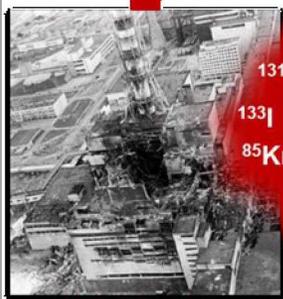
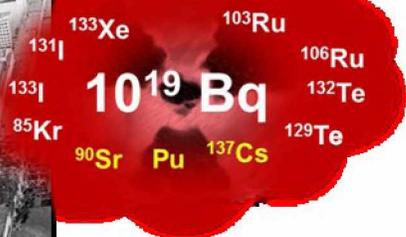


Behaviour of radionuclides in continental aquifers following a severe accident: theoretical, experimental and modelling approaches



26 April 1986 :
Explosion of reactor
n°4 of the Chernobyl
nuclear power plant



IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

H.L. Nguyen¹, A. Martin-Garin², J.M. Matray¹, C.
Courbet¹, C. Meurville¹, M. Bourgeois¹

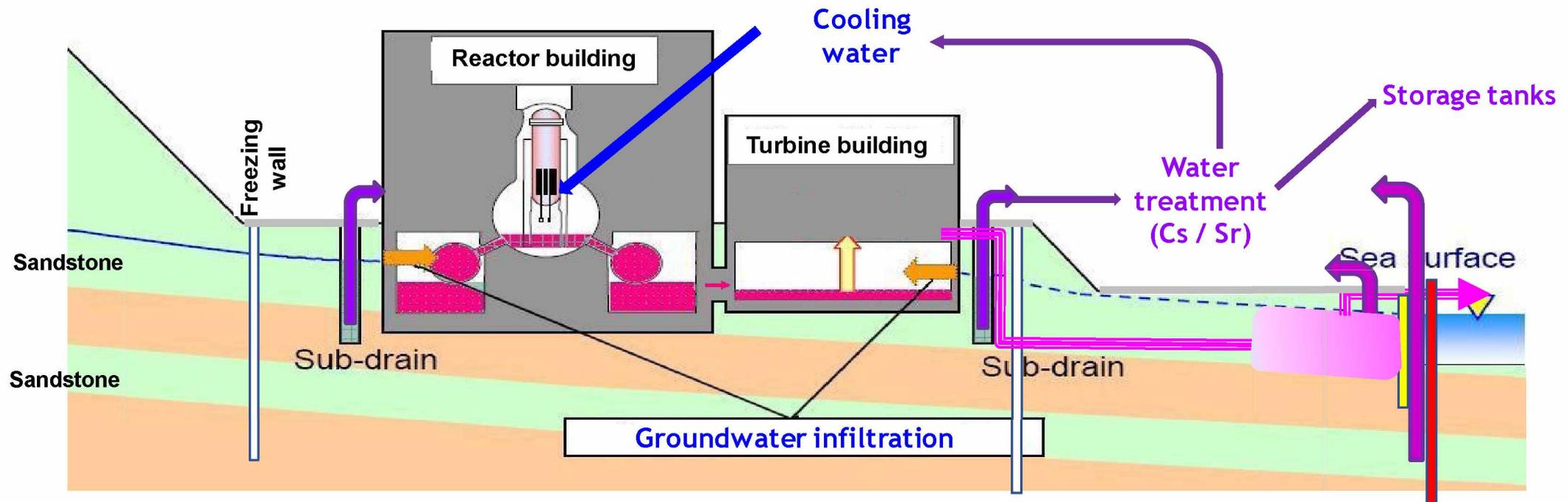
1, IRSN, Fontenay aux Roses, France

2, IRSN Cadarache, France

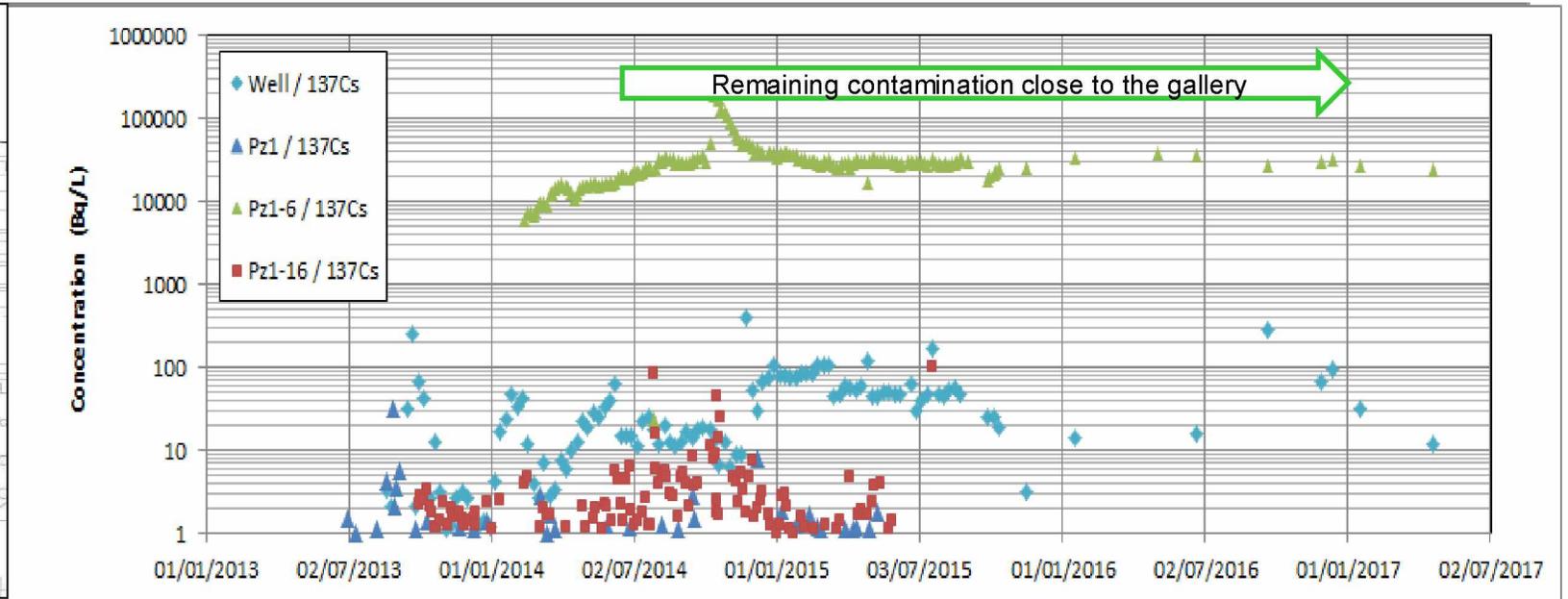
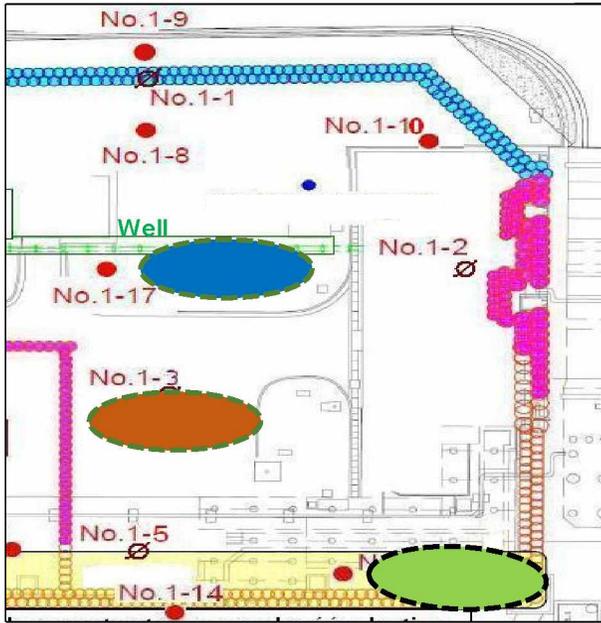


Introduction → post-accidental situation → Fukushima Dai-Ichi

- water injection for reactor cooling, groundwater infiltrations in the buildings
- water accumulation in buildings = dissemination to the ocean through technical galleries
- Contaminated water pumping: maintain a low level in the buildings to avoid *new* releases
- On site works : to stop releases of contaminated water to the ocean, to reduce infiltrations (and water storage), to control possible leakages from buildings



Highly contaminated water releases: tritium, caesium, strontium,...



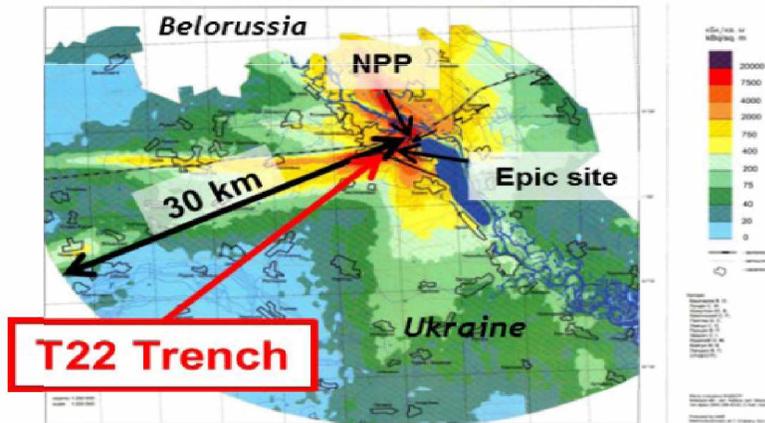
- Water from galleries: releases to the sea and soils (2011/04)
- GW pumping (2013/08): slow decontamination and spreading of contamination from galleries to pumping wells

Main radionuclides	Evolution (2013-2017)	Associated nuclides
^3H	$5 \cdot 10^5$ to $5 \cdot 10^4$ Bq/L	^{106}Ru
^{90}Sr (gross beta)	$9 \cdot 10^6$ to $3 \cdot 10^5$ Bq/L	^{125}Sb
^{137}Cs (^{134}Cs)	up to $2 \cdot 10^5$ Bq/L	^{60}Co

