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### Heterogeneity-assisted carbon dioxide storage in marine sediments

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#### HIGHLIGHTS

• CO<sub>2</sub> storage in heterogeneous marine sediments in investigated for the first time.

• CO2 storage is possible in shallower marine sediments than previously found.

• Physical and operational thresholds for secure CO<sub>2</sub> storage are presented.

#### ARTICLE INFO

Keywords: CO<sub>2</sub> storage Marine sediment Heterogeneity-assisted trapping

#### ABSTRACT

Global climate change is a pressing problem caused by the accumulation of anthropogenic greenhouse gas emissions in the atmosphere. Carbon dioxide  $(CO_2)$  capture and storage is a promising component of a portfolio of options to stabilize atmospheric CO<sub>2</sub> concentrations. Meaningful capture and storage requires the permanent isolation of enormous amounts of CO2 away from the atmosphere. We investigate the effectiveness of heterogeneity-induced trapping mechanism, in potential synergy with a self-sealing gravitational trapping mechanism, for secure CO<sub>2</sub> storage in marine sediments. We conduct the first comprehensive study on heterogeneous marine sediments with various thicknesses at various ocean depths. Prior studies of gravitational trapping have assumed homogeneous (deep-sea) sediments, but numerous studies suggest reservoir heterogeneity may enhance CO<sub>2</sub> trapping. Heterogeneity can deter the upward migration of CO2 and prevent leakage through the seafloor into the seawater. Using geostatistically-based Monte Carlo simulations of CO2 transport in heterogeneous sediment, we show that strong spatial variability in permeability is a dominant physical mechanism for secure CO<sub>2</sub> storage in marine sediments below 1.2 km water depth (less than half of the depth needed for the gravitational trapping). We identify thresholds for sediment thickness, mean permeability and porosity, and their relationships to meaningful injection rates. Our results for the U.S. Gulf of Mexico suggest that heterogeneity-assisted trapping has a greater areal extent with more than three times the  $CO_2$  storage capacity for secure offshore  $CO_2$  storage than with gravitational trapping. These characteristics offer CO<sub>2</sub> storage opportunities that are closer to coasts, more accessible, and likely to be less costly.

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#### 1. Introduction

Carbon dioxide (CO<sub>2</sub>) capture and storage (CCS) can help stabilizing greenhouse gas concentrations in the atmosphere and mitigate global climate change [1–7]. Multiple approaches have been proposed for the long-term storage of anthropogenic CO<sub>2</sub> emissions, including injection into deep geologic formations (e.g., depleted oil and gas reservoirs, coalbeds, saline formations, shale [8–16]), deep ocean waters [17,18], and storage via chemical transformations [19,20]. However, each of these options has drawbacks. In onshore geologic formations, CO<sub>2</sub> is buoyant and can leak from the reservoir through an overlying impervious formation [21,22], and overpressure due to CO<sub>2</sub> injection may fracture the overlying caprock or induce seismicity [23,24].

Deep ocean waters may be acidified by CO<sub>2</sub>, with concomitant impacts on marine ecosystems, and the CO<sub>2</sub> is prone to mixing with ocean currents and may be ultimately released to the atmosphere [25]. Further, industrial demand for chemical transformations is ~200 MtCO<sub>2</sub> per year, which is roughly 175 times less than the amount of CO<sub>2</sub> that was emitted from energy use worldwide in 2013 [22]. Offshore geologic formations could also be used for CO<sub>2</sub> storage, which may be beneficial because they are not beneath onshore populations, cannot negatively affect underground sources of drinking water [26], and regulatory considerations and ownership issues may be more easily reconciled than for onshore locations [27]. In fact, several offshore CO2 storage projects are underway, including CO2ReMoVe [28], SUCCESS [29], QICS [30], and NLECI [31], and NCIP [32]. Most of these projects are implemented in aquifers under shallow seas and, as in onshore geologic formations, there are hazards that arise from overpressure and the buoyancy of emplaced CO<sub>2</sub>, such as, CO<sub>2</sub> interaction with seawater.

We revisit in detail a more promising option for  $CO_2$  storage: emplacement in deep marine sediments [33–35].  $CO_2$  storage in deep marine sediments has the advantages of geologic storage and deep ocean storage [33]. For example,  $CO_2$  storage in marine sediments can benefit from continuous subsea pressure management and potential chemical transformations through the formation of  $CO_2$  hydrates [34]. While the costs are likely to be higher,  $CO_2$  storage in marine sediments could be more tenable than other options for  $CO_2$  storage, especially when risks are considered. We note that the cost evaluation and cost comparison to onshore  $CO_2$  storage are outside the scope of our work.

