Characterizing CO₂ plumes in deep saline formations: Comparison and joint evaluation of time-lapse pressure and seismic tomography

Linwei Hu¹, Joseph Doetsch¹, Ralf Brauchler², and Peter Bayer³

ABSTRACT

Monitoring the migration of sequestered CO₂ in deep heterogeneous reservoirs is inherently difficult. Geophysical methods have been successfully used, but flow conditions are only indirectly linked to the measured properties. Besides geophysical methods, pressure tomography (PT) is proposed as an alternative method to depict the structure of deep saline formations for CO₂ sequestration and to continuously delineate CO₂ plumes. In contrast to more established geophysical measurements, pressure transients are directly related to flow properties, which allows for the estimation of permeability. We investigate the influence of aquifer heterogeneity on PT performance, and we compare the PT results to crosshole seismic tomography (ST). Multilevel fluid injections and high-frequency P-wave pulses are induced in a simulated deep borehole, and the recorded signals at another well are processed by a traveltime inversion scheme. The reservoir structure is inferred by clustering the inverted hydraulic diffusivity prior to CO₂ injection, and the plume distribution is determined by clustering the tomograms of the inverted mixed-phase diffusivity difference and P-wave velocity difference. The clustered structures are then used for zonal calibration to acquire the saturation within the plumes. Modeling results indicate that PT provides clearer structural information on the CO₂-free aquifer due to its direct linkage to permeability. However, the plume depicted by PT can be ambiguous, whereas ST is less sensitive to the prevailing heterogeneity of permeability at postinjection and can thus image the plume more clearly. PT and ST can be complementary to each other through the joint clustering to improve plume shape identification and estimation of spatial CO₂ saturation.

INTRODUCTION

A recent report (Global CCS Institute, 2015) pointed out that CO₂ capture and storage (CCS) is the only countermeasure to lessen greenhouse gas emissions in a significant scale from industrial processes. Among various geologic storage media, deep saline aquifers are considered sound formations for sequestering CO₂, in which CO₂ is injected and stored in a supercritical state, with a large storage capacity. Safe disposal of CO₂ in saline aquifers demands favorable storage conditions, such as high porosities in the storage medium and an impermeable caprock. In case of unfavorable conditions, such as seal imperfections, preexisting faults or overpressure-induced fractures, an evolving CO₂ plume can escape toward shallower formations and, which might have an adverse impact on shallow groundwater quality (Wang and Jaffe, 2004). For minimizing the risk of CO₂ leakage and for formulating effective remediation strategies, appropriate site investigation and CO₂ plume monitoring techniques are required.

Geophysical exploration methods are extensively applied for depicting stratigraphy and CO₂ plume geometry in deep saline formations. Their applicability has been demonstrated at several CO₂ storage sites, such as Sleipner (Chadwick et al., 2010), Snøhvit (Shi et al., 2013), In Salah (Ringrose et al., 2009), Ketzin (Zhang et al., 2012), Cranfield (Doetsch et al., 2013), Frio brine pilot (Daley et al., 2011), and Nagaoka (Nakajima et al., 2014). The most common geophysical approaches for monitoring of CO₂ plumes are seismic surveys, electrical methods, and gravity measurements. The induced CO₂ phase alters the effective physical properties of the storage formation (e.g., seismic velocity, electrical resistivity, and density) over time, which is examined by time-lapse data sets.
Among established geophysical techniques, active seismic surveys are most commonly used. Usually, seismic measurements are conducted prior to CO₂ injection to obtain baseline information, and then measurements are repeated multiple times after CO₂ injection. CO₂ plume evolution is monitored by the traveltime delay or amplitude anomalies from different vintages. Depending on the configurations of sources and receivers, seismic-based approaches can be classified into the surface seismic survey, surface-downhole monitoring, and crosshole measurement. Typically, a 2D or 3D surface seismic survey is conducted for large-scale problems, and its spatial resolution is limited. In the case that the CO₂ layer thickness is less than the resolution, the uncertainty for evaluating the CO₂ mass becomes significant (Ivanic et al., 2015). Surface-downhole monitoring includes two different configurations, namely, vertical seismic profiling and moving-source-profiling. It is used for estimating the vertical expansion of the CO₂ plume and for improving the vertical resolution to complement surface seismic methods. For crosshole measurements, the seismic sources and receivers are installed in different boreholes, and the experiments are performed for obtaining insight in the reservoir and mapping CO₂ plumes between the borehole pair. The crosshole variant can provide high-resolution information between the boreholes, which are typically separated by a distance of tens to hundreds of meters. The main inversion algorithms for crosshole seismic tomography include travelt ime-based and full-waveform inversion (Pratt and Shipp, 1999).

Although successful at imaging migrating CO₂, two difficulties have been identified for established geophysical approaches. First, petrophysical models have to be formulated for analyzing the stored CO₂ in the subsurface, which is challenging mainly due to the uncertainty in model parameters. For instance, for a petrophysical model that couples a patchy fluid-saturation model, the patch size of CO₂ is difficult to determine precisely in practice (Daley et al., 2011). Second, favorable conditions are required for the repeatability of geophysical experiments, such as a high contrast of reservoir properties, identical experimental setups at different times, and low noise levels. Not having these ideal conditions can lead to artifacts during inversion of time-lapse data sets, or during conversion of geophysical parameters into CO₂ saturation.

As a complementary method for characterizing reservoir and monitoring CO₂ plumes during the early time of storage, as well as for detecting CO₂ leakage, pressure-based methods have recently been suggested. Because pressure directly relates to flow conditions and travels much faster than a CO₂ plume, pressure-based methods are recognized as an appropriate approach for evaluating the flow properties before and during CO₂ injection (Doughty et al., 2008; Wiese et al., 2010), as well as for early detection of CO₂ leakage (Birkholzer et al., 2009; Sun et al., 2016). However, few pressure-based methods can provide a spatial image of CO₂ plumes. Several available techniques can identify the plume shape to a certain degree. For instance, the upcoming of a CO₂ plume near the well or a CO₂ front can be inferred by some analytical or semianalytical solutions (Nordbotten et al., 2004; Chhan et al., 2011). The approach of Martínez-Landa et al. (2013) can estimate the proximal width of a CO₂ plume by analyzing pressure measurements in a single borehole. Nevertheless, most of these methods are based on the assumption that the reservoir is homogeneous, neglecting most of the involved physicochemical CO₂ transport processes and potential reservoir heterogeneity.

For resolving spatial variability of flow properties, hydraulic tomography (HT) was proposed by Gottlieb and Dietrich (1995) and has seen significant development since its introduction. Compared with conventional hydraulic/pressure tests, HT can delineate the spatial distribution of hydraulic parameters. Feasibility of HT has been studied in porous (Yeh and Liu, 2000; Hu et al., 2011; Cardiff et al., 2013; Paradis et al., 2015) and fractured media (Hao et al., 2008; Illman et al., 2009; Sharman et al., 2012; Zha et al., 2014) by numerical simulations (Zhu and Yeh, 2005; Jiménez et al., 2013; Schwede et al., 2014), laboratory experiments (Brauchler et al., 2007; Liu et al., 2007; Zhou et al., 2016), and field tests (Straface et al., 2007; Brauchler et al., 2013; Jiménez et al., 2015; Zha et al., 2015). The application scale of HT can vary from several meters to kilometers. The rationale of HT is conceptually analogous to geophysical tomographic techniques. In lieu of using geophysical sources (e.g., active or passive seismic excitations), HT or more generally pressure tomography (PT), requires a series of pressure stimulations in a tomographic array. Tomographic data sets are derived by conducting fluid injection or extraction tests in different intervals at one well (sources), with pressures measured at the response well at different observation levels (receivers). The pressure measurements are used for reconstructing hydraulic parameter heterogeneity by different inversion techniques, such as the sequential linear estimator (Yeh and Liu, 2000), quasilinear Bayesian geostatistical method (e.g., Nowak and Cirpka, 2004), ensemble Kalman filter (Schöninger et al., 2012), and the travelt ime-based approach (Brauchler et al., 2003). Most of the available HT inversion techniques are based on predefined geostatistical models with known or presumptive correlation lengths and variances of hydraulic parameters. These models are taken to generate hydraulic parameter fields and then to simulate the groundwater flow during an iterative inversion procedure. On the contrary, the time-lapse-based inversion approach does not require geostatistical estimates, and the inversion process does not involve flow modeling. Instead, the groundwater flow equation is approximated to an eikonal equation, which can be solved in a computational efficient way by the ray-tracing technique (Jackson and Tweeton, 1996). Structural information of the aquifer is then inferred from reconstructed diffusivity tomograms. To date, HT methods are mainly applied to the “static” single-phase flow condition (i.e., hydraulic parameters are considered not varying with time) and shallow aquifers. Hu et al. (2015) introduce the concept of “time-lapse PT.” It is proposed to use the travelt ime-based inversion strategy for identifying an evolving CO₂ plume in a homogeneous deep saline aquifer. In this case, replacement of the local brine by CO₂ induces an effect on the observed flow properties over time. Considering CO₂ and brine as a phase mixture, the mixed-phase diffusivity can be changed by up to two orders of magnitude due to the high compressibility of CO₂. Spatial diffusivity variations can be inferred from inspecting pressure transients at different times. Thus, the inversion of traveltimes derived from pressures offers time-lapse information of the plume. Furthermore, CO₂ saturation can be estimated by including a storativity-saturation model. In comparison with geophysical approaches, PT is not related to the CO₂ plume or patch size because it considers CO₂ and brine as a mixture (Hu et al., 2015). The properties of the CO₂–brine mixture are estimated from the density, viscosity, and temperature.

