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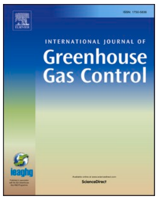

Development of CCUS clusters in Croatia

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ABSTRACT

Carbon Capture and Storage is a concept that is not yet fully implemented largely because of the high costs. Clustering of industrial stakeholders is imposed as a measure for cost reduction. All relevant emitters, possible transport routes, including existing gas pipeline corridors, and their geographic location in relation to potential storage locations are assessed in this paper. Site availability and CO_2 storage capacity are examined, summarizing all study results gathered under the Strategy CCUS project. The CO_2 enhanced oil recovery is being studied for CO_2 storage rather than extra oil recovery. As logical choices, three clusters were recognized. Only less expensive, onshore injection was taken in consideration for assessment of early (economic) feasibility in the Adriatic, Central, and Eastern clusters. Because of the shorter distance between CO_2 emitters and injection sites, the Eastern and Central clusters are being investigated in more detail, despite the fact that the largest point source emitter is in the Adriatic region. Because of small number of point source CO_2 emitters and huge theoretical storage capacity, further research is needed to better assess the storage capacities as well as possibilities for development of cross-border projects. Based on previous research (particularly regarding the emitters), the number of facilities (fewer facilities, with more concentrated emissions), and the availability of storage objects, the Eastern cluster is recommended to be further studied as the next stage of Carbon Capture, Utilization and Storage cluster research and development in Croatia and nearby cross-border regions.

1. Introduction

Because the reduction of CO_2 emissions depends on economic and energy efficiency of the capture, transport and storage/mitigation process, Carbon Capture and Utilization (CCU), and Carbon Capture Utilization and Storage (CCUS) became widely investigated options, primarily in the USA and Europe (IEA, 2020).

Mitigation of CO_2 through CCU and CCUS projects requires high investments. The emitters should involve new capital investments and increase operating costs, and without state support for mitigated CO_2, this will weaken stakeholders' interest in such technologies. The problem is identified in many published works. CCUS policies in China were reviewed and discussed that government should provide direct financial support for CCUS, and that subsidies at power plants with CO_2 capture are needed because of high capture costs (Jiang et al., 2020). The tax exemptions and mandatory market quotas or guaranteed purchases on energy (electricity, gas, oil) produced as a part of CCUS chain were recommended (Jiang et al., 2020).

Solar and wind power generation industries showed how government signals and market forces can positively influence the cost of these technologies, and that market forces, on their own, are not capable of ensuring the feasibility of CCS projects (Billson and Pourkashanian, 2017).

The CCS Directive (European Parliament, 2009) serves as the basis for the legal framework in EU member states. It suggests roles and responsibilities for storage in future CC(U)S projects. In practice (or when applied to individual countries such as Croatia), this framework enables the licensing of a CCS project, but at the same time introduces several uncertainties that will hinder project investments. The exploration phase, which can be costly, is not encouraging to further development of storage projects and investments. Liability costs, monitoring costs cannot be clearly defined - in the Croatian legal framework, the payment of monitoring and liability costs, after the CO_2 injection phase stops, is defined for several decades in advance. To ensure the implementation of CCS, legislators could try to remove these uncertainties as much as possible, while promoting the safety and feasibility of such projects (Haan-Kamminga et al., 2010).

The European Union Emissions Trading System (EU ETS) represents

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a fiscal policy instrument that should be well-aligned with legal framework and should provide annual CO_2 reduction of 2.2% from 2021. It should ensure an acceptable carbon price and stimulate cost-efficient greenhouse gas emission reductions (European Parliament, 2018) serving the objectives of the Paris Agreement. The main commodity that is traded on the EU ETS market is the European Union allowance (EUA), a tradable unit that allows the emission of one metric ton of CO_2. The number of auctioned allowances on the market is regulated by rule mechanism - Market Stability Reserve (MSR) (European Union, 2015; Osorio et al., 2021), which basically prevents auctioning through back-loading of EUAs if a threshold of total number of allowances in circulation is exceeded. Because of its restricting nature, the demand – supply relations could be disturbed and need for revisions of MSR is detected (EP, 2011; Marcu et al., 2021; Morgado Simoes Henrique Andre, 2022). As corporate banking practices develop, policy makers should be cautious because of the potential unintended implications of cap adjustment actions. Unless and until banking is promoted and substituted by some type of restriction, the price of permits may not rise as quickly as it should (Chaton et al., 2018).

From 2023, the quantity of EUAs in the MSR is limited to the quantity of EUAs auctioned in the previous year, which means that "surplus" allowances will be deleted from the system. The market reaction to, amongst other things, the envisaged introduction of this system leads to a significant increase in EUA prices (Bruninx et al., 2020).

In one of the impartially written review papers on the level of development of certain CCU technologies, it is emphasized that there is not much demand from those who can encourage the development of CCUS technologies and the market (Zimmermann and Schomäcker, 2017). For some industries, CO_2 utilization could bring some market advantages or even a lower raw material price, as (expensive) feedstock (of fossil origin) is replaced by CO_2. In such manner, CO_2 related markets are defined as "*market pull*", and authors note that besides market pull, much of research is focused on the commercialization of CCU technologies and supported by public funding in last years to make that "*technology push*", but those identified as "policy pushers" (i.e., policy makers) are the most important and influential drivers, motivating changes by climate mitigation and energy (resource) efficiency.

In EU, due to different levels of technological and economic development and differences in energy resources there is no unique definition of CO_2 utilization. Two interest groups can be distinguished with two different views of CO_2 utilization:

(1) Chemical CCU (CCCU) - CO_2 utilization is a process where CO_2 should change its chemical structure,
(2) Feasible or "Physical CCU" (PCCU) - CO_2 utilization is considered as any technology that utilizes CO_2 as a raw material, making the technology physically and technologically more feasible and cost effective.

Even though two definitions are not opposite at first sight, the big difference comes from different interest groups which are aware of importance of the "official definition" that will be included in EU and its member states policies. From such perspective, without strict definition of CCU and CCUS, as the part of H2020 "Strategy CCUS" project (Strategy CCUS, 2019), promising regions for CCUS are observed for each partner country. Tools for techno-economic analysis (TEA) and Life Cycle Analysis (LCA) are being developed, however huge differences between mentioned regions by means of industry size, amount of CO_2 that could be captured, CO_2 geological storage type and capacity are making development of such tools rather challenging. Notwithstanding variations in CO_2 emissions and CO_2 storage capacities, different technologies are viewed from a different perspective and with varying levels of advocation in different countries. This variation in views highlights that CO_2 utilization and storage in EU Member States comprises a number of options with technologies at different levels of maturity and acceptance in each country. This paper mainly considers CCS and PCCU in Northern Croatia region, which is selected because of ongoing CO_2 Enhanced Oil Recovery (EOR) projects that are achieving additional oil recovery from mature oil fields by CO_2 injection (and thus pressurizing the oil reservoirs and decreasing oil viscosity by mixing with CO_2, while the most of injected CO_2 is stored in the respective reservoir "as a consequence").

1.1. Capture technologies

The most important parts of CCU process (Pieri et al., 2018) are: (1) source characterization, (2) capture/separation, (3) purification, (4) compression, (5) transportation and (6) utilization. They review capture technologies, underlining as the most important (or the most widely considered and used):

(1) Pre-combustion CO_2 capture and separation by physical sorbents (commonly used in natural gas processing plants, but less mature capture technologies from coal and biomass are also considered).
(2) Post-combustion capture from exhaust gases and chemical absorption as separation process.
(3) Oxy-fuel combustion CO_2 capture.

Other possible separation/capture options include chemical looping combustion and direct air capture (Wang and Song, 2020). Additionally, biomass energy with carbon capture and storage should be mentioned here as CO_2 is removed through biomass creation process for further biofuel production (Warsi et al., 2020).

The main drawback of the pre-combustion technology is very low efficiency of the whole process. Post-combustion technology implies robust equipment and energy demanding cooling systems, which both make this technology expensive. Oxy-fuel combustion is a high temperature technology that results in high CO_2 concentration in the flue gas, making the separation/capture phase simple, but pre-treatment is necessary due to various impurities. Additionally, this technology is burdened with high oxygen supply costs (Omoregbe et al., 2020).

Considering post-combustion capture costs, natural gas power plants exhibit notably higher values compared to coal power plants (Schmelz et al., 2020).

Non-power generation industry CCS cost reflecting the CO_2 avoidance cost can be calculated by three commonly used methods that all rely on different assumptions – "exhaustive", "net present value", and "annualization" (Roussanaly, 2019).

