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## Dissolved CO<sub>2</sub> Storage in Geological Formations with low Pressure, low Risk and large Capacities

Michael Kühn<sup>a,b,\*</sup>, Thomas Kempka<sup>a</sup>, Marco De Lucia<sup>a</sup>, Magdalena Scheck-Wenderoth<sup>c,d</sup>

<sup>a</sup>GFZ German Research Centre for Geosciences, Fluid Systems Modelling, Potsdam, Germany

<sup>b</sup>University of Potsdam, Institute of Earth and Environmental Science, Potsdam, Germany

<sup>c</sup>GFZ German Research Centre for Geosciences, Basin Modelling, Potsdam, Germany

<sup>d</sup>RWTH Aachen, Faculty of Georesources and Materials Engineering, Aachen, Germany

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### Abstract

Geological CO<sub>2</sub> storage is a mitigation technology to reduce CO<sub>2</sub> emissions from fossil fuel combustion. However, major concerns are the pressure increase and saltwater displacement in the mainly targeted deep groundwater aquifers due to injection of supercritical CO<sub>2</sub>. The suggested solution is storage of CO<sub>2</sub> exclusively in the dissolved state. In our exemplary regional case study of the North East German Basin based on a highly resolved temperature and pressure distribution model and a newly developed reactive transport coupling, we have quantified that 4.7 Gt of CO<sub>2</sub> can be stored in solution compared to 1.5 Gt in the supercritical state.

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### 1. Introduction

Geological CO<sub>2</sub> storage is discussed as promising mitigation technology to reduce CO<sub>2</sub> emissions from combusted fossil fuels into the atmosphere [1-2]. This holds true even more after the United Nations Conference of Parties 21 (UN COP21) Climate Change Conference, which concluded that carbon capture and storage (CCS) is very

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\* Corresponding author. Tel.: +49-331-288-1594; fax: +49-331-288-1529.  
E-mail address: [michael.kuehn@gfz-potsdam.de](mailto:michael.kuehn@gfz-potsdam.de)

much part of the toolkit required to help keep greenhouse gases under control [3]. However, major concerns regarding CO<sub>2</sub> storage are the pressure increase and saltwater displacement in the mainly targeted deep groundwater aquifers [4-5]. Pressure increase could lead to disintegration of caprocks [6] or reactivation of faults [7-9] and subsequently to leakage of CO<sub>2</sub> [10]. Saltwater displacement is a threat for drinking water reservoirs in shallow groundwater systems above the storage complex, if migration pathways for the brines exist [11-13].

A solution for this issue could be storage of CO<sub>2</sub> in deep saline aquifers exclusively in the dissolved state, because this significantly reduces the volume required for typical reservoir conditions, compared to structural or stratigraphic trapping [14]. In that way the overall footprint of a storage project and its potential impact on groundwater resources is reduced. As much as the injection pressure is minimized is the risk, regarding incidents as described above. However, the operational costs of CCS in combination with surface dissolution are estimated to be 20% higher than the standard process and the capital costs are 50% greater, mainly due to a larger number of wells required. Altogether, the additional power consumption is estimated with 3% to 8% [15-16]. Finally, it is the question of available storage capacity which we address here for a first regional case study.

Exemplarily, we have chosen the North East German Basin (NEGB) and performed numerical simulations to quantify how much CO<sub>2</sub> could be theoretically stored in solution depending on pressure, temperature and salinity. Because the Buntsandstein provides the largest storage potential in the investigated area [17] this was our target formation as well. If the amount was significant, it would be an option to produce water from a suitable formation at depth, enrich it with CO<sub>2</sub> at the surface and reinject the solution into the reservoir again. Finally, we oppose the determined storage capacity of dissolved CO<sub>2</sub> for the entire area with the previously identified potential sites for storage of supercritical CO<sub>2</sub> in the respective region.

## 2. Numerical simulation method and model setup

The basis for our simulations is a 3D structural model of the NEGB developed by Scheck and Bayer [18]. It describes distribution and geometry of the stratigraphic layers as well as lithology-dependent physical rock properties and is constructed from well data, thickness as well as depth maps and integrates seismic and gravity data. The model covers an area of approximately 230 km by 330 km extending down to 33 km depth and its vertical resolution is determined by the number of stratigraphic layers. It provides us the depth map for the Buntsandstein and a temperature distribution of high precision therein for the following steps (Fig. 1).

