

# Dissolved CO<sub>2</sub> injection to eliminate the risk of CO<sub>2</sub> leakage in geologic carbon storage

Victor Vilarrasa<sup>1</sup>, Maria Pool<sup>2</sup>, Silvia De Simone<sup>2,3</sup> and Jesus Carrera<sup>2</sup>

**Abstract.** Geologic carbon storage is usually viewed as injecting, or rather as storing, CO<sub>2</sub> in supercritical phase. This view is very demanding on the caprock, which must display: (1) high entry pressure to prevent an upward escape of CO<sub>2</sub> due to density effects; (2) low permeability to minimize the upwards displacement of the brine induced by the injected CO<sub>2</sub>; and (3) high strength to ensure that the fluid pressure buildup does not lead to caprock failure. We analyze the possibility of injecting dissolved CO<sub>2</sub> and, possibly, other soluble gases for cases when the above requirements are not met. The approach consists of extracting saline water from one portion of the aquifer, reinjecting it in another portion of the aquifer and dissolving CO<sub>2</sub> downhole. Mixing at depth reduces the pressure required for brine and CO<sub>2</sub> injection at the surface. We find that dissolved CO<sub>2</sub> injection is feasible and eliminates the risk of CO<sub>2</sub> leakage because brine with dissolved CO<sub>2</sub> is denser than brine without dissolved CO<sub>2</sub> and thus, it sinks towards the bottom of the saline aquifer.

**Keywords:** CO<sub>2</sub> storage, dissolved CO<sub>2</sub>, leakage, caprock, buoyancy.

## 1.1 Introduction

Geologic carbon storage is usually considered as injecting CO<sub>2</sub> in free phase under supercritical or liquid (cold) conditions, which will eventually become supercritical

---

<sup>1</sup> V. Vilarrasa (✉)

Institute of Environmental Assessment and Water Research, Spanish National Research Council (IDAEA-CSIC), Jordi Girona 18-26, 08034 Barcelona, Spain  
e-mail: [victor.vilarrasa@idaea.csic.es](mailto:victor.vilarrasa@idaea.csic.es)

Associated Unit: Hydrogeology Group (UPC-CSIC), Jordi Girona 1-3, 08034 Barcelona, Spain

<sup>2</sup> M. Pool, S. De Simone, J. Carrera

Institute of Environmental Assessment and Water Research, Spanish National Research Council (IDAEA-CSIC), Jordi Girona 18-26, 08034 Barcelona, Spain

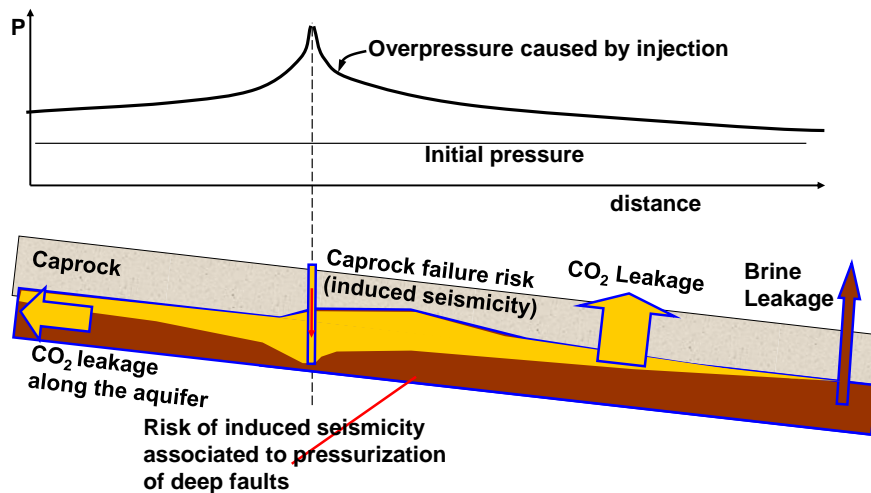
Associated Unit: Hydrogeology Group (UPC-CSIC), Jordi Girona 1-3, 08034 Barcelona, Spain

<sup>3</sup> S. De Simone

Dept. of Earth Science and Engineering, South Kensington Campus, Imperial College, London, UK

under pressure and temperature reservoir conditions (Vilarrasa et al., 2013). This injection scheme leads to a CO<sub>2</sub>-rich phase which floats on resident brine. Thus, CO<sub>2</sub> remains in the upper portion of the saline formation, which may lead to leakage across the caprock. While the concept is robust and simple, its application is hindered by two sets of difficulties (Fig. 1.1):