Capture cost is the most expensive component of the CCS commercialisation chain, and also hard to generalise because it depends not only on the emitter type and size, but it also takes into account the transport option dictating the operating conditions of the capture phase. Rough estimates of captured CO_2 costs in different process plants and for different capture technologies are shown in Figs. 1 and 2, respectively (Budinis et al., 2018).

Despite different assumptions, reference years, and calculation methods in the literature, Naims (2016) gave an overview of the benchmark capture costs (Fig. 3), including only separation and compression at a single facility, according to source type for the largest point CO_2 sources. Although Petroleum to power and Waste combustion emitting sources were also analysed, no capture costs were reported for those two source types.

Pieri and Angelis-Dimakis (2021) decomposed and investigated capturing costs in power related industries but also in non-power related industries such as metal, cement and fluid catalytic cracking considering different capture technologies. Only the chemical and physical absorption models showed economies of scale, while all other models exhibited reverse economies of scale.

Aforementioned technologies imply CO_2 removal from point emission sources, but air capture could be implemented to mitigate aeroplane and automobile emissions as well as capturing CO_2 from air has been successfully practiced in spaceships and submarines for a long

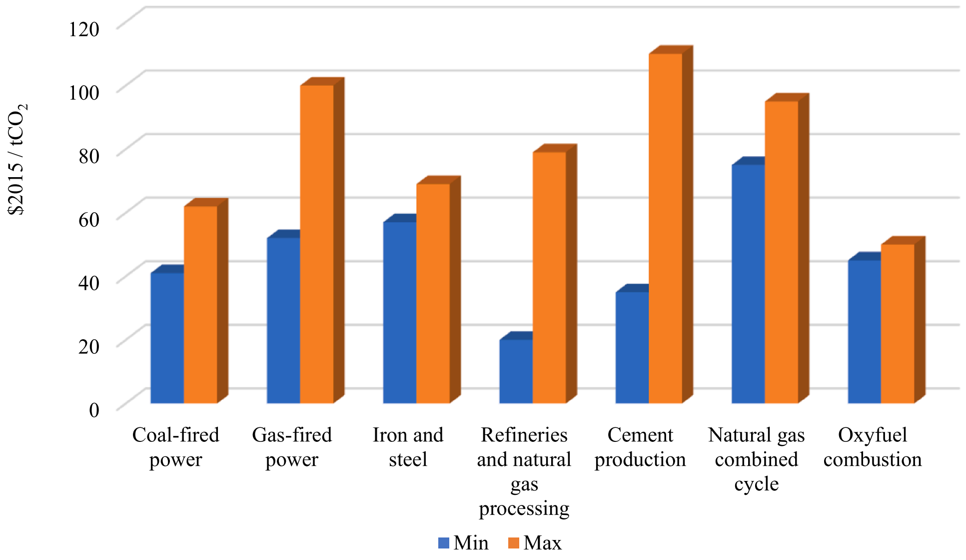

Fig. 1. Cost ($2015/tCO$_2$) of captured CO_2 for different process plants (Budinis et al., 2018).

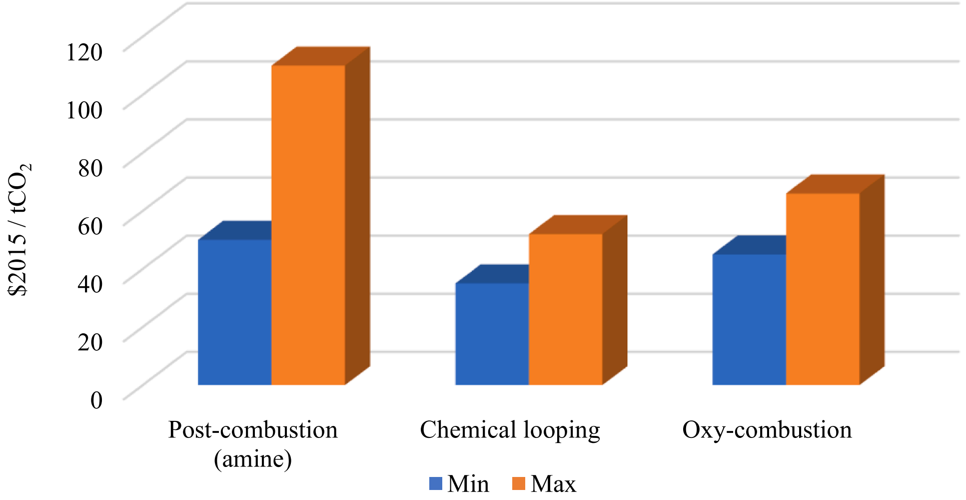

Fig. 2. Costs ($2015/tCO$_2$) for different CO_2 capture technologies (Budinis et al., 2018).

time. Air capture also enables removal of point-source residual emissions and fugitive emissions from the transport and storage phases of the CC(U)S chain. However, to capture notable amounts of CO_2 it is necessary to process large volumes of air, leaving absorption or adsorption as the only feasible solutions (Lackner et al., 2012).

Finally, one of the main challenges hindering carbon capture technology development is the appropriate material selection for efficient and cost-effective separation (Khosroabadi et al., 2021), and recent publications dealing with this issue (Maniarasu et al., 2021; Osman et al., 2021) are proof that the interest in CCS commercialization is on the rise.

1.2. Checmical carbon capture and utilization concepts

Galadima and Muraza (2019) state that the EU countries projected applications in almost all areas of the petrochemical industry, however, CCU fuels are two to three times more expensive than fuels from oil and gas reservoirs. They critically discuss the three important CCU options:

(1) CO_2 Methanation or CO_2 to Gasoline Hydrocarbons technologies, where price of catalysts along with heat management and energy requirements are the main issues. Technology is always connected with low carbon to hydrogen ratio, that considers CO_2 for CH_4 production instead of the desired C5+ compounds. The CO_2 hydrogenation can be achieved with Na–Fe_3O_4 and zeolite catalysts (Wei et al., 2017) as the reduction of CO_2 to CO through reverse water-gas shift reaction and successive hydrogenation of CO to hydrocarbons through Fischer–Tropsch synthesis.

(2) CO_2 to Methanol - high levels of CO_2 conversion are hard to achieve and the industry prefers catalysts that should be improved to be closer to the "green approach", which should include CO_2 and (renewable-based, (Nyári et al., 2020)) hydrogen produced through the water electrolysis by using renewable energy.

(3) biofuels from algae - microalgae can be used to produce organic carbon by CO_2 conversion during photosynthesis.

(4) Bioenergy carbon capture and storage (BECCS) – organic material (i.e. biomass) is used to create heat, electricity, biomethanol or biogas. The process includes CO_2 capture, after which CO2 storage is required. Storage is possible by injecting into geological formations or by embedding into other products. As CO_2 is

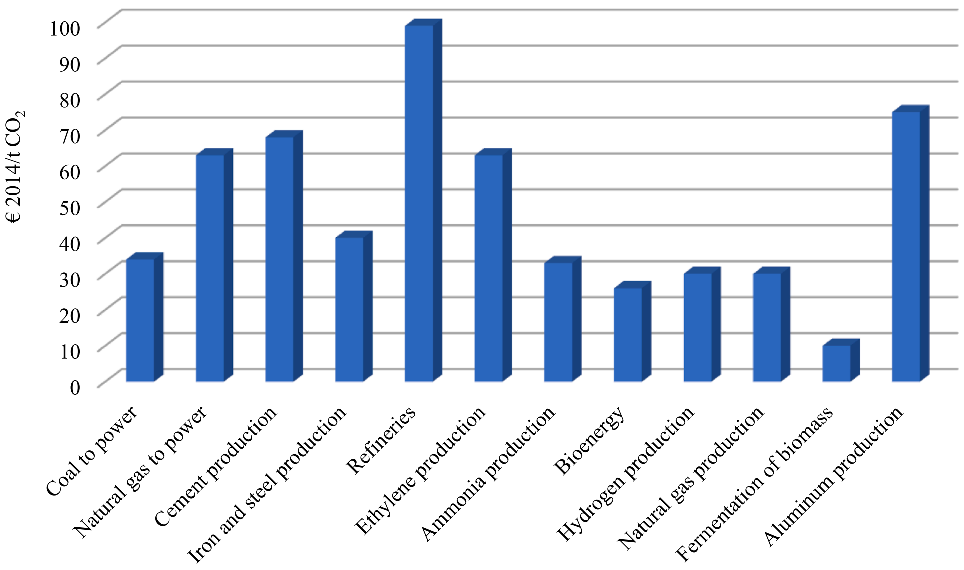

Fig. 3. Literature-based CO_2 benchmark capturing costs in € 2014/t CO_2 (Naims, 2016).

initially taken up (stored) by new growing plants (biomass), the process can be considered as use and storage.

