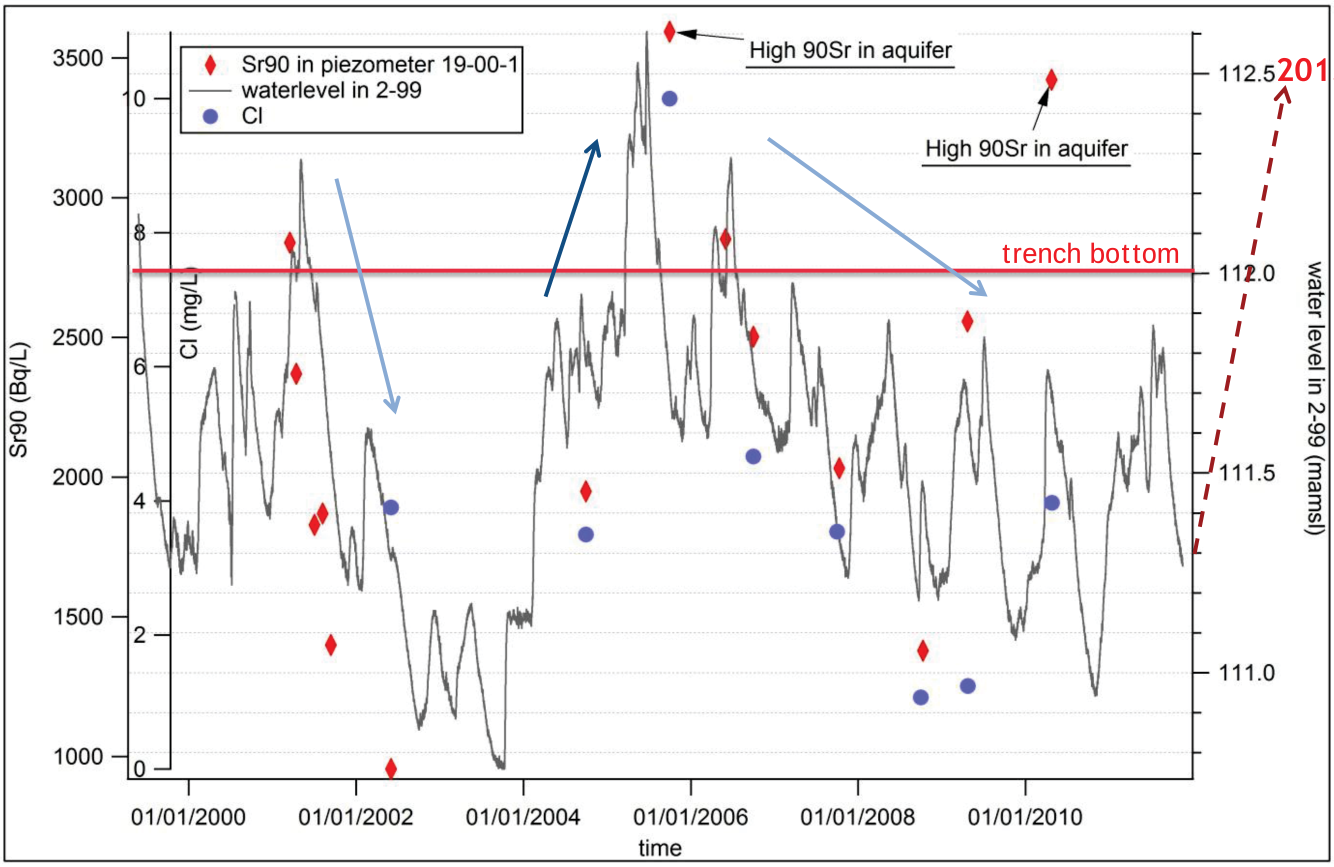
Most of the organic compounds easily degradable have been transformed
→ *Mainly the less degradable compounds are left (ex.: trunks)?*