- 1) Large volumes of CO<sub>2</sub>, in the order of millions of tons per year, will have to be injected for industrial scale geologic carbon storage projects. Such injections will displace the resident brine, possibly polluting shallow drinking water aquifers (Birkholzer and Zhou, 2009; Birkholzer et al., 2012). Additionally, a large pressure buildup will be generated, which may reactivate fractures or faults, potentially inducing felt earthquakes (Zoback and Gorelick, 2012; Vilarrasa and Carrera, 2015),
- 2) Supercritical CO<sub>2</sub> is lighter than the resident brine, so it may leak upwards along sloping aquifers, across the caprock, through faults or wells (Lindeberg, 1997; Pruess, 2008; Nordbotten et al., 2009; Humez et al., 2011).

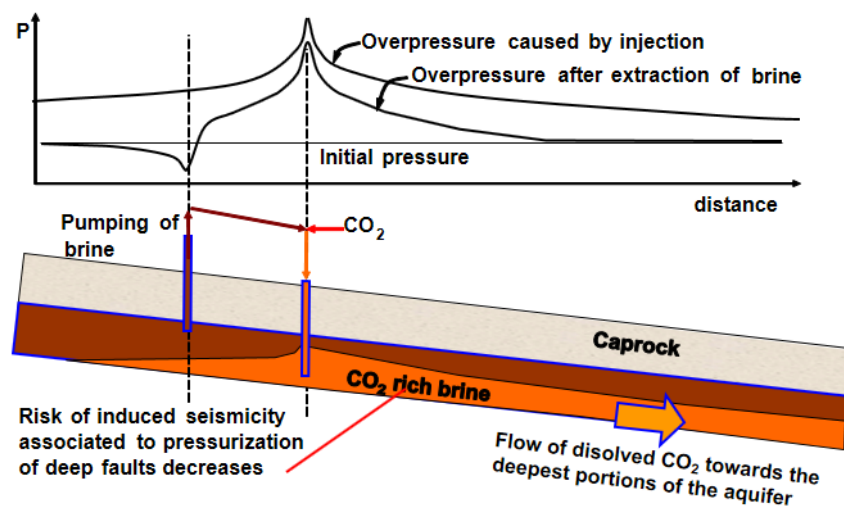


**Fig. 1.1** Schematic representation of supercritical CO<sub>2</sub> storage and some of the difficulties it must overcome. Note that overpressure (above) can be large and extend over a large area, thus risking caprock sealing capacity and the occurrence of felt induced seismicity

These difficulties have motivated approaches for fluid pressure control. The main idea behind these approaches consists in pumping native brine from the storage formation where CO<sub>2</sub> will be injected. The pumped brine can be disposed of (e.g., in sites close to the sea), or desalinated (e.g., in water scarce regions) and the residual brine reinjected into a saline formation (Court et al., 2011; Bergmo et al., 2011). This concept includes approaches such as the “Active CO<sub>2</sub> Reservoir Management” (Buscheck et al., 2011, 2012) or the “Passive injection” strategy (Dempsey et al., 2014). These approaches increase CO<sub>2</sub> storage capacity and reduce the

induced pressure buildup and associated risks. Nonetheless, the risk of leakage persists with these two storage strategies because the injected CO<sub>2</sub> is still buoyant and thus, stays at the top of the storage formation.

To overcome this issue, CO<sub>2</sub> can be injected dissolved into the brine. Dissolved CO<sub>2</sub> injection can be regarded as a special case of pressure control. By injecting CO<sub>2</sub> dissolved in the pumped brine, the risk of buoyant escape of CO<sub>2</sub> is eliminated. This is because the density of CO<sub>2</sub>-rich brine is higher than that of resident brine and thus, the injected CO<sub>2</sub>-rich brine will tend to sink towards the bottom of the storage formation (Fig. 1.2).



**Fig. 1.2** Schematic representation of the dissolved CO<sub>2</sub> concept. Brine is pumped from the storage formation and reinjected after dissolving CO<sub>2</sub> in it. Note that both maximum pressure buildup and the area affected by overpressure are significantly reduced. Note also that the concept can be extended to other flue gases (Carrera et al, 2011)

It may be argued that CO<sub>2</sub> will dissolve naturally in the aquifer without the need of engineered actions. However, dissolving the whole CO<sub>2</sub> plume may require centuries. Actually, dissolution rates are slow for relatively low permeability media and only becomes quite fast in porous media with a high vertical permeability once convection develops under the CO<sub>2</sub> plume (Riaz et al., 2006, Hidalgo and Carrera, 2009, Pau et al., 2010; MacMinn et al., 2012; Elenius and Johannsen, 2012). Even though CO<sub>2</sub> dissolution can be accelerated by fluctuating the injection rate (Bolster et al., 2009) and/or by alternating CO<sub>2</sub> and brine injection either in the well or at some distance (Leonenko and Keith, 2008, Hassanzadeh et al., 2008; Zhang and Agarwal, 2012), some of the injected CO<sub>2</sub> will continue in free-phase.