Broader list of CCU technologies that is substantially covered in the literature and involves:

- Carbonation (carbonate mineralisation by reaction of CO2), usually in waste-to-energy plants, for producing the construction materials, i.e., CCU cement (e.g., concrete curing to make concrete building materials, (Ravikumar et al., 2021) which could mitigate up to 1.4 Gt of CO2 within the facility lifespan (e.g., in 30 years, (Hepburn et al., 2019)).
- Cement production with CO2 utilization – One of the most promising approaches is to use and store (or prevent) the emissions related with cement manufacture in the production of concrete. Capturing CO2 from exhaust gases and purifying it results in a rich CO2 stream that may subsequently be transformed to methane, methanol, or other useful chemicals.
- Food and carbonated beverages - CO2 is currently used for carbonation of drinks, food freezing
- Horticulture – utilization of CO2 to enhance the plant growing and increase the production yield of crops with small part of CO2 absorbed (temporarily stored), while the most (more than 75%) is released into the atmosphere.
- Polymer production - CO2 is used to produce different types of polymers (e.g., polyurethane). Up to 50% CO2 can be temporarily stored in the material.

When it comes to chemical industry, the term defossilization is more appropriate than decarbonization, and the schemes for achieving this defossilization include carbon capture and storage based on fossil fuels usage, carbon capture and utilization in new chemical processes with "green" hydrogen, and the use of biomass for production of chemicals (Gabrielli et al., 2020). The selection of one of these options depends on the trade-off between the necessary energy consumption, land use, and the resulting emissions.

1.3. Physical carbon capture and utilization technologies

The most important (by quantity, TRL and economic feasibility) feasible CCUS method is miscible CO_2-EOR. By injection of CO_2 to oil fields, after some distance from injection well(s), it can mix with oil by successive vaporization of lighter and medium components from oil, which promotes transport of "light phase" (CO_2 plus lighter and medium weight hydrocarbon components) further in the reservoir and finally dissolution of "light phase" in oil. During that process, which is called multiple contact miscibility process (condensing and vaporizing mechanism), oil reservoir is pressurized, increasing oil flow potential, the oil viscosity is reduced (greater oil mobility) and additional important effect is increase of oil (volumetric) saturation in pore space, which is called "swelling effect", and which increases relative permeability of oil (compared to other fluids in the reservoir, such as brine and petroleum gas). The result of such increased reservoir potential and oil mobility is additional recovery of oil, where increase of (dissolved) CO_2 in produced fluid composition should be monitored, accounted, and recovered through (some form of) CO_2 taxes.

Economic feasibility of physical CO_2 utilization for enhanced oil recovery has been confirmed both in many studies and commercial projects. In a study by Suicmez (2019), a long-term development option using Enhanced Oil Recovery (EOR) in the Danish sector of North Sea was investigated for its technical and economic viability. The study focused on the subsurface aspects, such as estimating the amount of CO_2 required and the additional oil to be recovered, as well as the offshore facilities required to gain additional recovery. Based on these estimates, the author performed a cash flow analysis by running stochastic simulations by changing certain parameters (oil price, discount rate, CO_2 cost, hydrocarbon tax). The author concluded that to reach the threshold NPV ($244 million), either the oil price should increase, or the discount rate and the cost of CO_2 should decrease. The hydrocarbon tax does not have a significant impact because it is only effective when the NPV of the project is positive. Since the CO_2 price is considered a disadvantage to the economic feasibility of CO_2-EOR, there is room for further study discussion since some of the injected CO_2 is permanently stored in the reservoir and the rest is produced with additionally produced oil. Arnaut et al. (2021) combined the same parameters with oil reservoir parameters (such as permeability and pressure), water-alternating gas (WAG) ratios, and well-pattern in their sensitivity study and also confirmed that CO_2-EOR can be economically feasible, even at lower oil prices, but with large CO_2 utilization – if CO_2 retention in the reservoir is sufficient and the CO_2 price is high enough.

2. Recent research and worldwide development of carbon capture, utilization, and storage projects and technologies

According to the Global CCS Institute (Global CCS Institute, 2021), there are 168 facilities for capturing and sequestration worldwide in different development statuses and categories. Most of the projects are pilot and demonstration CCS facilities with an equal share in Europe and America. However, commercial projects are mainly located in America, while less than 20% of the total number of commercial projects in the world are located in Europe (Fig. 4 Share of CC(U)S projects by category and location, modified from Global CCS Institute CO2RE data (Global CCS Institute, 2021)).

According to the project's status, there are six different categories: (1) advanced development, (2) completed, (3) operational, (4) early development, (5) operation suspended, (6) in construction (Fig. 5).

The first CO_2 injection project was developed in Texas, the United States of America, in the early seventies, where CO_2 captured at the natural gas processing plant is transported via pipeline to mature oil fields for enhanced oil recovery (Speight, 2019). Almost 25 years later, the first implementation in Europe - Sleipner CO_2 Storage started (Baklid et al., 1996; Furre et al., 2017; Hermanrud et al., 2009).

Since 2000, there has been a significant increase in the number of such projects globally, and linear growth is projected until 2035. In the next 15 years, the development of more than 50 such projects is planned. The cumulative number of CC(U)S projects expanded practically exponentially from the 1970s to 2020, and it is expected to rise at least linearly from 2021 to 2035 (Fig. 6).

3. Overview of carbon capture, utilization, and storage options in Croatia

An overview of largest CO_2 point sources in the Northern Croatia is given along with the analysis of their emissions. The possible transport routes from identified emitters to potential storage objects are indicated. Regarding potential storage objects, two types of storage objects are presented: nearly depleted hydrocarbon reservoirs that are well characterized, and regional deep saline aquifers that need to be further investigated for structural and stratigraphic traps. Also, CO_2 utilization options are analysed, mostly focused on the deployment of the CO_2-EOR method.

3.1. *CO_2 sources/emitters*

The Northern Croatia promising region comprises ten emission sources (Fig. 7), with two additional sources outside the considered region added to the analysis, due to existing transport connections with Northern Croatia and their significant emissions exceeding 1 Mt/y: "TE Plomin" coal power plant, (marked with 11 on Fig. 7), and Rijeka refinery (Rafinerija nafte Rijeka, marked with 12 on Fig. 7), with CO_2 emissions of 1.2 Mt/y and 1.0 Mt in 2018, respectively. Overall, six power plants and two refineries define the dominating sectors, both regarding the number of facilities and the percentage of emissions, accounting for 67% of the emissions in the area (Fig. 8). Other industrial sectors are represented by a single facility, although in some cases with important emissions, particularly the cement plant at Našice (marked with 2 on Fig. 7), which emitted 0.65 Mt CO_2 in 2018, and the "Petrokemija" fertilizer plant (marked with 1 on Fig. 7) near Kutina, accounting for 0.75 Mt/y. The oil & gas processing sector has one important facility at Virje: Molve Natural Gas Processing Plant (marked with 5 on Fig. 7), with emissions of 0.29 Mt/y. There is also a glass production facility (marked with 9 on Fig. 7), with emissions just above 0.1 Mt/y.

The emission trends for seven sources have been increasing, with some of the larger sources, such as the "NEXE" cement factory, the Rijeka refinery, and the "TE-TO Zagreb" (combined heat and natural gas power plant). Apart from the Sisak refinery, where operation is to be terminated (with facilities being converted for biorefinery and bitumen production), two other facilities show a decreasing trend in emissions: "EL-TO Zagreb" (combined heat and power plant), and the third largest emitter in the area, the fertilizer production "Petrokemija" with the main driver being consumer behaviour. The largest CO_2 emitter in the region, the "TE Plomin" coal power plant shows a stabilized trend in emissions, and has the permit until 2040 to operate as energy balancing system with 210 MWe (Croatia, 2021; Jerković, 2022; The Ministry of Economy and Sustainable Development of the Republic of Croatia, 2022). Regarding the number of emission points, the emitters show significant variations. The number of emission points is very high in the Sisak refinery (19 points) and at the gas processing facility at Virje (14), increasing the difficulty and costs of CO_2 capture. No indication is provided about the number of emission points at the Rijeka refinery, but it is likely to be high. In all other sources, the number of emission points varies from one (the cogeneration biomass plant "Viridas") to six ("TE-TO Zagreb").

CO_2 produced from biomass combustion is not only the predominant in the cogeneration power plant at Babina Greda but may also be important at the "TE-TO Sisak" power plant in Sisak which uses biomass in the fuel mix. The cement sector is where process induced emissions are most relevant, with 65% of the emissions not related to fuel combustion, but it is possible that the fertilizer facility "Petrokemija" also has an important component of process emissions.

