DE LA RECHERCHE À L'INDUSTRIE

Détection et Migration des gaz dans les milieux géologiques: Expériences et simulations au Laboratoire Naturel de Roselend

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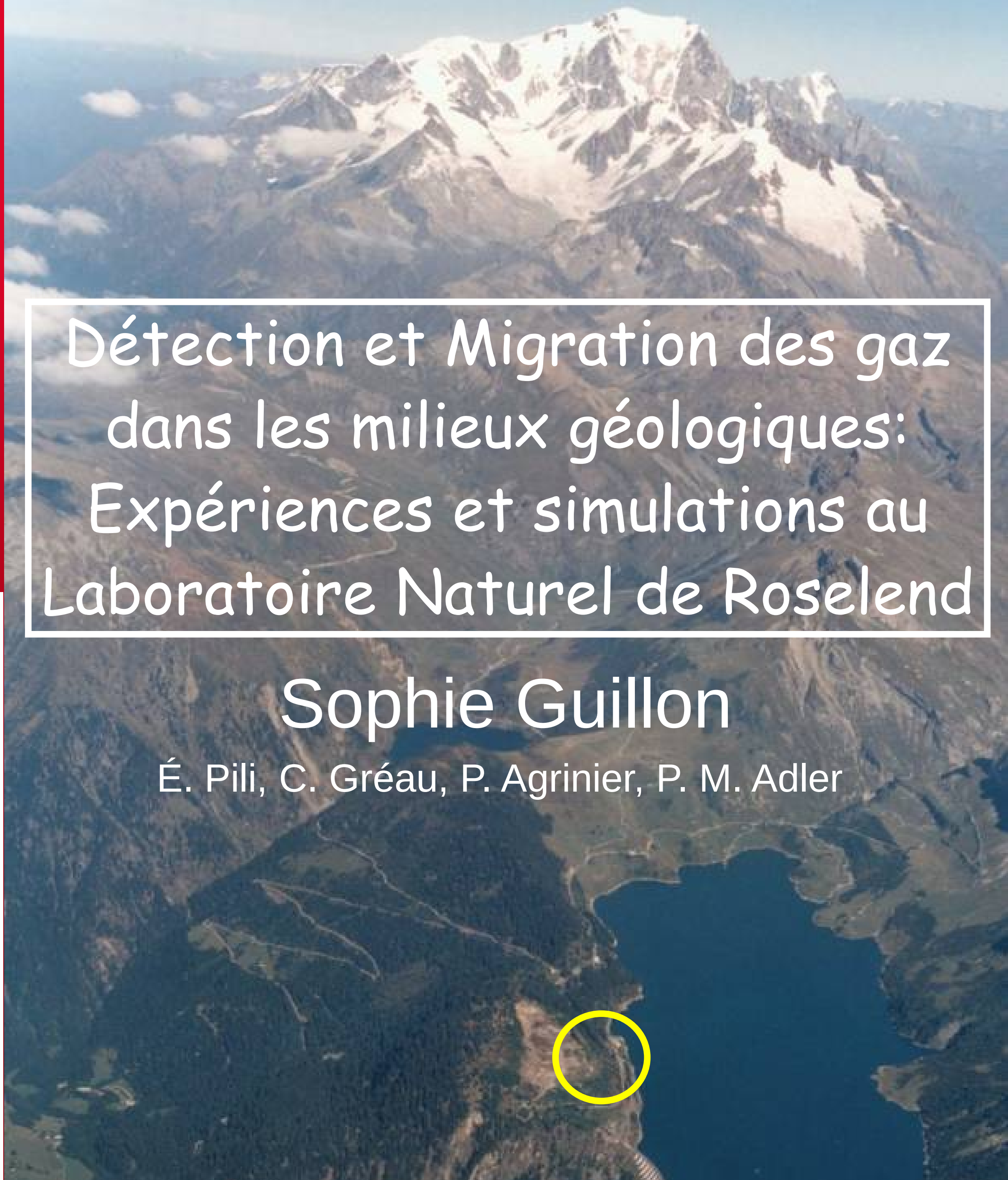
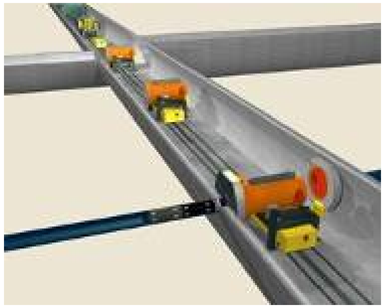


Gas migration in rocks matters for…

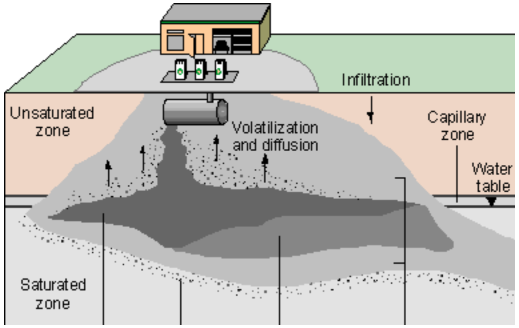
Monitoring of CO_2 Carbon Sequestration

Safety of nuclear waste disposal



Precursory signals of natural hazards

(earthquakes, volcanic eruptions)



Remediation of VOCs pollution

Recording of paleoclimates in archives

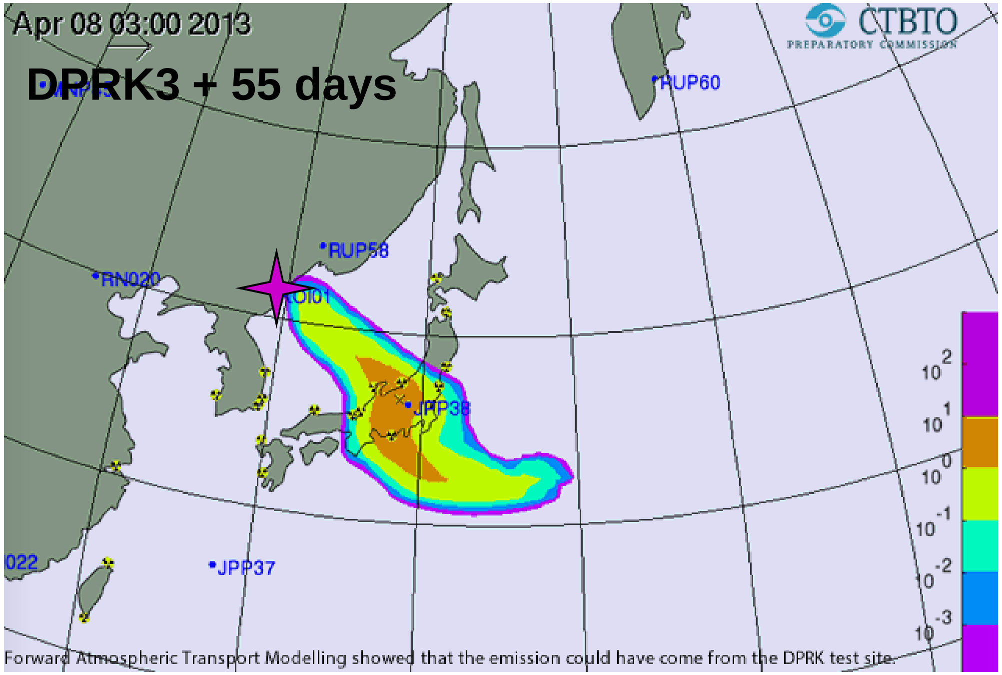
Greenhouse gas budget

Gas migration matters for the detection of Undergound Nuclear Explosions

- Comprehensive Nuclear-Test-Ban Treaty
- Detection & localization from **seismicity**
- Confirmation of nuclear origin by **radioactive noble gases** (Xe, Kr, Ar)

Gas venting following Baneberry event (1970, Nevada Test Site, USA)

✓ Early venting



Radioxenon migration following North Korea nuclear test in 2013

✓ Late-time seepage

Verification regime of the Comprehensive Nuclear-Test-Ban Treaty

- **International Monitoring System (IMS)**

Monitoring network for radioxenons in the atmosphere

SPALAX

Système de Prélèvement Automatique en Ligne avec l'Analyse du Xénon

- **On-Site Inspection (OSI)**

Gas migration matters for the verification of the Comprehensive Nuclear-Test-Ban Treaty

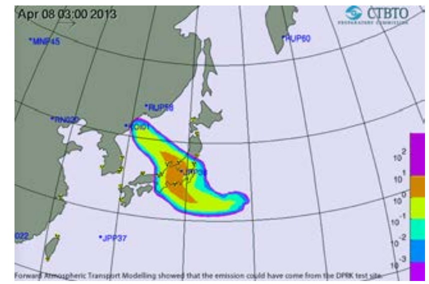
On-Site Inspection

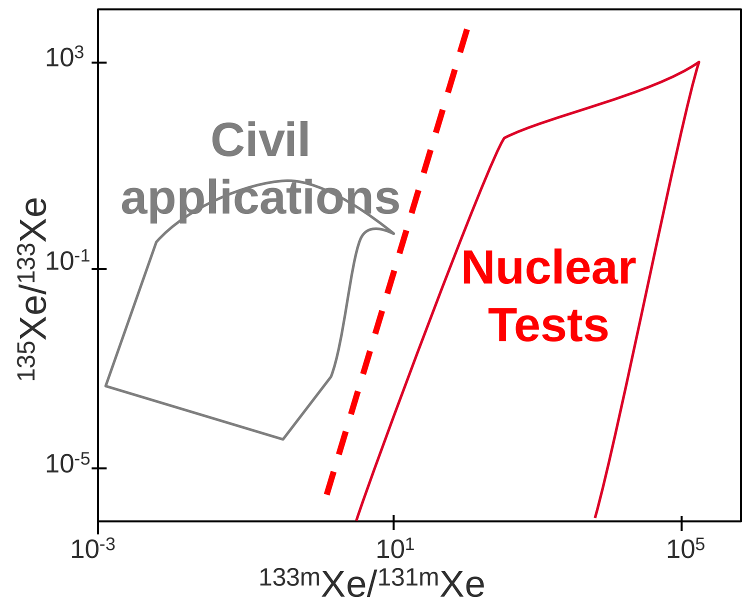
International Monitoring System

Atmospheric transport

Transport in the geosphere

- ⇒ **Delay ?**
- ⇒ **Dilution ?**
- ⇒ **Fractionation ?**





From Sun, Carrigan & Hao (2013)

Questions to be solved

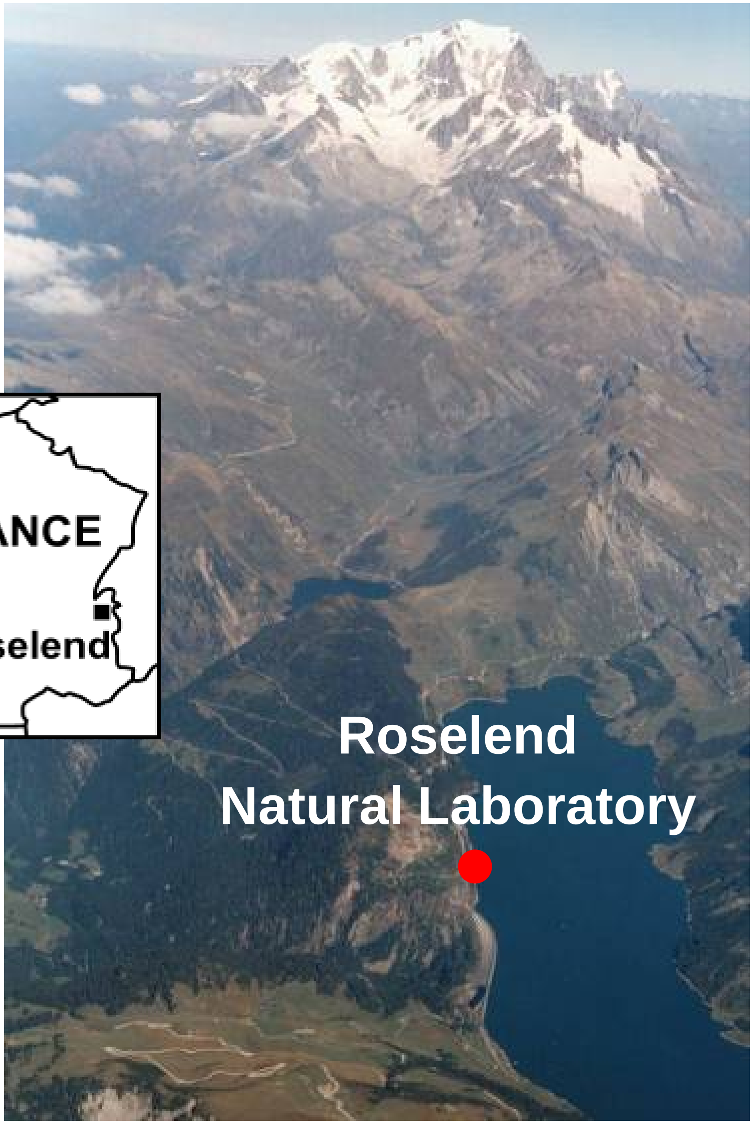
- What are the **driving forces** of gas migration and their respective influences ?

- What are the **dilution** and **temporal delays** between production of a tracer at depth and breakthrough at the surface?

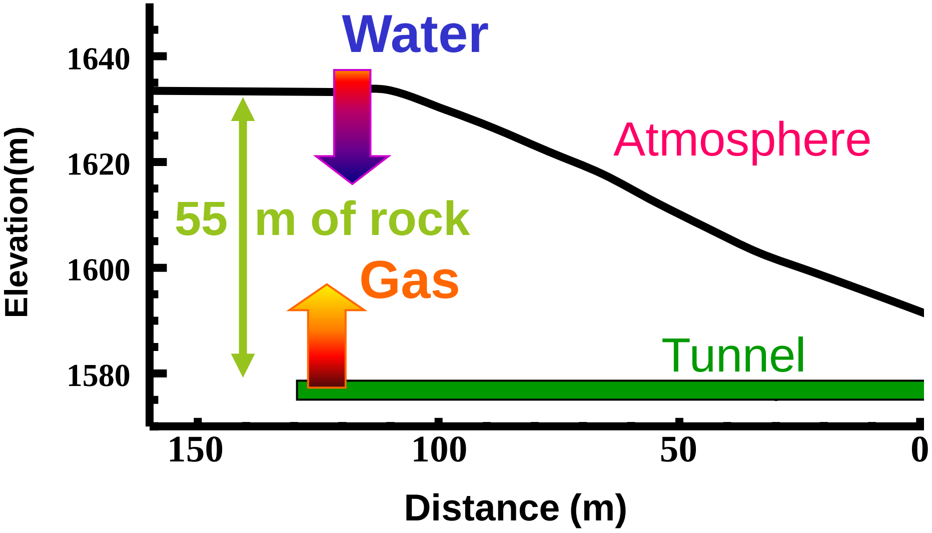
- How do **water fluxes** affect gas migration in the unsaturated zone?

- How to measure and understand **biogenic** gas dynamics?

