Probabilistic Analysis of CO₂ Storage Mechanisms in a CO₂-EOR Field Using Polynomial Chaos Expansion

Wei Jia^{1,2*}, Brian J. McPherson^{1,2}, Feng Pan^{1,2}, Ting Xiao^{1,2}, Grant Bromhal³

¹Department of Civil & Environmental Engineering, the University of Utah, Salt Lake City, UT 84112, USA

²Energy & Geoscience Institute, the University of Utah, Salt Lake City, UT 84108, USA

³National Energy Technology Laboratory, Morgantown, WV 26507, USA

*Corresponding author: Tel: +1 801 651 2837; Fax: +1 801 587 9148; Email address:

wei.jia@utah.edu

Submitted to International Journal of Greenhouse Gas Control

October 2015

1 ABSTRACT

2 Oil fields are already used for storing carbon via CO₂-enhanced-oil-recovery
3 (CO₂-EOR). Such storage is an outcome of CO₂-EOR, albeit not necessarily by design.
4 A next step would be intentional storage via post-EOR CO₂ injection. **Trapping**
5 **mechanisms in such post-EOR operations would include the same mechanisms as in**
6 **CO₂-EOR, including and especially hydrostratigraphic trapping and oil/aqueous**
7 **dissolution.** Forecasting the nature of trapping and the ultimate CO₂ distribution in a
8 reservoir is hindered by uncertainty in reservoir properties. The purpose of this study is to
9 develop and apply reduced order models (ROMs) integrated with Monte Carlo
10 simulations to quantify oil solubility trapping (oil phase), aqueous solubility trapping
11 (aqueous phase), and hydrodynamic trapping (CO₂ in supercritical phase). The case
12 study site for this analysis is the SACROC unit in western Texas. A Polynomial Chaos
13 Expansion (PCE) technique was used to develop the ROMs. The sources of uncertainty
14 considered are porosity and permeability. Model results of interest include dissolved
15 mass of CO₂ in oil phase, mass of CO₂ in supercritical phase, dissolved mass of CO₂ in
16 aqueous phase, and oil saturation in the rock formation. Reduced order models are
17 developed for all cells in five selected layers of the model, which are adjacent to injection
18 wells, at three specific time points of interest, including the end of a simulated CO₂-EOR
19 period, the end of a post-EOR continuous CO₂ injection period, and the end of a post-
20 injection monitoring period. Results of regression fit and validation analysis yield high R²
21 values and low NRMSE values, indicating that the ROMs derived from PCE are capable
22 of meaningful predictions (compared to conventional reservoir models) and effectively
23 represent relationships between model parameters (inputs) and model results (outputs) of

1 interest. Results of 1000 Monte Carlo simulations indicate a dominantly upward transport
2 of CO₂ during injection, driven by buoyancy, and a dominantly downward transport of
3 dissolved CO₂ after injection stops, caused by increases in both oil and brine density. At
4 the end of the 100-year simulation of the SACROC case study, the results forecast that
5 the storage layers of interest store between 54% and 61% of total trapped CO₂ in the
6 entire domain in the oil phase, between 21% and 24% in the supercritical phase, and
7 between 11% and 15% in the aqueous phase. These layers are forecasted to store between
8 89% and 98% of total trapped CO₂ in the field by the end of simulation.

9

10 **Keyword:** CO₂ enhanced oil recovery (CO₂-EOR); CO₂ trapping mechanisms;
11 Uncertainty quantification; Polynomial chaos expansion (PCE)

12

1 INTRODUCTION

Geological CO₂ Sequestration (GCS) is considered a promising approach to reduce CO₂ atmosphere concentration and mitigate associated climate change (Bachu et al., 1994; Metz et al., 2005). Among many potential sequestration targets, CO₂ storage in oil reservoirs is believed to be a very feasible and economic option because of well-characterized geology, existing infrastructure, and economic incentive in the form of incremental oil production. The carbon intensity of using CO₂ for enhanced oil recovery (CO₂-EOR) could be lowered if anthropogenic CO₂ is injected. Moreover, continuous CO₂ injection after CO₂-EOR ceases would definitively provide net positive CO₂ storage in the subsurface.

Many previous studies investigated impacts of CO₂ injection on reservoir performance and oil recovery (e.g., Blunt et al., 1993; DeRuiter et al., 1994; Holm and Josendal, 1974). Few studies focus on CO₂ sequestration in EOR fields. Practical experience suggests approximately 40% of injected CO₂ can be produced and re-injected (Hadlow, 1992; Shaw and Bachu, 2002), and the rest (60%) of CO₂ is permanently stored in the formation. However, there remains much to learn about CO₂ transport and trapping mechanisms. While CO₂ buoyancy-driven transport is addressed in studies of both CO₂ deep saline sequestration and CO₂-EOR (Bryant et al., 2006), CO₂ transport driven by density increases in the oil phase is typically ignored (Ahmed et al., 2012), despite experimental data that suggest its significance (DeRuiter et al., 1994; Grigg, 1995; Lansangan and Smith, 1993). A recent study (Ahmed et al., 2012) indicates a significant impact of oil density changes on forecasted CO₂ spatial transport in synthetic models.

1 Therefore, a secondary but important goal of this analysis is to evaluate specifically the
2 impacts of buoyancy changes associated with oil density increases with CO₂ dissolution.

3 One particular CO₂ trapping mechanisms in oil reservoirs is that CO₂ dissolves in
4 oil (Iglauer, 2011). Han et al. (2010) estimated that oil dissolution trapping accounts for
5 the highest proportion of CO₂ storage in oil reservoir, much more than that trapped as a
6 supercritical phase (hydrodynamic trapping) and as an aqueous phase (aqueous solubility
7 trapping). The primary reason for this disparity is the much higher solubility of CO₂ in
8 oil, but in addition, continuous CO₂ injection may lead to higher dissolution in oil
9 compared to CO₂-WAG (water alternating gas) injection schemes. Continuous CO₂
10 injection (non-WAG) such as evaluated by Han et al.(2010) also neglects much three-
11 phase (gas-oil-water) interaction due to lack of water injection. Therefore, in oil
12 reservoirs that are known to employ WAG but are simulated and analyzed using constant
13 CO₂ injection, it is possible that the amount of CO₂ trapped in the aqueous phase is
14 underestimated and CO₂ trapping in the oil phase is overestimated.

15 Uncertainty quantification of CO₂ storage in oil reservoirs is essential for
16 effective assessment of storage performance and associated risk, and perhaps the greatest
17 source of uncertainty is that associated with heterogeneity of reservoir properties. Monte
18 Carlo simulation is a straightforward method for uncertainty analysis of CO₂
19 sequestration and other subsurface fluid flow processes (Dai et al., 2013; Pan et al., 2009;
20 Ye et al., 2007). However, Monte Carlo methods usually require a very high
21 computational cost (Wriedt et al., 2014; Zhang and Sahinidis, 2012). It is this high cost
22 that has motivated development and application of reduced order models (ROMs), or
23 surrogate models, to replace fully coupled geo-cellular simulators typically used for

1 Monte Carlo simulation. The National Risk Assessment Partnership (NRAP) has made
2 ROMs a priority research objective, and this work is in collaboration with NRAP. In this
3 study, however, the ROMs are not applied to risk assessment, but rather to advanced
4 quantification of storage mechanisms in an active storage site.

