## INDUSTRIAL CARBON DIOXIDE EMISSIONS AND POTENTIAL GEOLOGICAL SINKS IN THE BALTIC STATES

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Industrial CO<sub>2</sub> emissions and geological storage opportunities in Estonia, Latvia and Lithuania are studied within the framework of EU GEO-CAPACITY and CO2NET EAST projects supported by European Commission through Framework Programme 6. The structure of the energy sector and socio-economic conditions vary considerably between these three countries. A total of 24 large (emitting more than 0.1 million tonnes (Mt) of  $CO_2$ ) industrial sources of  $CO_2$  emissions, registered in 2005 in the European Trading Scheme, consists of 11.5 Mt of CO<sub>2</sub> from Estonia, 1.9 Mt from Latvia and 5.6 Mt from Lithuania. The highest amount of  $CO_2$  emission in Estonia is related to the oil shale used as the main fuel for power generation; the two largest Estonian power plants - Estonian and Baltic - produced respectively 7.7 and 2.25 Mt of  $CO_2$  in 2005 and 9.4 and 2.7 Mt of  $CO_2$  in 2007.  $CO_2$ emission from oil shale combustion is significantly higher in comparison with other fossil fuels as energy sources. This is why  $CO_2$  emission per capita in *Estonia is about two times higher than the average value in Europe.* The three Baltic States are located within the Baltic sedimentary basin, the

thickness of which varies from 100 m in Northeast Estonia up to 1900 m in Southwest Latvia and 2300 m in West Lithuania. The most prospective formation for geological storage of  $CO_2$  is the Cambrian reservoir. 15 large structures have been identified in Latvia with a total capacity exceeding 300 Mt of  $CO_2$ . The tightness of structures is evidenced by 40 years of successful operation of the Inčukalns Underground Gas Storage. Due to shallow setting, geological conditions in Estonia are unfavourable for  $CO_2$ storage. Therefore an option of transporting  $CO_2$  from Estonia via pipelines

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to one of the Latvian storage structures could be considered. Alternatively, the technology of  $CO_2$  trapping by sorption of oil shale ash is under development in Estonia. In Lithuania, the capacity of  $CO_2$  storage in Cambrian and Devonian structures as well as in oil fields is negligible, but  $CO_2$  solubility and mineral trapping is a long-term option.

## Inroduction

Most of the energy used to meet human needs is derived from combustion of fossil fuels (natural gas, oil, coal, oil shale, etc.), which release carbon dioxide into the atmosphere. Combustion of oil shale, the main energy resource of Estonia, produces significantly higher  $CO_2$  emissions per energy produced than other energy sources. Globally about 23.5 Gt of  $CO_2$  was emitted from fossil fuel use in 2000 [1]. The atmospheric concentration of  $CO_2$ , a greenhouse gas, is increasing, causing trapping of solar heat and subsequent global warming. As a result of human activities, the  $CO_2$  concentration in the atmosphere has risen from pre-industrial 280 ppmv to 380 ppmv and might reach 1100 ppmv by 2100 [2]. Global warming studies predict that climate changes, resulting from increase in atmospheric concentration of  $CO_2$ , will adversely affect life on the Earth [3].

Carbon management consists of a broad portfolio of strategies to reduce carbon emissions via carbon capture and storage, enhanced efficiency of power generation and use, application of low-carbon fuels and the employment of renewable energy sources [4]. Carbon dioxide is already being captured in oil and gas and chemical industries. Once CO<sub>2</sub> has been captured, it would need to be stored securely for hundreds and thousands of years. CO<sub>2</sub> has been used for enhanced oil recovery (EOR) purposes since the 1950s [5]. Research concerning sequestration of CO<sub>2</sub> for environmental purposes began only 10-15 years ago. CO<sub>2</sub> sequestration in geological media can be safely undertaken within national boundaries in most countries, thus avoiding international political issues. Geological sinks for CO<sub>2</sub> include depleted oil and gas reservoirs, enhanced oil recovery, unminable coal seams, and deep saline porous formations. Together, these can hold hundreds to thousands of gigatonnes of carbon (GtC), and the technology to inject  $CO_2$  into the ground is well established. CO<sub>2</sub> is stored in geological formations by a number of different trapping mechanisms, with the particular mechanism depending on the formation type [4, 6-15].