The Roselend Natural Laboratory is all About Transfer Functions in Fractured Porous Media

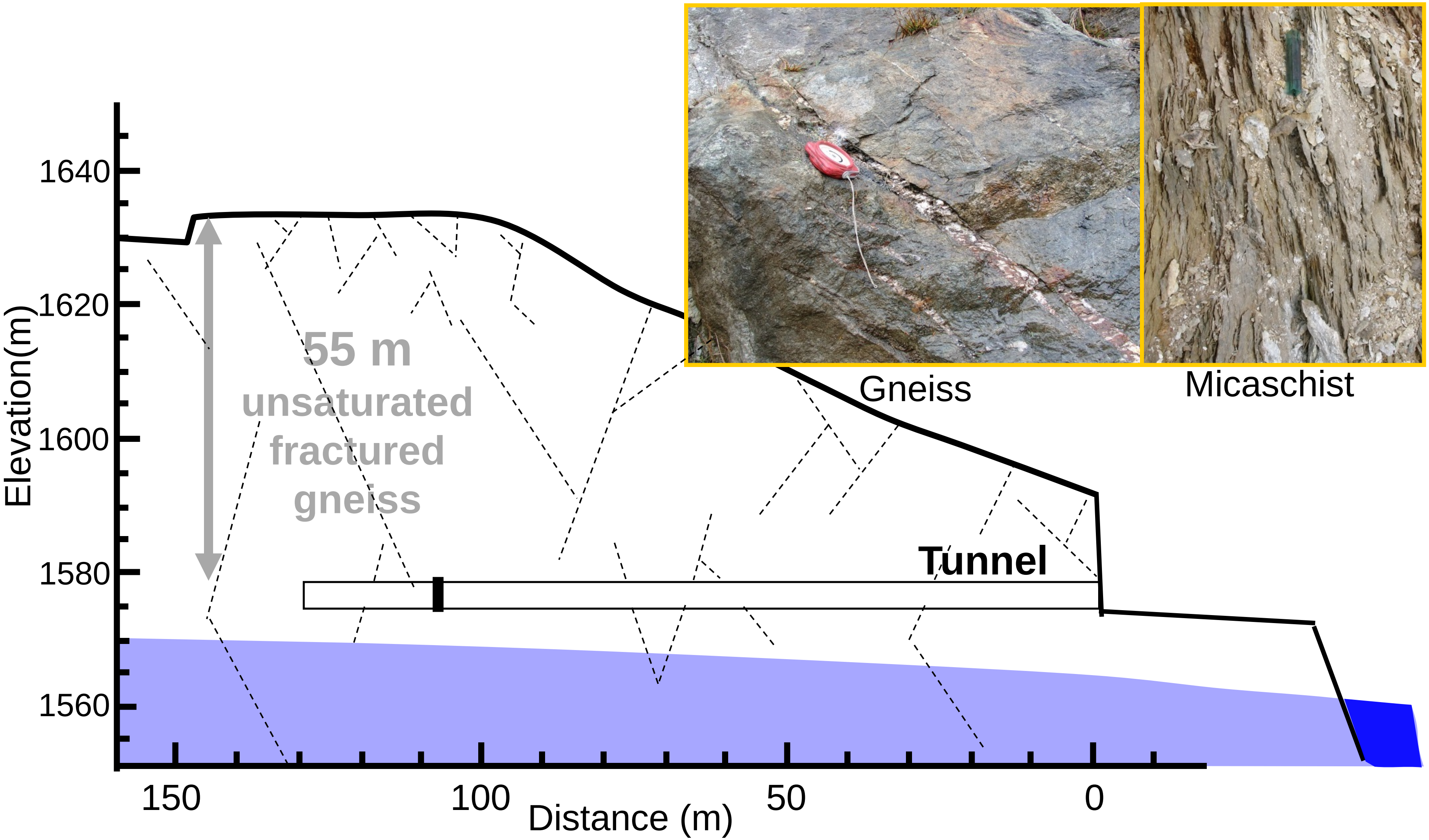


Long term, high resolution monitoring of fluids in unsaturated fractured rocks

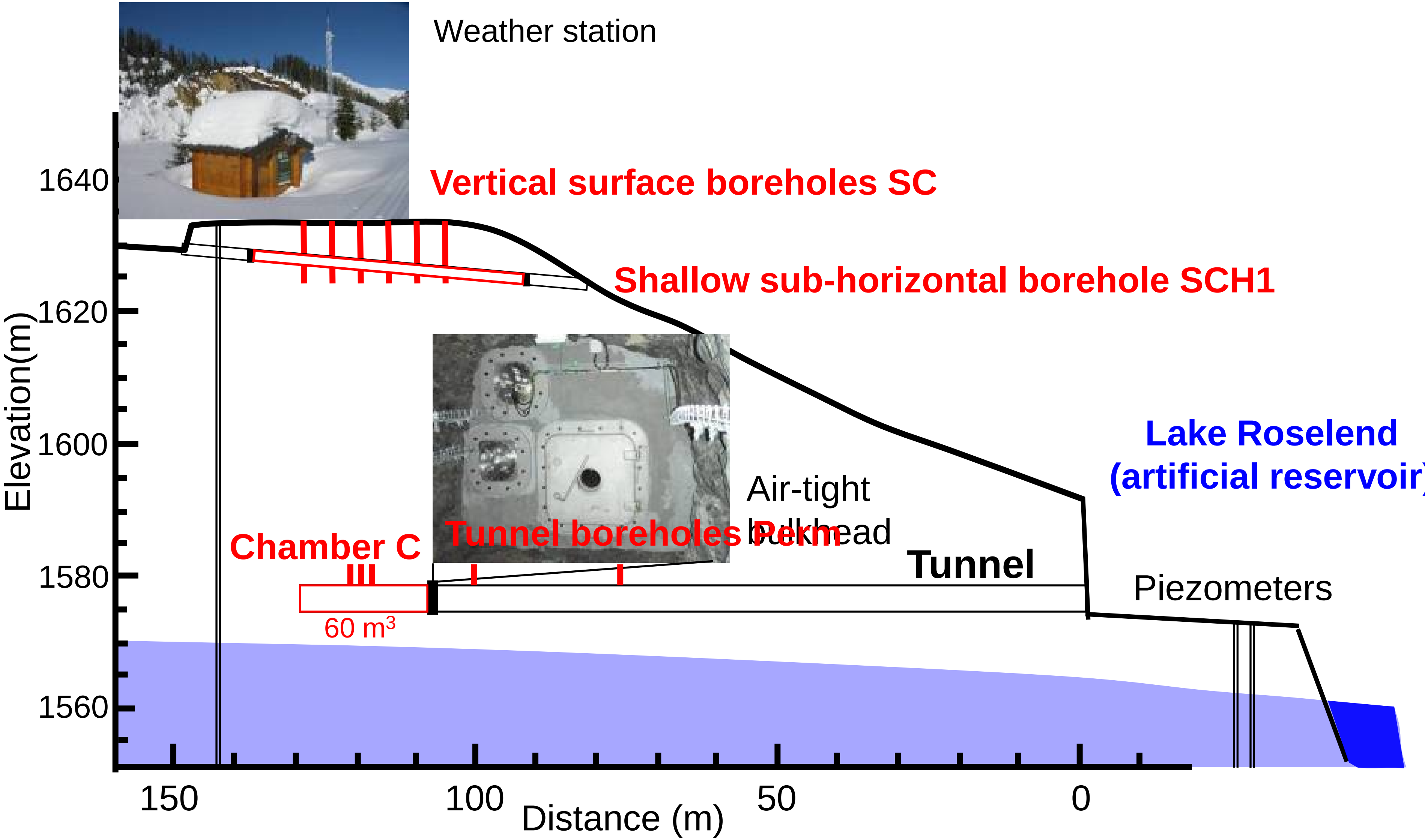


Perrier et al. (1998), Trique et al. (1999), Trique et al. (2002), Pili et al. (2004), Richon et al. (2005), Perrier et al. (2005), Patriarche et al. (2007),Pili et al. (2008),Richon et al. (2009), Perrier et al (2009), Wassermann et al. (2011), Pili et al. (2012)

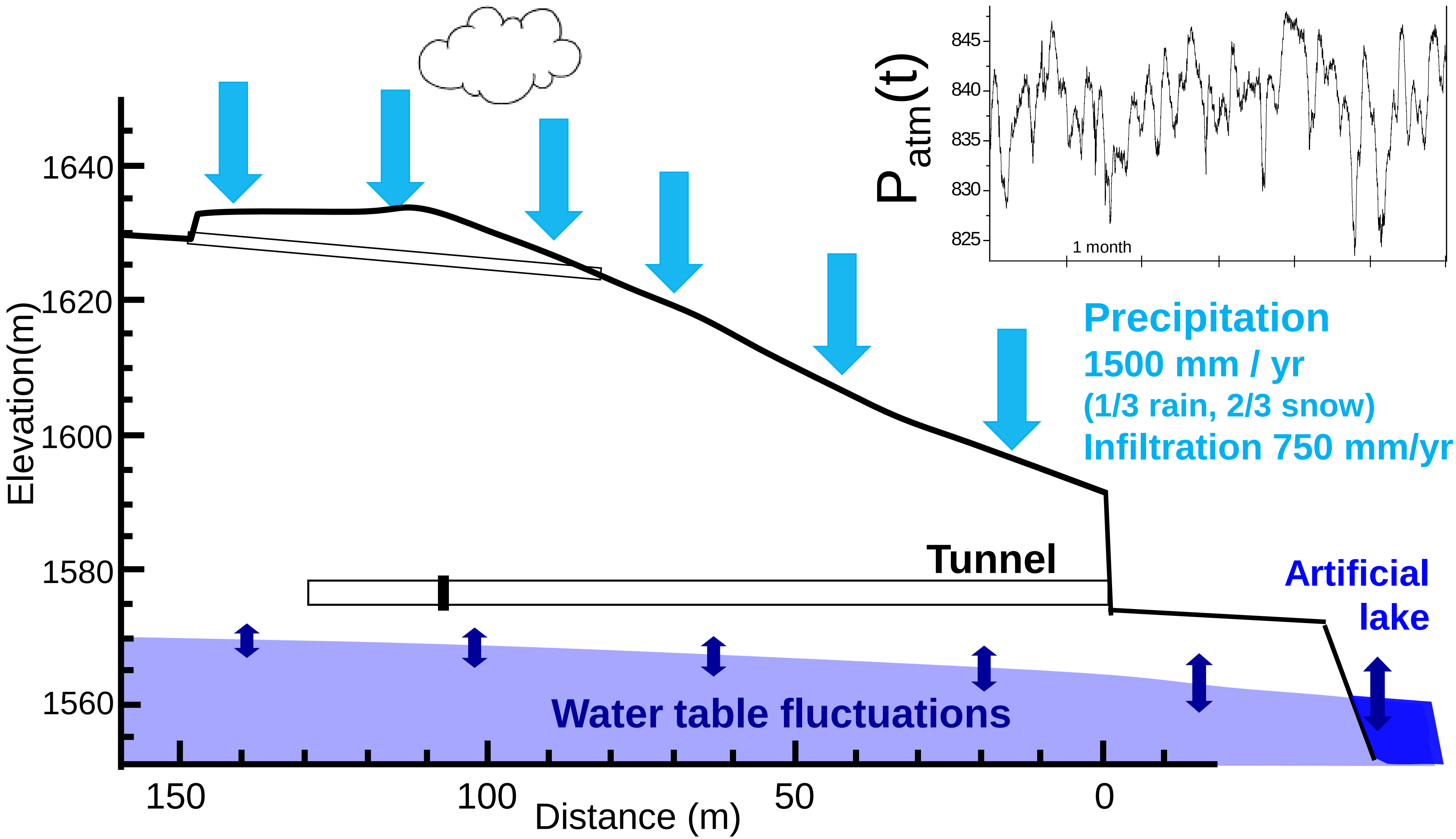
Geology of the Roselend Natural Laboratory



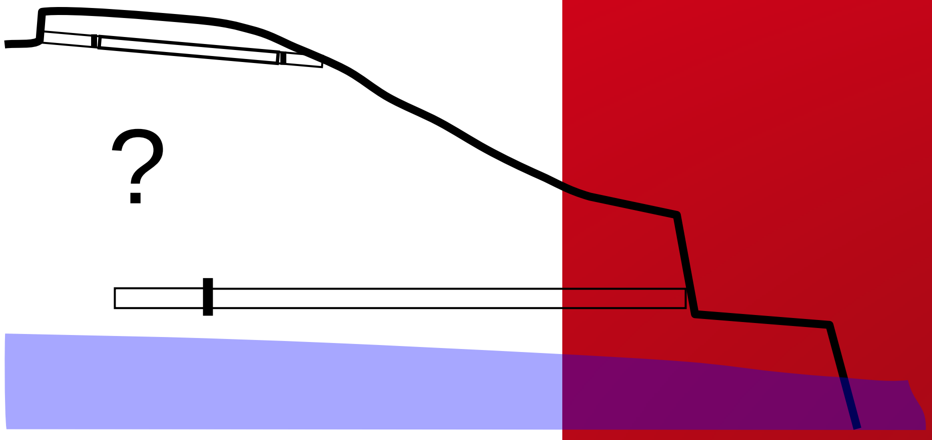
Infrastructures of the Roselend Natural Laboratory



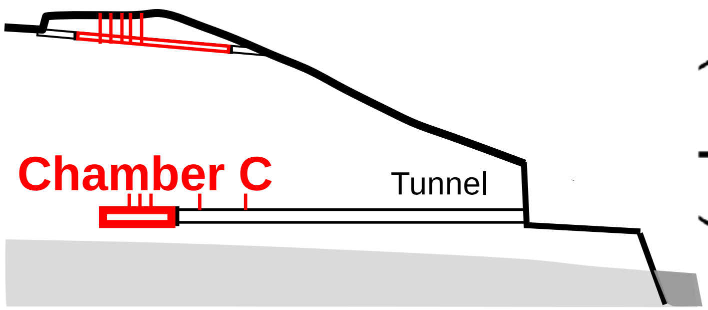
What is moving gases in the unsaturated zone?

