



Kaunas University of Technology
Faculty of Chemical Technology

Life cycle based environmental impact of CO₂ mineral carbonation using serpentine and phlogopite

Master's Final Degree Project

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Summary

As anthropogenic CO₂ emissions drive the urgent need for carbon capture, utilisation, and storage, mineral carbonation has emerged as a potential permanent storage solution. However, the technology is challenged by high energy intensity and a lack of standardised environmental assessments. This study addresses this gap by comparing life cycle assessments of serpentine- and phlogopite-based mineral carbonation routes in the Finnish context. Using unified functional unit and system boundaries, both conventional and integrated process routes are evaluated, incorporating chemical recovery to improve system realism.

This study uses SimaPro 9.0 software and Ecoinvent 3.5 database to perform life cycle assessment according to standards ISO 14044:2006 and 14044:2006. Following life cycle assessment structure, this study has established goal and scope, analysed the inventory, evaluated impacts and followed with interpretation. The functional unit was set as one kilogramme of CO₂ captured. System boundaries chosen were gate-to-gate focusing primarily on the mineral carbonation processes in Åbo Akademi University, Turku, Finland where this study partially took place under Magnex project funding. IMPACT 2002+ was chosen as life cycle impact assessment calculation methodology. Implementation of this calculation method has allowed to analyse scenarios based on four damage impacts such as human health, ecosystem quality, climate change and resources. Electricity parameter was used to perform sensitivity analysis.

During the research, three scenarios were established based on different mineral carbonation routes implementing dry/wet magnesium sulfate extraction and wet carbonation principles. While scenario 1 and 2 use serpentine as their feedstock, scenario 3 implements phlogopite. Life cycle assessment results show that, despite its higher magnesium content, phlogopite does not achieve greater CO₂ capture efficiency than serpentine, disproving the hypothesis. While all three scenarios show similar environmental performance, scenario 1 exhibits lowest environmental burdens due to smaller electricity demands during magnesium sulfate extraction phase. Calculations resulted in CO₂ estimations of -0.383, -0.329, -0.278 kg of CO₂ emitted per functional unit across scenarios one, two and three respectively. During this study, importance was placed on establishing the use of chemical recycling to minimise environmental burdens and after establishing fourth scenario, it was concluded that chemical recovery and reuse is crucial to keep environmental burdens low in mineral carbonation process. Sensitivity analysis indicates that energy use is the primary driver of environmental impact. The findings demonstrate that process configurations relying on heat as the main energy input yield the most favourable outcomes, and that mineralogical composition alone does not ensure improved capture efficiency.

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Santrauka

Kadangi antropogeninės CO₂ emisijos skatina skubų anglies dioksido surinkimo, panaudojimo ir saugojimo poreikį, mineralų karbonizacija iškilė kaip potencialus nuolatinio saugojimo sprendimas. Tačiau šiai technologijai iššūkis yra didelis energijos intensyvumas ir standartizuotų aplinkosauginių vertinimų trūkumas. Šis tyrimas sprendžia šią problemą, palygindamas serpentiną ir flogopitą pagrindų veikiančių mineralų karbonizacijos būdų gyvavimo ciklo vertinimus Suomijos kontekste. Naudojant vieningus funkcinis vienetus ir sistemos ribas, vertinami tiek įprasti, tiek integruoti procesų maršrutai, įtraukiant cheminį regeneravimą, siekiant pagerinti sistemos realizmą.

Šiame tyrime naudojama „SimaPro 9.0“ programinė įranga ir „Ecoinvent 3.5“ duomenų bazė, siekiant atlikti gyvavimo ciklo vertinimą pagal standartizuotus procesus pagal ISO 14044:2006 ir 14044:2006. Remiantis gyvavimo ciklo vertinimo struktūra, šiame tyrime nustatytas tikslas ir apimtis, išanalizuota inventorizacija, įvertintas poveikis ir pateikta interpretacija. Pasirinktos sistemos ribos buvo „nuo vartų iki vartų“, daugiausia dėmesio skiriant mineralų karbonizacijos procesams Åbo Akademi universitete Turku, Suomijoje, kur šis tyrimas iš dalies buvo atliktas finansuojant „Magnex“ projektą. Funkcinis vienetas buvo nustatytas kaip vienas kilogramas surinkto CO₂. IMPACT 2002+ buvo pasirinkta kaip gyvavimo ciklo poveikio vertinimo skaičiavimo metodika. Šio skaičiavimo metodo įgyvendinimas leido išanalizuoti scenarijus, pagrįstus keturiais žalos poveikiais, tokiais kaip žmonių sveikata, ekosistemų kokybė, klimato kaita ir ištekliai. Elektros parametras buvo naudojamas jautrumo analizei atlikti.

Tyrimo metu buvo sukurti trys scenarijai, pagrįsti skirtingais mineralų karbonizacijos būdais, įgyvendinant sauso/šlapio magnio sulfato ekstrakcijos ir šlapio karbonizacijos principus. 1 ir 2 scenarijuose kaip žaliava naudojamas serpentinas, o 3 scenarijuje naudojamas flogopitas. Gyvavimo ciklo vertinimo rezultatai rodo, kad nepaisant didesnio magnio kiekio, flogopitas nepasiekia didesnio CO₂ surinkimo efektyvumo nei serpentinas, o tai paneigia hipotezę. Nors visi trys scenarijai rodo panašų aplinkosauginį veiksmingumą, 1 scenarijus pasižymi mažiausia aplinkosaugine apkrova dėl mažesnio elektros energijos poreikio magnio sulfato ekstrakcijos etape. Skaičiavimų rezultatai parodė, kad vienam funkciniam vienetai pagal pirmąjį, antrąjį ir trečiąjį scenarijus išmetamo CO₂ kiekis yra atitinkamai -0,383, -0,329 ir -0,278 kg. Šio tyrimo metu buvo akcentuojamas cheminio perdurbimo naudojimas siekiant sumažinti aplinkos naštą, o įgyvendinus ketvirtąjį scenarijų, buvo padaryta išvada, kad cheminių medžiagų regeneravimas ir pakartotinis vartojimas yra labai svarbus siekiant sumažinti aplinkos naštą mineralų karbonizacijos procese. Jautrumo analizė rodo, kad energijos naudojimas yra pagrindinis poveikio aplinkai veiksnys. Rezultatai rodo, kad proceso konfigūracijos, kuriose pagrindinis energijos šaltinis yra šiluma, duoda palankiausias rezultatus ir kad vien mineraloginė sudėtis neužtikrina geresnio surinkimo efektyvumo.

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List of abbreviations and terms

Abbreviations:

ABS - Ammonium Bisulfate;

AS – Ammonium Sulfate;

Assoc. prof. – associate professor;

BP MED - Bipolar Membrane Electrodialysis;

CCS - Carbon Capture And Storage;

CCU - Carbon Capture And Utilisation;

CCUS - Carbon Capture, Utilisation And Storage;

CO₂ - Carbon Dioxide;

F-gases - Fluorinated Gases;

FU - Functional Unit;

GHG - Greenhouse Gas Emissions;

GWP - Global Warming Potential;

IEAGHG - International Energy Agency Greenhouse Gas R&D Programme;

IPCC - Intergovernmental Panel on Climate Change;

ISO - International Organisation for Standardisation;

LCI - Life Cycle Inventory;

LCIA - Life Cycle Impact Assessment;

LCA - Life Cycle Assessment;

MCH - Magnesium Carbonate Hydrates;

MgSO₄ - Magnesium Sulfate;

PFB - Pressurised Fluidised-Bed;

SA – Sensitivity Analysis;

TRL – Technology Readiness Levels;

VC – Virgin Chemicals;

ÅA - Åbo Akademi University.

Introduction

Anthropogenic carbon dioxide (CO₂) emissions remain the primary driver of climate change, stemming primarily from fossil fuel combustion, cement, iron, petrochemical and ammonia production sectors, despite ongoing efforts to reduce emissions at source and deploy mitigation technologies [1]. While decarbonisation of energy systems and industrial processes is essential, significant residual emissions from hard-to-abate sectors like chemicals or cement are expected to persist. This has increased the need for carbon capture, utilisation and storage (CCUS) technologies that are capable of delivering long-term CO₂ mitigation. CO₂ mineralisation and particularly mineral carbonation have emerged as a promising pathway for permanent carbon storage. It involves the reaction of CO₂ with alkaline minerals, forming thermodynamically stable carbonate phases [2]. Magnesium-rich (Mg-rich) silicate minerals are particularly attractive due to their favourable chemistry. Serpentine is used due to its abundance. Mineral carbonation is a promising technology, but it is often criticised due to the high energy and material requirements associated with mineral extraction and pre-treatment. While CCUS is considered as a potential climate change mitigation technology, the environmental burdens arising during carbonation process itself may offset some of the benefits [2].

Despite extensive research on technologies, there is a lack of consistent and comparable environmental assessments across different feedstocks and process configurations. Many of the life cycle assessment (LCA) studies often focus on individual materials or downstream product application limiting the systematic comparison of overall environmental performance. This becomes more evident when considering less studied minerals like phlogopite as a feedstock, keeping the gap in the literature. In this study, feedstocks like serpentine and phlogopite are evaluated using constant and unified functional unit (FU), system boundaries and impact assessment methods to allow for more equal route and process comparison. To further provide a more realistic representation of mineral carbonation systems, integration of chemical recovery is introduced. The analysis is further contextualised within the Nordic region, focusing on Finland due to its mineral resources and existing processing capacity. Evaluating conventional and integrated process routes allows this study to contribute to a better understanding of mineral and process selection based on environmental performance.

Project aim: To assess the environmental performance of serpentine and phlogopite based CO₂ mineral carbonation routes through a comparative life cycle assessment.

Project hypothesis: Phlogopite, due to its higher magnesium and potassium composition, exhibits higher CO₂ capture efficiency and lower overall environmental impact than serpentine.

Project objectives:

1. Review existing scientific literature on mineral carbonation technology, processes and routes, analyse current life cycle assessment studies on mineral carbonation.
2. Define functional unit, system boundaries and scenarios for CO₂ mineral carbonation based on serpentine and phlogopite minerals.

3. Perform life cycle inventory analysis and life cycle impact assessment of chosen scenarios evaluating their environmental impacts and burdens.
4. Identify environmental hotspots for the chosen scenarios and perform sensitivity analysis.

1. Literature review

1.1. Climate change and the role of carbon capture, utilisation and storage

Global efforts to mitigate climate change are driven by the need to achieve reductions in greenhouse gas (GHG) emissions. This requirement aligns with international and regional climate targets. It was further accelerated at the global level when the Intergovernmental Panel on Climate Change (IPCC) was established in 1988 [3]. IPCC enabled scientific teams to detail climate change information and shape future regulations. Since IPCC was established, global accountability was pushed through agreements and regulatory milestones like the Kyoto protocol [4], the Paris agreement [5] and the European Green Deal [6]. All of them focus on goals such as reducing GHG emissions and achieving climate neutrality.

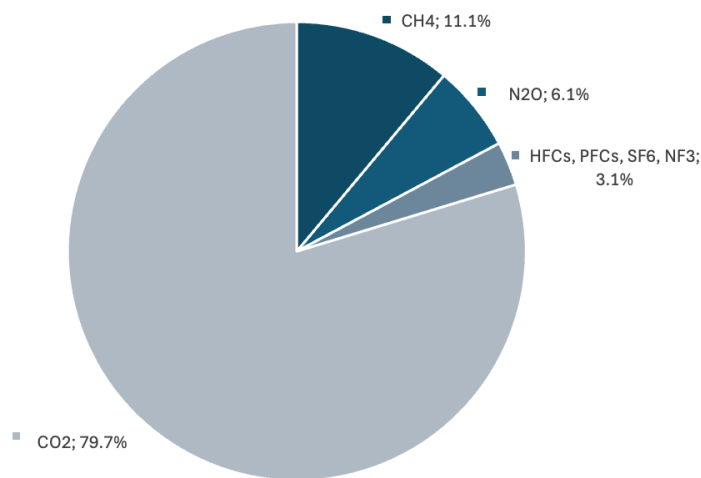


Fig. 1. Greenhouse gas emissions expressed by type (reproduced from [8])

GHG emissions consist of several gases. As shown in Fig. 1., the main ones are carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O) and fluorinated gases (f-gases) [7], which are all anthropogenic source gases. These gases come from direct human activities and are big contributors to climate change through the GHG effect. Both CH₄ and N₂O are directly linked to agriculture as the biggest sources, with CH₄ additionally released through waste management and biomass burning. F-gases stem from industrial processes and refrigeration. Even with additional gases, CO₂ remains the main culprit in the GHG emissions [1].

Fig. 2. visually represents the annual global CO₂ emissions rise despite the established international climate agreements and regulations [9]. In the past few years, emissions have reached nearly 40 billion tons per year. Emissions are still rising, showing that there is still a need for mitigation strategies. There's a strong pressure to decarbonise sectors that rely heavily on fossil fuels. But as rapid decarbonisation of energy systems and industrial processes remains the primary strategy for limiting global temperature rise, emissions reductions alone are unlikely to be enough to meet long-term climate objectives [10]. Additionally, residual emissions from hard-to-abate sectors such as heavy industry and long-distance transport, together with chemical processes, are expected to persist even under ambitious established mitigation scenarios [11]. The current rate of decarbonisation puts pressure on finding technologies that are capable of removing CO₂ from the atmosphere, as this would ensure its long-term storage. This step becomes an essential for reducing emissions.

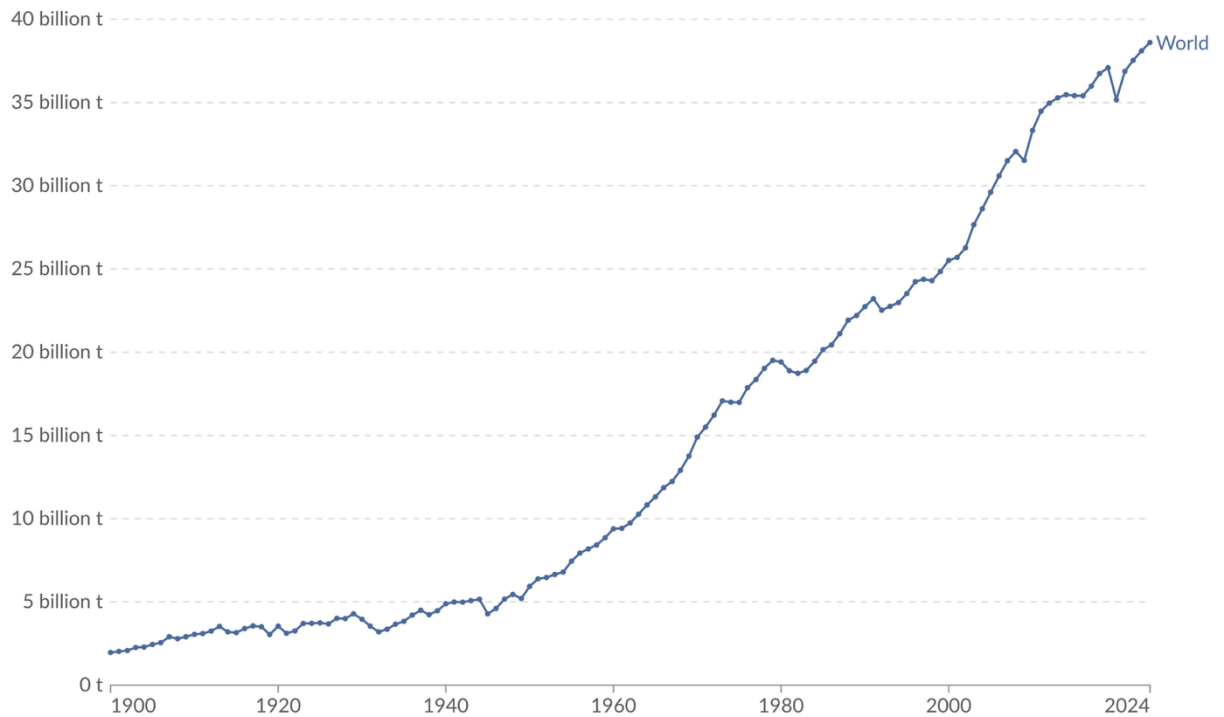


Fig. 2. Annual global CO₂ emissions from fossil fuels and industry [9]

CCUS have emerged as a broad category of approaches for managing CO₂ emissions and these technologies can be separated into several classifications by their purpose and effectiveness:

- **Carbon capture and storage (CCS)** – works by capturing CO₂ from large production facilities (power plants, industrial factories) and transporting it to be stored in underground geological formations.
- **Carbon capture and utilisation (CCU)** – instead of storing captured CO₂, focuses on creating a feedstock to be used in different industries (chemistry, fertilisers, construction materials, etc.).
- **Carbon capture, utilisation and storage (CCUS)** – connects both CCS and CCU under a single definition as its purpose is to both capture CO₂ from industries and transform it into usable feedstock [12].

The climate effectiveness of these technologies strongly depends on the stability of CO₂ storage, as many utilisation routes involve only temporary fixation of CO₂. Even though this still creates small risks of carbon being released back into the atmosphere, which would limit long-term mitigation potential [13], this technology is on the rise as conventional geological storage creates certain challenges requiring extensive site characterisation and long-term monitoring to minimise the risk of leakage. These challenges have increased interest in CO₂ storage options that could offer stability and minimise long-term environmental risks [14].

Mineral carbonation represents one pathway in which CO₂ reacts with alkaline minerals to form thermodynamically stable carbonate phases. Converting CO₂ into solid materials that are stable on the geological time scale [2] eliminates the risk of leakage and reduces the need for long-term monitoring. Favourably, it can be applied to both natural silicate minerals and industrial residues rich

in calcium or magnesium, making it a potentially scalable and versatile option for permanent storage, distinguishing it from other approaches amid the growing relevance of climate change mitigation research [15]. Despite its advantages, mineral carbonation is associated with technical and environmental challenges that may influence its overall sustainability. One of the challenges would include the energy requirements associated with mineral extraction and pre-treatment and this could cause challenges in material handling at larger scales while affecting the net environmental performance [2].

1.2. Mineral carbonation as a storage pathway

Conventional geological storage of CO₂ involves physically injecting and retaining it in subsurface formations. During that process, CO₂ is stored with the help of geological trapping mechanisms. In contrast, mineral carbonation is a CCUS pathway in which CO₂ reacts with alkaline minerals to form stable carbonate phases [16][17]. In Mg-rich silicate minerals, it results in the formation of thermodynamically stable magnesium carbonates over geological timescales [15].

Due to its technological potential and the ability to provide permanent, leak-free storage, mineral carbonation has been a significant research topic in climate mitigation, though current research indicates it is not yet practical for industrial implementation. This is mainly due to kinetic and process-related factors and limitations [18]. To better understand the difference, mineral carbonation should be reviewed as a storage pathway. Geological storage works by physically trapping CO₂ in subsurface formations through structural mechanisms. By contrast, mineral carbonation chemically transforms CO₂ into stable solid carbonates which reduces the risk of leakage by immobilising carbon in mineral form rather than geological structures. Additionally, this technology offers a more permanent storage pathway with less intensive long-term monitoring requirements when compared to geological storage [2].

Within natural systems, mineral carbonation occurs as part of the long-term silicate weathering cycle, during which atmospheric CO₂ dissolves in water to form weak carbonic acid. This acid promotes the dissolution of metal-bearing silicate minerals releasing cations. The released cations include Ca²⁺, Mg²⁺ or Fe²⁺ and react with dissolved carbonate species formed in solution to precipitate solid minerals. Common minerals include calcite (CaCO₃), magnesite (MgCO₃) and siderite (FeCO₃) [2].

Mineral carbonation has gained attention in engineering due to its ability to provide permanent carbon immobilisation inspired by natural processes. The ability to apply it to naturally occurring silicate rocks or alkaline industrial residues makes it more attractive for research allowing new applications to emerge. The importance lies in the degree of process enhancement, as mineral carbonation approaches can differ in classification. Particularly Fig. 3. [19] presents and separates mineral carbonation into passive and accelerated processes determining how CO₂ is captured. Accelerated mineral carbonation can be further broken down into in-situ and ex-situ applications while the passive mineral carbonation is a naturally occurring carbon sequestration process.

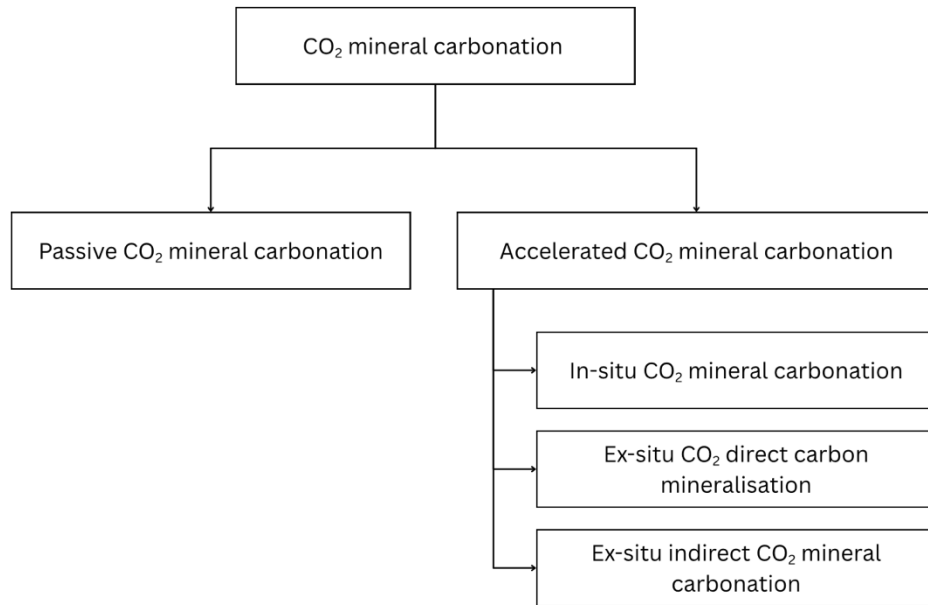


Fig. 3. CO₂ mineral carbonation scheme showing mineral carbonation methods (reproduced from [19])

During passive mineral carbonation, atmospheric or dissolved CO₂ reacts with reactive minerals to form stable carbonate solids. Unlike engineered CCUS, which may require significant energy inputs, passive mineral carbonation relies primarily on the natural exposure of CO₂ to suitable minerals in air and water, but is chemically limited by a low CO₂ content[2].