As mentioned above, previous study on time-lapse PT in Hu et al. (2015) was based on a homogeneous aquifer, without considering data noise and formation heterogeneity. The main objective of this study is to further explore the inversion performance of PT in heterogeneous formations and to compare the results with those from cross-
hole traveltime seismic tomography. In contrast to seismic traveltime tomography, two-phase PT is unusual for such conditions. Due to the direct and more deterministic relationship between CO₂ saturation and hydraulic properties, such as storativity, it is anticipated that PT can complement existing seismic approaches through the improved estimation of saturation. In the following section, we first briefly introduce the two tomographic inversion concepts used in a time-lapse manner. These are examined together in scenarios with different degrees of heterogeneity. For simplification, the expression “seismic tomography” mentioned in this study refers to the traveltime-based seismic tomography method.

**METHODOLOGY**

**Overview of the methodology**

In this work, the inversion methodology is tested using real site-based synthetic models. As a basis, we set up three scenarios for this synthetic model, which differ with respect to heterogeneity and model parameters. Subsequently, we investigate the changes of the mixed-phase diffusivity and P-wave velocity induced by CO₂ injection. The relationship between either diffusivity or velocity and CO₂ saturation provides the basis for the inversion.

Figure 1 presents the general flowchart of the forward simulation, inversion, and calibration procedures. The CO₂ sequestration process is simulated with a fully coupled two-phase simulator, PFLOTRAN (Hammond et al., 2014). PT and ST data acquisition is simulated prior to and after CO₂ injection. The derived hydraulic and seismic travel times are inverted separately to reconstruct the spatial distribution of diffusivity and velocity. The structure of the reservoir and the geometry of the CO₂ plume are obtained by individual or joint clustering (JT) of the tomograms at different times. Ultimately, we acquire the CO₂ saturation through the calibrated specific storage based on the clustered structure, and the calibration is conducted in a single-phase emulator. In the following sections, each step is explained in detail.

**Problem setup**

**Virtual site and three scenarios**

A simplified 2D cross-sectional model based on a virtual site is used for testing our method (as in Hu et al., 2015, 2016). The simulated regime is located at a depth of 1600 m, constituting three components: a storage reservoir, an overlying caprock, and an underlying bottom seal (Figure 2). The thickness of the reservoir and the two low-permeability components are 15 and 30 m, respectively. The reservoir is composed of sandstone, and the caprock and bottom seal are shale formations with very low permeability. We assume that initially no CO₂ exists in the reservoir. For improving computing efficiency, the caprock and seal bottom are assumed to be impervious and considered to be no-flow boundaries during flow simulation. The lateral extension of the entire model is 580 m, which is bounded with constant hydrostatic pressure at the east and west sides. The pressure at the model bottom is 14.76 MPa, with a pressure gradient of 0.01 MPa/m. The aquifer is initially fully saturated with brine, and its temperature and salinity are 67°C and 67 g/l, respectively. Under these conditions, CO₂ is injected and sequestered in a supercritical state.

**Figure 1.** Schematic flowchart of the forward simulation, inversion, and calibration strategy.

**Figure 2.** Conceptual model and source-receiver configurations of PT and ST. The left figure only presents part of the model. For details of the full model, the reader is referred to Figure A-1. Three variants of aquifer heterogeneity, one homogeneous, and two two-layer scenarios are considered. Note that sources and receivers for ST are illustrated schematically, and their true numbers are much higher (76).
The fluid injection well is at the center of the model (see Appendix A), fully penetrating the entire aquifer. An accompanying observation well is located 50 m away. The distance between the well pair is comparable to several practical injection sites such as Ketzin (Wiese et al., 2010) and Heletz (Niemi et al., 2016). The lateral grid size of the model increases telescopically, ranging from 0.09 m at the injector to 40 m at the side boundaries. The vertical discretization of the model is 0.6 m. Finally, the model is discretized by 287 and 75 grid cells in the horizontal and vertical directions, with 21,525 grid cells in total (see Appendix A).

For PT, five sources and five receivers are used. These are screened at injection and observation wells in the reservoir (Figure 2, red circles and crosses). The length of each source (fluid injection interval) is 0.6 m, and the distance between two adjacent sources or receivers is 3.6 m. To formulate a tomographic configuration, fluid is injected sequentially at the five sources. During each injection, pressure fluctuations at all five receivers are monitored simultaneously. Sources and receivers for ST are also assumed to be located at the two wells (sources are in the injection well, and receivers are in the observation well). The distribution of source-receiver configurations for ST experiments is much denser (Figure 2, red circles and crosses). Here, 76 sources and 76 receivers are installed referring to Ajo-Franklin et al. (2013). The number of seismic sources and receivers is much more than pressure sources and receivers. This is because the installation of borehole isolation systems (e.g., packers, multichamber systems) is needed for PT. Seismic sources and receivers are not only in the reservoir but are also in the caprock and bottom seal, which allows locating a reservoir prior to conducting pressure tests. This also yields larger angle tomographic arrays to improve spatial resolution. The vertical distance between two adjacent seismic sources or receivers is 1 m.

Three different scenarios are defined for exploring the influence of reservoir heterogeneity. In the first scenario, the reservoir is perceived as a homogeneous and isotropic aquifer. In the second and third scenarios, the reservoir consists of two “perfect” homogeneous and isotropic layers (Figure 2). The thickness of the upper and lower layers is 5 and 10 m, respectively. We discriminate these two scenarios by assigning different values of model parameters (permeability, porosity, and other parameters calculated by these) for the two layers. In the next sections, we refer to these three scenarios as “homogeneous,” “2layers_A,” and “2layers_B,” with 2layers_A having the higher permeability in the upper layer and 2layers_B in the lower layer.

### Model parameters

Model parameters are summarized in Tables 1 and 2. The values of model parameters for the homogeneous scenario are the same as in Hu et al. (2015). The intrinsic permeability $k$, porosity $\phi$, and rock density $\rho_r$ are $1 \times 10^{-13}$ m$^2$, 0.23, and 2550 kg/m$^3$, respectively. Rock density $\rho_r$ is estimated by the rock-matrix density $\rho_m$, brine density $\rho_w$, and porosity $\phi$ through equation B-4. The values of $\rho_m$ and $\rho_w$ are 3000 and 1052 kg/m$^3$, respectively.

The characteristic functions of permeability-saturation and capillary pressure-saturation are based on the Brooks-Corey-Burdine model (Burdine, 1953; Brooks and Corey, 1964). The pore-size distribution $\lambda$ and entry pressure $P_d$ are set to 0.76 and 4000 Pa, respectively. In the scenario 2layers_A, the permeability of the top layer is $1 \times 10^{-12}$ m$^2$ and the bottom layer is $1 \times 10^{-13}$ m$^2$. The corresponding $\phi$ is set to 0.28 and 0.22, which is calculated from $k$ according to an empirical equation for sandstone (Schön, 2011)

$$k = 3.95 \times 10^{-17} \cdot \exp(35.77\phi).$$

(1)

The pore-size distribution $\lambda$ is set to a constant value of 1.5 for the entire model. The $P_d$ is calculated by the Leverett scaling function (Leverett, 1941), which explains the difference in $k$, $\phi$, and $P_d$ between a reference and an unknown media

$$P_d = \sqrt{\frac{h_{\text{sat}}}{\phi}} P_{d,\text{ref}}.$$  

(2)