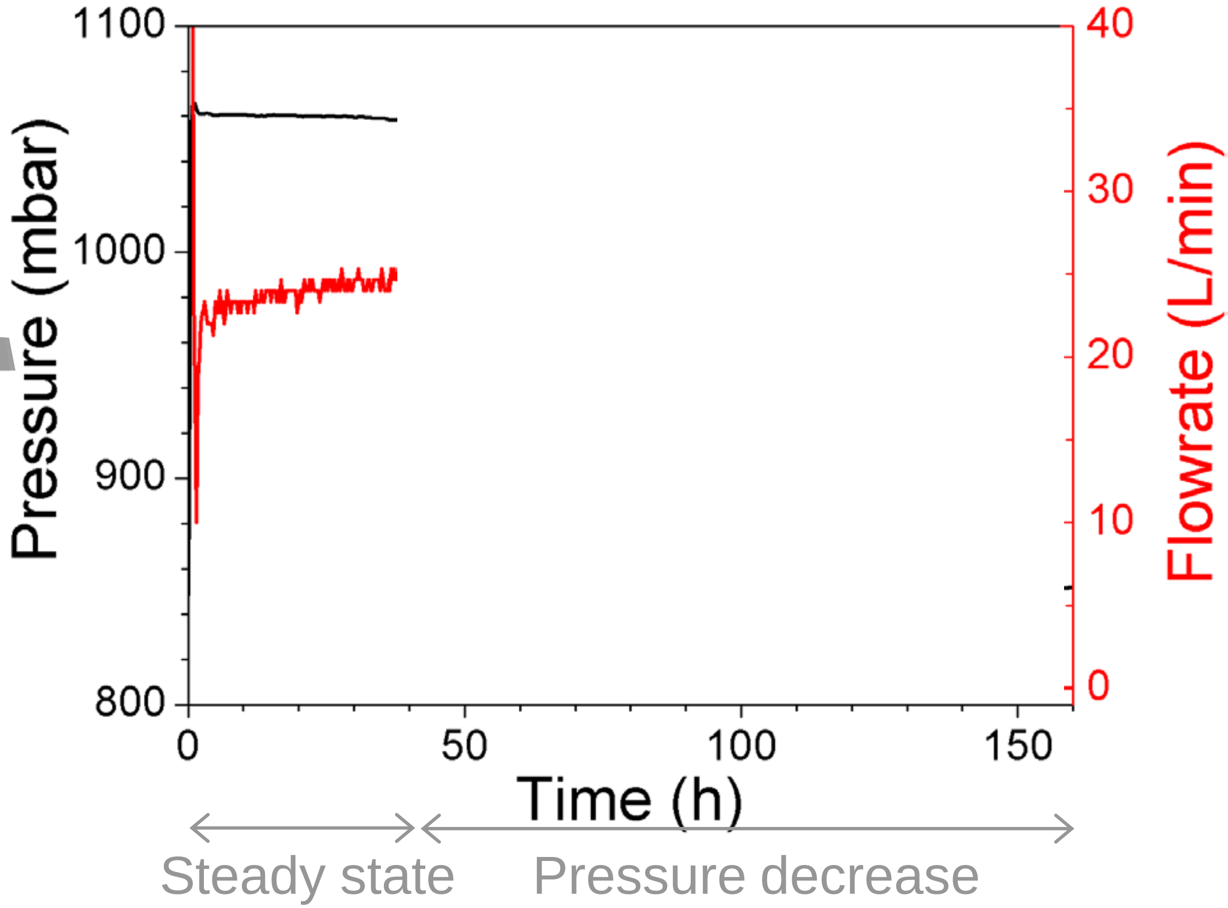


Pneumatic parameters: Permeability estimations from experimental and numerical approaches



Pneumatic injection tests



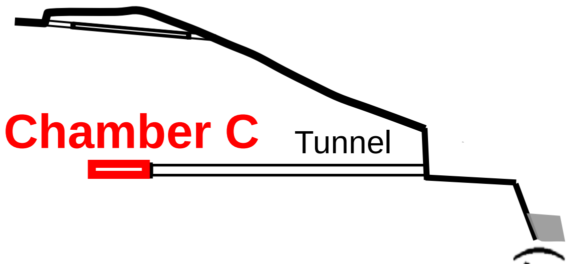


- Injection of air at constant flow-rate & steady state
- Exponential pressure decrease after injection stops

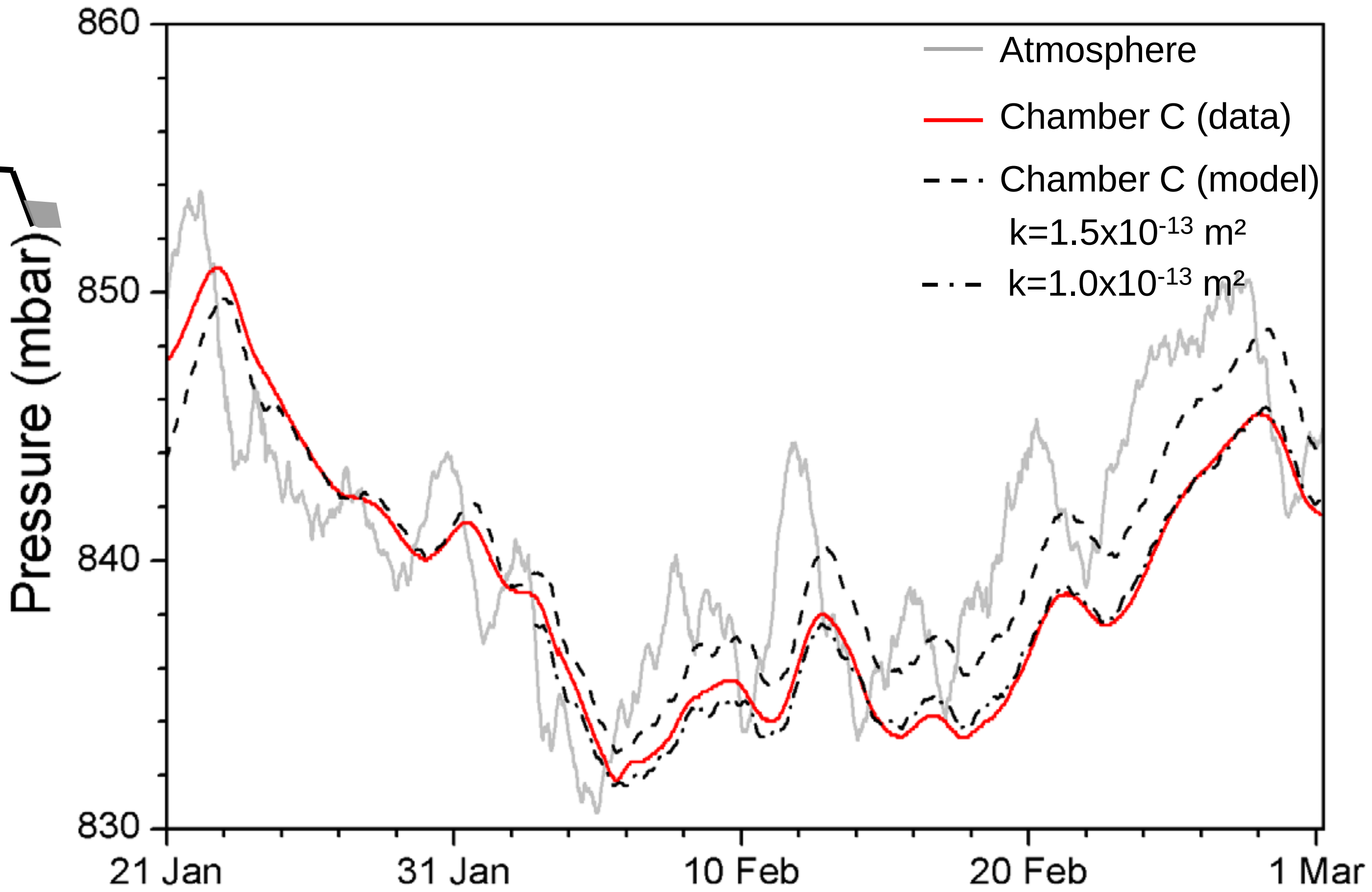
✓ Equivalent porous medium: $k \sim 8 \times 10^{-15}$ m²

Wassermann et al. (2011), Guillon et al. (2013)

Diffusion of atmospheric pressure fluctuations



$$\frac{\partial P}{\partial t} = \frac{k}{\varepsilon}\frac{P_0}{\mu}\nabla^2 P$$



✓ Estimation of diffusivity (k/ε) k~ 1.5×10^{-13} m² for ε~ 5.0%

Temporal variability of permeability (~ water content)

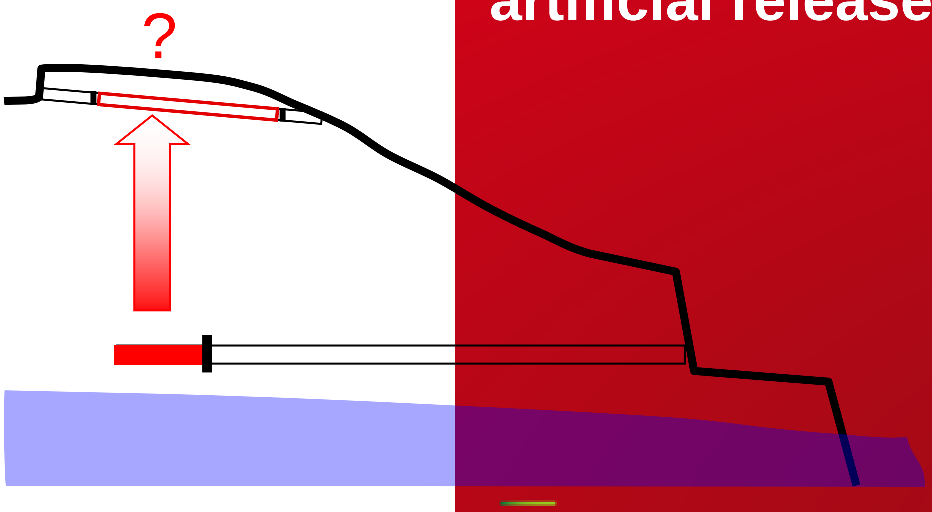
Large-scale value (55 m)

Guillon et al. (2013)

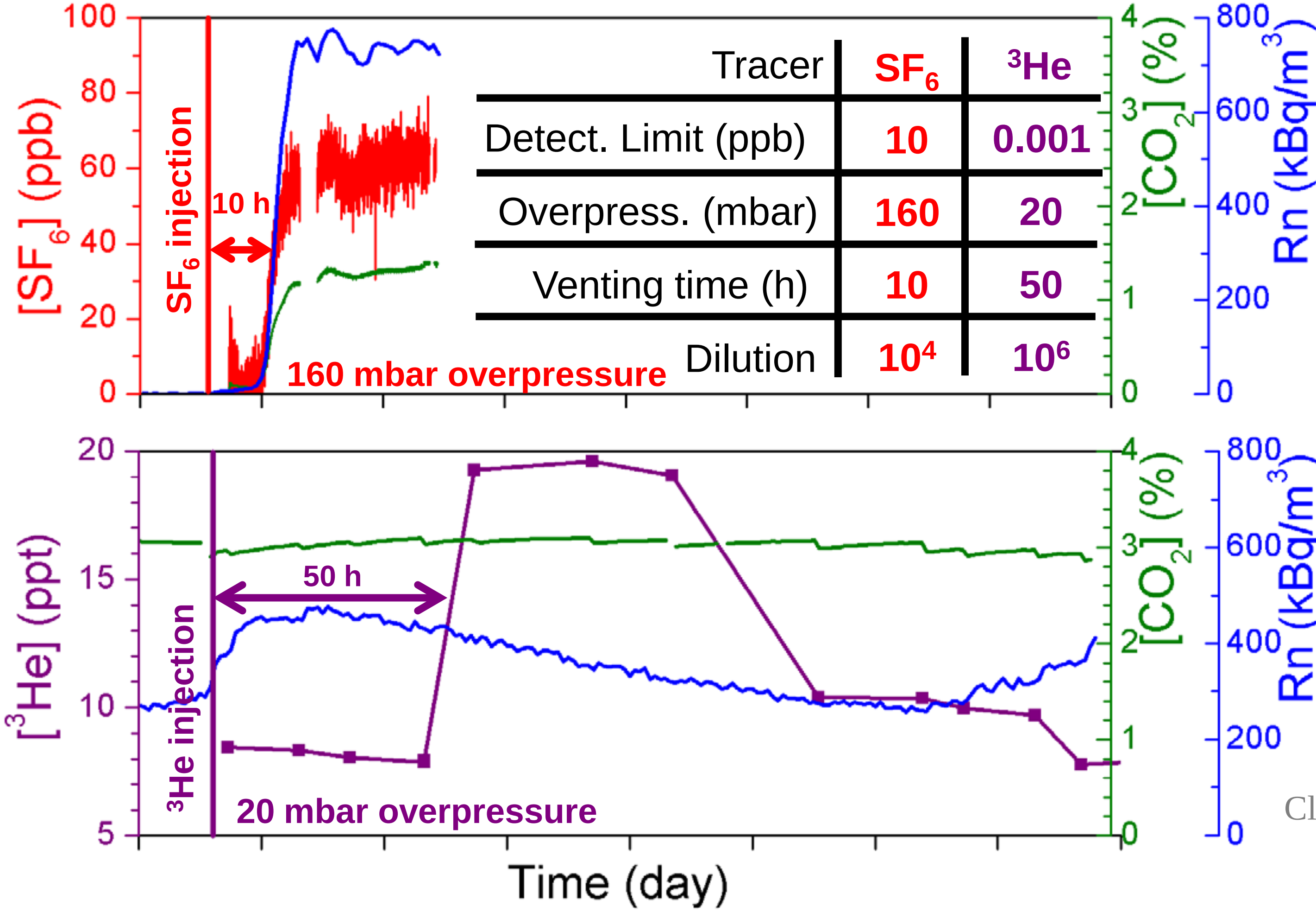
T R A C I N G E X P E R I M E N T S

Migration

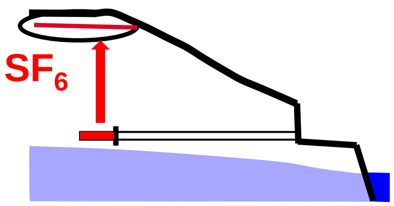
Learning from the transfer of gases after artificial releases at depth

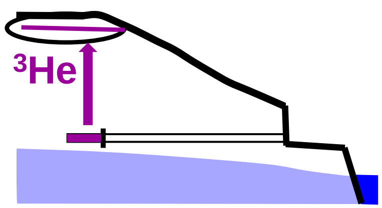


Rapid advection in few fractures following tracer injection in a deep cavity



Tracer	SF_6	^3He
Detect. Limit (ppb)	10	0.001
Overpress. (mbar)	160	20
Venting time (h)	10	50
Dilution	10^4	10^6

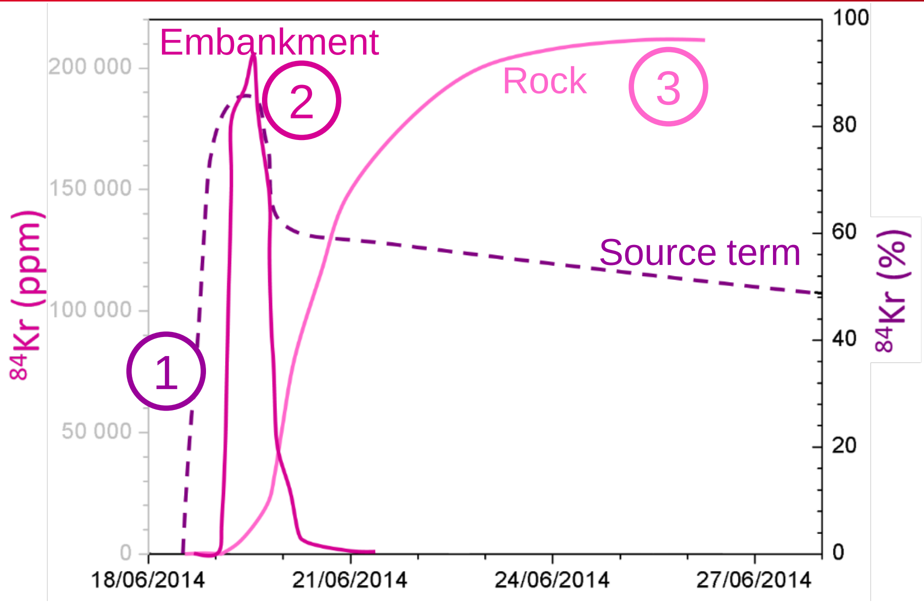


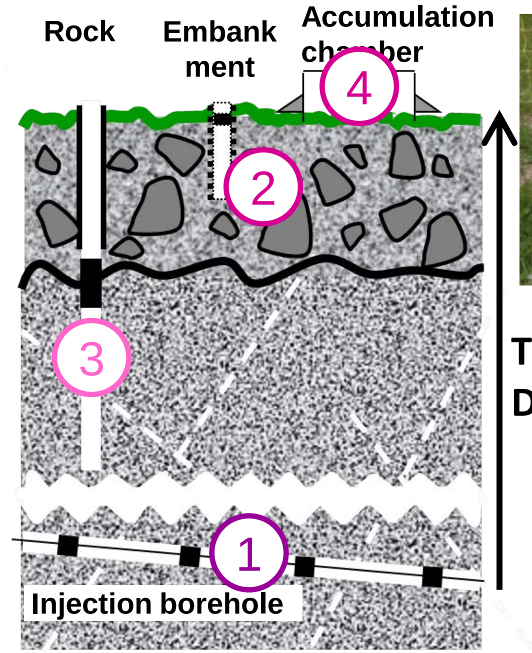


Claire Gréau is thanked for ^3He analyses

- ✓ Early venting with very large dilution
- ✓ Rapid advection in fractures driven by cavity overpressure
- ✓ Complex relation with naturally occurring gases (Rn, CO_2)