5 Response surface methodology (RSM) is a popular method to develop ROMs, and
6 has been applied to risk assessment for CO₂ sequestration research (Dai et al., 2013;
7 Ghomian et al., 2010; Liu and Zhang, 2011; Wriedt et al., 2014). However, application of
8 RSM to heterogeneous models is fundamentally hindered by the required design of
9 experiment (DoE) to select correct samples for generating ROMs. Details of DoE are
10 described elsewhere (Myers et al., 2016).

11 Polynomial Chaos Expansion (PCE) has been widely used to generate ROMs.
12 First introduced by Wiener (1938), the PCE method has been extended and improved to
13 provide high-order predictions with non-linear effects (Fajraoui et al., 2011; Foo and
14 Karniadakis, 2010; Zhang and Lu, 2004; Zhang and Sahinidis, 2012), to accommodate
15 various types of statistical distributions (Askey and Wilson, 1985; Wan and Karniadakis,
16 2006; Xiu and Karniadakis, 2002), and to adapt to arbitrary shapes and statistical
17 moments of probability distributions inferred from sparse data (Oladyshkin et al., 2011b).
18 The PCE approach was recently successfully applied for uncertainty quantification of
19 CO₂ storage in GCS (Oladyshkin et al., 2011a; Oladyshkin et al., 2011b; Oladyshkin et
20 al., 2013; Zhang and Sahinidis, 2012). And, the PCE method does not require strict DoE
21 for developing ROMs, (Zhang and Sahinidis, 2012), nor is it limited to homogeneous
22 models (Oladyshkin et al., 2013).

The specific objectives of this study are (1) to quantify transport of sequestered CO₂ during and after CO₂-EOR and specifically the impacts of CO₂ dissolution on oil density, (2) to quantify uncertainty of CO₂ trapping mechanisms using PCE-generated ROMs, and (3) to re-evaluate dissolution and hydrodynamic CO₂ trapping mechanisms in the SACROC unit using a CO₂-WAG injection scheme instead of constant CO₂ injection, as done in previous studies. For this study, we selected the Scurry Area Canyon Reef Operations Committee (SACROC) unit, undergoing CO₂-EOR since 1972 (Raines and Dobitz, 2001). Its geology and heterogeneity are reported in previous studies (Han et al., 2010; Lucia and Kerans, 2004). This study is the first application of the PCE method to an active CO₂-EOR site using site-specific data.

The rest of this paper is organized as follows: Section 2 describes the SACROC numerical model, the method to generate stochastic realizations for sources of uncertainty, and a detailed description of the PCE approach; Section 3 discusses simulations of CO₂ spatial and temporal transport, and associated forecasted uncertainty of CO₂ trapping among different phases at SACROC. The paper concludes with Section 4, which summarizes the findings of this study and associated recommendations.

2 MATERIAL AND METHODS

2.1 Study Site

The SACROC unit is located within the Permian basin in western Texas (Figure 1). Specifically, SACROC is in the southeast portion of the Horseshoe Atoll, which is a reef mound and consists of mixed types of boundstones and bioclastic debris (Vest Jr, 1970). The carbonate reef complex at SACROC represents the Strawn, Canyon, and

1 Cisco Formations of the Pennsylvanian. Oil is produced from the Canyon and Cisco
2 Formations (Figure 1(b)). The overlying Permian Wolfcamp Shale Formation is
3 designated the primary sealing formation. Detailed geological descriptions of SACROC
4 are reported in previous studies (Han, 2008; Han et al., 2010). In SACROC, the top of
5 Cisco and Canyon Formations is below approximately 1200 m depth, and the bottom is
6 below approximately 1400 m depth (Han, 2008).

7 **2.2 SACROC 3-D Reservoir Model**

8 The Bureau of Economic Geology at the University of Texas at Austin developed
9 a high-resolution geological model, which includes more than 9 million cells. The
10 heterogeneous porosity distribution was derived from 3-D seismic surveys and 368 well
11 logging analyses. The permeability distribution was determined based on porosity using
12 empirical equations for rock fabric classification (Lucia and Kerans, 2004). Han et
13 al.(2010) upscaled the high-resolution model to a coarser resolution using a
14 renormalization technique for reduction of computational cost, but heterogeneity is
15 necessarily smoothed (reduced) by upscaling (Han et al., 2010).. In the same interest of
16 computational cost saving, we used a modified version of the upscaled model from Han
17 et al.(2010). The modification we introduced is a uniform increase of all permeability
18 values 50 times to match the mean of the original high-resolution model; this change
19 appeared to facilitate a better hydrodynamic match of fluid flow with recorded production
20 rates. Grid permeability varies from 1 mD to 4710 mD with an average of 25.48 mD.
21 Average porosity is 0.05 with a range of 0.02 to 0.18.

22 The SACROC model is approximately 8200 m long in west-east direction (x-
23 axis), 4000 m wide in north-south direction (y-axis), and about 249 m thick (z-axis). The

upscaled model contains 13600 cells ($34 \times 16 \times 25$). A constant hydrostatic pressure is specified at the southern boundary to account for the connectivity of the model to the rest of southern reservoir, while no flow boundary condition is assigned to all other boundaries (Han et al., 2010). According to SACROC production history, we adopted the hydrostatic pressure (15.73 MPa to 17.90 MPa) and isothermal (56.78°C) conditions in Han et al. (2010) as the initial conditions of this study. The details of the rock-fluid information and oil composition are referred to Han et al.(2010).

The model domain includes 45 wells, with 23 production wells and 22 injection wells (Han et al., 2010). A CO_2 -WAG injection scheme is used to mimic the real CO_2 -EOR practice. While the CO_2 injection rate and production rate are adopted from Han et al. (2010), various water injection rates at different injection wells are assigned to achieve an approximately 40% CO_2 recycle rate (Hadlow, 1992; Shaw and Bachu, 2002). The simulation consists of three periods: (1) a 30-year CO_2 -EOR period with 1 year of CO_2 injection followed by 1 year of water injection in each CO_2 -WAG cycle; (2) a 20-year post-EOR CO_2 injection period, with all production wells shut in and continuous CO_2 only injection at 50% of CO_2 injection rate during the CO_2 -EOR period to avoid pressure buildup; and (3) a 50-year monitoring period, with all CO_2 injection ceased. All fully coupled reservoir simulations were performed using the CMG-GEM simulator (Computer Modeling Group, 2015).

2.3 Parameter Uncertainty

For sake of brevity, we limited the parameter uncertainty to what we consider the two most important hydrologic parameters, porosity and permeability. The high-resolution model includes a detailed distribution of porosity and permeability.

We generated synthetic well logs for each of the 45 wells within the model domain, based on the updated upscaled porosity and permeability distributions. We then estimated the variograms of porosity and permeability from the synthetic well logs. We found no trend (with respect to depth) in either the porosity or the permeability well logs, directing us to use the well logs directly in variogram analysis. Table 1 summarizes variogram parameters for porosity and permeability well logs. Based on the variogram models, 1000 random realizations were created for each parameter (porosity and permeability) via sequential Gaussian simulation.

2.4 Polynomial Chaos Expansion (PCE) and Monte Carlo Simulation

Generally, the two types of PCE methods are intrusive and non-intrusive. The former inserts PCE approximations into the governing equations, while the latter only requires a small number of model simulations to solve for expansion coefficients. Due to the complexity of subsurface CO₂ sequestration multiphase processes, we use the non-intrusive PCE in this study.

