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Life cycle and cost assessment of mineral carbonation for carbon capture and storage in European power generation

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ABSTRACT

Large-scale greenhouse gas (GHG) emission reductions are crucial for achieving the European goals for climate change mitigation. A frequently discussed option is carbon capture and storage (CCS), where CO_2 emissions from point sources are captured and stored in geologic structures. However, concerns about risks of leakages of CO_2 from geological storage have been raised. These risks could be avoided with ex situ mineral carbonation, where the captured CO_2 is stored in an inert and stable solid form after reacting with calcium and magnesium silicates. For a comprehensive assessment of the environmental and economic performance of this CO_2 storage option in fossil-fueled power generation chains, life cycle assessment (LCA) and levelized cost of electricity (LCoE) calculations are performed. The implementation of CCS using mineral carbonation leads to life cycle GHG emission reductions of 15–64% and LCoE increases of 90–370% on a per kWh_{el} basis compared to a reference power plant without CCS. The life cycle GHG emission reduction achievable with mineral sequestration is less substantial than with geological storage of CO_2 due to significant energy and chemical additives requirements. Accordingly, LCA results for other environmental indicators are worse than those of the reference plant without CCS and the geological CO_2 storage option.

1. Introduction

1.1. Motivation

A consistency of changes in physical and biological systems since 1970 is mentioned by the Intergovernmental Panel for Climate Change (IPCC, 2007b), which is directly connected to global temperature increases. In the same report it is stated that most of the observed increase in global average temperatures since the mid-20th century is very likely a result of the observed increase in anthropogenic greenhouse gas (GHG) emissions. The contribution of fossil fuel use for energy conversion to these GHG emissions is significant. Global GHG emissions need to be substantially reduced in order to mitigate global warming. The main options for emission reductions until 2050 include efficiency developments on generation and end-use consumption, increasing shares of renewable energies, fuel switching and development of carbon capture and storage (CCS) solutions (IEA, 2010).

Large point sources of carbon dioxide (CO_2), such as fossil fuel power plants, are ideal to implement the CCS technology. CCS includes three main stages: CO_2 capture from flue gas, transportation to the storage site and CO_2 storage (IPCC, 2005). Focusing on CO_2 storage considerable efforts are currently made in providing an efficient and sustainable mineral carbonation pathway.

Mineral carbonation is the reaction of CO_2 captured from fossil-fueled power plants with magnesium and calcium containing silicate minerals (e.g. wollastonite, serpentine, olivine) to form geologically stable and environmentally benign carbonate minerals. The clear advantage of this storage option is the stable, inert and solid form of the stored CO_2. Comparing the mineral sequestration with the geological storage pathway in deep saline formations or depleted gas and oil reservoirs, the zero risk of leakage of the stored CO_2 in a solid form is of great importance. By reducing the leakage risk, not only the public acceptance of CCS increases, but also insurance and financial risks are decreased. Potential disadvantages may derive from the technical complexity of mineral carbonation as well as additional environmental burdens and higher costs.

For a comprehensive comparative evaluation of fossil-fueled power generation with and without CCS both life cycle assessment (LCA) and levelized cost of electricity (LCoE) calculations are required. These assessments thus allow for the direct comparison of the mineral carbonation options, the geological storage option and the reference power plant without CCS.

1.2. Literature review

Mineral sequestration as a way to reduce CO_2 emissions from point sources such as power plants was first mentioned by Seifritz

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(1990). Since then numerous attempts were conducted for developing a complete and viable mineral carbonation process. In IPCC (2005) the main process development attempts are described. Based on this description the approach of the National Energy Technology Laboratory (NETL) in the United States is considered as one of the most comprehensive energy and economic evaluations for performing a single-step wet carbonation process (Gerdemann et al., 2004, 2007; Herzog, 2002; O'Connor et al., 2005) and was expanded by other research groups (Huijgen et al., 2006). Apart from the NETL process, a few more research attempts are also described in IPCC (2005). For instance, processes are described where strong acid (i.e. hydrochloric acid) or strong alkaline solutions (i.e. sodium hydroxide) are used to dissolve the natural minerals. Both cases are characterized by lack of experimental results and high energy intensity. Finally, a number of weaker acids (i.e. acetic, oxalic, orthophosphoric and ammonium bisulfate) are tested for reducing the energy requirements. However, a complete process scheme was only provided for using acetic acid by Kakizawa et al. (2001). But the process was only demonstrated for wollastonite and the experimental conversion levels have not exceeded 20%. Further developments in mineral carbonation after the year 2005 are described in Fagerlund (2012) and Torróntegui (2010). Most of this research was carried out by the Åbo Akademi University (ÅAU) in Finland. The research lead to a new carbonation process with some interesting findings and promising ideas (Fagerlund, 2012; Fagerlund et al., 2011; Nduagu et al., 2012b; Romão et al., 2012b; Zevenhoven and Fagerlund, 2011). Concluding, based on the literature reviewed, the most promising and best established mineral carbonation processes have been proposed by NETL and ÅAU. Both propose a complete carbonation process with sufficient carbonation efficiencies. The efficiency level is the most crucial point of the process for reducing the energy intensity and achieving a certain level of viability.

Performing an LCA for a large scale application of mineral carbonation is still a challenging task since there is no demonstration project available yet. Nevertheless, due to the importance of a LCA for this CCS option, especially in the development step, some studies have already been carried out (Khoo et al., 2011; Khoo and Tan, 2006; Kirchofer et al., 2012; Nduagu et al., 2012a). Khoo and Tan (2006) provide an LCA of mineral carbonation with the NETL process, focusing on the energy intensity of the process. The functional unit is defined as 1 MWh_{el} generated by a coal power plant, which provides also the required energy to the CO_2 mineralization processes. Wollastonite, olivine and serpentine are assumed to be the carbonation minerals. Khoo et al. (2011) evaluate the ÅAU process with serpentine as the only carbonation mineral focusing on a test case for mineral extraction in Australia and sequestration of flue-gas derived CO_2 by a natural gas combined cycle (NGCC) power plant in Singapore. The functional unit is again defined as 1 MWh_{el} generated by the NGCC, which also provides the heat and electricity requirements for the mineral carbonation. Results are provided for life cycle CO_2 emissions and costs, but other environmental burdens are not considered. Nduagu et al. (2012a) provide one of the most complete LCA on the topic by comparing both NETL and AAU processes but only on the serpentine basis. The functional unit is 1 ton of CO_2 stored in serpentine rock, i.e. there is neither consideration of the power plant, nor of the CO_2 capture and transportation units. For the first time the solvent recirculation is mentioned as a crucial parameter, and the electricity and heat requirements of the carbonation processes are defined on an exergy basis. Another detailed LCA on mineral carbonation is provided by Kirchofer et al. (2012), where the main focus is on comparing and optimizing the main trade-offs of the carbonation reaction based on the LCA results. The results presented are based on the NETL process using both natural minerals such as olivine and serpentine and industrial residues. Heat and power needed for the process are assumed to be provided by burning natural gas.

A few cost results are also available for power generation with mineral carbonation as a CO_2 storage option. Gerdemann et al. (2007) show that if the energy requirements of the mineral plant are covered by the coal-fired power plants generating the CO_2, the carbonation costs are between \$80 and \$120 per ton of CO_2 avoided for the olivine and the wollastonite cases respectively. For antigorite and lizardite serpentine cases they amount to roughly \$300 and \$500 per ton of CO_2 avoided. In addition, the IEA Greenhouse Gas R&D Programme (Newall et al., 2000) estimates the costs of mineral sequestration processes to be \$60–100 per ton of CO_2 sequestered where CO_2 capture and transportation costs are excluded. Finally, Khoo et al. (2011) estimate the costs for mineral carbonation for different serpentine scenarios between \$106 and \$180 per ton of CO_2 avoided.

1.3. Objectives of this study

Main goal of this study is to carry out an LCA of fossil power generation with CCS focusing on the mineral carbonation CO_2 storage pathway. In addition, a cost assessment is performed in a consistent way with the LCA on the basis of the levelized costs for producing 1 MWh_{el}. This allows for a consistent evaluation of both the environmental and economic performance of mineral carbonation processes as an option for permanent storage of CO_2. The study is carried out for the geographic region of Europe with the reference year 2025 as no mature technologies are expected to be available before. Since the most detailed research that can be found in literature is conducted based on the methods proposed either by the National Energy Technology Laboratory (NETL) or by the Åbo Akademi University (ÅAU), this study focuses on those two processes. The two mineral sequestration processes are examined in detail for all the carbonation minerals with research data available. The main uncertainties still open to research are examined in a sensitivity analysis, since they can be decisive for the final assessment of the processes. Results are provided on the basis of generating 1 MWh_{el} either by a hard coal or a NGCC power plant with post-combustion CO_2 capture and subsequent mineral carbonation.

The important questions to be answered are:

i. What are the environmental burdens of power generation with CCS based on mineral carbonation?
ii. What are the costs of power generation with CCS based on mineral carbonation?
iii. How do the environmental and economic performance of mineral carbonation compare to a non-CCS case and a geological CO_2 storage pathway?
iv. Which of the mineral carbonation options/processes layouts seem to be most promising from the environmental and economic perspectives?

2. Methodology and data used in the study

2.1. Technology characterization

CO_2 mineralization is a process that occurs also in nature. By weathering, rocks are breaking up into smaller fractions that more easily react with (mildly acidic) CO_2 saturated rain water. As a result, elements such as calcium and magnesium can react with the dissolved CO_2 to form solid carbonates. This reaction is not only thermodynamically favorable, but also exothermic at atmospheric conditions. The process is very slow and a variety of methods have been mentioned to accelerate the rate of reaction along with the CO_2 uptake rate of various minerals. As described by Fagerlund

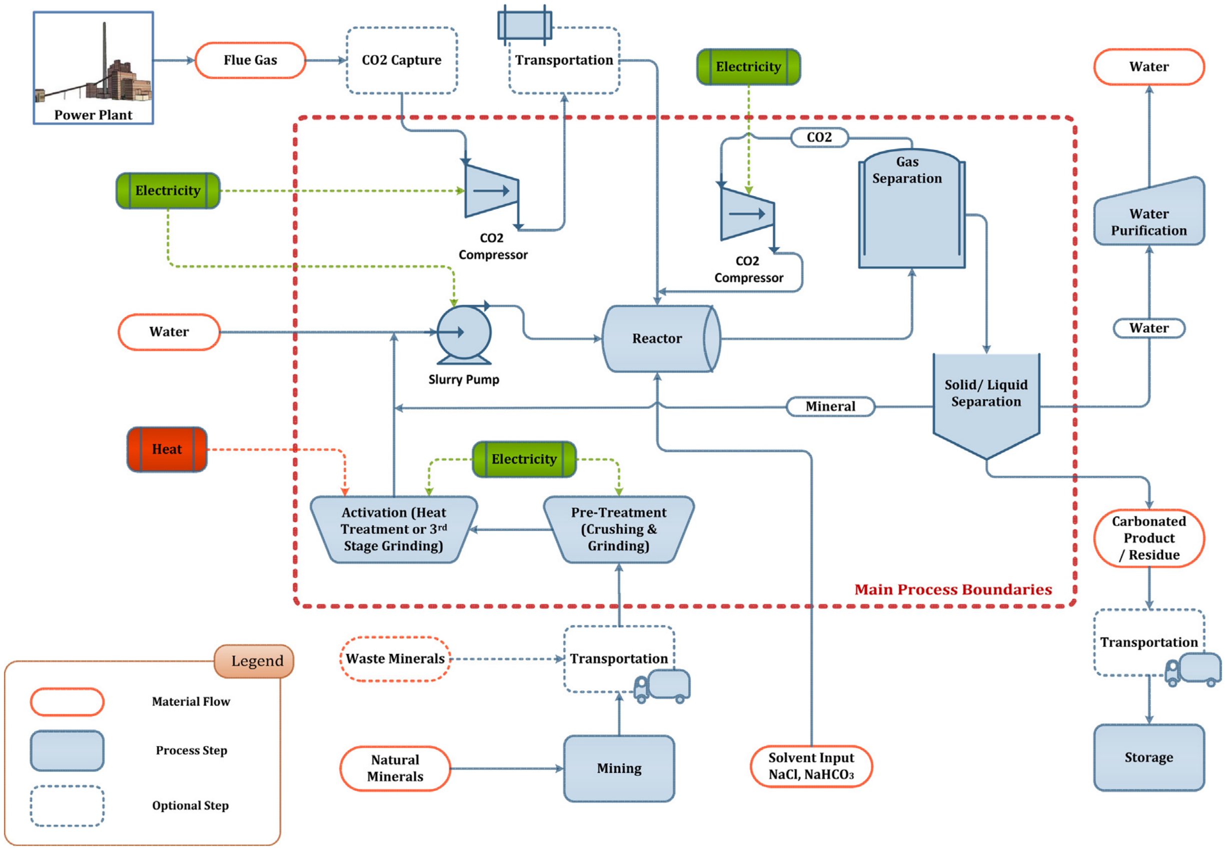

Fig. 1. Scheme of the process model considered for the LCA based on the National Energy Technology Laboratory (NETL) process, adapted from Gerdemann et al. (2007), Huijgen et al. (2006).

