AMPHOS 21
SCIENTIFIC AND STRATEGIC ENVIRONMENTAL CONSULTING

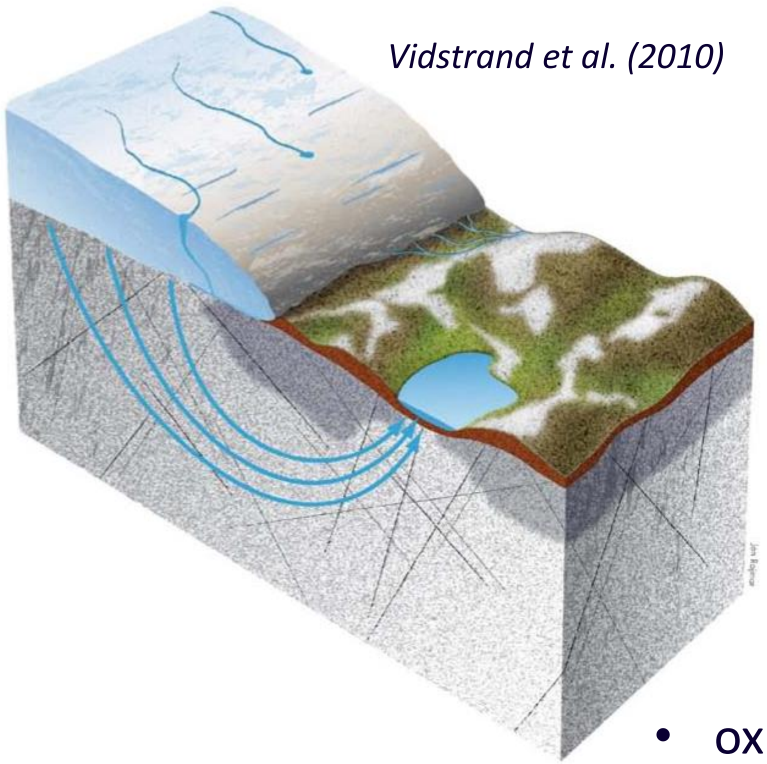
Oxygen intrusion in fractured rocks

How natural analogues and numerical numerical models can work together

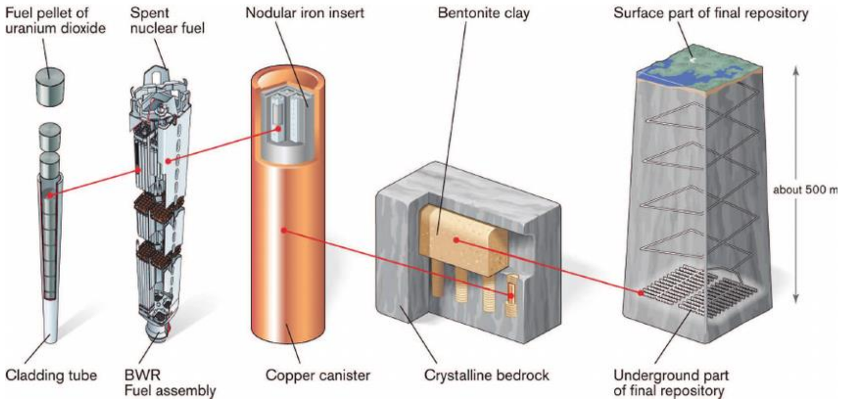
Jorge Molinero / NAWG 15 Workshop / Prague. May, 2017

Motivation

- The infiltration of **oxygen** (e.g. due to ingress of oxygenated glacial melt water) into **initially anoxic fractured media** is an issue of concern for the safety of **nuclear waste respositories**



Vidstrand et al. (2010)



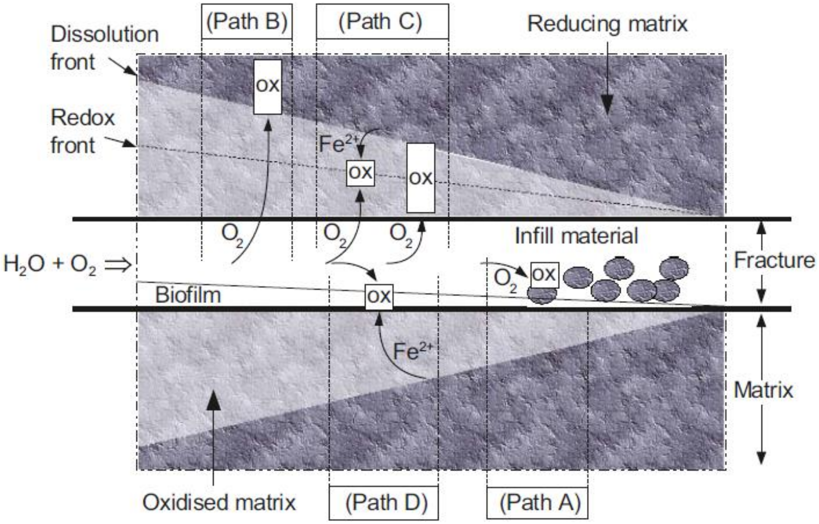
courtesy of SKB

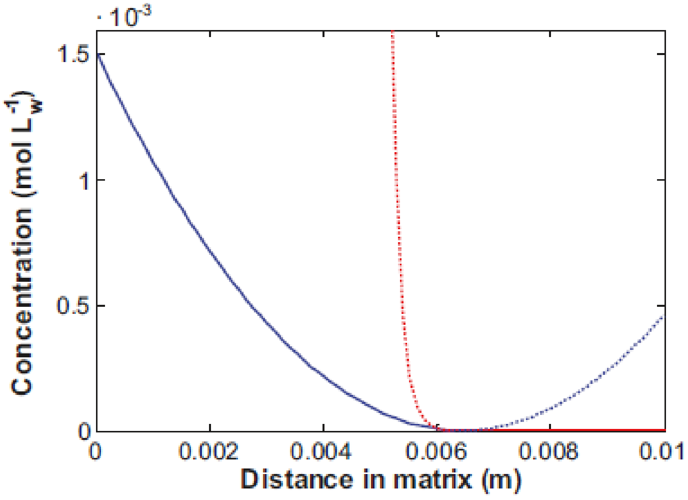
- oxidizing conditions would **increase the solubility** and **mobility** of many radionuclides and could **corrode** copper canisters

Background: Previous models

- Due to the **stiffness** of the underlying system of equations, existing numerical models are based on simplified **1D** or **2D geometries**…
- …and on **equivalent homogeneous** parameters

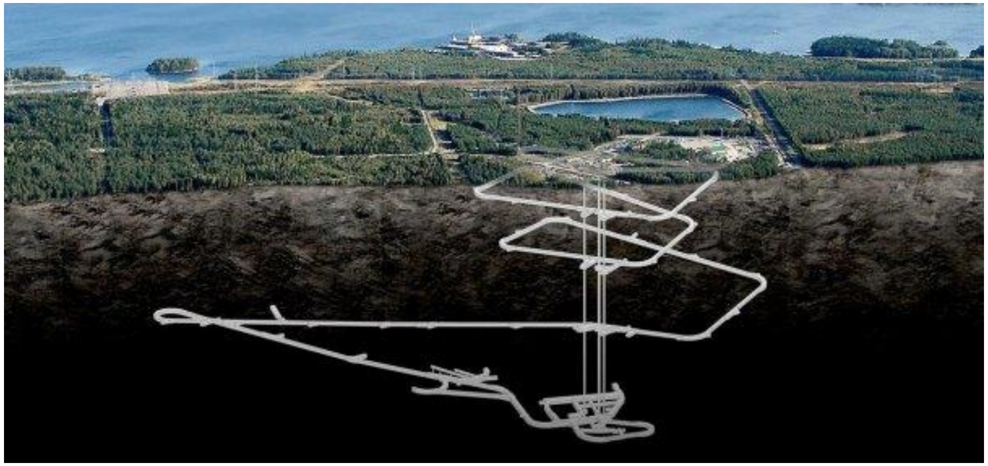
Sidborn et al. (2010)





