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12 4 On the detection of leakage pathways in geological CO₂ storage systems using pressure
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Abstract

In this study, we examine the effect of model parameter uncertainties on the feasibility of detecting unknown leakage pathways from CO₂ storage formations via inversion of pressure monitoring data, and discuss the strategies for enhancing detectability and reducing the impact of those uncertainties. We conduct a numerical study of leakage detection, using an idealized storage system consisting of a storage formation and an overlying aquifer separated by a caprock, with an injection well and a leaky well. Our uncertainty quantification analysis shows that (1) the anomalous leakage signals induced by the leaky well can be clearly detected in the overlying aquifer, with minimal impact of model parameter uncertainties, as long as the leaky well permeability is sufficiently large and the caprock permeability is small with the assumed aquifer and caprock thickness; and (2) the pressure monitoring data in the storage formation are not adequate for detecting leakage signals, because the model predictions can be significantly affected by the uncertainties of the model parameters (e.g., permeability and specific storativity of the storage formation and the overlying aquifer). Therefore, we propose an inverse-modeling methodology that combines leakage detection with model recalibration under conditions of model parameter uncertainties. Our results show that the combined leakage detection and model recalibration are most successful when pressure monitoring data from both the storage formation and the overlying aquifer are used, owing to the strong detectability in the overlying aquifer and the strong sensitivity of pressure in the storage formation to model parameters. The proposed methodology also shows that the effect of model uncertainties on leakage detection can be reduced by simultaneously estimating the leakage parameters and the uncertain model parameters, using long-term pressure data under various conditions of permeabilities and locations of the leaky well, and a wide range of uncertainties for the model parameters.

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44 Keywords: geological carbon storage; risk assessment; early leakage detection; pressure

45 monitoring; model parameter uncertainty

1. Introduction

For geological carbon sequestration (GCS) to have a sizable effect on mitigating climate change, large volumes of CO₂ must be injected into deep storage formations. Large-scale CO₂ injection may result in substantial increases in pressure within these formations and heighten the potential risks of GCS [1,2]. Brine or/and CO₂ may leak from these formations through unknown high-permeability leakage pathways (e.g., abandoned wells and faults) within the area of influence. In addition, the increased pressures in the storage formations could induce geomechanical alteration of the reservoirs and their surroundings, e.g., creating new fractures or reactivating larger faults [3]. These changes occurring in the caprock or overburden could result in new leakage pathways for brine and/or CO₂. If such leakage events cannot be properly assessed, GCS might cause undesirable environmental and safety consequences that may ultimately prevent future deployment of GCS. Therefore, it is essential to the success of GCS to have the ability to detect brine or CO₂ leakage through high-permeability pathways from the storage reservoirs, predict potential risk profiles, and manage the risks as early as possible.

Pressure-based detection for CO₂ and brine leakage in GCS is one of the techniques currently under investigation. At a storage site, pressure monitoring is usually conducted for hydraulic tests during characterization phase and for CO₂ injection during operation phase. The monitored pressure data can be used to estimate hydrogeological properties of the storage reservoir, assess CO₂ injectivity, monitor the system behavior during CO₂ injection, and calibrate a hydrogeological model by history-matching [4-6]. Pressure data may also be very useful for detecting leaks of brine or CO₂ (if any). This is because (1) the pressure pulse (buildup) induced by CO₂ injection travels much faster than the CO₂ plume toward a leakage pathway in the caprock, (2) the pressure anomalies induced by brine leakage through the leakage pathway (e.g.,

pressure increases in the aquifer overlying the storage formation) will also propagate fast to the monitoring wells, and (3) the monitored pressure anomalies can be detected through inverse modeling in advance of CO₂ leakage through the leaky pathway.

Several modeling studies evaluating the detectability of anomalous signals in pressure data showed that the pressure-based detection method could be useful for providing early warning of large leaks [7-9]. These studies focused on using pressure data from the overlying aquifer, obtained via so-called above-zone monitoring, as a cost-effective leakage detection method. Above-zone pressure monitoring has been applied at several storage sites, including the CO₂ storage pilot site at the Ketzin site in Germany [10,11], the Cranfield (Mississippi, USA) enhanced oil recovery (EOR) and sequestration site [12,13], CO₂-EOR sites in the Gulf Coast region [14], and the Illinois Basin–Decatur Project for large-scale demonstration of GCS [5]. The non-negligible pressure monitored in the above zone at these sites may show diffuse leakage through caprock [5], a focused leakage through wells [13], or a combination of both. However, with respect to well leakage, the existing analysis was not conclusive on leakage through *unknown* leaky wells [13], possibly because of the complexity of the real storage system and the lack of detection methodology.

A few studies have been conducted to investigate the feasibility of detecting *unknown* leaky pathways via inverse analysis of pressure data, using different synthetic scenarios of the storage system. It has been shown that the effectiveness and accuracy of an inversion methodology greatly depend on the number of monitoring wells available, their spatial configuration, and pressure data noise [15-19]. Most of the feasibility studies cited above used only above-zone pressure data for the inversion, and also assumed that the overlying aquifer was

