Physical, Chemical and Biological Controls on Surface-gas Fluxes Quantified With High-resolution Monitoring of Multiple Tracers

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Abstract

In the subsurface, water content, gas solubility, adsorption on minerals and chemical reactions control gas fluxes between soil and the atmosphere. Because these processes vary in intensity both in time and space, it is very challenging to quantify emissions, specifically when flux measurements are used for detection, identification or monitoring of a subsurface gas source. An experimental setup for gas percolation though soil column experiments under well-controlled conditions was developed and validated at the ECOTRON IleDeFrance research center. Its design included the effect of: i) watering/evaporation cycles, ii) barometric pressure, iii) injection pressure, iv) tracer behaviors and v) plant metabolism. To better understand subsurface processes controlling gas fluxes, we studied transport of multiple tracers across soil columns using long-term and high-resolution monitoring thanks to online low-flow mass-spectrometry. We injected tracer gases into columns containing different porous media, pure sillica sand and zeolite. This set-up allowed us to evaluate the relative contribution of diffusion, solubility and adsorption on various trace gases (SF6, noble gas including Xe). All the experimental data are discussed in conjonction with simulations using the NUFT unsaturated flow and transport code.

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I- BACKGROUND RESEARCH: Gas fluxes at the soil-atmosphere interface

Aim of the research

Gas transport in soils is **highly variable in space and time** leading to **modulations of gas fluxes** at the **soil-atmosphere interface** that must be understood

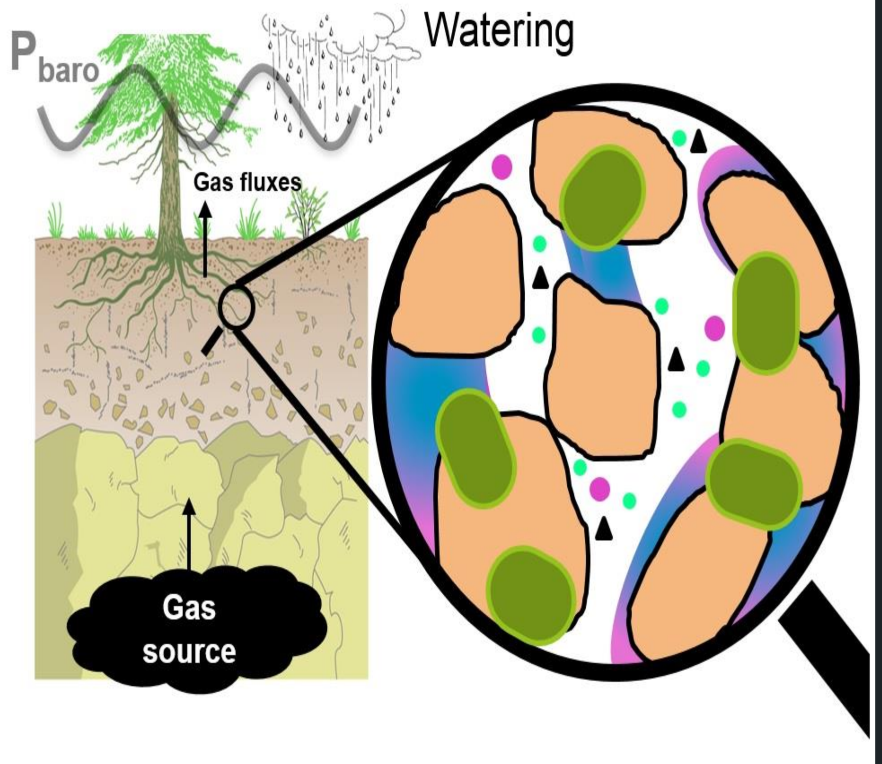
Flux variability due to

- Nature and localization of gas source
- Soil permeability and porosity
- Barometric pressure fluctuations
- Water content and capillary pressures
- Respiration and biomass degradation

Applications

Discrete **flux measurements are integrated in space and/or time to detect, identify or monitor subsurface gas sources** such as:

CO_2 sequestration reservoir, volcanic emissions, carbon release from permafrost thaw, volatile contaminant plumes, shale gas production or underground nuclear explosions



II- EXPERIMENTAL SET-UP: Long-term and high-resolution monitoring

Controlled experimental conditions

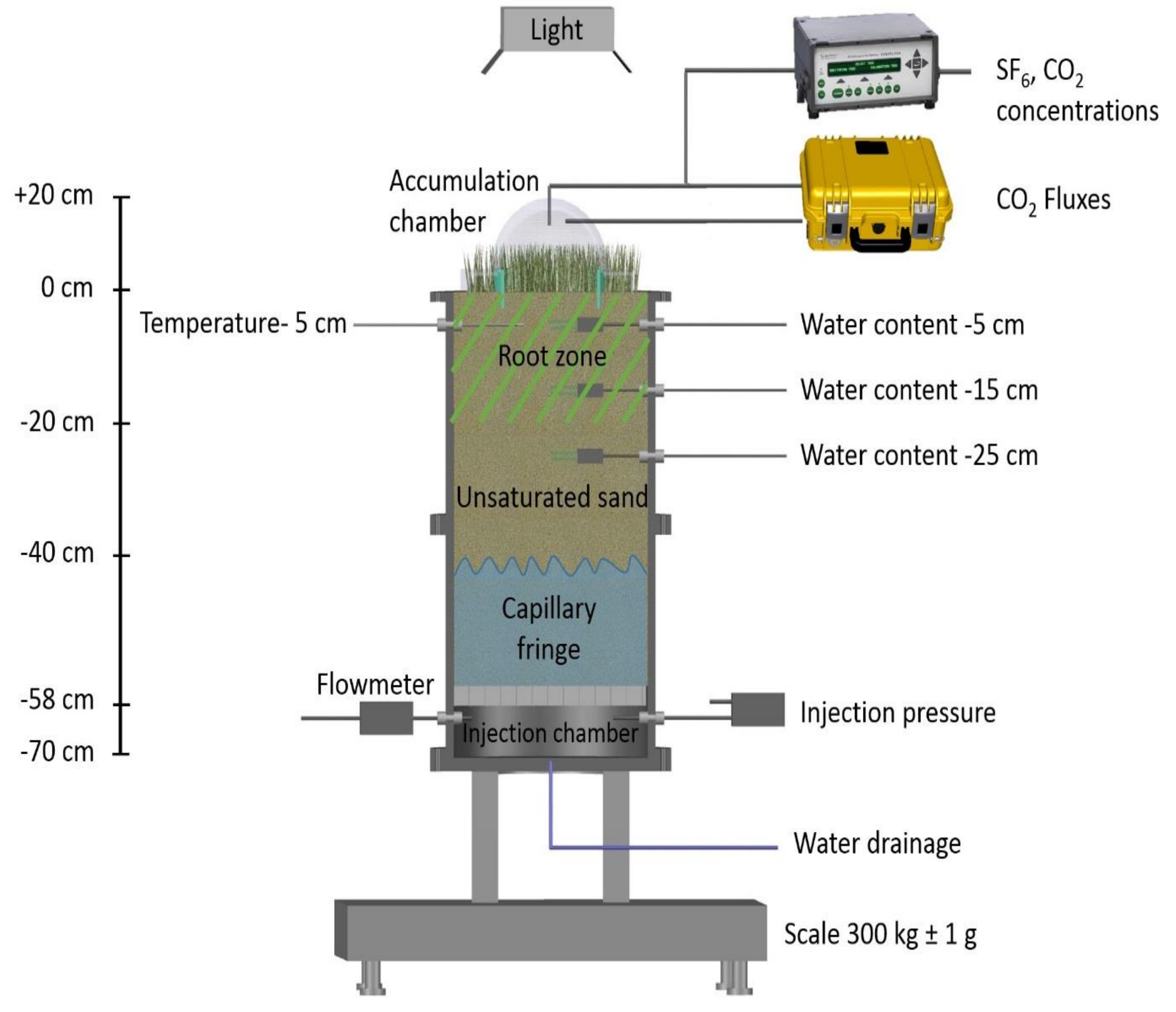
- In climatic chamber at the ECOTRON-IDF
- Constant temperature
- Barometric pressure
- Atmosphere renewal
- Diurnal light cycle and Periodic watering

