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EXPANSION AND MIGRATION OF GASEOUS AND DISSOLVED CO_2 IN A SITE SPECIFIC SHALLOW AQUIFER

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ABSTRACT

TOUGHREACT EOS2 was used to quantify the influence of aquifer parameters (e.g., formation pressure, permeability, and effective porosity), the CO_2 leakage rate, the groundwater flow velocity, and the aquifer topography on the extent and migration of CO_2 gas-phase plumes in shallow aquifers. Aquifer pressure and leakage rate are the parameters with greatest impact on the extent of the gas-phase plume. Groundwater flow stretches the gas phase plume in groundwater flow direction and influences the dissolution of CO_2 in the water. The dip of the aquifer controls the migration direction of the gaseous CO_2 along the steepest gradient at the top of the aquifer.

Site-specific impact scenarios show that the gaseous CO_2 migrates preferentially into the highest anticlinal structure at the site. The total impact area due to CO_2 leakage is the combination of the width of the gas-phase body perpendicular to the groundwater flow direction and the transport length of the dissolved CO_2.

Using field site data, we have developed a site-specific scenario simulation to simulate CO_2 leakage impact scenarios and to develop a database for site-specific risk-assessment methods using numerical simulations.

INTRODUCTION

One possibility for reducing the anthropogenic emissions of CO_2 into the atmosphere is carbon capture and storage (CCS). Using this method, however, there is the risk that CO_2 could leak into the subsurface and intrude into freshwater aquifers (Celina et al., 2009). If the CO_2 enters freshwater aquifers, it can reduce groundwater quality, due to the dissolution of minerals and the release of inorganic components like lead or arsenic (Wang and Jaffe, 2004; Zheng et al., 2009; Fahrner et al., 2012). Therefore governmental organizations like the European Union (EU) and the U.S. Environmental Protection Agency (EPA) have proposed regulations for the geological storage of CO_2 (EU Directive, 2009, EPA, 2010), including impact assessment and monitoring for shallow freshwater aquifers. The processes taking place during CO_2 leakage into a shallow aquifer can be identified in laboratory or pilot experiments. Such experiments also demonstrate which geochemical parameters can be used to detect CO_2 leakage and which detection methods are effective (Peter et al., 2012; Oldenburg et al., 2011; Fahrner et al., 2012). These experiments should not be performed at a CCS site, in order to prevent changes to the natural baseline. Onsite scaling additionally would require a site-specific monitoring strategy and risk assessment, but experiments on this scale are extensive and not feasible.

Given these site or field restrictions, numerical scenario simulations are particularly useful in developing suitable and cost-efficient risk assessments or monitoring strategies. They can be used to calculate many impact scenarios and prognosticate the development of CO_2 migration at potential leakage scenarios at large scales and over long time scales.

We expect that the migration of leaking CO_2 is influenced by the leakage geometry (point, line, or diffuse leakage), the leakage rate, hydrogeological parameters, groundwater flow, and geological structures such as aquifer topography. Large-scale numerical simulations are required to investigate the potential spreading of CO_2 with unknown leakage positions at the site-scale.

The migration of gaseous CO_2 has already been part of quantitative scenario simulations of deep reservoir conditions (Bauer et al., 2012; Carroll et al., 2009; Celina and Nordbotten, 2009; Kühn et al., 2012; Oldenburg and Lewicki, 2006; Oldenburg et al., 2010). For protected, shallow aquifers, only a few scenarios have been simulated (e.g., Esposito and Benson, 2012; Carroll et al., 2009; Celina and Nordbotten, 2009), and most of them have not been performed for specific site conditions.

The first aim of this work is to quantify the impact of various potentially influential factors (leakage rate, initial aquifer pressure, water temperature, permeability, effective porosity, and the van Genuchten parameters *m* and alpha) on the spreading of a CO_2 gas-phase plume. The second aim is to investigate the spreading of a gas phase below the top of a confined aquifer and the evolution of a dissolved CO_2 plume at site scale. These site-specific simulations of worst-case leakage are steps toward a database for developing a suitable and convenient CO_2-sequestration monitoring concept, as well as a basis for risk assessment.