We assume a physical (geo-cellular) model $y = Model(x)$, where $x = (x_1, x_2, \dots, x_M)^T$ is a vector of model input parameters, and $y = (y_1, y_2, \dots, y_N)^T$ is a vector of model outputs. If the vector x is uncertain, then an element y_i in the vector y can be represented by a polynomial chaos expansion (Wiener, 1938):

$$y_i = M(x) = \alpha_0 B_0 + \sum_{j=1}^M \alpha_j B_1(x_j) + \sum_{j=1}^M \sum_{k=1}^j \alpha_{jk} B_2(x_j, x_k) + \sum_{j=1}^M \sum_{k=1}^j \sum_{h=1}^k \alpha_{jkh} B_3(x_j, x_k, x_h) \dots, \quad \text{Eq. (1)}$$

where α are coefficients and B are multivariate polynomial basis functions. For normally distributed parameters, Hermite polynomials are employed to generate basis functions. When y_i is obtained from geo-cellular model simulations, the coefficients can be determined by solving $\alpha = (B^T B)^{-1} B^T y_i$, provided that $(B^T B)^{-1}$ exists. Then the calculated coefficients are substituted into Eq. (1) to build the ROM. A PCE can also be applied to parameters with other PDFs. The Askey scheme and generalized options (Askey and Wilson, 1985; Xiu and Karniadakis, 2002) extend PCE applicability to the most commonly known PDFs, such as Gamma, Beta, and uniform distributions. A recent generalization of PCE, the arbitrary polynomial chaos (aPC) expansion, makes better use of available data by adapting arbitrary probability distribution shapes of input parameters. Moreover, it also works for very limited data sets, which may be too sparse to develop any distribution shapes (Oladyshkin et al., 2011a).

A PCE is capable of rendering ROMs with higher orders (greater than 2), as long as appropriate polynomial basis functions are provided. Therefore, more accurate ROMs may be developed with proper goodness-of-fit targets, such as coefficient of determination (R^2) and leave-one-out cross validation error (Q^2) (Zhang and Sahinidis, 2012).

A small number (N_p) of model simulations are required to develop the ROMs. Hosder et al. (2007) recommend that N_p be twice of the number of the terms (N_t) in the expansion. Accordingly, N_t may be calculated as

$$N_t = \frac{(M+d)!}{M!d!}, \quad \text{Eq. (2)}$$

where M is the number of model input parameters, and d is the highest degree of the polynomial. For the two model parameters or inputs of interest, porosity and

1 permeability, assuming the highest degree d is 5, N_t is 21 from Eq. (2). Therefore, we
2 randomly selected 50 (N_p) realizations from the previously generated 1000 realizations
3 and simulated these with the CMG-GEM simulator to develop the ROMs.

4 The orthogonality of polynomial basis functions requires that model inputs are
5 independent of each other. Permeability in the high-resolution model is derived from
6 porosity; however, the correlation is weakened by the process of upscaling. Figure 2
7 shows the joint distribution and marginal PDFs for porosity and log-permeability for a
8 selected cell. It is clear that both distributions are Gaussian, and no obvious correlation is
9 exhibited between the two input parameters. Very weak correlation (<0.2) is observed
10 between the 1000 porosity values and the 1000 log-permeability values for all cells.
11 Moreover, we confirmed that both parameters are normally distributed with the
12 Anderson-Darling test (Anderson and Darling, 1954). Therefore, Hermite polynomial
13 basis functions are the optimum option for the PCE application in this study.

14 While analyzing the upscaled distributions of porosity and permeability, we
15 found that the distributions are close to uniform in both the upper layers (Layer 1 to
16 Layer 17) and bottom layers (Layer 23 to Layer 25), as shown in Figure 3. **Porosity and**
17 **permeability values in the upper and bottom layers are extremely low.** Han et al. (2010)
18 suggests that CO₂ upward transport is limited to Layer 18 and CO₂ downward transport
19 does not reach Layer 23 after 200 years of simulation. Simulation results of this study
20 confirms that majority of injected CO₂ is restricted between Layer 18 and Layer 22
21 (figures not shown). Therefore, we decided to focus on the five layers adjacent to the
22 injection wells, which are Layer 18, Layer 19, Layer 20, Layer 21, and Layer 22. **Total**

thickness of these five layers is about 40 m, with each layer having an average thickness of 8 m. The PCE will be applied only to cells in these layers to build the ROMs.

In this study we selected four specific model results or outputs of interest, including mass of CO₂ dissolved in the oil phase, mass of CO₂ in supercritical phase, mass of CO₂ dissolved in the aqueous phase, and oil saturation. Due to the relatively nominal amounts of residually trapped CO₂ and mineral trapped CO₂ (less than 1% and 0.1% of total sequestered CO₂, respectively) in a 200-year simulation (Han, 2008), we deliberately neglect these two trapping mechanisms in this study.

A total number of 1000 Monte Carlo simulations were conducted to evaluate the four model outputs of interest using ROMs for all cells in the five selected layers at three time points, Time 1 (end of the 30-year CO₂-EOR period), Time 2 (end of the 20-year post-EOR continuous CO₂ injection period), and Time 3 (end of the 50-year monitoring period). Outliers, including negative values and extremely high values that result in unreasonable CO₂ density at reservoir conditions (higher than 843 kg/m³) were culled.

3 RESULTS

3.1 Quality of ROMs

Although a target R² value was assigned in the PCE algorithms to develop ROMs, it is necessary to check the overall accuracy of the regression. Besides R² values, NRMSE (Normalized Root Mean Square Error) values were used to assess the quality of resulting ROMs. ROM-predicted values of four model outputs, mass of CO₂ dissolved in the oil phase, mass of CO₂ in supercritical phase, mass of CO₂ dissolved in the aqueous phase, and oil saturation, were compared to corresponding values obtained from the 50

simulations used to develop the ROMs. Because each cell is assigned its own ROM, we decided to superimpose and compare all ROM-predicted values to all original (full reservoir model) simulated values, i.e., combined plots of all values for all cells (Figure 4), because we rely on the overall quality of all ROMs for this study. Figure 4 presents comparison plots for the four model outputs of interest at Time 3 (end of the 50-year monitoring period). The R^2 values for mass of CO_2 dissolved in the oil phase, mass of CO_2 in supercritical phase, mass of CO_2 dissolved in the aqueous phase, and oil saturation are 0.9856, 0.9734, 0.9883, and 0.9139, respectively. The high R^2 values suggest a robust linear relationship between predicted and simulated values. For oil saturation, the data trend for lower saturation (0 to 0.5) deviates from a linear fit; however, the majority of data in the upper right of the plot dominate data fitting. The NRMSE values are 1.67% for mass of CO_2 dissolved in the oil phase, 2.58% for mass of CO_2 in supercritical phase, 0.97% for mass of CO_2 dissolved in the aqueous phase, and 6.82% for oil saturation. These values yield small residuals between predicted and simulated values. Table 2 summarizes R^2 and NRMSE values at Time 1 (end of the 30-year CO_2 -EOR period) and Time 2 (end of the 20-year post-EOR continuous CO_2 injection period). Similar to the comparison results at Time 3 (end of the 50-year monitoring period), the high R^2 values and low NRMSE values indicate accurate predictions from ROMs at both Time 1 and Time 2.

Before being used for Monte Carlo simulations, PCE-generated ROMs must be validated. Therefore, 100 realizations, which are different from the previous 50 realizations, were randomly selected from the 1000 realization pool and simulated with the CMG-GEM simulator. The 100 realizations were also calculated with the ROMs.

