



2020 SCCS Annual Affiliates Meeting

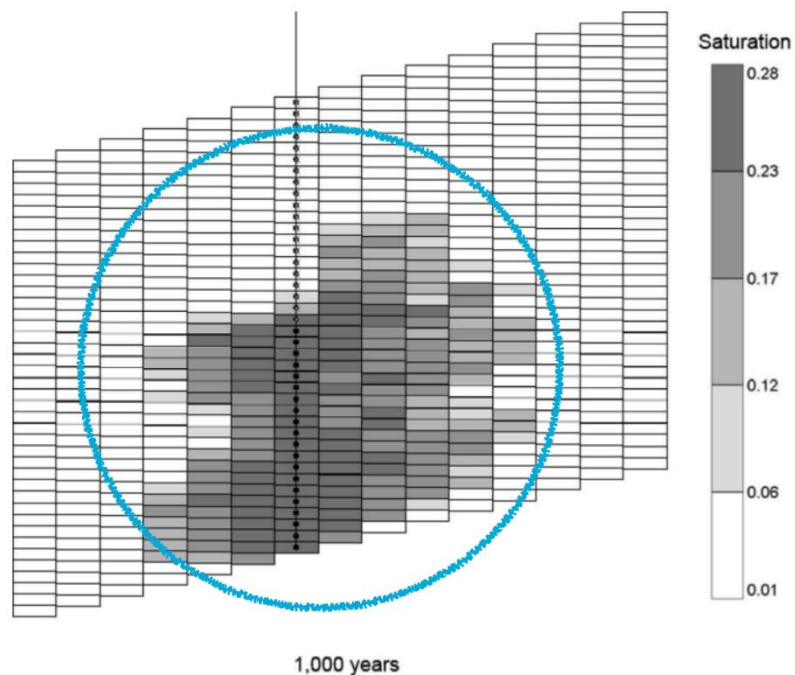
Diffusion-induced Gas Redistribution in Porous Media

Yaxin Li and Prof. Sally M Benson

Nov 19th, 2020

A large fraction of injected CO₂ are trapped due to residual trapping

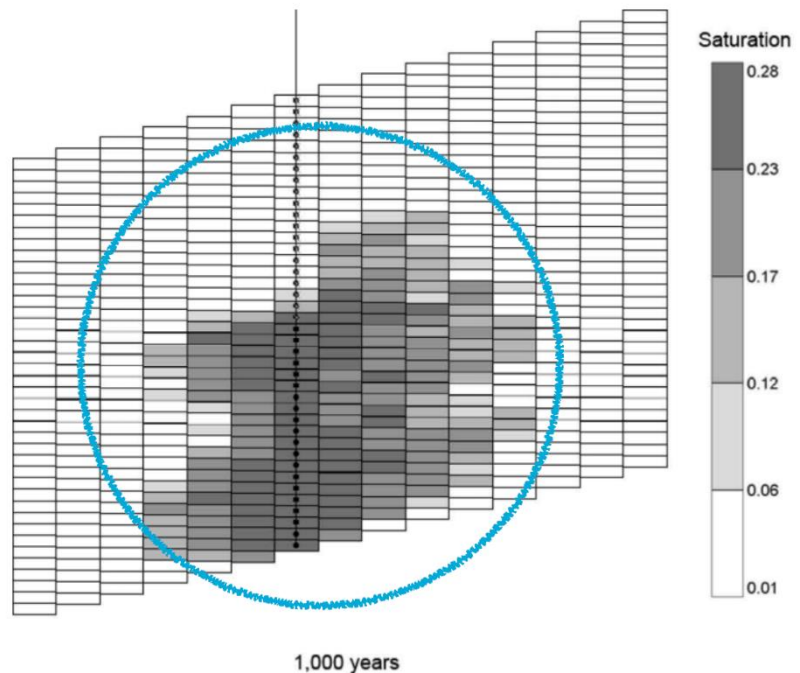
- Residually trapped CO₂ is crucial to long-term CO₂ entrapment



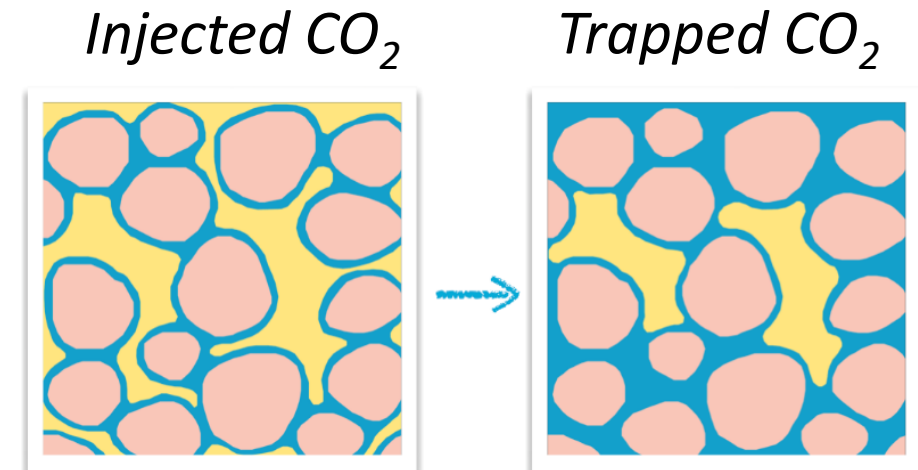
(Kumar et al., 2005)

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- Residually trapped CO₂ is in the form of CO₂ ganglia trapped in pore spaces by capillary forces

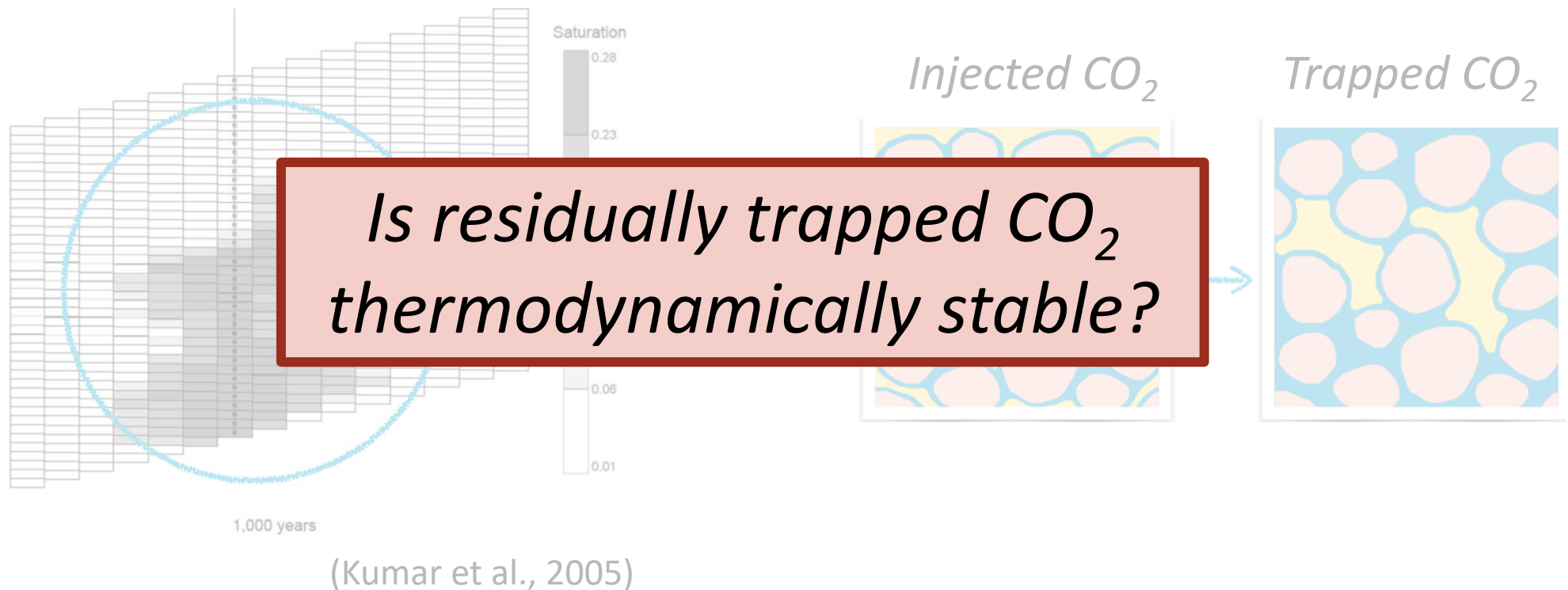


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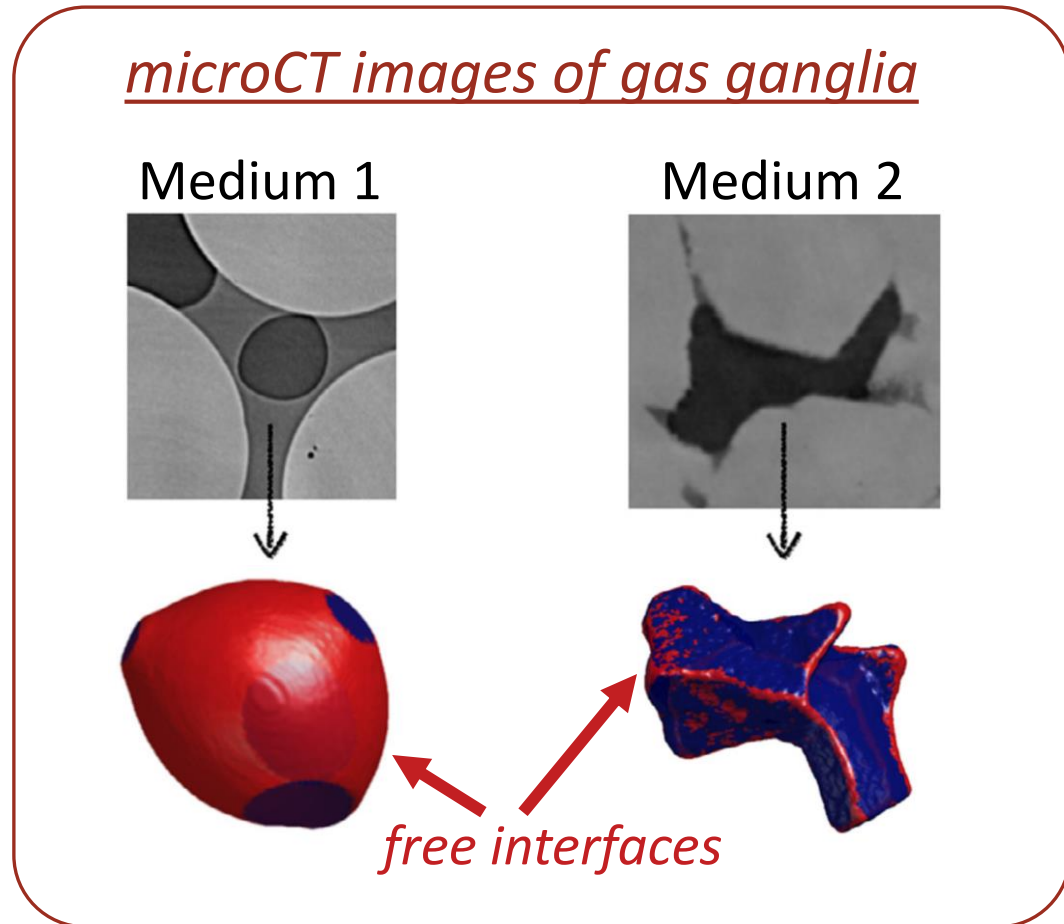


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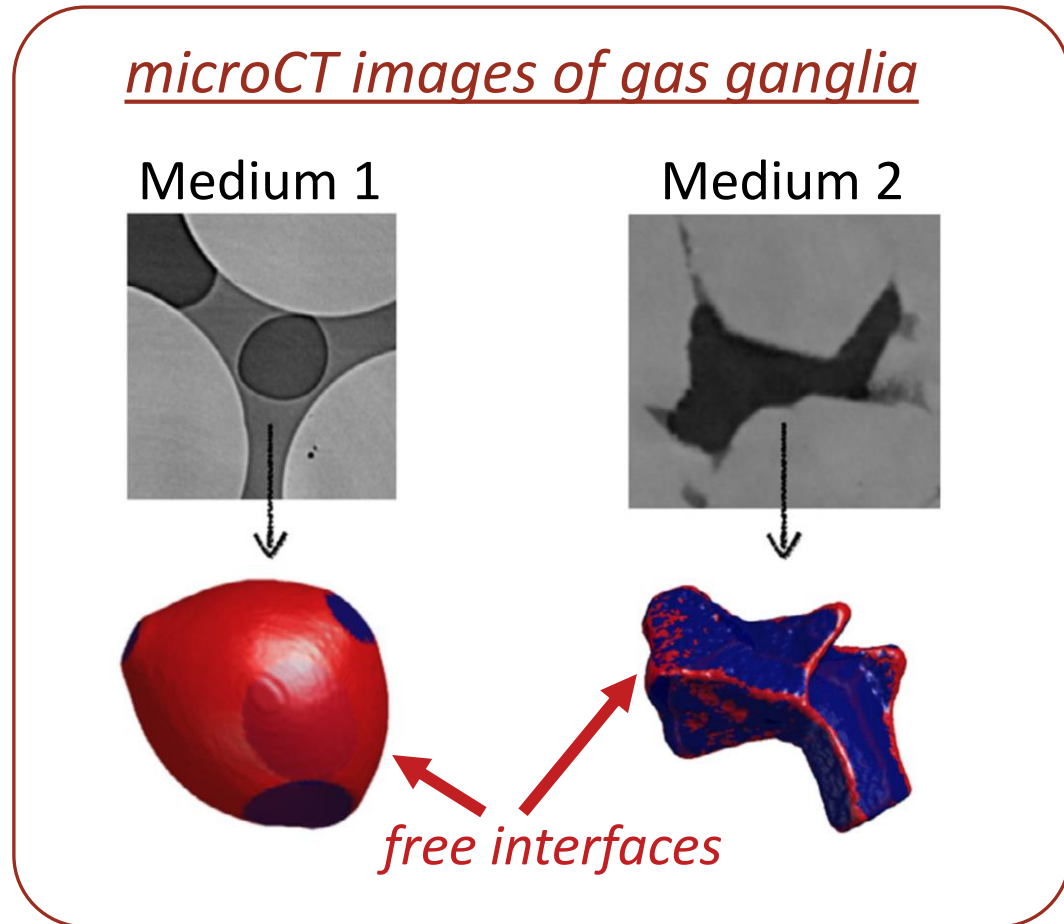


What do residually trapped gas ganglia look like?



(Modified from Garing et al., 2017)

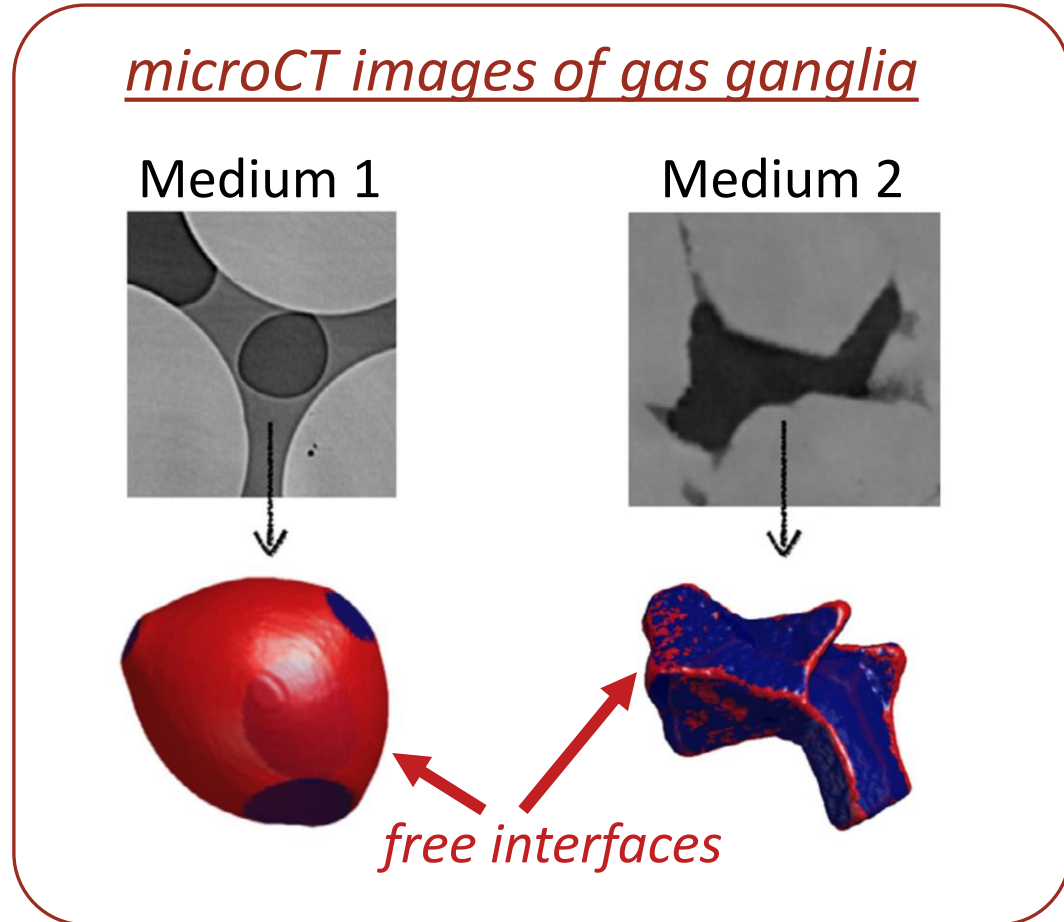
What do residually trapped gas ganglia look like?