BIOTIC MIGRATION OF RADIONUCLIDES

Vegetation on the top of the trench:
47 pines, 14 birch trees, 49 bushes
(at approx. 400 m^2 area)

RN activity in trees (2001)
^{90}Sr - 0.6 – 6.7 MBq/kg
^{137}Cs - 0.01 – 2.1 MBq/kg
(Kashparov *et al*., 2002)

Chronicle of 90Sr fluxes evolution in the aquifer vs. water-table fluctuations



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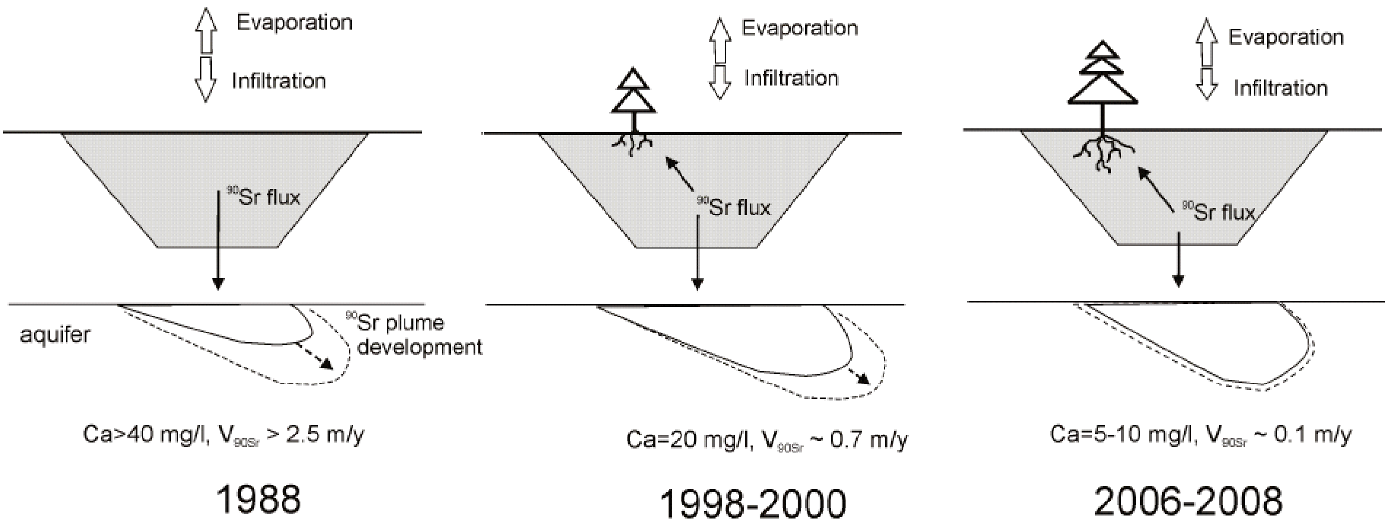
Evolution of ^{90}Sr fluxes coming out of the trench and to the aquifer



(Bugai et al., 2012)

Explicative processes:
- ↓ stock of soluble fuel particles
- ↓ stock of easily degradable organic matter
- modification of the physical-chemical environment
- ↑ of transfers (cations & RN) to the superior plants with ↑ of their biomass

Role of vegetation: - direct RN uptake; - nutrient element (Ca,K...) uptake, which influences GW geochemistry and hence - RN mobility

