Matray Jean-Michel - Tremosa Joachim

Non-diagonal transport phenomena in deep disposal facilities: contribution of osmotic processes to the interpretation of the far-field water pressure in the Tournemire argillite

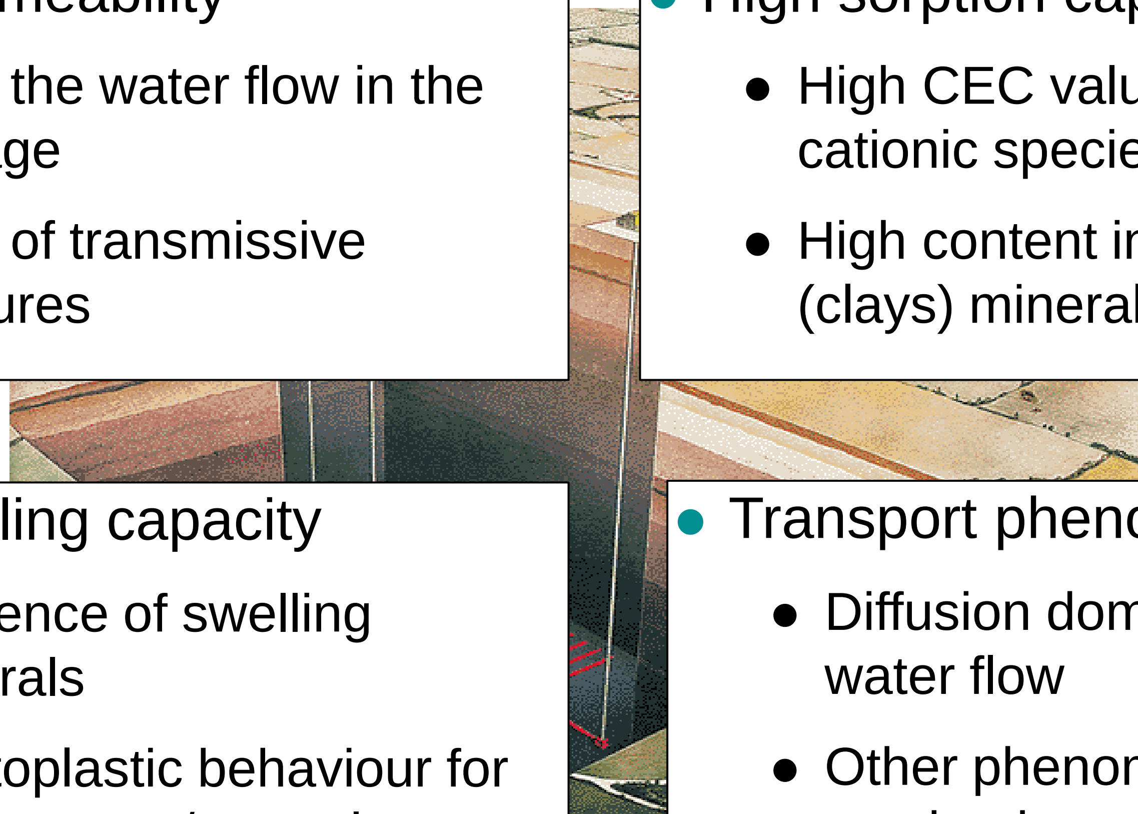
Brussels – 5 & 6 November 2012

Motivation (1)

- RFS III.2.f from 1991 by ASN through sets out objectives for a deep geological disposal:

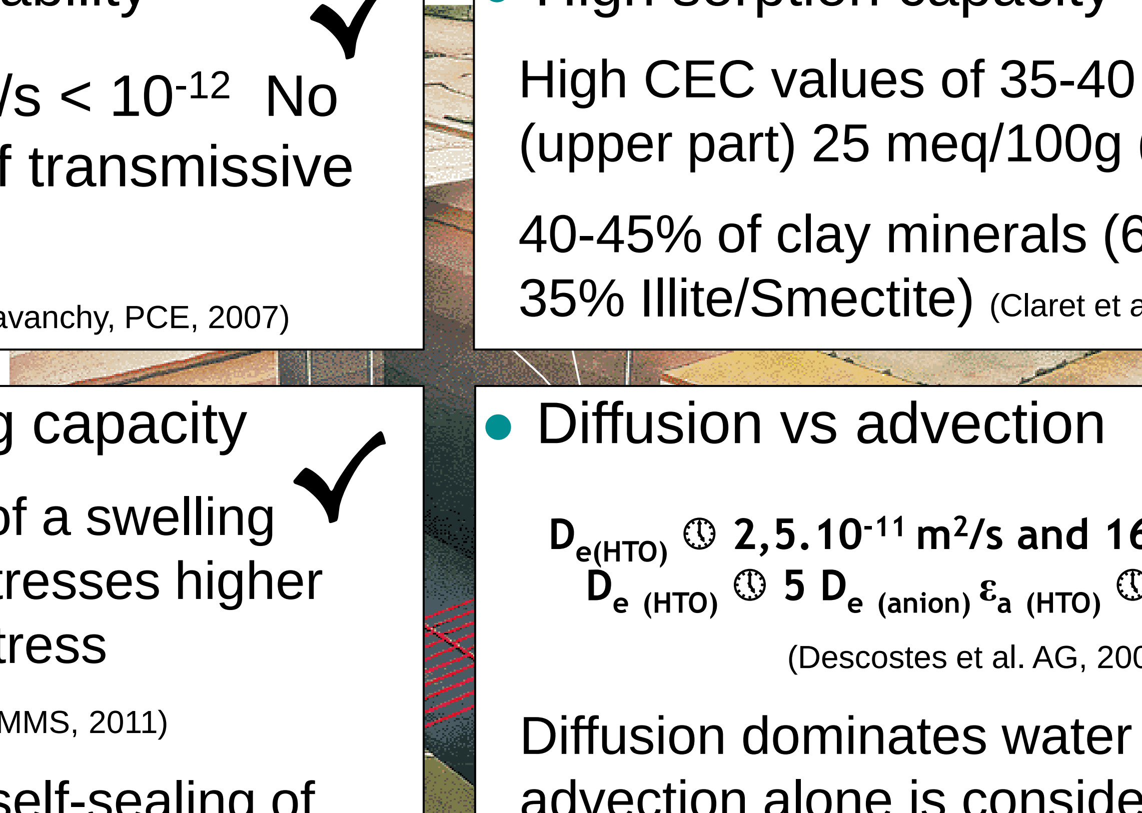
1) lack of long-term seismic risk,
2) absence of significant water flow in storage,
3) rock enabling the excavation of galleries in the facility,
4) confinement properties with respect to radioactive substances,
5) depth sufficient to put the wastes away from aggressions,
6) absence of scarce resources exploitable nearby.

Motivation (2): confinement properties of a clayrock



- Low permeability
  - Limit the water flow in the storage
  - Lack of transmissive fractures
- High sorption capacity
  - High CEC values for cationic species
  - High content in sorbing (clays) minerals
- Self sealing capacity
  - Presence of swelling minerals
  - Elastoplastic behaviour for the fractures/gaps closure
- Transport phenomena
  - Diffusion dominates over water flow
  - Other phenomena are not that important

Motivation (3): confinement properties of the Callovo-Oxfordian argillite



- Low permeability ✓

$10^{-14} < K$ m/s $< 10^{-12}$ No evidence of transmissive fractures

(Distinguin and Lavanchy, PCE, 2007)

- High sorption capacity ✓

High CEC values of 35-40 meq/100g (upper part) 25 meq/100g (lower part)

40-45% of clay minerals (65% illitic 35% Illite/Smectite) (Claret et al, CCM, 2004)

- Self-sealing capacity ✓

Occurrence of a swelling capacity at stresses higher than in situ stress

(Mohajerani et al, IJRMMS, 2011)

Evidence of self-sealing of fractures during resaturation

(TIMODAZ workshop, 2012)

- Diffusion vs advection ✓…

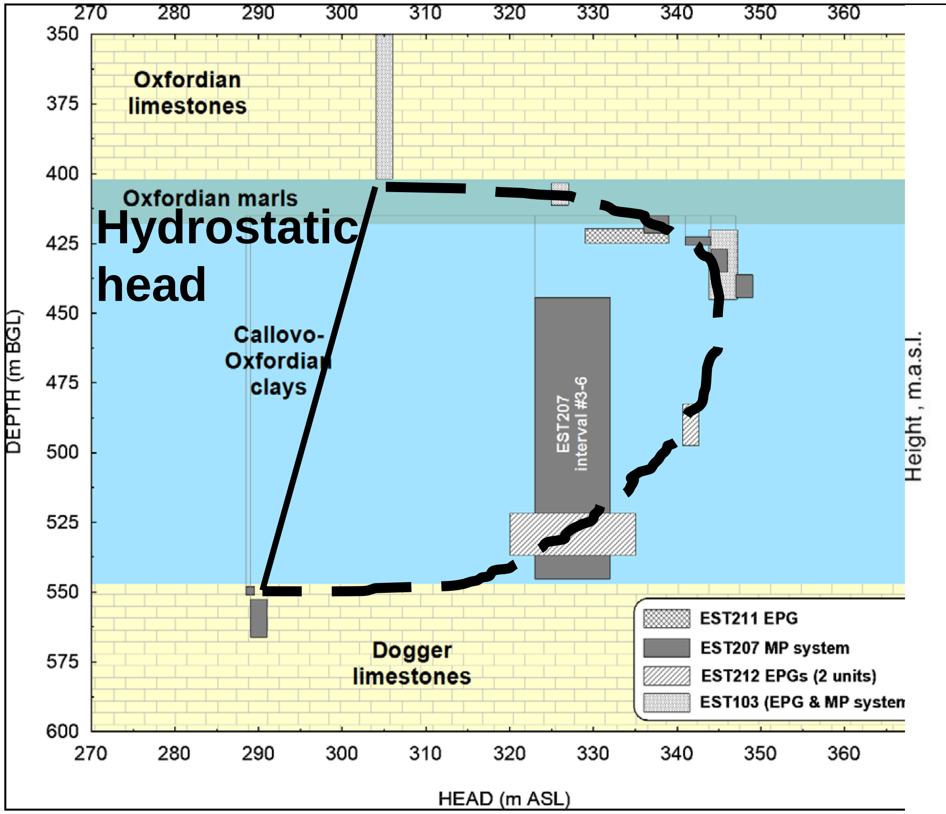
$D_{e(HTO)} \approx 2{,}5.10^{-11}$ m²/s and $16 < \varepsilon_a < 17$ %

