Migration of contaminants through the unsaturated zone overlying the Hesbaye aquifer in Belgium: Field investigation and modelling

S. Brouyère – A. Dassargues
Serge.Brouyere@ulg.ac.be

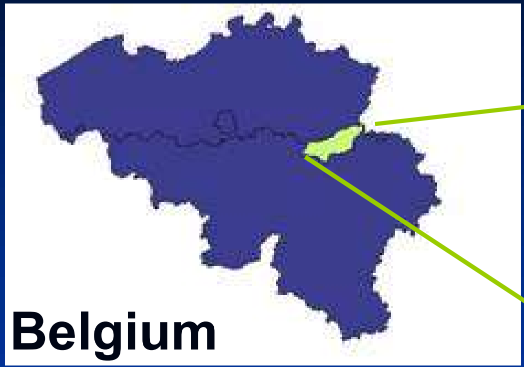
Geomac

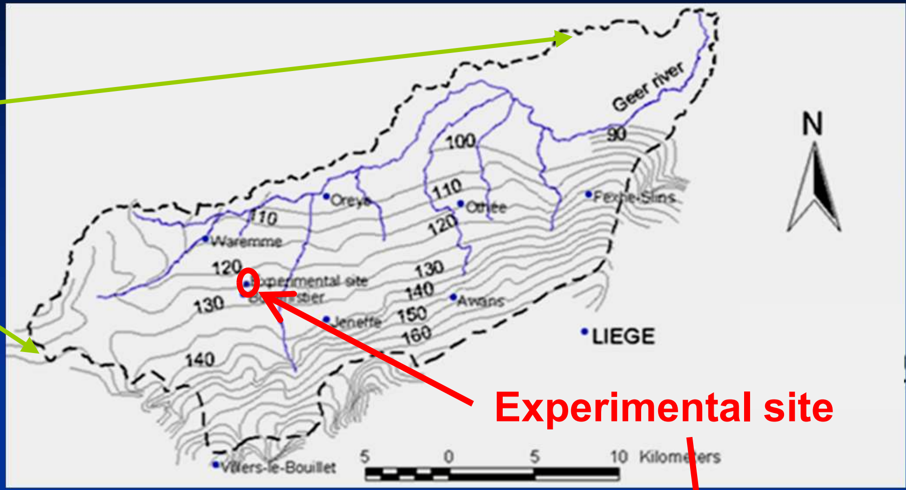
Hydrogéologie

Aquapôle

ULg
UNIVERSITÉ de Liège

General context

