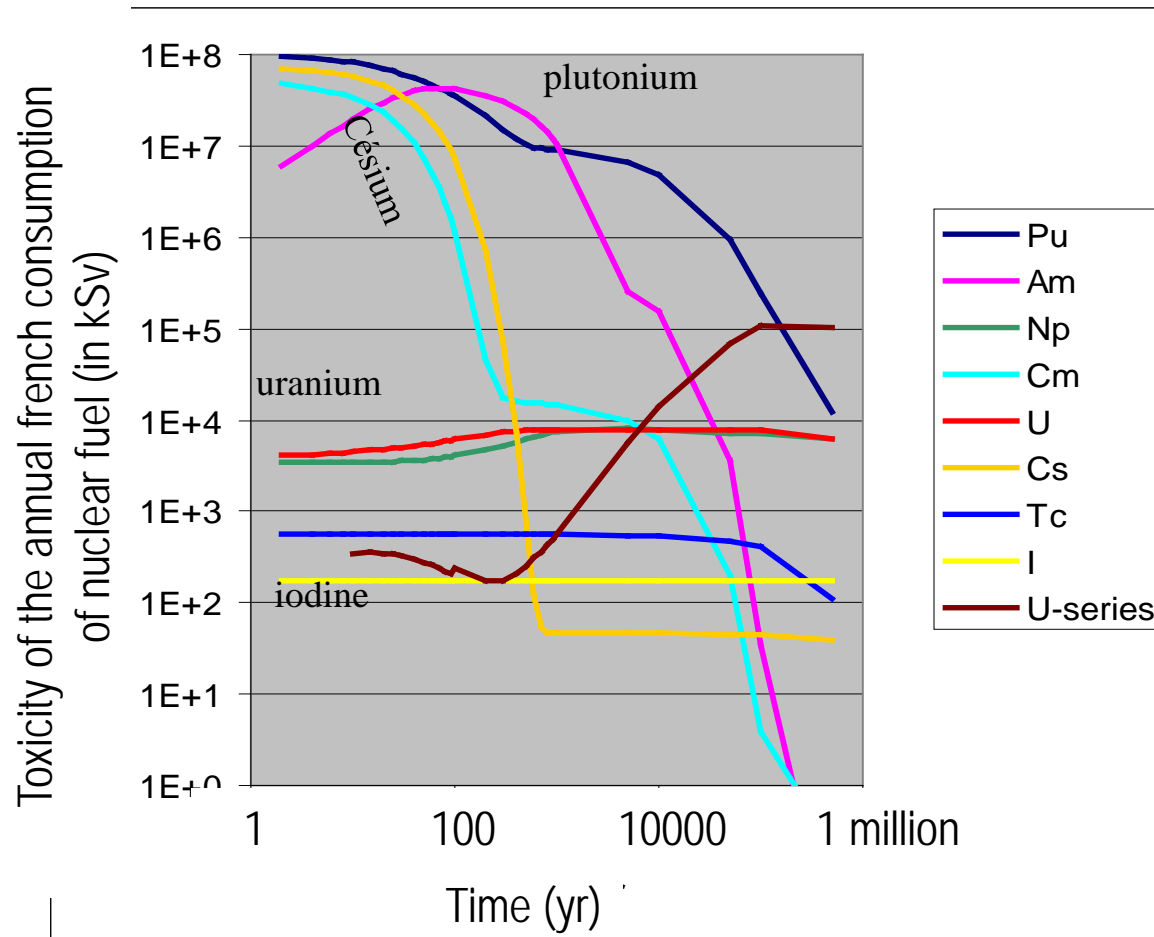


RADIOACTIVE WASTE DISPOSAL IN EUROPEAN CLAY FORMATIONS: SCIENCE, SAFETY AND SOCIETY

Bernd Grambow
SUBATECH
(University, Mines Nantes, CNRS-IN2P3)

Nuclear waste: an toxicity inventory about 5000 times higher than that released in Fukushima



Which protection? 1) Confinement in solid matrices stored at **interim storage sites**
2) Geological **Disposal**

We need to organize multigenerational safety

- ❑ Without disposal, interim storage becomes an ever lasting pre-occupation
 - implying an important transfer of radiological liabilities to future generations
 - Needs for long term stewardship
 - Need for stable political situation over hundreds of years : compared to disposal, an interim storage site increases the vulnerability, in particular in case of international crises, terrorism
 - Spent fuel storage tanks close to high population areas still remain a high security risk
 - The risk of abandon in far future, in particular if a country steps out of nuclear energy
 - Acceptable if looking for a better solution, like transmutation, but there is no solution for most radionuclides and it does not work for vitrified waste. Transmutation does not diminish the need for disposal
 - Acceptable for low level short lived wastes
- ❑ For high-level long lived waste one needs a form of long-term isolation that allows for maintaining control but that limits vulnerability and the consequences of abandon and ignorance
 - This is the ideal of the passive barrier system of disposal
 - The radiological consequences of abandon are lower than in interim storage
 - However, opponents often identify disposal with abandon



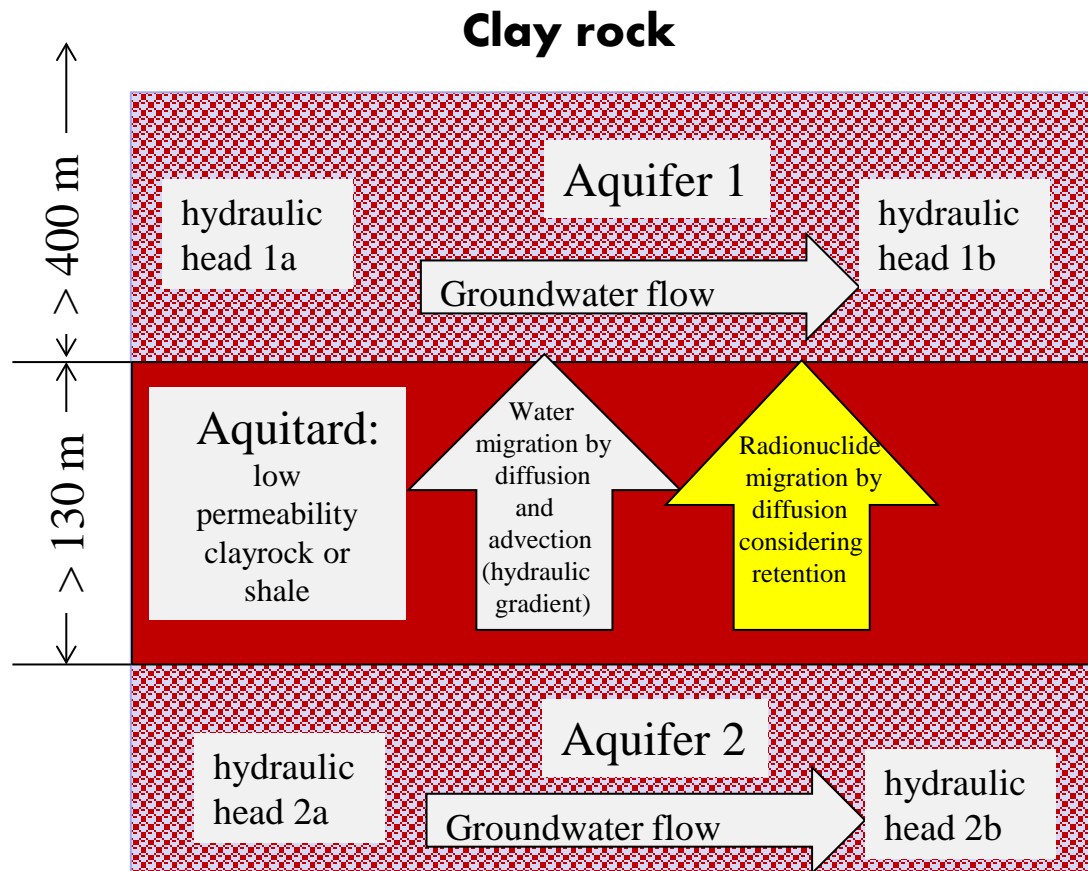
Creation of the european technology platform IGD-TP

- In 2009, several European waste management organizations have established a technology platform to accelerate the implementation of geological disposal of radioactive waste in Europe
 - Under EC auspices
- Industry-led to ensure relevancy of priorities and implementation of RD&D results
 - Members of the Executive Group shall be organizations either being responsible for implementing a disposal programme or being formally responsible for RD&D programme needed for implementation
- Open to all science providers
 - Consulting companies, academics, research centers...

