



GHGT-9

## Possibilities for geological storage and mineral trapping of industrial CO<sub>2</sub> emissions in the Baltic region

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### Abstract

Industrial CO<sub>2</sub> emissions and possibilities for geological storage of CO<sub>2</sub> in Estonia, Latvia and Lithuania were studied within the framework of EU GEOCAPACITY and CO<sub>2</sub>NET EAST projects supported by European Commission Sixth Framework Programme (FP6). Twenty-two large industrial sources produced 14.5 Mt of CO<sub>2</sub> in Estonia, 1.9 Mt in Latvia and 4.8 Mt in Lithuania in 2007. The two greatest Estonian power stations, using oil-shale, produced 9.4 and 2.7 Mt of CO<sub>2</sub>. The Baltic States are located within the Baltic sedimentary basin, the thickness of which varies from 100 m in NE Estonia up to 1900 m in SW Latvia and 2300 m in western Lithuania. The most prospective formation for the geological storage of CO<sub>2</sub> is the Cambrian reservoir, with an estimated potential of 300 Mt of CO<sub>2</sub> in 15 large structures located in Latvia. Geological conditions are unfavourable for CO<sub>2</sub> storage in Estonia, while mineral trapping with watered oil shale ash can bind up to 10–12% of CO<sub>2</sub> emissions produced by the above-mentioned two large stationary sources. In Lithuania the capacity of CO<sub>2</sub> storage in Cambrian and Devonian structures as well as in oil fields is negligible, but solubility and mineral trapping, including carbonation of the serpentinites hosting rich iron deposits can sink up to 18.6 Gt of CO<sub>2</sub>.

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Keywords: CO<sub>2</sub> emissions; Estonia; Latvia; Lithuania; geological storage; structural trapping; mineral trapping; serpentinites; oil shale ash

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### 1. Introduction

According to the Kyoto protocol signed by the Baltic countries in 2002, during the commitment period of 2008–2012 air-polluting greenhouse gases (GHG) emissions should be reduced by 8% compared to the 1990 level. The Post Kyoto Targets published in a European Strategic Energy Plan “Towards a low carbon future” aimed to reduce GHG emissions by 20% by 2020 and by 60–80% by 2050. Compared to 1990, the GHG emissions decreased in Latvia and Lithuania by more than 50% and in Estonia by 49%. However, the changing energy market and increasing industrial growth urge to evaluate different options of reducing CO<sub>2</sub> emissions, including the assessment of the potential of geological sinks and mineral trapping. In 2006 all three Baltic countries started an inventory of



In Latvia, the main CO<sub>2</sub> producers are situated in the western part of the country. The Liepaja metallurgical enterprise emitted 0.356 Mt of CO<sub>2</sub> and three electric power stations in the Riga area emitted 0.567, 0.386 and 0.23 Mt of CO<sub>2</sub> (year 2007). There are two CO<sub>2</sub> source clusters in Lithuania, situated respectively in the north-west and south-east of the country. The greatest GHG producer the Mažeikiai oil refinery reduced the emissions from 1.87 Mt in 2005 to 1.20 Mt of CO<sub>2</sub> in 2007, while the other largest source, the Akmenė cement plant, increased CO<sub>2</sub> production from 0.78 Mt in 2005 to 1.09 Mt in 2007 due to drastic growth of the building industry.

Table 1. CO<sub>2</sub> sources registered in the EU Emissions Trading Scheme (ETS) in 2005 and 2007

	Big sources (>100 000 t CO <sub>2</sub> )			All sources registered in ETS		
	Million tonnes	Number of sources	Share in all ETS emissions, %	Million tonnes	Number of sources	ETS share in total GHG emissions
	2005/2007			2005/2007		2005
Estonia	11.5/14.5	9/9	91.3/94.6	12.6/15.3	41/47	59.3
Latvia	1.9/1.9	6/5	63.8/65.7	2.98/2.89	89/89	26.7
Lithuania	5.6/4.8	9/8	84.8/80	6.6/6	89/93	32.5

### 3. Prospective CO<sub>2</sub> storage options

The Baltic countries are situated in the eastern part of the Baltic sedimentary basin that lies in the western periphery of the East European Craton. Upper Vendian and all Phanerozoic systems are represented in the basin. The thickness of the sediments is less than 100 m in northern Estonia, increasing to 1900 m in south-western Latvia and 2300 m in western Lithuania [16].