(2012), the main ex situ mineral sequestration options can be categorized according to:

i. Direct and indirect (multi-step) carbonation
ii. Aqueous and gas–solid process

According to the categorization above, NETL describes a direct aqueous process, whereas ÅAU investigates an indirect gas–solid one.

2.1.1. *Mineral carbonation based on the NETL process*

A general scheme of the NETL process as well as upstream and downstream processes as considered in the LCA is given in Fig. 1 where all the main material and energy flows are shown along with the boundaries between the carbonation process and the rest of the system. The electricity and heat consumptions are taken into consideration according to the available literature.

An efficient mineral carbonation reaction requires a high carbonation rate and re-use of the unreacted material. High efficiency can be achieved by crushing and grinding the mineral during pre-treatment steps, combined with increased pressure levels of CO_2 fed into the reactor. Experimental results on CO_2 mineral sequestration are available only for three different minerals: wollastonite, olivine and serpentine. Depending on the mineral used different amounts of electricity and heat are required for pre-treating and activating the material. According to Gerdemann et al. (2007), olivine and wollastonite achieve optimal reaction rates after an activation step of ultrafine grinding (3rd stage grinding). On the other hand, serpentine is activated by heat treatment. After the activation step the mineral is mixed with water (both recycled and fresh) and through a slurry pump the mixture is fed into the reactor.

Depending on the optimal CO_2 pressure level of the reaction, the CO_2 stream delivered by pipeline needs to be compressed further or not. Finally, the CO_2 stream is also fed into the reactor. The next steps after the carbonation reaction are the gas and solid/liquid separations. Through those steps the unreacted CO_2 and mineral on the one hand are recycled along with most of the water. On the other hand both, the carbonated product and the solid residue, leave the process to be transported to and stored at the mining site.

According to Gerdemann et al. (2007) the reaction rates for most minerals are considered to be low in distilled water. Based on the results from early tests, a buffered saline solution (0.64 M $NaHCO_3$ and 1 M NaCl)[1] became standard. The $NaHCO_3$ in the system serves the dual purpose of first modifying the solution to slightly alkaline pH which facilitates the carbonate precipitation and secondly it is acting as an effective CO_2 carrier. The only exception from the minerals examined is wollastonite which is the most reactive mineral and unlike with the magnesium minerals (olivine and serpentine) the reaction proceeds rapidly in distilled water.

2.1.2. *Mineral carbonation based on the ÅAU process*

By using recent research data (Fagerlund, 2012; Fagerlund et al., 2011; Nduagu et al., 2012b; Romão et al., 2012b; Zevenhoven and Fagerlund, 2011) the ÅAU process is modeled as a continuous process, which is shown in Fig. 2. Whereas the NETL process is achieving high reaction efficiencies for all three minerals (wollastonite, olivine and serpentine), the ÅAU process is currently only suited for serpentine (Zevenhoven et al., 2012).

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[1] M stands for the molarity, which is defined as the number of moles per 1 l of solution.

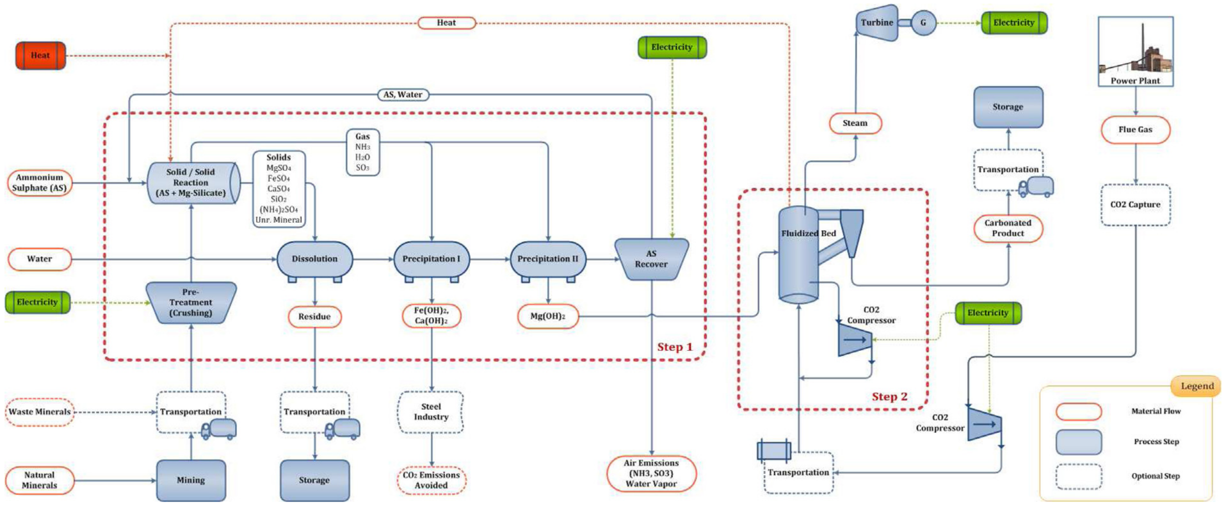

Fig. 2. Scheme of the process model considered for the LCA based on the Åbo Akademi University (ÅAU) process, adapted from Fagerlund (2012), Romão et al. (2012a), and Zevenhoven and Fagerlund (2011).

Compared to the one step aqueous NETL process, the ÅAU process is a multi-step process with two discrete main steps. In the first step the following processes are included:

1. Solid–solid extraction for reactive magnesium production from magnesium silicate mineral, using ammonium sulfate (AS) as solvent
2. Dissolution of reaction products
3. Precipitation (I) of calcium and iron in the form of hydroxides (OH)
4. Precipitation (II) of magnesium as magnesium hydroxide ($Mg(OH)_2$)
5. AS recovery

In the second step the $Mg(OH)_2$ reacts with gaseous CO_2 to form magnesium carbonate ($MgCO_3$) and water vapor in a pressurized fluidized bed reactor at 50 bar and 550 °C (Romão et al., 2012b). The main electricity consumptions of the process stem from the pre-treatment of mineral (crushing) and the recovery of AS. The power demand from external sources can be reduced by expanding the steam produced in the fluidized bed in a steam turbine for electricity generation Significant heat demand comes from the solid/solid Mg extraction step ($\Delta H_E = 234.6$ kJ/mol Mg extracted at 480 °C). On the other hand, a substantial amount of heat is produced in the $Mg(OH)_2$ carbonation step at the pressurized fluidized bed reactor ($\Delta H_C = -59.5$ kJ/mol Mg carbonated at 550 °C). The overall external heat requirements are calculated according to Eq. (1), where, X_E and X_C are defined as the Mg extraction and the $Mg(OH)_2$ carbonation efficiencies respectively (Zevenhoven et al., 2012):

$$Q\left(\frac{\mathrm{MJ}}{\mathrm{kg\ CO_2}}\right) = \frac{\Delta H_E}{X_E} - X_C \Delta H_C \qquad (1)$$

2.1.3. Mineral carbonation scenarios

For the NETL process, the main differentiation is based on the type of mineral used for carbonation. The available literature allows for an assessment of using three minerals (wollastonite, olivine and serpentine); therefore three different scenarios are defined (see Table 1). The olivine and serpentine scenarios are based on the optimal reaction parameters given by Gerdemann et al. (2007). The wollastonite scenario is based on the research results of Huijgen et al. (2006), who provide the most detailed study on wollastonite based on simulation results from Aspen Plus tool.

For the ÅAU process only use of serpentine is considered, since it is currently the only mineral reaching viable process efficiencies. Two additional scenarios are created, which differ in the heat requirement of the process (see Table 1). The first one can be described as a current (worst case) scenario, which assumes the currently achieved efficiencies of the process based on experimental data with heat requirements calculated from Eq. (1). The second scenario is based on increased efficiencies, which reflect the future optimization potential of the process according to Romão et al. (2012b).

Table 1
Mineral carbonation scenarios and optimal parameters overview.

Scenarios Mineral	NETL 1 Wollastonite	NETL 2 Olivine	NETL 3 Serpentine	ÅAU 1 Serpentine	ÅAU 2 Serpentine	Sources
Mineral on rock (wt%)[a]	84.3	90.0	83.0	83.0	83.0	Fagerlund (2012), Gerdemann et al. (2007), Huijgen et al. (2006)
$R(CO_2)$[b]	2.64	1.60	2.50	2.50	2.50	Gerdemann et al. (2007), Huijgen et al. (2006)
X_E (%)[c]	–	–	–	75	80	Romão et al. (2012b)
X_C (%)[c]	69	81	92	65	90	Romão et al. (2012b), Zevenhoven et al. (2012)
Mg extraction parameters						
P (bar)	–	–	–	1.013	1.013	Romão et al. (2012b)
T (°C)	–	–	–	480	480	Romão et al. (2012b)
Carbonation parameters						
PCO_2 (bar)	20	152	117	50	50	Gerdemann et al. (2007), Huijgen et al. (2006), Romão et al. (2012b)
T (°C)	200	185	155	550	550	Gerdemann et al. (2007), Huijgen et al. (2006), Romão et al. (2012b)
Carrier solution	Distilled water	$NaHCO_3$ and NaCl		Ammonium sulfate (AS)		Gerdemann et al. (2007), Huijgen et al. (2006), Romão et al. (2012b)

[a] $R(CO_2)$ is defined as the ratio of mineral ore needed per ton of CO_2 carbonated.
[b] $R(CO_2)$ is defined as the ratio of mineral ore needed per ton of CO_2 carbonated.
[c] X_E and X_C are defined as the Mg extraction efficiency (Step 1 in the ÅAU process) and the carbonation efficiency (NETL and Step 2 in the ÅAU process) respectively.

An overview of the scenarios described above is provided in Table 1 along with all the assumptions about the minerals, the reactions efficiencies and optimal parameters according to literature.

2.2. Life cycle assessment

Evaluating and comparing the environmental performance of mineral carbonation processes and – in more comprehensive terms – different electricity production requires the application of LCA methodology where all inputs and outputs are brought into one consistent framework wherever they occur during life cycles of products and services (ISO, 2006). LCA allows for consistent accounting of burdens to human health and the environment, crucial for the evaluation of new technologies like mineral carbonation and for the identification of trade-offs between potential impacts.

The functional unit is defined as 1 MWh_{el} at the busbar of the power plant, but results are also calculated on the basis of storing 1 ton of CO_2 enabling direct comparison with other LCA studies. The system boundaries encompass the entire energy chain including fuel extraction, processing and transport, power plant operation, CO_2 capture, compression, transport and storage, and waste disposal. Particularly for mineral carbonation as a CO_2 storage option mineral mining and processing, CO_2 compression and transport, electricity and heat supply, solvent feed-in, and residual and carbonated product disposal are considered.

ecoinvent v.2.2 (ecoinvent, 2012) is used as background life cycle inventory (LCI) database. LCI data of selected processes in the energy chains and in other economic sectors that are considered to be important in terms of contributions to cumulative LCA results are modeled according to developments that have been indicated by Blesl (2008), Bauer et al. (2012), EU (2012), and OECD/IEA (2009, 2010) for 2025; i.e. the electricity mixes are defined according to expected future scenarios, the natural gas and hard coal supply are adjusted according to the expected future shares of the producing countries, natural gas imports from the Middle East are newly included and leakage rates of long-distance natural gas pipelines for natural gas imports from Russia to Europe are reduced due to the improved quality of the pipelines. Metal production schemes and lorry transport emissions are redefined according to ESU-services/IFEU (2008).