Strongly deformed by
local pore matrices

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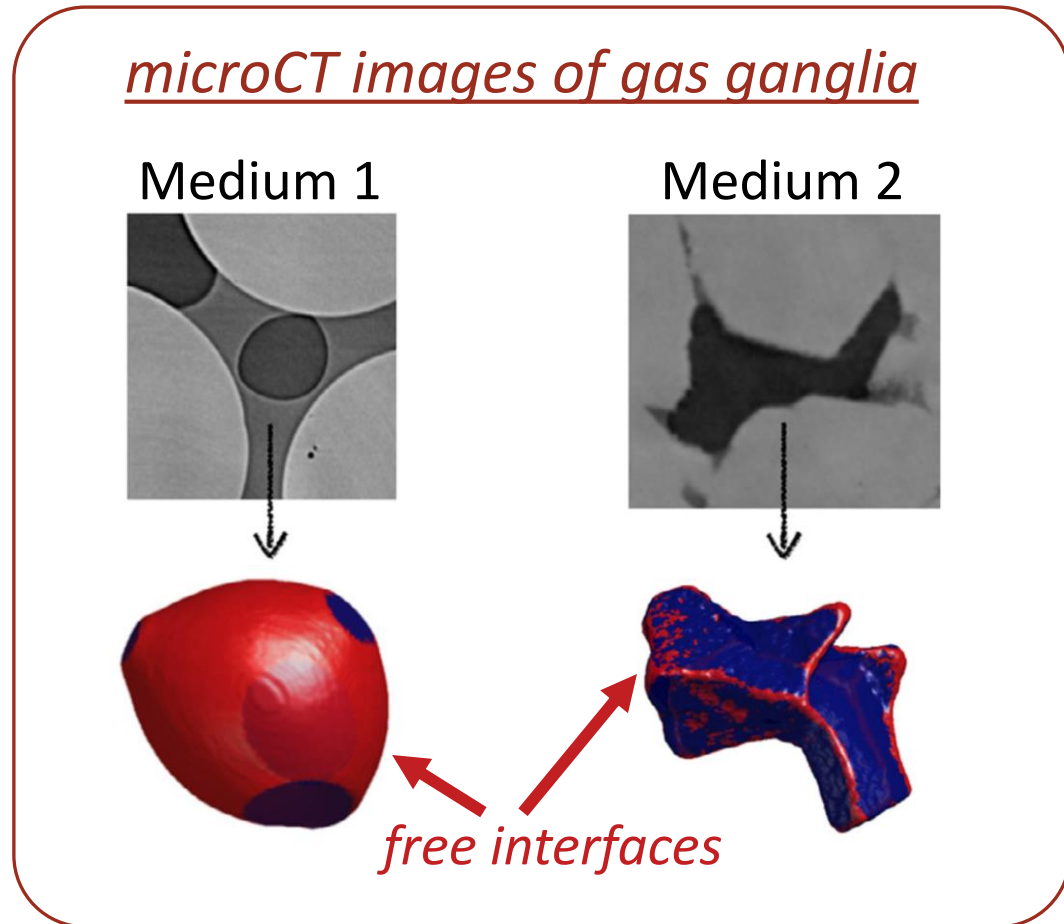
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Very different
interfacial curvatures

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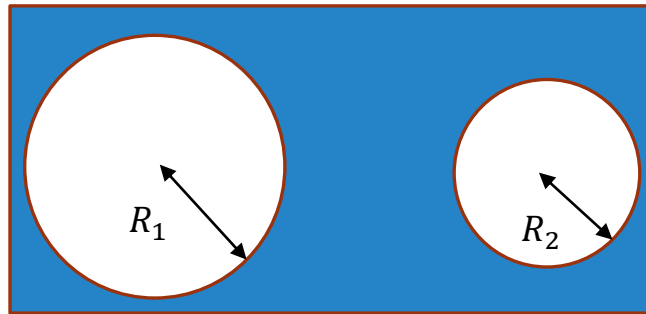


Ostwald ripening

Interfacial curvature difference drives Ostwald ripening – in a bulk fluid

Ostwald ripening in a bulk fluid

Initial condition



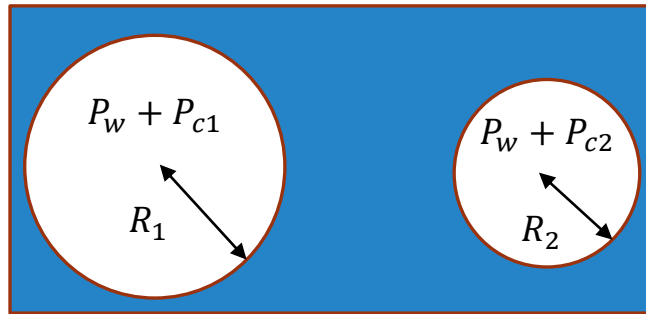
Interfacial curvature
difference



Interfacial curvature difference drives Ostwald ripening – in a bulk fluid

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Interfacial curvature
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$$P_c = \frac{2\sigma}{R}$$

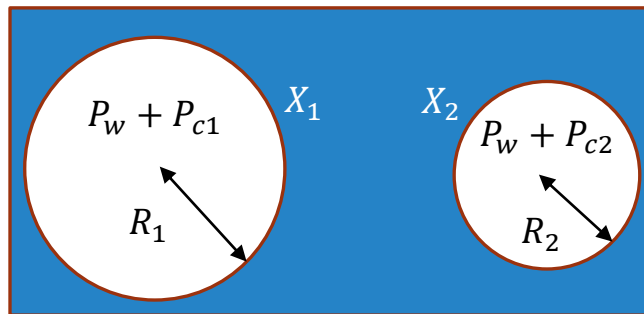
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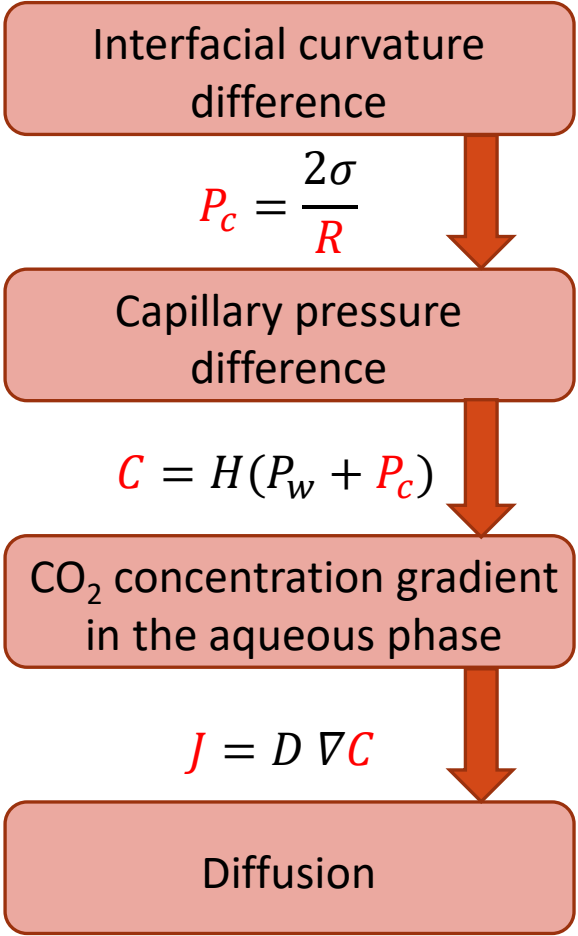
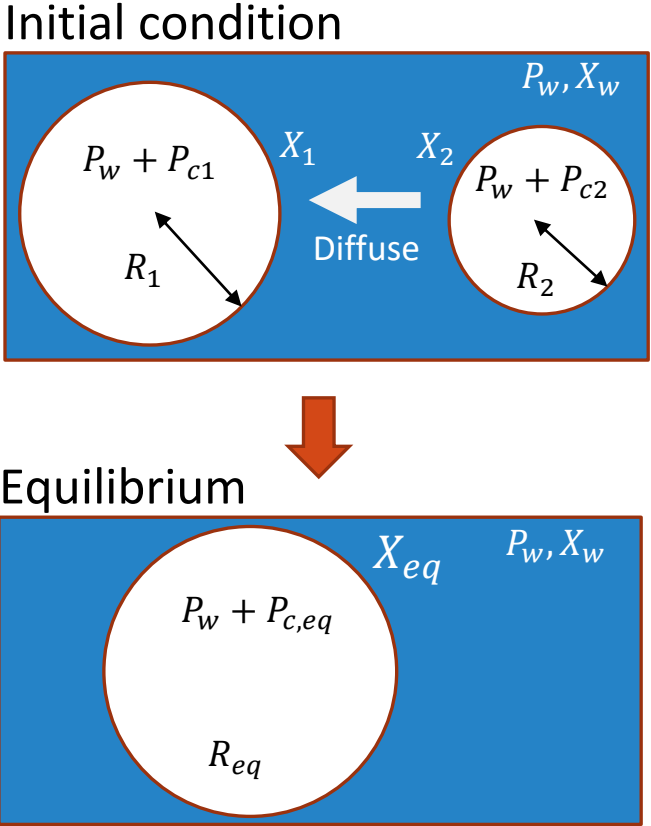
$$C = H(P_w + P_c)$$

CO₂ concentration gradient
in the aqueous phase

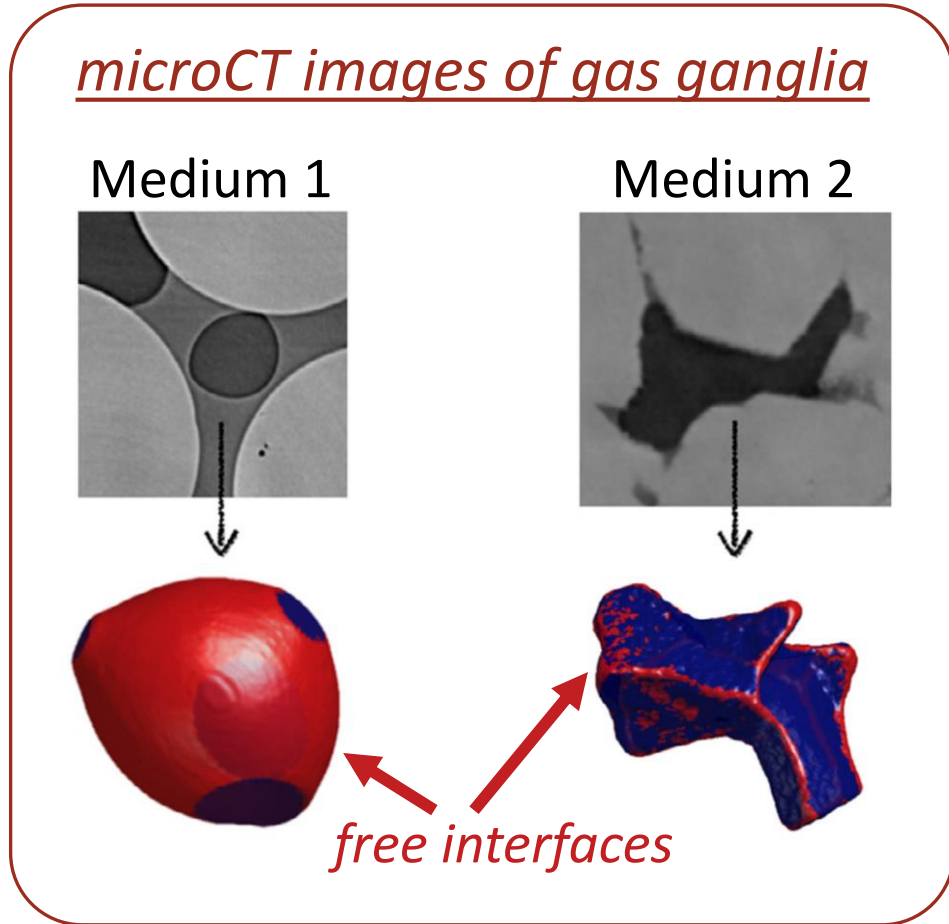


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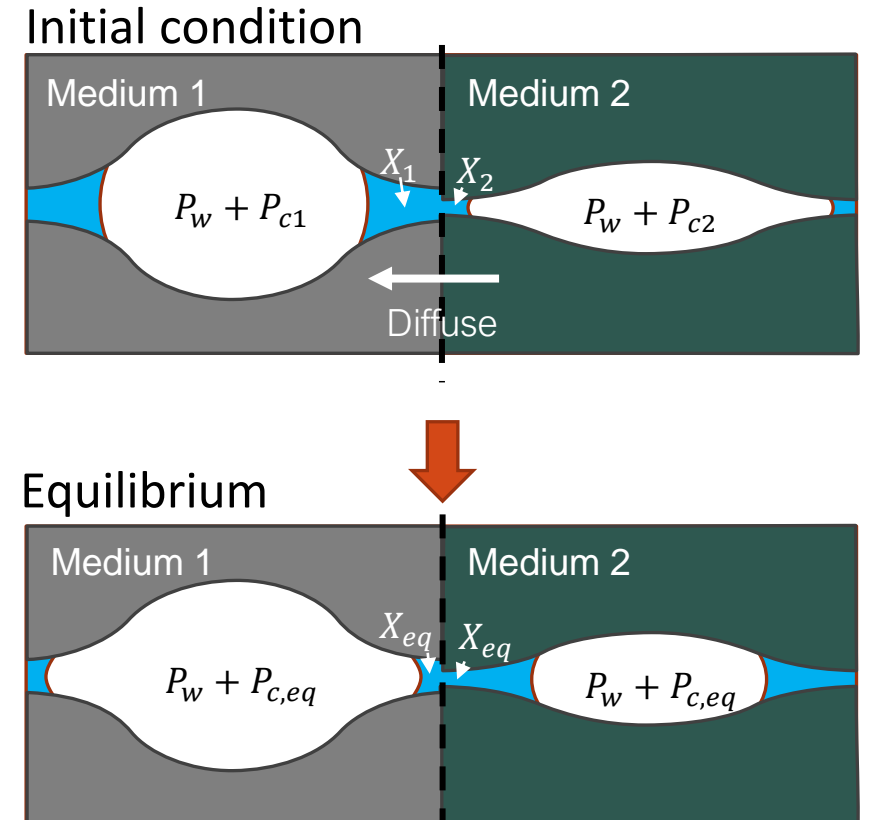
Ostwald ripening can also occur in porous media, where the ganglia approach the same interfacial curvature eventually



