Hydrogen storage in the Upper Rhine Graben

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Potential analysis

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1. Introduction

Energy generation from renewable sources will have to play a key role in meeting the climate targets set by the German government and the European Union for the years 2030 and 2050 (Deutscher Bundestag, 2019; European Council, 2014; European Parliament, 2020). For a 100% renewable and regional energy supply, intermediate energy storage becomes important for the security of supply (Mahlia et al., 2014). In addition to the possibilities of battery storage, hydropower storage, compressed air storage, thermal storage, and flywheel mass storage, power-to-gas storage is considered to have high potential (Blanco and Faaij, 2018; Götz et al., 2016). This technology uses electrolysis to convert excess energy into gas (hydrogen, methane), which can then be used to generate energy when needed. The gases can be temporarily stored both in pressurized containers above ground and in underground rocks. In order to store large amounts of energy, large volumes of gas are required, and geological storage is more suitable than above-ground storage media. This report, which was prepared within the framework of the INTERREG project RES-TMO (www.res-tmo.com), deals with the possibilities of geological hydrogen storage in the Upper Rhine region. It is largely based on data from the INTERREG project GeORG (GeORG-project team, 2013), which has worked out the geology of the Upper Rhine Graben (ORG) in cross-border detail and analyzed the potentials of CO2 storage in the ORG.

2. Geological storage of hydrogen

Basically, there are two different ways of storing gases safely underground: (1) In artificially created caverns in salt rocks, which are characterized by their very low permeability. (2) In the pore space of rocks with high porosity and permeability. Here it is key that these rocks are covered by a top layer of impermeable rock (barrier complex) so that the stored gases do not escape from the rock layer in which they are temporarily stored. In addition to aquifers, depleted oil and gas reservoirs can also be used as pore storage facilities. So far, there is only limited experience with both methods, cavern storage and pore storage, as far as the storage of hydrogen is concerned. However, both methods have been used extensively for decades for the temporary storage of natural gas. It should be noted that salt rocks in the subsurface are much less widespread than pore storage facilities that can be used for gas storage.

Salt caverns are artificially created in salt layers or salt domes using a borehole and circulating water. The size of the cavern can be precisely specified, depending on the geological conditions and future use, and ranges from a few 10,000 m³ to more than 1 million m³. The gas to be stored is injected or withdrawn through the well, so it also controls the maximum flow rates and thus the amount of energy that can be delivered when needed. Hydrogen has been successfully and safely stored in salt caverns in the United Kingdom since as early as 1972, and in the United States since the 1980s (Tarkowski, 2019).

For hydrogen storage in aquifers, there are currently only major field investigations, including in Argentina and Austria, but no active hydrogen storage facility is yet in operation. Experiences in aquifer storage of town gas obtained from coal gasification, which has a hydrogen content of about 50-60%, have shown that mineral reactions as well as microbial activity can occur in such storage (Heinemann et al., 2021; Tarkowski, 2019). On the one hand, this can lead to the formation of H2S, and on the other hand, hydrogen can be consumed and methane can be formed. The former has implications for the safety of storage operation, while the latter results in low energy content in the storage facility, but can also be used to produce "green methane" from hydrogen and carbon dioxide (Panfilov, 2010, www.underground-sun-conversion.com).

3. Geology of the Upper Rhine Graben

The Upper Rhine Graben is part of the Cenozoic rift system of western and central Europe. The rift has a length of about 300 km and a width of about 40-50 km and the rift axis is oriented approximately SSW-NNE. It is bounded by the Swiss Jura to the south and the Rhenish Massif to the north. The rift shoulder in the west is bordered by the Vosges Mountains and the Palatinate Forest, and in the east the Black Forest and the Odenwald form the rift boundary. The formation of the Graben can be traced back to the Alpine orogeny and the main phase of Graben formation took place from the Eocene to the Oligocene, about 45 to 25 million years ago (Geyer et al., 2011). The Mesozoic overburden has subsided to varying degrees in the interior of the graben and is found at depths greater than 5000 m below sea level in parts of the graben. On the trench shoulders, most of the overburden has been removed and the basement exposed. Tertiary and Quaternary sediments form partly thick sedimentary packages of up to 4000 m thickness in the trench interior. Tectonically, the Upper Rhine Graben is a complex fault zone affected by both east-west stretching and left-lateral longitudinal displacement. A variety of faults cross the graben, some of them are presumably reactivated basement structures while others are due to the graben formation itself. Due to tectonic movements, a complex pattern of tectonic blocks and floes is present today (Fig. 1).