### Table 1. Parameter values of numerical model based on a virtual site.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intrinsic permeability of the caprock and seal bottom ($k_{\text{cap}}$ and $k_{\text{seal}}$)</td>
<td>$1 \times 10^{-19}$ m$^2$</td>
<td>Wang and Small (2014)</td>
</tr>
<tr>
<td>Porosity of the caprock and seal bottom ($\phi_{\text{cap}}$ and $\phi_{\text{seal}}$)</td>
<td>0.05</td>
<td>Estimated</td>
</tr>
<tr>
<td>Salinity</td>
<td>67 g/l</td>
<td>Estimated</td>
</tr>
<tr>
<td>P-wave velocity of the caprock and seal bottom ($V_{\text{cap}}$ and $V_{\text{seal}}$)</td>
<td>3500 m/s</td>
<td>Mavko et al. (2009)</td>
</tr>
<tr>
<td>P-wave velocity of CO$_2$ ($V_n$)</td>
<td>292.9 m/s</td>
<td>NIST</td>
</tr>
<tr>
<td>Initial datum pressure ($P_d$)</td>
<td>14.76 MPa</td>
<td>Erlström et al. (2011)</td>
</tr>
<tr>
<td>Initial average brine density ($\rho_w$)</td>
<td>1052 kg/m$^3$</td>
<td>Duan et al. (2008)</td>
</tr>
<tr>
<td>Initial average brine viscosity ($\mu_w$)</td>
<td>$4.2 \times 10^{-4}$ Pa s</td>
<td>Span and Wagner (1996)</td>
</tr>
<tr>
<td>Average isothermal compressibility of brine ($c_w$)</td>
<td>$3.8 \times 10^{-10}$ 1/Pa</td>
<td>Duan et al. (2008)</td>
</tr>
<tr>
<td>Average CO$_2$ density ($\rho_n$)</td>
<td>520 kg/m$^3$</td>
<td>Span and Wagner (1996)</td>
</tr>
<tr>
<td>Average isothermal compressibility of CO$_2$ ($c_n$)</td>
<td>$9 \times 10^{-8}$ 1/Pa</td>
<td>Span and Wagner (1996)</td>
</tr>
<tr>
<td>Average CO$_2$ viscosity ($\mu_n$)</td>
<td>$3.9 \times 10^{-5}$ Pa s</td>
<td>Fenghour et al. (1998)</td>
</tr>
<tr>
<td>Bulk modulus of dry frame rock ($G_{\text{dry}}$)</td>
<td>$3 \times 10^9$ Pa</td>
<td>Caspary et al. (2011)</td>
</tr>
<tr>
<td>Bulk modulus of rock matrix ($G_n$)</td>
<td>$3.7 \times 10^{10}$ Pa</td>
<td>Makovsky et al. (2009)</td>
</tr>
<tr>
<td>Shear modulus of saturated rock ($N_{\text{sat}}$)</td>
<td>$2.5 \times 10^{10}$ Pa</td>
<td>Makovsky et al. (2009)</td>
</tr>
</tbody>
</table>
where the values of the reference permeability \( k_{\text{ref}} \), porosity \( \phi_{\text{ref}} \), and entry pressure \( P_{\text{entry}} \) are \( 1 \times 10^{-13} \) m², 0.25, and 8700 Pa, respectively (Rasmusson et al., 2014). In the scenario 2layers_B, the values of the model parameters are switched between the layers.

More realistic values of the aforementioned flow and elastic model parameters can be estimated by laboratory experiments on core samples taken from the storage site.

**Relationship between flow properties and fluid saturation.**

The initial single-phase hydraulic conductivity \( K_w \) and specific storage \( S_{sw} \) of the aquifer are related to the intrinsic permeability \( k \), porosity \( \phi \), brine density \( \rho_w \), and viscosity \( \mu_w \) in equations 3 and 4:

\[
K_w = \rho_w g \left( \frac{k}{\mu_w} \right),
\]

\[
S_{sw} = \rho_w g (\phi c_w),
\]

where \( \rho_w, \mu_w, \) and \( c_w \) are assumed constant by averaging the values within the reservoir. In equation 4, the rock compressibility is neglected because the value of a sandstone is usually one to two orders of magnitude smaller than that of CO₂ given the pressure and temperature conditions in this study (Vilarrasa, 2012). Furthermore, hydraulic diffusivity of the initial CO₂-free formation \( D_{\text{pre}} \) is defined as the ratio between \( K_w \) and \( S_{sw} \):

\[
D_{\text{pre}} = \frac{k}{\phi c_w \mu_w}.
\]

The transferred values of \( D_{\text{pre}} \) for the three scenarios are listed in Table 2.

After CO₂ injection, considering CO₂ and brine as a phase mixture, the mixed-phase diffusivity \( D_{\text{post}} \) is determined by the intrinsic permeability and porosity, the fluid properties of brine and CO₂ (density, viscosity, and compressibility), the relative permeability, and the saturation of the two phases (Hu et al., 2016):

\[
D_{\text{post}} = \frac{K}{S_n} = \left( \frac{k}{\phi} \right) \left[ \frac{k_{\text{rel, brine}}} {\mu_w} \rho_w + \frac{k_{\text{rel, CO2}}} {\mu_w} \rho_n} \right] \left( S_{sw} \rho_w + S_s \rho_n \right).
\]

where \( k_{\text{rel, brine}} \) and \( k_{\text{rel, CO2}} \) are the relative permeability of brine and CO₂, respectively. Equations 5 and 6 indicate that the diffusivity varies with CO₂ saturation. Figure 3 displays three different \( D - S_n \) models for the three scenarios. Model 1 (black solid line) is for the homogeneous scenario. Models 2 (red solid line) and 3 (blue solid line) are for the scenarios 2layers_A and 2layers_B, relating to the two layers. Model 2 is for the low-permeability layer, and model 3 is for the high-permeability layer (Figure 2 and Table 2). For all models, diffusivity is smaller than that prior to CO₂ injection, which provides a basis for the inversion of diffusivity changes. However, it is also noticeable that, due to the nonmonotonic \( D_{\text{post}} - S_n \) relationship, one diffusivity value could correspond to two different saturation values, except when the CO₂ saturation is very low \( (S_n < 0.05-0.1) \). This could introduce uncertainties for estimating of CO₂ saturation directly from diffusivity.

Relationship between the P-wave velocity and fluid saturation. To infer CO₂ saturations and their temporal or lateral variation from seismic-velocity tomograms or to calculate the change in seismic velocity due to a change in CO₂ saturation, a specific relationship between velocity and CO₂ saturation needs to be assumed. Usually, such a relationship is determined by laboratory tests of field samples (Vanorio et al., 2011). In this study, we apply the Gassmann-Wood theory (Wood, 1941; Gassmann, 1951), which is valid at low seismic frequencies, to characterize seismic velocity in a rock that is saturated with a fluid mixture (Casparsi et al., 2011). The Gassmann-Wood rock-physics model was chosen as one possibility out of the several realistic theories for different CO₂ patch sizes and local CO₂ distribution. Although the choice of rock-physics model and its calibration is critical for field studies to get a realistic estimation of CO₂ saturation, the choice of rock-physics model is somewhat arbitrary for synthetic studies. The parameters used

![Figure 3. Diffusivity D versus CO₂ saturation S_n. Model 1: homogeneous scenario; model 2: layer with small permeability, k; and model 3: layer with large k.](image-url)

**Table 2. Parameter values of the reservoir in the three scenarios.** Hydraulic conductivity, specific storage, diffusivity, entry pressure, and velocity are calculated based on the intrinsic permeability and porosity.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Zone</th>
<th>( k ) (m²)</th>
<th>( \phi ) (%)</th>
<th>( K_w ) (m/s)</th>
<th>( S_{sw} ) (1/m)</th>
<th>( D ) (m²/s)</th>
<th>( P_{\text{entry}} ) (Pa)</th>
<th>( V ) (m/s)</th>
<th>( c_p ) (J/kgK)</th>
<th>( \kappa_{\text{wet}} ) (W/mK)</th>
<th>( \kappa_{\text{dry}} ) (W/mK)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Homogeneous</td>
<td></td>
<td>1 \times 10^{-13}</td>
<td>0.23</td>
<td>2.46 \times 10^{-6}</td>
<td>9.1 \times 10^{-7}</td>
<td>2.7</td>
<td>4000</td>
<td>4465</td>
<td>930</td>
<td>3</td>
<td>4.5</td>
</tr>
<tr>
<td>2layers_A</td>
<td></td>
<td>1 \times 10^{-12}</td>
<td>0.28</td>
<td>2.46 \times 10^{-5}</td>
<td>1.1 \times 10^{-6}</td>
<td>22.36</td>
<td>2932</td>
<td>4579</td>
<td>860</td>
<td>2.5</td>
<td>4.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2 \times 10^{-13}</td>
<td>0.22</td>
<td>2.46 \times 10^{-6}</td>
<td>8.6 \times 10^{-7}</td>
<td>2.86</td>
<td>8143</td>
<td>4454</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2layers_B</td>
<td></td>
<td>1 \times 10^{-13}</td>
<td>0.22</td>
<td>2.46 \times 10^{-6}</td>
<td>8.6 \times 10^{-7}</td>
<td>2.86</td>
<td>8143</td>
<td>4454</td>
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<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>1 \times 10^{-12}</td>
<td>0.28</td>
<td>2.46 \times 10^{-5}</td>
<td>1.1 \times 10^{-6}</td>
<td>22.36</td>
<td>2932</td>
<td>4579</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
for the calculation of the velocities are given in Table 1, and for the equations used, see Appendix B.