Two different life cycle impact assessment (LCIA) methods are used for the evaluation of the environmental impacts of technologies: the life cycle GHG emissions are calculated using the IPCC GWP 100a method (IPCC, 2007a) version 1.02 as implemented in the LCA software SimaPro v7.3.3 (PréConsultants, 2011). Further environmental burdens are evaluated using the ReCiPe (Europe Hierarchist) method (Goedkoop et al., 2008) version 1.07 as implemented in SimaPro v7.3.3.

2.2.1. LCI of power plants, CO_2 capture, CO_2 pipeline transport and geological storage

Based on the main CCS stages being CO_2 capture, transport and storage (IPCC, 2005), each life cycle phase is modeled as a combination of individual processes, which can be removed or modified so as to create every desired CCS scheme in order to allow for comparable results for different mineral carbonation options, geological storage options and non-CCS options.

In this study the CO_2 source is either a pulverized hard coal (PC) or a NGCC power plant equipped with a post-combustion CO_2 capture unit. The performance parameters of the power plants with and without CO_2 capture are based on (Volkart et al., 2013) according to the expected technological improvements until the base year 2025 (Supplementary data; Tables S1 and S2). The post-combustion CO_2 capture process is defined for chemical absorption using 30 weight-% monoethanolamine (MEA) and the CO_2 capture rate is assumed to be 90% for both fuels.

The transport of CO_2 by pipeline is modeled according to Wildbolz (2007) with a transport distance of 400 km between the CO_2 source and the potential geological storage site and a life time of 30 years for the pipeline. The CO_2 mass flow is calculated according to the specifications of the respective plants. Scenarios of multiple CO_2 sources connected through a CO_2 pipeline network are not considered.

The geological CO_2 storage option requires supercritical conditions. In order to reach these conditions, minimum depths of around 1000 m are needed (IPCC, 2005). Saline aquifers are assumed to have the largest storage potential worldwide (IPCC, 2005). According to Volkart (2011) and the example of Germany, a saline aquifer at 1000 m depth is chosen as the CO_2 storage medium for geological storage.

2.2.2. LCI of the NETL process

The most important parameter in the LCI datasets for the different mineral sequestration scenarios is the energy consumption, i.e. heat and electricity demand. These demands are consistently specified for each carbonation scenario, either according to literature or calculations based on the schemes presented for the two processes.

As far as the NETL process is concerned all the pre-treatment and activation energy requirements in this assessment correspond to data from Gerdemann et al. (2007), Huijgen et al. (2006), Khoo and Tan (2006). The electricity consumption of the slurry pump is calculated according to the handbook published by Warman (2009) and that of the CO_2 compression is calculated based on McCollum and Ogden (2006). CO_2 remains in the gas phase between 1 bar and its critical pressure (73.8 bar), i.e. compression is required. Above 73.8 bar CO_2 is in the liquid/dense phase, i.e. a pump is used or further pressure increase. As a result the calculations are based on the electricity requirements of a 5-stage compressor and a pump. It is also assumed that the unreacted CO_2 from the carbonation reaction is fed back to the compressor/pump in order to react with the mineral. Accordingly, the electricity consumption is increased based on the reaction efficiency $X_{C.}$ No considerations are made for the post-reaction treatments such as the gas and solid/liquid separations assuming that their contribution is not significant on the total energy burden of the process. Finally, the heat required for increasing the water temperature in the NETL process is assumed to be balanced with the heat produced by the carbonation reaction based on the (Huijgen et al., 2006) results.

Use of solvent and potential re-use requires special attention in the LCA. The NETL process is modeled based on (Huijgen et al., 2006) with a liquid/solid ratio equal to 5 ton/ton and a fresh water feed-in stream equal to 1 ton of water per ton of $CO_{2\ \text{stored}}$. Nduagu et al. (2012a) translates the solution requirements as mentioned by Gerdemann et al. (2007) to 146.8 kg $NaHCO_3$ and 161 kg NaCl per ton of $CO_{2\ \text{stored}}$. Nduagu et al. (2012a) mentions that the solvents used in the process are not recovered, but it seems natural that up to a certain level the solvents need to be recovered for both environmental[2] (Farag and Harper, 2012) and economic reasons. Sensitivity analysis is carried out for three different cases where the losses are assumed to be equal to 0%, 50% and 100% of the solvent, respectively. The LCI dataset of NaCl production can be found in (ecoinvent, 2012) and the one of $NaHCO_3$ is created by modifying the ecoinvent sodium carbonate (Na_2CO_3) production dataset through mass allocation and by removing the final calcination step of the Solvay process described in the dataset.

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[2] Salinity increases of water disposed of will influence the aquatic life and $NaHCO_3$ can be fatal for the fish populations of rivers even in low concentrations according to recent findings by Farag and Harper (2012).

All the main LCI data and their references used for the assessment of the NETL mineral carbonation process are summarized in Table 2.

2.2.3. *LCI of the AAU process*

As pre-treatment stage in the ÅAU process the mineral crushing to diameters of 75 μm based on Nduagu et al. (2012a) is considered. From the same source it is clear that the main electricity consumption requirement stems from the AS recovery stage (equal to 290 kWh/ton $CO_{2\ stored}$). Due to the steam produced in the fluidized bed and expanded in a steam turbine, these demands can be reduced by 79 kWh/ton $CO_{2\ stored}$. The CO_2 compression requirements are calculated in the same way as for the NETL process. Focusing on the most important parameter of the ÅAU process, the heat consumption, the first scenario is calculated based on Eq. (1), so the heat demand is based only on thermodynamic and stoichiometric calculations and is equal to 6.23 MJ/kg $CO_{2\ stored}$. In the second scenario, based on the (Romão et al., 2012b) process modeling and optimal design of heat exchangers, the heat requirement is reduced to 2.22 MJ/kg $CO_{2\ stored}$.

The use of solvent in the ÅAU process is modeled according to Romão et al. (2012a) with an AS/serpentine/water ratio equal to 3:2:1. If 1% loss is assumed in the AS recovery step, a make-up stream of 400 kg of AS is needed for every 45 ton of AS processed (Romão et al., 2012b). So based on stoichiometric calculations for the reaction equations, a feed-in stream equal to 43.7 kg AS per ton of $CO_{2\ stored}$ is needed. If the AS was lost completely through air emissions, then the emissions would be equal to 11.27 kg NH_3 and 26.48 kg SO_3 per ton of $CO_{2\ stored}$. Especially for NH_3 emissions, this value would violate all the legal emission limits found in the EU countries (LDK-ECO, 2004). So NH_3 emissions have to be reduced at least to the limits imposed by the EU countries. In this study emissions of 12 kg NH_3 to air/per hour are assumed, what is equal to Ireland's emission regulations whose regulations are considered as representative for Europe. As a result, the NH_3 emissions to air are calculated as 28.96 g NH_3 per ton of $CO_{2\ stored}$ for hard coal and to 103.10 g NH_3 per ton of $CO_{2\ stored}$ for the NGCC power plant in this study.[3] The remaining NH_3 apart from the air emissions is assumed to be emitted as solid residues and as part of waste water. A reduction of the air emissions would is possible through flue gas treatment. The final decision on how to keep the air emissions below the limitations will be a matter of cost minimization.

All the main LCI data and their references used for the assessment of the AAU mineral carbonation process are summarized in Table 2.

2.2.4. *LCI of mining and backfilling*

The location of the mineral carbonation unit in relation to the power plant and the mining site is of crucial importance for the definition of the mode of transportation that is needed. Significant amounts of mineral rock need to be transported in order to operate a continuous and large scale CO_2 mineral sequestration plant. It was expected that it would be more preferable to transport the compressed CO_2 stream to the mineral carbonation plant, which would be placed next to the mineral mine. In order to validate this assumption a sensitivity analysis was performed on the basis of transporting either the natural minerals or the CO_2 stream. As displayed in the supplementary information (Fig. S1) the GHG emissions deriving from transporting the mineral rocks are much higher than those from transporting the pure CO_2 stream with an onshore pipeline. Thus the abovementioned assumption was confirmed.

Table 2
Summary of the LCI data for the different mineral sequestration scenarios.

Scenarios Mineral	NETL 1 Wollastonite	NETL 2 Olivine	NETL 3 Serpentine	ÅAU 1 Serpentine	ÅAU 2 Serpentine	Sources
Electricity requirements						
Pre-treatment (kWh/ton $CO_{2\ stored}$)	253	320	180	38	38	Gerdemann et al. (2007), Huijgen et al. (2006), Khoo and Tan (2006), Nduagu et al. (2012a)
Activation (kWh/ton $CO_{2\ stored}$)		333	–	–	–	Gerdemann et al. (2007), Huijgen et al. (2006), Khoo and Tan (2006), Nduagu et al. (2012a)
Slurry Pump (kWh/ton $CO_{2\ stored}$)	33	69	66	–	–	Warman (2009)
CO_2 compression (kWh/ton $CO_{2\ stored}$)	151	144	123	170	170	McCollum and Ogden (2006)
Solvent recovery (kWh/ton $CO_{2\ stored}$)	–	–	–	290	290	Nduagu et al. (2012a)
Steam expansion (kWh/ton $CO_{2\ stored}$)	–	–	–	−79	−79	Nduagu et al. (2012a)
Total (kWh/ton $CO_{2\ stored}$)	437	866	369	419	419	
Heat requirements (MJ/ton $CO_{2\ stored}$)	0.17	–	2.98	6.23	2.22	Huijgen et al. (2006), Khoo and Tan (2006), Romão et al. (2012b)
Mineral mining (ton/ton $CO_{2\ stored}$)	3.13	1.78	3.10	3.10	3.10	Own calculation and (Romão et al., 2012b)
$NaHCO_3$ input (ton/ton $CO_{2\ stored}$)	–	0.1468[a]	0.1468[a]	–	–	Nduagu et al. (2012a)
NaCl input (ton/ton $CO_{2\ stored}$)	–	0.161[a]	0.161[a]	–	–	Nduagu et al. (2012a)
AS input (ton/ton $CO_{2\ stored}$)	–	–	–	0.0437	0.0437	Romão et al. (2012b)
Infrastructure and equipment						
Cement plant (p/ton $CO_{2\ stored}$)	7.64E−08/1.30E−07[b]					Own calculation
Hard coal power plant (p/ton $CO_{2\ stored}$)	–	–	–	4.57E−10/7.31E−10[b]		Own calculation

[a] The values given in the table about the $NaHCO_3$ and the NaCl inputs are for 100% solvent losses in the NETL process.

[b] Values are given for the hard coal and the NGCC power plant respectively.

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[3] Assuming a hard coal PC power plant with a net electric capacity of 576 MW_{el} and a CO_2 capture ratio equals 0.719 ton/MWh_{el}, and a NGCC power plant with a net electric capacity equal to 366 MW_{el} and a CO_2 capture ratio equals 0.318 ton/MWh_{el}.

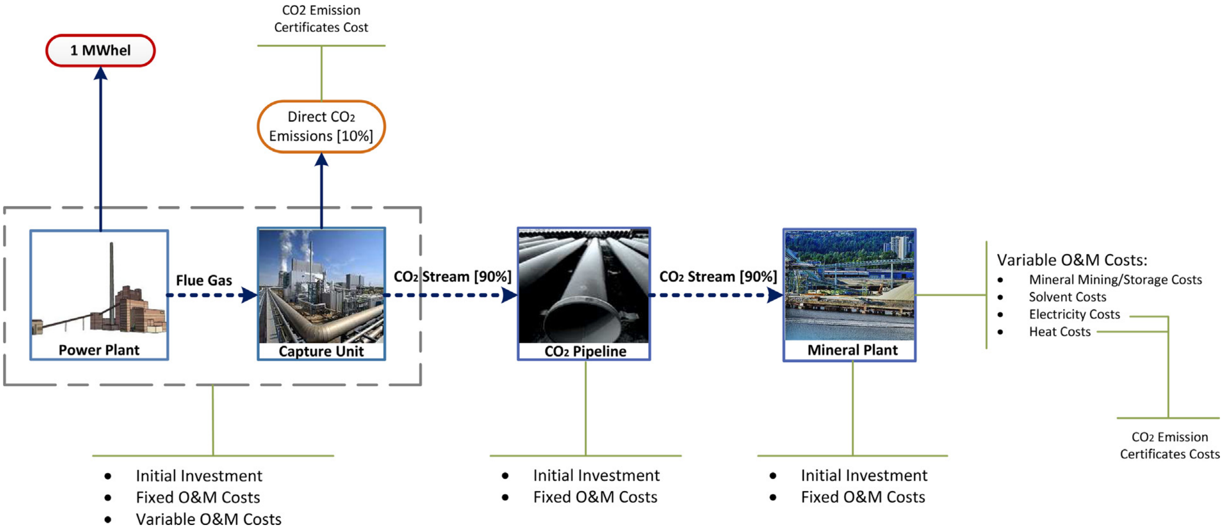


Fig. 3. Concept of the LCoE calculations for generating 1 MWh_{el}.