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4 91 separated from the storage formation by an impermeable caprock [15-18]. This means that an
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6 92 anomalous pressure signal in the overlying aquifer must be caused by a leakage pathway—
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8 93 either a leaky well or a natural pathway such as a fault—because the possible effects of slow
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10 94 pressure propagation through an intact caprock are neglected. Jung et al. [19] developed an
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12 95 inverse modeling approach to use pressure data from both the above-zone and storage aquifers
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14 96 for detection, with a forward model considering the effect of slow diffuse leakage through a low-
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16 97 permeability caprock formation. Their study showed that the accuracy of leakage detection
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18 98 greatly depends on the level of data noise, and that use of additional complementary data, such as
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29 101 In addition to the detectability of the leakage signals and the effectiveness and accuracy
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31 102 of the inverse modeling methodology, the method for inverse detection of leakage relies on an
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33 103 adequate forward model for simulating the pressure responses in the storage formation and the
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35 104 overlying aquifer. This forward model provides the “expected” system response without taking
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37 105 into account unknown leakage pathways, and the “expected” response is compared to the “actual”
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39 106 measured system response to define anomalous signals. To date, the inverse modeling studies
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41 107 have all assumed that the forward models are error-free. In other words, the anomalous signals
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43 108 introduced by leakage pathways of unknown location and properties would be obvious in
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45 109 comparison to the expected system responses simulated by a “perfect” forward model, in which
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47 110 the only deviation is caused by leakage. Of course, it is not practically possible to have a perfect
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49 111 model with no structural and parametric uncertainties, which could accurately predict pressure
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51 112 evolution profiles at monitoring wells at real storage sites, for various reasons: (1) the data from
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4 114 accurate model calibration, (2) multiscale heterogeneity of hydrogeological properties is difficult
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6 115 to perfectly capture in the model, and (3) the field data used for model calibration may already
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8 116 comprise signatures of leaky pathways near the injection zone. Zeidouni and Pooladi-Darvish
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10 117 [17], for instance, showed that the inverse problem of identifying leakage pathways using
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12 118 pressure data from measurements in the overlying aquifer is highly ill-conditioned and unstable,
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14 119 and therefore not only the noise in measurements but also the error in any model parameters can
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16 120 lead to inaccurate estimates of leakage. Considering that a small error in the model parameters
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18 121 can have a more significant impact on identifying leakage pathways than measurement noise, it
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20 122 is important to ascertain whether the idea of using pressure monitoring data for detecting
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22 123 unknown leaky pathways is achievable in cases when the forward model is not perfect, i.e., when
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31 125 In this study, we examine the impact of uncertainties in forward model parameters on the
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33 126 accuracy of leakage detection and estimation of potential leakage. For demonstration, we employ
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35 127 an idealized storage system with an injection well and a leaky well, and develop a forward model
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37 128 for pressure prediction using an analytical solution. We conduct an uncertainty analysis for a
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39 129 suite of model parameters, and discuss the effect of the model uncertainties on model prediction
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41 130 and leakage detection. Based on the uncertainty quantification analysis, we consider two
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43 131 synthetic examples of leakage detection via inverse analysis with different sets of pressure
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45 132 monitoring data: (1) pressure data exclusively from the above-zone (or overlying) aquifer, and (2)
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47 133 pressure data from both the above-zone aquifer and the storage formation—and evaluate the
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49 134 accuracy and convergence speed of leakage detection. Finally, we discuss strategies for
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2. Methodology

To examine the impact of model uncertainties on detection of unknown leaky pathways, we construct an idealized storage system and conduct a generic leakage detection study using a simplified pressure prediction model. We use a “perfect” forward model (having true model parameters) with a leaky well to generate “measured” monitoring responses from the system, and have an “imperfect” forward model (with uncertain values for some selected model parameters) with no leaky well to simulate the “expected” behavior of the system. We assume that the leakage pathway and the uncertainties in the model parameters are unknown during the inversion, and invert the calculated pressure anomalies—the difference between the measured and the expected pressure changes at monitoring locations—to detect and characterize the leakage pathways, while simultaneously revising and recalibrating the imperfect forward model. After the model recalibration, the imperfect forward model will have more accurately calibrated model parameters than initially uncertain values.

2.1. Forward model setup and parameters

The idealized CO₂ storage system consists of a target storage formation bounded by an impervious boundary at the bottom and overlain by a semi-pervious caprock formation and a permeable aquifer. The above-zone aquifer is bounded by an impervious boundary at the top (see Fig. 1a). This system is simplified by assuming that each aquifer/aquitard is homogeneous, with a uniform thickness and an infinite extent, and by assuming horizontal flow in the aquifers and vertical flow in the aquitard. In this study, we focus on single-phase brine flow outside of the CO₂ plume (e.g., Cihan et al. [20]), where all monitoring wells used for leakage detection are located. For this storage system, we develop a forward model for predicting pressure response in

the aquifer/aquitard system, using the semi-analytical solution in Cihan et al. [21]. The solution can account for both diffuse leakage through aquitards and focused leakage through multiple leaky wells.

The true model parameters of the storage formation, the overlying aquifer, and the caprock are summarized in Table 1. The permeability of the two aquifers is 10^{-13} m^2 , and the permeability of the aquitard is 10^{-19} m^2 . The specific storativity value for the aquifers corresponds to an aquifer pore compressibility of $4.5 \times 10^{-10} \text{ Pa}^{-1}$ and a porosity of 0.2, while the specific storativity value of the caprock corresponds to a pore compressibility of $9.0 \times 10^{-10} \text{ Pa}^{-1}$ and a porosity of 0.1. All formation properties are based on previous studies of large-scale CO_2 storage [22, 23].

Since a single-phase flow model is used for pressure simulation outside the CO_2 plume, CO_2 injection is represented by injection of an equivalent volume of brine, which has been demonstrated as a reasonable way to describe the brine pressurization and migration processes outside of the CO_2 plume [24]. We assume an industrial-scale CO_2 storage operation with a constant volumetric injection rate of $5700 \text{ m}^3 \text{ d}^{-1}$. The injection well is located at the center of the domain [0 km, 0 km]. To test the feasibility of inverse detection, we introduce an unknown leaky well 2 km away from the injection well at [2 km, 0 km] (see Fig. 1b). This leaky well location is used for all scenarios except the cases where different leaky well locations of [1 km, 0 km], [3 km, 0 km], [4 km, 0 km], and [-2 km, 0 km] are considered to examine the effect of the location of the leaky well relative to the monitoring wells. Through the leaky well, resident brine migrates into the overlying aquifer, resulting in anomalous pressure signals. The radius of the injection and the leaky well is 0.15 m, and the leaky well permeability is $k_L = 10^{-7} \text{ m}^2$. This high permeability is selected based on the dependence of the detectable region of leakage-induced

anomaly on k_L in the overlying aquifer (Jung et al. [19], Figure 6c). The detectable region, where the anomaly (i.e., the 0.1 bar of pressure difference between the cases with and without well leakage) can be detected, is very small when $k_L < 10^{-10} \text{ m}^2$, increases in sensitivity with the increase in k_L up to 10^{-8} m^2 , and then is relatively insensitive to k_L when $k_L > 10^{-8} \text{ m}^2$. To focus our discussion on the impact of model parameter uncertainties, we use the high permeability $k_L = 10^{-7} \text{ m}^2$ as the default value for most of the simulations unless otherwise stated. We also consider sensitivity-analysis cases with lower permeabilities ($k_L = 10^{-8} \text{ m}^2$ and 10^{-9} m^2), to test the effectiveness of the inversion when the forward model is imperfect and the leakage signal is relatively weak.

