

# Chapter 2

## Overview of Processes Occurring During CO<sub>2</sub> Geological Storage and Their Relevance to Key Questions of Performance

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**Abstract** The objective of this chapter is to provide an overview and discussion of the relevancy of various physical and chemical processes to be associated with the geological storage of CO<sub>2</sub> at a particular site, and thereby serve as a bridge between the detailed process descriptions and modeling techniques to be presented in the following chapters and the studying and simulation of site-specific physicochemical behavior of a potential CO<sub>2</sub> geosequestration site. The approach adopted is to address the relevancy of a given process in terms of the specific objectives, the technical issues of concern, or the key questions associated with CO<sub>2</sub> geological storage, in the context of the geological settings and characteristics of the storage site. The suggested approach is exemplified by application to two field cases.

### 2.1 Introduction

In recent literature, many of the physical and chemical processes associated with CO<sub>2</sub> geological storage have been extensively and intensely investigated and reported. In Chaps. 3–5 of this book, they are reviewed and described in detail, together with modeling techniques used to simulate them. These chapters also give a substantial list of references related to these processes and their modeling. The objective of the present chapter is to provide an introductory overview and a bridge between these process descriptions and modeling techniques and the studying and simulation of physicochemical behavior of a potential CO<sub>2</sub> geosequestration site, and, in particular, to identify the relevant physicochemical processes to site-specific study of CO<sub>2</sub> injection and sequestration in the deep subsurface. The focus will be on CO<sub>2</sub> geosequestration in saline formations.

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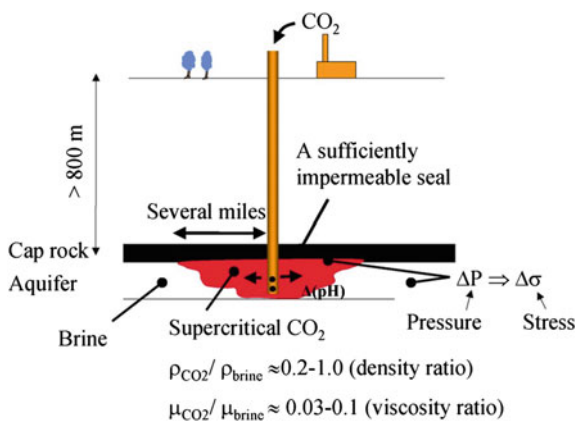
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The next section below gives an overview of the processes involved in a basic scenario of subsurface CO<sub>2</sub> injection and sequestration in a brine formation, followed by a general discussion of the geological settings and characteristics. Then the approach for discussing the relevancy of the physicochemical processes to CO<sub>2</sub> geosequestration at a specific site is presented. The implementation of this approach with an example of application to two field cases is described and discussed in the remaining sections of this chapter.

## 2.2 Overview of Processes in a Basic Scenario

For the sake of discussion in this chapter, it is useful to consider a basic scenario of injection and storage of CO<sub>2</sub> in brine formations as presented in Fig. 2.1, which shows a storage injection zone overlain by a caprock greater than 800 m in depth. Three main physicochemical processes are indicated. First, there is the hydrological process of buoyancy flow of the CO<sub>2</sub> with its factor-of-two lower density and an order-of-magnitude lower viscosity. Thus the plume of injected CO<sub>2</sub> will migrate outwards from the injection well and upwards towards the caprock by buoyancy. Other hydrologic factors also come into play, which will be discussed below. Second, both injection and buoyancy provide additional pressure on the rock matrix of the formation, which may thus be deformed, with changes in matrix porosity or fracture apertures. They in turn cause changes in flow permeability and, consequently, the flow field. This is what we call the hydromechanical effect. Finally, the injected CO<sub>2</sub> plume will, in general, chemically react with the formation minerals. This could give rise to porosity changes near the injection well, but, positively, formation matrix minerals can react with the injected CO<sub>2</sub> to form new minerals in the rock matrix, thus trapping CO<sub>2</sub> chemically. This is the mineral trapping process for sequestration of CO<sub>2</sub>. These main processes are discussed in more detail below.

**Fig. 2.1** Schematic diagram of CO<sub>2</sub> geosequestration in saline formation



CO<sub>2</sub> injected into a deep brine formation will be present in three forms: a dense supercritical phase; a dissolved state in pore water, and an immobilized state through geochemical reaction with in situ minerals. The dissolved part of estimated to be from 2 % in saturated NaCl brines by weight to 7 % in typical ground water. CO<sub>2</sub> immobilization in formation matrix minerals is a slow process and varies considerably with rock types. The amount of CO<sub>2</sub> sequestered through such mineral reactions can be comparable with CO<sub>2</sub> dissolution in pore waters. Among all the forms that the injected CO<sub>2</sub> takes in a brine formation, the liquid-like supercritical phase is the main storage form and it has properties quite different from those of the pore water. For example, for storage of CO<sub>2</sub> at 1000 m depth, CO<sub>2</sub> density is about 60–75 % that of water in the formation, while its viscosity is about a factor of 15–20 times less than that of water.

The lower density of the stored supercritical CO<sub>2</sub> will cause buoyant flow of CO<sub>2</sub> to the top of the injection zone below the caprock. The flow depends on the density difference as well as the vertical and horizontal permeabilities of the formation. Because of the tendency for buoyancy flow of CO<sub>2</sub> to the top of the injection formation, the areal extent of the injected CO<sub>2</sub> will be much larger than a buoyancy-neutral fluid. For example, storage of  $2.7 \times 10^{11}$  kg of CO<sub>2</sub>, injected at a rate of 350 kg/s for 30 years into a 100-m thick formation with  $k_x = k_z = 10^{-13} \text{ m}^2$ , has been estimated to have an increase in areal extent resulting from buoyancy flow by a factor of approximately 1.4. In this example, because of the large volume of CO<sub>2</sub> involved, the areal extent of the injected CO<sub>2</sub> can be as much as 120 km<sup>2</sup>.

Once the injected fluid is in place, what happens if it is next to a leakage path in the caprock, such as an abandoned borehole or a fault? The density of supercritical CO<sub>2</sub> at a depth of 1000 m is about 600–750 kg/m<sup>3</sup>, resulting in a significant buoyancy driving force causing an upward leakage of CO<sub>2</sub>. However the buoyancy pressure needs to be larger enough to overcome the gas entry pressure into the caprock pores. One can estimate the thickness  $h$  of the layer of CO<sub>2</sub> needed to provide enough buoyancy pressure to exceed the gas entry, which turns out to be 70–170 m for a pore radius of  $10^{-7}$  m. However, if there exists a fracture in the caprock, the effective pore radius in the fracture can be much larger and thus the thickness of CO<sub>2</sub> required to overcome the gas entry pressure of the fracture would be much less. In general, as CO<sub>2</sub> migrates upward, the flow involves evolving phases of CO<sub>2</sub> as well as the brine, presenting a complex phase interference effect.

The very low viscosity of supercritical CO<sub>2</sub> will give rise to flow instability at the CO<sub>2</sub>-brine interface as CO<sub>2</sub> is being injected into the storage formation. This flow instability results in fingering. In other words, instead of piston-like flow of the CO<sub>2</sub> front into the injection formation, parts of the front will flow much faster in the form of fingers. This phenomenon occurs in parallel with the buoyancy flow effect discussed above. However, viscous fingering of CO<sub>2</sub> may not be as significant in the presence of geologic heterogeneity.