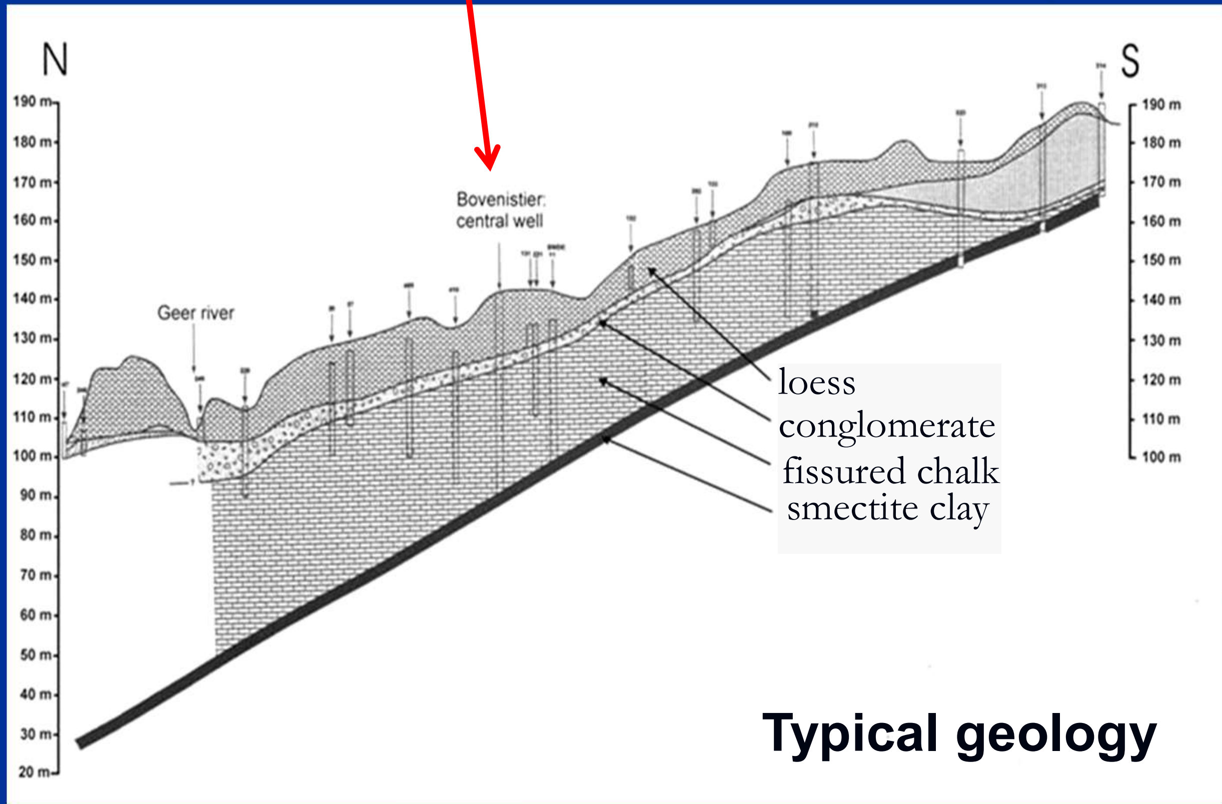




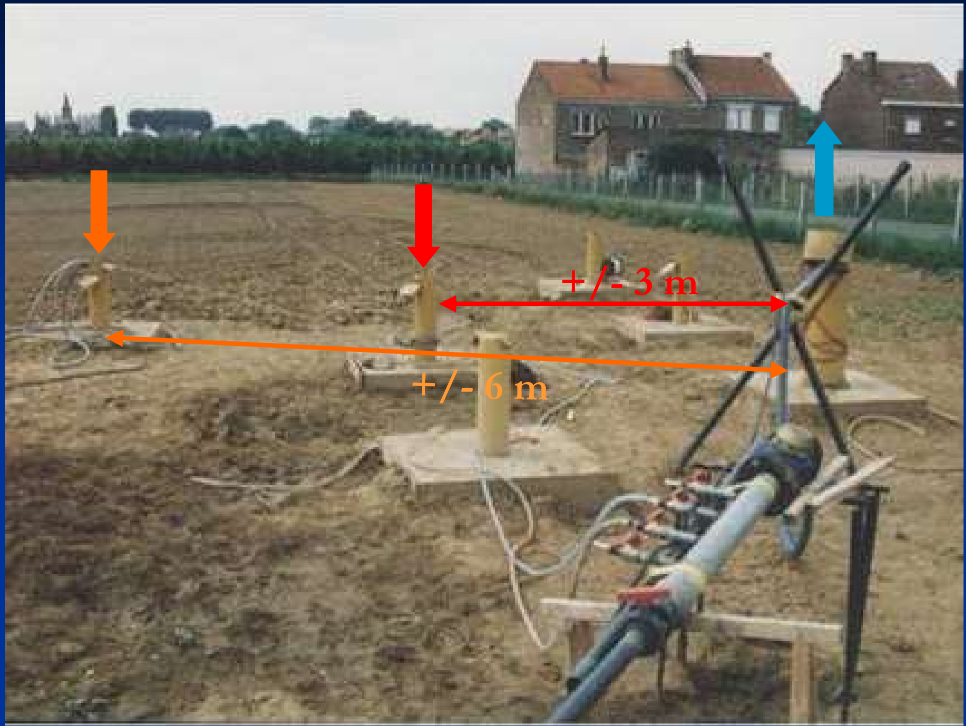
Main problem: continuous increase in nitrate content in the Hesbaye chalk aquifer

Objective: better understanding of recharge mechanisms and solute migration in the unsaturated zone

→ *Experimental study financed by the Walloon Ministry for Environment and Natural Resources*