The LCI data of mining is based on the assumption that gold and iron ore mining can be considered representative for the mining of olivine-rich rocks and the other minerals (Hangx and Spiers, 2009). This assumption is based on the fact that gold and iron ore are part of (ultra)mafic rocks or materials and thus have a similar hardness than the olivine-rich rocks assessed here. The energy requirements, infrastructure and emissions are considered as those given for iron mining (iron ore, 46% Fe) in the ecoinvent database for open pit mining with global average data (ecoinvent, 2012).

All solid materials produced during the carbonation process (carbonated and residue product) are assumed to be stored as backfill at the mining site. Modeling the storage of the carbonated product and the solid residues of the two processes is based on the mining dataset: The energy requirements for storage of carbonated solids back in the mine are estimated to be reduced by 50% compared to the mining process according to Kirchofer et al. (2012). Furthermore, 20% of land occupation, machinery and infrastructure usage of the mining process are considered for the backfilling. Emissions of particulates with a diameter of more than 10 μm are considered to be reduced by 50% in line with the energy consumption. Emissions of smaller particulates are not considered, since specific dust emissions from mining, i.e. from blasting and crushing, are not present during storage. The particulate emissions from material storage, truck loading and dumping are of importance, but they are expected to be of greater size (i.e. >10 μm). In addition to that, the diameter of the carbonated product particulates is rather a design parameter of the carbonation process than a fixed characteristic. In order to minimize potential environmental burdens it is thus expected that additional particulate emissions smaller than 10 μm (representing a burden to human health) will be avoided. The mass calculations are based on both the stoichiometry of Mg extraction (only for the ÅAU process) and the carbonation reactions, taking into account also the ratio $R(CO_2)$, the reaction efficiencies X_E and X_C and the mineral purity of the rock.

All the main LCI data for mining and backfilling are summarized in Table 2.

2.2.5. LCI of the mineral carbonation plant

Since the mineral carbonation plant is assumed to be next to the mining site and not close to the power unit, all the energy requirements for carbonation are assumed to be provided externally. All the heat demands of the process are covered by an industrial furnace which burns either hard coal or natural gas according to the fuel used in the power plant. In addition, the electricity requirements are covered by the grid for which a European mix for 2025 is considered based on Blesl (2008).

Based on the CO_2 mass processed at the mineral carbonation plant also the infrastructure and equipment of the plant are scaled. The mineral plant used in the NETL process is assumed to have the same infrastructure and land occupation as a cement production plant (ecoinvent, 2012). Considering the ÅAU process some additional infrastructure assumptions should be made for the fluidized bed used in the process. So an additional dataset is added, i.e. the one of a hard coal power plant, where the magnitude of that infrastructure is scaled based on the power production of the steam turbine that is followed by the fluidized bed.

All the main LCI data and their sources used for the assessment of the mineral carbonation process are summarized in Table 2.

2.3. Cost assessment

For the assessment of CCS in power generation in Europe in 2025 also cost calculations on the basis of LCoE are performed. Consistent with the LCA, the same technological changes are assumed for power generation efficiencies with and without CCS as stated in Volkart et al. (2013) and for the 2025 European electricity mix (Blesl, 2008). In addition, all the fuel, heat and electricity costs are projected for 2025.

The reference monetary year is 2010 and the reference currency is Euro (€). The discount rate is considered equal to 6% and the average interbank exchange rate for 2010 between Euro and American dollar is given by OANDA (2012) to be equal to 0.755 €/$. In addition the average 2010 electricity price for industrial consumers in the Euro area is taken from Eurostat (2012) being equal to 109 €/MWh_{el}. An overview of the CCS chain units considered for the calculations of the LCoE is shown in Fig. 3. Thus the modeling and specification of the process chains considered for cost assessment are performed in consistency with LCA.

As shown in Fig. 3 the cost calculations are split between the power plant, the CO_2 capture unit, the CO_2 pipeline and the mineral carbonation plant. In addition, different cost scenarios are assumed for the CO_2 emission allowances and the fuel prices according to ZEP (2011). Cost calculations for the power plant, the CO_2 capture unit as well as geological CO_2 storage are based on the reference plants in Volkart et al. (2013) and on cost estimates in ZEP (2011). All the CO_2 pipeline cost calculations are derived from NETL (2010) for an above ground

Table 3
Summary of the variable O&M Costs of the mineral carbonation plant.

			Sources
Mineral mining costs (€/ton)		6	Khoo et al. (2011), Schuiling and Tickell (2011)
Backfilling costs (€/ton)		3	Own assumption
European electricity mix (€/MWh$_{el}$)		143.5/147.3/154.7[a]	Own calculation and Eurostat (2012)
Heat (Industrial Furnace) (€/GJ)	Hard coal	4.8/6.7/10.3[a]	Own calculations and ecoinvent (2012), ZEP (2011)
	Natural gas	9.5/10.6/12.8[a]	
Solvent costs ($/ton)	AS	140	ICIS (2012)
	NaCl	45	
	$NaHCO_3$	460	

[a] The electricity and heat costs are given assuming a medium fuel price scenario for 2025 and the three different scenarios for CO_2 emissions allowances costs.

pipeline of a total length of 400 km. Different pipeline diameters are chosen according to the CO_2 mass flow requirements for either the hard coal PC or the NGCC power plant. Finally both the initial investment and the fixed Operation & Maintenance (O&M) costs for the mineral plant are derived from the O'Hara method cost estimation for processing plants (Mular et al., 2002).

All the infrastructure cost items are converted to a common basis by using the average 2010 Marshall & Swift M&S(M/M) equipment index (Zuwala, 2012). In order to account for the spreading of the costs over the construction time, all the overnight equipment costs are multiplied by the respective cost trajectory factor given in Eq. (2) to retrieve the present value. For the CO_2 pipeline the total construction time is considered as being two years and the costs are occurring to 50% in each year. The same construction time is considered also for the mineral carbonation plant, but the costs are split into 40% for the first and 60% for the second year as the more sophisticated equipment is assumed to be installed at the finalization of the plant.

$$\text{cost trajectory factor} = f_{\text{year-1}}{}^{*}(1+i)^{0.5} + f_{\text{year-2}}{}^{*}(1+i)^{1.5} + \cdots \quad (2)$$

$f_{\text{year-}x}$ is the share of year x on the total cost (%/a), i is the interest rate (%).

As it is assumed in the LCA, all the energy and material requirements of the mineral plant are covered externally and they are considered in this study as Variable O&M costs. Taking into account the EU Emission Trading System (ETS, 2012), a sensitivity analysis is performed on the influence of the future CO_2 emission costs on the LCoE. Three different emission cost scenarios are assumed equal to the ones used in the (ZEP, 2011) report, where 20, 40 and 80 € are assumed as the low, medium and high emission cost scenarios for emitting 1 ton of CO_2. The CO_2 emission costs are not only taken into consideration for the direct CO_2 emissions from the power plant, but also for the final electricity and heat price calculations for 2025. In the European electricity mix projection for 2025 also a small share of CCS technologies is considered (Supplementary information, Table S3).

The 2025 electricity price is calculated based on Eq. (3), where the 2010 price is projected to 2025 by assuming an annual increase of 1%. In addition, the costs of CO_2 emission allowances are added based on the CO_2 intensity of the electricity mix. But according to Reinaud (2007) the additional emission costs for producing 1 MWh of electrical energy do not imply that the power prices will increase also by that amount. In general, the more concentrated the market is, the smaller the total increase in prices is found. So overall prices will be higher under a monopolistic market in general, but the rise in prices due to the pass-through of allowance costs should be lower. Based on empirical studies on the topic (Reinaud, 2007), an average European pass-through ratio of 85% is considered reasonable for this study.

$$\begin{aligned} P_{\text{electricity}} = {} & \text{Electricity price projection}_{2025} \\ & + (CO_2 \text{ emissions allowances costs} \\ & \times CO_2 \text{ intensity electricity mix}_{2025} \times \text{Pass}_{\text{through}} \text{ ratio}) \end{aligned} \quad (3)$$

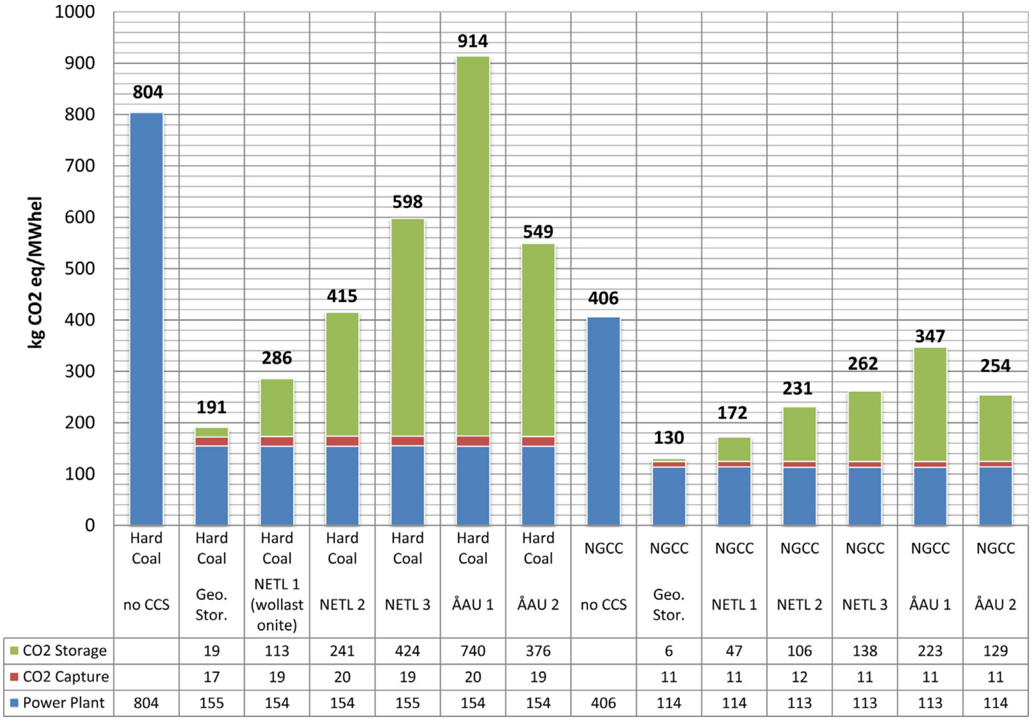
The future fuel prices are also considered in three different scenarios according to ZEP (2011). In detail those are defined as 2.0/2.4/2.9 €/GJ for hard coal and 4.5/8.0/11.0 €/GJ for natural gas. Based on those projections, the industrial furnace efficiencies, the CO_2 emission intensities (ecoinvent, 2012), the CO_2 emissions allowances cost scenarios and the heat costs are calculated. An overview of the electricity and heat prices along with the solvent and mining cost assumptions is given in Table 3. The costs for storing the carbonated product and the solid residues of the carbonation reaction are considered to be 50% of the mining costs in agreement with the assumption for energy requirement in the LCA.

Up to this point the cost considerations are in line with the LCA already performed. The mineral carbonation plant is treated as a unit separated from the power plant. This choice is based on preliminary LCA results suggesting environmental benefits of such a layout considering the estimated distance between the existing power plants and the mineral resources. From an environmental point of view transport of CO_2 through a pipeline is beneficial compared to transport of the substantial amounts of solid mineral. However, integration of both the carbonation plant and the power plant at one site might be advantageous from an economic point of view. In this case the electricity requirements of the carbonation plant are covered by the power plant itself. In order to perform a calculation for such a case in a consistent way with the functional unit of delivering 1 MWh$_{el}$ to the grid, the net electricity production and the amounts of CO_2 processed by the carbonation plant are adjusted taking into account both the process-specific internal power consumption of the mineral carbonation plant and the additional CO_2 emissions due to increasing fuel consumption for the same amount of net electricity generation. Since both the power and the carbonation plant are considered as one unit, costs of pipelines for CO_2 transport are omitted. The mineral resource is assumed to be available on site, i.e. no mineral transport is required.