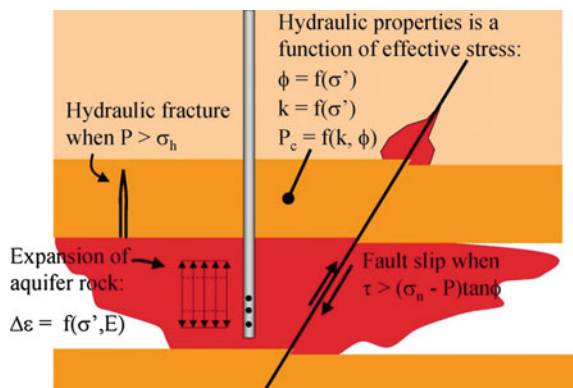
Heterogeneity of the injection formation gives rise to the fingering or channeling effect. The injected CO<sub>2</sub> will be channelized to follow the most permeable paths because of the spatial variation of permeability. The flow pattern will depend not

only on the permeability variability and its spatial correlation range, but also on the saturation level of  $\text{CO}_2$  in the different parts of the brine formation.

Mechanically, the main concern for liquid waste injection has been to ensure that injection pressure is safely below that which will cause hydraulic fracturing or affect well integrity (Fig. 2.2). For  $\text{CO}_2$  injection and storage, however, both the injection and buoyancy pressures need to be considered. While injection pressure is highest around the injection well and starts to decrease after the termination of injection, buoyancy pressure extends over the entire  $\text{CO}_2$  plume and lasts well beyond the injection period. An increase in formation fluid pressure, due to both injection and buoyant pressures, will cause local changes in the effective stress field, which, in turn, will induce mechanical deformations, possibly increasing the porosity and permeability and thus reducing the fluid pressure. However at the same time, increasing pressure may also cause irreversible mechanical failure in the caprock. This mechanical failure may involve possibly shear-slip along existing fractures and creation of new fractures (hydraulic fracturing), that reduce the sealing properties of the caprock system. In addition to these mechanical processes, replacing the native formation fluid with  $\text{CO}_2$  may cause changes in rock mechanical properties through chemical-mechanical interactions between the  $\text{CO}_2$  and the host rock, or through desiccation of fractures.

Chemically, at the  $\text{CO}_2$  front where  $\text{CO}_2$  is dissolved in water, the acidity of the groundwater is increased and many minerals comprising the host rock matrix minerals such as calcite, may dissolve readily, leading to an increase in permeability and porosity along the flow channel. This leads to a higher flow rate and increased dissolution, potentially forming what are known as wormholes. On the other hand, based on experience from enhanced oil recovery,  $\text{CO}_2$  has been known to reduce injectivity in some cases, but to increase permeability near injection wells in others. There are also data indicating that dissolved  $\text{CO}_2$  will cause a reduction in permeability where the carbonate minerals precipitate along the flow paths with a large pressure gradient. All these observations suggest the need for careful evaluation of the compatibility between supercritical  $\text{CO}_2$  and geochemistry of the brine formation. Such an evaluation may also yield information useful for the design of

**Fig. 2.2** Schematic diagram on hydromechanical changes due to  $\text{CO}_2$  injection and storage (from Jonny Rutqvist, private communication 2012)



injection operations, such as keeping injection pressure below a certain value so that there will be no severe pressure gradients to induce precipitation or dissolution.

With CO<sub>2</sub> storage at or below the depth of about 1000 m, there may well be a sequence of intervening strata of confining and permeable layers separating the injection zone and the lowest underground sources of drinking water (USDW). This sequence of strata can provide a compounded margin of safety to reduce CO<sub>2</sub> upward leakage. Thus, each high-permeability layer serves as an injection zone for CO<sub>2</sub> leaking into it from below and spreading in it. The next overlying confining layer will then act as the next caprock to prevent continuing CO<sub>2</sub> leakage.

Because of the large volume of CO<sub>2</sub> being injected and stored, the displacement of in situ brine is an issue of concern. The displaced brine may migrate to neighboring formations and/or diffuse into shallower hydraulically conductive units. Potential focused migration may also occur through abandoned wells or sub-vertical faults and connected fractures.

## 2.3 Geological Settings and Characteristics

A number of different types of geologic formations have been proposed for CO<sub>2</sub> geosequestration. These include saline formations, depleted oil and gas reservoirs, coal seams, and possibly organic-rich shale. In this chapter, we focus on saline formations. For such formations, CO<sub>2</sub> sequestration is mainly through four different trapping mechanisms: structural or stratigraphic trapping, capillary or mobility trapping, dissolution trapping, and mineralization or chemical trapping. The first two tend to occur earlier in time, whereas the latter two are much slower.

Structural or stratigraphic trapping depends on the local geology: a low-permeability, regionally extensive caprock with high gas entry pressure, sealing faults, and anticline structures all have the potential to trap buoyant CO<sub>2</sub>. Conversely, geologic heterogeneity that creates gaps in the low-permeability structures can promote concentrated vertical flow of CO<sub>2</sub> along these preferential flow paths.

Capillary or mobility trapping is due to phase interference between the immiscible CO<sub>2</sub> (the nonwetting gas-like phase) and the brine (the wetting phase). The mobilities of the flowing phases depend on phase distributions at the pore scale, as embodied in continuum-scale relative permeability functions. It is widely recognized that fluid distributions within the pore space differ for drainage (where CO<sub>2</sub> as the nonwetting phase displaces brine as the wetting phase, in the case of an advancing CO<sub>2</sub> plume), and imbibition (where brine as the wetting phase replaces CO<sub>2</sub> as the nonwetting phase, in the case of a retreating CO<sub>2</sub> plume). This process-dependence can be represented by using hysteretic relative permeability functions in which the nonwetting-phase residual saturation is small during drainage, but large during imbibition, leading to the trapping of significant quantities of CO<sub>2</sub>.

Dissolution trapping occurs when  $\text{CO}_2$  dissolves in brine. The  $\text{CO}_2$ -saturated brine density increases, making it heavier than the surrounding brine. Buoyancy forces then contribute to trapping of dissolved  $\text{CO}_2$  due to the tendency for  $\text{CO}_2$ -saturated brine to migrate downwards, often in the form of fingering flow. Mineralization or chemical trapping occurs when  $\text{CO}_2$  reacts with rock minerals to form carbonate compounds, effectively immobilizing the  $\text{CO}_2$ .

Among these four trapping mechanisms, structural trapping is important, especially in the early time frame. Saline formations may be embedded in several different large-scale and regional tectonic settings: (a) post-collisional, or post-orogenic, inland basins; (b) passive continental margins; e.g., paralic to shelf environment; and (c) fold and thrust belts. The inland basins contain mainly listric and normal faulting with minor periods of reverse faulting. Both inland basins and passive continental margins are assumed to have low tectonic or seismic activity. The fold and thrust belts as a whole are compressional, but they can exhibit large extensional domains, with the result that all types of faults, including large strike-slip faults, may be expected. An understanding of these large-scale  $\text{CO}_2$ -geosequestration site settings is useful because of the large amounts of  $\text{CO}_2$  to be stored; a proper evaluation of the large  $\text{CO}_2$  plume footprint and the even larger-scale effects of its displacement of in situ brine (see Fig. 2.1) require consideration of the characteristics of these large-scale settings.

Geological factors of importance for  $\text{CO}_2$  geosequestration in all the different large-scale settings include faults, folds, sedimentary facies, and various caprock characteristics. Faults may offset stratigraphic layers, bring permeable formations above the caprock and the storage formation closer together, or juxtapose them. They may also be either hydraulically conductive, providing a migration path, or sealed, providing a barrier. Thus, they play a significant role in potential migration, or conversely, in flow compartmentalization and storage capacity.