The software package Petrel [19] is used as pre- and post-processor and integrates the reservoir simulator TOUGH2-MP [20] as well as a newly developed reactive transport coupling between R and PHREEQC [21] in a workflow, which has been successfully applied and evaluated for predictions of long-term site stabilization regarding CO<sub>2</sub> storage in deep saline aquifers [22].

The structural model of 740,600 elements comprises the Buntsandstein which occurs at depth from 250 m to maximum 5,000 m in the investigated region. We assumed a constant ratio between permeable and impermeable layers based on site-specific knowledge [12]. Compared to a total thickness of the Buntsandstein of 680 m at this location, the hydraulically active horizon measures only 51 m, resulting in an equivalent effective porosity of 1.3% for the stratigraphic unit. The temperature range is between 15 °C and 150 °C. With an initial hydrostatic pressure gradient of up to 50 MPa at depth, the resulting pressure in the system is calculated in an iterative process depending on the given temperature range and salinity (Fig. 2). Data for water salinity increase from 0 g/L at the top of the model to 250 g/L at 1,750 m as given by Wolfgramm et al. [23]. Below 1,750 m salinity remains constant. Brine density is parameterised following Rowe and Chou [24] after tests against Mao and Duan [25], which indicated a maximum deviation of only 0.4% and at average even less than 0.01%. Temperature, pressure and salinity data are transferred to PHREEQC [26] and the maximum soluble amount of CO<sub>2</sub> is determined accordingly and with respect to the available amount of water in the formation depending on its porosity. The accuracy of the PHREEQC model was evaluated against the one from Duan and Sun [27]. Both solubility models differ by only 0.2% from each other. We assume that water-rock-interactions can be neglected within this first order approach.

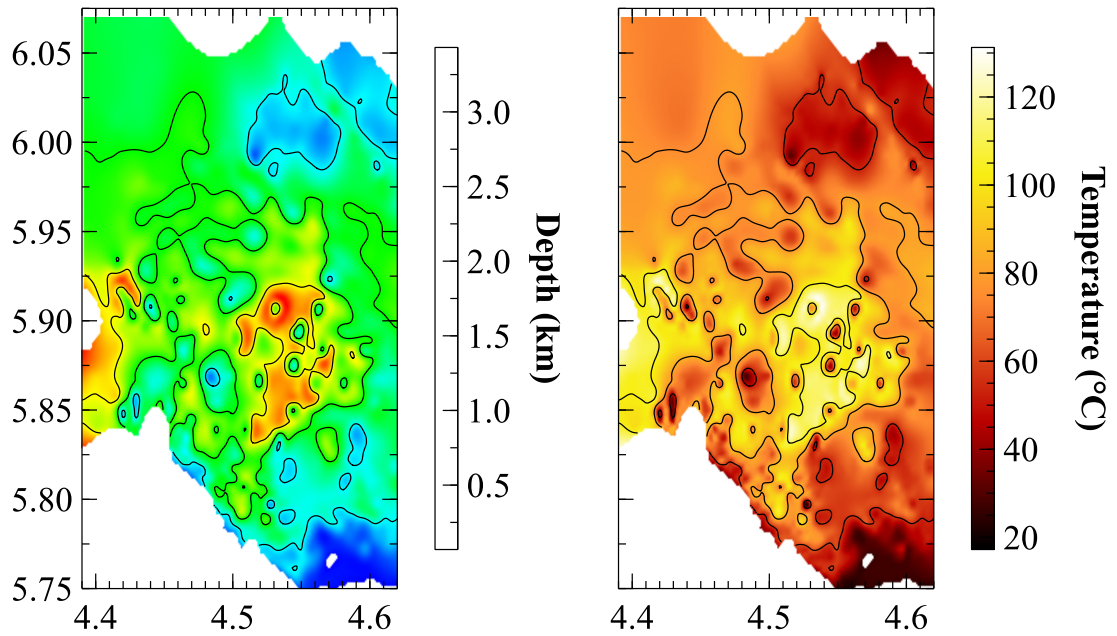


Fig. 1. Basic structural model from Scheck and Bayer [18] with the depth map (left) of the top of the Buntsandstein stratigraphic unit within the North East German Basin (NEGB) and the therein modelled temperature distribution (right) which shows a clear depth dependence. Axis labels (scaled by  $10^{-6}$ ) show Gauß-Krüger (Bessel, Potsdam) coordinates. Solid lines represent depth contours.