3. Results

3.1. Life cycle GHG emissions

Since the main goal of assessing CCS technologies in fossil-fueled power generation is the reduction of life cycle GHG emissions, the initial comparison should be performed on that basis. The results are categorized in Fig. 4 as contributions from the power plant, CO_2 capture and CO_2 storage emissions. The power plant emissions also include the upstream phase, i.e. the fuel supply chain. While 90% of the CO_2 direct emissions are captured and stored in the CCS cases, the other emissions per MWh$_{el}$ are increased due to the CO_2



	Hard Coal no CCS	Hard Coal Geo. Stor.	Hard Coal NETL 1 (wollastonite)	Hard Coal NETL 2	Hard Coal NETL 3	Hard Coal ÅAU 1	Hard Coal ÅAU 2	NGCC no CCS	NGCC Geo. Stor.	NGCC NETL 1	NGCC NETL 2	NGCC NETL 3	NGCC ÅAU 1	NGCC ÅAU 2
🟩 CO2 Storage		19	113	241	424	740	376		6	47	106	138	223	129
🟥 CO2 Capture		17	19	20	19	20	19		11	11	12	11	11	11
🟦 Power Plant	804	155	154	154	155	154	154	406	114	114	113	113	113	114

Fig. 4. Life cycle GHG emissions for electricity production from hard coal PC and NGCC in Europe in 2025 with and without post-combustion CCS (with geological storage and mineral carbonation scenarios), differentiated by the life cycle phases using the IPCC GWP 2007 method [kg CO_2-eq/MWh_{el}].

capture-induced efficiency loss at the power plant and the resulting increased fossil fuel consumption. The contributions from the power plant and the CO_2 capture stay the same for all CCS options. The CO_2 storage includes – besides the GHG emissions deriving by mineral carbonation – the electricity requirements for compressing and transporting the CO_2. This allocation choice is based on the fact that in the mineral carbonation scenarios the compression of CO_2 is required not only for transportation but also for performing the carbonation reaction.

Life cycle GHG emission reductions vary significantly when comparing the geological storage (68–76%) and the mineral carbonation scenarios (15–64%). This difference can be explained mostly by the significant heat and electricity requirements of the mineral sequestration. Especially for the ÅAU 1 scenario, where the heat requirements are extensive since they are not optimized, the implementation of CCS results in even worse life cycle GHG emissions than the non-CCS case for a hard coal PC power plant.

Since the contribution of mineral sequestration is decisive for the overall result, an additional categorization is performed in Fig. 5 specifying different contributions to GHG emissions within the mineral carbonation step. Electricity and heat requirements cause the highest contributions. They include the pre-treatment and activation steps of the mineral, the operation of the slurry pump (NETL process only), the solvent recovery (ÅAU process only) and the CO_2 compression requirements. Significant contributions to the life cycle GHG emissions also derive from the solvent losses (i.e. demand) in the mineral carbonation processes. It should be noticed that in NETL process 100% solvent losses and in ÅAU process only 1% are assumed according to literature (Nduagu et al., 2012a; Romão et al., 2012b), but the contribution of the solvent feed-in stream to the life cycle GHG emission results is of the same magnitude for both processes. So this indicates a significantly higher burden for the solvent (AS) production used in the ÅAU process. The mining of mineral rock and the storage of the carbonated product/solid residue do not produce significant GHG emissions. Moreover, the waste water treatment[4] as well as the infrastructure used for the mineral carbonation plant and the CO_2 transport have small shares in the total GHG emissions.

According to the results presented, a classification of the three natural minerals according to their performance is possible: wollastonite seems to be the most suitable among all three with minimum energy consumptions and without any chemical additives requirements. Also the mineral carbonation scenario NETL 1 is the only process that is on a similar level as geological storage in terms of GHG emissions. The olivine scenario NETL 2 achieves the second best life cycle GHG emission results and the serpentine scenarios (NETL 3 and ÅAU 1/2) the worst.

3.2. *Other environmental impacts*

Out of the eighteen midpoint impact categories of the ReCiPe method, six have been selected for more detailed analysis: human toxicity (HT, in kg 1,4-DB-eq/MWh_{el}), particulate matter formation (PMF, in kg PM10-eq/MWh_{el}), terrestrial acidification (TA, in kg SO_2-eq/MWh_{el}), marine eutrophication (ME, in kg N-eq/MWh_{el}), urban land occupation (ULO, in m_2a/MWh_{el}) and photochemical oxidant formation (POF, in kg NMVOC MWh_{el}). The selection is motivated by the substantial effect of the implementation of mineral carbonation storage option in those impact categories as well as by recent literature on LCA of CCS implementation (Corsten et al., 2013; Modahl et al., 2012; Singh et al., 2011; Volkart et al., 2013; Zapp et al., 2012). Impacts on ecosystems and human health due to climate change are not further discussed, since these results are equivalent to the life cycle GHG emissions. LCA results for all midpoint impact categories can be found in the supplementary

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[4] Simple waste water treatment process according to ecoinvent (2012); energy is required for pumping and infrastructure in sewers and municipal waste water plants. It is considered only in the NETL process, according to Farag and Harper (2012).

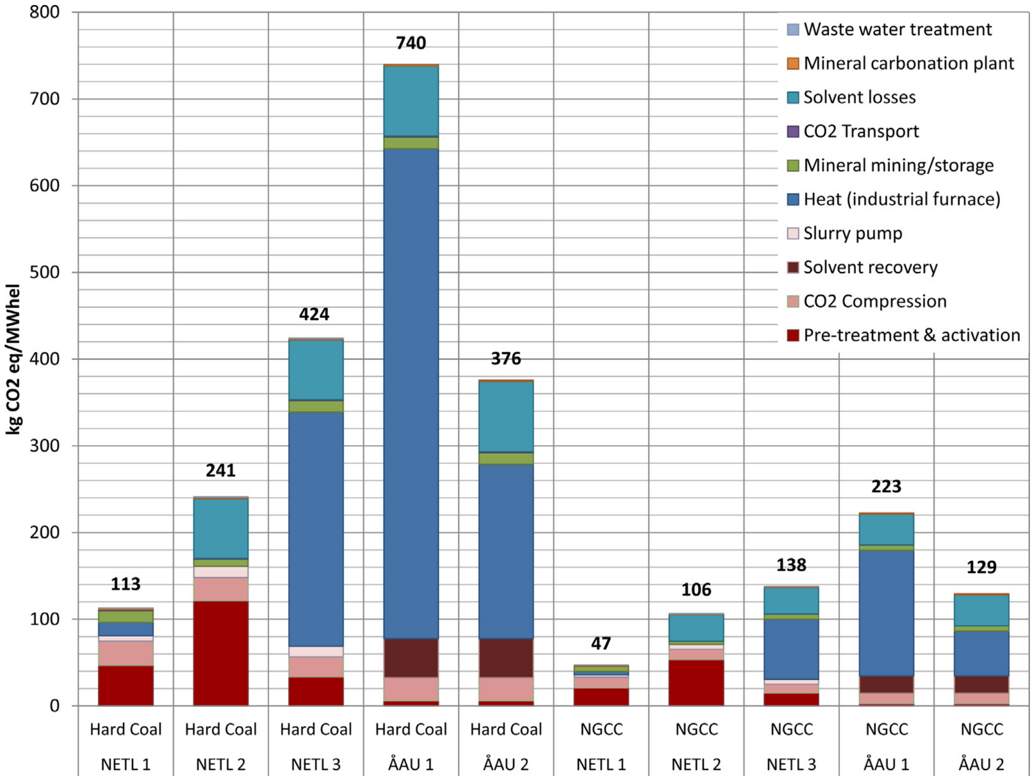

Fig. 5. Mineral carbonation contribution to life cycle GHG emissions for electricity production from hard coal PC and NGCC with post-combustion CO_2 capture in Europe in 2025, differentiated by the storage process phases using the IPCC GWP 2007 method [kg CO_2-eq/MWh_{el}].

information. The results for the various indicators are depicted relative to the maximum value determined for the respective midpoint impact category (Figs. 6 and 7).

Hard coal PC combustion power generation leads to significant impacts in the HT category, whereby NGCC power generation results in comparatively minor impacts. The HT burdens are dominated by the spoils and tailings from coal mining, the direct heavy metal emissions from operation and the emissions from the ash to be disposed of. HT is directly proportional to the amount of fuel burnt and therefore larger for electricity production with CCS. In addition, in cases where hard coal PC is burnt to provide the extensive heat requirements of the carbonation process (NETL 3,

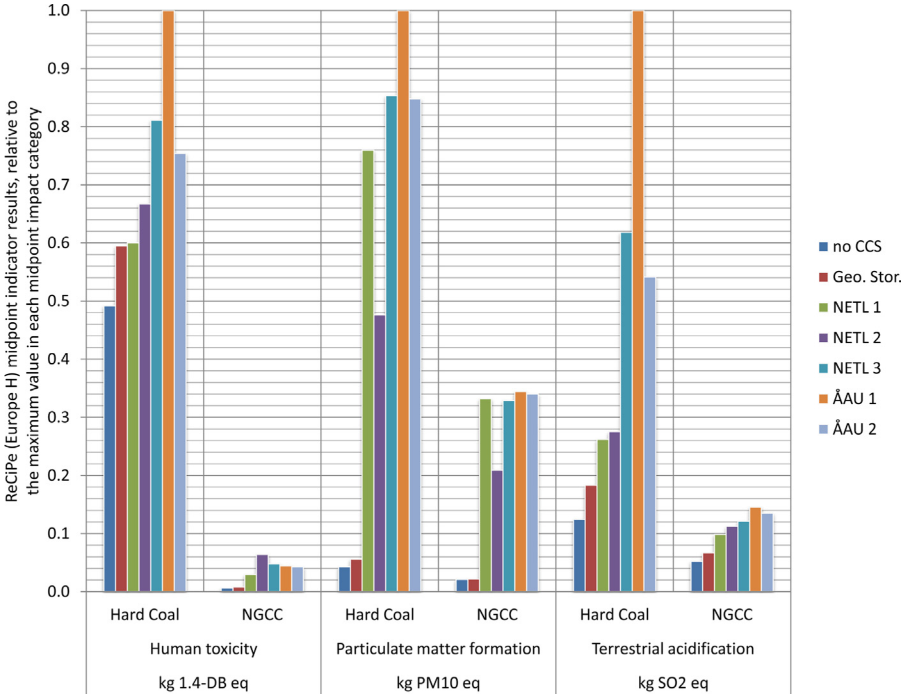

Fig. 6. Life cycle environmental burdens for electricity production from hard coal PC and NGCC in Europe in 2025, with and without post-combustion CO_2 capture with geological storage and mineral carbonation, respectively, differentiated by midpoint category using ReCiPe (Europe Hierarchist) method.

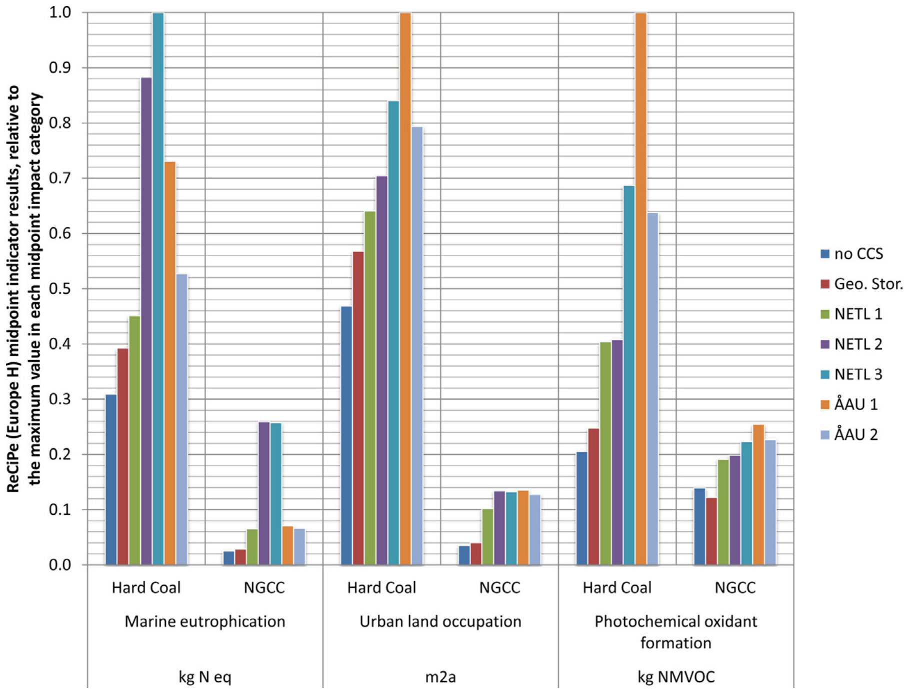

Fig. 7. Life cycle environmental burdens for electricity production from hard coal PC and NGCC in Europe in 2025, with and without post-combustion CO_2 capture with geological storage and mineral carbonation, respectively, differentiated by midpoint category using ReCiPe (Europe Hierarchist) method.