$D_{e\ (HTO)} \approx 5\ D_{e\ (anion)}$ $\varepsilon_{a\ (HTO)} \approx 3\ \varepsilon_{a\ (anion)}$

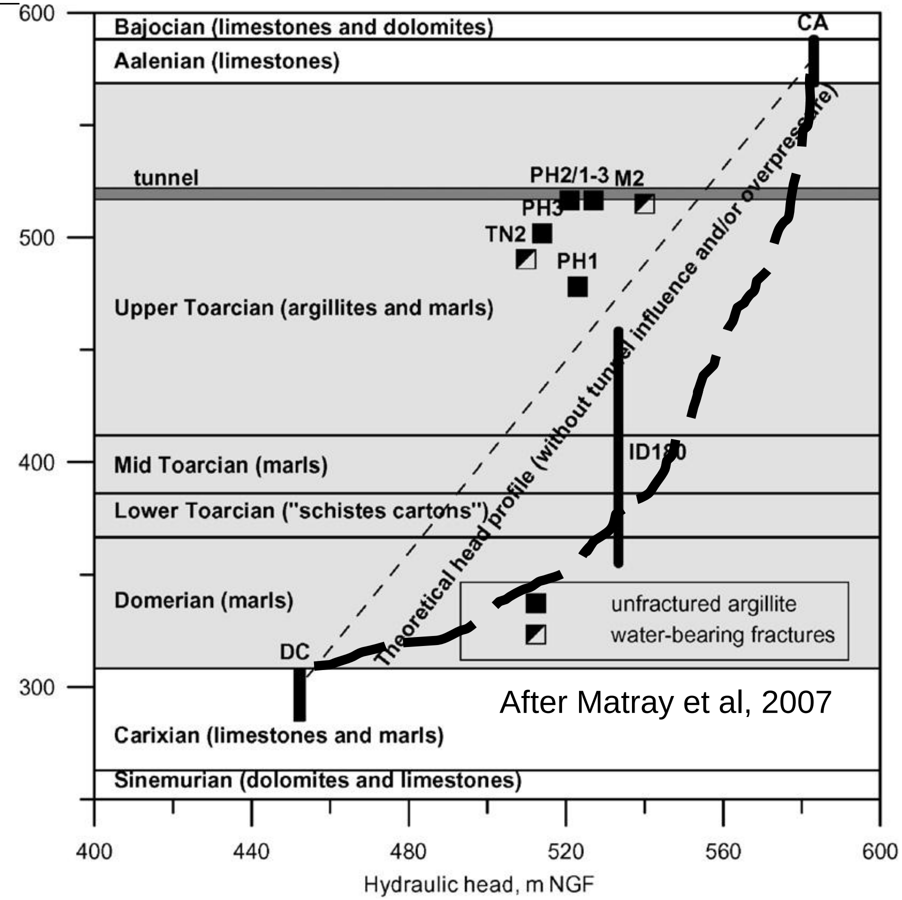
(Descostes et al. AG, 2008)

Diffusion dominates water flow when advection alone is considered (Dossier 2005) but what if other transport phenomena are considered ?

Motivation (4): excess-heads in the Callovo-Oxfordian and the Toarcian/Domerian argillites



After Dossier 2005, Distinguin and Lavanchy, PCE, 2007

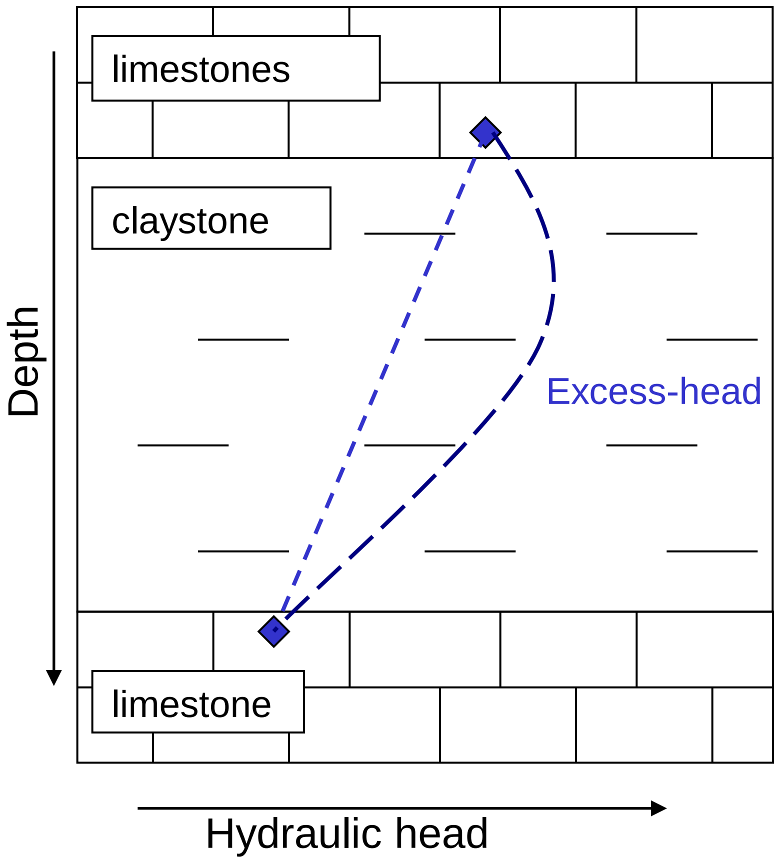


After Matray et al, 2007

~50m of excess head

~30m of excess head

Motivation (5): possible explanations on excess-heads



1. Hydromechanical processes
   - Variation of total stress (compaction disequilibrium, tectonic deformation, …)
   - Visco-plastic behaviour of clays (volumetric creep)
2. Change in hydraulic boundary conditions (erosional decompaction)
3. Diagenetic transformations
4. Osmotic processes
   4. Chemical osmosis
   5. Thermo-osmosis

Motivation (6): Conclusions on Dossier 2005 – Consequences and goals of the study

- Andra → dynamic causes (points 1 & 2) were excluded; chemical osmosis (point 4) alone could account for the observed excess-hydraulic heads;

What are osmotic processes?

Transport phenomena — phenomenological coefficient

$$\frac{1}{V}\frac{dS}{dt} = \sum_i J_i X_i \quad \Rightarrow \quad J_i = \sum_j L_{ij} X_i$$

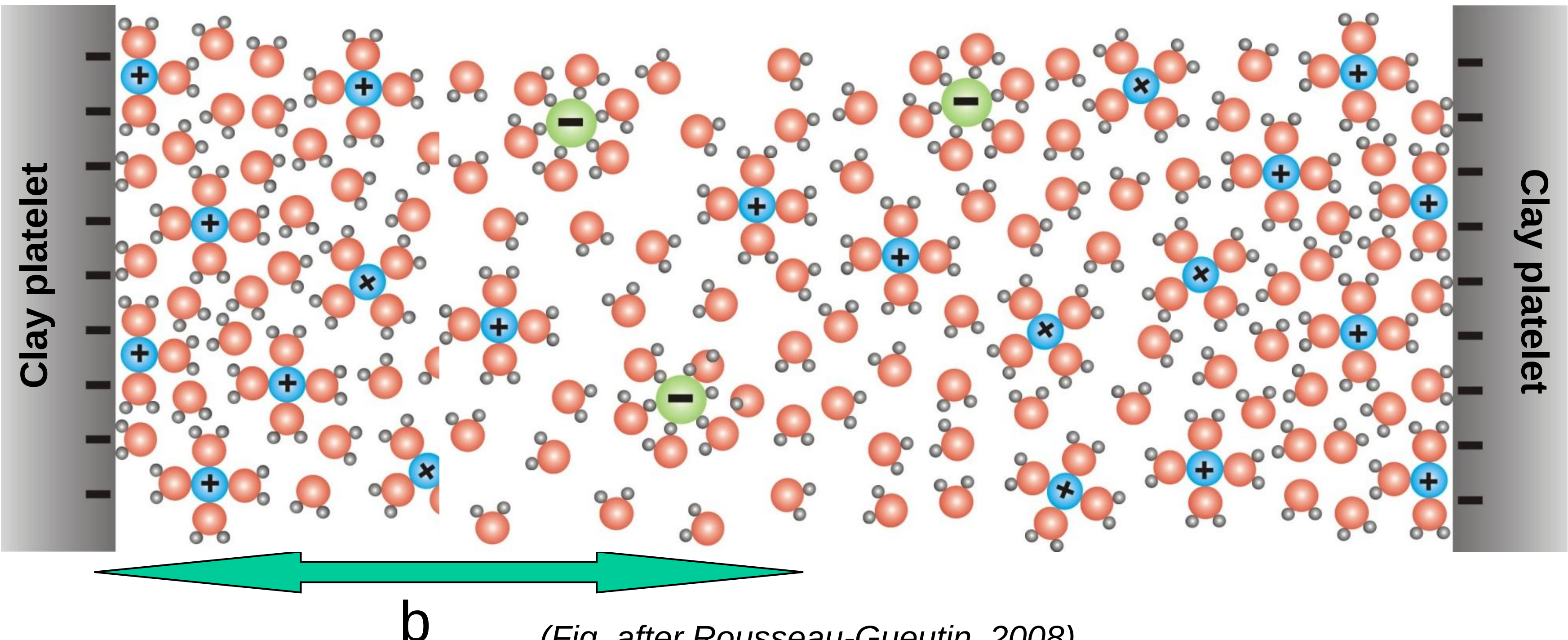
flow (J_i) — phenomenological coefficient (L_{ij}) — force gradient (X_i)

	Gradients X Onsager matrix (1931)			
Flows J	**Temperature**	**Hydraulic head**	**Chemical concentration**	**Electrical**
Heat	Thermal conduction (Fourrier)	Thermal filtration	Dufour effect	Peltier effect
Fluid	Thermo-osmosis	Advection (Darcy)	Chemical-osmosis	Electro-osmosis
Ion	Soret effect	hyperfiltration	Diffusion (Fick)	Electrophoresis
Current	Seebeck effect or Thompson	Rouss effect	Membrane potentiel	Electrical conduction (Ohm)

Fluid flow equation: $$q = -\frac{k}{\eta}\left(\nabla P + \rho g \nabla z\right) + \varepsilon \frac{k}{\eta}\nabla \Pi - \frac{k_T}{\eta}\nabla T + \beta \nabla \psi$$

Why do osmotic flows occur?

