

Referee #1

We thank the Reviewer for the careful review and comments, which will help us improve the manuscript. We are glad the Reviewer found the manuscript to be well-written, well-organized, and noted the topic is of high interest to the scientific community. In what follows, we respond in turn to each of the comments (included verbatim, in **bold**), attached also as a separate file.

- 1. I think the Introduction is a bit wordy and can be summarized in 4-5 concise paragraphs raising the main points.**

Following the Referee's comment, the introduction will be shortened while retaining important background information and necessary details for the presentation.

- 2. Considering the main model assumptions, how applicable/reliable is constant fluid density with temperature changes, in particular, for CO₂ as the working fluid? The same question will be raised for the assumption of having the same heat capacity for both confining rocks and the aquifer.**

We thank the Reviewer for noting these issues. The Lauwerier solution (Lauwerier, 1955) refers to a thin confined aquifer layer, in which no substantial vertical temperature gradients develop (lines 211-213). Consequently, under these conditions, no free convection or convection cells are expected to develop.

Regarding the potential effect of variable density on lateral flow, changes in density due to heating or cooling along the flow path can cause variations in flow rate. Specifically, fluid contraction during cooling can lead to a decrease in flow rate, while fluid expansion during heating can lead to an increase in flow rate along the path. However, in most cases, the overall temperature change does not exceed $\Delta T = 80$ °C, resulting in a low change in water density, typically around 2%. Consequently, this change does not have a substantial effect on flow and reactive transport (assuming there is no phase change). However, for the case of supercritical CO₂, the changes in density can be much larger, and in some cases the incompressibility assumption may not be appropriate.

Regarding the uniform heat capacities, the reactive Lauwerier solution, like the original Lauwerier solution, does not inherently assume uniform heat capacities. This assumption is made here for simplicity and to avoid cumbersome equations. By adopting alternative definitions of the parameters in Eqs. 10 and 19, non-uniform heat capacities of the confining layers and the aquifer can be considered in reactive Lauwerier solution (see Eq. 3.122 in Stauffer et al. (2014).

The above explanations, specifically noting the limitation of the assumption with respect to scenarios where CO₂ is the working fluid, will be integrated in the revised manuscript.

- 3. The authors have used a power-law relationship between the porosity and permeability of the aquifer to calculate the effective permeability. The authors have raised the limited predictive capability of these types of relations as well and mentioned that they solely used this type of relations to evaluate the general trends. Although this type of relationship could help evaluate the general trend of permeability evolution in mineral dissolution cases, they may not be the best choice for anti-correlated porosity-permeability changes in the systems when the percolating fluid has a weak capacity for dissolution or when we have mineral precipitation. Why did the authors not use other types of porosity-permeability relationships (e.g., a two-parameter exponential model) for sandstone aquifers where they expected to see mineral precipitation?**

We agree with the Reviewer that for specific cases and reactive flow duration (e.g., Garing et al., 2015) there may be more appropriate choices for porosity-permeability relations. However, we note this falls outside the scope of the current work. Here, we emphasize the different trends of aquifer permeability evolution due to dissolution in a carbonate aquifer and silica precipitation. Consequently, we selected a generic form of the porosity-permeability relation. The power-law relation demonstrates that permeability will typically rise within relatively short geological timescales in carbonate aquifers due to dissolution while substantial permeability changes due to silica precipitation occur on the order of tens of thousands of years. We argue, besides unique cases, the conclusions will remain valid regardless of the porosity-permeability relation used.

- 4. It seems that the last sentence in the Figure 1 caption is not complete. Please review the caption and correct it.**

Thank you for catching this oversight, the caption for Figure 1 will be corrected.

- 5. How reliable are the utilized values for the surface area of the carbonates and sandstones? Are they within the reported range for the surface area of these rocks in the literature?**

The values for the specific surface area of the sandstones utilized in the present work (i.e., $A_s = 10^4 \text{ m}^{-1}$) fall within the ranges reported in the literature for common sandstone formations (10^4 - 10^5 m^{-1} ; Hussaini & Dvorkin, 2021; Lai et al., 2015). However, it is important to note the ongoing research and discussions regarding the uncertainties in the estimation of reactive surface area in various rocks and under different reactive transport conditions (Noiriel & Daval, 2017; Recalcati et al., 2024; Seigneur et al., 2019). To address the Reviewer's comment, the range of values reported in the literature including additional references will be added in the revised manuscript.

For fractured carbonates, previous studies have demonstrated that in cases of large permeability contrast between fractures and the rock matrix, only the fracture surface area effectively participates in the reaction (i.e., constitutes the reactive surface area). This has been demonstrated by field case studies (e.g., MacQuarrie & Mayer, 2005; Pacheco & Alencão, 2006). In this case, the reactive surface area can be calculated using $A_s = 2 \cdot \kappa \cdot \text{RF}$ where κ is fracture density (defined as the number of fractures per unit volume), the factor of 2 accounts for the presence of two surfaces, and RF is the roughness factor (see Deng et al., 2018). Assuming $\kappa = 1/3^3 \text{ m}^{-3}$ and $\text{RF} = 1.35$, results in $A_s = 0.1 \text{ m}^{-1}$. Typical values of κ and fracture spacing can span a substantial range and may be higher or lower (Narr & Suppe, 1991; Scholz, 2019). Following the Reviewer's comment, this issue will be further clarified and references will be added to the revised manuscript.

References

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