According to the Kyoto protocol signed by the Baltic countries in 2002, the level of air-polluting greenhouse gases emissions should be reduced by 8% during the commitment period 2008–2012 compared to the 1990 level. Compared to 1990, the greenhouse gas (GHG) emissions decreased in Baltic countries for more than 50% [16–18], Table 1a. However, the changing energy market (e.g. closure of the Ignalina NPP) 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. Furthermore, the inventory of

geological storage of  $CO_2$  in the Baltic countries became an integrated part of the European-scale projects, like EU GEOCAPACITY and CO2NET EAST.

	Total GHG emissions			CO <sub>2</sub> emissions per head	
	In CO <sub>2</sub> equivalents, million tonnes		Reduction compared to 1990, %	Tonnes CO <sub>2</sub> /capita	Place in world rate
Year	1990	2005		2004	
Estonia Latvia Lithuania	42.6 26.4 48	20.9 10.9 22.6	50.9 58.9 53	14.1 3.87 3.07	16 90 100

Table 1a. Total greenhouse gas (GHG) emissions and CO<sub>2</sub> emissions per capita

The geological setting of the Baltic States is rather different from that of the other European countries that comprise a number of small sedimentary basins, while Lithuania, Latvia and Estonia are situated within one common Baltic sedimentary basin [19]. Therefore, a joint study is required for the assessment of geological sinks. The source types and emissions differ considerably in the Baltic countries, depending on the socio-economic conditions. The main energy and  $CO_2$  in Estonia comes from oil shale combustion, while CO<sub>2</sub> emissions in Lithuania and Latvia are significantly lower due to the utilisation of other main energy sources (nuclear and hydro-energy). Geological conditions are also different, as these countries represent different parts of the Baltic basin. The composition and properties of reservoir rocks of the Baltic Cambrian basin were studied some years ago in the framework of the Baltic-German project GEOBALTICA [20-24]. The estimation of carbon capture and storage capacity was recently started in Lithuania [25]. The capacity of Latvian structures for the purpose of underground gas storage was assessed in 2007 by Latvian Environment, Geology and Meteorology Agency (LEGMA) [26]. In 2006 all three Baltic countries started inventory of their CO<sub>2</sub> industrial sources and geological capacity in the framework of EU GEOCAPACITY project supported by EU Framework Programme 6 [27].

#### Distribution and types of stationary CO<sub>2</sub> sources

In 1990 (base year of the Kyoto Protocol) the Baltic countries produced 48 Mt of GHG emissions in  $CO_2$  equivalents in Lithuania, 42.6 Mt in Estonia and 26.4 Mt in Latvia. The emissions have reduced considerably since this basic year, due to large-scale socio-economic rearrangement of the Baltic countries. In 2005 these emissions were reduced for 53% in Lithuania, 51% in Estonia and 59% in Latvia compared to the basic year (Table 1a). However, GHG emissions have increased systematically since 1999–2000 owing to

economical growth [16–18]. A significant increase in emissions is forecasted in Lithuania due to the planned closure of the Ignalina NPP in 2009.

The largest GHG emissions in the Baltic countries are produced by the energy sector (Table 1b), while contribution from other sectors is much less significant [16–18].  $CO_2$  sources exceeding 100 000 tonnes/year are only considered economically feasible for geological sequestration. Twenty four large sources (Fig. 1) produced 11.5 Mt of  $CO_2$  in Estonia, 5.6 Mt in Lithuania and 1.9 Mt in Latvia. The stationary sources included into the European Union Emission Trading Scheme (EU ETS) produced 12.7 Mt of  $CO_2$  in Estonia (41 sources), 6.6 Mt in Lithuania (89 sources) and 2.98 Mt in Latvia (89 sources). In Estonia,  $CO_2$  emission per capita amounting 14.1 tonnes is one of the highest in Europe and in the world, while reaching 3.87 and 3.07 tonnes in Lithuania and Latvia, respectively (Table 1a). For the sake of comparison it should be noted that in 2004 average  $CO_2$  emissions per capita was 7.7 tonnes in Europe.

