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Towards Net-Zero and Carbon-Negative Transformations in Lime-Based Construction Materials

Agustin Laveglia

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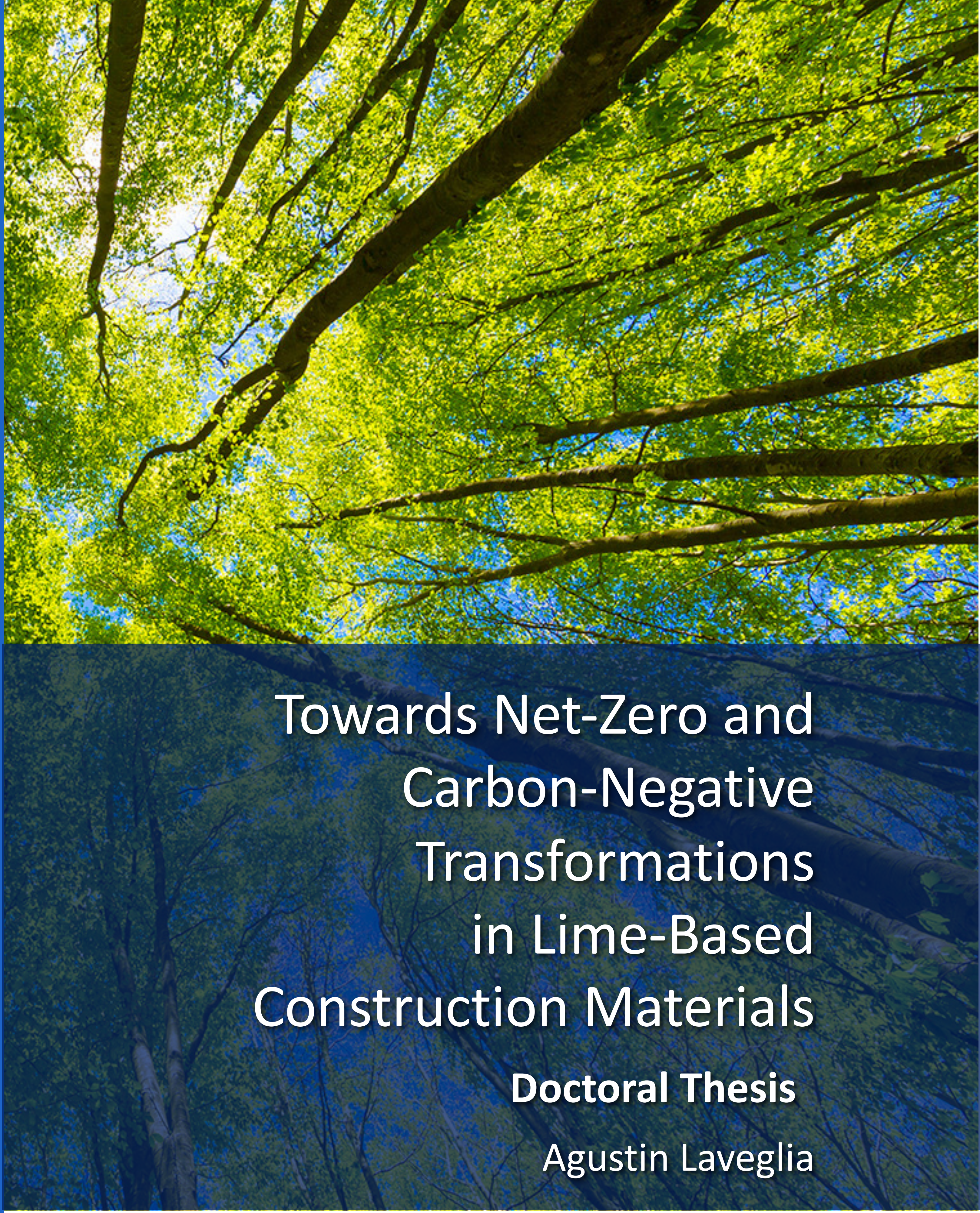

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Doctoral Thesis

Agustin Laveglia

GHENT UNIVERSITY

TECHNISCHE UNIVERSITÄT DARMSTADT

FACULTY OF ENGINEERING
AND ARCHITECTURE

Department of Civil
and Environmental
Engineering

Towards Net-Zero and Carbon-Negative Transformations in Lime-Based Construction Materials

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"You cannot hope to build a better world without improving the individuals. To that end each of us must work for his own improvement, and at the same time share a general responsibility for all humanity."

Marie Skłodowska-Curie (1867-1934)

Acknowledgements

After nearly three amazing years on this incredible journey, my time as a PhD student is drawing to a close. So much has unfolded, so much learning has taken place, and the support I have received from numerous individuals over the years has been overwhelming. It only feels right to kick off this book by sharing a piece of how I arrived here and expressing my deep gratitude to everyone who played a part in it.

In many instances, one is not fully aware of the path being traversed until you're distant enough to see how far you've come from the starting point. I was always passionate about chemistry, faced with two major choices at my hometown university in Olavarría: Medicine and Chemical Engineering. The path became clear after I attended an introductory medicine course, where I nearly fainted at the sight of an arm in a preservation liquid.

My first year at my hometown university in 2014 was enlightened by the introductory Chemistry course taught by one of my greatest mentors and supporters, Prof. Mónica Trezza. For those who know me, they are familiar with my curious mind and intensity. We often reminisce about my crazy questions and the afternoons spent in the lengthy laboratory sessions. Passing the course opened the door to my first foray into science the following year, thanks to an invitation from Prof. Trezza for an undergraduate scholarship. Our work focused on circular economy in cementitious systems, where she not only taught me the fundamentals of cement hydration but also instilled the values of honesty, dedication, and ethics. In 2017, she encouraged me to present my findings on waste glass as a pozzolanic addition at my first scientific congress of young researchers in materials in Buenos Aires. Little did I know that this experience would lead me to Germany, culminating in the start of my PhD. When I returned from the conference, I mentioned to her about a fully-funded DAAD study trip for master engineering students to spend a month in German Universities and specialized research centers. I hesitated, thinking there was no chance I'd be selected among 200 attendees, but she insisted: "*¡De ninguna manera! ¡Vos te presentas igual!*" (No way! You are going to apply!) – She said. To my surprise, I was chosen as one of the 15 engineering students from Argentina to receive the DAAD scholarship. Prof. Viviana Rahhal was equally important during this time, particularly as student and in my early stages as a researcher. Thank you Vivi for sharing with an incredible generosity your knowledge and life with me. Our afternoons working, *tomando mate*, and flying in our minds will be forever in my heart.

The trip in 2018 marked my first profound connection with my German roots, as my grandparents had fled Germany during World War II to settle in Argentina. Each of the 15 students formed a nomadic family, traveling from city to city, collecting indelible memories, savoring German beer and Schnitzel. One of the universities we visited was in Darmstadt, and the Institute of Construction and Building Materials (WiB), led by Prof. Eddie Koenders, hosted a full day of activities. I was captivated by the laboratories and equipment, momentarily envisioning myself working there, hoping that dreams do come true with dedicated effort. Three years later, in 2021, Prof. Koenders, *mein Doktorvater*, would indeed make that dream a reality for a guy living 12,000 km away from that institute, and for that opportunity, I will be forever grateful. The trip not only opened up a life full of opportunities but also brought four amazing friends into my life — Ignacio, Lucio, Florencia, and especially Josefina (Jose), who remain, even five years later, one of my dearest groups of friends.

In 2020, just a few months after completing my master's, TU Darmstadt reappeared on my LinkedIn feed. **An open call for Early-Stage Researchers (ESR) within the European consortium SUBLime caught my eye. SUBLime, Sustainable Building Lime Applications Via Circular Economy and Biomimetic Approaches, was a Marie Skłodowska-Curie Action European Training network – Innovative Training Network.** I was mesmerized by the project, not just for its prestigious funding but also for uniting outstanding universities, professors, and industries under a single project, driven by the challenge of contributing to a more sustainable world. It was everything I was looking for and more. From the outset, I knew I wanted to be part of it, and I decided to apply for the ESR9 position, focusing on *Cradle-to-grave sustainability of novel lime-based materials*. I felt I had enough experience in the chemistry of building materials, combined with my chemical engineering background, to make me a competitive candidate. The position promised a deep understanding of sustainability quantification.

The preparation for the application was far from a walk in the park. One of the initial challenges was language-related, as my English was not the brightest. My dear friend Nerea, an exceptional English teacher and the best baker of *torta de coco y dulce de leche*, helped me successfully tackle the TOEFL exam while enjoying her daily company. She and Jose were my pillars, supporting me from the moment I decided to apply until the end of my PhD journey. We spent long afternoons working on my application, from crafting my cover letter to conducting mock interviews, discussing, and refining my presentation and speech. Jose and Nere, I am blessed to have you as friends, and a part of my present success is because of your unwavering support. All the effort invested bore fruit, and I was shortlisted for an interview. There, I encountered Prof. Koenders again, and *unexpectedly*, one of my biggest science inspirations, Prof. Nele De Belie from Ghent University, was also present. The interview, which took place almost in summer, was intense. I remember Nele asked me many sharp questions, but nothing made me sweat more than the sole idea of meeting her, whose work I had read extensively. A few months later, at the supermarket making groceries, I received an email from the project manager of SUBLime. I could only read "***congratulations,***" "***you ranked first,***" "***would you accept the position?***" I dropped everything I had in my hands and started jumping and screaming "***Vamos locooooo!!***" in the middle of the supermarket. People stared as if I were crazy, but in that moment, I cared about nothing. It was the exact moment that my dream, which began in 2018, was becoming a reality three years later.

I was ecstatic and anxious about my future, but moving abroad is no easy feat. Early in 2021, I relocated to Darmstadt, and it has been a roller coaster of emotions ever since. My time here has been one of the most incredible experiences of my life, and I could not have asked for better colleagues.

In the first place, I would like to acknowledge that this research has been carried out within the **Marie Sklodowska-Curie project SUBLime**. I would like to thank the European Union's Horizon 2020 research and innovation programmee, for the funding that made SUBLime possible (**Grant Agreement n◦955986)**. I want to express my gratitude to all those who made SUBLime possible, especially those from whom I learned a great deal every time we had the chance to meet — Prof. Lourenço, Prof. Azehna, Prof. Navarro, Dr. Pereira, Frederik Verhelst, Dr. Oertel, and M.Sc. Gräf.

A big thank you to all my esteemed ESR colleagues from the SUBLime project, especially my dear *amigas* Dulce (how much you have supported me, and how lucky I am of being your friend!) and Teodora, Franco, Vadim, Monika, and Guilherme. Your friendship is one of the most valuable treasures from this time. The entire WiB team was equally important. Eddie, thank you for giving me the freedom and encouraging me to dare to think, always leaving the door to your office kindly open to discuss. Thank you, Neven, for your valuable feedback and engaging discussions. Together with Max, Maxi, Donglin, Peng, Nico, Liliya and Kira, you have collectively contributed to making our research group an enriching and wonderful place to work. A special thanks goes to our secretary, Aysen, for saving me so many times when I made mistakes, always being there with a kind word, a laugh, or some advice. You are invaluable to this team. I also want to thank all the Concrete & Environment research team at Magnel Laboratory in Ghent, my second home for six months during the SUBLime project. Nele, the only thing that could make me admire you even more was having the privilege of working with you. Thank you for your invaluable support. Roberto and Juan, my office mates at Magnel and dear Latino friends, brought joy to my days in Ghent and made me feel at home. I am so thankful that our lives crossed.

Lime is one of the oldest and most long-lasting building materials; even nowadays, lime plasters are found in Egyptian pyramids built around 4000 BCE. However, I am convinced that nothing in the world has more strength and durability than the love of my family — *Papa, Mama, y Valentina* — who supported me unconditionally. To them, my deepest appreciation, and thanks.

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Alkylbenzene sulfonate	**ABS**
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Aquatic Eutrophication	**AQE**
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Abstract

This doctoral thesis thoroughly explores the feasibility of attaining net-zero and carbon-negative transformations in lime-based construction materials, with a particular focus on masonry applications, employing advanced life-cycle assessment modelling.

The primary focus lies in the production stage, where the research seamlessly integrates circular economy practices, a transition towards eco-friendlier energy sources, and optimized production systems, leveraging innovative kiln technologies. By successfully implementing these decarbonization strategies throughout the entire life cycle, the study envisions scenarios in which lime-based rendering materials serve as a carbon sink during their service life, leading to carbon-negative transformations.

The research underscores the transformative nature of repositioning CO_2 from a critical emission to a valuable by-product, presenting a notable competitive advantage for the lime industry. Supported by quantitative evidence, the thesis envisages a future where CO_2 evolves into a valuable resource for green energy and sustainable products. To bring this vision to fruition, the thesis proposes a comprehensive action plan finely tuned to environmental objectives within the 2050 European Action Plan, specifically tailored for the lime-based building materials industry. This research not only provides essential insights into sustainable practices but also delineates a strategic roadmap for the lime sector to embrace a more environmentally conscious and economically viable future.

Summary

Since the late 1960s, various environmental issues, such as energy consumption, resource efficiency, air pollution, and waste management, have become integral parts of the public agenda. The mining industry, particularly for lime, significantly contributes to these problems. In 2020, approximately 400 million tons of calcite ($CaCO_3$) were extracted globally to produce calcium oxide (lime), resulting in substantial CO_2 emissions due to the high-temperature decomposition of $CaCO_3$ at around 1000°C. On a global scale, an average of 475 million tons of CO_2 were released, with nearly two-thirds being a consequence of unavoidable limestone decomposition, and the rest stemming from fuel combustion in the kiln. The extensive global impacts of lime manufacturing can be attributed to its versatility, serving diverse applications, from construction to steel production, sugar refining, and water purification.

Lime stands as one of the world's oldest and most vital construction materials, having been extensively applied in constructions worldwide, including ancient Israel (7000 BCE), Syria (4250 BC), China (2000 BC), and various civilizations such as the Mayan, Inca, and Aztec (500 BC). Over time, lime has played various roles, from structural functions to decorative techniques. It is a crucial material for conserving European built heritage and is widely employed in modern masonry systems, encompassing aerated and asphalt mortars, bedding mortars, rendering materials, and ready-mixes designed for specific applications like thermal insulation, fire resistance, and fungi prevention. The unique microstructure provided by lime as a binder allows masonry to accommodate small movements in building structure, gives smooth finishes, and porosity of the matrix enables buildings to breathe. Considering all its advantages, lime consumption is not expected to decrease; rather, it is anticipated to increase, exacerbating the environmental challenges associated with lime-based materials manufacturing.

Climate change, recognized as a global emergency transcending borders, has prompted international organizations such as the United Nations and the International Panel on Climate Change to issue warnings about the challenging prospect of limiting global warming to 1.5°C, as outlined in the Paris Agreement. This has led to stringent legislations imposing significant challenges on the building materials industry, both environmentally and economically due to carbon taxes. The European Climate Law sets the ambitious goal for Europe's economy and society to become climate-neutral by 2050. Despite the adverse context created by the current level of emissions, it is crucial to remember a quote attributed to Albert Einstein: "*In the midst of every crisis lies a great opportunity*". In such challenging times, each sector must develop its own decarbonization plan. For the lime-based building materials sector, important questions are raised:

- ► Is achieving net-zero emissions in lime-based construction materials feasible?
- ► Can carbon-negative transformations be realized?
- ► What changes should be implemented in the current production processes for these transformations to take place?
- ► How can we rigorously quantify net-zero and carbon-negative transformations in lime-based construction materials?
- ► Which life-cycle stages in lime-based materials must be included in these assessments?
- ► What measures are necessary to expedite the decarbonization process?

These pivotal questions form the foundation of this doctoral thesis, offering essential insights to design sustainable plans towards a climate-neutral scenario for the entire lime sector. A key consideration is the understanding that hydrated lime constitutes around 80% of the CO_2 emissions in ready-mix lime-based rendering materials. Successfully decarbonizing these materials requires a fundamental redesign of manufacturing processes. Achieving this redesign transforms the substantial CO_2 emissions from a significant challenge into an opportunity that can provide the sector with a competitive advantage. Adapting to new challenges and a dynamic context and applying an integrated set of decarbonization strategies are crucial for success.

This thesis rigorously evaluates and quantifies the feasibility of achieving net-zero and carbon-negative transformations throughout the life-cycle of lime-based rendering materials. An integral decarbonization strategy is implemented in life-cycle assessment calculations at the manufacturing stage. This strategy incorporates principles of circular economy, direct separation reactors coupled with carbon capture technologies, and the utilization of sustainable energy sources. Another critical factor integrated into the environmental and economic impact assessments of lime-based materials is the carbonation occurring during the application phase of the render. Lime distinguishes itself among other materials as its well-documented carbon uptake during its service life is not only scientifically well-established but also constitutes a vital reaction necessary for fulfilling its intended application as a building material.

To ensure a robust quantification of net-zero and carbon-negative scenarios, two primary challenges are identified. First, obtaining background data for modelling life-cycle inventories is challenging, as they are commonly sourced from generic, inflexible, and often unreliable databases. The quality of input data significantly influences the results and their reliability. The second challenge is the lack of comprehensive sustainability studies for modern lime-based construction materials, a critical factor in establishing a baseline for envisioning carbon reductions and benchmarking alternatives with enhanced sustainability.

The introductory section of Chapter 1 in the doctoral thesis establishes the motivation, hypothesis, and research objectives. The thesis is structured as a compilation of articles that address these objectives,

offering quantitative evidence to demonstrate the feasibility of achieving net-zero and carbon-negative scenarios in modern lime-based masonry rendering materials. In Chapter 2, a comprehensive state-of-the-art report provides context to understand the importance of decarbonizing lime manufacturing in the context of combating climate change. It delves into the specific characteristics of lime-based building materials, particularly in modern masonry applications such as renders or plasters. The chapter also elucidates the fundamental scientific principles governing the hardening mechanism of lime-based materials, with a focus on carbonation. Finally, the discussion includes a thorough exploration of identified research gaps in the environmental impact assessment of lime-based materials.

Chapter 3 introduces a parametrized life-cycle inventory modelling methodology, combining concepts from process engineering, materials science, and environmental engineering. This methodology, consistently applied throughout the thesis, incorporates scientific principles and transparent parameterization, enabling practitioners to calculate methodologically sound and well-grounded inventories. Through collaboration with Sievert, one of the largest ready-mix manufacturers in Germany and a key partner in the SUBLime network, the developed methodology was integrated with first-hand data from the industry. In Chapter 4, critical variables in the current production process are determined, alongside a comprehensive quantification of the impact of various components in modern mortars, renders, and plasters, establishing the most thorough analysis available in the literature of current lime-based masonry mortars and renders.

To evaluate the potential impact of each decarbonization strategy, individual aspects were thoroughly examined in dedicated chapters, identifying challenges and opportunities toward achieving net-zero and carbon-negative transformations.

Chapter 5 delves deeply into circular economy practices within lime-based render manufacturing, centering on the significance of closed and open-loop upcycling strategies. The chapter introduces a cradle-to-cradle design for a lime-based render, proposing a comparison between the technical and environmental performance of a completely recycled render and a render in its first life. Within the open-loop upcycling strategy, the environmental and economic performance of two distinct materials, namely paper mill sludge and carbide lime (a by-product of the acetylene industry), is rigorously compared from Cradle-to-Grave against modern lime-based renders. This comparative analysis extends beyond the reduction of CO_2 emissions to address the broader goals of minimizing raw materials extraction and reducing waste landfilling. The findings underscore the potential of these strategies as promising avenues for sustainability within the lime-based render manufacturing process.

Chapter 6 provides comprehensive insights into the environmental impact of current energy scenarios in the manufacturing of hydrated lime. Additionally, the chapter explores achievable improvements through

the utilization of low-carbon energy sources, shedding light on alternative pathways for reducing the carbon footprint associated with lime production.

An exhaustive comparison of current kiln technologies and energy sources is presented in Chapter 7, introducing an enhanced direct separator reactor coupled with carbon capture systems and decarbonized energy sources. This improved production system not only allows for the complete recovery of pure CO_2 but also facilitates its utilization in a diverse range of products and industries. The chapter integrates the current knowledge generated in the thesis, covering the entire lifecycle from Cradle-to-Grave, and underscores that achieving net-zero and even carbon-negative scenarios for lime-based renders is feasible in the framework of the scenario analysis. Such an approach not only contributes to environmental sustainability but also brings about positive economic impacts. Achieving rapid decarbonization of the lime-based construction materials industry requires a coordinated action plan that balances the needs of the private sector, environmental protection, and societal well-being within a regulatory framework that ensures equitable implementation of political decisions.

A final noteworthy contribution of this research lies in its quantitative assessment, supporting a transformative shift in mindset regarding CO_2 emissions, reframing them from a dramatic obstacle to a valuable by-product. Despite being traditionally perceived as a significant industrial challenge over the past decades, the lime sector stands out for its capacity to produce pure CO_2 through the application of a decarbonization framework, as previously discussed. This paradigm shift views the substantial CO_2 emissions not as a threat but as a competitive opportunity. Progress in establishing a robust CO_2 value chain is evident, with collaborative efforts spanning manufacturers, governments, policymakers, the market, and the scientific community. This concerted approach anticipates a future where CO_2 evolves into a precursor for green energy sources and sustainable products. The imminent prospect of CO_2 contributing to the realization of net-zero emissions signifies a shift from dreams to concrete reality in the short and medium term.

Zusammenfassung

Seit den späten 1960er Jahren sind verschiedene Umweltfragen wie Energieverbrauch, Ressourceneffizienz, Luftverschmutzung und Abfallwirtschaft zu integralen Bestandteilen der öffentlichen Agenda geworden. Die Bergbauindustrie, insbesondere für Kalk, trägt erheblich zu diesen Problemen bei. Im Jahr 2020 wurden weltweit etwa 400 Millionen Tonnen Calcit ($CaCO_3$) abgebaut, um Calciumoxid (Kalk) herzustellen, was zu erheblichen CO_2-Emissionen aufgrund der Hochtemperatur-Zersetzung von $CaCO_3$ bei rund 1000°C führte. Auf globaler Ebene wurden durchschnittlich 475 Millionen Tonnen CO_2 freigesetzt, wobei fast zwei Drittel eine Folge der unvermeidlichen Zersetzung von Kalkstein und der Rest aus der Verbrennung von Brennstoffen im Brennofen resultierten. Die umfangreichen globalen Auswirkungen der Kalkherstellung können auf seine Vielseitigkeit zurückgeführt werden, die von Bauwesen über Stahlproduktion, Zuckerreinigung bis zur Wasseraufbereitung reicht.

Kalk gilt als eines der ältesten und wichtigsten Baustoffe der Welt und wurde weltweit in Konstruktionen eingesetzt, einschließlich im antiken Israel (7000 v. Chr.), Syrien (4250 v. Chr.), China (2000 v. Chr.) und verschiedenen Zivilisationen wie den Maya, Inka und Azteken (500 v. Chr.). Im Laufe der Zeit hat Kalk verschiedene Funktionen von strukturellen Aufgaben bis zu dekorativen Techniken gespielt. Es ist ein entscheidender Werkstoff für die Erhaltung des europäischen Kulturerbes und wird weitreichend in modernen Mauerwerksystemen eingesetzt, darunter luft- und asphaltgebundene Mörtel, Bettungsmörtel, Verputzmaterialien und Fertigmischungen für spezifische Anwendungen wie Wärmedämmung, Brandschutz und Schimmelpilzprävention. Die einzigartige Mikrostruktur, die Kalk als Bindemittel bietet, ermöglicht es dem Mauerwerk, kleine Bewegungen in der Gebäudestruktur aufzunehmen, sorgt für glatte Oberflächen und die Porosität der Matrix ermöglicht Gebäuden zu atmen. Angesichts all seiner Vorteile wird nicht erwartet, dass der Kalkverbrauch abnimmt; vielmehr wird erwartet, dass er zunimmt und die mit der Herstellung von kalkbasierten Materialien verbundenen Umweltprobleme verschärft.

Der Klimawandel, der als globale Notlage ohne Grenzen anerkannt wird, hat internationale Organisationen wie die Vereinten Nationen und den Weltklimarat veranlasst, vor den herausfordernden Aussichten zu warnen, die mit der Begrenzung der globalen Erwärmung auf 1,5°C gemäß dem Pariser Abkommen verbunden sind. Dies hat zu strengen Gesetzgebungen geführt, die der Baustoffindustrie sowohl umwelt- als auch wirtschaftlich aufgrund von CO_2-Steuern erhebliche Herausforderungen auferlegen. Das Europäische Klimagesetz setzt das ehrgeizige Ziel, dass die Wirtschaft und Gesellschaft Europas bis 2050 klimaneutral werden sollen. Trotz des ungünstigen Kontexts aufgrund des aktuellen Emissionsniveaus ist es wichtig, sich an ein Zitat von Albert Einstein zu erinnern: "Inmitten jeder Krise liegt eine große Chance". In solch herausfordernden Zeiten muss jeder Sektor seinen eigenen Dekarbonisierungsplan entwickeln. Für den Sektor der kalkbasierten Baustoffe werden wichtige Fragen aufgeworfen:

- Ist die Erreichung von Netto-Null-Emissionen bei kalkbasierten Baumaterialien ein realistisches Ziel?
- Können kohlenstoffnegative Transformationen in diesem Kontext verwirklicht werden?
- Welche Veränderungen sind erforderlich, um in den aktuellen Produktionsprozessen diese Transformationen zu ermöglichen?
- Wie können wir Netto-Null- und kohlenstoffnegative Transformationen bei kalkbasierten Baumaterialien genau quantifizieren?
- Welche Lebenszyklusphasen von kalkbasierten Materialien sollten in diesen Bewertungen einbezogen werden?
- Welche Maßnahmen sind notwendig, um den Dekarbonisierungsprozess zu beschleunigen?

Diese entscheidenden Fragen bilden das Fundament dieser Doktorarbeit und bieten wesentliche Einblicke, um nachhaltige Pläne für ein klimaneutrales Szenario für den gesamten Kalksektor zu entwerfen. Eine zentrale Überlegung ist das Verständnis, dass gelöschter Kalk etwa 80% der CO_2-Emissionen in Fertigmischungen auf Kalkbasis ausmacht. Eine erfolgreiche Dekarbonisierung dieser Materialien erfordert eine grundlegende Neugestaltung der Herstellungsprozesse. Durch die Umsetzung dieser Neugestaltung werden die erheblichen CO_2-Emissionen von einer bedeutenden Herausforderung zu einer Möglichkeit, die dem Sektor einen Wettbewerbsvorteil verschaffen kann. Die Anpassung an neue Herausforderungen und einen dynamischen Kontext sowie die Anwendung eines integrierten Sets von Dekarbonisierungsstrategien sind entscheidend für den Erfolg.