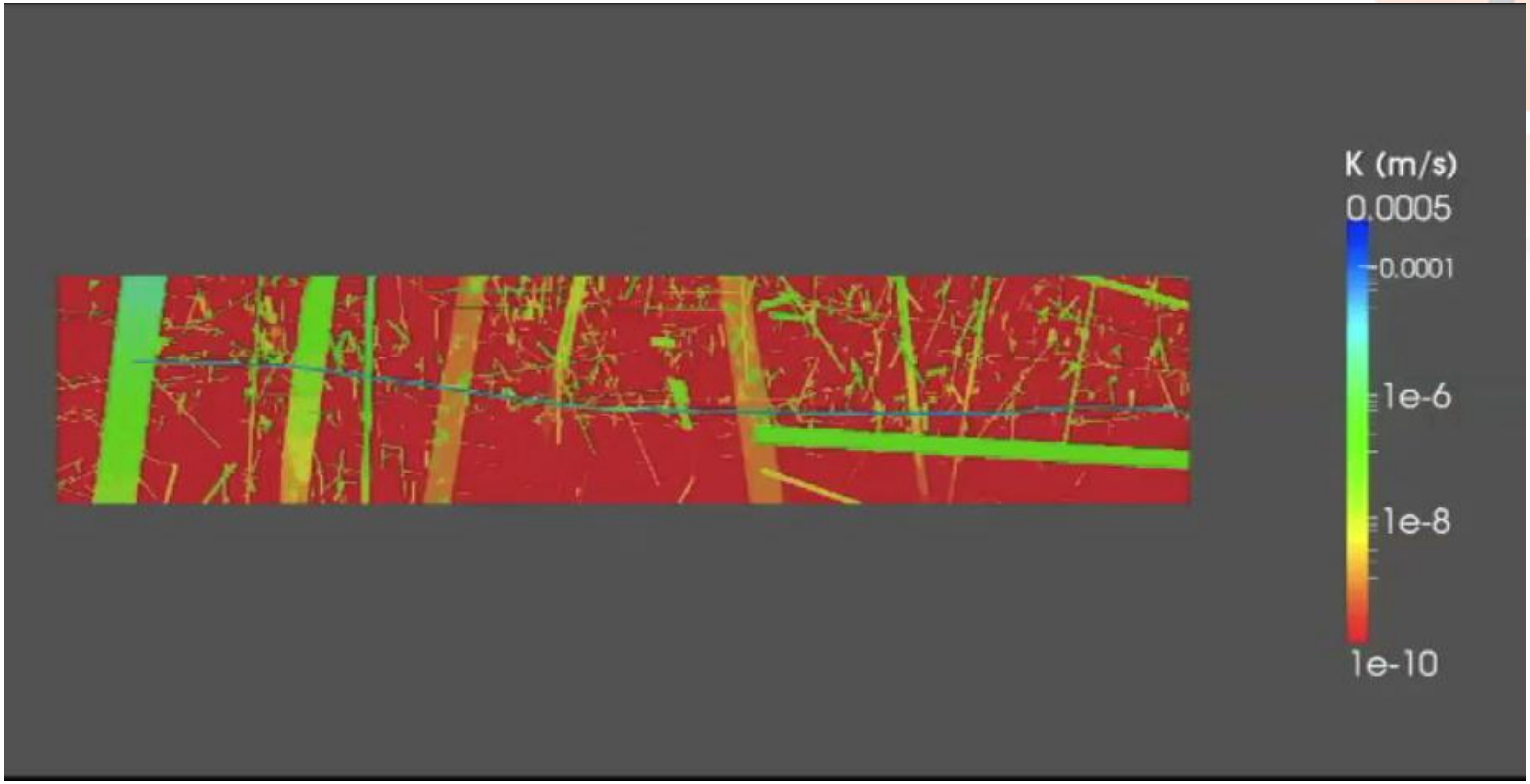
(a) $c_{1f} = 1.5 \cdot 10^{-3} [\mathrm{mol}_{O_2} L_w^{-1}]$

But we are telling the site looks like this:



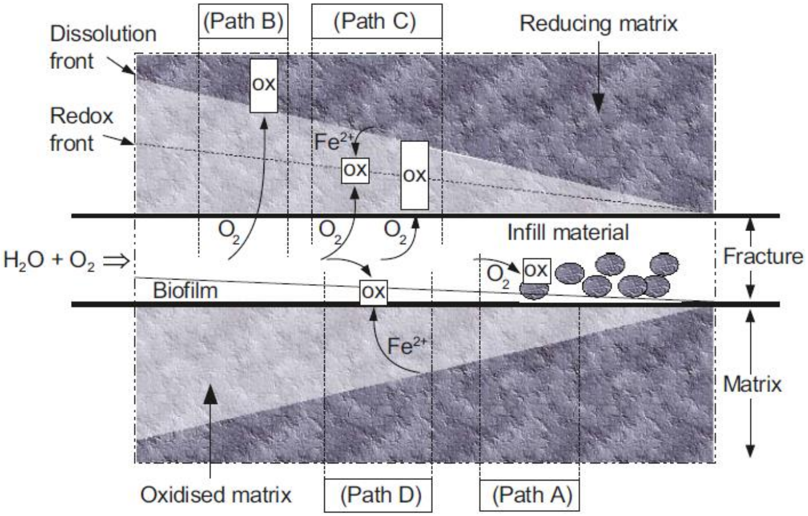
And we spent lots of money in site characterization…

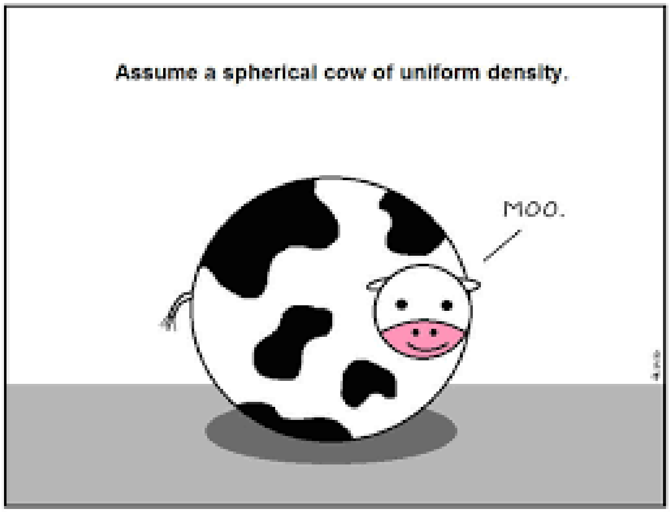
And we construct sofisticated SDM…



And finally we use something like this:

Sidborn et al. (2010)





We (science) know how oxygen consumption works

- **Chlorite dissolution** (Lowson et al., 2005)

$$k_{chl,diss}[mol/m_m^2 \cdot s] = 10^{-9.79} \cdot c_{H^+}^{0.49} + 10^{-13.00} + 10^{-16.79} c_{H^+}^{-0.43}$$

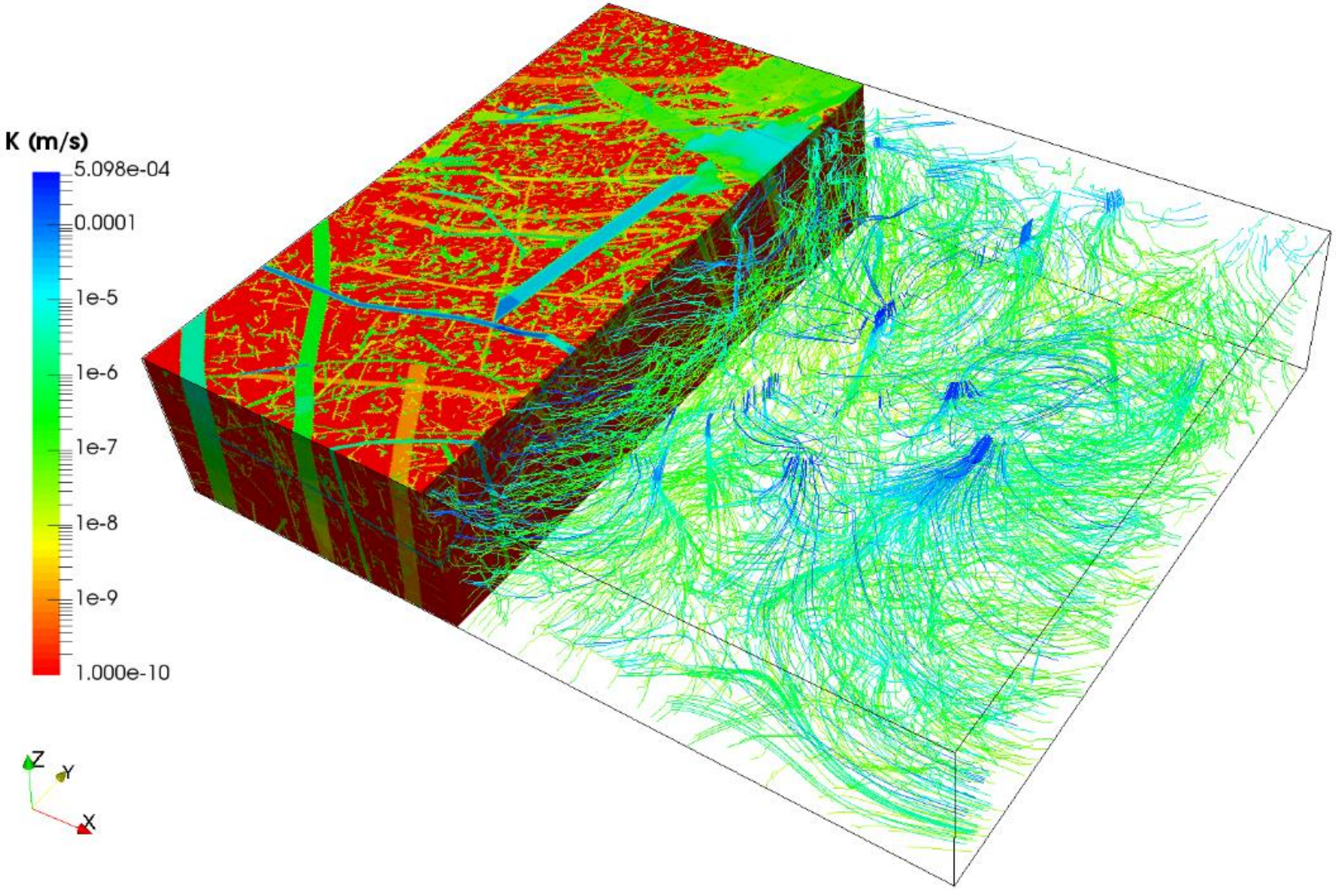
$$r_{Fe}\left[mol_{Fe}/m_b^3 \cdot s\right] = n_{stoich} \cdot A_s \cdot k_{chl,diss}\left(1 - \frac{c_{Fe}}{c_{Fe,sol}}\right)$$