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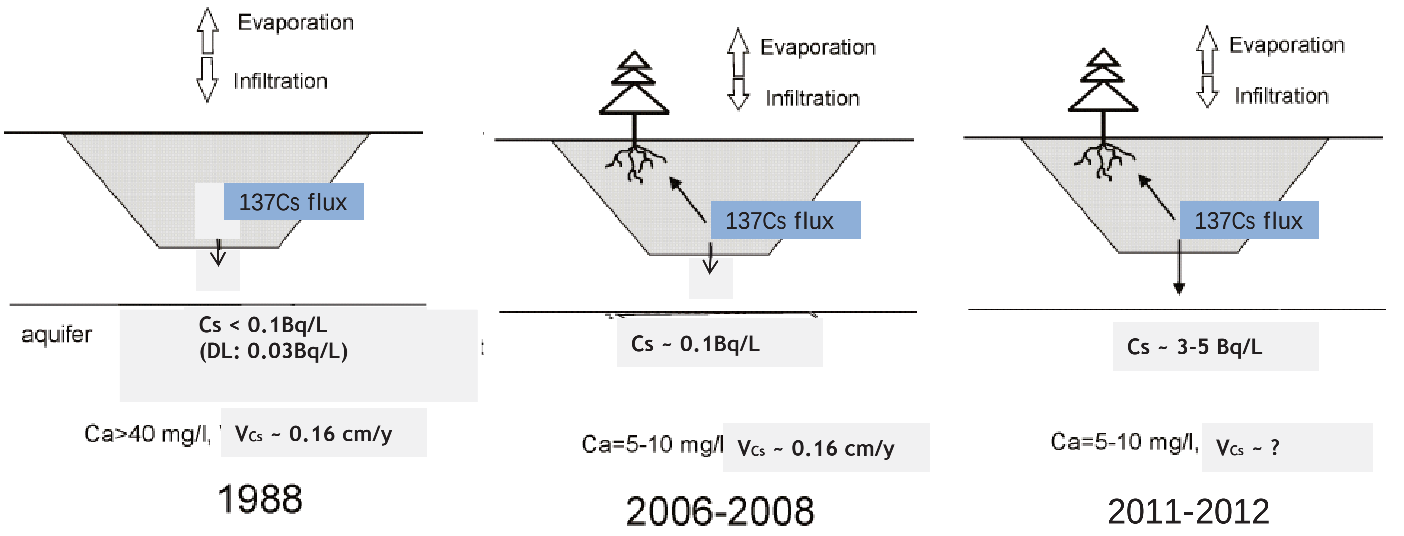

(based Bugai et al., 2012)

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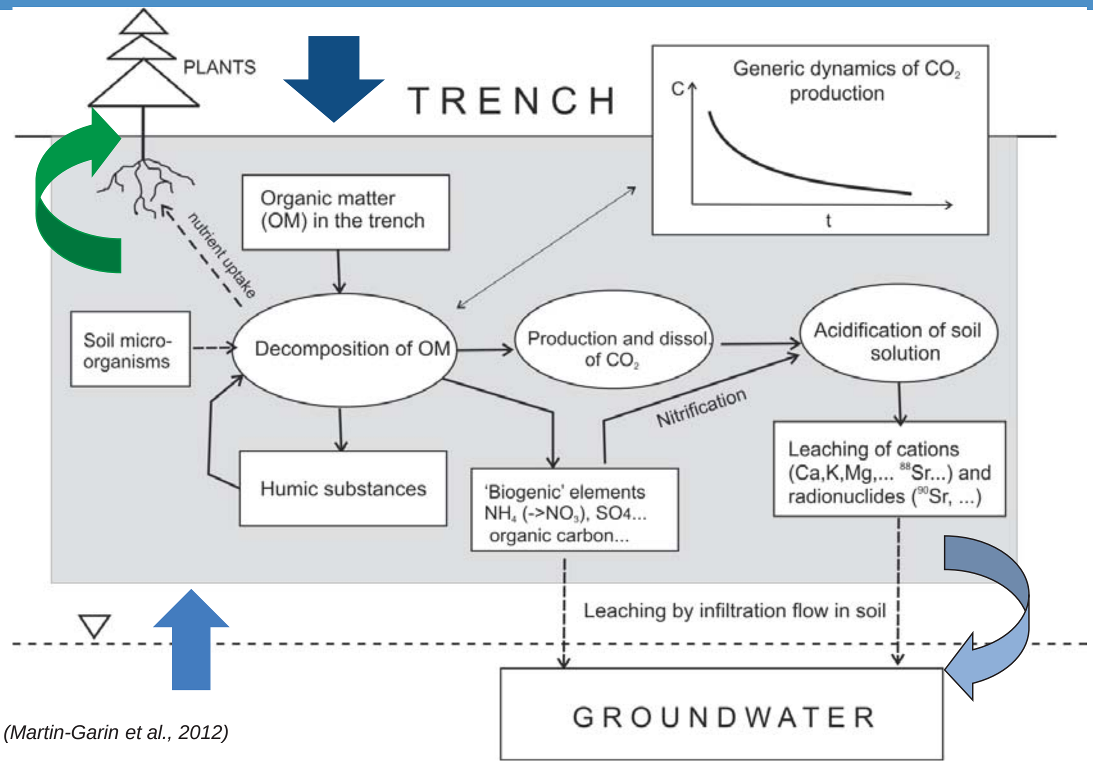