Diese Arbeit bewertet und quantifiziert eingehend die Machbarkeit von netto-null und kohlenstoffnegativen Transformationen im gesamten Lebenszyklus von auf Kalk basierenden Verputzmaterialien. Eine integrale Dekarbonisierungsstrategie wird in den Berechnungen der Lebenszyklusanalyse in der Herstellungsphase umgesetzt. Diese Strategie integriert Prinzipien der Kreislaufwirtschaft, direkte Separationsreaktoren in Verbindung mit Kohlenstoffabscheidungstechnologien und die Nutzung nachhaltiger Energiequellen. Ein weiterer kritischer Faktor, der in die Umwelt- und Wirtschaftsauswirkungsbeurteilungen von kalkbasierten Materialien integriert ist, ist die Carbonatisierung während der Anwendungsphase des Verputzes. Kalk zeichnet sich unter anderen Materialien aus, da seine gut dokumentierte CO_2-Aufnahme während seiner Nutzungsdauer nicht nur wissenschaftlich fundiert ist, sondern auch eine wesentliche Reaktion darstellt, die für die Erfüllung seiner beabsichtigten Anwendung als Baustoff notwendig ist.

Um eine robuste Quantifizierung von netto-null und kohlenstoffnegativen Szenarien sicherzustellen, werden zwei Hauptprobleme identifiziert. Erstens ist es herausfordernd, Hintergrunddaten für die Modellierung von Lebenszyklusinventaren zu erhalten, da sie häufig aus generischen, unflexiblen und oft

unzuverlässigen Datenbanken stammen. Die Qualität der Eingabedaten beeinflusst die Ergebnisse und deren Zuverlässigkeit erheblich. Die zweite Herausforderung besteht im Mangel an umfassenden Nachhaltigkeitsstudien für moderne kalkbasierte Baustoffe, ein entscheidender Faktor zur Festlegung eines Ausgangspunkts für die Vorstellung von Kohlenstoffreduktionen und zum Benchmarking von Alternativen mit verbesserter Nachhaltigkeit.

Die einführende Sektion von Kapitel 1 in der Doktorarbeit legt die Motivation, die Hypothese und die Forschungsziele fest. Die Arbeit ist als Zusammenstellung von Artikeln strukturiert, die diese Ziele behandeln und quantitative Beweise liefern, um die Machbarkeit von netto-null und kohlenstoffnegativen Szenarien in modernen auf Kalk basierenden Mauerwerksputzmaterialien zu demonstrieren. In Kapitel 2 bietet ein umfassender Bericht zum aktuellen Stand der Technik Kontext, um die Bedeutung der Dekarbonisierung der Kalkherstellung im Kontext des Kampfes gegen den Klimawandel zu verstehen. Er geht auf die spezifischen Merkmale von kalkbasierten Baustoffen ein, insbesondere in modernen Anwendungen wie Verputzen oder Stuck. Das Kapitel erläutert auch die grundlegenden wissenschaftlichen Prinzipien, die den Verfestigungsmechanismus von kalkbasierten Materialien regeln, mit einem Schwerpunkt auf der Carbonatisierung. Schließlich beinhaltet die Diskussion eine gründliche Erkundung der identifizierten Forschungslücken in der Umweltauswirkungsbeurteilung von kalkbasierten Materialien.

Kapitel 3 führt eine parametrisierte Modellierungsmethodik für den Lebenszyklus-Inventaransatz ein, die Konzepte aus Verfahrenstechnik, Werkstoffwissenschaft und Umwelttechnik kombiniert. Diese Methodik, die konsequent in der gesamten Arbeit angewendet wird, integriert wissenschaftliche Prinzipien und transparente Parameterisierung, wodurch Praktiker methodologisch fundierte und gut fundierte Inventare berechnen können. Durch die Zusammenarbeit mit Sievert, einem der größten Hersteller von Fertigmischungen in Deutschland und einem wichtigen Partner im SUBLime-Netzwerk, wurde die entwickelte Methodik mit aus erster Hand stammenden Daten aus der Branche integriert. In Kapitel 4 werden kritische Variablen im aktuellen Produktionsprozess bestimmt, zusammen mit einer umfassenden Quantifizierung der Auswirkungen verschiedener Bestandteile in modernen Mörteln, Verputzen und Stuck, was die gründlichste Analyse in der Literatur von aktuellen Kalk-basierten Mauerwerksmörteln und -verputzen darstellt.

Um das potenzielle Auswirkung jeder Dekarbonisierungsstrategie zu bewerten, wurden einzelne Aspekte in dedizierten Kapiteln gründlich untersucht, wobei Herausforderungen und Chancen zur Erreichung von netto-null und kohlenstoffnegativen Transformationen identifiziert wurden.

Kapitel 5 geht ausführlich auf Praktiken der Kreislaufwirtschaft in der Herstellung von Kalkverputzen ein und konzentriert sich auf die Bedeutung von geschlossenen und offenen Kreislauf-Upcycling-Strategien. Das Kapitel stellt ein Cradle-to-Cradle-Design für einen Kalkverputz vor und schlägt einen Vergleich zwischen der technischen und Umweltleistung eines vollständig recycelten Verputzes und eines Verputzes in seinem ersten Leben vor. Im Rahmen der offenen Kreislauf-Upcycling-Strategie wird die Umwelt- und Wirtschaftsleistung von zwei unterschiedlichen Materialien, nämlich Papiermühlenschlamm und Karbidkalk (ein Nebenprodukt der Acetylenindustrie), von der Wiege bis zur Bahre gegen moderne Kalkverputze rigoros verglichen. Diese vergleichende Analyse geht über die Reduzierung von CO_2-Emissionen hinaus und behandelt die breiteren Ziele der Minimierung von Rohstoffextraktion und der Reduzierung von Abfalldeponierung. Die Ergebnisse unterstreichen das Potenzial dieser Strategien als vielversprechende Ansätze für die Nachhaltigkeit in der Herstellung von Kalkverputzen.

Kapitel 6 bietet umfassende Einblicke in die Umweltauswirkungen aktueller Energie Szenarien bei der Herstellung von gelöschtem Kalk. Darüber hinaus untersucht das Kapitel mögliche Verbesserungen durch die Nutzung von kohlenstoffarmen Energiequellen und beleuchtet alternative Wege zur Reduzierung des CO_2-Fußabdrucks, der mit der Kalkproduktion verbunden ist.

In Kapitel 7 wird ein erschöpfender Vergleich aktueller Brennöfen und Energiequellen vorgestellt und ein verbessertes Direktseparationsreaktor in Verbindung mit Kohlenstoffabscheidungssystemen und dekarbonisierten Energiequellen eingeführt. Dieses verbesserte Produktionssystem ermöglicht nicht nur die vollständige Rückgewinnung von reinem CO_2, sondern erleichtert auch seine Verwendung in einer vielfältigen Palette von Produkten und Branchen. Das Kapitel integriert das in der Arbeit generierte aktuelle Wissen, das den gesamten Lebenszyklus von der Wiege bis zur Bahre abdeckt, und betont, dass die Erreichung von netto-null und sogar kohlenstoffnegativen Szenarien für Kalkverputze im Rahmen der Szenarioanalyse möglich ist. Ein solcher Ansatz trägt nicht nur zur Umwelt nachhaltigkeit bei, sondern bringt auch positive wirtschaftliche Auswirkungen mit sich. Die rasche Dekarbonisierung der kalkbasierten Baustoffindustrie erfordert einen koordinierten Aktionsplan, der die Bedürfnisse des Privatsektors, des Umweltschutzes und des gesellschaftlichen Wohlbefindens in einem Regelwerk ausbalanciert, das eine gerechte Umsetzung politischer Entscheidungen gewährleistet.

Ein letzter bemerkenswerter Beitrag dieser Forschung liegt in ihrer quantitativen Bewertung, die eine transformative Veränderung des Denkansatzes zu CO_2-Emissionen unterstützt, indem sie sie von einem dramatischen Hindernis zu einem wertvollen Nebenprodukt umgestaltet. Trotz ihrer traditionellen Wahrnehmung als bedeutende industrielle Herausforderung in den letzten Jahrzehnten zeichnet sich der Kalksektor durch seine Fähigkeit aus, reines CO_2 durch die Anwendung eines Dekarbonisierungsrahmens zu produzieren, wie zuvor diskutiert. Dieser Paradigmenwechsel betrachtet die erheblichen CO_2-Emissionen nicht als Bedrohung, sondern als Wettbewerbschance. Fortschritte bei der Schaffung einer robusten CO_2-Wertschöpfungskette sind offensichtlich, wobei kooperative Bemühungen Hersteller, Regierungen, politische Entscheidungsträger, den Markt und die wissenschaftliche Gemeinschaft umfassen. Dieser gemeinsame Ansatz lässt auf eine Zukunft hoffen, in der CO_2 zu einem Vorläufer für grüne Energiequellen und nachhaltige Produkte wird.

Samenvatting

Sinds eind jaren zestig zijn verschillende milieukwesties, zoals energieverbruik, grondstoffenefficiëntie, luchtverontreiniging en afvalbeheer, integrale onderdelen geworden van de publieke agenda. De mijnbouwsector algemeen en de ontginning van kalk in het bijzonder, draagt aanzienlijk bij aan deze problemen. In 2020 werd wereldwijd ongeveer 400 miljoen ton calciet ($CaCO_3$) gewonnen om calciumoxide (kalk) te produceren, wat resulteert in een aanzienlijke CO_2-uitstoot door de ontbinding van $CaCO_3$ bij hoge temperaturen, rond de 1000°C. Op wereldwijde schaal werd gemiddeld 475 miljoen ton CO_2 uitgestoten, waarvan bijna twee derde het gevolg was van deze onvermijdelijke chemische ontbinding van kalksteen en de rest afkomstig was van brandstofverbranding in de oven. Deze omvangrijke wereldwijde impact van kalkproductie kan worden toegeschreven aan het veelzijdige gebruik, met toepassingen variërend van bouw tot staalproductie, suikerraffinage en waterzuivering.

Kalk staat bekend als een van 's werelds oudste en meest vitale bouwmaterialen, uitgebreid toegepast in constructies over de hele wereld, waaronder het oude Israël (7000 v.Chr.), Syrië (4250 v.Chr.), China (2000 v.Chr.), en verschillende beschavingen zoals deze van de Maya's, Inca's en Azteken (500 v.Chr.). In de loop der tijden heeft kalk verschillende functies vervuld, van structurele functies tot decoratieve. Het is een cruciaal materiaal voor het behoud van het Europese gebouwenerfgoed en wordt veel gebruikt in moderne metselsystemen, waaronder cellenbeton en asfaltmortels, beddingmortels, pleistermaterialen en kant-en-klare mengsels die zijn ontworpen voor specifieke toepassingen zoals thermische isolatie, brandwerendheid en schimmelpreventie. De unieke microstructuur die kalk als bindmiddel biedt, stelt metselwerk in staat kleine bewegingen in de bouwstructuur op te vangen, zorgt voor gladde afwerkingen en de porositeit van de matrix maakt het mogelijk dat gebouwen "ademen". Gezien al deze voordelen, wordt niet verwacht dat het verbruik van kalk zal afnemen; eerder wordt verwacht dat het zal toenemen, waardoor de milieuproblemen die gepaard gaan met de productie van kalkhoudende materialen verergeren.

Klimaatverandering, erkend als een wereldwijde noodsituatie die grenzen overschrijdt, heeft internationale organisaties zoals de Verenigde Naties en het Intergouvernementeel Panel inzake Klimaatverandering aangezet tot het signaleren van de uitdagingen verbonden aan de beperking van de opwarming van de aarde tot 1,5°C, zoals uiteengezet in het Parijse Akkoord. Dit heeft geleid tot strenge wetgeving die aanzienlijke uitdagingen oplegt aan de bouwmaterialenindustrie, zowel op milieugebied als economisch vanwege de koolstofbelastingen. De Europese Klimaatwet stelt het ambitieuze doel dat de economie en samenleving van Europa tegen 2050 klimaatneutraal moeten zijn. Ondanks de ongunstige context die wordt gecreëerd door het huidige niveau van de emissies, is het cruciaal om een citaat van Albert Einstein te onthouden: "In elke crisis schuilt een geweldige kans." In dergelijke uitdagende tijden moet elke sector zijn eigen decarbonisatieplan ontwikkelen. Voor de sector van kalkhoudende bouwmaterialen worden belangrijke vragen gesteld:

- ► Is het haalbaar om netto -zero emissies te bereiken bij kalkhoudende bouwmaterialen?
- ► Kunnen koolstofnegatieve transformaties gerealiseerd worden?
- ► Welke veranderingen moeten worden doorgevoerd in de huidige productieprocessen om deze transformaties plaats te laten vinden?
- ► Hoe kunnen we netto zero en koolstofnegatieve transformaties in kalkhoudende bouwmaterialen nauwkeurig kwantificeren?
- ► Welke levenscyclusfasen van kalkhoudende materialen moeten worden opgenomen in deze beoordelingen?
- ► Welke maatregelen zijn nodig om het decarbonisatieproces te versnellen?

Deze cruciale vragen vormen de basis van dit proefschrift en bieden essentiële inzichten om duurzame plannen te maken voor een klimaatneutraal scenario voor de gehele kalksector. Een belangrijke overweging is dat gebluste kalk ongeveer 80% van de CO2-uitstoot door kant-en-klare kalkgebaseerde pleistermaterialen vormt. Het succesvol decarboniseren van deze materialen vereist een fundamentele herziening van productieprocessen. Het realiseren van deze herziening transformeert de aanzienlijke CO2-uitstoot van een uitdaging in een kans die de sector een concurrentievoordeel kan bieden. Het aanpassen aan nieuwe uitdagingen en een dynamische context en het toepassen van een geïntegreerde reeks decarbonisatiestrategieën is cruciaal voor het succes.

Dit proefschrift evalueert en kwantificeert grondig de haalbaarheid van het bereiken van netto zero en koolstofnegatieve transformaties gedurende de gehele levenscyclus van kalkgebaseerde pleistermaterialen. Een integrale decarbonisatiestrategie wordt toegepast in de levenscyclusbeoordeling tijdens de productiefase. Deze strategie omvat principes van de circulaire economie, directe scheidingsreactoren gekoppeld aan koolstofafvangtechnologieën en het gebruik van duurzame energiebronnen. Een ander cruciaal element dat is geïntegreerd in de milieu- en economische impactbeoordelingen van kalkmaterialen, is de carbonatatie die plaatsvindt tijdens de toepassingsfase van de pleister. Slechts voor weinig materialen is het potentieel voor koolstofopname goed gedocumenteerd, nochtans is dit een spontane reactie die essentieel is voor het vervullen van hun rol als bouwmaterialen.

Om een robuuste kwantificering van netto zero en koolstofnegatieve scenario's te waarborgen, worden twee belangrijke uitdagingen geïdentificeerd. Ten eerste is het verkrijgen van achtergrondgegevens voor het modelleren van levenscyclusinventarissen uitdagend, omdat deze vaak afkomstig zijn uit generieke, inflexibele en vaak onbetrouwbare databases. De kwaliteit van de invoergegevens beïnvloedt de resultaten en hun betrouwbaarheid aanzienlijk. De tweede uitdaging is het gebrek aan uitgebreide duurzaamheidsstudies voor moderne kalkgebaseerde bouwmaterialen, een cruciale factor bij het vaststellen van het referentieniveau voor het visualiseren van koolstofverminderingen en het benchmarken van alternatieven met verbeterde duurzaamheid.

De inleidende sectie van Hoofdstuk 1 in het proefschrift legt de motivatie, hypothese en onderzoeksdoelstellingen vast. Het proefschrift is gestructureerd als een compilatie van artikelen die deze doelstellingen behandelen en kwantitatief bewijs leveren om de haalbaarheid van het bereiken van netto-nul en koolstofnegatieve scenario's in moderne kalkgebaseerde pleistermaterialen aan te tonen. In Hoofdstuk 2 biedt een uitgebreid state-of-the-art rapport context om het belang te begrijpen van het decarboniseren van kalkproductie in de context van de strijd tegen klimaatverandering. Het gaat dieper in op de specifieke kenmerken van kalkgebaseerde bouwmaterialen, met name in moderne metselwerktoepassingen zoals pleisters of stucwerk. Het hoofdstuk verduidelijkt ook de fundamentele wetenschappelijke principes die de verhardingsmechanismen van kalkgebaseerde materialen regeren, met een focus op carbonatatie. Ten slotte omvat de discussie een grondige verkenning van geïdentificeerde onderzoekslacunes in de milieubeoordeling van kalkgebaseerde materialen.

Om het potentiële effect van elke decarbonisatiestrategie te evalueren, werden individuele aspecten grondig onderzocht in toegewijde hoofdstukken, waarbij uitdagingen en kansen werden geïdentificeerd om netto zero en koolstofnegatieve transformaties te bereiken.

Hoofdstuk 5 duikt diep in praktijken van de circulaire economie met betrekking tot de productie van kalkgebaseerde pleisters, met de nadruk op het belang van gesloten en open kringloop upcycling-strategieën. Het hoofdstuk introduceert de beschouwing van een kalkgebaseerde pleister van wieg tot wieg (cradle-to-cradle), waarbij een vergelijking wordt voorgesteld tussen de technische en milieuprestaties van een volledig gerecyclede pleister en een pleister in zijn eerste leven. Binnen de open kringloop upcycling-strategie wordt de milieu- en economische prestatie van twee verschillende materialen, namelijk papierslib en calciumcarbide (een bijproduct van de acetyleenindustrie), grondig vergeleken van wieg tot graf met moderne kalkgebaseerde pleisters. Deze vergelijkende analyse reikt verder dan de vermindering van de CO_2-uitstoot. Ook de doelstellingen rond het minimaliseren van de grondstoffenwinning en het verminderen van het afvalstorten worden bekeken.

Hoofdstuk 6 biedt uitgebreide inzichten in de milieueffecten van huidige energiescenario's voor de productie van gebluste kalk. Bovendien worden in het hoofdstuk haalbare verbeteringen door het gebruik van koolstofarme energiebronnen besproken, en werpt het licht op alternatieve wegen voor het verminderen van de koolstofvoetafdruk die gepaard gaat met kalkproductie.

Een doorgedreven vergelijking van huidige oventechnologieën en energiebronnen wordt gepresenteerd in Hoofdstuk 7, waarin een verbeterde directe scheidingsreactor wordt geïntroduceerd die is gekoppeld aan koolstofafvangsystemen en gedecarboniseerde energiebronnen. Dit verbeterde productiesysteem maakt niet alleen de volledige terugwinning van zuivere CO_2 mogelijk, maar vergemakkelijkt ook het gebruik ervan in een divers scala van producten en industrieën. Het hoofdstuk integreert de huidige kennis die in het proefschrift is gegenereerd, waarbij de gehele levenscyclus van wieg tot graf wordt behandeld, en benadrukt dat het bereiken van netto zero en zelfs koolstofnegatieve scenario's voor kalkgebaseerde pleisters haalbaar is binnen het kader van de scenarioanalyse. Een dergelijke benadering draagt niet alleen bij aan milieuduurzaamheid, maar brengt ook positieve economische impact met zich mee. Het snel decarboniseren van de kalkgebaseerde bouwmaterialenindustrie vereist een gecoördineerd actieplan dat de behoeften van de particuliere sector, milieubescherming en het welzijn van de samenleving in evenwicht houdt binnen een regelgevend kader dat een rechtvaardige uitvoering van politieke beslissingen waarborgt.

Een laatste opmerkelijke bijdrage van dit onderzoek ligt in de kwantitatieve beoordeling, die een transformerende verschuiving in denken ondersteunt met betrekking tot CO_2-uitstoot, waarbij deze wordt herschikt van een onoverkomelijk obstakel tot een waardevol bijproduct. Ondanks dat CO_2-uitstoot traditioneel werd gezien als een aanzienlijke industriële uitdaging in de afgelopen decennia, onderscheidt de kalksector zich door zijn vermogen om zuiver CO_2 te produceren door de toepassing van een decarbonisatiekader, zoals eerder besproken. Deze paradigmaverschuiving beschouwt de aanzienlijke CO_2-uitstoot niet als een bedreiging, maar als een competitieve kans. Het vestigen van een robuuste CO_2-waardeketen is nodig, met gezamenlijke inspanningen van fabrikanten, overheden, beleidsmakers, de markt en de wetenschappelijke gemeenschap. Deze gezamenlijke aanpak voorziet in een toekomst waarin CO_2 evolueert tot een basismateriaal voor groene energiebronnen en duurzame producten. Het op handen zijnde vooruitzicht dat CO_2 bijdraagt aan de realisatie van netto zero emissies betekent een verschuiving van dromen naar concrete realiteit op de korte en middellange termijn.

Resumen

Desde finales de la década de 1960, diversas problemáticas ambientales, como el consumo de energía, la eficiencia de recursos, la contaminación del aire y la gestión de residuos, se han convertido en partes integrales de la agenda pública. La industria minera, en particular la de la cal, contribuye significativamente a estos problemas. En 2020, se extrajeron aproximadamente 400 millones de toneladas de calcita ($CaCO_3$) a nivel mundial para producir óxido de calcio (cal), lo que resultó en emisiones sustanciales de CO_2 debido a la descomposición de $CaCO_3$ a altas temperaturas, alrededor de 1000°C. A nivel global, se liberaron en promedio 475 millones de toneladas de CO_2, con casi dos tercios como consecuencia de la inevitable descomposición de la piedra caliza y el resto derivado de la combustión de combustibles en el horno. Los extensos impactos globales de la fabricación de cal se atribuyen a su versatilidad, sirviendo a diversas aplicaciones, desde la construcción hasta la producción de acero, refinación de azúcar y purificación de agua.

La cal se erige como uno de los materiales de construcción más antiguos y vitales del mundo, habiéndose aplicado extensamente en construcciones a nivel mundial, incluyendo la antigua Israel (7000 a.C.), Siria (4250 a.C.), China (2000 a.C.) y diversas civilizaciones como la Maya, Inca y Azteca (500 a.C.). Con el tiempo, la cal ha desempeñado diversos roles, desde funciones estructurales hasta técnicas decorativas. Es un material crucial para la conservación del patrimonio arquitectónico europeo y se emplea ampliamente en sistemas modernos de albañilería, abarcando morteros aireados y asfálticos, morteros de asiento, materiales de revestimiento y mezclas listas para usos específicos como aislamiento térmico, resistencia al fuego y prevención de hongos. La microestructura única proporcionada por la cal como aglutinante permite que la albañilería se adapte a pequeños movimientos en la estructura de los edificios, brinda acabados suaves y la porosidad de la matriz permite que los edificios respiren. Considerando todas sus ventajas, no se espera que el consumo de cal disminuya; más bien, se anticipa que aumentará, exacerbando los desafíos ambientales asociados con la fabricación de este tipo de materiales.

El cambio climático, reconocido como una emergencia global que trasciende fronteras, ha llevado a organizaciones internacionales como las Naciones Unidas y el Panel Intergubernamental sobre el Cambio Climático a emitir advertencias sobre la difícil perspectiva de limitar el calentamiento global a 1.5°C, como se establece en el Acuerdo de París. Esto ha dado lugar a legislaciones estrictas que imponen desafíos significativos a la industria de materiales de construcción, tanto desde el punto de vista ambiental como económico debido a impuestos a la emisión de CO_2. La Ley Europea del Clima establece el ambicioso objetivo de que la economía y la sociedad de Europa se vuelvan climáticamente neutrales para 2050. A pesar del contexto adverso creado por el nivel actual de emisiones, es crucial recordar una cita atribuida a Albert Einstein: "*En medio de cada crisis yace una gran oportunidad*". En tiempos tan desafiantes, cada

sector debe desarrollar su propio plan de descarbonización. Para el sector de materiales de construcción a base de cal, se plantean preguntas importantes:

- ► ¿Es factible lograr emisiones netas cero en materiales de construcción a base de cal?
- ► ¿Se pueden realizar transformaciones carbono-negativas?
- ► ¿Qué cambios deben implementarse en los procesos de producción actuales para que estas transformaciones tengan lugar?
- ► ¿Cómo podemos cuantificar rigurosamente las transformaciones netas cero y carbono-negativas en materiales de construcción a base de cal?
- ► ¿Qué etapas del ciclo de vida de los materiales a base de cal deben incluirse en estas evaluaciones?
- ► ¿Qué medidas son necesarias para acelerar el proceso de descarbonización?

Estas preguntas fundamentales constituyen la base de esta tesis doctoral, proporcionando ideas esenciales para diseñar planes sostenibles hacia un escenario climáticamente neutro en todo el sector de la cal. Una consideración clave es reconocer que la cal hidratada constituye aproximadamente el 80% de las emisiones de CO_2 en los materiales de revestimiento a base de cal listos para usar. Descarbonizar con éxito estos materiales exige un rediseño fundamental de los procesos de fabricación, transformando las sustanciales emisiones de CO_2 de un desafío significativo en una oportunidad que puede brindar al sector una ventaja competitiva. Adaptarse a nuevos desafíos y a un contexto dinámico, y aplicar un conjunto integrado de estrategias de descarbonización, son cruciales para el éxito.

Esta tesis evalúa rigurosamente y cuantifica la viabilidad de lograr transformaciones neutrales en carbono y negativas en carbono a lo largo del ciclo de vida de los materiales de revestimiento a base de cal. Se implementa una estrategia integral de descarbonización en los cálculos de evaluación del ciclo de vida en la etapa de fabricación. Esta estrategia incorpora principios de economía circular, reactores de separación directa acoplados con tecnologías de captura de carbono y la utilización de fuentes de energía sostenibles. Otro factor crítico integrado en las evaluaciones ambientales y económicas de los materiales a base de cal es la carbonatación que ocurre durante la fase de aplicación del revestimiento. Pocos materiales acreditan un potencial tan bien documentado para la captura de carbono como la cal, constituyendo una reacción espontánea esencial para cumplir su función como materiales de construcción.

Para garantizar una cuantificación sólida de escenarios neutrales en carbono y negativos en carbono, se identifican dos desafíos principales. En primer lugar, obtener *background data* para modelar inventarios del ciclo de vida es desafiante, ya que comúnmente se obtienen de bases de datos genéricas, inflexibles y a menudo poco confiables. La calidad de los datos de entrada influye significativamente los resultados del modelo y, finalmente, en la toma de decisiones. El segundo desafío es la falta de estudios de sostenibilidad integrales para materiales modernos de construcción a base de cal, un factor crítico para establecer una línea base para proyectar reducciones de carbono y comparar alternativas con una mayor sostenibilidad.