1.2.1. Accelerated mineral carbonation

Accelerated mineral carbonation is an engineered approach designed to enhance the rate of CO₂ mineral carbonation beyond naturally occurring weathering rates and how can it be applied to further climate change mitigation. While passive mineral carbonation provides permanent carbon storage under ambient conditions, factors like reaction rates are generally too slow to address anthropogenic CO₂ emissions [20][21]. This limitation becomes particularly relevant when time scales are considered, leading to development of accelerated carbonation operating to overcome natural kinetic limitations during controlled process conditions. Depending on where the reaction occurs, these processes are commonly classified as in-situ or ex-situ [19].

In-situ carbonation involves injecting CO₂ into suitable geological formations containing reactive silicate minerals. Ultrafamic bodies containing serpentine represent promising targets throughout literature due to their high magnesium content allowing for the potential natural carbonation reactions over extended timescales [22]. Minimal material handling and the use of naturally occurring geological conditions becomes beneficial however limited control over reaction rates and process conditions becomes the main limitation. Slow reaction kinetics and long-term implementation uncertainties are another limitations in in-situ carbonation [23].

Field-scale mineral carbonation has been demonstrated in basaltic systems by the CarbFix project, where rapid conversion of injected CO₂ stabilises carbonate minerals. Although basalt differs mineralogically from serpentine, these findings demonstrate the feasibility of engineered acceleration of silicate mineral carbonation under appropriate conditions. Permeability, surface area and fluid transport properties are critical for carbonation rate in serpentine ultrafamic formations, while

phlogopite are less likely to exhibit significant in-situ carbonation. Structural stability and slower dissolution kinetics are major factors in. Mining is environmentally detrimental factor which plays an important role in mineral carbonation and in-situ approach eliminates such factor as mining is not requiring. Eliminating the need for mining minimises surface disturbance [24].

Ex-situ carbonation is conducted in engineered systems where reaction conditions can be systemically controlled and optimised in a laboratory or factory settings. Involving mining, crushing and processing of Mg-rich silicate minerals brings the environmental burdens associated with surface disturbance and material transportation. Slurry preparation and carbonation under controlled temperature and pressure conditions follow the ex-situ approach involving chemicals and additional materials [19]. Two principal approaches are commonly distinguished:

- **Direct aqueous carbonation** involves mineral dissolution and carbonate precipitation occurring in a single reactor system;
- **Indirect carbonation** involves two stages in which magnesium or calcium is first extracted into solution and then carbonate precipitation is induced.

Even though, ex-situ has more environmental burdens, when compared to in-situ approaches, this process offers significantly greater flexibility during process optimisation, enabling higher conversion efficiencies and integration with concentrated industrial CO₂ streams. Through particle size reduction and thermal activation reaction rates can be enhanced adding elevated pressure when required which isn't available with in-situ process. Chemical additives make reactions quicker and more efficient but brings out the disadvantages that consist of substantial energy and material requirements. Optimisation of the systems allow detailed control over various factors such as mass transfer and solution chemistry, while reactor design can be improved during the process. While mining operations and solid-liquid separation steps may contribute significantly to environmental burdens, the overall sustainability highly depends on how the process is integrated and what energy source is used [19].

This study focuses on comparing routes and feedstock for mineral carbonation. When it comes to comparing the feedstock for this type of carbonation, phlogopite is identified as a potentially attractive feedstock. Although comparatively underexplored, it is viable. Due to its aluminosilicate framework and interrelated potassium, it might be considered less favourable for direct carbonation [25]. Currently in literature, serpentinite is a more promising feedstock for engineered mineral carbonation due to its higher magnesium content and comparatively greater reactivity activation.

1.3. Silicate minerals for mineral carbonation

Mineral carbonation relies on reactive mineral feedstock. Silicate minerals are one of the most widely studied feedstocks for CO₂ mineral carbonation due to their abundance and capacity to supply alkaline earth metals required for carbonate formation. Within this group, magnesium- and calcium-bearing silicates are particularly relevant, as they provide the cations necessary for the formation of stable carbonate products [2]. The mineralogical composition and crystal structure strongly influence mineral reactivity. Although both calcium and Mg-rich silicates have been investigated for CO₂ mineral carbonation, this research exclusively focuses on Mg-rich systems.

Magnesium-bearing silicate minerals have been widely studied in recent research due to their ability to form stable magnesium carbonate phases, making them an attractive feedstock enabling permanent CO₂ storage. These minerals are widely distributed and occur in ultramafic and metamorphic rock formations. These include phases such as serpentine and magnesium-bearing mica minerals. In the mineral carbonation process, magnesium is the key cation that reacts with CO₂ to form thermodynamically stable carbonate products. The focus placed on Mg-rich silicates is motivated by the fundamentally different reaction pathways compared with calcium systems. These differences extend to carbonate products and overall process requirements [24]. Magnesium-based mineral carbonation typically exhibits slower reaction kinetics, often requiring energy-intensive extraction and activation steps to increase its availability, which may offset the long-term benefits of permanent CO₂ storage. These factors result in distinct energy and material tradeoffs compared with calcium carbonation systems. Magnesium-based mineral carbonation presents specific technical and environmental challenges that warrant separate investigation. Despite their advantages, slow kinetics and additional processing requirements remain a major barrier to large-scale deployment. Limiting the scope of the study to Mg-rich silicates enables a more consistent and meaningful comparison of mineral-specific routes within the LCA framework while avoiding confounding effects arising from different carbon chemistry and process assumptions [25].

1.3.1. Serpentine: structure, availability, reactivity

Commonly used serpentinite in CO₂ mineral carbonation refers to rock containing serpentine group minerals such as antigorite, chrysotile, lizardite. In earlier literature, the terms serpentinite and serpentine are often used interchangeably, while this research focuses on serpentine as a primary feedstock, commonly represented by the general formula (1.3.1.1) following the works of Zevenhoven et al.[26].



As serpentine is among the most widely studied Mg-rich silicates for CO₂ mineral carbonation, making it a suitable benchmark for comparison with phlogopite. Serpentine, as a feedstock, is abundant in ultramafic and mafic rock formations and globally available in significant quantities. Available quantities are estimated to be sufficient to bind all carbon from fossil fuels [27]. As a result, the mineral is attractive for large-scale CO₂ sequestration. Structurally, serpentine exhibits a layered phyllosilicate structure composed of alternating tetrahedral (T) silica sheets and octahedral (O) magnesium hydroxide layers [26], which are similar to those of brucite as expressed in Fig. 4.



Fig. 4. Molecular structure of lizardite which primarily consists of brucite and silica

The presence of structurally bound hydroxyl groups contributes to the stability of the crystal lattice. This contributes to lattice stability and reduces dissolution under ambient conditions. Throughout the research, serpentine has shown a high theoretical CO₂ sequestration due to its magnesium content. However, the natural reactivity under ambient conditions is limited [28][29]. To carbonise serpentine,

pretreatment is required as serpentine carbonation efficiency depends on its surface reactivity and dissolution rate. To advance mineral reactivity, several pretreatment options exist: thermal activation, chemical treatment, sonication activation. Thermal activation has been used most commonly. Pretreatment significantly improves reaction kinetics. However, it also introduces additional energy and material demands. Within the assessment of lifecycle analysis, which influences overall environmental performance and introduces burdens to the system [26][27].

1.3.2. Phlogopite: structure, availability, reactivity



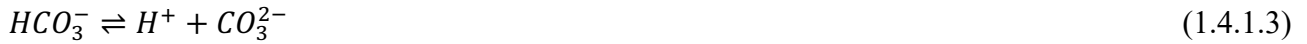
Phlogopite is a common Mg-rich mica mineral with the general formula expressed in the (1.4.3.1) formula. Similar to serpentine, phlogopite can be found in ultramafic rocks and bears a similarity to the general serpentine formula (1.4.2.1). Studies indicate that this mineral is less extensively studied in mineral carbonation settings, but could potentially be a relevant feedstock due to additional potassium (K) content. Phlogopite commonly occurs alongside serpentine in ultramafic rock formations. As part of the mica group, phlogopite exhibits a layered silicate structure in which tetrahedral sheets are bonded by interlayer potassium ions. Having a 2:1 layer arrangement (tetrahedral-octahedral-tetrahedral) improves phlogopite structural strength [30][31]. Compared to serpentine, phlogopite contains magnesium within a more complex alumina-silicate framework (AlSi₃). It particularly incorporates additional elements such as potassium (K) and aluminium (Al). The presence of interlayer potassium and aluminium contributes to greater structural stability and increases resistance to chemical weathering under ambient conditions [32]. Due to phlogopite's stability and bonded structure, can be expected to be less reactive than serpentine in direct carbonation. Reduced reactivity may limit viability for direct carbonation, making indirect routes not just preferable but potentially necessary. Additionally, structure breaks down at lower temperatures than serpentine [33]. Phlogopite shares reactivity factors with serpentine as under indirect or chemically assisted extraction routes, magnesium can be selectively mobilised for subsequent carbonation. This is preferable when compared with direct carbonation as it allows separation of extraction and carbonation, providing optimised reaction conditions [34]. The more complex elemental composition of phlogopite, particularly its potassium and aluminium content, may complicate downstream processes, including membrane-based systems or chemical looping approaches. Even though it is much less extensively studied than serpentine, phlogopite provides an interesting comparative case due to its distinct structural characteristics and potential integration within advanced mineral carbonation process routes [35].

1.4. Mineral carbonation fundamentals and process design

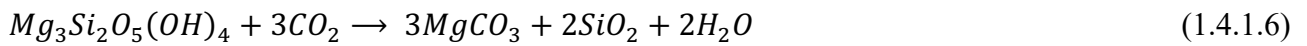
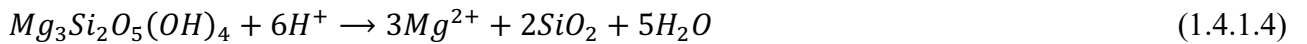
1.4.1. Reaction mechanisms and thermodynamics

The mineral carbonation of Mg-rich silicates goes through a series of chemical reactions which involve mineral dissolution, CO₂ dissolution and carbonate precipitation [36]. As the process begins with dissolution of CO₂ in water, all of the steps together control both the extent and rate of carbonation linking aqueous chemistry with solid-phase mineral reactivity. Carbonic acid and its dissociation products forms expressed in equations (1.4.1.1), (1.4.1.2), (1.4.1.3) [36][37].





An acidic environment is generated, and it provides conditions for mineral dissolution. Protons attack the silicate structure which breaks MgO bonds and releases magnesium ions into solution. During that process, hydroxyl groups contribute to water formation. Dissolution can be described by the equation in (1.4.1.4) for serpentine [36] and is commonly referred in literature as rate-limiting stage. The bond strength within silicate mineral structures is the one contributing to the limitation [38][39]. Following the dissolution, magnesium ions react with carbonate species in solution to form stable carbonate minerals like magnesite as shown in equation (1.4.1.5). The overall carbonation reaction is summarised in equation (1.4.1.6) [36][37].



These reactions are thermodynamically favourable and lead to the formation of stable carbonate phases becoming a promising approach for long-term CO₂ storage option under ambient conditions [36].

1.4.2. Mineral pre-treatment

Despite favourable thermodynamics, carbonation rate is often limited due to slow dissolution kinetics therefore pre-treatment often becomes an important step in mineral carbonation routes involving Mg-rich silicate minerals. Engineering optimal pre-treatment might directly influence magnesium availability and reaction kinetics. Due to the strong bonding within silicate lattices, untreated minerals typically exhibit low reactivity towards CO₂ under ambient conditions, and to overcome these limitations, various pre-treatment strategies have been proposed in the literature. Pre-treatment strategies include thermal activation, mechanical activation, chemical treatment and engineered activation including biochemical activation with the use of enzymes [40].

Thermal activation as a pre-treatment is a commonly studied approach in hydroxyl-bearing minerals, particularly serpentine. Serpentine group minerals contain up to 13% by weight in chemically bound water, which makes thermal activation particularly favourable [41]. Factors like heating temperature and duration, as well as particle size, influence the structural transformation of the mineral. Managing thermal activation with regard to structural transformation is essential to ensure subsequent carbonation. Heating minerals to high temperatures leads to dehydroxylation, releasing chemically bound water and increasing mineral reactivity. Studies consistently show that mechanical pre-treatment is critical for thermal activation. Reducing particle size allows heat to thoroughly penetrate and accelerate chemical water release [42]. However, thermal activation introduces significant limitations. When powered by fossil fuels, it becomes environmentally challenging. Focusing on heat from waste or renewable materials allows this process to stay viable. Additionally, complex processes and high energy requirements pose a deployment barrier for this pretreatment option [40].

Mechanical pre-treatment becomes an important step in mineral carbonation routes as it increases the reactivity of silicate minerals. This is done by milling or grinding. Grinding disrupts mineral

composition and order of crystal lattices. Disruption creates surface defects that accelerate the dissolution of magnesium-bearing silicates, promoting pore formation [43] and increasing the materials permeability and reactivity. Increasing the surface area of the mineral allows the reduction of energy required for carbonation. Optimising grinding and the mineral's parameters is critical. Excessive grinding of overly fine particles can reduce effectiveness [40]. The high energy demand has so far prevented large-scale deployment. Similar to thermal activation, the process depends on electricity for the grinding, which generates indirect CO₂ emissions. Managing high electricity demands and potential dust generation with noise pollution keeps mechanical pretreatment unfavourable for scalability opportunities [40].

Like previous pretreatment options, chemical treatment methods aim to selectively increase mineral solubility and accelerate reaction rates. By using acids, salts, and bases to extract magnesium and weaken silicate structures, crystallographic stability is utilised. Disrupting the stability of silicate structures increases the release of reactive cations, allowing carbonate formation [44]. Acid leaching is commonly applied as strong inorganic acids have the capabilities of dissolving silicate minerals. Typical acids include hydrochloric acid (HCl), sulphuric acid (H₂SO₄) and nitric acid (HNO₃) [45]. Salts are also commonly used in pretreatment to enhance reaction kinetics while providing milder leaching conditions resulting in more sustainable process [46]. Ammonium salts (AS) such as sulfate ((NH₄)₂SO₄), chloride (NH₄Cl) and ammonium bisulfate (NH₄HSO₄)(ABS) are common choices for salt pretreatment as research has established ammonium salts as effective magnesium extractors [47]. Bases play a critical role in pH adjustment. Although, it is not very commonly used, they provide support for closed-loop processes. Within the base usage, the most common are sodium hydroxide (NaOH), potassium hydroxide (KOH), and ammonium hydroxide (NH₄OH) [40]. Using potassium hydroxide could be beneficial for phlogopite due to its potassium content. That would provide a more sustainable approach and a potassium loop without introducing virgin chemicals. Using chemical pretreatment imposes significant limitations in real-world deployment settings. The use of chemicals introduces both the burdens of the infrastructure, which would require large scale handling systems and multi-stage reactors, and the burden of chemical waste or regeneration. The use of acids, bases and salt-based reagents creates an environment of hazards which would need to be managed with regulatory means and safety. On the environmental scale, it is important for mineral carbonation using chemical pretreatment to recover reagents. Azdarpour et al. [44] have established that without recycling of the chemicals, mineral carbonation sequestration gains are lost to environmental and cost burdens.

To minimise the burdens and limitations of each pre-treatment option, engineered activation is introduced. It can integrate various approaches to create an optimal pre-treatment strategy. Engineered activation can include mechano-chemical, mechano-thermal, thermo-chemical, mechano-thermo-chemical techniques. By combining the aspects of each pre-treatment strategy, particle reduction and reactivity are enhanced by eliminating factors that are negative in the use of just one pretreatment strategy. Considering mechano-chemical activation, combining milling, where mechanical forces are applied to particles, with the introduction of chemicals results in minerals dissolving more rapidly in aqueous environments. By eliminating the drawbacks of certain pretreatments, the process becomes more favourable in real-world applications. Studies show, particularly, Norouzpour et al. [40] propose that mineral carbonation should further focus on developing engineered activation routes which would improve process scalability and possibly save costs. This could potentially allow mineral carbonation to enter industrial applications. Apart from

improving scalability, using engineered activation could be beneficial in saving operational costs. Although engineered activation shows great potential for pre-treatment strategies, it can have additional limitations. Implementation still remains on laboratory scale as the system requires near-perfect coordination due to system complexity, while Norouzpour et al. [40] propose the possibility of performing LCA to validate larger scale industrial potential.

1.4.3. Mineral carbonation pathways

The pathways discussed will focus on ex-situ processes, as this research primarily focuses on ex-situ carbonation. Following pretreatment, carbonation reactions can proceed through different pathways depending on process configuration and reaction conditions. In magnesium-based systems, carbonation involves the reaction of the dissolved or extracted magnesium phases with CO₂ to form magnesium carbonate phases [47]. Carbonation reaction can proceed via either direct or indirect pathways. Direct mineral carbonation converts CO₂ into stable carbonates in one step. Being a single-step process, direct carbonation requires less complex infrastructure. The simplification reduces process complexity and infrastructure requirements. Easier carbonation only requires pre-treatment before CO₂ is carbonated in a single reactor. Even though a single-step process simplifies the carbonation, indirect carbonation pathways have been established in research as more efficient. Indirect carbonation is a two-step process in which reactive elements are extracted prior to carbonation. Direct carbonation is generally a slower process, whereas the indirect process achieve faster kinetics and higher conversion efficiency. Both pathways are highly dependent on mineral factors like composition and system factors like temperature or pressure [48].

The indirect carbonation route can be implemented through solid-gas or aqueous phases. The solid-gas phase represents the simplest reaction in the process design. It is designed to work with solid minerals that react with gas, which means that the CO₂ reaction happens directly with the mineral surface. To perform this carbonation, the mineral has to go through mechanical pre-treatment, which enhances reactivity through a larger surface area. Operating this type of carbonation, however requires extensive reaction conditions such as elevated temperature or pressure, possibly combining both. The temperature required for CO₂ to be preheated can range from 300 to 500 °C, while pressure has to be stable across the range of 20 bar. These conditions enable the reaction but significantly increase energy demand, limiting practical scalability and keeping this technology at low technology readiness levels (TRL) [48][49][51].

On the other hand, aqueous carbonation is a route where minerals react with CO₂ in the presence of water, creating a slurry where water within this process acts as a reaction medium. In this medium, CO₂ is dissolved and ion transport is facilitated. Aqueous carbonation involves three steps, out of which (1.4.3.1) magnesium ions (Mg²⁺) are released due to the presence of free protons (H⁺) while CO₂ dissolves in water and forms carbonic acid and bicarbonate ions (HCO₃⁻) (1.4.3.2) leading to metal and bicarbonate ions are combining to precipitate carbonation (1.4.3.3).



These reactions highlight the importance of proton availability and solution chemistry in controlling overall carbonation efficiency. The aqueous phase may allow for lower temperatures and lower pressure than the solid-gas phase process routes. The choice of carbonation pathway directly affects process efficiency, reactor design, and material handling and is strongly influenced by the characteristics of the mineral feedstock and the intended scale of operation [48][49].

1.4.4. Academic mineral carbonation process concepts

In addition to conventional carbonation pathways, several academic research groups have proposed integrated mineral carbonation process concepts that combine magnesium extraction, CO₂ mineral carbonation and chemical recovery within a single system. These concepts are often designed to address kinetic limitations and improve process efficiency by using looping systems, such as product recovery or alternative reaction sequences. Among the research conducted at Åbo Akademi University (ÅA), researchers have proposed mineral carbonation routes based on magnesium sulphate intermediates and an integrated chemical regeneration. During the process, magnesium is extracted from silicate minerals through chemical processing, which is followed by carbonation. The carbonation process forms stable magnesium carbonate phases, while additional chemicals are recovered and recycled through electrochemical or separation-based processes. Integrated routes are created to reduce reagent consumption and improve overall process sustainability [50].

Throughout ÅA research, serpentinite and serpentine are prioritised as a main feedstock for possible large-scale CO₂ mineral carbonation due to its high magnesium content and global abundance. Since the Zevenhoven research at ÅA has started in the early 2000s, the objective of process hasn't changed. ÅA has been working on developing an energy-efficient and technically feasible process capable of operating directly on CO₂ containing flue gas streams. Throughout the years, the development of the process routes was driven by several technical challenges in mineral carbonation: slow reaction kinetics, high energy demand for magnesium extraction, the need for CO₂ compression in high-pressure systems, requirement for efficient chemical recovery. Over time, five process configurations were proposed, with each addressing the technological evolution from heat-intensive, high-pressure carbonation towards more integrated and energy-flexible system [50-56].