Predictions using ROMs were compared to the CMG-GEM simulated values. Table 3 summarizes R^2 and NRMSE values for the four model outputs of interest at the three time points (Time 1, Time 2, Time 3, as defined previously). All R^2 values are greater than 0.8, with most values higher than 0.9, suggesting a meaningful linear fit between predicted values and simulated values. All ROMs for CO₂ mass in the three phases of interest exhibit NRMSE values less than 7%, and ROMs for oil saturation yield NRMSE values ranging from 8.42% to 8.90%. The validation effort produced lower R^2 values and higher NRMSE values than previous values (Table 2). Slight deterioration of goodness-of-fit measurements in the ROMs validation effort was also observed in a previous study (Wriedt et al., 2014). Nevertheless, this validation exercise yielded what appear to be fairly robust predictions from ROMs for the 100 random realizations.

3.2 Probabilistic Analysis of CO₂ Trapping Mechanisms

Cumulative distribution functions for CO₂ storage in the sub-domain (Layer 18 to Layer 22) were plotted in Figure 5 to illustrate storage in each phase at each time point (Time 1, Time 2, Time 3). The mass of CO₂ dissolved in oil at Time 1 ranges from 1.63 million tonnes (P5) to 1.92 million tonnes (P95). CO₂ storage at Time 2 increases dramatically due simply to continuous CO₂ injection, ranging from 2.53 million tonnes (P5) to 2.88 million tonnes (P95). By Time 3, the final value of CO₂ storage in oil varies from 2.74 million tonnes (P5) to 3.10 million tonnes (P95).

Less CO₂ was retained in supercritical phase at all three time points. Smaller and negative increments of supercritical CO₂ storage are observed at Time 2 and Time 3, respectively. It is also interesting to note that the range of P45 to P99 at Time 1 overlaps with the range of P1 to P83 at Time 2, indicating that the 20-year post-EOR CO₂ injection

1 may cause small or even negative increments of supercritical CO₂ storage in all five
2 layers next to the injection well.

3 The least amount of CO₂ was stored in the aqueous phase simply because it has a
4 smaller storage capacity compared to the other two phases. Resulting CDF plots for CO₂
5 mass stored in the aqueous phase (Figure 5) at the three time points of interest overlap
6 significantly, suggesting that storage of CO₂ in the aqueous phase is somewhat
7 irrespective of the time stage of the simulation.

8 The relative storage by each phase is analyzed by calculating the percentage of CO₂
9 storage in each phase in the five layers (next to the injection wells) to the overall CO₂
10 storage amount in the entire domain (3.66 million tonnes at Time 1, 5.06 million tonnes
11 at Time 2 and Time 3). The results in Table 4 indicate that the oil phase retains most of
12 CO₂, while the aqueous phase has the least amount of storage. Han et al. (2010) also
13 concluded that the oil phase stores most CO₂ (about 72%) by the end of a 200-year
14 simulation, followed by mobile CO₂ (supercritical phase) and aqueous CO₂ (aqueous
15 phase). Oil phase storage monotonically increases over time. Correspondingly, the
16 supercritical phase and aqueous phase storage monotonically decrease over time. In the
17 long term, sequestered CO₂ tends to move from supercritical phase and aqueous phase to
18 oil phase. Consequently, risk of CO₂ leakage is reduced due to the relatively high
19 viscosity of oil compared to brine and supercritical CO₂.

20 **3.3 Spatial and Temporal distribution of CO₂ in Oil Phase**

21 CO₂ storage in the oil phase was studied for five layers at the three time points
22 (Time 1, Time 2, Time 3, as defined previously). Figure 6 depicts results of 1000 Monte
23 Carlo simulations using the ROMs. For each box on Figure 6, the central mark indicates

1 the median; the upper and lower edges represent 75th and 25th percentiles respectively;
2 and the whiskers extend to 1.5 interquartile range.

3 At Time 1, Layer 19 has the most CO₂ storage in oil phase with a median value of
4 about 0.46 million tonnes. Layer 20, where most of wells inject or produce from the
5 reservoir, has the second largest extent of CO₂ storage at about 0.42 million tonnes. This
6 implies that injected CO₂ moves upward from the injection layer to overlying layers
7 within the 30 years of CO₂ -EOR injection. However, vertical transport of CO₂ is
8 restricted, exhibited by the significant difference between CO₂ stored in Layer 18
9 (median value 0.18 million tonnes) and in Layer 19 (median value 0.46 million tonnes).
10 This result is consistent with the previous SACROC CO₂ trapping analysis (Han et al.,
11 2010). While the permeability of Layer 18 is similar with the one of Layer 19 (as shown
12 in Figure 3), vertical transport retardation is due to the presence of the oil phase and low
13 relative permeability of supercritical CO₂. It is found that the oil phase has significantly
14 higher relative permeability than supercritical CO₂ in most portion of Layer 18 (figure
15 not shown). Therefore, relatively small amount of supercritical CO₂ could enter Layer 18
16 and dissolve in the oil phase.

17 At Time 2, CO₂ storage in oil has increased compared to that for Time 1. The
18 overall distribution of CO₂ in the five layers adjacent to the injection wells (Layer 18,
19 Layer 19, Layer 20, Layer 21, and Layer 22) is similar to that for Time 1. Layer 19
20 contains the greatest amount of CO₂, and has the largest CO₂ storage increase at about
21 0.29 million tonnes. The least amount of CO₂ storage increase is about 0.11 million
22 tonnes, in Layer 18. This result again suggests that supercritical CO₂ upward transport
23 from Layer 19 to the overlying layers is impeded.

After 50 years with no well operations, the amount of CO₂ stored in the oil phase within the five layers has slightly increased at Time 3. The relative amounts of storage among layers for Time 3 is identical to that in Time 1 and Time 2; specifically, Layer 19 contains the most CO₂, followed by Layer 20, Layer 21, Layer 22, and then Layer 18. There are CO₂ injection wells in Layer 21 and Layer 22; however, the injected amounts through these wells were insufficient to yield the observed CO₂ storage within these layers. For example, injection well I22 has injected 0.11 million tonnes of CO₂ into Layer 21 by Time 1, which is only about 1/3 of stored CO₂ in oil phase in this layer. It is similar in Layer 22, where cumulative CO₂ injection by Time 1 is 0.22 million tonnes. The extra amount of CO₂ in these two layers must come from other layers, such as Layer 20, into which most CO₂ was injected. It is interesting to note that Layer 21 receives more CO₂ (220000 tonnes) than Layer 22 (55000 tonnes), which also indicates that the origin of additional CO₂ is the overlying layers (Layer 20 and above) instead of the underlying layers (Layer 23 and below).

The downward transport of CO₂ in the oil phase is more evident by comparing results for Time 2 and Time 3 (Figure 6). An increase of CO₂ storage is observed in all layers, while there is no ongoing CO₂ injection in the entire domain. Moreover, evolution of oil saturation demonstrates the oil “sink” over time.

Figure 7 presents the mean of oil saturation at a vertical slice ($j=7$) at Time 1 (top), Time 2 (middle), and Time 3 (bottom) from Monte Carlo simulations. After 30 years of production, low oil saturation values are exhibited around Layer 20. At Time 2, oil saturation is starkly increased in Layer 18, Layer 19, and Layer 20. However, the low oil saturation at the lower right corner (Figure 7) has not similarly increased due to

1 inflow. The low oil saturation region continues moving downward, and almost moves
2 outside of Layer 22 at Time 3. Meanwhile, the front of high oil saturation on the left side
3 (Figure 7) extends from Layer 18 at Time 1 to Layer 20 at Time 2, and to Layer 22 at
4 Time 3, suggesting that oil influent is from the upper layers. It should be noted that while
5 the transport of supercritical CO₂ from lower to upper layers is restricted by its low
6 relative permeability, oil has much higher mobility to sink from upper layers to lower
7 layers.