The detailed evolution of geological disposal cannot be predicted, but a number of issues must be addressed :

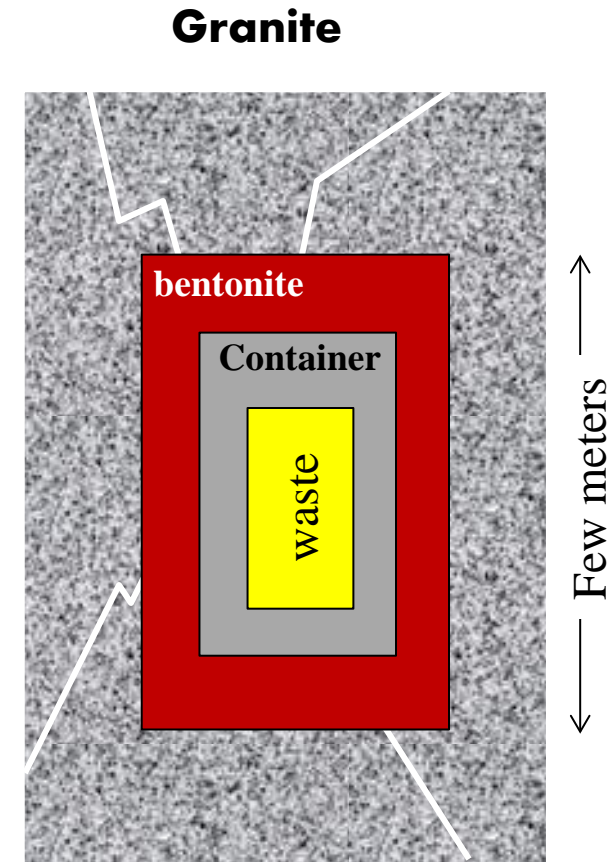
- ☐ Geological stability of a site (earth quakes?)
- ☐ Risks of human intrusion
- ☐ Time of container tightness facing corrosion by groundwater
- ☐ Radionuclide release rates from the waste into groundwater
- ☐ Degree of radionuclide fixation on rock surfaces
- ☐ Isolation times of the repository for radionuclides

Principle of isolation



Bure/France
Due to osmosis;
apparent hydraulic
head in clayrock
up to 40 m higher
than in adjacent
formations

Migration time in clay
 $> 10^5$ yr

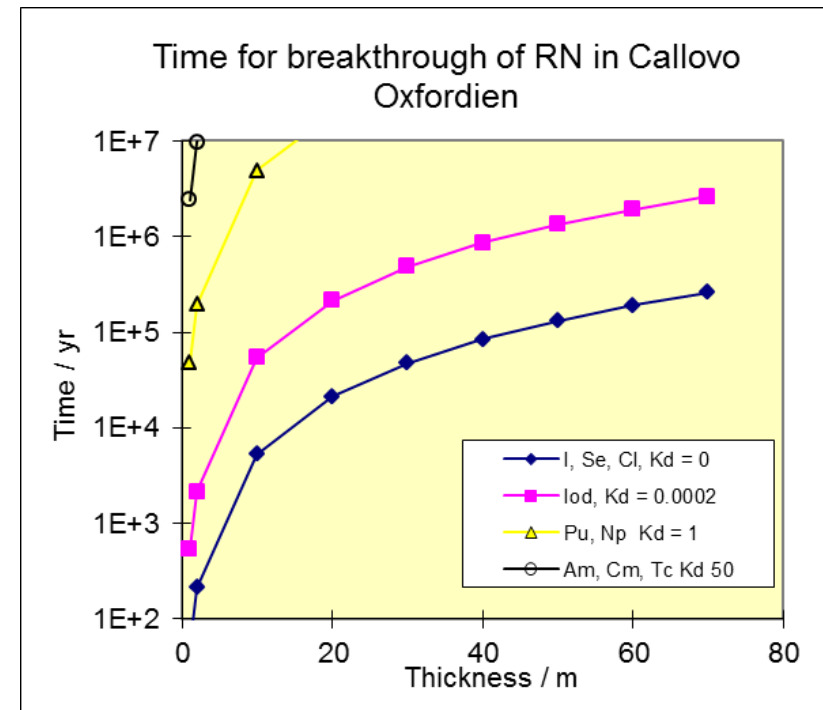


Container (Cu, Fe) life time
 $> 10^5$ yr

Safety based on near field

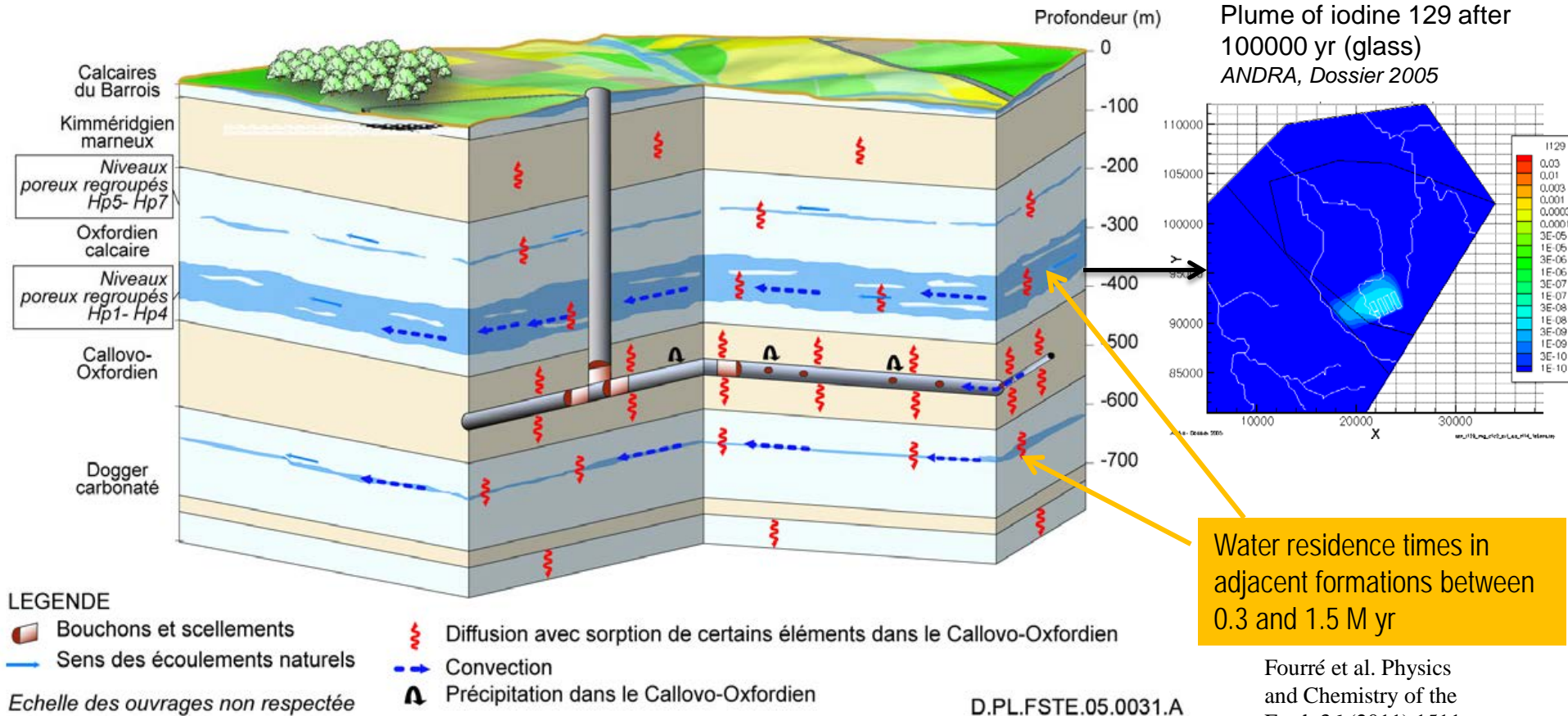
Clay formations are promising repository host formations

- Clay formations are repository reference concepts in France, Belgium and Switzerland
- Geological stability for hundreds of millions of years
- Very large extension and hundreds of meters of thickness
- Strong retention and low solubility of cations: Plutonium, Uranium and Technetium are expected to move only few meters prior to decay in hundreds of thousand to million of years
- Anions (I-129, Cl-36, ...) are the most mobile species