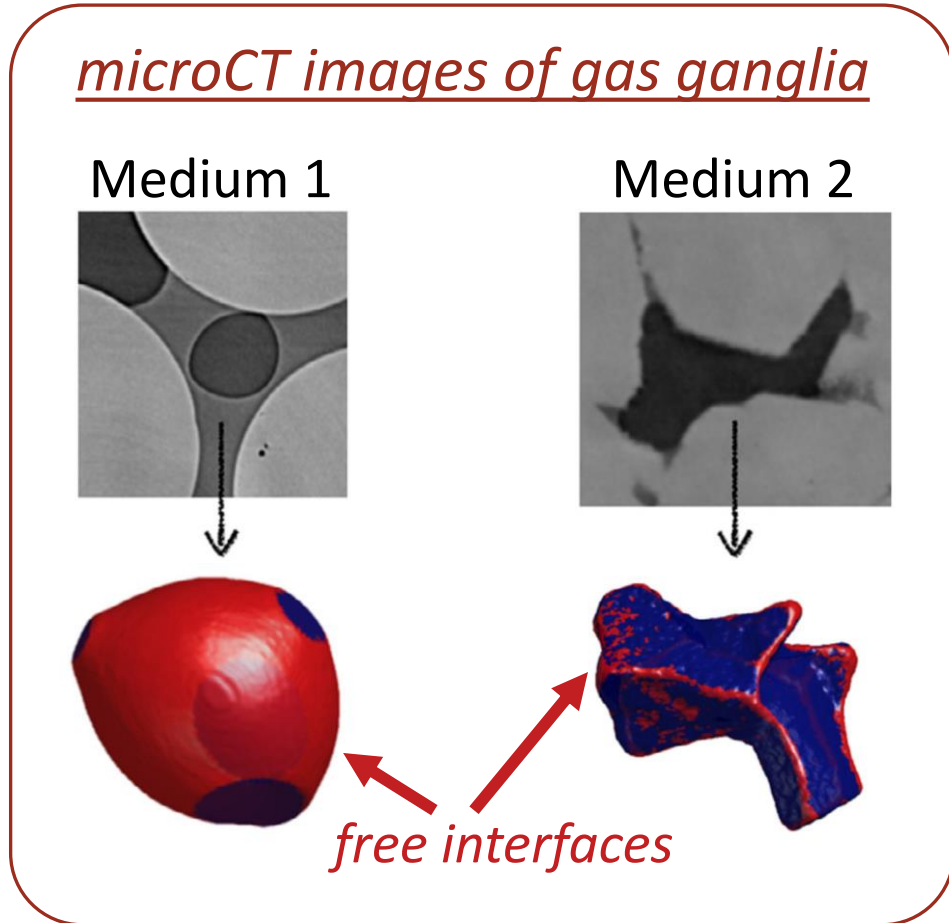
(Modified from Garing et al., 2017)

Conceptualize

Ostwald ripening in porous media

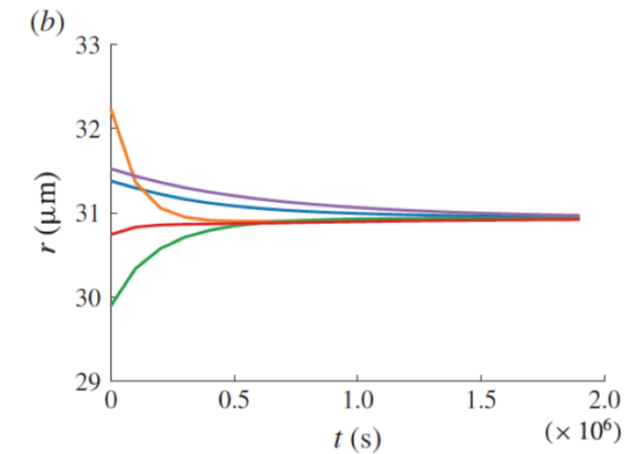
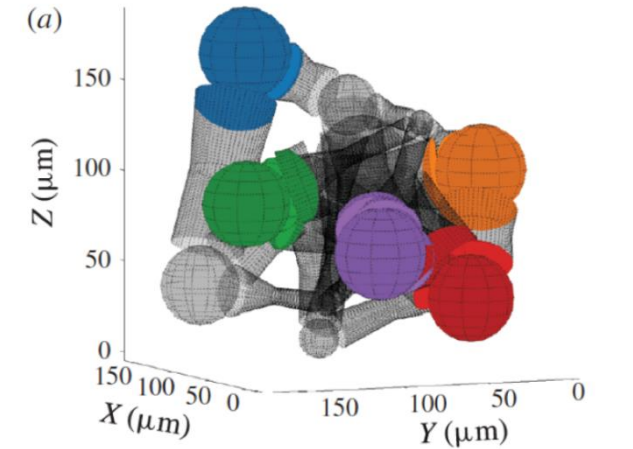


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Numerical
simulation



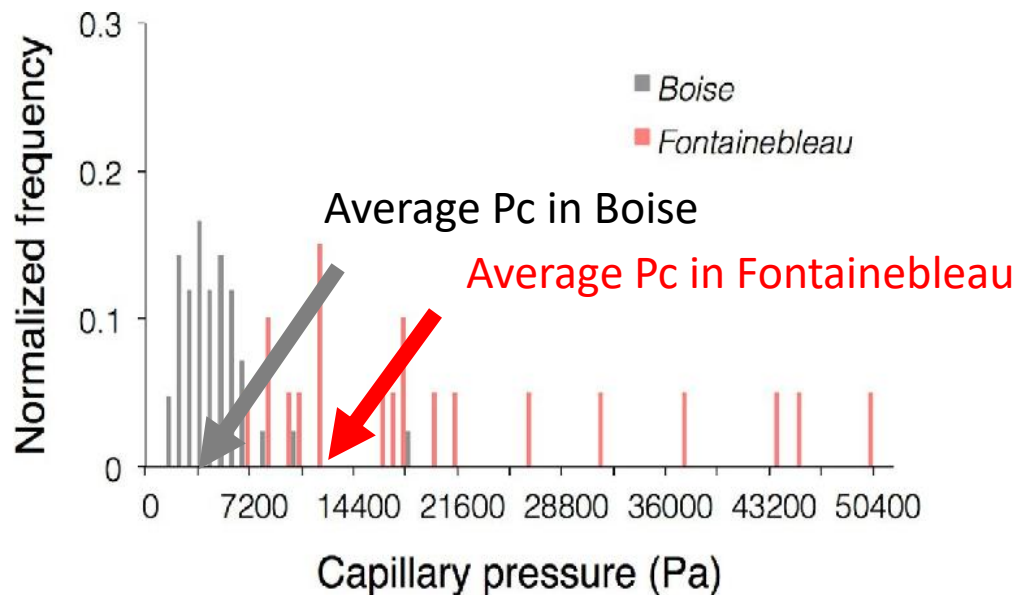
(de Chalendar et al., 2018)



The finding on equilibrium interfacial curvature allows us to explore the impact of Ostwald ripening at the continuum scale

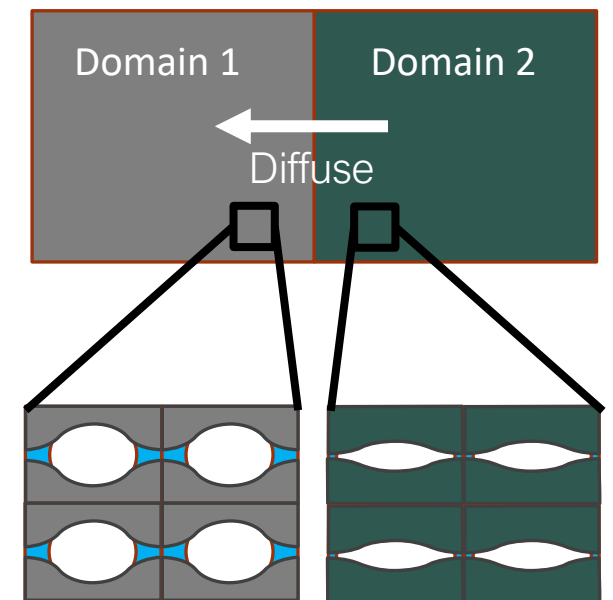
- This local **equilibrium capillary pressure varies** in different porous media, roughly at the same order of the entry pressure.
- We build a conceptual model to describe the impact of Ostwald ripening at the continuum scale

(Garing et al., 2017)



Conceptualize

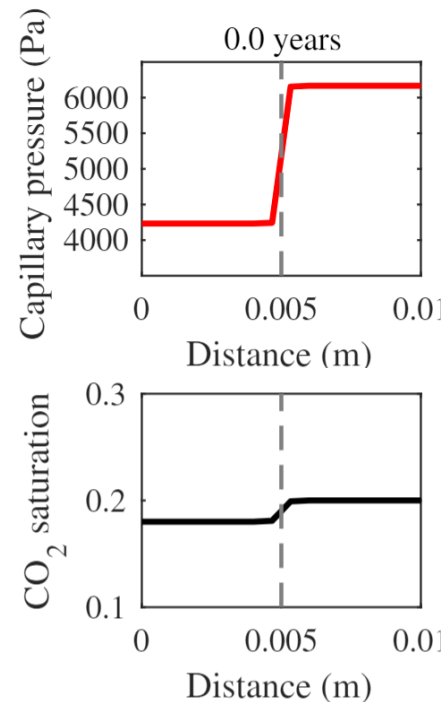
The conceptual model at the continuum scale



At the continuum scale, Ostwald ripening can change the gas saturation distribution while the gas remains trapped at a time scale of 10^5 years

Initial state

Equilibrium

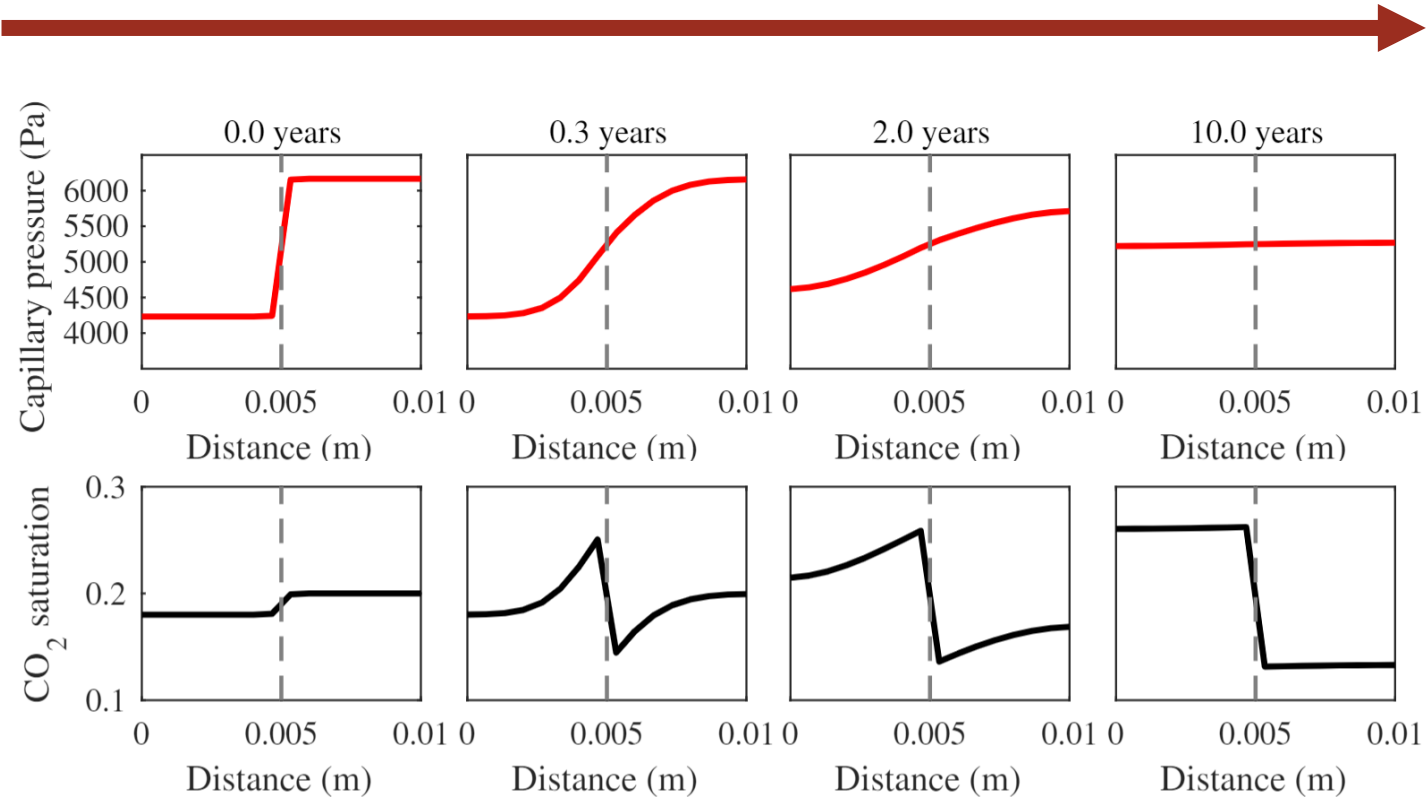


Li, Y., Garing, C., & Benson, S. (2020). A continuum-scale representation of Ostwald ripening in heterogeneous porous media. *Journal of Fluid Mechanics*, 889, A14. doi:10.1017/jfm.2020.53

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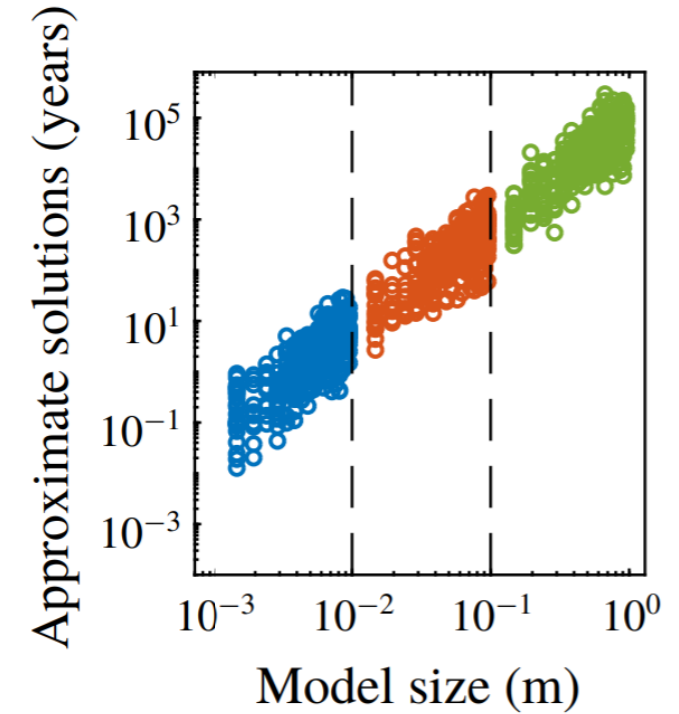
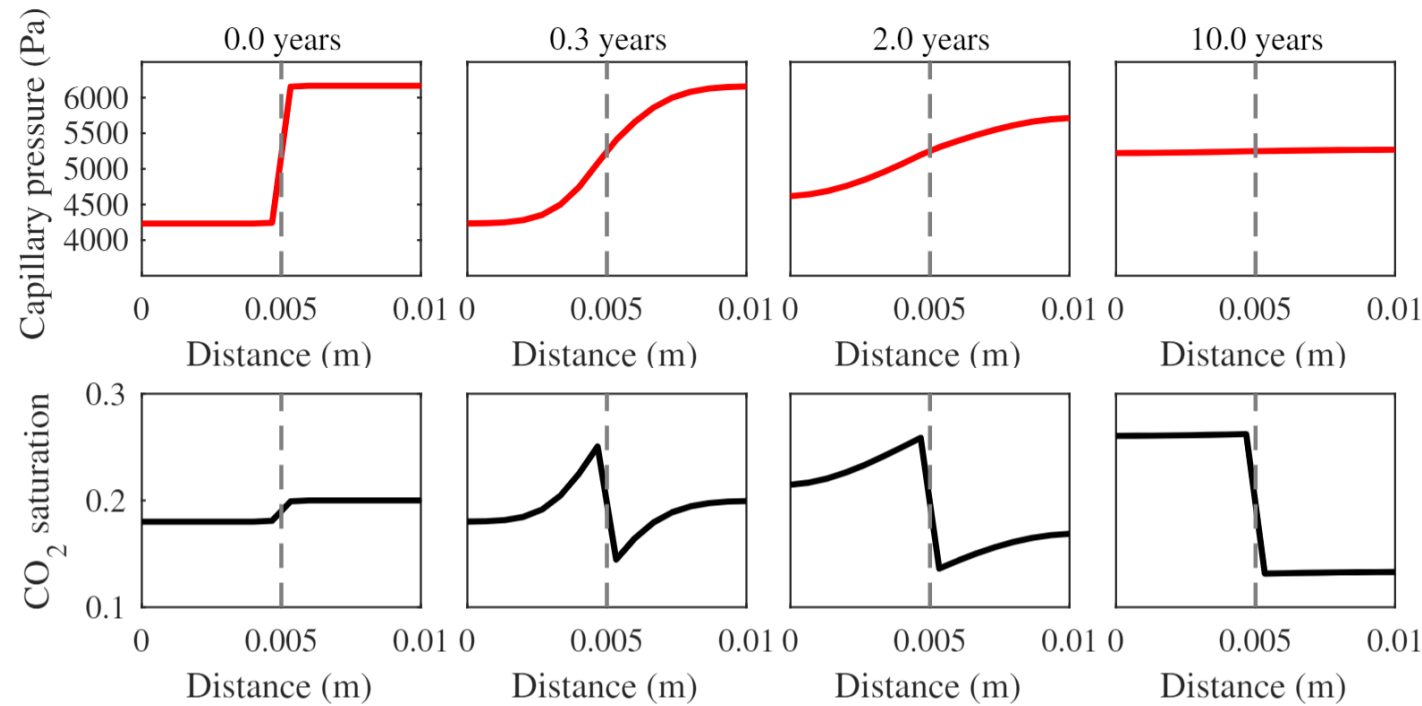