Table 1b. Share of sectors (%) in greenhouse gas (GHG) emissions in countries

Emissions	Estonia	Latvia	Lithuania
Energy	89	72	58
(fuel combustion and emissions from fuels in			
all sectors, including transport )			
Fuel combustion in transport	10	27.5	18.2
Agriculture	5.7	17.7	17.9
Industrial processes	2.7	2.5	16.6
Waste	2.5	7	6.8

*Table 1c.* CO<sub>2</sub> emissions registered in European Union Emission Trading System (EU ETS)

Big CO <sub>2</sub> sources(>100 thousand tonnes) registered in EU ETS						
	Million tonnes	Number of sources	Share in ETS emissions, %	Million tonnes	Share in ETS emissions, %	
Year	2005			2007		
Estonia Latvia Lithuania	11.5 1.9 5.6	9 6 9	91.3 63.8 84.8	14.5 1.9 4.8	94.6 65.7 80.0	
All registered in EU ETS industrial sources						
	CO <sub>2</sub> emissions, million tonnes	Number of sources of verified emissions	EU ETS share in total GHG emissions	CO <sub>2</sub> emissions, million tonnes	Number of sources of verified emissions	
Year	2005			2007		
Estonia Latvia Lithuania	12.6 2.98 6.6	41 89 89	59.3 26.7 32.5	15.3 2.89 6	47 89 93	



*Fig. 1.* Total industrial CO<sub>2</sub> emissions (produced in 2005) in Estonia, Latvia and Lithuania registered by European Union Emissions Trading scheme are shown by circles. The biggest circles show CO<sub>2</sub> emissions > 100,000 tonnes per year, the middle circles show emissions from 10,000 to 100,000 tonnes per year, little circles show sources < 10,000 tonnes per year. Solid lines show natural gas pipeline network connecting Baltic States with Russia. Little open circles show pipelines terminals. Filled rectangle shows Incukalns underground gas storage (UGS).

The high GHG emission rate in Estonia results basically from the application of oil shale for power production. Main CO<sub>2</sub> sources are located in northeast of the country, close to the oil-shale deposits. The largest CO<sub>2</sub> sources in the Baltic countries are *Estonian* and *Baltic* Power Plants, producing respectively 7.7 Mt and 2.25 Mt of CO<sub>2</sub> in 2005, and respectively 9.4 and 2.7 Mt of CO<sub>2</sub> in 2007. The Kunda Nordic cement plant produced 0.746 Mt of CO<sub>2</sub> in 2005 and 1.17 Mt of CO<sub>2</sub> in 2007 (Fig. 1) Another concentration of CO<sub>2</sub> sources has been determined in the Tallinn region. In Latvia, the main  $CO_2$  producers are located in the western part of the country. The Liepaja metallurgical enterprise emits 0.366 Mt of CO<sub>2</sub>. Three electric power stations in the Riga area emit respectively 0.62, 0.38 and 0.14 Mt of CO<sub>2</sub>. There are two CO<sub>2</sub> source clusters in Lithuania, situated in the northwest and southeast of the country. The greatest GHG producer is the Mažeikiai oil refinery, emitting 1.87 Mt of CO<sub>2</sub> per year. The other two largest sources are Akmene cement plant and Vilnius power station producing 0.78 and 0.70 Mt of CO<sub>2</sub>, respectively.

#### The geological setting and stratigraphy of the Baltic basin

The Baltic countries are located in the eastern part of the Baltic sedimentary basin that overlies the western periphery of the East European Craton (Fig. 2). Baltic basin contains the Upper Vendian at the base and all of the Phanerozoic systems as a result of protracted subsidence history. The basin is only weakly tectonized; the sedimentary layers are generally inclined to the southwest. The thickness of the sediments is less than 100 m in Northern Estonia, increasing to 1900 m in Southwestern Latvia and 2300 m in West Lithuania [19].

The oldest sediments are represented by up to 200 m thick Ediacaran (Vendian) siliciclastics aguifer and up to 120 m thick lowermost Cambrian Blue Clays that are distributed in the eastern half of the Baltic countries. The rest of Cambrian succession is composed of triple alternation of quartz sandstones, siltstones, and shales, occurring in different proportions across the basin. The thickness of the Cambrian aquifer is up to 170 m in West Lithuania. The Cambrian is overlain by a 40-250 m thick Ordovician shalycarbonaceous aquitard, except East Lithuania and Estonia dominated by limestones and dolostones. This shaly-carbonaceous succession grades upwards into up to 800 m thick package of shales of Silurian age, while carbonates predominate in the shallow periphery of the basin. Together with the Ordovician it composes the major basin-scale aquitard. Devonian sediments cover the whole territory of Latvia, Southern and Eastern Estonia, and most of Lithuania (Figs. 2, 4). The composition of Devonian sediments is variable in the section, the marly/carbonaceous packages alternating with sandy packages. Maximum thickness of 1100 m has been reported from West Lithuania. Some major aquifers are defined in the succession, i.e. Lowerlower Middle Devonian, Middle-lower Upper Devonian, and some smaller



aquifers, such as Stipinai, Žagarė, etc. They are distributed in most of Lithuanian and Latvian territories.