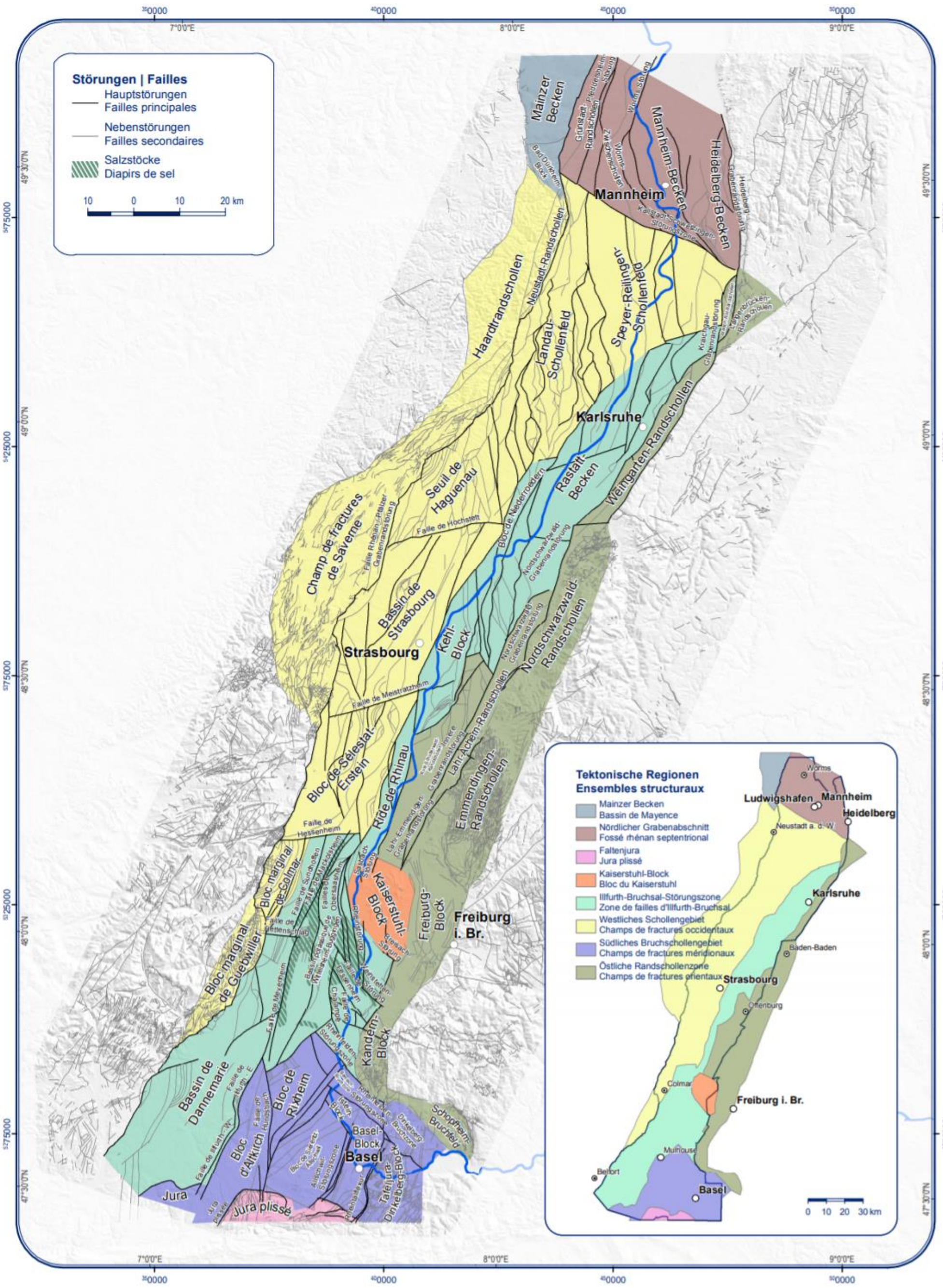

Figure 1: Tectonic overview map of the Upper Rhine region. The strong dissection into geological blocks and floe areas is very clearly visible. Aus GeORG Fachlich-Technischer Abschlussbericht des Interreg-Projekts GeORG Teil 4 - Atlas.

The geological structure of the Upper Rhine Graben comprises several stratigraphic levels (Fig. 2): above the deepest unit of the basement, which consists of metamorphic and igneous rocks, lie partly thick sediments of the Permocarbon. Above this, in the entire Upper Rhine Graben and partly also on the graben shoulders, Permian and Mesozoic sedimentary rocks of the so-called overburden follow. The younger Mesozoic units (younger Jurassic and Cretaceous) have not survived in the Upper Rhine Graben area. The overburden is followed by Tertiary rocks which form the trench fill. The youngest sediments in the Upper Rhine Graben are from the Late Tertiary (Pliocene) and Quaternary, but between these unconsolidated rocks and the other rocks of the Tertiary there is a layer gap of more than 10 million years. A detailed description of the individual stratigraphic sequences can be found in the GeORG final report (GeORG project team, 2013) and in Geyer et al. (2011).

Within the stratigraphic floors, several sedimentary sequences are present, which can be considered as reservoir-barrier complexes. These sequences consist (from oldest to youngest strata) of (GeORG project team, 2013):

- Permo-Carboniferous and Permo-Triassic sandstones (pore storage) and Lower and Middle Muschelkalk (barrier complex).
- Upper Muschelkalk (pore storage) and Keuper to Lower Jurassic (barrier complex).
- Middle Jurassic (main roe stone) as pore storage and Upper Middle Jurassic (barrier complex)
- Older Tertiary as both reservoir and barrier complex. Both pore storage and salt domes are present here.

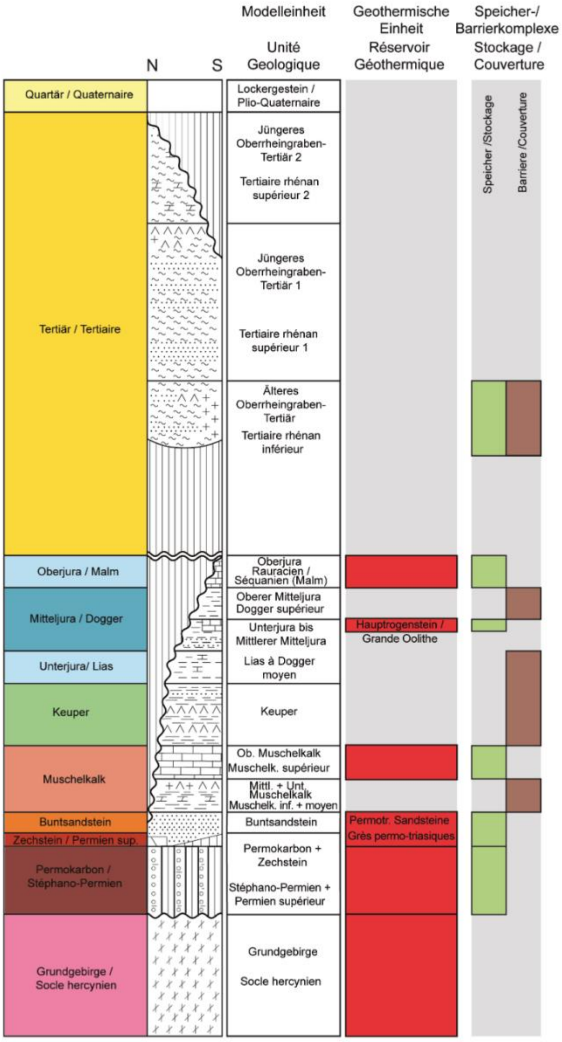

Figure 2: Sequence of strata in the Upper Rhine area. The storage and barrier complexes are shown on the right. From GeORG Technical Final Report Part 1 (GeORG project team, 2013).