METHODS

The modeling code TOUGHREACT EOS 2 (Xu et al., 2004) was used to evaluate the migration of gaseous and dissolved CO_2 after a possible CO_2 leakage. A radially symmetric sensitivity analysis was conducted to quantify the influence of aquifer and hydraulic parameters on the extent of a CO_2 gas-phase body after CO_2 leakage within a shallow groundwater aquifer. To quantify the impact of groundwater flow velocity and the dip of the top of the aquifer, we ran different scenarios in a simplified 3D simulation, using data from a real field site with real topography, Groundwater flow conditions were also developed to simulate impact scenarios for this field site.

Geological Settings

3D virtual shallow aquifer scenarios with data from the joint research project CLEAN were used to perform site-specific numerical simulations, employing realistic geological data on groundwater flow velocity and aquifer topography. The extent of the chosen area is nearly 14 km^2, with thickness varying between 30 m and 50 m. The geological structural model has four geological layers of quaternary sediments (Fig.1).

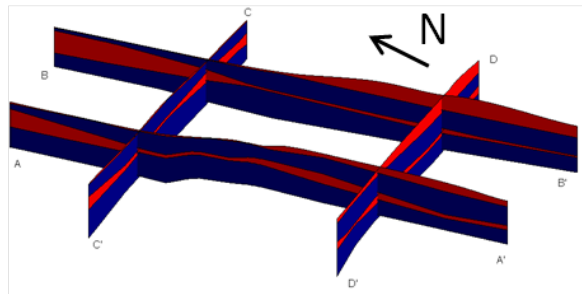

Figure 1. Geological structure model of the CLEAN site (Schäfer et al. in press). Blue: sandy aquifers, red: impermeable till layer

The base of this structural model is a tertiary impermeable clay layer. The lowest layer is a sandy aquifer from the Elster and Saale glacier (L4). The overlaying aquitard is a base moraine from the Saale 1 glacier and defined as a low-permeable till (L3). The till layer is overlain by another aquifer with sandy material. These deposits are from the Saale and Weichsel glaciers (L2). At the top of this aquifer system is yet another till layer, the base moraine of which comes from the Saale glacial and limnic Holocene deposits. This layer is defined as an aquitard (L1)—a detailed description can be found in Kühn (2012).

Conceptual Model

For the 3D multiphase flow scenario simulation, only layer L4 was considered. The interpolated topography for the top and bottom of this aquifer are implemented as input data from the site sampling. Because of the till sediments in layer L3 and the clay deposits at the model base, the top and the bottom of layer L4 are defined as impermeable, with the thickness of layer L4 varying between 5 m and 20 m. The sandy aquifer in the multiphase flow model is simplified as a homogeneous aquifer with a horizontal permeability of $4.2\times10^{-11}m^2$; the permeability in the z-direction is $4.2\times10^{-12}m^2$. The effective porosity is set to n_e= 20% approximated from literature data for the sampled sediment. Relative permeability and capillary pressure are described as a function of saturation using the van Genuchten - Mualem model (van Genuchten, 1980, Mualem, 1976). The parameters for these functions are

approximated from literature from similar sandy sediments (Esposito and Benson, 2011).