The Baltic basin contains several major aquifers that are viewed as prospective media for the storage of CO<sub>2</sub>. However, a prospective aquifer should meet certain requirements, among which the most important are the large volume of the reservoir, adequate depth and temperature, and the presence of a reliable seal (including structural tightness). Deep saline aquifers are by far the most suitable for large-scale CO<sub>2</sub> storage. Depending on the formation pressure and temperature, CO<sub>2</sub> can be stored either as compressed gas or in a supercritical state ( $P > 73.8$  bars,  $T > 31^\circ\text{C}$ ). No coal neither salt deposits are present in the sedimentary cover.

The Baltic basin contains also a number of oil fields related to Cambrian siliciclastic and Ordovician and Silurian carbonaceous reservoirs. Therefore the storage of CO<sub>2</sub> in depleted oil fields and the enhanced oil recovery (EOR) option are considered as the potential technology in the Baltic area. Oil fields are exploited in western Lithuania, Kaliningrad District and offshore Poland. In Lithuania, oil fields are confined to two major tectonic zones, the Telšiai fault zone and the Gargždai fault zone. Some oil shows (and a small Kuldiga oil field) were discovered in Cambrian and Ordovician reservoirs in Latvia [17]. Silurian reefs in Central Lithuania, contain small oil fields which are not exploited. In Lithuania, 10 oil fields are presently exploited with the production capacity from 16 000 to 1 400 000 t. The storage potential of the largest oil fields of western Lithuania does not exceed 2 Mt of CO<sub>2</sub>, the total estimated potential is as much as 7.6 Mt of CO<sub>2</sub>, which is just a little more than the annual stationary CO<sub>2</sub> emissions in the country. Another option is the utilization of carbon dioxide for enhancement of the oil recovery. Most of the oil fields have reached the tail phase and EOR can prolong the lifetime of oil fields. The oil is light and exceeds 35 API° [18], meaning that CO<sub>2</sub> could be injected in miscible conditions – a favourable factor for CO<sub>2</sub> sequestration. The estimated total EOR net volume of CO<sub>2</sub> is 5.6 Mt [2].

### 4. Deep saline aquifers

Only two large aquifers of the Baltic sedimentary basin, the Lower–Middle Devonian (Pärnu–Kemerī formations) and Middle Cambrian aquifers are buried to depths exceeding 800 m (Figs. 3–5). The Cambrian reservoir is distributed in all Baltic countries. Its depth varies from outcrops in Estonia to more than 2 km in west

Lithuania (Figs. 2, 3). The depth of the reservoir exceeds 800 m in western Latvia, western Lithuania, northern Poland, and in the Baltic offshore, but beyond the limit of the supercritical state of CO<sub>2</sub> in Estonia. The reservoir is composed of quartz sandstones with subordinate siltstones and shales. The thickness of the aquifer is in the range of 20–70 m. Due to considerable variations in depth and temperature, the porosity of sandstones changes dramatically across the basin, from 20–30% in the northern and eastern shallow parts of the basin to less than 5% in the central and western parts of the basin [19]. The Middle Cambrian aquifer is sealed by a 500–900 m thick Ordovician–Silurian shale cap rock.

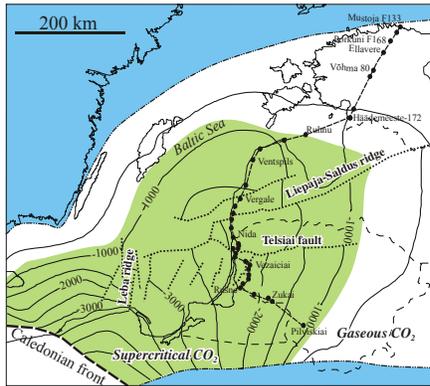


Figure 2. Depths of the top of the Cambrian aquifer. The contour lines indicate the depth of the top of the Cambrian. The hatched lines show major faults. The P–T fields of gaseous (white) and supercritical (green) state of CO<sub>2</sub> are indicated. The line of the geological cross-section shown in Fig. 3 is indicated.