Significant GW contaminants: ^{137}Cs , ^{90}Sr , ^{239}Pu , ^{240}Pu and ^{241}Am

^{90}Sr , the most mobile in soils and GW

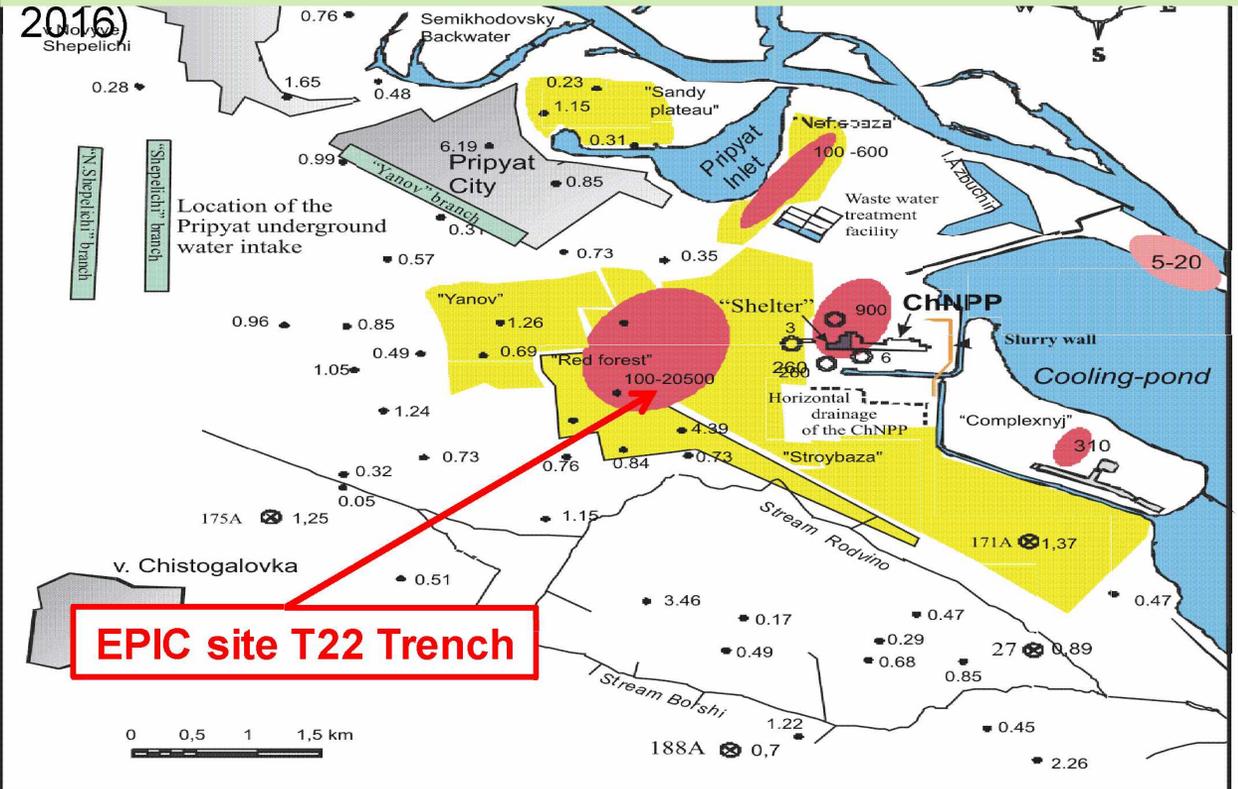
Extent of ^{90}Sr contamination and delineation of the 30-km exclusion zone (UIAR, 1997)



4 main sources of GW contamination:

- Radioactive fallouts,
- Shelter (damaged Unit 4 of ChNPP),
- Radioactive waste dump sites (“Red Forest” ...),
- Cooling pond of ChNPP

^{90}Sr in GW nearby the NPP in 1998 (Bugai et al,



LEGEND

4 •	- IGS's wells and volumetric activity of strontium- 90, Bq/l;	188	- SSPCREMDC's wells and volumetric activity of strontium- 90, Bq/l;
	- areas of increased contamination of groundwater by strontium-90		- SSSPCREMDC wells of for the aquifer in the Eocene deposits (to the right - well , 'to the left volumetric activity of strontium- 90, Bq/l);
	- sites of radioactive waste localization;		

Introduction → post-accidental situation → Radiological risk caused by GW contamination

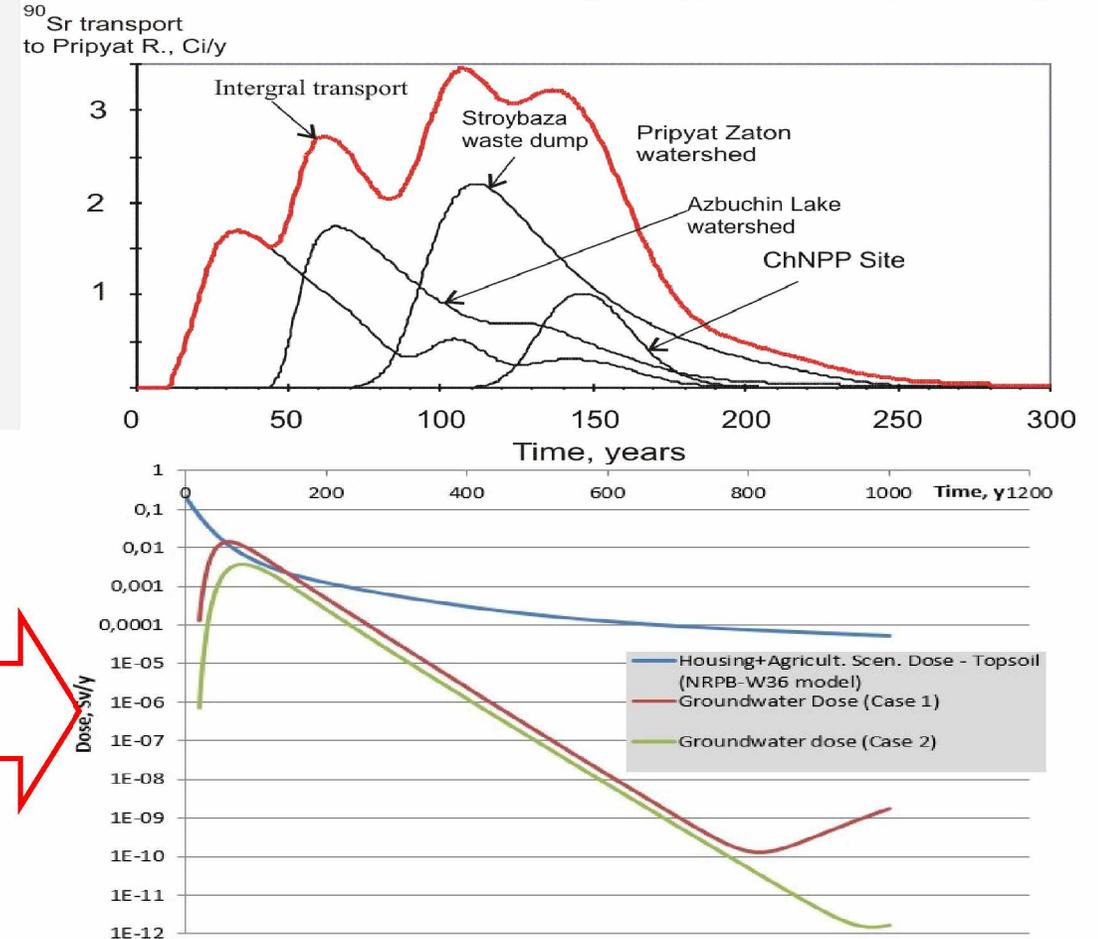
Off-site risks

- Projected max. ^{90}Sr release rate from the ChNPP Site is approx. $\sim 0.1 \text{ TBq/y}$ or 0.02% per year from initial watershed inventory
- Surface water transport of ^{90}Sr ($\sim 10\text{-}20 \text{ TBq/y}$ in 1989-1991) was dominated by wash-off of radionuclides from floodplains and from wetlands
- Water pathway contributed only 6% of dose to the resident of Kiev Region (in 1993); doses induced by water pathway are estimated at $10^{-2}\text{-}10^{-3} \text{ mSv/y}$ [Berkovsky et al., 1993]

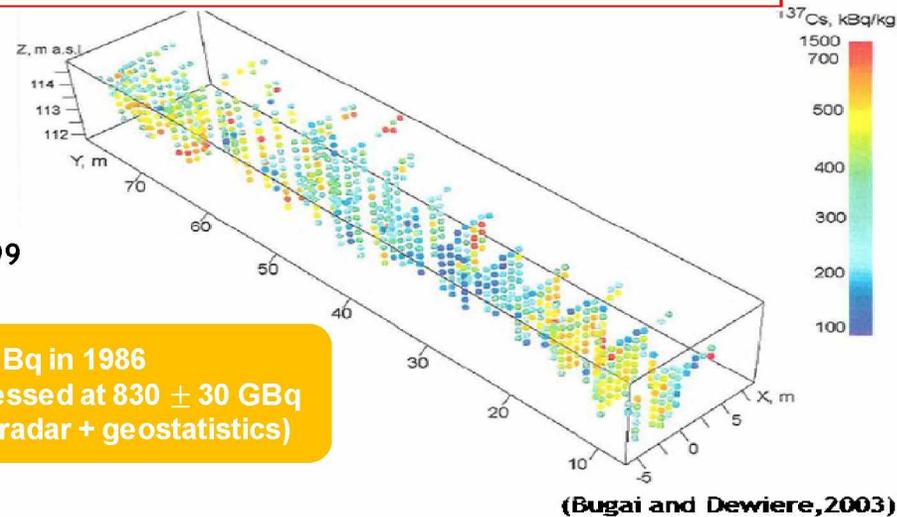
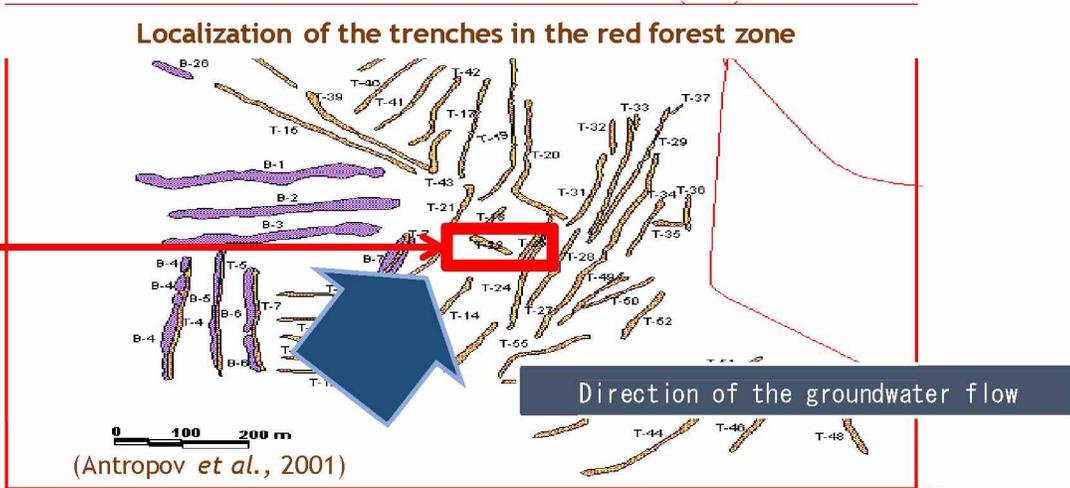
On-site risks