ÅAU 1&2), additional HT burdens are visible. Comparing the NGCC cases, the NETL 2 mineral carbonation process generates the highest HT impacts, caused by the significant electricity requirements for olivine grinding which is covered by a partially hard coal PC based electricity mix.

Significant increases in PMF can be noticed for all the mineral carbonation CCS scenarios in comparison with geological storage and non-CCS cases. Those derive mainly from direct and indirect sources: Firstly, the large amounts of mineral that need to be mined lead to significant direct PMF due to direct particulate emissions: i.e. the ratio of mineral ore needed per ton of CO_2 carbonated $R(CO_2)$, the purity of the mineral ore in the rock and the efficiency of the carbonation process are the key parameters concerning PMF. This clearly demonstrated comparing the wollastonite (NETL 1) and the olivine (NETL 2) scenarios, where olivine has lower $R(CO_2)$ requirements. Secondly, since heat is provided by an industrial furnace, in the hard coal PC cases the contribution to PMF from heat supply is important; this becomes obvious comparing the two ÅAU cases with and without optimized heat requirements and by comparing the hard coal PC and NGCC scenarios. Thirdly, for the ÅAU processes there are additional secondary PMF due to NH_3 emissions. Those emissions derive from the AS production (externally in chemical industry) and the carbonation process itself.

The TA impact category for hard coal PC scenarios is dominated by the direct NO_x and SO_2 emissions of hard coal PC combustion for both power and heat generation. The heat demanding serpentine scenarios (NETL 3, ÅAU 1&2) generate the highest TA impacts. The NGCC scenarios are influenced only by the formation of nitrogen oxides (NO_x) and NH_3 due to mineral mining (blasting) and thus have smaller increases in the TA burden.

The most important contributor in the ME impact category is the feed in of $NaHCO_3$ solvent in NETL scenarios 2 and 3. During the production of $NaHCO_3$, NH_3 emissions are dissolved in the water, which lead to important impacts for ME. Additional ME burdens derive from hard coal mining. Freshwater eutrophication impact results not presented here follow the exact same pattern found in the HT results.

Concerning ULO results once again the hard coal PC contribution for both power and heat generation is dominant due to increased hard coal PC mining. One the other hand, the NGCC mineral carbonation scenarios have increased ULO burdens deriving from multiple sources such as mineral mining, chemical plant for solvent production, mineral carbonation plant itself, industrial furnace infrastructure and infrastructure requirements for the electricity grid.

Finally, the POF impact results in PC cases are influenced mainly by NO_x and NMVOC emissions from hard coal PC combustion to cover the heat requirements of the mineral plant. Additional contribution derives from the share of PC plants in the European electricity mix. Burning natural gas for the same reasons gives lower POF results, making it more favorable also in this impact category. An additional contribution in this impact category derives from NO_x formation due to mineral mining (blasting).

3.3. Cost assessment

3.3.1. Cost results for a process scheme consistent with LCA (transport of CO_2 between power plant and mineral carbonation plant)

The cost results are provided for all the cases as described in ZEP (2011) for direct comparison. The main results are provided for average fuel cost scenarios and all three CO_2 emission cost scenarios. The cost results are categorized as follows (Fig. 8):

i. capital expenditures (CAPEX) for power plant and capture unit
ii. fixed & variable O&M costs for power plant and capture unit
iii. fuel costs for power generation and CO_2 capture
iv. total underground storage costs
v. CAPEX for CO_2 pipeline and mineral carbonation plant
vi. fixed O&M costs for CO_2 pipeline and mineral carbonation plant
vii. variable O&M costs for mineral carbonation plant
viii. CO_2 emission costs based on the CO_2 emission allowances price scenarios (20/40/80 €/ton CO_2 emitted).

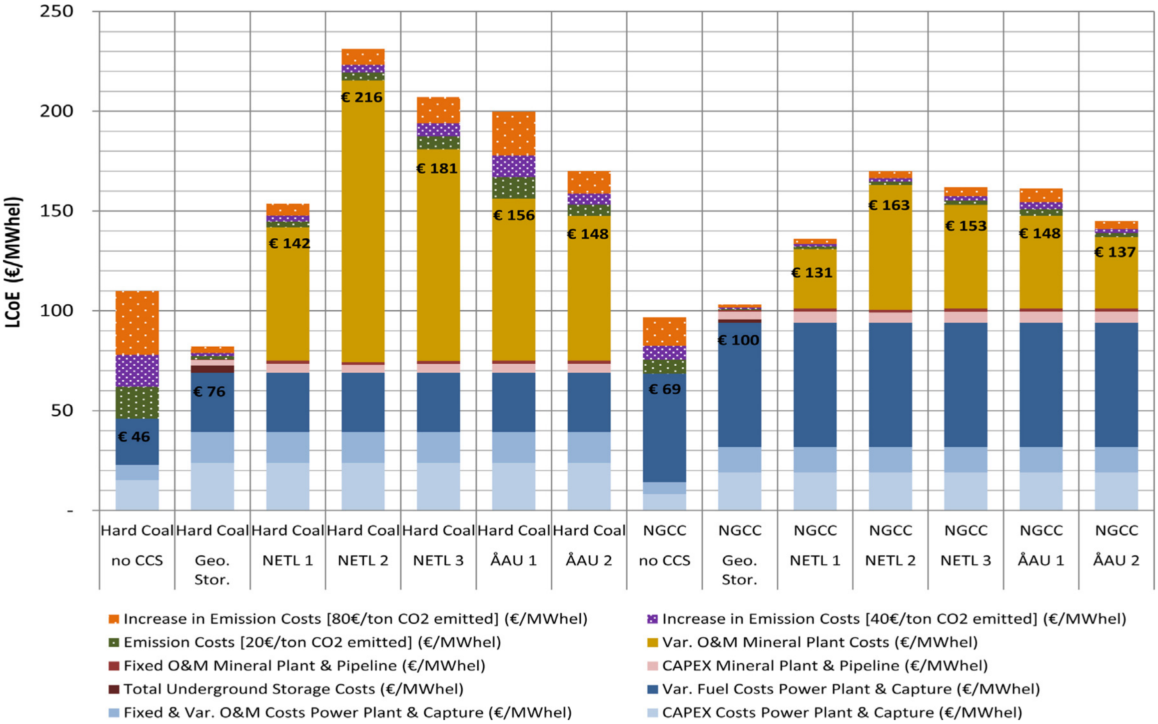

Fig. 8. Levelized cost of electricity (LCoE) from hard coal PC and NGCC in 2025 with and without post-combustion CO_2 capture with geological storage and mineral carbonation, respectively, differentiated by cost components. The numbers indicate the LCoE for the medium fuel price scenario and without CO_2 emission allowances costs.

The number indicated for each scenario in Fig. 8 includes all the costs described above except of CO_2 emission costs, in order to allow for a direct comparison of the cost increases for the CCS cases. All the mineral carbonation scenarios are less economic than the geological CO_2 storage one. If the CO_2 emission costs are not accounted for, starting from the non-CCS case for hard coal PC power generation and total power generation costs of 46 €/MWh$_{el}$, the costs increase to 76 €/MWh$_{el}$ for geological storage and between 142 and 216 €/MWH$_{el}$ for the mineral carbonation scenarios. Power generation from a NGCC power plant results in electricity production costs of 69 €/MWh$_{el}$ for the non-CCS case rising up to 100 €/MWh$_{el}$ for geological storage and between 131 and 163 €/MWh$_{el}$ for mineral sequestration storage.

The high costs for the mineral carbonation scenarios are mainly caused by the significant variable O&M costs of the carbonation plant: energy requirements (heat and electricity), mineral mining and the solvent additives represent the main cost drivers. Taking into account the EU Emissions Trading System (EU ETS) and a scenario of 80 €/ton of CO_2 emitted, the only CCS scenario that seems economically viable is the geological storage for power generation with a hard coal PC power plant. Mineral sequestration as CO_2 storage option still shows much higher costs and would not be competitive.

A more detailed comparison for each cost category shows the following: Adding the post-combustion CO_2 capture unit to the power plant causes considerable cost increases at both the CAPEX and the fixed O&M costs... Those derive from the fact that due to the implementation of the CO_2 capture unit the overall electric efficiency of the power plant drops and the fuel consumption increases per 1 MWh$_{el}$ produced. On the other hand, the CAPEX and the fixed O&M costs for geological storage, CO_2 pipeline and mineral carbonation plant do not substantially contribute; they are more significant for the NGCC cases, since they are levelized over a shorter life time period (25 years in comparison to 40 years assumed for the hard coal PC power plant). The high share of the variable O&M mineral carbonation plant costs is lower for the NGCC cases in comparison to the hard coal PC ones since the generation of 1 MWh$_{el}$ from NGCC leads to smaller amounts of CO_2 that need to be carbonated and stored. Finally, the CO_2 emission costs are also found to be higher in the mineral carbonation scenarios in comparison with geological storage due to heat and electricity consumptions which add up to direct CO_2 emissions and need to be included in a consistent cost analysis. Moreover, these emissions are more significant for the hard coal PC cases due to the increased direct emissions from burning hard coal PC to provide the heat requirements of the carbonation process.

3.3.2. *Results of an integrated "power plant – mineral carbonation" system*

The results presented in the previous chapter were consistent with the system consideration made in the LCA. As discussed in Section 2.3 it is of interest to analyze a test case where the mineral carbonation plant is not considered as a separate unit, but as an integrated CO_2 emissions reduction solution. In such a case, both the power and the carbonation plant are placed next to the mineral source, consequently no CO_2 pipeline is considered and the power demand of the mineral carbonation is covered by the power plant.

Comparing the results in Figs. 8 and 9 shows quite similar cost results for the carbonation cases, despite of the differences found in the contributions from each cost category. The integrated "power plant – mineral carbonation" system results in substantially higher costs of LCoE only for the NETL 2 hard coal case as a consequence of the high electricity consumption of this case. A more detailed analysis points out the following:

- CAPEX, Fixed & Variable O&M costs of the power plant and capture unit increase considerably. Integrating the carbonation plant with the power plant leads to a decreased net power output. In

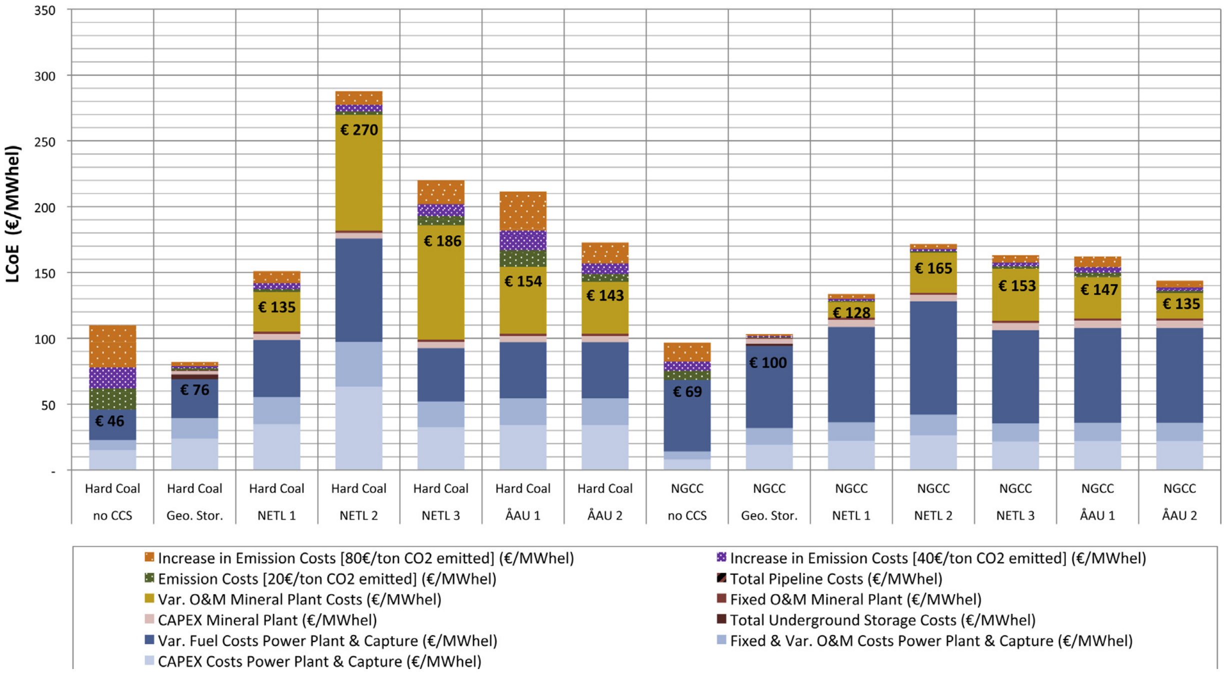

Fig. 9. Levelized cost of electricity (LCoE) from hard coal PC and NGCC in 2025 with and without post-combustion CO_2 capture with geological storage and mineral carbonation, respectively, differentiated by cost components. Results for an integrated "power plant – mineral carbonation system", i.e. CO_2 pipeline costs are excluded for all the carbonation cases. The numbers indicate the LCoE for the medium fuel price scenario and without CO_2 emission allowances costs.

addition, the CO_2 stream that needs to be captured from the flue gas, per 1 MWh_{el} delivered to the grid, increases.