Unsaturated soil column experiment

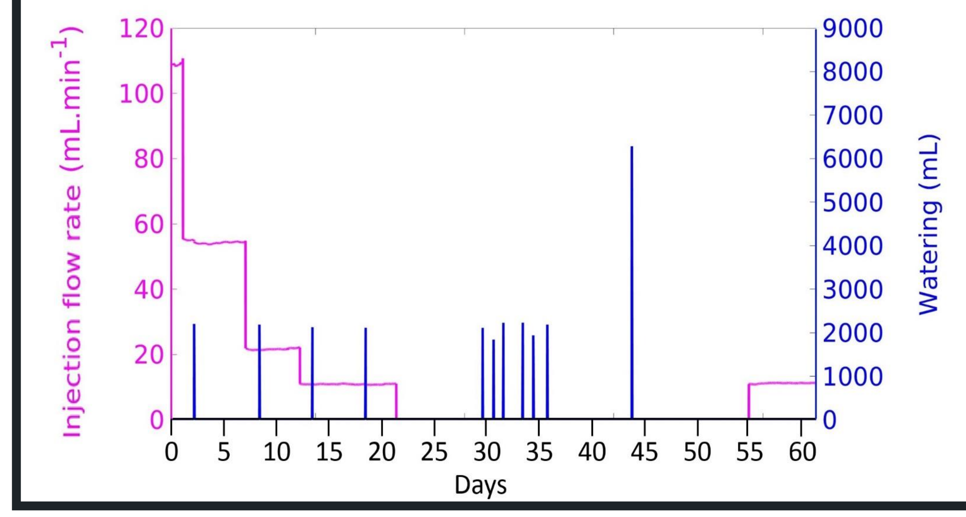
- Pure Fontainebleau sand
- 2 Columns: 1 with bare soil, 1 with plants
- Unsaturated porous media with water
- Water content profile monitoring

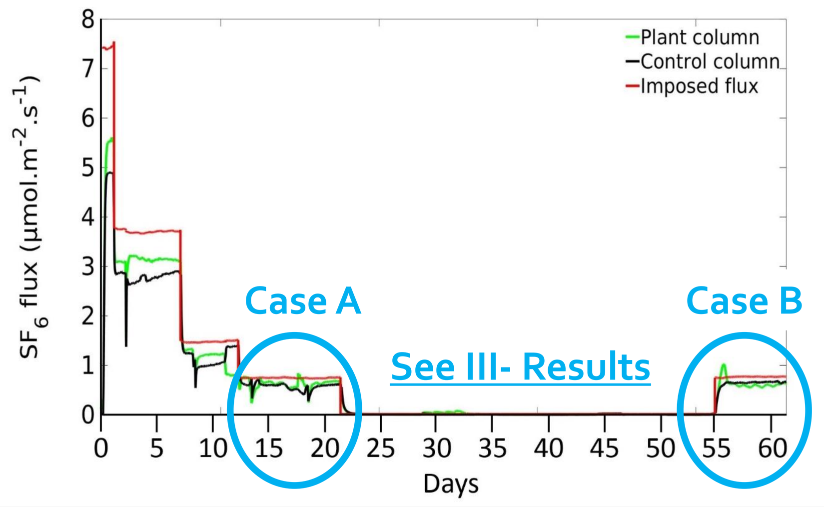
Gas percolation and flux measurements

- Constant injection flow-rate of 10,000ppm SF_6 in N_2,O_2
- Pressure gradient monitoring between injection chamber and atmosphere
- Flux measurements of SF_6 and CO_2 by accumulation chamber



More than 60 days of experimentation, Flux measurements at 1 hour time-step, Experimental conditions monitoring at 5 minutes time-step