Table 1. Assumed parameter for capillary pressure and relative permeability

Capillary Pressure (P_c)	van Genuchten
Van Genuchten parameter	$\lambda = 0.457$
Residual liquid saturation	$S_{lr} = 0.10$
Van Genuchten parameter	$\alpha = 1.48E\text{-}04\ Pa^{-1}$
Max. Pressure	$P_{max} = 5E5$ Pa
Max. liquid saturation	$S_{ls} = 1$
Relative Permeability (k_r)	van Genuchten
Residual liquid saturation	$S_{lr} = 0.15$
Residual gas saturation	$S_{gr} = 0.05$

We implemented a point source as a CO_2 source, which simulates a gas-migration pathway along a borehole into the shallow groundwater (Nordbotton, 2009). Because there has been no observed CO_2 leakage from geological storage sites, assessing a possible leakage rate is not possible. Therefore, we used a range of leakage rates from natural analogues to define leakage rates for our scenarios. Leakage rates at natural analogues have a wide range, 1×10^{-8}kg/s up to 1×10^{-1}kg/s.

Numerical Model Setup

Three different types of 3D multiphase flow simulations have been performed with TOUGH 2 ECO2 to evaluate the expansion of CO_2 gas-phase bodies below the impermeable aquifer top. To evaluate the influence of various geological parameters—permeability (K), effective porosity (n_e), initial aquifer pressure (P), aquifer temperature (T) (aquifer pressure and temperature are defined by the depth of the geological formation), the leakage rate (L), and the van Genuchten parameters alpha and P_{max}—we used a radial symmetrical model setup. Constant CO_2 leakage is established in the center of the cylindrical model domain at the bottom of the aquifer. The thickness of the aquifer is 10 m and the radius is 500 m. For discretization, 1 m×1 m cells are chosen. A finer discretization of 0.5 m×0.5 m is chosen over a width of 2 m within the model domain, where the CO_2 migrates upwards from the leakage to the top of the aquifer, and 2 m below the top of the aquifer, where the CO_2 spreads radially. The parameters for the sensitivity analysis are approximate, due to the uncertainty of site parameters. The applied parameters can be found in Table 2. To quantify the results and to choose parameters for the 3D models, we define a base case (BC), as described in Table 2.

Within a simplified 3D simulation with a horizontal planar aquifer geometry, we varied leakage rates and groundwater flow velocity to evaluate their influence on the migration and extent of the CO_2 gas-phase body and the dissolved CO_2. For the model domain, we defined an area 100 m × 100 m. The thickness of the aquifer is only 2 m, because the area of interest for the migration and extension of CO_2 gas phase bodies is that region directly below the top of the aquifer. From some pretests, we learned that the thickness of the gaseous CO_2 body would not be more than 0.5 m. The horizontal discretization is 1 m × 1 m in the inner zone, 15 m around the leakage location, and 1.5 m × 1.5 m outside that zone. Three grid layers are used for the aquifer depth. At the bottom of the aquifer model domain, we set a layer of 1 m thickness; at the top of the model domain, we set two layers with 0.5 m thickness. The applied input parameters are the same as used in the Base Case scenario from the cylindrical simulations (Table 1 and Table 2).

Table 2. Parameters used for the scenario simulations, m and alpha are van Genuchten parameters. The base case is used to compare the effects of the parameters and for the site scenarios (except for the leakage rate)

Variables	Base Case	Sensitivity analysis
Leakage rate [kg/s]	$1*10^{-4}$	$1*10^{-2}$-$1*10^{-9}$
Initial aquifer pressure [m]	10	0 – 500
Temperature [C°]	11	11 -30
Permeability [m^2]	$4.2*10^{-11}$	$4.2*10^{-9}$ $4.2*10^{-13}$
Eff. Porosity [%]	20	10 -40
m[-]	0.457	0.2 – 0.8
Alpha [m^{-1}]	$5.1*10^{-4}$	$2*10^{-4}$-$8*10^{-4}$

Scenario leakage rates are 1.16×10^{-5}kg/s; groundwater pore velocities are 0.01 m/d, 0.05 m/d, and 0.07 m/d, respectively. In a second simulation series with a pore velocity of 0 m/d, we varied the dip of the aquifer, using dip angles of 1°, 2°, and 3°. Water can leave the model domain to all sides of the model using a constant pressure boundary condition.