- Homogeneous **oxidation** of **ferrous** ions (Stumm & Lee, 1961)

$$4\mathrm{Fe}^{2+} + \mathrm{O}_2 + 4\mathrm{H}^+ \leftrightarrow 4\mathrm{Fe}^{3+} + 2\mathrm{H}_2 0$$

$$r_{ox}[mol_{Fe}/L \cdot s] = k_r \cdot c_{Fe} \cdot P_{O_2} \cdot c_{OH^-}^2$$

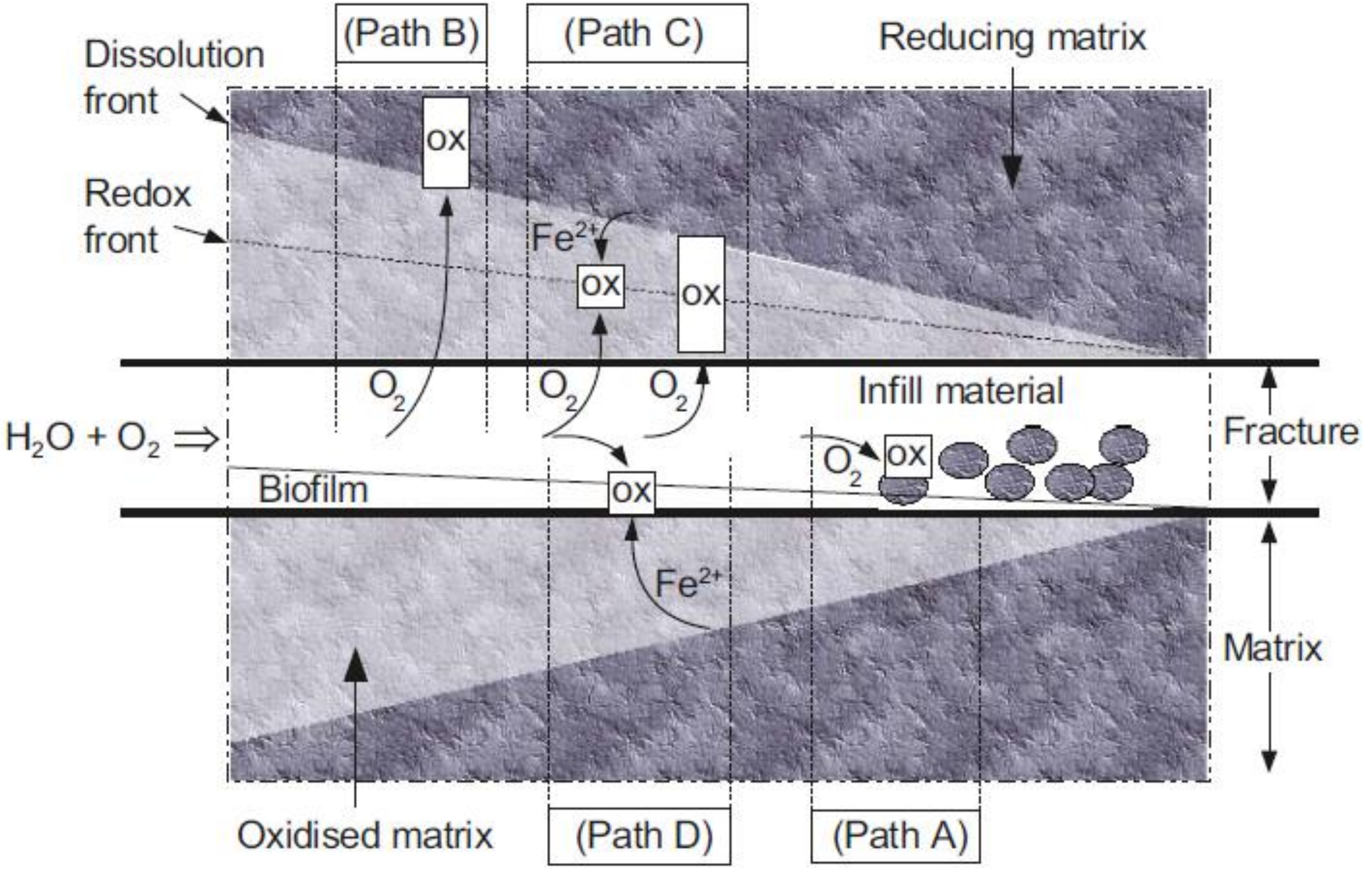
We (science) know how to represent hydrogeology in FR



- regional hydrogeological model of Forsmark

So again… why?

Sidborn et al. (2010)



It is just (or mainly) a technological barrier

Software: iDP

interface DarcyTools PFLOTRAN

FLOW

DarcyTools

REACTIVE TRANSPORT

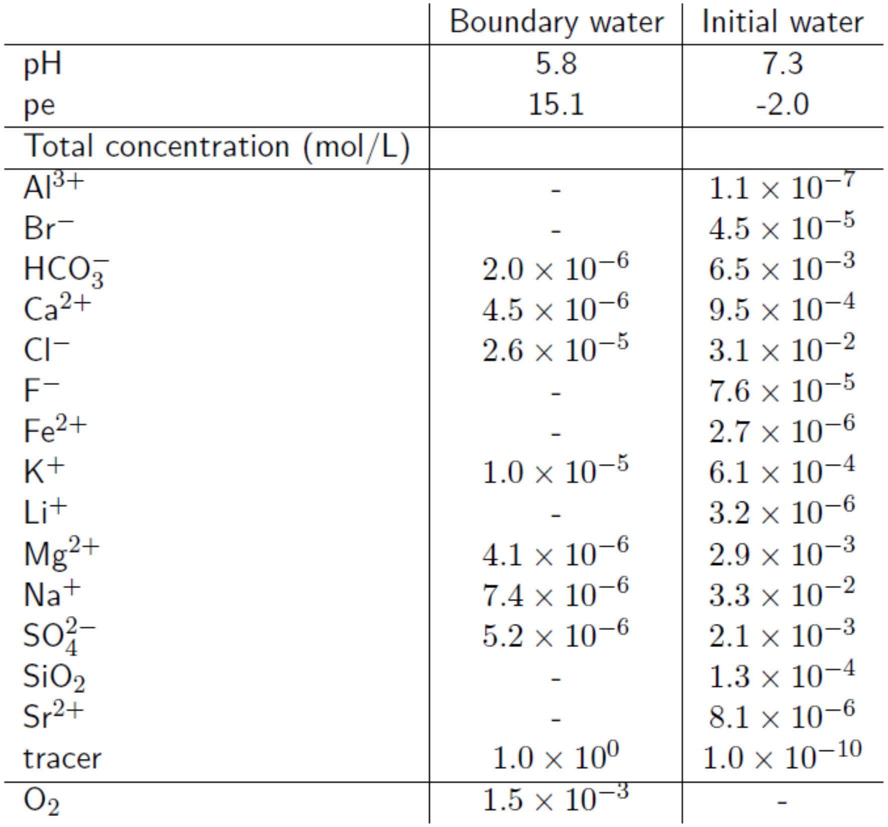
PFLOTRAN

A Massively Parallel Reactive Flow and Transport Model for describing Surface and Subsurface Processes

Hardware:

JÜLICH
FORSCHUNGSZENTRUM

Then, we incorporate our best knowledge

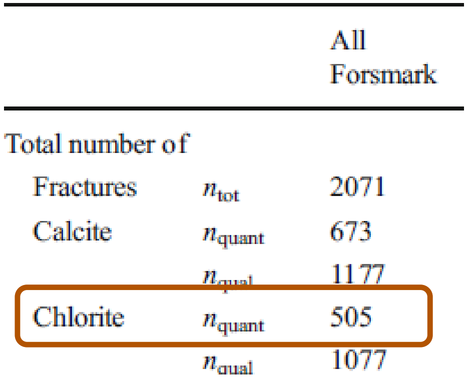


	Boundary water	Initial water
pH	5.8	7.3
pe	15.1	-2.0
Total concentration (mol/L)		
Al^{3+}	-	1.1×10^{-7}
Br^{-}	-	4.5×10^{-5}
HCO_3^{-}	2.0×10^{-6}	6.5×10^{-3}
Ca^{2+}	4.5×10^{-6}	9.5×10^{-4}
Cl^{-}	2.6×10^{-5}	3.1×10^{-2}
F^{-}	-	7.6×10^{-5}
Fe^{2+}	-	2.7×10^{-6}
K^{+}	1.0×10^{-5}	6.1×10^{-4}
Li^{+}	-	3.2×10^{-6}
Mg^{2+}	4.1×10^{-6}	2.9×10^{-3}
Na^{+}	7.4×10^{-6}	3.3×10^{-2}
SO_4^{2-}	5.2×10^{-6}	2.1×10^{-3}
SiO_2	-	1.3×10^{-4}
Sr^{2+}	-	8.1×10^{-6}
tracer	1.0×10^{0}	1.0×10^{-10}
O_2	1.5×10^{-3}	-