Considering fuel utilization, natural gas is the main one, except for the coal power plant and the biomass cogeneration power plant "Viridas". Refinery gas and other gases are used as fuels in the Rijeka and Sisak refineries, while the cement factory utilizes petroleum coke, coal, and natural gas.

From the CO_2 capture point of view, it is worth mentioning that no high CO_2 concentration sources were inventoried in Northern Croatia, the highest being at the cement plant at Našice. It should be noted that no issues are expected regarding space requirements for construction of the CO_2 capture facilities at most of the emitters' locations due to their

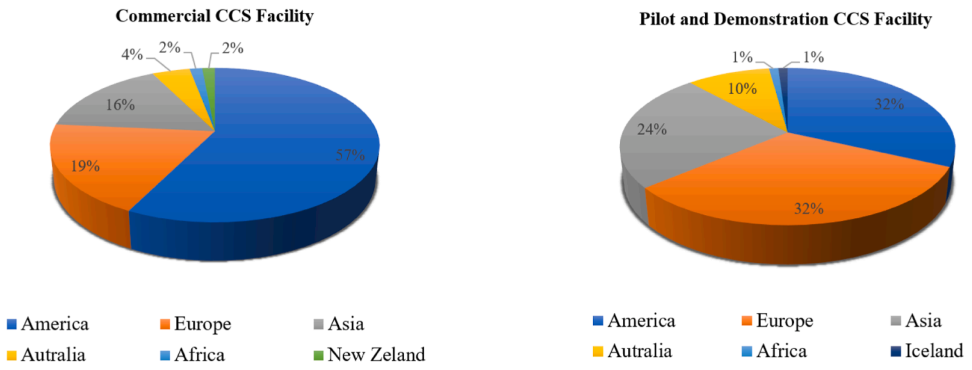

Fig. 4. Share of CC(U)S projects by category and location, modified from Global CCS Institute CO2RE data (Global CCS Institute, 2021).

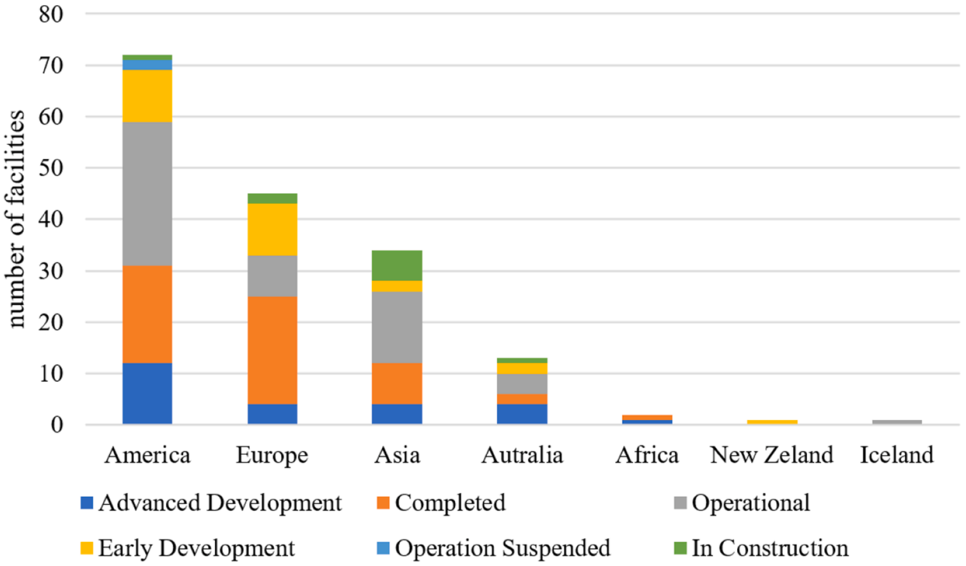

Fig. 5. Facility Status per location modified from Global CCS Institute CO2RE data (Global CCS Institute, 2021).

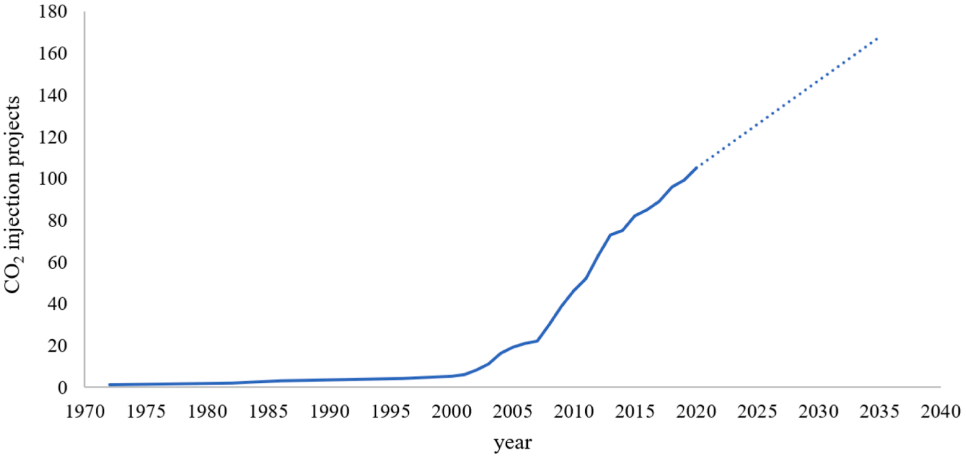

Fig. 6. Total cumulative number of CC(U)S projects in the world source of data: Global CCS Institute CO2RE data (Global CCS Institute, 2021).

distant position from urban areas. Exception is the EL-TO power plant in Zagreb, which is located in the densely inhabited city district, so building any additional facilities on site is expected to be challenging.

3.2. CO_2 transport options

Fig. 7 depicts transport options which include: natural gas pipelines, railways and well-maintained roads including motorways and county roads. Additionally, emitters and storage units are also presented on the map to evaluate the most efficient transport method of the captured CO_2 (Fig. 7), CO_2 emitters and storage sites with possible existing transport modes (Strategy CCUS, 2019).

Existing natural gas pipeline network covers almost all of the emitters and storage units (Fig. 7) locations. It is well known that for large enough CO_2 quantities, transportation via pipeline is the most cost-efficient method. However, the scenario in which the existing natural gas pipeline network is used only for CO_2 transport is not taken in consideration; the pipelines are currently used for natural gas transport and the economics of the retrofitting of the existing pipeline system is not feasible. Therefore, the routes shown on Fig. 7 indicate the possibility of construction of the new pipeline for CO_2 transport following the route of the existing natural gas pipeline. This imposes as the optimal solution due to already resolved property issues and other legal issues in the mentioned area.

Amongst the emitters which are not located near the existing natural gas pipeline network is the biomass cogeneration power plant “Viridas” (marked with 10 in Fig. 7). Furthermore, the emitter is not connected to the railway network. Hence, the transport option for this specific case is by roads or by purposely built pipeline. Since the CO_2 emissions are low in quantity (0.1 Mt/y), transport by road should be considered as most promising in the technoeconomic scenarios.

Additionally, since TE-TO and EL-TO power plants in Zagreb (marked with 3 and 7 on Fig. 7, respectively) are located within the city boundaries, CO_2 transport options remain unclear. In authors’ opinion, additional effort is needed in order to accomplish CO_2 transport via pipeline for these two emitters due to their CO_2 emissions of 0.7 Mt/y.

Similar to the cogeneration power plant “Viridas” in Babina Greda, the glass production facility “Vetropack Straža” in Hum na Sutli has low CO_2 emissions of around 0.1 Mt/y. Unlike the cogeneration plant, it is connected to the existing natural gas pipeline, which means that purposely built pipeline for CO_2 transport is an option, but due to the low emission quantities, transport by road seems to be an optimal solution.