8 The simulated downward oil movement is attributed to an increase in oil density
9 due to CO₂ dissolution. Previous experiments indicate that as CO₂ concentration in oil
10 increases, oil density increases because of enhanced intermolecular Columbic interactions
11 between CO₂ and hydrocarbon molecules (Lansangan and Smith, 1993). Density
12 increases caused by increasing CO₂ concentration has also been observed in oil samples
13 (Bennion, 1993; DeRuiter et al., 1994). More specifically, a 2% oil density increase was
14 measured for west Texas crude oil (Grigg, 1995) when CO₂ was added to oil samples.
15 However, few simulation studies include density effect of CO₂ dissolution in oil; Ahmed
16 et al. (2012) reported an 11.7% underestimation of oil density if the density effect is
17 ignored (for specific conditions of 11.7 MPa and 46.7°C).

18 Simulation results illustrate that injected supercritical CO₂ moves upward due to
19 buoyancy, and dissolves into the oil phase within the upper layers as shown in the top and
20 middle plots of Figure 8 ($x < 5000$ m, and $x > 10000$ m). As CO₂ concentration increases,
21 oil in upper layers becomes denser and moves downward. Comparing Figures 7 and 8,
22 transport of CO₂ in the oil phase is in accordance with oil movement. Regions storing less
23 CO₂ in the oil phase (between 6000 m and 8000 m) have relatively smaller oil saturation

increases, while regions storing more CO₂ (between 2000 m and 4000 m, and between 10000 m and 15000 m) reflect more significant oil saturation increases over time.

3.4 Spatial and Temporal distribution of CO₂ in Supercritical Phase

Monte Carlo simulation results of the amount of CO₂ stored as supercritical phase are presented in Figure 9. By the end of the CO₂ -EOR period (Time 1), Layer 19 contains the most supercritical CO₂ (median: 0.30 million tonnes) relative to other layers. Median values of the three underlying layers, Layer 20, Layer 21, and Layer 22, are 0.27 million tonnes, 0.21 million tonnes, and 0.14 million tonnes, respectively. Similar to CO₂ dissolved in oil, Layer 18 stores the least amount of CO₂ as supercritical, at about 83000 tonnes. The range of Monte Carlo simulation results, i.e. the distance between whiskers, follows the same order, that Layer 19 has the greatest uncertainty while Layer 18 has the least uncertainty.

After 20 years of continuous CO₂ injection, storage of CO₂ as a supercritical phase increases in all layers except Layer 22. It is the first time in the overall simulation time period that Layer 18 stores more CO₂ than another layer. The largest increase happens within Layer 19, which stores about 72000 tonnes of CO₂ during the 20 years. The main injection layer, Layer 20, has retained about 53000 tonnes of injected CO₂ in supercritical phase. Driven by buoyancy, 52000 tonnes CO₂ moved upward into Layer 18. Compared with the upper layers, lower layers have little or negative CO₂ accumulation. While Layer 21 has about 15000 tonnes of CO₂ stored within the post-EOR CO₂ injection period, Layer 22 has lost 5000 tonnes of CO₂ that had been sequestered as supercritical CO₂.

By the end of the simulation, the amount of CO₂ in the supercritical phase has decreased in all layers except Layer 18, where about 46000 tonnes of CO₂ was stored in

1 supercritical phase during 50 years. The other four layers have lost a similar amount
2 (~25000 tonnes) of CO₂ that was retained as supercritical. A simple mass balance
3 calculation suggests that more than 50000 tonnes of previously sequestered CO₂
4 partitioned from the supercritical phase and dissolved into other phases, i.e., the oil phase
5 or aqueous phase.

6 Supercritical CO₂ is only observed within the model grid where oil and brine are
7 fully saturated with CO₂ (Figure 10). When oil from the upper layers moves into lower
8 layers, supercritical CO₂ dissolves into oil phase until saturation is reached. The decrease
9 of supercritical CO₂ over time is marked (Figure 10), especially for two regions (between
10 2000 m and 4000 m, and between 10000 m and 15000 m) where a significant increase of
11 oil saturation and CO₂ dissolved in oil is exhibited (Figures 7 and 8).

12 **3.5 Spatial and Temporal distribution of CO₂ in Aqueous Phase**

13 Monte Carlo simulations of the amount of CO₂ in all five layers next to the
14 injection well at Time 1, Time 2 and Time 3 illustrate clear patterns. At Time 1, while
15 Layer 21 has the most amount of CO₂ in the aqueous phase (median value: 0.15 million
16 tonnes), Layer 19, which contains the highest CO₂ mass (Figures 6 and 9), retains 0.13
17 million tonnes of CO₂ in the aqueous phase and also falls behind Layer 20 (0.14 million
18 tonnes). Layer 18 still contains the least amount of CO₂ at about 60000 tonnes. All five
19 layers have relatively smaller amounts of CO₂ stored in the aqueous phase compared to
20 that in the oil phase or supercritical phase.

21 The amount of CO₂ sequestered in the aqueous phase increases in all layers at
22 Time 2 due to continuous CO₂ injection. While Layer 21 still retains the largest amount
23 of CO₂, Layer 22 has the greatest increase at about 21000 tonnes, which is almost twice

the increase in Layer 21. Almost 18,000 tonnes CO₂ is stored in Layer 18 between Time 1 and Time 2, which is attributed to dissolution of supercritical-phase CO₂.

Compared to Time 2, almost all layers experienced reduction of CO₂ in the aqueous phase by Time 3. Layer 18 was the sole exception, and exhibited a slight increase (to about 5000 tonnes). The mean of Monte Carlo simulations for aqueous phase CO₂ in a vertical slice (j=7) is illustrated in Figure 12 (see also Figures 7, 8, and 10 for other results corresponding to this slice). At Time 3, CO₂ mass dissolved in the aqueous phase decreased within those five layers of interest next to the injection well. We speculated that perhaps the CO₂-laden brine moved to deeper layers due to its higher density (Li and Firoozabadi, 2009), but further analysis of the simulation results confirmed that dissolved CO₂ exsolved from the aqueous phase and dissolved into the oil phase, as exhibited by Figures 8 and 12 (between 2000 m and 4000 m, and between 10000 m and 15000 m). It is well established that CO₂ solubility in oil is much higher than CO₂ solubility in water (DeRuiter et al., 1994; Firoozabadi and Cheng, 2010; Iglauer, 2011).

3.6 Probabilistic Analysis of CO₂ Storage

Total amounts of CO₂ storage in the 5 layers are listed in Table 5, as well as ratios of those amounts to amounts of CO₂ storage in the entire domain. By the end of CO₂ - EOR period, 86.65% to 98.76% of total CO₂ was retained in the five layers. There are two possible reasons for the high percentage, restricted upward transport and production activities, both of which help injected CO₂ to stay close to injection layers. Although the amount of CO₂ storage in the five layers increases after 20 years of CO₂ injection, the storage percentage decreases slightly. Due to continuous CO₂ injection and shut in of production wells, a small portion of supercritical phase CO₂ moves upward into upper

1 layers (Layer 17 and above). While some CO₂ dissolves in oil and brine, the rest stays in
2 supercritical phase. Then downward oil transport caused by its higher density leads to an
3 increase of total storage percentage in the five layers. At the end of the 100-year
4 simulation, 88.71% to 97.99% of total stored CO₂ is retained in the five layers. As time
5 increases, the percentage is likely to grow slightly and then level off due to gradually
6 attenuation of downward oil transport from upper layers. It is also possible to see a
7 decrease of the percentage at an even larger time scale, as CO₂ dissolved oil continues to
8 move down to the bottom of the domain (or close to the water-oil contact), which is
9 favorable for long-term CO₂ sequestration.