- **ÅA route 1 (ÅA1)** - hot solid/solid Mg extraction + hot gas/solid carbonation
- **ÅA route 2 (ÅA2)** - hot solid/solid Mg extraction + aqueous carbonation
- **ÅA route 3 (ÅA3)** - hot solid/solid Mg extraction + membranes for ions separation and acid/base pair production from salt + aqueous carbonation
- **ÅA route 4 (ÅA4)** - aqueous Mg extraction + membranes for ions separation and acid/base pair from salt + aqueous carbonation
- **ÅA route 5 (ÅA5)** - aqueous Mg extraction + membranes for ions separation and acid/base pair from salt + hot gas/solid carbonation

ÅA1, visualised in Fig. 5, represents the first fully proposed integrated indirect carbonation route developed at the university, in which the process is based on two main steps:

1. Production of magnesium hydroxide (Mg(OH)₂) [g]

2. Carbonation of $Mg(OH)_2$ in pressurised reactor, producing magnesite ($MgCO_3$) [s]

The first step of producing $Mg(OH)_2$ by extraction is achieved by reacting ground serpentinite with AS. The extraction step is conducted at elevated temperatures of approximately 400 to 450°C, which enables conversion of magnesium-bearing phases into water-soluble magnesium sulphate ($MgSO_4$). To produce a solution containing $MgSO_4$, reactive material is dissolved in water while precipitation of $Mg(OH)_2$ is implemented through purification. Ammonia (NH_3) is used to increase pH. Carbonation of $Mg(OH)_2$ occurs after filtration in a pressurised fluidised-bed (PFB) reactor at 500°C over 10-15 minutes at elevated CO_2 partial pressure, resulting in relatively fast reaction kinetics compared to direct silicate carbonation. By-product pressurised steam is formed during the reaction.

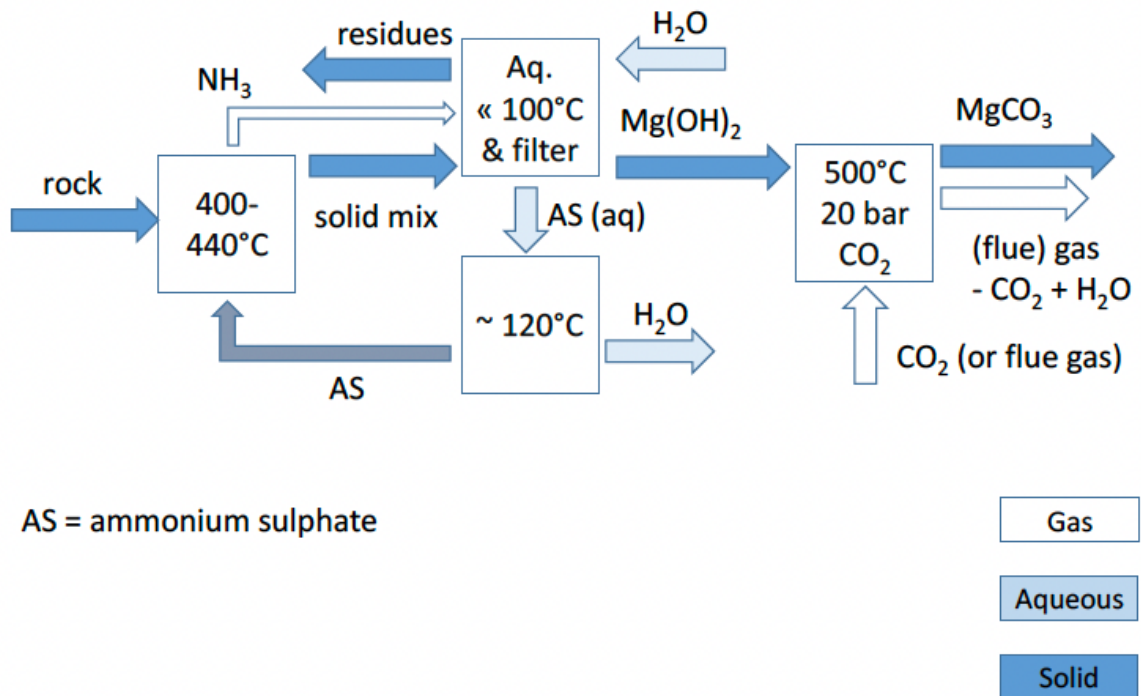


Fig. 5. ÅA1 route focuses on hot/solid Mg extraction combined with hot gas/solid carbonation [50]

Experimental studies report magnesium extraction efficiency in laboratory settings reaches approximately 80%, resulting in a carbonation conversion level of 70-80%. Rapid carbonation rate is considered one of the main advantages in ÅA1 routes as it is enabled by direct operation with CO_2 containing gas streams. It was noticed that this route is experiencing considerable energy demands due to the requirement for high-temperature heat input and significant CO_2 compression. Even though partial heat recovery from the carbonation reactor is possible, the process remains heat-intensive. ÅA1 demonstrates technical feasibility but highlights the need to reduce pressure requirements and overall energy use [50][51][52].



The ÅA2 route (scheme presented in Appendix 1, Fig. 27) was developed to address the high energy and compression demands associated with solid-state carbonation in ÅA1 route. Magnesium is extracted similarly as $MgSO_4$ following the ÅA1, but carbonation occurs in aqueous solution rather than in the high-temperature PFB reactor. CO_2 reacts with the $MgSO_4$ solution to form magnesium carbonate hydrates (MCH), such as nesquehonite (formula 1.4.4.1). To implement ÅA2 process and significantly lower temperatures while avoiding the need for high CO_2 pressure, carbonation approach

is changed in the aqueous phase. The ÅA2 route improves compatibility with diluted f-gas streams and eliminates electricity consumption for compression. Comparing with ÅA1 conversion efficiencies of magnesium to carbonate hydrate are generally higher due to improved mass transfer in solution, moving from 70-80% to reaching 90% [53]. The aqueous process introduces additional complexity in solution handling and chemical recycling. Thermal energy demand is lower than in ÅA1, while the management of salt and the recycling of AS become increasingly important and signifies a shift from heat-intensive carbonation to a more moderate, solution-based approach [54].

After both ÅA1 and ÅA2 identified main burdens of the process and the scientific attention shifted toward improving chemical circularity and reducing the requirement for fresh AS input, ÅA3 was developed (visualised in a scheme attached with Appendix 1, Fig. 28). Marking a further change in the route development, membrane separation technology is introduced in ÅA3 transitioning ÅA research. Considering incorporation of monovalent ion-selective membranes and bipolar membrane electro dialysis (BPMED), ÅA3 shifts towards recover and regeneration of AS. The integration of membrane technologies enables partial closure of the chemical loop and reduces material losses, leading towards a more sustainable approach to mineral carbonation. This configuration allows AS and ammonium hydroxide to be recovered and recycled to support magnesium extraction and pH adjustment. This route benefits from reduced dependency on external chemical supply. In the process, membrane separation introduces additional electricity demand, shifting part of the energy burden from thermal to electrical energy. This route specifically marks the transition toward electrified process optimisation and highlights the trade-off between chemical recovery and energy use [55].

ÅA4 (scheme attached with Appendix 1, Fig. 29) was further developed to support the concept of energy flexibility by investigating magnesium extraction using an aqueous mixture of ABS and AS at lower temperatures. This carbonation approach avoids the need for full thermal decomposition of AS, which reduces high-temperature heat demand. This route relies more heavily on membrane technologies and on the electricity-driven regeneration of chemical agents. BPMED used in ÅA4 produces sulfuric acid and ammonium hydroxide while allowing the process to enable magnesium extraction and pH control without extensive thermal input. ÅA4 is particularly suited to locations where waste heat is limited, but electricity is available. This choice of energy input raises additional considerations, as reliance on renewable energy would be crucial to the process, making the solution more environmentally friendly. The shift from heat-dominated to electricity-dominated energy input reflects broader energy system trends toward electrification, which requires evaluation of electricity-related environmental impacts [56].

After continuously developing ÅA routes, ÅA5 route represents the most integrated and energy efficient configuration developed at the university. This route combines elements of high-pressure carbonation (like in ÅA1) with membrane-based chemical recovery (like ÅA3 and ÅA4), visualised in Fig. 6. enabling adaptation to site-specific energy and CO₂ conditions. In cases where CO₂ concentration is high and compression heat can be recovered, the use of the PFB reactor may remain energetically viable, although alternatively, membrane technologies can be integrated to improve chemical recovery, reducing external region demand. Optimisation based on the availability of heating options and electricity prices, as well as flue gas composition is allowed due to the concept of the hybrid configuration. ÅA5 route represents a mature progress concept emphasising system integration and operational flexibility, but it increases capital intensity and system complexity [50-56].

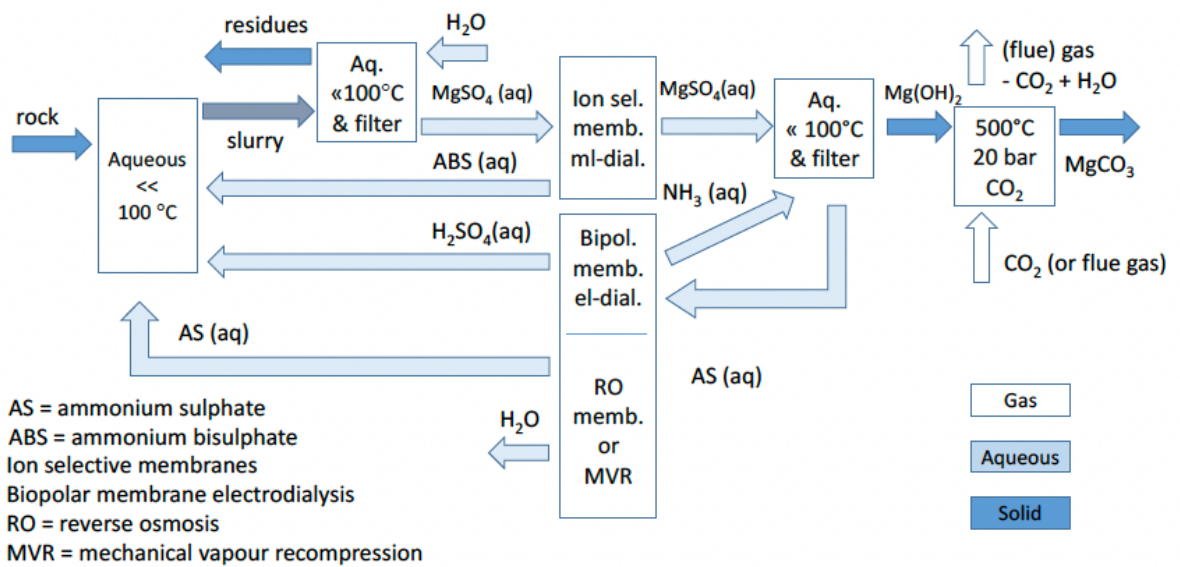


Fig. 6. ÅA5 route represents the integration of previously developed routes optimising the process [50]

Across the routes, the primary engineering focus shifts from proving carbon feasibility to adjusting the efficiency of the process while further minimising energy demand, improving chemical recovery and adapting to different energy infrastructures. The evolution of the routes at ÅA and further research of them in the literature reflects on the increasing importance of system integration and environmental performance in mineral carbonation design. More broadly, academic process concepts often emphasise closed-loop operation and selective extraction with CO₂ sources or waste heat streams. Even though these approaches are typically demonstrated at laboratory or conceptual scale and stay at a low technology readiness levels (TRL<6), they provide valuable insight into how technology processes may be optimised showcasing the diversity of route configurations reported in the literature. These studies form an important basis for comparative assessment, particularly when evaluating mineral-specific routes under consistent assumptions.

1.4.5. By-products, recycling, waste management

During the mineral carbonation waste streams and by-products are generated that can influence both technical feasibility and environmental performance. In magnesium-based routes, the main by-products include silica-rich solids and process solutions containing dissolved salts or secondary materials. The management of these streams is an important step in process design as they can represent either environmental burdens or opportunities for resource recovery.

Silica (SiO₂) is a major by-product of the Mg-silicate carbonation process. During serpentine carbonation, it remains in the form of amorphous or crystalline silica while magnesium is converted to carbonate. The accumulation of it as a by-product in the fine particle phase becomes a valuable residue that potentially can be treated as part of the material and reduce overall solid waste generation [2].

In aqueous carbonation processes, the extraction of magnesium is done with chemical reagents. Saline solutions and AS are generated as the main liquid by-products and brings the need for effective reagent recycling. However, recycling and recovery steps may introduce additional energy and

infrastructure requirements. Recycling chemicals through BPMED introduces both higher financial burdens and greater energy demands. The treatment of byproducts and waste streams is an integral component of mineral carbonation process assessment and plays a significant role in determining the overall sustainability of different process routes [57]. By converting process residues into usable materials waste stream is minimised. It allows the process to function both as CCUS and a pathway to waste utilisation. Integration of waste management is important when mineral carbonation is considered at a large-scale deployment.

1.4.6. Mineral carbonation process requirements

Mineral carbonation processes are influenced by a set of operational requirements. Temperature, pressure, mining and material handling, energy and chemical environment all make a big impact on environment and costs associated. Optimising these can make the process quicker, more efficient and contribute to a net-negative carbon balance.

Temperature and pressure are often used as tools to accelerate reaction kinetics in aqueous processes. According to the study by O'Connor et al. [36] and the report by the International Energy Agency Greenhouse Gas R&D Programme (IEAGHG)[57], elevated temperatures are needed to overcome activation barriers, however industrial feasibility is heavily constrained by energy penalties that are established in the studies. A temperature range from 150°C to 200°C is recommended [58][59]. Increasing pressure improves conversion rates in the commonly used aqueous phase, however it also increases energy demands. To achieve commercially relevant conversion rates, it is necessary to establish conditions that enable trade-offs between reaction efficiency and system sustainability [59].

Chemical additives, such as sodium bicarbonate (NaHCO_3), AS or ABS, could enhance the mineral solubility. They work through shifting pH. However, they introduce additional material flows and create potential recovery challenges. Introducing chemical recovery can substantially minimise the environmental impact associated with virgin chemicals [60].

Mining and material handling involve high requirements. It is established that the stoichiometric requirement for silicate minerals is substantial, and about 1.6 - 3.7 tonnes of rock may be required to sequester 1 ton of CO_2 [2]. The amount of rock needed depends heavily on mineral composition. At this scale, mining and processing demands on such scale begin to compare to existing global mining industries, while the IPCC report notes that such a scale can raise concerns. IPCC [2] raises concerns in the environmental areas that impact local ecosystems. This process on a large-scale leads to significant land disturbance and water usage; however, waste management can be monitored and reused in industrial processes.

Energy demand becomes vital in mineral carbonation, although it is energy-heavy throughout. For mineral carbonation to compete with other CCUS technologies, the evaluation needs to go beyond reaction efficiency and include a system-level considerations such as resource use and environmental trade-offs [61]. Most importantly, upstream energy demand and material inputs have to be considered as both CO_2 capture and transformation have high energy requirements although implementation of direct f-gas operations, removes the expensive part of CO_2 capture step [2][19].

1.4.7. Mineral carbonation limitations

There are several limitations that can affect the performance of mineral carbonation processes. The stability becomes crucial part and despite thermodynamics being favourable during the process, slow reactions kinetics are restraining. Similarly, mineral composition and structure is important for determining dissolution rates and reactivity.

Slow dissolution is one of the most significant challenges in the mineral carbonation process is the slow dissolution rate of Mg-rich silicates such as olivine and serpentine, which can directly limit carbonation efficiency [36][2].

Mineral structure plays an important role in the process. It affects overall reactivity. Magnesium in the minerals is tightly bound to the crystalline silicate lattice. This makes it kinetically resistant to leeching under ambient conditions. It was determined that in aqueous carbonation of forsterite (Mg_2SiO_4), which is a Mg-rich olivine member, the solution often becomes the limiting step in the overall carbonation pathway [36][62].

For direct routes, passivation layers is a significant barrier to reactions. Magnesium leaches from the silicate lattice and silica-rich amorphous layer forms on the surface of the mineral grains. The layer that has formed acts as a barrier for diffusion. The flux of protons (H^+) is reduced with unreacted mineral, hence preventing the migration of Mg^{2+} ions. Secondary precipitation of carbonate minerals on the particle surface further protects the mineral and slows down the reaction. Further research conducted by Dufourny et al. [63] analyses how the transition to industrial scale has to overcome the passivation barrier. It is done through continuous mechanical attrition that can peel away silica-rich layers, proving that maintaining high reaction rates is possible [36].

Kinetics and industrial scalability are interconnected. Slow reaction kinetics have been widely reported in assessments, particularly by IPCC, identifying mineral reactivity as a central issue in larger-scale mineral carbonation deployment [2]. That can also be confirmed by kinetic studies, which demonstrate that without enhancement, especially direct mineral carbonation processes become too slow for industrialisation. Due to these limitations, mineral carbonation routes often introduce a mandatory pre-treatment step [3] which will be discussed later in this study [36].

1.5. Mineral carbonation LCA review

LCA has become a widely used methodological framework for evaluating the environmental performance of CO_2 mineral carbonation systems. Given to the technical complexity of mineral carbonation and the potential trade-offs between CO_2 sequestration efficiency and process-related environmental burdens, LCA offers a structured approach for assessing impacts across the full life cycle of a given process configuration [64][65]. For climate change mitigation, LCA is particularly important for assessing whether mineral carbonation systems deliver net environmental benefits when upstream and downstream processes are considered. Considering CO_2 sequestration processes, permanent storage of CO_2 alone might not guarantee sustainability [66].

For a more analytical LCA, mineral carbonation activities are commonly categorised into upstream (raw material sourcing and preparation) and downstream (processing, transport, and disposal) [67]. Upstream processes like pre-treatment represent a significant carbon penalty due to high energy demand, becoming detrimental to the overall environmental assessment performance [68]. Choosing

which activities to include in an LCA system boundaries is important for determining how efficient it is at CO₂ sequestration falls on the methodological choices. Evaluating only the amount of CO₂ mineralised is insufficient to assess overall sustainability, which is why a life-cycle perspective is required to account for the cumulative impacts across the entire system [17].

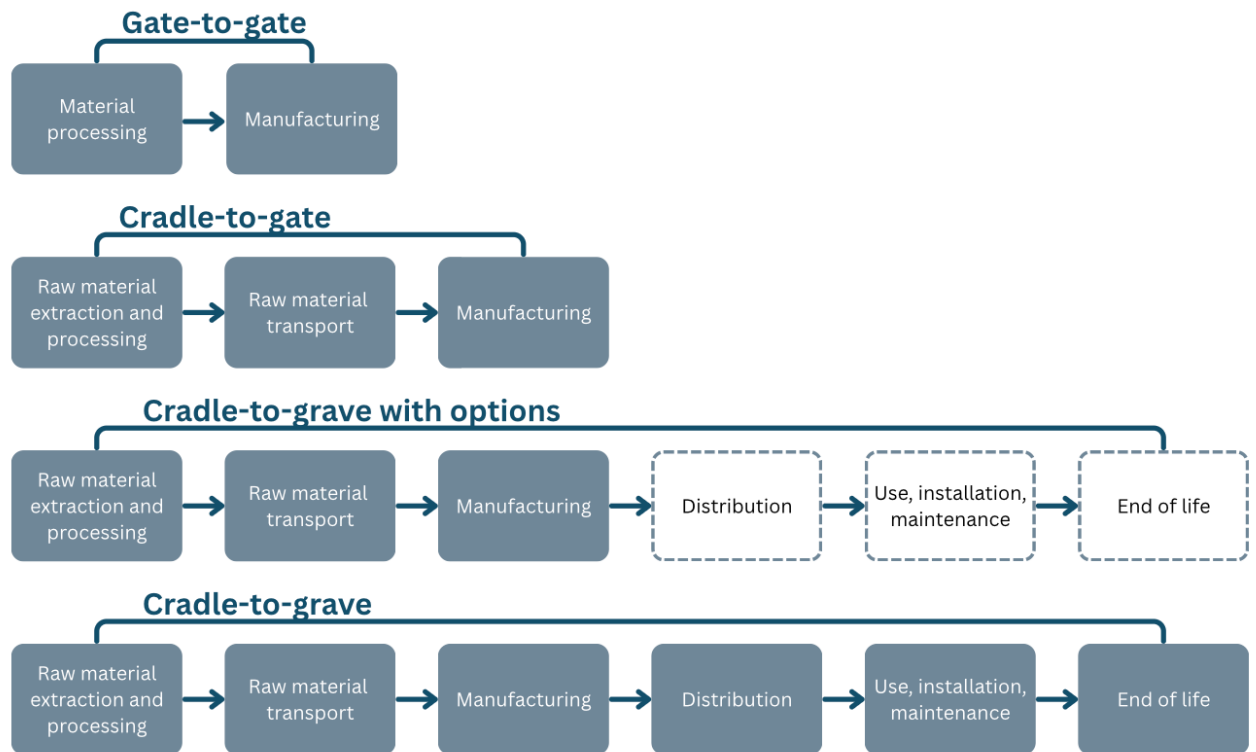


Fig. 7. LCA system boundaries expressing the choices made for evaluation (reproduced from [69])

As shown in Fig. 7, system boundaries determine which processes are included in the assessment. The most common types are cradle-to-gate and cradle-to-grave, while cradle-to-gate with options offers the opportunity to adjust certain factors in the calculations [69]. However, there is an option of using gate-to-gate which is limiting and only focuses on the manufacturing itself. Choosing system boundaries is based on the needs of the research and what needs or can be included in the modelling. Selecting FU is equally important as it provides transparent basis for impact assessment and comparison between modelled processes. The FU is defined by the ISO 14040/44 standard [70].