La sección introductoria del Capítulo 1 en la tesis doctoral establece la motivación, la hipótesis y los objetivos de investigación. La tesis está estructurada como una compilación de artículos que abordan estos objetivos, ofreciendo evidencia cuantitativa para demostrar la viabilidad de lograr escenarios de emisión neta cero y carbono negativo en materiales modernos de revestimiento de albañilería a base de cal. En el Capítulo 2, un informe exhaustivo del estado del arte proporciona contexto para comprender la importancia de descarbonizar la fabricación de cal en el contexto de la lucha contra el cambio climático. Se adentra en las características específicas de los materiales de construcción a base de cal, especialmente en aplicaciones modernas de albañilería como revestimientos o enlucidos. El capítulo también aclara los principios científicos fundamentales que rigen el mecanismo de endurecimiento de los materiales a base de cal, con un enfoque en la carbonatación. Finalmente, la discusión incluye una exploración exhaustiva de las brechas en la literatura científica identificadas en la evaluación del impacto ambiental de los materiales a base de cal.

El Capítulo 3 presenta una metodología parametrizada de modelado de inventario del ciclo de vida, que combina conceptos de ingeniería de procesos, ciencia de materiales e ingeniería ambiental. Esta metodología, aplicada de manera consistente a lo largo de la tesis, incorpora principios científicos y parametrización transparente, permitiendo a los profesionales calcular inventarios metodológicamente sólidos y bien fundamentados. A través de la colaboración con Sievert, uno de los mayores fabricantes de morteros listos para usar en Alemania y un socio clave en la red SUBLime, se integró la metodología desarrollada con datos de primera mano de la industria. En el Capítulo 4, se determinan variables críticas en el proceso de producción actual, junto con una cuantificación integral del impacto de diversos componentes en los morteros, revestimientos y enlucidos modernos a base de cal, estableciendo el análisis más exhaustivo disponible en la literatura hasta el momento de los morteros y revestimientos de albañilería a base de cal actuales.

Para evaluar el impacto potencial de cada estrategia de descarbonización, se examinaron a fondo aspectos individuales en capítulos dedicados, identificando desafíos y oportunidades hacia la consecución de transformaciones neutrales y negativas en carbono.

El Capítulo 5 se sumerge profundamente en prácticas de economía circular en la fabricación de revestimientos a base de cal, centrando la atención en la importancia de las estrategias de reciclaje cerrado y de bucle abierto. El capítulo introduce un diseño de cuna a cuna para un revestimiento a base de cal, proponiendo una comparación entre el rendimiento técnico y ambiental de un revestimiento completamente reciclado y un revestimiento en su primera vida. Dentro de la estrategia de reciclaje de bucle abierto, se compara rigurosamente la performance ambiental y económica de dos materiales distintos, la escoria de pulpa de papel (residuo de la industrial del papel) y la cal de carburo (un subproducto de la industria del acetileno), desde la cuna hasta la tumba, frente a revestimientos modernos a base de cal. Este análisis comparativo se extiende más allá de la reducción de las emisiones de CO_2 para abordar los objetivos más amplios de minimizar la extracción de materias primas y reducir el vertido de residuos. Los resultados destacan el potencial de estas estrategias como vías prometedoras para la sostenibilidad dentro del proceso de fabricación de revestimientos a base de cal.

El Capítulo 6 brinda una visión integral del impacto ambiental de los escenarios energéticos actuales en la fabricación de cal hidratada. Además, el capítulo explora mejoras alcanzables mediante la utilización de fuentes de energía bajas en carbono, arrojando luz sobre vías alternativas para reducir la huella de carbono asociada a la producción de cal.

Una comparación exhaustiva de las tecnologías actuales de hornos y fuentes de energía se presenta en el Capítulo 7, introduciendo un reactor de separación directa mejorado acoplado con sistemas de captura de carbono y fuentes de energía descarbonizadas. Este sistema de producción mejorado no solo permite la recuperación completa de CO_2 puro, sino que también facilita su utilización en una amplia gama de productos e industrias. El capítulo integra el conocimiento actual generado en la tesis, cubriendo todo el ciclo de vida desde la cuna hasta la tumba, y destaca que lograr escenarios neutrales en carbono e incluso negativos en carbono para revestimientos a base de cal es factible dentro del marco del análisis de escenarios. Este enfoque no solo contribuye a la sostenibilidad ambiental, sino que también conlleva impactos económicos positivos. Lograr una descarbonización rápida de la industria de materiales de construcción a base de cal requiere un plan de acción coordinado que equilibre las necesidades del sector privado, la protección del medio ambiente y el bienestar social dentro de un marco normativo que garantice la implementación equitativa de decisiones políticas.

Una contribución final destacada de esta investigación radica en su evaluación cuantitativa, respaldando un cambio transformador en la mentalidad con respecto a las emisiones de CO_2, replanteándolas de un obstáculo dramático a un subproducto valioso. A pesar de ser percibidas tradicionalmente como un desafío industrial significativo en las últimas décadas, el sector de la cal destaca por su capacidad para producir CO_2 puro mediante la aplicación de un marco de descarbonización, como se discutió anteriormente. Este cambio de paradigma considera que las sustanciales emisiones de CO_2 no son una amenaza, sino una oportunidad competitiva. Se evidencia un progreso en el establecimiento de una cadena de valor robusta para el CO_2, con esfuerzos colaborativos que abarcan fabricantes, gobiernos, responsables políticos, el mercado y la comunidad científica. Este enfoque concertado anticipa un futuro en el que el CO_2 se convierta en un precursor de fuentes de energía renovable y productos sostenibles. La perspectiva inminente de que el CO_2 contribuya a la realización de emisiones netas cero permite pensar en transformaciones de la industria que dejarán de quedar en sueños para pasar a ser una realidad concreta en el corto y mediano plazo.

Chapter 1: Introduction

The introduction of the doctoral thesis comprehends the establishment of the motivation and hypothesis, as well as research objectives.

The thesis, is structured as a compilation of articles responding to the stated goals, providing quantitative evidence to demonstrate the feasibility of achieving net-zero and carbon-negative scenarios in modern lime-based masonry rendering materials.

1.1 Motivation and Hypothesis

Since the late 1960s different environmental issues related to energy consumption, resources efficiency, air pollution, waste management, among others, became part of the public agenda. The mining industry, particularly for lime, is still a significant contributor to all these problems, as in 2020 around 400 Mt of calcite were extracted worldwide to produce calcium oxide (lime). As a result, an average of 475 Mt of CO_2 were released globally, with almost two thirds of the emissions being completely unavoidable (limestone decomposition) and the rest due to fuel combustion in the lime kiln. Being calcium oxide the precursor for the manufacturing of lime-based construction materials (ranging from aerated concrete to mortars, renders and plasters), one essential next step to meet to achieve an enhanced sustainability in the sector is the redesign of the manufacturing processes.

A quote attributed to Albert Einstein says, *“in the midst of every crisis lies a great opportunity”*. Indeed, the decarbonization of the lime industry can be seen, instead of a problem, as an opportunity that provides the sector a competitive advantage. For this, it is necessary to remain adaptable to the new challenges and dynamic context and apply an integrated set of Decarbonization Strategies (DS), envisioning the lime plant of the future, and covering from relevant life-cycle phases of building materials (Figure 1.1). As a first strategy, the principles of circular economy to reduce raw materials extraction should be incorporated in the manufacturing process (DS1, Fig. 1.1). Furthermore, the calcination process in which the CO_2 emissions are concentrated, needs redesigning. In the first place, more sustainable energy sources, both fuel and electricity, will help to reduce the avoidable portion of the CO_2 emissions (DS2, Fig. 1.1). The contribution of the energy sources to carbon emissions are the avoidable portion of the total CO_2 released, because the fuel types in the lime kiln can be optimized.

Yet there is still a significant amount of unavoidable emissions, as a result of calcite decomposition, that cannot be tackled in a technical and cost-effective way by using the most widespread current kiln technologies. Most promising future technology is the direct separation reactor coupled with carbon capture systems (DS3, Fig. 1.1). Lime manufacturing has the unique advantage of producing pure CO_2 when calcite is decomposed (unavoidable emissions). If these emissions can be captured and not released to the environment, they will stop being a global challenge to be a business opportunity, reducing not only the environmental impact but also enhancing the economic sustainability by avoiding carbon taxes that European industries must pay within the European Trading System. Furthermore, the captured CO_2 holds potential to be used as a raw material in various industrial processes, from polymers to energy production, as well as to improve the properties of building materials through accelerated carbonation. Under this framework, CO_2 would transform from an emission into a valuable by-product [97]. However, due to the novelty of this technology, there is no existing record in the literature of a comprehensive LCA modelling study, and this serves as a primary motivating factor for undertaking this doctoral thesis.

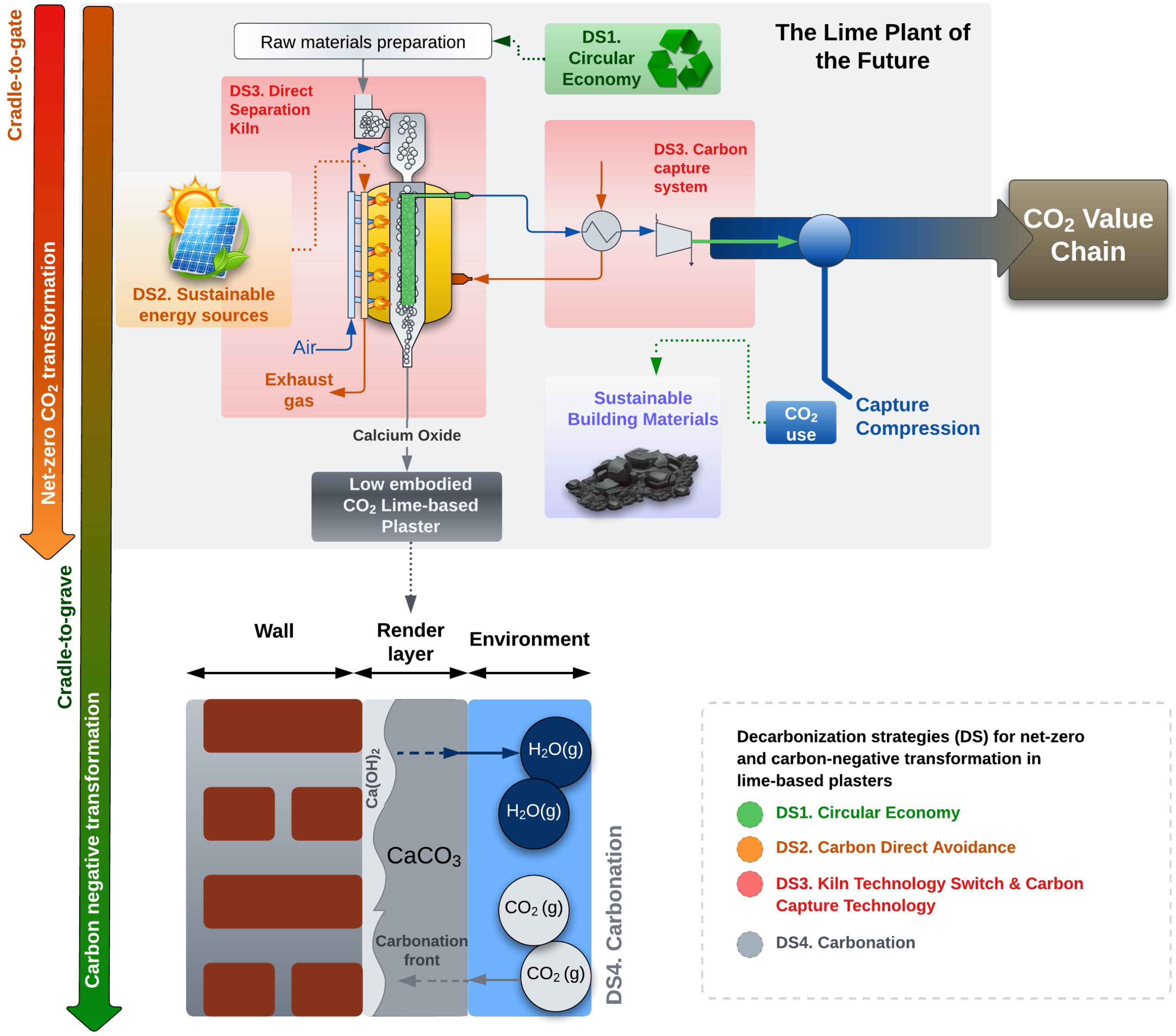

Figure 1.1 Image of the decarbonization strategies for net-zero and carbon-negative transformation in a lime-based plaster applied to a masonry unit. The scheme is not on scale, the render layer has been enlarged for illustration purposes.

By improving the manufacturing process of lime, net-zero transformations might be possible. This can in turn be beneficial for the lime-based construction industry, as the initial embodied emissions of the binder are being reduced. As a particular application of lime-based materials, the ones used for plastering purposes in masonry units are studied within this thesis. The final decarbonization strategy (DS4) implies the accounting of the CO_2 sequestration during the use phase of the plaster. The second biggest motivation of the doctoral thesis is to prove if it is feasible (to what extent) to achieve net-zero and carbon-negative transformations by optimizing the production process of lime, used for the manufacturing of lime-based building materials, incorporating in this assessment the specific performance during the use phase of a lime-based plaster layer.

This leads to the following **hypothesis**: by implementing the integral decarbonization strategies (DS1 to DS3) at the emission point (i.e., the lime manufacturing plant), net-zero transformations are feasible; and when the CO_2 sequestration by the lime-based plaster during the use phase is accounted for in the LCA (DS4) carbon-negative transformation can be achieved.

To illustrate the expected results, Figure 1.2 shows the carbon profile (through the Global Warming Potential indicator, GWP), measured in CO_2 emissions/Functional Unit (FU) from Cradle-to-Grave of two lime-based plasters manufactured through two different manufacturing processes. The FU used to compare the two scenarios is the amount of plaster with similar technical specifications (thermal conductivity, bond strength, etc.) required to keep 1m^2 of wall covered over the same period of time (End of Life).

Cradle-to-Gate emissions account for the manufacturing of hydrated lime, aggregates, and additives. In the business-as-usual (BAU) scenario, hydrated lime is produced in a traditional way, using limestone as main raw material, the current kiln technology is the Parallel Flow Regenerative Kiln (PFRK) and is powered by fossil fuel energy sources. The plaster is applied and during the use phase absorbs CO_2 from the environment. Even though the CO_2 indicator can be reduced due to carbonation during the service life of the plaster, relying on the use phase to achieve carbon neutrality is far from realistic. The real problem is that the starting point of emissions is so high, that even if the material could completely carbonate, it could never off-set the Cradle-to-Gate emissions. In a scenario with decarbonizing process (DP) in which the starting point of the emissions is reduced sufficiently by implementing strategies DS1 to DS3 (including capturing and storage or usage of the process bound CO_2), and the same rate of carbonation during the service life (DS4), net-zero and carbon-negative scenarios are more plausible.

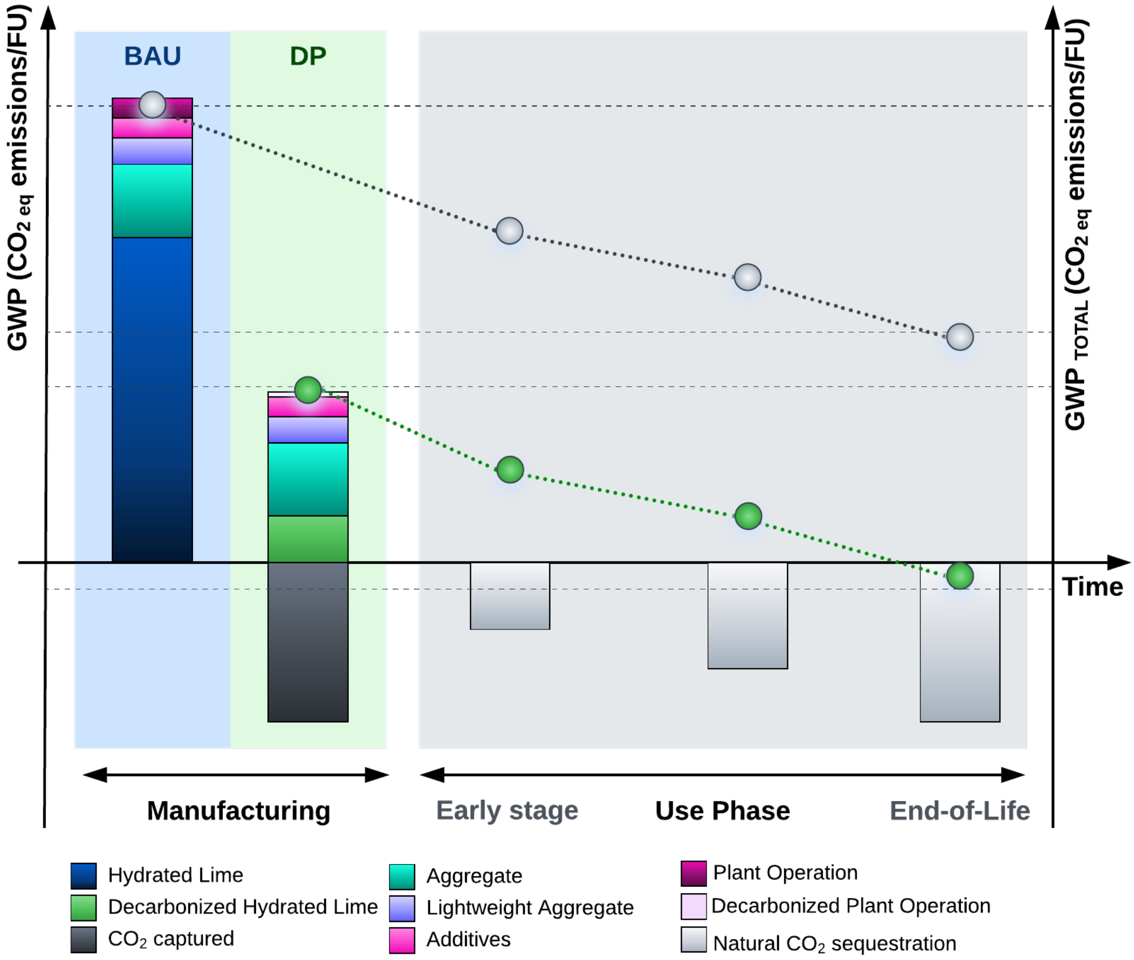

Figure 1.2 Expected carbon profile over the life-cycle of the plaster for the Business-as-Usual (BAU) and Decarbonized Process (DP) for hydrated lime manufacturing. The left axis shows the CO_2 emissions/FU at a specific stage of the life-cycle and the right axis shows the accumulated effect of each life-stage on the GWP indicator.

Building upon the formulated hypothesis, the following section outlines the general and specific objectives of this doctoral thesis.

1.2 Objectives

General objective

To demonstrate the feasibility of achieving net-zero and carbon-negative transformations for lime-based rendering material, an advanced life-cycle assessment methodology throughout its entire life cycle is proposed. This will be accomplished by implementing an integrated decarbonization strategy, incorporating principles of the circular economy, advancements in kiln technology featuring carbon capture systems, sustainable energy sources, and natural carbonation during the use phase.

Secondary objectives

Objective 1. To develop a robust LCA framework for calculating a methodologically sound and well-grounded life-cycle inventory, parameterized to facilitate the implementation of decarbonization strategies during the manufacturing stage of the life cycle.

Objective 2. To establish a robust baseline environmental profile for the manufacturing of modern lime-based renders in Germany used in contemporary masonry systems.

Objective 3. To assess the impact of circular economy principles, specifically closed-loop and open-loop upcycling, on the decarbonization of lime-based rendering throughout its life cycle.

Objective 4. To quantify the impact of both current and decarbonized energy sources on the manufacturing of hydrated lime, aiming to assess the potential for decarbonization in the production process.

Objective 5. To quantitatively compare the impact of a combined decarbonization strategy, which includes direct separation technologies with carbon capture and decarbonized energy sources, with the current scenario throughout the Cradle-to-Grave life cycle of lime-based rendering.

Objective 6. To comprehensively assess the feasibility of achieving net-zero and carbon-negative scenarios within the proposed decarbonization strategy and to discuss the necessary measures to expedite the overall decarbonization process.

1.3 Structure of the Thesis

The thesis is structured as a compilation of research articles, both in published and submitted states at the time of thesis submission (Table 1.1).

Table 1.1. Publications of the thesis and contribution of each manuscript to the specific goals of the doctoral thesis

	Research Articles	Contribution
Paper 1 ***Status*** *Published*	Laveglia A., Ukrainczyk N., De Belie, N., Koenders, E. (2023). ***"Decarbonizing the Lime-based Construction Materials Industry: A practical Guide for Cradle-to-Gate Life Cycle Inventory"*** Journal of Sustainable Production and Consumption (Elsevier, ISSN: 2352-5509). doi.org/10.1016/j.spc.2023.06.021	**Objective 1** Methodology development
Contribution statement	**Agustin Laveglia:** Conceptualization, Methodology, LCI calculations and analysis, discussion, Writing – original draft, paper preparation, Writing – review & editing, results, and discussion. **Neven Ukrainczyk:** Resources, Writing – review & editing, results and discussion, Supervision. **Nele De Belie:** Writing – review & editing, results and discussion, Supervision. **Eddie Koenders:** Resources, Writing – review & editing, results and discussion, Supervision.	
Paper 2 ***Status*** *Published*	Laveglia A., Sambataro, L., Ukrainczynk N., Oertel, T., De Belie, N., Koenders, E. (2023). ***"How to Improve the Cradle-to-Gate Environmental and Economic Sustainability in Lime-Based Construction Materials? Answers from a Real-life Case-study"***. Developments in the Built Environment (Elsevier, ISSN: 2666-1659). doi.org/10.1016/j.dibe.2023.100186	**Objective 2** Baseline scenario establishment
Contribution statement	**Agustin Laveglia:** Conceptualization, Methodology, materials, and processing, LCI, LCA, discussion, Writing – original draft, paper preparation, Writing – review & editing, results and discussion**. Luciano Sambataro:** Review & editing. **Neven Ukrainczyk:** Resources, Writing – review & editing, results and discussion and Supervision. Tina Oertel: Writing – review & editing, results, and discussion. **Nele De Belie:** Writing – review & editing, results and discussion, Supervision. **Eddie Koenders:** Resources, Writing – review & editing, results and discussion, Supervision.	
Paper 3 ***Status*** *Under review*	Laveglia A., Ukrainczyk N., De Belie, N., Koenders, E. ***"Circular Design, Materials Properties, Service Life and Cradle-to-Cradle LCA of Lime-based Building Materials"***. Resources, Conservation and Recycling (Elsevier, ISSN: 1879-0658).	**Objective 3** Decarbonization strategy 1&4
Contribution statement	**Agustin Laveglia:** Conceptualization, Methodology, Investigation, Formal Analysis, Validation, Data Curation, Writing – Review and Editing, Results and Discussion. **Dulce Valdez Madrid:** Methodology, Investigation, Formal Analysis, Validation, Data Curation, Writing – Review and Editing, Results and Discussion. **Neven Ukrainczyk:** Writing – Review and Editing, Results and Discussion, Resources, Supervision. **Veerle Cnudde:** Writing – Review and Editing, Results and Discussion, Resources, Supervision. **Nele De Belie:** Writing – Review and Editing, Results and Discussion, Resources, Supervision. **Eddie Koenders** Writing – Review and Editing, Results and Discussion, Resources, Supervision.	

Table 1.2. Publications of the thesis and contribution of each manuscript to the specific goals of the doctoral thesis *(continued)*

	Research Articles	Contribution
Paper 4 ***Status*** *Revised version submitted*	Laveglia A., Ukrainczyk N., De Belie, N., Koenders, E. ***"Cradle-to-Grave Environmental and Economic Sustainability of Lime-based Plasters Manufactured with Upcycled Materials".*** Journal of Cleaner Production (Elsevier, ISSN: 1879-1786).	**Objective 3** Decarbonization strategy 1&4
Contribution statement	**Agustin Laveglia:** Conceptualization, Methodology, LCI/LCA calculations and analysis, investigation, discussion, Writing – original draft, paper preparation, Writing – review & editing, results, and discussion. **Neven Ukrainczyk:** Writing – review & editing, results and discussion, Supervision, Resources. **Nele De Belie:** Writing – review & editing, results and discussion, Supervision. **Eddie Koenders:** Resources, results and discussion, Supervision.	
Paper 5 ***Status*** *Published*	Laveglia A., Sambataro, L., Ukrainczyk N., De Belie, N., Koenders, E. (2022). ***"Hydrated Lime Life-cycle Assessment: Current and Future Scenarios in four EU Countries"*** Journal of Cleaner Production (Elsevier, ISSN: 1879-1786). doi.org/10.1016/j.jclepro.2022.133224	**Objective 4** Decarbonization strategy 2
Contribution statement	**Agustin Laveglia:** Conceptualization, Methodology, LCI/LCA calculations and analysis, investigation, discussion, Writing – original draft, paper preparation, Writing – review & editing, results, and discussion. **Luciano Sambataro:** LCI calculation. **Neven Ukrainczyk:** Writing – review & editing, results and discussion, Supervision, Resources. **Nele De Belie:** Writing – review & editing, results and discussion, Supervision. **Eddie Koenders:** Resources, results and discussion, Supervision.	
Paper 6 ***Status*** *Under review*	Laveglia A., Ukrainczyk N., De Belie, N., Koenders, E. ***"From Quarry to Carbon Sink: Process-based LCA Modelling of Lime-based Construction Materials for Net-zero and Carbon-negative Transformations".*** Green Chemistry (Royal Society of Chemistry, ISSN 1463-9270)	**Objective 5 and 6** Integration of Decarbonization Strategies 2, 3 & 4
Contribution statement	**Agustin Laveglia:** Conceptualization, Methodology, LCI/LCA calculations and analysis, investigation, discussion, Writing – original draft, paper preparation, Writing – review & editing, results, and discussion. **Neven Ukrainczyk:** Writing – review & editing, results and discussion, Supervision, Resources. **Nele De Belie:** Writing – review & editing, results and discussion, Supervision. **Eddie Koenders:** Resources, results and discussion, Supervision.	

Serving as the backbone of the thesis, these research articles were meticulously crafted with the aim of contributing to each specified objective and, ultimately, advancing the overarching goal of the thesis. Each paper in this collection has played a distinct role and relates to the chapters of the thesis, as outlined in Figure 1.3.