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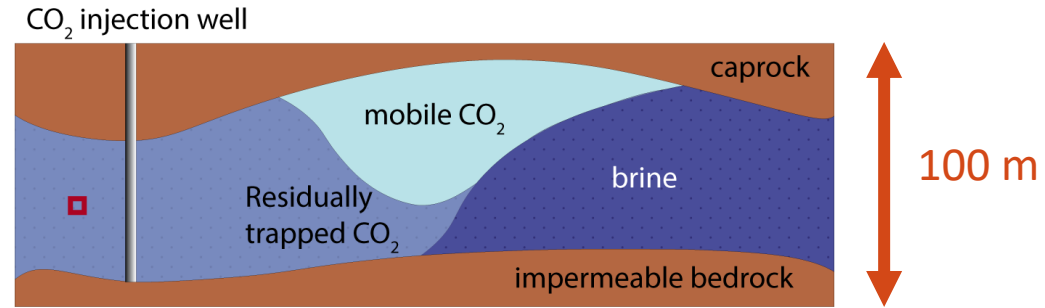
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 - Capillary heterogeneity

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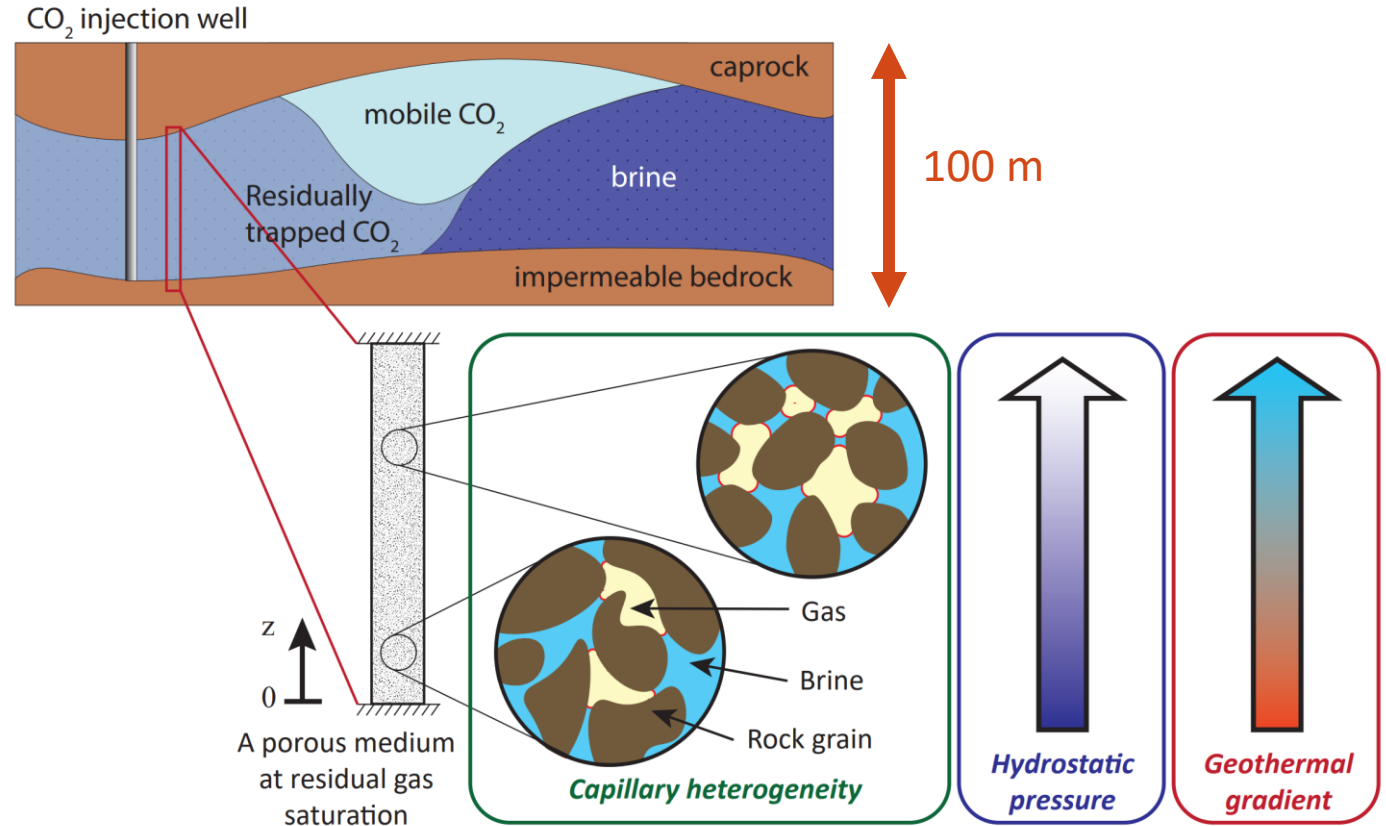


Li, Y. & Benson, S., Diffusion-induced gas redistribution in porous media, *under review*

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 - Capillary heterogeneity
 - Hydrostatic pressure
 - Geothermal gradients

At a larger scale



Li, Y. & Benson, S., Diffusion-induced gas redistribution in porous media, *under review*

Generalized Fick's law – from concentration to energy

Fick's law - concentration

$$J_{CO_2}^{aq} = -D \nabla c_{CO_2}$$

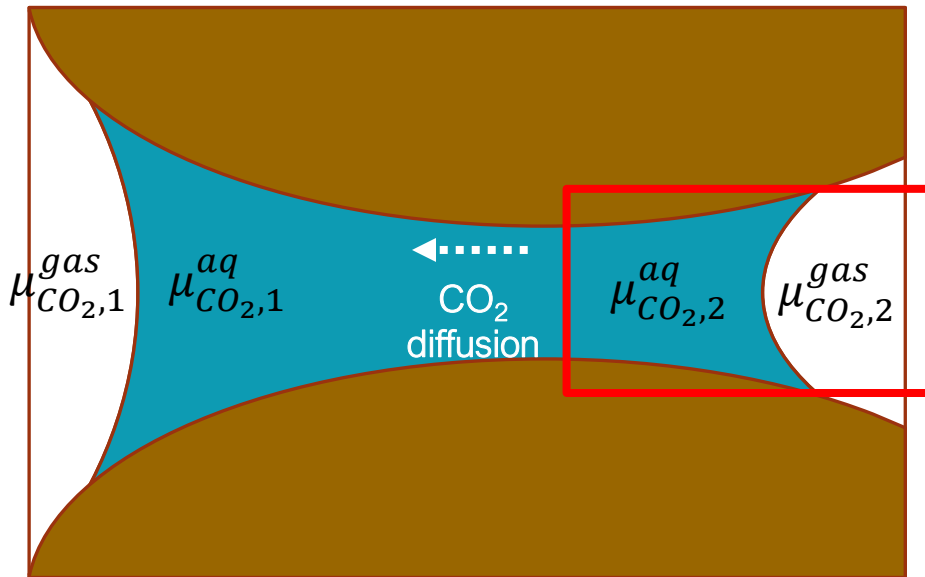
Generalized Fick's law - chemical potential

$$J_{CO_2}^{aq} = -\frac{D c_{CO_2}}{RT} \nabla \mu_{CO_2}^{aq}$$

Generalized Fick's law – other thermodynamic potentials?

$$J_{CO_2}^{aq} = -\frac{D c_{CO_2}}{RT} \nabla F_{CO_2}^{aq}$$

Diffusion between two CO₂ ganglia



Local equilibrium near interface:

$$\mu_{CO_2}^{gas} = \mu_{CO_2}^{aq}$$

For an ideal mixture:

$$\mu_{CO_2}^{aq} = \mu_0 + RT \ln(x_{CO_2})$$

The driving force for diffusion consists of three sub-forces

Generalized Fick's law – other thermodynamic potentials?

$$J_{CO_2}^{aq} = -\frac{Dc_{CO_2}}{RT} \nabla F_{CO_2}^{aq}$$

$\nabla F_{CO_2}^{aq}$

$$\nabla F_{CO_2}^{aq}(x_{CO_2}, x_w, P, z, T) = \frac{\partial F_{CO_2}^{aq}}{\partial x_{CO_2}} \frac{dx_{CO_2}}{dz} + \frac{\partial F_{CO_2}^{aq}}{\partial x_w} \frac{dx_w}{dz} + \frac{\partial F_{CO_2}^{aq}}{\partial P} \frac{dP}{dz} + \frac{\partial F_{CO_2}^{aq}}{\partial z} \frac{dz}{dz} + \frac{\partial F_{CO_2}^{aq}}{\partial T} \frac{dT}{dz}$$

Solubility-driven diffusion

$$J_{CO_2,x}^{aq} = -\frac{Dc_{CO_2}}{RT} \left(\frac{RT}{x_{CO_2}} \frac{dx_{CO_2}}{dz} \right)$$

Gravitational segregation

$$J_{CO_2,z}^{aq} = -\frac{Dc_{CO_2}}{RT} \left(-M_{CO_2}g \frac{\rho_w - \rho_{CO_2}}{\rho_{CO_2}} \right)$$

Ludwig-Soret effect

$$J_{CO_2,T}^{aq} = -\frac{Dc_{CO_2}}{RT} \left(R\alpha_T x_w \frac{dT}{dz} \right)$$

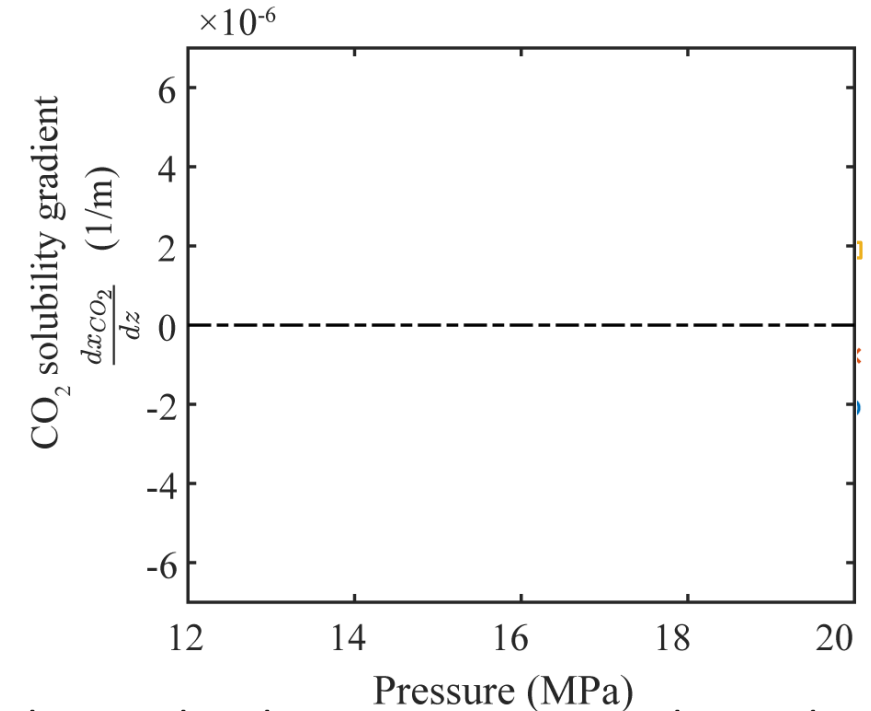
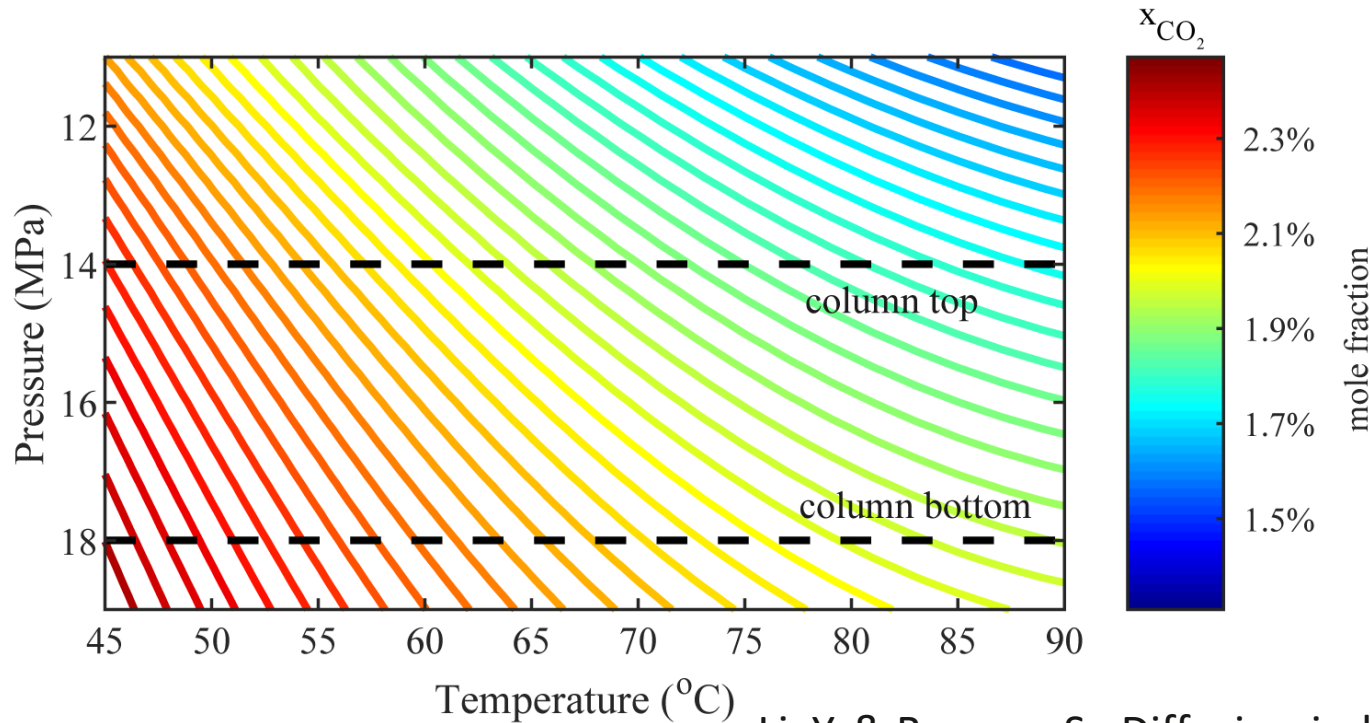
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Solubility-driven diffusion – the solubility gradient strongly depends on the reservoir conditions

- CO₂ solubility is non-uniform under hydrostatic pressure and geothermal gradients
- Geothermal gradients can reduce or even alter the solubility gradient