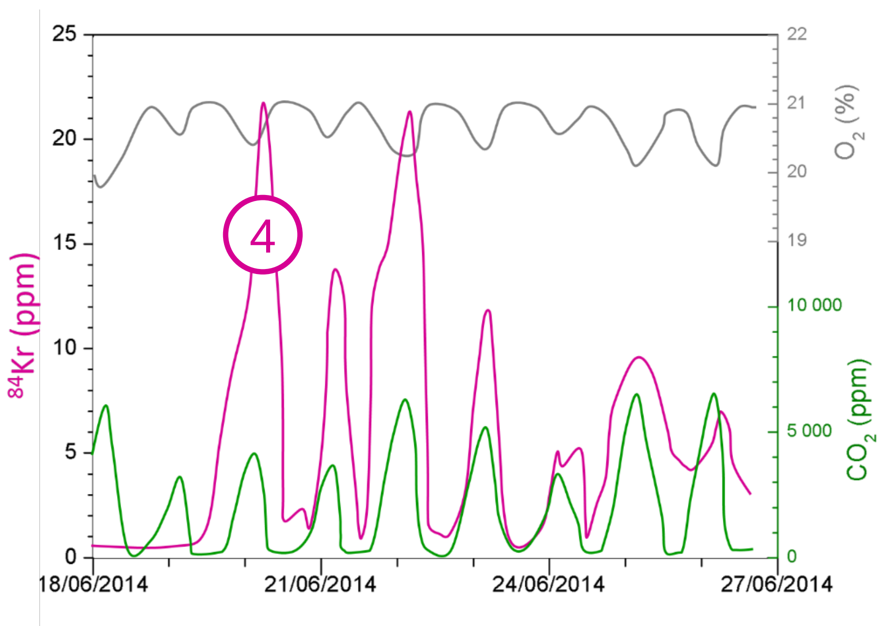
Krypton injection in the sub-surface

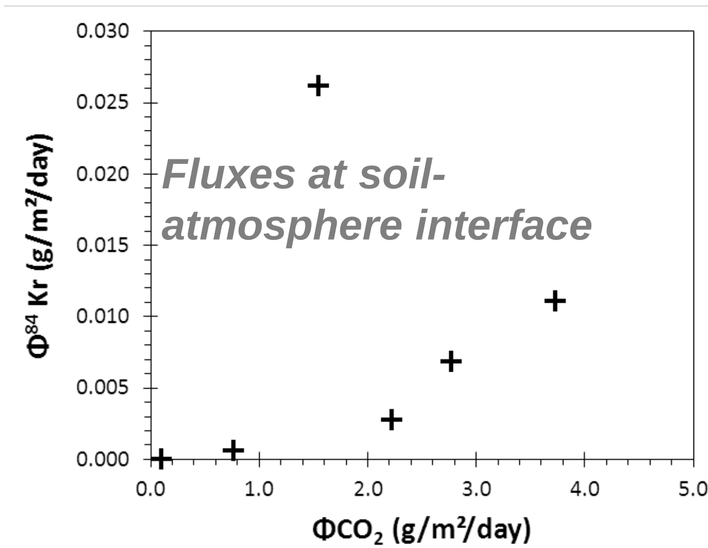




Time: 2 -30 h

Dilution : 10^3





SF_6 CO_2

^{222}Rn

R134a

^{3}He

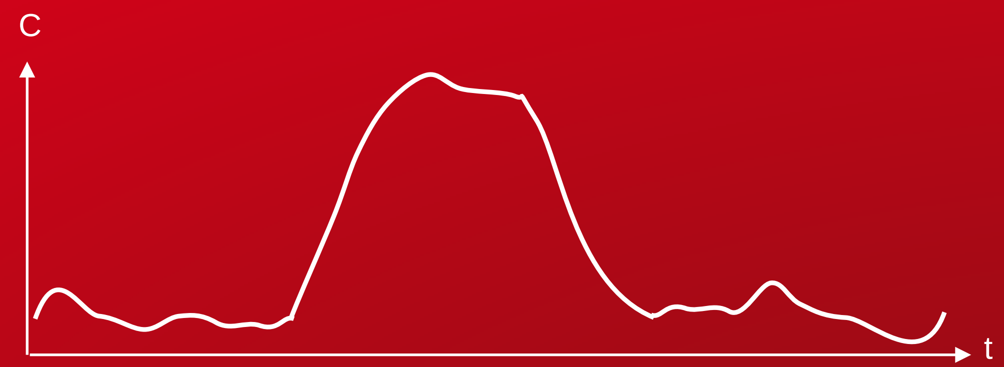
Xe

Kr

N_2O

CH_4

Detection:
Learning from the natural dynamics of gases in the unsaturated zone



High-resolution and long-term monitoring of gases in the unsaturated zone

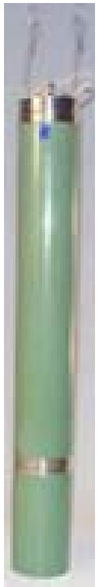
CO_2, $\delta^{13}C$, $\delta^{18}O$

LGR DLT-100
Isotope-Ratio Infrared Spectroscopy

Gas Bench / DeltaPlus^{XP}
Isotope-Ratio Mass Spectrometry (IPGP)

SF_6, CO_2, R134a, CH_4, NH_3
Photo-acoustic spectroscopy

Innova 1412



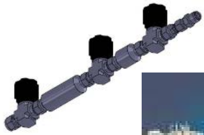
^{222}Rn
α spectroscopy

BMC2 & AlphaGuard

SF_6, CO_2, Xe, Kr

Prima Pro
& UGA200

$^3He/^4He$



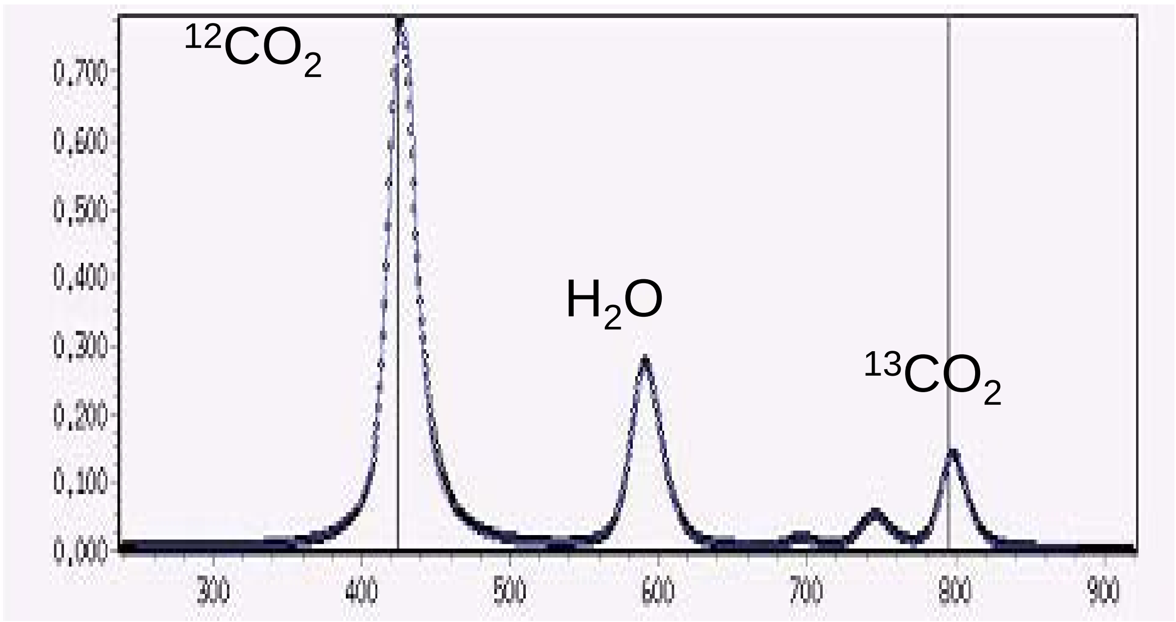
Noblesse (IPGP)

Mass spectrometry

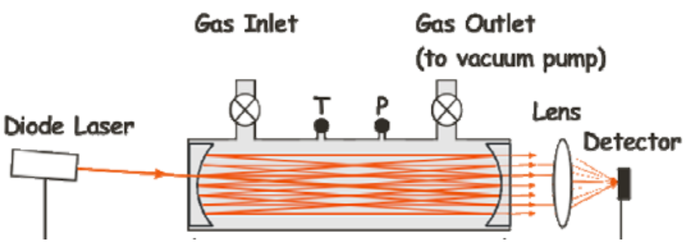
+ Meteorological and hydrological parameters

Absorbance

Mid infrared spectrum



Wavelength (Arbitrary Unit)



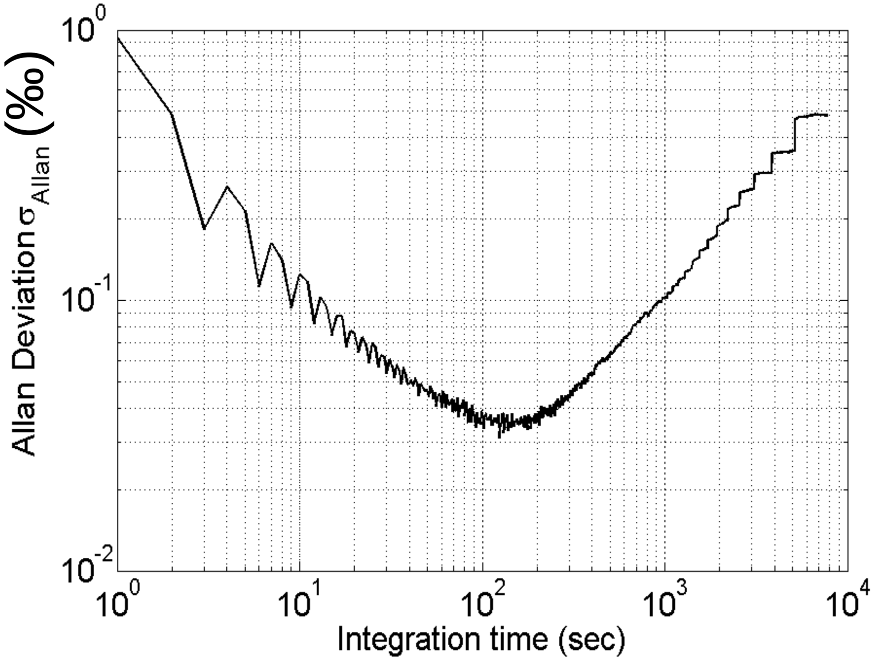
(from MacAlexander et al. 2010)

Los Gatos Research DLT-100

$$\delta^{13}C_{CO2}(‰)=\left(\frac{\left({}^{13}CO_2/{}^{12}CO_2\right)_{sample}}{\left({}^{13}CO_2/{}^{12}CO_2\right)_{VPDB}}-1\right)\times 1000$$

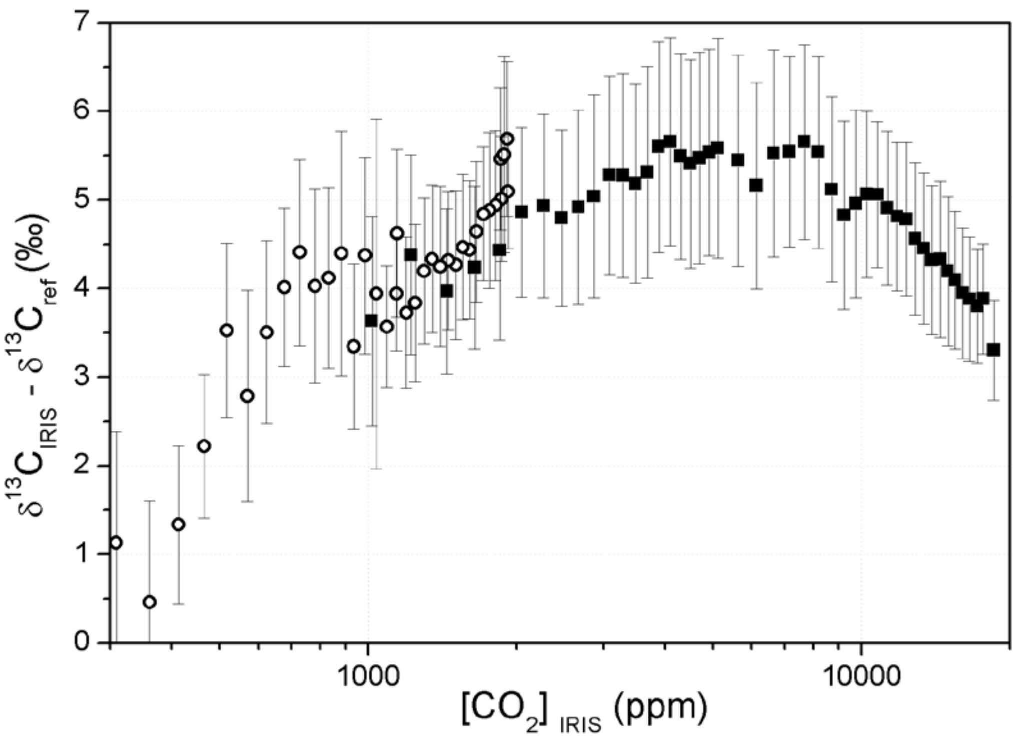
Isotope Ratio Infrared Spectroscopy
Performance assessment

✓ Accuracy depends on integration time, linearity and external temperature



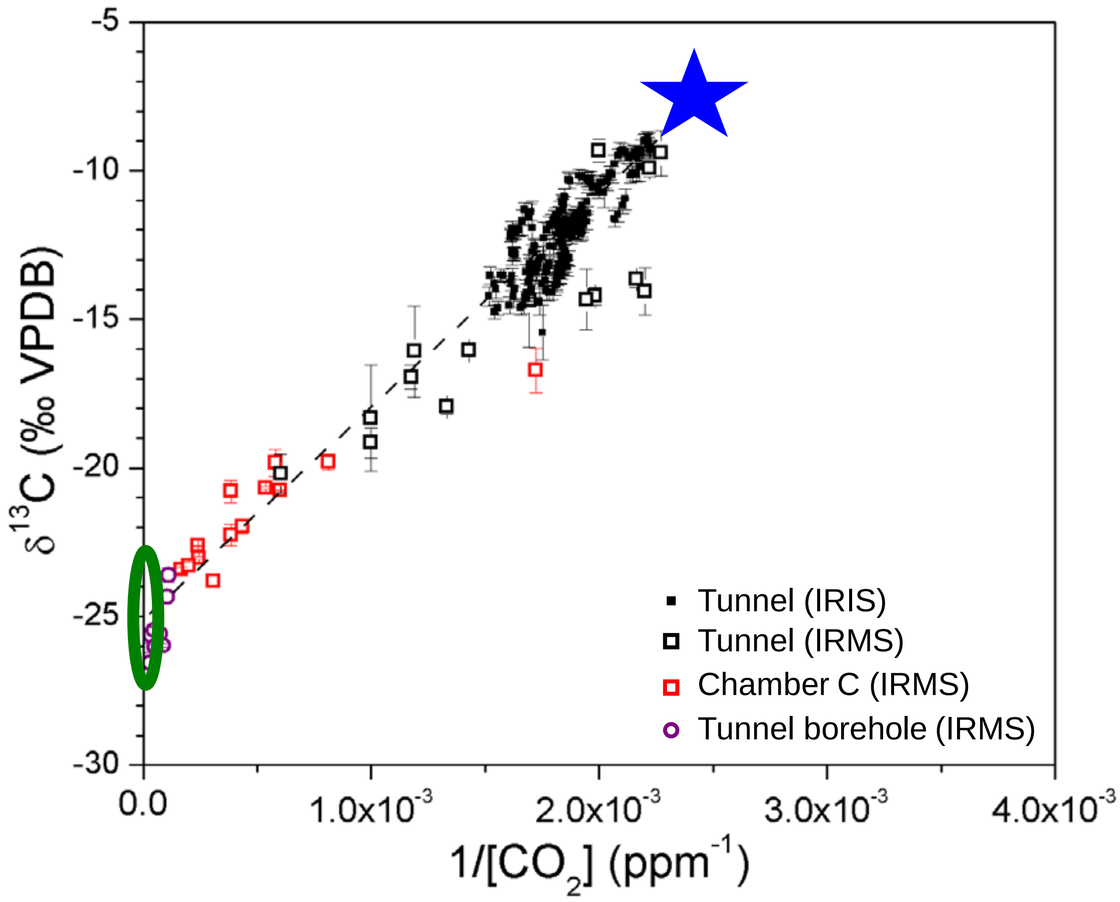
Integration time	Precision
1 s	0.9 ‰
60 s	0.05 ‰
200 s	0.04 ‰