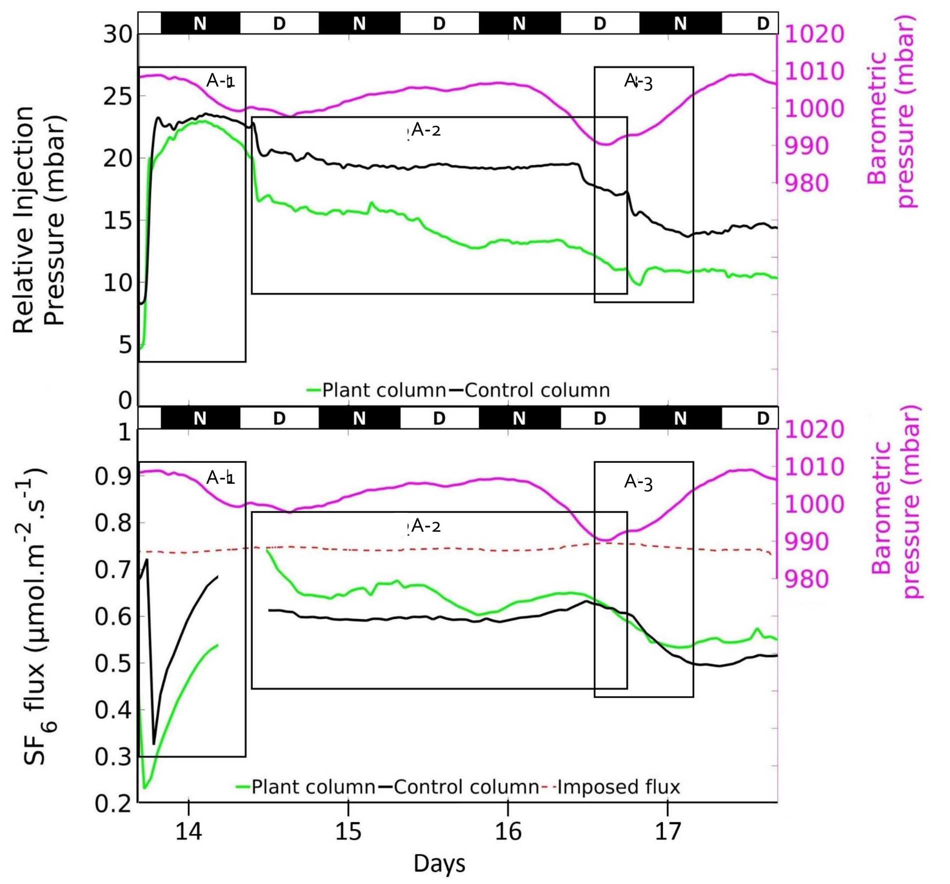




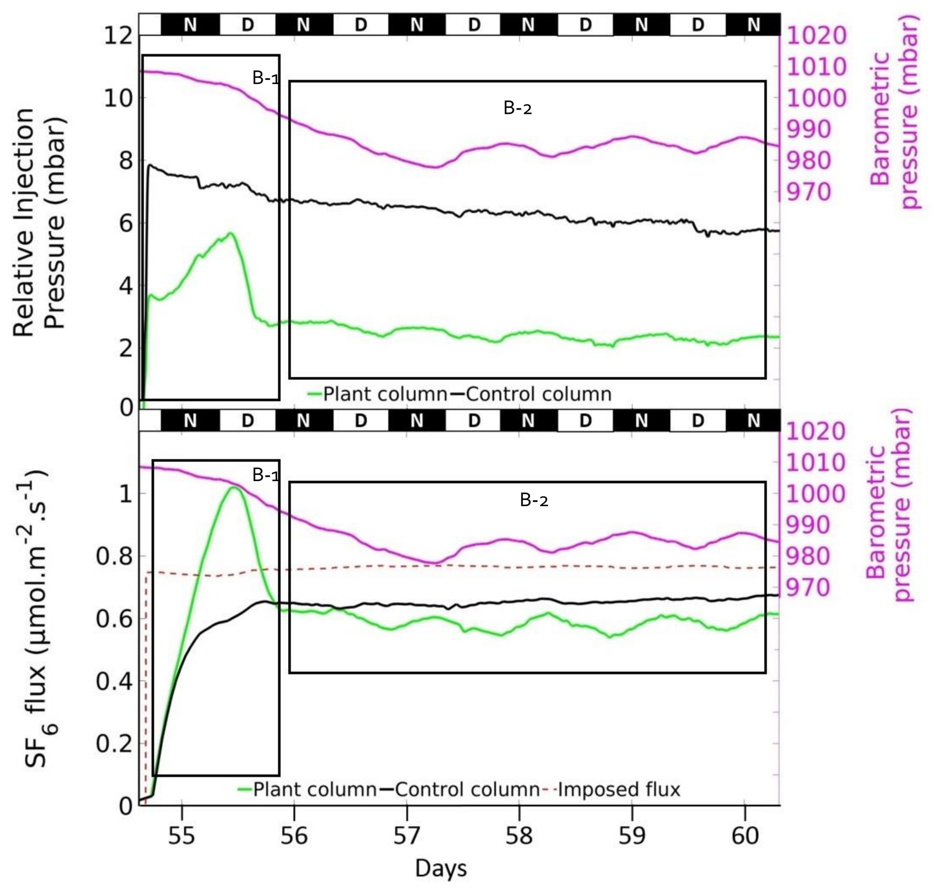
III- RESULTS

First order variations mainly physical and chemical

Case A



Case B



- A-1: Watering + overflow
- A-2: Evapotranspiration or evaporation