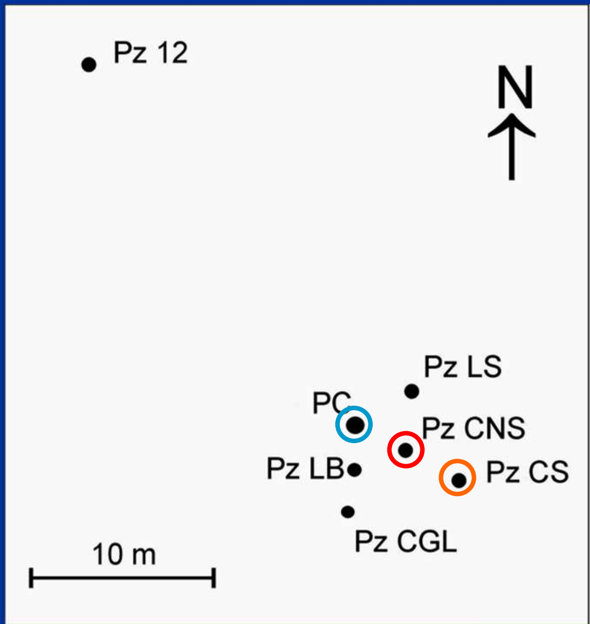


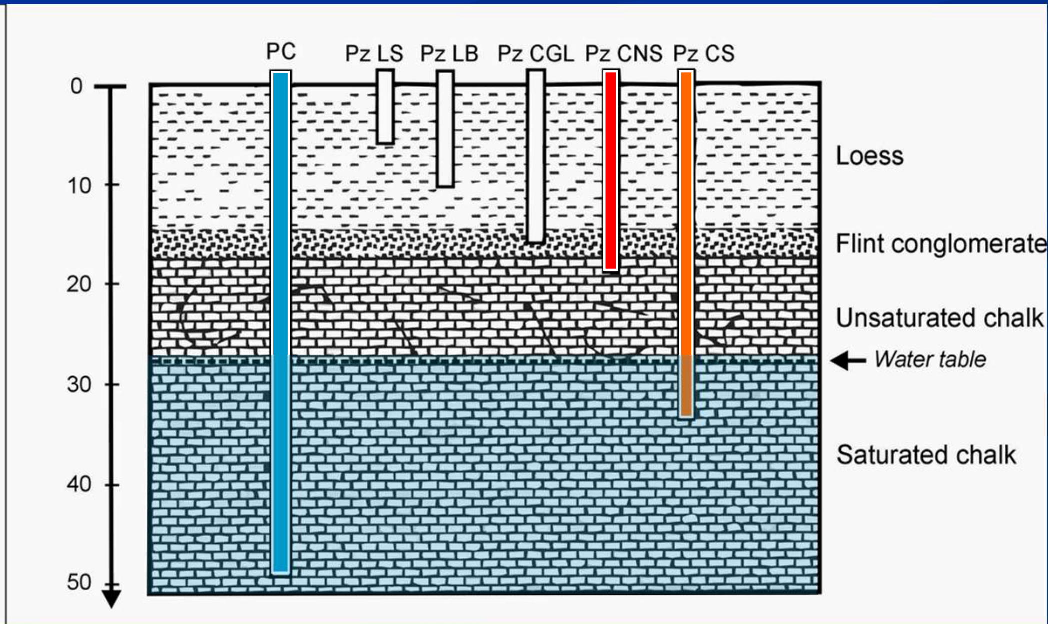
Experimental site



- Boreholes drilled → various depths
- Core samples taken for lab measurements
- Well logging
- Infiltration tests in the unsaturated zone
- Pumping tests in the saturated zone
- Tracer experiments in the saturated zone
- Tracer experiments in the unsaturated zone

Brouyère et al. (2004) in *J. Contam. Hydrol.*

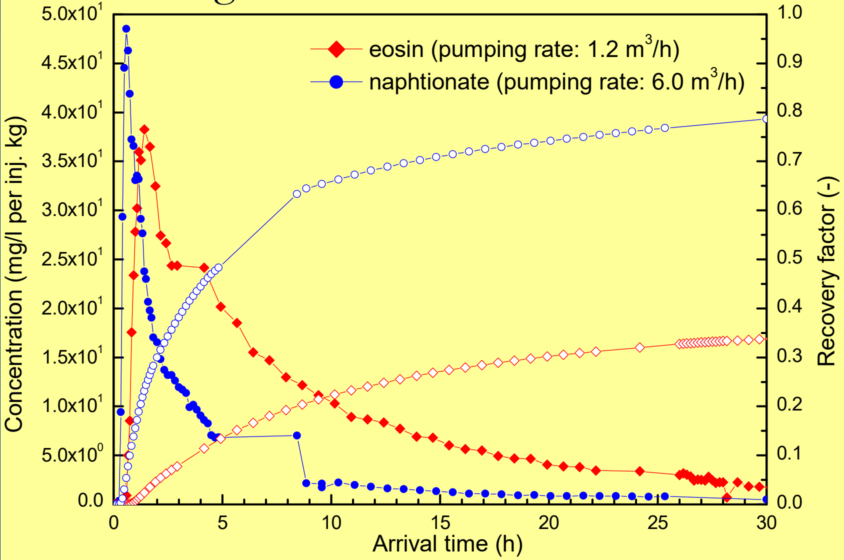




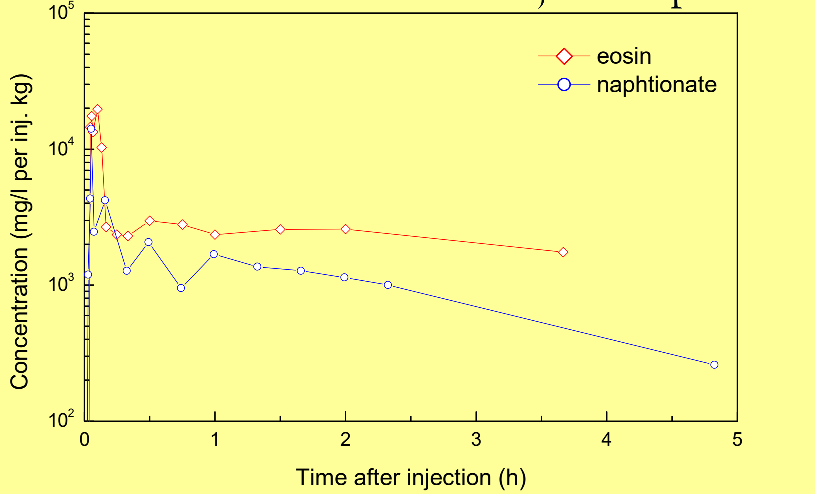
Tracer experiments in the saturated zone

Tracing distance: 6m	Phase 1	Phase 2
Tracer	**eosin Y**	**naphtionate**
Date of injection	2/7/1998 14:50	4/7/1998 9:40
Pumping rate (m^3/h)	**1.2**	**6.0**
Injected quantity (kg)	0.0053	0.0051
Tracer volume (m^3)	0.010	0.010
Tracer injection duration (h)	0.031 (1min53s)	0.036 (2min11s)
Water flush volume (m^3)	0.127	0.132
Water flush duration (h)	0.29 (17min20s)	0.22 (12min56s)
Date of 1st arrival	2/7/1998 15:25	4/7/1998 9:55
Minimum transit time (h)	**0.58 (35min)**	**0.25 (15min)**
Maximum velocity (m/h)	10.2	24
Date of modal restitution	2/7/1998 16:15	4/7/1998 10:15
Modal transit time (h)	**1.42 (85min)**	**0.5 (30min)**
Modal velocity (m/h)	4.2	1.2
C_{mod}/ C_{inj} (-)	1.93×10^{-3}	3.44×10^{-3}
Recovery factor (%)	**35 (after 40 h)**	**87 (after 77 h)**