3.3. CO_2 storage options

Storage objects in the North Croatia region are identified in the Sava

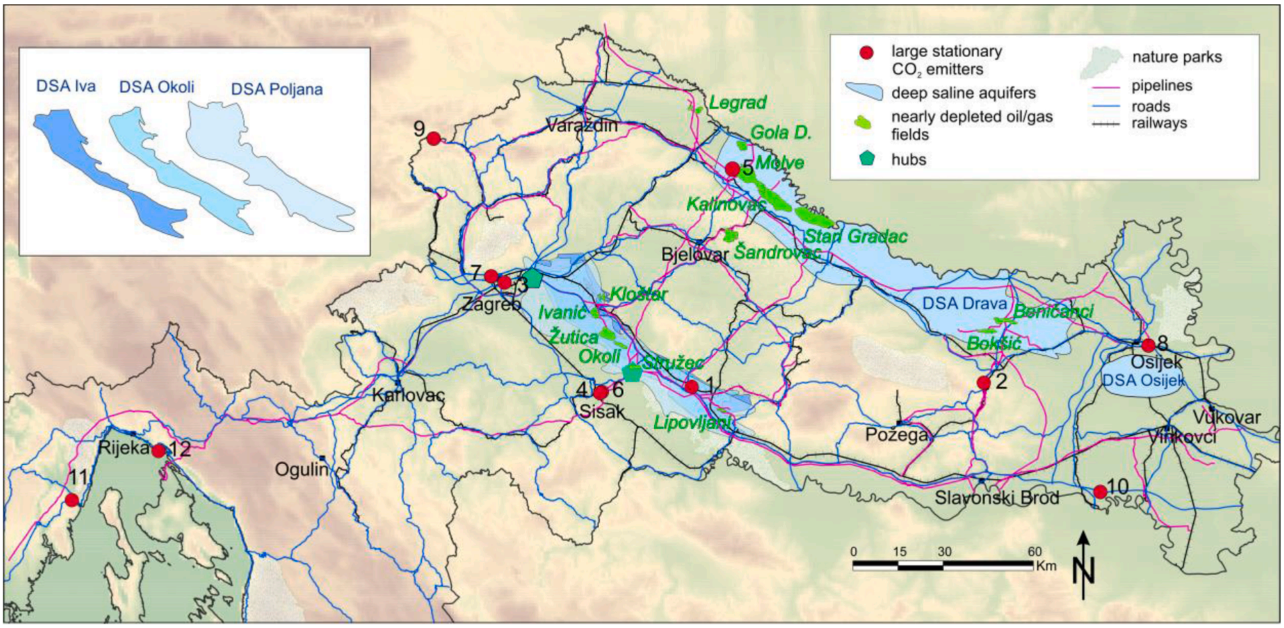

Fig. 7. The largest CO_2 emitters, potential transport routes and potential CO_2 storage objects identified by authors within the Strategy CCUS project (emitters are as follows: 1-"Petrokemija" fertilizer facility, 2-"NEXE" cement factory; 3-"TE-TO Zagreb" Heat and Power Plant (HPP); 4-Sisak Oil Refinery; 5-Natural Gas Processing Plant (NGPP) Molve; 6-"TE-TO Sisak" power plant; 7-"EL-TO Zagreb" Heat and Power Plant; 8-"TE-TO Osijek" Heat and Power Plant; 9-"Vetropack Straža" glass factory; 10- Cogeneration Biomass Plant "Viridas"; 11-" TE Plomin" Power Plant; 12-Rijeka Oil Refinery).

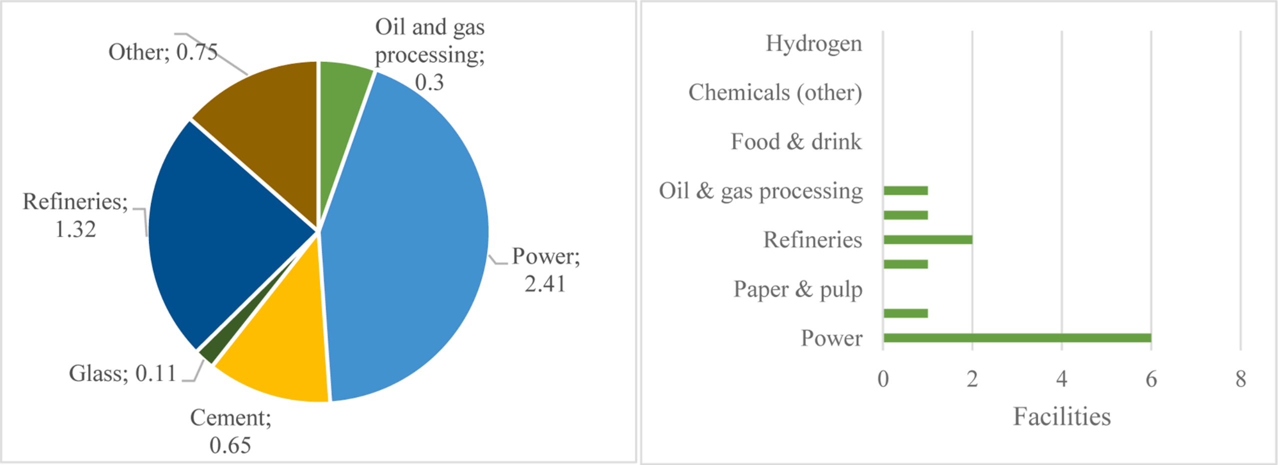

Fig. 8. Emissions (in Mt/y) and facilities per sector (Strategy CCUS, 2019). The sources are quite dispersed across the region (Fig. 7), with Zagreb and Sisak being the only cities with more than one source: two heat and power plants are present in Zagreb: "TE-TO Zagreb" (marked with 3 on Fig. 7) and "EL-TO Zagreb" (marked with 7 on Fig. 7), and in Sisak the "TETO Sisak" power plant (marked with 6 on Fig. 7) and a refinery can be found (marked with 4 on Fig. 7). All other sources are isolated, i.e., tens of kilometres distant from each other.

and Drava depressions which are located in the SW part of the Pannonian basin system (PBS). The depressions are characterized by a rather thick succession of Neogene sediments whose deposition resulted from Mid-Miocene rifting and subsequent thermal subsidence in PBS (Lučić et al., 2001; Malvić and Cvetković, 2013; Pavelić and Kovačić, 2018; Saftić et al., 2003). A brief tectono-stratigraphic overview of depressions' development is given below to enable a better understanding of the storage objects' features. The lithological column is given in Fig. 9.

Opening of Drava and Sava depressions is associated with the continental rifting that began in Ottnangian age and the main extension stage lasted until Badenian age (Lučić et al., 2001). Older and Middle Miocene epoch is generally characterized by a syn-rift sedimentation of coarse-grained clastics deposited in alluvial to lacustrine environments, with some pyroclastics (Saftić et al., 2003); see Fig. 9). Main marine transgression occurred in Badenian age (Ćorić et al., 2009; Pavelić and Kovačić, 2018). During the latest Badenian age, the sea-level fall caused erosion of the newly formed islands and resulting deposition of shallow-water gravel, calcarenites and limestones (Pavelić and Kovačić, 2018). General shallowing trend occurred during Sarmatian age, resulting from local compression, is indicated by a heterogenic lithological composition, from limestones and calcitic marls to sandstones and conglomerates in latest Sarmatian age (Pavelić and Kovačić, 2018).

In Pannonian age, displacement along the normal faults on southern margins of the Sava and Drava depressions enabled widening of half-grabens, reaching all the way to the Slavonian Mountains (Jamičić, 1995; Lučić et al., 2001). The expansion of accommodation area was accompanied by increase of sediment supply, resulting in deposition of

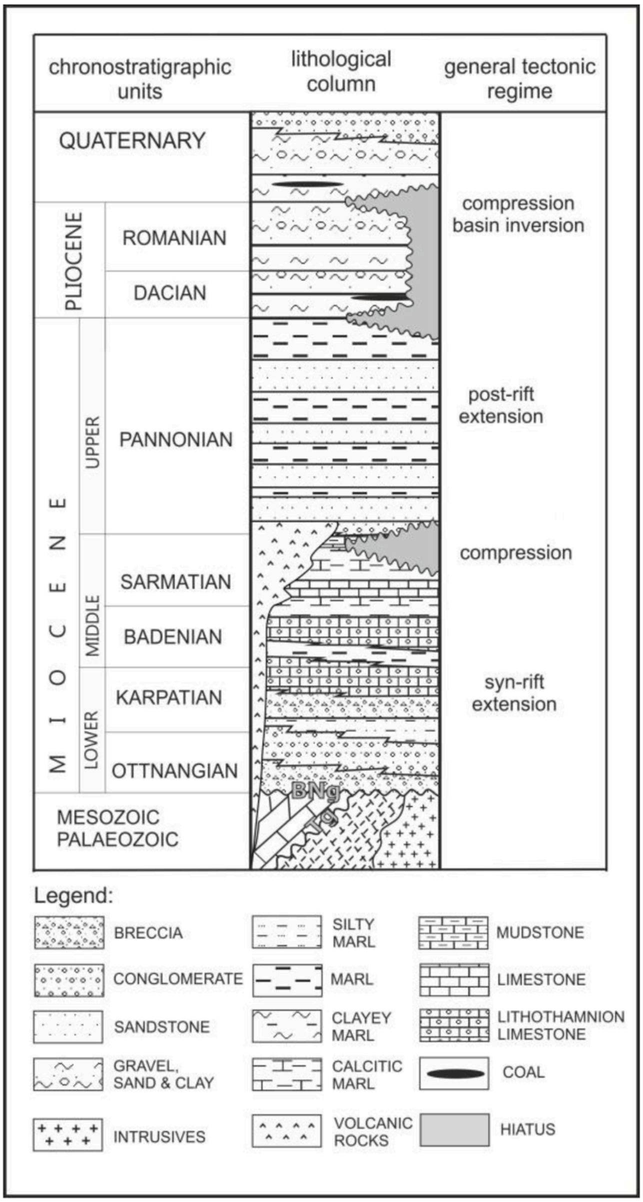

Fig. 9. Schematic lithological column of subsurface in Sava and Drava depressions (modified after (Malvić and Cvetković, 2013; Saftić et al., 2003; Saftić and Kolenković, 2011).

rather uniform sandstone–marl sequence (Pavelić and Kovačić, 2018; Saftić et al., 2003; Šimon, 1973).