To generate synthetic pressure monitoring data, we run the “perfect” forward model (with true model parameters in Table 1) with the leaky well for a period of 10 years after the start of injection, and record the pressure responses as the “measured” data. Two monitoring wells are available in addition to the injection well (IW): MW1 at [-60 m, 80 m] and MW2 at [1.5 km, 1.5 km]. For the cases using only the above-zone pressure data, the monitoring data include the transient pressures in the overlying aquifer at IW, MW1, and MW2. For the cases using the data from both the above-zone aquifer and the storage formation, the monitoring data additionally include the transient pressure at MW2 in the storage formation. The pressure data from the storage formation at IW and MW1 are disregarded, because the assumption of single-phase flow cannot be justified for these locations during the injection period. Since noise in the pressure data is typically proportional to the measurements themselves, different noise is assumed for the pressure data measured in the storage formation and the overlying aquifer. A zero-mean Gaussian distribution is used for the noise, with a standard deviation of 0.1 bar in the storage formation, and a standard deviation of 0.01 bar in the overlying aquifer.

2.2. Errors in model parameters

The “imperfect” forward model is developed by introducing errors within some selected model parameters. Here, we consider several factors that can introduce such errors during site characterization, and use the parameters obtained (calibrated) from this setup as the initial values for the imperfect forward model. For the synthetic examples of leakage detection, we assume that characterization and estimation of subsurface properties comes with errors (insufficient data), that the collected pressure data contain random noise as described in the previous section (noise in data), and that the leakage pathways are unknown (lack of knowledge regarding potential leakage pathways). In terms of subsurface properties, let us consider that among other site characterization activities, a pumping test has been conducted in the storage formation before CO₂ injection.

We run the “perfect” forward model to generate the pressure response in MW1 from pumping at IW, and use these pressure data for the calibration of the imperfect forward model before injection. A constant pumping rate of $24 \text{ m}^3 \text{ d}^{-1}$ is applied for three days, and pressure data are monitored at MW1 for one week, which includes the pumping and the recovery period. The permeability (K_1) and specific storativity (S_1) of the storage formation estimated from the pumping test are $9.78 \times 10^{-14} \text{ m}^2$ and $2.14 \times 10^{-6} \text{ m}^{-1}$, and are 2.2% underestimated and 13.8% overestimated, respectively. On the other hand, assuming that no pumping test data are available for the overlying aquifer, we postulate that the permeability (K_2) and specific storativity (S_2) of the overlying aquifer are estimated from other measurements or best guess, based on other literatures or experiences, and include 50% error for the base case. Different K_2 and S_2 , which are

up to one order magnitude larger than the true values, are considered in a sensitivity analysis as well, to examine the effect of error size on the inversion. All the other parameters are assumed to be known and error-free.

2.3. Uncertainty Quantification

Before investigating the effect of model uncertainties on leakage detection, we perform an uncertainty quantification analysis to understand how likely model predictions are, and how potential uncertainties in the pre-calibrated model parameters propagate to the system responses (e.g., pressures at monitoring wells in the storage formation and the overlying aquifer). For the synthetic examples of leakage detection considered in this study, we assume that the uncertainty of the pre-calibrated model parameters (K_1 , S_1 , K_2 , and S_2) is unknown, as well as the reliability of model prediction. We consider the standard deviations of 10%, 20%, and 50% in K_1 , S_1 , K_2 , and S_2 of the imperfect forward model, which reflects the uncertainty associated with their initial estimates. The uncertainty of the model prediction is assessed using the First-Order-Second-Moment (FOSM) uncertainty propagation analysis [25].

2.4. Inverse Modeling Strategy

Several scenarios and strategies for inverse modeling are considered to assess the impact of model parameter uncertainty and data availability on leakage detection, as summarized in Table 2. For the base-case permeability of the leaky well, three cases are considered for the combination of leakage detection and model recalibration. In the first case, we estimate the leakage parameters (the location and the permeability of the leaky well) by assuming that all pre-calibrated model parameters are correct. In the second case, we include K_2 and S_2 (which are not

estimated from direct field measurements and may not properly represent field conditions) as the parameters to be estimated along with the leakage parameters. In the final case, we take into account the uncertainties in the estimated K_1 and S_1 , as well as in leakage detection and model recalibration. We also examine the effect of leaky well permeability ($k_L = 10^{-7}$, 10^{-8} , and 10^{-9} m²) in addition to model parameter uncertainties. For each inverse modeling case, two different data sets are considered: (1) pressure data exclusively from the above-zone (or overlying) aquifer, and (2) pressure data from both the above-zone aquifer and the storage formation.

For each case in Table 2, we generate the “monitoring” data by running the perfect forward model, including the leakage pathway, with the model parameters in Table 1 and the given permeability of the leaky well. For the leakage parameters, four different initial guesses (RUNs) of the leaky well location at [± 1 km, ± 1 km] are used to examine the uniqueness and accuracy of the inversion solution, along with the initial guess of the leaky well permeability of 2×10^{-9} m². For the uncertain model parameters considered (i.e., the permeability and specific storativity of the storage formation and the overlying aquifer), their initial guesses, described in Section 2.2 (i.e., $K_1 = 9.78 \times 10^{-14}$ m², $S_1 = 2.14 \times 10^{-6}$ m⁻¹, $K_2 = 1.5 \times 10^{-13}$ m², and $S_2 = 2.82 \times 10^{-6}$ m⁻¹, see Table 1), are used for model recalibration. In the cases of leakage detection only, all the uncertain model parameters are fixed at their initial guesses, because these values are to our best knowledge available. We use the imperfect forward model (with the uncertain model parameters) to predict the system responses, and execute the inversion to detect possible leakage pathways. In the cases of the combined leakage detection and model recalibration, the forward model is run with newly updated model parameters for each iteration of inverse modeling, with the goal to minimize the mismatch between predicted and measured system responses.