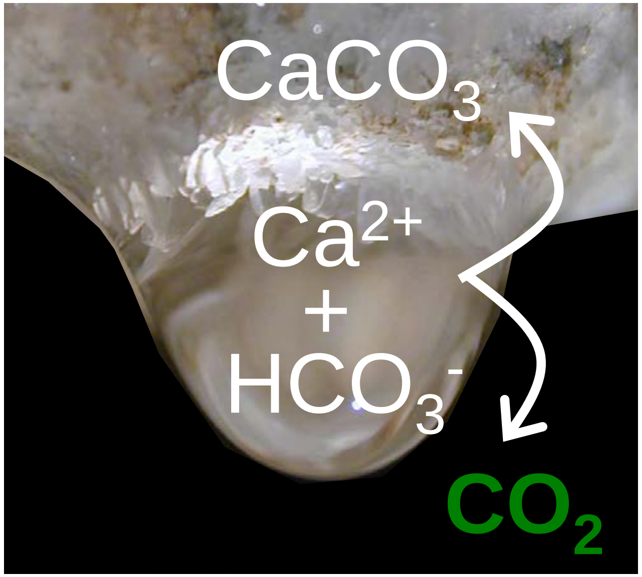
Concentration dependence



Guillon, S., Pili, E. and Agrinier, P., 2012. Using a laser-based CO_2 carbon isotope analyser to investigate gas transfer in geological media. Applied Physics B: Lasers and Optics, 107: 449-457.

Identification of CO_2 sources

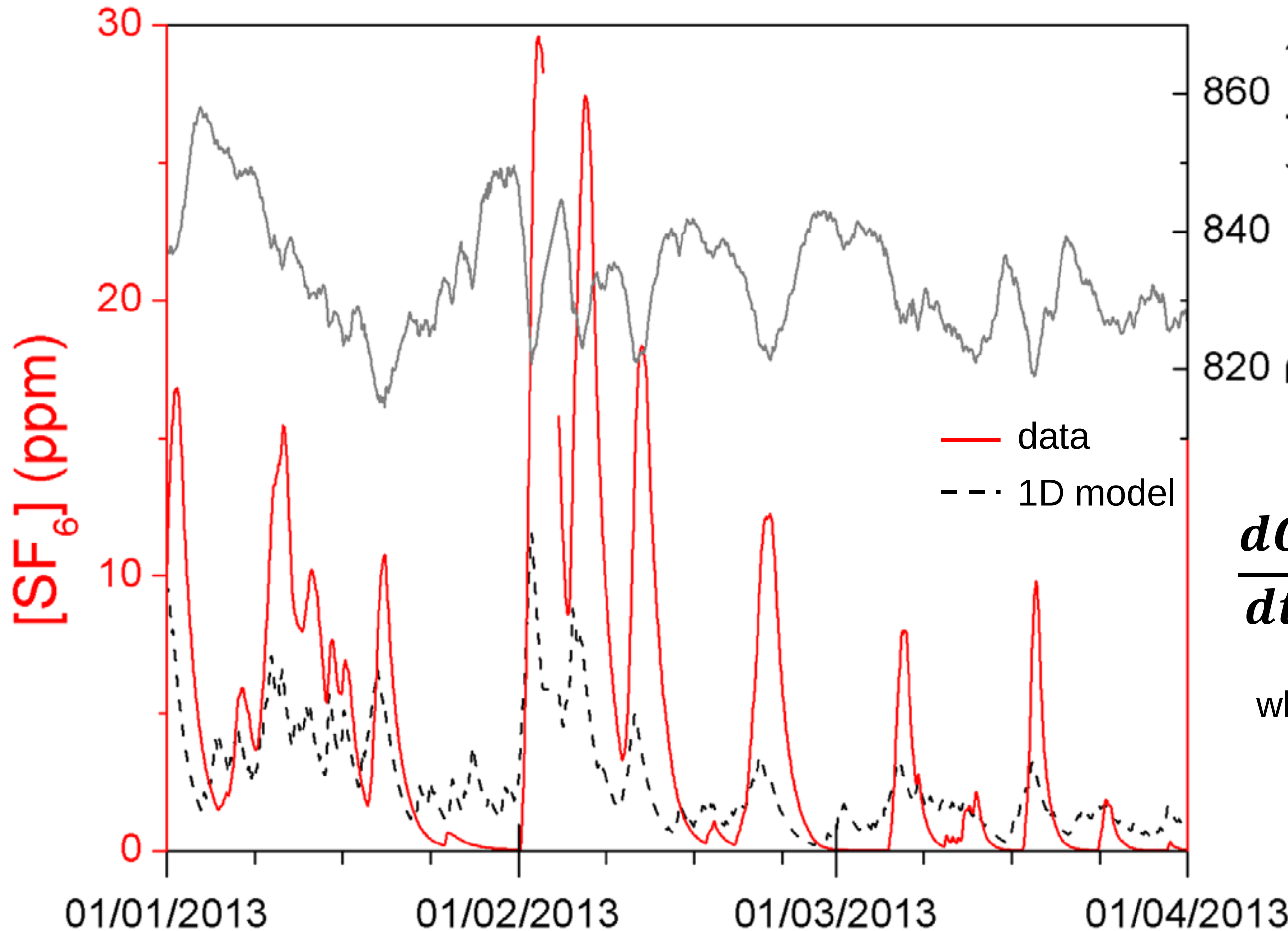


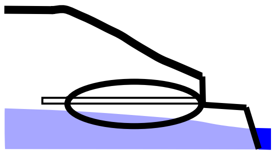


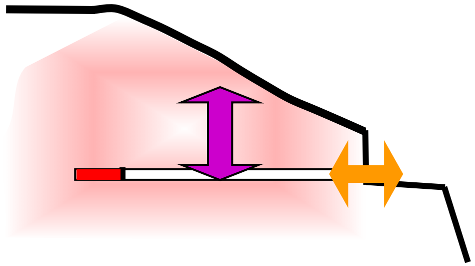
- ✓ Mixing: Atmosphere and Pore Space ([-27; -23 ‰])
- ✓ CO_2 degassing during calcite precipitation

Guillon et al. (2012)

Atmospheric pressure fluctuations modulate gas concentrations in the tunnel







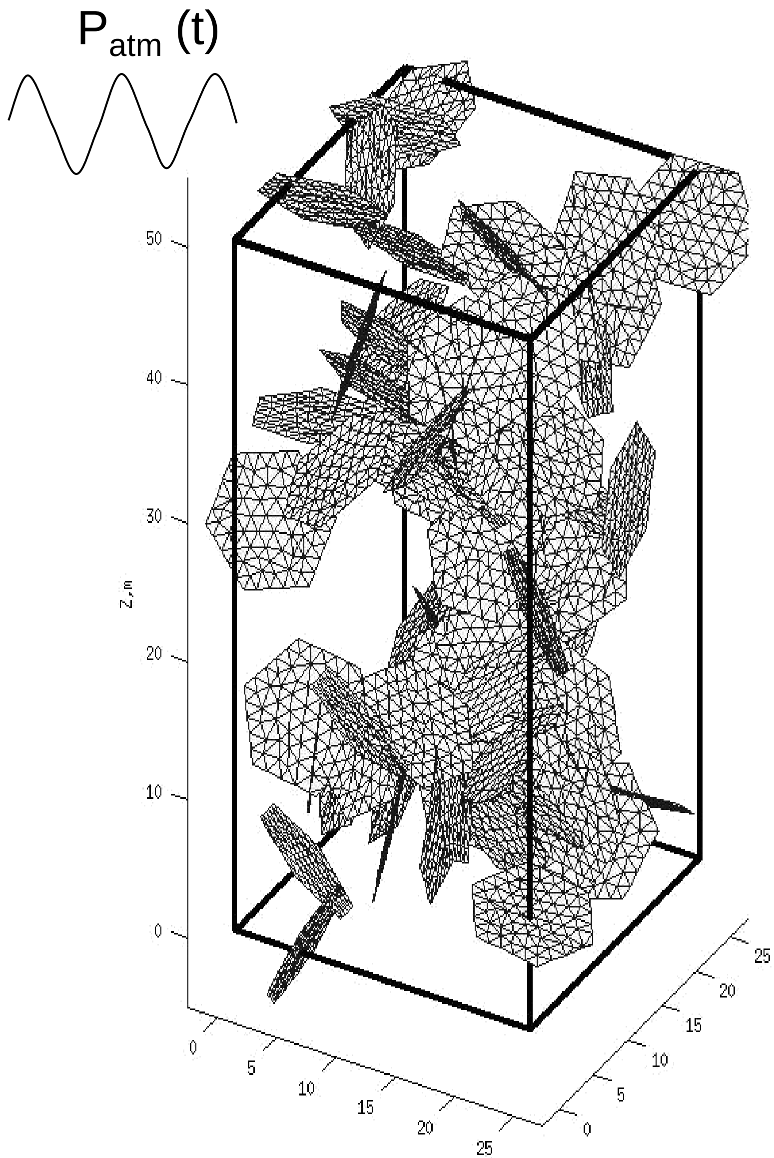
$$\frac{dC}{dt} = \frac{S}{V}\frac{k}{\mu}C_r\frac{\Delta P(t)}{L} - \lambda_V C(t)$$

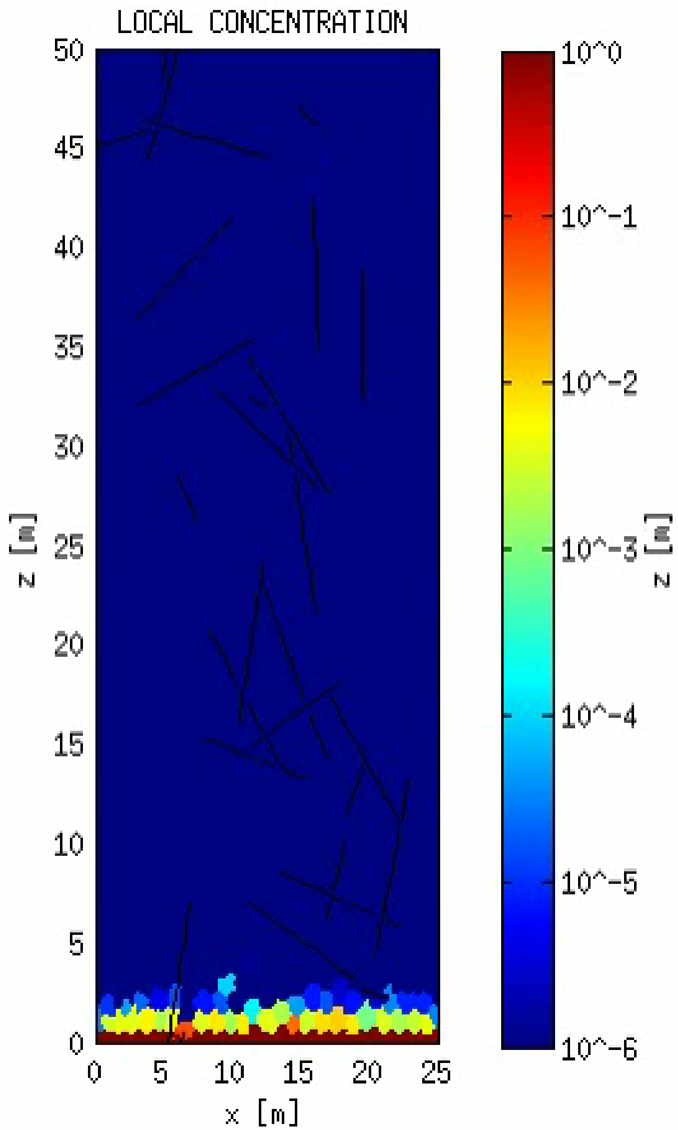
where $\Delta P(t) = P_r(t) - P_{atm}(t)$

and $\frac{\partial P_r}{\partial t} = \frac{kP_0}{\varepsilon\mu}\frac{\partial^2 P_r}{\partial z^2}$

- ✓ SF_6 peaks driven by pressure lows
- ✓ Advection due to barometric pressure, tunnel ventilation (1D model)

Barometric pumping drives tracer migration





From Patriarche et al. (2007)

From P.M. Adler (pers. com.)