4. Storage potentials Upper Rhine Graben

4.1 Pore storage

Since so far there has been very little experience with the storage of gases in carbonate rocks and these rocks often have a complex porosity and also permeability distribution, only the siliciclastic sandstones of the Permian and Triassic and Tertiary are considered below as pore storage in the ORG.

4.1.1 Permotriassic sandstones

Permotriassic sandstones are widespread in the ORG and range in thickness from a few meters in the south to more than 450 m in the central ORG (Fig. 3). The bedding sequence consists mainly of red and variegated sandstones with intercalated mudstones and siltstones. The medium to coarse sandstones are overlain by the base of the Lower Muschelkalk, which is marked by evaporites and carbonates. These form the barrier complex for the sandstones. Some of the sandstones show good porosity as shown in table 1, but permeabilities are mostly quite low (Fig. 4), which is also due to the partly high depth of injection of up to 5500 m (Fig. 3). Higher reservoir qualities (high permeability and porosity) are in essence due to the presence of fractures (GeORG-project team, 2013). Along faults, there are offsets of more than 1000 m and the sandstones are sometimes strongly tilted within individual blocks.

Table 1: Porosities and permeabilities of red sandstone samples from the ORG (GeORG-project team, 2013).

	Anzahl Proben	**Minimum**	**Maximum**	**Median**
Porosität (%)	254	1,4	24,2	9,51
Permeabilität (mD)	211	$1 * 10^{-3}$	1524	2,33

Hydrogen storage in permotriassic sandstones in the ORG is possible in principle, since the rocks have a (low) reservoir quality, are overlain by cap rocks, and have a high thickness. However, the low permeability has significant implications for use as pore storage for hydrogen, since rapid injection as well as production of hydrogen is not possible or possible only to a limited extent in reservoirs with low permeability. For long-term storage of CO2, which was the primary consideration in the evaluation of the geologic model during the GeoORG project, low permeabilities are not as problematic as for hydrogen storage, which is why the permotriassic sandstones were identified in this project as basically suitable for gas storage. Positive for a potential water storage is the tilting of the layers whereby structural trap structures are formed. When using such trap structures, however, the permeability of the faults has to be investigated in detail to avoid fluid loss. The greatest depth of the sandstones is in principle also rather negative for storage operations, because with increasing drilling depth also the costs for injection and production wells increase strongly.

Assuming that about 1% of the geographic spread of the Buntsandstein (with barrier complex, shaded area in Figure 3) can be used for hydrogen storage, and the average thickness of this reservoir is 200 m, porosity 9.51%, temperature 135°C, and reservoir pressure 31.5 MPa, then a volume of about 1.47*109 m^3 of hydrogen can be stored (Amid et al., 2016). This corresponds to an energy of about 4412GWh, which is about 24% of the renewable energy produced in Baden-Württemberg in 2019.