10 While most of injected CO₂ is sequestered in the five layers, significant amount of
11 CO₂ transports to other layers in the model domain. Table 5 suggests that the other layers
12 store 1.24% to 13.35% of total stored CO₂ in the entire model domain at Time 1, 4.99%
13 to 14.05% of entire CO₂ storage at Time 2, and 2.01% to 11.29% at Time 3. Since the
14 1000 Monte Carlo simulations were conducted for only the five layers, it is difficult for a
15 direct comparison of CO₂ storage in the three subdomains, which are the upper layers
16 (Layer 17 and above), the five layers, and the bottom layers (Layer 23 and below).
17 However, simulation results of the 50 realizations, which were used to build ROMs,
18 could be used to investigate CO₂ storage in the upper and bottom layers. Table 6 presents
19 storage percentages of three subdomains derived from average of the 50 simulation
20 results. Percentages of CO₂ storage in the five layers (Layer 18 to Layer 22) at three time
21 points are within the predicted range (P5 to P95) shown in Table 5. The bottom layers
22 always sequester more CO₂ than the upper layers. At the end of simulation, besides the
23 92.47% of CO₂ (or 4.68 million tonnes) that is sequestered in the five layers, 5.27% of

total CO₂ (or 26700 tonnes) is in the bottom layers, and only 2.27% of total CO₂ (or 11486 tonnes) is in the upper layers. The bottom layers are very likely to store more CO₂ in a larger time frame because of the downward transport of CO₂-dissolved oil.

Density increase in oil caused by CO₂ dissolution has been shown as a favorable factor for CO₂ storage in oil reservoirs. In this study, injectors are located at lower layers. To maximize CO₂ storage capacity, CO₂ could be injected into upper layers at appropriate permeability and porosity conditions to take advantage of the adverse density gradient (Kim, 2014). However, it should be noted that reservoir heterogeneity has important impact on fluid flow.

4 CONCLUSIONS

The PCE method is integrated with Monte Carlo simulations in this study to quantify uncertainty of CO₂ storage in different phases in the SACROC unit. ROMs were developed for cells in five selected layers, which are adjacent to injection wells, to represent the relationship between model input variables (porosity and permeability) and model outputs of interest (mass of CO₂ in three phases and oil saturation). ROMs were generated for three time points, including end of the 30-year CO₂-EOR period, end of the 20-year post-EOR continuous CO₂ injection period, and end of the 50-year monitoring period. Goodness-of-fit measurements show accuracy of ROMs in both regression fit of 50 random realizations and validation of 100 random realizations. A total number of 1000 Monte Carlo simulations were conducted via ROMs derived from PCE. The main conclusions are drawn as follows.

- (1) During CO₂ injection, majority of injected CO₂ moves upward to Layer 19, which is immediately above the major injection layer (Layer 20). The buoyancy-driven

1 upward transport causes that Layer 19 contains the greatest proportion of CO₂ in
2 oil phase and supercritical phase. However, the upward transport is restricted in
3 Layer 19, suggested by the significant difference of trapped CO₂ between Layer
4 18 and Layer 19.

5 (2) After CO₂ injection stops, there is a dominant downward transport of dissolved
6 CO₂ in oil phase. It is believed to be associated with oil density increase due to
7 CO₂ dissolution. The density increase of oil results in a reverse density gradient
8 and consequently downward transport of heavier oil. Due to the high CO₂
9 solubility in oil and the high oil saturation, sequestered CO₂ partitions from
10 supercritical phase and dissolved aqueous phase and dissolves into oil. Therefore,
11 amount of CO₂ trapped in oil phase monotonically increases over time, while
12 amount of CO₂ trapped in supercritical phase and aqueous phase monotonically
13 decreases over time.

14 (3) Hydrodynamic trapping and aqueous solubility trapping are more important at
15 Time 1 (end of the CO₂-EOR period) than at other time points (Time 2 and Time
16 3). Oil solubility trapping is more significant at Time 3 (end of the 50-year
17 monitoring period) than at earlier time points (Time 1 and Time 2).

18 (4) Oil solubility trapping accounts for the largest proportion of CO₂ storage at three
19 selected time points, followed by hydrodynamic trapping, and aqueous solubility
20 trapping. While the relative importance of each trapping mechanism follows
21 conclusion of previous study using constant CO₂ injection, the CO₂-WAG
22 injection scheme yields more CO₂ trapped in aqueous phase and less CO₂ trapped
23 in oil phase.

1 (5) At the end of the 100-year simulation, the selected five layers store between
2 88.71% and 97.99% of CO₂ in the entire domain. Due to the downward transport
3 of trapped CO₂, the selected five layers are expected to store more CO₂ in the
4 entire domain in the long term. Given the overlaying Wolfcamp Shale Formations
5 as a seal layer, a shallower injection layer is recommended for the SACROC unit
6 during post-EOR CO₂ injection period to maximize CO₂ storage.

7 **ACKNOWLEDGMENT**

8 This work has been supported by the Phase III Southwest Partnership on Carbon
9 Sequestration (SWP) funded by the U.S. Department of Energy through the National
10 Energy Technology Laboratory.

11 **REFERENCES**

- 12 Ahmed, T., Nasrabadi, H., Firoozabadi, A., 2012. Complex flow and composition path in
13 CO₂ injection schemes from density effects. *Energy & Fuels* 26, 4590-4598.
- 14 Anderson, T.W., Darling, D.A., 1954. A test of goodness of fit. *Journal of the American*
15 *Statistical Association* 49, 765-769.
- 16 Askey, R., Wilson, J.A., 1985. Some basic hypergeometric orthogonal polynomials that
17 generalize Jacobi polynomials. *American Mathematical Soc.*
- 18 Bachu, S., Gunter, W., Perkins, E., 1994. Aquifer disposal of CO₂: hydrodynamic and
19 mineral trapping. *Energy Conversion and Management* 35, 269-279.
- 20 Bennion, D.B., 1993. The use of carbon dioxide as an enhanced recovery agent for
21 increasing heavy oil production, Paper for Presentation at the Joint
22 Canada/Romania Heavy Oil Symposium, March, pp. 7-13.
- 23 Blunt, M., Fayers, F.J., Orr, F.M., 1993. Carbon dioxide in enhanced oil recovery. *Energy*
24 *Conversion and Management* 34, 1197-1204.
- 25 Bryant, S.L., Lakshminarasimhan, S., Pope, G.A., 2006. Buoyancy-dominated multiphase
26 flow and its impact on geological sequestration of CO₂, SPE/DOE Symposium on
27 Improved Oil Recovery. Society of Petroleum Engineers.
- 28 Computer Modeling Group, 2015. GEM user guide, compositional & unconventional
29 reservoir simulator (version 2015).

1 Dai, Z., Middleton, R., Viswanathan, H., Fessenden-Rahn, J., Bauman, J., Pawar, R., Lee,
2 S.-Y., McPherson, B., 2013. An integrated framework for optimizing CO₂
3 sequestration and enhanced oil recovery. *Environmental Science & Technology*
4 *Letters* 1, 49-54.

5 DeRuiter, R., Nash, L., Singletary, M., 1994. Solubility and displacement behavior of a
6 viscous crude with CO₂ and hydrocarbon gases. *SPE Reservoir Engineering* 9,
7 101-106.