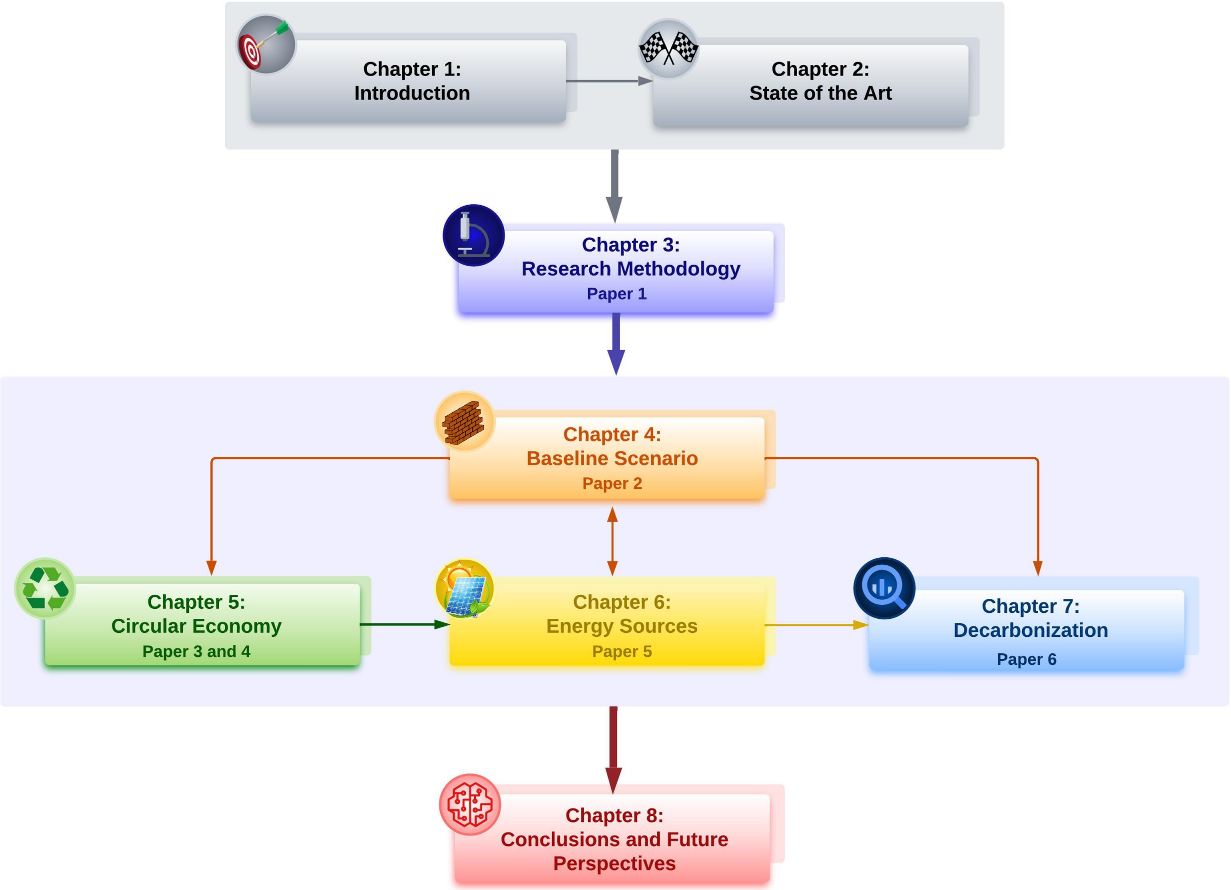

Figure 1.3. Structure of the doctoral thesis

A summary of the focal points addressed in each chapter is provided below, outlining their unique contributions to the overall research objectives and the organizational framework of the thesis.

Chapter 1: Introduction

The introduction of the doctoral thesis comprehends the establishment of the motivation and hypothesis, as well as research objectives. The thesis, is structured as a compilation of articles responding to the stated goals, providing quantitative evidence to demonstrate the feasibility of achieving net-zero and carbon-negative scenarios in modern lime-based masonry rendering materials.

Chapter 2: State of Art

The state of art provides a context to comprehend the significance of decarbonizing lime for combating climate change. Lime, an incredibly versatile material, not only holds a crucial role in modern construction and European cultural heritage but also is a key enabler to produce steel, clean water, building materials, and even sugar. This exceptional versatility underscores lime's importance in addressing a spectrum of societal needs, highlighting the relevance of this doctoral thesis dedicated to quantifying decarbonization strategies in its manufacturing process. Furthermore, the specific characteristics of lime-based building materials, specifically in modern masonry applications, as renders or plasters are discussed. The main scientific principles of the hardening mechanism of lime-based materials, carbonation, are described. Finally, identified research gaps in environmental impact assessment of lime-based materials, are thoroughly explored.

Chapter 3: Research Methodology | Contribution: Specific Goal 1 | Paper 1

This section proposes a methodology for the parametric calculation of Cradle-to-Gate life-cycle inventories based on process engineering concepts, mass and energy balances, materials science, and environmental engineering. The methodology is described in the form of a flow diagram and is illustrated in simple case-studies to show how to include in the inventory the decarbonization strategies proposed in the thesis (effect of energy sources, new kiln technologies, carbon capture systems, etc.). The calculation of the parametric inventories is used throughout the chapters.

Chapter 4: Cradle-To-Gate Life-Cycle Assessment of Current Lime-Based Rendering Materials | Contribution: Specific Goal 2 | Paper 2

To make comparisons with future scenarios with enhanced sustainability, the baseline scenario of the current situation should be established for lime-based rendering materials. In this chapter a real-case scenario is described in which a real manufacturing plant of lime-based mortars, renders and plasters was studied. An unprecedent life cycle inventory analysing different binders, aggregates and additives was built and used for the life cycle assessment calculation. A life cycle cost calculation was also made to evaluate the effect of carbon pricing on the total cost of the material.

Chapter 5: Applying the Concepts of Circular Economy in Lime-Based Rendering Materials | Contribution: Specific Goal 3| Papers 3 and 4

The circular economy strategy studied in this thesis has two approaches (i.e., two parts): closed-loop and open-loop upcycling.

The closed-loop upcycling is based on the philosophy of keeping materials in closed cycles (Cradle-to-Cradle) for as long as possible. As an application, the complete recyclability at the end of life of a lime-based render was experimentally addressed, evaluating relevant technical and durability properties of second life renders in comparison with first life renders. The decarbonization potential of the circular economy "Cradle-to-Cradle" against the linear economy "Cradle-to-Grave" is quantified through LCA. The results (Paper 3) are presented in the first part of this chapter.

When by-products from other markets or waste materials are introduced as secondary resources (SR) in a different manufacturing process to produce an added value, the strategy is called open-loop upcycling. A Cradle-to-Grave LCA was conducted for the case of lime-based renders manufactured with a by-product from the acetylene industry, carbide lime, and paper mill sludge, a residue from the paper industry. The intermediate treatments for the conditioning of the SR were designed based on the proposed methodology and the results were compared against the traditional scenario. The results (Paper 4) are presented in the second part of the chapter 3.

The chapter aims at giving an overview of potential decarbonization contributions when circular economy principles are applied over the life-cycle of a lime-based render.

Chapter 6: The Role of Energy Sources in Hydrated Lime Production | Contribution: Specific Goal 4 | Paper 5

The environmental impact of producing hydrated lime, a crucial precursor for lime-based construction materials, is markedly affected by the energy sources employed in the manufacturing plant. The selection of an energy source is intrinsically linked to the geographic location of the facility. In this chapter, a comprehensive scenario analysis and quantitative impact assessment are undertaken, utilizing projected trends in sustainable energy production for both energy and fuel up to the year 2050 in Germany, Belgium, Portugal, and Spain. The objective was to rigorously quantify the repercussions of this transition on the Cradle-to-Gate environmental impact. Subsequently, the potential decarbonization scenario for energy is juxtaposed with the existing scenario as of 2020.

Chapter 7: Towards Net-zero and Carbon-negative Transformations in Lime-Based Rending Materials | Contribution: Specific Goal 5 and 6 | Paper 6

This chapter reports a comprehensive decarbonization approach, involving Direct Separation Reactors (DSR) with different operation modes and eco-efficient energy sources in the production of hydrated lime. Environmental and economic impacts are calculated through an in-depth life-cycle Cradle-to-Grave assessment for a lime-based render. The feasibility of net-zero and carbon-negative scenarios in lime-based plasters, by combining the decarbonization strategies at the manufacturing plant with the natural carbonation, are proven. To realize a swift and effective decarbonization of the lime industry, a harmonized effort is imperative and involves balancing interests of the private sector, environmental protection, and promoting societal well-being, all within a supportive regulatory framework. The challenges and coordinated actions across the value chain are discussed.

Chapter 8: Conclusions and Future Perspectives | Contribution: General Objective

The findings of this thesis are synthesized to provide comprehensive insights into the environmental implications of the analysed decarbonization strategies. This section aims to highlight the key outcomes and their significance in advancing sustainability goals within the lime sector and, particularly, lime-based construction materials industry. Additionally, it delves into the broader implications of the research, discussing potential avenues for further investigation and exploring future perspectives on the continued decarbonization of manufacturing processes. The section serves as a critical reflection on the study's contributions while paving the way for future research directions and practical applications in the pursuit of more sustainable construction practices.

Chapter 2: State of the Art

The state of art provides a context to comprehend the significance of decarbonizing lime manufacturing for combating climate change.

Lime, an incredibly versatile material, not only plays a crucial role in modern construction and European cultural heritage but also has the unique capability to produce steel, clean water, building materials, and even sugar. This exceptional versatility underscores lime's importance in addressing a spectrum of societal needs, highlighting the relevance of this doctoral thesis dedicated to quantifying decarbonization strategies for its manufacturing process.

Furthermore, the specific characteristics of lime-based building materials, specifically in modern masonry applications, as renders or plasters are discussed. The main scientific principles of the hardening mechanism of lime-based materials, carbonation, are described. Finally, identified research gaps in environmental impact assessment of lime-based materials, are thoroughly explored.

2.1 A Real Environmental Crisis

The current environmental crisis urgently calls for a shift towards a green and circular economy, which requires rethinking the way humanity consumes virgin raw materials and produces and uses products and services. We are at a pivotal point in which further unsustainable decisions can lead to irreparable consequences for humanity [1].

Climate change is one of the most important environmental challenges of modern society. According to the Intergovernmental Panel on Climate Change (IPCC), Climate Change is *“a change in the state of the climate that can be identified… by changes in the mean and/or the variability of its properties, that persists for an extended period, typically decades or longer”* [2]. The climate itself is a long-term summary of weather (meteorological) conditions, which considers not only average conditions of rain, snow, wind, temperature, etc. but also its fluctuation in time at a specific place.

The Earth’s climate has been through significant variations (ice ages, sea-level changes, etc.) over many thousands of years. However, scientific evidence shows that global temperatures have risen with unusual speed over the last few decades. Human influence has been the dominant factor of observed warming since the middle of the 20th century, during the start of the industrial age. Figure 2.1 shows the distribution of average temperature increases in each country of the world according to their level of development. Human-induced warming in highly industrialized countries has reached approximately 1°C (likely between 0.8°C and 1.2°C) above pre-industrial levels in 2017, increasing at 0.2°C (likely between 0.1°C and 0.3°C) per decade [3].

The 2015 Paris Agreement seeks to limit the global temperature increase to 1.5°C, with the overarching goal of mitigating the potential impacts linked to climate change. These impacts span from the vulnerability of ecosystems and communities to the risks of sea-level rise and extreme weather events. According to the assessments by the Intergovernmental Panel on Climate Change (IPCC), the transition toward net-zero carbon dioxide scenarios is deemed crucial for achieving this objective and as a pivotal strategy in the collective effort to address climate change and protect the planet from its detrimental consequences.

Temperature rise to date has already resulted in profound alterations to human and natural systems, including increases in droughts, floods, and some other types of extreme weather; sea level rise; and biodiversity loss – these changes are causing unprecedented risks to vulnerable persons and populations. The most affected people live in low- and middle-income countries, some of which have experienced a decline in food security, which in turn is partly linked to rising migration and poverty. Small islands, megacities, coastal regions, and high mountain ranges are likewise among the most affected. Worldwide, numerous ecosystems are at risk of severe impacts, particularly warm-water tropical reefs and Arctic ecosystems [4].

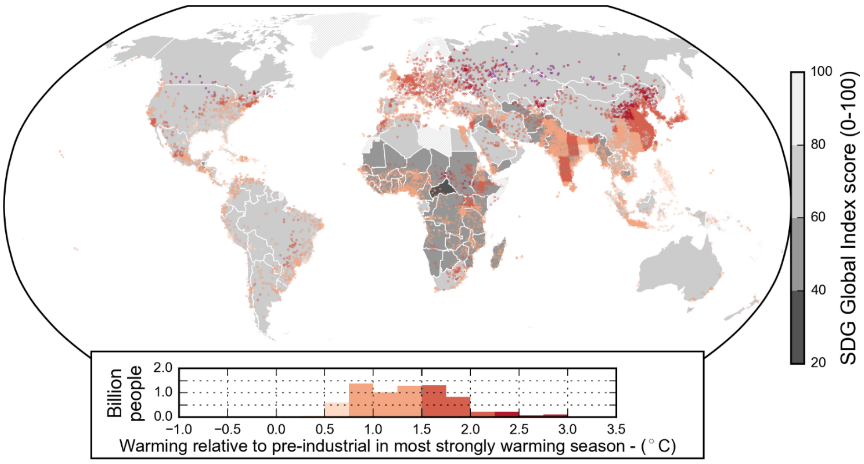

Figure 2.1. Classification of each country in the world according to the Sustainable Development Goals index (SDG), as the gray scale shown in the right axis. The histogram shows the amount of population living in regions experiencing different levels of warming (at 0.25°C increments). The country-specific data of the histogram is plotted on the world map on a colour scale. Extracted from: [2]

Three main factors govern the temperature of a planet: the solar energy influx, the albedo of the surface and the chemistry of the atmosphere (Figure 2.2). In an equilibrium state, the average net radiation at the top of the atmosphere is zero. A change either in the solar radiation or in the infrared radiation caused by a change in albedo of the surface or the chemistry of the atmosphere leads to an imbalance. If this imbalance is positive, the earth receives more incoming energy from sunlight than it irradiates to space. This net gain of energy causes an increment in the temperature of the Earth (i.e., global warming) [5].

Notably, humans have a direct effect on the last two factors. The albedo of the surface is a measure of how strongly it reflects the light from the sun and is modified as we extract virgin raw materials from the earth, create buildings, and dispose of residues, among others [6]. Of vital importance is the chemical composition of the atmosphere, mostly composed of nitrogen (78%) and oxygen (21%), which do not interact with the infrared radiation emitted by the Earth. There is around 0.1% of trace gases which do absorb and emit infrared radiation, called greenhouse gases (GHGs), and play an essential role in the Earth's energy budget. Among them, carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O) and ozone (O_3) are critical GHGs [2], [7]. Each of these gases have a global warming potential (GWP), which is a relative measure of how of how much heat GHG traps in the atmosphere. It is comparative to the amount of heat trapped by a similar mass of $CO_{2,}$ and it is calculated over a specific time interval.

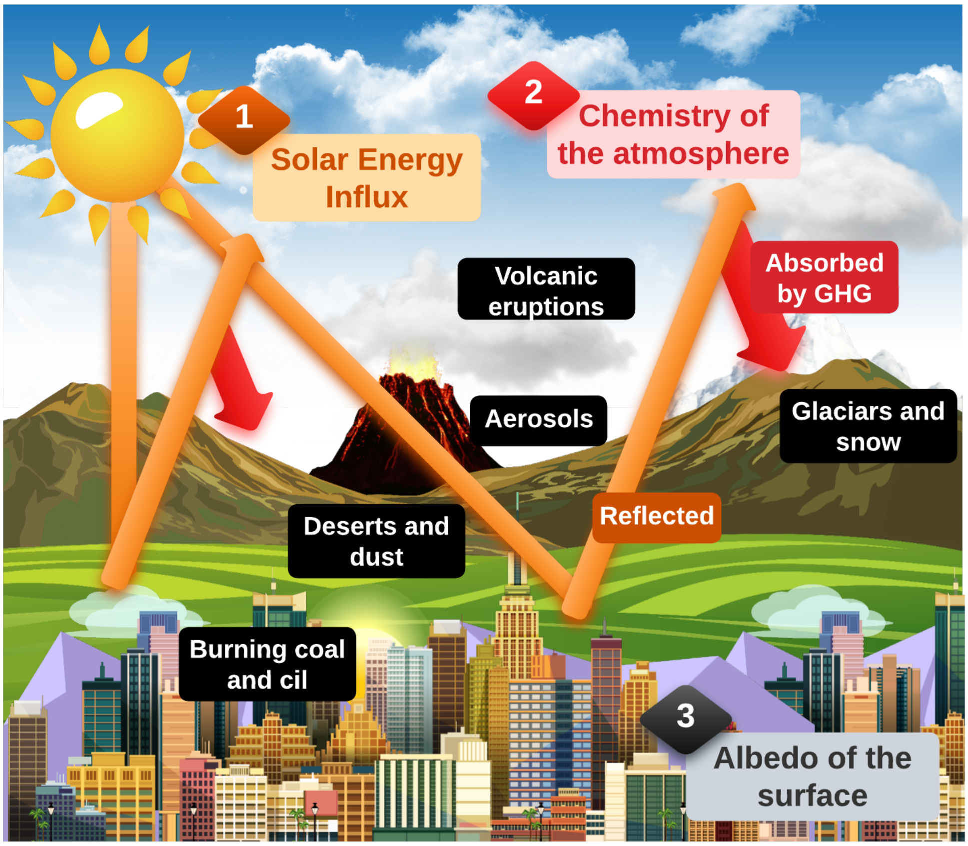

Figure 2.2. Factors affecting the temperature of a planet: solar energy influx, chemistry of the atmosphere and albedo of the surface

According to the European Environmental Agency, around 80% of the GHGs emissions are dominated by CO_2, followed by 11% CH_4, 6% N_2O, 2% of hydrofluorocarbons (HFCs) and 1% O_3. The sources of these emissions are distributed between the energy sector (77%), industrial processes (9%), agriculture (11%) and waste management (3%). The 77% attributed to the energy sector is distributed as follows: 31% corresponds to energy and heat, 12.4% to manufacturing and construction, 15% to transportation, 8.4% to other fuel combustions and 5.2% to fugitive emissions [8]. The statistics show that a significant amount of GHGs originated because of industrial processes dedicated to construction and building materials. The production of cement and lime (main binders present in these materials) can contribute from 9 to 12% of the anthropogenic CO_2 emissions [9], [10]. From this analysis, it becomes evident that the fight against climate change requires a decarbonization strategy for the construction and building materials sector.

2.2 Lime: An Essential but Often Unseen Material

While the primary focus of this thesis centers on the utilization of lime to be applied in the manufacturing of building materials, it is essential to recognize that the decarbonization of lime has far-reaching implications beyond the construction sector. Lime, an indispensable yet often overlooked material, plays a vital role in numerous daily activities undertaken by humanity. Consequently, reducing CO_2 emissions at the production stage can exert a substantial influence on several markets that rely on lime. This section is dedicated to comprehending the sources of emissions in lime production and examining the current progress in advancing towards the decarbonization of the manufacturing process.

2.2.1Origin of the Emissions

Lime stands as one of the world’s oldest and most vital construction materials. It has been found to be extensively applied in constructions all over the world, from ancient Israel (7000 BCE), Syria (4250 BC), China (2000 BC), to Mayan, Inca, and Aztec civilizations (500 BC) [11]–[13]. Since then, it has played distinct roles, from structural functions to decorative techniques [14]. However, besides its use as building material, lime is a critical precursor used in a wide variety of industries and applications. It is a material that can be used to produce steel, masonry, and sugar, as well as to purify water [15].

Because of its versatility, in 2020 alone, the lime industry represented a global market value of approximately 42 billion USD, accompanied by a total lime production of 396 million metric tons (Mt). The industrial CaO production contributed 475.2 Mt of CO_2, with 24 Mt allocated to the European market, lagging behind Asia, the leading producer boosted by a rapidly expanding sector [15]. Predictions indicate a substantial 21% growth in the market and a 25% increase in lime production by 2028, posing significant challenges to environmental sustainability. Consequently, the projected sustained high production of lime threatens to escalate environmental impacts, endangering the realization of the United Nations' goal to limit global warming to 1.5°C [2]. Coupled with the 90 €/t CO_2 carbon pricing under the European Trading System (ETS) in 2022, these factors pose substantial hurdles to achieving environmental sustainability goals and maintaining market competitiveness [16], [17].

The chemical name for lime is calcium oxide (CaO), and it is produced through a relatively straightforward industrial process, illustrated in Figure 2.3. Lime manufacturing requires the extraction of calcium carbonate ($CaCO_3$) from the Earth crust, which goes through several crushing and grinding operations to produce a fine powder. The next step is the thermal treatment of the powder in an industrial kiln. The calcination process is the most energy intensive step in the lime production process, accounting for around 90% of the total energy consumption and 99% of the specific impact of the entire process on global warming ([15], [18], [19]). Depending on the kiln the energy demand can vary from 3 to 9 MJ/kg CaO, with the Parallel Flow Regenerative Kiln (PFRK) being the most used technology in Europe (Figure 2.3) [20].

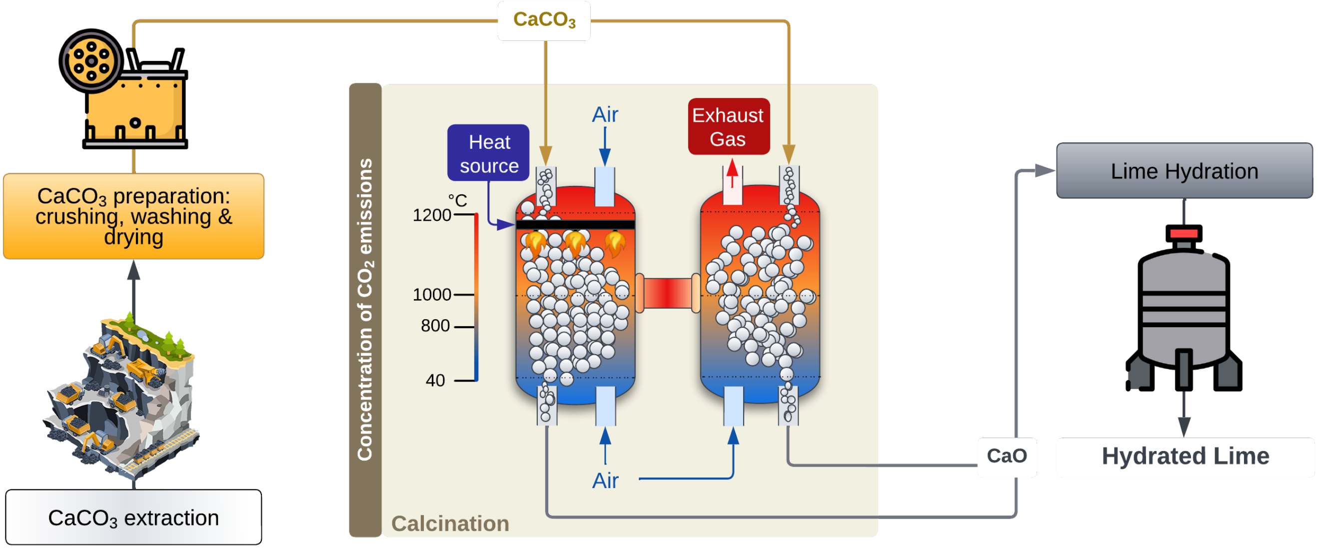

Figure 2.3. Industrial manufacturing of lime and hydrated lime from Cradle-to-Gate of the factory. The figure highlights the concentration point of CO_2 emissions at the calcination operation, because of the heat source combustion and limestone decomposition.

The PFRK has proven to be successful in the lime industry for approximately two decades [21]. This technology comprises two interconnected units, namely a pre-heater, burning, and cooling zones (Fig. 2.3). At the top of the first calcination unit, burners are strategically positioned, and the combustion gases travel in the opposite direction to the limestone feed, thus improving the energy use in the kiln. During this process, limestone undergoes decomposition into lime and carbon dioxide, occurring within a temperature range of 900 to 1100°C. Through the cross-over channel, the combustion gases and the CO_2 resulting from calcite decomposition are injected into the second unit, continuing their counter-directional movement to the limestone introduced at the top. This design ensures highly efficient calcination.

The production of 1 ton of CaO generates around 1.2 tons of CO_2, most of it generated by the calcination operation, as highlighted in Figure 2.3 [15]. Almost two thirds of the emissions generated in the kiln are categorized as *unavoidable emissions*, as they are related to the chemical reactions during the calcination of calcium carbonate ($CaCO_3$) to produce CaO (0.78 kg CO_2/kg CaO) (Reaction 2.1). The rest are considered *avoidable emissions* and are associated with electricity consumption and fuel combustion during the manufacturing processes [10]. While the PFRK stands out as a highly efficient technology, a notable drawback is that it combines avoidable and unavoidable emissions, releasing them together. This blending of emissions makes the technology both technologically challenging and economically costly. Hydrated Lime ($Ca(OH)_2$) used in the construction industry is produced by mixing calcium oxide and water in a unit process called slaking (Reaction 2.2).

$$CaCO_3(s) \rightarrow CaO\ (s) + CO_2(g) \qquad \textbf{Reaction 2.1}$$

$$CaO\ (s) + H_2O\ (l) \rightarrow Ca(OH)_2\ (s)$$ **Reaction 2.2**

The examination of the manufacturing process underscores that achieving decarbonization in the lime industry necessitates addressing both avoidable and unavoidable emissions. Over the past decade, there have been gradual, yet consistent strides made in adapting the production processes of cement and lime to meet the challenges of the contemporary era.

2.2.2 Recent Advances in the Decarbonization of Lime Manufacturing

As demonstrated in the preceding section, the pivotal focus for decarbonizing lime-based building materials lies in addressing lime manufacturing. A significant milestone in this endeavour is the European Union's ambitious goals, targeting a net emissions reduction of at least 55% by 2030 compared to 1990 levels [22]. To achieve this objective, numerous EU-funded projects have been launched, engaging in close collaboration with the industrial sector to explore process emissions and potential decarbonization strategies. A summary of some of the most important projects in the European/UK region is depicted in Figure 2.4 (adapted from [23]).