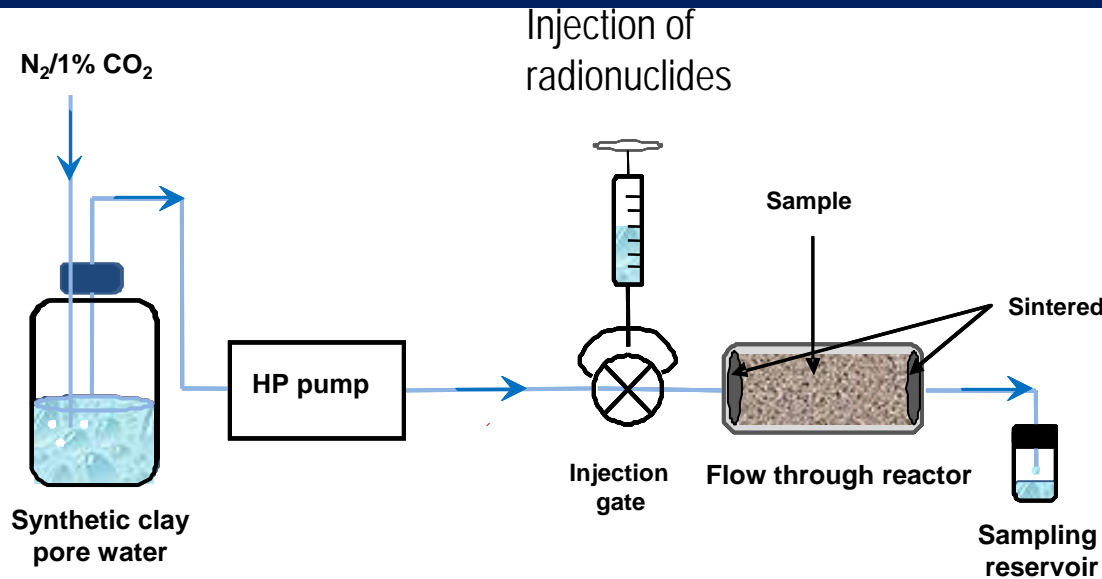
Is it possible to isolate the radioactivity in deep clay formations so that it will never contaminate soils, aquifers and food chains ?

Example: site in Callovo-Oxfordian clayrock in France proposed by ANDRA



Simulating the repository performance in percolation tests with 10000 times increased hydraulic gradients

Work financed by ANDRA



Sample	Drilling	Depth (m)	Content in clay minerals (% w/w)	Content in carbonate phases (% w/w)	Lithofacies
EST 25695	PAC 1002	474	60 ± 16	18 ± 4	C2b1
K119	EST 205	477	45 ± 3	25 ± 2	C2b1
EST 26480	FOR 1118	490	53 ± 21	24 ± 5	C2b1
EST 27861 (AB)	EST 423	501	17 ± 7	37 ± 8	C2c
EST 27861 (B)			26 ± 7	17 ± 4	

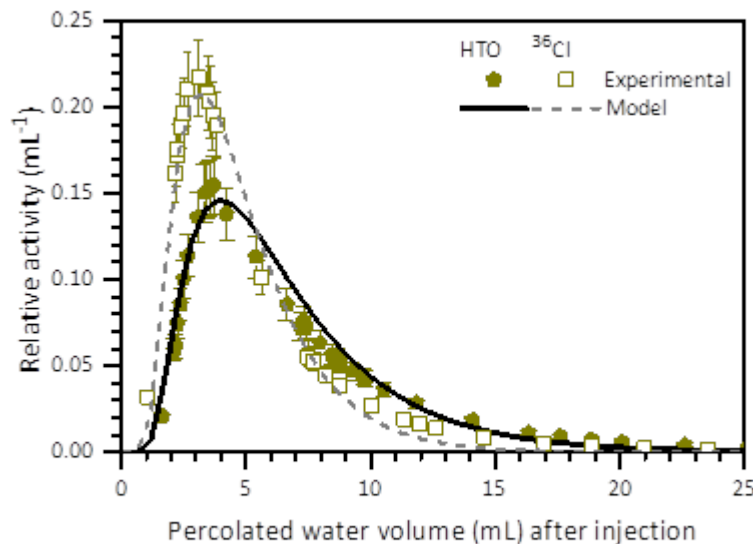
Modell buildup (1D diffusion, advection, mineral solubility, radionuclide speciation...)

Observed hydrodynamic parameters (injection of HTO or ^{36}Cl):

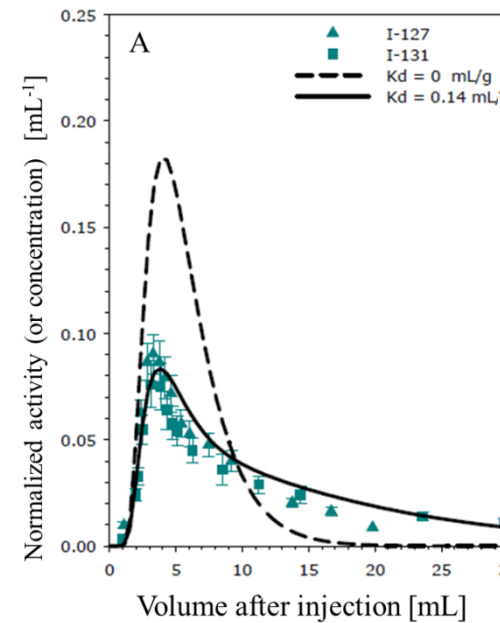
Anion accessible porosity 50% (clay rich) to 75% (carbonate rich) of total porosity

Permeability: 10^{-12} to 10^{-14} m/s

Diffusion coefficient for I-129 2×10^{-12} m²/sec



Some retention for iodine



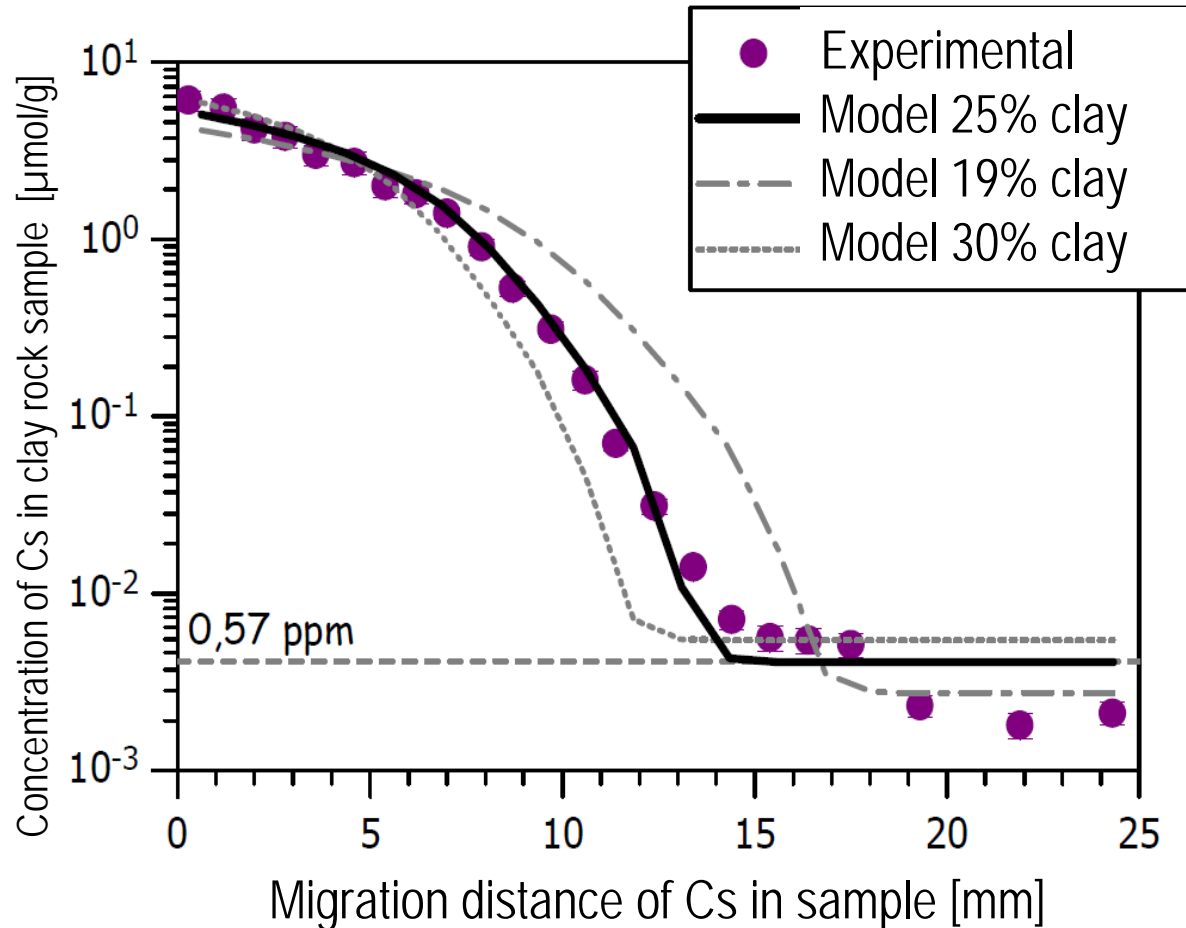
These data suggest:

that break through of anions in 70 m of callovo oxfordian clayrock takes more than 200000 yr and that diffusion is the dominant transport mechanism