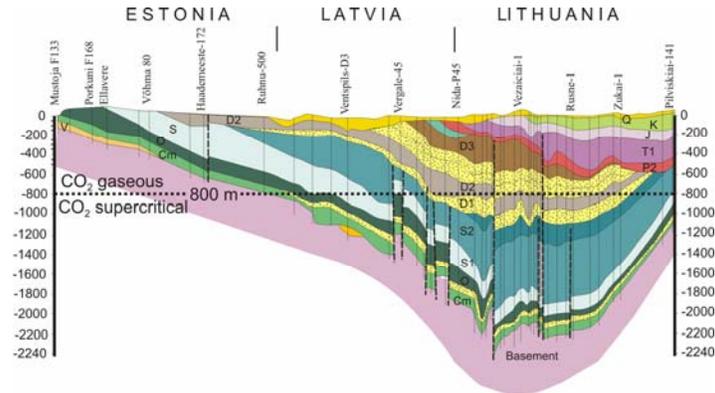


Figure 3. Geological cross-section across Estonia, Latvia and Lithuania. Major aquifers are indicated in yellow and by dots. V – Vendian (Ediacaran), Cm – Cambrian, O – Ordovician, S – Silurian, D1, D2 and D3 – Lower, Middle and Upper Devonian, P2 – Middle Permian, T1 – Lower Triassic, J – Jurassic, K – Cretaceous, Q – Quaternary.

The depth of the Pärnu–Kemeri aquifer, distributed in the central part of the basin, exceeds 800 m only in western Lithuania and the south–eastern part of the Baltic Sea. The aquifer is composed of arkosic sandstones containing 20–30% siltstone and shale layers [20]. The average porosity of sandstones is 26%; permeability is in the range of 0.5–4 Darcy. The thickness of the aquifer is in the range of 100–160 m in western Lithuania. The aquifer is covered by 80–120 m thick marlstones of the Narva Formation, representing a basin-scale aquitard.

Upon injection into saline aquifers, carbon dioxide may be stored by (1) hydrodynamic (structural) trapping, (2) solubility trapping (carbon dioxide dissolved in aquifer water), (3) residual trapping, and (4) mineral trapping. Solubility and mineral trapping are the most important long-term solutions to carbon dioxide sequestration in geological media. However, these processes involve a larger proportion of injected CO<sub>2</sub> only 100 years after the injection, whereas hydrodynamic trapping becomes effective immediately and can be compared to existing natural analogues.

#### 4.1. Structural trapping potential

Fifteen major structures, with estimated storage capacity exceeding 10 Mt CO<sub>2</sub>, have been identified in western Latvia (Fig. 1), [21], while only small-scale uplifts are known in Lithuania [20]. The storage capacity of a structural trap was estimated, according to the formula

$$M_{CO_2} = A \times h \times \varphi \times \rho_{CO_2r} \times S,$$

where  $M_{CO_2}$  is the storage capacity (kg),  $A$  is the area of a closure (m<sup>2</sup>),  $h$  is the net thickness of reservoir sandstones (m) (typically is 20–40 m in Latvia and Lithuania),  $\varphi$  is the porosity (typically from 0.25–0.20 in central Latvia and central Lithuania to 0.06 in western Lithuania),  $\rho_{CO_2r}$  is the in situ CO<sub>2</sub> density in reservoir conditions

(ranges from 600 kg/m<sup>3</sup> in western Lithuania to 750 kg/m<sup>3</sup> in central Lithuania and central Latvia),  $S$  is the sweeping efficiency, often also referred to as the storage efficiency (assumed 0.35).

The total capacity of large structures of Latvia is estimated to exceed 300 Mt of CO<sub>2</sub>, with the potential of the greatest uplifts of 40–80 Mt and more of CO<sub>2</sub>. The depths range from 650 to 1200 m. The thickness of Cambrian sandstone reservoir rock is of 40–60 m, with average porosity of 22% and permeability of 3–7 Darcy. The structures are similar to the Inčukalns underground gas storage (UGS) operating in Latvia since 1968. The potential of the gas storage is estimated to be 5.7 billion m<sup>3</sup>, of which 4.46 billion m<sup>3</sup> is filled now by natural gas [21]. The major CO<sub>2</sub> emitting sources are located close to major uplifts in Latvia. Furthermore, the CO<sub>2</sub> sources and potential traps are located close to the existing gas supply pipelines, which potentially reduce the cost of CO<sub>2</sub> transportation.