The Cambrian-lowermost Devonian succession is referred to as the Caledonian structural complex, whereas the overlying Devonian-lowermost Carboniferous succession is attributed to the less deformed Variscan structural complex. The end of the Caledonian tectonic stage was marked by the extensive faulting of the basin (Fig. 3).

The Permian, Mesozoic, and Cenozoic sediments are attributed to the Alpine structural complex. Their thickness and depths increase to southwest, reaching 600 m in Southwest Lithuania. Those sediments are absent in Estonia and most of Latvia, while covering the southwestern part of Lithuania. Upper Permian deposits consist of a 100 m thick sequence of carbonates and evaporates. The Naujoji Akmenė formation comprises an important aquifer for



*Fig.* 3. Depths of top of Cambrian aquifer. The contour lines indicate the depth of the top of Cambrian. The hatched lines show major faults. The P-T fields of gaseous (white) and supercritical (dotted) state of  $CO_2$  are indicated. The line of the geological cross-section shown in Fig. 4 is indicated.





the water supply of north Lithuania. The Permian is overlain by up to 250 m thick Lower Triassic red-bed mudstones representing the regional-scale aquitard. The shales grade into up to 120 m thick Jurassic sandstones, clays, and limestones. The Cretaceous is composed of glauconite sand in the lower part and chalk and chalky marl in the upper part of the section of 140 m thick. In some areas of Lithuania it is used for the water supply. Cenozoic terrigenous aquifer reaching 80 m in thickness is known only in the southwestern part of Lithuania. The sedimentary cover is topped by the Quaternary glacial sediments varying in thickness from centimetres to a few hundred meters.

## **Prospective formations for CO<sub>2</sub> storage**

#### Physical properties of the prospective geological formations

The technology for geological  $CO_2$  sequestration in sedimentary basins has already been developed by the mining and petroleum industry. Still, there are several uncertainties concerning the volumes available for sequestration, safety, liability, and the cost associated with the  $CO_2$  transport and injection [6].  $CO_2$  can be sequestered in geological media by:

- methane displacement in coal beds [14, 15],
- storage in salt caverns [7],
- storage in depleted hydrocarbon reservoirs [8], in particular when applying the enhanced oil recovery techniques [9],
- storage in deep saline aquifers through hydrodynamic trapping and mineral immobilization [10].

The Baltic basin contains a number of different formations. However, the prospective media should meet certain requirements, among which the most important are the large volume of the reservoir, suitable depth and temperature, and presence of a reliable seal (including structural tightness).

## Coal and salt

No coal seams exist in the Baltic area, but thin lignite layers have been identified in Jurassic succession of Lithuania. Salt has accumulated in the Zechstein lagoon in the Kaliningrad district, while only one small salt pillow is found in Southwestern Lithuania. Consequently, these types of formations are not prospective in the Baltic region.

## Deep saline aquifers

Deep saline aquifers are by far the most popular proposal for large-scale  $CO_2$  storage. These are water-saturated porous layers in the subsurface of sandstone or limestone, at present not used for any other purpose. The high water salinity renders these layers unsuitable for use as drinking water or for watering plants. 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 °C). Carbon dioxide, injected in a supercritical state, has a much lower density and viscosity than the liquid brine it displaces. In situ, the supercritical CO<sub>2</sub> partitions between an immiscible gas-like phase and dissolution in the aqueous phase, according to an extended version of Henry's Law, yielding a multi-phase, multi-component system. At depths greater than 800 m the carbon dioxide will be in a supercritical state, which enables an efficient injection method in both pipeline engineering and in filling deep pore space. It is important to realize that in the deep subsurface there is no vacant space – all pores within sandstones and limestones are filled with fluid (usually pore water). Therefore, the thermobaric conditions P = 73.8 bars, T = 31 °C are considered the lower limit for the geological storage of CO<sub>2</sub>.