Iodine retention (ignored in safety calculations) would block this nuclide for 10^6 yr

Analysis of retained Cs in sliced clay rock sample at test termination

Agreement of model and experiment: $K_d(\text{trace})$ between 500 and 1500 L/kg



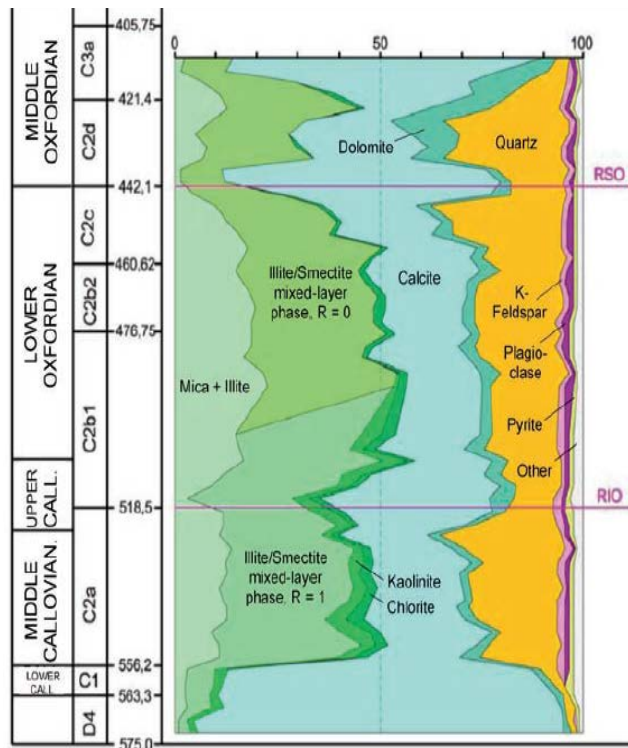
Our results show that Cs isotopes are effectively retained:

Cs137 can never leave the glass container due to decay prior to container breaching

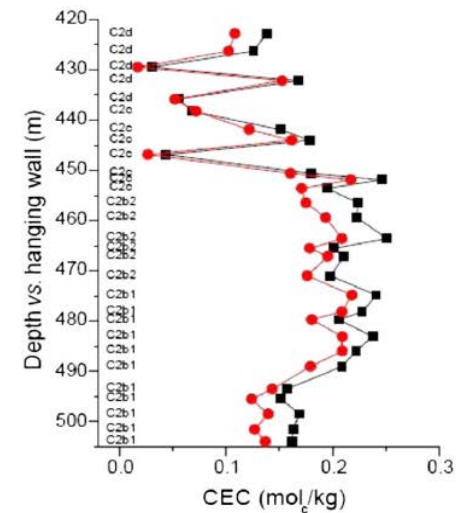
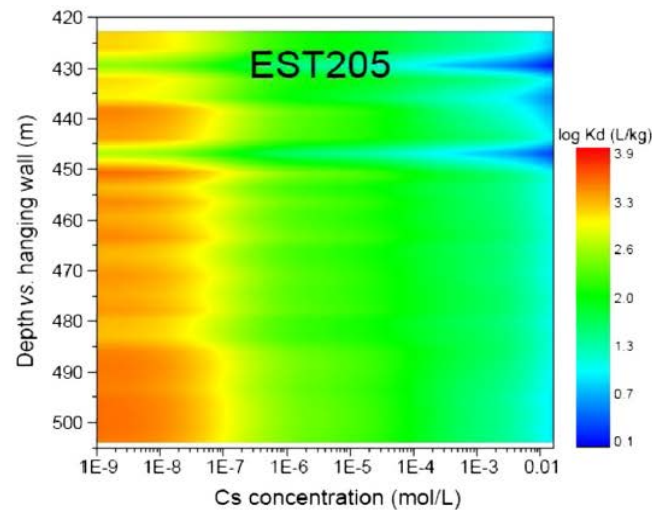
Cs135 can not migrate over a distance much more than some 10 m in the clayrock

Cs135 migration: How can one deduce from short-term experimental data, measured on centimeter- to decimeter-sized rock samples, the long term behavior of geologic units over a scale of hundreds of meters?

- Step 1: batch test assessment of sorption of Cs on individual minerals ($=f(\text{pH}, T, I, \text{pCO}_2\dots)$)
- Step 2: surface complexation modelling
- Step 3: assessment of additivity
- Step 4: assess applicability of batch data derived models to compact clay (percolation tests)
- Step 5: apply to heterogeneous clay rock column



From Rebours *et al.* (2004)



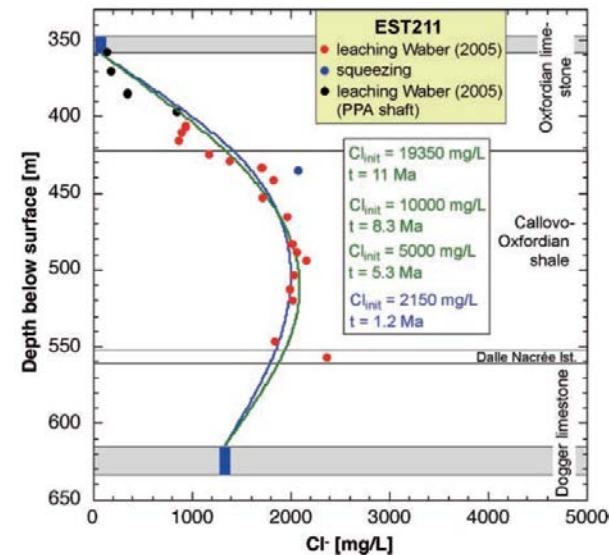
Z. Chen, G. Montavon, S. Ribet, Z. Guo, J.C. Robinet, K. David, C. Tournassat, B. Grambow, C. Landesman, Key factors to understand in-situ behavior of Cs in Callovo–Oxfordian clay-rock (France), *Chemical Geology*, 387, 2014, 47-58

How can one be assured that a scientific model or theory derived from experimental data is applicable under larger-scale repository conditions? Natural tracer profiles in clay formations

➤ Chloride

- Diffusion alone can explain the data, but uncertainties in paleohydrological data are large. In some cases (high salinity base aquifer) steady state diffusion (time 10-20 Myr)
- High initial concentration of between today's value (2150 mg/L) at 1.2 Myr and seawater (19350 mg/L) 11 Myr ago

Data: Mazurek et al. Natural tracer profiles across argillaceous formations, the claytrac project. OECD/NEA 2009



➤ Water isotopes

- Stable isotope data indicate water travel time of about 1.2 to 2 Myr (max 4 Myr)

➤ Helium

- Production can be estimated from measured Uranium and Thorium contents and formation age of 155 Myr. Data are in agreement with transient diffusion over 4 Myr