- 17 **primary species**, 29 **secondary species**, two reacting minerals (**calcite** and **chlorite**) and a homogeneous **aqueous oxidation** reaction
- 70,125,000 transport **degrees of freedom**

And parameterization based on SC data:

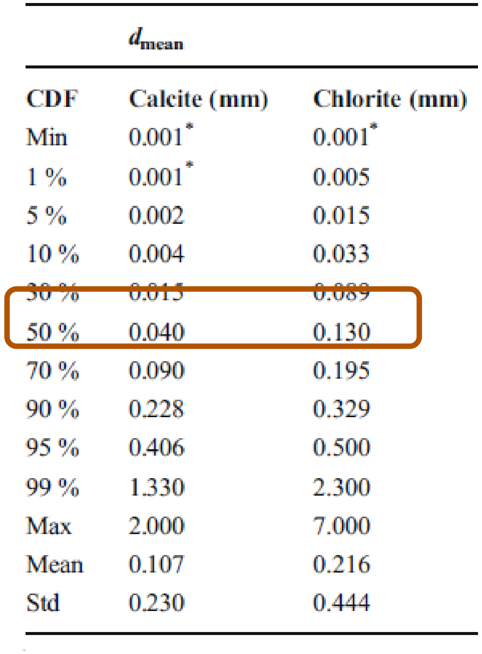
- Löfgren and Sidborn (2016) mapped minerals in **2071 open fractures** and provided statistics of **mineral coverage** and **mineral thickness** for chlorite, calcite and pyrite



		All Forsmark
Total number of		
Fractures	n_{tot}	2071
Calcite	n_{quant}	673
	n_{qual}	1177
Chlorite	n_{quant}	505
	n_{qual}	1077

$$\alpha = 0.24$$

$$d_{mean} = 1.3 \cdot 10^{-4}\ m$$

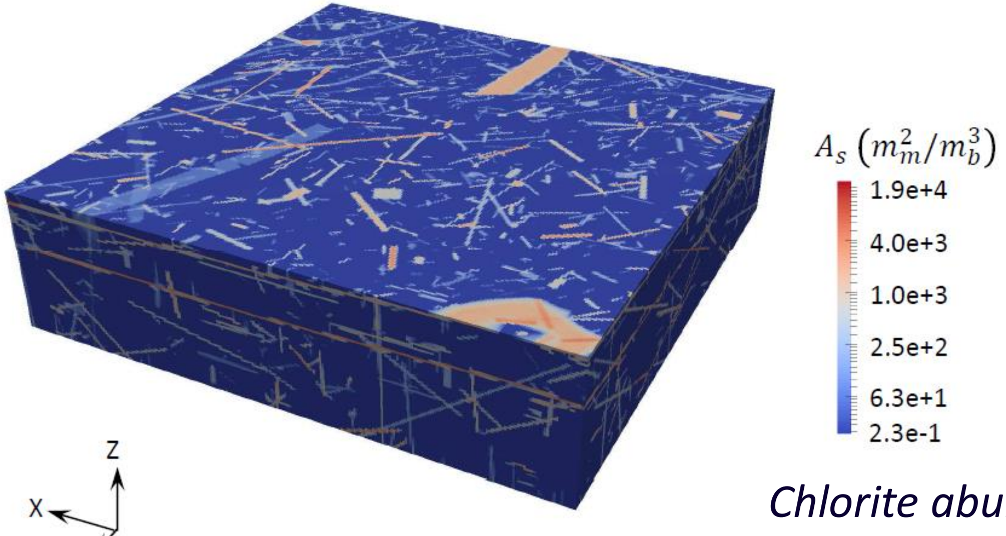


	d_{mean}	
CDF	**Calcite (mm)**	**Chlorite (mm)**
Min	0.001*	0.001*
1 %	0.001*	0.005
5 %	0.002	0.015
10 %	0.004	0.033
30 %	0.015	0.089
50 %	0.040	0.130
70 %	0.090	0.195
90 %	0.228	0.329
95 %	0.406	0.500
99 %	1.330	2.300
Max	2.000	7.000
Mean	0.107	0.216
Std	0.230	0.444

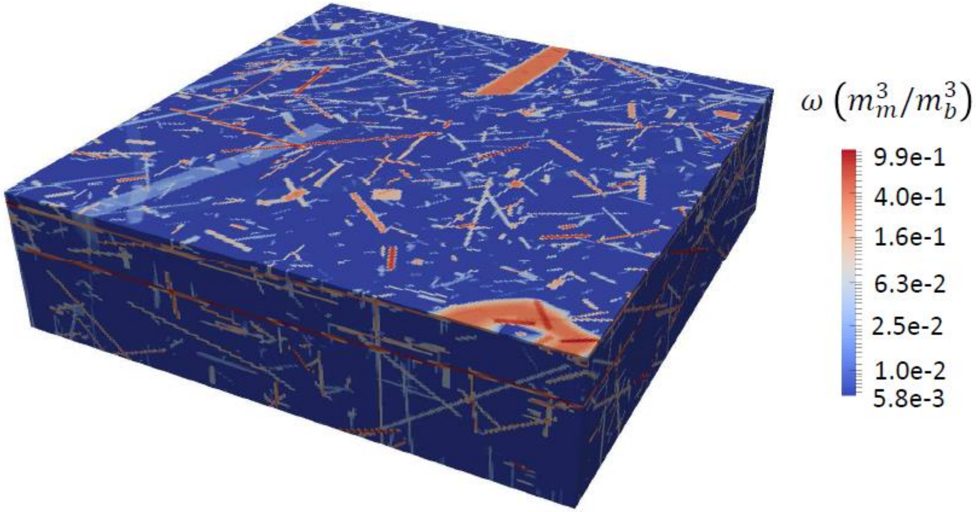
Löfgren, M., Sidborn, M., 2016. Quantitative mapping and statistical evaluation of fracture minerals in the granitic bedrock at Forsmark, Sweden. Mineralogy and Petrology, 110, 663-680.

Geochemical parameterization looks like this:

Chlorite surface area

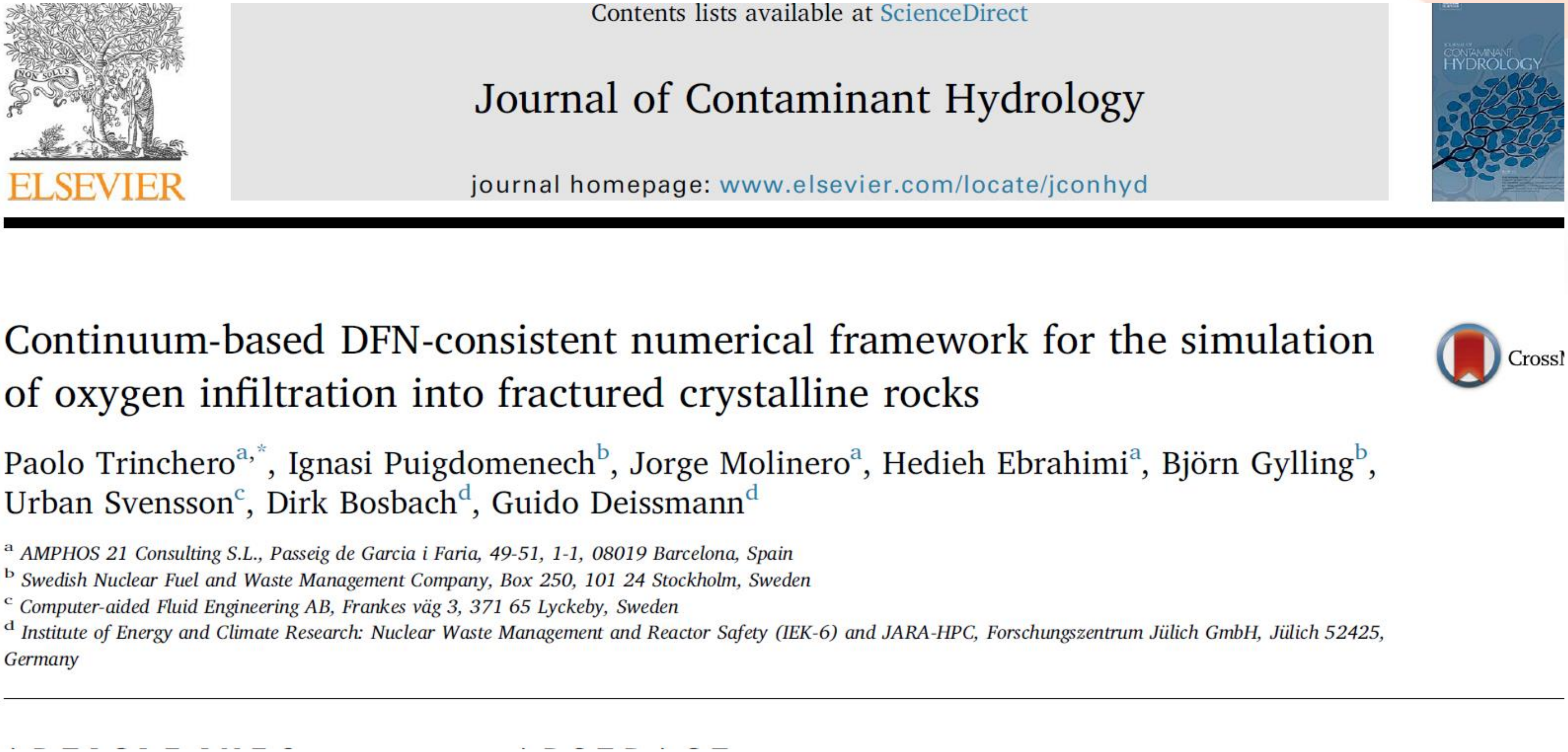


Chlorite abundance



- Reactive transport calculation ran out to **340 y**
- A total of about **33,000 h** (3.7 years) of **supercomputing time** were consumed

All details of the model:



Contents lists available at ScienceDirect

Journal of Contaminant Hydrology

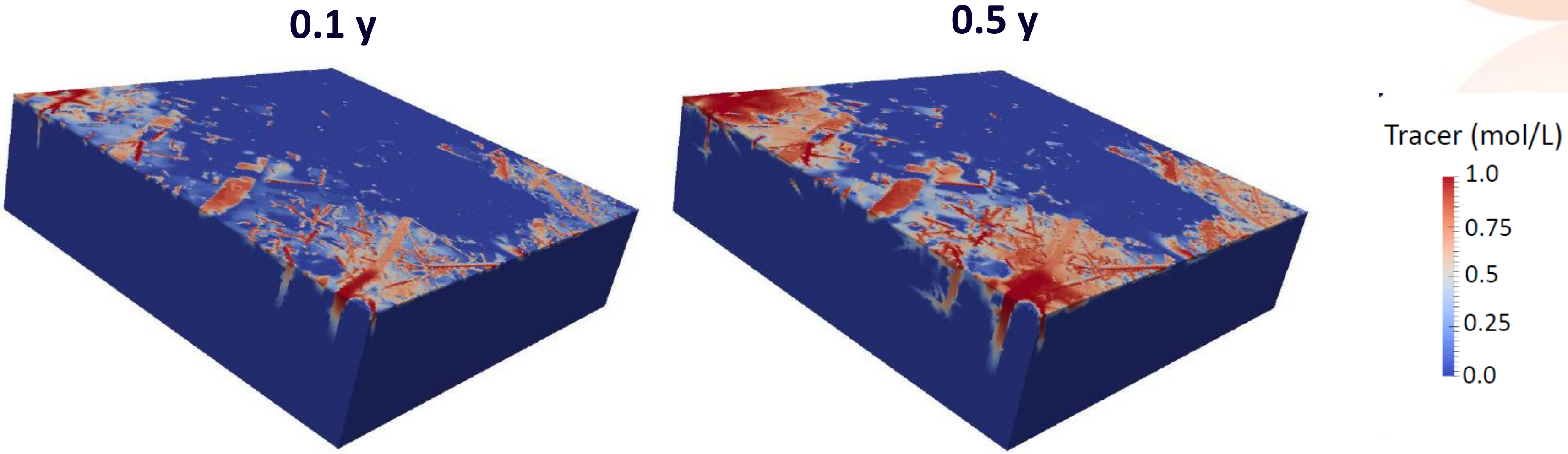
journal homepage: www.elsevier.com/locate/jconhyd

Continuum-based DFN-consistent numerical framework for the simulation of oxygen infiltration into fractured crystalline rocks

Paolo Trinchero[a,*], Ignasi Puigdomenech[b], Jorge Molinero[a], Hedieh Ebrahimi[a], Björn Gylling[b], Urban Svensson[c], Dirk Bosbach[d], Guido Deissmann[d]

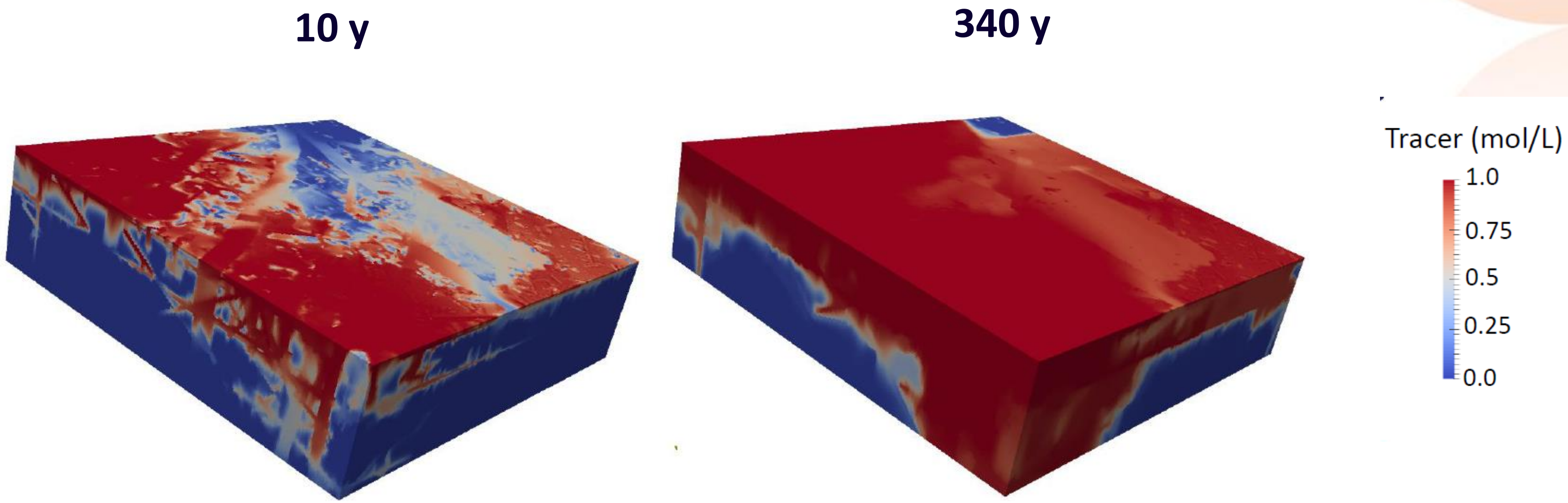
[a] AMPHOS 21 Consulting S.L., Passeig de Garcia i Faria, 49-51, 1-1, 08019 Barcelona, Spain
[b] Swedish Nuclear Fuel and Waste Management Company, Box 250, 101 24 Stockholm, Sweden
[c] Computer-aided Fluid Engineering AB, Frankes väg 3, 371 65 Lyckeby, Sweden
[d] Institute of Energy and Climate Research: Nuclear Waste Management and Reactor Safety (IEK-6) and JARA-HPC, Forschungszentrum Jülich GmbH, Jülich 52425, Germany