- Due to the decreased net power output in the integrated approach, also the Variable fuel costs of the power plant and the capture unit increase substantially.
- A slight increase is noticed for CAPEX and Fixed O&M costs of the carbonation plant. Such an increase derives from the increased CO_2 that needs to be processed due to increasing fuel demand for a net power output of 1 MWh_{el}.
- On the other hand, the electricity costs for externally purchased power are no longer contributing to the Variable O&M costs of the carbonation plant, which balances in general the previously described cost increases.

3.4. Sensitivity analysis

In order to track down the parameters which are most crucial for the validity of the results a sensitivity analysis was performed. Sensitivity analysis is crucial due to the uncertainties that derive from using the experimental carbonation results found in literature for the assessment of a large-scale continuous process.

3.4.1. Life cycle GHG emission sensitivities

Since the life cycle GHG emission results of mineral carbonation (Fig. 5) indicate that the most significant contributors are heat, electricity and solvent consumptions, the focus is on these three parameters. The relative changes are presented compared to the ÅAU 2 (Figs. 10 and 12) and ÅAU 1 (Fig. 11) respectively. Those two processes are selected for sensitivity analysis since the level of uncertainty about their final heat, electricity and solvent requirements is considered significant.

It can be seen in Fig. 10 for the ÅAU 2 carbonation process that the amount of heat required and supplied by hard coal PC combustion is the most sensitive parameter for the GHG results among the three and therefore, this heat demand needs to be minimized. The results are less sensitive to the electricity and solvent consumption.

Further sensitivity analysis was performed on the heat requirements and their source so as to show their influence on the life cycle GHG emission results of the mineral carbonation CO_2 storage option. In Fig. 11 the worst case scenario (in terms of GHG emission results), which is the ÅAU 1 process CO_2 storage option with heat requirements covered by a hard coal PC industrial furnace, is defined as the reference case. If the optimized ÅAU 2 scenario is assumed with increased reaction efficiencies and optimal heat requirements, it is clear that the life cycle GHG emissions are reduced by almost 50% even by keeping the heat supply from hard coal PC. If the heat requirements are covered by a natural gas industrial furnace then further reductions of the total GHG emissions can be achieved (−12%). Heat could also be available as a by-product from other processes, e.g. waste heat from a waste incineration plant. This choice leads to additional GHG emission reductions of 13%. Finally, a scenario of combined heat and power (CHP) plant,[5] which provides all the heat and electricity requirements in the carbonation plant, is evaluated. This scenario in comparison with the second and third shows a reduction of the GHG emissions from heat consumption, but also a significant increase of those deriving from electricity consumption. The reason for this is the high exergy content of electricity which is taken as a basis for the allocation of the burdens of the CHP plant (exergy allocation).

3.4.2. LCoE sensitivities

In contradiction to the environmental results, the sensitivity analysis for the LCoE (Fig. 12) shows that the electricity required in the carbonation process(es) is more important than the heat and fuel. To make this result even more clear once more the ÅAU 2 process is selected for sensitivity analysis as it is highly heat and relatively low electricity intensive. Consequently, Fig. 12 shows that

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[5] ecoinvent (2012) dataset: "electricity, at mini CHP plant, allocation exergy, CH" is used to model the CHP plant which burns natural gas to cover the heat and electricity requirements of the mineral plant.

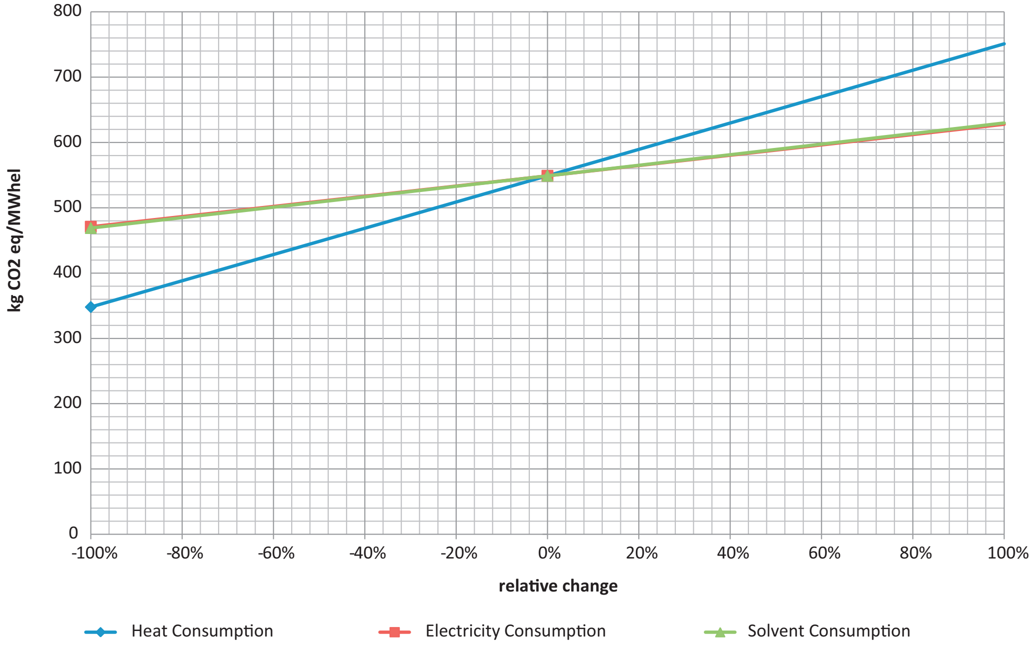

Fig. 10. Sensitivity analysis of the life cycle GHG emissions for electricity production from hard coal PC with CCS in Europe in 2025, based on the mineral carbonation demands of the ÅAU 2 process, using the IPCC GWP 2007 method [kg CO_2-eq/MWh_{el}].

the high costs for European electricity contribute the most. The effect of electricity costs can be seen also in the final results for LCoE in Fig. 8, where the NETL olivine option (NETL 2) with significant electricity requirements due to olivine grinding performs worst from an economic point of view. On the other hand, the solvent consumption is relatively insignificant from a cost perspective, which is in contrast to the significant environmental burdens of the solvent losses.

4. Discussion

Research on mineral sequestration as an option for CO_2 storage is of great interest for both reducing the GHG emissions of fossil fueled power generation and avoiding the CO_2 leakage risks that may occur in the CO_2 geological storage option. In order to compare it to other CO_2 mitigation options its environmental and economic performance must be evaluated. This

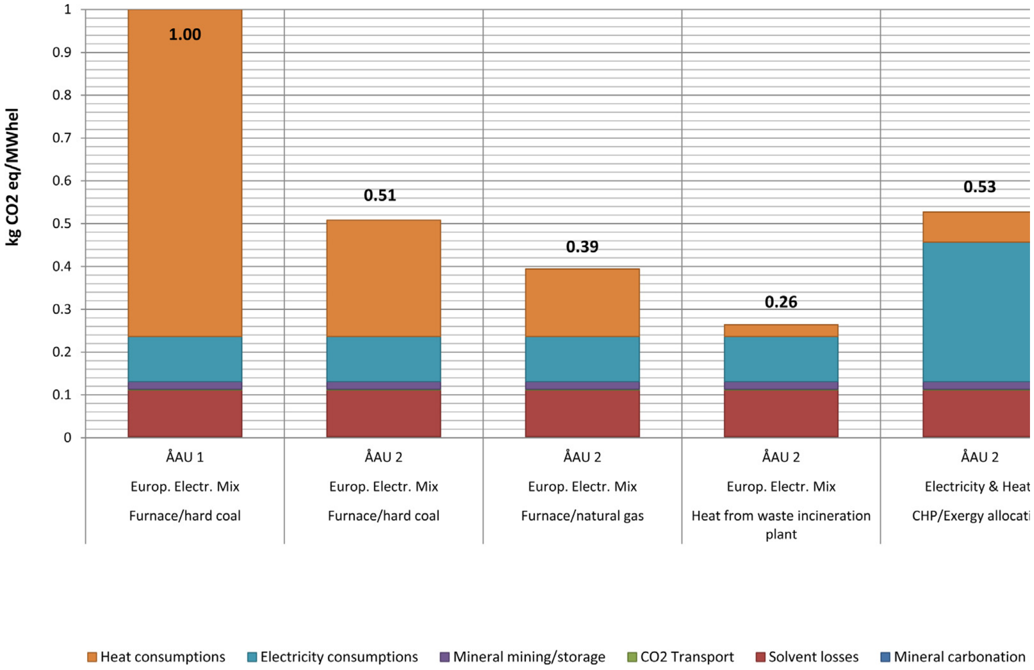

Fig. 11. Mineral carbonation contribution to life cycle GHG emissions of electricity production from hard coal PC with CCS in Europe in 2025 using different heat and electricity sources for the CO_2 mineral carbonation, differentiated by the life cycle phase using the IPCC GWP 2007 method [kg CO_2-eq/MWhel]. The results are presented relative to the ÅAU 1 scenario. CHP: combined heat and power generation, natural gas-fueled.

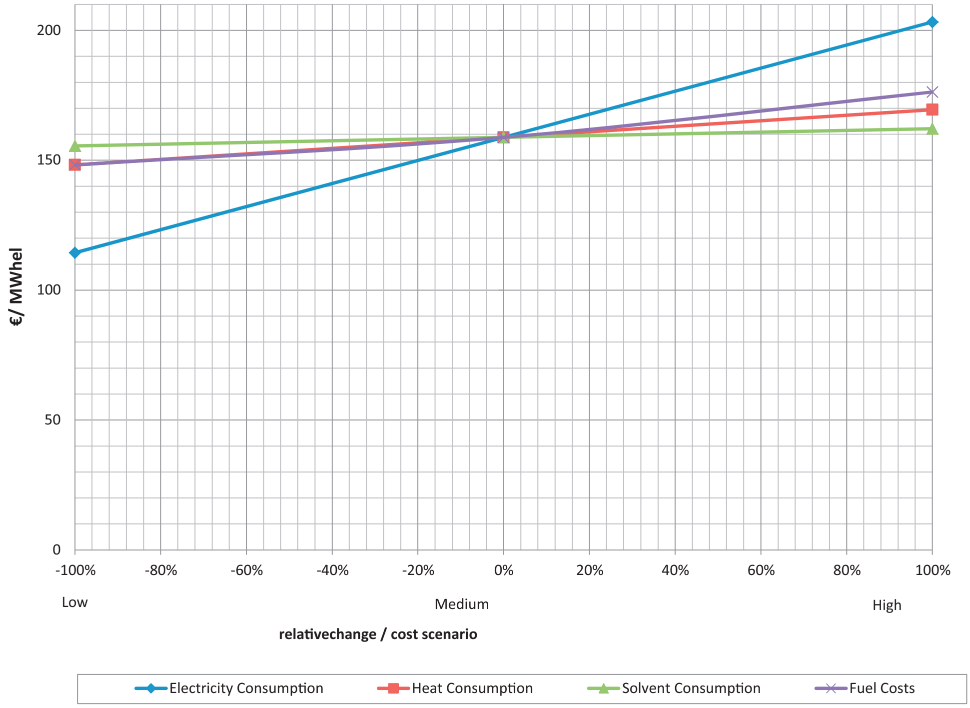

Fig. 12. Sensitivity analysis of the LCoE from hard coal PC with CCS in Europe in 2025, based on the mineral carbonation demands of the ÅAU 2 process. The medium fuel cost price scenario is selected and no CO_2 emission allowances costs are included.

study provides numerous results for various mineral carbonation scenarios allowing for the direct comparison with the non-CCS power generation systems and the geological CO_2 storage option. Compared to geological storage of CO_2, the mineral carbonation pathway leads to higher impacts on human health and ecosystems as well as higher electricity production costs. These results demonstrate trade-offs between climate change mitigation and storing CO_2 in a stable and inert form on the one hand and environmental and economic drawbacks on the other hand.