8 Fajraoui, N., Ramasomanana, F., Younes, A., Mara, T.A., Ackerer, P., Guadagnini, A.,
9 2011. Use of global sensitivity analysis and polynomial chaos expansion for
10 interpretation of nonreactive transport experiments in laboratory - scale porous
11 media. *Water Resources Research* 47.

12 Firoozabadi, A., Cheng, P., 2010. Prospects for subsurface CO₂ sequestration. *AIChE*
13 *journal* 56, 1398-1405.

14 Foo, J., Karniadakis, G.E., 2010. Multi-element probabilistic collocation method in high
15 dimensions. *Journal of Computational Physics* 229, 1536-1557.

16 Ghomian, Y., Sepehrnoori, K., Pope, G.A., 2010. Efficient investigation of uncertainties
17 in flood design parameters for coupled CO₂ sequestration and enhanced oil
18 recovery, *SPE International Conference on CO₂ Capture, Storage, and Utilization*.
19 *Society of Petroleum Engineers*.

20 Grigg, R.B., 1995. Dynamic phase composition, density, and viscosity measurements
21 during CO₂ displacement of reservoir oil, *SPE International Symposium on*
22 *Oilfield Chemistry*. *Society of Petroleum Engineers*.

23 Hadlow, R., 1992. Update of industry experience with CO₂ injection, *SPE annual*
24 *technical conference and exhibition*. *Society of Petroleum Engineers*.

25 Han, W.S., 2008. Evaluation of CO₂ trapping mechanisms at the SACROC northern
26 platform: site of 35 years of CO₂ injection. *New Mexico Institute of Mining and*
27 *Technology, Socorro, New Mexico*, p. 465.

28 Han, W.S., McPherson, B.J., Lichtner, P.C., Wang, F.P., 2010. Evaluation of trapping
29 mechanisms in geologic CO₂ sequestration: Case study of SACROC northern
30 platform, a 35-year CO₂ injection site. *American Journal of Science* 310, 282-324.

31 Holm, L., Josendal, V., 1974. Mechanisms of oil displacement by carbon dioxide. *Journal*
32 *of petroleum Technology* 26, 1,427-421,438.

33 Hosder, S., Walters, R.W., Balch, M., 2007. Efficient sampling for non-intrusive
34 polynomial chaos applications with multiple uncertain input variables,
35 *Proceedings of the 48th AIAA/ASME/ASCE/AHS/ASC Structures, Structural*
36 *Dynamics and Materials Conference*, *AIAA paper*.

37 Iglauer, S., 2011. Dissolution trapping of carbon dioxide in reservoir formation brine-a
38 carbon storage mechanism. *INTECH Open Access Publisher*.

39 Kim, M.C.K., 2014. Onset of buoyancy-driven convection in a variable viscosity liquid
40 saturated in a porous medium. *Chemical Engineering Science* 113, 77-87.

- 1 Lansangan, R., Smith, J., 1993. Viscosity, density, and composition measurements of
2 CO₂/west Texas oil systems. SPE reservoir engineering 8, 175-182.
- 3 Li, Z., Firoozabadi, A., 2009. Cubic plus association equation of state for water -
4 containing mixtures: Is “cross association” necessary? AIChE journal 55, 1803-
5 1813.
- 6 Liu, B., Zhang, Y., 2011. CO₂ modeling in a deep saline aquifer: a predictive uncertainty
7 analysis using design of experiment. Environmental science & technology 45,
8 3504-3510.
- 9 Lucia, F.J., Kerans, C., 2004. Permeability estimation using porosity logs and rock fabric
10 stratigraphy: an example from the SACROC (Pennsylvanian) field, Scurry
11 County, Texas. PUBLICATIONS-WEST TEXAS GEOLOGICAL SOCIETY 4,
12 271.
- 13 Metz, B., Davidson, O., De Coninck, H., Loos, M., Meyer, L., 2005. IPCC special report
14 on carbon dioxide capture and storage. Prepared by Working Group III of the
15 Intergovernmental Panel on Climate Change. IPCC, Cambridge University Press:
16 Cambridge, United Kingdom and New York, USA 4.
- 17 Myers, R.H., Montgomery D.C., Anderson-Cook C.M., 2016. Response surface
18 methodoloty: processa and product optimization using designed experiments.
19 John Wiley & Sons.
- 20 Oladyshkin, S., Class, H., Helmig, R., Nowak, W., 2011a. A concept for data-driven
21 uncertainty quantification and its application to carbon dioxide storage in
22 geological formations. Advances in Water Resources 34, 1508-1518.
- 23 Oladyshkin, S., Class, H., Helmig, R., Nowak, W., 2011b. An integrative approach to
24 robust design and probabilistic risk assessment for CO₂ storage in geological
25 formations. Computational Geosciences 15, 565-577.
- 26 Oladyshkin, S., Class, H., Nowak, W., 2013. Bayesian updating via bootstrap filtering
27 combined with data-driven polynomial chaos expansions: methodology and
28 application to history matching for carbon dioxide storage in geological
29 formations. Computational Geosciences 17, 671-687.
- 30 Pan, F., Ye, M., Zhu, J., Wu, Y.-S., Hu, B.X., Yu, Z., 2009. Numerical evaluation of
31 uncertainty in water retention parameters and effect on predictive uncertainty.
32 Vadose Zone Journal 8, 158-166.
- 33 Raines, M., Dobitz, J., 2001. A review of the Pennsylvanian SACROC Unit.
34 PUBLICATIONS-WEST TEXAS GEOLOGICAL SOCIETY, 67-74.
- 35 Shaw, J., Bachu, S., 2002. Screening, evaluation, and ranking of oil reservoirs suitable for
36 CO₂-flood EOR and carbon dioxide sequestration. Journal of Canadian Petroleum
37 Technology 41.
- 38 Vest Jr, E., 1970. Oil fields of Pennsylvanian-Permian horseshoe atoll, west Texas.
- 39 Wan, X., Karniadakis, G.E., 2006. Multi-element generalized polynomial chaos for
40 arbitrary probability measures. SIAM Journal on Scientific Computing 28, 901-
41 928.

- 1 Wiener, N., 1938. The homogeneous chaos. American Journal of Mathematics, 897-936.
- 2 Wriedt, J., Deo, M., Han, W.S., Lepinski, J., 2014. A methodology for quantifying risk
3 and likelihood of failure for carbon dioxide injection into deep saline reservoirs.
4 International Journal of Greenhouse Gas Control 20, 196-211.
- 5 Xiu, D., Karniadakis, G.E., 2002. The Wiener--Askey polynomial chaos for stochastic
6 differential equations. SIAM journal on scientific computing 24, 619-644.
- 7 Ye, M., Pan, F., Wu, Y.-S., Hu, B.X., Shirley, C., Yu, Z., 2007. Assessment of
8 radionuclide transport uncertainty in the unsaturated zone of Yucca Mountain.
9 Advances in water resources 30, 118-134.
- 10 Zhang, D., Lu, Z., 2004. An efficient, high-order perturbation approach for flow in
11 random porous media via Karhunen--Loeve and polynomial expansions. Journal
12 of Computational Physics 194, 773-794.
- 13 Zhang, Y., Sahinidis, N.V., 2012. Uncertainty quantification in CO₂ sequestration using
14 surrogate models from polynomial chaos expansion. Industrial & Engineering
15 Chemistry Research 52, 3121-3132.