The neotectonic phase in the Pliocene and Quaternary epoch is characterised by marked compression of the area (Lučić et al., 2001). Remnants of the Lake Pannon have been filled during Pliocene epoch and Quaternary period with coarse clastics mixed with clay, with occurrences of lignite seams (Saftić et al., 2003).

The structural settings are illustrated by cross-section through depressions (Fig. 10) showing inherited pre-Neogene paleorelief resulting from Late Cretaceous-Paleogene compression/transpression. In the southern parts of Sava depression and Bjelovar subdepression, normal, mostly listric faults are present, that have accommodated extension, i.e., opening and deepening of the depressions during Early to Middle Miocene epoch. Some of the initially normal faults reflect the compressional phase that took place in Sarmatian as well as the recent Pliocene-Quaternary basin inversion (they have been reactivated with reverse character). In analyses in Strategy CCUS project, classification of storage site suitability criteria has been used:

- Tier 1 - Regional assessment; equivalent to prospective (theoretical),
- Tier 2 - Discovery assessment;equivalent to low contingent (effective),
- Tier 3 - Prospect assessment; equivalent to pending/on hold (practical),
- Tier 4- Site assessment; equivalent to justified/approved/on injection (matched), project.

Both depressions (Sava and Drava) include 19 assets with 14 Tier 2 depleted hydrocarbon fields (DHF) and 5 Tier 2 deep saline aquifers (DSA) (Fig. 7). All DSAs are associated with Pannonian sandstones, as well as most of the depleted hydrocarbon reservoirs, making them the most promising storage objects. This is due to the fact that they can be reliably correlated and usually in the convenient depth range. At some locations, identified storage objects are in the Neogene breccia-conglomerate bodies (Beničanci field), and particularly where those reservoirs are hydraulically connected with the underlying Mesozoic or Palaeozoic rocks (Molve, Stari Gradac gas fields).

3.3.1. *Deep saline aquifers (DSA)*

DSA Drava and DSA Osijek, both defined in the Drava Depression in the eastern part of the promising region, have an estimated storage capacity of 2049 Mt CO_2 (Table 1). The boundaries and characteristics of DSA Osijek are defined based on work by Brezovac (2021). These two aquifers are well located with respect to the NGPP Molve, the cement factory at Našice and the “TE-TO OSIJEK” power plant, and at about 40 km from the Biomass power plant “Viridas” at Babina Greda (Fig. 7).

The deep saline aquifers in the Sava Depression, DSA Poljana, DSA Okoli and DSA Iva are represented by three members of Pannonian sandstones – Poljana Sandstones, Okoli Sandstones and Iva Sandstones, respectively. The boundaries and characteristics of DSA Poljana are defined based on work of Kolenković et al. (2013) and DSA Okoli and Iva were defined after Vrbanac (Vrbanac, 1996). Their joint storage capacity amounts to 536 Mt. These three DSAs are located in the centre of the Northern Croatia region, at distances less than 20 km to the remaining sources, except for the Rijeka refinery and the TE Plomin coal power plant, which are at 170 km and 140 km distance. The glass factory is also at around 60 km (Fig. 7).

The arrangement of these three regional storage units can be particularly favourable for the flexibility of the storage system, with the storage units of larger capacity (Poljana and Okoli, each with more than 200 Mt) compensating for the smaller capacity site (Iva, with 55 Mt).

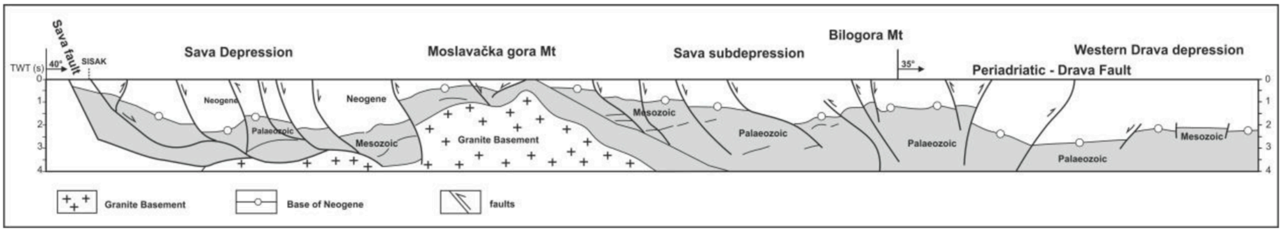

Fig. 10. Cross section through the Sava depression, Bjelovar Sag and SW part of Drava depression (Lučić et al., 2001).

Table 1
Main features of potential DSA storage sites*.

Regional Deep Saline Aquifer	Age	Lithology	Setting	Depth to top (m)	Unit thickness (m)	Storage Capacity (Mt)
DSA Poljana	Pannonian	Sandstone	Onshore	1450	150	251.6
DSA Okoli	Pannonian	Sandstone	Onshore	2000	250	229.0
DSA Iva	Pannonian	Sandstone	Onshore	2050	250	55.1
DSA Drava	Pannonian	Sandstone	Onshore	900	1000	1938.9
DSA Osijek	Pannonian	Sandstone	Onshore	1000	105	109.9

[*] For age and general lithological composition see Fig. 9.

Given that the emission levels in the twelve operational sources is 5.53 Mt/y it is likely that here is enough storage capacity in the deep saline aquifers to meet the maximum possible demand of CO_2 capture in the region. However, it should be emphasized that capacities of these units are highly theoretical and further research is needed in order to define the traps as potential storage objects within them.

3.3.2. *Depleted hydrocarbon fields (DHF)*

Hydrocarbon fields represent another storage opportunity in Northern Croatia, with seven fields producing oil and seven fields producing natural gas. The full storage capacity in the fourteen fields amounts to 146 Mt (Fig. 11), based on the total recoverable volume of oil/gas under reservoir conditions, and considering that CO_2 could replace the volume that was previously occupied by the hydrocarbons in the reservoirs. Several of the producing fields are almost depleted. In fact, the Bokšić and Legrad fields should have stopped producing already in 2021, but they are still in operation while the Okoli field will be depleted by 2023 (Table 2).

Geographically, the hydrocarbon fields overlap those of the deep saline aquifers, since many of them are situated within the same Sava group, increasing the flexibility of a storage system. There is a group of nine hydrocarbon fields located in the Eastern Croatia, with a full capacity of 113 Mt, and a second group of five fields in the Central Croatia, with a capacity estimated at 31 Mt. These capacities are considerable and, most of all, reflect a higher level of confidence in the storage adequacy of the formations, as containment conditions are proved and the level of both detail and reliability of characterization is much higher than for the DSAs. Hence, the hydrocarbon fields are likely to be the initial preferential targets in the Croatian promising region. Still, storage capacity in the hydrocarbon fields is not evenly distributed; in Western part of Drava depression only two fields provide 74 Mt capacity, the Molve and Kalinovac fields. Capacity in all other fields range from 3 Mt to 13 Mt. Field availability will obviously vary, and although some fields were expected to become available for CO_2 injection in 2022, most of them are still in operation, and will continue producing until 2030. Thus, the alternative for storage in these fields is CO_2-EOR, which is already ongoing in Croatia.

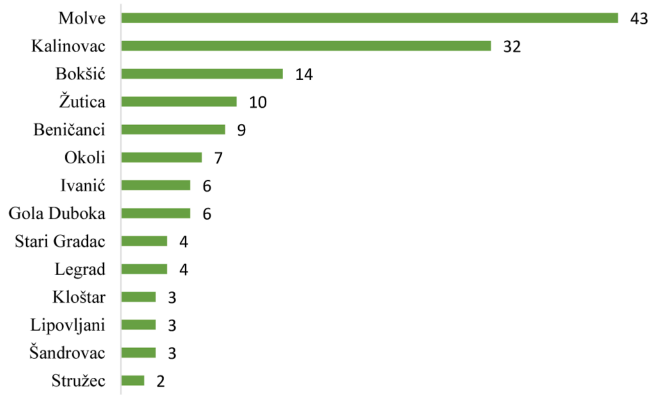

Fig. 11. Distribution of storage capacity (in Mt of CO_2) for DHF storage units.