Breakthrough curves:



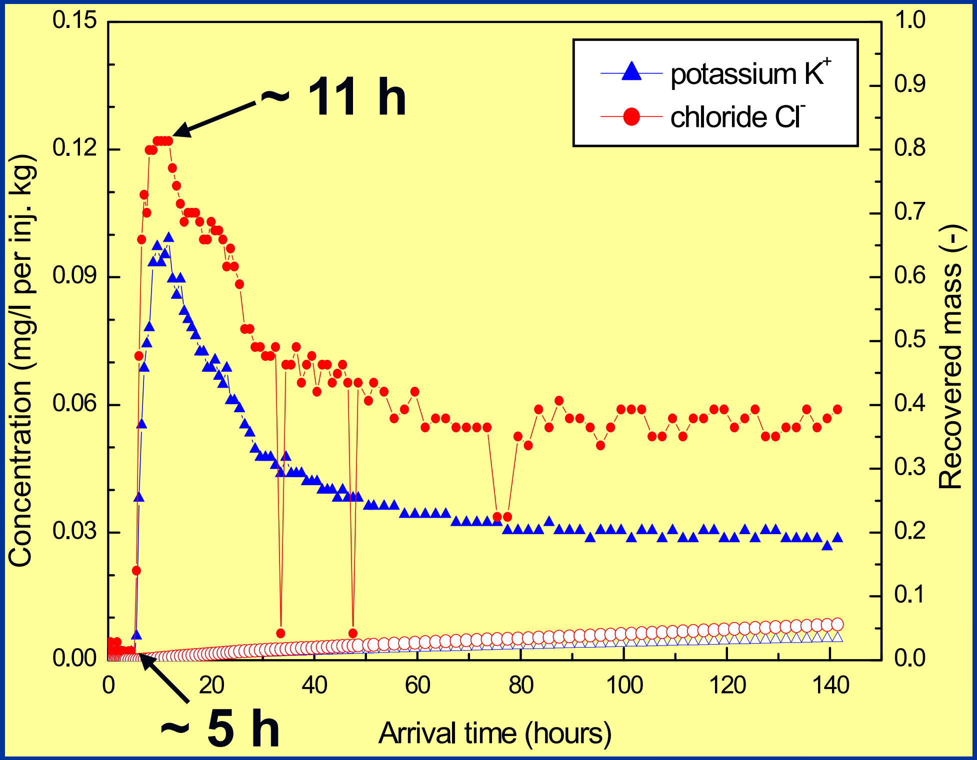
Concentration evolutions at injection point:

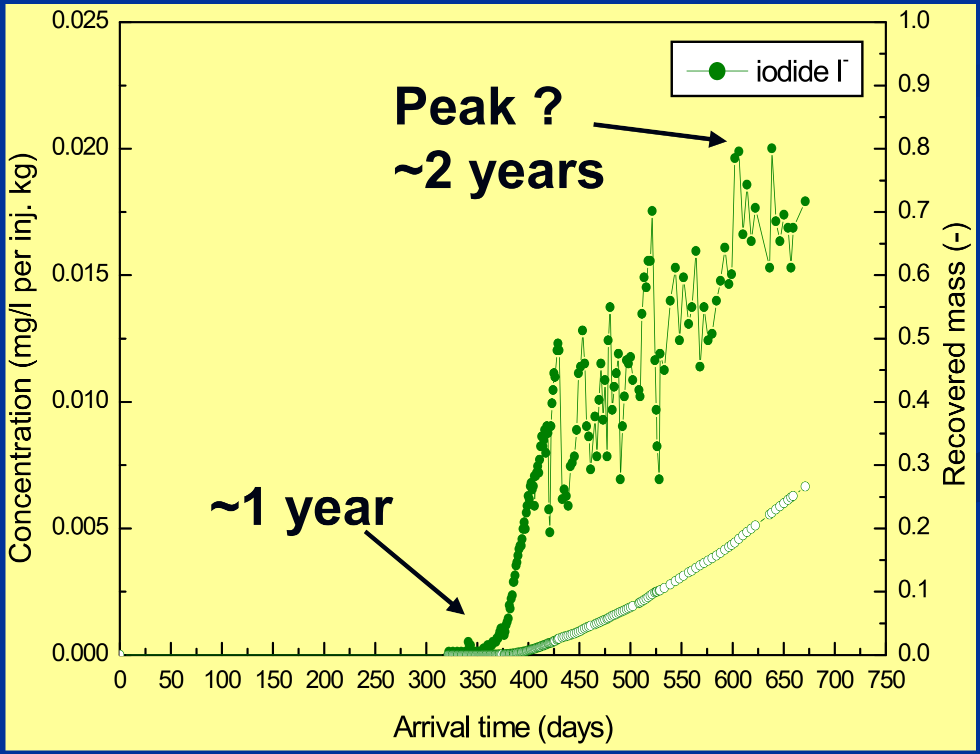


Tracer experiments in the unsaturated zone

- "Intense" recharge conditions
  - Tracer: KCl
  - Minj: 100 kg
  - Vinj: 300 l
  - Tinj: ~1 h
  - $Q_{recharge}$: ~0.3 m³/h
  - $Q_{pumping}$ = 6.4 m³/h
  - Vertical distance ~10m