Existing LCA studies on mineral carbonation cover a wide range of process configurations and mineral feedstocks, with the majority focusing on ex-situ mineral carbonation routes while using tailings for the process. Additionally, research focuses on the processes which create products like cement to be used in the industry. These routes explicitly include mining, mineral processing, carbonation reactions, and product handling within the defined system boundaries. Evaluation of climate change impacts is usually expressed as global warming potential (GWP) per unit of CO₂ captured and serves as the primary impact category in most studies. Several studies extend beyond GHG emissions and include additional environmental indicators such as cumulative energy demand or resource depletion, allowing for a broader analysis of the environmental impacts. Despite the volume of research [71], there are still gaps in methodological inconsistency, process integration and uncertainty analysis which stems from assessments that examine isolated process concepts without a direct comparison across multiple mineral feedstocks under identical methodological conditions [71]. That approach creates differences throughout the reported mineral carbonation performance, and as

a result, it may reflect methodological choices as results rather than material or process characteristics. Similarly, process integration is uncommon, with rare studies like Nduagu et al. [72] that have integrated process routes combining extraction, separation, carbonation and closed-loop configurations. These systems may exhibit different environmental trade-offs compared to conventional direct carbonation approaches. There is a common gap in the literature as not all studies apply uncertainty and sensitivity analysis that are needed to validate the research calculations. These limitations underscore the importance of comparative and systematically structured studies that apply consistent assumptions across alternative mineral systems and process configurations.

1.6. Summary of the literature review

The literature review investigated the role of CO₂ mineral carbonation in climate change mitigation, while focusing on its use as a pathway for permanent CO₂ storage. The main mechanisms of this technology, which include reaction thermodynamics, limitations and process configurations, are discussed to establish the scientific basis for mineral-based CO₂ mineral carbonation. Mg-rich silicate minerals are identified as a key feedstock due to their abundance in the world and ability to form stable carbonate phases. Serpentine and phlogopite represent two distinct mineral systems that are similar in chemistry but with different structural and reactivity characteristics.

This review further examined mineral selection which has an important influence on process design. It includes pre-treatment that are combined for more enhanced process concepts, pathways focusing on different systems and by-product management. All these process combined further determine environmental impact of the mineral carbonation system. Academic processes (at the TRL 4-5) that may include integrated routes proposed in the literature, show a wide range of approaches aimed at overcoming kinetic limitations while improving process efficiency. Academic routes, particularly ones proposed by ÅA, differ in terms of energy demand and material inputs, creating different system complexity and further showing the need for systematic environmental assessment. To address these considerations, LCA emerged as a critical tool to evaluate beyond theoretical CO₂ uptake and fully identify key environmental tradeoffs.

Despite the growing number of LCA studies on mineral carbonation, several gaps remain in the existing literature. Many published assessments focus on single minerals or individual process routes. Having just single process with one mineral limit the comparability of results across different possible mineral feedstocks. Considered together with inconsistencies in FU, system boundaries and methodological assumptions make direct comparison between studies difficult. Comparative LCA that evaluate different Mg-rich silicate minerals under equal system assumptions remain sparse, resulting in a limited understanding of how mineral-specific properties influence the overall environmental performance of mineral carbonation routes.

The research reported in this thesis, therefore, aims to contribute to the existing literature by addressing the limitations and supporting the development of mineral carbonation as a viable climate change mitigation strategy. Addressing these gaps requires a comparative LCA framework that applies consistent methodological choices to evaluate serpentine- and phlogopite-based mineral carbonation routes. This approach enables systematic comparison of mineral scientific pathways and provides insights into the role of mineral selection in determining the sustainability of CO₂ mineral carbonation processes.

2. Life cycle assessment research methodology

This study applies the Life Cycle Assessment (LCA) framework in accordance with the standards of the International Organisation for Standardisation (ISO). Standards that are consistently referenced in the study are 14040:2006 and 14044:2006. These standards establish principles, requirements, and guidelines for conducting and reporting of an LCA [73][74]. A systematic structure is used to implement LCA, which consists of:

- I. Goal and Scope Definition
- II. Life Cycle Inventory Analysis
- III. Life Cycle Impact Assessment
- IV. Interpretation

Fig. 8. further emphasises the stages of LCA, which are interconnected by two-way lines with other phases, allowing constant iterative analysis. Allowing continuous cyclical improvement from feedback of phase changes during the performance of the LCA furthers the tool's accuracy [75].

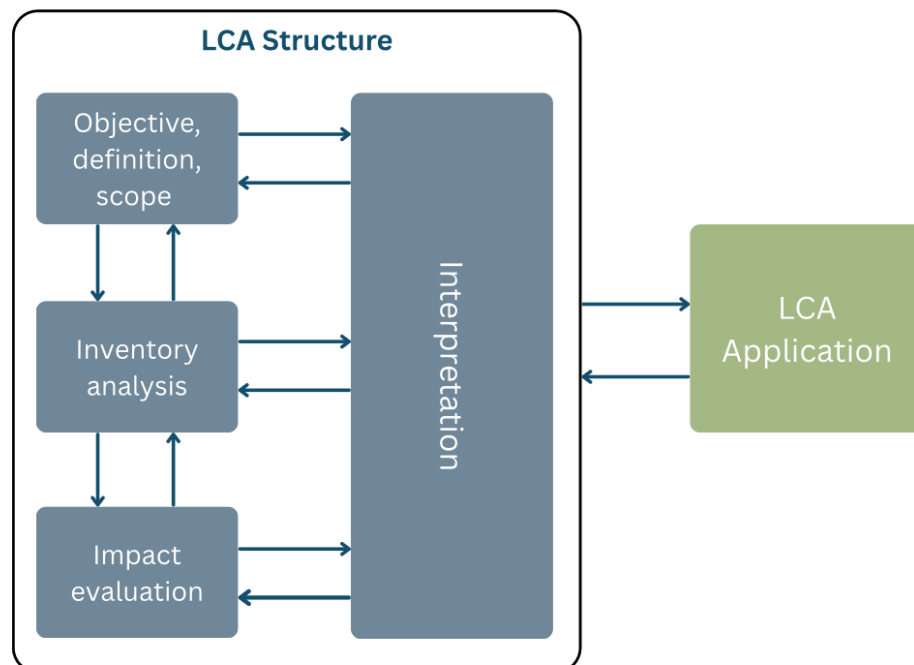


Fig. 8. LCA structure emphasising connectivity of the phases (reproduced from [75])

SimaPro 9.0 software is used in the study to perform LCA, as it is a software used for environmental modelling and scenario analysis [76]. Data required for calculations, including energy, materials, and transport, were sourced from the Ecoinvent database, as it demonstrates regional representativeness for European production conditions and enables modelling that is closer to reality. For the environmental impact modelling, the IMPACT 2002+ method was chosen as it integrates both midpoint and endpoint indicators across categories. They include human health, ecosystem quality, climate change and resource depletion and prioritised over the more widely used method ReCiPe which doesn't include climate change as a separate endpoint. Methodology ensures consistency and enables reproducible comparison of the environmental performance [5] while sensitivity analysis validates the viability of results. Following the Life Cycle Impact Assessment stage, using both deterministic parameter variation guarantees that conclusions drawn are scientifically sound and statistically supported [73].

2.1. Defining Goal and Scope

2.1.1. Goal and Scope

Defining the goal and scope is the first and fundamental step in performing an LCA, as this step establishes the study's purpose, context and methodological framework [73][74]. The goal of the study explains why the LCA is conducted and its intended audience, while the scope defines the system boundaries for the FU. Furthermore, the scope assumptions and impact methods are considered and combined to ensure consistency across all phases of the LCA. A clearly defined goal in scope ensures that conclusions align with the study's objectives and intended applications [73][74].

The goal of the study stems from a gap in the literature: a lack of comparison of the environmental performance of different feedstocks across different CO₂ mineral carbonation routes. Serpentine and phlogopite are the feedstocks evaluated in this study. Both feedstocks use various processes to produce magnesium sulfate (MgSO₄), which is later carbonised. This is implemented through a multi-stage process that includes extraction, carbonation, Bipolar Membrane Electrodialysis (BPMED), and by-product metal oxides purification. Notably, this demonstrates that comparisons aim to identify environmental hotspots and quantify potential trade-offs to determine which route and feedstock offer the greatest potential for the most sustainable CO₂ utilisation. A key consideration in this study is that the work is based on laboratory operations reflecting Nordic European conditions, as mineral carbonation and feedstocks are located in Finland. Specifically, mineral carbonation is occurring at ÅA University, while feedstocks are sourced from Vammala (serpentine) and Siilinjärvi (phlogopite) in Finland. No by-products in the study are currently requiring waste allocation, as they are not generated in a way that would result in waste or reuse in new product creation.

2.1.2. Defining scenarios for LCA

Defining scenarios becomes a critical part of the LCA when different mineral carbonation approaches are expected to be compared. For this study, three distinct process configurations (scenarios) are being modelled based on the ÅA mineralisation routes established earlier. Scenarios will include two alternative serpentine base routes and one phlogopite base route. Having two serpentine scenarios which vary in the mineral extraction step approach allows us to establish the most environmentally friendly route. As for the phlogopite-based route, it will follow a single optimised pathway that heavily relies on the already-developed serpentine route and will adopt the main parts of it.

Scenarios that were chosen:

Scenario 1 is the only one which focuses on heat usage during the dry extraction phase based on ÅA3 (discussed in page 25 with a scheme visualised in Appendix 1, Fig. 27) and is performed with serpentine. BPMED is implemented in this route to allow for recirculation of salts and reagents.

Scenario 2 focuses on serpentine, but implements ÅA4 (discussed in page 25 with a scheme visualised in Appendix 1, Fig. 28) with the use of a wet extraction step, which uses electricity instead of heat. BPMED is implemented in this route to allow for recirculation of salts and reagents.

Scenario 3 is the only scenario with the use of phlogopite. It uses the same extraction and carbonation approach as scenario 2, based on ÅA4. Using the same approach as in the other

scenario, here, phlogopite, as a mineral, is tested while eliminating other variables. BPMED is implemented in this route to allow for recirculation of salts and reagents.

All of the scenarios will go through the same phases, eliminating factors that could further affect the calculations. The main differences that will affect performance are heat use in scenario 1 during the mineral extraction phase and phlogopite use in scenario 3. After the main calculations are performed, a new scenario will be introduced:

Scenario 4 will focus on the best-performing scenario from established scenarios 1-3 (selected from the results). This scenario will eliminate the use of BPMED and recirculation of materials, focusing on virgin chemicals (reagents and salts bought and discharged for every mineral carbonation cycle) in the process and calculates on MgSO_4 extraction and mineral carbonation phases. Having only virgin chemicals in the process allows for comparison with already established calculations for BPMED inclusion [78][79].

2.1.3. Functional unit identification

For methodological consistency between the mineralisation routes and reproduction of environmental performance evaluation, it is essential to determine the FU [73][74][80]. It provides a quantitative reference that all inputs and outputs are based on the same criteria, reinforcing the credibility of comparing different systems on an equivalent basis. The FU in this study is defined as “1 kg of CO_2 captured”. This FU was selected because the study's purpose and scenarios are to capture and chemically stabilise CO_2 as solid magnesium compounds. By implementing the FU based on the amount of CO_2 captured, rather than the mass of product (MgSO_4) allows for a direct and fair comparison of carbon capture efficiency. Unifying FU to “1 kg of CO_2 captured” allows for the evaluation of energy demand per unit of CO_2 sequestered and environmental burdens.

2.1.4. System boundaries

While the more common approaches to determining system boundaries in LCA are expressed in Fig. 6 as cradle-to-gate, cradle-to-gate with options and cradle-to-grave [69], it is hard to apply either of these to the LCA performed in this study. Given that the LCA is for laboratory-scale mineral carbonation rather than the industrial application, it is important to identify the correct system boundaries. Defining system boundaries within LCA establishes the link between the product and the environmental exchange. At this stage of the analysis, the gate-to-gate system boundaries are most appropriate for the expected LCA. It stems from critical aspects of the study of real-life situations. Preferably, for this type of LCA, cradle-to-gate should be used, however, neither phlogopite nor serpentine is mined or transported for this study. Including mining and all associated effects of the process are not calculated, hence limiting the system boundaries. The feedstock is only crushed, and ÅA provides information on the energy required for grinding. Both serpentine and phlogopite routes are modelled in four stages and shown in Fig. 9.

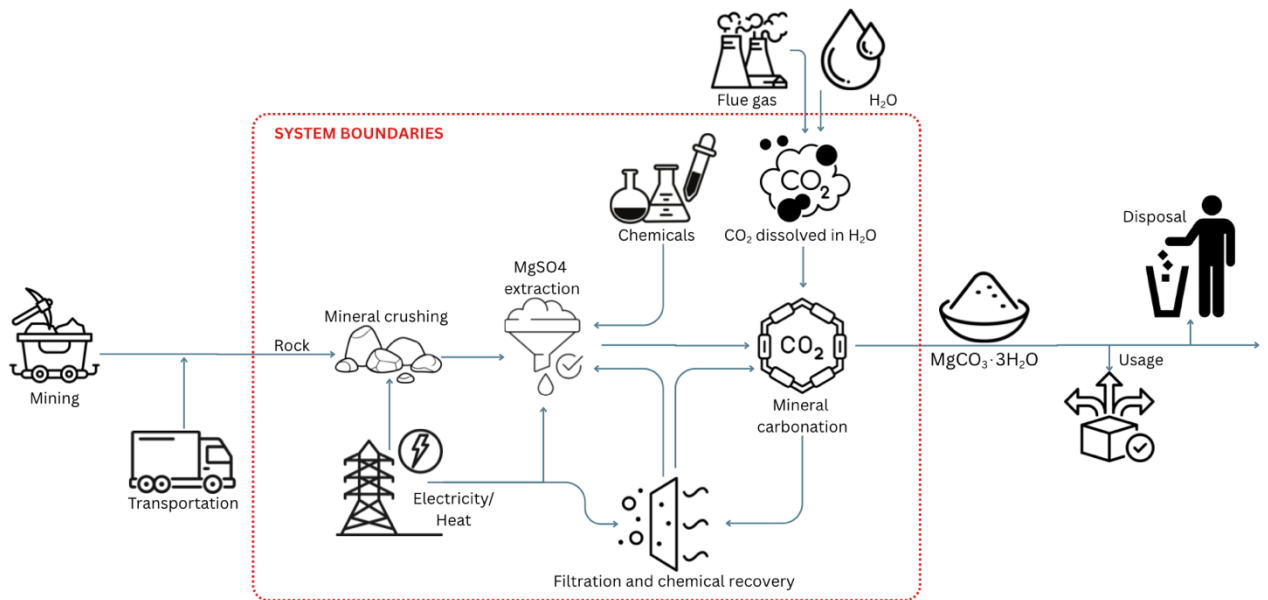


Fig. 9. System boundaries focusing on stages included in LCA focused on gate-to-gate approach

Mineral carbonation is the main process of the study, included as the second stage in the LCA. This phase consists of combining different stages into one process and has these process included:

MgSO₄ extraction represents the initial stage in the LCA and is a major contributor to energy use. It includes feedstock (rock) with crushing process, water and energy source as inputs, while silica (SiO₂) and MgSO₄ are outputs. Recirculated BPMED chemicals are also considered as an input;

BPMED looping to avoid purchasing virgin chemicals with its main input being electricity required to separate chemicals;

Metal precipitation is included separately as a last step in mineral carbonation LCA and is modelled separately.

2.2. Life cycle inventory analysis

Life Cycle Inventory (LCI) is the second step of LCA. It involves a systematic collection and quantification of all inputs, material and energy flows, machine and waste allocations. This is done within the system boundaries defined in the first step. It forms the foundation and directly influences the accuracy of the results. The LCI was established to ensure completeness and transparency in line with the requirements and framework of ISO 14044:2006 [74]. As this study uses two feedstocks, serpentine and phlogopite as magnesium sources, it is vital to determine what inputs and outputs are required for each feedstock and route.

Table 1 shows a preliminary view of the inputs required for the scenarios established earlier. Due to scenario similarities, everything presented in Table 1 will be continuously used throughout all of the scenarios. However, the chemical that is used in the mineral extraction stage differs and will be adjusted according to the chemical used. Scenario 1 use AS for dry mineral extraction, while scenarios 2 and 3, which involve wet mineral extraction uses sulfuric acid (H₂SO₄). Calculations were performed during the internship at ÅA using laboratory data and subsequently adjusted based on stoichiometric calculations.

Table 1. Expected inventory table for inputs and outputs for mineral carbonation of expected primary scenarios

PRODUCT	UNIT	SCENARIO 1	SCENARIO 2	SCENARIO 3
NESQUEHONITE	kg			
CO ₂ , CAPTURED	kg			
INPUT ITEM				
MGSO ₄	kg			
AMMONIA, LIQUID	kg			
WATER	kg			
CARBON DIOXIDE, LIQUID	kg			
TREATMENT OF (NH ₄) ₂ SO ₄ , BY BPMED	kg			
TREATMENT OF METALS	kg			
ELECTRICITY MIX	kWh			
OUTPUT ITEM				
CO ₂ FOSSIL EMISSIONS	kg			

2.2.1. Energy mix limitations

During this study, Ecoinvent database was used for LCA, however it was noted that database for energy mixes were from 2012. Häggqvist and Zevenhoven overview the impact of the LCA using data from 2014 in Ecoinvent v3.5 accentuating the difference between 2014 (247g CO₂ eq/kWh) and 2025 (72g CO₂ eq/kWh) data [86]. Basing calculations off of 2012 database in this study would create discrepancies across final interpretation due to the change in energy sources. Following the released data from official statistics of Finland (OSF) [81] it is noticeable how using data from 2012 could be inefficient and inaccurate across this study. Based on the Table 2 in 2012 fossil fuels were at 46.1% of the whole energy share which leads to higher emissions when compared with 28.9% in 2024. Additionally, renewable energy and nuclear energy went from 31.3% and 17.5% respectively in 2012 to 43.4% and 26.2% respectively in 2024 leading to cleaner energy mix with lesser emissions.

Table 2. Total energy use and source share comparison for Finland focusing on 2012 and 2024 (recreated from [81])

	2012		2024	
	Quantity (TJ)	Share (%)	Quantity (TJ)	Share (%)
Total energy use	1375591	100	1296115	100

	2012		2024	
	Quantity (TJ)	Share (%)	Quantity (TJ)	Share (%)
1 Renewable energy	429957	31.3	561987	43.4
1.1 Hydro power	60001	4.4	50876	3.9
1.2 Wind power	1780	0.1	72852	5.6
1.3 Wood fuels	330549	24	355576	27.4
1.4 Other renewable energy	37627	2.7	82683	6.4
2 Fossil fuels and peat	634540	46.1	374960	28.9
2.1 Oil (fossil)	322374	23.4	242254	18.7
2.2 Coal	122651	8.9	53413	4.1
2.3 Natural gas	114985	8.4	45854	3.5
2.4 Peat	66355	4.8	19968	1.5
2.5 Other fossil fuels	8175	0.6	13471	1
3 Nuclear energy	240685	17.5	339582	26.2
4 Net imports of electricity	62796	4.6	11452	0.9
5 Others	7614	0.6	8134	0.6

2.3. Life cycle impact assessment

Life Cycle Impact Assessment (LCIA) is a mandatory phase of LCA, which converts the elementary flows collected in LCI into indicators [73][74]. Depending on the chosen method, indicators describe potential environmental impacts, focusing on human health, ecosystem quality, climate change, and resource depletion. Throughout this step, emissions and resource use are closely linked to environmental mechanisms as they are derived from scientific characterisation factors, where results represent potential impacts rather than actual. It can be inferred that the scale and direction of pressure on the environment, but not precise real-world outcomes, are considered. Within LCA, the LCIA stage follows the guidelines of ISO 14044:2006 [74], which defines the structure of characterisation, normalisation, and weighting within the framework. It shows that characterisation is a mandatory step, while subsequent ones may be optional depending on the goal and scope. Within this study, all of the steps are taken into account to strengthen the reliability of the results.

LCA can be done through different calculation methods, and for this study, IMPACT 2002+ was chosen and implemented because it integrates both midpoint and endpoint approaches, allowing for a detailed analysis of effects specific to the category alongside damage level results as it gives climate change as a separate endpoint. This structure makes it easier to identify environmental problem points at the midpoint level while supporting overall performance comparisons of the factors. This is important to analyse serpentine and phlogopite-based CO₂ mineralisation routes at the endpoint level [76][77][78].

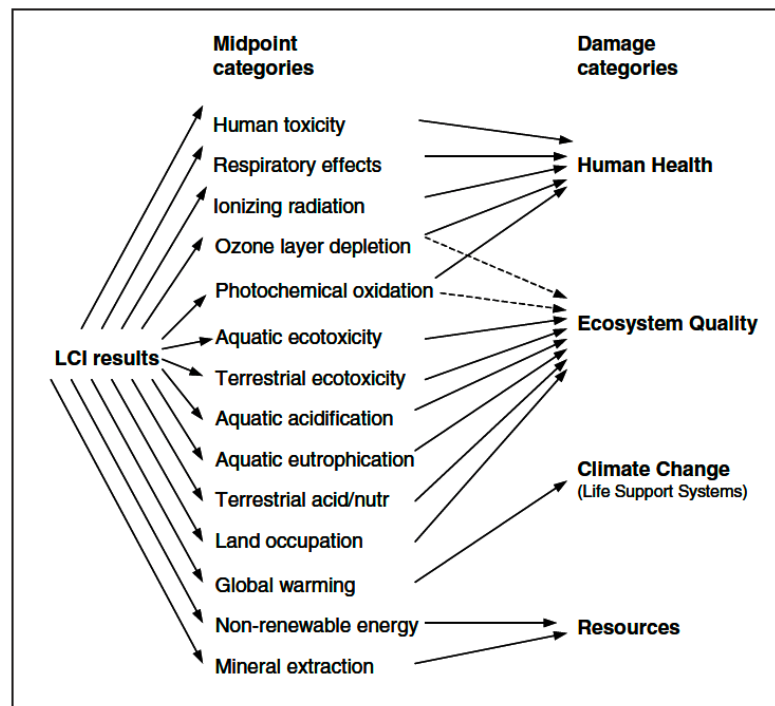


Fig. 10. IMPACT 2002+ categorisation with 14 midpoint categories of impact, grouped into four endpoint categories [76]

Fig. 10, the IMPACT 2002+ method comprises of 14 midpoint categories of impact, which are grouped into four endpoint categories [76]. These categories are described as damage categories:

- I. Human health, measured in *Disability Adjusted Life Years (DALYs)*;
- II. Ecosystem Quality, measured in *Potentially Disappeared Fraction of species over area and time (PDF·m²·year)*;
- III. Climate Change, measured in *kg of CO₂ equivalent*s;
- IV. Resources, measured in *MJ of primary energy*.