For the 3D field-site-scenario simulation, we used the relief data from the CLEAN site structural model for aquifer top and the bottom (Fig. 2). For the aquifer thickness, we used a discretization of five layers. The thickness of these layers varied due to the variation in the aquifer thickness. In the X- and Y- directions, 100 and 50 cells are used, respectively. To evaluate the influence of the groundwater flow velocity, we set pore velocities to 0 m/d, 0.1 m/d, and 0.6 m, respectively, which are approximated to the measured groundwater flow velocities at the CLEAN site.

The hydraulic parameters are the same as in the scenarios described before (Tables 1 and 2). A leakage rate of 1.16×10^{-2}kg/s is chosen. The assumed positions of the leakages are shown in Figure 7.

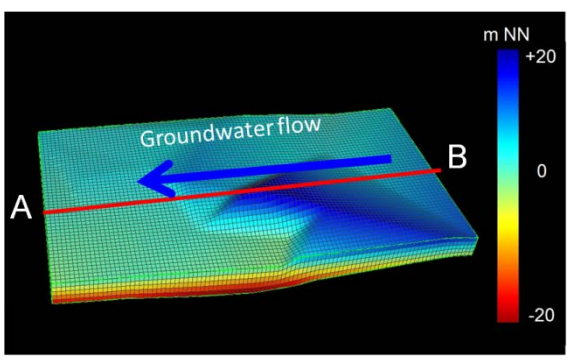

Figure 2. Topography of the layer 4 of the CLEAN site model domain (cf. fig. 1).

RESULTS

Radial symmetric simulations

If CO_2 leaks into the aquifer, it migrates upwards, buoyancy driven, and spreads radially below the impermeable top of the aquifer. The gas phase created is only a thin layer. The radius of the gas-phase body ranges from 10 m to 450 m after a one-year simulation time, depending mainly on the leakage rate and therefore on the volume of inflowing CO_2. The variation in the initial aquifer pressure causes a radius ranging from 8 to 51 m. This is an effect of the gaseous CO_2 solution in the groundwater. Increasing the pressure increases the solubility as well (Duan, 2006). The second reason is the compression of the gas volume, which can be described by the ideal gas law. The pressure contributes only 3% to the decrease in the gas-phase body.

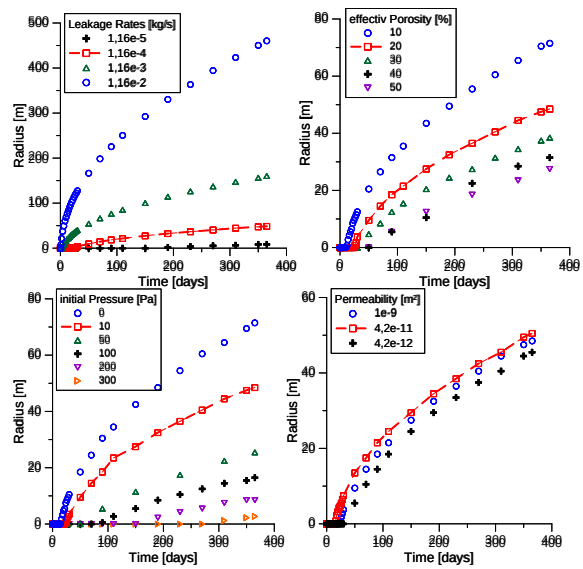

Figure 3. Development of the radius of the gas-phase body under the top of the aquifer over time, due to variation in aquifer parameters.

The variation in porosity results in the radius ranging from 31 m to 48 m. If the pore volume increases, more gas can be stored per volume unit in the aquifer and therefore the extension of the gaseous CO_2 under the impermeable layer becomes smaller. Note that in our simulations, the changes in permeability have no significant effect on the extension of the gas-phase body (Fig.3).