Folds in large-scale reservoirs are rather a local feature, occurring in the vicinity of faults, and can be accompanied by many fractures. If a caprock layer laps on a buried fold or pre-existing inclining structure, the caprock may thin out (or “pinch” out), with potential migration of stored  $\text{CO}_2$  or displaced brine, at that location.

For sedimentary facies, the concern is with spatially varying grain size or effective pore-size distribution, and pore structure. This spatial variability will impact estimates of storage capacity and pressure buildup. Spatially varying mineralogy also leads to different dissolution and precipitation reactions and different reaction rates of rock with the stored supercritical  $\text{CO}_2$ , in situ brine, and brine with dissolved  $\text{CO}_2$ .

With respect to the caprock, the geological properties of interest are rock type, facies distribution, thickness, and (more specifically) its integrity under different thermal and geomechanical processes, such as those involved in induced or natural seismic events.

## 2.4 Approach

The approach adopted in this chapter for discussing the relevancy of the physico-chemical processes to CO<sub>2</sub> geosequestration is to identify process relevancy in terms of specific objectives, technical issues of concern, or key questions associated with CO<sub>2</sub> geosequestration, in the context of the geological settings and characteristics of the geosequestration sites. We shall refer to the CO<sub>2</sub> geosequestration-related objectives, issues, and questions all as key questions (KQ), which will be discussed below.

In our discussion of relevant processes, we shall consider not only physical and chemical processes (P), but also geologic features or structures (F). The latter include such features as the presence of fractures and faults, as well as rock-property heterogeneity, such as spatial variations in grain size, porosity and permeability. It is often impossible to discuss processes without also discussing the features, because features are the framework within which the processes operate. Furthermore, the separation of processes and features is, in many instances, not clear-cut. Sometimes a so-called process is defined to represent some underlying physical processes acting on “smeared-out” or averaged features, especially when detailed information on these features is not available. One example of this is dispersion, which results from solute transport through pathways of different velocities in the rock pores, with diffusion among the paths. Dispersion is then a flow and migration process in a continuum representation of this pore-scale variability. Nevertheless, on a practical level, one may separate processes (P) and features (F) by the fact that processes are represented as a term in the governing equations of a model that simulates rock physical and chemical behavior, while features are accounted for by the mesh design and material property values in the internal or boundary elements of the model.

Definitions or representations of processes and their characteristics depend also on the conceptual model used for their representations. For example, the relative permeability functions used to describe multiphase flows are used to represent the flow interference between the multiple phases in the pore or fracture aperture structure. They may display hysteretic effects, and are a function of scale. Such hysteresis and scale-dependence will also be included in our discussions as part of the processes.

As can be seen in the brief discussion above, there are no clear distinctions between features, processes, and process representations, and often this kind of grouping is somewhat arbitrary. It is probably fruitless to pursue a better definition of such distinctions, and the present chapter proposes the use of these categories only as a convenience to guide our discussion on their relevancy to CO<sub>2</sub> geosequestration.

Two further remarks are needed concerning our approach. First, the matter of temporal and spatial scales is critical and needs to be kept in mind in any consideration of processes and features. In fact, this applies not only to processes, but also to the key questions, objectives, or technical issues of concern related to CO<sub>2</sub>

geosequestration. For example, the potential or risk of CO<sub>2</sub> migration is an issue of concern, but processes related to focused migration from a particular location and those related to average migration over a large area may be quite different.

Secondly, some of the key questions can and need to be addressed through operational strategy, which includes such activities as site selection, injection well spacing, injection schedule, monitoring plan, and associated operational re-adjustment or optimization. Furthermore, for consideration of operational strategy, there is a need of data, both generic and site-specific. Since it is impossible to have a “complete” set of data, one issue is the evaluation of an optimal set of desired data, while another issue is research towards defining and determining uncertainty due to data gaps.

In Sect. 2.5, the main objectives, technical issues of concern, or key questions associated with CO<sub>2</sub> geosequestration will be identified. For simplicity, they are all referred to as key questions (KQs).

Section 2.6 will provide a number of design alternatives related to operational strategy. Different alternatives involve different costs, and costing level is an important factor in any practical CO<sub>2</sub> geosequestration projects, but this will not be addressed in the present chapter. In Sects. 2.7 and 2.8 we shall list the main features (F), and processes (P), respectively. For this, we draw on information from Chaps. 3–5 of this book, and the current state of knowledge. Particular attention will be paid to the coupled processes, which are those processes coupling the effects of thermal, hydrologic, mechanical, and chemical processes.

These sections are then followed by Sect. 2.9 with an attempt (in the form of tables) to associate each KQ with various F and P, along with a few remarks on related operational strategy and data needs. The tables are not presumed to be complete, but may be useful as an initial step in the planning, evaluation, and development of a CO<sub>2</sub> geosequestration project.

Finally, in Sect. 2.10 one particular large-scale study of two potential CO<sub>2</sub> geosequestration sites in the literature will be reviewed, to point out the KQs considered and the features and processes included in the study. A few remarks on the application of the present work then conclude this chapter.

## 2.5 Key Questions

Key questions (KQs) associated with CO<sub>2</sub> geosequestration addressed in this chapter are those from a technical and scientific perspective. Thus, we do not discuss public acceptance, cost-benefit, or regulatory and legal aspects of the problem. We may divide the KQs in two groups: those related to the performance of CO<sub>2</sub> geosequestration and those related to its risk.



### ***2.5.1 Performance-Related Key Questions***

In line with current thinking in this field, we may define the performance-related KQs to be of three categories: namely, capacity of CO<sub>2</sub> that can be sequestered at a given site, injectivity of CO<sub>2</sub> into deep saline formations, and containment of CO<sub>2</sub> without significant migration.

Capacity is by definition a large-scale issue, since for CO<sub>2</sub> geosequestration to be useful, large quantities of CO<sub>2</sub> will have to be stored. Capacity includes contributions from structural trapping, residual or capillary trapping, dissolution and mineralization trapping, and mechanical deformation of pore space. One controlling parameter that limits storage capacity is the pressure rise in the CO<sub>2</sub> storage formation. A maximum limit to the pressure rise is often required by regulatory agencies to stay safely below the level that could potentially cause hydraulic fracturing of the caprock or significant seismic events (Rutqvist et al. 2007). Another limit to pressure rise is the gas entry pressure into the overlying sealing formation, with its low permeability and high capillarity.

Injectivity, on the other hand, is probably a local issue concerning the capability of the formation to receive the injected CO<sub>2</sub> without unwarranted effects, such as gas entry into caprock and hydraulic fracturing leading to leakage near the injection well or above the CO<sub>2</sub> plume. This KQ may be partially addressed by selecting a storage saline formation with sufficiently high permeability, using horizontal injection wells, or changing the separation of injection areas over the storage formation, which are part of operational strategy.

Containment in the performance context involves the general question, how effective is the containment of CO<sub>2</sub> sequestration in the saline formation? In some assessments, it has been suggested that a small percentage of leakage can be tolerated for a system still considered to be effective in sequestering a significant amount of CO<sub>2</sub>. Although the mechanisms of potential migration through faults and abandoned wells have been much studied, how large and significant the migration volume would be for a given geosequestration site remains an open issue to consider. Furthermore, separately from the containment question, a focused leakage causing environmental damage and danger at the leakage locations on the land surface is an important issue.

### ***2.5.2 Risk-Related Key Questions***

These may be identified as (a) induced seismicity, (b) focused migration, (c) diffused migration, (d) large-scale flow, brine displacement, and pressure changes, and (e) leaching and transport of minerals and chemicals to shallow groundwater systems.