IMPACT 2002+ allows for each endpoint category to be made of one or more midpoint categories. Characterising converts emissions into equivalence within the relevant categories using impact factors, and normalisation allows for the comparison of impacts that were received to reference values, while weighting will support the reflection of the importance of each damage category. Moreover, the single score aggregation step collects all results into a single overall indicator, expressed in points (Pt). By doing so, it allows to have comparison between factors and maintain transparency through categories that are important [76]. LCIA results vary depending on the method chosen, as different models may yield varying values. Interpretations in this study emphasise relative differences between alternatives rather than absolute numerical values. Acknowledging that while the survey incorporates scientifically established models, it represents a potential impact assessment rather than a prediction and correctness depends on good interpretation. This phase in the study provides the foundation for identifying environmental hotspots and quantifying potential damage. Together, they support the comparative interpretation of feedstock-based CO₂ mineralisation routes.

2.4. Interpretation of results

Interpretation is the final step of LCA, integrating the results of the inventory and impact assessment to derive conclusions. Consistent with ISO 14044:2006 requirements [74], it is ensured that the outcome of LCA is reliable and includes three main steps:

- I. Identifying significant issues;
- II. Evaluating completeness and sensitivity;
- III. Formatting conclusions and recommendations.

Interpretation can be performed through contribution analysis of both midpoint and endpoint categories, as this identifies the dominant life cycle stages and environmental hotspots within feedstock-based CO₂ mineralisation routes. The evaluation stage includes checking the data and verifying the consistency across system boundaries. Sensitivity analysis of key parameters, such as energy mix and extraction efficiency, along with chemical usage, is conducted as part of the final interpretation step. Within LCIA, this stage plays a crucial role by connecting software results to decision-making insights. By continually testing the validity of results and highlighting key impact drivers, this interpretation will allow the laboratory to improve process routes and minimise CO₂ emissions and environmental impact overall. This allows the findings across different mineralisation routes and feedstocks to be coherent and to maintain uniform assumptions across scenarios [82].

2.5. Sensitivity analysis

Last step for the LCA would be Sensitivity Analysis (SA), as it is mentioned in ISO 14044:2006 being a key methodological step and is done to evaluate how correctly the study was performed [74]. Based on SA applications, it determines which inputs influence the final results in the study and is used to evaluate how variations in input data or assumptions influence the final results. Such modelling enables the study to produce transparent, scientifically defensible results, as LCA models rely heavily on assumptions. By doing so, the study captures that comparative conclusions remain valid under sensitivity [74]. To test the stability of results between feedstock-based CO₂ mineralisation routes SA is performed after the LCIA stage. One-at-a-time SA conducted by systematically varying selected parameters (electricity mix, extraction, carbonation, chemical recovery) within predefined ranges, while holding all other parameters consistent to assess their individual influence over the whole range of modelling.

3. Results

3.1. Goal and scope definition results

During the methodology stage of this research, FU was set as „1 kg of CO₂ captured“, during the LCI stage, several reference units were added which eventually account for the main FU. It was established in this study, that mineral carbonation LCA cannot be performed in one stage so to perform a LCA correctly, process-stages were created. Both serpentine and phlogopite routes are modelled in four stages, and in each stage different reference unit was set internally as to simplify initial modelling. FU that were established throughout the stages:

Mineral carbonation with the FU of the whole study set as “1 kg of CO₂ captured“ and within itself includes these processes:

MgSO₄ extraction in which reference unit was set while calculating 1kg of MgSO₄. Creation of this step was based of ÅA established calculations and LCA modelling [83];

BPMED looping to avoid purchasing virgin chemicals in with reference unit of 1kg of treatment of NH₄OH sent to BPMED;

Metal separation, which focused on removing metals from MgSO₄ extraction, and is calculated on the unit of 1kg of metals sent to waste sludge.

Fig. 11, 12 and 13 show the flow diagrams of each scenario established in methodology section. Flow charts are recreated from existing ÅA route diagrams [50] with the adjustments needed for LCA breaking down processes into stages.

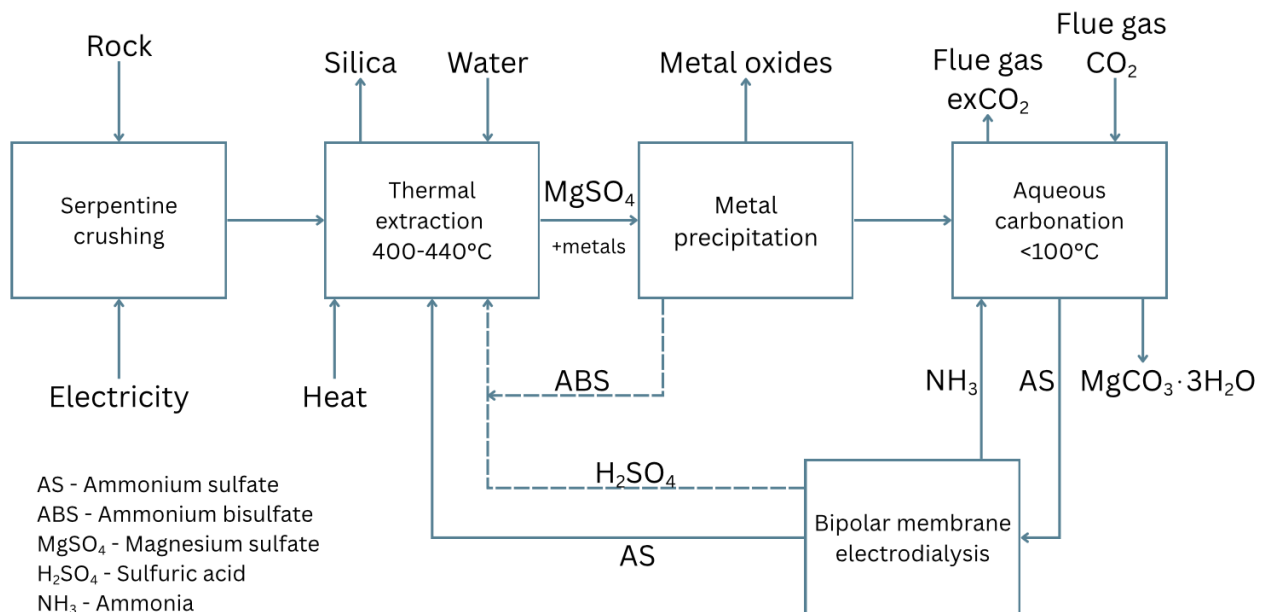


Fig. 11. System boundaries of defined scenario 1 (adjusted based on the LCA stages from [50])

Visualised scenario 1 uses heat for MgSO₄ extraction through thermal activation, while both scenarios 2 and 3 have electricity input and an aqueous activation process. Transport was considered, but it was established as barely negligible as the locations from which feedstock originates don't exceed long distances (within a 400 km radius), and it doesn't alter the impact of the LCA significantly [54].

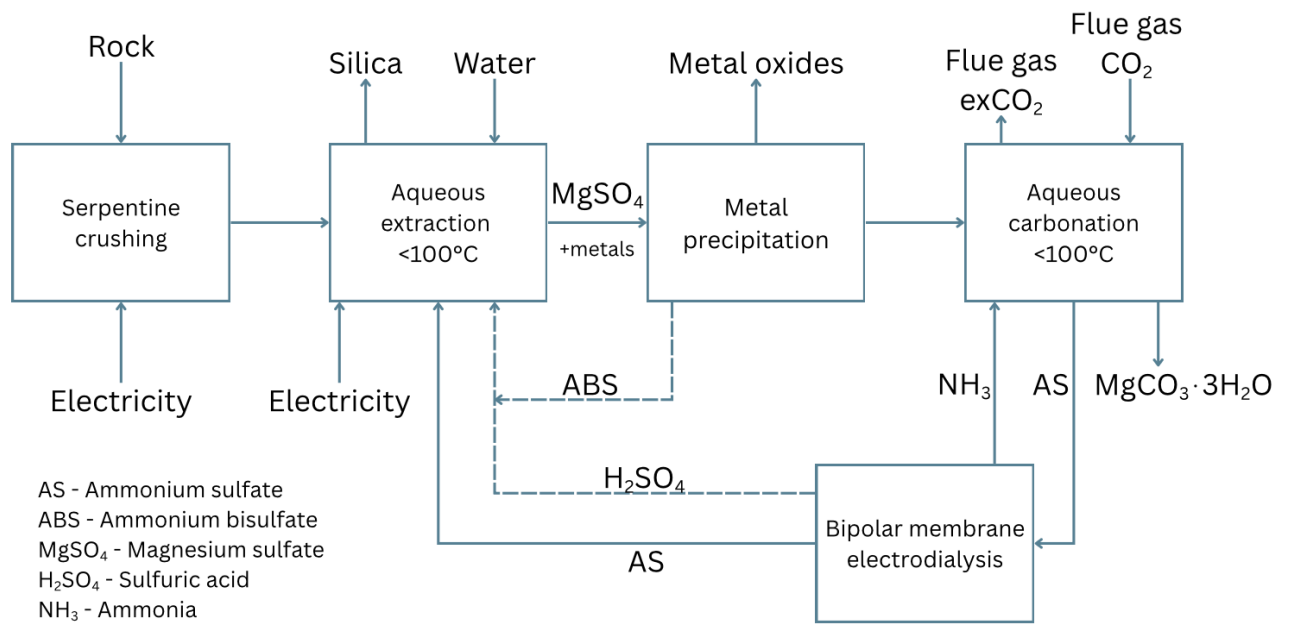


Fig. 12. System boundaries of defined scenario 2 (adjusted based on the LCA stages from [50])

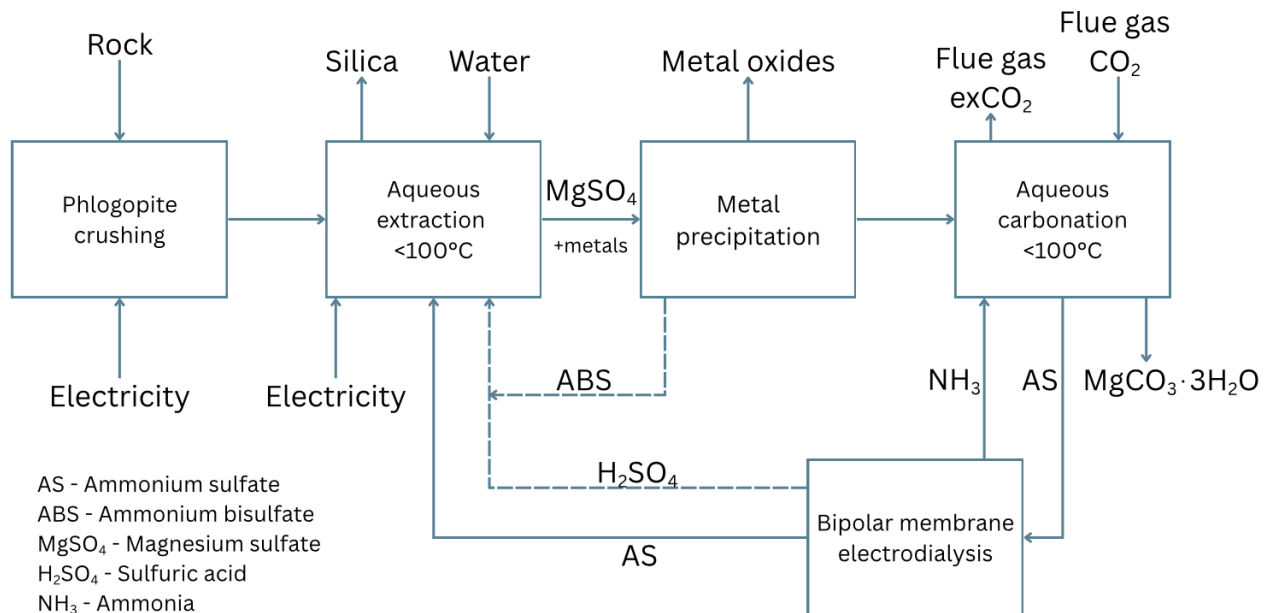


Fig. 13. System boundaries of defined scenario 3 (adjusted based on the LCA stages with phlogopite as main mineral from [50])

3.2. Life cycle inventory analysis results

During the research process, it was noted that Ecoinvent database has products from nature and they include feedstocks but inputs are empty bringing to decisions how these feedstocks will be modelled and what has to be included in the system boundaries. As the calculations and modelling are implemented from a project in ÅA laboratories, feedstocks are received from Vammala (serpentine) and Siilinjärvi (Phlogopite). Mining isn't done by the research team, hence including the mining process in LCA would be detrimental to the realities of this study. Serpentine and phlogopite crushing are included with the data received from ÅA and represent 1 tonne of material crushed. ÅA has determined that to crush 1 tonne of feedstock, 19 kWh of electricity is used.

Inventory was finalised based on the phases and their modelling sequence and are represented in the Tables 3, 4, 5 and 6. Data was gathered and calculated based on the information provided by ÅA during internship under Research connect of Finland - Academy Project 34785 (Viable magnesium ecosystem: Exploiting Mg from magnesium silicates with carbon capture and utilization) (MAGNEX) (2022–2025) and the Business Finland project REMIS (2025-2027) for phlogopite. Finalisation of calculations and inventory focused on idealistic approach to mineral carbonation under laboratory conditions and stoichiometric calculations.

Table 3. Finalised inventory for mineral carbonation with calculations based on FU “1 kg of CO₂ captured“

PRODUCT	UNIT	SCENARIO 1	SCENARIO 2	SCENARIO 3
NESQUEHONITE	kg	3.144	3.144	3.144
CO ₂ , CAPTURED	kg	-1	-1	-1
INPUT ITEM				
MGSO ₄	kg	2.735	2.735	2.735
AMMONIA, LIQUID	kg	0.744	0.744	0.744
WATER	kg	0.819	0.819	0.819
CARBON DIOXIDE, LIQUID	kg	1	1	1
TREATMENT OF (NH ₄) ₂ SO ₄ , BY BPMED	kg	3.003	3.003	3.003
TREATMENT OF METALS	kg	0.1	0.1	0.1
ELECTRICITY MIX	kWh	0.1	0.1	0.1
OUTPUT ITEM				
CO ₂ FOSSIL EMISSIONS	kg	1	1	1

Table 4. intermediate inventory for stage 2 focusing on MgSO₄ extraction

PRODUCT	UNIT	SCENARIO 1	SCENARIO 2	SCENARIO 3
MGSO ₄	kg	1	1	1
INPUT ITEM				
CRUSHED SERPENTINE	kg	0.767	0.767	-
CRUSHED PHLOGOPITE	kg	-	-	1.156
H ₂ SO ₄	kg	-	0.816	0.951
AS	kg	1.099	-	-
HEAT AT 400–440°C	kWh	0.94	--	-
ELECTRICITY MIX	kWh	-	0.91	0.91

OUTPUT ITEM	UNIT	SCENARIO 1	SCENARIO 2	SCENARIO 3
SILICA	kg	0.333	0.333	0.499
WATER	kg	0.249	0.249	0.05

Table 5. intermediate inventory for phase 3 of BPMED looping

PRODUCT	UNIT	ALL SCENARIOS
TREATMENT OF (NH ₄) ₂ SO ₄ , BY BPMED	kg	1
INPUT ITEM		
WATER	kg	0.273
(NH ₄) ₂ SO ₄ TO BPMED	kg	1
ELECTRICITY MIX	kWh	0.849
OUTPUT ITEM		
H ₂ SO ₄ - AVOIDED PRODUCT	kg	0.742
AMMONIA - AVOIDED PRODUCT	kg	0.258

Table 6. intermediate inventory for phase 4 of Metal separation

PRODUCT	UNIT	ALL SCENARIOS
TREATMENT OF METALS	kg	1
INPUT ITEM		
WATER	kg	0.140
AMMONIA, LIQUID	kg	0.133
TREATMENT OF (NH ₄) ₂ SO ₄ , BY BPMED	kg	0.515
ELECTRICITY MIX	kWh	0.05

3.3. Life cycle impact assessment results

This chapter focuses on results received after modelling each scenario based on the determined methodology prior in this study. Characterisation of the results will be the main aspect as it is a mandatory part while damage assessment and normalisation will be included to support the collected results. Characterisation results are presented in the Fig. 9 and represent raw environmental data translated into standardised environmental impact scores. In results section, the primary importance for this modelling is placed on global warming and non-renewable energy midpoint categories as they

collectively focus on products' possible impacts while it contributes to climate change and ties with the mineral extraction and land use.

3.3.1. Scenario 1

After finalising inventory results, scenario 1 was modelled with the material flows illustrated in Appendix 2 Fig. 30. From the characterisation results, presented in Fig. 14, it is noticeable that chemical inputs and energy use with BPMED as well as MgSO₄ are the highest contributors for environmental burdens. BPMED across all of the midpoint categories stays consistent between 35% and 65% while MgSO₄ at 20% to 40% of the impact. Having such high percentages in characterisation of the process solidifies them as primary environmental drivers.

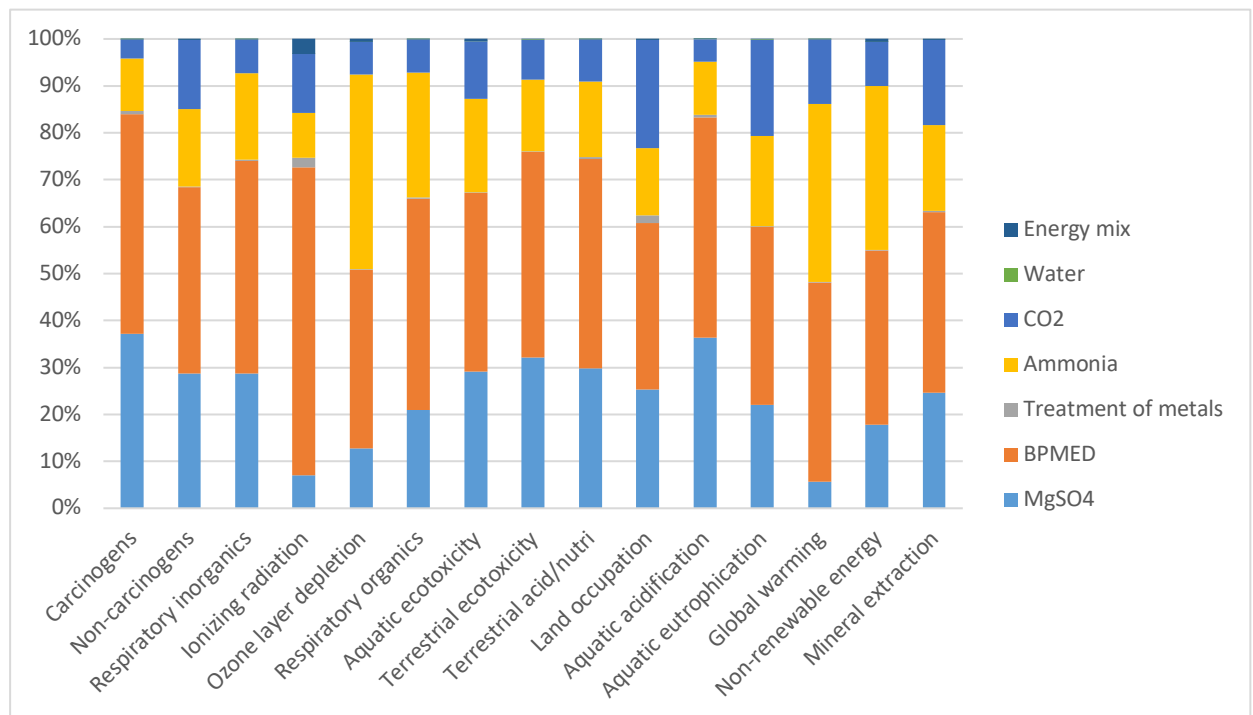


Fig. 14. IMPACT 2002+ characterisation results for scenario 1

Although, electricity is hardly seen from the data, it could suggest the sensitivity to consumption of it, particularly when the BPMED process heavily relies on it. Similarly, CO₂ like electricity, is non dominating across the data consistently staying below 20% with more midpoint categories leaning less than 10%. It is important to note that BPMED is dominating across multiple midpoint categories. Particularly, ionising radiation reaches 66% of the impact. Even though, BPMED is heavily influencing process environmental burdens, there are multiple process drivers indicated by land occupation and global warming.

Previously mentioned MgSO₄ at 20% to 40% becomes a big influence in scenario 1 which can be further broken by characterisation expressed in Appendix 2 Fig. 33. There is a systematic dominance of AS across all of the impact categories reaching almost 80% in categories like carcinogens, non-carcinogens, respiratory inorganics, aquatic ecotoxicity, terrestrial ecotoxicity, terrestrial acid, land occupation and aquatic acidification. Acknowledging such dominance of AS brings the conclusion of environmental burdens being driven by chemical recovery systems. Previously mentioned electricity burdens are noticeable in high energy demands noted in literature for production and recovery of the chemical [2][19]. Subsequently, ammonia used in the process consistently stays at 20% to 40%

indicating dependence on chemical inputs within the process while other inputs expressed in characterisation suggest the need for optimisation and prioritisation of chemical recovery rather than physical processing adjustment.