Computed results: Conservative tracer



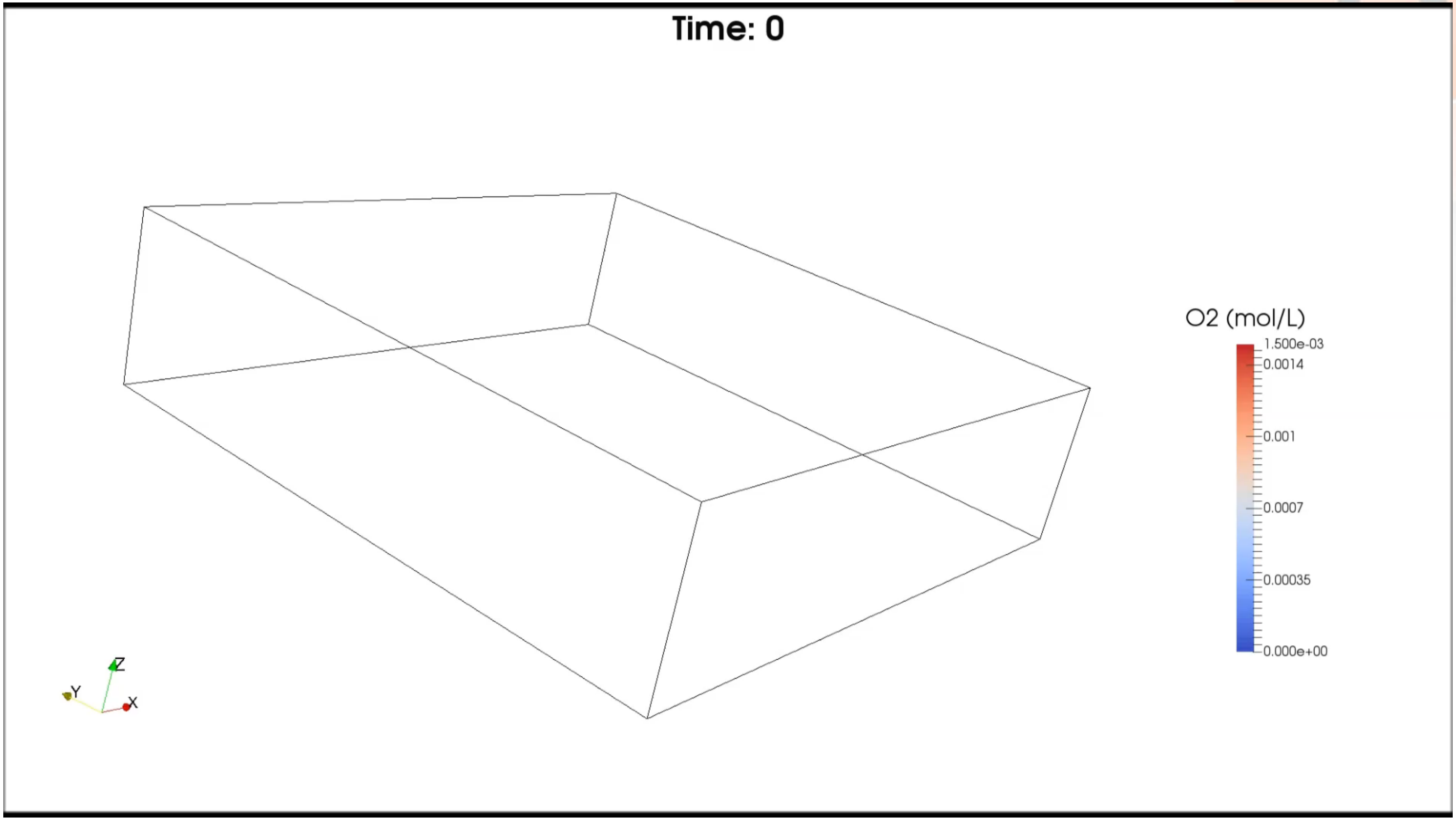
- At early times, the tracer infiltrates from the recharge areas and is **channeled** along **transmissive fractures** and **deformation zones**

Computed results: Conservative tracer

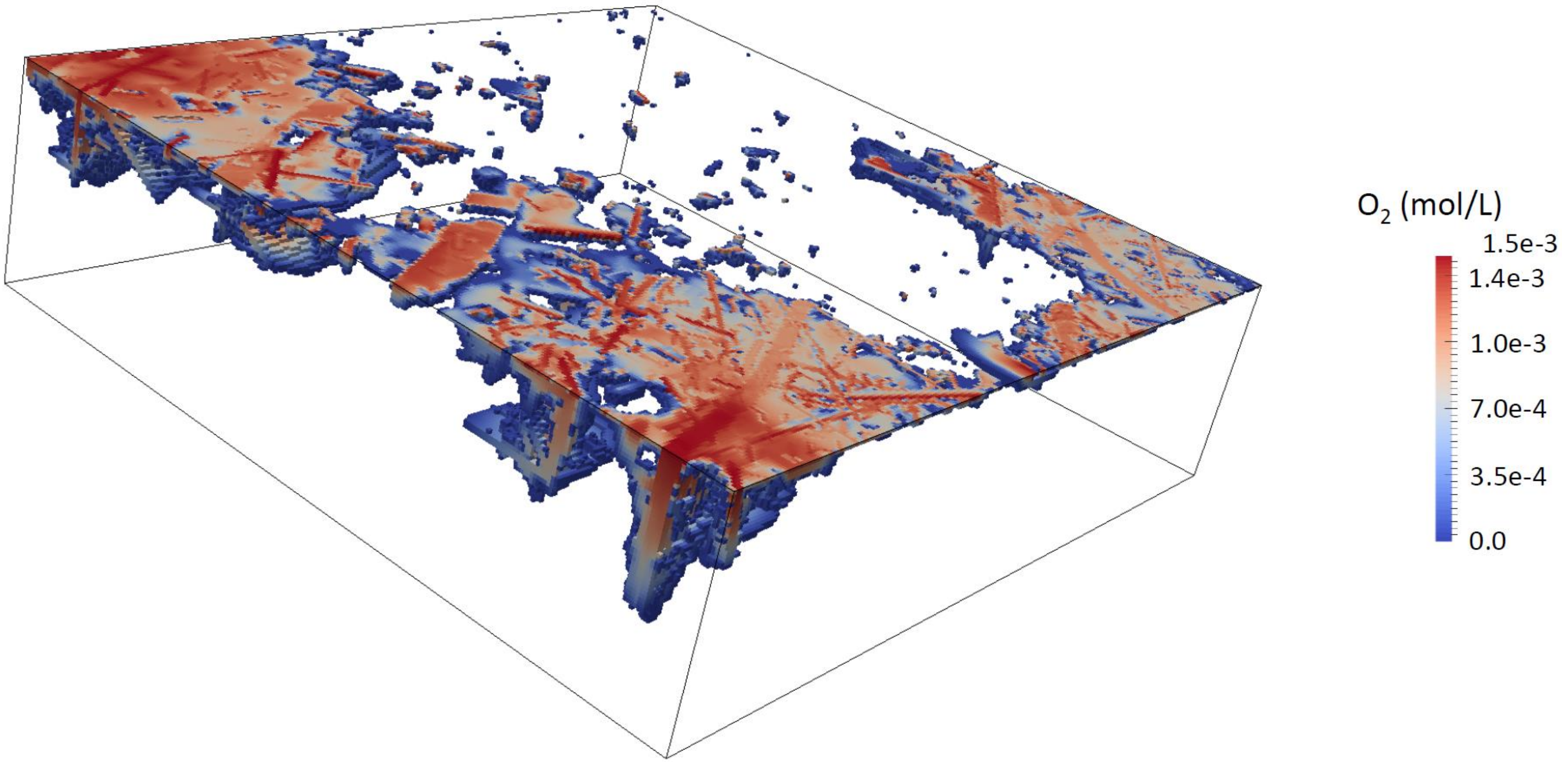


- It progressively infiltrates deep into the bedrock and reaches most of the **discharge areas**