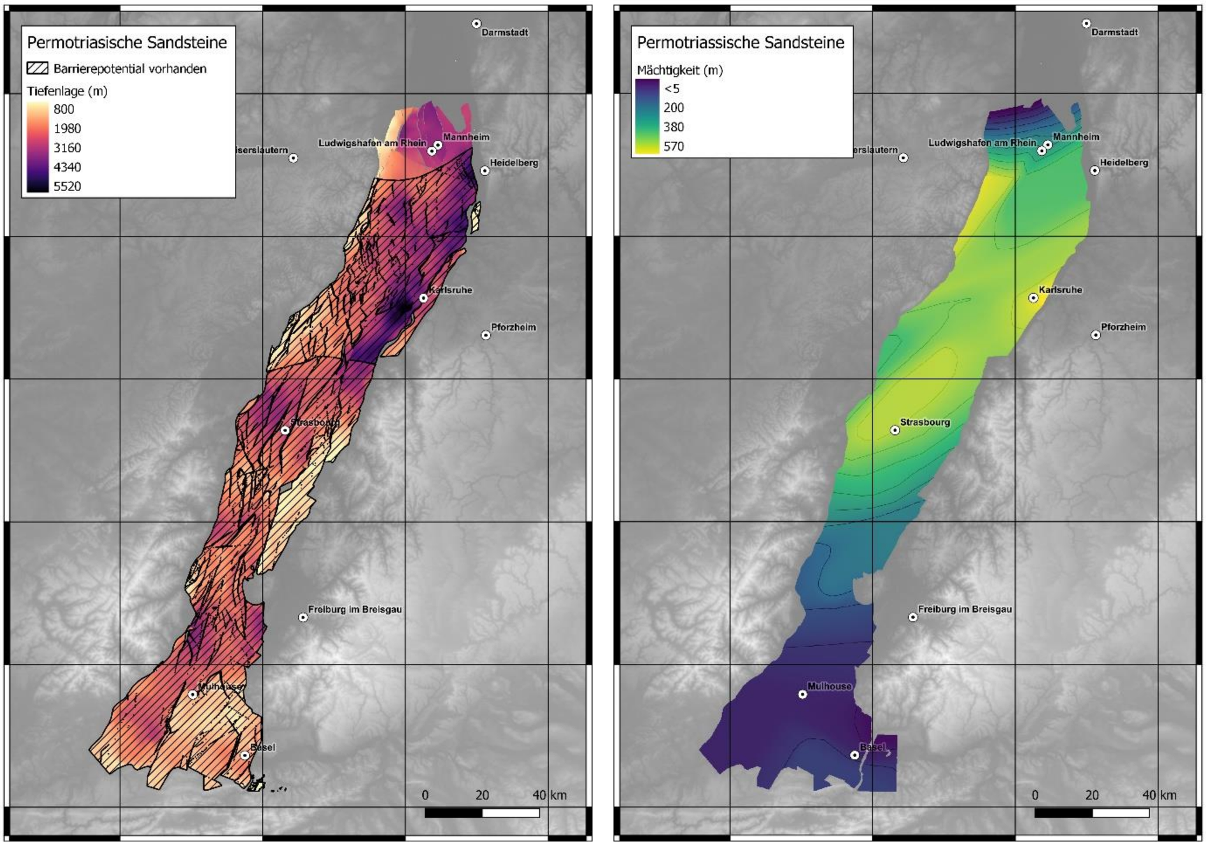

Figure 3: Left: Depth location of permotriassic sandstones in the ORG. Barrier potential is present in the shaded areas. Right: Thickness of permotriassic sandstones in the ORG, contours in 50m increments. Based on GeORG project team, 2013.

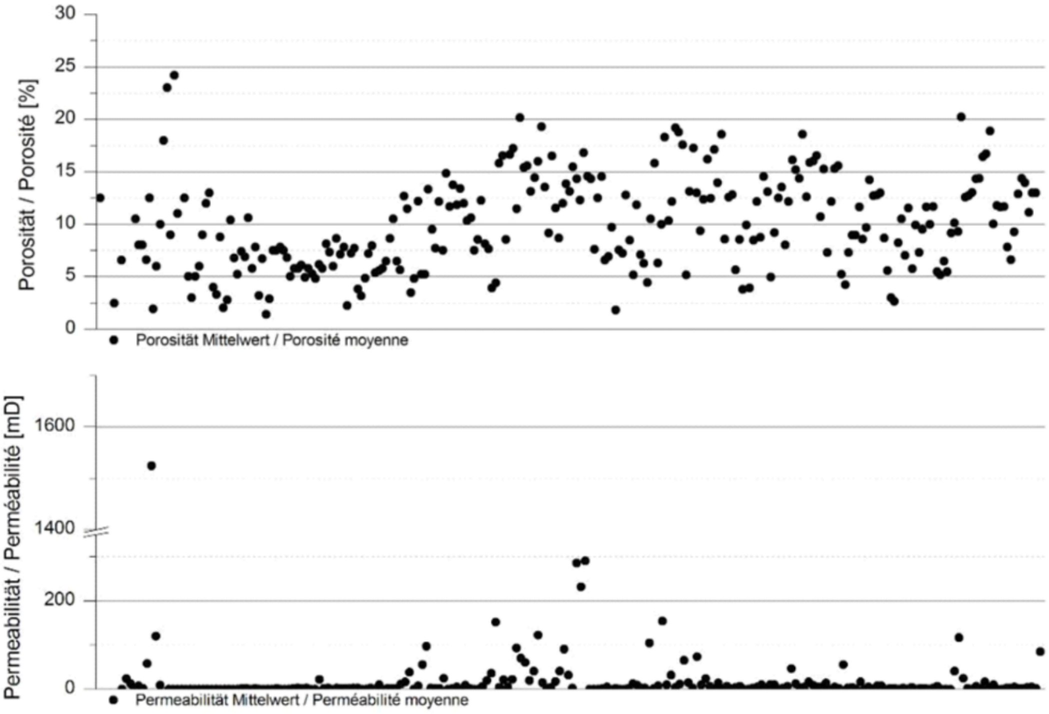

Figure 4: Porosity and permeability distribution of red sandstone samples from the ORG.

4.1.2. Lower Tertiary sandstones

In the Older Upper Rhine Graben Tertiary, the complex of the Pechelbronn Formation can be regarded as a reservoir and barrier complex due to the vertically and laterally changing lithological formations (GeORG-project team, 2013). These rock formations are widespread in almost the entire ORG (Fig. 6) and reach maximum thicknesses of up to 2300 m near Karlsruhe. Present-day depths range from a few meters to as much as 4800 m. Depending on location within the ORG and age, the Pechelbronn Formation varies in expression: The Lower Pechelbronn Formation consists mainly of alternating sand and marlstones, the Middle Pechelbronn Formation consists of clay marlstones, to the margin of depressional areas with dolomites, and the Upper Pechelbronn Formation has intercalations of fine to coarse grained sandstones in an alternating bedding of dolomitic marlstone and clay marlstone. In subisdence centers predominantly gray clay marlstones. The sandstone layers (mainly Lower and Upper Pechelbronn Formation) could be suitable as reservoir rocks, but they account for only about 0.3 to 1.1% of the total thickness of the formation. Porosities of the unit vary widely between 0.2 and 45.4%, with a median at 15.7% (Fig. 5). Permeabilities range from 0.01 to 5000 mD, with a median of 27 mD. It should be noted that the existing data set from the GeORG project does not include lithofacies, so it is not clear which of these rock parameters apply to the sandstone layers. However, it is reasonable to assume that the sandstones tend to be at the upper end of these distributions based on the grain size distribution.

Although hydrogen storage in the sandstones of the Older Tertiary is possible in principle, this is very dependent on the local rock parameters, especially the presence of thick sandstone packages, and this requires extensive preliminary exploration. Interestingly, terranets BW operates a pore storage facility for natural gas in Tertiary sandstones near Sandhausen (near Heidelberg). The pore storage facility is located at a depth of about 600 m and has a working gas volume of 30 million m^3. This example clearly shows that the storage of gases in the tertiary sediments of the ORG is possible.