With induced seismicity, we have both the potential for significant or major seismic events involving fault shear slippage (with possibly the creation of new

migration paths or enhancement of existing paths) and induced seismic swarms of small magnitudes. The risk for focused migration refers specifically to migration through discrete flow paths along faults and connected fractures through the caprock. The caprock may be of a single layer or have a multilayered structure. Included in this category is also leakage through imperfectly sealed, abandoned wells or improperly constructed injection wells. Such focused migration may involve free-phase  $\text{CO}_2$  or  $\text{CO}_2$ -rich brine. For the migration of brine with dissolved  $\text{CO}_2$ , the  $\text{CO}_2$  may degas with reduced pressure as the fluid moves upwards. Diffused migration, on the other hand, is migration of mainly brine and  $\text{CO}_2$ -rich brine through caprock layers as a whole without discrete well-defined flow paths. It thus tends to be slow migration that covers a large horizontal area and flows vertically through a number of low-permeability layers to the shallower groundwater system, the vadose zone, and to the land surface.

The key question regarding large-scale flow, brine displacement, and pressure change arises from concerns over how the large quantity of  $\text{CO}_2$  injected and stored underground would affect the groundwater system. The brine displaced by the stored  $\text{CO}_2$  may enter into shallower groundwater formations, increasing their salinity, and the associated increases in pressure may also cause significant changes in the groundwater flow patterns. Finally, the leaching and transport of minerals and chemicals from  $\text{CO}_2$ -rock interactions, and from interactions between deeper brine and shallower formations during brine migration, are subjects of concern if they significantly affect the shallow aquifers.

## 2.6 Operational Strategy

Before we discuss features and processes involved in  $\text{CO}_2$  geosequestration, it is useful to consider operational strategy used in  $\text{CO}_2$  injection. Some of the key questions discussed in the last section can and should be addressed by appropriate design and planning of  $\text{CO}_2$  geosequestration projects. Based on the operational strategy used, the impact of some of the features and processes may be reduced, while others may become more important and require careful study. Below, we identify a number of issues related to operational strategy.

Usually, the first step within operational strategy is site evaluation or characterization, based on which a site selection can be made. For example, the performance-related key question of  $\text{CO}_2$  storage capacity can be partially addressed by selecting a site with an extensive saline formation under a good caprock layer. Similarly the risk-related key question of induced seismicity can be partially addressed by selecting a site with a low potential for seismic activity and moderate in situ stress fields. Such site selection depends on previously available data and new data that can be obtained at reasonable cost and time. Scale is also an issue: sometimes, it may be possible to select a “good” site around an injection well or even over the expected  $\text{CO}_2$  plume footprint, but the site may not be so good over the vast area of pressure changes and brine migration induced by  $\text{CO}_2$

injection and buoyancy flow. An added aspect of site characterization is the establishment of the baseline conditions (including dynamic conditions) of the site, such as pressure and salinity distributions, as well as natural seismic activities. Such information is needed to understand the impact of CO<sub>2</sub> injection and storage at the site and also the monitoring results during storage operation.

The choice of multiple vertical or horizontal CO<sub>2</sub> injection wells and placement of such wells is also part of operational strategy. With horizontal wells and a wide separation between injection areas, the injection pressure at the injection site may be less for the same total injection rate, but the cost may be dramatically higher. An injection strategy to target certain storage formations or to promote certain trapping mechanisms has also been explored. For example, alternating CO<sub>2</sub> and water injection has been suggested as a way to enhance capillary trapping of CO<sub>2</sub>. Constant-rate injection versus variable-rate injection has also been studied, with some studies indicating an advantage for variable-rate injection in promoting CO<sub>2</sub> dispersion and dissolution in brine.

Concerns for pressure rise caused by CO<sub>2</sub> injection and buoyancy can probably be partially addressed by schemes such as brine withdrawal from the storage formation. In this case, however, treatment and disposal of the produced brine become an issue. In general, pressure-management methods can be considered to be part of the operational strategy. Finally, strategies for monitoring potential leakage and its mitigation are important steps in CO<sub>2</sub> geosequestration. How to best design and implement monitoring and mitigation is an important operational issue.

## 2.7 Features

Some of the large-scale regional features have been described in Sect. 2.3 (on regional geological setting). Of relevance to the key questions discussed in Sect. 2.5 are a number of local and large-scale geological features and structural characteristics.

First, the geometry of the saline storage formation and caprock needs to be defined and characterized. This activity includes accounting for the lateral extent of the storage formation and the boundary conditions, whether they are open, closed, or partially open. The geometry and permeability structure of the caprock has great relevance to CO<sub>2</sub> containment. For example, there may be an advantage in having the caprock be composed of multiple low-permeability, high-capillarity layers rather than a single low-permeability layer. The roughness of the caprock-saline formation interface has also been proposed to have a significant effect: it may provide effective traps for CO<sub>2</sub>, since the CO<sub>2</sub> flows up and spreads at the caprock interface due to its buoyancy.

The CO<sub>2</sub> storage formation is heterogeneous with a permeability correlation structure, and this heterogeneity may subject flow to hydraulic compartmentalization, so that not all of the storage formation is utilized for CO<sub>2</sub> storage. Other features that affect the distribution of CO<sub>2</sub> flow are faults (sealed or open), folds,

and fracture zones. All these complications could result in preferential flow paths, which may divert CO<sub>2</sub> flow to farther-away regions without fully occupying the storage formation in the near field, and which, furthermore, may also create CO<sub>2</sub> migration paths to shallower formations.

Not only is formation heterogeneity an issue, but also the formation property anisotropy, which is a feature that is essential for addressing some of the key questions. Not all of the geological features can be known a priori. This will give rise to uncertainty because of lack of knowledge. Some research effort has been devoted to the definition and estimation of uncertainty: it is hoped that the uncertainty can be bounded, based on site-specific data on a large scale.

## 2.8 Processes

Processes involved in CO<sub>2</sub> geosequestration are discussed in detail in Chaps. 3–5 of this book. Below, they are presented in an outline way under processes, process representations, and coupled processes.

### 2.8.1 Processes

Under processes, we identify below some significant hydrological, chemical, mechanical, and thermal processes relevant to CO<sub>2</sub> geosequestration:

- Flow of supercritical CO<sub>2</sub>, gaseous CO<sub>2</sub>, brine with dissolved CO<sub>2</sub>, and in situ brine, including “impurities” injected with the CO<sub>2</sub>.
- Pressure changes due to both injection pressure and buoyancy (gravity) pressure.
- Buoyancy-driven flow of supercritical and gaseous CO<sub>2</sub>, and, in general, gravity-driven flow among fluids of different densities.
- Pore-scale capillarity, which contributes to entrapment of CO<sub>2</sub>.
- Multicomponent flow-viscosity fingering.
- Heterogeneity-induced channeling or fingering and exchange between fast and neighboring slower paths.
- Macro-scale trapping of CO<sub>2</sub>/brine due to heterogeneity and flow compartmentalization.
- Interface behavior between CO<sub>2</sub>, brine with dissolved CO<sub>2</sub>, in situ brine, and low-salinity fluids at shallower formations. For example, such behavior includes the capillary fringe effect at the CO<sub>2</sub>-brine interface and potential flow instability at the interface.
- Displacement of formation brine and its interaction with shallow groundwater and rock minerals.
- Large-scale CO<sub>2</sub> spreading, mixing, fingering, and dissolution.

- Dissolution of CO<sub>2</sub> in brine and transport of dissolved CO<sub>2</sub> through convective flow.
- Evaporation of water into CO<sub>2</sub> phase and subsequent salt precipitation.
- Molecular diffusion of dissolved CO<sub>2</sub> between flow zones of different velocities or between fractures and the adjacent rock matrix.
- Reactions of CO<sub>2</sub> and supercritical CO<sub>2</sub> with rock minerals.
- Thermal transfer.
- Mechanical stress and displacements/deformations.
- Fracture shear slippage and propagation; induced seismicity.