3.3.2. Scenario 2

Scenario 2 was modelled with the material flows illustrated in Appendix 2 Fig. 31. Scenario 2 characterisation visualised in Fig. 15 shifts from scenario 1 with one primary environmental driver, to having different fairly equal environmental burdens that stem from process differences in route based carbon mineralisation technology. Throughout almost all categories, there is a noticeable balance of MgSO₄ at 25% to 50% and BPMED as 30% to 50% which indicate co-dominance.

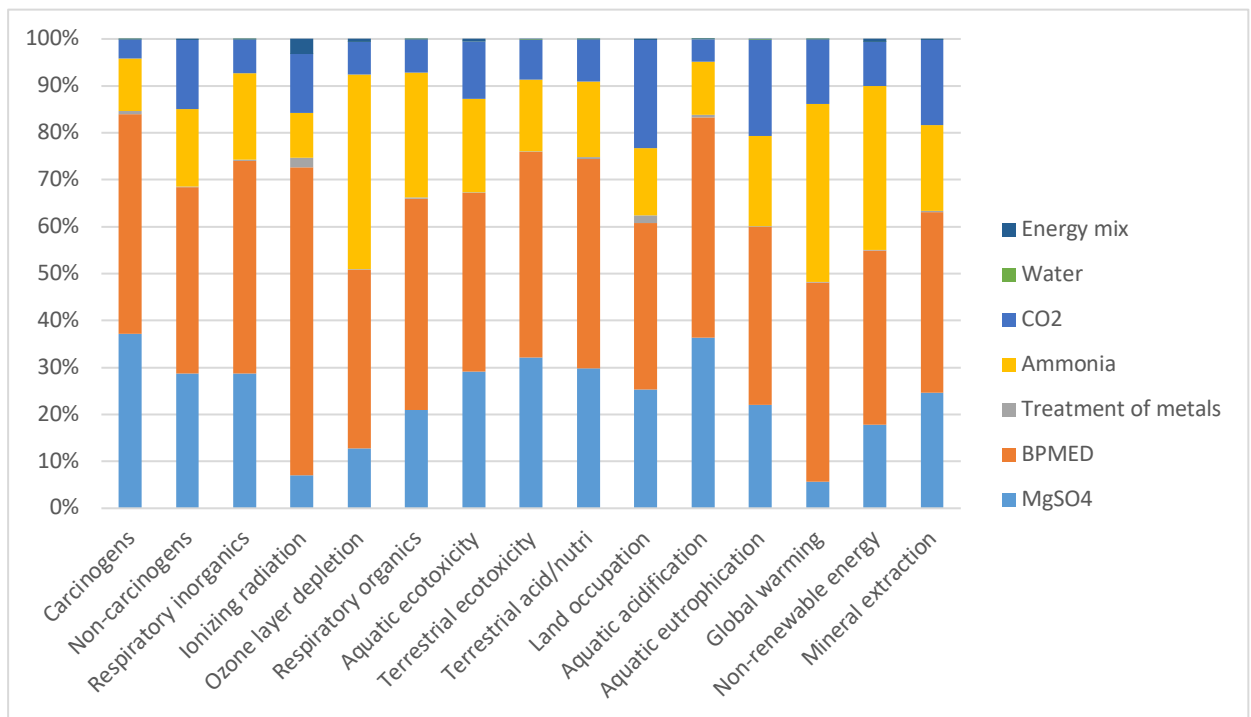


Fig. 15. IMPACT 2002+ characterisation results for scenario 2

There is a noticeable pattern across the data indicating trade-off between chemical recovery process and mineral process as whenever MgSO₄ is at higher levels of environmental burdens, BPMED is lower. The same is noticeable whenever BPMED is at higher levels. Having shifting burdens may lead to optimisation faults across the system. Throughout scenario 1, ammonia was indicated as secondary contributor which is noticeable in scenario 2 leading to importance of chemical looping. Scenario 2 exhibits higher levels of CO₂ emissions as a burden but it does not exceed 40% while peaking in climate related categories such as global warming, eutrophication and land occupation but does not dominate overall performance.

Separating the process characterisation and analysing MgSO₄ alone as it was done with scenario 1, allows to further identify dominant environmental impact burdens. Visualised in Appendix 2 Fig. 34, sulfuric acid becomes the dominant category rather than electricity as it was hypothesised due to scenario 2 relying on electricity to extract MgSO₄. Although, the expected physically dominated systems highlight energy demand burdens, chemically intensive routes continue shifting environmental burdens towards reagent production and recirculation.

3.3.3. Scenario 3

Scenario 3 follows similar route composition as scenario 2 and was modelled with the material flow illustrated in Appendix 2 Fig. 32. Due to scenarios sharing main structure of the mineral carbonation, Fig. 16 becomes an identifying aspect indicating that the environmental performance during LCA performed, is not governed by mineral type like it was expected in hypothesis. Process configuration becomes crucial while similarities in data support this statement. Throughout almost all categories, following the scenario 2, there is a noticeable balance between the trade-off's of MgSO₄ at 30% to 50% and BPMED at 30% to 45% which further indicate co-dominance. The observed process trade-off between MgSO₄ and BPMED continues across different mineral types further emphasising process and system interactions and not material specific effects.

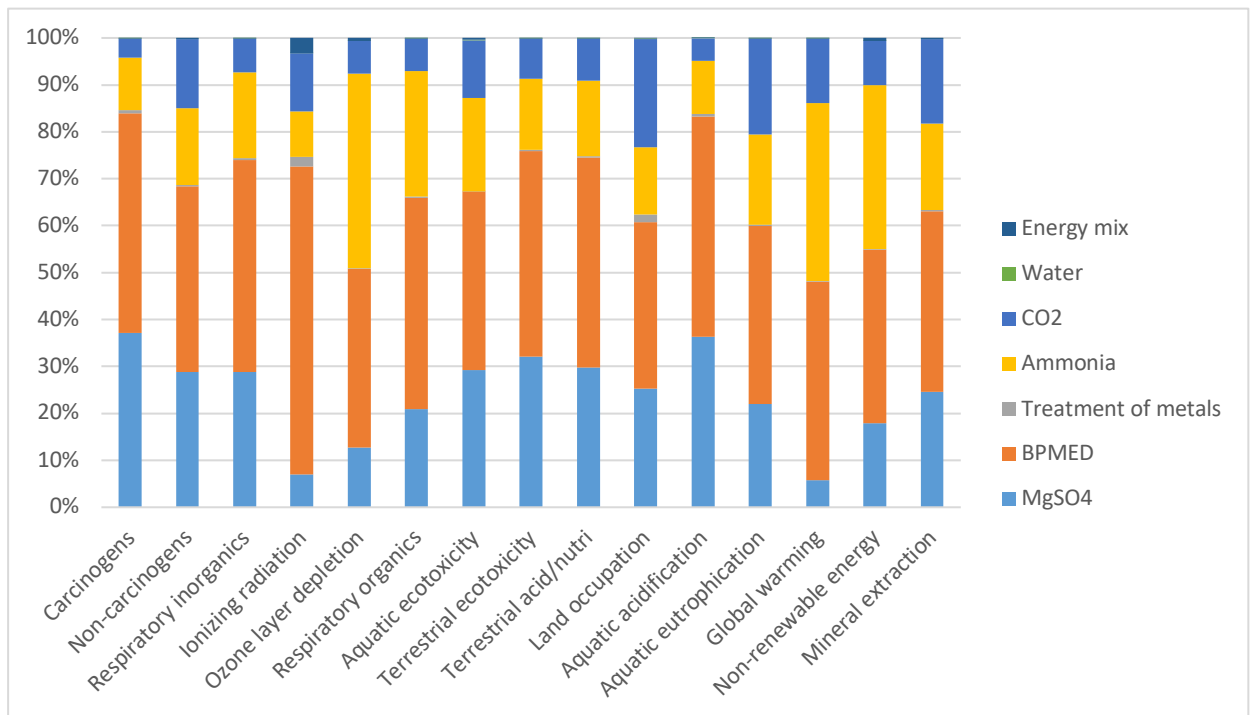


Fig. 16. IMPACT 2002+ characterisation results for scenario 3

MgSO₄ extraction stage for phlogopite is where the differences between the routes, sharing the same mineral carbonation route, appears. Appendix 2, Fig. 35 visualises the characterisation of the wet extraction phase and when comparing with a setup in scenario 2 (Fig. 33), it is noticeable that scenario 3 exhibits higher levels of mineral persistence across three biggest impact categories – ionizing radiation, land occupation and global warming. These increases can be attributed to the higher demand for mineral itself. Scenario 3 needs almost 50% more phlogopite, than scenario 2 with serpentine. Similarly scenario 3 requires more than 15% of H₂SO₄ as a reagent while it emits more than 50% of silica than scenario 2. Continuing symmetry with previous scenario, sulfuric acid stays as the dominant category rather than electricity bringing the attention to BPMED need across mineral carbonation to counteract emissions stemming from mineral extraction.

3.3.4. Normalised life cycle impact assessment of three scenarios

Comparative analysis of all three scenarios was performed implementing IMPACT 2002+ method for consistency. Across all impact categories (visualised in Fig. 17.), scenario 3 consistently carries the highest environmental burdens. Scenario 2 which is modelled similarly to scenario 3, consistently

stays second while scenario 1 across all categories, except aquatic eutrophication, shows lowest impacts. Consistent ordering of scenarios across nearly all impact categories may indicate a strong difference in environmental performance of system configurations. Scenario 3 closely follows scenario 2 which is consistent with the scenario 3 dependency on ÅA4 which is a base for scenario 2. Performed LCA and results portray scenarios as chemical and energy based rather than mineral. Biggest differences created stem from ionising radiation and non-renewable energy which confirms the suggested sensitivity to energy demands and its upstream inputs.

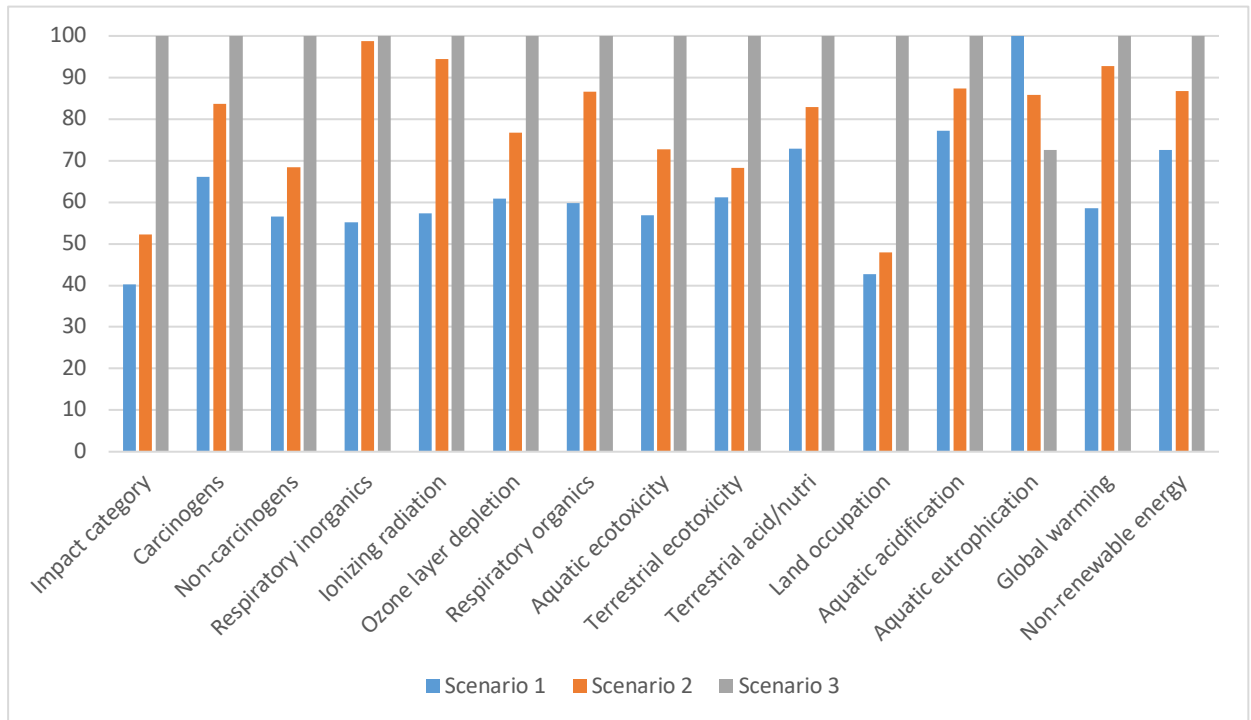


Fig. 17. IMPACT 2002+ characterisation results for scenario comparison

Normalisation is an optional LCIA stage which is defined by ISO 14044 [74]. Normalisation calculates relative magnitude of environmental impacts by division of raw results with reference value allowing for diverse units being read into common scale. Across all of the endpoint categories (Fig. 18), scenario 1 symmetrically remains the least burdened. While scenario 2 and 3 share similar process route, throughout human health category, scenario 3 which uses phlogopite as the main feedstock, exhibits over 77% higher impact than scenario 1 and over 42% when it is compared with scenario 2. Scenario 2 is higher than scenario 1 as it reaches almost 25% higher environmental burdens. Relatively, ecosystem quality environmental impact category exhibits smallest values but when scenarios are compared between each other, scenario 1 stays consistent while scenario 2 shows over 25% higher impacts while scenario 3 reaches over 70%. Comparing scenario 3 against scenario 2 allows for over an 35% higher impact. Resources has shown to be most impactful category in this analysis although comparing scenarios between each other allows for the insight of scenario 3 exhibiting only under 8% higher environmental impacts by normalisation when compared with scenario 2. The use of chemicals and electricity are cumulated in the resources with BPMED becoming a crucial part in mineral carbonation. Koivisto et al. has proposed [84], that changing the configuration of BPMED from three- to two-compartment stack allows for lower electricity consumption. This could be implemented in the mineral carbonation as a part of emission lowering approach.

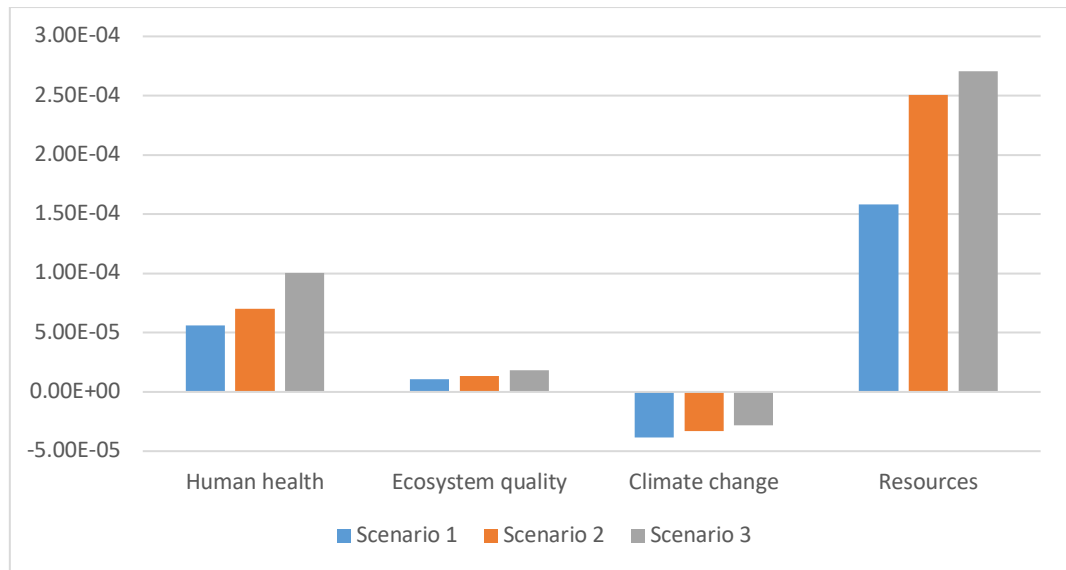


Fig. 18. IMPACT 2002+ normalisation results for scenario comparison

Focusing on IMPACT 2002+ damage assessment results for scenario comparison, climate change emerges as an important consideration both in characterisation calculations and damage assessment due to its connection with FU of the study. Fig. 19. Shown negative climate change results for all scenarios indicates that the CO₂ amounts that are permanently stored in nesquehonite exceeds the emissions stemming from the process. The negative calculations do to directly remove CO₂ from the atmosphere but acts a a net carbon sink within chosen system boundaries. Changes between the scenarios are attributed to variations in energy and material requirements. Scenario 1 exhibits the highest calculated CO₂ sequestration while scenario 2 is 14% weaker, then again scenario 3 is 27% higher than scenario 1.

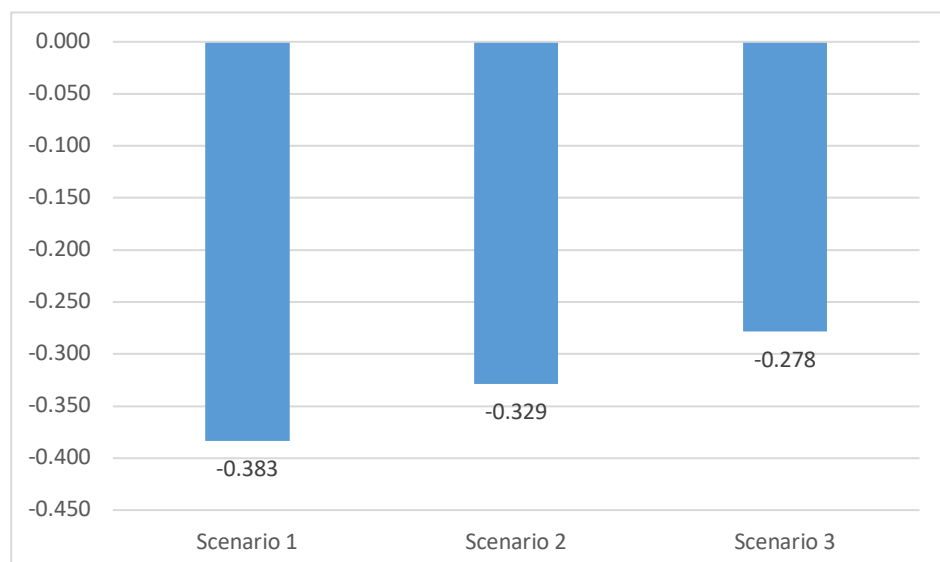


Fig. 19. IMPACT 2002+ damage assessment results for scenario comparison focusing on climate change expressed in kg CO₂ eq

The LCA study performed by Khoo et. al. [85] concludes similar calculations but they have to be taken into consideration cautiously due to differing system boundaries. Their FU was set as tonne of CO₂ input to CO₂ mineralisation, while system boundaries focus on mine gate to incineration plant

which includes full lifecycle of the product with transportation and incineration. Although the calculation method is not identified, it is focused on 100 year time horizon (GWP) which allows for cautious comparison with IMPACT 2002+ Climate change damage category expressed in *kg of CO₂ equivalents*. While scenario 1, 2 and 3 has showed consistent capture capabilities (Fig. 19), study by Khoo et. al. has gathered the results reaching 0.937 kg of CO₂ eq when the FU is equalized to 1 kg of CO₂ captured. The capture capabilities are sparse but that could be attributed to various factors across the system boundaries.

3.3.5. Energy mix change impact across scenarios

During chapter 2.2.1 it was established that current Ecoinvent data is inefficient to purposely calculate mineral carbonation results that are close to current energy situation in Finland. Basing calculations on 2012 data allows for observation how impactful energy mix is to the process. Fig. 20. represents the normalisation data in LCA which highlights the shift in impact categories. Across all scenarios several damage categories has shown a rise in emissions way above 100% if 2012 data is used. Across the ecosystem quality damage category scenario 2 impact increases by over 240% as the calculations go from 0,1816 PDF.m².yr with 2024 data to 0,6187 PDF.m².yr. with 2012 data. Climate change rises by over 300% with -0,3287 kg CO₂ equivalent in 2024 to 0,7608 kg CO₂ equivalent across 2012 data. Scenario 3 follows scenario 2 closely with ecosystem quality impact category going from 0,2471 PDF.m².yr with 2024 data to 0,6811 with 2012 data while climate change over with -0,2782 kg CO₂ equivalent in 2024 to 0,8033 kg CO₂ equivalent across 2012 data .