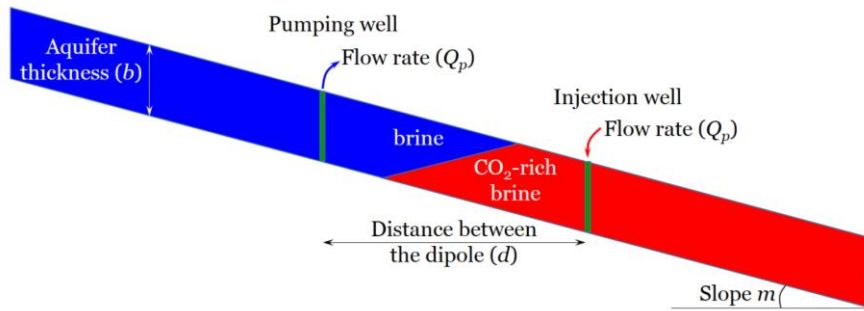
The goal of this paper is to present the dynamics of a dipole system in which brine is pumped from one well and brine with dissolved CO<sub>2</sub> is injected in the other well. We aim to assess the feasibility of dissolved CO<sub>2</sub> injection as an effective

storage strategy. We also investigate the effects of this storage approach on the rock geomechanical stability.

## 1.2 Dynamics of the Dipole System

The dynamics of a dipole system of dissolved CO<sub>2</sub> injection and brine pumping is non-trivial. The injected CO<sub>2</sub>-rich brine is denser than the resident brine and, as a result, it tends to sink. However, this sinking tendency of CO<sub>2</sub>-rich brine is counter-balanced by the high pressure at the injection well and by the drag generated by the pumping well. Consequently, part of the injected CO<sub>2</sub> may be pumped and returned to the surface, which reduces the efficiency of the system. The critical pumping rate,  $Q_c$  [L<sup>3</sup> T<sup>-1</sup>], can be defined as the maximum flow rate that can be pumped with negligible CO<sub>2</sub> concentration (0.1% of saturation) (Pool et al., 2013).

To understand the system dynamics and thus, being able to optimize its design, we consider a homogeneous sloping aquifer of constant thickness  $b$  [L] and slope  $m$  [-] (Fig. 1.3). The dipole system consists of an injection-pumping well pair. A fully penetrating pumping well pumps brine from the storage formation at a constant flow rate,  $Q_p$  [L<sup>3</sup> T<sup>-1</sup>]. The pumped brine, CO<sub>2</sub> and, possibly other gases, are injected separately at the injection well, which is located at a distance  $d$  [L] downslope, and mixed downhole. The resulting CO<sub>2</sub>-saturated brine is denser than the native brine, leading to a safe storage in which the CO<sub>2</sub>-saturated brine will sink to the bottom of the storage formation.



**Fig. 1.3** Model set-up for the analysis of dissolved CO<sub>2</sub> injection in a dipole system in which brine is pumped and reinjected with CO<sub>2</sub> in the same saline formation through vertical wells in a sloping homogeneous aquifer

To illustrate the dynamics of the system, we show the case of a confined saline aquifer, with 20% slope, at a depth ranging from 1000 to 1800 m. The aquifer is assumed homogeneous and isotropic. The extent of the model is 4000 x 1000 x 50 m<sup>3</sup> and the distance between the fully penetrating injection-pumping well pair is

500 m. Water viscosity,  $\mu$  [ $\text{M L}^{-1} \text{T}^{-1}$ ], is assumed to be constant and equal to 0.4 mPa·s. The buoyancy factor is taken as  $\varepsilon=0.027$  ( $\varepsilon=(\rho_s-\rho_b)/\rho_b$ , where  $\rho_s$  [ $\text{M L}^{-3}$ ] is the density of CO<sub>2</sub> saturated brine and  $\rho_b$  [ $\text{M L}^{-3}$ ] is the density of resident brine).