The inverse modeling, as well as sensitivity and uncertainty quantification analyses, is conducted with iTOUGH2-PEST [26]. As a computer program for parameter estimation, sensitivity analysis, and uncertainty propagation analysis, iTOUGH2-PEST inherits all the capabilities of iTOUGH2 [27], developed for use with the TOUGH2 forward simulator for non-isothermal multiphase flow in porous and fractured media [28]. However, by utilizing the PEST protocol [29] as a way to communicate with any generic forward model (e.g., the semi-analytical pressure model used in our study), iTOUGH2-PEST can also be used model-independently. As mentioned before, the forward model communicating with iTOUGH is a semi-analytical solution for pressure perturbation and fluid leakage developed by Cihan et al. [21].

3. Inversion Results and Discussion

3.1. Potential Impact of Parameter Uncertainties on Model Prediction

The uncertainty in model parameters is usually unknown, or difficult to estimate, before any further data are available. The accuracy of a site-specific model developed for a CO₂ storage site may vary strongly, depending on the regulations, resources, and techniques applied for site characterization. The heterogeneity of the site is also a factor contributing to the reliability of the model developed. For our scenarios of leakage detection and pressure monitoring, the uncertainty in the forward model is also unknown. Therefore, an uncertainty quantification analysis is first performed to understand how the uncertainty in the forward model could affect the model prediction.

Figure 2 shows 95% FOSM error bands of the pressure increase at MW2, along with the “expected” pressure increase (blue line) in the storage formation and the overlying aquifer, using the imperfect forward model, with the initial guesses for the uncertain model parameters as shown in Table 1. These results are compared with the “measured” data of the pressure increase

(red line, obtained with the perfect forward model with the leaky well). The shaded areas from the innermost (darkest blue) to the outermost (lightest blue) represent 95% FOSM error bands for 10%, 20%, and 50% uncertainties in the model parameters of interest, respectively.

In the storage formation, the expected pressure responses are close to the measured data up to about one year, and then slowly deviate from the measurements with time. This means that the forward model predicts the pressure evolution fairly well, despite the small to moderate errors in the model parameters, and that the pressure anomalies induced by the presence of the leaky well are not strong enough at early monitoring times, making the detection of leakage signals difficult. However, the impact of model parameter uncertainties on model prediction is significant. The magnitude of the error band depends on the pressure and increases with time. A moderate uncertainty ($> 10\%$) in the model parameters can result in noticeably different predictions. The large uncertainty range of the expected pressure may disguise any pressure anomaly induced by the presence of the leaky well. Only in the case of 10% uncertainty in the model parameters are the expected pressure responses distinct from the measured ones at the later time of injection and monitoring (i.e., the measured pressure is below the 95% FOSM error band of the expected pressure). That is, with the pressure data from the storage formation only, it would be difficult to tell whether an anomalous pressure signal originates from the potential presence of a leaky well or is simply a result of parameter uncertainty, unless the uncertainty in model parameters is small enough (i.e., $< 10\%$).

In the overlying aquifer, the calculated “expected” pressure is below the detection limit throughout the monitoring time of 10 years. Since the leaky well is not accounted for in the pre-calibrated forward model, the diffuse leakage through the caprock results in negligible pressure buildup in the overlying aquifer, and the uncertainties in the model parameters have virtually no

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4 321 impact on the expected pressure. On the contrary, the measured pressures provide a clear leakage
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6 322 signal as early as 50 days after start of injection, even though the pressure changes in the
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14 325 presence of a leakage pathway can still be detected, as long as the leakage rate is sufficiently
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16 326 large, inducing anomalous pressure buildup in the overlying aquifer. Therefore, having a
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19 327 monitoring well in the overlying aquifer is a critical asset for early detection of anomalous
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21 328 leakage signals. We can hypothesize the presence of a leaky pathway from this pressure anomaly
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24 329 observed in the overlying aquifer, and conduct an inversion analysis to locate the leaky pathway
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26 330 and estimate its properties.

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29 331 This finding—that model parameter uncertainties have minimal impact on detecting
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31 332 anomalous signals in the overlying aquifer—is useful when applying the above-zone pressure
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33 333 monitoring technique and diagnosing the caprock integrity and potential leakage through a
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36 334 known leaky well. Indeed, this insensitivity of the “expected” above-zone pressure is combined
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41 336 will impact the behavior of leakage and thus the accuracy of leakage detection. In contrast, the
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3.2. Effect of Model Parameter Uncertainties on Leakage Detection

In the first inversion attempt, we disregard all uncertainties in the model parameters by assuming their “best-known” values are correct, and merely estimate the leakage parameters using the data from the overlying aquifer (Case A1 in Table 2), and then using the data from both the storage formation and the overlying aquifer (Case B1). The match between the measured and simulated pressures is poor and not random in either case (not shown), indicating that the imperfect forward model does not properly capture the actual system responses and needs to be adjusted. The location and permeability of the leaky well are poorly estimated for both data sets used for inversion, and the estimates are not improved with additional monitoring data available with time, reaffirming the inadequacy of the forward model. In Case A1, the estimated well location is [1.8 km, 0.2 km], and the estimated well permeability is more than two orders of magnitude higher than the true well permeability. In Case B1, the estimates are no better than those in Case A1.

In the second attempt, K_2 and S_2 are also included in the parameters to be estimated through inversion. The pressure data of the incremental monitoring periods of 1, 2, 3, and 6 years are used, and Cases A2 and B2 are compared for the effectiveness of model recalibration and leakage detection. In Case B2, the misfits (not shown) between the measured and simulated system responses are not random, and are not appraised appropriately, indicating that the estimates of the leaky well are not accurate. In short, the leakage detection fails even with the 6-year monitoring data for both aquifers, when the uncertainties of the model parameters in the storage formation are not considered.