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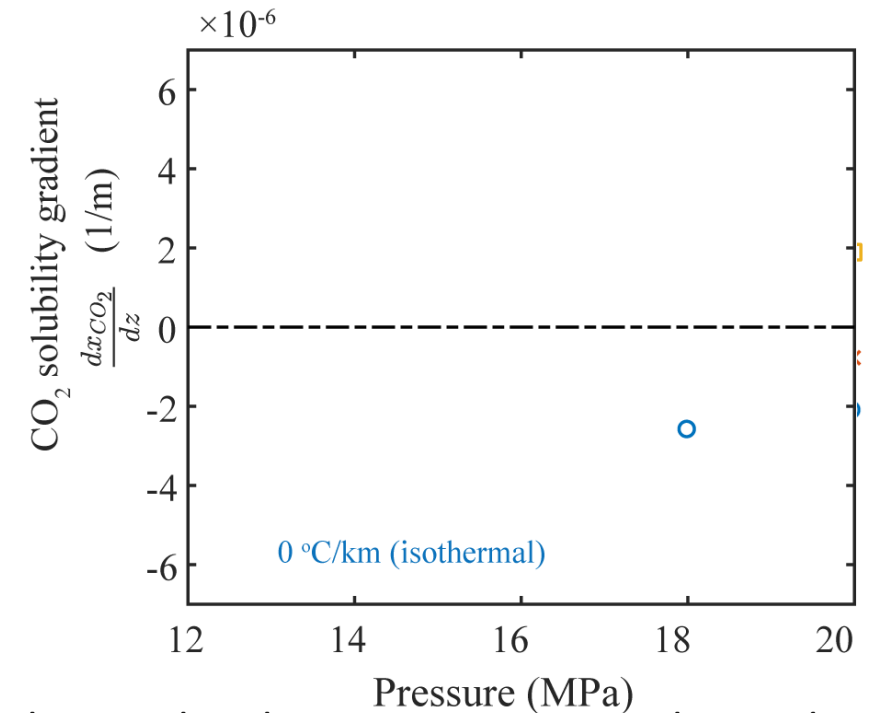
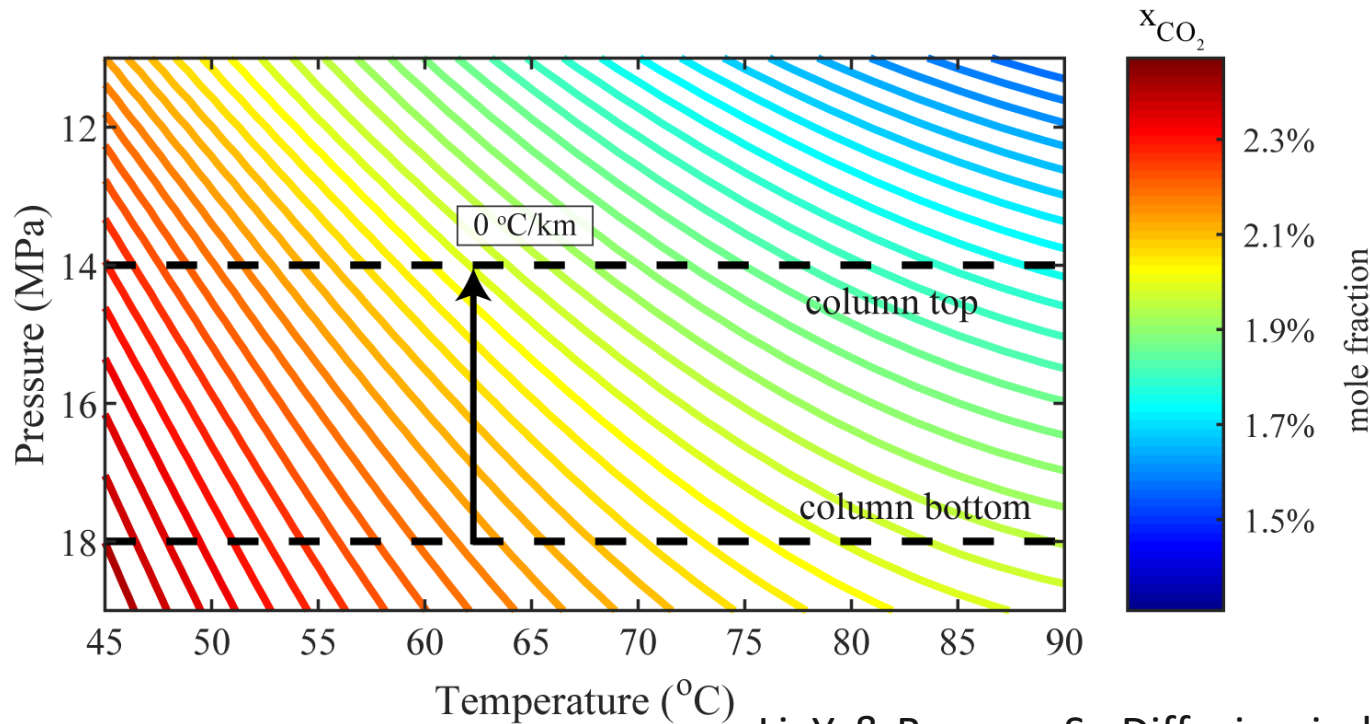
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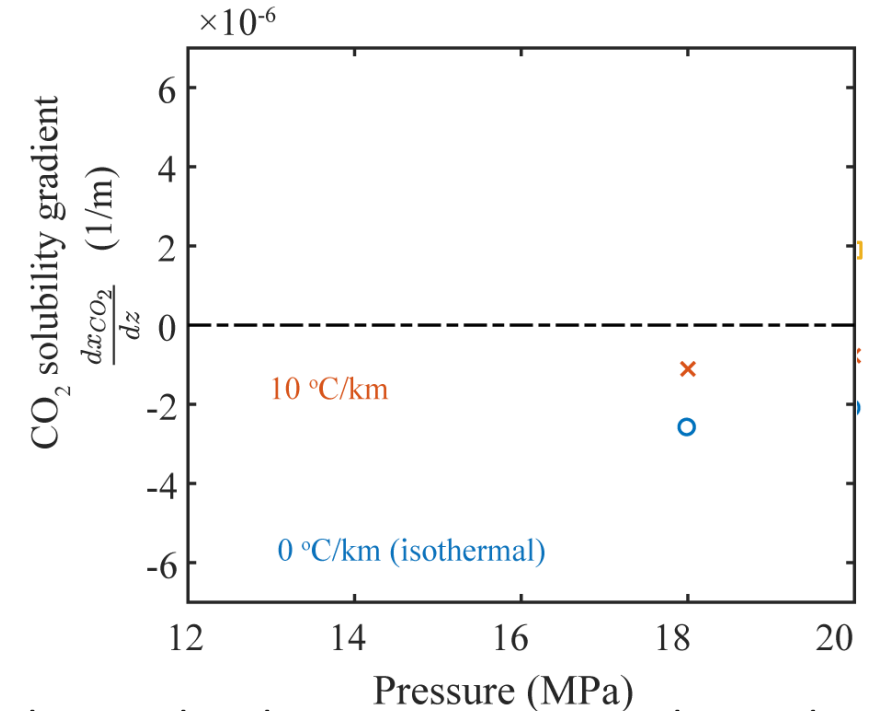
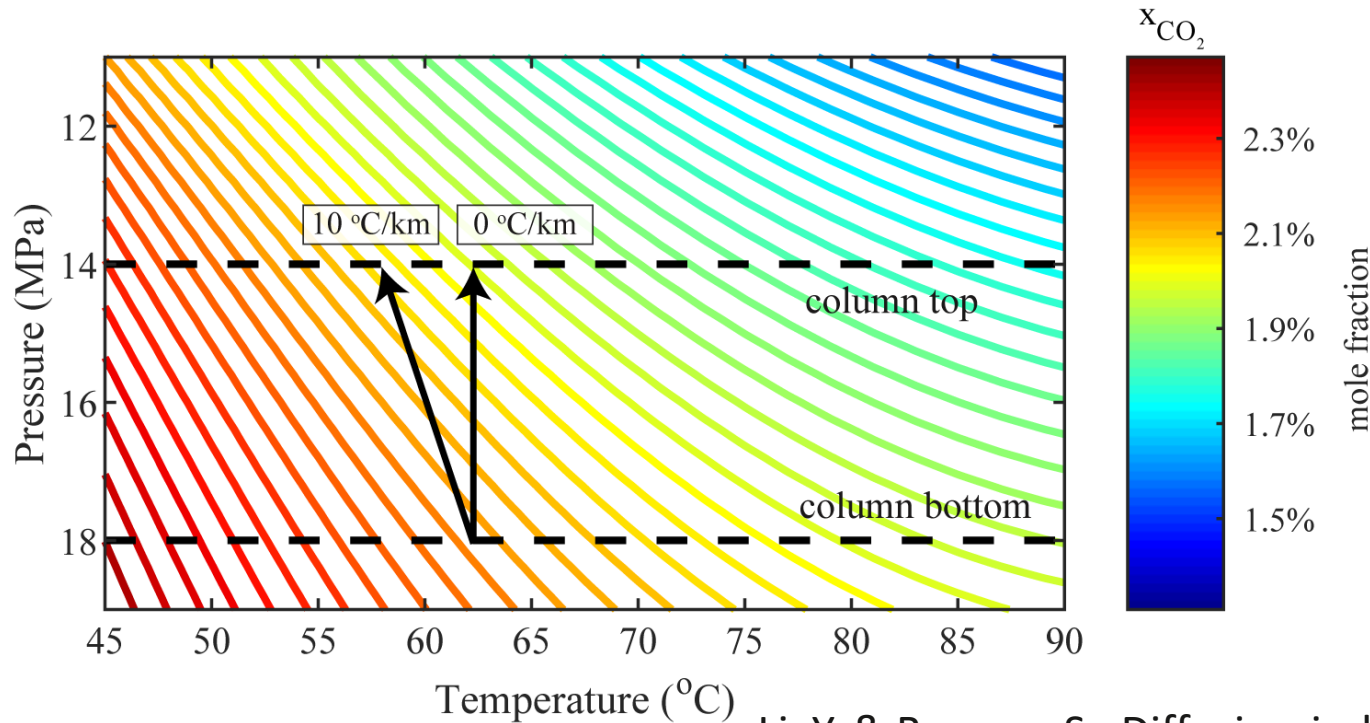
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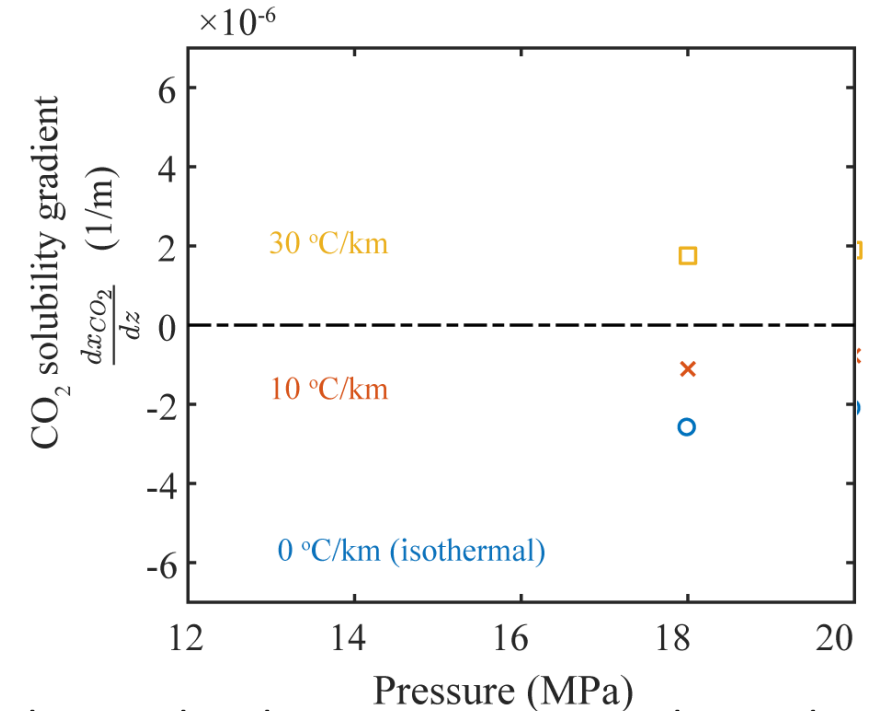
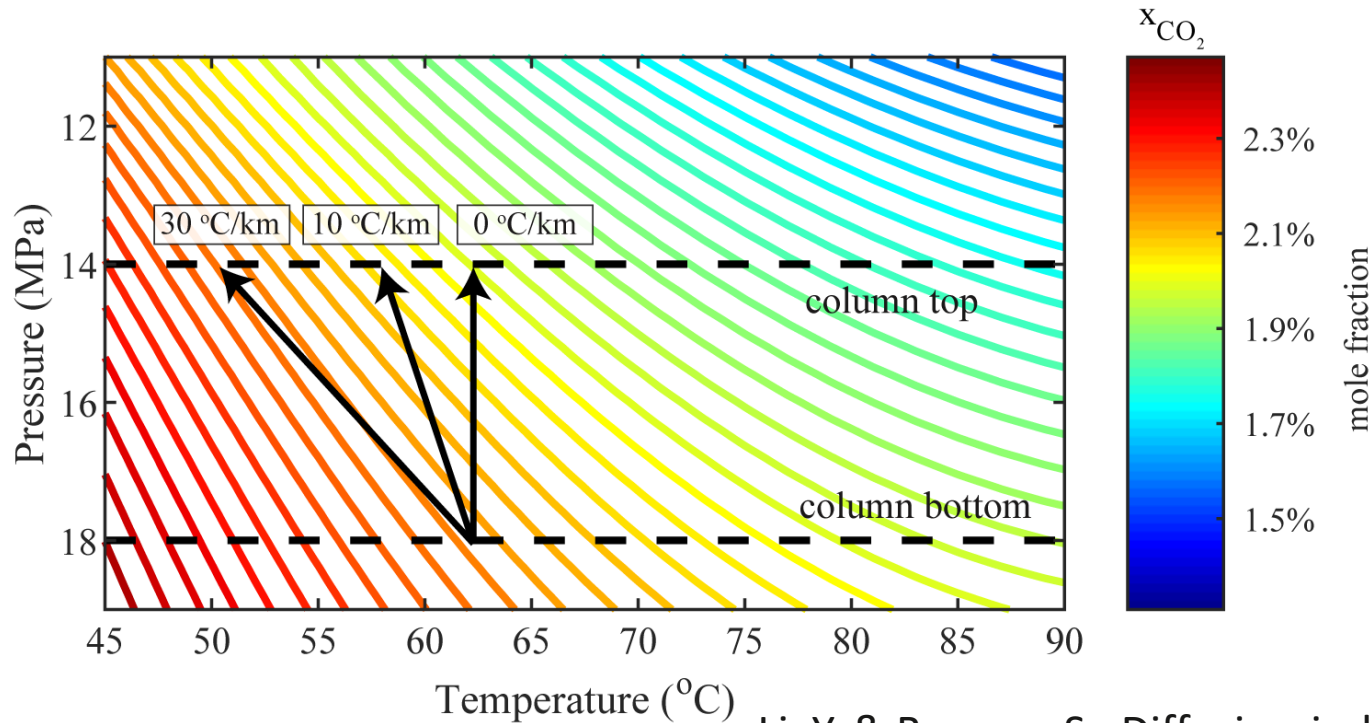
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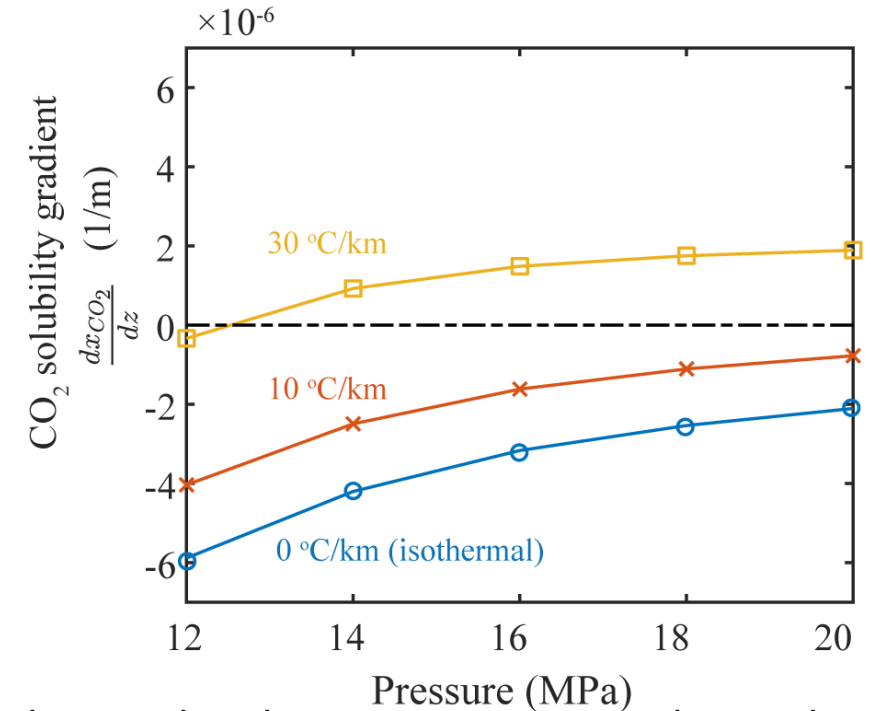
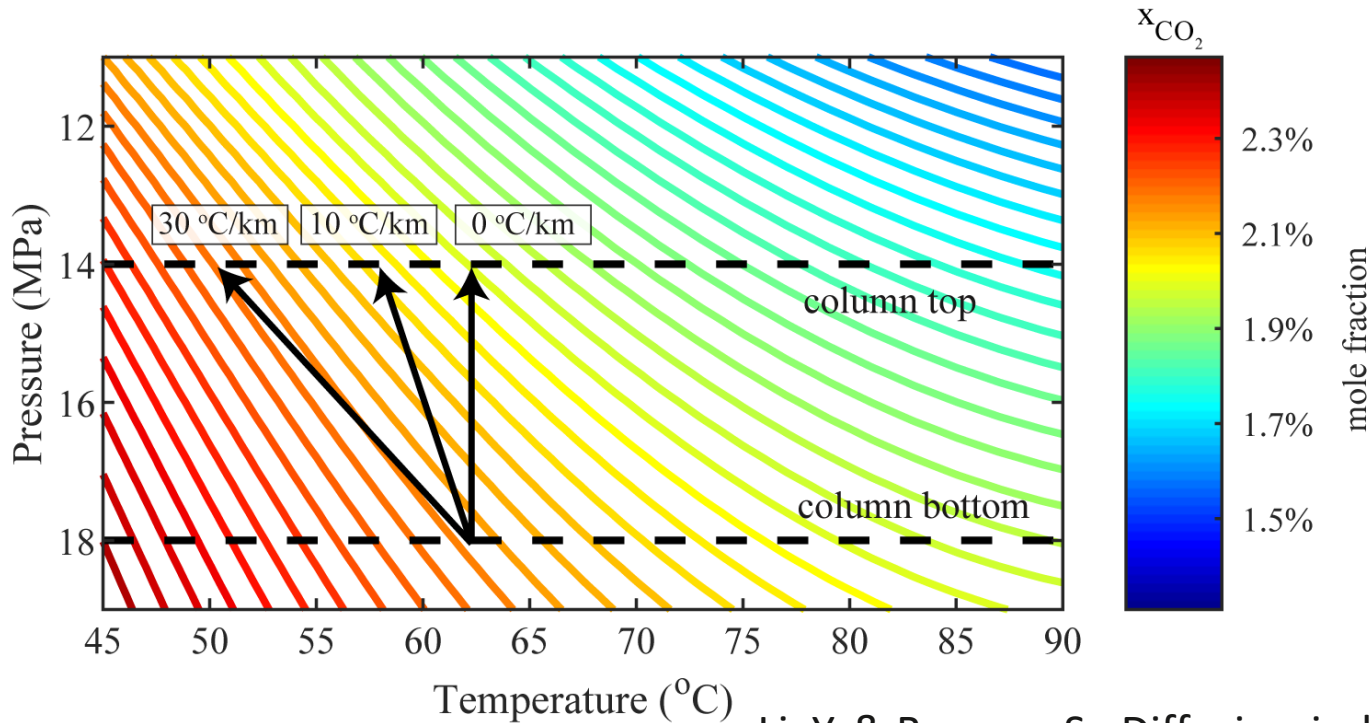
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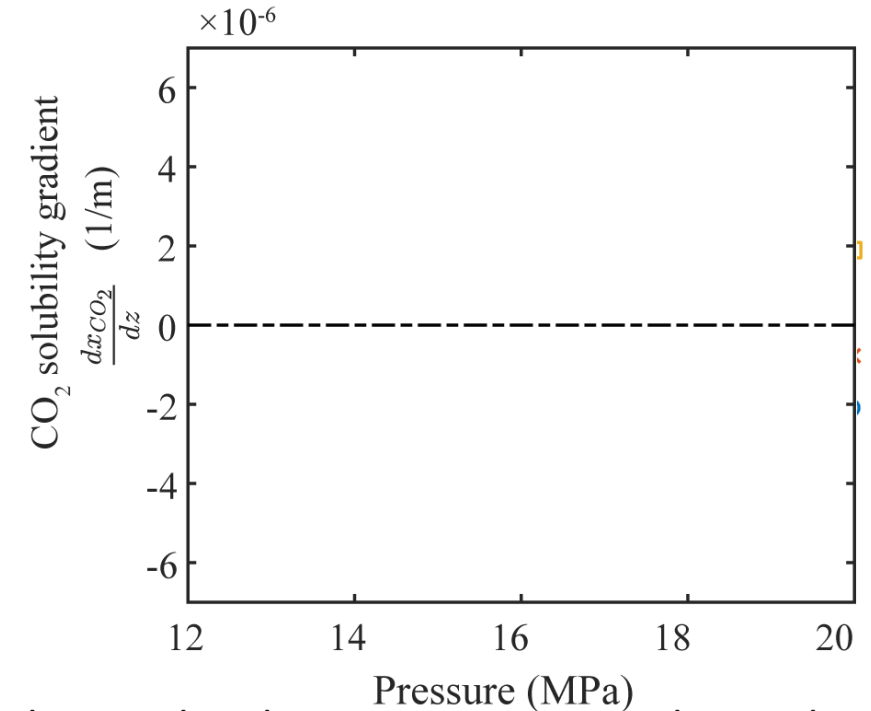
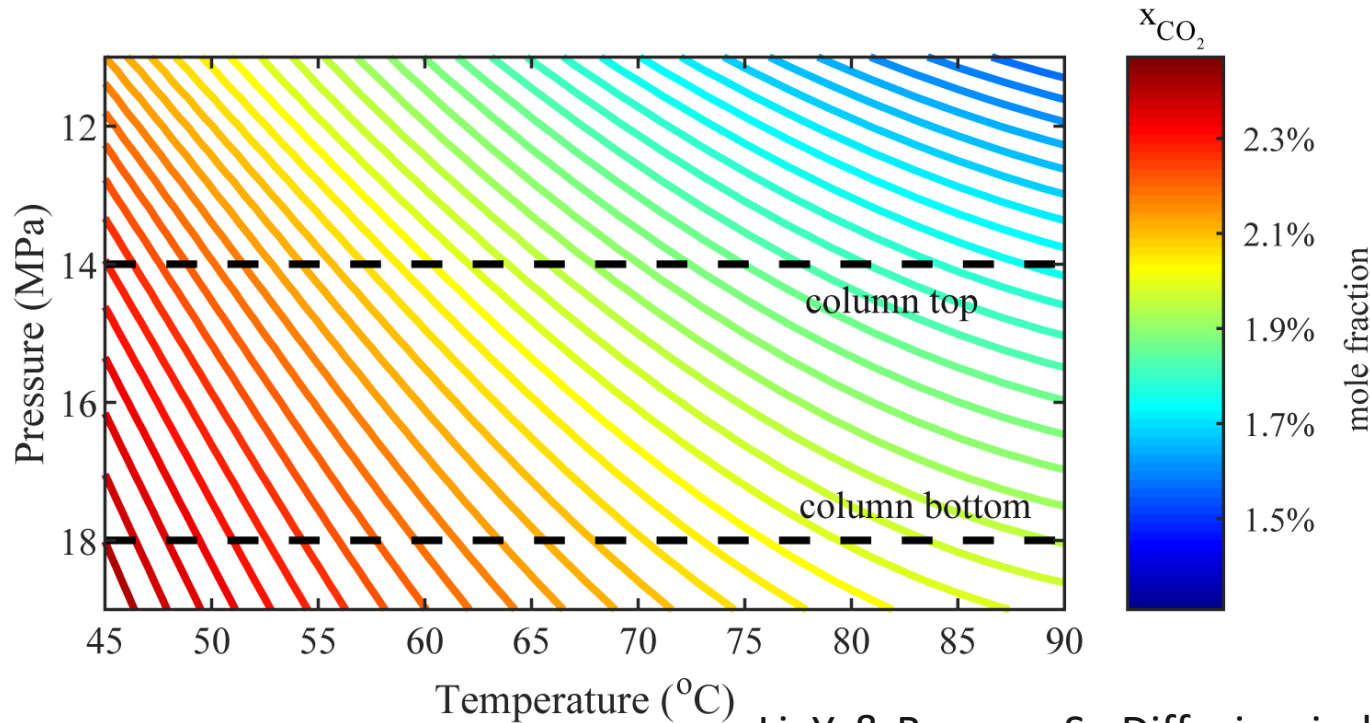
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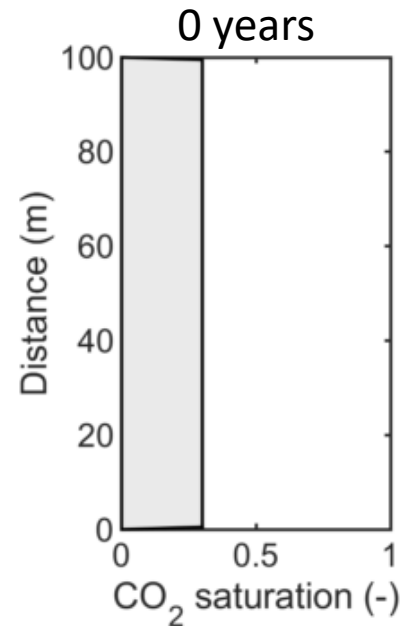
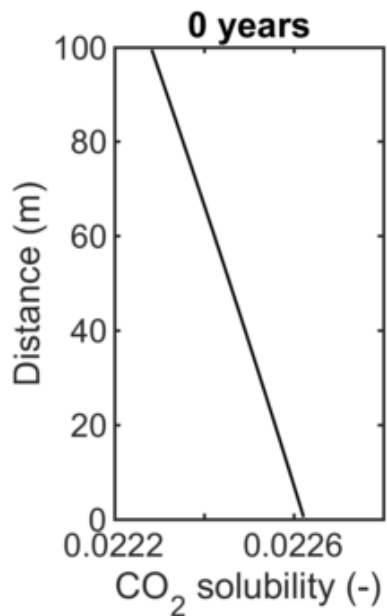