At sufficient ocean depths, the pressure is high enough and the temperature is low enough for liquid CO<sub>2</sub> (CO<sub>2(l)</sub>) to be more dense than the surrounding less compressible seawater [36]. If CO<sub>2</sub> was injected above the seafloor, it would sink and form a lake of gravitationally stable  $CO_{2(l)}$  on the seafloor. But if CO<sub>2</sub> was injected into the sediments below the seafloor, the emplaced  $CO_{2(l)}$  would descend deeper into the sediment. As it descends, the increase in temperature due to the geothermal gradient would cause the density of the CO<sub>2(l)</sub> to pronouncedly

decrease. At the neutral buoyancy depth (NBD) into the sediments, the  $CO_{2(l)}$  would stop descending because its density becomes equal to the density of the surrounding pore fluid, and thus the  $CO_{2(l)}$  isolates itself within the sediment pores. Gravitational trapping occurs in the "negative buoyancy zone" [33] in marine sediments, located between the seafloor and the NBD, where the emplaced  $CO_2$  is more dense than the pore fluid. This gravitational trapping exists at water depths  $>\sim 2.5$  km and sediments within several hundred meters beneath the seafloor [37]. Also, if  $CO_2$  were injected beneath the NBD, it would ascend until it reaches the NBD, where it would then be gravitationally trapped.

Despite the advantages of  $CO_2$  storage in marine sediments, the suitability of this option is poorly understood, and thus the global storage capacity, while potentially enormous, is highly uncertain [35,38]. Prior studies of  $CO_2$  storage via gravitational trapping have assumed *homogeneous* sediments [33,35], but experimental, numerical, and field studies suggest that reservoir permeability heterogeneity may enhance  $CO_2$  trapping by hindering the upward migration of buoyant  $CO_2$  [39]. As such, models that incorporate permeability heterogeneity more accurately account for  $CO_2$  fate and transport in the reservoir [40,41].

In this work, we perform reservoir simulations of CO<sub>2</sub> injection and its interaction with sediment pore water, based on fluid and sediment data from four sites in the U.S. Gulf of Mexico (GOM), shown in Fig. 1(a). The marine sediments at these sites are sufficiently thick and permeable for CO<sub>2</sub> storage. The sediment porosity and permeability have variable degrees of heterogeneity and anisotropy, which reflect the original and post-depositional processes [42-44]. We incorporate these heterogeneities and anisotropies at the reservoir level into our models, where we explicitly model solubility and gravitational trapping processes. Given the various temperatures and pressures in the marine sediments, and the uncertainty in fluid flow parameters, we develop and implement a statistical framework to quantify the uncertainty in CO<sub>2</sub> storage capacity and leakage into the seawater during the heterogeneity-assisted trapping of CO<sub>2</sub> in marine sediments. To best of our knowledge, this work is the first numerical simulation study on CO<sub>2</sub> sequestration in heterogeneous marine sediments.

We find a large  $CO_2$  storage potential due to heterogeneity-induced trapping, which is also effective in combination with gravitational trapping for physico-gravitational trapping (PGT). Marine sediment reservoirs must also accommodate sufficient  $CO_2$  injection rates, which under homogeneous permeability conditions also requires that the sediments be thick and permeable, but such characters are uncommon in most deep-water settings [34,35]. We further show that these constraints can be relaxed through heterogeneity-induced trapping, such that marine sediments in much shallower seas can be viable  $CO_2$  storage options.

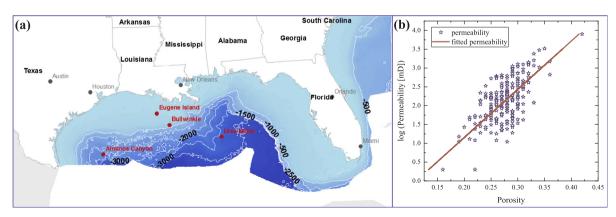


Fig. 1. Data from the Gulf of Mexico (GOM) within the U.S. Exclusive Economic Zone. (a) Site locations in the Gulf of Mexico (GOM), (b) Permeability-porosity correlations.

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