- Clays have a negatively charged surface
- Cation accumulation at the clay surface to respect electroneutrality
- Partial exclusion of anions which have access to a lower porosity
- semipermeable membrane behaviour: restricts the passage of some elements, due to their size or electrical size



(Fig. after Rousseau-Gueutin, 2008)

Motivation (6): Conclusions on Dossier 2005 – Consequences and goals of the study

- Andra ➔ dynamic causes (points 1 & 2) were excluded; chemical osmosis (point 4) alone could account for the observed excess-hydraulic heads;

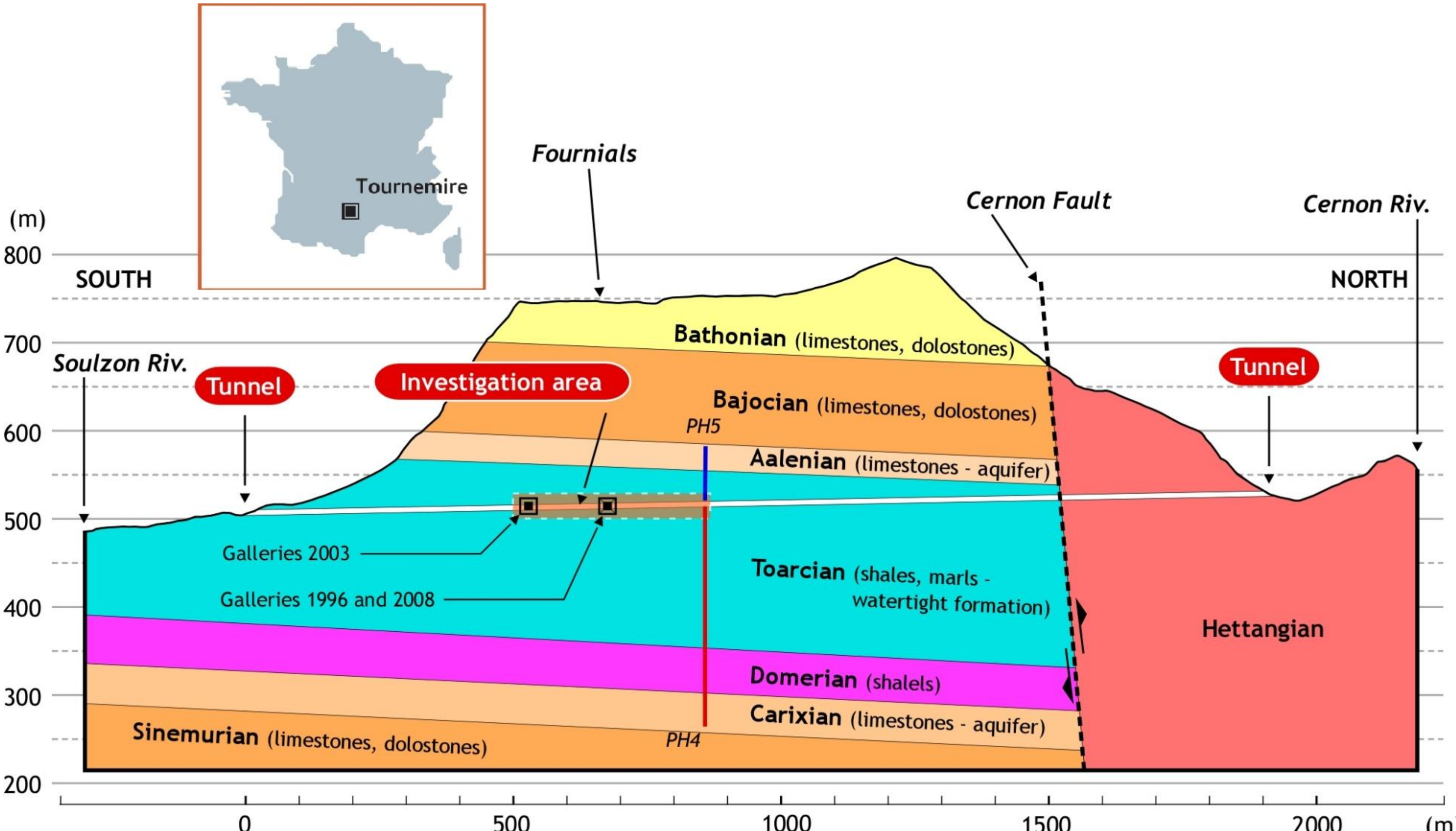
- IRSN ➔ dynamic causes need to be properly estimated; no conclusion can be drawn on osmotic processes due to unsolved scientific issues.

- IRSN and Andra started 2 separate PhD work on their URL:
  - Can we assess the actual contribution of Chemical osmosis and other transport phenomena to the flow?
  - Can coupled transport phenomena alone explain the measured excess hydraulic head in a clayrock?
  - Is Diffusion the dominant transport phenomenon?

Application to the Tournemire case study: Outcome

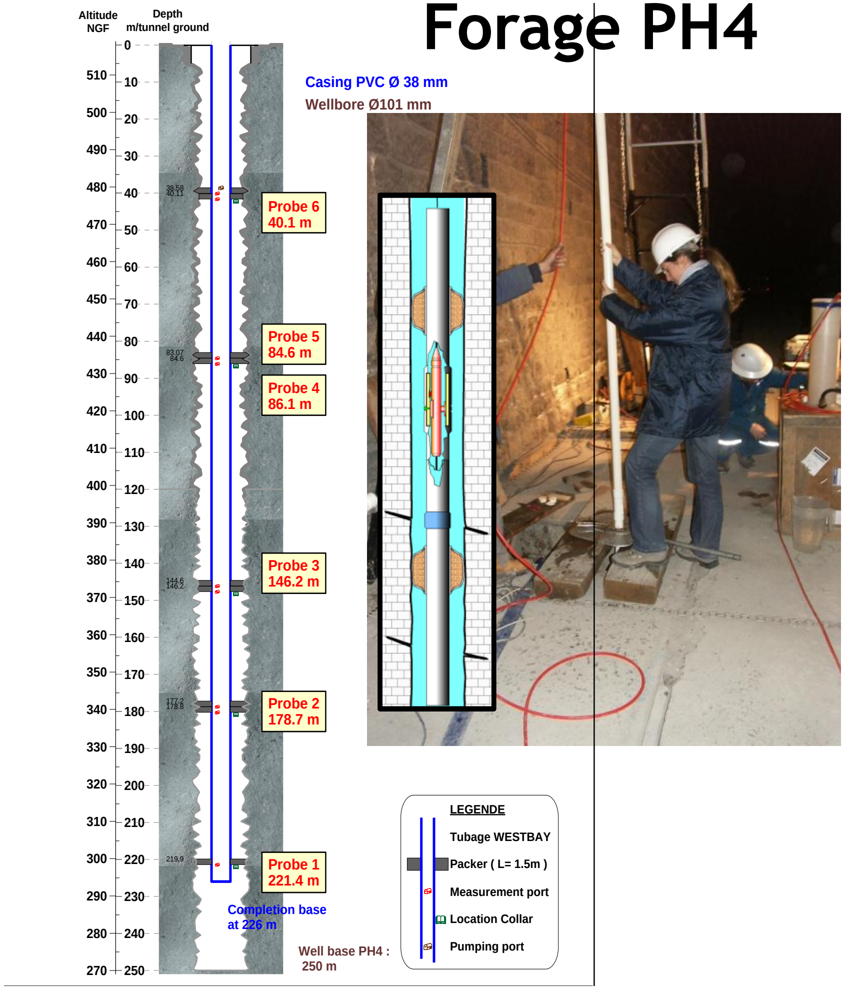
- I- Acquisition of force gradients (P/T/C)
- II- Acquisition of phenomenological parameters ($\boldsymbol{k}/\varepsilon/\boldsymbol{k_T}$)
- III - Modelling the head profile across the clayrock
- IV- Conclusions

I. Acquisition of force gradients: Realization of 2 boreholes

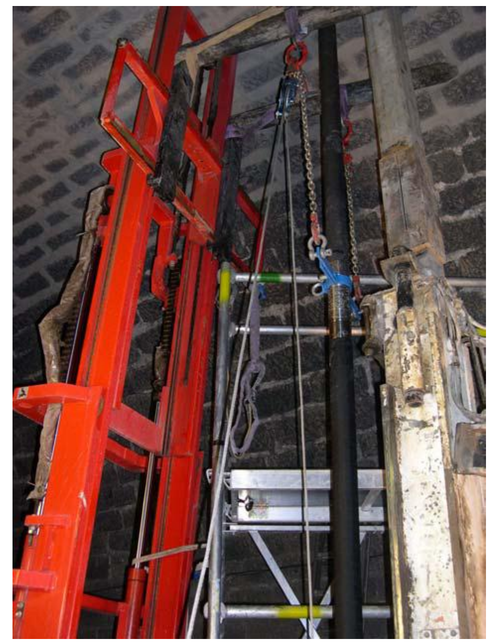


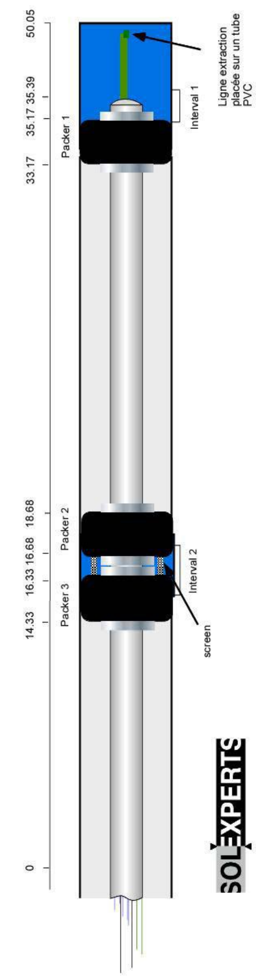
I. Acquisition of force gradients: Installation of hydraulic devices for P/T profiles