- On-site radiological risks for hypothetical residents in Chernobyl Zone are dominated by risks caused by surface contamination
- Probabilistic analysis has shown low risk of contamination of water wells exploiting Eocene aquifer [Bugai and Smith, 1996]

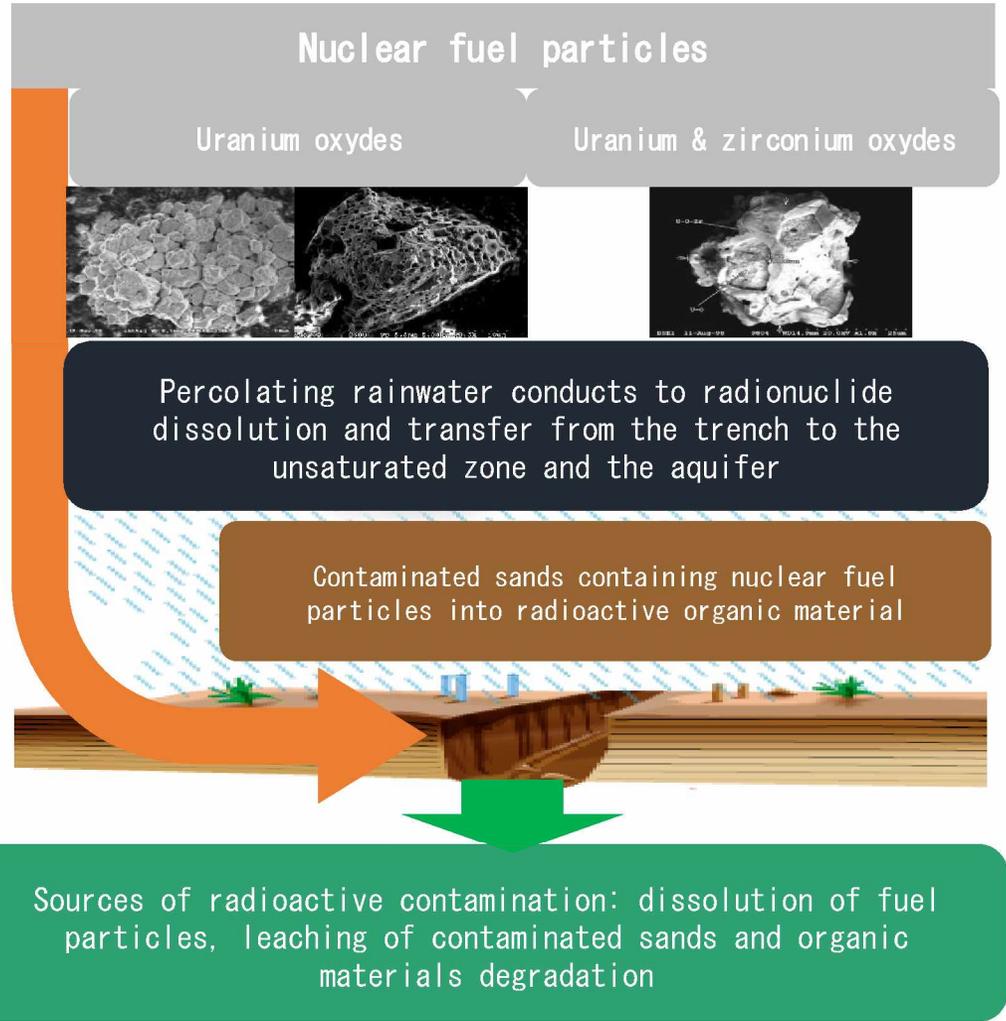
Screening forecast of ^{90}Sr transport from sources at ChNPP Site [Bugai et al., 1998]



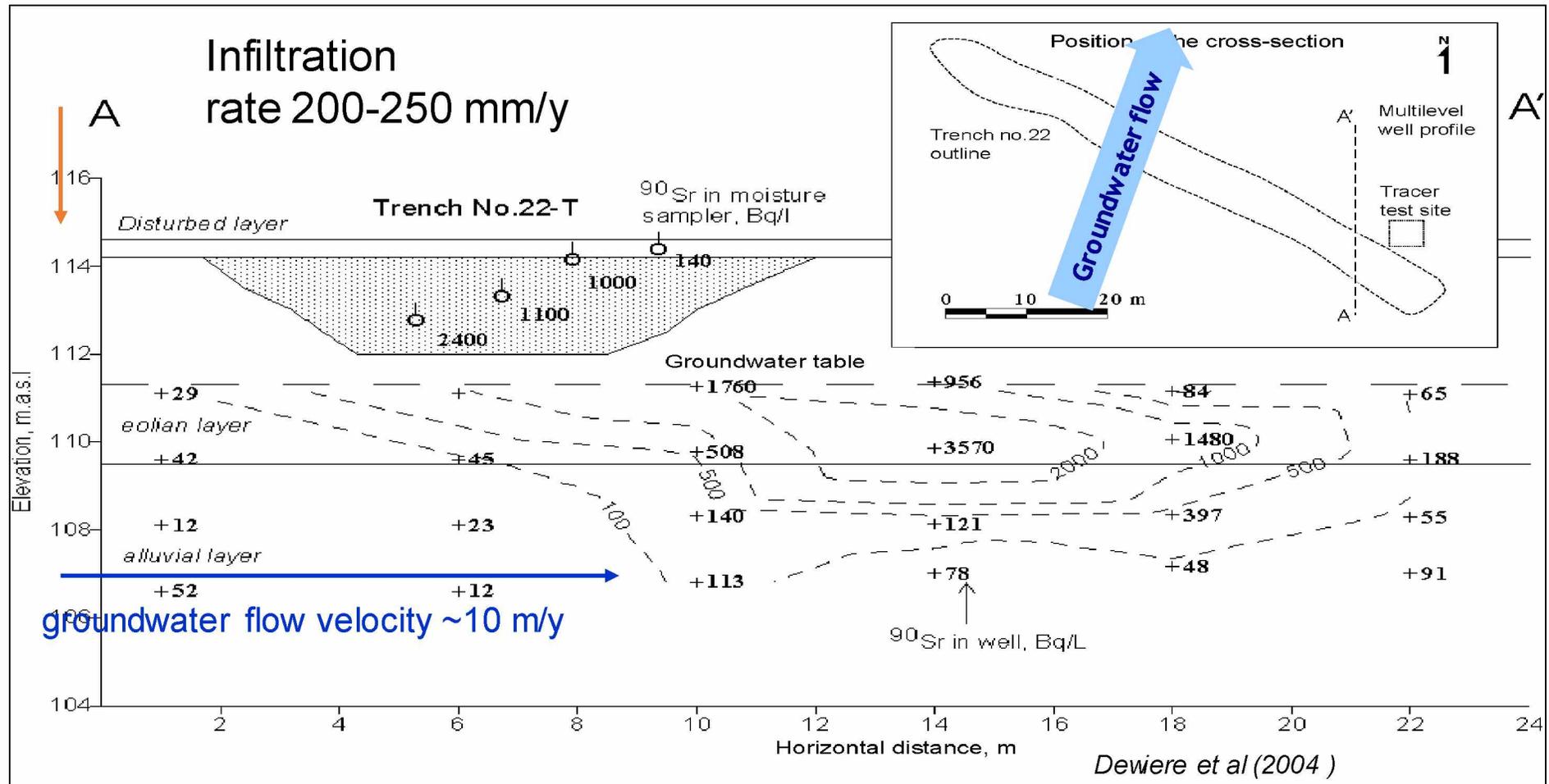
T22 Trench



^{137}Cs : 600 ± 250 GBq in 1986
 (Kashparov et al, 2012) reassessed at 830 ± 30 GBq
 (Nguyen, 2017: all data + georadar + geostatistics)



⁹⁰Sr distribution in the aquifer cross-section at Trench no.22 (2002)