Figure 3 shows the estimated location and permeability of the leaky well and the recalibrated K_2 and S_2 in Case A2, using two initial guesses for well location (RUN1 at [1 km, 1 km] and RUN4 at [1 km, -1 km]). For these two runs, the converged location estimates are

relatively close to the actual well location compared to those in RUN2 and RUN3 (not shown).

In general, when the initial guesses are closer to the actual well location, the estimates are more accurate.

In Figure 3, we observe that the estimated well location converges to the actual value with additional available monitoring data: from [1.3 km, 0.4 km] with the 1-year monitoring data to [1.9 km, 0.05 km] with the 6-year data. On the other hand, the accuracy of the estimated well permeability is in general poor. In most trials, the estimated value is off by 1–3 orders of magnitude; even with the 6-year monitoring data, the estimated well permeability is $2.2 \times 10^{-9} \text{ m}^2$, in comparison to the true leaky well permeability of 10^{-7} m^2 . Moreover, when the leaky well permeability is overestimated, the marginal standard deviation of the estimate is so large that the range cannot be properly shown in Figure 3b (marked with the asterisk symbol, *). This high uncertainty in the permeability estimate is caused by the insensitivity of the pressure response (and the leakage rate) to the leaky well permeability when the leaky well permeability is sufficiently large (i.e., $> 10^{-8} \text{ m}^2$) (Jung et al. [19]). The recalibrated K_2 and S_2 using the 6-year monitoring data converge to $3.3 \times 10^{-14} \text{ m}$ and $5.9 \times 10^{-7} \text{ m}^{-1}$, respectively, which are 33% and 31% of their true values, respectively.

The result of Case A2 shows that the leaky well location can be quite accurately estimated by using the 6-year monitoring data, even though the other parameter estimates are not accurate (i.e., the estimated leaky well permeability is nearly two orders of magnitude less than the true value, and the recalibrated K_2 and S_2 are underestimated by ~30%). The simulated pressure changes, using the estimated leakage parameters and the recalibrated K_2 and S_2 , are in a good agreement with the measured ones at all pressure monitoring locations. That is, if the errors in other parameters (here, K_1 and S_1) are sufficiently small (K_1 is only 2.2% underestimated, and

388 S_1 is 13.8% overestimated)—in other words, if the site characterization of the storage reservoir
389 were done accurately—successful leakage detection might be feasible with above-zone pressure
390 monitoring, and risk assessment and management decisions based on the estimate might be
391 acceptable.

392 In the final inversion attempt, we also include K_1 and S_1 in the parameters to be estimated.
393 We compare the accuracy and convergence speed of leakage detection between the case using
394 pressure data exclusively from the overlying aquifer (Case A3) and the case using pressure data
395 from both the overlying aquifer and the storage formation (Case B3). In both cases, the model
396 parameters K_1 , S_1 , K_2 , and S_2 are simultaneously estimated with the leakage parameters using
397 incremental pressure monitoring data. The simulated pressures using the estimated leakage and
398 model parameters are in a good agreement with the measured ones at all monitoring locations for
399 both Cases A3 and B3. Figure 4 shows the accuracy and convergence speed of the estimation of
400 the leaky well location. In Case B3, the estimated well location converges quickly to the true
401 location, with a small standard deviation (see Figure 4b). The initial guesses for well location
402 have only a small effect on the convergence speed. In Case A3, the estimate of the leaky well
403 location converges to the true location as the monitoring period increases in RUN 1, with a
404 significantly higher standard deviation than Case B3 (see Figure 4a), and does not converge after
405 6 years of monitoring in RUN4.

406 The estimated leaky well permeability shows a similar trend of convergence (see Figure
407 5). In Case B3, the leaky well permeability converges to $3.3 \times 10^{-8} \text{ m}^2$ with the 6-year monitoring
408 data, and the marginal standard deviation is less than half an order magnitude. However, in Case
409 A3, the leaky well permeability is still underestimated by two orders of magnitude (7.8×10^{-10}
410 m^2 for RUN1 and $2.7 \times 10^{-9} \text{ m}^2$ for RUN4) at six years, and the uncertainty of the estimate is

large. For both cases, the estimate is inaccurate, and its uncertainty remains large during the first three years, which may be attributed to the small sensitivity of the pressure responses to the well permeability at the tested leaky well permeability of 10^{-7} m^2 . This insensitivity is also reflected in the leakage-rate prediction. Even though the estimated leaky well permeability is two orders of magnitude higher (or lower) than the true value with the 2-year monitoring data in Case B3 (or with the 6-year monitoring data in Case A3), the predicted cumulative leakage volume is only 7% higher (about 30% smaller) than the actual leakage volume in six years.

The estimated model parameters (K_1 , K_2 , S_1 , and S_2) in Case B3 are in general more accurate than those in Case A3 (see Figure 6). Specifically, the accuracy of the model parameters for the storage formation (K_1 and S_1) is significantly improved in Case B3 by including the pressure monitoring data from the storage formation. The errors in the calibrated K_1 and S_1 are only 5% at one year, and decrease even more with monitoring time. The standard deviations for these estimated model parameters are less than 10% of their values. In Case A3, however, the estimated model parameters are not accurate, and their uncertainties are large (i.e., the standard deviations of the estimated K_1 and S_1 from RUN4 are too large to be shown in Figures 6a and 6e, respectively).

The inverse modeling analysis for Cases A3 and B3 shows that the accuracy of the estimated leakage parameters and the calibrated model parameters can differ greatly with data availability. The addition of pressure data from the storage formation effectively improves the estimation accuracy of the model parameters, which in turn improves the estimation accuracy of the leakage parameters. In fact, without these additional data, the attempt of taking into account the uncertainties in K_1 and S_1 in Case A3 (by simultaneously estimating these model parameters with K_2 , S_2 and the leakage parameters) results in over-parameterization and increases the

uncertainties in the estimated parameters, since more parameters need to be estimated with the same pressure data used in Case A2.

