## Regional Capacity Estimates for Geological Storage of CO<sub>2</sub> in Deep Saline Aquifers – Upper Miocene Sandstones in the SW part of the Pannonian Basin

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## ABSTRACT

Deep saline aquifers are regarded as the most promising objects for  $CO_2$  geological storage, mainly due to their large storage capacities and extensive spatial distribution in most sedimentary basins. To estimate the storage capacity in this type of sinks presents a problem due to the lack of subsurface data.

Significant step from regional towards local capacity estimation is redefinition of regional storage capacities applying modified methodology for integrated studies of hydrocarbon reservoirs. The suggested procedure was investigated by detailed mapping of the West Sava aquifer. First, the cap rock was chosen based on its depth, thickness and lateral continuity, and then the target reservoir –Upper Miocene Poljana sandstone layers underlying the regional cap rock.

Their depth and effective thickness, as well as the subsurface pressure, temperature and resulting density of  $CO_2$  were mapped based on the well data. The aquifer body was then divided into square elements and the storage capacity was calculated for each of them.

The resulting map of specific storage capacity shows the areas of greater potential for geological storage that should be further investigated for detailed definition of the potential storage objects.

## **INTRODUCTION**

In order to mitigate growing negative effects of global climate change, significant reductions of anthropogenic greenhouse gas emissions are needed. Model simulations performed by Matthews and Caldeira [1], suggest that to obtain climate stabilization, emissions of  $CO_2$  from anthropogenic activities must be decreased to nearly zero. Moreover, models of Solomon et al. [2] suggest that changes in surface temperature, rainfall, and sea level are largely irreversible for more than 1000 y after  $CO_2$  emissions are completely stopped. According to Pacala and Socolow [3], efficient mitigation can be obtained only by implementation of various measures including fuel switching, increase of energy efficiency and energy conservation, use of renewable energy sources, enhancement of natural sinks and carbon capture and geological storage.

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Republic of Croatia became a party to the United Nations Framework Convention on Climate Change (UNFCCC) in 1996. Croatia has also signed and later ratified the Kyoto Protocol, taking the obligation to reduce the greenhouse gas emissions by 5% in the 2008-2012 period.

Major part of  $CO_2$  emissions in Croatia is from thermal power plants, municipal/district heating plants, and oil industry activities (about 2800 kt  $CO_2/yr$ ) as well as other industrial processes [4]. Total power plant emissions amount around 6000 kt  $CO_2/yr$ . Another large point source is the natural gas processing plant "Molve" where almost 700 kt of  $CO_2$  are separated from the natural gas streams and released into the atmosphere every year.

The fact that the emissions from point sources in Croatia amount to 26% of total CO<sub>2</sub> emissions allows significant reductions to be made by carbon capture and storage systems.

After Bachu [5], geological media suitable for  $CO_2$  storage must have sufficient capacity, adequate confinement in order to prevent the migration of buoyant  $CO_2$  to shallow aquifers and satisfactory injectivity to take in  $CO_2$  at the rate that is supplied from the emitter.

Hence, when investigating natural conditions for geological storage of  $CO_2$ , it is necessary to make as reliable capacity estimates as possible.

In the southern Pannonian basin, Upper Miocene sandstones are the most frequent type of reservoir rocks that might be used for geological storage of  $CO_2$  because they are numerous, they can be reliably correlated and usually are situated in the convenient depth range (800-2500 m). The first estimates of storage capacities in these formations were made within FP6 projects Castor and EU GeoCapacity, using simplified methodology based on a volumetric approach and with the single average value of thickness, temperature, pressure and porosity for each of the objects. Therefore, the obtained capacities were only theoretical, declared as the total pore space volume to make it clear that only part of it could eventually be used for  $CO_2$  storage.

Next step in regional capacity estimation would be to delineate the most perspective areas within aquifers and to really define the distribution of the cap rock and reservoir rock properties, applying modified methodology for integrated studies of hydrocarbon reservoirs. The most important aspects of this procedure are investigated by detailed mapping of the West Sava aquifer. The cap rock is defined based on its depth, thickness and lateral continuity. As the target reservoir the Upper Miocene Poljana sandstone unit underlying the regional cap rock is chosen.