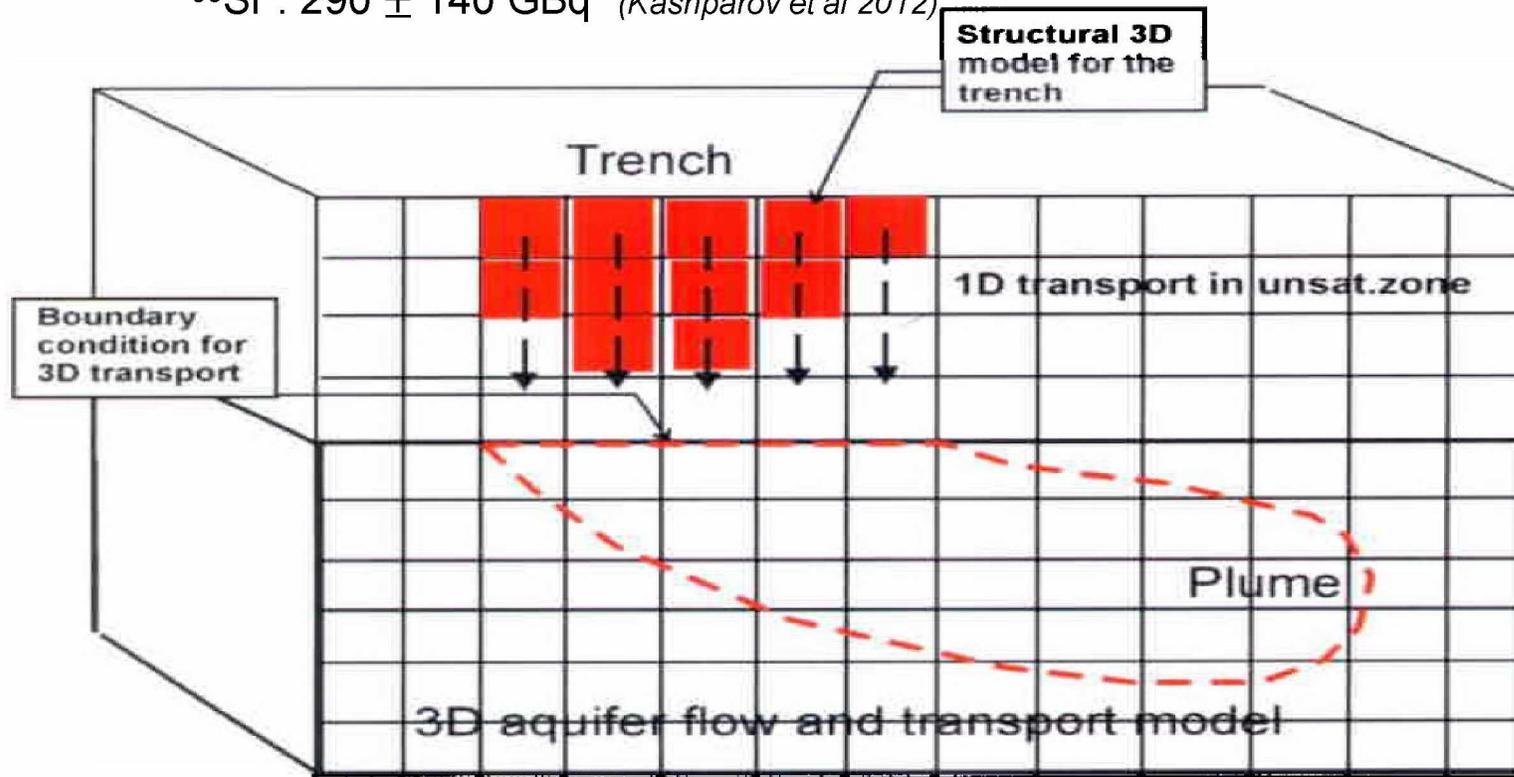


Introduction → post-accidental situation → Tchernobyl EPIC site

1st Modelling approach of the ⁹⁰Sr plume at Trench no.22 - Combined 1D unsaturated/3D saturated flow and transport model at steady state

Initial inventory in T22 in 1986

⁹⁰Sr : 290 ± 140 GBq (Kashparov et al 2012)

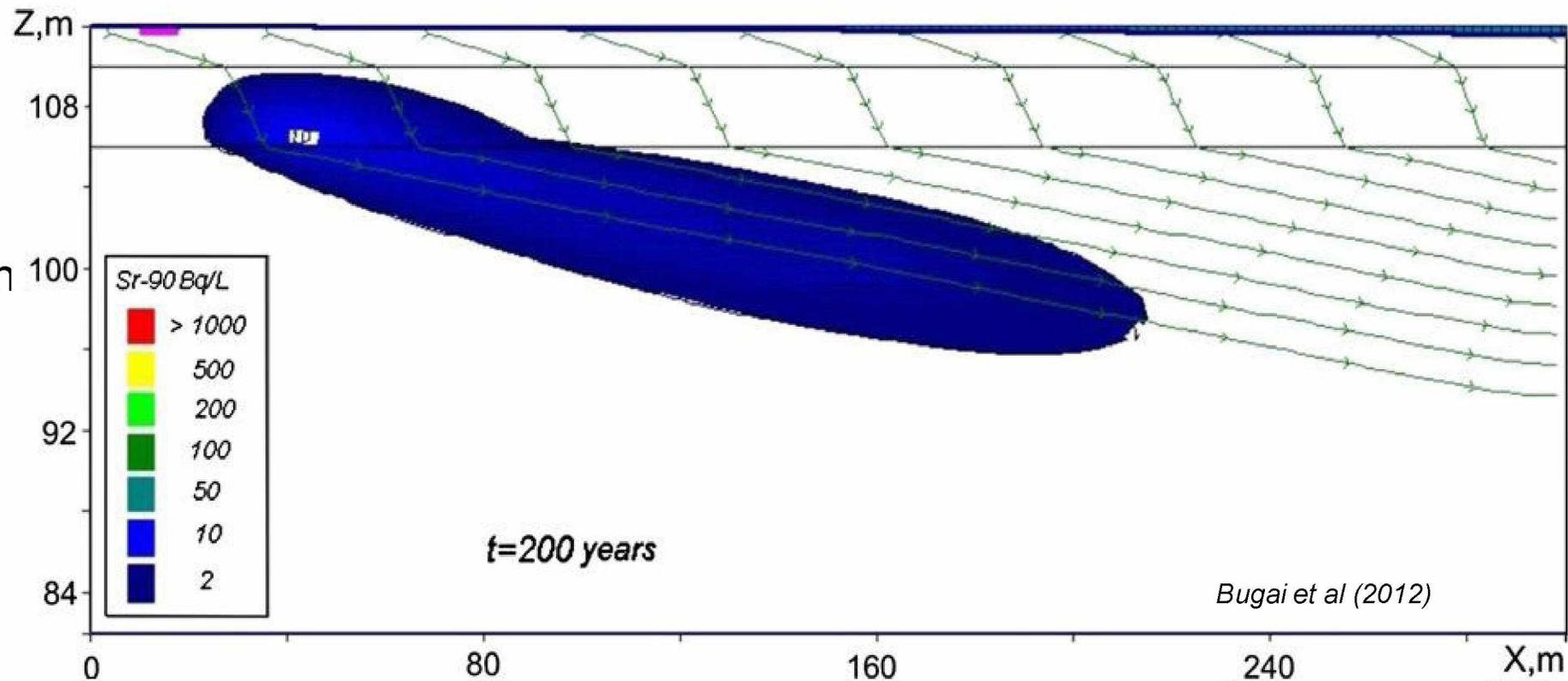


- Hydrogeologic parameters
- Fuel particle dissolution parameters
- Sorption parameters for ⁹⁰Sr (after in situ measurements)

Bugai et al (2012)

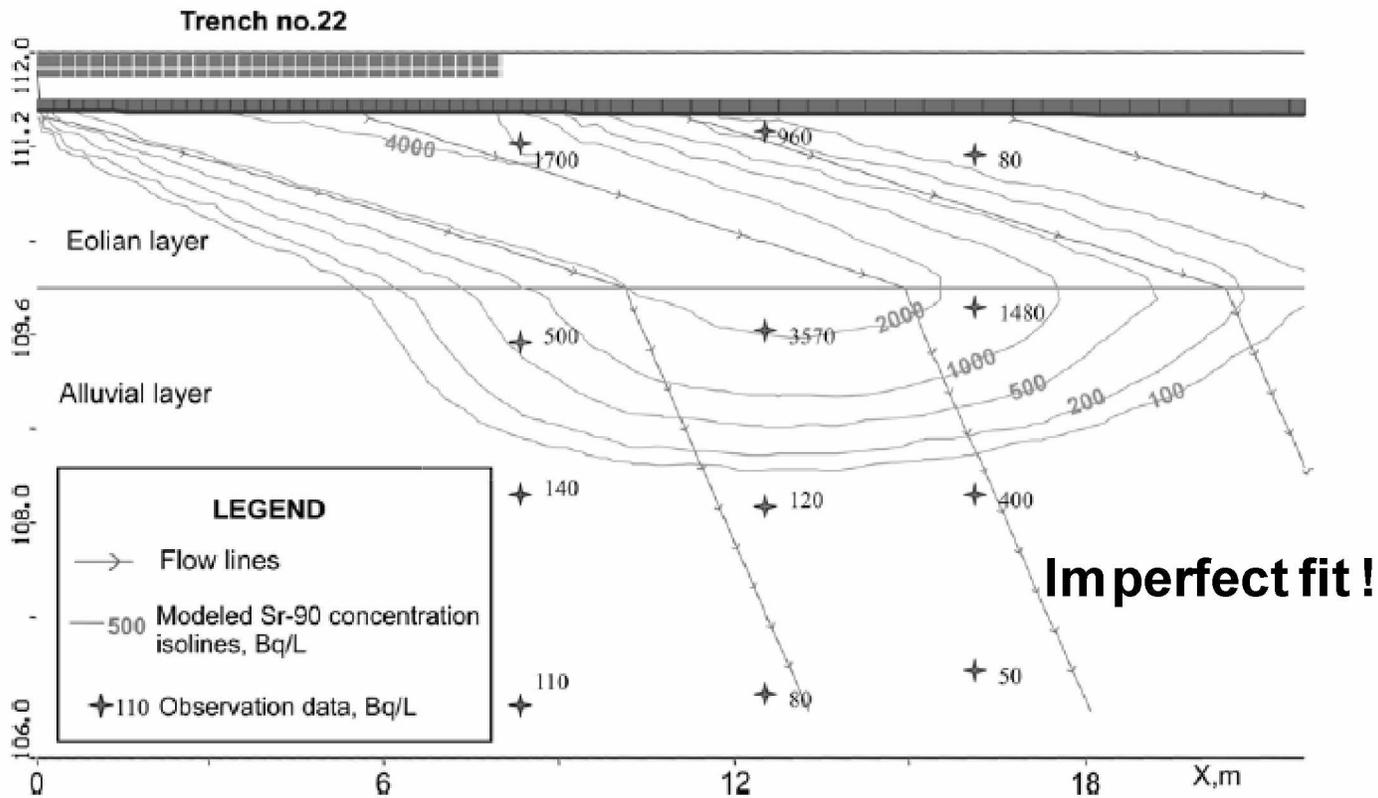
Introduction → post-accidental situation → Chernobyl EPIC site