Table 2
Main features of potential DHF storage sites.

Storage Unit	Lithology	Depth to top (m)	Storage Capacity (Mt)	Field Availability
Kloštar	Sandstone/Granite	973	2.7	2027
Ivanić	Sandstone	1619	5.5	2032
Žutica	Sandstone	1699	10.1	2032
Okoli	Sandstone	1750	7.3	2023
Stružec	Sandstone	727	1.7	2040
Lipovljani	Sandstone	1026	3.2	2023
Gola Duboka	Carbonate	2521	5.8	2030
Molve	Breccia/ Carbonates/ Metamorphic rocks	3100	42.8	2027
Kalinovac	Carbonates/ Metamorphic rocks	3054	31.6	2035
Stari Gradac	Clastics/ Carbonates/ Metamorphic rocks	3450	3.5	2035
Beničanci	Carbonate breccia	1700	8.8	2035
Bokšić	Sandstone	1519	13.6	2021
Legrad	Sandstone	1635	4.1	2021
Šandrovac	Sandstone	750	2.9	2037

3.4. *CO_2 utilization options*

The utilization of the captured CO_2, instead of solely storing it, is of great importance for the feasibility of any CCUS project since it creates an additional revenue stream. Therefore, for Northern Croatia region, enhanced oil recovery (EOR) is considered as the most appropriate CO_2 utilization method due to the maturity of the technology and ongoing CO_2-EOR operations in Croatia.

It should be noted that CO_2-EOR screening campaign is already ongoing in republic of Croatia, and that more candidates are already listed and published (Goričnik, 2001; Smontara and Bilić-Subašić, 2014).

After the initial screening (based on oil density ρ_o, oil viscosity μ_o, oil composition C_N, saturation S_O, effective thickness h_{eff}, permeability k, depth h, and temperature T_r), the evaluation of CO_2-EOR potential was carried out. It was performed by numerical simulation parameter sensitivity analysis (by use of conceptual models that have the volumetric, rock, and pVT properties of observed oil reservoirs) to assess details about the most suitable CO_2-EOR process (e.g., WAG ratios, produced water and oil, gravity drainage, CO_2 retention during the injection, final CO_2 capacity, etc.).

Based on the available data, possibility of CO_2 utilization as a part of CO_2-EOR was examined in several steps:

1 Extended available data (i.e., more detailed, compared to those for depleted hydrocarbon
2 fields) required for CO_2-EOR evaluation was collected.
3 CO_2-EOR screening has been performed, based on Taber et al. (Taber et al., 1997a, 1997b) criteria and validated based on Smontara and Bilić-Subašić (Smontara and Bilić-Subašić, 2014) for Croatian oil fields (Table 3).

Table 3
Screening criteria for CO_2-EOR candidates.

ρ_o	μ_o	C_N	S_o	h_{eff}	h	k	T_r	porosity	heterogeneity
kg/m^3	**mPas**	**%**	**%**	**m**	**m**	**mD**	**°C**		
<920	<10	C_5-C_{12}	>20	wide range	>760	not critical	not critical	>11	secondary porosity is unfavourable

4 Conceptual numerical models for seven Croatian oil fields were developed. Neither detailed static models, nor detailed data per wells were included, because of data confidentiality.

The criteria for screening along with the results are presented in Table 3. The explicit values of screening parameters are not shown due to the confidentiality of the data and certainty of the data is given instead. Additionally, the table provides only the data for CO_2-EOR candidates that are also good candidates for storage. CO_2-EOR projects will provide larger storage volumes and quicker development of an injection project due to economic parameters.

After the screening criteria have been validated, generic numerical models (without detailed geological models) were developed to simulate CO_2 injection. Production data and final pressure on a reservoir level were matched with simulation results in conceptual compositional Eclipse model. The production (decline and waterflood) to year of 2025 was simulated for all fields while the CO_2-EOR process was simulated until 2040.

Detailed PVT CO_2-EOR studies for Ivanić and Žutica were available (Vulin et al., 2018) and based on those studies PVT conditions of the model are defined, while for other fields estimated minimum miscibility pressure conditions were used. All seven analysed oil fields in the Northern Croatia region are feasible CO_2-EOR candidates with their data certainty being at a satisfactory level.

4. Carbon capture, utilization, and storage clustering and scenario development in Croatia

In Northern Croatia, the spatial distribution of sources indicates three potential clusters based on the 175 km distance (from a perspective storage unit) criteria: the Eastern cluster (including emitters marked with green circles), the Central cluster (emitters marked with yellow circles), and the Adriatic cluster (emitters marked with blue circles, Fig. 12).

The sources in the Eastern cluster are the cement factory, the biomass plant and the Osijek Power plant, which jointly emit about 0.86 Mt/y. Consolidation hubs do not seem to be necessary for this cluster, since the most obvious candidates for CO_2 injection are the CO_2-EOR candidate field Beničanci and depleted natural gas field Bokšić, with a total storage capacity of 23 Mt, enough to store several years of the emissions from the three sources. These fields are located from 18 km to 65 km from the sources in the cluster, but with a spatial arrangement that does not require for a consolidation hub right until the injection site.

The Central cluster includes seven sources which together emit 2.5 Mt/y. The cluster is spread geographically in an area of about 8500 km^2, with the Molve power plant quite distant from any other source. Two scenarios of consolidation hubs for collecting CO_2 from the Central cluster can be envisaged. The first scenario considers two hubs. The first hub is located near Ivanja Reka (Zagreb area), gathering CO_2 from the glass factory, TE-TO Zagreb, and EL-TO Zagreb power plant. Trunk transport from the hub to the storage sites could then follow the pipeline corridors of the existing natural gas network. The other hub is located near Stružec (15 km from Sisak), gathering captured CO_2 from the Sisak Refinery, the TE-TO Sisak power plant, the fertilizer production plant in Kutina and the Molve NGPP in Virje. CO_2 is expected to be trunk transported by pipeline from the hubs in this cluster to storage sites in the western part of the Sava depression, firstly to be injected into oil reservoirs for EOR operations, in connection to the ongoing CO_2-EOR projects in that area. In the second phase, the transport would be directed toward the large gas fields expected to become depleted in a few years in the western part of the Drava depression, although it should be noted that the largest emitter in that area is the Molve NGPP, whose emissions will decrease as the gas production from the mentioned gas fields declines.

The second scenario for the Central cluster assumes that, in the first phase, CO_2 coming from all sources except from NGPP Molve would be

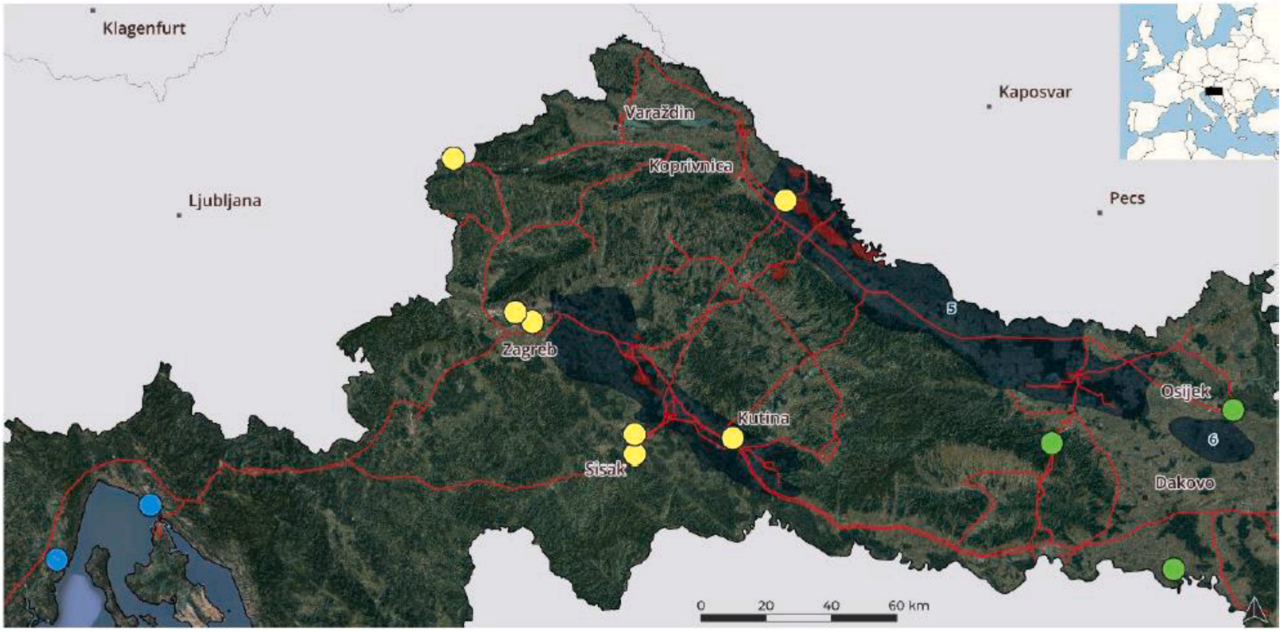

Fig. 12. Clustering of CO_2 emitters and location of existing natural gas pipelines (Strategy CCUS, 2019).

injected into oil reservoirs for EOR operations while CO_2 from NGPP Molve would be injected into reservoirs of Šandrovac oil field for EOR. Nowadays, there is an operational pipeline transport from Molve to western Sava, making the transport in opposite direction also viable. In any case, CO_2 from sources within the Central Cluster is expected to be injected firstly in oil fields reservoirs as part of EOR operations and afterwards into depleted gas fields.