- A-3: Increase of barometric pressure

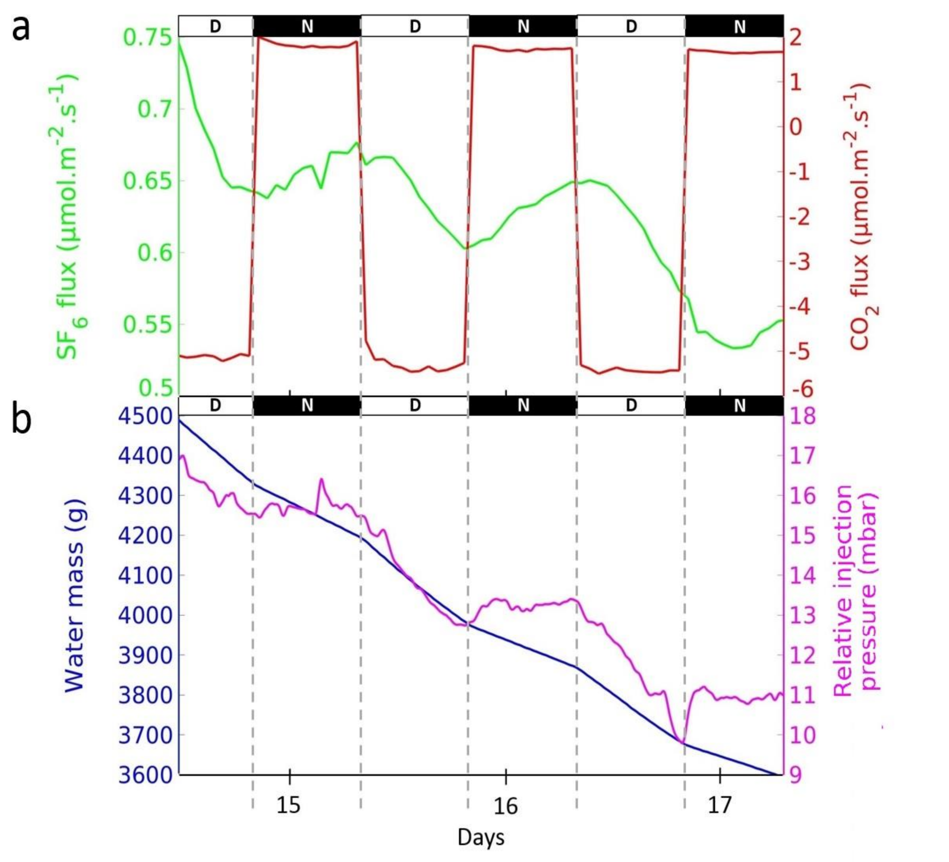
- B-1: Sudden overflow due to switching injection from diffusion to advection regime
- B-2: Steady state regime reached