The initial velocity of the storage formation (Table 2) is calculated based on equation B-4 by assuming a CO₂-free reservoir ($S_n = 0$). The velocity difference $\Delta V$ is defined as the difference between the velocity before ($V_{pre}$) and after ($V_{post}$) CO₂ injection

$$\Delta V = V_{post} - V_{pre}. \quad (7)$$

Analogous to the relationship of $D = S_n$ (Figure 3), $\Delta V - S_n$ is plotted in Figure 4 for five different models (see above). Similar to $D - S_n$, the relationship between $\Delta V$ and $S_n$ is also not monotonic. Slight differences among these models are because of the different porosities for each of them. For all models, initial velocity declines due to the CO₂ injection. The $\Delta V$ decreases steeply along with the increased $S_n$ because $S_n$ is smaller than 0.15–0.25. When $S_n$ is larger than 0.15–0.25, $\Delta V$ increases gently along with $S_n$. The maximum and minimal velocity changes are shown in models 2 and 3, with the value of approximately −350 and −200 m/s, respectively. However, changes of $V$ along with $S_n$ are obviously smaller than those of $D$.

![Figure 4. Velocity difference $\Delta V$ versus CO₂ saturation $S_n$. Model 1: homogeneous scenario; model 2: layer with small $k$; and model 3: layer with large $k$.
](image)

### Table 3. Fluid-injection rate and duration of the test sequence.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Time</th>
<th>$Q_c$ (stage 2)</th>
<th>$Q_c$ (stage 4)</th>
<th>$T_{inj}$</th>
<th>$T_{rec}$</th>
<th>$T_{S1}$</th>
<th>$T_{S2}$</th>
<th>$T_{S3}$</th>
<th>$T_{S4}$</th>
<th>$T_{S5}$</th>
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<tr>
<td></td>
<td></td>
<td>(kg/s)</td>
<td>(kg/s)</td>
<td>(h)</td>
<td>(h)</td>
<td>(h)</td>
<td>(h)</td>
<td>(h)</td>
<td>(h)</td>
<td>(h)</td>
</tr>
<tr>
<td>Homogeneous</td>
<td>Short</td>
<td>0.01</td>
<td>0.02</td>
<td>120</td>
<td>240</td>
<td>4</td>
<td>4</td>
<td>4</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>Long</td>
<td>360</td>
<td>240</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>2layers_A</td>
<td>Short</td>
<td>0.01</td>
<td>0.01</td>
<td>120</td>
<td>40</td>
<td>0.7</td>
<td>0.7</td>
<td>0.4</td>
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<tr>
<td></td>
<td>Long</td>
<td>320</td>
<td>110</td>
<td>1.1</td>
<td>1.1</td>
<td>1.1</td>
<td>0.7</td>
<td>0.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2layers_B</td>
<td>Short</td>
<td>0.015</td>
<td>0.01</td>
<td>90</td>
<td>40</td>
<td>0.4</td>
<td>0.4</td>
<td>0.7</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Long</td>
<td>230</td>
<td>60</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
<td>1.1</td>
<td>1.1</td>
<td></td>
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</tr>
</tbody>
</table>

### Forward simulation

**Single-phase and two-phase flow simulation**

An open source code, PFLOTTRAN (Hammond et al., 2014), is used for the forward simulation of the single-phase and two-phase flow processes. For the forward simulation, we focus on the early-stage injection procedure. Residual trapping, chemical reactions with the rock matrix, and any geomechanical processes are neglected. The entire simulation is divided into four stages: baseline study (stage 1), CO₂ sequestration (stage 2), shut-in period (stage 3), and repetition of interference fluid injection tests (stage 4).

**Stage 1: Baseline study.**—At this stage, brine is injected from the bottom to the top sources in the borehole. For all three scenarios, each injection lasts for 2 h following a recovery period of 15 h, allowing the pressures to fall back to the initial hydrostatic condition. Pressure transients during the injections are used for depicting the structure of the aquifer, as well as the initial hydraulic conductivity and specific storage.

**Stage 2: CO₂ sequestration.**—At this stage, for each scenario, CO₂ is injected in a depth-integrated way, that is, at the injection well, it is injected over the entire aquifer. Injection durations are different (Table 3) to generate two plumes of different size. We discriminate the two injections by naming them the “short injection” and the “long injection,” respectively. The injection rate for the homogeneous scenario and 2layers_A is 0.01 kg/s, and for 2layers_B, it is 0.015 kg/s. The durations of the short and long injections for the three scenarios are listed in Table 3.

**Stage 3: Shut-in period.**—This stage is included for recovering the pressure to a quasisteady state. Durations are summarized in Table 3. In applications in practice, the experiments of the next stage can be prepared now.

**Stage 4: Repetition of interference fluid injections (stage 1).**—As soon as the pressure recovers to a quasisteady state, the CO₂ is injected sequentially comparably with previous brine injections. Pressure fluctuations are recorded at the observations for tomographic inversion. The injection rate for the homogeneous scenario, 2layers_A, and 2layers_B is 0.02, 0.01, and 0.01 kg/s, respectively. Durations of one injection and the following recovery period are set to be equivalent (Table 3).

### Hydraulic traveltime calculation

The pressure transients at the observation well derived from stages 1 (baseline) and 4 (after CO₂ sequestration) are used for calculating the hydraulic traveltimes, following the work of Brauchler et al. (2003) and Hu et al. (2015). For the continuous fluid injections, the hydraulic traveltime is determined by the first time derivative of the pressures. The time at which the maximum derivative appears at a receiver is defined as the peak time. However, here we use the early traveltime diagnostic for the inversion because it resolves the preferential flowpaths better than the late diagnostic. We use a 20% traveltime di-
Seismic-traveltime calculation

Based on the flow simulations discussed in the section “Single-phase and two-phase flow simulation,” seismic traveltimes (i.e., the first arrival times of P-wave pulses) are calculated for stage 1 (baseline) and stage 4 (after the first arrival times of P-wave pulses) are calculated for stage 1 and the parameters of Tables 1 and 2. The traveltimes are computed with 1% of Gaussian noise prior to the inversion, which is a realistic noise level for field studies (see, e.g., Ajo-Franklin et al., 2013).

Traveltime-based inversion

Inversion of hydraulic traveltimes

The inversion scheme of the hydraulic traveltimes is based on Brauchler et al. (2003). The hydraulic traveltime \( \hat{t} \) is related to the reciprocal value of the square root of hydraulic diffusivity \( \sqrt{D} \) by a line integral

\[
\hat{t} = \int_{s_1}^{s_3} \frac{ds}{\sqrt{D(s)}}. \tag{9}
\]

A staggering technique is applied to the inversion procedure for improving the resolution of the final tomogram, and to weaken the effect of positioning (Vesnaver and Bölhm, 2000; Somogyvári et al., 2016). The base model used for the inversion is discretized by five columns and four rows. By shifting the underlying model nine times in the horizontal direction and three times in the vertical direction during the inversion, the final tomogram reaches a resolution of 50 \( \times \) 16 cells.

Repetition of the inversion to the time-lapse data sets delineates the diffusivity distribution at different times. For removing the influence of the preinjection structure, we define the diffusivity difference \( \Delta D \) as follows:

\[
\Delta D = \log_{10} D_{\text{post}} - \log_{10} D_{\text{pre}} = \log_{10} \left[ \frac{c_n P_{\alpha w} (k_{rn} P_{\mu w} + k_{rn} P_{\mu w})}{(S_{ncw} + S_{nCn}) (S_{n} P_{\mu w} + S_{n} P_{\mu w})} \right]. \tag{10}
\]

The seismic traveltime data sets that are simulated for the different scenarios and at the different times (stage 1 and stage 4) are inverted using the algorithm of Doetsch et al. (2010b). It implements an Occam’s type inversion with stochastic regularization (chosen integral scales of \( I_1 = 16 \) m, \( I_2 = 8 \) m), in which the regularization strength is decreased until the data are fit to the error level. The assumed error on the traveltimes is 1%, in accordance with the noise contamination level, and all inversion results fit the data to that assumed error level.

The inversion results of the baseline inversion are used as starting and reference models for the time-lapse inversions of data acquired during the multilevel CO\(_2\) injections. A difference inversion scheme is being used that inverts for the changes to the baseline inversion result (Doetsch et al., 2010a), so that even small changes to the baseline model can be resolved and inversion artifacts are minimal. The results of the time-lapse inversions are analyzed and shown as a change in velocity compared with the baseline inversion result.

Clustering and zonal calibration

Time-lapse 1D and 2D clustering

The goals of clustering are to determine the baseline structure and to delineate the extent of CO\(_2\) plumes at different times. The unlabeled data of inverted tomograms are partitioned into homogeneous groups, in which the hydraulic conductivity and specific storage are constant. We first apply individual clustering (i.e., 1D clustering) to the diffusivity tomograms of the CO\(_2\)-free aquifer. Spatial heterogeneity is determined by the clusters that are used in the following calibration. Individual and JT (i.e., 2D clustering) are used for the tomograms of the diffusivity difference and velocity difference obtained after CO\(_2\) injection. Compared with individual clustering of PT or ST, we hereafter refer to joint clustering as “JT.”