### 2.8.2 *Process Representations in Macro-Scale Models*

Some processes are defined based on macro-scale conceptual models used to represent the underlying physicochemical processes occurring in a structural framework of a smaller scale. Because of this, there may be significant scale dependence in these processes, both temporally and spatially. Although we refer to them as simply “processes”, it is useful to list them here so that their dependence on conceptual models of the geological system is highlighted.

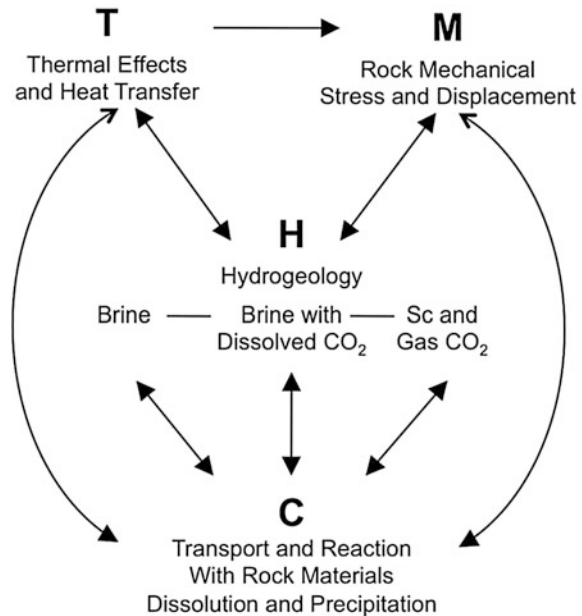
- Flow and transport in the hydrogeological system with underlying heterogeneity of different scales. The representative permeability values depend on the existence of representative elementary volume. Furthermore, the representative elementary volume may be different for different processes. For example, it will be very different among pressure (a very diffusive process), thermal transport (a moderately diffusive process), and solute transport (a process much more sensitive to local-scale heterogeneity). In this respect, methods for determining parameter values for particular applications require careful consideration of the related scale involved.
- Flow anisotropy. The differences in flow permeability for different flow directions depend on the characteristics of permeability distribution in the different directions. It may also be a spatially varying quantity.
- Flow dispersion is a representation of lower-level velocity variations of CO<sub>2</sub> (or other fluids) in the pore structure at the scale of interest, such as the scale of the calculation element within a numerical model mesh. If the calculation element is small, a dispersion coefficient (in the context of the conventional Darcy’s equation) may not be defined, and other types of governing equations involving possible non-local spatial and temporal terms may need to be used.
- Relative permeability effects on CO<sub>2</sub>-brine flow, as represented, for example, by van Genuchten equations or Brooks–Corey curves. They may also display hysteresis effects.
- Linear sorption and higher-order kinetic effects.

### 2.8.3 Coupled Thermo-Hydro-Mechanical-Chemical Processes

Coupled processes are illustrated in Fig. 2.3, which shows the possible coupling between thermal effects and heat transfer (T), rock mechanical stress and deformation (M), chemical transport and reactions with rock minerals resulting in dissolution and precipitation (C), and hydrological processes (H). For hydrological processes, it is useful to consider different types of fluids; i.e., brine, brine with dissolved  $\text{CO}_2$ , and supercritical and gaseous  $\text{CO}_2$ , since they not only interfere with each other, but they also have different chemical reactions and characteristics. In Fig. 2.3, the solid arrowheads indicate a strong coupling direction, while the open arrowheads indicate a coupling direction of weaker strength. Possible coupled processes include

- Induced flow and transport of heat through thermal convection.
- Dissolution and transport of rock minerals in brine, brine with dissolved  $\text{CO}_2$ , and supercritical and gaseous  $\text{CO}_2$ .
- Flow-permeability changes due to precipitation or dissolution, changing pore size and structure.
- Chemical reactions with injected  $\text{CO}_2$  containing impurities such as  $\text{H}_2\text{S}$ ,  $\text{SO}_2$ , etc.
- Fluid-pressure-induced rock stress changes, causing porosity or fracture aperture changes, leading to changes in flow permeability.

**Fig. 2.3** Coupled THMC processes, coupling thermal (T), hydrological (H), rock mechanical (M) and chemical (C) effects occurring in saline formations used for  $\text{CO}_2$  geosequestration



- Hydraulic fracturing or shear slippage.

Figure 2.3 may be useful as a framework to identify additional coupled processes, in the context of the various features presented in Sect. 2.7, which are of relevance to particular key questions associated with CO<sub>2</sub> geosequestration.

**2.9 An Attempt to Associate Features (F) and Processes (P) with Key Questions (KQ)**

This section is composed of a series of tables (Tables 2.1, 2.2, 2.3, 2.4, 2.5, 2.6, 2.7 and 2.8) that attempt to associate those F and P discussed in Sects. 2.7, and 2.8, respectively, with the KQ presented in Sect. 2.5. No claim of completeness is made here; the tables may serve as a starting point in the initial evaluation, planning, predictive modeling, design and implementation of a CO<sub>2</sub> geosequestration project. Details about the individual processes may be found in the other chapters of this book.

**2.10 An Example Application to a Study of Large-Scale CO<sub>2</sub> Geosequestration at Two Potential Sites**

This section provides an example of applying the above formalism on a recently published large-scale modeling of two potential sites for CO<sub>2</sub> geosequestration. After a summary of this study, we shall identify the key questions addressed and the important features and relevant processes included in this work.

In a series of papers (Birkholzer and Zhou 2009; Zhou et al. 2010; Zhou and Birkholzer 2011), Birkholzer, Zhou, and coworkers studied the storage capacity of

**Table 2.1** KQ: Capacity

F	Porosity; lateral and vertical extent of storage formation; boundary conditions; caprock structure—structural trapping; presence of sealed or conductive faults and of migration paths; heterogeneity, and flow compartmentalization
P	Buoyancy flow, flow fingering at CO <sub>2</sub> -water interface and at CO <sub>2</sub> -saturated brine and formation-brine interface; CO <sub>2</sub> solubility in brine and solution rate; mineralization and rate; residual saturation and hysteresis in relative permeability characteristics
Coupled process	Hydromechanical effects on porosity and on fault or fracture permeability; hydromechanical effects on dissolution and mineralization
Remarks on operational strategy	Site characterization and selection are needed. The controlling parameter is pressure rise in the storage formation due to CO <sub>2</sub> injection and storage; it must be kept below a regulatory maximum. Pressure management methods may be applied

**Table 2.2** KQ: Injectivity

F	Short-term
	Local permeability and permeability structure; potential presence of faults and abandoned wells
	Long-term
	Lateral and vertical extent of storage formation; boundary conditions; presence of sealing or nonsealing faults
P	Injection pressure and flows; two-phase flow effects
Coupled process	Hydromechanical couplings that allow porosity changes; hydrofracturing process
Remarks on operational strategy	Controlling injection peak pressure through adjustments of injection well spacing; use of horizontal wells, pressure management through brine production (where brine treatment and disposal become an issue); controlled hydrofracturing

**Table 2.3** KQ: Containment

F	Boundary conditions; faults and fractures; caprock geometry and properties; anticlinal structures, presence of multiple caprock layers; abandoned wells
P	Buoyancy flow; flow channeling; multiphase flow through potential migration paths; effectiveness of structural trapping, capillary or residual trapping, and dissolution and mineralization trapping
Coupled process	Hydromechanical effects on fracture and fault permeabilities; hydrochemical effects on dissolution and precipitation, changing permeability and permeability structures
Remarks on operational strategy	Site evaluation and selection are important; development of monitoring plans and response strategies