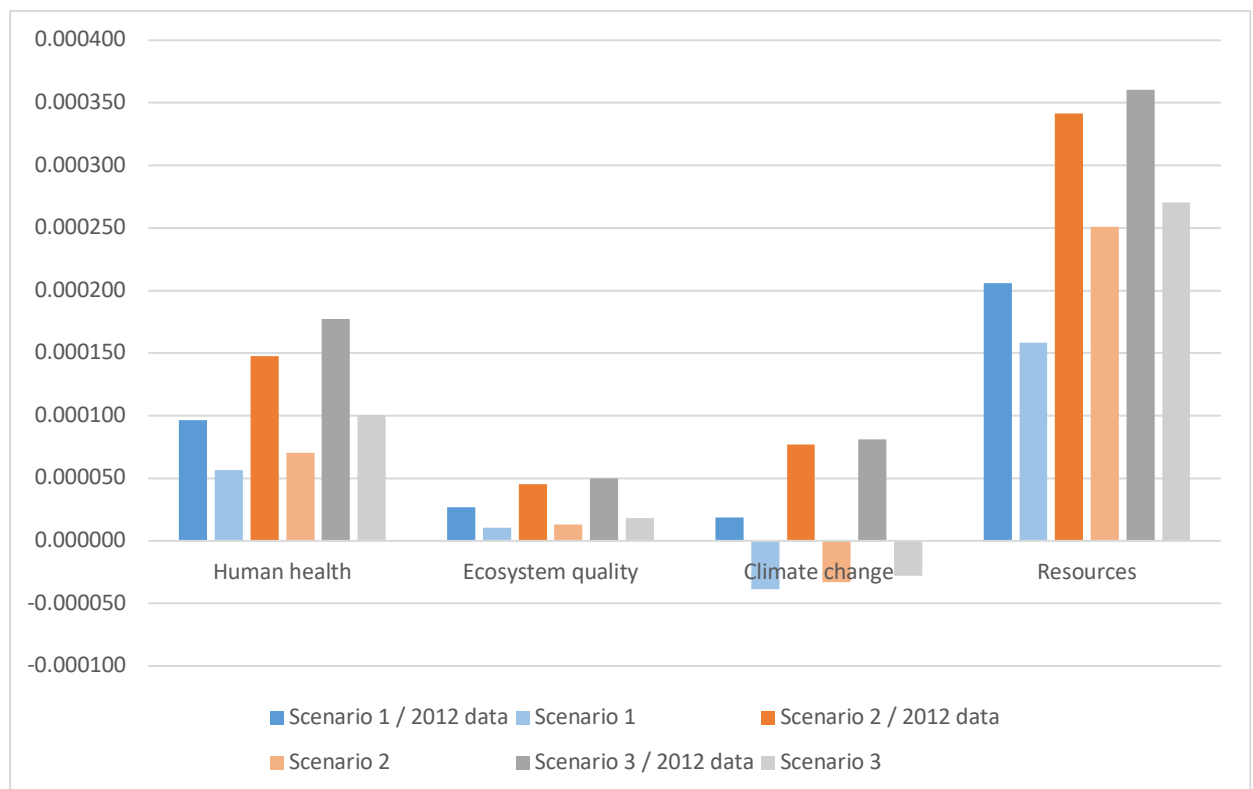


Fig. 20. Normalisation of data while comparing established scenarios through old 2012 data base and new energy mix created based on 2024 data

Collectively when considered average calculations, scenario 2 is mostly affected when electricity input is changed while scenario 1 lowest. Scenario 1 dependency to electricity is lowest due to mineral carbonation process which applies heat rather than electricity throughout MgSO₄ extraction phase.

While the drastic change to electricity variation shows scenario's environmental performance dependency on electricity sources, there's an additional consideration when electricity generation is using fossil fuels besides renewable or nuclear energy. While performing LCA this study established the equivalent of CO₂ emissions per FU of mineral carbonation process gathering -0.3833, -0.3287 and -0.2782 kilograms of CO₂ equivalent emitted per scenario 1,2 and 3 respectively. Calculations with 2012 data has shown 0.184, 0.768 and 0.811 kilograms of CO₂ equivalent emitted per FU per scenario 1,2 and 3 respectively. Allowing for consideration of using less renewable energy, results in inefficient mineral carbonation which could potentially lead to higher emissions emitted during the process than captured.

3.3.6. Scenario 4 analysis with BPMED

After all three scenarios were modelled and analysed, a fourth one, using serpentine, was chosen based on the best performing mineral carbonation scenario. Due to BPMED high energy demand, there is concern on whether applying membrane looping to filter reagents needed for mineral carbonation, scenario 4 was established. Scenario 1 consistently showcased the overall performance when analysed through various damage categories. As it was established during methodology, scenario 4 will focus on implementation of BPMED and the usage of virgin chemicals (VC).

Table 7. Finalised inventory for newly established scenario 4 that eliminates the use of BPMED

PRODUCT	UNIT	SCENARIO 4
CO ₂ , CAPTURED	kg	1
NESQUEHONITE	kg	3.144
INPUT ITEM		
MGSO ₄	kg	2.735
AMMONIA, LIQUID	kg	1.593
WATER	kg	0.819
CARBON DIOXIDE, LIQUID	kg	1
TREATMENT OF METALS	kg	0.1
ELECTRICITY MIX	kWh	0.1
OUTPUT ITEM		
CO ₂ FOSSIL EMISSIONS	kg	-1

Table 7 and Fig. 21 shows finalised inventory table and system boundaries for newly established scenario 4 which uses best performing scenario 1. As established in previous sections, allowing for BPMED elimination throughout environmental assessment allows to analyse the efficiency of the energy demanding BPMED process and highlight whether the high energy demand is environmentally acceptable.

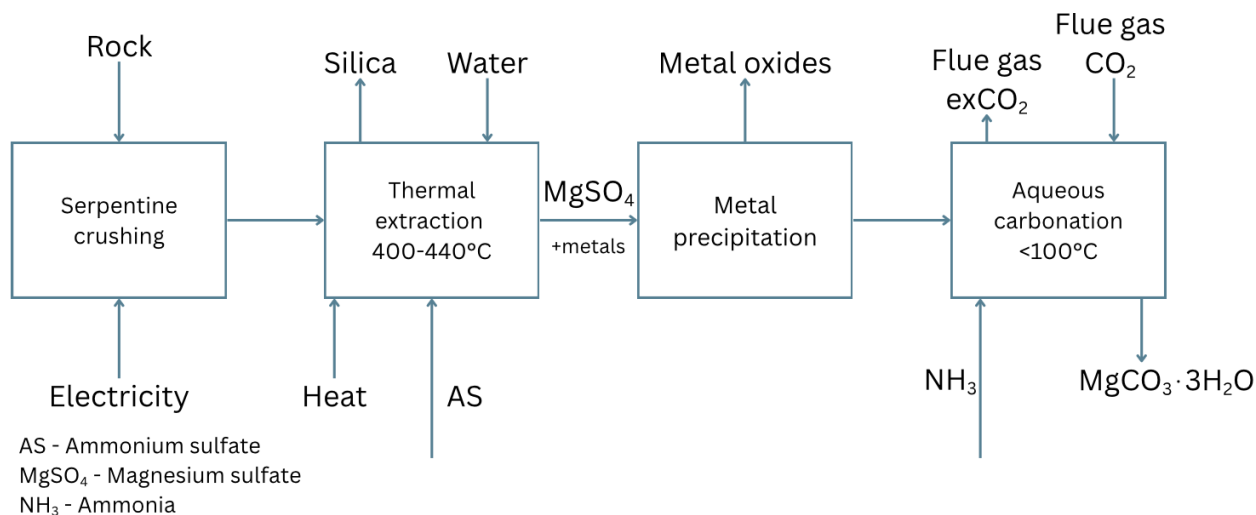


Fig. 21. Adjusted system boundaries for newly established scenario 4 focusing on BPMED elimination

Fig. 22. Visualises characterisation results for scenario 1 and 4 comparison highlighting the importance of BPMED implementation in process design established in ÅA3 route development. Scenario 4 continues with electricity as a main environmental impact factor. Significant increases are consistently observed in electricity-sensitive categories like ionising radiation and non-renewable energy. Chemical regeneration processes are identified as secondary impact drivers further visible in global warming results. Furthermore, BPMED importance can be highlighted through a perspective of global warming impact category. Scenario 4 follows process design of scenario 1 with adjustments to inputs requiring only VC and eliminating membrane recirculation of the chemicals while FU for both is „1kg of CO₂ captured“.

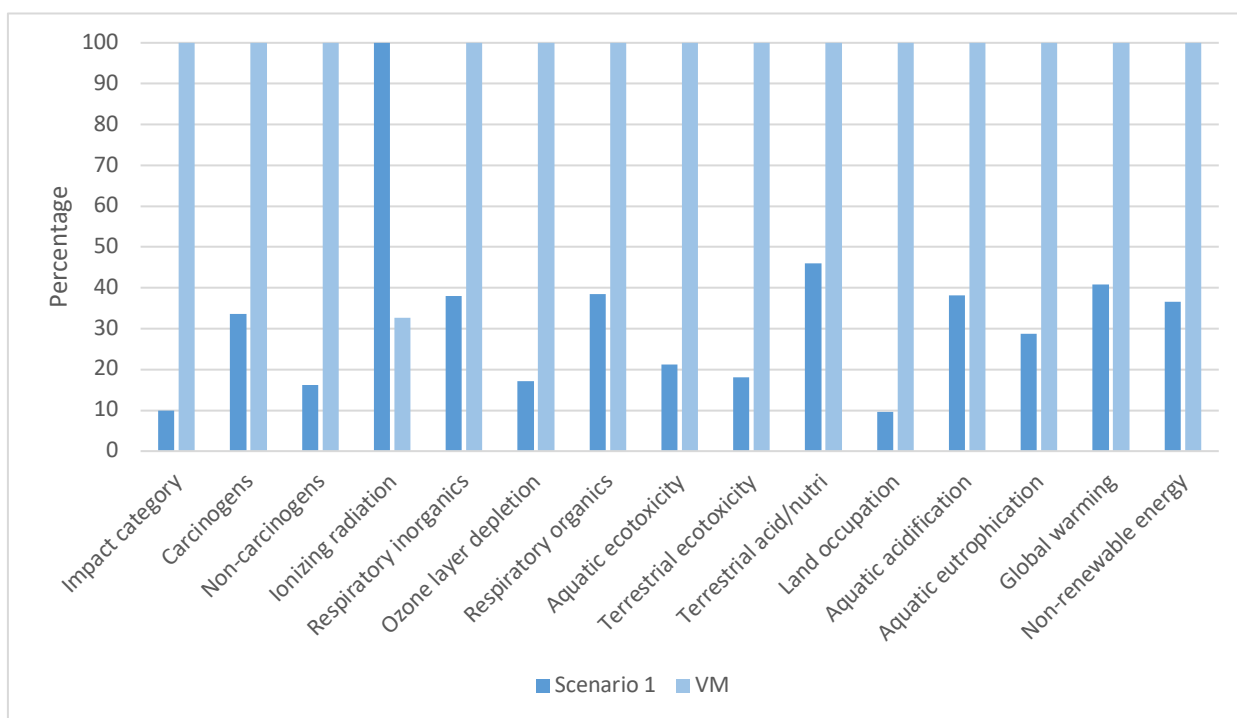


Fig. 22. IMPACT 2002+ characterisation results for scenario comparison focusing on climate change

Fig. 23 focuses on normalization results of climate change endpoing category linking characterisation results to overall environmental performance. Expressed in kilograms of CO₂ emitted equivalent,

scenario 1 has a negative impact per FU while scenario 4 is estimated to emit 1.33 kg of CO₂ per FU highlighting BPMED as functional necessity to achieve net CO₂ capture. Based on the calculations, mineral carbonation process designs without the implementation of BPMED gives rise to additional environmental burdens. While improving climate performance, high energy demands lead trade offs between increased process intensity and climate benefits.

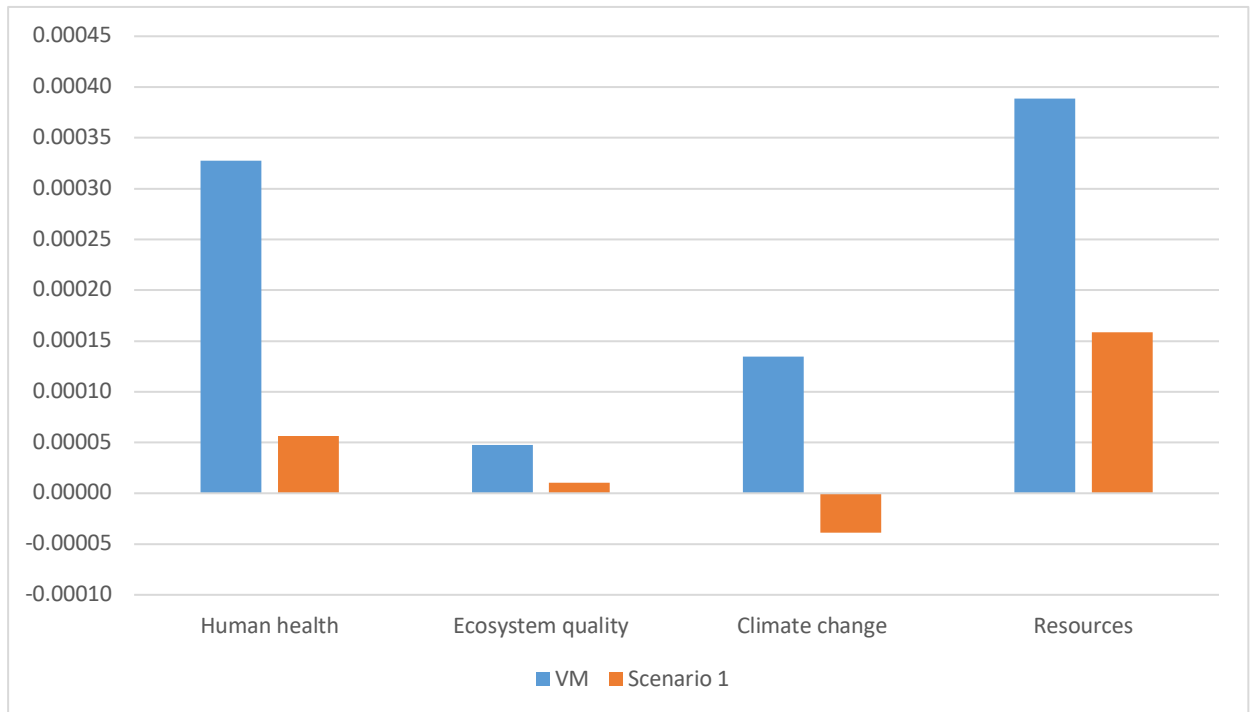


Fig. 23. IMPACT 2002+ normalisation results comparing scenario 1 and its variation when only VC are used

3.4. Sensitivity analysis

To determine whether the results of LCIA are reliable, SA was conducted across three main scenarios as a last step in this study. Per ISO 14044:2006 SA becomes a key methodological step to evaluate how correctly the study was performed and which inputs influence the final results the most [74]. Once-through SA was conducted by systematically varying a selected parameter within predefined ranges, while holding all other parameters consistent to assess their individual influence over the whole range of modelling. Selected parameter was chosen as electricity and heat input across mineral carbonation scenarios phases due to its continuous impact across both all scenarios observed in characterisation and normalisation results. Results obtained while comparing 2012 and 2024 energy inputs further finalised energy as main parameter for SA. The data that was obtained during the inventory analysis step was varied by 10% while other parameters stayed consistent. Due to MgSO₄ extraction and BPMED stages being most energy intensive, both were adjusted by 10% from original calculations while the mineral carbonisation stage which only consumes 0.1 kWh per FU was varied to stay consistent with energy demand. Table 8 finalises MgSO₄ stage calculations which has scenario 1 with heat input that was varied due to its energy source while scenario 2 and 3 share electricity calculations. Values in tables 9 and 10 are calculated based on all scenarios while considering the identical stages.

Table 8. Electricity input across MgSO₄ stage calculated for SA. Scenario 1 uses heat while scenario 2 and 3 are using the same input

	SCENARIO 1	SCENARIO 2	SCENARIO 3
BASELINE	0.94		0.91
10%	1.039		1.001
-10%	0.846		0.819

Table 9. Electricity input across BPMED stage SA calculated across all scenarios

ALL SCENARIOS	
BASELINE	0.849
10%	0.9339
-10%	0.7641

Table 10. Electricity input across mineral carbonation stage SA calculated across all scenarios

ALL SCENARIOS	
BASELINE	0.1
10%	0.11
-10%	0.09

To plot the SA results, normalization results were transformed and presented as a relative percentage of change stemming from baseline scenario. Changes in Fig. 24, 25 and 26 are expressed in relative change from baseline by percentage. Comparing the percentage change from baseline allows for improved comparability across damage categories. Moreover, comparing on a scale normalized through baseline, calculations are adjusted through the use of different units.

SA based normalisation for scenario 1 is expressed in Fig. 24. which shows fairly balanced outcomes across all endpoint categories and their performance with both increased and decreased 10% of energy inputs. Climate change has shown the least sensitivity to the parameter while resources are mostly affected reaching approximately 6.20 - 6.30 % result change. Model behaviour through stable symmetric changes may suggest that there are no major numerical instabilities. Considering that energy use and material consumption dominated the results, it is adequate to have sensitivity to the inputs related to them. Scenario 2 and 3 with graphs in Fig. 24 and 25 are a continuing with resources being the most sensitive impact category reaching around 7 - 8%. There is a noticeable shift across both scenario 2 and 3 when it is compared with scenario 1 which could be attributed to overall lower environmental impact of scenario 1.

SA was performed following the same methodology as established earlier using Simapro 9.0 with the same IMPACT 2002+ impact calculation method. Every other parameter stayed the same from inventory tables. When considering results from the study and SA, there is a repeating outcome noticeable across scenarios with scenario 1 consistently outperforming the other scenarios modelled.

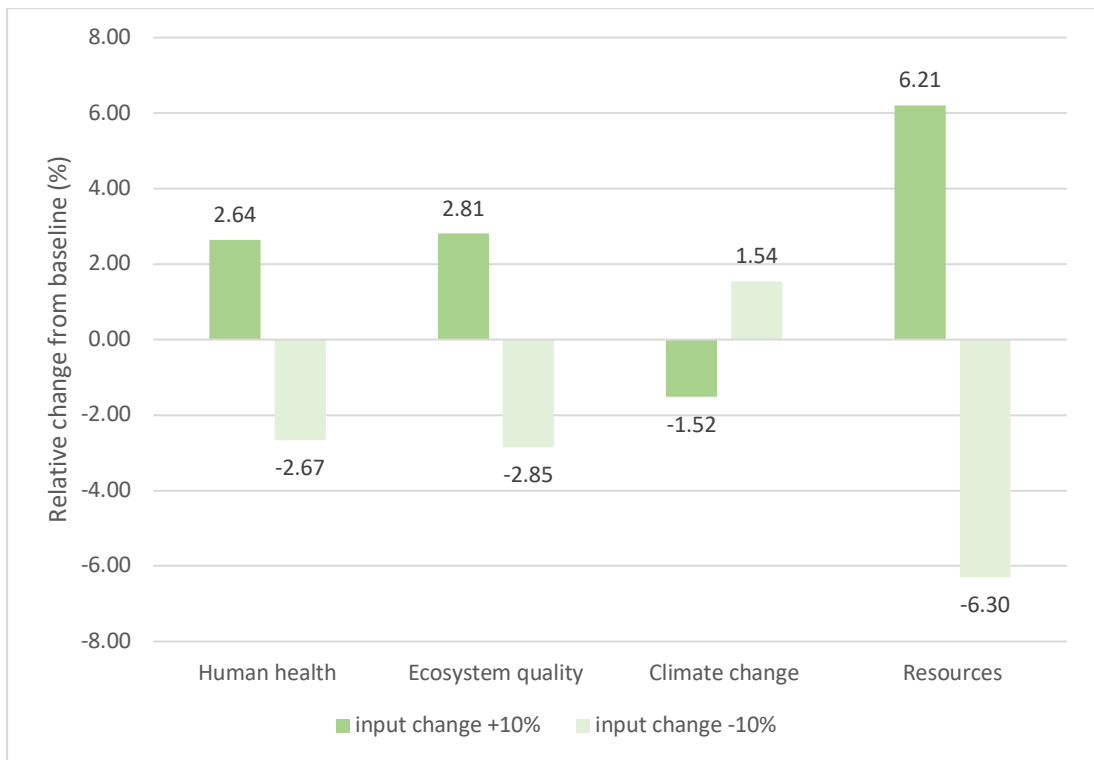


Fig. 24. SA for scenario 1 expressed under normalisation using $\pm 10\%$ parameter variation

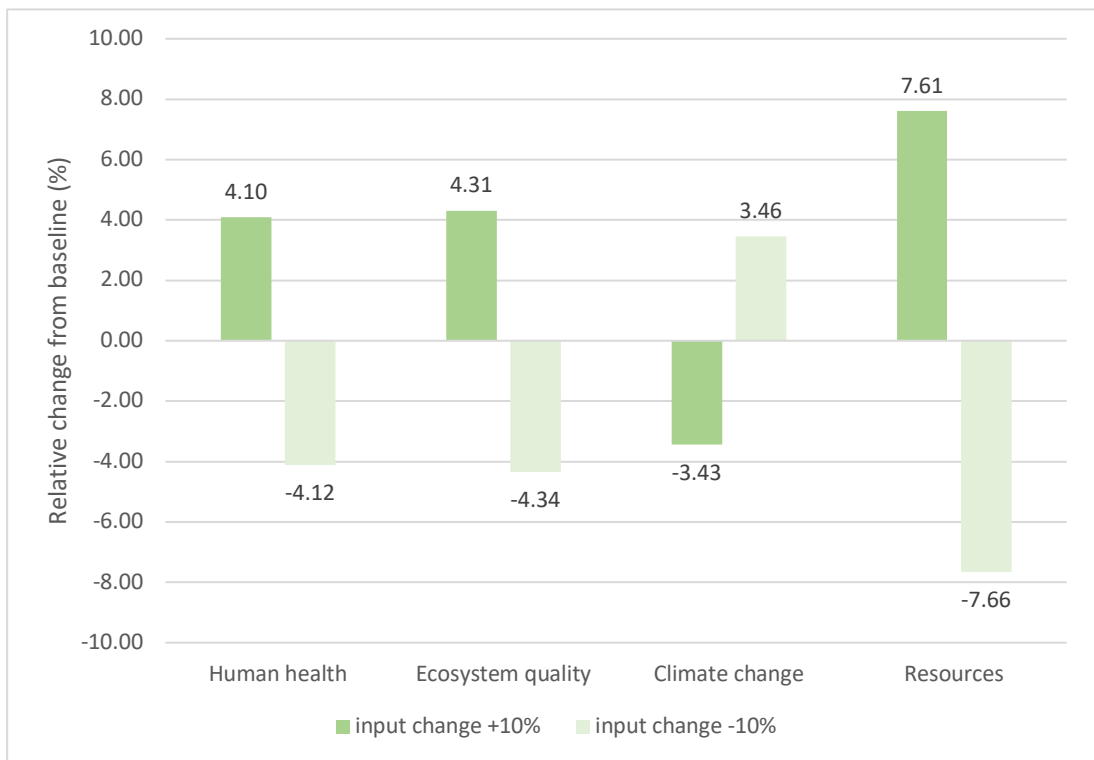


Fig. 25. SA for scenario 2 expressed under normalisation using $\pm 10\%$ parameter variation