The two large emitters from the northern coastal part of Croatia, the Rijeka refinery and the TE Plomin coal power plant, compose the Adriatic cluster. This cluster is quite distant from the storage sites, but it comprises the largest emitters in the region, with the Rijeka refinery having its CO_2 in a ready-to-transport state. A consolidation hub at Rijeka should gather the CO_2 from the power plant and the refinery, before trunk transport along the natural gas magistral pipeline corridor to the west, i.e., to the storage sites in Central Croatia. In the first instance, the hydrocarbon fields would be the target for CO_2 injection, but since this cluster emits more than 2 Mt/y, it is likely that in the long run, storage in the DSA of Central Croatia would be considered. Alternatively, the hydrocarbon fields in eastern Croatia with the largest capacity could be considered.

A scenario is also possible in which the CO_2 from this Adriatic cluster is connected via pipeline to the hubs in the Central cluster, either at the Zagreb hub or to Sisak hub and then trunk transported together to the injection sites in the hydrocarbon fields.

4.1. Quantification of the CO_2 enhanced oil recovery storage scenarios

CO_2-EOR injection predictions for the period from 2025 to 2040 were modelled for all selected reservoirs. However, the moment when EOR project starts is very important and the additional recovery and CO_2 utilization factor are not optimized, because all CO_2-EOR is simulated from the same year (2025). In other words, the most favourable moment for the start of the EOR project was not individually selected, but this approach makes further analysis easier and uniform.

The CO_2 utilisation estimates are based on the selected scenarios. Field oil production and both injected and produced quantities of CO_2 vary from year to year, so that minimum, maximum and average values are indicated in the (Table 4). Note that the amount of CO_2 required on average varies from 0.28 Mt/y for the Lipovljani oil field to 8.5 Mt/y for the Beničanci oil field. Any combination of two or three fields being exploited simultaneously would be enough to store all the CO_2 that is currently being produced in the Central and Eastern clusters.

On average, 40% to 60% of the injected CO_2 is produced back and emitted to the atmosphere, resulting in net reductions that range, on average, from 0.2 Mt/y for the smallest fields to 3.2 Mt/y for the largest fields. Still, the overall net reduction would be around 40%. In CO_2-EOR processes the re-emission of CO_2 could be prevented if the produced CO_2 is separated from the natural gas stream and then re-injected. That option would imply a higher CO_2 avoidance in the CO_2-EOR process, but it would also require smaller volumes of CO_2 to be captured.

The numbers from the Table 4 represent the most efficient scenarios for each hydrocarbon field. Since the CO_2 emissions from the Central and Eastern cluster sum up to 3.36 Mt per year, or 5.5 Mt/y if Adriatic cluster is included, a need for an outsourced CO_2 supply is essential for the realization of the optimal utilization potential for each field. Potential streams of the outsourced CO_2 may come from the industries of EU members which are part of the EU emission trading system (EU ETS). It is worth noting that EOR is not currently recognised as an emissions avoidance or reduction option under the ETS or the Green Taxonomy, so emitters would not be eligible for credits under the ETS as it currently stands.

5. Conclusions

Considering the distances between CO_2 emitters and potential storage sites, including their capacities, clustering of those links in the CC(U)S chain is a logical step toward feasible and efficient emission mitigation system in Croatia. Commercialization of CO_2 injection projects highly depends on the CO_2 market development, and beside the CO_2 price, a push toward more intensive projects implementation would be strengthening of the capture, transport, utilization, and storage network.

Taking into consideration site availability (a year in which CCS can be fully activated) and capacity for CO_2 storage and summarising all the study results collected in the Strategy CCUS project, the CO_2-EOR option is evaluated as the most viable, focusing on CO_2 storage rather than additional oil recovery.

Three clusters have been identified as logical options: the Adriatic cluster, the Central cluster and the Eastern cluster. To achieve earlier (economic) feasibility, only the less costly onshore injection was chosen as an option.

Major conclusions drawn from this analysis:

- Clustering is possible in Croatia, however the distance between certain emitters exceeds 100 km (which was used as one criterion within Strategy CCUS).
- Capturing costs represent the biggest challenge for the CC(U)S commercialization, which implies that clustering would be beneficial from the cost reduction point of view.
- Grouping (clustering) of emitters would undoubtedly reduce total CC(U)S costs, hence data for cross-border emitters should be gathered and incorporated in future research.
- The considered clustering options include twelve CO2 emitters, transport routes, 14 well-characterized, nearly depleted onshore hydrocarbon fields where CO2 can be utilized and consequently stored, and two onshore deep saline aquifers with huge theoretical capacity but low level of characterization.
- Injection (storage) site availability is an important factor, and, despite controversial elucidations what should be considered as utilization, CO2-EOR, with CO2 retention (rather than oil production) as the main objective helps to bridge the period between availability of less investigated storage options (mainly DSA).
- Because of the level of research from the emitter side, close proximity to injection sites, the ability to use all three concepts (CO2-EOR, injection into depleted hydrocarbon fields, injection into one of two available DSA), and CO2 storage availability, the Eastern Cluster in Croatia is most likely the earliest cluster to be formed.

Table 4
Minimum, maximum and average CO_2 used (injected) and emitted, along with oil produced.

DHF	CO_2 used (kt/year)			CO_2 emitted (kt/year)			Oil produced (kt/year)		
	min	max	average	min	max	average	min	max	average
Beničanci	3 880	11 820	8 542	828	11 368	5 291	100	600	288
Ivanić	3 743	8 214	5 890	14	7 815	3 501	100	300	138
Kloštar	2 639	5 713	3 957	1 046	5 490	2 519	58	155	91
Lipovljani	151	433	275	0 23	197	80	155	155	155
Šandrovac	2 206	3 940	2 979	605	2 735	1 274	100	117	102
Stružec	4 373	7 683	5 892	911	7 403	3 593	100	400	250
Žutica	1 223	2 797	1 849	29	2 257	769	100	300	181

Considering low CO_2 emissions in Croatia on one side, and huge theoretical storage capacity on the other, possibilities arise for CO_2 imports from countries with emissions exceeding their own domestic storage capacities. Having said that, further research is needed, particularly more studies on the cost-effectiveness of various cluster scenarios, with consideration of level of research (primarily focused on CO_2) capture from the capture side, and even more detailed studies of dynamics of CO_2 injection into storage sites. It might be useful to compare the feasibility of converting a depleted hydrocarbon reservoir to a pure storage site after certain (enhanced) recovery is achieved considering that this recovery implies some extra pore space available for storage (which might encourage acceleration of clusters and infrastructure development) to CO_2 storage in deep saline aquifers which usually have huge theoretical capacity.

CRediT authorship contribution statement

Domagoj Vulin: Conceptualization, Methodology, Investigation, Visualization, Formal analysis, Validation, Supervision, Writing – original draft, Writing – review & editing. **Iva Kolenković Močilac:** Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing – original draft, Writing – review & editing. **Lucija Jukić:** Investigation, Visualization, Writing – review & editing. **Maja Arnaut:** Investigation, Formal analysis, Visualization, Writing – original draft. **Filip Vodopić:** Visualization, Writing – original draft. **Bruno Saftić:** Validation, Supervision. **Daria Karasalihović Sedlar:** Writing – review & editing. **Marko Cvetković:** Investigation, Formal analysis, Visualization, Validation, Writing – original draft.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

Data availability

Public data and previously published data was used, so the most of the paper has reproducible or visible data. The rest is available in references, primarily in Strategy CCUS deliverables/reports

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