Forage PH4



Forage PH5





I. Acquisition of force gradients: Core sampling and analysis

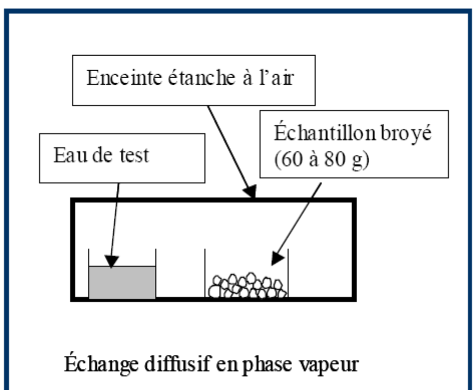
Core mapping

Petrophysical measurements

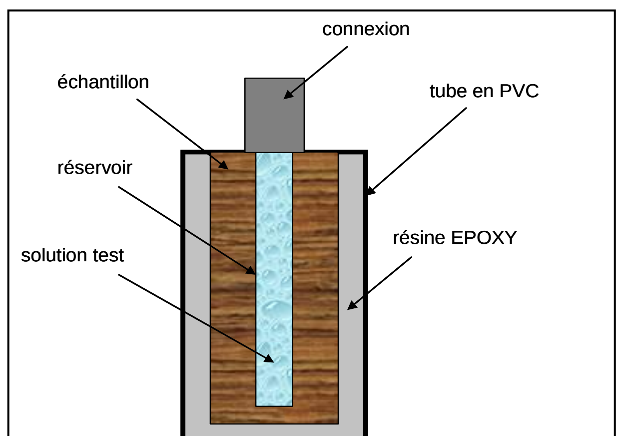
140	TOARCIEN INFERIEUR (SCHISTES CARTONS)		NG	AM	MP		DRA	MP+PV	DRB	MP	TD		ANR	MO	IBA			
141			NG	AM	MP		double	MP+PV	double	MP	double		standard	MO	IBA			
142		STD																
143		STD																
144		STD																
145			NG	AM	MP		DRA	MP+PV	DRB	MP	TD			MO	IBA			MGM
146			NG	AM	MP		double	MP+PV	double	MP	double			MO	IBA			MGM
147		STD																
148		STD														PCO2		
149		STD																
150			NG	AM	MP	CLIS	DRA	MP+PV	DRB	MP	TD		ANR	MO	IBA			
151			NG	AM	MP	CLIS	double	MP+PV	double	MP	double		standard	MO	IBA			
152		STD																
153		STD																
154			NG	AM	MP		DRA	MP+PV	DRB	MP	TD			MO	IBA			MGM
155			NG	AM	MP		double	MP+PV	double	MP	double			MO	IBA			MGM
156		STD															EOA/EOB	
157		STD																
158			NG	AM	MP		DRA	MP+PV	DRB	MP	TD		ANR	MO	IBA			
159			NG	AM	MP		double	MP+PV	double	MP	double		standard	MO	IBA			

Example of sampling sequence

Core preservation



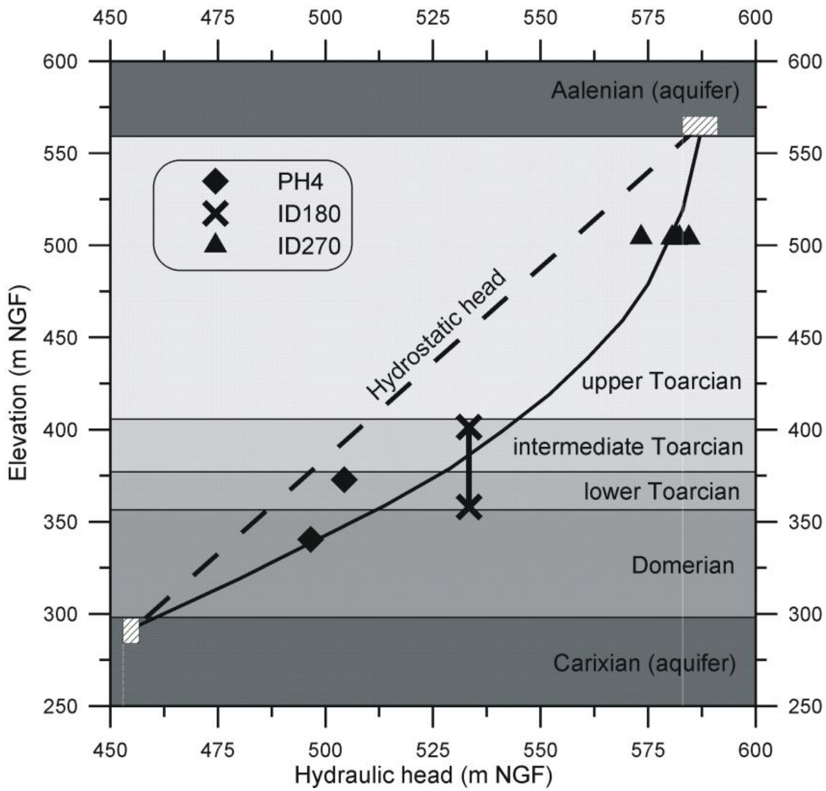
Through diffusion cell



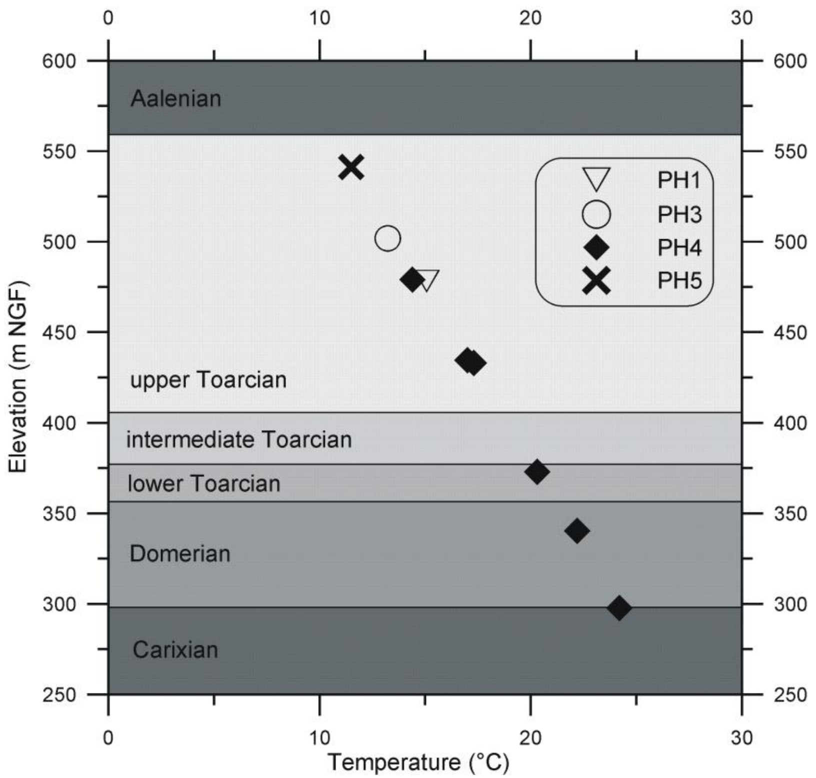
Radial diffusion cell

Degassing cells

I. Acquisition of force gradients:
Head $(\nabla P + \rho_f g \nabla z)$, Temperature (∇T)



Excess-head of ca 30m verified



Temperature gradient 5.2°C/100m

I. Acquisition of the Chemical gradient($\nabla\Pi$): geochemical Interaction model

(Tremosa et al, 2012)

Exchangeable cations

Mineralogy and Petrology

Interaction model

→ equilibrium with mineral phases

→ multisite cation-exchange model

- mineralogy dependent (illite,I/S)
- 3 sites for illite, 2 with strong affinity *(Bradbury et B., 2000)*
- 1 site for smectite (Tournassat et al, 2009)

Mobile anions and porosity *(Bensenouci, 2010)*

Equilibrium with carbonates (pCO_2)

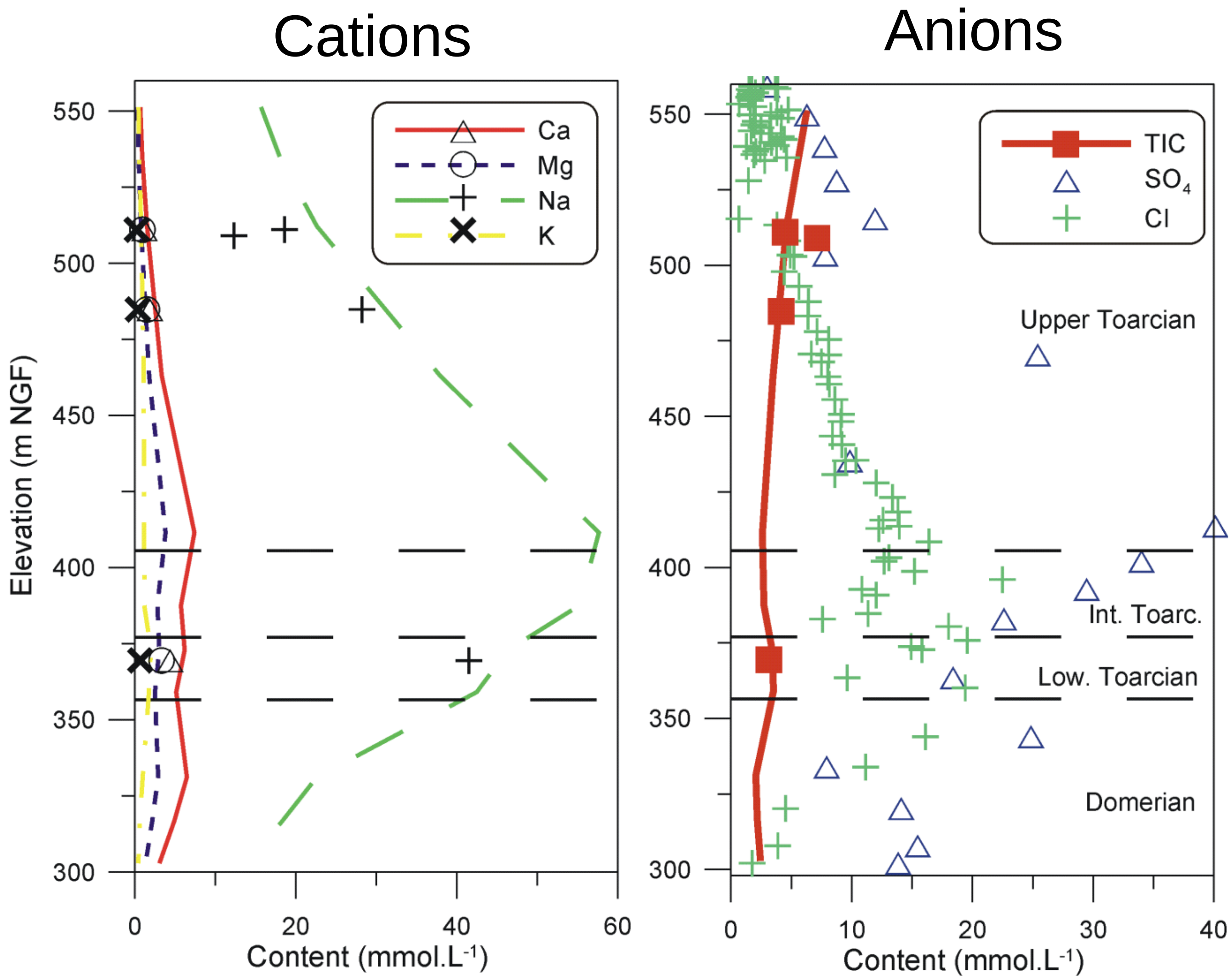
Composition of water in equilibrium with the clayrock