The integration of the new life cycle inventories for different mineral carbonation scenarios into recently published LCA of CCS (Volkart et al., 2013) allows for a consistent comparative environmental evaluation of the different systems. Including the upstream and downstream effects of CCS implementation provides an indication of the “real” reduction of life cycle GHG emissions achieved compared to the one implied by the CO_2 capture rate. Assessment based on additional LCA indicators provides a broader picture of the environmental performance. Moreover, by introducing LCI for different mineral carbonation scenarios based on experimental results available in literature, the present assessment of the various mineral sequestration options represents the current and potential future state of the carbonation process.

Table 4

Summary of life cycle GHG emission results of assessing the mineral carbonation CO_2 storage option. For direct comparison between this study and the available literature, the results are given in both the functional units being the generation of 1 MWh_{el} and the storage of 1 ton of CO_2. Comparable results are indicated with equal color patterns.

Own Results		**NETL 1**	**NETL 2**	**NETL 3**	**ÅAU 1**	**ÅAU 2**	**Fuel type of power generation plant**	**Heat source of carbonation process**
	Mineral	**Wollastonite**	**Olivine**	**Serpentine**	**Serpentine**	**Serpentine**		
	Process Efficiency	69%	81%	92%	(75%) 65 %[a]	(80%) 90%[a]		
	kg CO2-eq/MWhel	286	415	598	914	549	Hard coal	Hard coal
	kg CO2-eq/MWhel	172	231	262	347	254	Natural gas	Natural gas
	kg CO2-eq/ton CO2 stored	202	400	682	1170	608	Hard coal	Natural gas
	kg CO2-eq/ton CO2 stored	198	402	507	798	478	Natural gas	Natural gas
Nduagu et al., 2012a	kg CO2-eq/ton CO2 stored			720			Hard coal	Natural gas
	kg CO2-eq/ton CO2 stored					580		
Khoo et al., 2011	kg CO2-eq/MWhel					300	Natural gas	Natural gas
Kirchofer et al., 2012[b]	kg CO2-eq/ton CO2 stored		260-400	680			-	Natural gas

[a] For the ÅAU process scenarios two efficiencies are given one for each step of the process. The efficiency inside the brackets is indicating the Mg extraction and the other the carbonation step.

[b] Very slow reaction times and extrapolated experimental data are assumed. So a direct comparison with this study is considered difficult.

In order to validate the results, Table 4 shows a summary of the life cycle GHG emission results found in this and in other studies which also focused on evaluating the environmental performance of mineral sequestration as a CO_2 storage option. A meaningful comparison with LCA results from up-to-date literature is enabled by the broadness of this study by addressing mineral carbonation in a general European case for 2025, for both hard coal PC and NGCC power plants, for two carbonation processes (NETL and ÅAU), for all three natural minerals for which experimental data are available and for two functional units (generation of 1 MWh_{el} and storage of 1 ton of CO_2).

Since one of the most complete LCA found on the topic is given by Nduagu et al. (2012a), a comparison with these results is crucial. As seen in Table 4 the life cycle GHG emissions derived from in the two studies for storing 1 ton of CO_2 are on a similar level (700 kg CO_2-eq/t CO_2 stored). Also similar results are found when comparing the life cycle GHG emissions for generating 1 MWh_{el} by a NGCC power plant from the present study with the study of Khoo et al. (2011). The results provided by Kirchofer et al. (2012) are in the same range, but since their study has considered different assumptions on the reaction times the comparison is not straightforward. Most of the differences found between the studies derive from different assumptions on the efficiency levels of the processes, the means of heat and electricity delivery and the depth of analysis in different aspects such as the solvent losses and the mineral mining.

In addition some main differences can be found about the system boundary conditions and the LCA background data assumed for the three studies mentioned. Nduagu et al. (2012a) consider a hard coal power plant placed in British Columbia (BC, Canada) with a CO_2 MEA capture unit as a CO_2 source unit for the mineral carbonation process. The ecoinvent v.1.3 database is used for the LCA with key modifications to substitute the European data with Canadian and US power plant data. On the other hand, Khoo et al. (2011) evaluate the CCS system where natural minerals are mined in Western Australia/Tasmania and shipped to Singapore for mineral carbonation. The pure CO_2 stream derives from a NGCC power plant with a MEA CO_2 capture unit. The ecoinvent database (2009) is used along with other sources for the LCA. Finally, Kirchofer et al. (2012) excludes the separation and compression from the system boundaries and pure compressed CO_2 stream is assumed as an input. A hybrid process model and an economic input–output LCA approach is used (hybrid EIO-LCA), where the direct consumptions in energy, materials and services are converted into life cycle consumptions. The study focuses in USA, so US commodity price indexes are used in the model.

The results of this study allow identifying the most suitable natural mineral and process for CO_2 carbonation. Wollastonite shows the best performance from the environmental and economic perspectives. Its high reactivity, which allows avoiding the usage of chemical additives, along with the optimized electricity and heat requirements provides a clear advantage compared to serpentine and olivine. Olivine seems to perform better than serpentine in the life cycle GHG emissions results and in most of the ReCiPe midpoint results since, with the electricity and heat sources chosen in this study, the increased electricity requirements for olivine carbonation cause less environmental burdens than the high heat requirements for serpentine carbonation. On the other hand, the cost results show that it is more profitable to use serpentine than olivine due to the fact that heat is cheaper than electricity. However, according to O'Connor et al. (2002) and USGS (2011) the availability of natural mineral in terms of global reserves shows the opposite trend than the environmental performance: Wollastonite is considered to be a relatively scarce mineral resource, whereas serpentine is abundant, along with olivine which is common as well.

Comparing the two carbonation processes assessed (NETL and ÅAU) with serpentine as carbonation mineral allowing for high reaction efficiencies in both processes – leads to the following conclusions: If ÅAU process can be implemented with the future potential efficiencies of the process, the minimized heat requirements through an optimized heat exchangers system and low solvent losses (1%) as assumed in the second ÅAU process scenario, it performs better than the NETL process in terms of all the environmental and cost results (life cycle GHG emissions, ReCiPe midpoint results and LCoE). However, the difference in the life cycle GHG emissions between the two processes is less substantial as stated in Nduagu et al. (2012a). Also, even if the ÅAU process achieved the abovementioned high reaction efficiencies and minimum heat requirements, the difference between the two processes about the solvent losses would still be an open question. If in NETL process achieved a reduction in solvent losses up to a level of 10%, the life cycle GHG emission results would be reverse.

None of the mineral carbonation scenarios seems economically viable even with an EU ETS scenario of 80 €/ton of CO_2 emitted. Similar to the environmental burdens, also the main cost increases derive from the extensive energy requirements of the carbonation process, the mineral mining and the solvent losses. Further research should be performed on those topics in order to increase the reaction efficiencies of the processes and minimize the energy and chemical additives requirements. Only then the mineral sequestration can increase its competitiveness compared to geological storage regarding GHG emission reductions and LCoE for fossil-fueled power generation.

As mentioned in literature review (Section 1.2) the cost results that are available on the topic are calculated only on the basis of carbonating 1 ton of CO_2. So overall cost results on the basis of generating 1 MWh_{el} as presented here cannot be directly compared. But if only the carbonation costs of 1 ton of CO_2 are calculated (excluding CO_2 capture and transport) for the different scenarios presented, the following results are derived: between €97 and €200 per ton of CO_2 avoided for the PC hard coal cases (heat provided by burning PC hard coal and EU electricity mix) and between €102 and €202 per ton of CO_2 avoided for the natural gas cases (heat provided by burning natural gas and EU electricity mix). So comparing with the available literature cost results (Gerdemann et al., 2007; Khoo et al., 2011; Newall et al., 2000), a broad range is also found between $80 and $500 per ton of CO_2 avoided for mineral carbonation scenarios.

Additional improvements have been proposed by the ÅAU which are not considered in the inventories of the current study and the results presented. Fagerlund (2012), Nduagu et al. (2012a) and Romão et al. (2012b) analyze the integration of the ÅAU process with the sintering industry. Using the iron and calcium by-products of the ÅAU process would allow for a reduction of the CO_2 emissions of the sintering process because no CO_2 is released when these by-products are oxidized (Romão et al., 2012b). In the same study additional positive aspects of this integration are mentioned, such as reduced heat demand in sintering and reductions in coke, sinter and pellets consumption. Screening calculations were performed about the potential CO_2 mitigation due to such an integration (assuming the ÅAU 1 and 2 scenarios). Based on the assumptions made for these scenarios a total additional mitigation of 19 kg of CO_2/ton of CO_2 carbonated could be achieved corresponding to around 2–7% of total GHG emissions. Since that amount is not significant, this integration is not addressed in the present study.

Finally, Romão et al. (2012a) propose to use direct flue gas in the fluidized bed so as to avoid the energy intensive step of CO_2 capture. But in order to perform the same carbonation reaction with partial pressure for CO_2 equal to 20 bar and high reaction efficiency, significant amounts of electricity are required to compress the total amount of flue gas. Calculations were performed on such a scenario, but first results showed that more electricity would be consumed for the flue gas compression than being generated by the power plant. In order to ensure the viability of such a process, it would have to achieve high carbonation reaction efficiency with low CO_2 partial pressures being as close as possible to the atmospheric conditions. Also additional problems should be considered when assuming very high total pressures for the flue gas which are connected to the reactor infrastructure and the limited distances that flue gas of such high pressure could be transported.

5. Conclusions

The main conclusions from the results of this study are:

- Implementation of mineral carbonation as option for CO_2 storage for hard coal PC and natural gas-fueled power generation shows a reduction of life cycle GHG emissions of 15–64% compared to electricity production without CCS. The large range indicated is a result of the assessment of two carbonation processes with several process layout scenarios, which substantially vary in their performance. These reductions are considerably below those achievable with geological CO_2 storage calculated with consistent boundary conditions. The life cycle GHG emission reductions are far below the initial CO_2 capture rate of 90%. Energy (electricity and heat) and solvent requirements of the carbonation processes are the main contributors to GHG emissions associated with mineral CO_2 sequestration.
- The LCA results for other environmental impacts show a similar pattern: mineral carbonation generally generates higher impacts than geological storage of CO_2 and mostly substantially higher impacts than power generation without CCS. Mineral carbonation is penalized by the high energy requirements of the carbonation processes. In addition, the mineral mining seems to cause significant health issues related to particulate formation.
- The calculations of LCoE show significant cost increases in power generation due to CCS implementation. Following a medium fuel price scenario for 2025 and without additional costs for CO_2 emission certificates, generating 1 MWh_{el} from hard coal PC costs 46€ for the reference case without CCS. Implementing CCS with geological CO_2 storage increases LCoE by 65%, while mineral carbonation results in an increase of 209–370%. For NGCC, power generation costs increase from 69€/MWh_{el} for a NGCC plant without CCS by 45% for geological CO_2 storage and by 90–136% for mineral carbonation. Mineral carbonation as CO_2 storage option seems to be more favorable for NGCC due to the reduced amounts of CO_2 to be stored per MWh_{el}. The energy and material requirements of carbonation processes (especially the electricity costs) are the main contributors to the cost increases compared to geological storage.
- Similar LCoE results were produced when the carbonation plant was considered as part of the power plant and not a separate unit. Only one test case led to substantially increased final costs, due to considerable electricity requirements for operating the carbonation plant (NETL 2 case).
- Among the natural minerals analyzed for the CO_2 sequestration process, wollastonite seems the most favorable from both environmental and economic perspectives. Olivine is more favorable than serpentine in terms of environmental results, but using serpentine results in lower power generation costs.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.ijggc.2013.12.002.

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