Influence of groundwater flow velocity and aquifer dip

Simplified 3D scenarios considering groundwater flow showed the distinct influence of groundwater flow on the shape of the gas-phase body. Whereas the CO_2 gas-phase body is nearly circular at low groundwater flow velocities, the CO_2 gas-phase body stretches in the flow direction under heavy groundwater flow velocities, and consequently the area of the

gas-phase body decreases (fig 4). In our scenarios, the area of the gas-phase body, with a groundwater flow velocity of 0.01 m/d, after a one-year simulation time, is 772 m², with a flow velocity of 0.07 m/d, it is 15m^2 (Fig. 4).

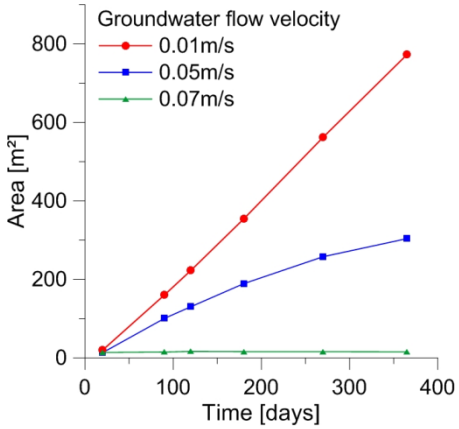

Figure 4. Development of the area of the CO_2 gas-phase body below the impermeable layer of the aquifer top.

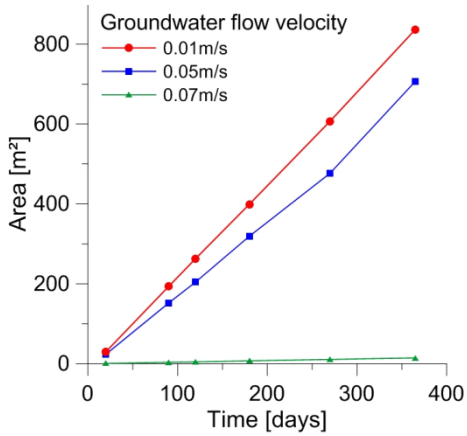

Figure 5. Development of the area of the dissolved CO_2 body below the impermeable layer of the aquifer top.

The gas-phase body gets smaller, because more groundwater saturated with CO_2 is transported downstream with increasing groundwater flow velocity, uncharged water flows from upstream to the phase body, and more gaseous CO_2 dissolves (fig. 5).

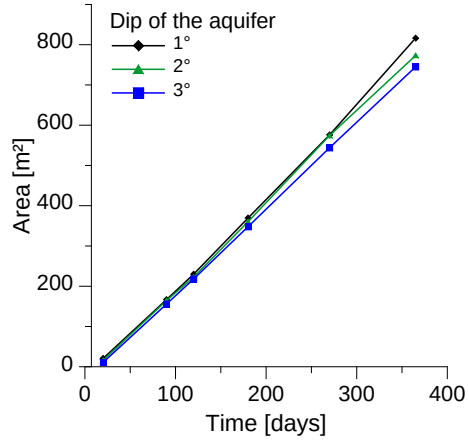

Figure 6. Development of the area of the CO_2 gas-phase body due to the angle of the aquifer dip.

In an aquifer with a dip, the gaseous CO_2 migrates upwards under the top of the aquifer along the steepest gradient. With increasing dip, the CO_2 migrates faster, and the gas-phase body is stretched in the dip direction. With increasing dip, the stretching effect increases. Due to this stretching, the transverse width of the gas-phase body decreases. The area of the gas-phase body does not change significantly (Fig. 6); the difference in area is only 40 m².

3D site scale simulations with a natural relief of the upper aquifer boundary

When the CO_2 enters the model aquifer, it migrates upwards (Fig. 7), and the gas-phase body in the water column is tilted in the flow direction (due to groundwater flow) before it reaches the top of the aquifer. After reaching the impermeable layer, the CO_2 migrates along the steepest gradient of the aquifer topography accumulates in the anticlinal structures (Fig.7).