Barometric pumping is not always efficient

after 50 days

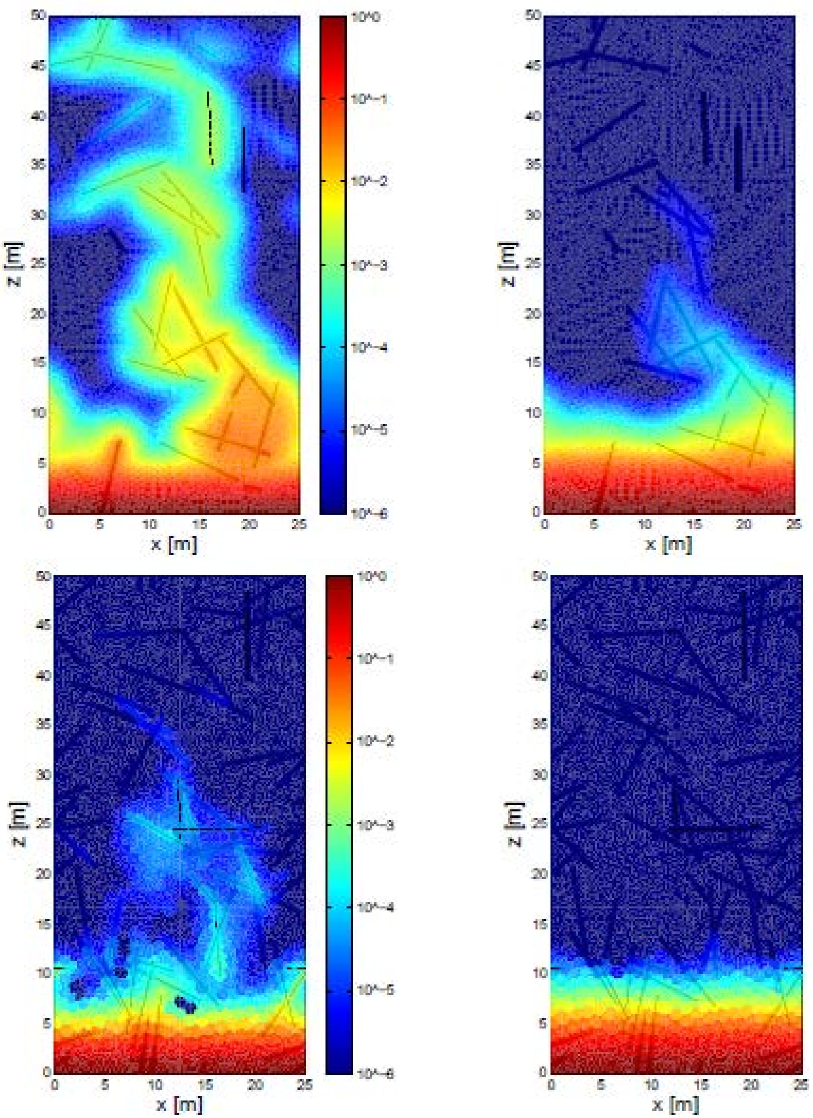
Matrix porosity

ε=1%   ε=5%

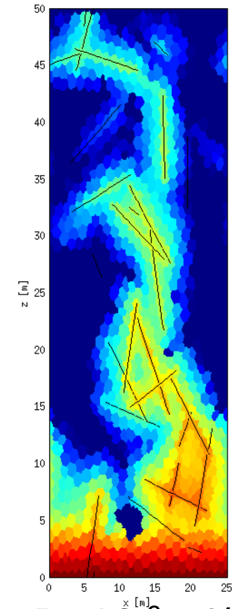
Fracture density

ρ=2.3

ρ=6

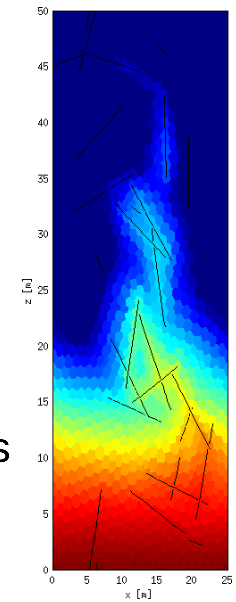


Diffusion coefficient



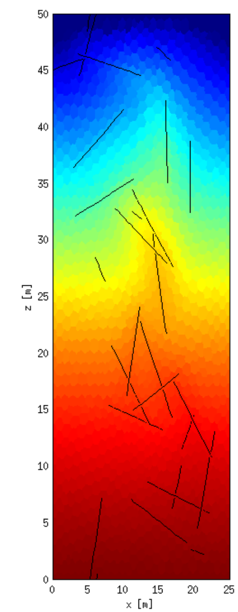
$D=10^{-6}\ m^2/s$

SF_6, Xe



$D=10^{-5}\ m^2/s$

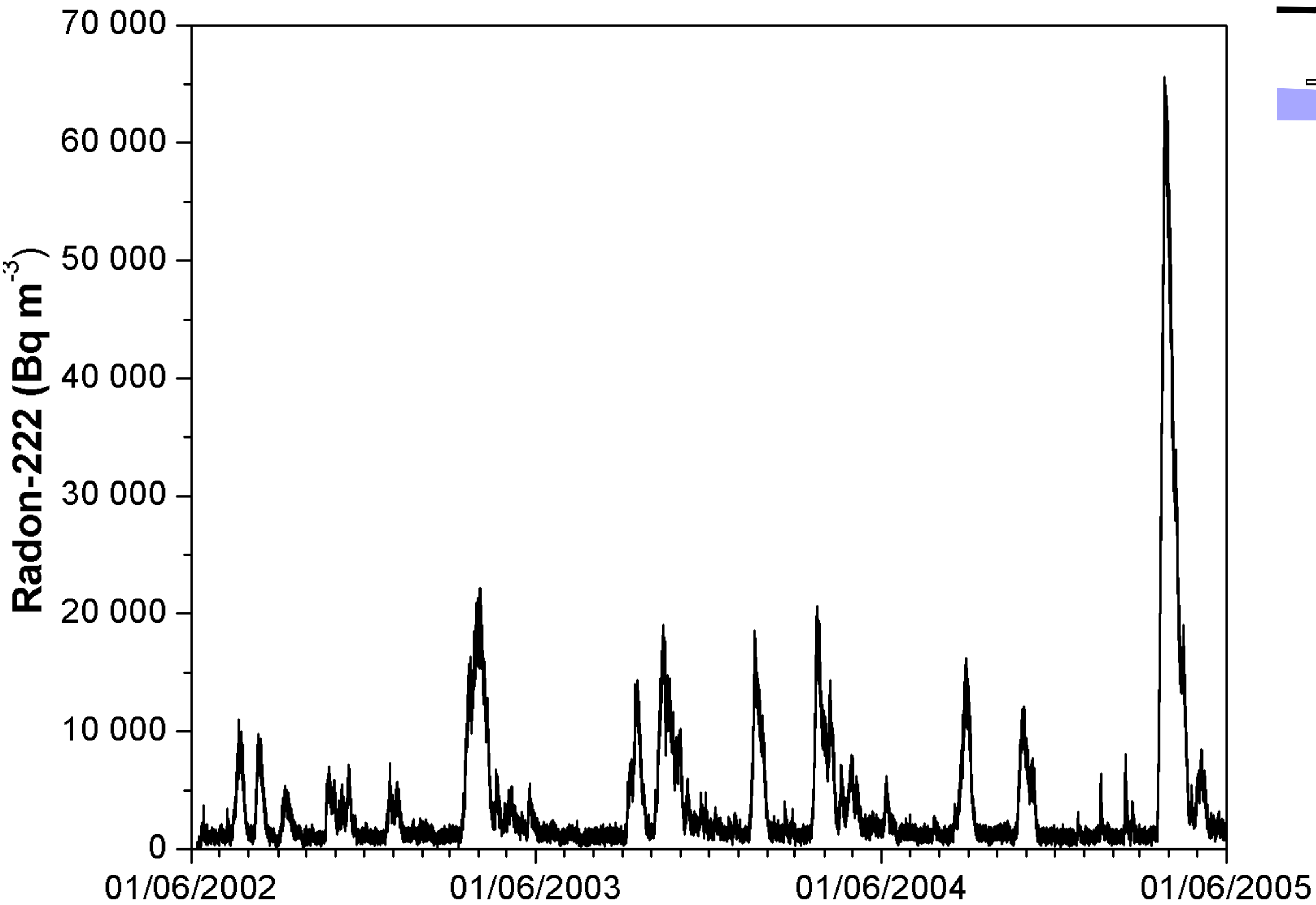
He

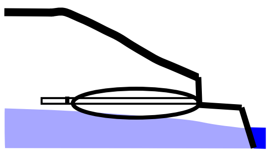


$D=6.10^{-5}\ m^2/s$

From Mourzenko et al. (2014)

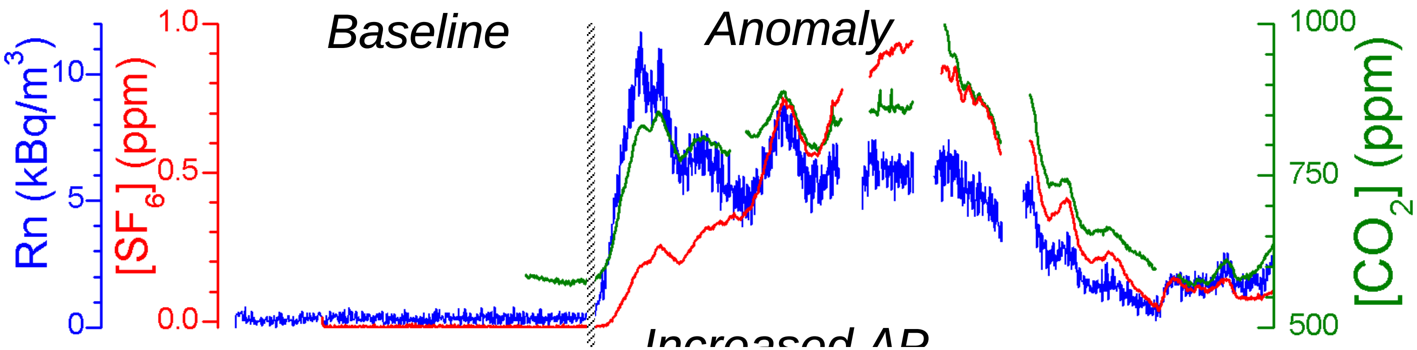
Background dynamics vs. large gas anomalies

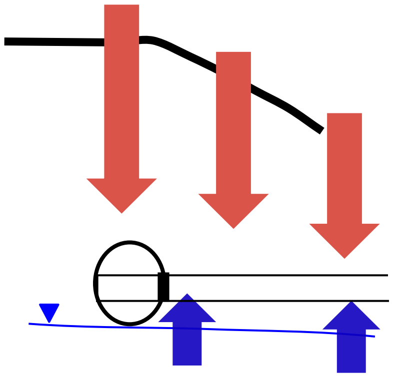




P. Richon (pers. com.)

Anomalies result from an increased gas flow



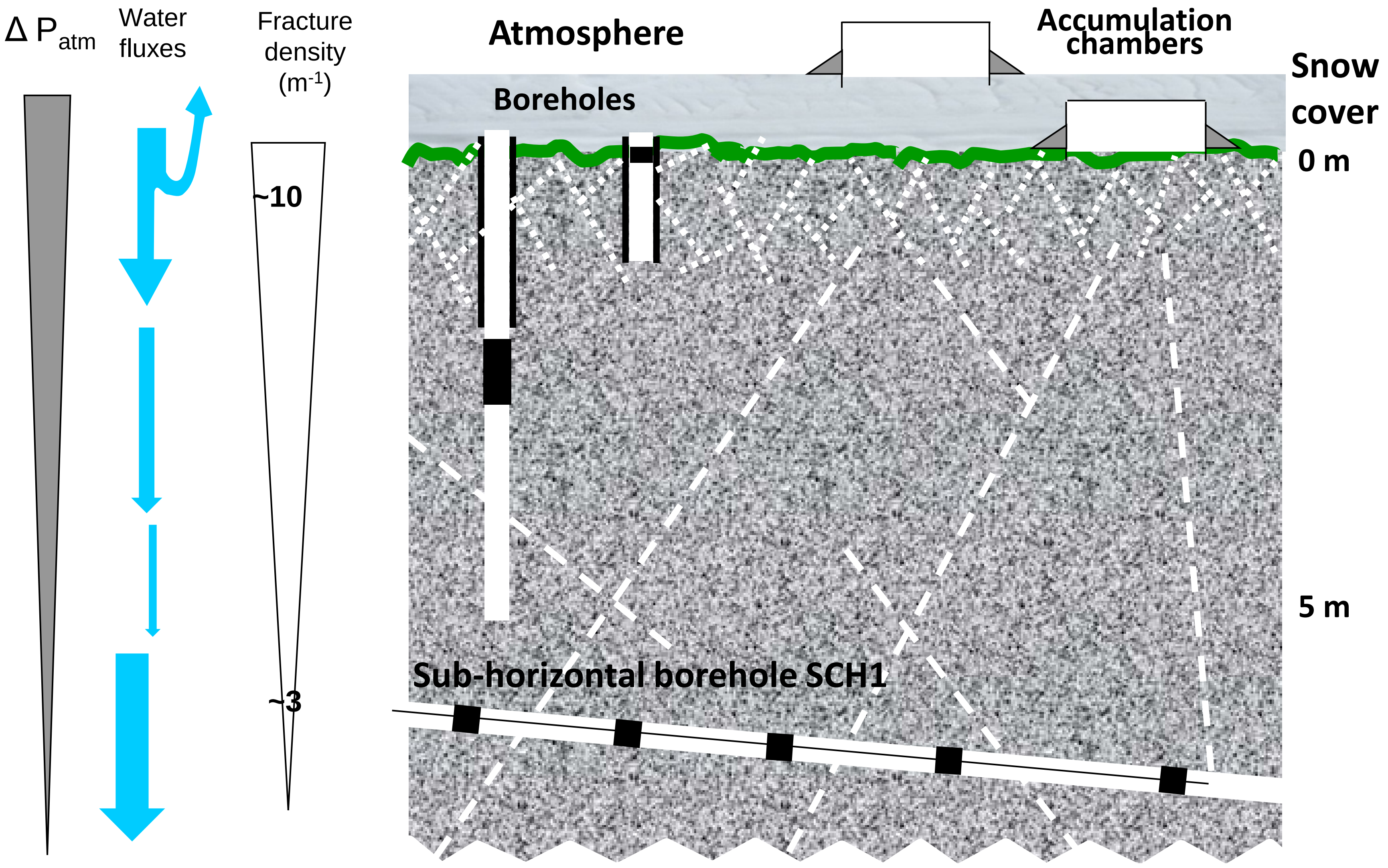


- ✓ Anomaly is not source dependent = flow increase (k or ΔP ?)
- ✓ Increased ΔP, driven by water movements

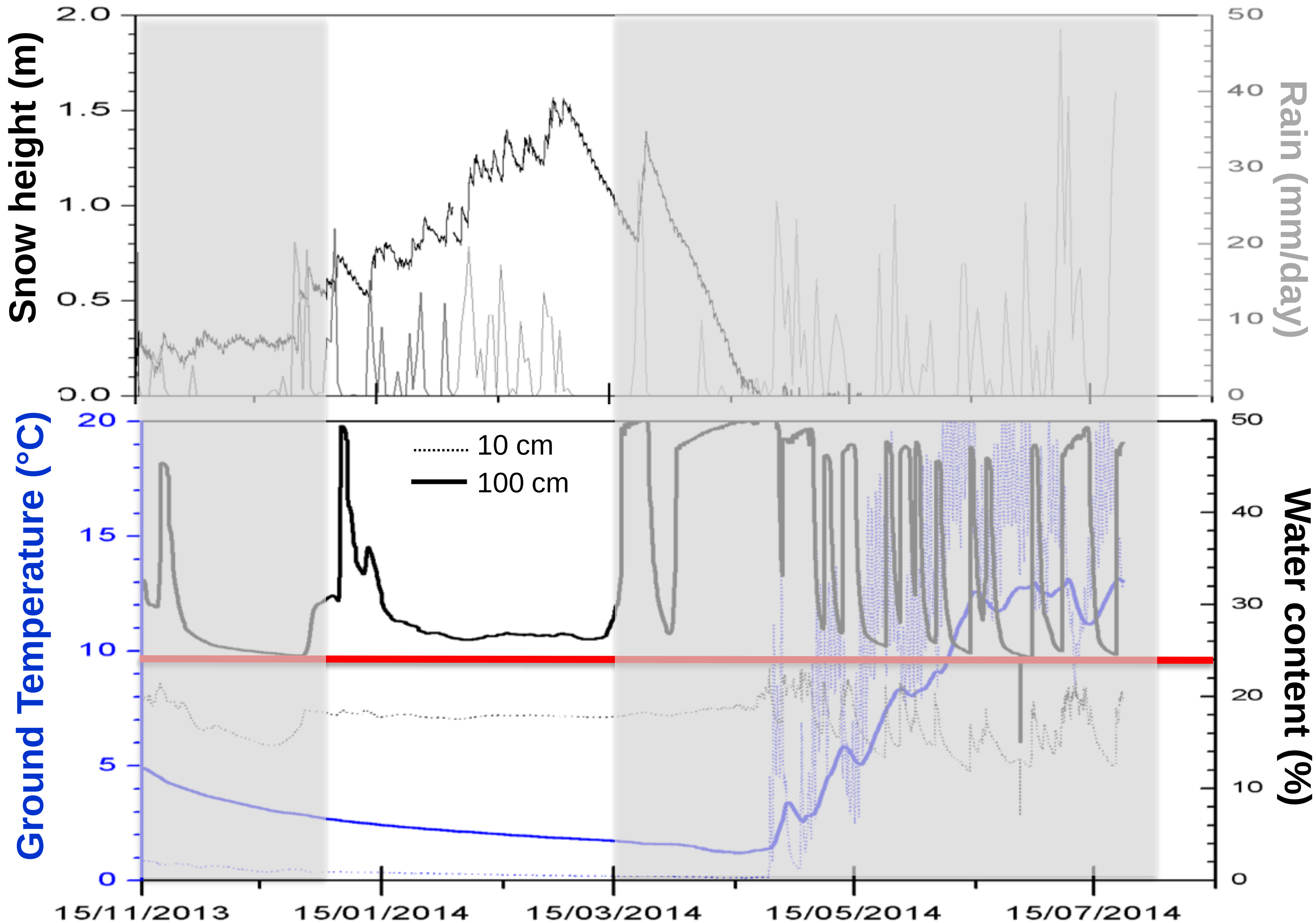
Gas migration at the Geosphere-Atmosphere interface:

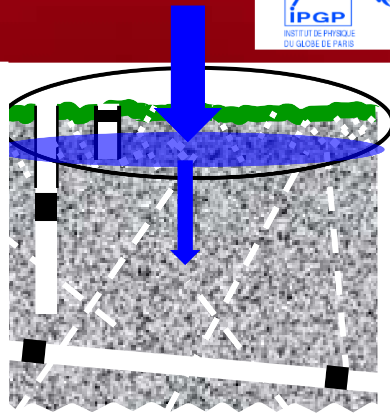
Bio- meteo- geo- hydrology

Gases in the Geosphere-Atmosphere interface



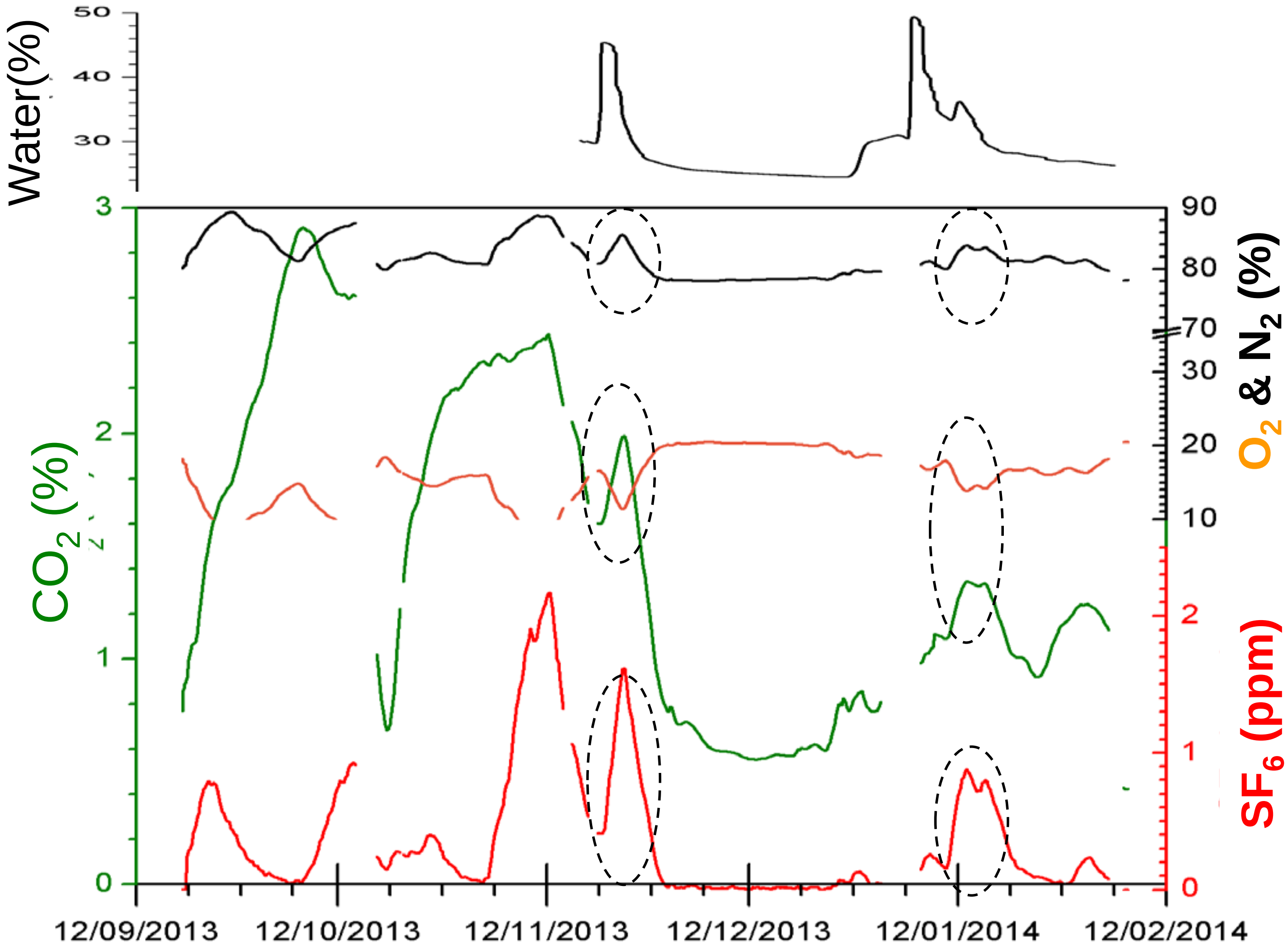
Water fluxes in the sub-surface and gas migration

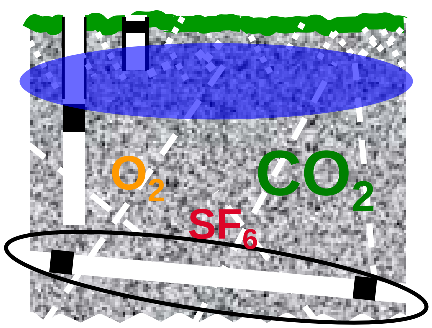




- ✓ Thick snow cover, unfrozen soil with limited water infiltration
- ✓ Water and Heat fluxes

Biological activity produces / consumes gases





+ **CH_4** (5-10 ppm)
N_2O (1 - 9 ppm)

- ✓ Dynamics of O_2 / CO_2
  control by moisture and O_2 availability (constant T at depth)
- ✓ Consequences on inert gas migration (noble gases, SF_6 tracer gas)

Migration of gases in unsaturated fractured porous media

- What are the **driving forces** of gas migration and their respective influences ?
  - ✓ Advection, Diffusion & Barometric pumping
  - ✓ Water infiltration
- What are the **dilution** and **temporal delays** between production of a tracer at depth and breakthrough the surface?
  - ✓ Only 10 to 50 h for 50 m migration
  - ✓ **Dilution** in the range $10^3 – 10^6$ for 50 m migration
- How do **water fluxes** affect gas migration in the unsaturated zone?
  - ✓ Piston like displacement
  - ✓ Solubility & degassing
- How to measure and understand **biogenic** gas dynamics?
  - ✓ Temperature, moisture, reactive / inert gases
  - ✓ **Oxygen depletion** and partial pressures

Transferts d'eau et de gaz sous couvert neigeux

Sophie Guillon, Florent Barbecot

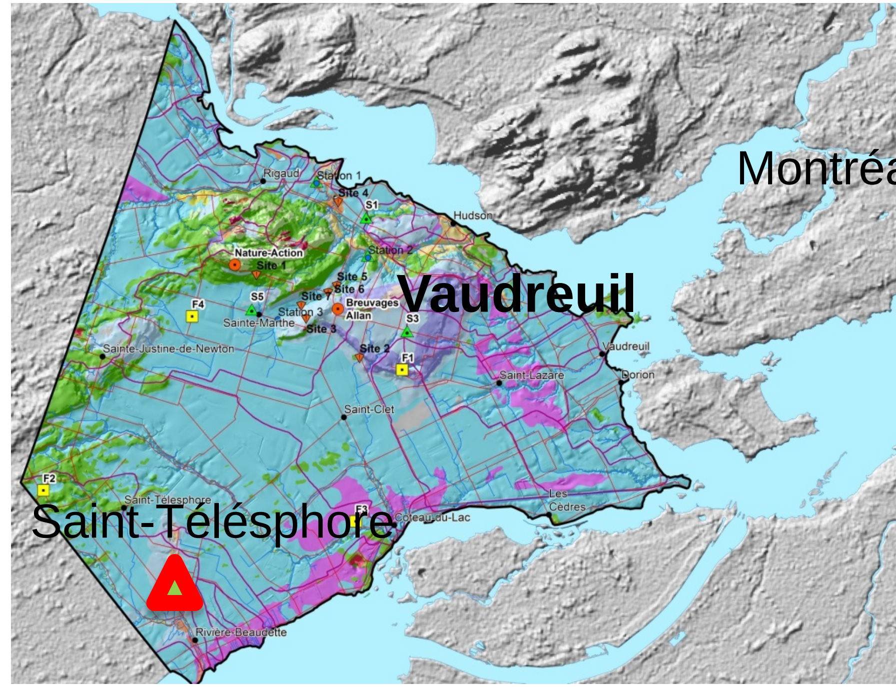
Marie Larocque, Daniele Pinti, Éric Pili

Site expérimental Sablière St-Télesphore

- Infrastructure de recherche sur la recharge des eaux souterraines (IRRES)

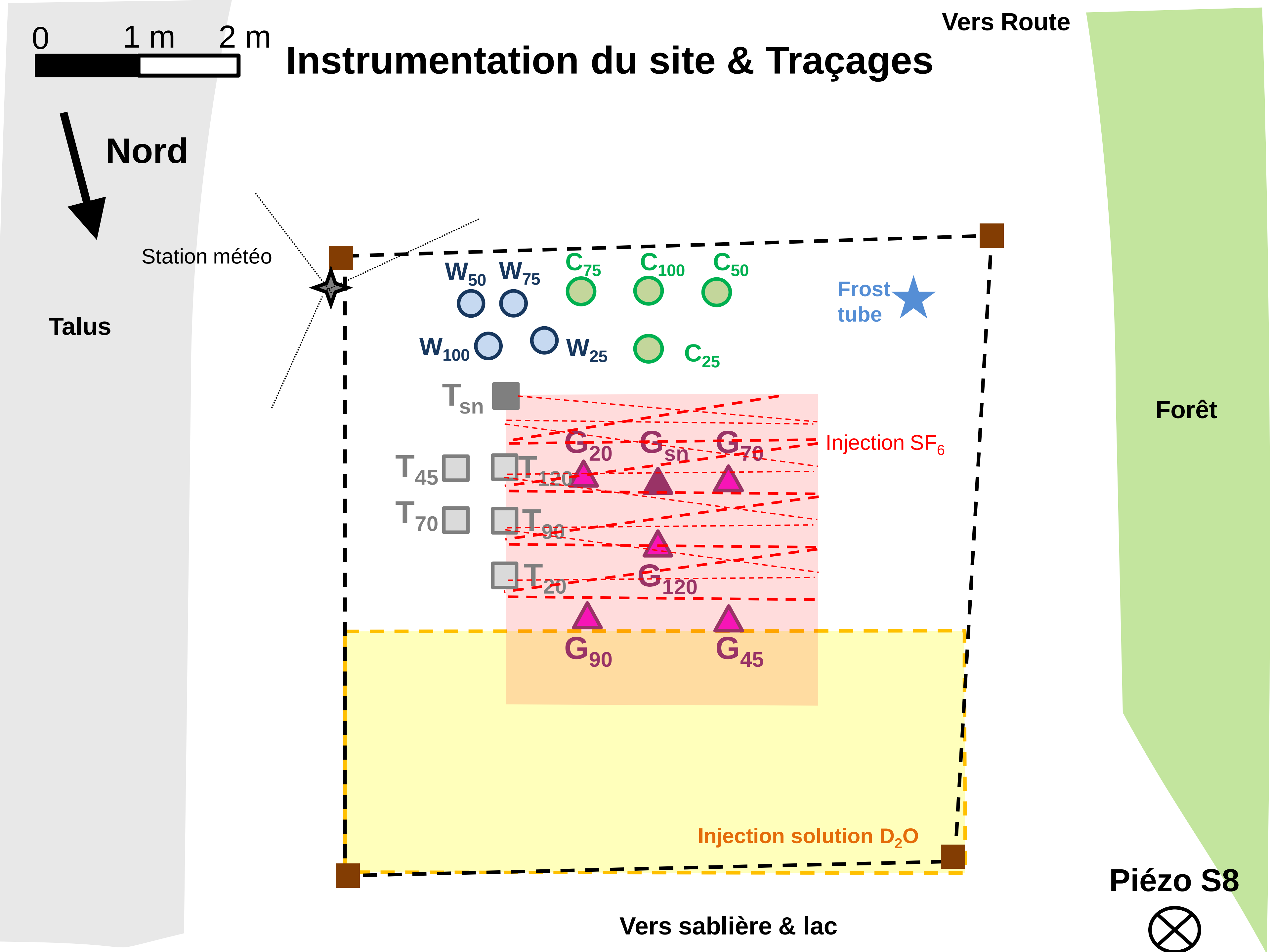
- **Esker**

dépôts fluvio-glaciaires, sable ~ homogène



Piézomètre PACES (S8)