# DEFINITION OF REGIONAL DEEP SALINE AQUIFER IN THE WESTERN PART OF SAVA DEPRESSION

Subsurface geological relations are the first critical factor to estimate the geological storage potential. The complex geological history of Pannonian basin resulted in a highly differentiated subsurface structure. Elongated basement highs and narrow depressions were developed during the Mid-Miocene rifting [6]. These narrow depressions, including Sava depression, are filled with the Neogene rocks. Thickness of Neogene and Quaternary rocks in western part of Sava depression is shown in Fig. 1. (exploration area is marked with rectangle). While their thickness reaches more than 5000 m in the central part of depression, in the western part of depression maximum thickness is barely over 3500 m.

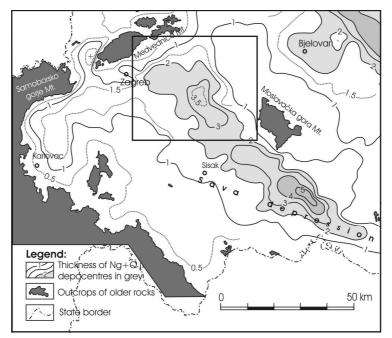


Fig. 1. Thickness of Neogene and Quaternary basin fill in the western and central part of Sava depression [7]

Generalised stratigraphic column of the southern Pannonian basin is given in Fig. 2. Basin fill is mostly composed of sediments. The oldest Neogene sediments in Sava depression are marine tuffaceous sandstones interpreted to be of Eggenburgian age, interlayered with gravely sandstones, marls and clays [8]. Otnangian is represented by coarse-grained clastics deposited in transitional, brackish to freshwater environment. According to LUČIĆ et al. [8], molasse sediments characteristic for beginning of basin opening also occur in Otnangian. Karpatian sediments were deposited in conditions of marine transgression that resulted in sedimentation of sandy and clayey marls with tuff intercalations [7]. Marine transgression continues during Early Badenian with sedimentation of basal conglomerates, coarse sandstones and lithotamnion limestones [8]. The end of Middle Miocene is characterised mostly by finegrained deposition in the starving brackish-water basin. Late Miocene age is characterised by the post-rift thermal subsidence of the Pannonian basin [7]. Lake Pannon deposits were formed in brackish (Pannonian) to freshwater environment (Pontian). Apart from local variations due to pre-Pannonian topography the sedimentary succession begins with littoral limestones and nearshore transgressive lag overlain by hemipelagic calcareous and clayey marls basin-wide. The deepest depressions were filled by lacustrine turbidite lobes and channel fills of considerable thickness, thus initial basin floor topography gradually became levelled. Turbiditic successions are overlain by shale-prone delta slope and sandy delta front to coastal plain sediments [7].

Pliocene and Quaternary rocks are sediments which were deposited in the remnants of Lake Pannon and in the subsequent fluvial systems [7]. These are mostly sands and sandy gravels with some clay and silt that have the largest thickness of 1000-1500 m in areas of continuous subsidence (Fig. 2.).

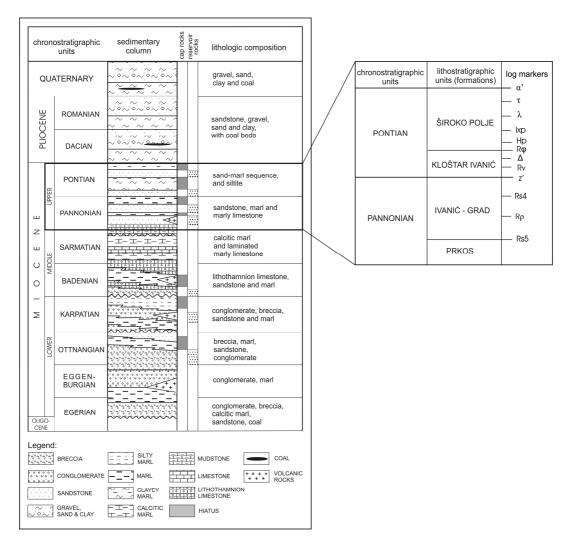


Fig. 2. Stratigraphic column of the Neogene basin fill of southern Pannonian basin (after [9]) with lithostratigraphic units and log markers within Upper Miocene sediments of Sava depression (after [10])

Taken in consideration the criteria of regional extension, depth and petrophysical properties, the most promising reservoir rocks that could be regarded as regional deep saline aquifers are the mentioned Upper Miocene sandstones of turbiditic and deltaic origin. Although not homogenous, these sandstones in Sava depression show relatively good reservoir characteristics: effective porosity is often higher than 20% and average permeability amounts to around  $60x10^{-3} \mu m^2$  [11].