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Solubility-driven diffusion – the gas phase redistributes driven by diffusion and accumulates at the low-solubility end

- We compute the gas redistribution by numerically solving Fick's second law
- Initial condition – $S_{\text{CO}_2} = 0.3$ everywhere
- Reservoir condition – 50°C and 16 MPa

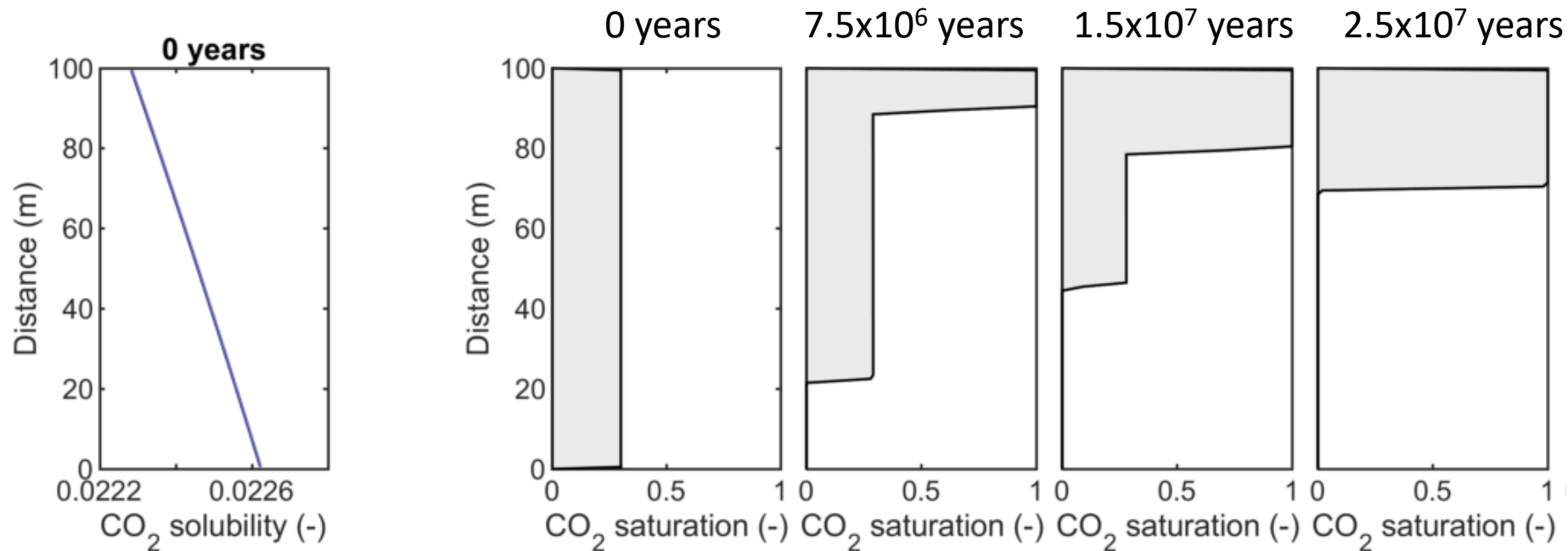
$$\frac{\partial m}{\partial t} = \frac{\partial J}{\partial z}$$



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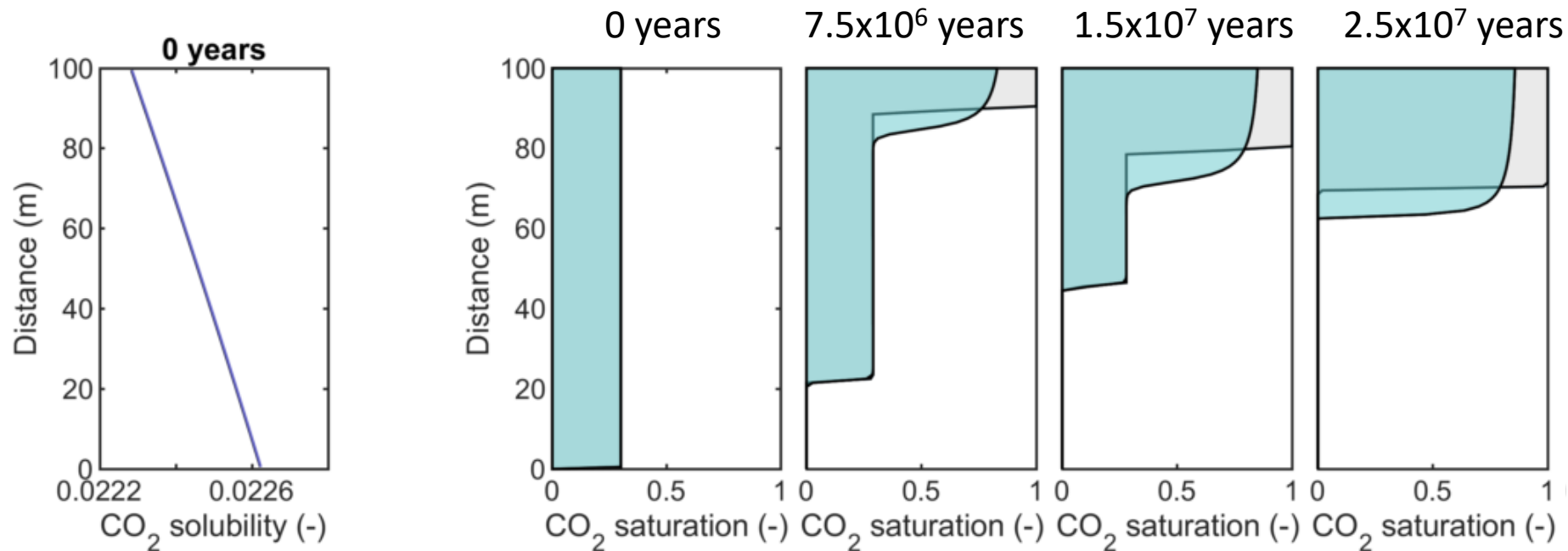
- Isothermal reservoir condition, without capillary pressure
- Isothermal reservoir condition, homogeneous with capillary pressure
- Isothermal reservoir condition, heterogeneous with capillary pressure
- Non-isothermal reservoir condition, homogeneous with capillary pressure