Assuming that about 1% of the geographic spread of the Older Tertiary (with barrier complex, shaded area in Figure 6) can be used for hydrogen storage, and the average thickness of this reservoir is 10 m, porosity 25%, temperature 50°C, and reservoir pressure 10 MPa, then a volume of about 1.4*108 m^3 of hydrogen can be stored (Amid et al., 2016). This corresponds to an energy of about 419 GWh, which is about 2% of the renewable energy generated in Baden-Württemberg in 2019.

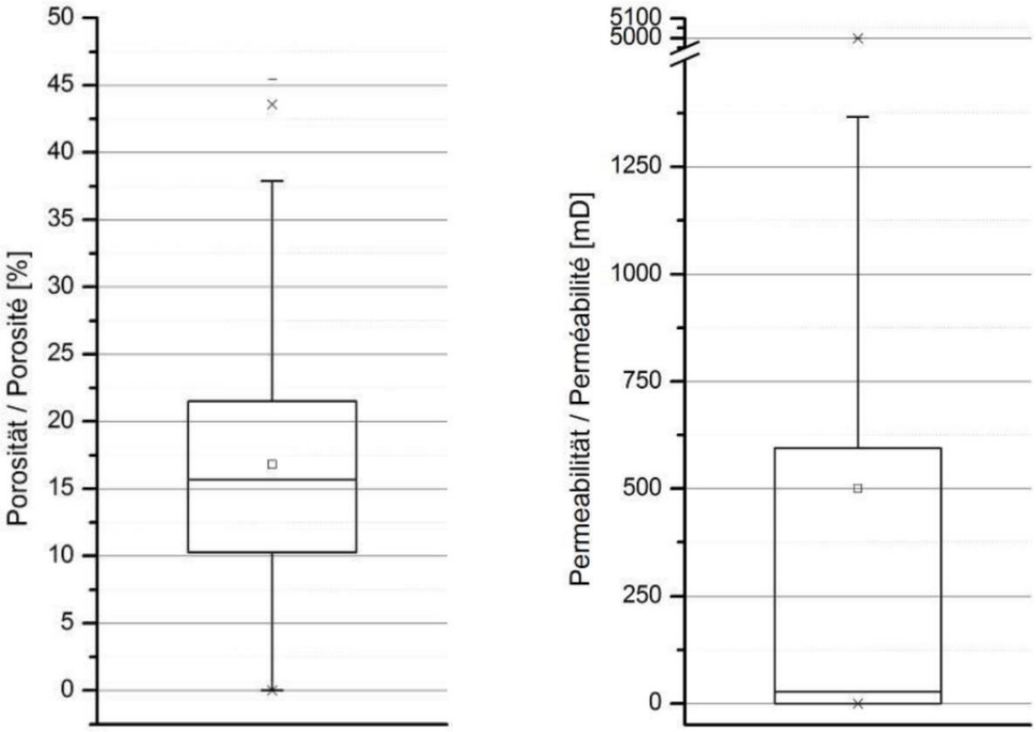

Figure 5: Boxplots of porosities and permeabilities in the Older ORG Tertiary. From (GeORG-project team, 2013).