^{90}Sr distribution in the aquifer at Trench no.22 (2002)



- 20y – only 5-7% of the inventory released to GW
- 200y – the maximum distance travelled by ^{90}Sr from the waste site at conc. > 2 Bq/L (^{90}Sr drinking standard in Ukraine) is about 200m for a maximum depth of 15 m

Modelling results vs monitored data (period 1999-2002)



Problems linked to shortcomings of the flow-transport model:

- T22 volume and inventory in ^{90}Sr reassessed for 1988 at 412 ± 15 GBq (Nguyen, 2017: all data + georadar + geostatistics) vs initial estimate of 290 ± 140 GBq (Kashparov et al, 2012)
- Heterogeneities not fully considered
- Assumption of a 1D model for the unsaturated zone
- Assumption of steady state hydraulic and geochemical conditions

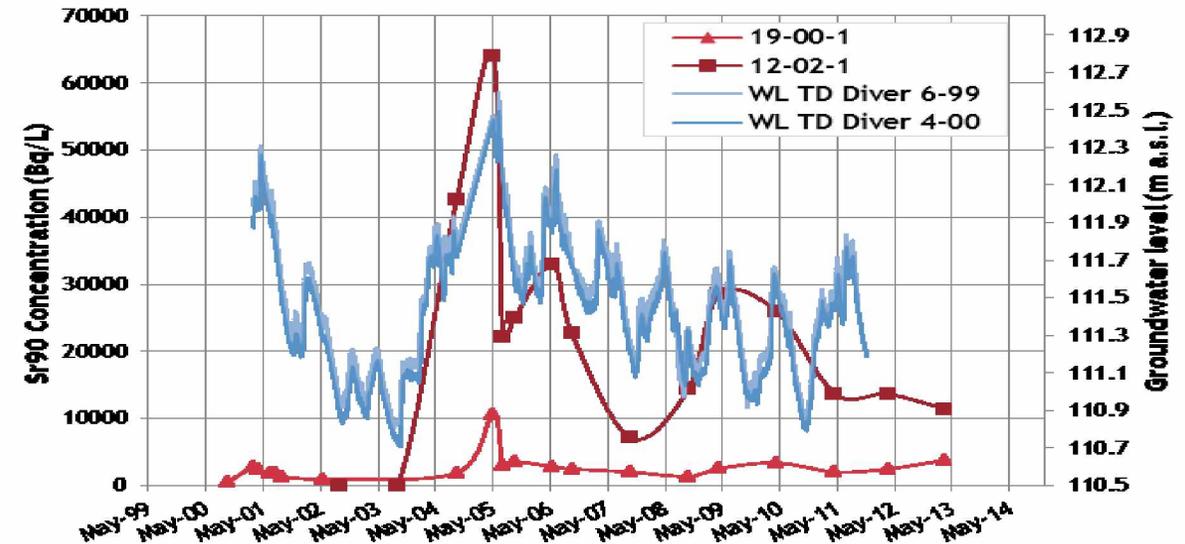
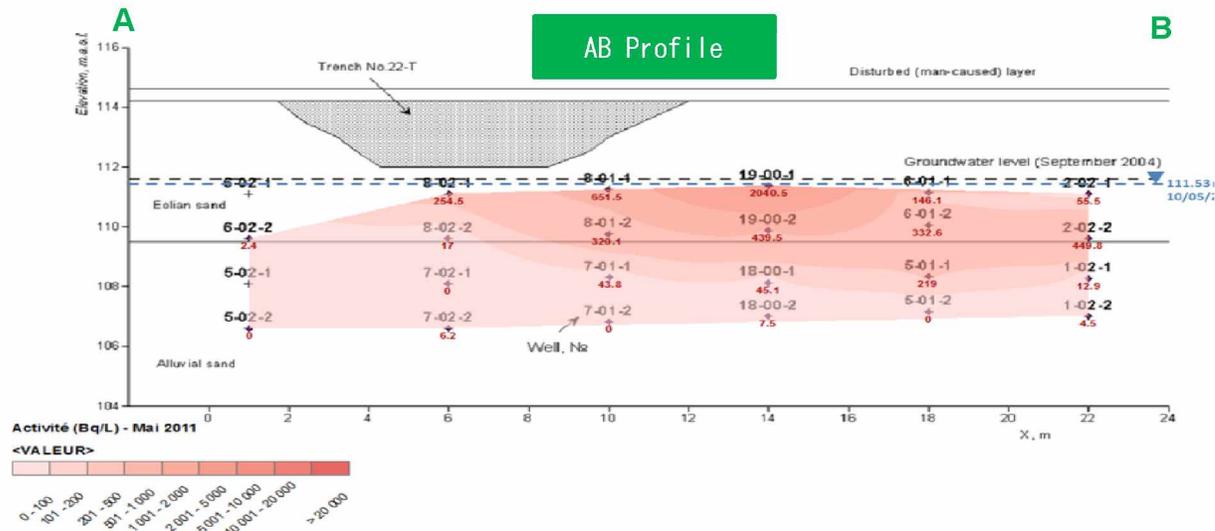
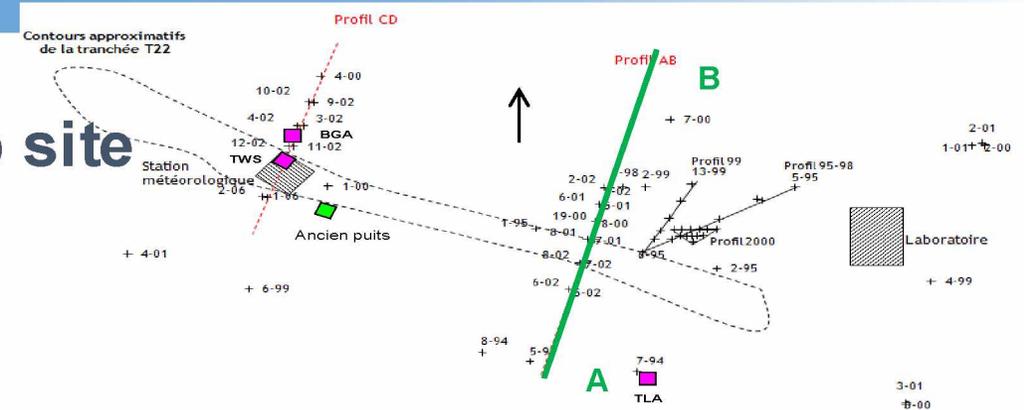
Main findings

- Years after accidents aquifers are still contaminated mainly by medium-lived fission (^{137}Cs , ^{90}Sr) and activation (^3H) products with rate transfers: $^3\text{H} = \text{water flow} > ^{90}\text{Sr} \text{ (weakly sorbed)} \gg ^{137}\text{Cs} \text{ (highly sorbed)}$
- Heterogeneities in aquifer flow and transport properties together with the transient conditions influence the RN distribution in hydrosystems
- Retention likely dependent on spatial heterogeneities and vegetation uptake
- Modelling are limited to steady state calculations as transient conditions should imply a high computation cost and running time

Main research topics

- Topic #1: Sensitivity of models to transient conditions and spatial heterogeneities
- Topic #2: Retention model at a given context
- Topic #3: Optimizing flow-transport modelling through mesh refinement

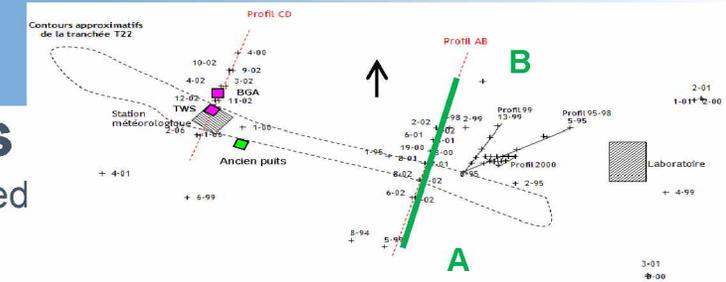
Case study of Trench 22 at the Waste Dump site in the Chernobyl Exclusion Zone



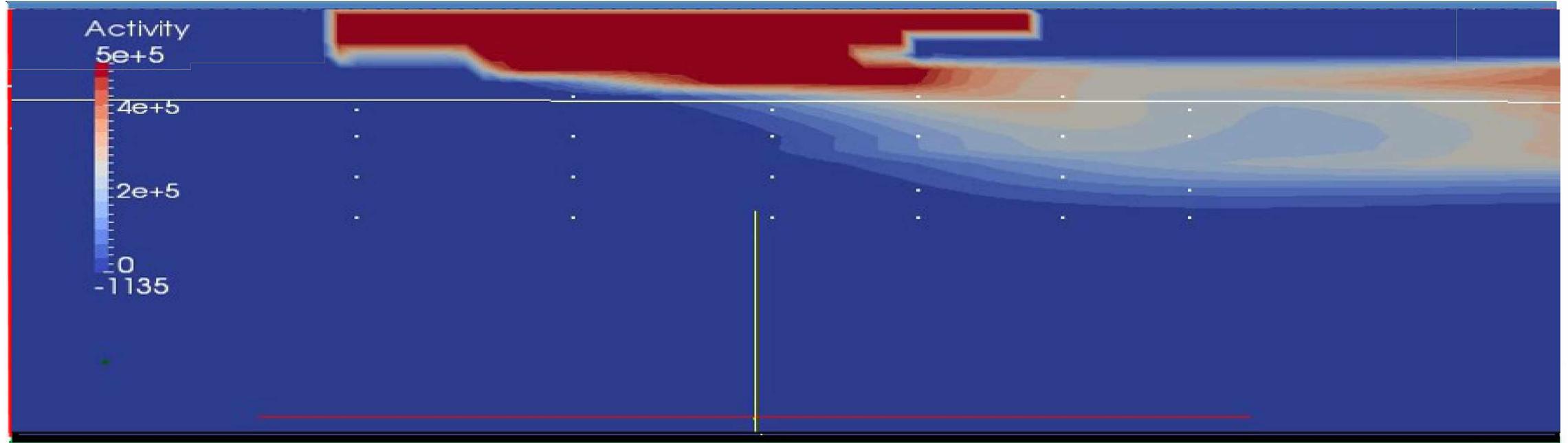
Strong variations in volumetric activity with time and space: what are the effects of transient conditions and of the spatial heterogeneities on ⁹⁰Sr transfer?