From 2010 to 2015, a substantial portion of projects aimed to leverage renewable energy sources such as biomass, biogas, solar panels, and minor kiln adjustments to improve overall thermal efficiency. Despite these efforts, it is crucial to acknowledge that around 30% of total CO_2 emissions still stem from avoidable emissions, emphasizing the need for additional solutions to address the remaining unavoidable emissions. Regrettably, there has been limited progress in the literature regarding unavoidable emissions from lime production between 2010 and 2020. Only a few articles have explored strategies involving solvents, sorbents, mineralization, and alkalinization of waste materials, resulting in relatively modest technological advancements that are also expensive [24]–[27].

Since 2020, we have witnessed revolutionary innovations in traditional lime manufacturing, with a particular emphasis on kiln designs incorporating direct separation technology (DST). This technology allows for the direct capture of CO_2 during the calcination of $CaCO_3$ [28], overcoming the main limitation of the PFRK, in which avoidable and unavoidable emissions are mixed. Notable European Union initiatives, such as LEILAC1 and LEILAC2 (https://www.leilac.com/), with a combined budget of 55 million euros, have been dedicated to the development of the CALIX kiln equipped with DST technology. Their ambitious goal is to scale up production to 400 t/d [29]. Figure 2.5 illustrates the installed kiln demonstrator and a schematic representation of the working principle of this ground-breaking technology.

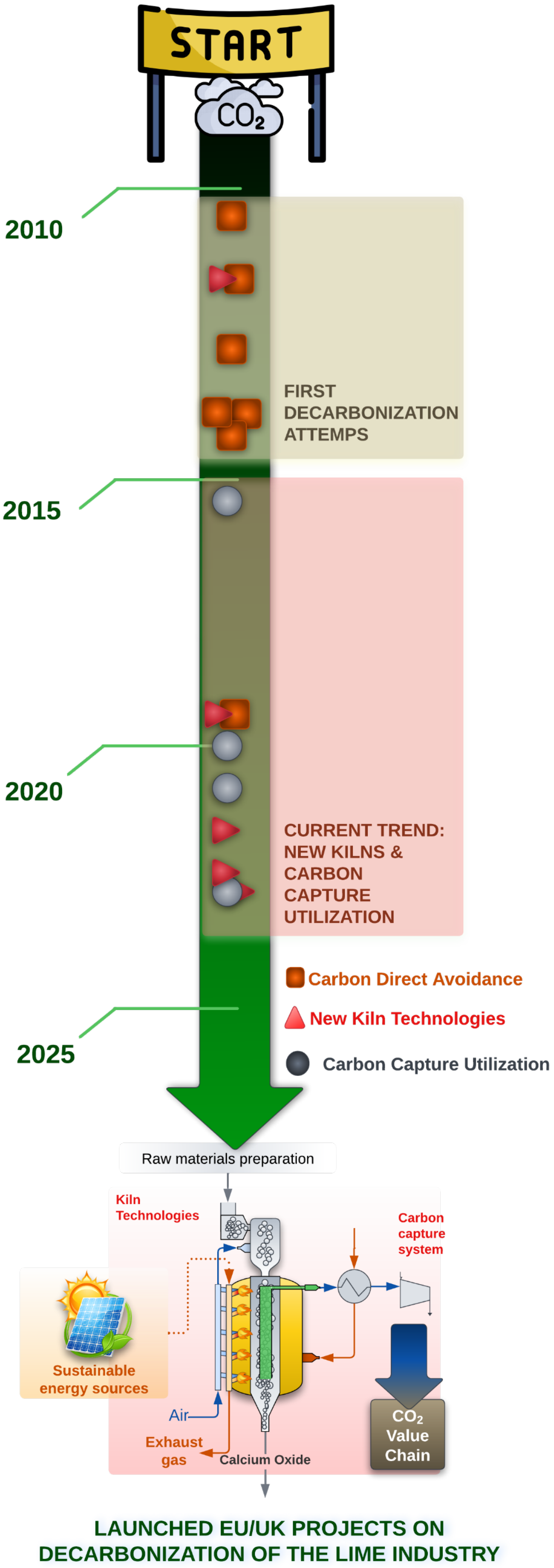

Figure 2.4. Launched European/UK projects on decarbonization of the lime industry (adapted from [23]).

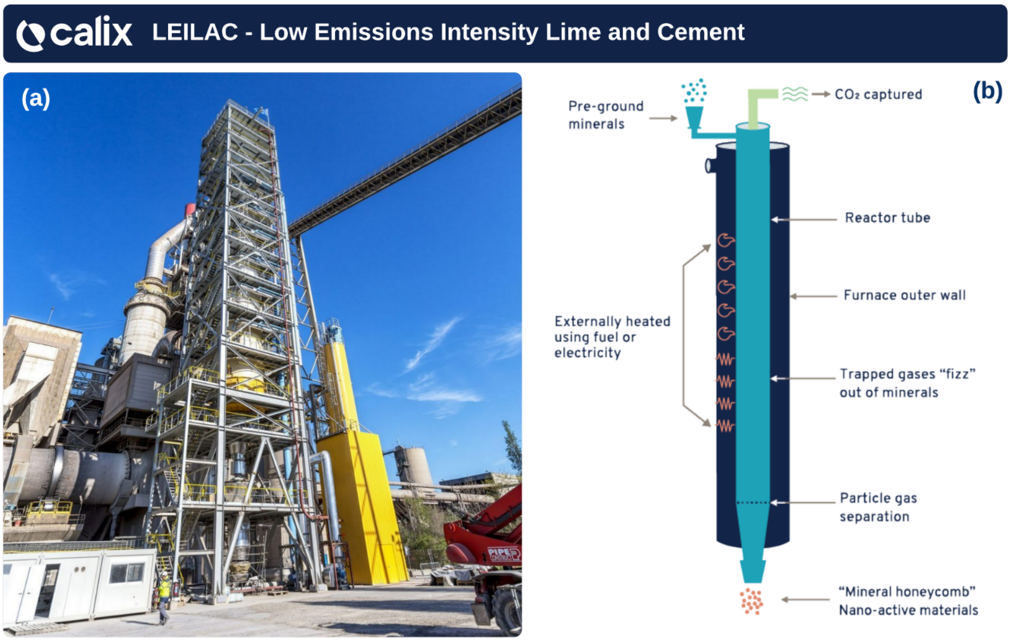

Figure 2.5. CALIX's LEILAC Technology. Installed direct separator reactor demonstrator (a) and scheme of the working principle of the technology (b) (adapted from https://calix.global/industries/industry-lime/).

While various kiln types are in operation globally, Calix's LEILAC Technology stands out for its demonstrated ability to capture CO_2 during the lime burning process. This technology is adaptable for retrofitting of existing plants and is currently technically available [28]. Governments and industries heavily reliant on lime production are increasingly recognizing the substantial environmental issues associated with traditional kilns. Consequently, they are introducing incentives to drive the adoption of cleaner manufacturing processes [22], [30], [31].

Securing the necessary funding and partnerships for the establishment of sustainable full-scale calciner plants for lime poses a challenge. Calix, however, has a commendable track record of securing government and industry funding for numerous pilot and demonstration plant . The success of these plants and their trials, coupled with the use of renewable electricity, indicates that lime manufacturing has potential to achieve near-zero emissions. This presents an appealing vision of the future for both operators and funders.

Unlike any other binder material used in the construction industry, lime production has the distinctive competitive advantage of generating pure CO_2 (Reaction 2.1). Nevertheless, an additional challenge involves ensuring that the captured CO_2 finds a suitable final destination. Effective management and utilization of this separated CO_2 becomes paramount, for which it is necessary to collaborate with partners across the CO_2 value chain to identify appropriate transport, storage, and/or utilization options. This collaborative effort is crucial for fully realizing the climate benefits offered by the direct separation technology.

Captured CO_2 can be either sequestered in storage sites (Carbon Capture and Storage, CCS) or, alternatively, utilized to produce value-added products (Carbon Capture and Use, CCU). CCS encounters various challenges, including high costs, limited financial incentives, and the difficulty of securing adequate storage space [32], [33]. Conversely, CCU, particularly when combined with renewable energy sources, holds immense potential as a component for a sustainable circular economy [34], [35]. Numerous projects are actively pursuing this direction (Figure 2.4). One of the most recent and cutting-edge endeavours is COLUMBUS (https://columbus-project.com/), which secured a substantial budget of 150 million Euros in 2022 to produce e-methane from hydrogen and CO_2. Progress, however, remains ongoing. In the EU ETS Innovation Fund's 2023 call, a staggering 1.4 billion euros have been allocated for eight decarbonization projects, with five of them specifically dedicated to CCU in the lime and cement industry [36]. This rapid development in the field of CCU underscores the emergence of various market alternatives to redirect the captured CO_2, providing it with a valuable destination as a by-product. This paradigm shift is actively supported by the EU in the framework of the Fit-for-55 package, establishing targeted incentives for CCU routes [30].

The lime industry finds itself at a pivotal juncture, offering a unique opportunity to play a decisive role in the sustainable transformation of multiple markets while concurrently decarbonizing its own production processes. By embracing CCU technologies and aligning with the European Union's sustainability goals, the lime industry has the potential to position itself as a frontrunner, contributing to both environmental stewardship and economic viability. A logical consequence of this decarbonization process is the enhanced sustainability of lime-based construction materials.

2.3 Lime-based Construction Materials

2.3.1Uses, Properties and Typical Composition

Calcium Oxide (CaO) and Calcium Hydroxide ($Ca(OH)_2$) serve as the main precursors for lime-based construction materials [37]. As mentioned before, they were extensively used in the past in old constructions. Therefore, they are fully compatible with ancient masonry, a key issue in conservation of heritage buildings and they fulfil the criteria for sustainable construction [38], because their chemical composition, transport properties, etc. are compatible with the majority of Europe’s built heritage [39], [40].

Buildings pre 1900 would not have been built with cement-based mortars but with mortars that have lime as a binder. Preferably, for the conservation of these structures it is critical to employ similar materials, because introducing a cement-based mortar/render would result in an irreversible damage of the historical building due to the difference in chemical and physical properties such as composition and water transport properties [14]. Therefore, hydraulic lime-based mortars, plasters and renders as well as lime-based putty are implemented in the process of restoring the majority of Europe’s built heritage. It is relevant to point out the importance of restoration, since it enhances the cultural value of buildings, because by attracting millions of tourists every year they represent an important source of income for various European countries.

Lime-based materials are also widely employed in modern constructions. According to the European Lime Association (EuLA), the construction and civil engineering sectors are major users of lime-based products, ranging from aerated concrete and asphalt to bedding mortars, renders and plasters [15]. Lime mortars have maintained extensive applicability in the European and worldwide scenario of masonry construction, both as joint material in between masonry units (bricks and blocks) and as a plastering (renders and plasters) material. When compared with cement, the benefits of using lime-based mortars/plasters include: low water penetration, increased breathability and moisture control, increased bond strength and reduced cracking (long-term "plastic" behaviour and crack healing due to dissolution-precipitation of $CaCO_3$), lower efflorescence, easier and cheaper building maintenance, workability and water retention resulting in optimum material use and productivity, unique aesthetics and texture, high compatibility with external insulation systems, excellent weather resistance, among others [41]–[44].

However, the compositions of modern renders and mortars incorporate -besides lime- cutting edge materials, specifically in terms of lightweight aggregates and additives to be used in diverse masonry applications. Typical compositions of lime-based mortars, renders and plasters are shown in Table 2.1. The unique microstructure provided by lime as a binder allows the masonry to accommodate to small movements in building structure and finishes and the porosity allows the building to breath [44]. However, the compositions of modern renders and mortars incorporate -besides lime- cutting edge materials, specifically in terms of lightweight aggregates and additives to be used in diverse masonry applications. Typical compositions of lime-based mortars, renders and plasters are shown in Table 2.1. The ready-mix material is easy to use and depending on the type of aggregates and additives can incorporate technologies such as fungi prevention, thermal isolation, fire resistance, self-cleaning properties and so on [13], [45]–[48].

Table 2.1. Components of typical lime-based mortar, render and plaster and ranges of variation. [49]

	Compound	Range (%wt.)	Comment
Aggregates	Aggregate	50-70	Natural sands, which contain natural minor and trace minerals along with the main minerals quartz (SiO_2) or calcite ($CaCO_3$)
	Fine aggregate	15-65	Limestone dusts which arise because of the preparation of natural sand to produce aggregates, as well as very fine sands.
	Mineral Lightweight aggregate	0-14	Natural or artificial inorganic lightweight aggregates to reduce the dry bulk density. Can be natural (pumice) or artificial (perlite).
	Polymer-based lightweight aggregate	0-1	Obtained by foaming manufactured organic, expanded polystyrene in spherical or particle form for reduction of the dry bulk density
Binders	Cement	0-22	As per EN 197-1 cement is used as a binder and is mainly made from limestone, marl or a mixture of limestone and clay.
	Hydrated Lime	5-20	As per EN 459 hydrated lime is used as binder and is made by treating at high temperature natural limestone, followed by a slaking process.
Additives*	Synthetic dispersion	<2.5	Polymer powder for improving the adhesive bond, elasticity, mechanical properties, etc. in thin-bed mortar.
	Water retention agent	<0.20	Cellulose ether, made from cellulose, which prevents dehydration from occurring in the fresh mortar too quickly.
	Air entraining admixture	<0.02	These reduce the bulk density of fresh mortar, improve workability, and reduce the tendency of contraction and stress cracking.
	Mineral pigments	<0.5	Mineral or synthetic powder-form colouring materials which are produced by mechanical processing of the relevant mineral substances such as chalk, clay, etc.
	Hydrophobic agent	<0.3	Water-soluble sodium oleate or zinc and calcium stearate for reducing the capillary absorption of the solid mortar.

**Ranges of concentration with respect to the total amount of binder*

2.3.2 Carbonation

Carbonation is a key feature of lime-based materials, specifically relevant in rendering/plastering applications. A scheme showing the carbonation of a lime-based coating is provided in Figure 2.6. This process takes place during the use phase of the material and can help to reduce the CO_2 emissions over its life-cycle through chemical bonding of CO_2.

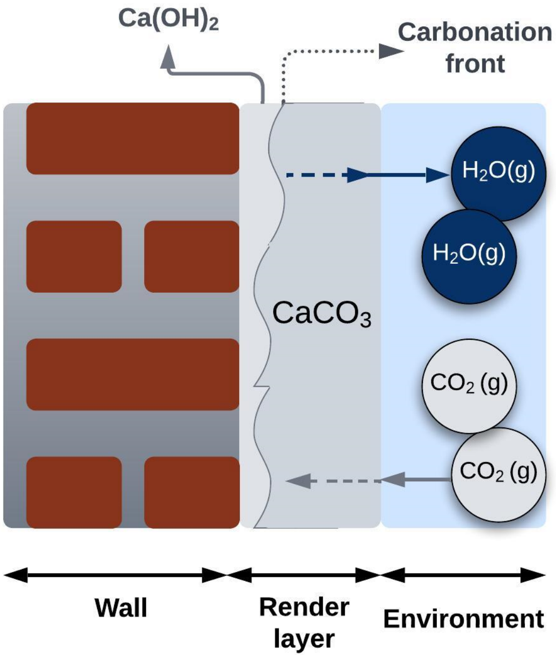

Figure 2.6. Carbonation of a lime-based coating in a masonry wall. The scheme is not on scale, the render layer has been enlarged for illustration purposes.

The carbonation process of $Ca(OH)_2$ has been extensively studied, as it is the main setting mechanism taking place spontaneously in these materials, because of the reaction with atmospheric CO_2 (Reaction 2.3) [50]. Reactions 2.1 to 2.3 (Pag. 20) are usually known as the *lime-cycle*. However, this overall reaction is the consequence of a complex mechanism of dissolution and precipitation of species, as shown in Reactions 2.4 to 2.8.

$$Ca(OH)_2\ (aq) + CO_2(g) \rightarrow CaCO_3(s) + \mathrm{H}_2O\ (aq)$$ **Reaction 2.3**

$$Ca(OH)_2\ (aq) \rightarrow Ca^{2+}\ (aq) + 2OH^-\ (aq)$$ **Reaction 2.4**

$$CO_2(g) \rightarrow CO_2(aq)$$ **Reaction 2.5**

$$CO_2(aq) + OH^-\ (\mathrm{aq}) \rightarrow HCO_3^-(aq)$$ **Reaction 2.6**

$$HCO_3^-(aq) + OH^-(\mathrm{aq}) \rightarrow CO_3^{2-}(\mathrm{aq}) + \mathrm{H}_2O(aq)$$ **Reaction 2.7**

$$Ca^{2+}(aq) + CO_3^{2-}(\mathrm{aq}) \rightarrow CaCO_3(s)$$ **Reaction 2.8**

The current knowledge in the area [51]–[53] indicates that the following mechanisms take place during the carbonation:

(i) dissolution of calcium hydroxide in the pore water with dissociation of Ca^{+2} and OH^- ions (Reaction 2.4).

(ii) dissolution of gaseous CO_2 into the alkaline pore solution to form a loosely hydrated aqueous form (Reaction 2.5).

(iii) hydration of CO_2 with OH^- ions to form carbonic acid (H_2CO_3) followed by its (nearly instantaneous) dissociation into bicarbonate (HCO_3^-) (Reaction 2.6) and carbonate (CO_3^{2-}) ions (Reaction 2.7) and, finally,

(iv) the reaction between Ca^{2+} and CO_3^{2-} ions forming a calcium carbonate precipitate through nucleation and subsequent growth resulting in an interconnected microstructure (Reaction 2.8).

From a practical application, when the coating is applied to the wall, it takes automatically contact with the atmospheric CO_2 and the carbonation reaction begins (Figure 2.6). The diffusion of CO_2 through the plaster (i.e., CO_2 sequestration) can be described by a simplified equation representing a diffusion-like process (Equation 2.1). Equation 2.2 can be employed to calculate the kg of CO_2 sequestrated per functional unit at a given time.

$$x = k\sqrt{t}$$ **Equation 2.1**

$$SC = 0.594 * FCH * \frac{x}{X_{total}}$$ **Equation 2.2**

where x (mm) is the carbonated thickness of the plaster at a given time t (days), k (mm/day$^{0.5}$) is the diffusion coefficient of CO_2, SC (kg CO_2/m^2) is the mass of CO_2 sequestrated per area of coated wall, 0.594 is a conversion factor (molecular weight ratio $CO_2/Ca(OH)_2$), FCH (kg $Ca(OH)_2/m^2$) is the amount of hydrated lime per area of coated wall, X_{total} (mm) is the total thickness of the plaster.

SC is calculated until the time of maximum carbonation (Eq. 2.1) is reached. The coefficient k can vary from 0.8 to 1.2 mm/d$^{0.5}$ depending on the mix composition [50], [54], [55]. In thick coatings (20 mm), a complete carbonation of the render/plaster can be expected in a moderate amount of time (1 year), even at atmospheric concentrations of CO_2 (421 ppm) [56].

The combination of decarbonizing the manufacturing process and harnessing the natural carbon sequestration of lime-based coatings holds the potential for achieving net-zero and even carbon-negative transformation throughout the Cradle-to-Grave life-cycle. However, it is crucial to rigorously quantify and compare the environmental impact of this solution against current manufacturing scenarios through the application of Life-cycle Assessment (LCA) methodology.

2.4 Environmental Impact Quantification

Life-cycle assessment (LCA) is a widely employed methodology that aids decision-making in selecting environmentally sound alternatives. Despite its widespread use, LCA is a relatively young and evolving standardized procedure, continuously adapted by the scientific community to address the dynamic complexities of current times. Numerous challenges, primarily associated with inventory data calculations, need to be overcome to rigorously quantify the environmental benefits of decarbonization strategies for lime-based construction materials from Cradle-to-Grave. This section introduces the general principles of LCA methodology and outlines the main challenges associated with environmental impact assessment in building materials.

2.4.1 Life-cycle Assessment

2.4.1.1 Generalities

LCA is a relatively young methodology from the field of environmental science. The first LCA concept was born and applied in 1969 under a study carried out by the Coca-Cola Company. Since then, it took around 30 years to develop a first standard procedure to carry out LCAs, by the International Standardization Organization (ISO). The methodology as such, was included in the ISO 14000 standard, related to environmental management. In 1997 the ISO 14040 published the first "Principles and Framework" standard. From 1997 until 2000, the ISO 14041, 14042 and 14043 were developed addressing the main characteristics of the methodology, from how to design the goal and scope of the study until how to evaluate and interpretate the results.

It was not until 2006 that the whole methodology was integrated in the ISO 14040-44 [57], [58]. At the moment of writing this thesis, end of 2023, the methodology has been fully standardized for 17 years. This is a relatively short period of time to enjoy full acceptance by the scientific community, and even more so by the private sector. As will be discussed more in detail below, the results are severely affected by the assumptions of the LCA practitioners, the quality of the data and the rigor of the inventory calculations. Furthermore, several standardized concepts are very general, which provides a significant flexibility to apply the methodology, but at the same time this freedom raises concerns about the significance of the results.

The current ISO14040 standard defines Life-cycle Assessment as *"the compilation and evaluation of inputs, outputs, and the potential environmental impacts of a product system throughout its life-cycle"* [57]. When carried out in a professional manner, the methodology can be a link between the scientific community, private sector and government and policy makers to design a more sustainable society (Figure 2.7).

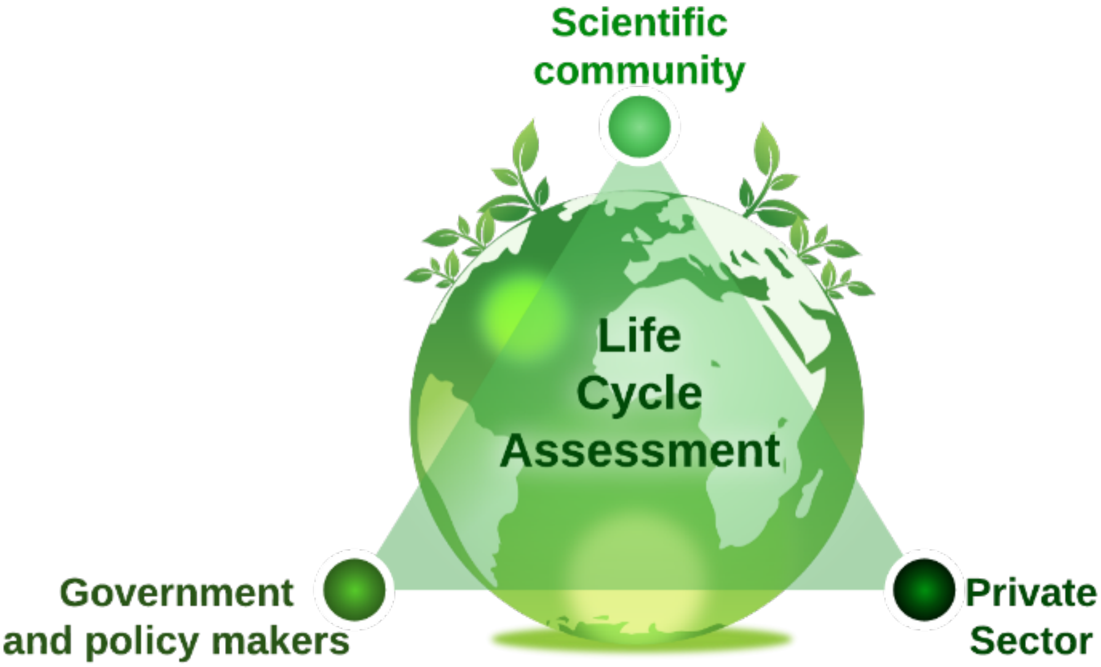

Figure 2.7. Life-cycle assessment as a link between the scientific community, the private sector and policy makers

As a primary objective, LCA has the function of supporting the decision process about which alternative of product or service is environmentally more performant. The methodology can provide reliable information for design and optimization of processes and products, assistance in several stages of the life-cycle of a product as well as to help efficiently communicating to a diverse public, as stated in Table 2.2.

2.4.1.1 Steps of the Life-cycle Assessment Methodology

There are four main steps to be performed under the standardized ISO 14040 Life-Cycle Assessment methodology:

(a) Goal and scope definition,
(b) Inventory analysis,
(c) Life-cycle impact analysis, and
(d) Interpretation of results

Figure 2.8 provides the general connection among the steps and specific clarification on each one is provided below.

Table 2.2. Main function and advantages of LCA methodology across different applications

Area	Specific function/advantage
Production plant	• Improve the efficiency of processes in the plant, from energy consumption and fuel emissions to virgin raw materials extraction minimization.
Design phase of the product	• Make decisions about which recipes would lead to a better environmental performance complying with an equivalent function. • Anticipate and avoid potential environmental impacts optimizing recipes by smart sustainable design. • Project in the future potential savings in environmental impacts related to life-stages besides the manufacturing. • Identify opportunities to improve the environmental performance of products at various points in their life cycle.
Certification/Environmental Product Declarations	• To calculate environmental profiles and communicate transparently the impact of production to the interested stakeholders. This is especially important due to o the context of product certification according to European Regulations o increasing interest on building design integrating not only technical aspects but also environmental ones
Communication	• Informing decision-makers in industry, government, or non-government organizations for the purpose of strategic planning, priority setting, product or process design or redesign.
Marketing	• Implementing an ecolabelling scheme, making an environmental claim, or producing an environmental product declaration.
Economic sustainability	• European industries -specifically mining and mineral processing industries- must pay carbon taxes because of the CO_2 emissions. Improving environmental sustainability will lead to economic sustainability as well.