**Table 2.4** KQ: Induced seismicity

F	Faults and fracture distributions; rock mechanical conditions in the neighborhood of faults and fractures In situ stress fields
P	Changes in rock stresses and deformations; fracture dilation; shear displacements; fracturing and fracture propagation
Coupled process	Injection-pressure-induced mechanical changes; buoyancy-pressure-induced mechanical changes; effects of stress dissolution on hydraulic and mechanical properties at mechanically stressed points
Remarks on operational strategy	Analysis of the potential for induced major seismic events versus multiple minor events; monitoring plan and understanding of possible occurrences of seismic swarms needed

CO<sub>2</sub> at two sites with different geological characteristics, based on a model study of the pressure rise and brine migration due to an industrial-scale CO<sub>2</sub> injection of 5 Mt per year over 50 years. The first site is the Illinois Basin, Mount Simon



**Table 2.5** KQ: Focused migration

F	Fault and fracture structure; connected fractures; heterogeneity-induced channeling; multiple caprock layers; abandoned wells and imperfectly constructed injection wells, formation pinch-out areas
P	Multiphase flow in faults, connected fractures, and heterogeneity-induced channeling paths; vertical migration through multilayer caprock; potential for accelerating migration rate, degassing
Coupled process	Coupled thermohydrological effects (e.g., cooling) on multiphase flow through vertical migration paths; coupled thermo-hydro-chemical and thermo-hydro-mechanical effects on permeability and migration paths
Remarks on operational strategy	Site evaluation and selection; Monitoring plan and response strategy needed

**Table 2.6** KQ: Diffused migration

F	Permeability distribution and geometry of storage formation and caprock system; structure of shallower geologic formation and aquifers; regional variations in rock hydrologic properties
P	Flow of brine with or without dissolved CO <sub>2</sub> through caprock and multilayered caprock; effects of spatial variations in porosity and gas-entry pressure; long-term diffusion and retardation processes; impacts of capillary or residual trapping, dissolution and mineralization trapping
Coupled process	Effects of hydrochemical processes on flow properties and vertical diffused migration
Remarks on operational strategy	Estimate of diffused migration and its impact needed; Monitoring plan and response strategy needed

**Table 2.7** KQ: Large-scale flow, brine displacement, and pressure changes

F	Regional geological structure and permeability distributions; current pressure distributions; properties and conditions at the boundaries between aquifer and aquitard systems both vertically and laterally; major faults and their permeabilities; geological features, such as folding and stratigraphic offsets; layer thinning; spatially varying pore structure; flow compartmentalization
P	Multiphase flow in near field or CO <sub>2</sub> injection well; flow of brine with dissolved CO <sub>2</sub> ; pressure and flow of regional brine across boundaries; interface flow behavior; effects of heterogeneity at different scales
Coupled process	Coupled thermo-hydro-chemical effects that may change permeability structure, especially at boundaries or flow constriction points
Remarks on operational strategy	Establishment of current and transient flow conditions prior to CO <sub>2</sub> geosequestration is important

**Table 2.8** KQ: Leaching and transport of minerals and chemicals from rock matrix to shallow groundwater systems

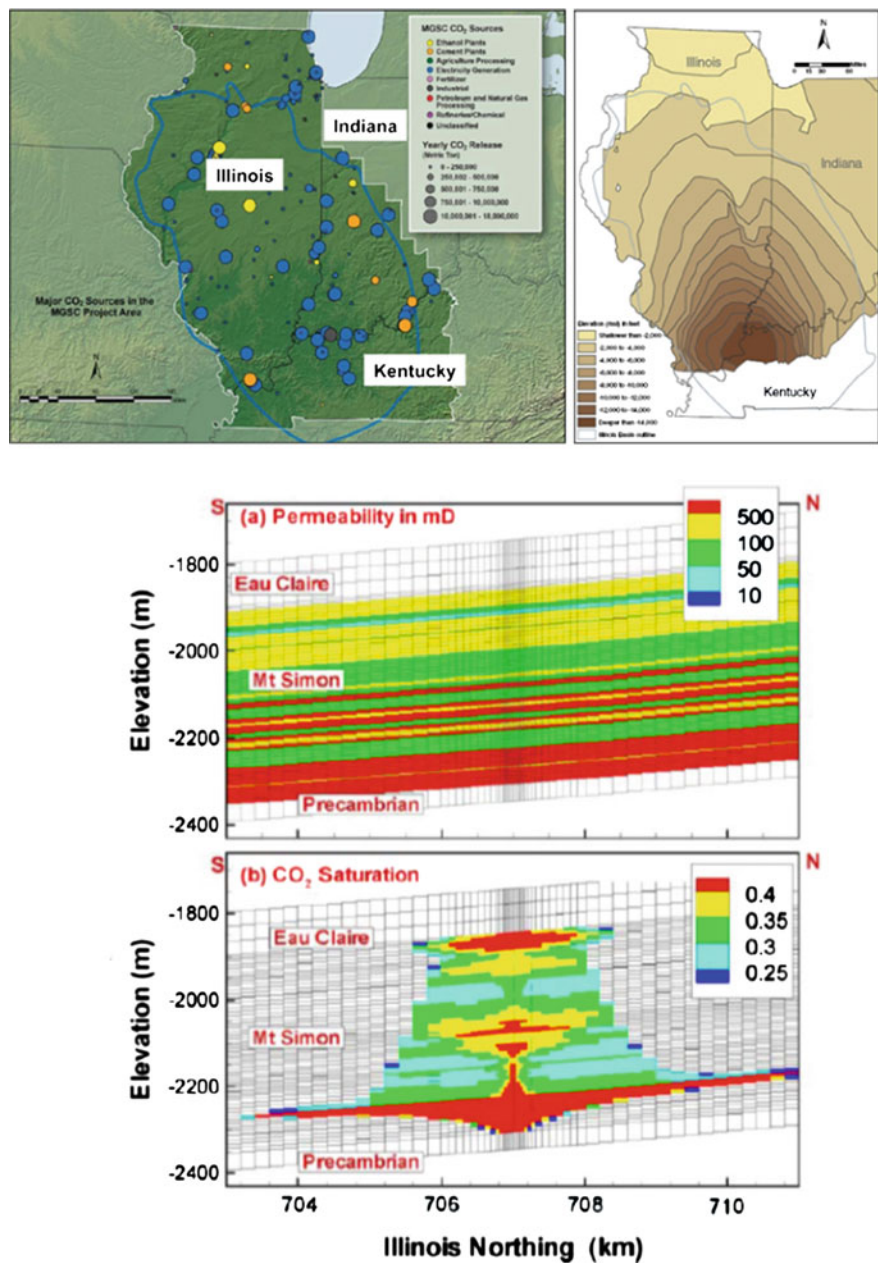
F	Local scale
	Hydraulic structure in the storage formation and spatial rock-mineral characteristics; chemical properties of injected CO <sub>2</sub> stream (with or without impurities)
	Regional scale
	Regional geological structure and permeability distributions; current pressure distributions; properties and conditions at the boundaries between aquifer and aquitard systems both vertically and laterally; major faults and their permeabilities; geological features, such as folding and stratigraphic offsets; layer thinning; spatially varying pore structure; flow compartmentalization
P	CO <sub>2</sub> -rock-water interactions, including effects of CO <sub>2</sub> in supercritical state, gaseous phase, and dissolved phase In the far field, chemical interactions between displaced brine into shallower or neighboring groundwater and rock systems
Coupled process	Effects of coupled thermo-hydro-chemical processes on flow patterns; mineral dissolution and precipitation
Remarks on operational strategy	Establishment of current and transient hydrochemical conditions prior to CO <sub>2</sub> geosequestration is important

Sandstone formation, which represents a large “open” system with continuous sealing caprock and few known fault zones. The second is the Southern San Joaquin Basin in California, with multiple sealing faults, which give rise to flow and storage volume compartmentalization. In this case, the caprock displays pinch-out in several directions. Details may be found in the references.