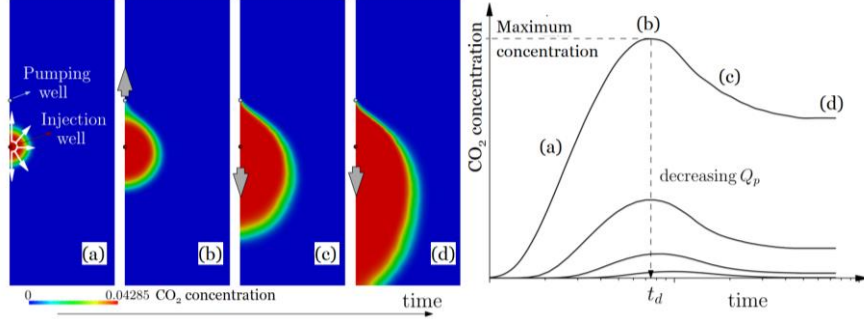
Simulation results show that the movement of the injected CO<sub>2</sub>-saturated brine plume is governed by two factors: (1) viscous forces induced by the pumping-injecting wells which produce a dipole flow field, and (2) buoyancy forces caused by the difference between the density of the injected CO<sub>2</sub>-saturated brine and that of the resident brine. Buoyancy,  $f_b$  [ $\text{M L}^{-2} \text{T}^{-2}$ ], is quantified by (Pool et al., 2013)

$$f_b = V' \Delta \rho m g \quad (1.1)$$

where  $V'$  [ $\text{L}^3 \text{L}^{-3}$ ] is the dimensionless injected volume, which is given by  $V' = Q_p t / (\pi d^2 b)$ ,  $t$  [T] is time,  $g$  is gravity [ $\text{L T}^{-2}$ ] and  $\Delta \rho = \rho_s - \rho_b$  [ $\text{M L}^{-3}$ ]. Viscous forces,  $f_v$  [ $\text{M L}^{-2} \text{T}^{-2}$ ], are given by (Pool et al., 2013)

$$f_v = \frac{Q_p \mu}{2 \pi d b k} \quad (1.2)$$

where  $k$  [ $\text{L}^2$ ] is the intrinsic permeability. While buoyancy forces grow with the plume size because they depend on the injected volume  $V'$ , viscous forces remain constant over time. This difference explains the time dependent role of density. During the initial stage (Fig. 1.4a), viscous forces mainly control flow and transport and thus, CO<sub>2</sub>-saturated brine migrates radially from the injection well and towards the pumping well. Similar effects would be obtained assuming constant density. Once the CO<sub>2</sub>-saturated brine plume reaches the pumping well, CO<sub>2</sub> concentration in the pumping well rises gradually until an equilibrium between upslope forces caused by the pumping well and downslope forces driven by buoyancy effects is attained (Fig. 1.4b). At this instant of time ( $t_d$  [T]), the CO<sub>2</sub> mass fraction at the pumping well reaches its maximum. Afterwards, density-driven flow leads to a growth of the plume downslope along the bottom of the aquifer because buoyancy forces increase and start to dominate over viscous forces. As a result, the extent and weight of the plume rise, pulling the plume downslope. This pull retreats the CO<sub>2</sub>-saturated brine plume towards the injection well, which leads to a decrease in the CO<sub>2</sub> mass fraction at the pumping well (Fig. 1.4c). Finally, steady state is reached (Fig. 1.4d). It should be highlighted that the first arrival decreases with the pumping rate, but the peak time remains constant, which is surprising and requires further analysis.



**Fig. 1.4** The four stages (a-d) of dissolved CO<sub>2</sub> injection in a sloping aquifer: left, plan view of vertically averaged concentration; right, time evolution of CO<sub>2</sub> concentration at the pumping well (modified from Pool et al., 2013)

The first arrival time of the CO<sub>2</sub>, which is controlled by the dipole nature of the flow field, can be computed by integrating the travel time along the flow line between the two wells, giving (Grove and Beetem, 1971)

$$t_0 = \frac{\pi\phi bd^2}{3Q_p} \quad (1.3)$$

where  $\phi$  [L<sup>3</sup> L<sup>-3</sup>] is porosity. This implies that the first arrival depends on the distance between the two wells, the aquifer thickness and the pumping rate. However, it is independent of the aquifer permeability. On the other hand, the time for which the CO<sub>2</sub> mass fraction at the pumping well peaks ( $t_d$ ) is controlled by the balance between buoyancy and viscous forces. Since both are proportional to  $Q_p$  and inversely proportional to  $b$ ,  $t_d$  becomes independent of both the flow rate and aquifer thickness. But  $t_d$  is affected by the aquifer slope and the aquifer permeability. On the one hand, an increase in the aquifer slope causes  $f_b$  to increase and thus, the time to balance viscous forces decreases. On the other hand, a decrease in permeability leads to an increase in  $f_v$ , which increases the time to reach equilibrium.