1 **List of Figures:**

2 Figure 1. (a) Location of SACROC and structural contour map of top of carbonate reef,
3 (b) Structure and stratigraphic cross-section of profile A-A' (modified from Han, 2007)

4 Figure 2. Marginal PDF plots and joint distribution of porosity and log-permeability for a
5 selected cell

6 Figure 1 Cross-sectional view of permeability distribution of the updated upscaled model
7 (j=7)

8 Figure 4. ROMs regression fit at Time 3 for the four model outputs of interest: (a) Mass
9 of CO₂ stored in oil phase, (b) Mass of CO₂ stored in supercritical phase, (c) Mass of CO₂
10 stored in aqueous phase, and (d) oil saturation

11 Figure 5. CDF plots for mass of CO₂ in oil, supercritical and aqueous phase at Time 1,
12 Time 2, and Time 3

13 Figure 6. Boxplot for mass of dissolved CO₂ in oil phase at Time 1, Time 2, and Time 3

14 Figure 7. Cross-sectional view of mean values of oil saturation (j=7) at (a) Time 1, (b)
15 Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted as flat layers and
16 thickness of layers is exaggerated)

17 Figure 8. Cross-sectional view of mean values of CO₂ mass in oil phase (j=7) at (a) Time
18 1, (b) Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted as flat
19 layers and thickness of layers is exaggerated)

20 Figure 9. Boxplot for mass of CO₂ in supercritical phase at Time 1, Time 2, and Time 3

21 Figure 10. Cross-sectional view of mean values of CO₂ mass in supercritical phase (j=7)
22 at (a) Time 1, (b) Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted
23 as flat layers and thickness of layers is exaggerated)

24 Figure 11. Boxplot for mass of dissolved CO₂ in aqueous phase at Time 1, Time 2, and
25 Time 3

26 Figure 12. Cross-sectional view of CO₂ mass distribution in aqueous phase (j=7) at (a)
27 Time 1, (b) Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted as
28 flat layers and thickness of layers is exaggerated)

1 **List of Tables:**

2 Table 1. Variogram parameters for porosity and permeability well logs

3 Table 2. ROMs regression fit results

4 Table 3. ROMs validation results

5 Table 4. Probabilistic assessment of CO₂ storage percentage in oil, supercritical and
6 aqueous phase at Time 1, Time 2, and Time 3

7 Table 5. Probabilistic assessment of CO₂ storage in five layers at Time 1, Time 2, and
8 Time 3

9 Table 6. Average CO₂ storage percentage in three subdomains0

10

Figure 2. (a) Location of SACROC and structural contour map of top of carbonate reef, (b) Structure and stratigraphic cross-section of profile A-A' (modified from Han, 2007)

1

2 Figure 3. Marginal PDF plots and joint distribution of porosity and log-permeability for a
 3 selected cell

4

1

2 Figure 4 Cross-sectional view of permeability distribution of the updated upscaled model
3 (j=7)

4

1

2

3 Figure 5. ROMs regression fit at Time 3 for the four model outputs of interest: (a) Mass
 4 of CO₂ stored in oil phase, (b) Mass of CO₂ stored in supercritical phase, (c) Mass of CO₂
 5 stored in aqueous phase, and (d) oil saturation

6

Figure 6. CDF plots for mass of CO₂ in oil, supercritical and aqueous phase at Time 1, Time 2, and Time 3

1

2

3 Figure 7. Boxplot for mass of dissolved CO₂ in oil phase at Time 1, Time 2, and Time 3

1

2

3

4 Figure 8. Cross-sectional view of mean values of oil saturation ($j=7$) at (a) Time 1, (b)

5 Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted as flat layers and

6 thickness of layers is exaggerated)

7

Figure 9. Cross-sectional view of mean values of CO₂ mass in oil phase ($j=7$) at (a) Time 1, (b) Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted as flat layers and thickness of layers is exaggerated)

Figure 10. Boxplot for mass of CO₂ in supercritical phase at Time 1, Time 2, and Time 3

Figure 11. Cross-sectional view of mean values of CO₂ mass in supercritical phase (j=7) at (a) Time 1, (b) Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted as flat layers and thickness of layers is exaggerated)

1

2 Figure 12. Boxplot for mass of dissolved CO₂ in aqueous phase at Time 1, Time 2, and
 3 Time 3

Figure 13. Cross-sectional view of CO₂ mass distribution in aqueous phase (j=7) at (a) Time 1, (b) Time 2, and (c) Time 3 (for better visual effect, actual layers are plotted as flat layers and thickness of layers is exaggerated)

1 Table 1. Variogram parameters for porosity and permeability well logs

	Porosity	Permeability
Major direction	60	315
Minor direction	330	225
Dip	0	0
Type	Spherical	Spherical
Sill	1	1
Nugget	0.191	0
Major range	3310.34	1027.68
Minor range	2145.59	1020.28
Vertical range	16.54	17.19

2

1 Table 2. ROMs regression fit results

Model outputs of interest		R^2	NRMSE
CO ₂ mass in oil phase	TIME 1	0.9876	1.17%
	TIME 2	0.9860	1.55%
	TIME 3	0.9856	1.67%
CO ₂ mass in supercritical phase	TIME 1	0.9860	1.15%
	TIME 2	0.9833	2.48%
	TIME 3	0.9734	2.58%
CO ₂ mass in aqueous phase	TIME 1	0.9910	0.91%
	TIME 2	0.9911	0.96%
	TIME 3	0.9883	0.97%
Oil Saturation	TIME 1	0.8919	6.34%
	TIME 2	0.8980	6.55%
	TIME 3	0.9139	6.82%

2

1 Table 3. ROMs validation results

Model outputs of interest		R^2	NRMSE
CO ₂ mass in oil phase	TIME 1	0.9891	5.49%
	TIME 2	0.9819	5.73%
	TIME 3	0.9666	5.98%
CO ₂ mass in supercritical phase	TIME 1	0.9751	5.75%
	TIME 2	0.8398	6.93%
	TIME 3	0.8932	6.17%
CO ₂ mass in aqueous phase	TIME 1	0.9891	4.54%
	TIME 2	0.9906	4.14%
	TIME 3	0.9801	3.93%
Oil Saturation	TIME 1	0.8561	8.42%
	TIME 2	0.8285	8.90%
	TIME 3	0.8596	8.56%

2

1 Table 4. Probabilistic assessment of CO₂ storage percentage in oil, supercritical and
2 aqueous phase at Time 1, Time 2, and Time 3

	Time 1		Time 2		Time 3	
	P5	P95	P5	P95	P5	P95
Oil	44.44%	52.54%	50.01%	56.89%	54.23%	61.22%
Supercritical	24.76%	30.08%	21.74%	25.41%	20.56%	24.41%
Aqueous	14.33%	19.32%	11.79%	15.55%	11.28%	15.18%

Table 5. Probabilistic assessment of CO₂ storage in five layers at Time 1, Time 2, and Time 3

	Time 1			Time 2			Time 3		
	P5	P95	Mean	P5	P95	Mean	P5	P95	Mean
CO ₂ Storage (million tonnes)	3.17	3.61	3.38	4.35	4.81	4.58	4.49	4.96	4.72
Storage Percentage	86.65%	98.76%	92.39%	85.95%	95.01%	90.46%	88.71%	97.99%	93.25%

1 Table 6 Average CO₂ storage percentage in three subdomains

	Layer 1 -17	Layer 18-22	Layer 23-25
Time 1	3.32%	88.64%	8.04%
Time 2	2.06%	93.11%	4.83%
Time 3	2.26%	92.47%	5.27%

2

3