The clustering approach is a modified \( k \)-means method. Unlike the usual \( k \)-means method (MacQueen, 1967), the centroids of the clusters are determined by fitting the data histogram with the summation of multiple 1D or 2D Gaussian functions. The 1D and 2D clusterings are performed in a time-lapse strategy. The centroids are determined by the data histogram at long injection, and then they are applied at short injection as well. The inverted shapes are compared with the true plumes and their ambient aquifers by analyzing the dissimilarity in their shape. Three metrics can be used for obtaining this goal: over-
estimation rate, underestimation rate, and total pixel misclassification error rate. These metrics can loosely depict the dissimilarity of two binary images, which in our study are the true plume and inverted plume. They are detection performance measures, regardless of the pixel positions and intensity. Definitions of these metrics are given in the following (Baddeley, 1992).

Let \( A, B, \) and \( X \) be the true plume, inverted plume, and the pixel raster (i.e., the entire inversion model), respectively. Pixels that belong to \( B \) but not \( A \) are called type I errors; pixels that belong to \( A \) but not \( B \) are called type II errors.

Type I errors (\( \alpha \)), also called the “overestimation rate,” are calculated by

\[
\alpha(A, B) = \frac{n(B \setminus A)}{n(X \setminus A)}, \tag{11}
\]

Type II errors (\( \beta \)), or the underestimation rate, are calculated by

\[
\beta(A, B) = \frac{n(A \setminus B)}{n(A)}, \tag{12}
\]

where \( n(A) \) and \( n(B) \) are the number of pixels in the true plume \( A \) and inverted plume \( B \). Here, \( n(X) \) is the total number of pixels in the raster. Based on these two error rates, the total pixel misclassification error rate (\( e \)) for binary images is defined as

\[
e(A, B) = \frac{n(A \Delta B)}{n(X)} = \alpha(1 - r) + \beta r. \tag{13}
\]

**Zonal calibration and estimation of saturation**

Calibration of \( \text{CO}_2 \) saturation in a multiple-process-coupled forward simulator usually is computationally demanding. In our study, one run of the 2D two-phase flow model in PFLTRAN costs 1 h to several days depending on the model heterogeneity (data are based on a workstation, 32× Xeon 3.1 GHz, 64 GB RAM). For reducing the model complexity and accelerating calibration, we use MODFLOW (Harbaugh, 2005) to run the forward simulation during the inversion procedure, considering \( \text{CO}_2 \) and brine as a mixed phase. Compared with PFLTRAN, one run of the mixed-phase flow model merely costs 2–4 min (data are based on a PC, Intel i7-3370S 3.1 GHz, 16 GB RAM). This greatly reduces the computing time and thus improves the calibration efficiency. Figure 5 presents a summary of two different zonal calibration steps carried out sequentially.

First, for the preinjection, we merely calibrate the effective hydraulic conductivity and specific storage assuming that the aquifer is homogeneous (Figure 5a). The calibrated values of \( K_w \) and \( S_{sw} \) are assigned to the clustered aquifer zone for the postinjection (Figure 5b). As pointed out in Hu et al. (2015), the mixed-phase conductivity \( K \) of the plume zone shows only minor changes during \( \text{CO}_2 \) injection, and thus this value is kept the same as \( K_w \) for the postinjection. Only the mean mixed-phase specific storage \( S_{n} \) of the plume zone needs to be calibrated (Figure 5b). The mean \( S_{n} \) of the plume is then transferred from \( S_{n} \) (Figure 5c) by the following equation:

\[
S_{n} = \phi g(p_w S_w + \rho_n S_n)(c_w S_w + c_n S_n). \tag{14}
\]

Second, the calibration is conducted considering the heterogeneity of the reservoir. For the two layered scenarios, the aquifer structure is determined by clustering the inverted \( D_{pec} \) and the \( K_w \) and \( S_{sw} \) of each cluster are calibrated primarily for the preinjection (Figure 5d). For the postinjection, the plume zone can be divided into two secondary plumes based on the aquifer structure. The functions \( K_w \) and \( S_{sw} \) of each cluster in the aquifer zone are fixed, as well as \( K \) in the plume zone (\( K = K_w \)) (Figure 5e). The mixed-phase specific storage \( S_{n} \) of the two secondary plumes is calibrated and converted to the \( S_{n} \) of the two plumes in equation 14.

To reduce the potential nonuniqueness of the calibration results in the second step, the acquired values of \( S_{n} \) in the first step are used as prior information for the following calibration. Here, we make an assumption that the \( S_{n} \) in the high-permeability layer is larger than the low-permeability layer. The mean \( S_{n} \) (Figure 5c) is transferred back to the two \( S_{n} \) values for the layered structure, considering the porosities are different for the two layers. These two values are used for constraining the calibration. For instance, presuming \( K_{w,1} \) is larger than \( K_{w,2} \) in Figure 5d, parameters \( S_{n} \) of the two layers transferred by the mean \( S_{n} \) are expressed as \( S_{n,1} \) and \( S_{n,2} \), respectively. Then, the calibrated \( S_{n,1} \) should be larger than \( S_{n,1} \), and \( S_{n,2} \) is smaller than \( S_{n,2} \). The validity of this assumption depends on whether the initial aquifer structure can be identified properly.

Figure 6 shows five different models for converting \( S_{n} \) back to \( S_{n} \). Models 1, 4, and 5 (black solid line and green and pink dashed lines) show the relationship between the effective \( S_{n} \) and \( S_{n} \) for a one-plume structure in three different scenarios. Models 2 and 3 (blue and red solid lines) are used for obtaining the saturation within each secondary plume for the two layered scenarios.

The performance of the calibration is evaluated by calculating the saturation error \( \xi \). It is estimated by the difference between the calculated saturation (\( S_{n}^{cal} \)) and the arithmetic mean of the true saturations within the same inverted plume or secondary plume structure (note, not the true plume extent) normalized by \( S_{n}^{true} \).

\[
\xi(\%) = \frac{S_{n}^{cal} - S_{n}^{true}}{S_{n}^{true}} \times 100. \tag{15}
\]
RESULTS

The procedure described above is tested step by step on the three reservoir scenarios to compare the performance of PT and ST.

PT and ST traveltimes

For eliminating the effect of different source-receiver configurations for PT and ST, we only compare the traveltimes of the tomographic arrays in the horizontal direction. Note that the seismic inversion is based on the full data set of the traveltimes (i.e., the total 5776 traveltimes). Supplemental Table S1 (see the supporting information) lists the statistics of these traveltimes (noise-free and with noise) in the horizontal direction derived for PT and ST. The horizontal hydraulic traveltimes vary from 12.7 (in scenario 2layers_B) to 173.4 s \(0.5\) (in the homogeneous scenario). The hydraulic traveltimes increase by 154%–486% after CO\(_2\) injection. The most affected horizontal seismic traveltimes through the reservoir increase from 11.2 ms before CO\(_2\) injection to 11.76 ms after injection, corresponding to an increase of 5%. Overall, changes of the seismic traveltimes are much smaller than for hydraulic traveltimes, with changes ranging from 0% to 5%.

To compare the results for PT and ST, the relative spread of the horizontal traveltimes (i.e., the standard derivation normalized by the mean) is examined (see Supplemental Table S1). At preinjection, the relative spread of the noise-free traveltimes in the homogeneous scenario is zero. For the traveltimes with noise, the relative spread is consistent with the noise level (1%). In the two layered scenarios, the relative spread at preinjection of the hydraulic traveltimes (0.23 for 2layers_A and 0.1 for 2layers_B) is much greater than the seismic traveltimes (approximately 0.01–0.02). At postinjection, the relative spread for the seismic traveltimes is approximately 0.01–0.02 for all three scenarios, whereas for the hydraulic traveltimes, it reaches from 0.1 to 0.7. In the following, we only present and discuss the results derived from the data with noise, thus considering the more realistic cases.

Diffusivity and velocity tomograms

The results obtained from two-phase flow simulation for the different scenarios are considered as the “truth,” and these serve as a reference for assessing tomographic inversion. The values of true diffusivity, velocity, and their differences for pre-, short, and long injections are calculated based on the simulated true CO\(_2\) saturation, according to equations 3–7 and 10. The true profiles and inverted tomograms are depicted in Figure 7, and a complete list can be found in the supporting information (Supplemental Tables S2 and S3).