- "Natural" recharge conditions
  - Tracer: I-
  - Minj: 10 kg
  - Vinj: 30 l
  - Tinj: ~ 6 min
  - $Q_{recharge}$: ---
  - $Q_{pumping}$ = 3 to 6 m³/h
  - Vertical distance: 3-4 m





Complementary information on chalk

- Infiltration and pumping tests

$$K_{sat} = 10^{-5} \rightarrow 10^{-4}\ m/s$$

- Laboratory measurements on chalk matrix samples
  - K_{matrix} =~10^{-8} m/s
  - Capillary rise in the matrix ~ > 10m
  - Matrix porosity ~ 30 – 40%
- Tracer tests in the saturated zone
  - Effective porosity θ_m ~< 1% (fissures)

→ Dual-porosity / dual-permeability of chalk

- Chalk matrix
  - High porosity (n_M ~ 30 → 45 %)
  - Microporosity (~1μm) → high capillary tensions (h_c ~10 – 30 m)
  - Low saturated hydraulic conductivity $K_{s,M}$ ~1×10^{-8} m/s
- Fissures
  - Low associated porosity ($n_F \leq 1\%$)
  - Relatively large opening → low capillary tensions
  - High saturated hydraulic conductivity $K_{s,F}$ ~$1\times10^{-5} \rightarrow 1\times10^{-4}$ m/s

Flow and transport conceptual model

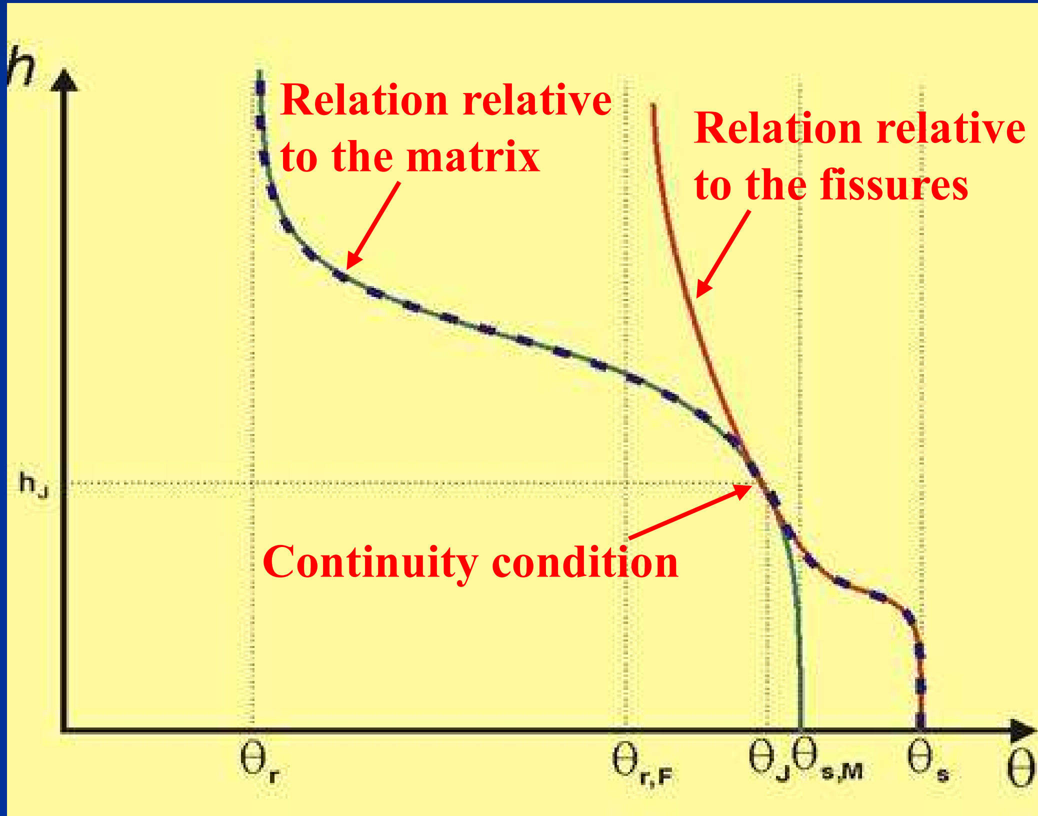
	Saturated zone and unsaturated zone under "intense" recharge conditions	Unsaturated zone under "normal" recharge conditions
Groundwater flow	$K_{s,M} << K_{s,F}$ → water storage in the matrix + water drainage through the fissures network	$K = K_{s,M}$ → Quick desaturation of the fissure network with groundwater flow restricted in the matrix
Solute contaminant transport	$K_{s,F}$ >> and n_F << → Fast migration along the fissures + physical retardation in the matrix (immobile water)	$K_{s,M}$<< and n_M >> → Slow migration of contaminants across the matrix