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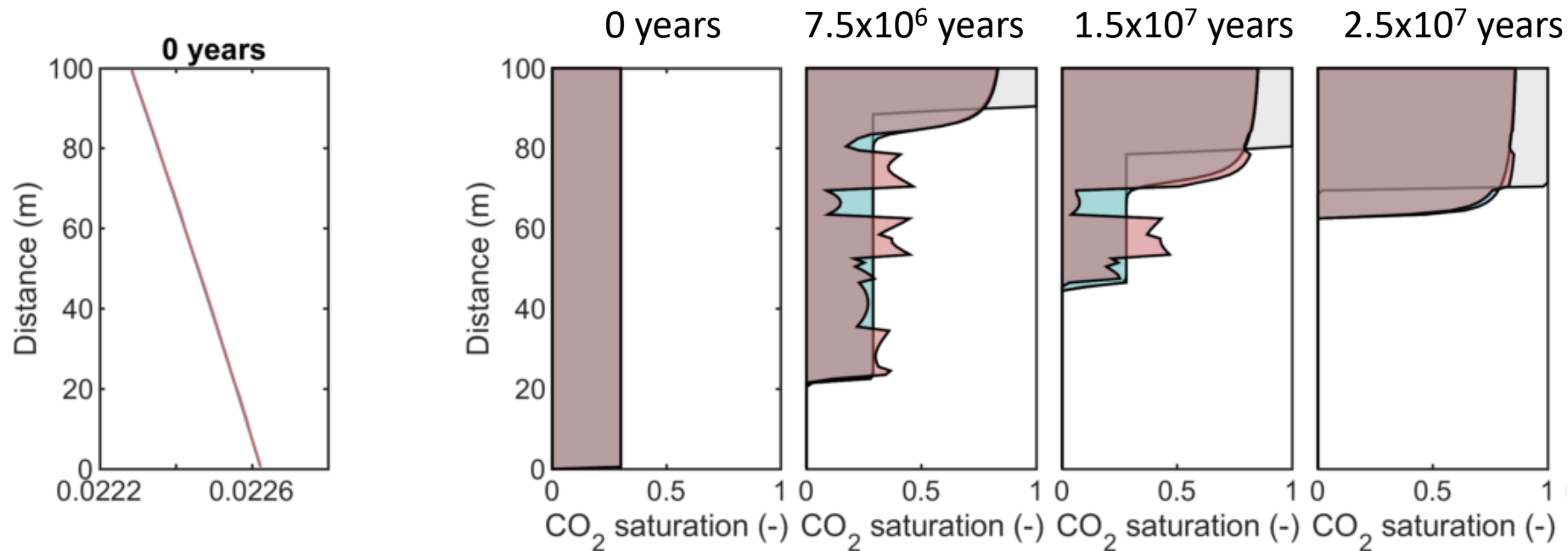
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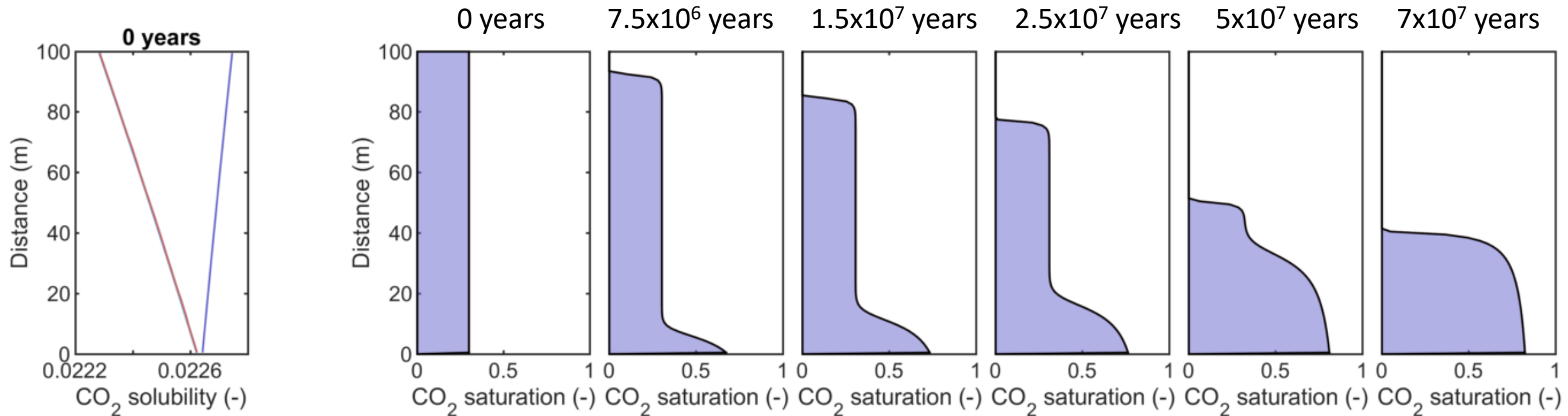
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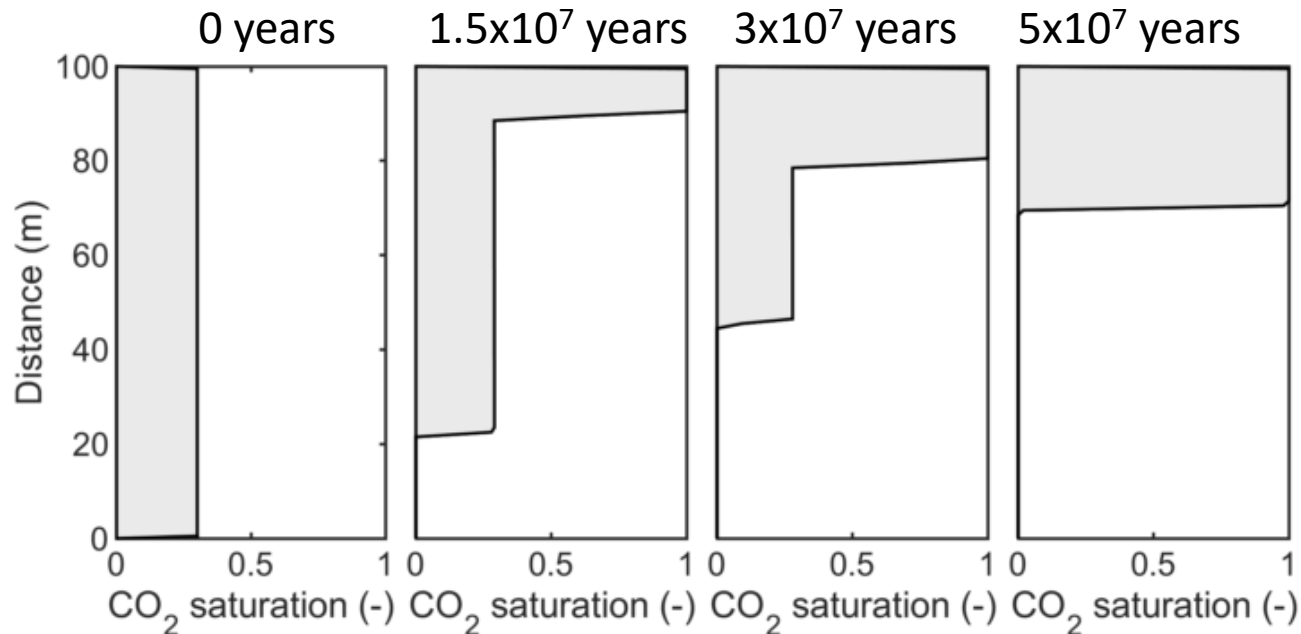
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Gravitational segregation – the gas phase always accumulates under the seal

- The diffusive flux is always upward under reservoir conditions
- The characteristics of gas redistribution is similar to solubility driven without capillary pressure

Gravitational segregation

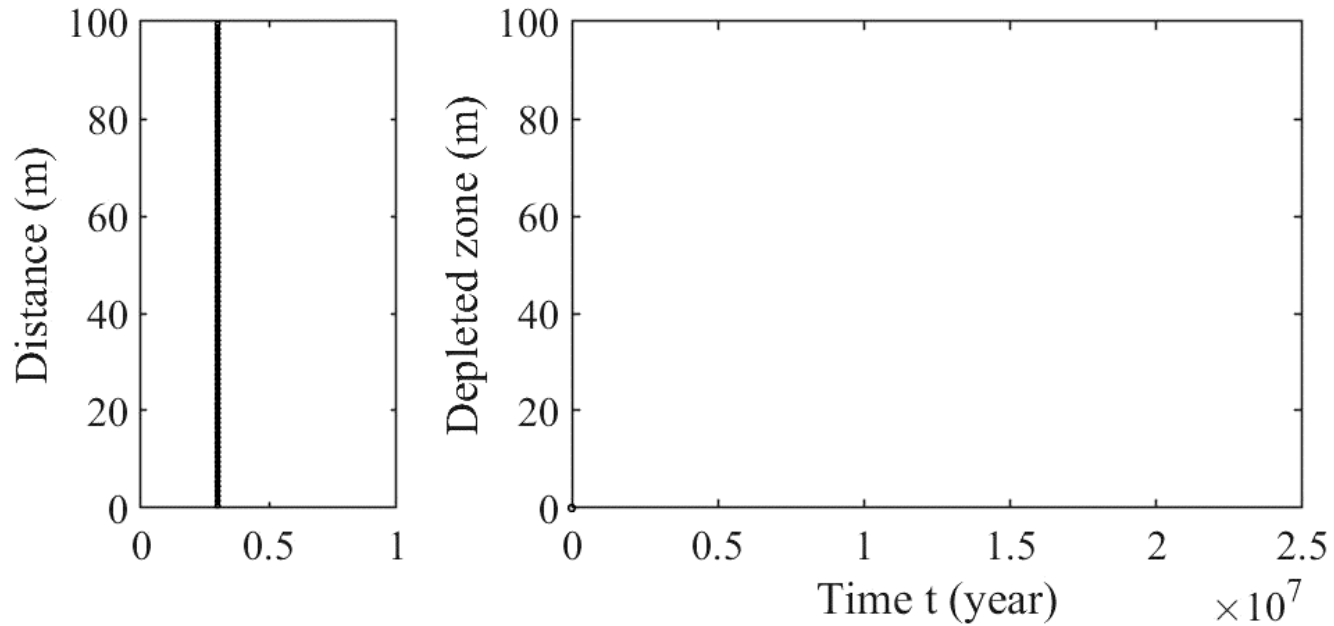
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The time scale of gas redistribution is at the order of 10^5 years/m

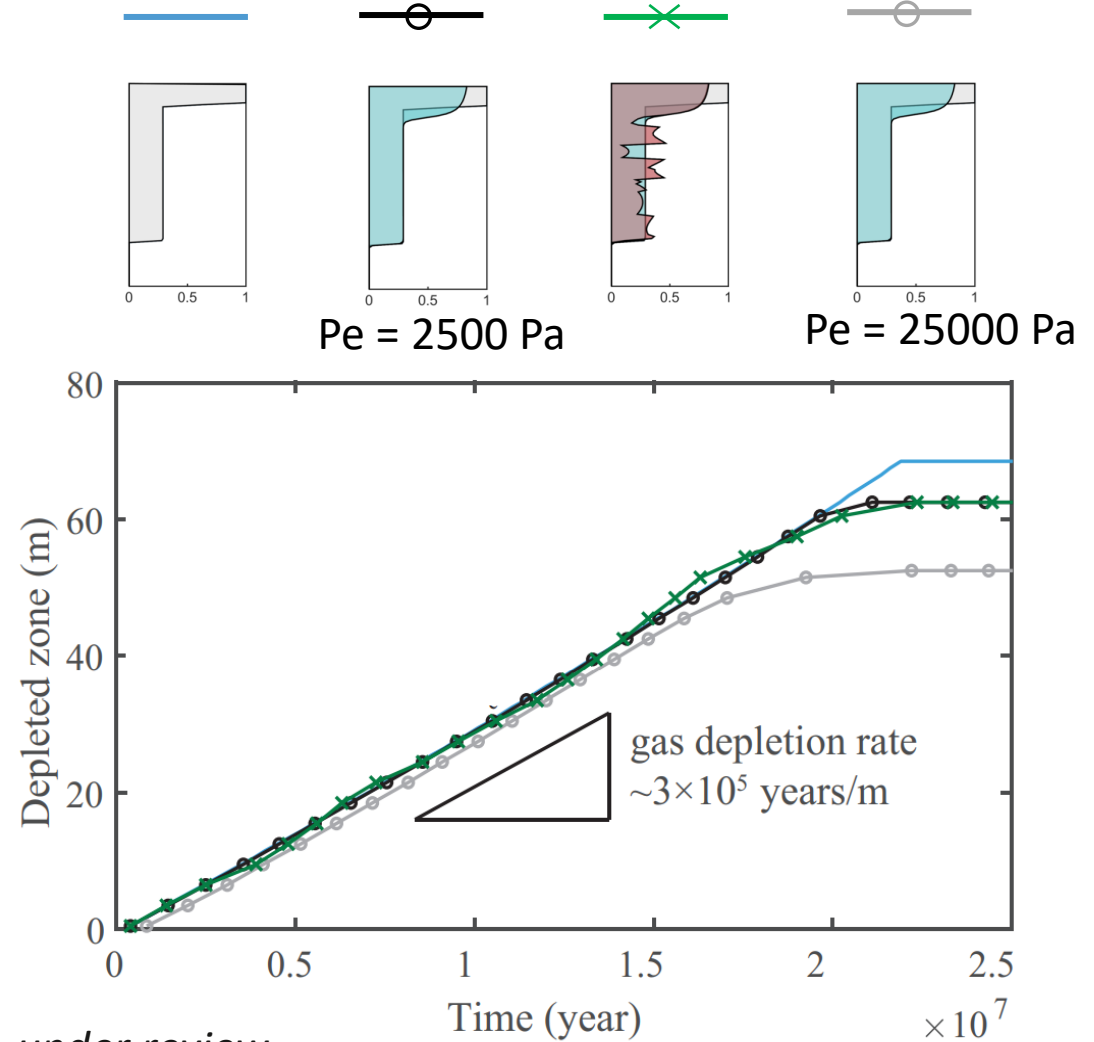
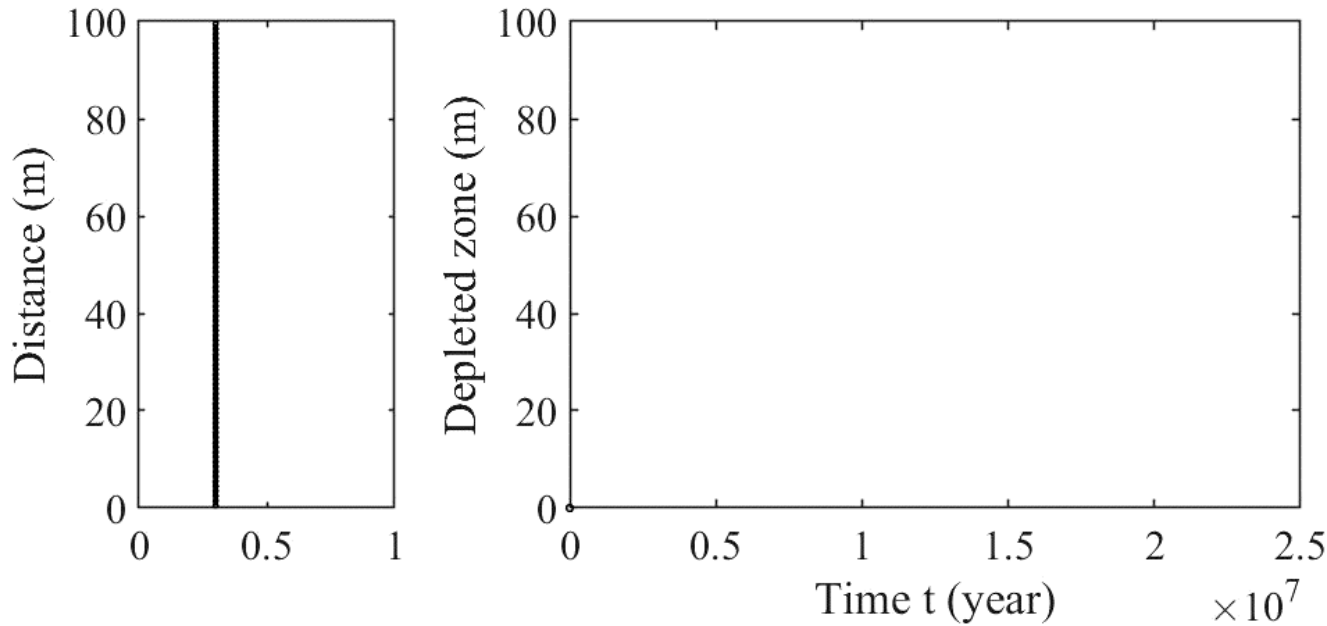
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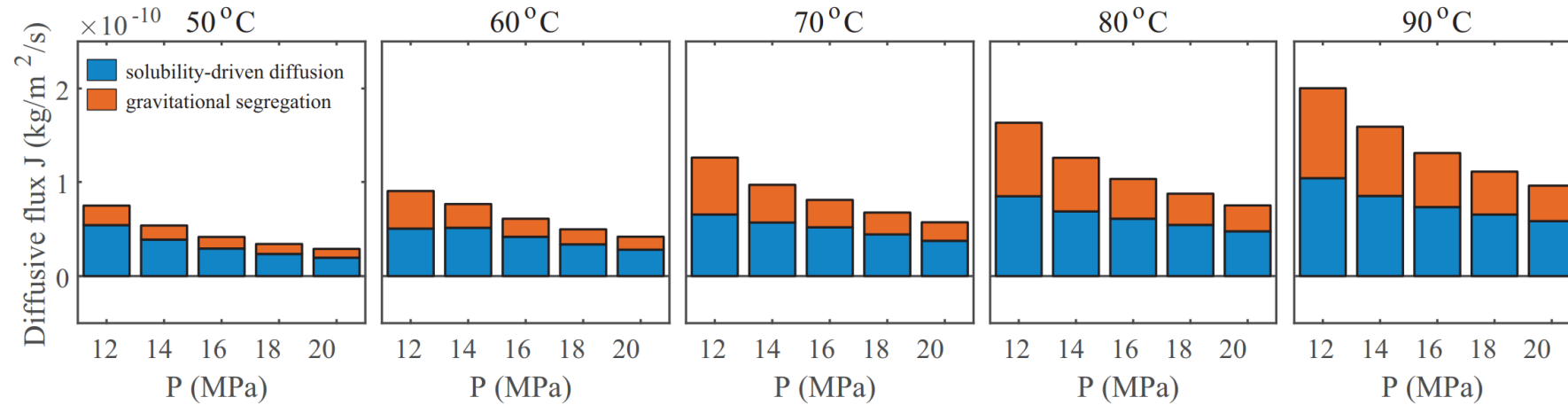
- We define “gas depletion rate” as the characteristic time
- The time scale of gas redistribution is at the order of 10^5 years per meter