The response of the system is also affected by the well characteristics, i.e., vertical or horizontal, because pressure buildup evolution differs when injecting through horizontal and vertical wells. CO<sub>2</sub> injection induces a smaller overpressure when injecting through vertical than horizontal wells (Zhang and Agarwal, 2012; Vilarrasa, 2014). In contrast, it is the other way around for water injection (Zhang and Agarwal, 2012). For CO<sub>2</sub> injection through a vertical well, pressure buildup peaks at the beginning of injection, but subsequently drops (Vilarrasa et al., 2016). However, pressure builds up continuously when injecting CO<sub>2</sub> through a horizontal well, which induces a higher pressure buildup in the long term (Zhang and Agarwal, 2012). In contrast, water injection leads to higher pressure buildup when injecting through a vertical than a horizontal well. As a result, caprock and fault stability may be compromised. Even though caprock integrity is not a concern when injecting CO<sub>2</sub> dissolved in brine because it sinks, fault stability may be an issue because of

potential felt induced seismicity (De Simone et al., 2017). Thus, horizontal wells are preferable for injection of CO<sub>2</sub>-rich brine.

### 1.3 Conclusions

We have shown that dissolved CO<sub>2</sub> injection is feasible to store CO<sub>2</sub> in deep saline formations. This CO<sub>2</sub> storage concept displays a number of advantages over conventional supercritical CO<sub>2</sub> storage. First, it is easy to control and generates little overpressure, which reduces (virtually eliminates) induced seismicity risk. Second, since CO<sub>2</sub>-rich brine is denser than native brine, CO<sub>2</sub> tends to sink towards the deepest portions of the storage formation, reducing (virtually eliminating) the risk of CO<sub>2</sub> leakage. Last but not least, for the same reason, there is no need for a caprock.

### 1.4 Acknowledgments

The authors acknowledge financial support from the "TRUST" project (European Community's Seventh Framework Programme FP7/2007-2013 under grant agreement n 309607) and from "FracRisk" project (European Community's Horizon 2020 Framework Programme H2020-EU.3.3.2.3 under grant agreement n 636811).

### 1.5 References

- Bergmo P, Grimstad A, Lindeberg E (2011) Simultaneous CO<sub>2</sub> injection and water production to optimise aquifer storage capacity. *International Journal of Greenhouse Gas Control* 5(3):555–564
- Birkholzer JT, Zhou Q (2009) Basin-scale hydrogeologic impacts of CO<sub>2</sub> storage: Capacity and regulatory implications. *International Journal of Greenhouse Gas Control* 3:745–56
- Birkholzer J, Cihan A, Zhou Q (2012) Impact-driven pressure management via targeted brine extraction – conceptual studies of CO<sub>2</sub> storage in saline formations. *International Journal of Greenhouse Gas Control* 7:168–80
- Bolster D, Dentz M, Carrera J (2009) Effective two phase flow in heterogeneous media under temporal pressure fluctuations. *Water Resources Research* 45, doi:1029/2008WR007460
- Buscheck TA, Sun Y, Hao Y, Wolery TJ, Bourcier W, Tompson AF, et al. (2011) Combining brine extraction, desalination, and residual-brine reinjection with CO<sub>2</sub> storage in saline formations: Implications for pressure management, capacity, and risk mitigation. *Energy Procedia* 4(0):4283–90
- Buscheck A, Sun Y, Chen M, Hao Y, Wolery TJ, Bourcier W, et al. (2012) Active CO<sub>2</sub> reservoir management for carbon storage: Analysis of operational strategies to relieve pressure buildup and improve injectivity. *International Journal of Greenhouse Gas Control* 6:230–45
- Carrera J, Silva O, Ayora C (2011) Method and system for the storage of soluble gases in permeable geological formations. European patent application; N EP11382321.5