The capacities of more than 100 Cambrian local uplifts identified in Lithuania were evaluated recently [20]. The two largest Vaskai and Syderiai aquifer structures can store only 3.5 and 5.4 Mt of CO<sub>2</sub> respectively, while the rest structures are of much lesser volume. Therefore, the hydrodynamic trapping in Cambrian aquifer structures has no prospects in Lithuania. No structural traps have been identified in the Pärnu–Kemeru aquifer, neither in Lithuania nor in Latvia due to low-intensity tectonic deformation of the Variscan structural complex [22].

#### 4.2. Solubility trapping

The solubility trapping is not restricted to a particular structure. The solubility of CO<sub>2</sub> ranges from 2% to 6%, depending on the brine salinity, temperature and pressure [23]. A large volume of a regional-scale aquifer provides an attractive alternative for CO<sub>2</sub> disposal. The solution time is of order of 10<sup>2</sup>–10<sup>3</sup> years, which is considerably longer than the hydrodynamic trapping process [24]. Before dissolving, the CO<sub>2</sub> phase migrates towards the basin margins, which may cause the risk of gas escape either through the faults or shallow margins of the basin. Therefore, the safe distance of gas migration should be evaluated before selecting prospective sites for CO<sub>2</sub> injection. The solubility trapping potential has been calculated using the approach presented in [23]. It accounts for the brine salinity, temperature, pressure and reservoir properties that vary considerably across the Baltic basin. The solubility of CO<sub>2</sub> in Cambrian aquifer varies from 25–30 kg/m<sup>3</sup> in western Lithuania to 40–50 kg/m<sup>3</sup> in eastern Lithuania and Latvia (Table 2). The CO<sub>2</sub> storage potential changes westwards from 0.4 Mt/km<sup>2</sup> to 0.05 Mt/km<sup>2</sup>. The calculated total solubility trapping capacity is about 11 Gt of CO<sub>2</sub> within the area of the supercritical state of carbon dioxide. The Pärnu–Kemeru aquifer is characterized by better reservoir properties, but has a smaller area than the Middle Cambrian reservoir. CO<sub>2</sub> solubility ranges from 36 kg/m<sup>3</sup> in the deep part of the basin to 60 kg/m<sup>3</sup> in shallow periphery. In western Lithuania the storage capacity of the reservoir is about 1 Mt of CO<sub>2</sub> in 1 km<sup>2</sup> area. The estimated total onshore potential of this formation is as high as 1 Gt of CO<sub>2</sub>.

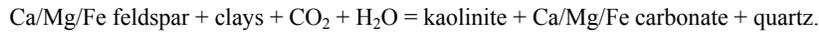
Table 2. Solubility trapping in the Cambrian reservoir in western, central and eastern Lithuania

Parameters	Western Lithuania	Central Lithuania	Eastern Lithuania
Aquifer temperature, °C	75	55	40
Aquifer Pressure, MPa	20	15	10
Salinity, g/l	160	110	90
CO <sub>2</sub> solubility, kg/m <sup>3</sup>	28.1	35.6	40.9
Effective thickness, m	20	35	40
Porosity, %	10	20	25
<b>Solubility storage in 1 km<sup>2</sup> area, Mt of CO<sub>2</sub></b>	<b>0.056</b>	<b>0.249</b>	<b>0.409</b>

## 5. Mineral trapping

### 5.1. Mineral trapping in aquifers

The mineral trapping that involves a series of interactions between the formation mineralogy and CO<sub>2</sub>-enriched aquifer waters can convert CO<sub>2</sub> to carbonate, an immobile and harmless mineral that will be stored for millions to hundreds of millions of years. Reactions with Ca/Mg/Fe-bearing silicate minerals are the most promising for carbon sequestration because these silicates neutralize the added acidic CO<sub>2</sub> and provide alkali metals that trap CO<sub>2</sub> through the precipitation of carbonate [24]. These reactions can be summarized as follows [25]:



The Middle Cambrian reservoir comprises quartz sandstones that are practically not reactive to carbon dioxide. The Pärnu–Kemeru sandstones contain clay admixture (up to 10%) and feldspar grains (up to 15%). Therefore they have a potential for permanent immobilization of carbon dioxide in mineral form. Assuming the rock capacity of 10 kg/m<sup>3</sup> [23], the sequestration potential can be evaluated to reach 5.6 Gt of CO<sub>2</sub> (onshore).