➔ 2 key factors:

- ➔ Unsaturated behaviour of the dual porosity chalk
- ➔ Variations in transport parameters (θ_m, θ_{im}) (matrix ←→ fissure) in function of the rock water content θ

Unsaturated properties of the chalk

- Bi-modal porosity of the chalk → bi-modal retention curve model required
  - Two van Genuchten relationships (one for the matrix, one for the fissures
  - Originality: continuity of the relationship at the matrix-fissure transition
  - $k_r(\theta)$ derived analytically from $\theta(h)$ using Mualem's pore distribution model
  - Possibility to compute analytically the derivatives $d\theta/dh$ and $dk_r/d\theta$



Unsaturated properties of the chalk

- Retention curve for the matrix

$$\Theta_M = \frac{\theta - \theta_r}{\theta_{s,M} - \theta_r} = \left(1 + (\alpha_M h)^{n_M}\right)^{-m_M}$$

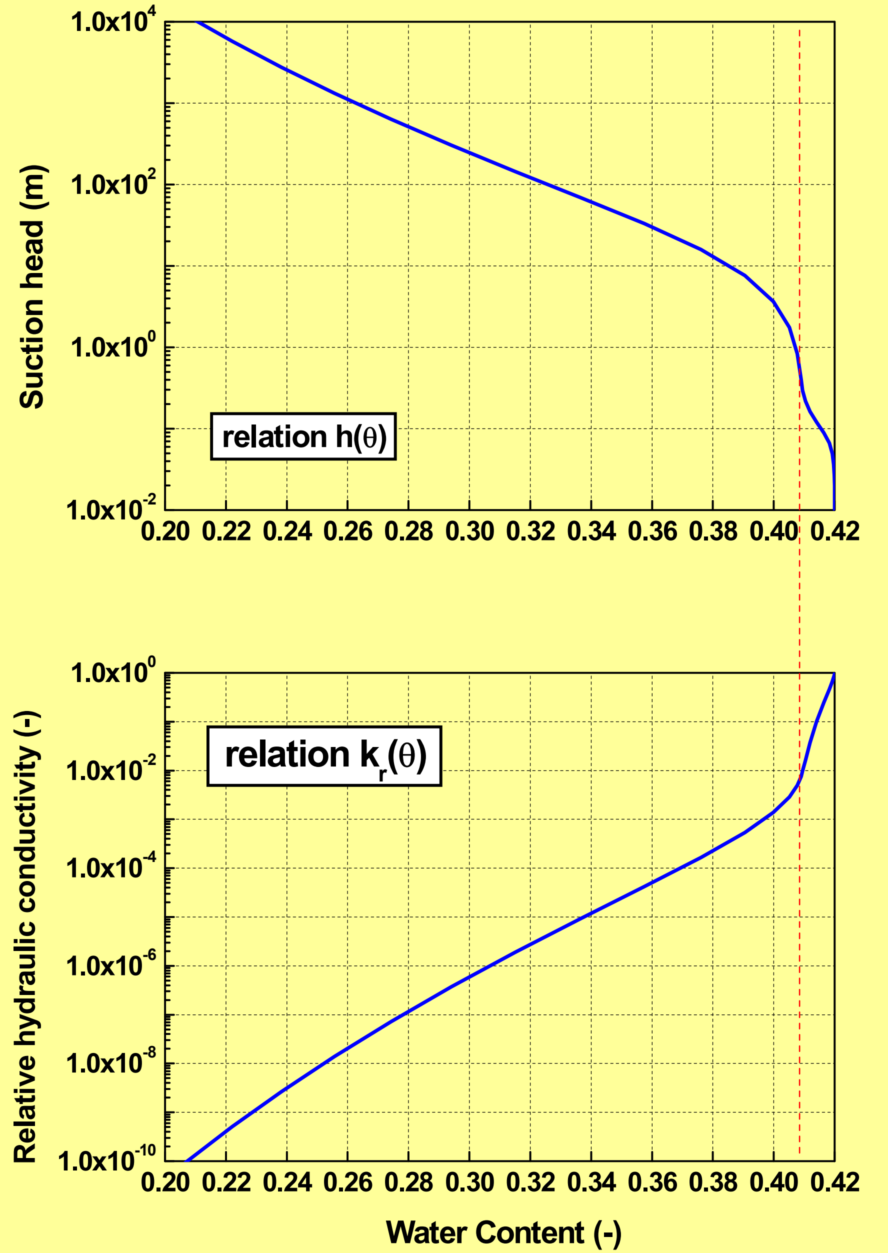
- Retention curve for fissures

$$\Theta_F = \frac{\theta - \theta_{r,F}}{\theta_s - \theta_{r,F}} = \left[1 + (\alpha_F h)^{n_F}\right]^{-m_F}$$