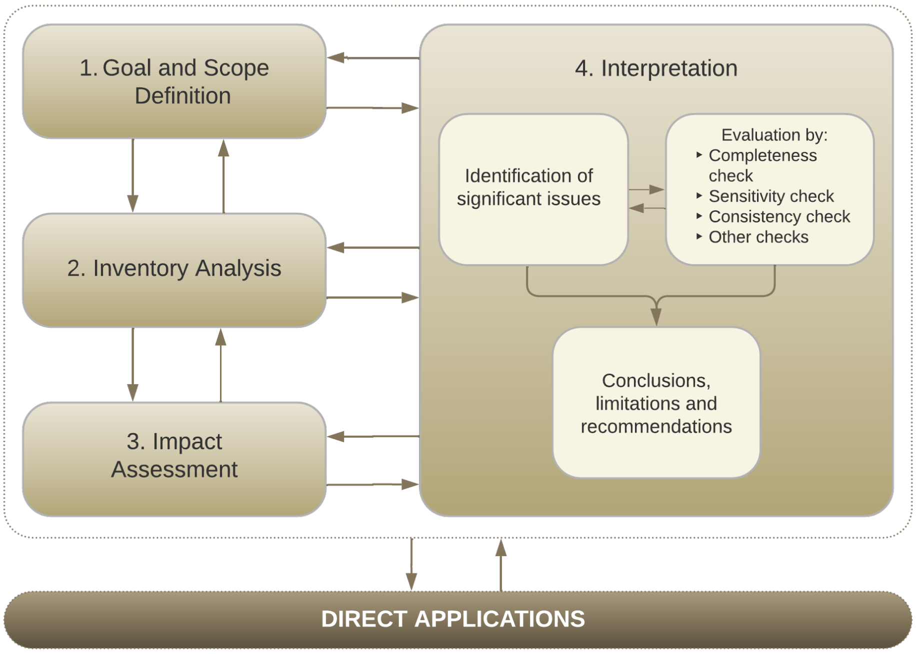

Figure 2.8. Steps of the LCA methodology according to ISO 14044:2006 Standard [57]

(a) Goal and scope definition

The goal and scope definition are the first phase of any LCA and decisive for all subsequent steps of the methodology. The goal definition guides all the detailed aspects of the scope definition, which in turn sets the frame for the life-cycle inventory (LCI) calculation and life-cycle impact assessment (LCIA).

During this stage the system boundaries of the study (Scope) are determined, which are a set of criteria specifying which unit processes are part of a product system. Two definitions are of relevance:

- A process is a set of interrelated or interacting activities that transforms inputs into outputs and the Unit Process is the smallest element considered in the life cycle inventory analysis for which input, and output data are quantified.
- A product system is a collection of unit processes with elementary and product flows, performing one or more defined functions, and which models the life cycle of a product.

The system's function and functional unit (FU) are central elements of an LCA. An LCA is always anchored in a precise, quantitative description of the function(s) provided by the analysed system. This is generally done by using the FU that names and quantifies the qualitative and quantitative aspects of the function(s) along the questions "what", "how much", "how well" and "for how long". In summary, the FU represents a quantified performance of a product system for use as a reference unit [59].

The impact assessment method is also defined during this phase, in concordance with the goal and scope of the study. The impact method selected depends on the type of assessment that the LCA is targeting. For instance, Impact 2002+ is an impact assessment method that addresses damage categories relevant for the mining industry such as Resources, Ecosystem quality, Climate Change and Human Health.

(b) Inventory Analysis

The inventory analysis comprises the collection of relevant input and output data included but not limited to use of energy and raw materials, emissions to air, water and soil, production of waste, etc. at each analysed stage of the life-cycle, as defined in the scope of the study.

The ISO Standard 14044 establishes that the data should address: time-related coverage, technology coverage, precision, completeness, representativeness, consistency, reproducibility and (un)certainty of the information [58]. Two types of data must be differentiated: foreground data calculated by the LCA performer and background data which include energy and materials that are delivered to the foreground system as aggregated data sets in which individual plants and operations are not identified [60].

Foreground data are usually available or can be calculated for the product that is being analysed. For example, a specific recipe for the production of a lime-based render composed by 25% hydrated lime, 74% lightweight aggregate and 1% additive. The production of hydrated lime in the specific inventory (background data) is usually modelled using data from Environmental Product Declarations, Environmental Reports, and literature (i.e., papers and theses). While a certain level of truthfulness is missed, generic databases are a powerful source for process modelling, among which EcoInvent [61] and Gabi [62] are considered the most complete available for the construction sector [63]. However, they present several disadvantages regarding data manipulation and lack of clear understanding of included activities, among others.

A significant challenge during the inventory calculation is the identification of multifunctional processes, which produce more than one function, and this requires a fair treatment of the data to assign in a fair way the environmental impact to the analysed functional unit. An example is lime manufacturing, because several products can be generated along with hydrated lime (i.e., milk of lime, lime putty, etc.). The ISO standard 14044 indicates as a first measure to avoid allocation by sub-dividing the system, although it is not often the case [58]. The second alternative should be system expansion/substitution, in which unit processes are mathematically treated to include the multifunctionality in the FU. The last, but most widely employed alternative is the allocation procedure, in which through a specific criterion the environmental impact is assigned to each product.

(c) Life-cycle Impact Assessment

Life-cycle Impact Assessment (LCIA) is the phase in an LCA where the inputs and outputs of elementary flows that have been collected and reported in the inventory are translated into impact indicator results related to human health, natural environment, and resource depletion.

Three main steps take place during the environmental impact calculation: classification, characterization, and aggregation. In the classification stage, the individual elementary flows from the inventory are classified according to the relevant impact category that they contribute to. Characterisation refers to the calculation of the magnitude of the contribution of each classified input and output to their respective impact categories. Furthermore, in the aggregation step, the contributions of the characterized elementary flows are aggregated within each impact category. This is done by multiplying the elementary flow values by the relevant characterisation factor in each impact category considered. The characterisation factors are substance- or resource-specific. They represent the impact intensity of a substance relative to a common reference substance for an impact category (and they are used to calculate the relative impact category indicator(s)) [58], [59]. For instance, the emission of sulfur hexafluoride has a characterization factor of 32400 kg CO_2/kg according to Impact2002+ [64] (See Section 1.1 for clarification). Table 2.3 provides an overview of the midpoint indicators and damage categories included in the Impact 2002+ impact method utilized in this thesis.

It can be observed in Table 2.3 that the Impact2002+ method proposes an implementation of a combined midpoint/damage approach, linking all types of life-cycle inventory results via several midpoint impact categories to several damage categories. In this sense, it is located in between two typical main schools of methods: problem oriented and damage oriented [59]. The first ones are focused on the impact at early stages in the cause-effect chain, such as the emission of a certain amount of CO_2, having an impact on global warming (increase of temperature). The second group focuses on the actual environmental damage of releasing a certain amount of CO_2 (i.e., climate change). For a differentiation between *global warming* and *climate change* concepts the reader is referred to Section 2.1. The selection of a relevant midpoint indicator or damage category to be communicated depends on the goal and scope definition of the study.

Table 2.3. Overview of the main midpoint indicators and damage categories in the Impact 2002+ impact method [64]. *DALY: Disability-Adjusted Life Years; PDF: Potentially Disappeared Fraction of Species; PM: Particulate Matter; -eq: equivalents; y: years*

Midpoint indicator	Midpoint reference substance	Damage category	Damage unit
Human toxicity (carcinogens + non-carcinogens)	kg Chloroethylene into air$_{-eq}$	Human health	DALY
Respiratory (inorganics)	kg $PM_{2.5}$ into air$_{-eq}$		
Ionic radiations	Bq Carbon-14 into air$_{-eq}$		
Ozone layer depletion	kg CFC-11 into air$_{-eq}$		
Photochemical oxidation [= Respiratory (organics) for human health]	kg Ethylene into air$_{-eq}$	Human Health/ Ecosystem quality	DALY/ n/a
Aquatic ecotoxicity	kg Triethyleneglycol into water$_{-eq}$	Ecosystem Quality	PDF*m^2*y
Terrestrial ecotoxicity	kg Triethyleneglycol into soil$_{-eq}$		
Terrestrial acidification/nitrification	kg SO_2 into air$_{-eq}$		
Aquatic acidification	kg SO_2 into air$_{-eq}$		
Aquatic eutrophication	kg PO_4^{3-} into water$_{-eq}$		
Land occupation	m^2 Organic arable land$_{-eq}$*y		
Water turbined	Inventory in m^3		
Global warming	kg CO_2 into air$_{-eq}$	Climate change (life support system)	kg CO_2 into air$_{-eq}$
Non-renewable energy	MJ or kg Crude oil$_{-eq}$ (860kg/m^3)	Resources	MJ
Mineral extraction	MJ or kg Iron$_{-eq}$ (in ore)		

(d) Interpretation

As mentioned previously, the main goal of an LCA is to support decision-making processes. In this context, the interpretation phase allows to evaluate the quality and robustness of the obtained results, identify critical processes and inventories that need to be refined, in relation to the FU defined for the study. Some of the elements of the interpretation (namely completeness and sensitivity analysis, as well as potentially uncertainty analysis for the determination of precision) are hence also applied throughout the LCI/LCA study (See Figure 2.8).

This phase has two main objectives. First, during the iterative steps of the LCA (see below), the interpretation phase serves to steer the work towards improving the LCI modelling to meet the needs derived from the goal and scope of the study. Secondly, it provides the opportunity to derive conclusions and recommendations. Conducting an LCI/LCA study is often an iterative process. After defining the goal of the work, initial scope settings are established, outlining the requirements for the subsequent phases.

However, as more information becomes available during the life cycle inventory phase, and later during impact assessment and interpretation, the initial scope settings may need refinement and, in some cases, revision, as illustrated in Figure 2.9. The interpretation phase is a crucial step that allows for the necessary modifications to enhance the quality of the results, facilitating the decision-making process. This iterative nature ensures that the study remains responsive to emerging data and insights throughout its progression [59].

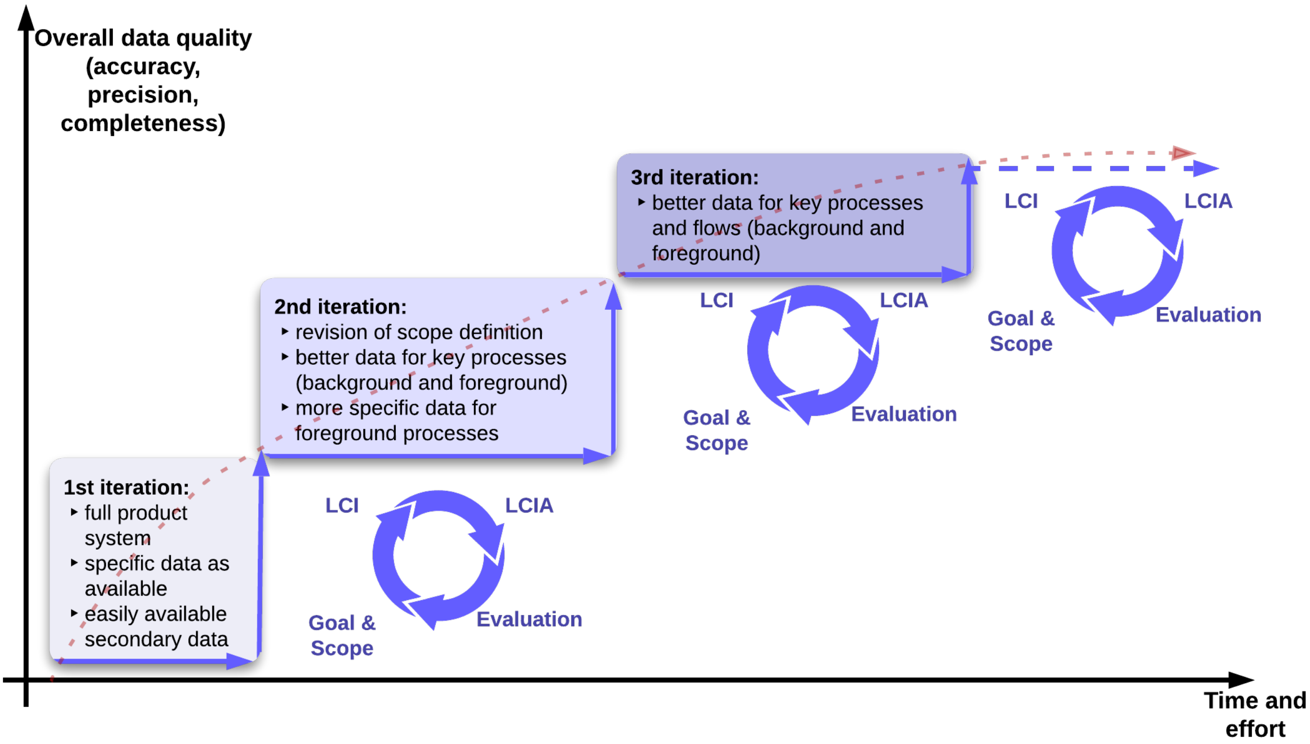

Figure 2.9. Schematic iterative nature of LCA, improving the overall data quality as the goal and scope and LCI/LCIA are evaluated, interpreted, and refined. Adapted from [59]

The preceding paragraphs provide a concise overview of the LCA, a versatile methodology applicable for calculating and comparing the environmental impact of diverse processes, products, and services. This thesis focuses on the environmental impact of building materials, particularly those based on lime. The next section will delve into specific considerations and research needs in this domain, addressing particularities associated with lime-based building materials.

2.4.2 Environmental Impact in Building Materials

The application of the LCA requires not only a solid understanding of the methodology itself but also of the context of application. In this way, the background knowledge required to properly assess the environmental impact of a certain fertilizer is not the same as the one required for a masonry unit. Each area of knowledge or field of application has its own particularities, and a proper LCI/LCA modelling requires a sufficient understanding of them, not only to make fair comparisons but to identify critical points in the LCI model. The next paragraphs address these issues in the specific context of building materials.

2.4.2.1 Background on LCA of Building Materials

A building material is any material that can be used for construction purposes, from wood to steel, cement, (hydrated) lime, bricks, sand, etc. These materials enable the creation of infrastructure necessary for economic development and well-being of people.

Although many building materials are naturally obtained (e.g., river sand) it is (almost) always necessary that they go through a certain industrial process before they can be used. Considering the key role that they play in society; it is not surprising that the construction sector is a relevant contributor to the current environmental crisis. Particularly, cement is the largest manufactured product on Earth by mass and the second most used substance in the world after water [9]. Projections even show that cement production must grow in the next 20 years to meet the demand for a decent built environment from citizens in developing, low-income countries.

Such context has driven scientific innovation in materials technology for the development of ‘green’ cement and concrete, centred on the replacement of clinker with other materials. Basically, any amorphous or imperfectly crystalline material containing silica, alumina and/or lime can be a potentially reactive supplementary cementitious material (SCM) [65]. For instance, granulated blast furnace slag from pig-iron production was used in cement before 1900. Fly ash resulting from electricity production by burning coal is also a well-known SCM . One of the most promising technologies nowadays is the so-called Limestone Calcined Clay Cement (LC3) [9]. In general, there is a robust knowledge about basic properties such as hydration, hardening and durability against aggressive environments in concrete manufactured with SCM [66].

Despite the long journey on concrete technology development, an objective quantification of the environmental impact of such strategies is an issue that remains open in the scientific community. To solve this issue, a new generation of scientists need to be the bridge between building materials technology and sustainability assessment. As the field consolidates, every year new research shows positive advances towards reliable calculations. In the construction industry, LCA can be performed under different system boundaries, as will be discussed in the next paragraph. A significant portion of research articles focus on the environmental impact at a manufacturing stage, of producing cements with a variety of SCMs (e.g., waste glass, fly ash, blast furnace slag), different types of aggregates and additives among many others [67]–[72]. A critical issue is the selection of the FU, which in many cases is based on the unit mass or volume of a product. This is acceptable if the comparison is between production processes of the same material. However, the FU should capture relevant performance design properties (compressive strength, durability, etc.) of the compared alternatives to fulfil an equivalent function. An alternative proposed by Van den Heede et al. is the amount of concrete needed in a simple structural element with a given mechanical load and a predefined service life in a given environment [63].

A further critical aspect is the selection of the scope (system boundaries) of the LCA. At a building scale, the system boundaries are well defined in the CEN TC350 standards [73]. Although they cannot be directly applied when the analysis is carried out at a materials level (or small-scale application), it provides guidance on relevant life-stages to consider. Figure 2.10 shows the life-cycle stages according to CEN TC350.

Modules A1 to A3 are usually called Cradle-to-Gate system boundaries. It includes all processes from the extraction of raw materials for the production of all buildings materials that are used in the foundations and structure of the building, until the gate of the factory. Data for these modules are usually provided from the Bill of Materials (BoM) of the building. At material scale, mix design compositions or production processes can be evaluated under these boundaries [49], [74]–[76]. The allocation of impacts in any necessary case (such as when using SCMs) is critical and can significantly influence the results [77]. Eventually, modules A4 and A5 can be integrated in an extended Cradle-to-Gate stage to consider transportations and machinery used on the application site.

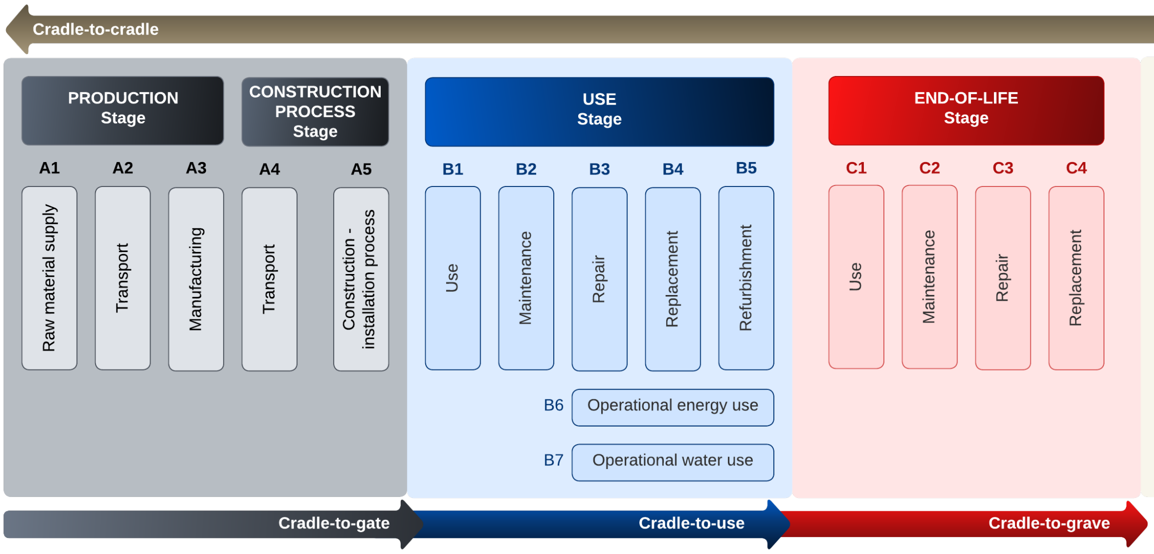

Figure 2.10. System boundaries at a building scale according to the CEN TC350 standard. Adapted from [73]

The next life-stage is the use phase, accounted for in the modules B1-B5. These modules include relevant data regarding the maintenance, repair, and refurbishment of the building. In connection with the material level, relevant design properties to be achieved during the service life should be accounted for.

These properties will depend on the application of the building material, such as specific resistance or exposure class. In this way, the need for maintenance can be quantified at materials level [63], [78]. When the LCA is performed at a building level, it can be coupled through developed algorithms with operational energy simulation to consider module B6 in the environmental impact assessment [79].

When the end of life is achieved, the last life-cycle stage is called 'grave' and if the system boundaries start from the cradle, the resulting scope is from Cradle-to-Grave. Modules C1 to C4 include relevant data from the decommissioning of the structural system of the building until the final disposition of the resulting materials [80], [81]. However, nowadays it is desirable to recirculate materials at the end of life to act as a raw meal for new materials or to be used in diverse applications. This system boundary is called Cradle-to-Cradle (Module D). At a material level, concrete can be designed to be completely recycled, improving the overall sustainability [82], [83]. Further applications also include the production of recycled aggregate from construction and demolition waste [72], [84], [85].

2.4.2.2 Lime-based Building Materials: Stage of Knowledge

It becomes evident that with the preponderance of cement-based materials in the modern world, the development of lime-based materials, traditionally used in the old constructions did not get the same level of engagement from the scientific community. If the consolidation of the scientific knowledge on concrete sustainability is still in process, in lime-based materials there is still a long way to go. And this knowledge is necessary because as discussed in Section 2.3, these are relevant materials in modern masonry structures.

Little is known about a sustainable assessment of lime-based materials. This lack of knowledge extends from the Cradle-to-Gate of the factory until the use phase. The environmental impact assessment of cement production has been thoroughly analysed in the research literature [86]–[88], which is not the case for hydrated lime manufacturing. This is a critical challenge to improve the sustainability in the sector, mostly if new production systems with different kiln technologies, use of alternative energy sources, carbon capture systems, etc. need to be benchmarked against current manufacturing scenarios (See section 2.2). This is key to understand up to what point embodied emissions (avoidable and unavoidable) from binder manufacturing can be reduced (Figure 2.10, Product stage).

Furthermore, only selected research articles describe a shallow LCA analysis focusing on the production of lime-based mortars, renders and plasters ([37], [89]–[91]). However, a comprehensive Cradle-to-Gate LCA of basic compositions in lime-based materials has not yet been thoroughly researched. This assessment should consider factors such as mix designs, the type of coarse and lightweight aggregates, additives, and so forth. Each component can have a distinct impact on the overall environmental and economic performance of the material, and it is essential to understand the extent of their contributions. This information is required to identify potential improvement scenarios, such as selecting one component over another, determining ranges of variation in indicators, and assigning proportions of the total impact to each component. In summary, there is no clear baseline scenario for this specific type of material, hindering the projection of sustainability improvements.

In section 2.3.2 it was shown that a main mechanism for the hardening of lime-based materials is the carbonation process. During this phase, the natural carbonation of portlandite occurs, leading to the sequestration of carbon dioxide and the production of calcium carbonate. These carbonation products serve as the main binding agent, holding all the components in the matrix together. Theoretically, 0.59 kg CO_2/kg $Ca(OH)_2$ can be sequestrated during the material's use phase. The natural carbon sink provided by lime-based mortars, renders and plasters should be considered in the environmental impact assessment since carbonation is an inherent and necessary reaction for the material to fulfil its intended function. This is a distinguishing phenomenon that must be included in the LCI/LCA modelling. Still, most of the existing research focuses primarily on the production stage [54], [55], [92]–[94], neglecting the crucial importance of the use phase.

2.4.2.3 Lime-based Building Materials: Research Needs

The challenges to improve the sustainability in the lime sector can be distinguished in two different categories: *data availability* and *LCA methodology*.

With respect to the data availability, the lack of environmental impact studies in lime-based materials was already discussed. The background data used in these calculations are always modelled by generic databases such as EcoInvent. During the literature review it was revealed that first-hand data have rarely been implemented during the calculation of the LCI [37], [95] and when using models to complement the lack of data, calculations are not consistently described. As a result, there is no thorough assessment of the Cradle-to-Gate manufacturing and, in consequence, there is a lack of baseline scenario from which improvements can be made. In terms of carbon capture and sequestration, given that it is a relatively new concept, the topic is even more unexplored. A very complete and recent state-of-art research article (and one of the very few) on decarbonization of the lime industry describes current and in-development processes and carbon capture technologies but does not address how to take them into account for the LCA calculation [96].

The second challenge is related to LCA methodology and two main points are to be addressed: the calculation of the LCI and the extended uses of LCA. The results of the impact assessment are strongly dependent on the quality of the input data and therefore, the way in which they are obtained and modelled in the LCI. A very common practice has been to model production systems, using background data provided from generic databases such as EcoInvent [61] and Gabi [62]. In many cases these datasets do not clearly describe the way in which they were built, how the data were collected and what are the assumptions (providers of electricity, transport distances of materials, efficiency of the devices, etc.). Such a range of uncertainties can evidently have a consequence on the results of the LCA and an alternative should be found to minimize the impact of the background data.

Besides the uncertainty in the assumptions, there is a problem of manipulation of the datasets, because it is not always possible to modify the processes included in them to evaluate for instance, changes in the system boundaries, unit operations, efficiency of the devices, use of alternative energy sources, etc. Therefore, the researcher or LCA practitioner do not have full control of the study and they are subjected to background data limitations. A less traditional approach to build the LCI model of the system process would be to design and calculate, at a certain level of detail, the plant that will deal with the product of interest. This is not always easy to do, nor recommended to all LCA practitioners, because it requires an integral background of transport phenomena (mass, energy, and momentum) as well as the process engineering and materials science criteria to select the adequate devices to model the process accurately.

Last, it is worth mentioning the extended uses of LCA. Since it was first born in 1969 until it has been formally instrumented as a methodology under the ISO Standard 14040/44, its application has changed a great deal and the products under assessment are every day more complex. Because of the already explained crisis, residues must be recirculated, other technologies need to be developed, new by-products can be the outcome of changes to more sustainable recipes, etc. and the ISO Standard 14040/44 does not establish clear guidelines on how to deal with them. In the case of building materials, and those with binding properties, this is even more complex, since because of different chemical reactions and physical phenomena, they change over time. On the same level of importance, accounting for credits in LCI is a pending topic in the standard (and very briefly mentioned). When it comes to considering credits from carbon capture or from the use of supplementary raw materials, there is a lack of clear guidelines and agreement on how to calculate and include them in the LCI.

All in all, the available literature in sustainability assessment of lime-based materials suggests that the topic is highly unexplored, which makes it very difficult for all stakeholders to make well informed scientific-based decisions to improve both, production systems and materials design.

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Chapter 3: Research Methodology

This section proposes a methodology for the parametric calculation of Cradle-to-Gate life-cycle inventories based on process engineering concepts, mass and energy balances, materials science, and environmental engineering.

The methodology is described in the form of a flow diagram and is illustrated in simple case-studies to show how to include in the inventory the decarbonization strategies proposed in the thesis (effect of energy sources, new kiln technologies, carbon capture systems). The calculation of the parametric inventories is used throughout the Chapters 4 to 7.

Publication 1. Decarbonizing the Lime-Based Construction Materials Industry: A Practical Guide for Cradle-to-Gate Life-cycle Inventory

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3.1 Introduction

Life Cycle Assessment methodology [1] has been applied to cement-based materials to calculate the environmental consequences of manufacturing different types of concrete with a wide variety of supplementary binders and/or applying different types of aggregates and additives [2]–[9]. However, studies related to the environmental impact of lime-based materials are less developed. During the last six decades the amount of produced cement has increased about 30 times, while the world's population grew only by a factor 3 [10]–[12]. Consequently, many resources have been invested mostly in designing more eco-efficient concretes rather than other construction materials. However, lime is widely utilized in new constructions, from concrete blocks to plasters and mortars, as well as in the preservation of cultural heritage [13].

The production of quicklime (CaO), the main precursor of these materials, in 2020 amounted to 20 Mt/y, with the construction sector representing 19% of the market [14] [15]. Its production emits 1.2 t CO_2/t of CaO, meaning that in Europe in one single year 4.6 Mt of CO_2 are generated only because of CaO production [16]. These emissions contribute to around 1% of the global anthropogenic emissions and have two main sources or origin: chemical decomposition of limestone (approx. 65% of the total) and other process emissions [17]–[19]. The European regulations require industries to adapt their production systems and technologies to achieve net-zero emissions by 2050 in response to the environmental crisis. The urgency to act is increasing due to the European Trading System's carbon price of 90 € per metric ton of carbon (2022) for emissions generated by industries, which has increased by 9 times from 2018, making the economic motivation as important as the environmental one [20].