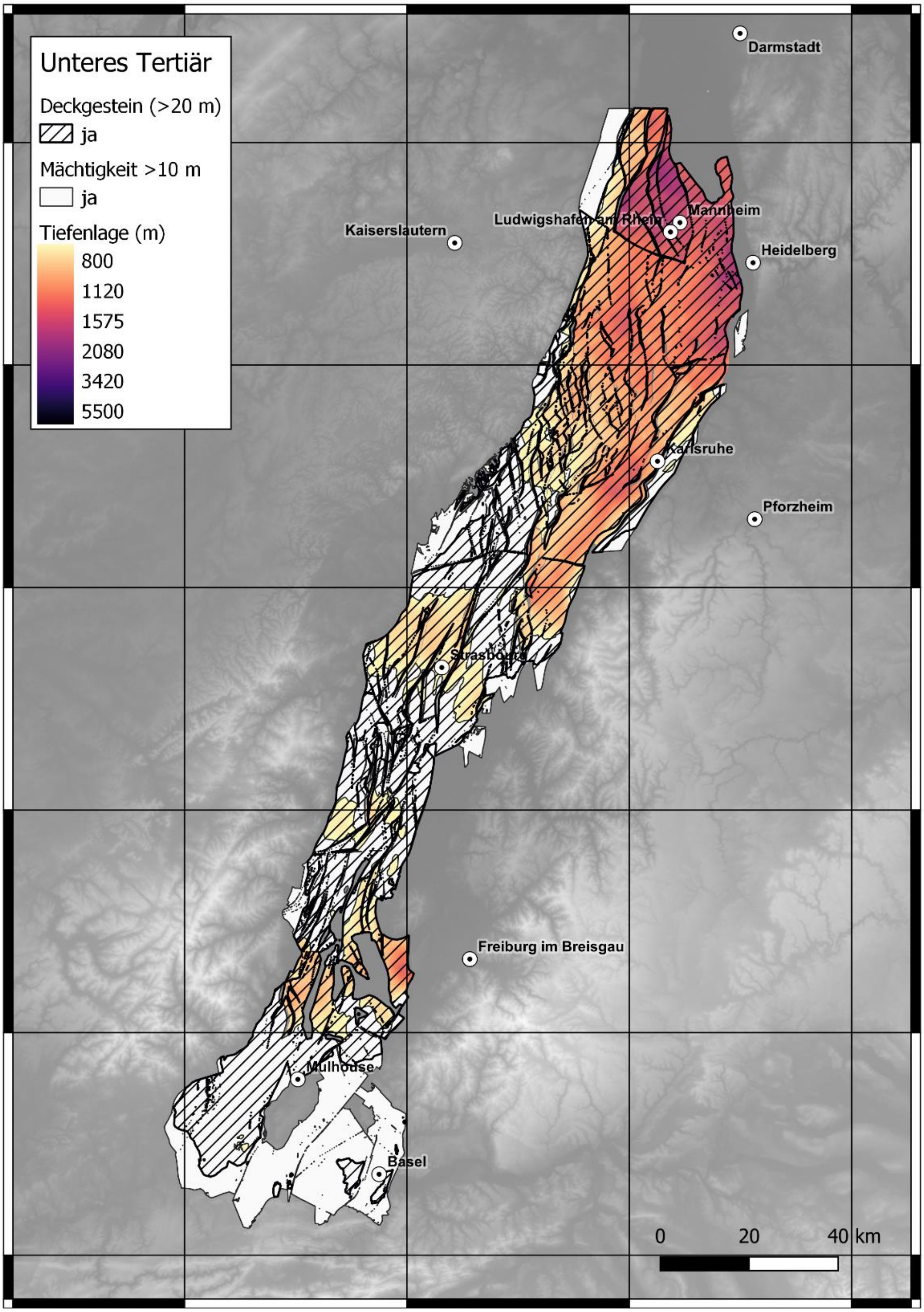

Figure 6: Distribution and depth of the storage and barrier complex Older (Lower) ORG Tertiary. In the white areas, the complex is located at a depth of less than 800 m, which, however, does not represent a fundamental hurdle for hydrogen storage.

4.2 Salt cavern storage

In the southern Upper Rhine Graben, precipitation of rock salt deposits several 100s of meters thick, often interbedded with anhydrite, gypsum, and dolomitic marlstones, occurred during the late Eocene and early Oligocene in basin structures such as the Dannemarie Basin and the Potassium Salt Basin (Hinsken et al., 2007). In some of these basin structures, the formation of salt diapirs up to 2000 m thick occurred during the Neogene (Fig. 7; BRGM, 1977; Hinsken et al., 2007; Lagneau-Herenger, 1965). These diapirs can be considered for cavern storage of hydrogen, but the shallow-bearing salinar formations can also be used economically for small-scale caverns. A minimum thickness of 70 m was specified as the minimum thickness of the salt layers in the InSpEE-DS project, which investigated the storage potential in salt layers in northern and central Germany in detail (InSPEE-DS, 2020).

A detailed calculation of potential cavern volumes would require a high-resolution seismic dataset and a good understanding of the internal structure of the diapirs, but only a 2D representation and profile of the salt diapirs is available from the GeORG project (Figs. 7 &8; GeORG-project team, 2013). Assuming that the diapirs are internally homogeneous in structure and consist of rock salt (no weaker horizons), an estimate of cavern storage potential can be made. Caverns with a volume of about 700,000 m³ are used as a basis; these have a height of 180 m and a radius of 25 m. In order not to affect the geomechanical stability of the diapir, the caverns must have a horizontal distance of at least two cavern diameters (100 m) from each other and from the edge of the diapir (Fig. 9). Thus, about 3200 caverns can be placed in the salt diapirs in the ORG (Fig.10). If the faults running through the diapirs were taken into account (Fig. 7), this number would be much smaller, but the height of the caverns could be doubled to 350 m. In total, hydrogen with an energy in the order of several thousand GWh could be stored in salt caverns in the ORG (Tab. 2).

Table 2: Hydrogen storage potential in salt caverns in the ORG.

Höhe (m)	Radius (m)	Volumen (m³)	Anzahl	Speicherpotential (GWh)
180	25	706.858	3200	6785
350	25	1.374.446	1000	4123