- Hydraulic conductivity curve

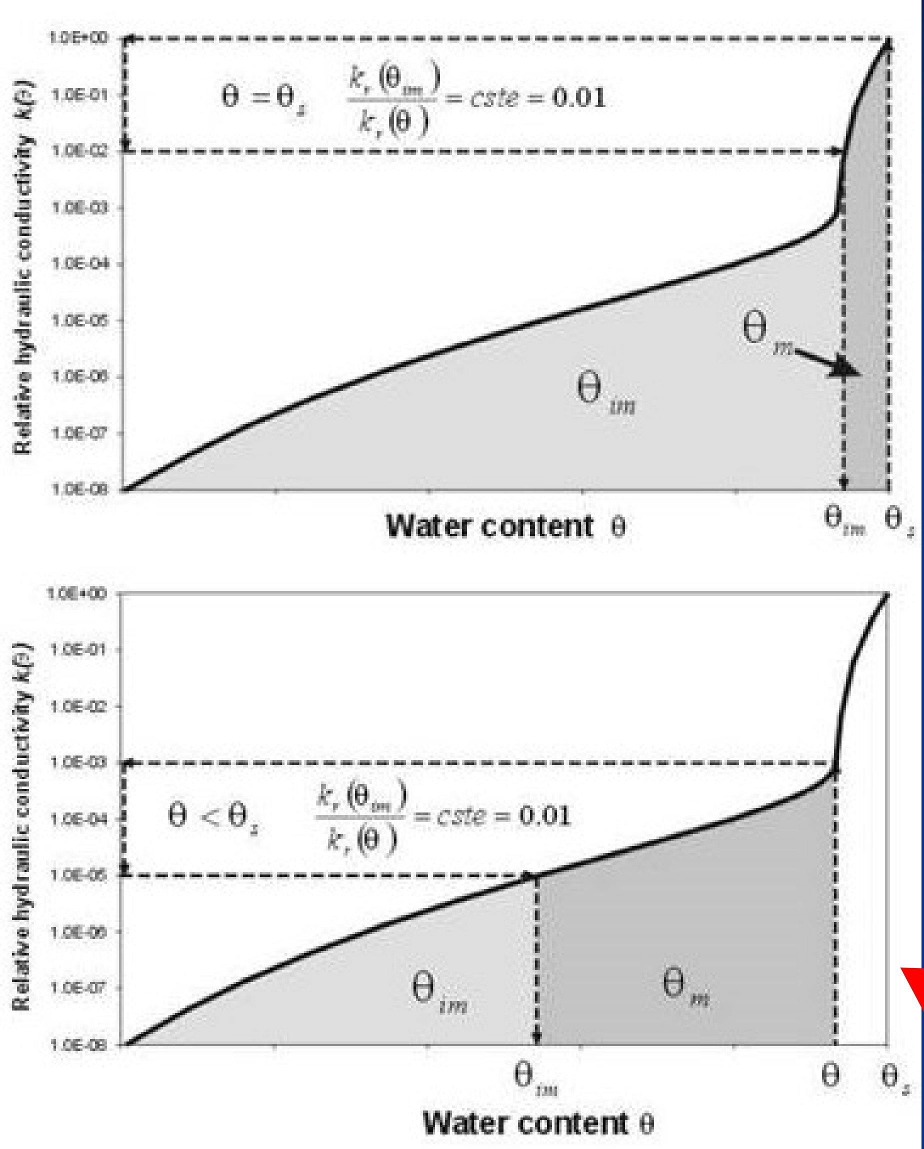
$$k_r(\theta) = S_e^P \left[\frac{f(\theta)}{f(\theta_s)}\right]^2 = S_e^P \left[\frac{\int_0^{\theta} d\theta / h}{\int_0^{\theta_s} d\theta / h}\right]^2$$

Details in Brouyère (2004), manuscript in preparation



Example with measurements obtained
On chalk matrix samples + in the field

Solute transport in the variably saturated chalk



- Classically: 1st order transfer, dual-porosity transport model (Mobile-Immobile water Model)
  - Transfer coefficient α (T^{-1})
  - Dual porosity θ_{im} (-) = constant
- MIM Not suited here as θ_{im} and θ_m vary with the water content θ of the chalk rock
- Adapted MIM approach (close to Zurmühl & Durner, WRR 1996) with dynamic variation of θ_{im} and θ_m using a partitioning coefficient:

  $C_{part}=k_r(\theta_{im}) / k_r(\theta)$

  (~capillary bundle approach)

Solute transport in the variably saturated chalk

- Adapted equations for MIM Model
  - Mobile water

$$\theta_m \frac{\partial C}{\partial t} + \frac{\partial \theta_{im}}{\partial t} C^* = -\underline{v}_D \cdot \underline{\nabla} C + \underline{\nabla} \cdot \left(\theta_m \underline{\underline{D}}_h \cdot \underline{\nabla} C \right)$$

$$+ q\left(C' - C\right) - \lambda \theta_m C - \alpha\left(C - C_{im}\right) + \left(F \frac{\partial h}{\partial t} - \frac{\partial \theta_m}{\partial t} \right) C$$