Rather than being a problem, the decarbonization of the lime-based construction materials industry can be an opportunity that provides the sector with a competitive advantage. There are five main points that are essential to achieving this goal:

(1) Power the manufacturing process with eco-efficient energy sources (such as biomass or electricity).
(2) Implement the concept of circular economy to minimise raw materials extraction.
(3) Change current technologies by using more efficient kilns with direct emission separation technologies.
(4) Implement carbon capture technologies.
(5) Combine point 3 and 4 to transform CO_2 from an emission into a co-product.

Last point is critical, because it can lead to minimize emissions per ton of produced material, along with a potential added revenue from the reduced cost of emissions and from selling CO_2 as by-product to other industries. By implementing these integral and interactive strategies, combined with the natural effect of carbonation during the use phase, it is possible to improve the overall sustainability performance of lime-based materials, while reaching even a carbon negative scenario.

To illustrate potential scenarios, in Figure 3.1 a scheme is provided showing as an example the carbon profile of a lime-based render over its entire life cycle. In both cases, a binder consisting of a mix of hydrated lime and cement is used, along with aggregates and additives to manufacture a dry rendered powder. In the “business-as-usual” scenario, hydrated lime is produced in a traditional way, using limestone as the main raw material, and solid fossils as the main fuel in the lime kiln representing an energy supply with a limited amount of renewable sources. Over the life cycle, the material naturally absorbs CO_2 from the environment in a process known as carbonation [21]–[23]. Even though this use phase could be considered as a carbon sink, the room for improvement is rather limited at this stage and relying on this carbonation phase to achieve carbon neutrality is far from realistic.

The manufacturing stage has such high levels of emissions that even if the material could fully carbonate during its life-cycle, it would be unable to offset all Cradle-to-Gate emissions. However, in a scenario of enhanced sustainability, by-products and/or waste materials are utilized as raw materials whilst supplying the same functionality to a mix as hydrated lime. Eco-efficient energy sources are utilized to power the devices and carbon capture technologies are implemented in such a way that not only CO_2 is captured but also employed to prepare a co-product that can substitute part of the energy requirements of the limekiln. In this case, the same rate of carbonation is considered as for the business-as-usual scenario, while carbon neutrality and even carbon negative are more plausible.

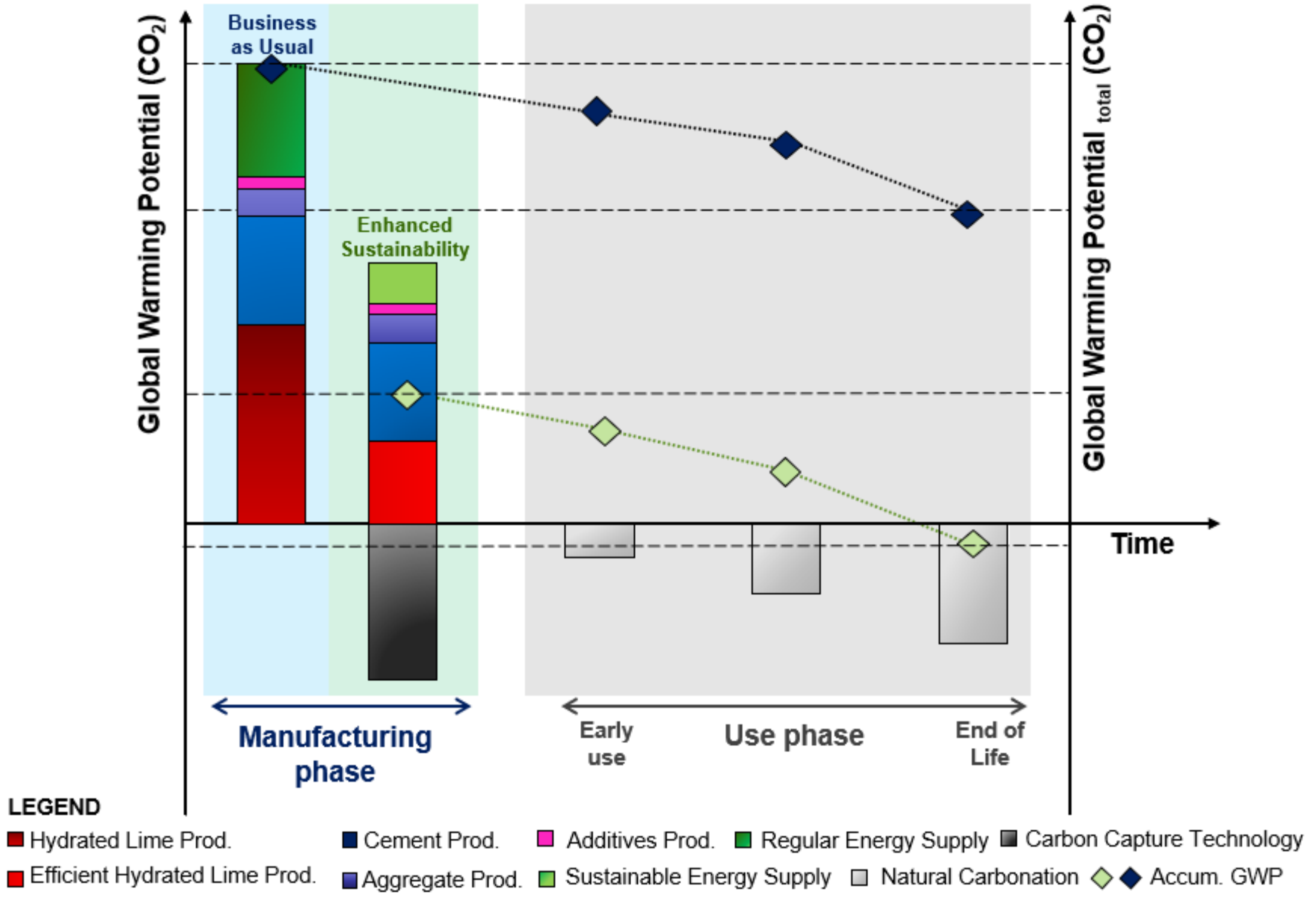


Figure 3.1. Carbon profile of a lime-based render over its life cycle under a business-as-usual and an enhanced sustainability scenario.

For a more sustainable scenario to take place, a change in mindset is required and accurate information to support decision making is mandatory. One of the main problems to face is the lack of environmental impact studies in this area, because so far only a few research articles have addressed the environmental profiles through LCA ([13], [24]–[27]). These studies are mainly focused on the effect of mix designs on the environmental impact through generic databases while no assessment of materials manufacturing is done. A rather complete research article (and one of the very few) on decarbonization of the lime industry describes processes and carbon capture technologies but does not address the opportunities of circular economy for LBM and the particularities on how to evaluate them through life cycle assessment [28]. All in all, the limited research on sustainability assessment of lime-based materials indicates a significant knowledge gap, posing challenges for stakeholders in making informed decisions that enhance production systems and materials design based on scientific evidence.

The assessment of decarbonization strategies in the lime-based construction materials industry requires a reliable transparent, robust, and flexible life cycle inventory (LCI). While parametrized process-based life cycle inventory has been used in evaluating scenarios at the building scale (e.g., operational energy and environmental flows), there is a need for further development of an LCI design specifically tailored to the challenges of producing sustainable construction materials [29]–[31]. Currently, the energy use in kiln technology and energy sources contributes up to 35% of CO_2 emissions in lime production, making it crucial to explore alternatives for significant reductions [16]. However, limitations arise from the inability to modify existing datasets in commercial databases used for modelling building materials production, hampering the implementation of different scenarios. Additionally, while carbon capture technologies hold promises for emission reduction in the industry, the lack of depth in inventory calculation and insufficient framework support [29], [32]–[34], raise concerns about inventory quality and potential missed opportunities for additional credits achievable through a parametrized inventory approach.

In building materials manufacturing, the scarcity of resources due to extensive raw material consumption is a critical issue. Circular economy emerges as an alternative to reduce resource extraction and waste landfilling. Nevertheless, the environmental impact of using waste-based materials as replacements for cement/lime needs careful evaluation. Currently, it is common practice to overlook the environmental impact of their production or omit reporting the intermediate treatments required for their use as supplementary materials [35]–[38]. Properly assessing their contribution in the manufacturing phase requires a comprehensive inventory considering intermediate treatments, integrating material physical and chemical properties, materials flows and energy requirements. Moreover, in the dynamic context of advancing technologies and energy sources, there are multiple alternatives to analyse for enhanced sustainability. Implementing these alternatives while ensuring robust and transparent assessment of scenarios can be facilitated by a parametrized inventory approach.

This paper presents an integral parametrized framework to calculate the life-cycle inventory (LCI) in lime-based construction materials manufacturing, utilized for implementing decarbonization strategies in the LCI. Three decarbonization strategies are studied: carbon direct avoidance, carbon capture and circular economy. The application of this methodology is demonstrated through three case studies. First, the inventory calculation of a single unit process in hydrated lime manufacturing is presented, allowing for a parametric study on different kiln technologies and energy sources. Second, the inventory calculation includes a CO_2 capture system with energy recovery in the lime kiln. Lastly, the modelling of the intermediate treatment of paper mill sludge as supplementary lime material in hydrated lime manufacturing is illustrated, along with an equation to determine the maximum substitution amount.

3.2 Methodology

3.2.1 Main Framework of the Life-cycle Assessment Methodology Applied to Building Materials

According to the ISO Standard 14044:2006 [39], there are four main steps to be considered in LCA: goal and scope definition, inventory analysis, life-cycle impact analysis and interpretation of the results [1], [39]. This paper is specifically focused on the life-cycle inventory modelling of the Cradle-to-Gate production, in which all the emissions as a result of the materials transformation to obtain the products take place. However, when aiming at decarbonization in the context of lime-based materials it is relevant to see how all stages of the life cycle of a material can influence the overall environmental performance. The different phases are well established in the CEN TC350 standards for the case of a complete life cycle of buildings [40], while not all of them are directly applicable when the scope is to analyse a single material or a combination of them (Figure 3.2). As shown in Figure 3.2a, the system boundaries are organized in 4 modules: (A) Product Stage and Construction Process; (B) Use Stage; (C) End of life Stage; and (D) Benefits beyond the system boundaries.

A modified proposal covering the scope of the CEN TC350 is presented in Figure 3.2b, to specifically consider in the life cycle the potential impact of carbon capture technologies on the manufacturing stage (A4) and the carbon sequestration during the use phase (B1) [41]. With regards to the use stage, carbon sequestration (B1) acquires a high relevance in lime-based materials, in contrast to what happens in cement-based materials. The main benefits of lime-based materials are that the carbonation proceeds in general faster than in cement-based materials and it is not unwanted phenomena like in concrete, which can lead to the corrosion of the reinforcement, increasing the need for maintenance (B2). In Module C, C1 is mainly focussing on the deconstruction approach rather than the traditional demolition. Finally, aiming at highlighting the importance of the circular economy, Module D is relocated at the bottom of the figure showing how each stage of the life cycle can interact with this module.

In this paper we propose a methodology to calculate a parametric life cycle inventory to be applied in the production stage A of building materials (Figure 3.2b). During this phase it is necessary to determine the energy requirements (i.e., to power a device), material inputs (raw materials and ancillary inputs as well as other physical inputs) and material outputs (products, co-products, and waste; and emissions to air, water, and soil) normalized per functional unit [1].

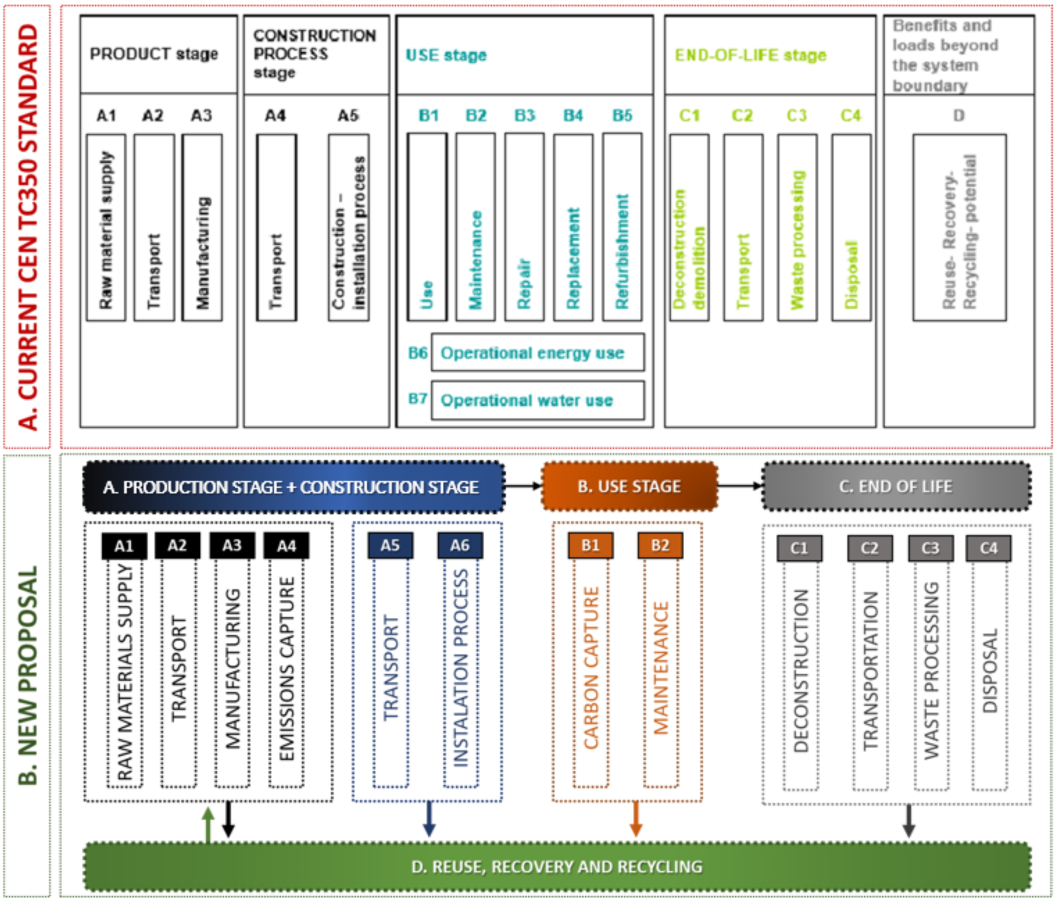


Figure 3.2. Scope of the LCA of buildings according to CEN TC350 standards [40] and modified scope of the LCA in lime-based materials.

Inventory modelling is crucial in LCA as it affects the accuracy of impact calculations. To obtain reliable results on the impact of decarbonization strategies for the lime industry, rigorous calculation of the inventory is essential. With regards to the data, a distinction between foreground data and background data should be made. The first are data from a foreground system of primary concern to the analyst. It can be collected, measured, calculated, or estimated to quantify inputs and outputs of a unit process (i.e., the specific electricity consumption of a device used in a unit process).

Background data in LCA consist of aggregated energy and materials inputs to the foreground system, without specifying individual plants or operations. Generic databases like EcoInvent [42] and Gabi are widely used for process modelling in the construction materials sector [7] due to their comprehensiveness. However, many datasets are unavailable, nonmodifiable or lack transparency, making it challenging to identify the specific unit processes and estimation sources. As a result, environmental scientists and LCA practitioners often have to rely on the provided data without the ability to make modifications or fully understand its origins.

Applying a process-oriented parametric methodology for life cycle inventory modelling can enhance transparency and results reliability. This approach, outlined in ISO Standard 14044 [1], involves calculating inputs and outputs through mass and energy balances, validating, and refining them with first-hand data. However, it is not widely used in the LCA community due to the need for process engineering expertise to ensure coherence in decision-making and calculations. The subsequent sections will provide a concise overview of the fundamental steps for constructing an inventory based on process calculations.

3.2.2 Process-oriented Methodology to Calculate the Life-cycle Inventory

The procedure of process-oriented design of LCI is shown in Figure 3.3. The main parts of the diagram are numbered for a better identification. The calculation methodology of the Unit Process Mass Inventory (UPMI) and Unit Process Energy Inventory (UPEI) is a general guideline to build the parametrized LCI. The aggregation of both constitutes the LCI of the calculated unit process. With respect to the UPMI, a specific substitution modelling approach is proposed when waste or by-products are used as supplementary lime materials. For simplicity, in the Results section the most relevant calculation steps for each case study are shown. Figure 3.3 also shows where the focus has been placed in each case study.

3.2.2.1 General Calculation of a Unit Process

After determining the goal and scope of the study, manufacturing process, functional unit and unit operations, the mass inventory, followed by the energy inventory, are calculated.

- **Calculation of the Unit Process Mass Inventory**

In the traditional Cradle-to-Gate scenario (i.e., no circular economy included), the calculation starts by identifying the physical/chemical transformation taking place in each unit. Physical transformations are those in which the chemical composition of matter remains unaltered (such as a size reduction operation, washing or drying). Usually, simple mass balances can be used to determine the flows. On the contrary, whenever the chemistry of material is modified, a chemical transformation takes place (i.e., limestone calcination). This information serves to establish the mass or energy balances that provide the set of equations to define every mass flow on the unit. These flows can be either emissions or intermediate/final products. The aggregated material flows constitute the Unit Process Mass Inventory (UPMI).

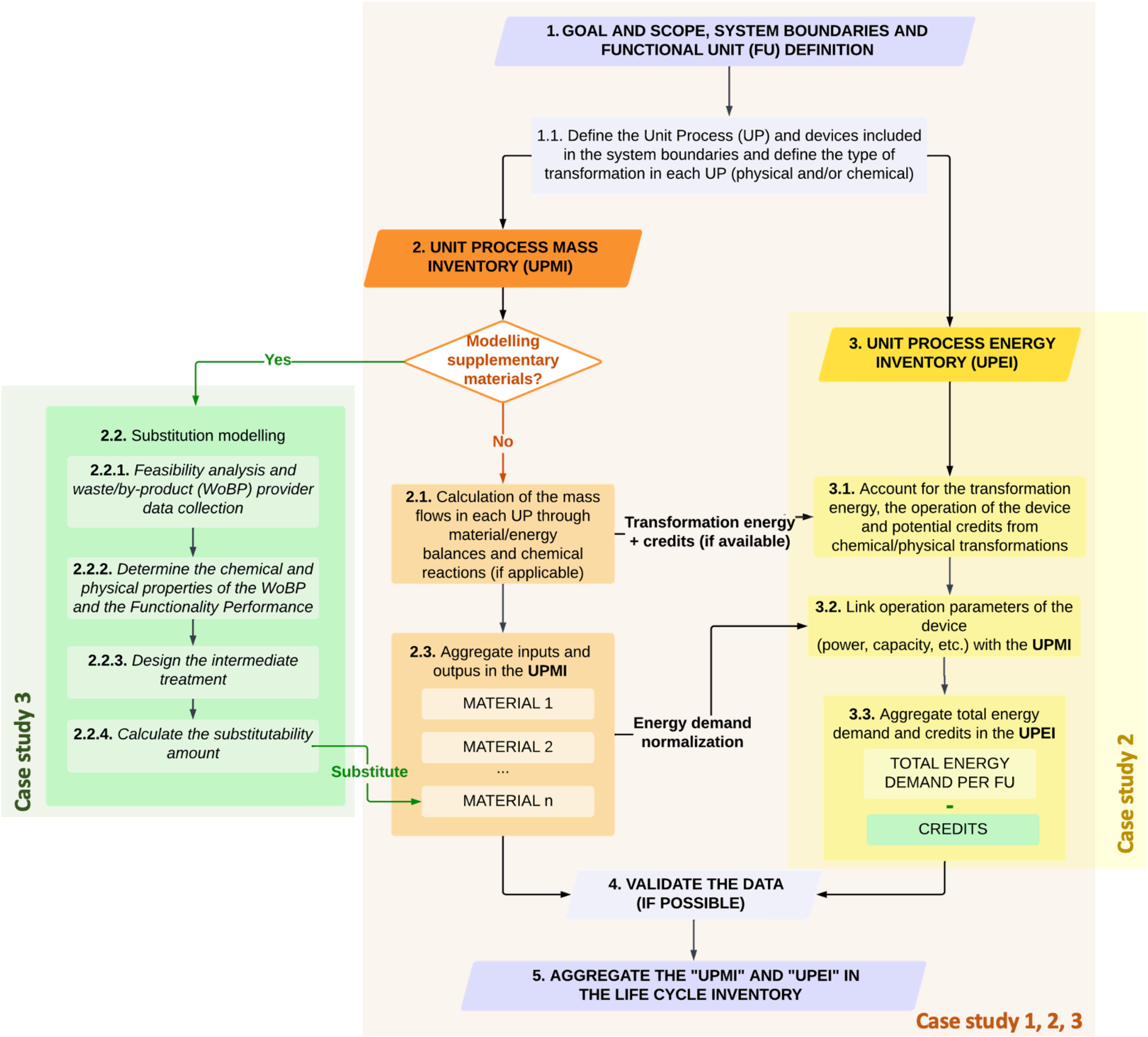


Figure 3.3. The process-oriented methodology for calculating the Cradle-to-Gate life cycle inventory of a manufacturing process and specific parts: the presented workflow is demonstrated in each case study.

- **Calculation of the Unit Process Energy Inventory**

The Unit Process Energy Inventory (UPEI) can only be calculated after the UPMI. The first reason behind this, is that the energy requirements are attached to the amount of material that needs to be processed (i.e., the energy demand is normalized to the mass flows). The second reason is that in many cases, there are interchanges of energy as result of chemical reactions or changes of state, which can be credited to the overall energy demand of the unit process.

The energy requirements depend on the physical or chemical process carried out and, on the technology used. The total energy consumption (Eq. 3.1) in the unit process can be expressed as:

$$E_{total} = E_{transformation} + E_{operation}$$ **Equation 3.1**

With regards to the first term $E_{transformation}$, the transformation of the materials involves interchange of energy (change of state, formation of new compounds, etc.). It is always helpful to model these processes using the chemical reactions or state change involved and using tabulated enthalpy data. The transformation energy needs to be complemented by the operational energy ($E_{operation}$), which is mostly defined by the type of device (i.e., technology) and its efficiency. Naturally, it is always a good practice to validate the data with literature and producers (Point 4 in Fig. 3.3). The application of these steps leads to a quantification of materials and energy required per functional unit in the inventory (Point 5).

3.2.2.2 Use of Supplementary Materials in the Cradle-to-Gate Production

When it comes to sustainable production and consumption, there are two problems to be addressed: the generation of waste and the consumption of virgin raw materials. The final disposal of waste materials is the less preferred option according to the waste hierarchy. Therefore, recycling waste materials for the manufacturing of valuable products under the so-called circular economy, emerges as a significative alternative to solve both problems [43]. Under this paradigm, there are at least two life cycles to consider: LC1 which generates the waste material/by-product (e.g., paper mill sludge) and the LC2 in which the determining product (e.g., lime-based plaster) is manufactured. The connection between both life cycles is what we call intermediate treatment, which has the function to treat the material to give a certain degree of equivalence between the displacing material and the material that will be displaced. The degree of equivalence is based on the substitution/system expansion concept, under which two or more products are considered interchangeable alternatives to fulfil a specific function [44].

The proposed methodology goes from a feasibility analysis of using the secondary material to the calculation of the substitution potential, linking the initial properties of the waste with the final properties required for the substitution. This methodology can be applied to model the inventory of a wide variety of waste treatments, independent of its initial properties. The availability of such datasets in generic databases is rather limited and by applying this approach the LCA practitioner can be independent of them. Moreover, given that the LCI is parametrized, it is also possible to evaluate different technologies and energy sources to power the devices. Further explanation of the steps for the application of the methodology is given below.

- **Feasibility Analysis and Waste or By-product Provider Data Collection**

The feasibility analysis includes the production process of the potential provider of the supplementary material and a market analysis to determine if the material should be considered as a waste or must be considered as a by-product. According to the EU directive 2008/98EC to qualify for the by-product status the following conditions should be met: (a) further use of the substance is certain (e.g., as cement replacement); (b) the substance or object is produced as an integral part of a production process; (c) the substance or object can be used directly without any further processing other than industrial normal practise; and (d) further use is lawful [45]. Therefore, part of the environmental impact associated to the production process should be somehow allocated to the by-products as well. However, if the material is usually disposed or if it does not have any economic value, the material can be considered as a waste and no allocation is required.

Furthermore, the local availability of the waste should be determined. On the one hand the rate of generation of the waste of interest by the first cycle process, should be sufficient to satisfy a predefined demand. On the other hand, the production plant of the first cycle process should be at a reasonable distance from the production plant of the determining product.

- **Determine the Properties of the Waste (or by-product)**

Along with the feasibility analysis, a waste or by-product can be introduced in the LC2 if its properties are to some extent related to the properties of the material that they would displace. Among relevant physical properties are identified the state (solid or liquid), density, humidity (if solid), average particle size (if solid); while chemical properties include chemical and mineralogical composition, pH, content of organics, etc.

- **Determine the Functionality Performance**

When looking for alternatives to replace the original materials, it is important to have clarity about the function they are fulfilling in the system. The aim of the supplementary material is to replace one of the original materials in the inventory. Therefore, the properties of the waste or by product should help to identify which material can be potentially substituted, according to a certain function that the original material provides. The functionality performance is an integral concept that requires the identification of a property of interest, in clear connection with the material that will be replaced and representative of it.

- **Design the Intermediate Treatment**

The intermediate treatment requires the selection and design (in the same way as described in Section 3.2.2.1) of the unit processes required to transform chemically or physically the waste or by-product in order to produce the secondary material that will be used as substitute. The design is linked to the definition of the functionality performance and generally would require scientific data as inputs for the design (calcination temperatures, materials, fineness requirements, etc.).

- **Potential Substitution Calculation**

The Equation 3.2 is inspired on the work by Vadenbo et al. [44] but the concepts behind the parameters are redesigned and adapted to be used in construction materials. The Equation can be used to calculate the potential amount of a specific resource that can be substituted (γ, in the Equation 3.2) [44].

$$\gamma = \alpha^{sr:disp} \cdot {R_p}^{sr} \quad \textbf{Equation 3.2}$$

The Equation integrates the data previously collected, considering the following technical aspects:

Substitutability ($\alpha^{sr:disp}$): is the link between the materials properties, the treatment process design and the final secondary resource that will displace the original material in the determining life cycle. The recovered resource is treated in a designed way according to the experiments performed to achieve a similar performance than the material that will be displaced. It is calculated as the ratio of a set of properties of the transformed secondary resource (sr) and the original displaced (disp) material. The degree of substitutability expresses to which extent the secondary resource can fulfil the function of the displaced material in the system.