Profile of the water chemical composition through the claystone

Comparison And validation

Corrected fracture waters *(Beaucaire et al., 2008)*

I. Acquisition of the chemical gradient ($\nabla\Pi$): measured vs modelled composition



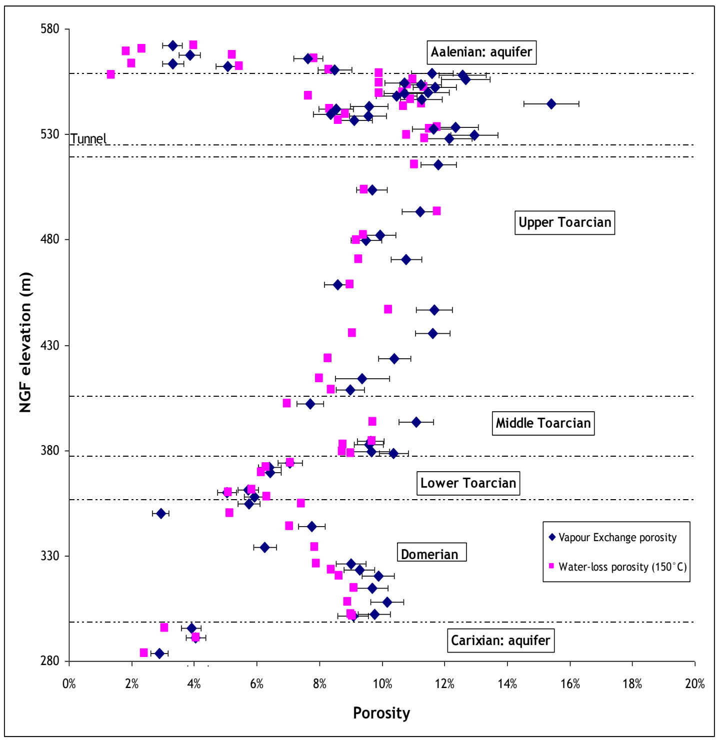
II. Acquisition of phenomenological parameters: intrinsic permeability *k*

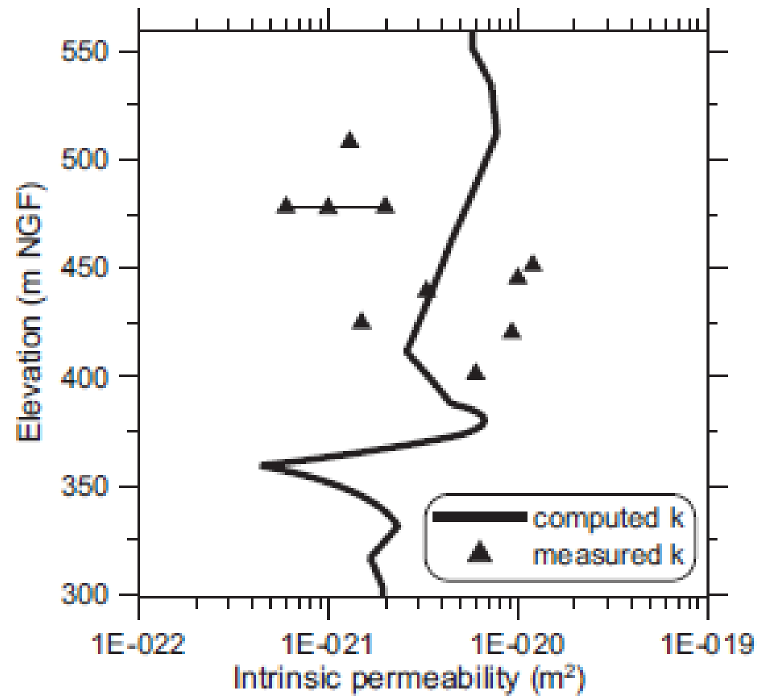
- Advective flow

$$q = -\frac{k}{\eta}\left(\nabla P + \rho_f g \nabla z\right)$$

- Poiseuille type law

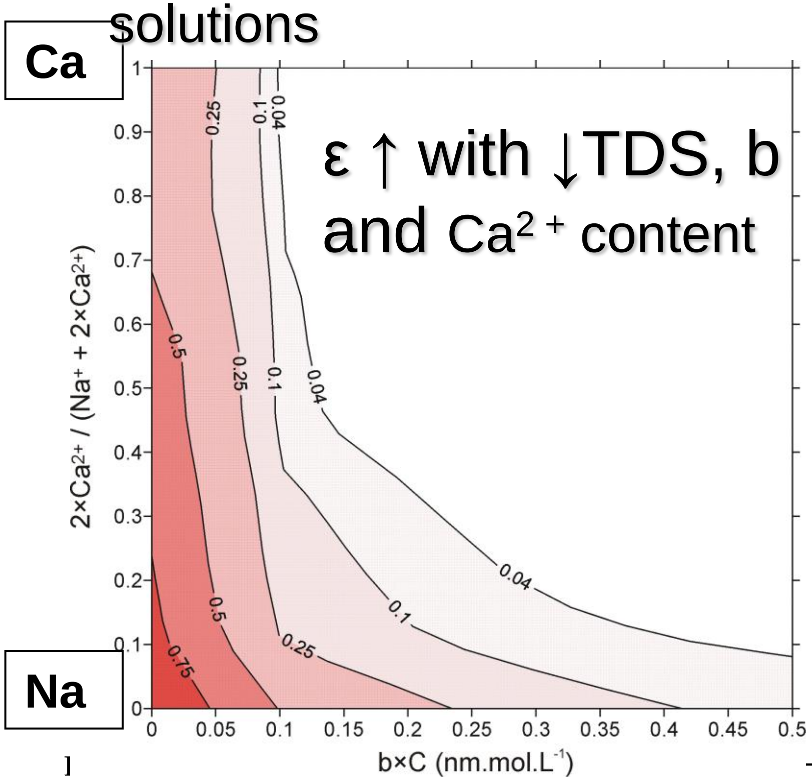
$$k = \frac{b^2}{3\omega^{-m}} \qquad b = \frac{\omega}{\rho_s A_s (1-\omega)}$$

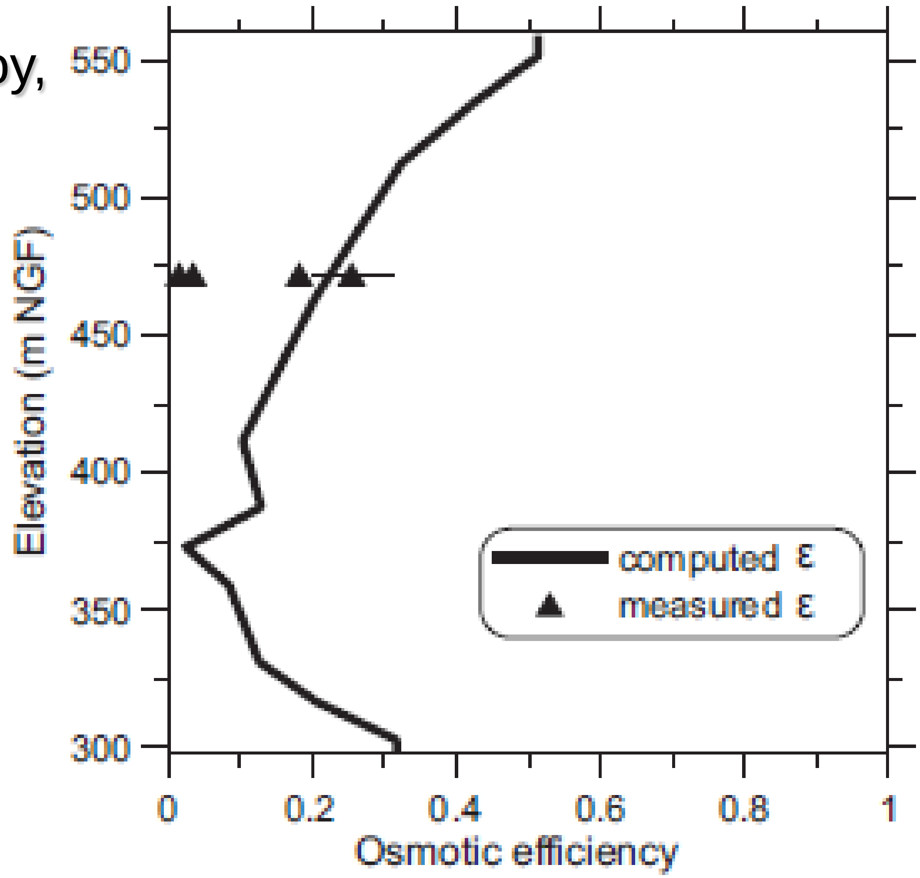




II. Acquisition of phenomenological parameters: osmotic efficiency ε

- Chemo-osmotic flow $q = +\varepsilon \frac{k}{\eta} \nabla \Pi$
- Bolt model 1979 $\varepsilon = 1 - \frac{\langle C - \rangle}{C^f}$
- Triple layer Model (Revil and Leroy, 2004) modified by Tremosa et al (2012) to account for multi-ionic solutions