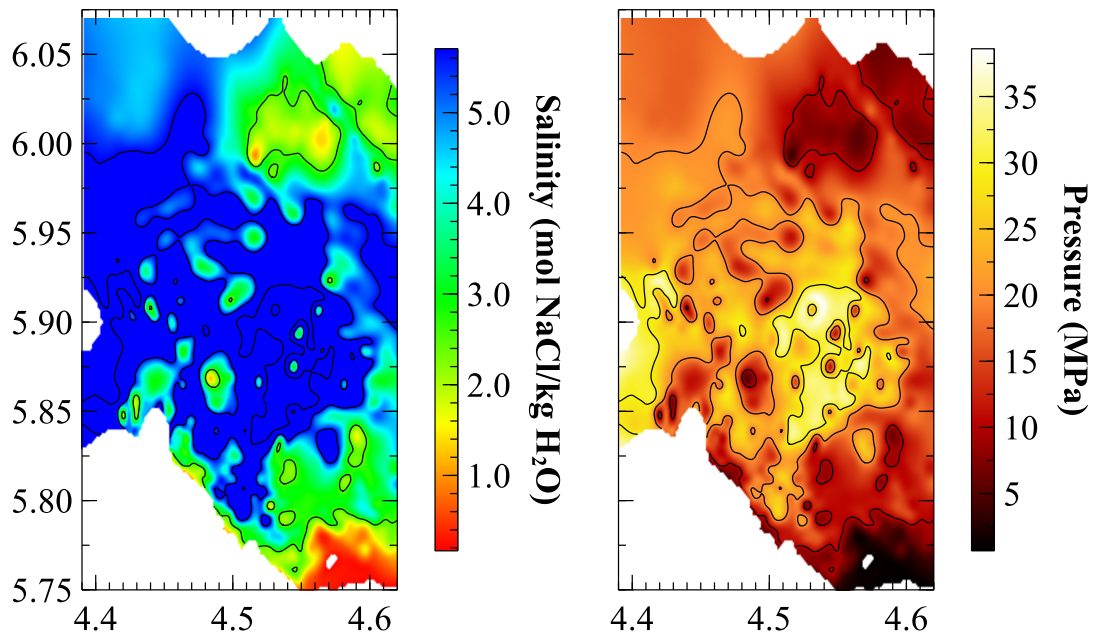


Fig. 2. Salinity distribution (left) and pressure depending on temperature and salinity (right) of the Buntsandstein. Axis labels (scaled by  $10^{-6}$ ) show Gauß-Krüger (Bessel, Potsdam) coordinates. Solid lines represent depth contours.

### 3. Simulation results

Following the workflow as described above, the resulting amount of CO<sub>2</sub> which depends on temperature, pressure and salinity ranges from around 0.1 kg to 0.5 kg per m<sup>3</sup> of rock (Fig. 3). The total storage capacity for CO<sub>2</sub> in the dissolved state within the Buntsandstein is 4.7 Gt under the given constraints for the investigated area of 230 km by 330 km within the NEGB. For the North German Basin (NGB), 408 potential storage sites are detected with a total capacity of 6.3 to 12.8 Gt [17] of which 32 are located within the investigated area of the NEGB. They provide a storage capacity of 1.5 Gt for supercritical CO<sub>2</sub>, which is only a third of the CO<sub>2</sub> determined here to be stored in the dissolved state.

We deduced as well that the control of the salinity gradient on CO<sub>2</sub> dissolution within the model is far stronger than the pressure gradient. The result is that significantly more CO<sub>2</sub> is dissolved within the areas of the region where the Buntsandstein is shallower (Fig. 3).