Within the FP6 EU Geocapacity project, the generalised geometry of the Upper Miocene aquifer Sava West was delineated after the regional subsurface maps that were based on the data coming from the numerous exploration wells located in the area. Top of the aquifer is represented by the top of the Upper Miocene sandstone-marl sequence (Base Pliocene), while the base of the aquifer represents the contact with underlying older Miocene sediments (Base Pannonian). Defining the regional deep saline aquifer in this way is satisfactory only for the regional capacity estimates, often also referred to as the "theoretical capacity".

In order to enable more detailed characterization of reservoir rocks and more reliable regional capacity estimates that should serve as guidance to further exploration on local scale, regional deep saline aquifers defined within EU Geocapacity project need to be redefined. Namely, the

Sava West aquifer should be divided into several, most likely three aquifers, each comprising one lithostratigraphic unit (member) – Poljana sandstone of the Kloštar Ivanić formation, and Okoli and Iva sandstones of the Ivanić Grad formation.

To correctly delineate the regional aquifer, a regional cap-rock needs to be defined in terms of its extension, lateral continuity and thickness. Based on interpretation of 30 well logs (measurements of spontaneous potential and resistivity), the Graberski marl unit overlying the Poljana sandstone is chosen because it can be traced on all well logs and shows thickness varying from around 20m to more than 70m. It should be noted that this unit has already proved to be efficient cap rock for hydrocarbons. Further laboratory analysis is needed to prove its sealing properties for carbon dioxide.

#### ESTIMATES OF STORAGE CAPACITIES IN DEEP SALINE AQUIFERS

#### Overview of the existing methodologies

Methodology used to estimate storage capacities in deep saline aquifers significantly differs depending on the scale. For estimates on the regional scale static models based on volumetric approach or compressibility of fluids and rocks are commonly used. Volumetric method requires reservoir geometry, including its area, effective thickness, porosity and storage capacity coefficient or storage efficiency factor – number defining how much of pore space could be available for storage of  $CO_2$ . Compressibility method takes into account change of pore pressure with change of water volume and pore volume. Namely, during  $CO_2$  injection, pore pressure will rise, resulting in decrease of water volume and increase of pore volume [12].

Significant contribution to development of methodology for estimation of storage capacity in deep saline aquifers gave the studies of Task Force for Review and Identification of Standards for  $CO_2$  Storage Capacity Estimation of Carbon Sequestration Leadership Forum [13, 14]. Somewhat different approach was introduced by the Capacity and Fairways Subgroup of the Geologic Working Group of the DOE Regional Carbon Sequestration Partnerships [15].

Authors of CSLF studies emphasize that the geological storage of  $CO_2$  is achieved through a combination of physical and chemical trapping mechanisms. Physical trapping occurs when  $CO_2$  is immobilized as a free gas or a supercritical fluid. It includes static trapping in stratigraphic and structural traps and residual-gas trapping in the pore space at irreducible gas saturation. Chemical trapping occurs when  $CO_2$  dissolves in subsurface fluids (solubility and ionic trapping) and may then be involved in chemical reactions with the rock matrix (mineral trapping). For theoretical estimates on regional scale authors of CSLF studies [13, 14] propose calculation of capacity by static trapping in stratigraphic and/or structural traps present within the regional aquifer. For that purpose, it is necessary to calculate the volume of every trap, taking in consideration that part of pore water will not be substituted by  $CO_2$  during the injection, regardless of pressure applied:

 $V_{CO2t} = V_{trap} \times \Phi \times (1 - S_{wirr}) \equiv A \times h \times \Phi \times (1 - S_{wirr})$ (1)

where A represents average trap area, h is average thickness,  $\Phi$  is average porosity and  $S_{wirr}$  irreducible water saturation.

For each trap, obtained volume should be multiplied with corresponding average density of  $CO_2$  under conditions of pressure and temperature assumed for estimated average depth of a trap. This approach assumes that  $CO_2$  storage in saline aquifers demands stratigraphic and/or structural traps of significant dimensions. Also, it assumes that geometries of these traps are known, as well as irreducible water saturation, which is often not the case.

After Bentham and Kirby [16], storage of CO<sub>2</sub> can also be performed in aquifers with no specific large structural or stratigraphic closures as a target. In such aquifers injected buoyant

 $CO_2$  would fill small domes and undulations underneath the cap rock, thus effectively trapping a proportion of the injected  $CO_2$ . Depending upon the average "roughness" of the contact surface between the aquifer and the cap rock, the volume of the  $CO_2$  trapped in this way could be significant if the  $CO_2$  is distributed across the large area. Evidently, for such regional aquifers, formula suggested by CSLF can not be applied and different approach is required.