The difference in the estimation accuracy and uncertainty between Case A3 and B3 can also be understood by examining how sensitive the pressure data are to each model parameter, and how those sensitivities affect the model inversion. Figure 7 shows the temporal changes of the dimensionless sensitivity coefficient for K_1 , K_2 , S_1 , and S_2 at MW2. This dimensionless sensitivity coefficient represents the local sensitivity of the pressure data to each parameter, scaled by the inverse of the respective standard deviations. Among the four model parameters, K_1 is in general the most influential parameter affecting pressure signals in the storage formation, while S_1 is more important than K_1 at very early times (about up to 0.5 year). In the overlying aquifer, while pressure data are more sensitive to S_1 and S_2 for the first three years, and to K_1 and K_2 for the rest of the monitoring period, no parameter has a dominant influence on pressure signals.

In addition, the difference in the sensitivity coefficients between the model parameters is small. That is, the uncertainties in K_1 and S_1 have a considerable impact on the model prediction of pressure changes, not only in the storage aquifer but also in the overlying aquifer.

These results are consistent with the global sensitivity analysis in Jung et al. [19]. Jung et al. [19] assessed the sensitivity of pressure responses at the monitoring wells to changes in the model parameters varied over wide ranges. K_1 was the most influential parameter affecting pressure signals in the storage formation, and K_1 and K_2 had a similar impact on pressure anomalies in the overlying aquifer. Thus, when the pressure data from the storage formation are considered in the inversion, the estimation uncertainties of K_1 and S_1 can be remarkably reduced because of their strong sensitivity to K_1 and S_1 (see Figures 6b and 6f). This simultaneously

improves the estimation accuracy of the other parameters (K_2 and S_2) as well. However, without any data from the storage formation (such as Case A3), most of the model parameters cannot be correctly recalibrated, due to the strong correlations among the parameters. The resulting adverse impact on leakage parameter estimation is inevitable, as shown in Figures 4 and 5.

The results discussed above are based on four inverse modeling runs with regularly spaced initial guesses of the leaky well location. Here, for Cases A3 and B3 and using the 3-year period of monitoring data, we additionally test 50 initial leakage parameter sets (the X- and Y-coordinate of the leaky well and leaky well permeability), which are generated using Latin Hypercube Sampling, with the assumption of uniform distribution for all parameters. The lower and upper limits of both the X- and Y-coordinates are -2.5 km and 2.5 km, respectively, and the lower and upper limits of the leaky well permeability are 10^{-9} m^2 and 10^{-6} m^2 , respectively.

The results of the Latin Hypercube Sampling analysis indicate that only 20 runs on the initial guesses of (X, Y) and permeability of the leaky well have a good match, with random residuals, between the measured and computed pressures for both Cases A3 and B3. For most of the 20 runs, the initial leaky well location is close to the actual location compared to that for the other unsuccessful 30 runs. The initial leaky well permeability appears not to have much influence on the inversion result. Figure 8 shows the distribution of the estimated leaky well locations for those 20 runs, with each bar representing the frequency of the locations in a 100 m by 100 m grid of the domain. In Case A3, only one inverse modeling run correctly locates the leaky well, whereas in Case B3, 16 out of 20 runs are successful. For these successful runs, the estimated leaky well permeability is $2 \times 10^{-6} \text{ m}^2$ in Case A3, and ranges from $4 \times 10^{-7} \text{ m}^2$ to $6 \times 10^{-6} \text{ m}^2$ in Case B3, which is reasonably accurate, considering the insensitivity of the pressure response to the leaky well permeability in this range. This result embodies our observation from

the inverse modeling exercise: having pressure monitoring data for the storage formation matters, and aids leakage detection when model uncertainties cannot be completely ruled out.

3.3. Sensitivity Analysis of Large Uncertainties in Model Parameters

In the previous section, a 50% error in K_2 and S_2 is considered in the inverse modeling. When site characterization for the overlying aquifer does not provide much information on these parameters, such an error might be much larger than that considered for the base case. Here, K_2 and S_2 are assumed to be one order of magnitude larger than their true values. With these larger uncertainties, the unknown leaky well is still located accurately in Case 3B, similar to the previous section. This means that as long as K_2 and S_2 are parameterized and estimated along with the leakage parameters (the results are not shown), the developed methodology can provide accurate leakage detection and model recalibration, for a large range of model parameter uncertainties.

3.4. Sensitivity Analysis of Moderate to Small Leaks

All the above cases have a leaky well permeability of 10^{-7} m^2 to maximize the magnitude of pressure anomalies for leakage detection. Here, we examine the accuracy of detecting more moderate leaks through a leaky well of smaller permeability ($k_L = 10^{-8}$ or 10^{-9} m^2). For each of the well permeabilities, we conduct the inversion first using pressure data from the overlying aquifer exclusively (Cases A4 and A5 in Table 2), and second using the pressure data from both the overlying aquifer and the storage formation (Cases B4 and B5). All other conditions, including data availability and inverse modeling strategy, remain the same as for Cases A3 and B3.

Figure 9 shows the distance between the estimated and actual location and the estimated permeability of the leaky well as a function of monitoring time in RUN4. (The differences between RUN1 and RUN4 are not significant.) Also shown is the marginal standard deviation of the estimated leaky well permeability. Marginal standard deviations in the estimated X- and Y-coordinates of the leaky well are shown in Table 3 for the estimates with the 3-year monitoring data as a representative case.

When the pressure data in the storage formation are additionally included, the effect of the leaky well permeability—in other words, the leakage rate—becomes evident in the case with $k_L = 10^{-9} \text{ m}^2$ (see Figure 9b). Compared to the cases with $k_L = 10^{-7} \text{ m}^2$ and $k_L = 10^{-8} \text{ m}^2$, the convergence speed of the estimated leaky well location is slower, with a higher uncertainty in the estimate (see Table 3). The estimated leaky well location in Case B4 is similar to that in Case B3 at all monitoring times. This results from the small change in the leakage rate (and the pressure buildup at the monitoring wells) with the decrease in leaky well permeability from 10^{-7} m^2 to 10^{-8} m^2 . In the cases using the pressure data only in the overlying aquifer, the distance between the estimated and true leaky well location in general decreases (i.e., the estimated leaky well location converges to the true location) as the monitoring time increases. However, the convergence speed in Case A4 (and A5) is much slower than that in Case B4 (and B5), and the uncertainty in the estimate is also much larger. Specifically, in Case A5 with the 1-year monitoring data, the standard deviation in the estimated X- and Y-coordinates of the leaky well is 14.4 km and 8.5 km, respectively. This huge uncertainty makes the estimates not credible.