Transient 2D modelling with variable water level and heads

MELODIE v5.1 deterministic software for modelling flow and transport in porous saturated and unsaturated



Recharge



A

B

The model is much conform to the temporal dynamic but differs from one cross section to another one → need for a 3D modelling ...

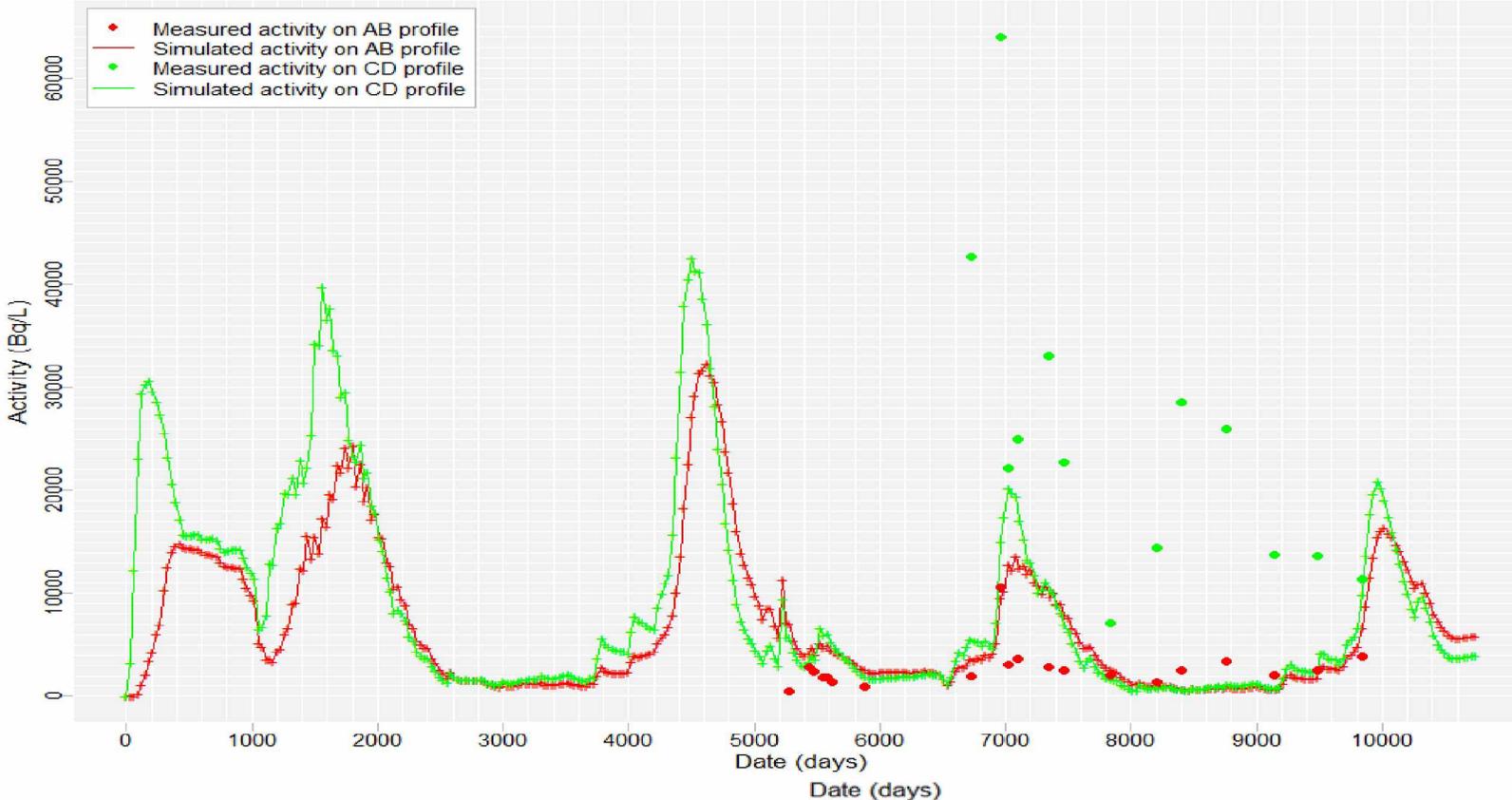
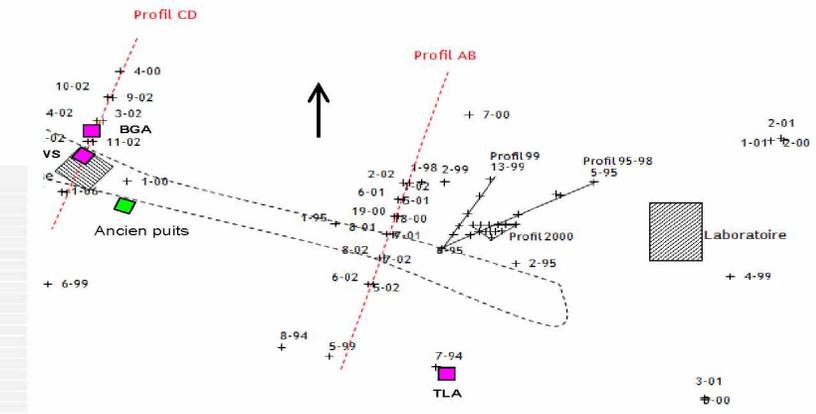
Sensitivity to spatial distribution of permeability

Heterogeneous inventory

$K_{eol} = 200.0$ m/day - $Kd_{eol} = 5.0$ mL/g

$Kh_{all} = 1.0$ m/day - $Kv_{all} = 0.01$ m/day - $Kd_{all} = 5.0$ mL/g

Contours approximatifs de la tranchée T22



Transient model with former K values overestimates activities

Best fit for K_{eol} 40 times higher to that proposed in the former model

➔ Need for a full spatial characterization of flow parameters (eg permeability field)

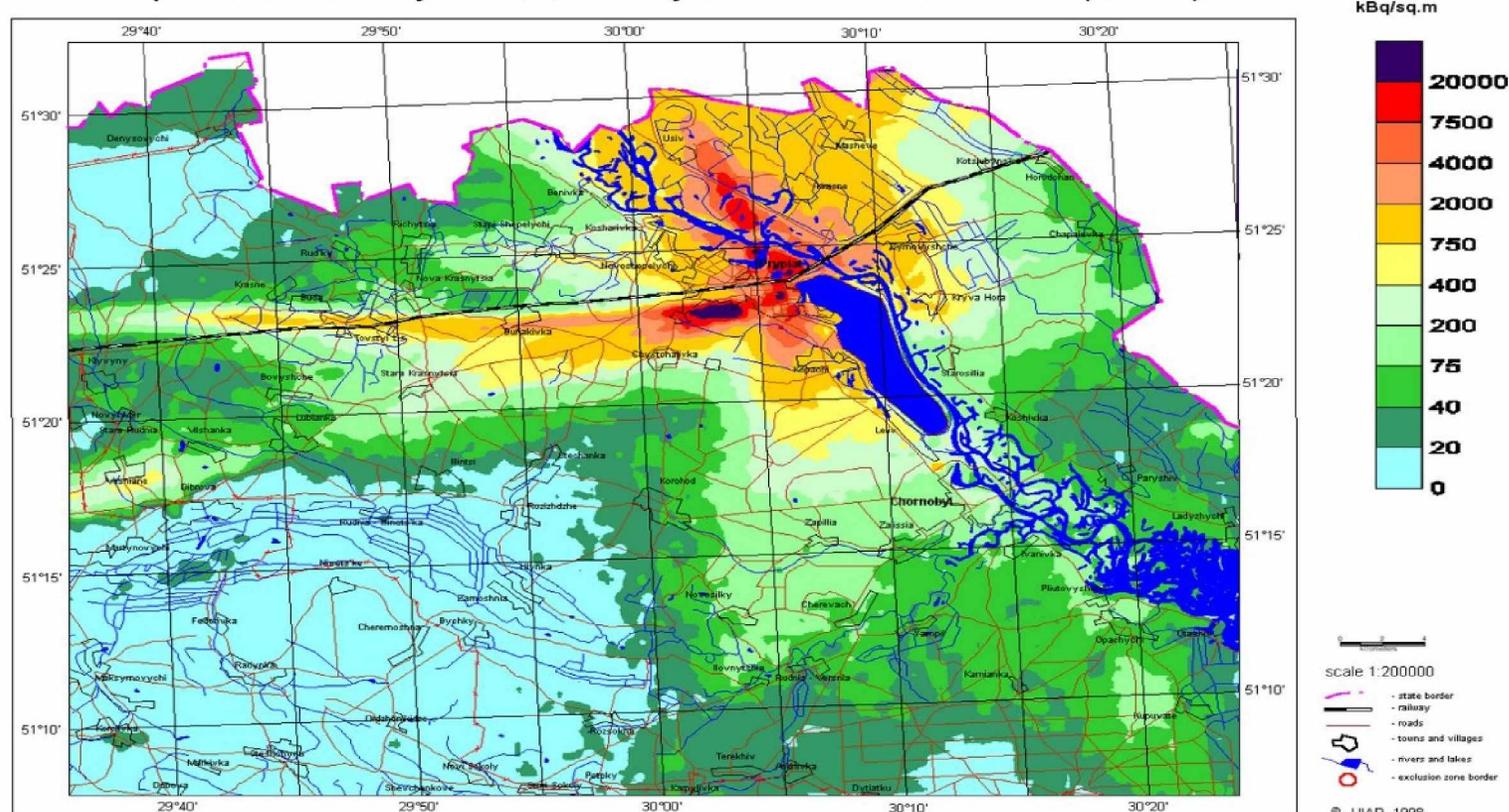
Topic #2: retention model → Ground surface vs free aquifer ⁹⁰Sr contamination at EPIC site