(Martin-Garin et al., 2012)

^{90}Sr fluxes from the trench may be controlled by:
- Dynamic of bio-physical-chemical modifications linked with stocks of buried OM and with their kinetic of degradation
- Transfers towards plants which lead to an increase with time of plant biomass
- Seasonality

SUMMARY & PERSPECTIVES

WHAT DID WE LEARN?

• Complex relationships between hydrogeological, geochemical and biological processes observed in the "real world" of a contaminated site such as the Red Forest (EPIC site) require the achievement of **interdisciplinary researches**

• The realization of relevant predictive calculation requires the use of a **GLOBAL MODEL** coupling transfers and RN migration in every compartment of interests (atmosphere, UZ, SZ, vegetable cover), and to consider the influence of the main geochemical and biological factors

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WHAT IS NEXT?

• Global numerical modeling coupling biogeochemistry-transport for Sr, Cs

• Evolution of the buried source term 25 years after: reevaluation of inventories for FP and OM. What is the main process that rule the dynamic of their evolution?; What is the role of microorganisms and their influence?; Water-table fluctuation influence (flood of the bottom of the trench) vs. precipitation influence on the dynamic of RN release?

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• Modeling of the most complex cases (Pu, U, etc.)? These last will need the use of other coupling (e.g. colloids role; interaction with microorganisms; factors controlling speciation variations, …)

• Dynamic of the RN uptake by plants (bioavailability, translocation, bioaccumulation)

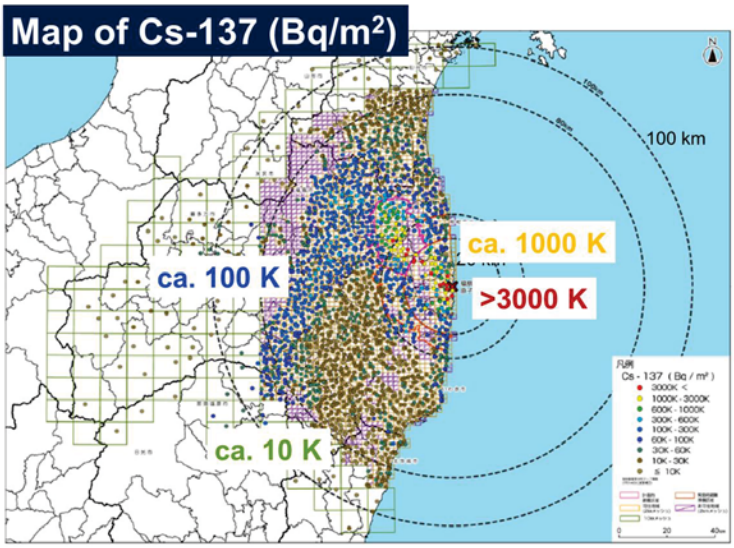
• The importance and the impact of a new source term, more diffuse on soil surface (contaminated liters); modification of RN speciation and their reactivity?