Computed results: Oxygen

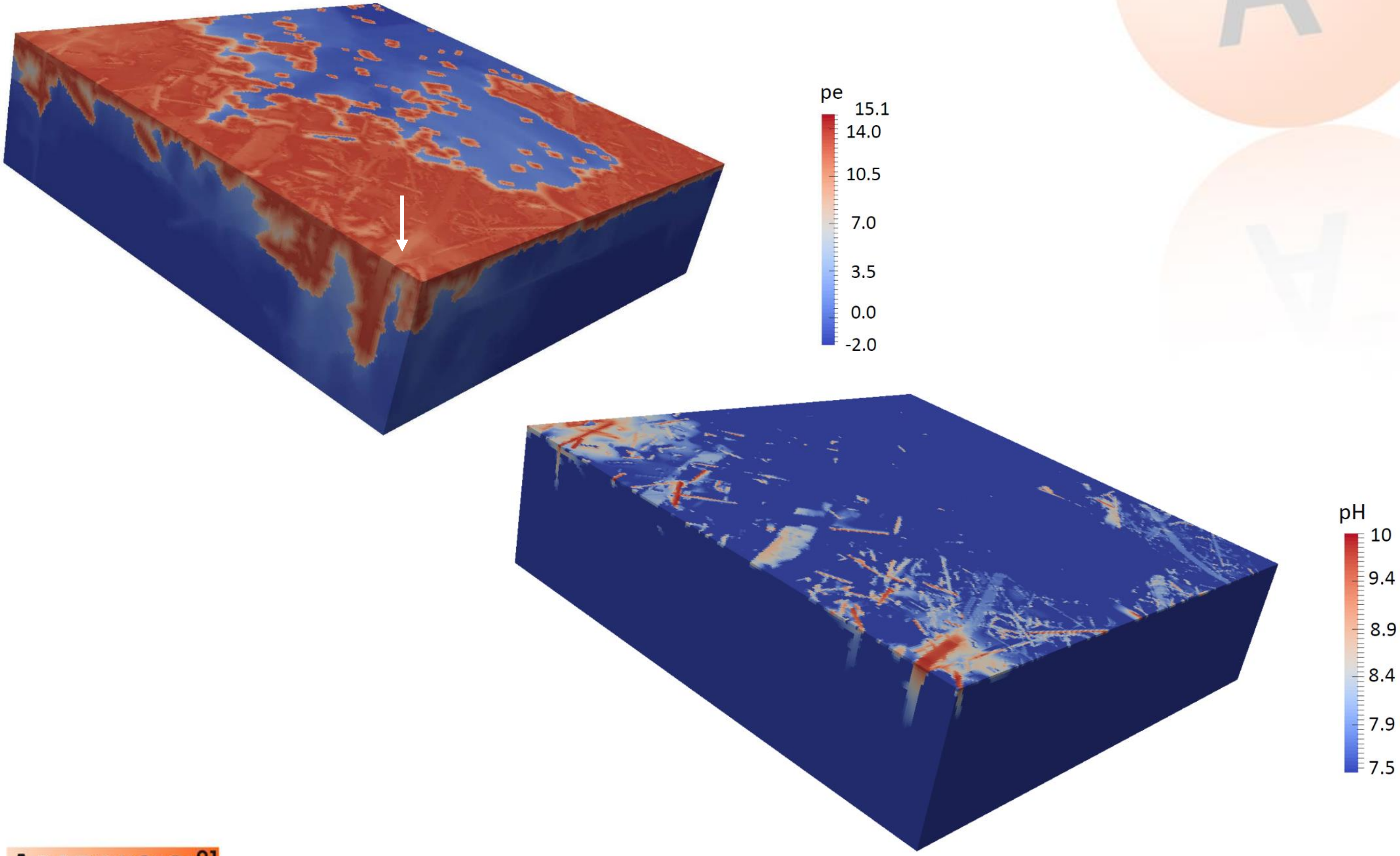


Computed results: Oxygen

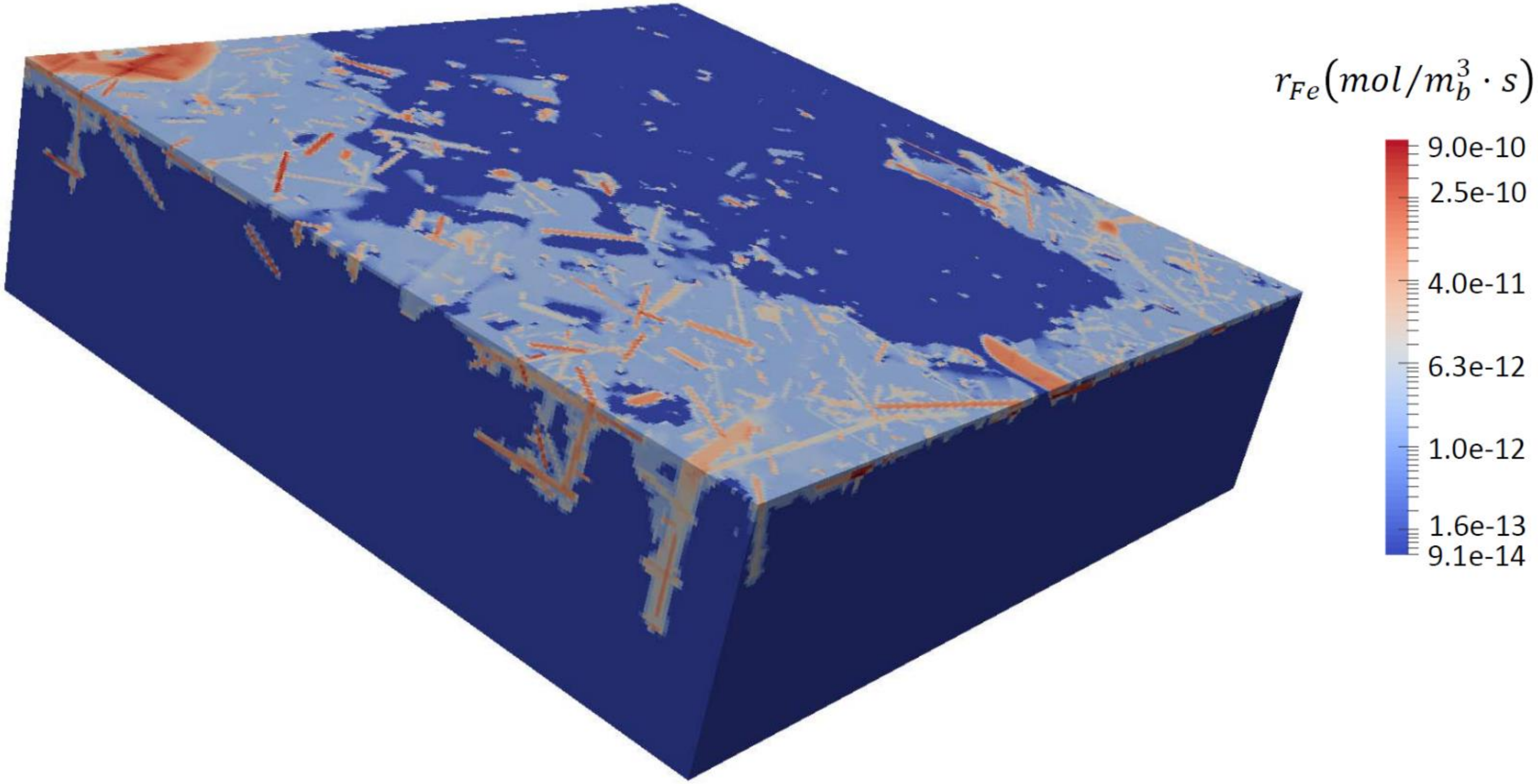


- Oxygen infiltrates predominantly along transmissive zones => the rest of the domain remains **anoxic**
- Maximum **penetration depth** is **90 m** approximately.