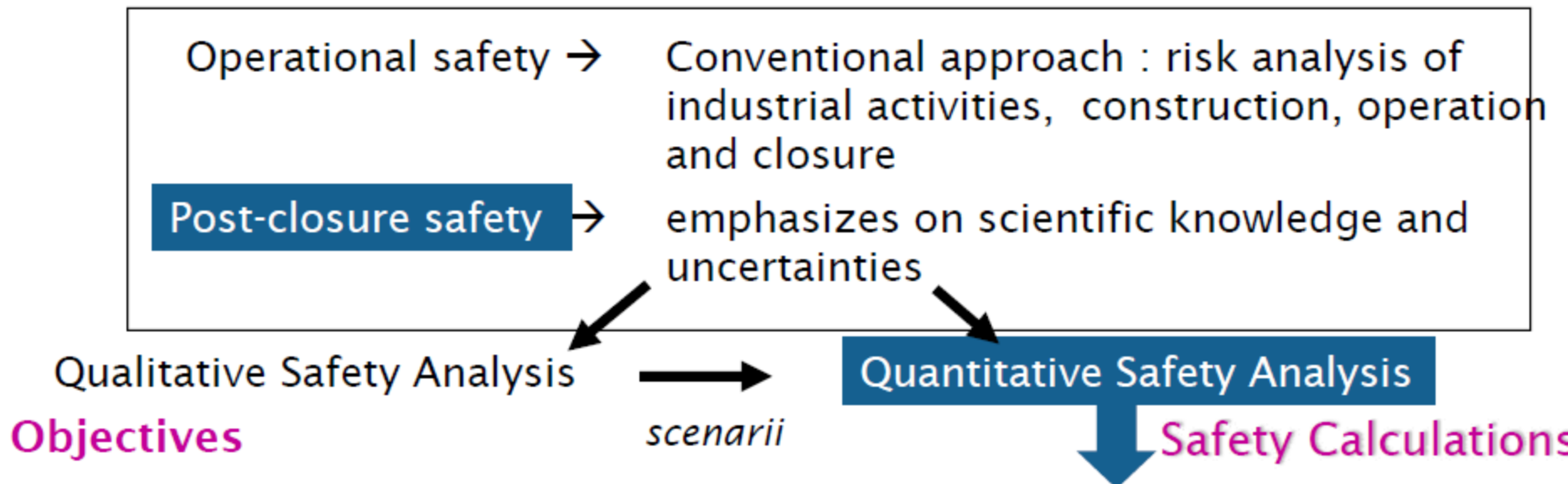
➤ All data indicate that callovo oxfordian clay can fix the most mobile species for million years

From data and field observation to safety assessment

- ❑ The European Community Project PAMINA (27 European organizations) studied the impact of uncertainties on the safety
- ❑ Model, data and scenario uncertainties
- ❑ Safety assessment is now a routine exercise used at various stages in the development of repository projects,
 - to compare alternatives
 - for demonstration of compliance with regulations
- ❑ But the principal role of safety analyses is not the numerical result (dose, risk) but the identification of key factors influencing safety.

The vision of the french nuclear waste agency

Safety Analysis



Objectives

- ☐ Evaluate the radiological and toxic impact for different scenarii,
- ☐ Check that the performances of the design components are reached to ensure the overall safety,
- ☐ illustrate the understanding of the behavior of the disposal, **particularly with regards to RN transfer** in near field and far field, from the wastes to the outlets
- ☐ Contribute to give a feed-back for research priorities and design orientations.

Kinds of calculation performed

❑ Deterministic calculations → mono and multi-parametric

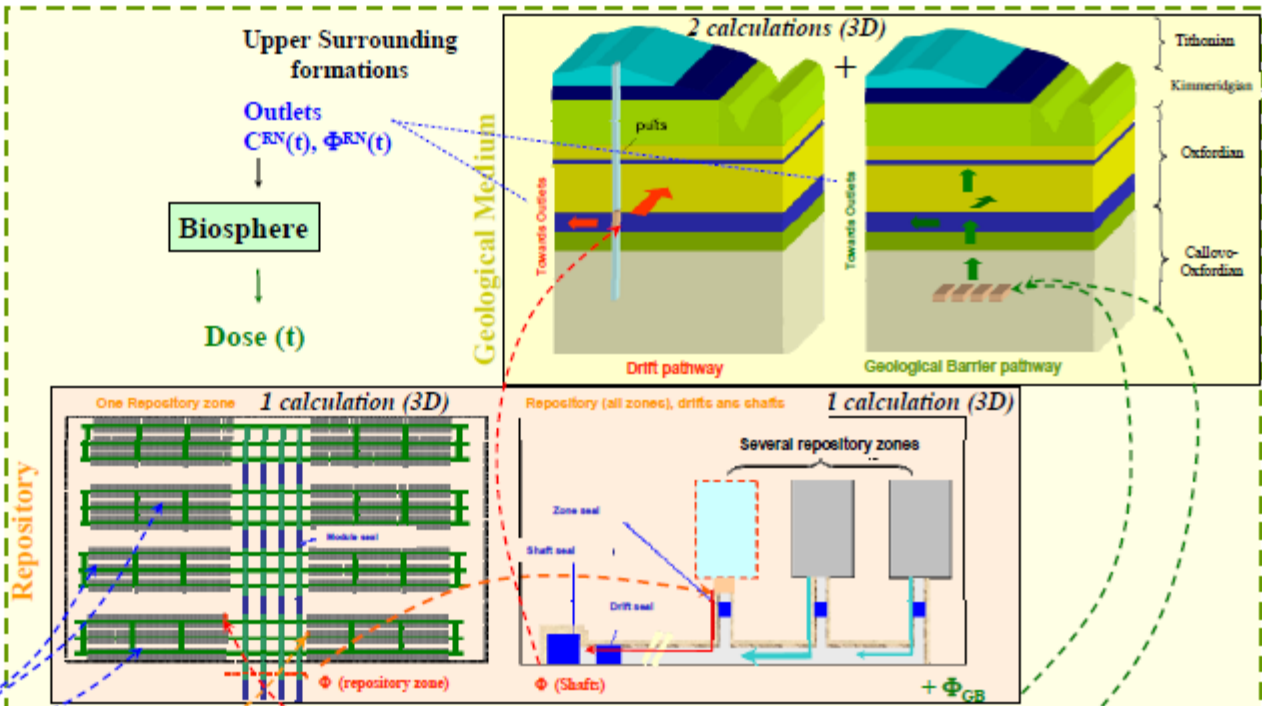
- **Reference** → models/parameters « phenomenological » (« best-estimate »)
or « conservative » in some cases
(*{pheno/techno} {high/low} value in range*)
- **Sensitivity** → models/parameters « conservative »
or « pessimistic » in some cases
(*conventional or physical limit value*)
or « alternative models » in some other cases

❑ Probabilistic calculations (Monte-Carlo) → multi-parametric

- **Sensitivity analysis** → classification of input data whose uncertainty is relevant
- **Uncertainty analysis** → distribution of the result

Safety calculations architecture (SEN)

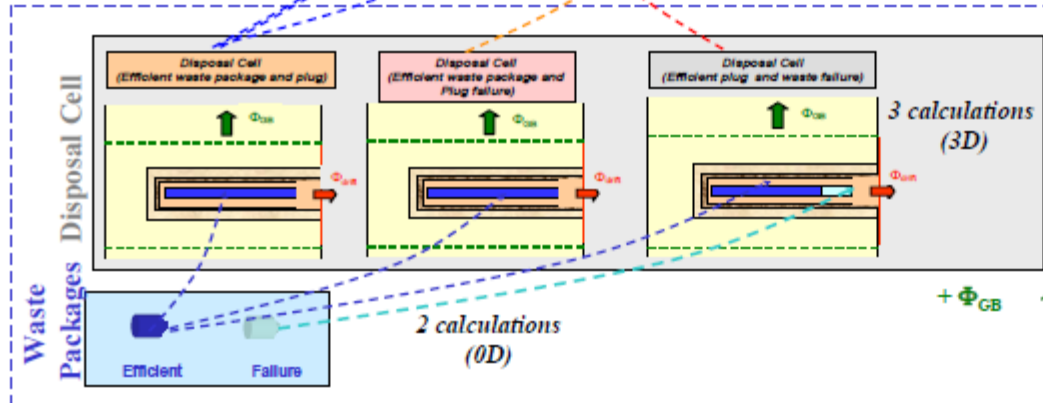
FAR FIELD



Indicators :

Molar fluxes
Concentration
Peclet Number
travel times
...