In the homogeneous scenario, the true \(D\) and \(V\) at preinjection are 2.7 m\(^2\)/s and 4465 m/s, respectively (Figure 7I-a and 7I-g). The inverted \(D\) varies from 2.8 to 3 m\(^2\)/s (Figure 7I-d), which is slightly different to previous results by Hu et al. (2015) for the same scenario due to the noise for hydraulic traveltimes that is included here. The seismic tomography includes the caprock above and below the reservoir, and the reservoir can be clearly identified in the tomograms (see Figure C-1). Inside the reservoir, the inverted values of \(V\) range from 4064 to 4755 m/s (Figure 7I-j). The relative spread is greater than that observed for the inverted \(D\). The inverted velocity in caprock/bottom seal is much less than the reservoir, which varies from 3413 to 3800 m/s. In the scenarios 2layers_A and 2layers_B, the true \(D\) and \(V\) of the initial CO\(_2\)-free formation are 22.4 m\(^2\)/s and 4579 m/s for the high-permeability layer, and 2.9 m\(^2\)/s and 4454 m/s for the low-permeability layer (Figure 7II-a, 7II-g, 7III-a, and 7III-g). The inverted \(D\) for 2layers_A and 2layers_B is within a range smaller than the true values (3.6–10.2 m\(^2\)/s for 2layers_A and 7.2–12.2 m\(^2\)/s for 2layers_B, Figure 7II-d and 7III-d). The inverted \(V\) shows a similar range for the two scenarios, which is 3998–4930 m/s and 4067–4854 m/s, respectively (Figure 7II-j and 7III-j).

For the two postinjection periods, both of the true \(D\) and \(V\) follow a nonmonotonic change along with \(S_n\). (Figures 3 and 4). The true \(D\) decreases by up to two orders of magnitude, resulting in a minimum \(\Delta D\) (i.e., the difference of logarithm \(D\) at pre- and postinjection) value of approximately −2 for all three scenarios (Figure 7I-b, 7I-c, 7II-b, 7II-c, 7III-b, and 7III-c). The minimum \(\Delta V\) is −340.9 m/s for the homogeneous scenario, and it is −356.4 m/s for 2layers_A and 2layers_B. It is noticeable that for each scenario, the minimum \(\Delta D\) and \(\Delta V\) are the same during the short and long injection because they do not correspond to the maximum \(S_n\) value (Supplemental Figure S1 and Figure 4). Likewise, in the true profiles of the flow and seismic parameters, the smallest values are shown within the plume, where \(S_n\) has a moderate value. The inverted \(\Delta D\) for the homogeneous scenario and 2layers_A span a smaller range compared with the truth (Figure 7I-e, 7I-f, 7II-e, and 7II-f). In contrast, the inverted \(\Delta D\) has a larger range than the truth for 2layers_B (Figure 7III-e and 7III-f). In general, the absolute values of the inverted \(\Delta V\) are smaller than the true values for all the three scenarios.

1D and 2D clustering structure

Prior to the zonal calibration, clustering was implemented based on the inverted results to obtain the plume shape at different times. First, according to the inversion performance of the baseline, the inverted \(D\) derived prior to CO\(_2\) injection was clustered to determine the structure of the reservoir. Subsequently, the inverted \(\Delta D\) and \(\Delta V\) were clustered individually (1D clustering), and then jointly (2D clustering). To judge the different approaches, the plume shapes derived from 1D and 2D clustering processes were compared with the true plumes according to the three aforementioned metrics (overestimation rate, underestimation rate, and total misclassification rate, calculated in equations 11–13). Figure 8 depicts the clustering.
results, with the metrics shown as the numbers in the same figure. In terms of misclassification, ST outperforms the PT and JT (i.e., joint clustering), but the numbers are generally similar.

Zonal calibration and calculated saturations

Based on the identified aquifer and plume zones by 1D and 2D clustering, zonal calibration was conducted. The effective $K_w$ and $S_{sw}$ of the original formation (ignoring its heterogeneity) were calibrated primarily in MODFLOW with PEST (Doherty, 2010), using the pressure observations derived from the full model. Because of the proxy, the calibrated $K_w$ and $S_{sw}$ at preinjection are slightly smaller than the true values (the errors are approximately 10%). The calibrated $K_w$ and $S_{sw}$ were then used as the prior information for the aquifer zone, and the calibrated $K_w$ for the plume zone was also fixed for the postcalibration. The calibrated $S_s$ of the plume zone (see

\[
\begin{align*}
-3.4 & -3.0 & -2.6 & -2.2 & -1.8 & -1.4 & -1.0 & -0.6 & -0.2 & 0 \\
-3.5 & -2.87 & -2.24 & -1.61 & -0.98 & -0.35 & 0.35 & 0.98 & 1.61 & 2.87 & 3.5
\end{align*}
\]

Velocity difference (m/s)

\[
\begin{align*}
2.9 & 6.72 & 10.54 & 14.36 & 18.18 & 22 \\
4000 & 4186 & 4372 & 4558 & 4744 & 4930
\end{align*}
\]

Velocity (m/s)

\[
\begin{align*}
-2.4 & -2.1 & -1.8 & -1.5 & -1.2 & -0.9 & -0.6 & -0.3 & 0 \\
\end{align*}
\]

Diffusivity difference (-)

\[
\begin{align*}
2.1 & 2.2 & 2.3 & 2.4 & 2.5 & 2.6 & 2.7 & 2.8 & 2.9 & 3 \\
4060 & 4198 & 4336 & 4474 & 4612 & 4750
\end{align*}
\]

Diffusivity (m/s)

Figure 7. True profiles versus inverted tomograms in the three scenarios (the true profiles are on the left, and the inverted tomograms are on the right). For ST, the model extends 30 m above and below the reservoir (see Figure C-1) to include the caprock and bottom seal ($V = 3500$ m/s). CO$_2$ is injected at the left of the model, and the migrating plume can be seen in PT and ST by the inverted tomograms. The internal structure of the aquifer can only be resolved using PT because internal variations in seismic velocity are too small compared with the contrast to caprock.
the supporting information, Supplemental Table S5) was then converted to \( S_n \) in the homogeneous scenario, as well as 2layers_A and 2layers_B by models 1–3 (Figure 6), respectively. The calibration results of the 1- and 2-plume structures for the three scenarios are presented in Figure 9 and Supplemental Figure S3 and Supplemental Tables S4 and S6, respectively.

Calibration quality was evaluated by calculating the error of \( S_n (\xi) \) using equation 15 (see Supplemental Table S5). For 1-plume structure, in the homogeneous scenario, \( \xi \) varies from \(-3\%\) to \(11\%\) except for the short injection, case in which the plume structure was derived from ST (\( \xi \) is \(89\%\)). In scenario 2layers_A, \( \xi \) ranges from \(36\%\) to \(51\%\) for short injection, which is generally higher than for long injection (\(23\%–29\%\)). In scenario 2layers_B, \( \xi \) remains low (<10\%) except for the case of PT for the long injection (46%). Overall, the JT results are not always the best, but they are robust. The estimation errors are reduced, maintaining errors at less than 36\% for all the scenarios. For the 2-plume structure, \( \xi \) changes from \(-24\%\) to \(127\%\) in 2layers_A, and which varies from 0\% to 63\% in 2layers_B (see Supplemental Table S6).

**DISCUSSION**

**PT and ST traveltimes**

Changes in PT and ST traveltimes are a direct measure of the sensitivity to relevant changes in the reservoir. Our results indicate that variability of hydraulic traveltimes is generally much larger than seismic traveltimes. A great variability of traveltimes is favorable because this potentially allows for better resolution of the subsurface. Through the comparison of relative spread in three scenarios, it is implied that relative spread correlates with the degree of heterogeneity. No relative spread indicates noise-free homogeneous conditions. Prior

![Figure 8](https://www.example.com/figure8.png)

Figure 8. True clustering structures (subgraphs a-c) versus 1D (subgraphs d-i) and 2D (subgraphs j-l) clustering structures in the three scenarios. Numbers in blue: overestimation rate (\(\alpha\)); numbers in green: underestimation rate (\(\beta\)); and numbers in red: total misclassification rate (\(\varepsilon\)). The performance among PT, ST, and JT is comparable in the homogeneous scenario and 2layers_A, whereas in 2layers_B, JT shows a combination of the 1D clustering results from PT and ST.
to CO$_2$ injection, the highest relative spread is obtained for 2layers_A, in which small-scale contrasts in permeability are simulated by a relatively thin conductive layer. After CO$_2$ injection, the highest relative spread is in 2layers_B, indicating the largest contrasts in permeability between the relatively thick conductive layer and the CO$_2$ plume. In addition, it is remarkable that the imposed noise has a small impact on the relative spread of the hydraulic travel times because the spread of the noise-free data is much larger than the noise level.

**Diffusivity and velocity tomograms**

In Figure 7, two-phase simulations show that the fronts of the plumes in the two heterogeneous scenarios have more complicated geometries (Figure 7II-b, 7II-c, 7III-b, and 7III-c) in comparison with the true plumes in the homogeneous scenario (Figure 7I-b and 7I-c). The high-permeability layer largely controls the plume. The CO$_2$ migrates preferentially within the highly conductive layer, whereas the migration is also controlled by buoyancy, complicating the plume geometry. In 2layers_A, the plume distribution is representative for a multilayer system (i.e., two or more continuous layers) in which the top layer has the highest permeability. Here, the plume ultimately assembles at the top of the reservoir, which is caused by a combined effect of the high permeability of the upper layer and buoyancy (Figure 7II-b and 7II-c). However, the less permeable lower layer hinders expansion of the plume. The scenario 2layers_B exemplifies conditions in which a highly conductive channel exists between the two wells. Here, the plume travels faster at the bottom layer, forming a striking finger-like shape at the boundary of the two layers (Figure 7III-b and 7III-c). The finger is not delimited strictly below the boundary because of buoyancy effects.