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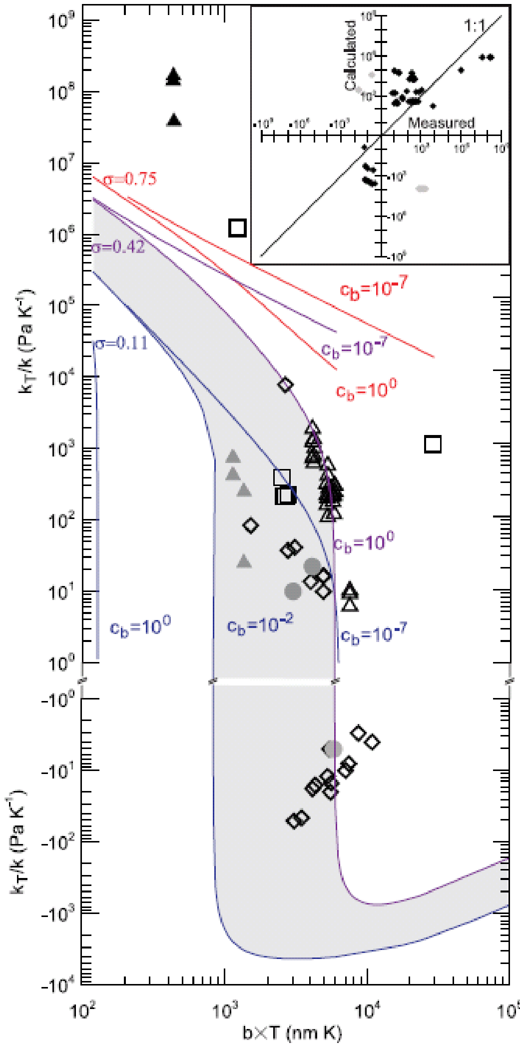
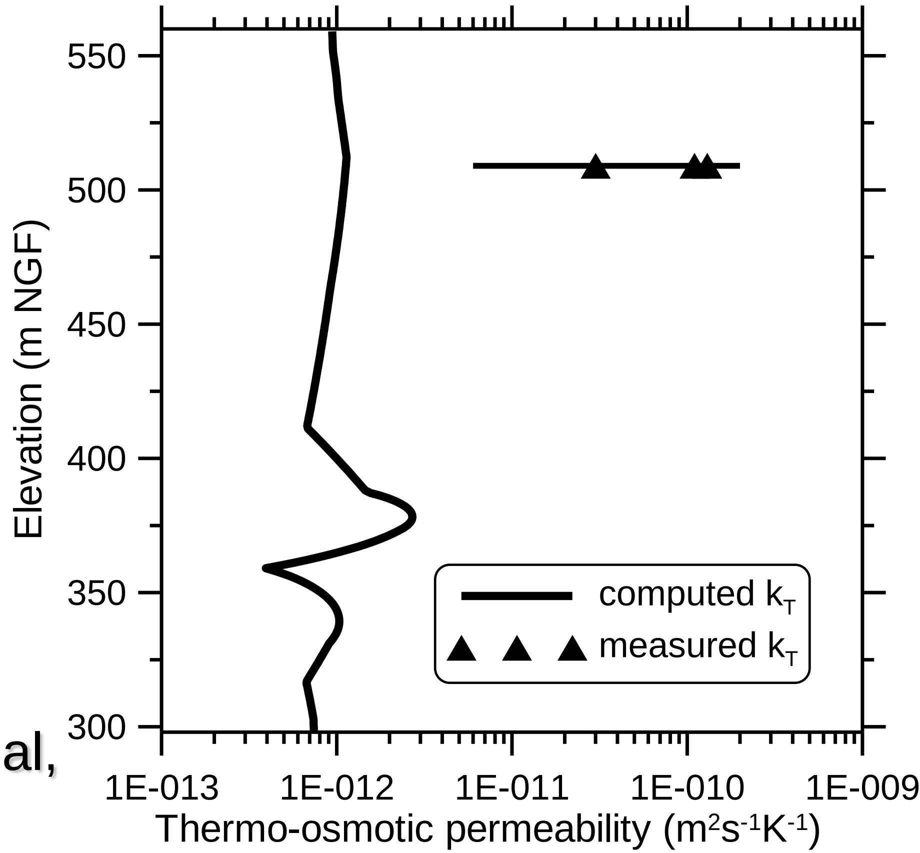
II. Acquisition of phenomenological parameters: thermo-osmotic coefficient k_T

- Thermo-osmotic flow $q = -\frac{k}{\eta}\frac{\Delta H}{T}\nabla T$



ΔH the enthalpy change is due to an alteration of the hydrogen bonds HB at the clay surface.



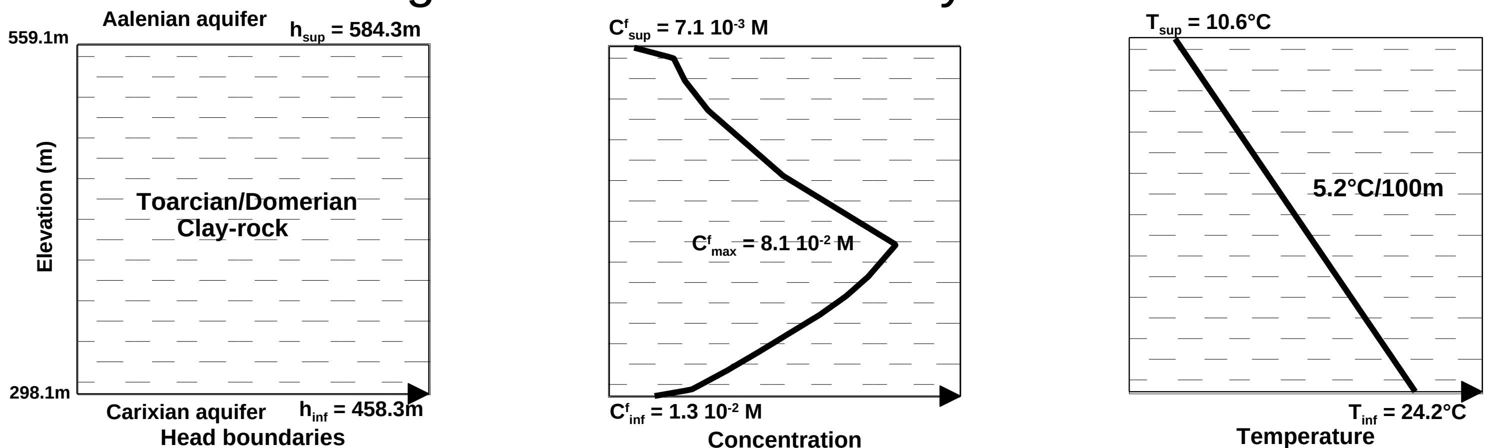
- Gonçalvès et al, 2012) model

$$\Delta H = (C_{HB}^{b} - C_{HB})\Delta H_{HB}$$

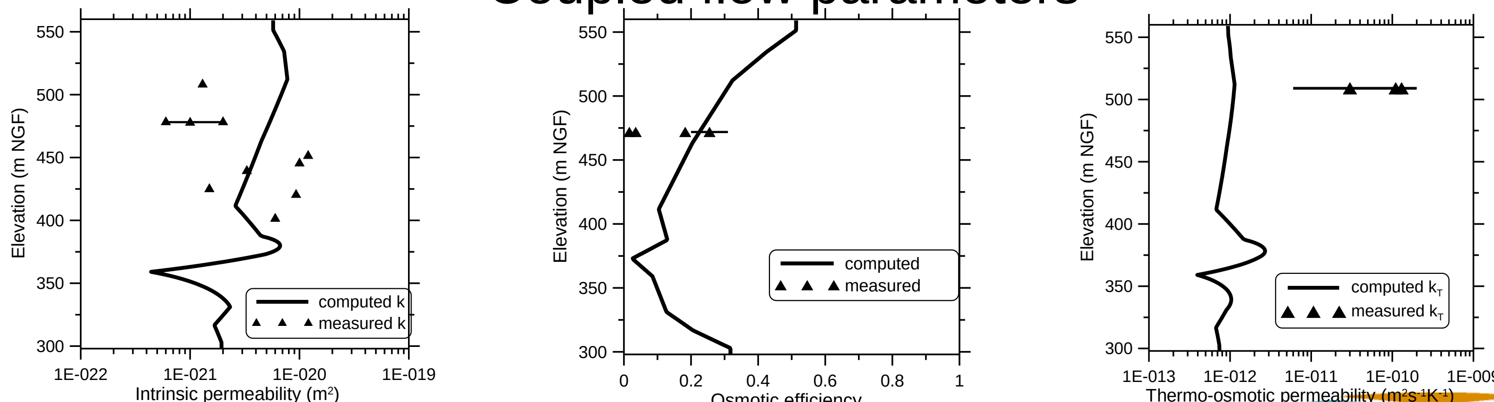
III - Modelling the head profile through the clayrock: concept and input parameters

$$\frac{\partial}{\partial z}\left(\rho_f \frac{k}{\eta}\left(\nabla P + \rho_f g \nabla z\right) - \rho_f \varepsilon \frac{k}{\eta}\nabla \Pi + \frac{k}{\eta}\frac{\Delta H}{T}\ \nabla \mathrm{T}\right) = 0$$

Force gradients and boundary conditions

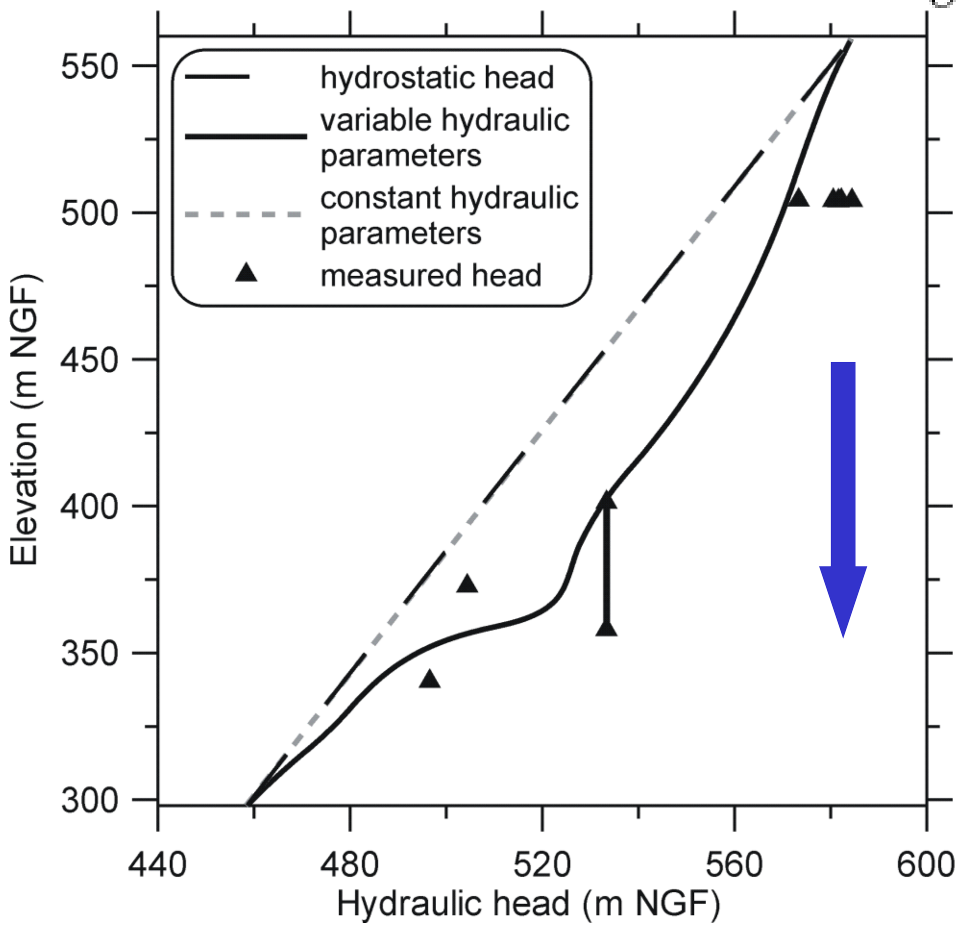


Coupled flow parameters



III - Modelling the head profile through the clayrock: Results for a pure hydraulic flow

$$\frac{\partial}{\partial z}\left(\rho_f \frac{k}{\eta}(\nabla P + \rho_f g \nabla z)\right) = 0.$$