The accuracy of the estimated leaky well permeability improves as the leaky well permeability decreases (see Figures 9c and 9d). This improvement is expected, since the pressure responses at monitoring wells are sensitive to the leaky well permeability of $\sim 10^{-9} \text{ m}^2$ (Jung et al.

[19]). The standard deviation of the estimated value is, however, much bigger when the inversion uses pressure data exclusively from the overlying aquifer than when the inversion uses pressure data from both the overlying aquifer and the storage formation. This reiterates the importance of pressure monitoring in the storage formation. The calibrated model parameters in the case where $k_L = 10^{-9} \text{ m}^2$ are in general less accurate than those in the cases where $k_L = 10^{-7} \text{ m}^2$ and $k_L = 10^{-8} \text{ m}^2$, and the accuracy is poorer when using data exclusively from the overlying aquifer than when using the data from both the storage formation and the overlying aquifer.

It should be noted that for these cases with smaller leaky well permeabilities, the inversion of anomalous leakage signals induced by the leaky well is still achieved, because the leakage signals are sufficiently distinct. However, when the leaky well permeability is very small (e.g., $k_L = 10^{-10} \text{ m}^2$), the inversion will be limited by low detectability of leakage signals (Jung et al. [19]). In fact, the pressure anomaly in the overlying aquifer at the monitoring wells available in this study barely reaches 0.1 bar by six years. Since the leakage signal itself is not strong enough to affirm the presence of a leakage pathway, the inversion analysis cannot be performed.

These results show that the detectability of leakage signals (strength of signal anomalies relative to data noise and model uncertainties) is the key factor for locating and characterizing unknown leakage pathways via the developed pressure-based detection methodology. Therefore, to improve the accuracy of model prediction and leakage detection for moderate leakage events, either stronger anomalous signals should be detected (i.e., having monitoring wells close to leakage pathways) or additional monitoring data that could complement pressure monitoring data (e.g., surface deformation data) should be available.

3.5. Sensitivity Analysis of the Leaky Well Location

We consider several locations of the leaky well to examine the effect of the configuration of the injection wells, leaky wells, and monitoring wells on leakage detection, while keeping all the other conditions unchanged from Cases A3 and B3. Figure 10a shows the distance of the estimated leaky well location from the actual well location, as a function of monitoring data from the overlying aquifer only. For cases of [1 km, 0 km], [3 km, 0 km], and [-2 km, 0 km], the leakage detection converges to the true well locations after 3-year monitoring data are used. For the other two cases of [2 km, 0 km] and [4 km, 0 km], however, the convergence of leakage detection is not achieved after 6 years of monitoring. It seems that the accuracy and convergence speed of leakage detection is not necessarily related to the distance of the leaky well from the injection well. For instance, the estimated leaky well location is more than 0.5 km away from the actual location with the 3-year pressure monitoring for the case of [2 km, 0 km], while it is only 27 m away for the case of [-2 km, 0 km]. In the former case, the pressure anomalies at MW2 are noticeably smaller than those at IW and MW1, providing the distance-sensitive information for the inversion, while the magnitudes of pressure anomalies at IW, MW1, and MW2 are similar in the latter case. This indicates that the configuration of the monitoring wells relative to the leaky well and the injection well has a great influence on the accuracy of leakage detection.

When the pressure data in both the overlying aquifer and the storage formation are used, the leakage detection quickly converges to the true well location with monitoring time for all cases except the [4 km, 0 km] case. This insensitivity of leakage detection to the leaky well location and the relative configuration of the monitoring wells can be attributed to the additional pressure data from the storage formation in the inverse modeling. Note that for the [4 km, 0 km]

case, the leakage detection fails in both cases of data availability, because of the small pressure anomalies at all monitoring wells used in the inverse modeling.

4. Concluding Remarks

Pressure-based methods of leakage detection have been shown to be promising for “early” detection of large brine leaks in geological carbon storage. However, the prerequisite for success in using these methods is a suitable forward model, one that can capture the hydrogeological features of a storage site and accurately predict pressure signals at monitoring wells in the storage formation and overlying aquifers. That is, detection of CO₂ or brine leakage depends not only on the sensitivity of system responses (monitored pressure) to leakage parameters (location and permeability of leakage pathways), but also on the accuracy of forward model parameters. Since these model parameters are always uncertain even after calibrated using field data, the impact of these uncertainties on leakage detection should be assessed and reduced.

In this study, we explored the impact of model uncertainties on detecting leakage signals (i.e., pressure anomalies) and locating leakage pathways, and proposed a methodology for combining leakage detection with model recalibration to reduce such impact. Our study shows that when caprock permeability is relatively small, with small diffuse leakage through the caprock, anomalous pressure induced by a leaky well can be detected in the overlying aquifer, with minimal impact of uncertainties in the model parameters, as long as the permeability of the leaky well is sufficiently large ($k_L > 10^{-9} \text{ m}^2$). However, with error in the uncertain model parameters, inversion methods using pressure data exclusively from the overlying aquifer may not successfully locate the unknown leaky well. On the other hand, uncertainties in model parameters, particularly the parameters of the storage formation, can significantly affect the

model prediction of pressure changes in the storage formation. Due to this strong sensitivity, pressure monitoring data from the storage formation may not be adequate to detect leakage signals, but are very useful in recalibrating the model parameters and thereby improving leakage detection. The combined use of pressure monitoring data from both the storage formation and the overlying aquifer enables the leakage parameters to be accurately estimated and the model parameters to be recalibrated, making early leakage detection possible. Our further sensitivity analysis, on the permeability and location of the leaky well and the magnitude of the model parameter uncertainties, confirm that the proposed methodology of leakage detection and model recalibration using pressure data from both aquifers works well for a range of conditions.