Main processes : **1) Water budget, 2) Barometric pressure, 3) Injection pressure, 4) Solubility ?**

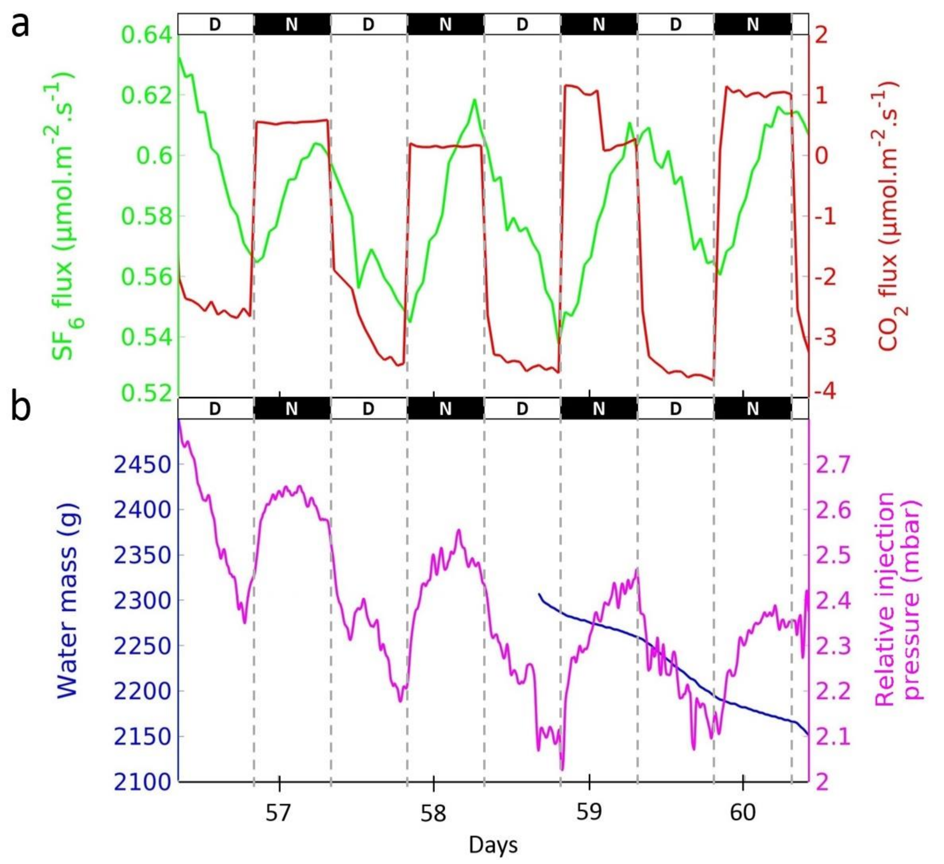
What are those **modulations** appearing on fluxes for the **plant column** ?

Second order variations due to diurnal biological activities: evapotranspiration and respiration

Case A



Case B



DAYTIME: Water loss by evapotranspiration -> Decrease of pressure gradient -> Decrease of SF_6 fluxes

increase in gas porosity and relative air permeability. This leads to more dispersion and storage of gases in the porous medium

NIGHTTIME: Respiration -> Consumption O_2 and production of CO_2 -> Increase of SF_6 fluxes

i) Dissolution of CO_2 higher -> local decrease of partial pressure -> increase of pressure gradient between injection and root-zone

ii) Possibility of scavenging due to CO_2 fluxes.