 $CO_2$  can be stored in the hydrodynamic traps (structural, stratigraphic). Some of the injected  $CO_2$  will dissolve in water or will be trapped by matrix particles. The capability of an aquifer to transmit and store  $CO_2$  is controlled by the depositional environment, structure, stratigraphy and pressure/temperature conditions. Critical factors are:

1) the regional water flow system,

2) the thickness, lateral extent and continuity of the aquifer,

3) the tightness of the seal above the aquifer, including the faults,

4) the capability of overburden layers above the reservoir seal to delay or diffuse leakage.

#### Depleted oil reservoirs and enhanced oil recovery

Depleted oil reservoirs are attractive as  $CO_2$  storage locations because they are known to have trapped and stored hydrocarbon fluids for many millions of years. The key advantage of depleted oil fields is that site-specific data for evaluating reservoirs and capability of top seals were already collected during petroleum exploration and production.  $CO_2$  has been used for enhanced oil recovery (EOR) purposes since the 1950s. Use of  $CO_2$  in EOR operations actually represents a form of both utilization and sequestration.

The Baltic basin contains a number of oil fields related to Cambrian siliciclastic and Ordovician and Silurian carbonaceous reservoirs. Therefore the storage of  $CO_2$  in the depleted oil fields and the EOR option are considered the potential technology in the Baltic area.

#### Prospective aquifers in the Baltic region

A number of aquifers have been identified in the Baltic sedimentary basin. Deep saline aquifers, not suitable for the drinking water supply, are considered prospective ones for  $CO_2$  storage.

Only two large aquifers of the Baltic States meet requirements listed above, i.e. the Lower-Middle Devonian (Pärnu-Kemeri formations) and Middle Cambrian aquifers buried to depths exceeding 800 m in the central and western parts of the Baltic basin (Fig. 3). The Cambrian reservoir is distributed in all Baltic countries. Its depth varies from outcrops in Estonia to more than 2 km in West Lithuania (Fig. 3). The depth of the reservoir exceeds 800 m in West Latvia, West Lithuania, North Poland, and in the Baltic offshore, while Estonia is beyond the limit of the supercritical state of CO<sub>2</sub>. 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 [19]. Due to considerable variations in depth and temperature, the porosity of sandstones changes drastically across the basin, from 20–30% in the northern and eastern shallow part of the basin to less than 5% in the central and western parts of the basin [24]. The Middle Cambrian aquifer is sealed by a thick (500–900 m) shaly package of Ordovician-Silurian age representing a reliable seal rock.

The Pärnu-Kemeri aquifer is distributed in the central part of the basin (Fig. 5). Its depth exceeds 800 m only in West Lithuania and the southeastern part of the Baltic offshore where it reaches 1100 m. The aquifer is composed of arkosic sandstones containing siltstone and shaly layers. The net-to-gross ratio is of order of 0.7–0.8 [25]. Average porosity of sandstones is 26%; permeability is in the range of 0.5–2 D. Total thickness of the aquifer varies from 100 to 160 m in West Lithuania. The aquifer is covered by 80–120 m thick marlstones attributed to the Narva Formation constituting a basin-scale aquitard.



*Fig. 5.* Depths of top of Pärnu-Kemeri aquifer. The contour lines indicate the depth of the top of Devonian. The hatched lines show major faults. The P-T fields of gaseous (white) and supercritical (dotted) state of  $CO_2$  are indicated.

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 larger proportion of injected  $CO_2$  only 100 years after the injection, whereas hydrodynamic trapping becomes effective immediately and can be compared to existing natural analogues.

#### Structural trapping

Structuring of the Cambrian reservoir varies across the basin. The most intense faulting and formation of associated local uplifts (potential traps) took place in Central Latvia (e.g. Liepaja-Saldus ridge) [28] (Fig. 3). A dense network of faults has been identified in West Lithuania and Estonia, but these faults are of much lower order, and the related uplifts are much smaller in size.

Fifteen major structures, with estimated storage capacity exceeding 10 Mt  $CO_2$ , have been identified in West Latvia (Fig. 6). One of these structures has been used for underground gas storage for several decades, which proves the tightness of the structural traps in Latvia (Inčukalns UGS).