Only exist in the ~10 km zone around ChNPP

⁹⁰ Sr, kBq/m ²	C _{gw} , Bq/L
10	0,06
100	0,6
1000	6,1
10000	61,3

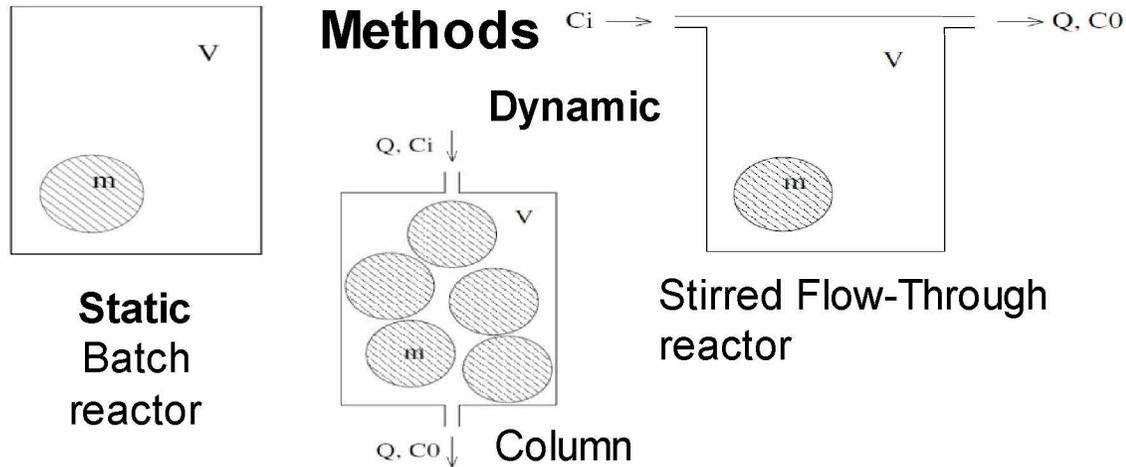
[Bugai et al., 1996]

The map of the 30-km Chernobyl zone terrestrial density of contamination with strontium-90 (on 1997)



⁹⁰Sr concentrations in groundwaters depend on the surface contamination density

Topic #2: retention model → Sorption parameters at the EPIC site



Distribution coefficients

$$K_d \left(\frac{L}{kg} \right) = \frac{[RN]_{soil/solid}}{[RN]_{solution}}$$

Retardation factors

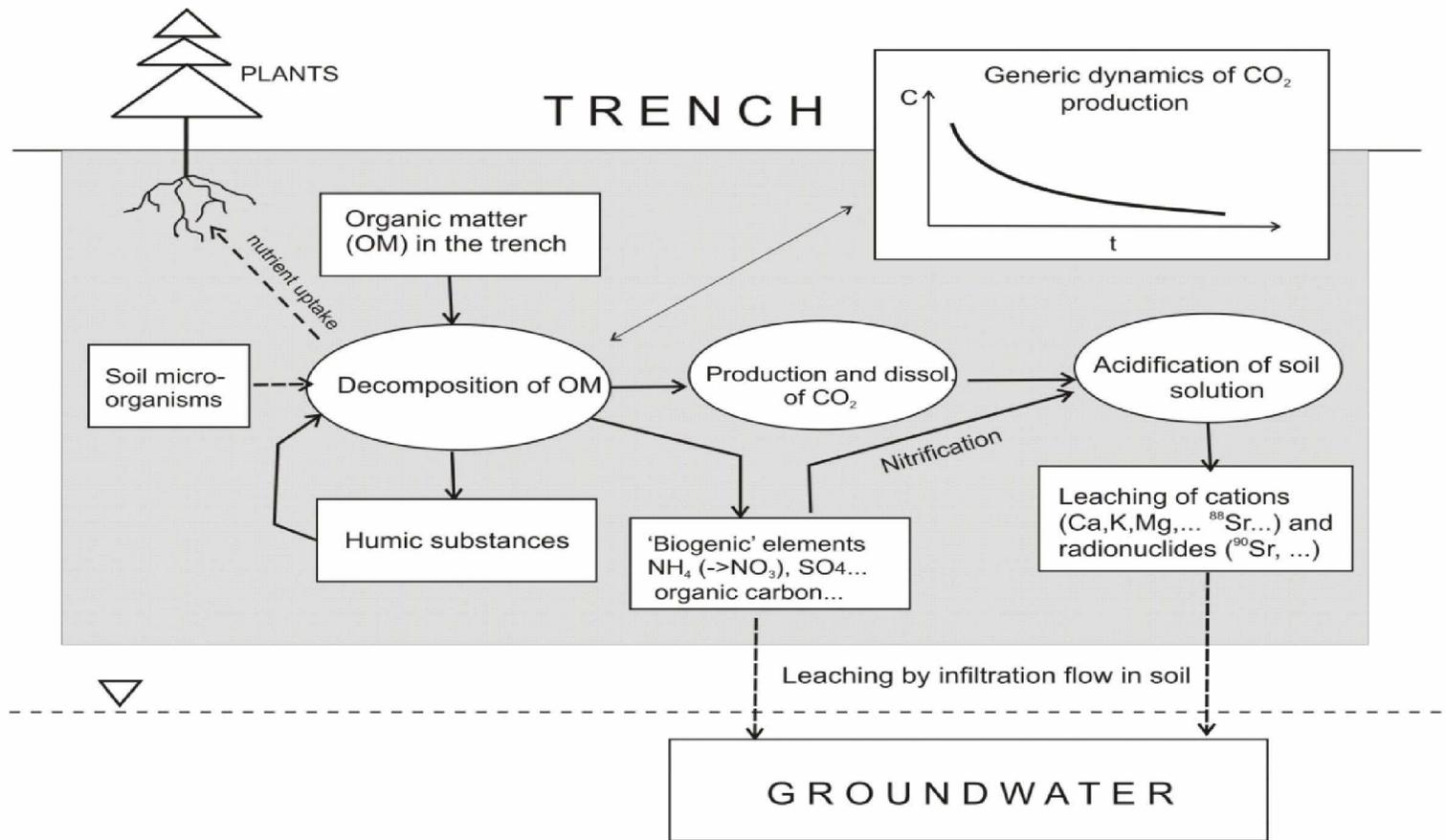
$$R_{sat} = 1 + \frac{(1 - \omega)}{\omega} \rho_s Kd_{sat}$$

$$R_{uns} = 1 + \frac{\rho_s}{\theta} Kd_{sat}$$

$$\text{div} (D \overline{\text{grad}} A - U A) = \omega R \frac{\partial A}{\partial t} + \lambda \omega (R_j A_j - R_i A_i)$$

Radio nuclide	Kd, L/kg IAEA TR 472, 2010	Measured Kd L/kg, R - (eolian sands)	Sorption from dynamic+static experiments [Szenknect, 2003]
Sr-90	1...25	0.25/5 (batch/column) 3 (field+flow-through) R ~ 17 (~ 0.6 m/a)	“fast” (< 1 min) reversible (desorption 100%) and non linear sorption process by Cation exchange with influence of porewater composition (pH, Ca)
Cs-137	20...5000	100-600 (lab tests) R > 6600 (~ 2 mm/a)	Sorption faster (< 10 min) than desorption (< 20h), quasi reversible and strongly non linear due to specific sorption sites. Kinetic of sorption evidenced from dynamic column tests

Topic #2: retention model → Conceptual bio-geochemical radionuclide migration model for the “Red Forest” waste dump site



Martin-Garin et al, 2012



⁹⁰Sr fluxes from the trench inventory:

Biogenic flux

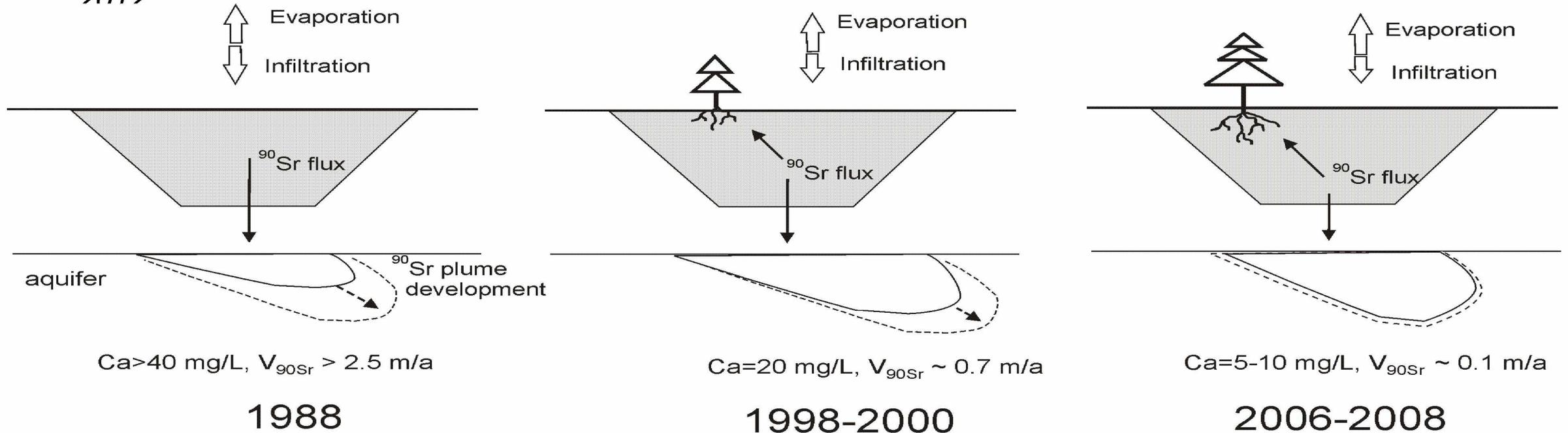
0.82% a⁻¹ from trench inventory in 2005 [Kashparov et al, 2012]