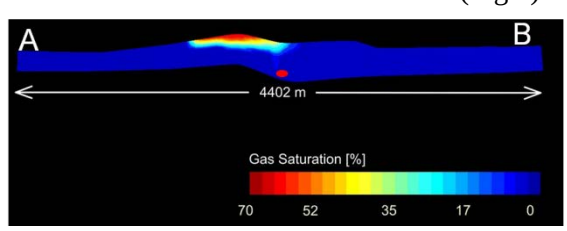

Figure 7. Profiles through the model domain of the site scenario (see Fig. 2). At the bottom of the aquifer is a CO_2 leakage (red point).

After a one-year simulation time, a groundwater flow velocity of 0.1 m/d, and a leakage rate of $1{,}16\times10^{-2}$kg/s below the impermeable top of the

aquifer, the gas-phase body has an area of 3000 m² (Fig. 8). The extent of the gas-phase body is controlled largely by the topography of the aquifer top, and to a lesser extent by the groundwater flow velocity. The groundwater flow influences the dissolution of the CO_2 into the groundwater (see 3D simplified simulations) and the transport of the dissolved CO_2 (Fig. 9).

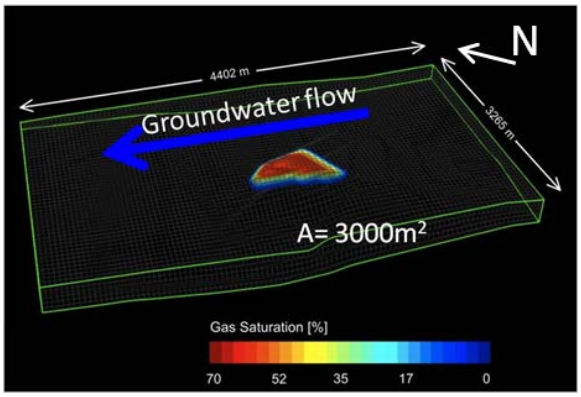

Figure 8. Extent of gaseous CO_2 at the site specific scenario.

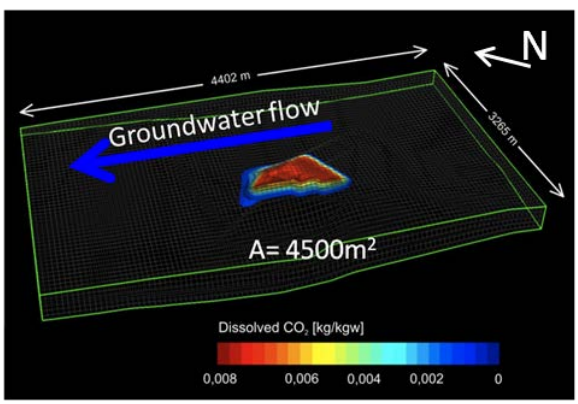

Figure 9. Extent of dissolved CO_2 at the site specific scenario.

The area influenced by dissolved CO_2 is 4500 m². Therefore, the total affected area as a result of CO_2 leakage in an aquifer is controlled by the width of the gas-phase body diagonally to the groundwater flow direction, and the length of the transport of the dissolved CO_2 along the groundwater flow direction.

Figure 10 shows the position of four arbitrarily chosen leakage positions (left) and the results after 10 years simulation time for the saturation of gaseous CO_2 (right). We can see that the gaseous CO_2 migrates from the different leakage positions by following the relief of the aquifer. A comparison between the pathway of the CO_2 migration and the topography of the aquifer top (Fig. 2 and Fig. 6) shows that the gaseous CO_2 migrates along the steepest gradient in the aquifer topography. The main migration direction for leakage A is to the northwest, for leakage B to the north, for leakage C to the northwest, and for leakage D first to the west, into the anticlinal structure and in the anticlinal structure.