Approach proposed by USDOE [15] takes into account the pore space of entire aquifer and does not make distinction between CO<sub>2</sub> stored by various mechanisms:

(2)

 $M_{CO2} = A \cdot h \cdot \Phi \cdot \rho_{CO2} \cdot S_{eff}$ 

where A is surface and h is average effective thickness of regional aquifer,  $\Phi$  is average effective porosity,  $\rho_{CO2}$  is average density of CO<sub>2</sub> at conditions of pressure and temperature anticipated for a regional aquifer and S<sub>eff</sub> is a storage efficiency factor that reflects the total pore volume that could be filled with CO<sub>2</sub>. Storage efficiency factor includes variety of different parameters: fraction of the saline aquifer that is suitable for CO<sub>2</sub> storage, fraction of the geological unit that has the porosity and permeability required for CO<sub>2</sub> injection, fraction of net aquifer thickness contacted (occupied) by CO<sub>2</sub> as a result of CO<sub>2</sub> buoyancy and sweep efficiency (comprising the areal displacement efficiency, vertical displacement efficiency and pore-scale displacement efficiency).

## Description of the proposed methodology

Methodology proposed by USDOE [15] is here modified in a way that allows making of a more detailed numerical estimate. Area of a regional aquifer is divided into square elements and storage capacity is then calculated for each of them. As was mentioned previously, Upper Miocene sandstones were deposited in turbiditic and deltaic facies that caused the specific morphology of the sandstone bodies. Namely, they are characterised by considerable variations of thickness and porosity. On the other hand, structure of sedimentation basin (in this case Sava depression) causes variability of depth. Naturally, central part of depression is deeper and it shallows towards rims, so the deposits in the central part are situated in larger depths and the depth is decreasing going towards the margins.

The proposed methodology is tested on a regional aquifer Poljana (corresponds to lithostratigraphic member Poljana sandstone of the Kloštar Ivanić formation), the shallowest of the three regional aquifers assumed to be present in the western part of Sava depression.

One of the important parameters of regional aquifer is its thickness, which is actually effective thickness of Poljana sandstone. Map of the regional aquifer thickness, constructed based on interpretation of E-logs from 125 wells, is shown in Fig. 3. It shows that sandstone layers are best developed in the central, deepest part of depression, where their total thickness generally varies between 100 and 200m. Boundaries of deep saline aquifer shown on the map are set with regard to three criteria: pinch-out line of Poljana sandstones, depth of aquifer top (800 m is regarded as depth boundary that provides pressure and temperature conditions needed for  $CO_2$  to be in supercritical state) and boundary of estimation area mostly defined by data availability, i.e. lack of data to the southeast. While the first two criteria depend upon natural conditions, the third one should be looked at as the margin of the area that was mapped in this phase (corresponding to the license block in the upstream petroleum exploration).

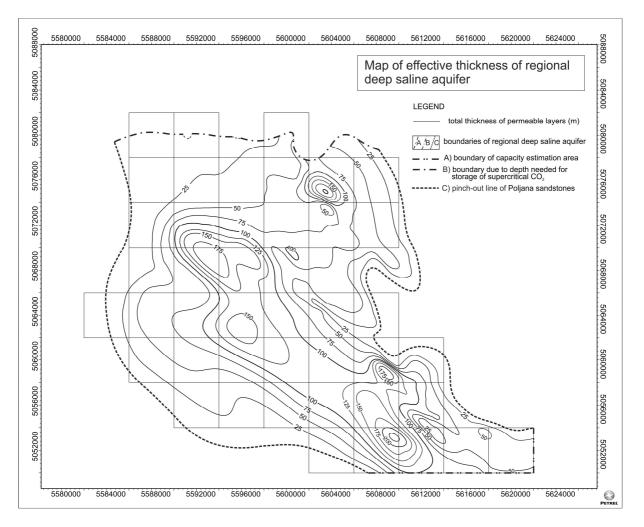


Fig. 3. Map of effective thickness of the Poljana regional deep saline aquifer

In order to obtain the values of pressure (assuming hydrostatic gradient) and temperature required for calculation of  $CO_2$  density, it was necessary to map depth of the regional aquifer. Depth of the aquifer has been approximated with average depth of Poljana sandstones member. Thus, the average depth taken into calculation is in fact average depth to middle of interval between E-log markers Rv and z'.