At preinjection, the small range of inverted D still nicely reflects the homogeneous properties of the aquifer in homogeneous scenario. The strong velocity variation (3500 versus approximately 4500 m/s) between caprock/bottom seal and reservoir enables identification of the reservoir, but makes it difficult to judge if the reservoir itself is homogeneous or heterogeneous. In the two layered scenarios, the inverted D generally has a higher value and less data spread in 2layers_B, which, in this scenario, is due to the larger high-permeability area and also to shorter travel times. In addition, the inverted D tomograms in the two scenarios display a layered distribution, which is consistent with the true aquifer structure to some extent. However, the “perfectly” horizontal boundary between the two layers was not accurately reconstructed due to nonhorizontal tomographic rays as well as regularization, which both cause smearing between the inversion cells. Comparison of the three scenarios indicates that PT resolves the internal structure and especially the hydraulic properties of the aquifer better because it is related to permeability and porosity (equations 3–5). Usually, permeability shows much larger spatial variability than porosity. In contrast, ST is able to delineate the structure of the reservoir, but it fails to identify additional variations within the reservoir. Seismic velocity mainly depends on the
porosity and rock density, which might only have slight variations within an aquifer. Velocity variations within the reservoir are only approximately 120 m/s prior to CO₂ injection, which is difficult to recover simultaneously with the 1000 m/s variation between reservoir and caprock/bottom seal.

At postinjection, the true profiles show that the contrast of ΔV caused by CO₂ is much smaller compared with ΔD. Inversion results of ΔD and ΔV are not consistent with the true values. Nevertheless, the information about the plume can still be inferred from the small values in the ΔD and ΔV tomograms. The ΔD tomograms of the two layered scenarios indicate that the variability of permeability has an adverse impact because PT is applied to the postinjection. The PT resolves the secondary plume in the lower permeability layer better. In the case that the contrast of diffusivity is sufficiently large at preinjection, the secondary plume in a higher permeability layer can still be identified (e.g., in 2layers_A, Figure 7II-e and 7II-f). Conversely, in 2layers_B, the finger at the layer boundary is masked in the ΔD tomograms (Figure 7II-e and 7II-f). This is mainly because the variations of the inverted D at preinjection are comparably small. The ST can capture the main front of the plume from the ΔD tomograms of all the scenarios because it is not influenced by the permeability. However, the lower relative spread of the seismic travel-times limits the capability of ST to identify the small-size plumes in the low-permeability layer (Figure 7III-k and 7III-I).

Overall, the inverted values from the three scenarios at pre- and postinjection indicate that neither diffusivity nor velocity values can be precisely reproduced by the inversions. Direct transformation of inverted values to CO₂ saturation leads to an incorrect estimation. There are several reasons that can explain this. First, for PT and ST, the loose density of the trajectories or rays in the low-diffusivity or -velocity parts can cause a nonuniqueness of the inversion solution in the tomograms. Second, these errors can also be attributed to inaccuracies introduced by using the single-phase proxy. Besides, the nonmonotonic relationship between the diffusivity or velocity and CO₂ saturation hampers deriving an exact CO₂ saturation value directly from the inverted diffusivity or velocity.

1D and 2D clustering structure

In the homogeneous and 2layers_A scenarios, clustering of D before CO₂ injection shows a homogeneous and two-layer aquifer structure (Figure 8I-d and 8II-d). The clustering results after CO₂ injection from 1D and 2D clustering are of comparable quality. Because of the similar distribution of the inverted ΔD and ΔV, they show a strong correlation (see the supporting information, Supplemental Figure S2a). Even 2D clustering does not significantly improve the results. In some cases, the 1D clustering results based only on PT or ST are better than the 2D clustering results. For instance, if we compare the total misclassification rate (Figure 8, red numbers) to assess the quality of the results, in the homogeneous scenario, the clustering result based on PT at short injection provides the best agreement with the true plume (Figure 8I-e). On the contrary, the clustering result from ST (Figure 8I-h) is the worst; thus, it has a negative impact on the final JT result (Figure 8I-k). In addition, in 2layers_A, during the short injection, the clustered ΔD and ΔV display a discontinuous distribution near the injection location (Figure 8II-e and 8II-h), yet this continuity vanishes in the jointly clustered plume (Figure 8II-k).

In 2layers_B, the two-layer structure also can be indicated by the 1D clustering of D (Figure 8III-d). However, for the two postinjection periods, the plumes show a significant difference from the 1D clustering results. The geometries of the plumes delineated by PT are more vertical (Figure 8III-e and 8III-f), whereas those derived from ST show a more lateral distribution (Figure 8III-h and 8III-I). The ΔD and ΔV show less correlation during the 2D clustering process (see the supporting information, Supplemental Figure S2b). The dissimilarities between ΔD and ΔV hamper the acquisition of the 2D centroids for clustering the time-lapse data sets. Therefore, we cluster ΔD and ΔV in another way. The 2D histogram was fitted by a model composed of multiple Gaussian functions (the aquifer zone) and a uniform distribution (the plume zone). The cutoff of the aquifer and plume zones was at the edge of the Gaussian functions, in which the values are equal to the mean value of the uniform distribution. This was applied for the short- and long-injection runs. Consequently, the 2D clustered structures are deemed to be a superposition of the plumes from PT and ST (Figure 8III-k and 8III-I), but in a more systematic way.

Clustering works best for the homogeneous scenario according to the total misclassification rate ϵ (smaller than 0.1 for all the three scenarios). The heterogeneous scenarios show a higher misclassification due to the additional complexity. Values of ϵ indicate that the clustering performance in 2layers_B (ϵ: 0.07–0.15) is better than 2layers_A (ϵ: 0.13–0.18). The overestimation rate α shows a similar trend as ϵ. The plume extents are most overestimated in 2layers_A compared with the other scenarios. The underestimation rate β of the plumes is relatively high as the true plumes near the injection well or if the plume fronts are not accurately characterized (e.g., Figure 8I-h and 8III-I). In general, JT reduces the underestimation of the plume extent (e.g., Figure 8I-k, 8II-k, 8III-k, and 8III-I).

Saturation errors

The underestimation rate β is considered the most crucial criterion to assess proper spatial classification. In Figure 10, the saturation error ξ is plotted with β. It is clearly shown that, for each scenario, the increased β generally provokes a higher saturation error. This is be-

![Figure 10. Underestimation rate β versus saturation errors ξ. The red and blue symbols are the calibrated results based on individual clustering structures derived from PT and ST, respectively. The green symbols represent the results derived by the JT. Each symbol exists twice, representing the short and long injection, respectively.](http://library.seg.org/)
cause the hydraulic conductivity and specific storage in the underestimated part of the plume are assigned the same values as the aquifer. As discussed in the section “Model parameters,” the specific storage of a plume can be approximately 1–2 orders of magnitude larger than the ambient aquifer. Therefore, the underestimation of the plume size leads to a higher value of specific storage within the inverted plume.

In comparison with homogeneous and 2layers_B, 2layers_A shows higher estimation errors. This might be due to the overestimation of the plume extent. The true saturation \( S_t \) is derived by averaging the saturations within an inverted plume, that is, it is the arithmetic mean of these saturations. As \( S_t \) increases nonlinearly with \( S_n \) (Figure 6), the transferred \( S_t \) might be different from the averaged \( S_{t,INV} \), even though they correspond to the same calibrated \( S_n \).

For 2-plume structures in 2layers_A and 2layers_B, in general, the estimated \( S_n \) values are consistent with the fact that \( S_n \) is larger in the high-permeability layer. However, the estimation errors span a broader range in comparison with the previous results from the 1-plume structure. As discussed above, these errors can be due to the misclassification of the secondary plume in each layer. Moreover, they can also be attributed to the pressure discrepancy between the full model and the proxy. To obtain the specific storage of each secondary plume, more pressure measurements were used for the 2-plume structure than the 1-plume structure, and thus the calibration involved more pressure errors. This can be improved in future work by quantifying the errors between the two-phase forward simulation and the single-phase proxy under different conditions.

CONCLUSION

We investigate the feasibility of PT and compare the inversion performance with crosshole seismic tomography (ST) for homogeneous and heterogeneous reservoirs. Relations between the inverted parameters (the mixed diffusivity and P-wave velocity) and the CO₂ saturation as used by these two methods are comparable. Both are fast and computationally efficient because they are eikonal-based. However, because different signals are processed, these two approaches can be complementary to each other for characterizing an evolving CO₂ plume shape and for evaluating the CO₂ saturation.