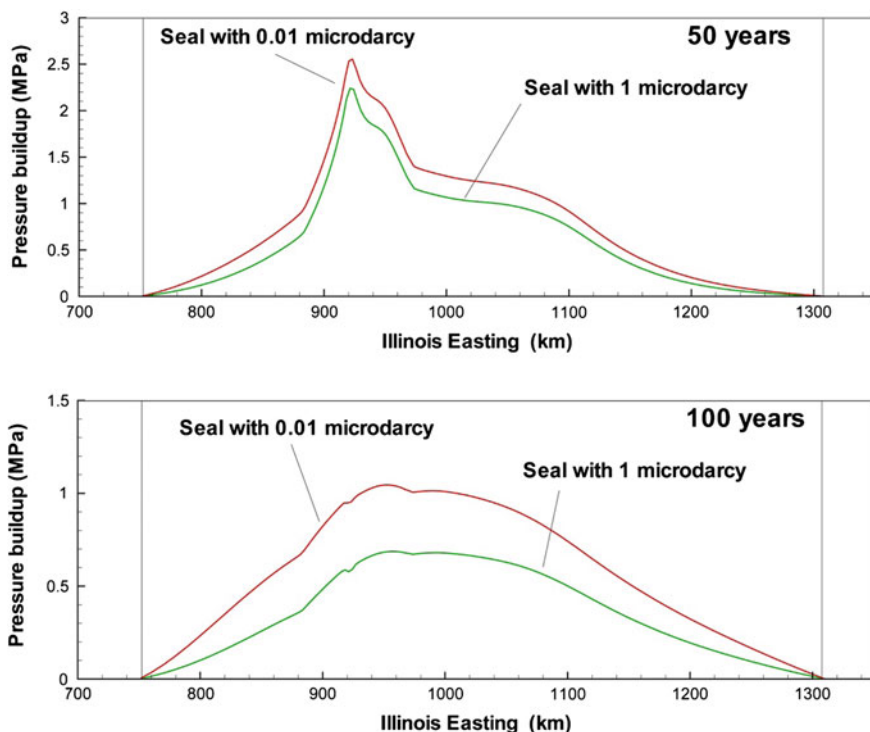
**2.10.1 Case of Illinois Basin, Mount Simon Sandstone Formation**

The upper left subfigure in Fig. 2.4 shows the location of the first site considered, the Mount Simon Sandstone formation. It has an area of roughly 570 km by 550 km, extending over the U.S. state of Illinois and parts of neighboring states. The depth of Mount Simon is shown in the upper right subfigure in Fig. 2.4. The formation is extensive laterally and continues to be present beyond the Illinois basin, thus allowing brine to escape into neighboring basins to the north, west, and east during a long-term CO<sub>2</sub> injection and migration. The southwestern model boundary is formed by the Ozark Uplift in Missouri, where the Mount Simon becomes thin or disappears.

The lower subfigure in Fig. 2.4 presents (a) the permeability distribution in a vertical cross section at roughly the middle of the Illinois Basin, showing layering in the vertical permeability with the Eau Claire serving as a caprock; and



**Fig. 2.4** The *upper subfigures* show the location and depth (in feet) of the Mount Simon Sandstone at Illinois Basin. The *lower subfigure* shows **a** the permeability (in mD) in a vertical cross-section at about the middle of the sandstone formation shown in the *upper figure* and **b** calculated CO<sub>2</sub> saturation after 50 years of CO<sub>2</sub> injection in the Arkosic layer into a depth interval from about 2240–2300 m. From Birkholzer and Zhou (2009)

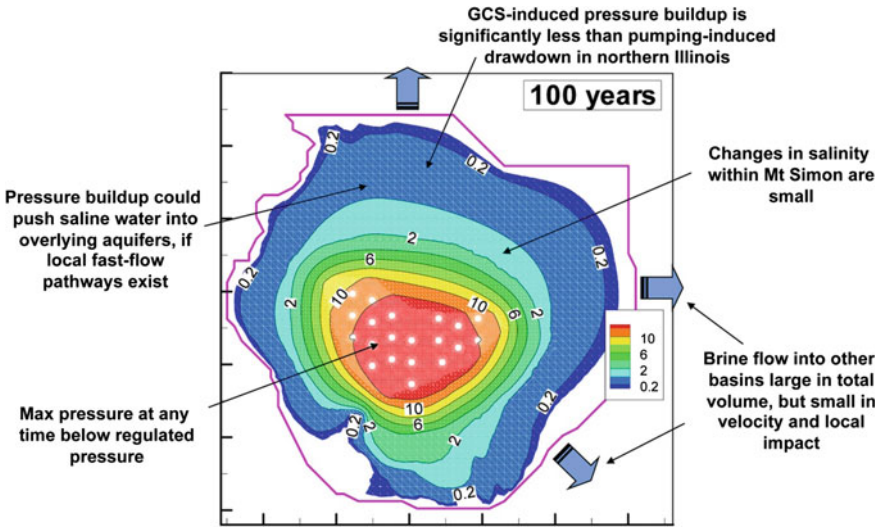


**Fig. 2.5** Profiles of pressure buildup (in MPa) in the Mount Simon Sandstone formation along the east-west direction for two cases of the caprock (Eau Claire) permeability of 1  $\mu\text{D}$  (lower curve) and 0.01  $\mu\text{D}$  (upper curve) at 50 years and 100 years, respectively. Results are for injection of 5 Mt of  $\text{CO}_2$  per year over the first 50 years. From Birkholzer and Zhou (2009)

(b) calculated  $\text{CO}_2$  saturation after 50 years of  $\text{CO}_2$  injection, showing the impact of vertical permeability variation.

Modeling was conducted using a 3D unstructured mesh, with progressive mesh refinement (down to the order of 10 m) in the core injection areas to very large grids (order of 10 km) in the far region, so that both the details of  $\text{CO}_2$  plume multiphase flow and its spatial variability in the near field and brine migration processes in the basin scale are properly simulated. The parallel version of the TOUGH2/ECO2N simulator (Pruess et al. 1999; Zhang et al. 2008; Pruess 2005) was used to solve the multiphase flow and multicomponent transport of  $\text{CO}_2$  and brine in response to  $\text{CO}_2$  injection. In the simulation, full accounts are taken of changes in brine density and viscosity; changes in  $\text{CO}_2$  density and viscosity;  $\text{CO}_2$  solubility in brine; brine solubility in  $\text{CO}_2$ , and their dependence on fluid pressure, temperature, and salinity.