Aqueous-migration

0.14-0.5% a⁻¹ (1987-2000) [Bugai et al., 2001]

Topic #2: retention model → Natural attenuation process in red forest GW

Evolution of ^{90}Sr migration fluxes from trench T22 in 1988-2008 *Martin-Garin et al., 2012*



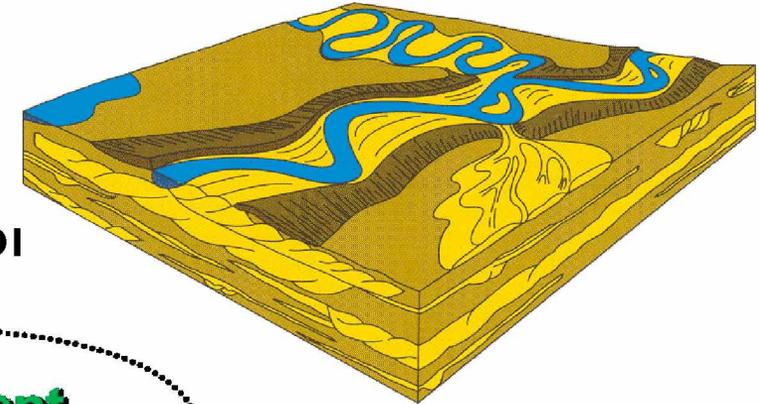
“Biogenic” factors contributing to natural attenuation in GW:

- Direct ^{90}Sr and ^{137}Cs uptake by vegetation leading to their incorporation to biological cycle;
- Variation of GW geochemistry (pH, Ca, K, Sr...) due to level of organic matter degradation and/or variations of uptakes by vegetation, which influences the ^{90}Sr mobility

■ Motivation

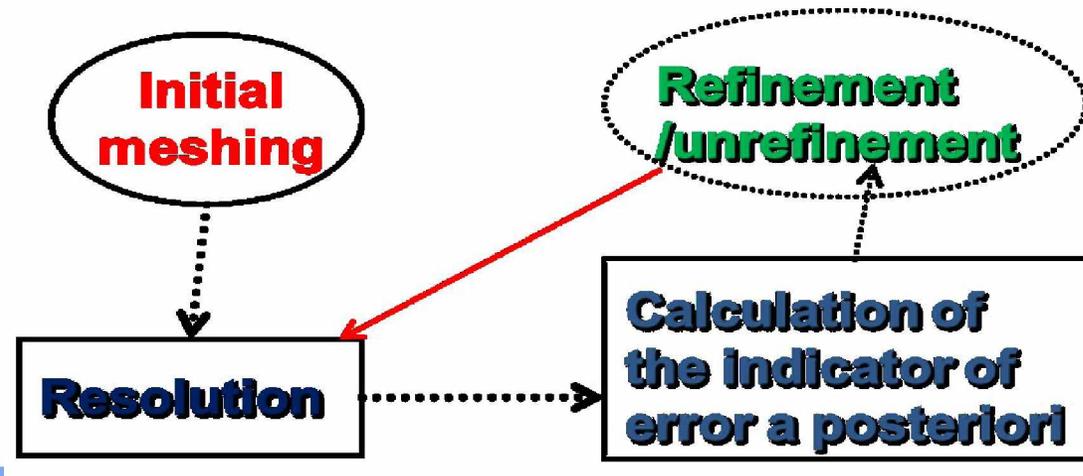
- To obtain a more accurate solution in case of transient state for flow and transport and for an extreme heterogeneous and anisotropic context as are alluvial plains

Mariethoz et al., 2009



- To optimize the mesh and computation cost (runtime, memory)

■ How does it work?



Heterogeneous and anisotropic model

Mesh	CPU time, s	Nb cells
Dynamic	50	3 000
Static	200	13 000

Flow boundary condition :

Hydraulic head =

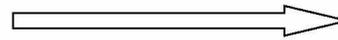
left side : 30 m

right side : 0 m

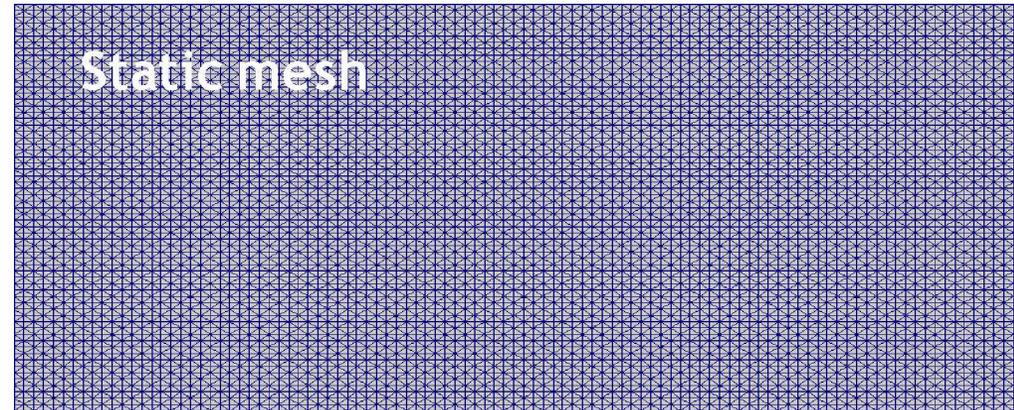
Injection pulse condition :

Duration : 0.01 unit of time

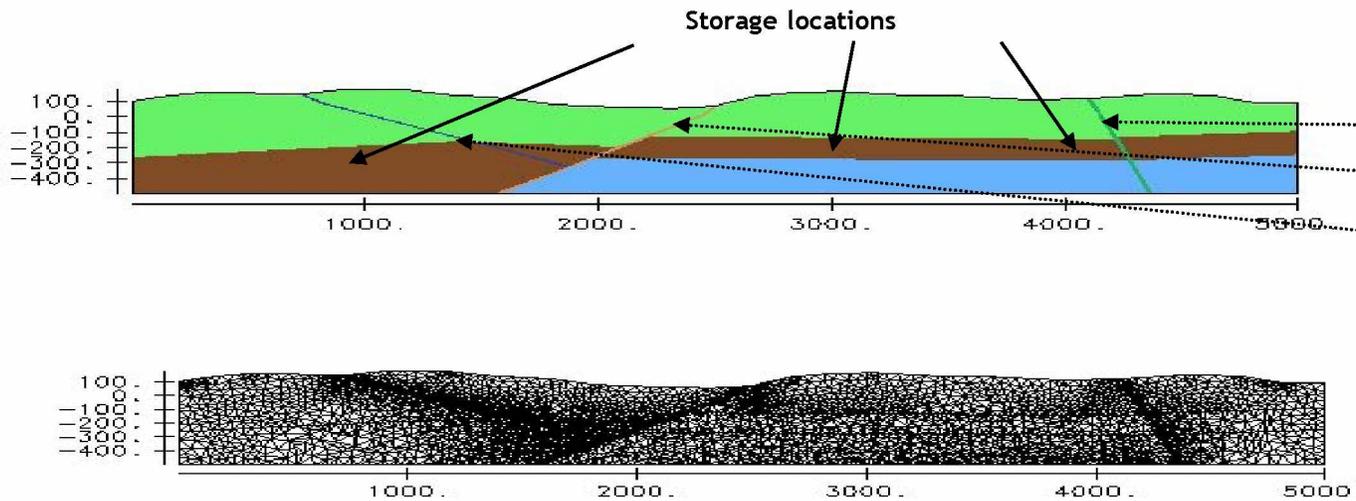
Quantity : 600 Bq



Flow direction



	Permeability		Porosity	Dispersion		Diffusion
	X	Y		α_l	α_t	
	5.	5.	0.1	1.	5.e-4	0
	1.	1.	0.05	0.5	0.5	0



	$K \text{ m}\cdot\text{year}^{-1}$	ω	Dispersion m		Diffusion $m^2\cdot\text{year}^{-1}$
			α_l	α_t	
Dark Brown	10^{-4}	0.3	1.	1.	0.01
Bright Green	10.	0.15	0.5	0.5	0.05
Light Blue	50.	0.17	10.	1.	0.05
Dark Green	5.	0.15	8.	0.4	2.
Yellow	0.05	0.2	5.	0.5	1.
Blue	500.	0.2	12.	6.	0.5

Injection pulse condition :
Duration : 0.1 year
Quantity : 600 Bq
Simulation : 0 - 10 000 years

Conclusions

- In case of severe accident, groundwaters are systematically contaminated. The radionuclides of primary concern are strontium-90 and sometimes tritium. Other radionuclides such as caesium-137, Pu isotopes and Am-241 usually show lower mobility due to higher retention properties of soils and aquifers.
- Modelling the fate and transport of radionuclides by groundwater pathway remains a challenge due to the complex interplay between the hydrogeological, geochemical and biotic processes. Of primary importance are to better estimate: 1) The inventory of the source term by using all available on-site data and geostatistics; 2) Representative properties for transport by considering transient conditions (in response of rain inputs, water level fluctuations, uptake by vegetation...) and retention including effects of biotics and ground heterogeneities properties at different scales (from watershed to atome).
- Improvements in predictive modeling should imply further works by combining, geophysical acquisitions, geostatistics and new developments in modeling works especially to minimize acquisition and high computation runtime, memory,..) by mesh optimizing.
- Minimizing the restricted areas next to severe accidents must therefore imply maximizing characterization where lands are assumed to be given back to people (eg CEZ)