The proposed methodology also shows that the parameterization of some uncertain model parameters in inverse modeling can greatly reduce the misfits between the observed and calculated pressures, by avoiding the systematic errors commonly occurring in the forward model and biased parameter estimates on leakage parameters. As demonstrated, a detailed residual analysis of multiple complementary data may help identify the systematic errors that produce non-Gaussian errors in pressure misfits, even for most practical storage sites.

It should be noted that locating unknown leakage pathways is feasible only if anomalous pressure signals are sufficiently strong relative to data noise and model uncertainties. When the leakage rate is small and the signal is not distinct, inverse analysis for leakage detection will not be effective, and the impact of model uncertainties could be substantial.

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Table 1. The “true” geometric and hydrogeologic parameters of the storage formation (SF), the aquitard, and the overlying aquifer (OA), as well as the base-case values of the “imperfect” forward model

Parameters		“True” values	“Imperfect” model values
Thickness of SF (m)	B_1	60	
Thickness of OA (m)	B_2	60	
Permeability of SF (m^2)	K_1	10^{-13}	9.78×10^{-14}
Permeability of OA(m^2)	K_2	10^{-13}	1.5×10^{-13}
Specific storativity of SF(m^{-1})	S_1	1.88×10^{-6}	2.14×10^{-6}
Specific storativity of OA(m^{-1})	S_2	1.88×10^{-6}	2.82×10^{-6}
Thickness of aquitard (m)	B'	100	
Permeability of aquitard (m^2)	K'	10^{-19}	
Specific storativity of aquitard (1/m)	S'	1.47×10^{-6}	

Table 2. Various scenarios and strategies of inverse modeling considered for leakage detection and model recalibration

Case		Pressure data		Parameters estimated			Leaky well permeability (m^2)
		OA	SF	Leakage parameters	K_1 & S_1	K_2 & S_2	
A	1	✓		✓			10^{-7}
	2					✓	
	3				✓	✓	
	4				✓	✓	10^{-8}
	5				✓	✓	10^{-9}
B	1	✓	✓	✓			10^{-7}
	2					✓	
	3				✓	✓	
	4				✓	✓	10^{-8}
	5				✓	✓	10^{-9}

Table 3. The estimated X- and Y-coordinates of the leaky well with the 3-year period of monitoring data

Case		X (m)		Y (m)	
		Estimate	Marginal standard deviation	Estimate	Marginal standard deviation
A	3	2.5×10^3	2.0×10^3	-2.9×10^2	1.1×10^3
	4	2.5×10^3	3.5×10^2	-2.9×10^2	1.8×10^2
	5	2.6×10^3	3.4×10^3	-4.4×10^2	1.9×10^3
B	3	2.0×10^3	1.2×10^2	4.4	50.2
	4	1.9×10^3	1.5×10^2	29.1	66.3
	5	1.7×10^3	5.7×10^2	1.2×10^2	3.0×10^2

Figure 1. An idealized storage system, consisting of a storage formation, a caprock (aquitard), and an overlying aquifer, bounded by impervious boundaries at the top and the bottom, with an injection well (IW) and a leaky well (LW) (2 km apart), in (a) the vertical section along the injection well and the leaky well, and (b) a plan view with the locations of two monitoring wells (MW1 and MW2) used in the sensitivity analysis and inverse modeling.

Figure 2. Comparison between the measured (red line) and expected (blue line) pressure increases at MW2 in (a) the storage formation (SF) and (b) the overlying aquifer (OA). The shaded areas from the innermost (darkest blue) to the outermost (lightest blue) represent 95% FOSM error bands for 10%, 20%, and 50% uncertainties in the model parameters of interest, respectively. These FOSM error bands in OA are too small to be visibly distinguished from the expected pressure.

Figure 3. Convergence of the estimated leakage parameters and model parameters: (a) leaky well location, (b) leaky well permeability, (c) K_2 , and (d) S_2 , using the incremental pressure monitoring datasets of 1, 2, 3, and 6 years in Case A2. Also shown are the marginal standard deviations of the estimates. The number on each point in (a) represents the end year of the incremental monitoring period. The asterisk symbol (*) means that the marginal standard deviation of the estimate is omitted, because it is too large to be included in the figure.

Figure 4. Convergence of the estimated leaky well location using the incremental pressure monitoring datasets of 1, 2, 3, and 6 years in (a) Case A3 and (b) Case B3. Also shown are the marginal standard deviations of the estimated well location. The number on each point represents the end year of the incremental monitoring period.

Figure 5. Convergence of the estimated leaky well permeabilities using the incremental pressure monitoring datasets of 1, 2, 3, and 6 years in (a) Case A3 and (b) Case B3.

Figure 6. Convergence of the estimated model parameters (K_1 , K_2 , S_1 , and S_2) using the incremental pressure monitoring datasets of 1, 2, 3, and 6 years in Case A3 (left column) and Case B3 (right column): (a) and (b) K_1 , (c) and (d) K_2 , (e) and (f) S_1 , and (g) and (h) S_2 .

Figure 7. Transient dimensionless sensitivity coefficients to the model parameters for pressure at MW2.

Figure 8. Distribution of the estimated leaky well location from 20 initial leakage parameter sets generated using Latin Hypercube Sampling: (a) Case A3 and (b) Case B3. The asterisk symbol (*) indicates the true leaky well location.

Figure 9. Convergence of the estimated leakage parameters using the incremental pressure monitoring datasets of 1, 2, 3, and 6 years with smaller leaky well permeabilities: (a) and (b) the distance of the estimated leaky well location from the true location and (c) and (d) the estimated leaky well permeability.

Figure 10. Convergence of the estimated leaky well location using the incremental pressure monitoring datasets of 1, 2, 3, and 6 years for different well locations: (a) using the pressure data in the overlying aquifer exclusively, and (b) using the pressure data in both the overlying aquifer and the storage formation.