In our scenarios, the upper and lower boundary of the reservoir can only be detected using ST. The PT cannot be used in the impermeable caprock. The ST is less suitable to resolve the smaller internal contrasts in the layered reservoir. Better results when reconstructing the heterogeneity of the reservoir can be obtained by PT because it directly links to aquifer permeability. The capability for resolving the plume shape is distinct for PT and ST due to the different features of the traveltimes. First, the contrast of diffusivity is much larger than seismic velocity. Therefore, the hydraulic traveltime used for the inversion change by orders of magnitude during CO₂ injection, rather than a few percent as for seismic traveltimes. This gives PT better sensitivity to the CO₂ plume. Moreover, due to the much larger traveltimes, PT requires less strict repeatability than does ST. In other words, the theoretical tolerance of the noise of PT is better. However, for PT, reservoir heterogeneity can alleviate the diffusivity contrast caused by CO₂ injection, and thus the front of the plume is hard to delineate by the diffusivity difference to-mogram. ST can better resolve the lateral spreading part of the plume. Consequently, the best results are generally derived from the presented joint inversion.

By clustering and subsequent zonal calibration, the mean saturation of the plume can be determined. We demonstrate by sequential calibration strategies how the saturation of different layers can be distinguished. Again, jointly clustered structures provide best results for the various scenarios and conditions examined. However, it is not surprising that improper spatial delineation of the plume makes it difficult to properly estimate the saturation.

This study provides an insight into the capability of PT for application in heterogeneous formations and its potential for complementing the geophysical approaches. One crucial point remains: the transfer of this approach to the field. The main challenges of a field application include the technical implementation of sources and receivers in deep reservoirs, conducting interference injection tests during the course of CO₂ injection, and interpreting PT or joint PT-ST results given nonideal conditions in the field. From the field-injection tests in several CO₂ storage sites (e.g., Ketzin and Cranfield), it is clear that it is possible to conduct multilevel CO₂ injection tests and to obtain useful pressure signals. Application of such tests in tomographic arrangements to complement seismic measurements is thus a promising area for future study. Also, more geophysical approaches used for CO₂ sequestration will be considered for comparing and being combined with PT, such as seismic full-waveform inversion and electrical resistance tomography.

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APPENDIX A

DISCRETIZATION OF TWO-PHASE FLOW-SIMULATION MODEL

Figure A-1 displays the 2D model used for forward simulations and pressure calibration. An extension of the model in horizontal...
direction (x-direction) is 580 m, and in the vertical direction (z-direction), it is 15 m. The injector for CO₂ and brine is placed at \( x = 290 \) m, and a pressure monitoring well is 50 m away from the injector. In the x-direction, the model is discretized into 287 grid cells. Between \( x = 240 \) and 340 m, the grid size is 0.5 m, with a progressive refinement to 0.09 m toward the injector. Outside of this area, the grid size increases exponentially. The largest grid size is 40 m at two lateral boundaries. In the vertical direction, the model is discretized into 25 layers, and for each, the thickness is 0.6 m.

**APPENDIX B**

**GASSMANN-WOOD ROCK-PHYSICS MODEL**

The low-frequency Gassmann equations (Gassmann, 1951) are widely used for calculating rock and fluid elastic properties in fully saturated media. The saturated bulk modulus \( G_{\text{sat}} \) is given by

\[
G_{\text{sat}} = G_{\text{dry}} + \left( \frac{1}{G_{\text{dry}}} - \frac{1}{G_{\text{m}}} \right) \frac{G_{\text{f}}}{\phi} + \frac{1 - \phi}{\rho_{\text{r}} G_{\text{m}}} - \frac{G_{\text{dry}}}{G_{\text{m}}},
\]

where \( G_{\text{dry}} \) and \( G_{\text{m}} \) are the bulk modulus of dry frame rock (drained of pore fluid) and rock matrix, respectively. Here, \( G_{\text{f}} \) is the bulk modulus of the pore fluid, which can be single phase or multiphase.

Bulk modulus of mixing pore fluid can be calculated by Wood’s (1941) equation

\[
G_{\text{f}} = \left( \frac{S_{\text{w}}}{G_{\text{w}}} + \frac{S_{\text{n}}}{G_{\text{n}}} \right)^{-1},
\]

where \( G_{\text{w}} \) and \( G_{\text{n}} \) are the bulk modulus of brine and CO₂, respectively. Here, \( G_{\text{m}} \) equals the product of the density and P-wave velocity of CO₂. The P-wave velocity in saturated rock is then estimated by the following equations:

\[
V = \sqrt{\frac{G_{\text{sat}} + \frac{4}{3} N_{\text{sat}}}{\rho_{\text{r}}}},
\]

where \( \rho_{r} \) is the rock density. For a CO₂-brine system, it is calculated through the linear relationship

\[
\rho_{r} = \phi (S_{\text{w}} \rho_{\text{w}} + S_{\text{n}} \rho_{\text{n}}) + (1 - \phi) \rho_{\text{m}},
\]

where \( \rho_{w} \) and \( \rho_{n} \) are the brine and CO₂ densities, and \( \rho_{m} \) is the rock-matrix density.

**APPENDIX C**

**FULL-VELOCITY (DIFFERENCE) TOMOGRAMS**

Figure C-1 shows the full velocity or velocity difference tomosgrams derived from ST inversion (an example of homogeneous scenario, corresponding to Figure 7I-j–7I-l). The grid size of the inversion model is 1 × 1 m. The dashed line delineates the reconstructed reservoir. Note that the results depicted here are slightly different in comparison with Figure 7. This is because the model grid shown in Figure 7 (ST results) is interpolated to 1 × 0.9375 m, to be consistent with the PT results and to implement the JT.

*Source (ST)  × Receiver (ST)*

Figure C-1. Full velocity (preinjection) and velocity difference (postinjection) tomosgrams in homogeneous scenario. The area outlined by the dashed line indicates the reconstructed storage reservoir. Note that the sources and receivers are illustrated schematically, and their true numbers are much higher (76).
**NOMENCLATURE**

- $A$ = “true” plume
- $B$ = inverted plume
- $c_n$ = CO$_2$ compressibility ($1/$Pa)
- $c_w$ = brine compressibility ($1/$Pa)
- $c_p$ = heat capacity ($J/$Kg)
- $D$ = mixed-phase diffusivity ($m^2$/s)
- $D_{pre}$ = diffusivity at preinjection ($m^2$/s)
- $D_{post}$ = diffusivity at postinjection ($m^2$/s)
- $\Delta D$ = diffusivity difference
- $G_{dry}$ = bulk modulus of dry frame rock (Pa)
- $G_f$ = bulk modulus of mixing pore fluid (Pa)
- $G_{m}$ = bulk modulus of rock matrix (Pa)
- $G_{sat}$ = saturated bulk modulus (Pa)
- $I_x$ = integrated length in the $x$-direction
- $I_z$ = integrated length in the $z$-direction
- $k$ = intrinsic permeability ($m^2$)
- $k_{cap}$ = permeability of the caprock ($m^2$)
- $k_{ref}$ = intrinsic permeability of reference media ($m^2$)
- $k_{perm}$ = relative permeability of brine ($m^2$)
- $k_{red}$ = relative permeability of CO$_2$ ($m^2$)
- $k_{post}$ = permeability of the bottom seal ($m^2$)
- $K_{w}$ = single-phase hydraulic conductivity ($m/s$)
- $N_{dry}$ = shear modulus of dry frame rock (Pa)
- $N_{sat}$ = saturated shear modulus (Pa)
- $P_0$ = pressure datum (MPa)
- $P_d$ = entry pressure (Pa)
- $P_{d,ref}$ = entry pressure of reference media (Pa)
- $Q_{inj}$ = CO$_2$ injection rate (kg/s)
- $S_n$ = CO$_2$ saturation
- $S_{true}$ = calculated CO$_2$ saturation
- $S_{true}$ = “true” CO$_2$ saturation
- $S_{mix}$ = mixed-phase specific storage (1/m)
- $S_{nw}$ = single-phase specific storage (1/m)
- $S_{w}$ = brine saturation
- $T_{inj}$ = duration of injection at CO$_2$ sequestration stage (h)
- $T_{inj}$ = duration of injection for multilevel CO$_2$ injection (h)
- $T_{rec}$ = duration of recovery at CO$_2$ sequestration stage (h)
- $V$ = P-wave velocity (m/s)
- $V_{post}$ = P-wave velocity at postinjection (m/s)
- $V_{pre}$ = P-wave velocity at preinjection (m/s)
- $\Delta V$ = velocity difference (m/s)
- $W$ = Lambert’s function

**GREEK SYMBOLS**

- $\alpha$ = overestimation rate
- $\beta$ = underestimation rate
- $\epsilon$ = total misclassification rate
- $\kappa_{dry}$ = dry thermal conductivity ($W$/mK)
- $\kappa_{wet}$ = wet thermal conductivity ($W$/mK)
- $\lambda$ = pore-size distribution
- $\mu_n$ = CO$_2$ viscosity (Pa s)
- $\mu_w$ = brine viscosity (Pa s)
- $\xi$ = saturation error
- $\rho_m$ = rock-matrix density ($kg$/m$^3$)
- $\rho_n$ = CO$_2$ density ($kg$/m$^3$)

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