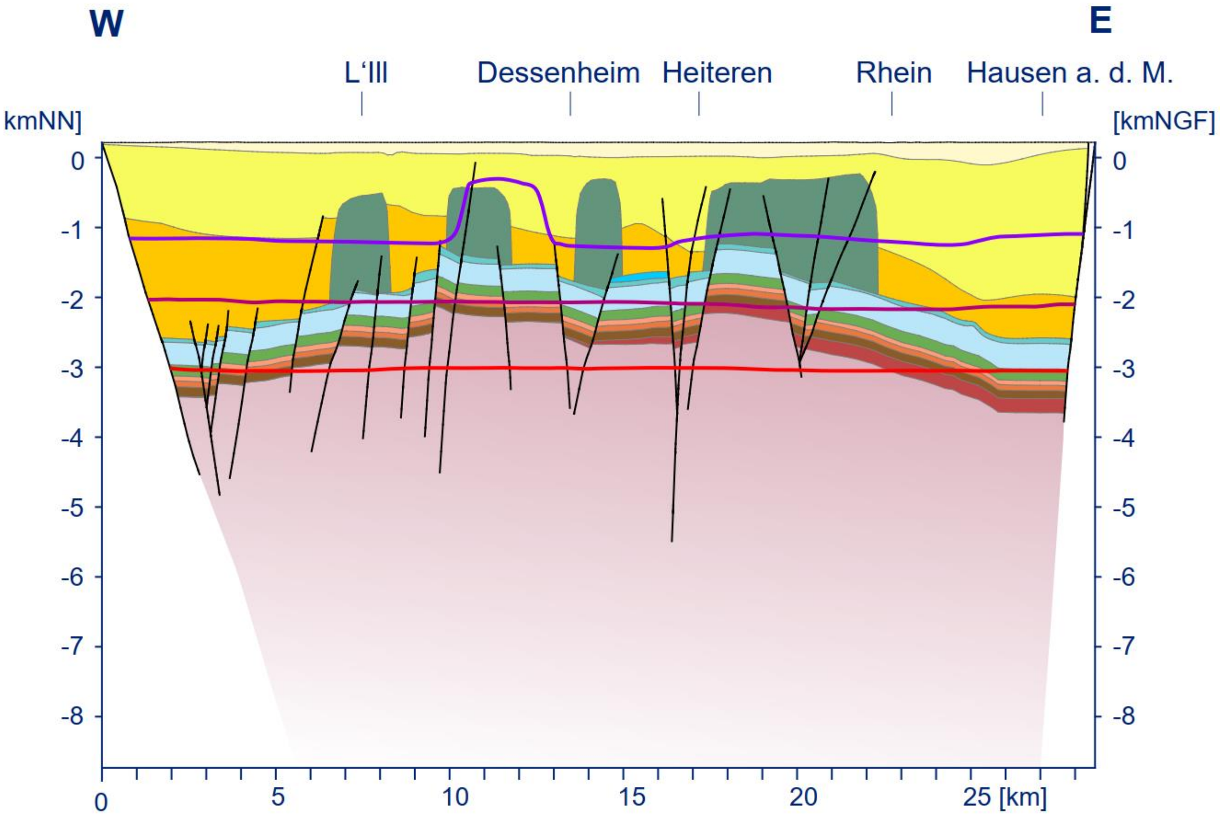

Figure 7: West-east profile through the ORG slightly north of Bad Krozingen (see Fig. 8). The salt diapirs are clearly visible and reach a thickness of more than 1.5 km. From GeORG-project team (2013).

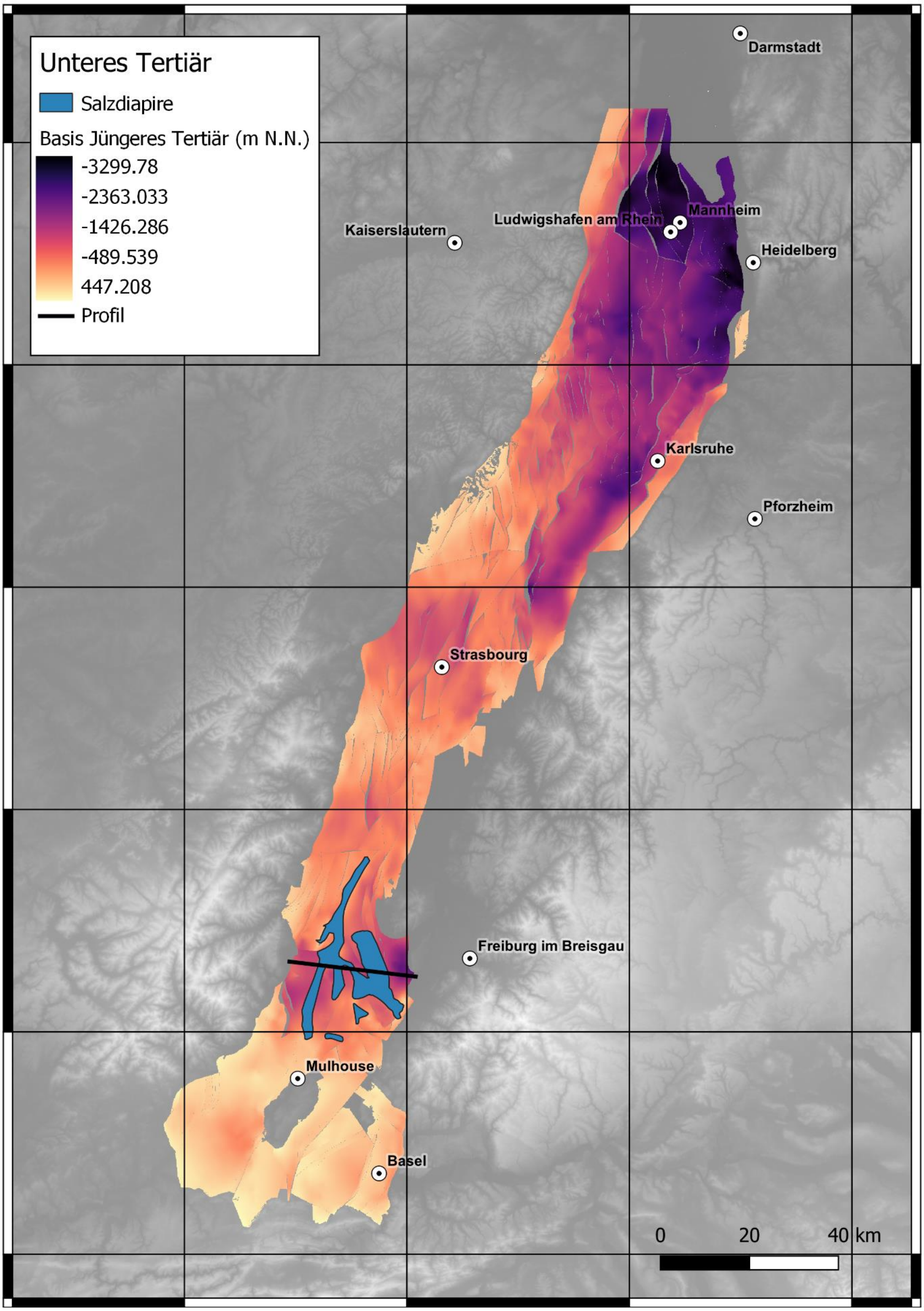

Figure 8: Location of the salt diapirs in the ORG. The profile line marks the location of the profile in Figure 7. Based on GeORG-Project team (2013).