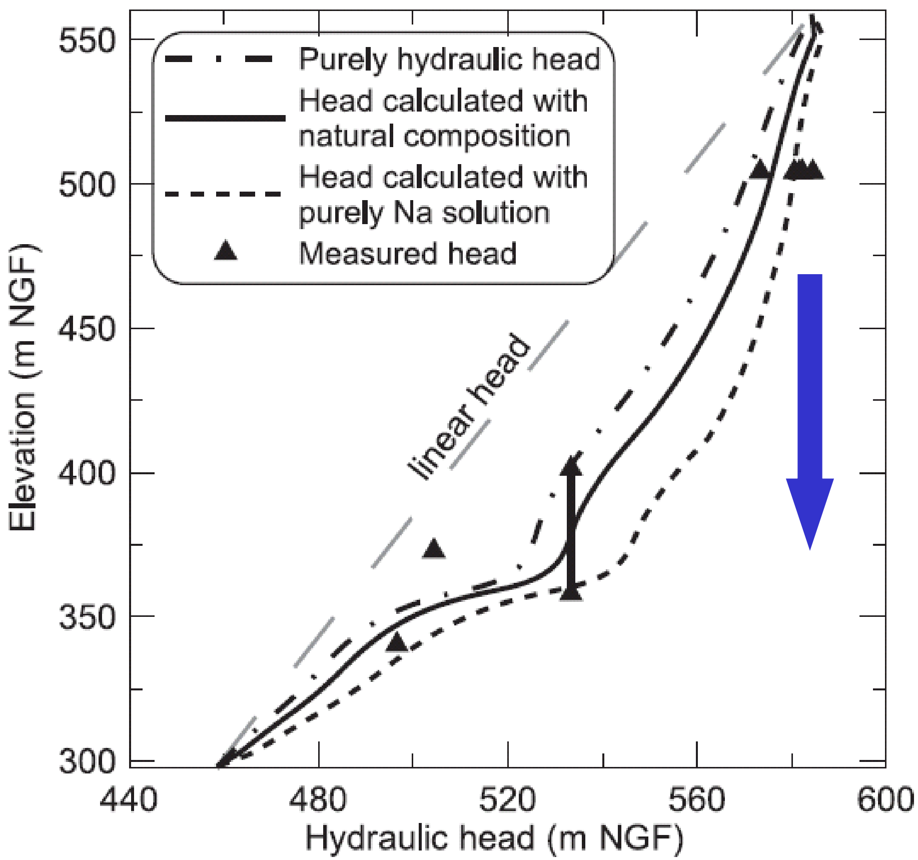
a. The excess head is partly explained by permeability variations

b. Downward flow

$$q = -1.3 \times 10^{-14}\, m.s^{-1}$$

III - Modelling the head profile through the clayrock: Results for a coupled chemo-osmotic and Darcy flow

$$\frac{\partial}{\partial z}\left(\rho_f \frac{k}{\eta}(\nabla P + \rho_f g \nabla z) - \rho_f \varepsilon(b, C)\frac{k}{\eta}\nabla \Pi\right) = 0.$$



a. **Fairly good agreement with the natural composition simulation except in the lower Toarcian**

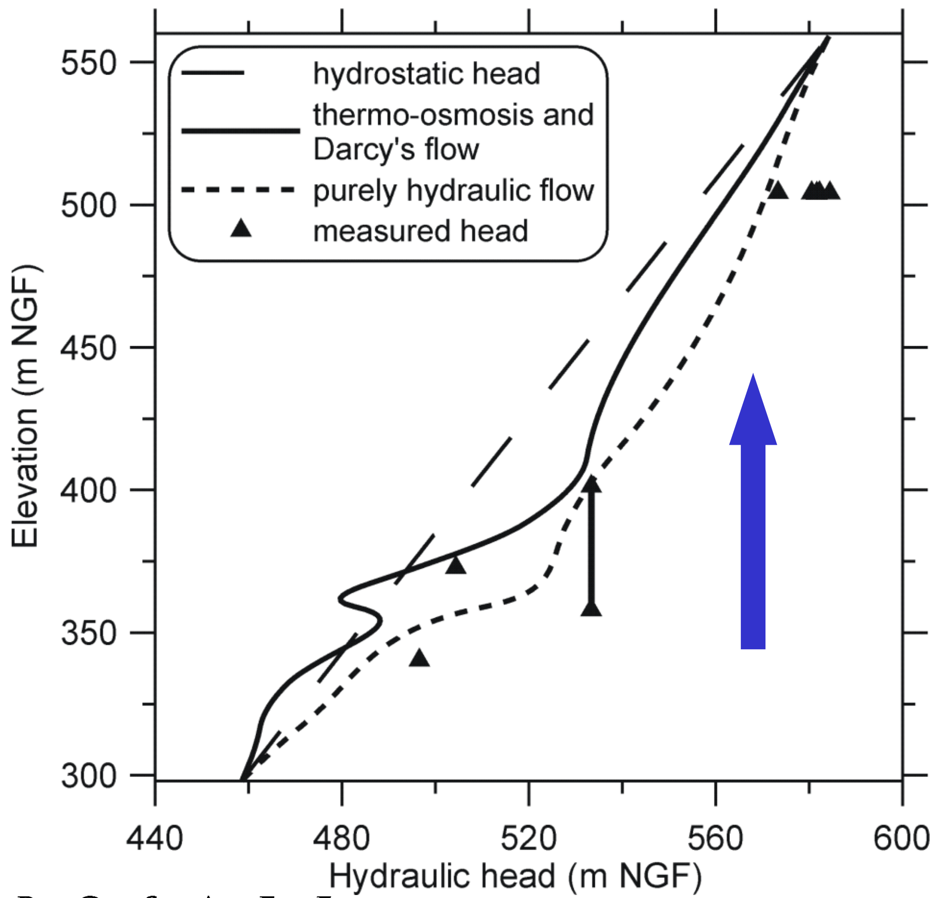
b. **Pure NaCl solution overestimate the head especially in the lower Toarcian/Domerian**

c. **Downward flow**

$$q = -1.4 \times 10^{-14}\, m.s^{-1}$$

III - Modelling the head profile through the clayrock: Results for coupled thermo-osmosis and Darcy flows

$$\frac{\partial}{\partial z}\left(\rho_f \frac{k}{\eta}(\nabla P + \rho_f g \nabla z) + \frac{k}{\eta}\frac{\Delta H}{T}\nabla T\right) = 0.$$

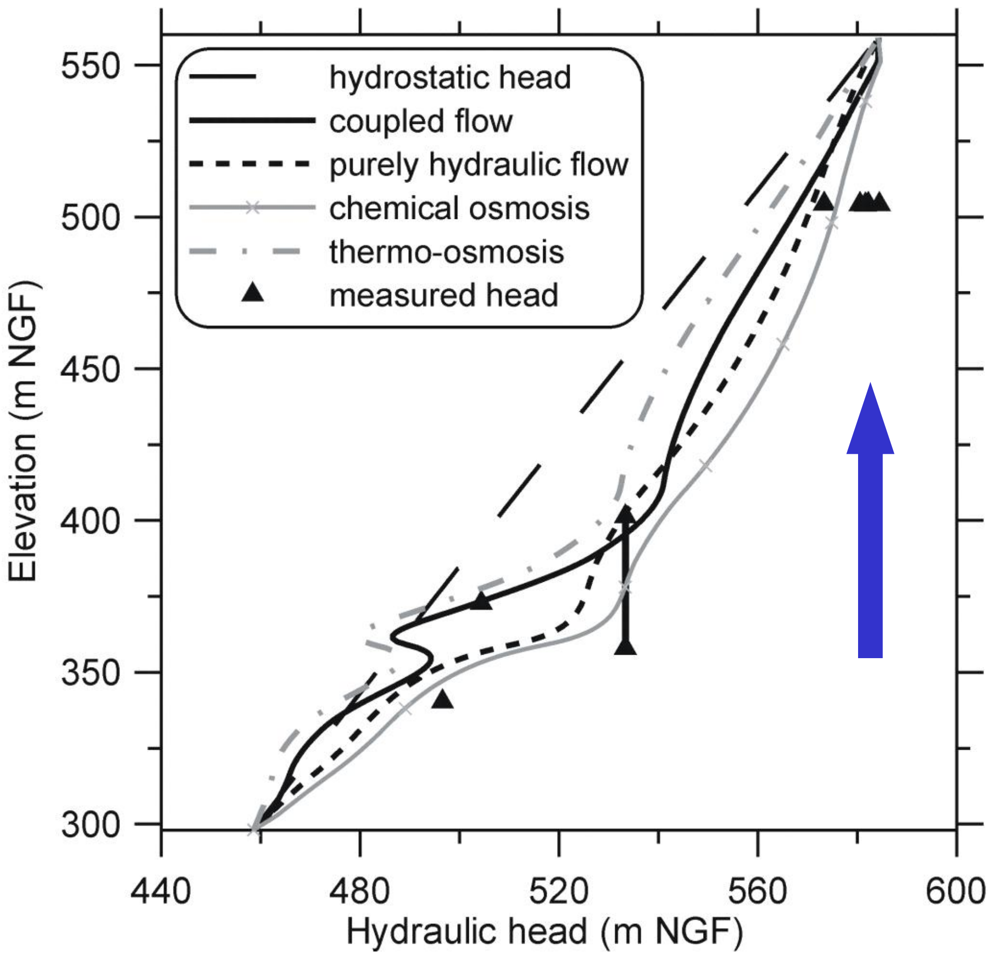


a. **Only explain the head in the lower Toarcian**

b. **Upward flow**

$$q = 3.3 \times 10^{-14}\ m.s^{-1}$$

III - Modelling the head profile through the clayrock: Results for all coupled flows



a. **Slight underestimation of the heads in the upper part and good fit in the lower part➔ the coupled flows mostly explain the head profile in the clayrock**

b. **Upward flow**

$$q = 3.2 \times 10^{-14}\, m.s^{-1}$$

III - Modelling the head profile through the clayrock: Diffusion vs Advection

- The Peclet number Pe= $\frac{\tau_D}{\tau_a}$ is the ratio of characteristic times of Diffusion to Advection (Diffusion dominates when Pe< 1).