Figure 2.5 shows the calculated pressure rise along one of the transects of the model. The pressure buildup in this large regional system reaches 2.5 MPa in 50 years and will be about 1 MPa after 100 years if  $\text{CO}_2$  injection is terminated after 50 years. Note that the pressure rise is sensitive to the permeability value of



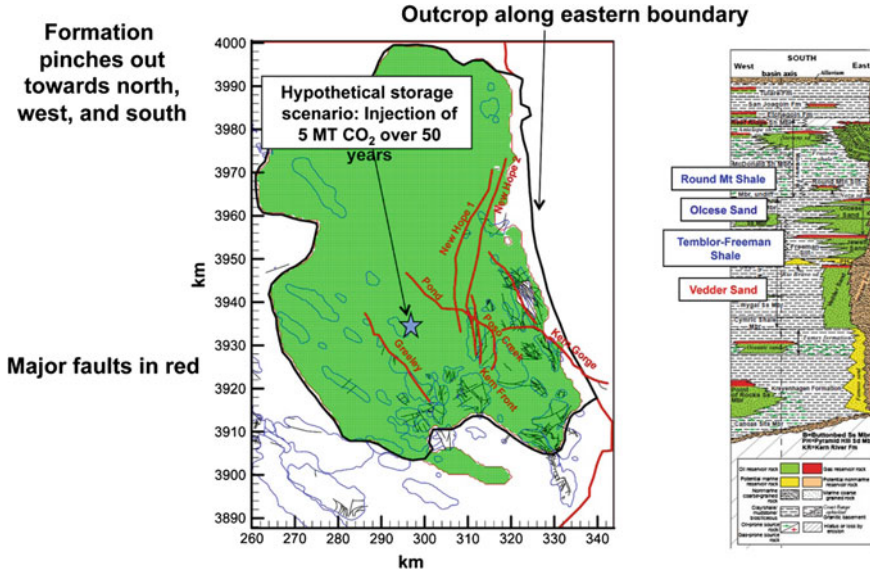
**Fig. 2.6** Conclusions from study of pressure rise in Mount Simon Sandstone due to CO<sub>2</sub> geosequestration. Here the contours are in bars, or 0.1 MPa, and the color area is about 500 km across, with scale tick marks at 100 km intervals (from Birkholzer private communications 2012)

the sealing caprock. Even though there is a significant contrast between the permeability of the Mount Simon Sandstone (about 10–100 mD) and the caprock permeability of 1  $\mu$ D, there is sufficient diffused migration of brine through the caprock to result in a 30–40 % decrease in pressure rise at 100 years, as compared with a zero-permeability caprock case, as represented by the 0.01  $\mu$ D results in the figure.

The conclusions from the Mount Simon study by Birkholzer and Zhou indicate (Fig. 2.6) that the maximum pressure rise at any time in the storage formation is below the regulated limit associated with potential hydrofracturing (Rutqvist et al. 2007). Changes in salinity within the Mount Simon are small, and brine flow into neighboring basins are large in total volume but small in velocity and local impact. However, the pressure buildup could push saline water into overlying conductive formations if fast-flow pathways exist between them.

### 2.10.2 Case of Southern San Joaquin Basin

In contrast to Mount Simon Sandstone in the Illinois Basin, the potential CO<sub>2</sub> storage formation in California, Vedder Sand, in the Southern San Joaquin Basin (Fig. 2.7), has a number of major sealing faults as determined from extensive petroleum exploration studies, so that the formation is partially compartmentalized hydrologically. The formation pinches out towards the south, north, and west.



**Fig. 2.7** On the *left* is the site map of the Vedder Sand formation in the San Joaquin Basin. Major faults are indicated as *red lines*. On the *right* is a vertical profile indicating the caprock, Temblor-Freeman shale separating the storage formation, Vedder Sand from an overlying conductive layer, Olcese Sand. From Zhou and Birkholzer (2011)

To the east, it outcrops along the edge of the Sierra Nevada mountain range. Vertically, the caprock is the Temblor-Freeman shale, which is overlaid by a hydraulically conductive formation called Olcese Sand. In fact, the Vedder Sand and the Olcese Sand connect in the northern area of the domain shown in Fig. 2.7.

Using the same methodology as in the modeling of  $\text{CO}_2$  geosequestration in the Mount Simon Sandstone, the pressure rise was calculated for the Vedder Sand at the Southern San Joaquin Basin. Figure 2.8 shows the pressure rise after 50 years of  $\text{CO}_2$  injection. These results indicate the significant effects of the sealing faults at the site, diffused water migration through the caprock, focused water migration through the caprock pinch-out and water discharge into the outcrop area.

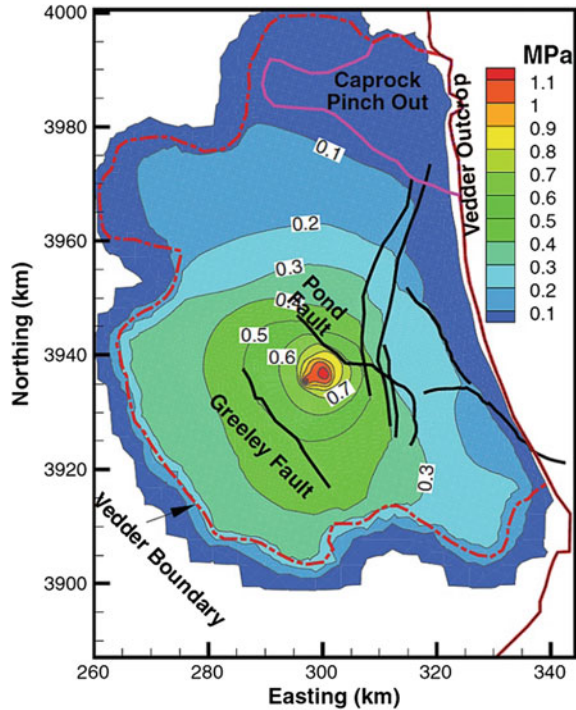
### 2.10.3 Discussion of the Two Cases

In the context of the present chapter on relevant processes for  $\text{CO}_2$  geosequestration, we may summarize the above study (with much more details in the references) as follows.

This study addresses the key question of “storage capacity” as indicated by the maximum pressure rise, which must be kept below a regulatory limit to avoid damage to caprock integrity. It also addresses the key issues of large-scale flow,



**Fig. 2.8** Simulated pressure rise in MPa after 50 years of CO<sub>2</sub> injection into the Vedder Sand formation at the Southern San Joaquin Basin. From Zhou and Birkholzer (2011)



brine displacement, and pressure changes at large scale. Results from this study will be useful as input in addressing the key issues of induced seismicity and focused and diffused migration.

Features in the study include the geometry of the storage formation, the definition of boundary conditions, vertical permeability variations, including the effects of multi-layered caprock, the possibility of caprock pinch-out at the boundary of the storage formation, occurrence of sealing faults, and flow and CO<sub>2</sub> storage compartmentalization.

Processes of relevance in this study include pressure rise due to injection and buoyancy, lateral pressure propagation and brine displacement, diffused or focused vertical brine migration into overlying and underlying formations, two-phase flow and multicomponent transport of CO<sub>2</sub> and brine, changes in brine and CO<sub>2</sub> density and viscosity, CO<sub>2</sub> solubility in brine and (inversely) brine solubility in CO<sub>2</sub> and their dependence on fluid pressure, temperature, and salinity. All these processes have been incorporated into the modeling work for the study of the two sites.

## 2.11 Concluding Remarks

The present chapter presents a framework for discussing the relevant processes in CO<sub>2</sub> geosequestration in saline formations. It is suggested that relevant processes can be usefully discussed in the context of certain key questions associated with CO<sub>2</sub> geosequestration, including the objectives of a site-specific project and key issues of concern. A list of key questions has been identified; features within which the processes occur are also described. Tables are provided for each of the key questions to give a list of relevant features and processes. These tables, however, are not presumed to be complete, and indeed may have to be revised as additional information emerges from further research and experience. Nevertheless, it is hoped that they may serve as a starting point for considering the relevant processes, features, operational strategy, and other factors in addressing key questions related to CO<sub>2</sub> geosequestration at a particular site.

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