• Scale changing: from the pilot site to the exclusion zone (water-basin scale)

Post-accidental situation: What can we learn? Contribution to F-TRACE

How to:

- Manage soil decontamination in a post-accidental situation?
- Remove radionuclides from soils?
- Manage remediation wastes?
- Organize the return of population and under what exposure conditions?
- Manage land reuse when population returns?
- Manage/organize monitoring after the return of population?



Which tools can we use?

Laboratory Studies & Field Observations → Integrated Modeling

How geochemistry and hydrology can help?

How geostatistics can help?

How biochemistry can help?

How modeling can help?

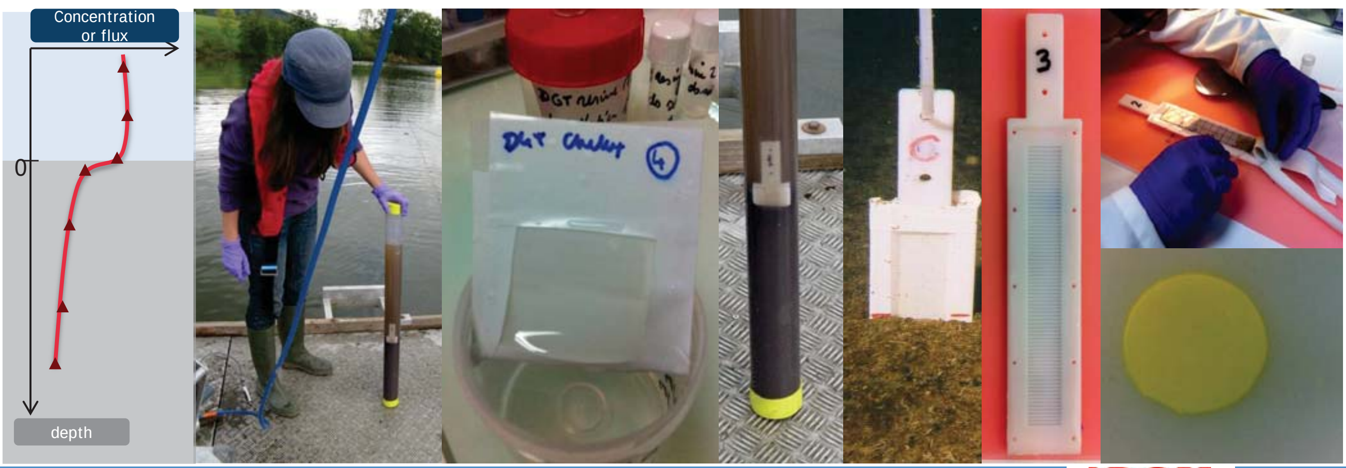
→
- Long-term predictions of RN transfers
- Impact assessment
- Upscaling
- *Remediation solutions*
- *Population return and use of land*

DGT can be used for many different purposes, including:
- In situ measurements
- Monitoring (time averaged concentrations)
- Speciation (labile inorganic and/or organic species)
- Bioavailability (effective concentration)
- Trace metals, phosphate, sulphide and RN
- Fluxes & conc. in sediments/soils & fresw./seaw.
- Kinetic and thermodynamic constants
- High spatial resolution measurements (sub-mm)
- 2D concentration images

DET can be used for in situ measurements of solute concentrations at high spatial resolution