Where $\tau_D = \frac{L^2}{D_p}$, $\tau_a = \frac{L}{u}$ with $u = q/\omega_k$ $Pe = \frac{Lq}{D_p \omega_k}$

For Cl^- : $D_e = 4\ 10^{-12}\ m^2\ s^{-1}$ *and* ω_k=6.6%, L = 125m and q determined previously, the Peclet number is:

0.27 for a pure Darcy flow

0.3 for darcy flow coupled to chemical osmosis

0.7 for all coupled flows

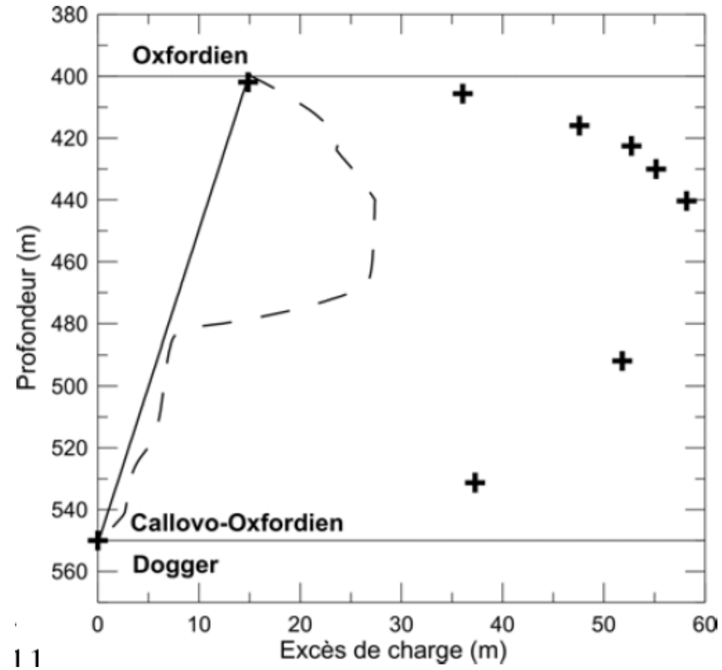
Peclet numbers ar sligthly lower than 1 indicating that diffusion still dominates the transport of anions

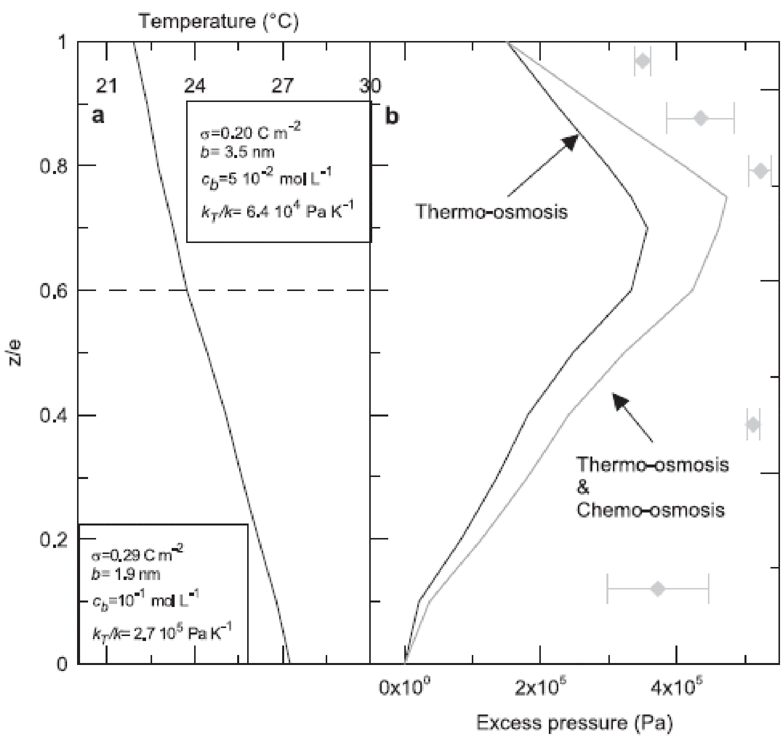
IV. Main conclusions on coupled flows at the Tournemire URL

- The measured head profile has confirmed the occurrence of an excess-head in the Toarcian/Domerain clay rock of about 30m.
- The force gradients and the coupled flows parameters were acquired and enabled us a full modelling approach by coupling a pure Darcy flow, to chemical osmosis and thermo-osmosis.
- Results indicate that the full coupled flows mostly explain the excess-head measured in the clayrock. They also show the role played by thermo-osmosis which inverse the flow direction which turns upward.
- Coupled flows including chemo-thermo osmosis can contribute to the convective transport of dissolved species even though the the Peclet numbers are still slightly in favour of a dominant diffusive regime.

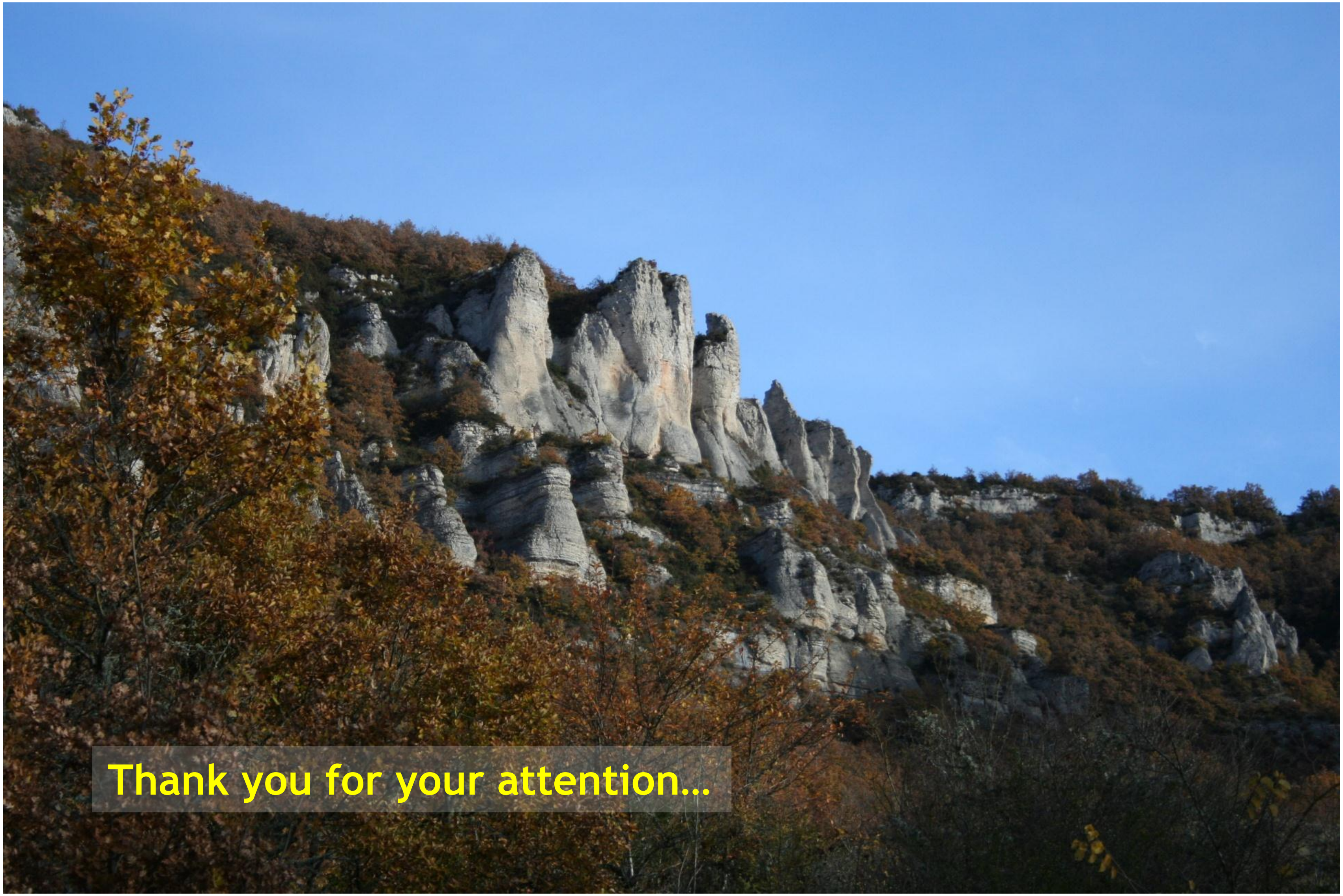
IV. Main conclusions for the Callovo-Oxfordian at Bure

- Rousseau-Geutin (2008) The coupled flows (Darcy + Chemical osmosis) only explain an excess-head of about 18m over the 50m ➔ conclusions of Andra given on Dossier 2005 were false.





- Gonçalvès et al (2012) by coupling all flows calculated an excess-head close to the measured one in the upper part of the Callovo-Oxfordian but could not explain the lower part. What about dynamic causes?


Thank you for your attention…

Influence des effets hydromécaniques sur le profil de charge

$$\frac{\partial}{\partial z}\left(\rho_f \frac{k}{\eta}(\nabla P + \rho_f g \nabla z)\right) = \frac{S_s}{g}\frac{\partial P}{\partial t} - \rho_f \alpha \frac{\partial \sigma}{\partial t} - \rho_f \frac{\sigma}{\eta_s}.$$

Variation de la contrainte totale

a. Déséquilibre de compaction : bassin ancien

b. Compression tectonique latérale : cause peu probable

Comportement visco-plastique des argiles

Effet très limité sur le profil de charge