The influence of temperature on the dissolved amount of CO<sub>2</sub> was tested with a model in which we implemented a constant depth dependent thermal gradient instead of the simulation results from Scheck and Bayer [18]. Due to generally lower temperatures in the Buntsandstein of the region, the amount of storable CO<sub>2</sub> is overestimated by around 10%.

Even more important than pressure, temperature and salinity is the estimate for the effective porosity within the Buntsandstein. Based only on one location, we presumed an equivalent value of 1.3% for the stratigraphic unit, fixed for the entire investigated area. The storage capacity for CO<sub>2</sub> is directly proportional to porosity. With an equivalent porosity of around 2.5%, the dissolvable amount of CO<sub>2</sub> increased up to around 10 Gt.

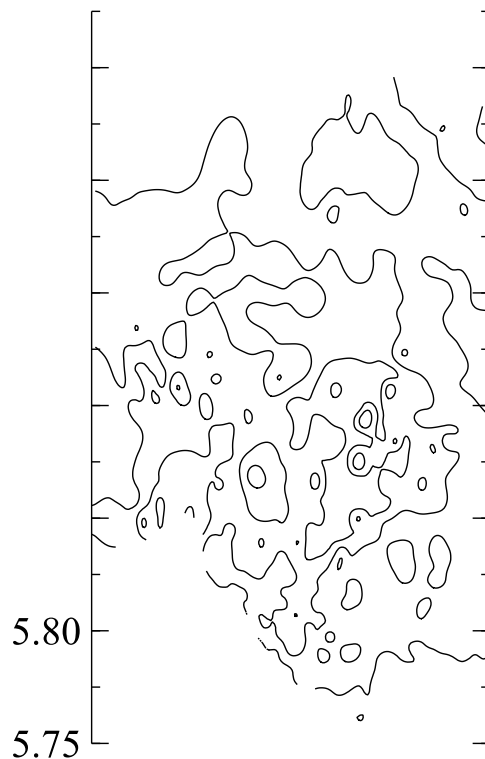


Fig. 3. Tons of dissolved CO<sub>2</sub> in the formation water of the Buntsandstein per volume of rock. Shallower parts carry more CO<sub>2</sub>. Axis labels (scaled by 10<sup>-6</sup>) show Gauß-Krüger (Bessel, Potsdam) coordinates. Solid lines represent depth contours.

#### 4. Conclusions

Major concerns with regard to geological CO<sub>2</sub> storage are pressure increase due to injection which could disintegrate formation caprocks or reactivate faults and subsequently lead to leakage of CO<sub>2</sub> in the extreme. However, salt water displacement will certainly occur and is a threat for drinking water reservoirs in shallow groundwater systems if migration pathways exist [4-13]. A suggested solution could be storage of CO<sub>2</sub> in deep saline aquifers exclusively in the dissolved state [14-16]. For that purpose, water from a suitable formation could be produced, enriched with CO<sub>2</sub> at the surface and reinjected into the reservoir. This would minimize pressure increase during re-injection and in conjunction reduce storage risks.

In our exemplary regional case study of the NEGB, we quantified the storage capacity for the Buntsandstein based on a stratigraphic model with highly resolved temperature distribution [18], coupled within a newly developed workflow with reactive transport [20-22]. Our simulation results imply that an amount of 4.7 Gt of CO<sub>2</sub> can be stored in solution over an area of 75,900 km<sup>2</sup>, compared to 1.5 Gt in supercritical state as proposed by the capacity estimates elaborated by the German Geological Survey [17]. A highly resolved temperature distribution of an investigated stratigraphy ensures that storage capacity is not overestimated. Nevertheless, it is shown that salinity dominantly governs solubility compared to temperature and pressure. Therefore, less CO<sub>2</sub> can be stored with depth if salinity increases. However, most inaccurate is the required estimate for the effective porosity, needed to finally quantify the storage capacity. A deviation of 10% would increase or decrease the calculated storage by the same amount, respectively. Altogether, we conclude that storage of CO<sub>2</sub> in the dissolved state in the subsurface is a promising option to minimize injection pressure and related risks, and furthermore provides significant storage capacities. The major advantage is the fact that CO<sub>2</sub> dissolved in brine can be injected with very little pressure increase, compared to a gas inj

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