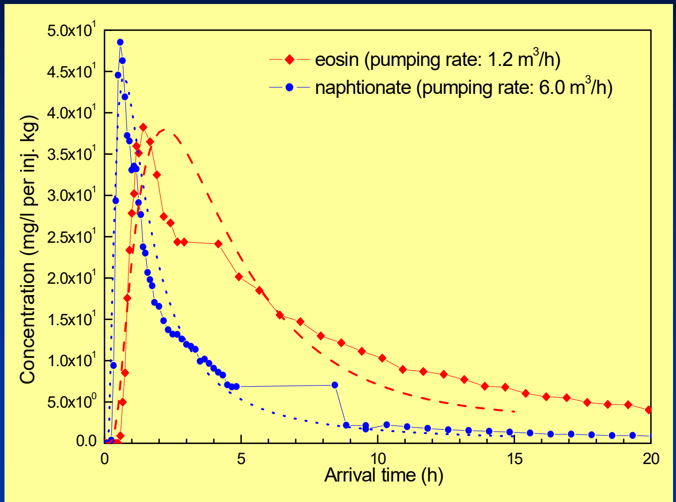
  - Immobile water

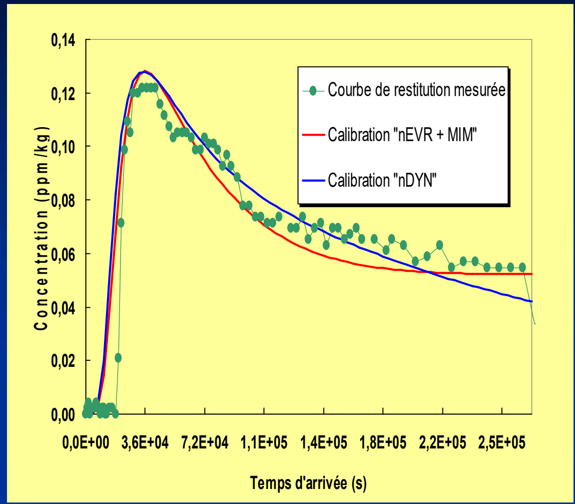
$$\theta_{im} \frac{\partial C_{im}}{\partial t} = \alpha\left(C - C_{im}\right) - \lambda \theta_{im} C_{im} + \left(C^* - C_{im}\right) \frac{\partial \theta_{im}}{\partial t}$$

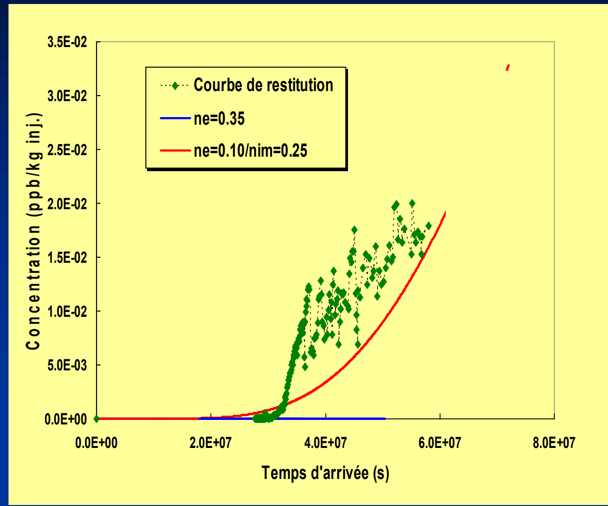
$C^* = C$ or C_{im} *according to the direction of mass flux exchange between mobile and immobile water*

- Finite Element Code SUFT3D
  - Semi-analytical solution on the computation time step applied to the mass-conservation equation in the immobile water
  - Solution back-substituted in the MC Equation in the mobile water which is solved using the CVFE method

Modelling Results







Injection well	Experimental conditions	Modelling approach	θ_m (-)	θ_{im} (-)	α_L (m)	α (s^{-1})
Pz CS	Saturated zone	CDPM	0.004	0.05	1.0	2.0×10^{-7}
PsCNS	Intense recharge	CDPM	0.01	0.08	1.0	2.3×10^{-7}
		DDPM	$c_{part} = 0.02$		1.0	9.0×10^{-8}
	Natural infiltration	CDPM	0.10	0.15	1.0	2.3×10^{-7}
		DDPM	no tracer arrival observed during the simulated period			

CDPM: Classical MIM approach DDPM: Dynamic MIM approach

Conclusions

- Tracer experiments → new insight on recharge mechanisms and solute migration in the chalk + importance of the dual-porosity/ dual-permeability of the chalk in the unsaturated and saturated zones
- Unified conceptual and mathematical approach to model the hydrodynamic and hydrodispersive behaviour of chalk in variably saturated conditions
  - New functionality for the unsaturated properties of structured geological formations (fissured chalk, macroporous soils …); hydraulic conductivity directly and analytically derived from the retention curve
  - « Easy-to-use » MIM model generalized to the case of variably saturated flow conditions
- Vulnerability of chalk to contamination occurring at the land surface: strongly dependent of recharge conditions at the top of the unsaturated chalk (protective cover…)
- Conceptual model could be used for different practical aspects: agricultural contaminants migration in the unsaturated – saturated system, trend analysis, fast flow in unsaturated rock without pressure disequilibrium, …

Thank you !

Unsaturated properties of the chalk

- Groundwater flow equation in variably saturated conditions
  - Single continuum approach considered (one pressure field)

$$\frac{\rho}{\rho_0} F \frac{\partial h}{\partial t} = \underline{\nabla} \cdot \left[\underline{\underline{K}}(\theta) \cdot \underline{\nabla} \left(h + \frac{\rho}{\rho_0} z \right) \right] + \frac{\rho^*}{\rho_0} q \quad \text{with} \quad F = S_s + \frac{d\theta}{dh}$$