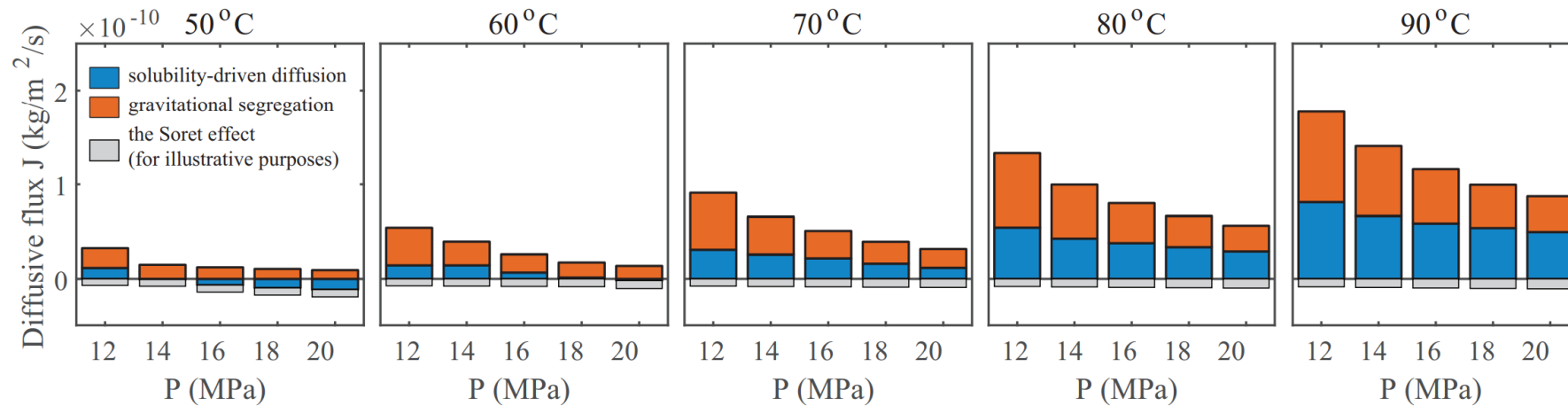
Li, Y. & Benson, S., Diffusion-induced gas redistribution in porous media, *under review*

The diffusive flux due to solubility gradient and gravity segregation at roughly the same order of magnitude

(a) isothermal



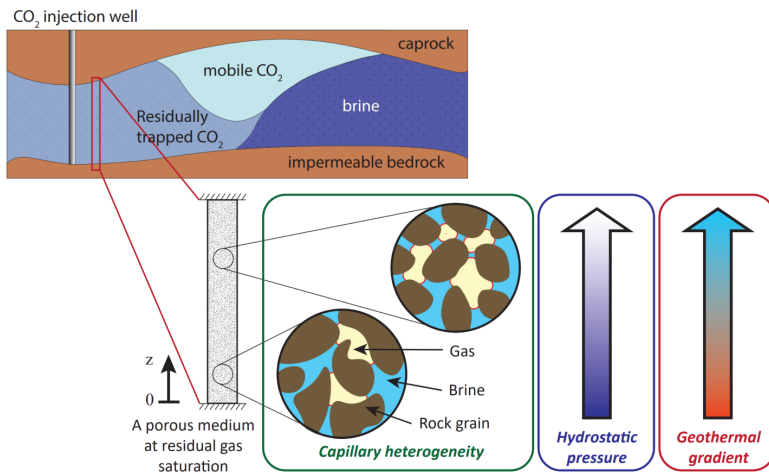
(b) non-isothermal (25 °C/km)



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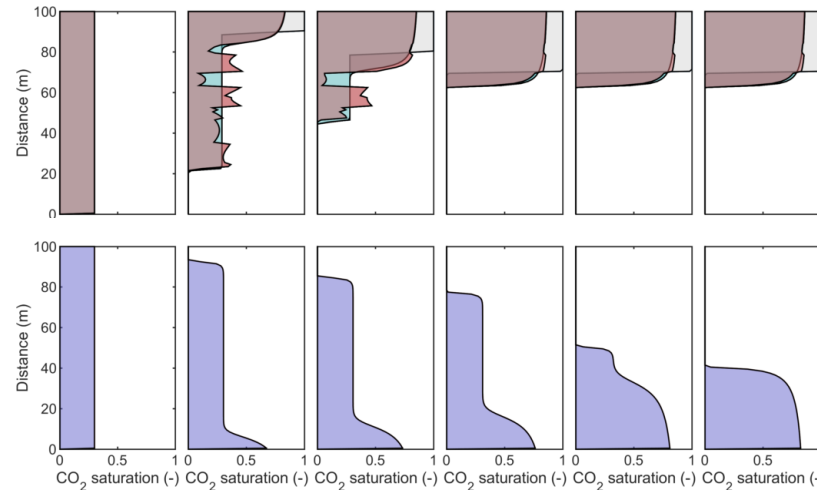
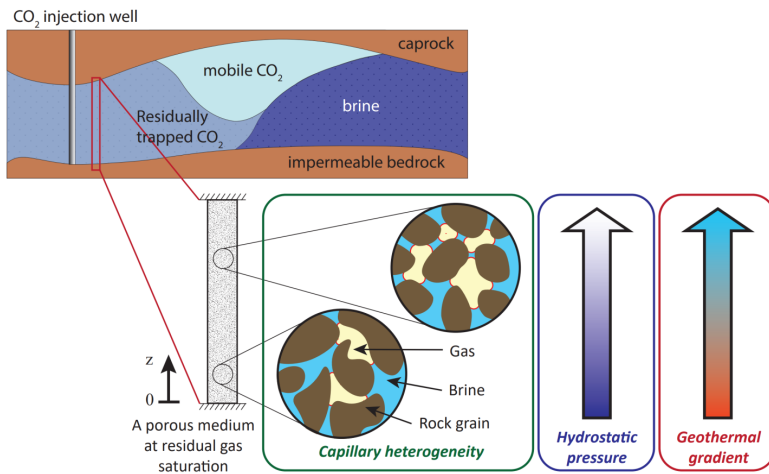
Implications and Conclusions

- Residual gas in porous media is thermodynamically unstable
 - hydrostatic gradients, geothermal gradients and capillary heterogeneity



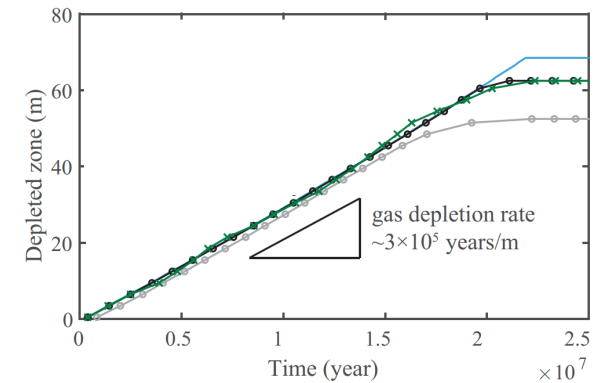
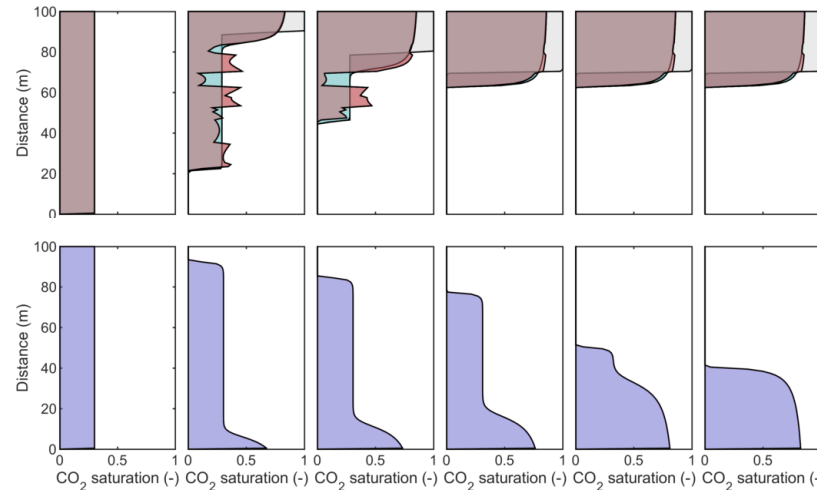
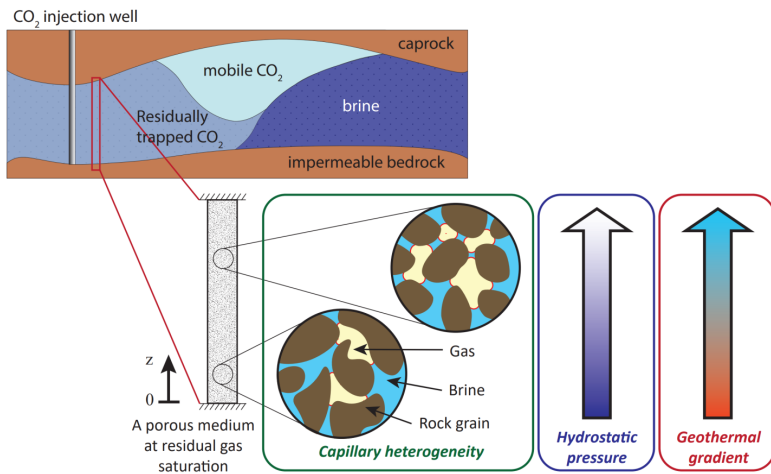
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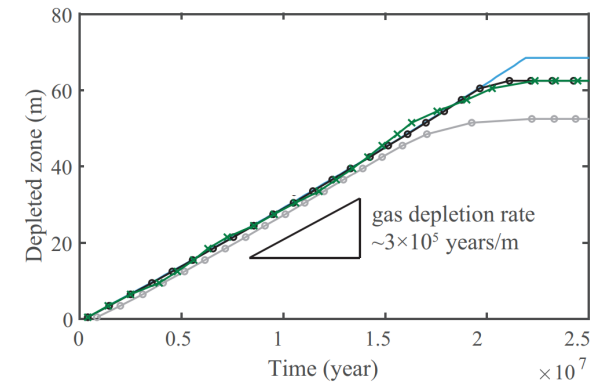
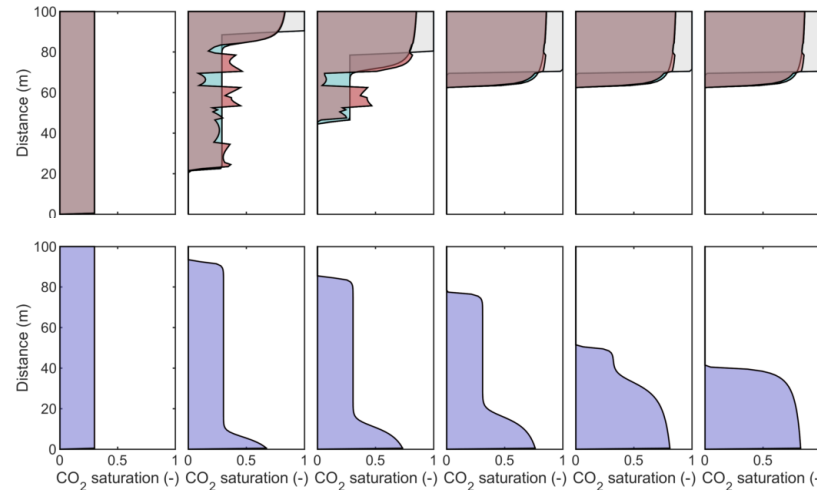
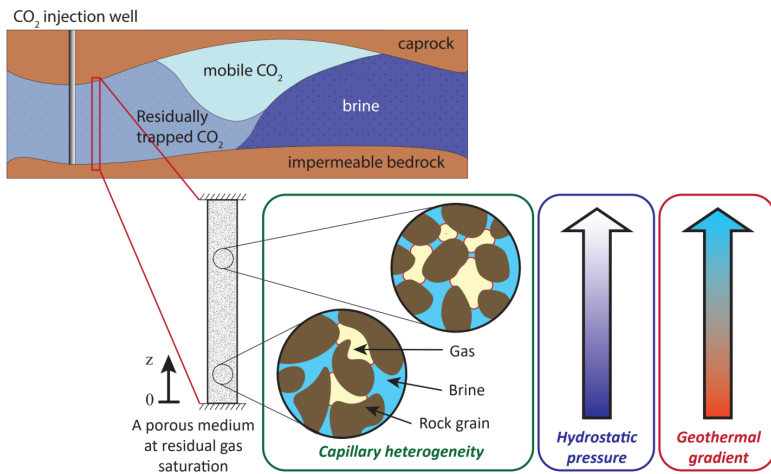
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- Timescales for gas redistribution (10^5 years/m) show that residual gas can remain stable over periods relevant for CO₂ storage. For example, it can take 20 million years for a 100-meter thick reservoir
- No mobilization of the gas phase is considered yet. Future work is to include mobilization.



Diffusion-induced gas redistribution in porous media

Yaxin Li and Prof. Sally M Benson

Scientific Achievement

We show that residually trapped CO₂ is thermodynamically unstable and thus can be redistributed by diffusion while remaining trapped.

Significance and Impact

We develop a comprehensive theory to evaluate the long-term stability of residually trapped CO₂, paving the path for systematically evaluating the long-term reliability of CO₂ entrapment in CCS projects.

Research Details

- Hydrostatic pressure, geothermal gradients and capillary heterogeneity can create energy gradient to cause residual CO₂ thermodynamically unstable
- Thermodynamic potential gradients can induce diffusion to redistribute residual gas across the system while the gas phase remains trapped
- Timescales for gas redistribution (10⁵ years/m) show that residual gas can remain stable over periods relevant for CO₂ storage