The storage capacity of a structural trap is estimated:

$$M_{\rm CO2} = A \times h \times \varphi \times \rho_{\rm CO2r} \times S,$$

where  $M_{\rm CO2}$  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 20–40 m in Latvia and Lithuania),  $\varphi$  is the porosity (typically ranges from 0.25–0.20 in Central Latvia and Central Lithuania to 0.06 in West Lithuania),  $\rho_{\rm CO2r}$  is the in situ



*Fig.* 6. Major Cambrian aquifer structures ( $CO_2$  storage potential exceeding 10 Mt) of Latvia (black circles) and Inčukalns underground gas storage (grey circle). Hatched line shows gas pipelines.

 $CO_2$  density at reservoir conditions (ranges from 600 kg/m<sup>3</sup> in West 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 for both reservoirs).

The total capacity of large structures of Latvia is estimated to the more than 300 Mt of  $CO_2$ , with the potential of the greatest uplifts reaching 40-80 Mt of  $CO_2$ . The major  $CO_2$  emitting sources are located close to major uplifts. Furthermore, the  $CO_2$  sources and potential traps are located close to the existing gas supply pipelines, which potentially reduces the cost of  $CO_2$  transportation.

The capacities of more than 100 Cambrian local uplifts identified in Lithuania were evaluated recently [25]. The two largest Vaskai and Syderiai aquifer structures can store only 3.5 and 5.4 Mt of  $CO_2$  respectively. The rest of the 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-Kemeri aquifer, neither in Lithuania nor in Latvia due to low-intensity tectonic deformation of the Variscan structural complex [25, 28, 30].

#### Solubility trapping

The solubility trapping is not restricted to particular structures. The solubility of CO<sub>2</sub> ranges from 2% to 6%, depending on the brine salinity, temperature, and pressure [12, 31, 32]. However, the large volume of a regional-scale aquifer provides an attractive alternative for CO<sub>2</sub> disposal. The solution time is of order of  $10^2-10^3$  years, which is considerably longer than the hydro-dynamic trapping process [33, 34]. 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 [12]. 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 formation water varies from 25–30 kg/m<sup>3</sup> in West Lithuania to 40–50 kg/m<sup>3</sup> in East 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 as high as 11 Gt of CO<sub>2</sub> within the area of the supercritical state of the carbon dioxide.

The Pärnu-Kemeri aquifer is characterised by better reservoir properties, but has a smaller area of extent than the Middle Cambrian reservoir.  $CO_2$  solubility ranges from 36 kg/m<sup>3</sup> in the deep part of the basin to 60 kg/m<sup>3</sup> in the shallow periphery of the basin. In West Lithuania the storage capacity of the reservoir is about one Mt of  $CO_2$  in one km<sup>2</sup> area. The total onshore potential of this formation is estimated as high as one Gt of  $CO_2$ .

*Table 2.* Solubility trapping in Cambrian reservoir in West, Central, and East Lithuania

Parameters	West Lithuania	Central Lithuania	East Lithuania
Aquifer temperature, °C	75	55	40
Aquifer Pressure, MPa	20	15	10
Salinity, g/l	160	110	90
$CO_2$ solubility, kg/m <sup>3</sup>	28.1	35.6	40.9
Effective thickness, m	20	35	40
Porosity, %	10	20	25
Solubility storage in 1 km <sup>2</sup>	0.056	0.249	0.409
area, Mt of CO <sub>2</sub>			

Furthermore, the mineral trapping that involves a series of interactions between the formation mineralogy and  $CO_2$ -enriched aquifer waters, can convert  $CO_2$  to carbonate, an immobile and harmless mineral that will be stored for millions to hundreds of millions of years [13, 35]. Reactions with Ca/Mg/Fe-bearing silicate minerals are the most promising for carbon sequestration because these silicates neutralize the added acidic  $CO_2$  and provide alkali metals that trap  $CO_2$  through the precipitation of carbonate [14]. These reactions can be summarized as follows [36]:

Ca/Mg/Fe feldspar + clays +  $CO_2$  +  $H_2O$ = kaolinite + Ca/Mg/Fe carbonate + quartz

The Middle Cambrian reservoir comprises quartz sandstones that are practically not reactive to carbon dioxide. The Pärnu-Kemeri sandstones contain clay admixture (up to 10%) and feldspar grains (up to 15%). Therefore they have a potential for permanent immobilisation of carbon dioxide in mineral form. Assuming the rock capacity of 10 kg/m<sup>3</sup> [12], the sequestration potential can be evaluated to reach 5.6 Gt of CO<sub>2</sub> (onshore).