- Court B, Celia MA, Nordbotten JN, Elliot TR (2011) Active and integrated management of water resources throughout CO<sub>2</sub> capture and sequestration operations. 10<sup>th</sup> International Conference on Greenhouse Gas Control Technologies GHGT-10, Energy Procedia 4:4221–4229
- Dempsey D, Kelkar S, Pawar R (2014) Passive injection: a strategy for mitigating reservoir pressurization, induced seismicity and brine migration in geologic CO<sub>2</sub> storage. *International Journal of Greenhouse Gas Control* 28:96-113
- De Simone S, Carrera J, Vilarrasa V (2017) Superposition approach to understand triggering mechanisms of post-injection induced seismicity. *Geothermics* 70:85-97
- Elenius MT, Johannsen K (2012) On the time scales of nonlinear instability in miscible displacement porous media flow. *Computational Geosciences* 16(4):901-911
- Grove DB, Beetem WA (1971) Porosity and dispersion constant calculations for a fractured carbonate aquifer using 2 well tracer method. *Water Resources Research* 7(1):128–34
- Hassanzadeh H, Pooladi-Darvish M, Keith D (2008) Accelerating CO<sub>2</sub> dissolution in saline aquifers for geological storage - mechanistic and sensitivity studies. *Energy & Fuels* 23:3328–36
- Hidalgo J, Carrera J (2009) Effect of dispersion on the onset of convection during CO<sub>2</sub> sequestration. *Journal of Fluid Mechanics* 640:443–54
- Humez P, Audigane P, Lions J, Chiaberge C, Bellenfant G (2011). Modeling of CO<sub>2</sub> leakage up through an abandoned well from deep saline aquifer to shallow fresh groundwaters. *Transport In Porous Media* 90:153–81
- Leonenko Y, Keith D (2008) Reservoir engineering to accelerate the dissolution of CO<sub>2</sub> stored in aquifers. *Environmental Science and Technology* 42:2742–7
- Lindeberg E (1997) Escape of CO<sub>2</sub> from aquifers. *Energy Conversion and Management* 38:S235–40
- MacMinn C, Neufeld J, Hesse MA, Huppert H (2012) Spreading and convective dissolution of carbon dioxide in vertically confined horizontal aquifers. *Water Resour Res* 48, doi:10.1029/2012WR012286
- Nordbotten J, Kavetski DC, Celia MA, Bachu S (2009) A semi-analytical model estimating leakage associated with CO<sub>2</sub> storage in large-scale multi-layered geological systems with multiple leaky wells. *Environmental Science Technology* 43(3):743–9
- Pau G, Bell J, Pruess K, Almgren A, Lijewski M, Zhang K (2010) High resolution simulation and characterization of density-driven flow in CO<sub>2</sub> storage in saline aquifers. *Advances in Water Resources* 33(4):443–55
- Pool M, Carrera J, Vilarrasa V, Silva O, Ayora C (2013) Dynamics and design of systems for geological storage of dissolved CO<sub>2</sub>. *Advances in Water Resources* 62:533-542
- Pruess K (2008) Leakage of CO<sub>2</sub> from geologic storage: role of secondary accumulation at shallow depth. *International Journal of Greenhouse Gas Control* 2:37–46
- Riaz A, Hesse MA, Tchelepi HA, Orr JF (2006) Onset of convection in a gravitationally unstable diffusive boundary layer in porous media. *Journal of Fluid Mechanics* 548:87–111
- Vilarrasa V, Silva O, Carrera J, Olivella S (2013) Liquid CO<sub>2</sub> injection for geological storage in deep saline aquifers. *International Journal of Greenhouse Gas Control* 14:84–96
- Vilarrasa V (2014) Impact of CO<sub>2</sub> injection through horizontal and vertical wells on the caprock mechanical stability. *International Journal of Rock Mechanics and Mining Sciences* 66:151-9
- Vilarrasa V, Carrera J (2015) Geologic carbon storage is unlikely to trigger large earthquakes and reactivate faults through which CO<sub>2</sub> could leak. *Proceedings of the National Academy of Sciences* 112(19):5938-5943
- Vilarrasa V, Carrera J, Olivella S (2016) Two-phase flow effects on the CO<sub>2</sub> injection pressure evolution and implications for the caprock geomechanical stability. In *E3S Web of Conferences* (Vol. 9, p. 04007). EDP Sciences
- Zhang Z, Agarwal R (2012) Numerical simulation and optimization of CO<sub>2</sub> sequestration in saline aquifers for vertical and horizontal well injection. *Computational Geosciences* 16:891–9
- Zoback MD, Gorelick SM (2012) Earthquake triggering and large-scale geologic storage of carbon dioxide. *Proceedings of the National Academy of Sciences* 109(26):10164-10168