NEAR FIELD



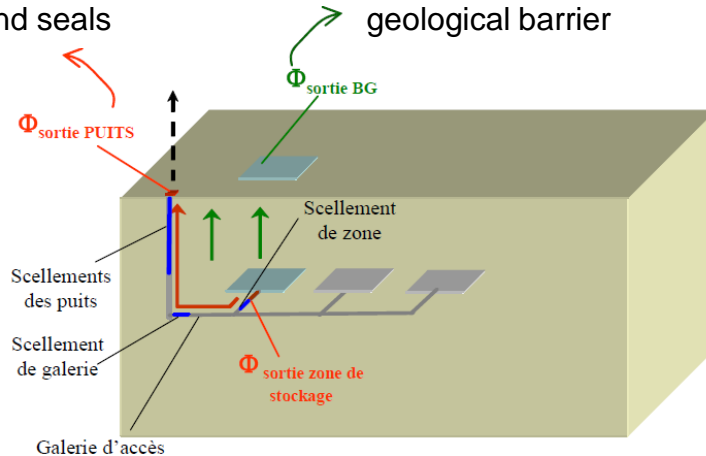
For each
{Radionuclide /
Waste Package},
→ 7 calculations (3D)

Evolution scenario

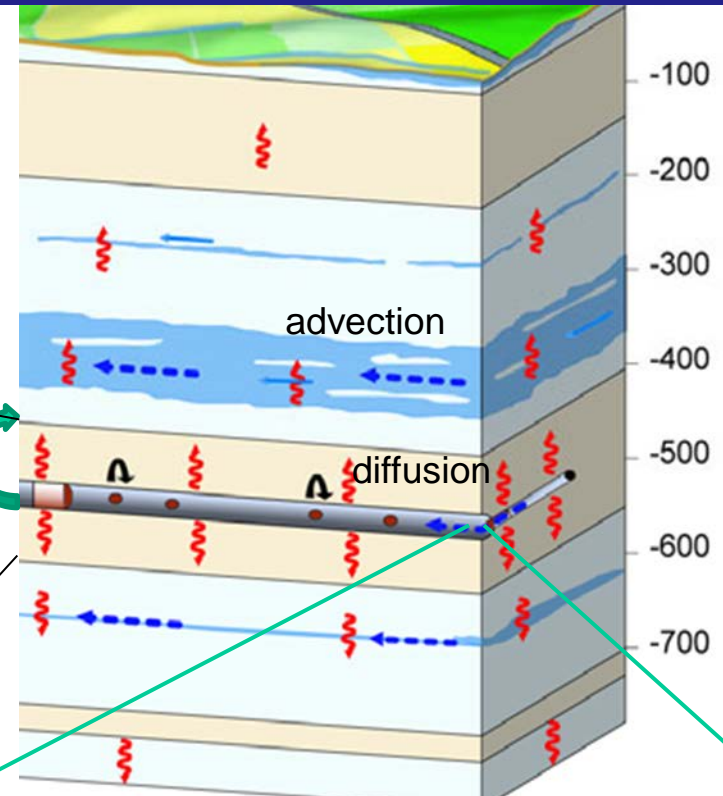
- ⑤ Decay of actinides, Tc, Cs etc.. They cannot leave the clayrock
- ⑥ Due to slow advection, diffusion is the principal transport mode the transport of Iodine out of the clayrock takes > 200000 yr
- ⑦ Iodine is principal dose contributor

B) Transfer through galleries and seals

A) Transfer through geological barrier

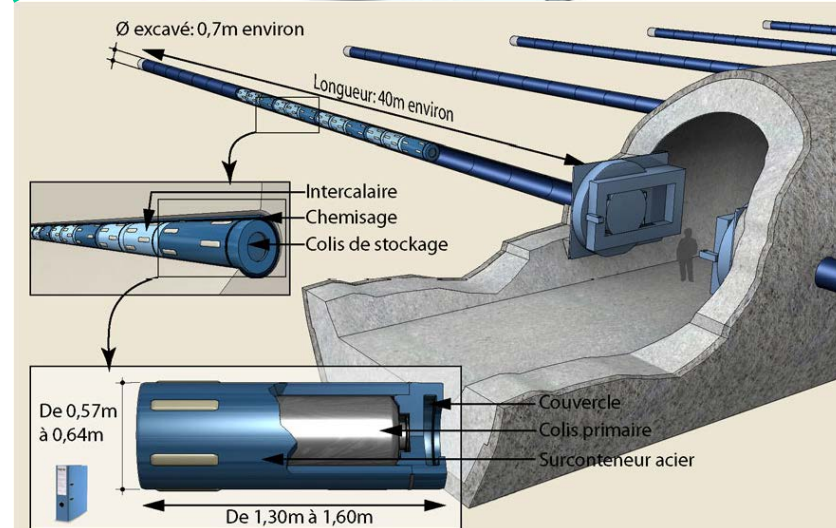


If galleries are properly filled and plugs and seals are tight, $A \gg B$



- ① Pore water migration to waste (gas/water system)
- ② Corrosion: penetration of conteneur after > 1000 yr
- ③ Glass protected from water for > 1000 yr

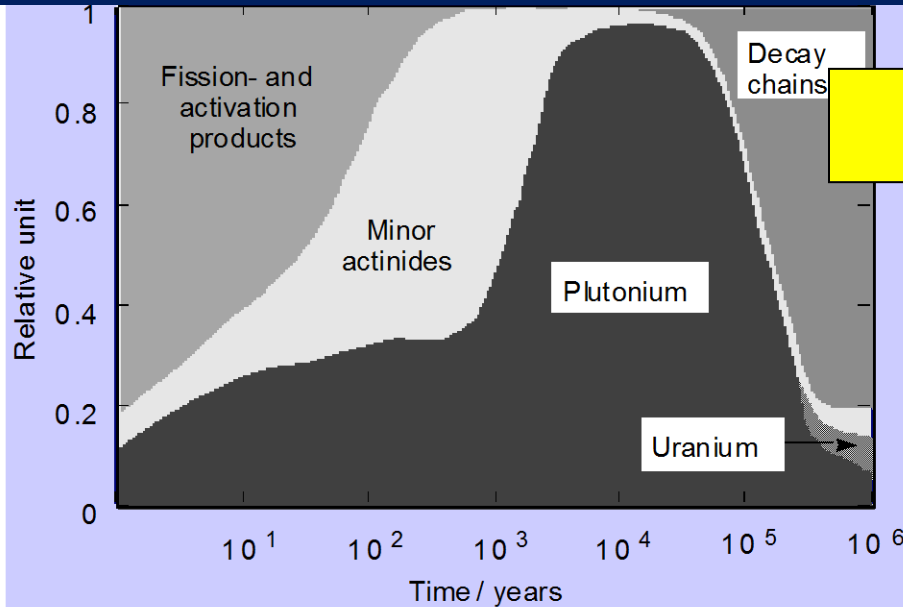
- ④ Complete glass corrosion takes > 200000 yr



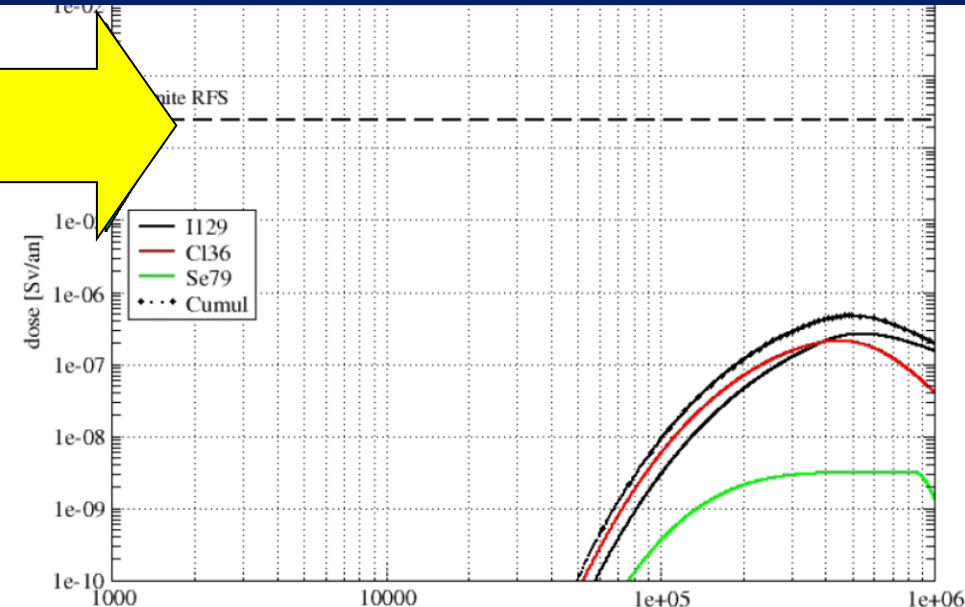
Performance indicators: toxicity, dose, risk not to confuse « radiotoxicity » with « risk » which includes the exposure scenario

Toxicity=hazard potential

risk= potential impact on health in future



spent nuclear fuel 50 MWd/kgU



Nuclear waste glass, Dossier 2005 ANDRA

Safety case

: **Low contribution of actinides to disposal risk**

- Radioactive decay of actinides in some meters from the waste due to weak solubility and strong retention on clay rock
- 100% protection for 50000 years and thereafter only anionic species (I129, Cl36) contribute to potential dose

From safety analyses to the safety case

- ❑ The fact of demonstrating compliance with dose or risk criteria does not mean that all stakeholders share this view on safety
- ❑ Fukushima is a good example for that we cannot only rely on quantitative criteria
- ❑ The safety case includes qualitative safety indicators as base for a dialog with stakeholders. The results of quantitative safety analyses are one indicator among many. Other indicators can be natural analogues, natural tracer profiles etc.