The technology of  $CO_2$  mineral trapping with waste oil shale ash is under development in Estonia. Investigations of Tallinn University of Technology performed within the framework of Nordic-Baltic project NoCO<sub>2</sub> and supported by SC Eesti Energia and Ministry of Environment of Estonia show that the amount of  $CO_2$  which is possible to bind with oil shale ash will cover about 10–12% of the large industrial  $CO_2$  emissions in Estonia (37–41).

#### **Depleted** oil fields

A number of oil fields have been discovered in the Baltic basin, mainly in the Middle Cambrian reservoir [42, 43]. Oil fields are exploited in West Lithuania, Kaliningrad District and offshore Poland. In Lithuania, oil fields are confined to two major tectonic zones, i.e. 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 [43]. The Silurian reefs contain small oil fields in Central Lithuania, which are not exploited.

The Cambrian oil-bearing structures are commonly as large as  $3\times5 \text{ km}^2 \times 20-40 \text{ m}$ . In Lithuania, ten oil fields are presently exploited. The size of oil fields ranges from 16,000 tonnes to 1,400,000 tonnes of the recoverable oil. The storage potential of the largest oil fields in West Lithuania reaches two Mt of CO<sub>2</sub>. The total potential in Lithuania is estimated at 7.6 Mt of CO<sub>2</sub>, which is just a little more than the annual stationary CO<sub>2</sub> emissions of the country.

Another option is the utilisation of carbon dioxide for oil recovery [5]. Most of the oil fields have reached the tail phase, and EOR can prolong the lifetime of those oil fields. The oil is light and exceeds 35 API° [44] meaning that  $CO_2$  could be injected in miscible conditions – a favourable factor for  $CO_2$  sequestration. The estimated total EOR net volume of  $CO_2$  is 5.6 Mt.

## Conclusions

CO<sub>2</sub> sources are distributed unevenly in the Baltic countries and the types of CO2 sources vary considerably. Major emissions are concentrated in the coastal area of the Gulf of Finland in Estonia. Due to the utilisation of oil shale for energy production, CO<sub>2</sub> emissions produced by two largest Estonian power plants exceed the CO<sub>2</sub> produced by all of the Lithuanian (6.6 Mt) and Latvian (2.98 Mt) stationary sources. During 2007, CO<sub>2</sub> emissions from Estonian industrial sources increased for about 18% owing to increased energy production of these two largest in the region power plants. Baltic countries are situated within the Baltic sedimentary basin that contains a number of regional-scale aquifers. However, only two of these, the Lower-Middle Devonian and Middle Cambrian reservoirs, meet the basic requirements for CO<sub>2</sub> storage. The Cambrian prospective area encompasses West Lithuania and West Latvia and most of the Baltic Sea territory, while the aquifers in Estonia are too shallow, they have no reliable seals and they all could be used for drinking water supply. The transportation of carbon dioxide from Northern Estonian sources to Latvian structures could be an alternative option (250-400 km distance to the potential sites in Latvia).

All major  $CO_2$  sources of Latvia are located within the prospective Cambrian area. The most prospective storage of  $CO_2$  is related to 15 large uplifts the total capacity of which exceeds 300 Mt that amounts more than 150 years of country's  $CO_2$  stationary emissions. Furthermore, the structures are rather close to existing pipelines.

Only the north-western cluster of  $CO_2$  sources is located within the prospective area of the Cambrian reservoir and only one source is within the Devonian prospective area in Lithuania. The structural trapping is not an option for Lithuania, as the uplifts are too small. Alternatively, the solubility trapping could be considered as having a high potential. Together with the mineral trapping it should cover industry needs for hundreds of years. However, these technologies are still immature. Basic problems are the poor

knowledge on the migration velocity of the  $CO_2$  plume and solubility rate that are important parameters for the safety assessment. Also, several problems still have to be solved to activate this potential, such as dissolution enhancement, monitoring, etc.

The Inčukalns underground gas storage operating in Latvia, which is used for the supply of natural gas to Latvia, Estonia and Lithuania, is an example of collaboration in the region [45]. The existing infrastructure of pipelines, already connecting the large Baltic  $CO_2$  sources with Latvian prospective structures, provides a possibility of reducing the price of the  $CO_2$  pipelines and some prospect for geological storage of the substantial Baltic industrial  $CO_2$  emissions in the most favourable geological conditions available in Latvia.

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