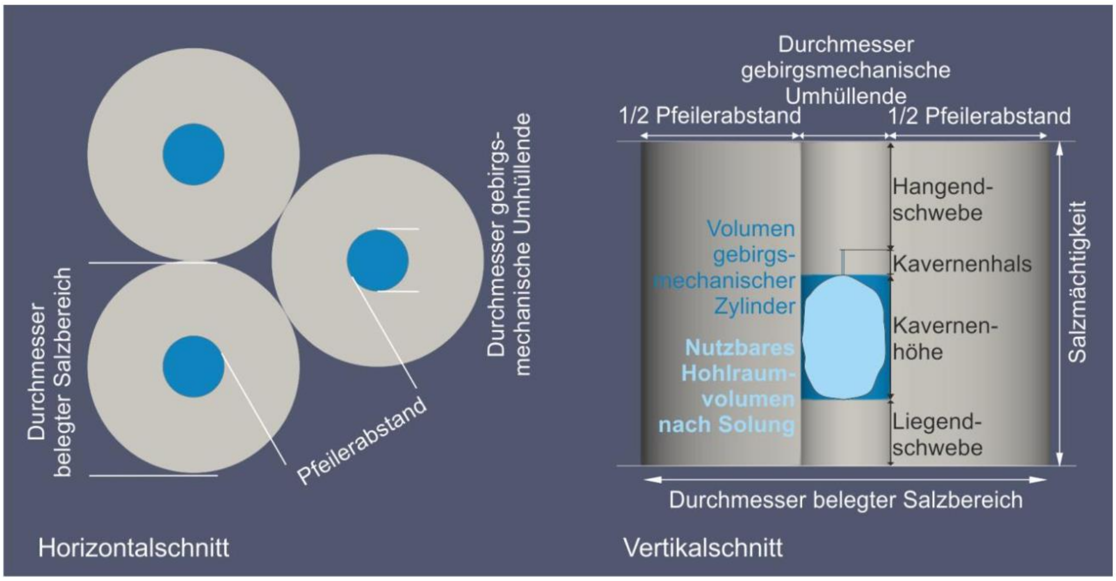

Figure 9: Required salt cylinder for the construction of a cavern. From (InSPEE-DS, 2020).

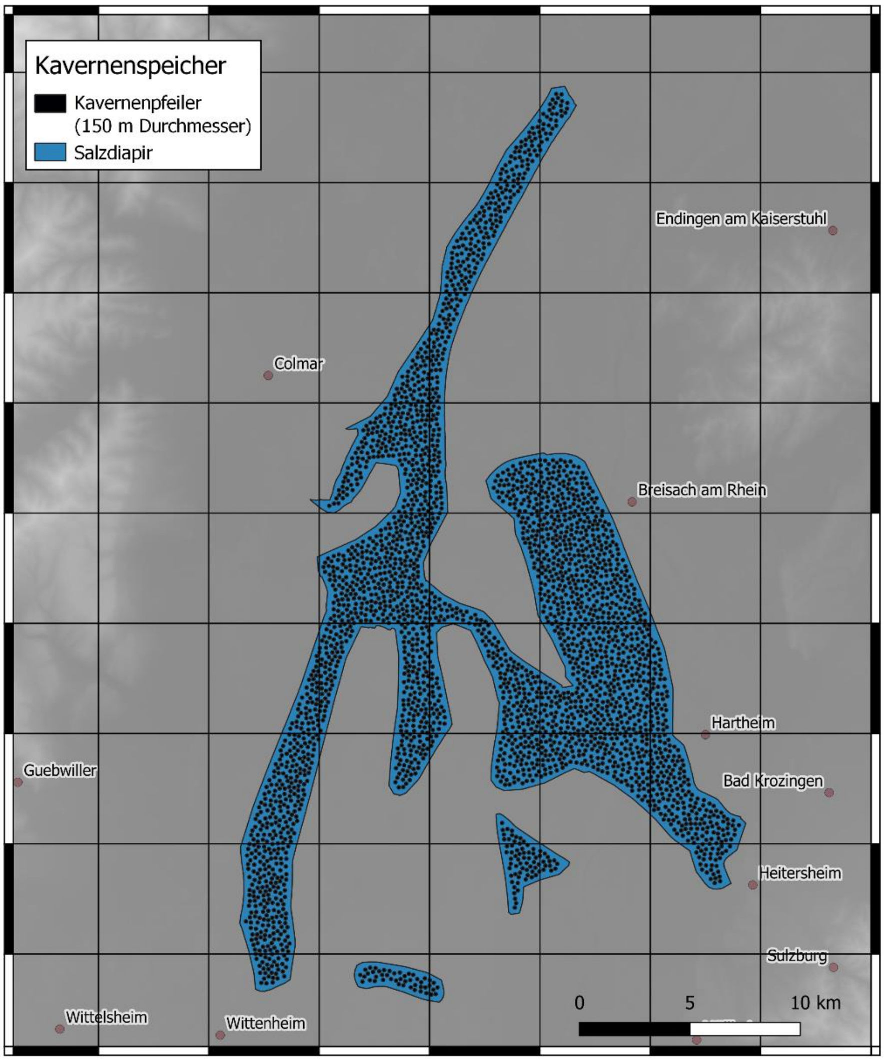

Figure 10: Cavern pillars for caverns with a diameter of 50 m in the salt diapirs of the southern ORG.A maximum of 3200 caverns could be created in total.

5. Recommendations

As explained in chapter 4, hydrogen storage in geological units of the ORG is possible in principle and the storage of energy up to 11 TWh would be feasible. While storage in pore reservoirs is very much dependent on local geological conditions and requires wide-area detailed (preliminary) investigations, the area in which salt cavern storage is possible can be spatially well delimited. Therefore, the following sequence is recommended for future geological energy storage in the ORG:

1. salt caverns in the salt diapirs of the southern ORG (Bad Krozinge-Colmar-Wittenheim).
2. salt caverns in the salt diapirs in the southern ORG (Wittelsheim-Staffelfelden).
3. pore storage facilities in Tertiary sandstones (marginal areas of the ORG, also in the northern OR (see existing gas storages)
4. pore storages in the permotriassic sandstones of the ORG

For a detailed potential analysis of the salt diapirs and salt layers in the southern ORG a high resolution 3D model is needed which should be based on (new) high resolution seismic data as well as existing borehole data. Such a project should be approached together with partners from the industry (energy companies/grid operators, salt cavern manufacturers (e.g. DEEP.KBB) as well as the corresponding geological state offices.

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