IV- ON GOING EFFORT

New experimental design to determine new processes:

Allows concentration profiles and multi-tracer gas experiments

Freeze and thaw cycle possible

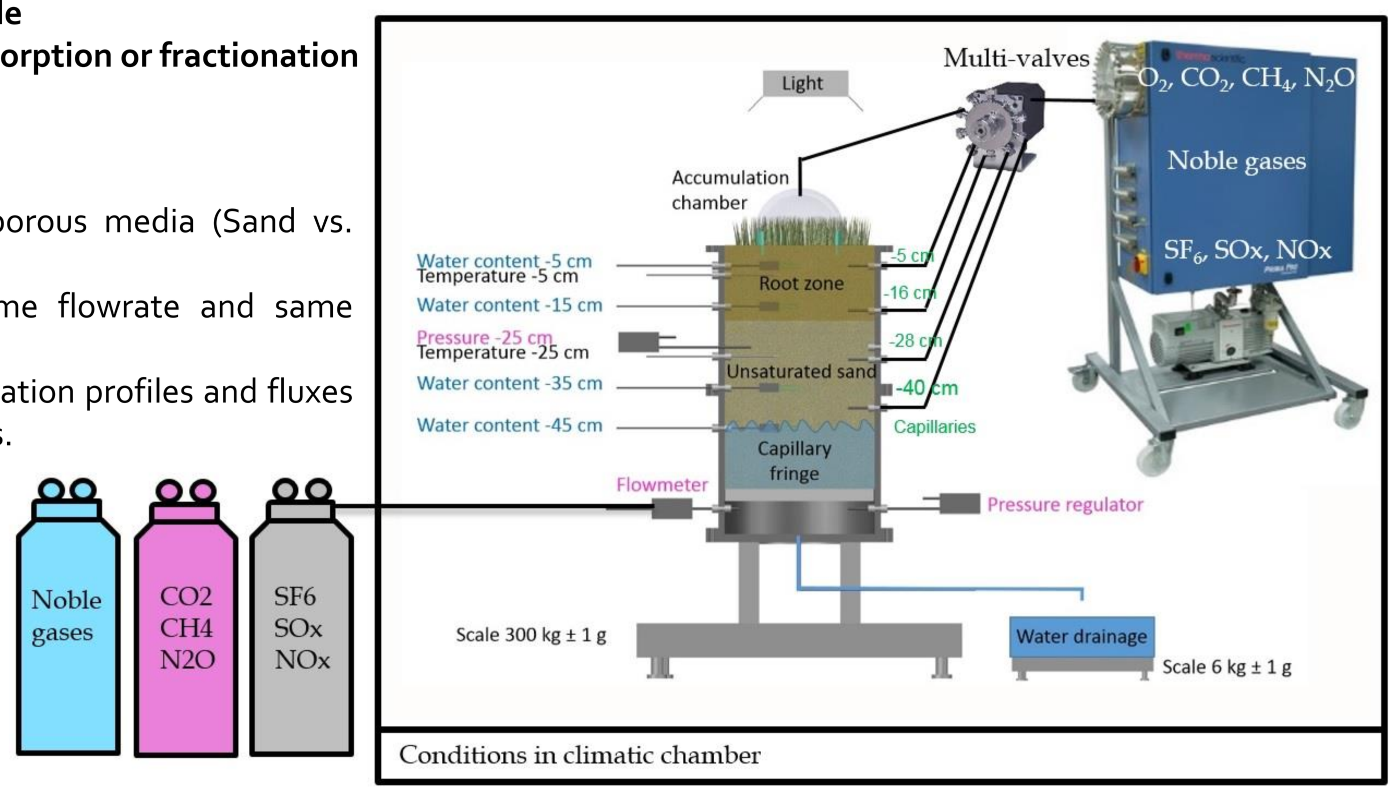
Observation of solubility, adsorption or fractionation effects

In-coming experiment:

2 columns with 2 different porous media (Sand vs. zeolite).

Injection of SF_6 and Xe: same flowrate and same concentration.

Observe evolution of concentration profiles and fluxes by comparing the two columns.