DET: Diffusive Equilibration in Thin films
DGT: Diffusive Gradient in Thin-films

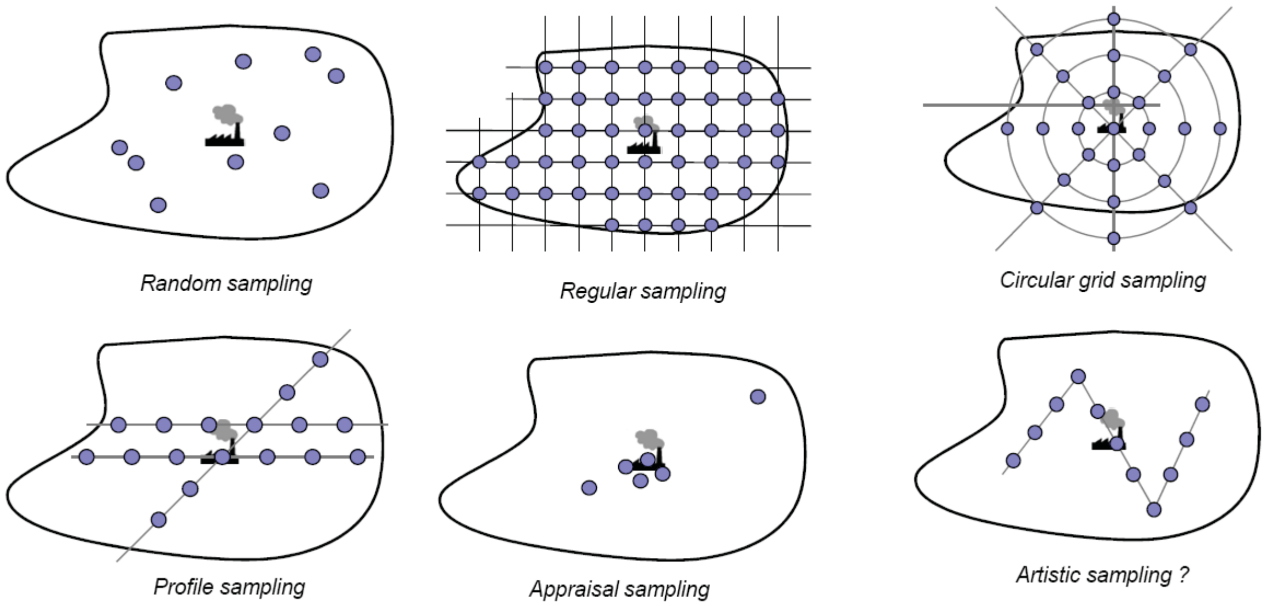
DGT & DET were invented by ***Bill Davison and Hao Zhang***



Which tools can we use?

How geostatistics can help?

- How to improve RN mass estimate from an optimized sampling plan?
- How to assess uncertainty on contaminated volumes of soils?



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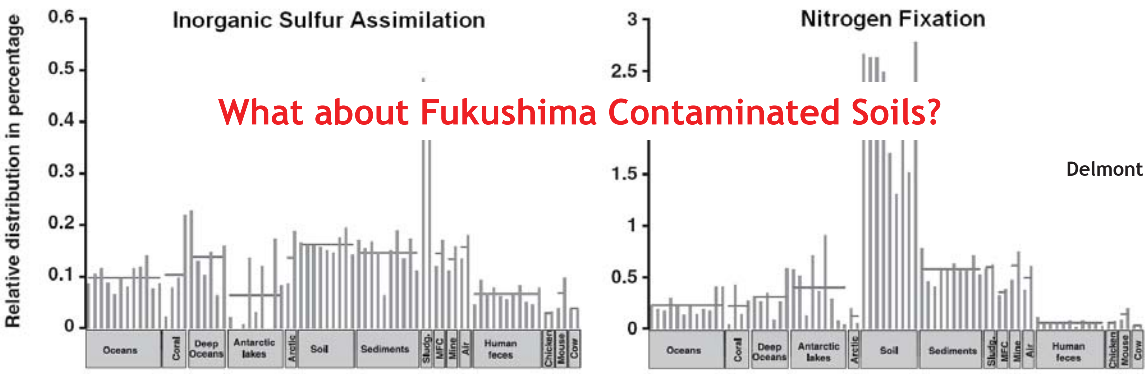
Which tools can we use?

How biochemistry can help?

Which biochemical functions are lost in a contaminated soil?

Metagenomic analysis (i.e. analysis of the DNA of all the soil microorganisms) could help to:
- assess the contamination impact
- better identify biochemical reactions occurring in the contaminated soils and whether they impact RN fate



What about Fukushima Contaminated Soils?

Delmont *et al.* 2011

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THANK YOU
VERY MUCH
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