Since the measured data were scarce and unevenly distributed, the pressure values were calculated for each well on the basis of estimated depths, assuming hydrostatic pressure gradient. This assumption is reasonable considering the fact that pressure measurements from oil and gas reservoirs in the Poljana sandstones show values similar to values calculated with hydrostatic gradient. Temperature values were also calculated on the basis of estimated average depth, using regional map of geothermal gradient [17], assuming average annual temperature of 10.7°C calculated from average monthly values of temperature measured on meteorological station Maksimir (Zagreb) in year 2009. Estimated values of pressure and temperature were put in the density diagram [18] from which density values for given pressure-temperature conditions were read.

Another important parameter that needs to be considered when estimating storage capacity of a regional deep saline aquifer is porosity. Data on porosity values are available only from oil and gas fields located in the area (Kloštar, Žutica, Ivanić and Dugo Selo), where the average porosity values amount between 16 and 25% [19, 20, 21]. After JÜTTNER et al. [22], for the Okoli field average porosity value amounts to 18%. Since no other data on porosity were available, the porosity value of 18% is chosen as average for the whole regional aquifer.

Effect of variability of porosity values should certainly be implemented in future detailed estimates of storage capacity.

Map of specific storage capacities of Poljana sandstone deep saline aquifer is shown in Figure 4. Aquifer area is divided into quadratic blocks and in the centre of each block is its number and estimated specific storage capacity in  $t/km^2$ . It is visible that the largest specific storage capacities, exceeding 200000 tCO<sub>2</sub>/km<sup>2</sup>, are estimated in four blocks in the central and southeastern part of the deep saline aquifer. Also, a significant storage potential can be assigned to eight blocks with storage capacities ranging between 150000 and 200000 tCO<sub>2</sub>/km<sup>2</sup>. The blocks of these two categories can be regarded as the most promising areas where the further research should be undertaken, mostly focusing on more detailed characterization of cap rock and reservoir rock properties.

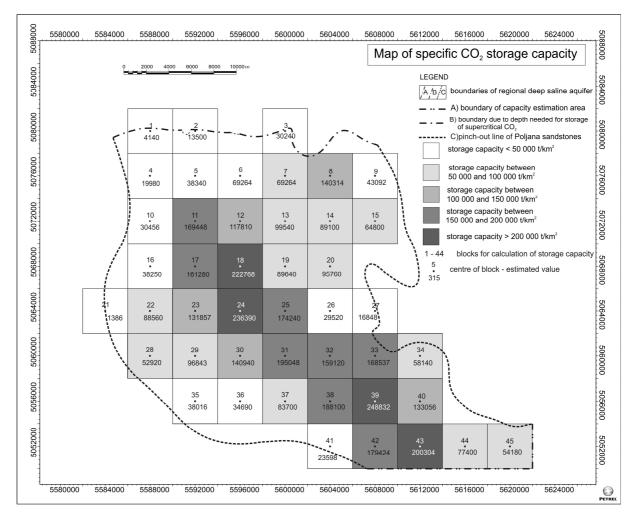


Fig. 4. Map of specific CO<sub>2</sub> storage capacity of the Poljana regional deep saline aquifer

## CONCLUSION

Due to a fact that rather large part of  $CO_2$  emissions in Croatia is coming from a few large point sources (almost 7000 kt  $CO_2/yr$ ), implementation of  $CO_2$  capture and geological storage should be regarded as an option for reduction of  $CO_2$  emissions.

In order to make reliable storage capacity estimates for deep saline aquifers in the Croatian part of the Pannonian basin a system will have to be designed according to the recommendations of DOE [15]. With our specific composition of the sedimentary basin fill,

i.e. the multiple and rather thin layers of finegrained sandstones interlayered with silty marls, and with marked neotectonic activity (and resulting structural deformation), and considering the fact that a number of old exploration wells is available, a tailored methodology is suggested which might cover all of the main critical parameters. They can be totally prohibitive, such as non-existing cap rock, or an open fault, or they may significantly influence the storage capacity, such as underground pressure, porosity or permeability. All of them will have to be identified and quantified at a specific storage location but before such locations are defined, a regional mapping of the storage capacity will have to be completed just to enable the authorities to plan the regions in which concessions will be issued for the more detailed exploration that will eventually lead to planning and construction of the subsurface storage facility for  $CO_2$ . To correctly plan the development of a CCS system, this regional exploration must not be omitted in every potential area and for each of the prospective units.

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