### 5.2. Mineral trapping by ultramafic rocks

Ultramafic rocks have been identified as the most suitable rock media for mineral trapping of carbon dioxide. However, they are rather rare and can provide only limited reserves for CO<sub>2</sub> sequestration. Alternatively, the serpentinite rocks are abundant in the world and can serve for immobilization of CO<sub>2</sub>. A large serpentinite province has been discovered in the Palaeoproterozoic crystalline basement of southern Lithuania. Serpentinites associate with the high-quality iron ore, which provides an opportunity for cascade utilization of these formations (Fig. 4). More than a dozen serpentinite bodies have been identified. Owing to their association with iron ore deposits, these bodies were extensively studied by drilling. They subcrop at the top of the basement and are covered by 280–500 m thick platform sediments. Serpentinites are located close to the south-eastern cluster of emission sources, with the distance to the particular sources varying from 50 to 150 km.

Carbon dioxide can be immobilized by reacting with serpentinite to form stable minerals. Roughly, the assumed ratio of immobilized CO<sub>2</sub> to serpentinite is assumed 1:2. The estimated volume of serpentinites of the largest Varena Iron Ore Deposit is 1–2 Gt. Consequently, the sequestration potential is evaluated to be as high as 0.5–1 Gt [26]. It equals to CO<sub>2</sub> production during 200–500 years in the southeastern CO<sub>2</sub> emission cluster.

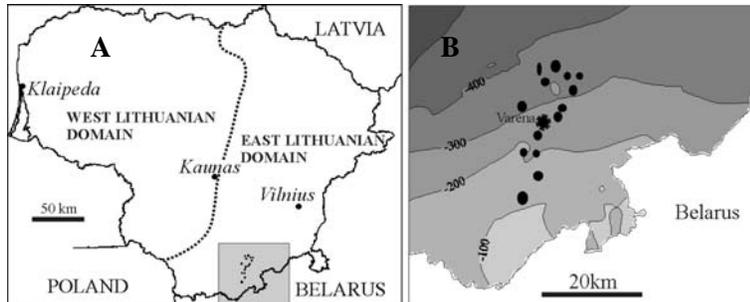


Fig. 4 A. Major lithotectonic domains of the crystalline basement of Lithuania. The Varena Geological Province is shaded. B. The Varena Geological Province. The Varena Iron Ore deposit is distinguished by black dots. The depths of the top of the crystalline basement are indicated.

### 5.3. Mineral trapping by oil shale ash

The technology of CO<sub>2</sub> mineral trapping with waste oil shale ash is under development in Estonia. The concept for abatement of CO<sub>2</sub> emissions in power production, based on oil shale ash as sorbent for CO<sub>2</sub> mineralization, has been proposed and elaborated (Fig. 5), [6, 7]. Estonian oil shale is a carbonaceous fine-grained sedimentary rock of Ordovician age containing 10–60% kerogen (solid organic matter), 20–70% carbonates represented by limestone, or more rarely by dolomite, and 15–60% siliciclastic minerals. During combustion of one tonne of oil shale 450–550 kg of ash is produced (in case of mineral coal only 100 kg of ash is produced). About 77% of the mined oil shale with a lower calorific value is used as boiler fuel in large power plants. During combustion of oil shale CO<sub>2</sub> is formed not only as a burning product of organic carbon, but also as a decomposition product of the carbonate part of ash. Therefore the total content of carbon dioxide increases up to 25% in flue gases of oil shale [13]. Oil shale ash contains up to 20–25% free Ca–Mg oxides. Portlandite Ca(OH)<sub>2</sub>, forming from free lime during hydraulic transportation and wet deposition of ash, can bind CO<sub>2</sub> also from air. This natural weathering process could be