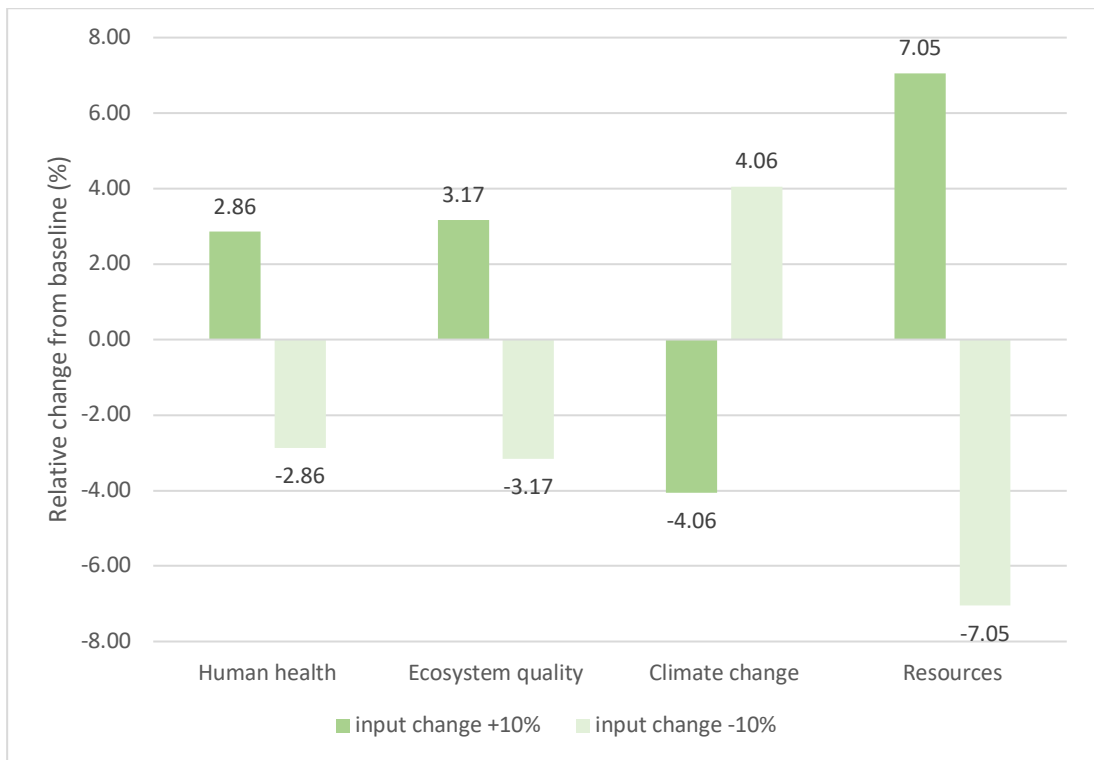


Fig. 26. SA for scenario 3 expressed under normalisation using $\pm 10\%$ parameter variation

Conclusions

1. The literature review overviews the role of CO₂ mineral carbonation in climate change mitigation, while focusing on its use as a pathway for permanent CO₂ storage. Serpentine and phlogopite are identified as a key feedstock due to their abundance in the world and ability to form stable carbonate phases. Academic routes (technology readiness level 4-5), particularly ones proposed by Åbo Akademi, allow for creation of different system configurations it shows the need for further systematic environmental assessment where life cycle assessment emerges as a critical tool to evaluate beyond theoretical CO₂ uptake and fully identify key environmental tradeoffs.
2. Data for the life cycle assessment was collected from Åbo Akademi laboratories, scientific literature and ecoinvent database. System boundaries chosen were gate-to-gate focusing primarily on the mineral carbonation processes in Åbo Akademi. The functional unit was set as one kilogramme of CO₂ captured. IMPACT 2002+ was chosen as life cycle impact assessment calculation methodology focusing four damage impacts: human health, ecosystem quality, climate change and resources.
3. Based on the established scenarios, the first one consistently showed its superiority across all of the impact categories due to the lower energy input. Characterisation resulted in identification of ionizing radiation, land occupation and global warming potential as primary impact categories. Bipolar membrane electrodialysis symmetrically stayed consistent as the primary environmental burden across all scenarios. After identifying the best performing scenario out of three, the fourth scenario was established that focused only on inputs that do not go through bipolar membrane electrodialysis which allows for less waste and recirculation of existing chemicals. Scenario four identified bipolar membrane electrodialysis as a crucial feature for mineral carbonation bringing estimated CO₂ emissions per functional unit from 1.33 kg to -0.38 kg.
4. Normalisation resulted in symmetrical results showcasing overall scenario 1 performance when compared with scenario 2 and 3. While scenario 2 and 3 show relative consistencies due to process configuration, scenario 3 which uses phlogopite exhibits higher environmental burdens across human health and ecosystem damage categories reaching almost 40% higher impact than scenario 2. Damage assessment for climate change damage category resulted in equivalent CO₂ estimations of -0.383, -0.329, -0.278 kg across scenarios one, two and three respectively with these calculations expressed per functional unit.
5. During sensitivity analysis, in which energy consumption was confirmed as the primary driver of environmental impact, resource damage category was the most sensitive across all scenarios. Symmetrically, damage categories were affected similarly throughout all of the scenarios and when measured against baseline, did not exceed moderate sensitivity metrics.
6. While all three established scenarios performed relatively similarly regarding mineral carbonation across all of the damage categories, scenario 1 has showed the importance of energy demand and possibilities of heat usage to lower environmental impacts. Additionally the importance falls on bipolar membrane electrodialysis when establishing an efficient mineral carbonation process while electricity source enhances process possibilities.

Scientific participation

The results of the final master's project research were presented:

14th young scientist conference „Fizinių ir technologijos mokslų tarpdalykiniai tyrimai“.

Cestauskaite, J. Comparative Life Cycle Assessment of Serpentine and Phlogopite-Based CO₂ Mineral Carbonation Routes // 14-a jaunųjų mokslininkų konferencijoje „Fizinių ir technologijos mokslų tarpdalykiniai tyrimai, April 17, 2026, Vilnius.

International conference „Chemistry and Chemical technology“.

Cestauskaite, J., Stasiulaitiene, I., Zevenhoven, R. Comparative Life Cycle Assessment of Serpentine and Phlogopite-Based CO₂ Mineral Carbonation Routes // Chemistry and Chemical technology: international conference CCT-2026, May 22, 2026, Kaunas.

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Appendices

Appendix 1. Supplementary figures for literature review

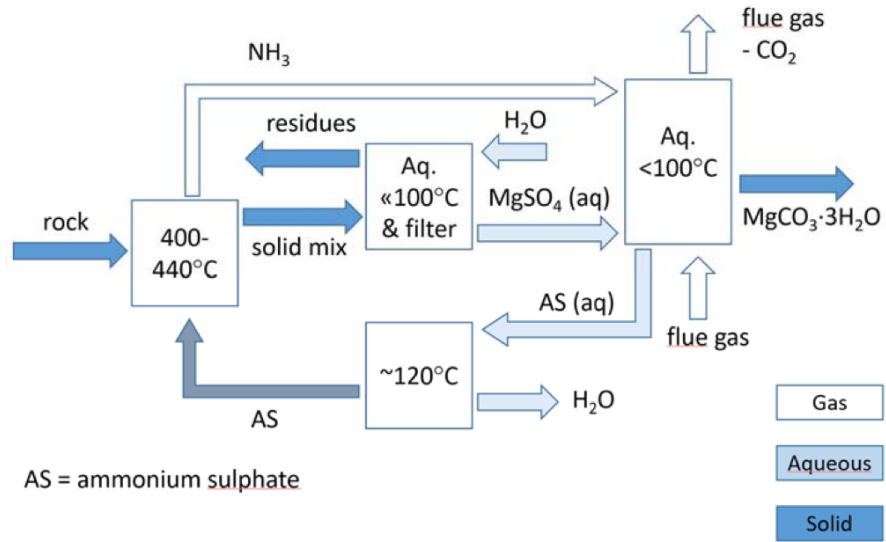


Fig. 27. ÅA2 route that continues with dry extraction process while introducing wet carbonation [50]

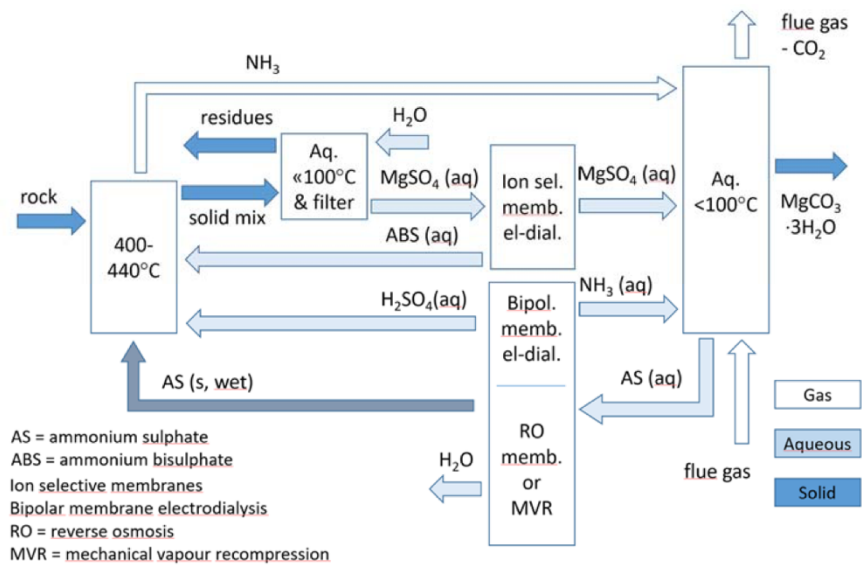


Fig. 28. ÅA3 route which follows ÅA2 with extraction and carbonation but introduces BP MED separation [50]

Appendix 2. Supplementary figures for LCIA

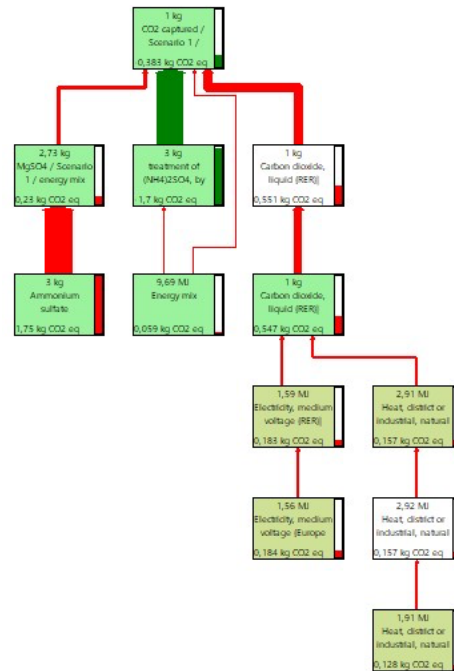


Fig. 30. Material flow for scenario 1

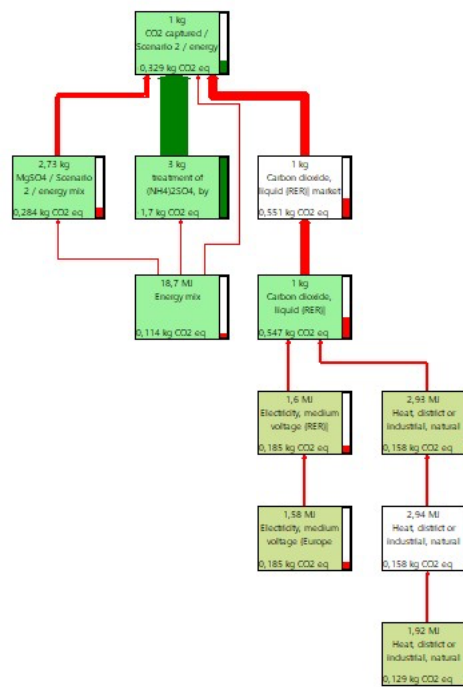


Fig. 31. Material flow for scenario 2

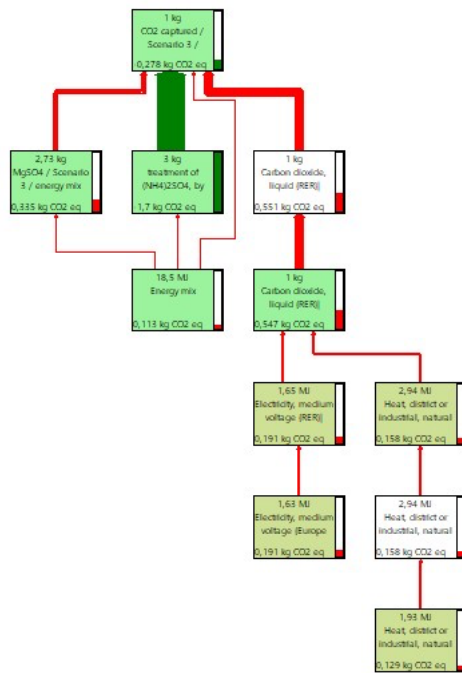


Fig. 32. Material flow for scenario 3

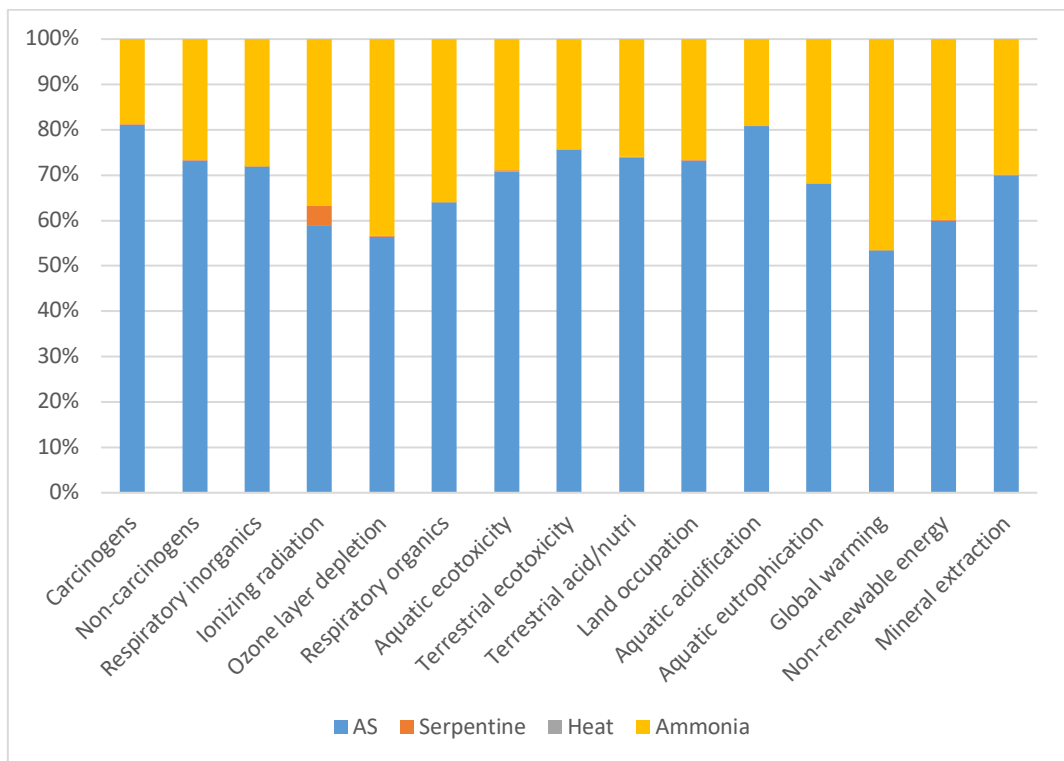


Fig. 33. IMPACT 2002+ characterisation results for scenario 1 focusing only on the process of MgSO4 extraction

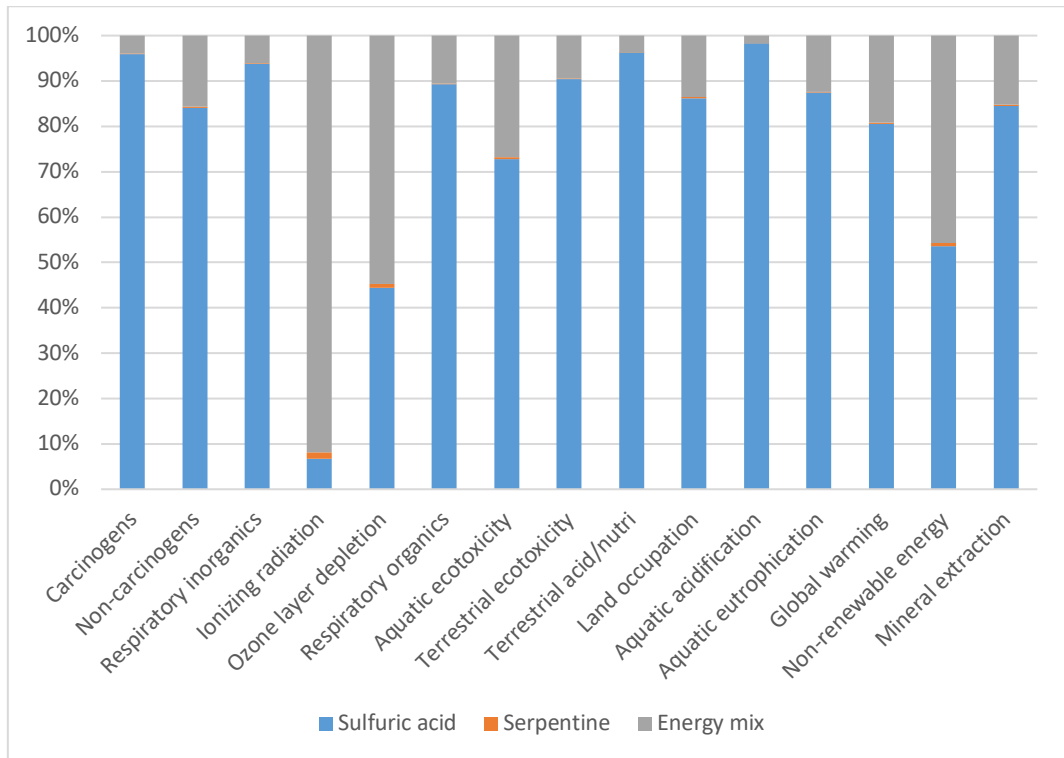


Fig. 34. IMPACT 2002+ characterisation results for scenario 2 focusing only on the process of $MgSO_4$ extraction

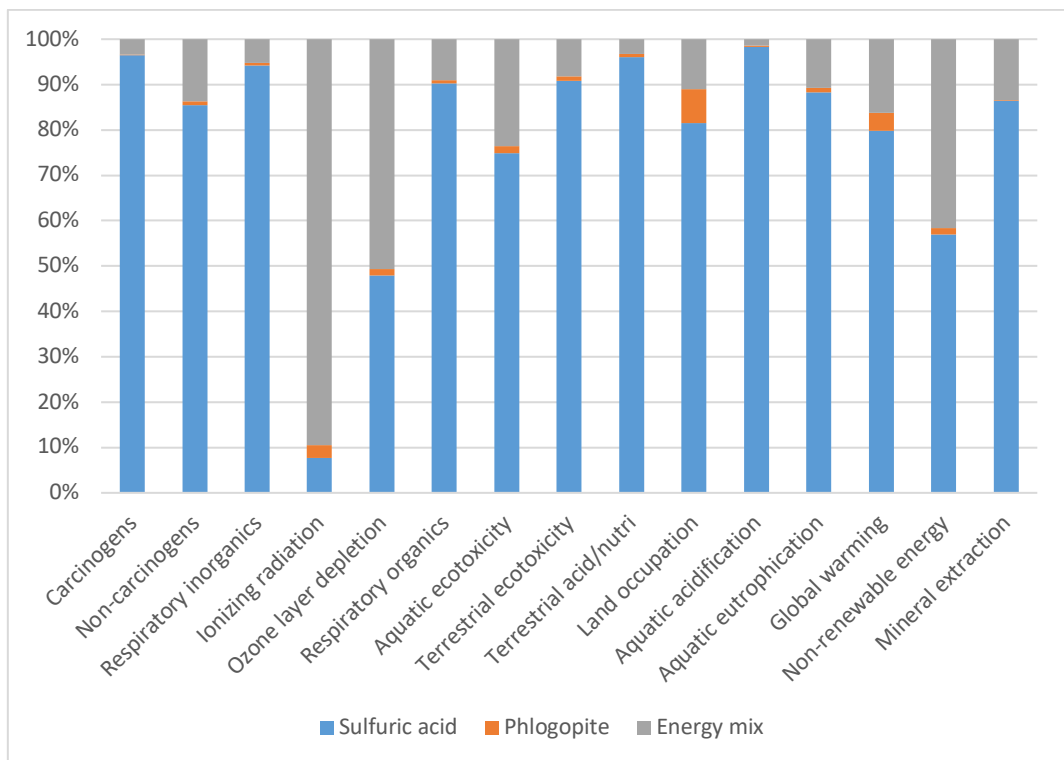


Fig. 35. IMPACT 2002+ characterisation results for scenario 3 focusing only on the process of $MgSO_4$ extraction