The safety case needs to acknowledge that the scientific approach is limited when predicting quantitative compliance results for very long time

- ☐ Model validation
- ☐ Credibility of models
- ☐ What about extrapolations?
- ☐ Bounding cases
- ☐ Uncertainties in evolution scenarios

Model validation

- How do we know that a model validated for short term lab data is valid for millions of years in the field?
 - According to Oreskes et al. 1994 validation is impossible and data comparison only validates partial
 - Others claim that a simple comparison demonstrates validity (e.g. Burkholder 1979)
 - In context of theory of truth, Nordstrom (2012) suggests to think in terms of usefulness of models
- Example:
 - Our **pore water model** is partial, non unique but useful
 - Non-unique, because other mineral phase assemblages can explain the same data
 - Partial, because many processes and components (trace elements) are not considered
 - Useful as it describes correctly million year old solution composition, its non-trivial evolution in the experiment and pH

Credibility of predictions

- ❑ It is the nature of empirical science that it does not provide absolute truth, but only analyses of different degree of credibility
- ❑ We propose to distinguish between strong and weak predictions
- ❑ Strong predictions (credible):
 - Natural law, deduction, complete induction, interpolation
 - The confidence in science since the time of the greeks and renaissance is based on the ability to predict
 - The standard model of particle physics predicted the Higgs Boson
 - Example for waste disposal:
 - geological records for repository stability, radioactive decay laws for radionuclide inventories in 10^5 yr, Ficks law for predicting diffusion, Darcys law for water flow, Fourriers law for predicting thermal gradients, solubility for predicting maximum radionuclide concentrations
 - Can be used to constrain bounding analyses for complex systems, but does not allow to predict complex system
- ❑ Weak predictions :
 - Analogie, extrapolation, induction

Bounding cases predictions

- ❑ In nuclear waste disposal, as an open natural system, it is neither possible nor necessary to do an exact prediction of radionuclide transfer.
- ❑ It is sufficient to demonstrate that bounding dose/risk criteria are never exceeded
 - Many future evolution may be compatible with such a bounding value
- ❑ Example: actinide migration
 - Solubilities of tetravalent actinides at neutral pH are highly uncertain, yet a maximum solubility values are sufficient for fixation in clay rock
- ❑ Bounding case prediction provide certainty of absence of dose contributions of actinides in reducing clay rock (exception: human intrusion). A very important conclusion since actinides are the most toxic waste components
 - However: actinide migration may be faster in the vicinity of organic MLW (complexation)

The challenge is to maintain credibility in bounding case assessment of a complex disposal system

- ❑ There exists only few people (if any) which have an overall view on the interlinkage of the key processes influencing geological disposal
- ❑ There is no academic but only an engineering view on the overall system
- ❑ Credibility in safety analyses rely strongly on the possibility of simplifying the system by identification of subsystems, that can carry a large part of the burden of proof for safety
 - A thermodynamically stable container (Cu, case Sweden, Finland)
 - A homogene thick clay barrier with a retardation potential demonstrated by natural tracer profiles (case of France, Switzerland, ..)
 - Low solubility and strong retention of actinides (reducing conditions) even if water flow is faster than predicted (all european repository sites)
 - A very stable wasteform (release $<10^{-6}/\text{yr}$)

Safety and society

Safety and risk are not only and not even primary technical quantifiable terms but they are societal ones

Disposal is only feasible if all stakeholders feel more safe with it than without it.

- ☐ The social dimension of scenarios
- ☐ Risk and temporality
- ☐ The social dimension of radiological risk
- ☐ The public debate

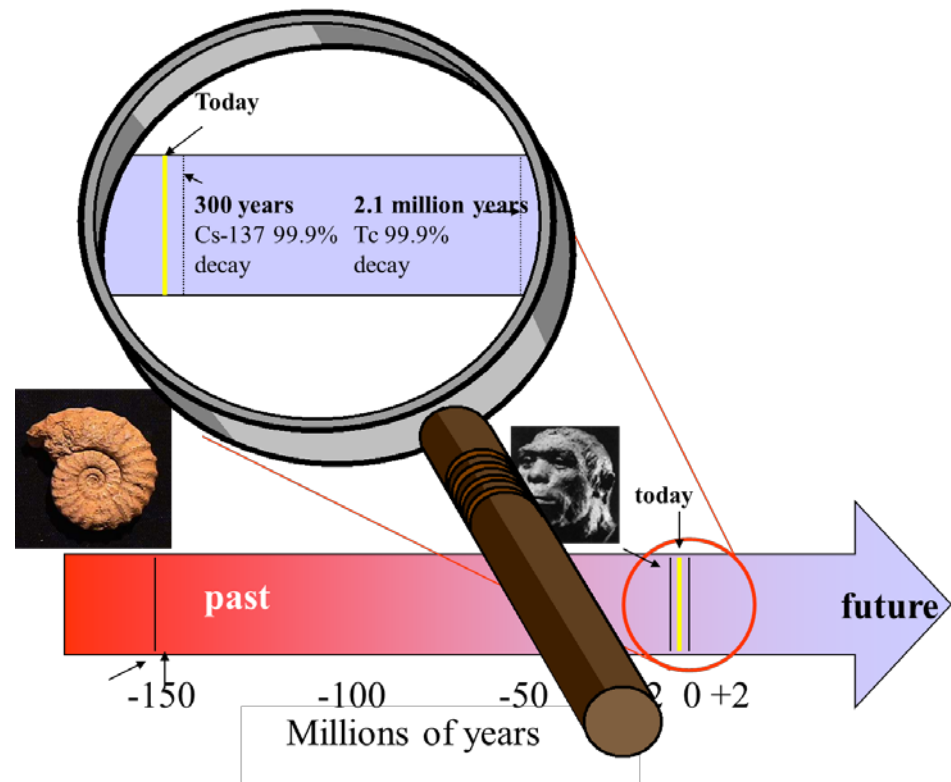
Uncertainties when combining technical and societal evolution scenarios