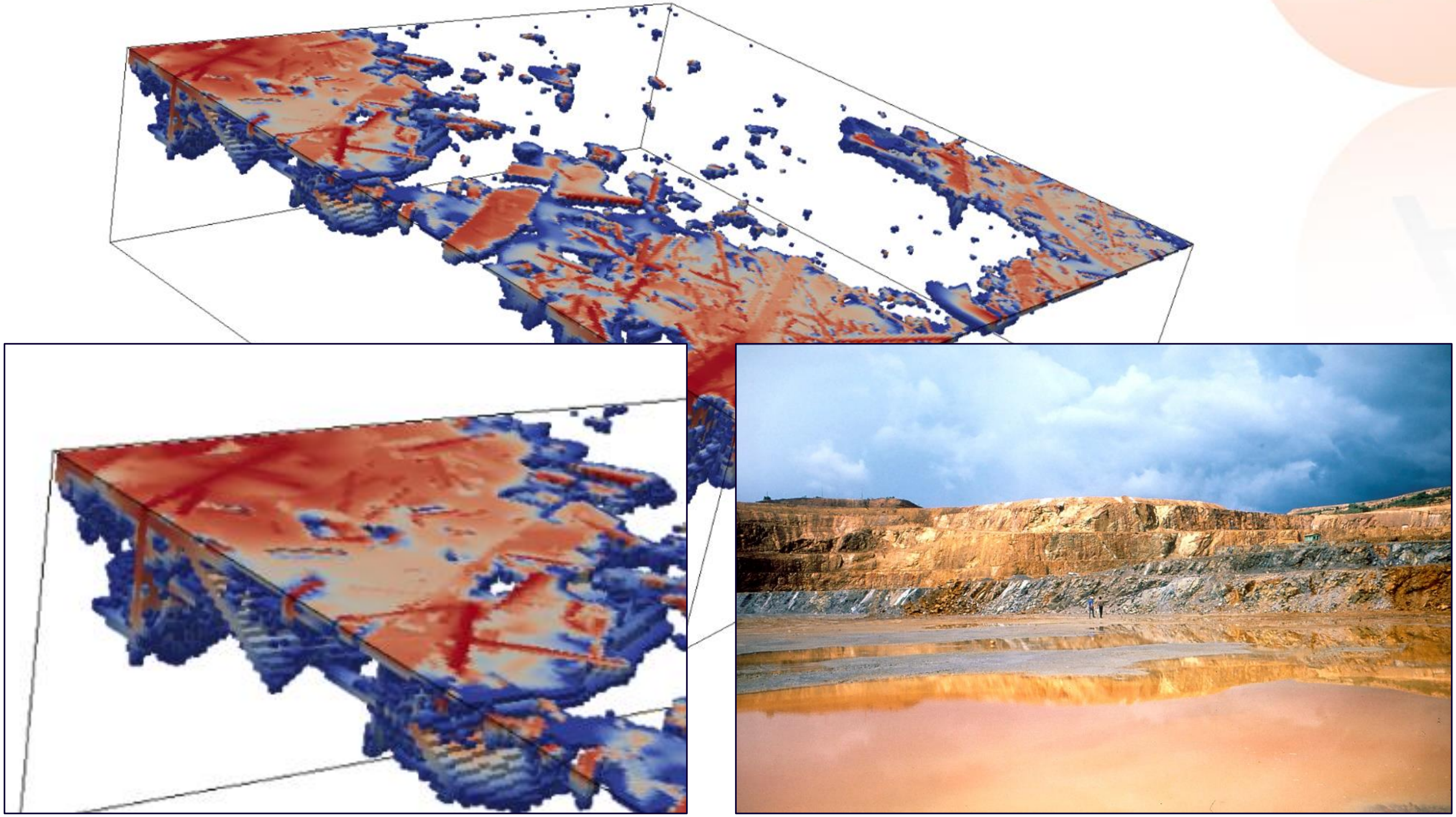
Computed results: pe & pH



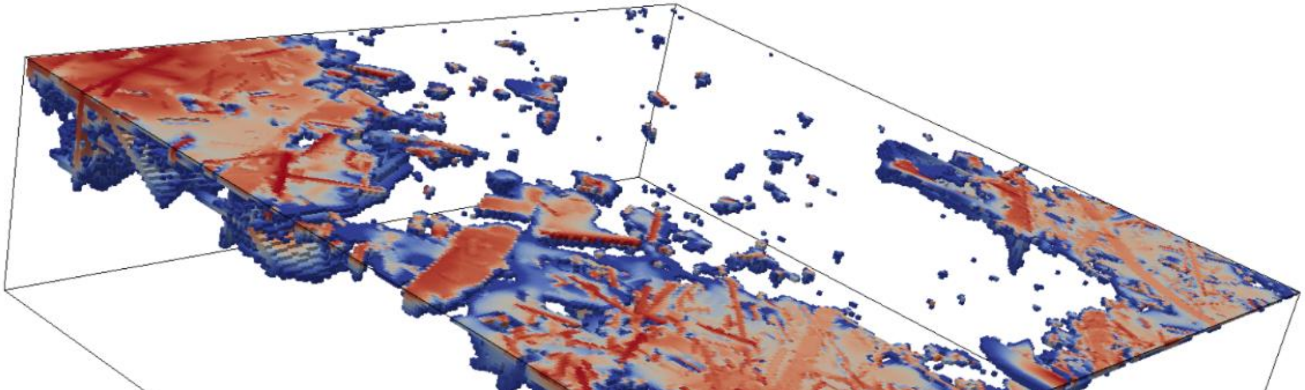
Computed results: Chlorite dissolution (rate)

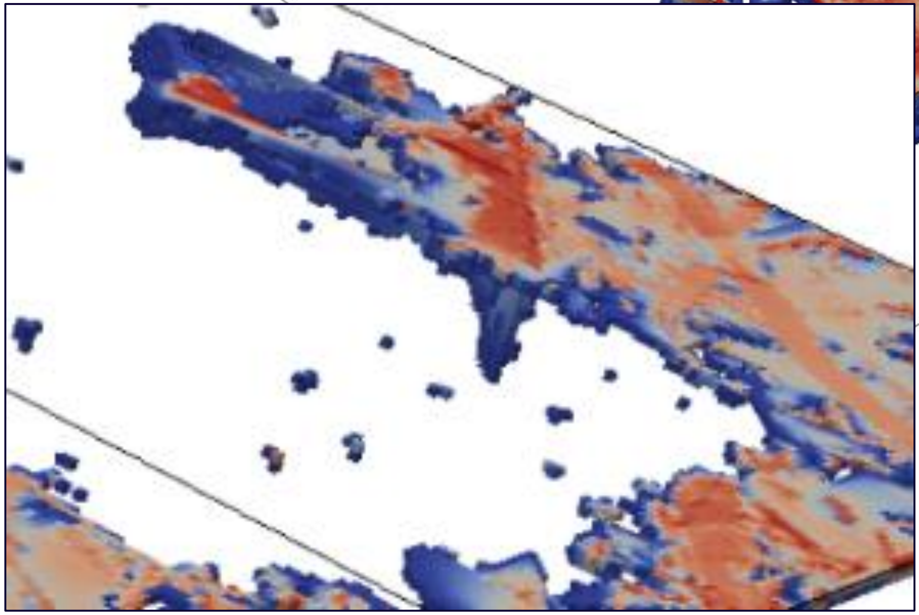


So now we have a better model… should we trust it?

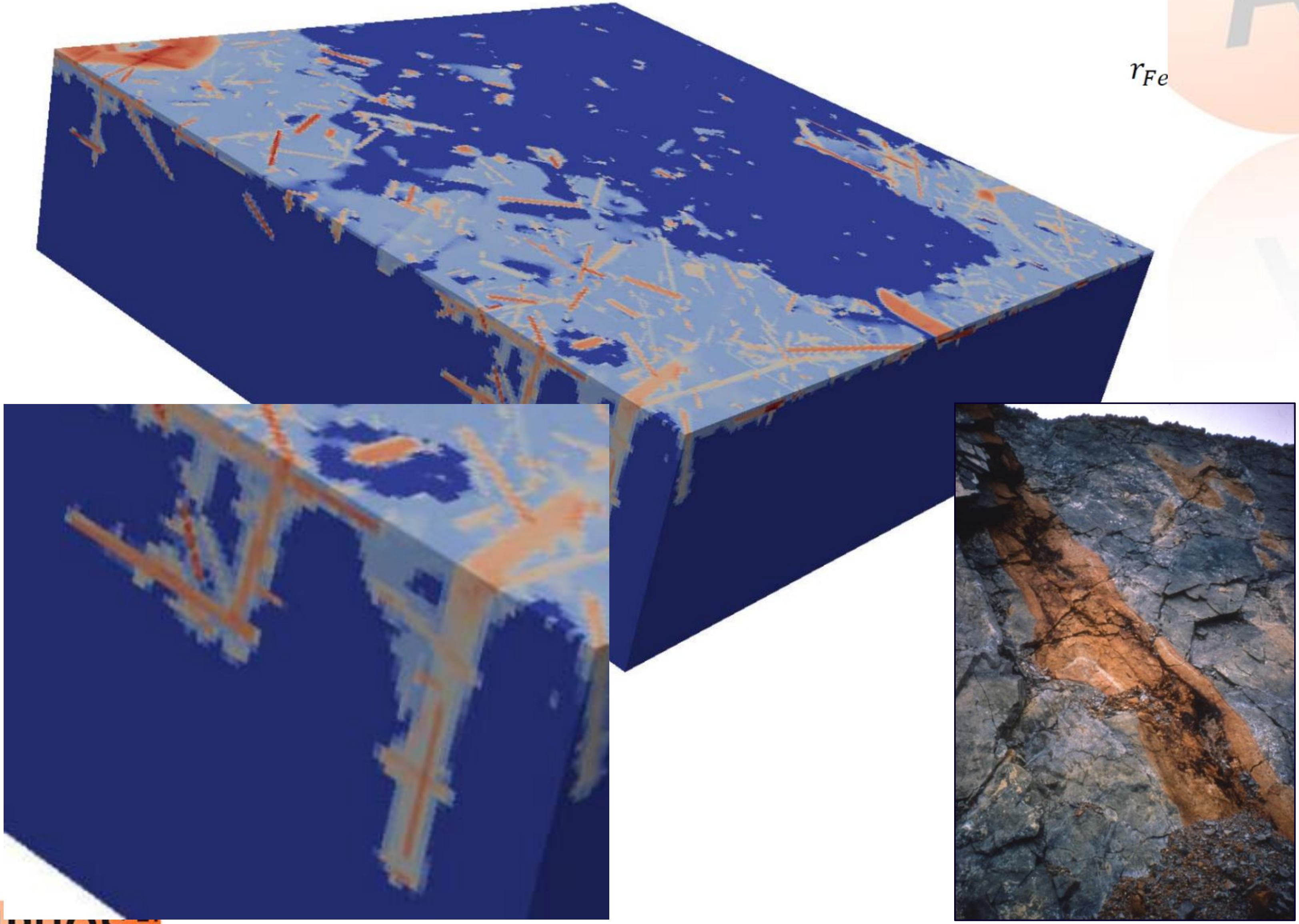


So now we have a better model… should we trust it?





So now we have a better model… should we trust it?



r_{Fe}

Conclusions

The results of the Forsmark model show that **quick penetration** of oxygen is observed only **along transmissive zones**

Steady state conditions is attained after around 10y => maximum penetration depth of **90 m**

The numerical model show results **highly consistent** with the qualitative observations of a **natural analogue** of the same processes we want to simulate (Poços de Caldas – Oxidative fronts)

Models must be adapted to the **best available technology**.

Realism and confrontation with actual **observations in nature** are the keys for building **model confidence.**

Credits

- Paolo Trinchero, Jorge Molinero & Hedieh Ebrahimi => Amphos 21
- Ignasi Puigdomenech & Björn Gylling => SKB
- Urban Svensson => CFE
- Guido Deissmann & Dirk Bosbach => Forschungszentrum Jülich

The authors gratefully acknowledge the computing time granted by the **JARA-HPC Vergabegremium** and provided on the JARA-HPC Partition part of the supercomputer **JUQUEEN** at Forschungszentrum Jülich.

This work has been **financed by SKB**. Also, all the pictures of the **Poços de Caldas Natural Analogue** are property of SKB

AMPHOS 21
SCIENTIFIC AND STRATEGIC ENVIRONMENTAL CONSULTING

ESPAÑA
Paseo de García Faria, 49-51
08019 Barcelona
Tel.: +34 93 583 05 00

Paseo de la Castellana 40, 8ª Planta
28046 Madrid
Tel.: +34 620634729

CHILE
Avda. Nueva Tajamar, 481
WTC – Torre Sur – Of 1005
Las Condes, Santiago
Tel.: +562 2 7991630

PERÚ
Jr. Pietro Torrigiano 396
San Borja, Lima 41
Tel.: +51 1 592 1275

FRANCE
92 Rue du Sergent Blandan
54000 Nancy
Tel.: +33 645 766 322

- Thanks for your attention -

www.amphos21.com