accelerated by simple methods. Batch and continuous mode experiments have demonstrated that by processing the ash–water suspension by flue gases, the CO<sub>2</sub> binding ability of ash could be utilized completely. The results of these experiments show that watered oil shale ash can bind 80–160 kg and more of CO<sub>2</sub> per one tonne of ash, and 30–80 kg of CO<sub>2</sub> can be bound by alkaline wastewater used for transportation of one tonne of ash [6, 7]. From the annual production of about 16.3 million tonnes of oil shale in Estonia in 2007, 14.3 million tonnes (88%) was combusted for energy production. As 450–550 kg of ash is produced from one tonne of combusted oil shale, about 7 million tonnes of ash was produced in 2007. The amount of CO<sub>2</sub> bound by oil shale ash in the wet mineralization process from flue gas can reach 560–1120 thousand tonnes and that bound by the alkaline wastewater neutralization process in reactor can reach 210–560 thousand tonnes. The maximum amount of CO<sub>2</sub> bound from flue gas can be estimated as 770–1680 thousand tonnes. It is about 10–12% of CO<sub>2</sub> emissions produced by power plants in 2007. Carbonates that formed as result of the binding process could be separated and used as independent by-product, but it would be more useful to store them in closed oil-shale mines. The latter solution will permit filling underground mining cavities and prevent environmental problems arising from ash heaps.

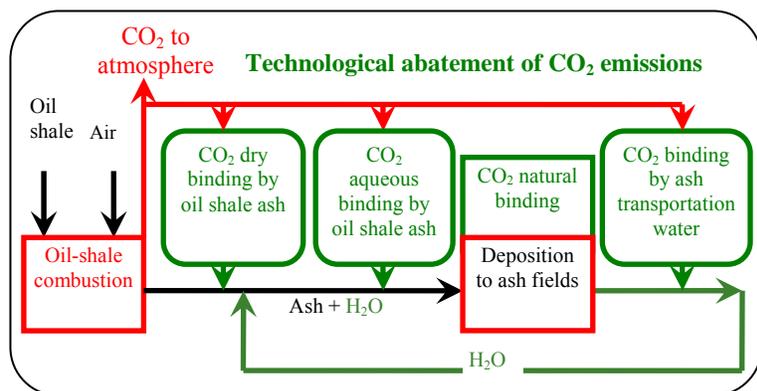


Figure 5. Concept for CO<sub>2</sub> binding in oil shale–based power production.

## 6. Conclusions

CO<sub>2</sub> sources are distributed unevenly in the Baltic countries and the types of CO<sub>2</sub> sources vary considerably. Major emissions are concentrated in the coastal area of the Gulf of Finland in Estonia. Due to the utilization of oil shale for energy production, CO<sub>2</sub> emissions produced by the two largest Estonian power plants (12.1 Mt in 2007) exceed the volume of CO<sub>2</sub> produced by all Lithuanian (6 Mt) and Latvian (2.89 Mt) stationary sources.

The Baltic countries are situated within the Baltic sedimentary basin. Two prospective Lower–Middle Devonian and Middle Cambrian aquifers meet the basic requirements for CO<sub>2</sub> storage media. However, only Latvia has structural traps large enough to store the industrial CO<sub>2</sub> emissions. The capacity of 15 large uplifts exceeding 300 Mt is sufficient to hold Latvian CO<sub>2</sub> stationary emissions during 150 years of production.

For Lithuania and Estonia, the transportation of CO<sub>2</sub> to the Latvian storage site is a potential option, which, however, can meet serious political and public problems. Alternative approaches should therefore be considered. The in–situ solubility and mineral trapping in deep saline aquifers are long-term technologies still to be developed. The mineral trapping of CO<sub>2</sub> from flue gas with alkaline ash produced by oil shale combustion and ash transportation water can bind up to 10–12% of large industrial CO<sub>2</sub> emissions in Estonia. Application of this technology can solve several environmental problems in Estonia: decreasing CO<sub>2</sub> emissions and ash residues, filling of the closed oil-shale mines with produced minerals and reducing the risk of surface collapses, saving natural landscape, etc. In Lithuania, the prospective formation for the mineral trapping is represented by serpentinites located in the crystalline basement at shallow depths. Still, the technologies are immature and need further development.

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