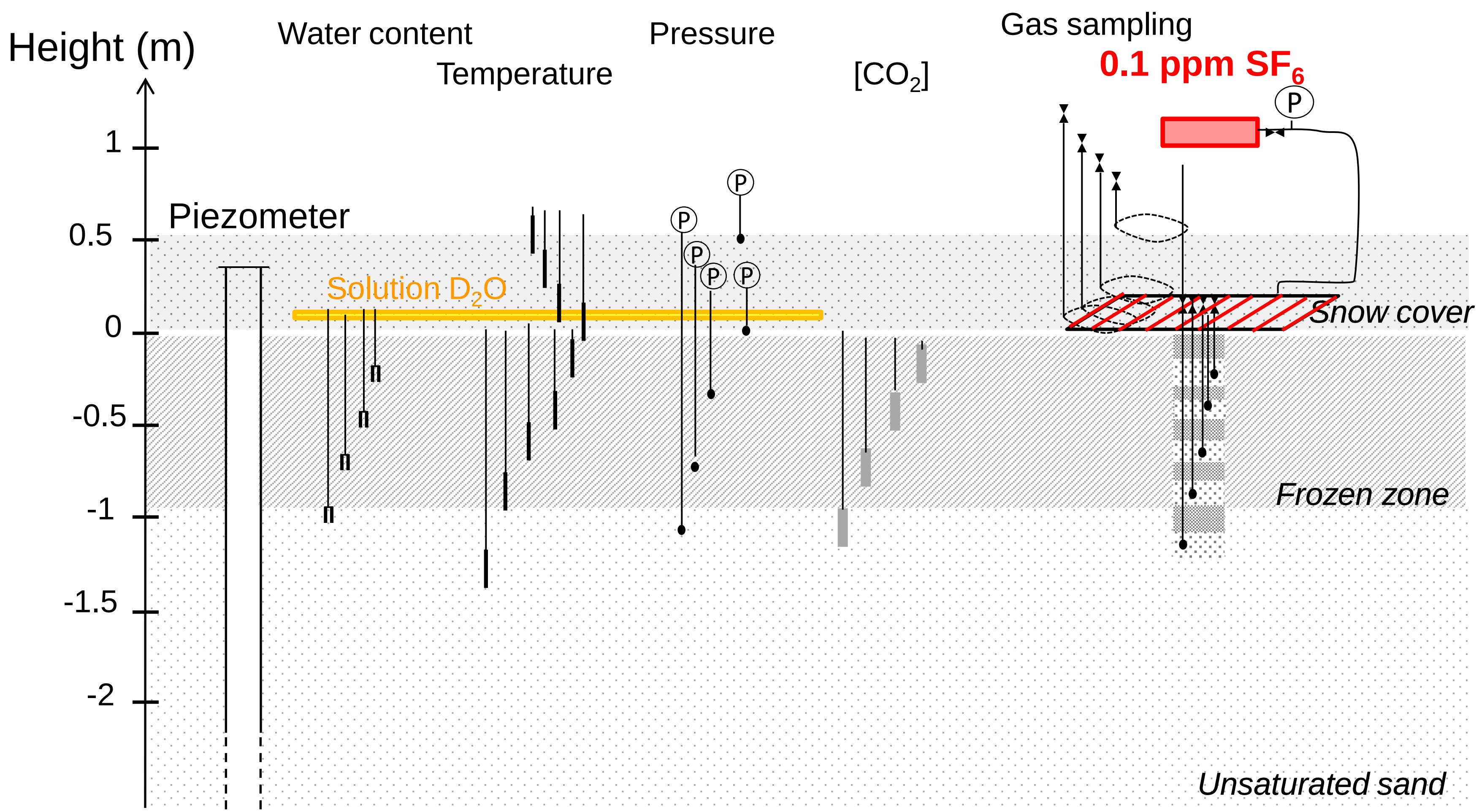
K ~ 10^{-5} m/s
k ~10^{-12} m²



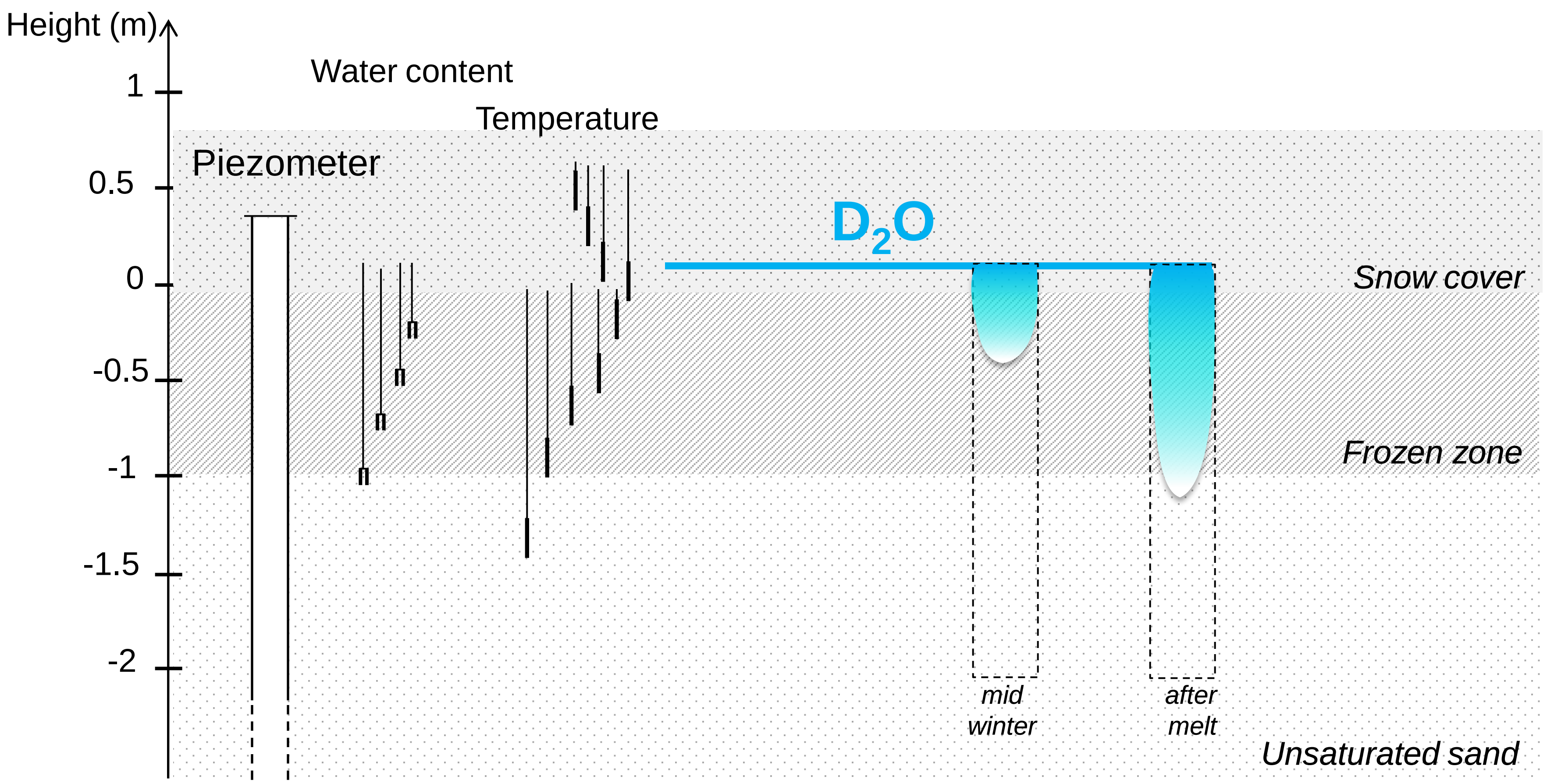
0
1 m
2 m
Instrumentation du site & Traçages
Vers Route
Nord
Station météo
Talus
W_{50}
W_{75}
C_{75}
C_{100}
C_{50}
Frost tube
W_{100}
W_{25}
C_{25}
T_{sn}
G_{20}
G_{sn}
G_{70}
Injection SF_6
T_{45}
T_{120}
T_{70}
T_{90}
T_{20}
G_{120}
G_{90}
G_{45}
Forêt
Injection solution D_2O
Piézo S8
Vers sablière & lac

Instrumentation du site & Traçages

Bilan Instrumentation & Traçages

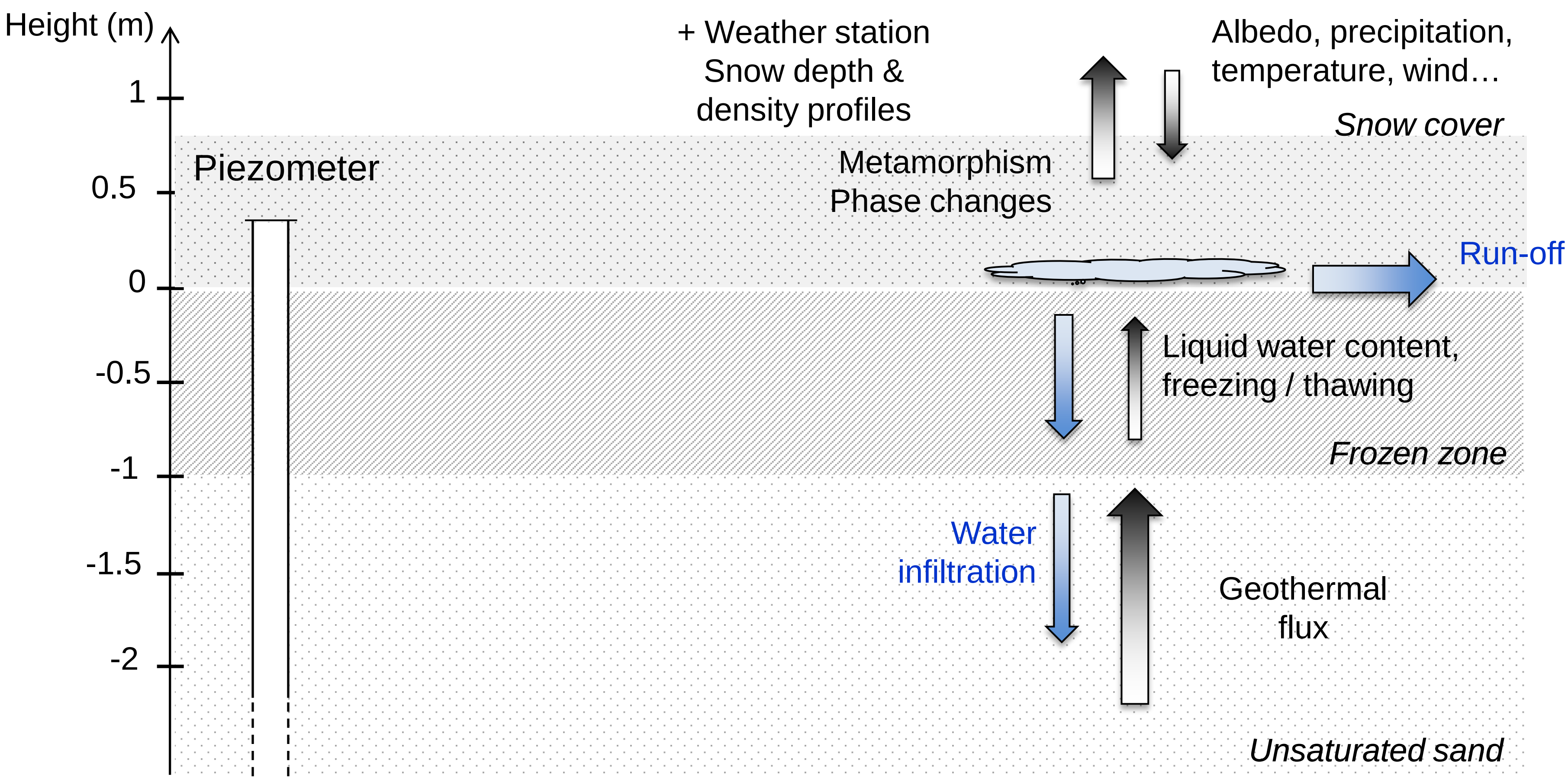


Traçage au D_2O: infiltration d'eau sous couvert neigeux



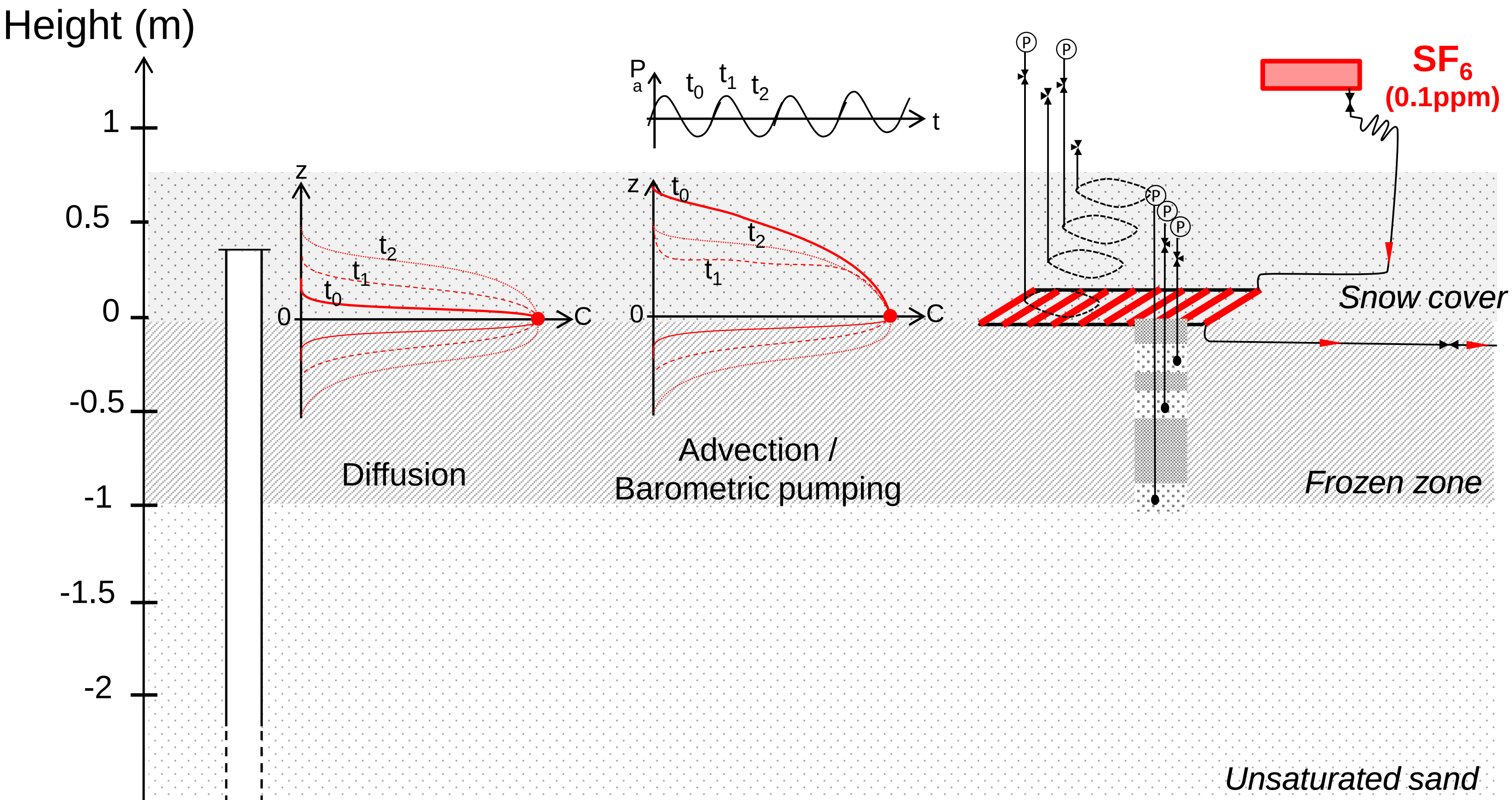
- ✓ Existence d'une infiltration diffuse pendant l'hiver
- ✓ Profondeur du front d'infiltration pendant l'hiver / au printemps
- ✓ Bilan isotopique et redistribution de la vapeur d'eau dans la neige

Interprétations: bilans des flux d'eau & d'énergie



- ✓ Flux géothermique, échanges avec l'atmosphère, isolation par la neige
- ✓ Fonte de la neige et/ou du sol
- ✓ Eau disponible pour l'infiltration et/ou le ruissellement
- ✓ Influence sur le cycle des nutriments et la qualité de l'eau souterraine

Traçage au SF_6: flux de gaz dans la neige et le sol gelé



- ✓ Migration du SF_6 par diffusion ou pompage barométrique ?
- ✓ Evolution propriétés pneumatiques (perméabilité, porosité) de la neige au cours de l'hiver?
- ✓ Evolution temporelle des flux de GES (CO_2, N_2O, CH_4) sous couvert neigeux, existence d'une bouffée de gaz lors de la fonte?

Conclusion

✓ **Gases migrate in rocks…**

through conductive fracture network

due to barometric pumping,
liquid phase displacement
biological reactions

✓ **Gas migration in unsaturated fractured rocks matters for…**

- biochemistry of recharge and water resources
- recording of climate and recharge conditions in gas and liquid phases
- detection of a gas leakage from a deep source
- greenhouse gas fluxes

✓ **Water and gas fluxes through snow cover and frozen / thawed soil**

- timing and intensity of groundwater recharge
- greenhouse gas budget