□ Example: migration of radionuclides to the biosphere

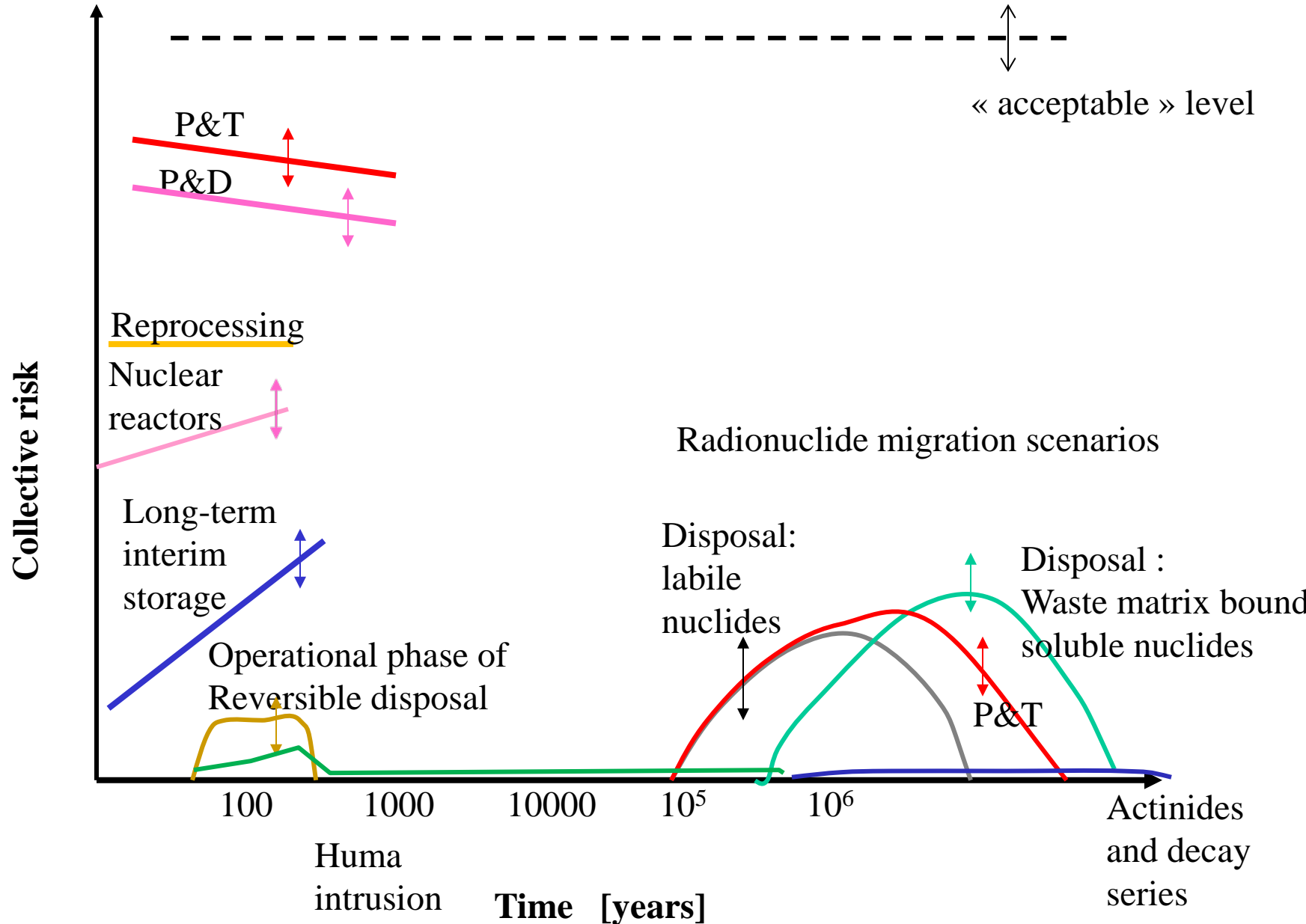
- The long isolation time of the clayrock formation for anions of 200 000 yr are valid only if transport occurs through the clayrock
- This is only the case if plugs and seals are tight and galleries are backfilled as planned
- Technical and societal evolution scenarios may intermingle
 - Earliest closure of the repository is planned some 100 yr after first waste emplacement (assuring reversibility), stepwise closing is under discussion
 - Cost of closing are high: what if a future government in 100 yr decides to leave the repository open, or if it does not decide at all to fill up the galleries ?
 - Then migration path ways may be more rapid through galleries
- Still many tens of thousands of years of isolation are to be expected, but the 200000 yr for iodine fixation may be unrealistic
- Such scenario could be avoided if galleries are closed immediately after waste implacement, but this would violating reversibility provisions

Does it make sense to assess risks for 10^6 yr?

- The projection into the future is constitutive of modern man beyond his own existence
- Although our ability to forecast is limited: our responsibility is not limited and our ability to predict risk is accompanied by our duty to provide protection
- Radioactive waste extend to an extreme the distance between the everyday and the time horizon of our responsibility.



A schematic non quantitative view on relative risks: the dilemma of decision making is the absence of an overall view. Risk analyses are partial and not based on system analyses



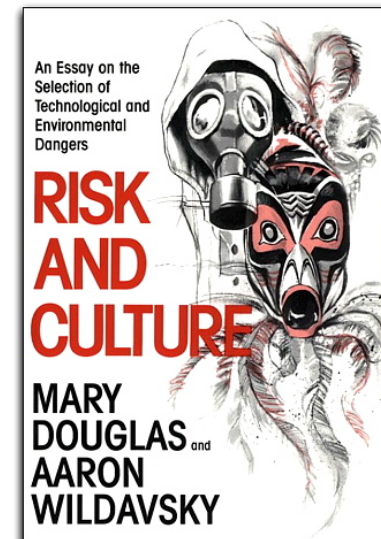
The social dimension of risk of disposal

❑ Radiological risk is not a pure « scientific fact »

- The debate on nuclear waste is an example for risks as social constructions where science is used in conflicting views on the definition of risk (Beck)
- The quantitative radiological risk criteria have poor scientific foundation:
 - « fatality at low dose » validated only at Hiroshima...
 - Example: 100 or 10^6 fatalities in Chernobyl
 - Risk as « average probability of fatality » or « Global burden of disease » (WHO)



only a

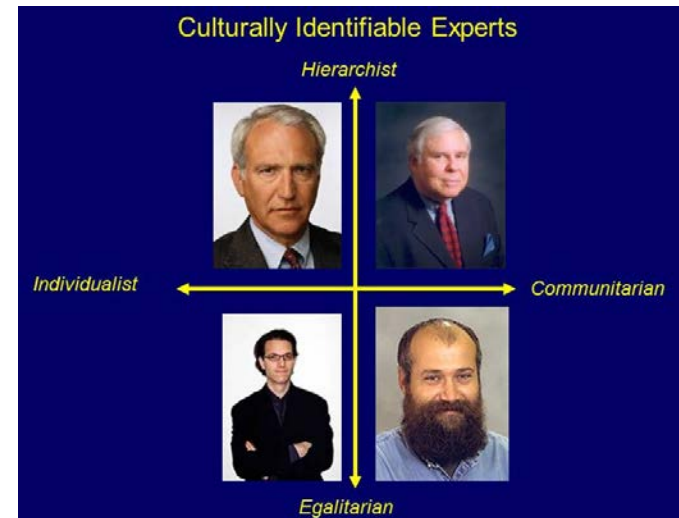


❑ Risk perception

- Even though safety analyses shows that there is small risk, opposition remained strong
- This often is presented as a problem of perception of risk with sharp cleavage between rational experts and general public
- Control over risk is important in risk perception (Fischhoff). Disposal is considered by certaines as loss of control
- Cultural theory: attitudes towards risk are based on different world views on social organization (Douglas, Wildavsky)

Cultural cognition of scientific consensus on risk of nuclear waste disposal (Kahan)

- ❑ Why do members of the public disagree—sharply and persistently—about facts on which expert scientists largely agree?
- ❑ Individuals overestimate the degree of scientific support for positions they are culturally predisposed to accept:
 - Individualistic positions accept the low risk view as an expert view,
 - while communitarian position consider the high risk view as expert opinion.
- ❑ Whether a prediction is credible or not will as well depend on cultural disposition



asn Public debate (organization)

In France, public debate :

- is organized by the national commission for public debate (CNDP);

It was not possible to hold public meetings :

- 12 meetings cancelled;
- 2 meetings stopped after few minutes.

It was then decided to :

- Hold 9 online debate;
- Put in place a “**citizen conference**”.

- ***The “conference of citizens”***

- *Selection of 60 “naïve” citizens representing ages, sexes, education, distance to site by an organism responsible for public polls out of which 17 were disponible*
- *formation of these citizens prepared by a steering committee of proponents and opponents over 2 weekend at a secret place and deliberation in a public meeting*
- *Participation of many high rank representatives of industry, state, civil society...*
- *Formation in antagonistic manner on issues like energy policy, waste inventories and definitions of what is considered waste, risks during construction, operation and long term closure of repository, ethics and role of the media*
- *Citizens gave a final statement like a jury in court but without any institutional role*

- Despite institutional role of the “national commission of public debate, members of parliament were reluctant to participate
- Philosophers refused as well
- A key result: “discussion is possible” but requires respect of opposing positions
 - Too long, the debate is characterized by sterile positions
 - Opponents changed position when they realized that citizens accept opposition but ask for proposition of alternatives
- The agenda of disposal concept was considered too tight
- Suggestion of a prototype disposal:
 - For a small fraction of real high level waste
 - A new decision after some years to continue disposal or to stop and to remove the waste
- ANDRA has included the prototype approach in its new strategie
 - Still, a real repository is necessary, fully equipped to control all risks
 - Well instrumented
 - Potential to continue disposal after a new decision towards a full scale repository
- ASN has changed the agenda to allow for submission of an authorization file in 2017 (instead of 2015) and the prototype approach is included as well

A final remark

- ❑ Scientific analyses clearly indicates that clay rock can isolate nuclear waste for hundreds of thousands to millions of years
- ❑ But there is certainly no direct link between increased scientific understanding and a public position on nuclear waste disposal
 - The voice of the scientist is difficult to hear, not due to a miss-informed public but due to cultural cognition of expertise and historical and cultural perception of hazards to territories destined for disposal,
- ❑ One should avoid cultural relativism, where everything becomes subjective and a social construction, and where an increase of scientific knowledge does not lead to an increase in rationality towards radiological risks.
- ❑ One should not confuse the diminishing credibility of experts with a diminishing role of science.
 - Just the opposite: scientific results become more and more a common object in the conflicting views on the definition of risk (Beck, 2007)
 - However, despite large public interest, scientific results on nuclear waste disposal are still poorly known among non-technical stakeholders in the field of waste disposal.