



Multiple caprock layers offer confidence in permanent geologic CO₂ storage at the gigatonne scale

Iman R. Kivi^{1,2,3}, Roman Y. Makhnenko⁴, Curtis M. Oldenburg⁵, Jonny Rutqvist⁵, and Victor Vilarrasa³

¹Institute of Environmental Assessment and Water Research, Spanish National Research Council (IDAEA-CSIC), Barcelona, Spain

²Associated Unit: Hydrogeology Group (UPC-CSIC), Barcelona, Spain

³Global Change Research Group (GCRG), IMEDEA, CSIC-UIB, Esporles, Spain

⁴Department of Civil & Environmental Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois, USA

⁵Energy Geosciences Division, Lawrence Berkeley National Laboratory, CA, USA

Widespread deployment of Geologic Carbon Storage (GCS) at the gigatonne scale is projected to play a vital role in reaching carbon neutrality and mitigating the climate crisis. One major concern with GCS scale-up is the ability of geologic formations to retain CO₂ deep underground, at least over several thousand years. Of particular importance to this issue is the sealing capacity of the ideally low-permeability, high-gas-entry pressure caprock(s). Existing simulation studies to address the long-term fate of the injected CO₂ could barely exceed multi-century time scales due to high computational costs. This work aims to provide an improved understanding of the extent to which the potentially leaked CO₂ from basin-wide GCS may rise through a multi-layered system of laterally uniform aquifers and shale caprocks over geological time scales (million years). To this end, we develop a one-dimensional CO₂ flow and transport model, which is arguably capable of capturing the dynamics of basin-scale upward CO₂ migration. We consider two sets of caprock properties: (1) low intrinsic permeability (10^{-20} m²) and high capillary entry pressure (2.5 MPa), obtained from laboratory measurements on intact clay-rich shales, and (2) high permeability (10^{-16} m²) and low entry pressure (0.1 MPa), representative of pervasively fractured shales at regional scales. On the one hand, we find that the free-phase CO₂ can hardly penetrate more than a few centimeters into the intact caprock directly overlying the storage reservoir. CO₂ leakage in this scenario is exclusively governed by molecular diffusion with an estimated migration rate of 1 meter over thousands of years. On the other hand, the high permeability and low entry pressure of fractured caprocks enable CO₂ to break through the whole primary caprock during the injection and through the secondary one(s) in the post-injection period. However, following the gradual CO₂ pressure decline, brine imbibition back into caprocks suppresses CO₂ leakage and the percolating path is cut by an overlying caprock. Once the pore fluid of upper aquifers becomes CO₂-saturated, secondary CO₂ accumulations form and may host a significant portion of the injected CO₂. The extreme leakage scenario, which allows for further CO₂ rise of nearly one hundred meters becomes eventually diffusion-dominated and hence relatively safe. Our model results suggest that the presence of multiple shaly caprock layers, even if pervasively fractured, provides secure CO₂

containment in the subsurface over millions of years.

Reference

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