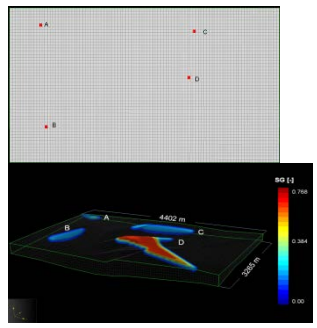

Figure 10. Position of four arbitrarily chosen leakage position (left). Extent of the CO_2 gas phase bodies after 10 years leakage time.

The differences in the extent of gas-phase body leakage depend on the different depths of the aquifer locations and (therefore) on the solubility of the CO_2. This effect can also be seen in the maximum gas saturations. At leakage scenarios A, B, and C, we see the maximum gas saturation of 30%, and in leakage scenario D, we see the maximum gas saturation of 80%. These values result from the accumulation in the fold structure. We showed, in the radial simulations, that the extent of the gas-phase body decreases due to increasing aquifer pressure. In this simulation, the CO_2 first fills the anticlinal structure in the relief, before it migrates in a horizontal direction. The second reason for the higher CO_2 gas saturation of leakage D is the minor dissolution of the gas phase into the aquifer.

CONCLUSION

The simulation scenarios demonstrate that the most important parameters for the extent of the gas-phase body are the leakage rate, the effective porosity, and the hydrostatic pressure of the target aquifer. The volume of the gas-phase body and the gas saturation depend on the pressure and therefore on the aquifer depth, and on

the groundwater velocity, which influences the dissolution of gaseous CO_2 in water as well.
The topography of the aquifer top defines the migration pathway of the gaseous CO_2 along the steepest gradient into anticlinal structures, where the gas accumulates. Heterogeneous permeability distributions may have an influence similar to topography, and can control the migration pathway, but this has not been simulated.

The gas-phase-body shape is controlled by aquifer topography and groundwater flow. Because the gas-phase body is the source of the dissolved CO_2, its width perpendicular to the groundwater flow defines the width of the impact area, while the length of the impact is determined by the pore velocity of groundwater flow. Hence, the area where geochemical reactions are expected is a combination of the extent of the gas-phase body and the extent of the dissolved CO_2.

In a worst-case leakage accident, in which the CO_2 has already leaked into a protected potable aquifer, impact scenario simulations can be used to develop high-resolution monitoring methods at the impacted area. Furthermore, the impact scenarios can be used for the conceptual design of remediation strategies.

The simulations allow some initial conclusions regarding risk assessment at the CLEAN site: For the investigated aquifer, the highest risk for an impact due to a CO_2 leakage is in the vicinity of the anticlinal structure. Hence, this area should be monitored more intensively. A possible remediation measure, e.g., that proposed by Esposito and Benson (2012), could be installed at the highest point of the anticlinal structure. Similar simulations should be performed for deeper potable aquifers.

In general, sensitivity analyses for the leakage rates and locations should be done as part of site-specific scenarios. If there is no obvious leakage pathway due to geological structures, like faults, or anthropogenic pathways, like bore holes (Nordbotten, 2009), the probability of a CO_2 leakage location would be the same for the entire model domain. For risk assessment, numerous scenarios of a site with different leakage locations and rates should be simulated.

Combining the results of the numerous leakage scenarios shows the areas with the highest probability for gas-phase-body accumulation. Accounting for the transport of dissolved CO_2 would indicate the area of highest risk for CO_2 leakage in the target aquifer.

Further work needs to be done on site-specific numerical simulations to transfer the results to real field sites, such as:

- General research geared toward evaluating the influence of hydraulic heterogeneities on gas-phase-body migration.
- Investigation into heterogeneities at large field sites.
- The influence of geochemical reactions on the migration behavior of gaseous CO_2: the coupling of geochemical reactions and multiphase flow in a site-scale simulation.
- Investigation of how to handle geochemical heterogeneities at large scales.

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