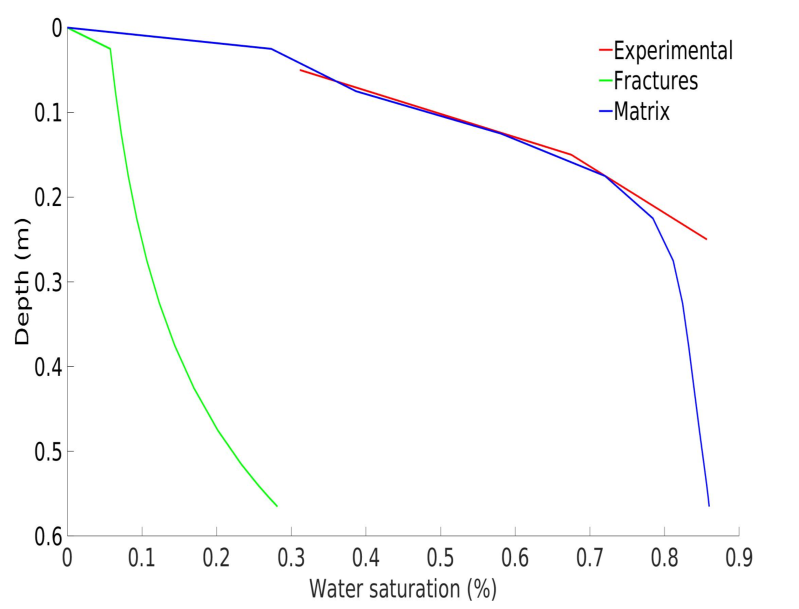


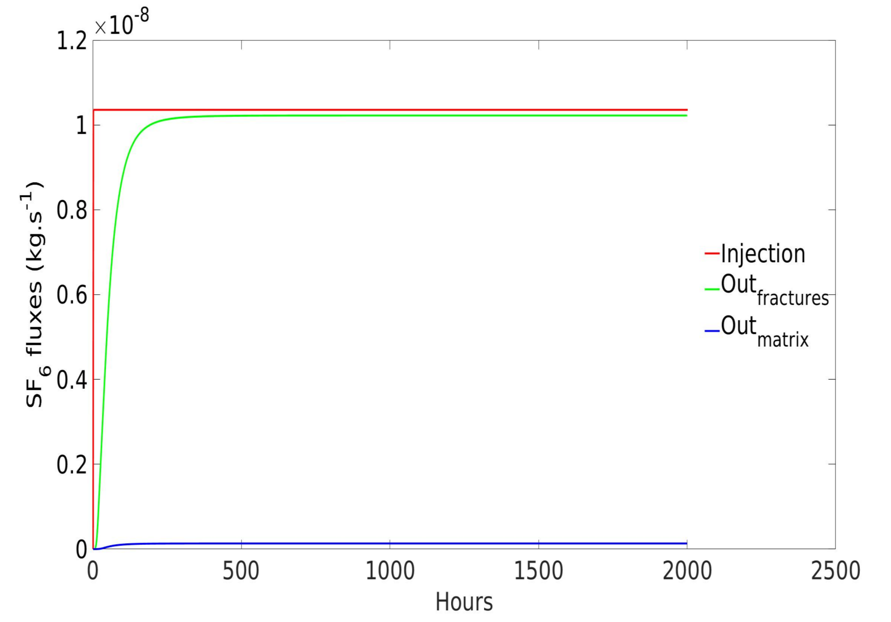
Modelisation with NUFT code:

Quantify processes brought to light with experiments (water budget variations, barometric pressure variations, solubility).

Double-permeability approach to mimic gas preferential path

First results obtained for a steady-state regim and unsaturated sand column at equilibrium





V- CONCLUSION

- New experimental set-up for long-term and high-resolution monitoring of gas percolations under controlled conditions in unsaturated columns, including plant growth.
- Large dynamical response of gas fluxes at the soil-atmosphere due to combined physical, chemical and biological controls acting mainly on pressure gradient.
- Nighttime-daytime gas flux modulations due to the combined effects of plant root respiration and photosynthesis-related evapotranspiration.

REFERENCE AND ACKNOWLEDGEMENTS

Alibert C., Pili E., Barre P., Massol F., Chollet S. (2019) Biologically-controlled gas fluxes revealed by high-resolution monitoring of unsaturated soil columns. *Vadose Zone Journal*. (Submitted and under review)

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