Resource Potential (Rp^{sr}): represents the maximum amount of the secondary resource -after the intermediate treatment- that is being produced per FU of the secondary life cycle. It is important to accurately calculate the real amount that will be used as displacing product because the substitutability would only have a real effect in practise if the provision were enough in the conditions of a real plant operation. Otherwise, a benefit that cannot be provided is being accounted for.

3.3 Results and Discussions

This section presents the results of the application of the methodology on three case studies focusing on hydrated lime production. Figure 3.4 illustrates which part of the production process is covered in each case-study for clarity.

The first case-study focuses on the production of CaO, a precursor for hydrated lime production. It demonstrates the calculation of the parametrized LCI for a single unit process, as outlined in Figure 3.3 (Section 3.2.2). The second case study explores carbon savings and carbon capture in the production of calcium oxide, utilizing the parametrized LCI from case study 1. Finally, case study 3 demonstrates the application of a substitution approach for a lime-based mortar production by upcycling paper mill sludge, a waste of the paper industry.

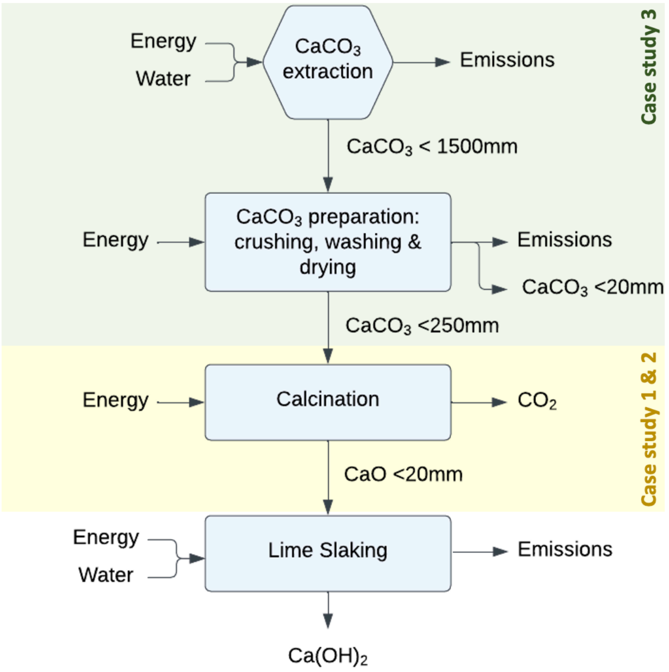


Figure 3.4. The flow diagram of $Ca(OH)_2$ production, along with the focus of each case study addressed.

3.3.1 Case-study 1: Calcium Oxide Production

Calcium oxide is the main precursor for hydrated lime manufacturing and is produced through the calcination process (Figure 3.4). Prior to calcination, the extraction and processing of calcite are required, involving activities such as crushing, washing, drying. While these operations are modelled using the same approach, for practicality and relevance, the methodology is applied specifically to calculate the inventory of the unit process "calcination".

Following Figure 3.3 (Step 1.1), as the calcination operation is being modelled, the functional unit is the production of 1 ton of calcium oxide and the system boundaries include the lime kiln, in which the chemical transformation from calcite to calcium oxide occurs. The calculation of the UPMI and UPEI is shown below.

3.3.1.1 Unit Process Mass Inventory (UPMI)

To calculate the mass flows in the unit process, the decomposition reaction of calcite in the lime kiln is first considered (Reaction 3.1). To produce 1 ton of CaO (FU) the amount of $CaCO_3$ that should be supplied, the amount of CO_2 generated and transformation energy to be supplied need to be calculated. For this calculation, the following assumptions are considered (1) 100% purity of $CaCO_3$, (2) 100% conversion of the reaction, (3) steady state of the process and (4) no mass accumulation in the process.

$$CaCO_3(s) + \Delta H \rightarrow CaO\ (s) + CO_2(g) \qquad \Delta H_{rCaCO_3}{}^{\circ} = 177.3\ kJ/mol$$ **Reaction 3.1**

Chemical reactions increase the complexity of a process. When analysing a system with chemical reactions, we need first to determine some important factors like stoichiometric coefficients, limiting reactant, and conversion percentage. Then we can choose the material balance equations, say, mass balance or element balance. The global mass balance can be written as follows (Eq. 3.3).

$$m_{\dot{CaCO_3}} = m_{\dot{CO_2}} + m_{\dot{CaO}}$$ **Equation 3.3**

After replacing the m_{CaO} (1 ton) in the Equation 3.3, there are two unknowns and therefore another equation is needed. The second type of balance that can be used is by chemical elements (Ca, C and O). To apply these balances, the molecular weight of the reactants and products are used ($CaCO_3$ = 100.08 g/mol; CaO = 56.07 g/mol; CO_2 = 44.01 g/mol). The balance can be done following any element, in this case Ca. Then, after making the calculations, the second equation needed to solve the system is found (Eq. 3.4).

$$0.0099 m_{CaCO_3} = 0.0178 m_{CaO}$$ **Equation 3.4**

Using Equations 3.3 and 3.4 the following results are found: 1.79 t $CaCO_3$/t CaO and 0.79 t CO_2/t CaO. These results mean that the “$CaCO_3$ preparation” unit process (Figure 3.4) should provide 1.79 t $CaCO_3$ to satisfy the function of the calcination process. All the material flows are now specified and can be aggregated in the UPMI.

3.3.1.2 Unit Process Energy Inventory (UPIE)

After completing the calculation of the UPMI, follows the energy inventory, which is calculated as specified in Eq. 3.1. Continuing with the calcium oxide production case, the term $E_{transformation}$ includes the chemical energy required for the decarbonation (Reaction 3.1). Here the UPMI and UPEI are integrated because the mass flow of $CaCO_3$ (1.79 t) and the enthalpy of the reaction (177.3 kJ/mol $CaCO_3$) are used and 3160 MJ/t CaO is the calculated value for the transformation energy term.

The second term in the UPEI is the operation energy, which highly depends on the operation principle and technology. As always, first-hand data is desired, but Best Available Technologies (BAT) published by the European Commission are a comprehensive source of information for the design, from which current technologies being used in the market, types of fuels used, the direction followed by the market and specific data such as efficiencies, average energy consumptions of devices, etc. can be obtained [19]. In the case of the lime industry, Parallel Flow Regenerative Kilns (PFRK) are the most efficient current technology for the manufacturing of lime and the literature research shows a total energy consumption by these kilns (E_{total} in Eq. 3.1) is 3900 ± 0.5 MJ/t CaO [19], [47]–[51]. With the calculated energy of the transformation (3160 MJ/t) and the total energy consumed by the device (3900 MJ/t), the energy of operation can be obtained (Eq. 3.1), amounting to around 740 ± 0.5 MJ/t. These values validate the calculation performed previously (Figure 3.3, Point 4).

3.3.1.3 Aggregated Data

The aggregated inventory of the unit process "Calcination" is summarised in Table 3.1, including the results of the UPMI and UPEI (Figure 3.3, Point 5).

Table 3.1. Inventory to produce 1 ton of calcium oxide

	PROCESSED AMOUNT		Comments
INPUT	**AMOUNT**	**UNIT**	
$CaCO_3$ from "$CaCO_3$ preparation"	1.79	t	See Figure 3.4
PFRK Operation	3900	MJ	(3160 MJ $E_{transformation}$ + 740 MJ $E_{operation}$)
OUTPUT	**AMOUNT**	**UNIT**	
CaO **(determining product)**	1	t	
CO_2 **(as emission)**	0.79	t	Chemical decarbonation of $CaCO_3$

The input $CaCO_3$ from "$CaCO_3$ preparation" brings along the environmental impact associated with all the upstream processes (from the extraction of the raw materials). An important remark is that the CO_2 produced during the calcite decomposition is accounted as an emission to the environment. Furthermore, by having the inventory parametrized, several alternatives for the decarbonation of the lime-industry can be implemented, as shown in the next case study.

3.3.2 Case-study 2: Carbon Savings/Capture in the Lime Industry

In the previous section, through the inventory calculation of the kiln, the case study evidences one of the main reasons for the high CO_2 emissions of the lime industry, namely decarbonation of limestone (0.79 tCO_2/t CaO). The second main reason are the emissions associated to the combustion of the fuels, which provide the energy for the chemical transformation. The mitigation of both impacts requires different strategies and here we show how the parametrized LCI calculated in Section 3.3.1 can help to assess these alternatives.

The main focus of this case study is then in the UPEI component of the inventory (Figure 3.3, Step 3). First, we discuss carbon direct avoidance as a mean to reduce the emissions of the fuel's combustion, which basically is oriented to the transition to more eco efficient energy sources [16]. Second, to reduce the emissions of the calcite decarbonation, a combination of improved kiln technologies coupled with energy recovery and carbon capture system is presented. Under this approach, CO_2 can be captured by a certain technology and furthermore be sold to vertical-related markets [52], [53].

3.3.2.1 Carbon Direct Avoidance

Carbon direct avoidance (CDA) requires the use of improved technologies in the lime kilns as well as eco-efficient fuels as energy source. The question that arises is how to include both scenarios in the parametrized LCI, because they will have a direct effect on the UPEI. To discuss this case study, we start from the parametrized LCI calculated in Table 3.1 and particularly focus, on the $E_{operation}$ term of the UPEI. More efficient kilns can reduce the heat loss through the walls of the kiln, can improve the efficiency of the fuel use and overall reduce the specific energy demand of the kiln (MJ/kg of product). The last point is of direct interest for the UPEI, as basically different specific heat demands are obtained for each technology.

Some kiln technologies used for quicklime production are shown in Figure 3.5. The Figure shows the contribution of the $E_{transformation}$ and $E_{operation}$ in the total energy demand (MJ/t) of each kiln (Eq. 3.1).

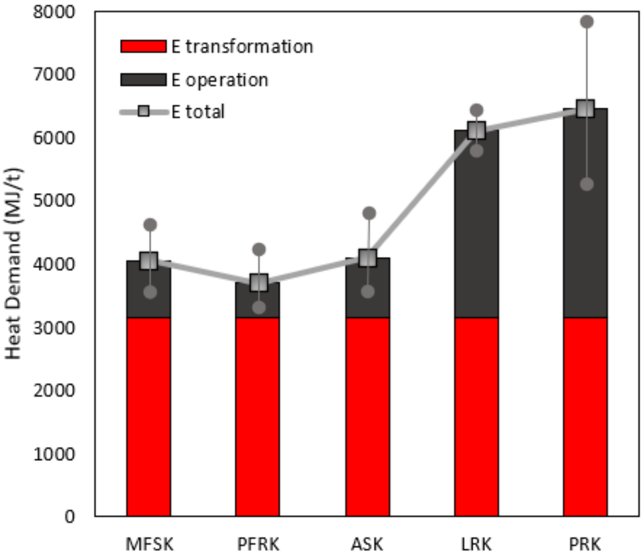


Figure 3.5. Contribution of the $E_{transformation}$ and $E_{operation}$ to the total Heat demand in lime kilns

Here it is observed that the contribution of the transformation energy is equal in all technologies, but the operational energy differs because all of the kilns have different efficiency. The minimum total energy demand is found for the PFRK kiln, which also explains why in Europe this the preferred technology is. Compared to the less efficient kiln LRK, around 40% reduction in the total energy is achieved. Both technologies can be implemented in the parametrized LCI by changing the PFRK operation line (heat requirement) in Table 3.1 from 3900 to around 6100 MJ/t on average. In this way, the effect of different technologies on the inventory can be analysed. A decrement in the heat requirement, will definitely lead to carbon direct emission avoidance. However, the exact values depend on the fuel mix used in the kiln because they have different CO_2 intensity per unit of energy released [16].

Moving towards eco-efficient fuels is a must to reduce the emissions, by minimizing the use of fossil fuels (hard coal, lignite, and petrol coke) [54]. According to EuLA report 2019 they represented around 51% of the fuel mix in the European Lime industry [48]. The main problem associated with fossil fuels is their high carbon intensity factor: 103.8 tCO_2/TJ for lignite, 74 tCO_2/TJ for light fuel oil and 55.9 tCO_2/TJ for natural gas [55]. The parametrized inventory developed in Table 3.1 helps to evaluate different energy sources, because different datasets can be selected from the used database (e.g., Ecoinvent) to model the production of the energy. Considering the energy demand of the PFRK (3900 MJ) and the carbon intensity factors of each fuel, different scenarios of fuel mixes can be evaluated, as shown in Table 3.2. For instance, if the used fuel is lignite or natural gas, it would lead to 0.23 $tCO_2/tCaO$ and 0.41 $tCO_2/tCaO$ respectively (44% increment). These scenarios are shown in Table 3.2. As can be observed in the Table, in the case of the use of natural gas, a total of 1.02 $tCO_2/tCaO$ are generated and 1.2 $tCO_2/tCaO$ for lignite. If natural gas is used instead of lignite, a 17% of carbon avoidance is obtained.

Table 3.2. Inventory to produce 1 ton of calcium oxide: Effect of two different energy sources

	FUEL: LIGNITE		**FUEL: NATURAL GAS**		**Comments**
INPUT	**AMOUNT**	**UNIT**	**AMOUNT**	**UNIT**	
$CaCO_3$ from "$CaCO_3$ preparation"	1.79	t	1.79	t	See Figure 3.4
PFRK Operation	3900	MJ	3900	MJ	(3160 MJ $E_{transformation}$ + 740 MJ $E_{operation}$)
OUTPUT	**AMOUNT**	**UNIT**	**AMOUNT**	**UNIT**	
CaO **(determining product)**	1	t	1	t	
CO_2 **(as emission)**	0.79	t	0.79	t	Decarbonation of $CaCO_3$
CO_2 **(as emission)**	0.41	t	0.23	t	Fuel combustion

Naturally, when building the LCI, coherent decisions regarding what is possible should be made. In principle, there is a mandate from the European Commission to reduce emissions in the building materials sector and therefore measures should be taken accordingly. However, the way in which they can be reduced is subjected to the availability of resources in each country. Not all countries can achieve this goal in the same way, and this should be reflected in the LCI.

For the specific case of the effect of changing the energy sources to power a production system of hydrated lime, the reader is referred to Chapter 6 [16]. In this work, a parametrized inventory of the plant was calculated following the methodology in Figure 3.3. The environmental impact of current and potential future energy sources used for hydrated lime production was calculated and the implementation of the scenario analysis in the parametrized LCI follows the same principle as explained before. The research was complemented by a geographic localization of the plant in which the availability of different energy sources was thoroughly analysed. It was found that as the proportion of fossil solid fuel decreases and natural gas and biomass increases in the fuel mix, a reduction of up to 22% in the Global Warming

Potential is achieved [16]. A detailed LCI and the Ecoinvent databases used in the analysis for the modelling of the energy sources used can be found in the research article [16] and in Chapter 6.

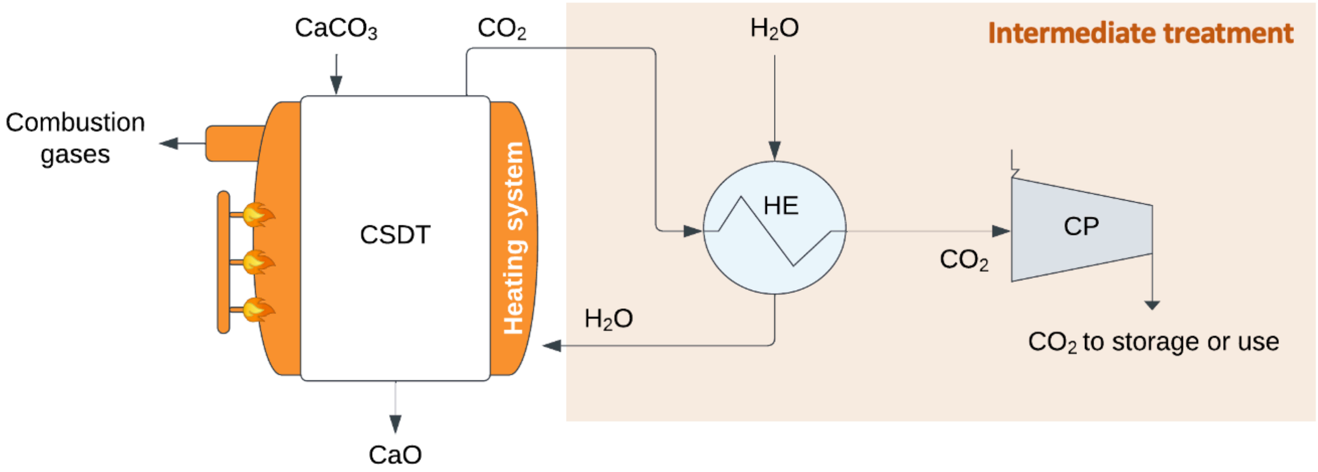
3.3.2.2 Smart Carbon Separation and Carbon Capture Technologies (SCS-CCT)

The current scenario on Smart Carbon Separation and Carbon Capture Technologies (SCS-CCT) is still underdeveloped to be applied to large scale projects, because of poor expectations of financial returns, inconsistent and insufficient policy support, technological uncertainties, and a complex value-chain system [56]. However, while CO_2 emissions from the combustion of the fuel can only be saved if they are replaced by greener fuels (such as biomass), CO_2 emissions resulting from the decomposition of the limestone (around 65% of the total emissions), can only be avoided by carbon capture technologies (CCT). Post pyrolysis CCT are of specific interest in the case of the lime industry, and they are being developed to reduce the emissions of the calcination process. There are different routes for carbon capture that can be applied in the lime industry, ranging from very well-developed solvent-based technologies to the indirect heating of the raw meal to capture the unavoidable CO_2 emissions (Calix's Direct Separation Technology, CDST) [57]. For a specific review of several available CCT the reader is referred to Simoni et al. [28].

The CDST kiln is a very promising technology as it requires minimal changes to the conventional process in the lime industry [57]. Regarding the working principle, calcite powder enters at the top of the kiln and the calcined product leaves the reactor at the bottom, while the CO_2 gaseous stream because of the Reaction 2.1 (Chapter 2) exits in counter-current flow the reactor. The main benefit of the technology is that the combustion gases are not mixed with the CO_2 from the decomposition of calcite, and mostly CO_2 is generated, avoiding complicated purification steps.

Recently, the International Energy Agency has discussed the alternative of using CO_2 in several industries. They state that the carbon in CO_2 enables the conversion of hydrogen into a fuel that is easier to handle and use and that CO_2 can also replace fossil fuels as raw material in chemicals and polymers [58]. Therefore, there is no doubt that if the purity of the obtained CO_2 is sufficient, not only avoided emissions but also revenues can be obtained. Even though the market for CO_2 at this moment is still in development, early opportunities can be cultivated, and CDST is an interesting technology for that purpose.

To illustrate this scenario through the process-oriented methodology, consider the CDST shown in Figure 3.6 in which an intermediate treatment for the CO_2 is provided. In this case study, the parametrized LCI presented in Section 3.3.2.1 in Table 3.2 (case of natural gas) is expanded to consider the extra unit processes required for the purification of the CO_2 from calcite decomposition. Here, we show how to apply the methodology to model the intermediate treatment of Figure 3.6 (Following Figure 3.3). It is relevant to mention that there are no available databases to model these kinds of treatments and therefore, the feasibility of modelling the LCI through the proposed methodology is a significant advantage.



*CSDT: Calix's direct separation technology; HE: Shell and Tube Heat Exchanger; CP: 2-stage compressor

Figure 3.6. Combined carbon capture system for CO_2 purification and heat recovery

The system boundaries include now the kiln, the heat exchanger and the compressor, being the last two devices the intermediate treatment to prepare CO_2 to storage or use (Figure 3.6). The functional unit is still the production of 1 ton of CaO. The first unit process of the intermediate treatment is a heat exchanger, used to cool down the CO_2 gaseous stream leaving the kiln at 1173.15 K and to recover part of the intrinsic thermal energy of the stream. The heat exchanger is designed considering (1) the flow of CO_2 (790 kg/h, Table 3.2) leaves the reactor at 1173.15 K and needs to be cooled down to 273.15 K, and (2) water at 273.15 K is used as heat exchanger medium, changing from liquid to vapor state (2260 kJ/kg) and entering the heating system of the kiln at 1173.15 K. Below the calculation of the UPMI and UPEI for the intermediate treatment are shown.

UPMI and UPEI calculation

Following the Figure 3.3, first the material flows are determined. To calculate the required amount of water input to cool down the CO_2 stream, the energy balance assumes that all the heat from the CO_2 stream is transferred to the water.

$$Q_{H_2O} = Q_{CO_2}$$

$$Q_{H_2O(l)_{sensible}} + Q_{H_2O(l)_{latent}} + Q_{H_2O(v)_{sensible}} = Q_{CO_2(g)_{sensible}}$$

$$m_{H_2O}cp_{H_2O}\Delta T + m_{H_2O}\lambda_v + m_{H_2O}cp_{H_2O}\Delta T = m_{CO_2}cp_{CO_2}\Delta T$$

$$m_{H_2O}\left[4.18\,\frac{kJ}{kgK}(373.15-298.18)K + 2260\frac{kJ}{kg} + 1.99\frac{kJ}{kgK}\,(1173.15-373.15)K\right] = 790kg\;0.84\,\frac{kJ}{kgK}\,(1173.15-298.18)K$$

$$m_{H_2O} = 128\;kg$$

In the heat exchanger, 790 kg of CO_2 are cooled down by 128 kg H_2O and all material flows are determined. For the calculation of the UPEI, the water vapor is reinserted in the heating system of the kiln. The total energy that can be theoretically transferred is:

$$Q_{CO_2} = m_{CO_2} cp_{CO_2} \Delta T = 790 kg\ 0.84\ \frac{kJ}{kgK}\ (1173.15 - 298.18)K = 580.63\ MJ$$

Besides the thermal energy that can be credited, the UPEI can be complemented by the electricity consumption of the heat exchanger (Point 3.1, Figure 3.3). Considering that the heat exchanger should deal with high pressures because of the gaseous states of the streams, a Shell and Tube Heat exchanger is recommended. Such heat exchangers can deal with biphasic flows; high and low pressures and they are compact and efficient. For simplicity we do not elaborate further but by knowing basic parameters such as material flows, state, etc. electricity consumption of the device can be obtained from a catalogue (e.g., https://www.funke.de/).

In compressor design, material balances are not necessary, and the focus is primarily on determining the UPEI. After the CO_2 is cooled down, 790 kg of CO_2 needs to be further processed for storage. For the specified flow, the electricity consumption of a 2-stage compressor is 1.3 MWh/Mt CO_2 (Point 3.1, Figure 3.3) according to an ASCO device (https://www.ascoco2.com/). The total electricity requirement considering the amount of processed CO_2 is 1.027 kWh.

Aggregated LCI data

Table 3.3 presents the parametrized LCI of the CSDT with heat recovery system (Figure 3.6). Despite its seemingly simplistic nature, this example yields several significant insights regarding the decarbonization of the lime industry. First, around 65% of the total emissions can be captured ([16]) and the total energy consumption of the lime kiln can be reduced by around 15% through the recovery of 580 MJ of energy from the CO_2 stream. The immediate result is that the fuel required, and the consequent CO_2 emissions of the fuel combustion diminish as well (as discussed in Section 3.3.2.1). In particular, with the traditional PFRK (heat requirement 3900 MJ) fuelled by natural gas, 0.23 $tCO_2/tCaO$ were released. However, with the savings of the CSDT system, the emissions are 0.20 $tCO_2/tCaO$, representing a 13% reduction in comparison to the previous scenario. Further decarbonization could be achieved by changing the fuel mix (Section 3.3.2.1).

Finally, the proposing system shows potentiality to obtain pure CO_2 with further economic value as discussed before. Under this hypothesis, the CO_2 from $CaCO_3$ decomposition could be potentially considered no longer as an emission but as a by-product of the CaO production [58]. As a result, part of the environmental load of CaO production could be allocated to CO_2 production. For instance, if a mass allocation procedure is applied, all the emissions from “$CaCO_3$ extraction” until “Calcination” (Figure 3.4) would be allocated 56% to CaO and 44% to CO_2 (Table 3.3).

Table 3.3. Inventory of the combined calcination and CO_2 recovery of lime production

			PROCESSED AMOUNT		**ALLOCATION**	**COMMENT**
	OPERATION	**FLOW**	**AMOUNT**	**UNIT**		
INPUT	**Calcination**	$CaCO_3$ from "$CaCO_3$ preparation"	1.79	t	-	See Figure 3.4
		Shaft kiln Operation	3320	MJ	-	3900 MJ **-580 MJ credit heat exchanger**
	Heat exchanger	Water	0.13	t	-	
	Compressor	Electricity consumption	1.027	kWh	-	
OUTPUTS	**OPERATION**	**FLOW**	**AMOUNT**	**UNIT**	**MASS**	
		CaO **(determining product)**	1	t	0.56	
		CO_2 **(as co-product)**	0.79	t	0.44	Decarbonation of $CaCO_3$
		CO_2 **(as emission)**	0.20		-	Fuel combustion (Natural gas)
		Emissions heating system			-	
		Water vapor **(as emission)**	0.13	t	-	

This limited example aims at showing how to model the unit processes through the methodology described in Figure 3.3, while emphasizing the ground-breaking idea of considering CO_2 no longer as a problem but as an opportunity of cost reduction. With regards to the costs, within the EU emissions allowance, the cost of emitted carbon by the industries can be significant and in the last 4 years, it has incremented 9 times [59]. According to the European Central Bank in 2018 it was 10 € per metric ton of carbon and in 2022 90 € per metric ton of carbon[20]. Considering the carbon price in 2022, for the case in which natural gas is used the carbon cost would be 91,8 €/tCaO (Table 3.2). However, by applying the CDST system (Table 3.3) a reduction of around 85% in the carbon taxes can be obtained. Furthermore, the recovery of pure CO_2 encourages the exploration of further market integration with potential revenues for the lime industry.

3.3.3 Case-study 3: Use of Wastepaper Mill Sludge in a Lime Plaster Manufacturing

In this section we focus mainly on the modelling of supplementary materials as specified in Figure 3.3, Step 2.2. In the context of circular economy, the idea of supplementary materials is to reduce the extraction of virgin raw materials (Figure 3.4), by looping in a closed or open cycle the materials at the end of life. In particular, the case study deals with the open upcycling of paper mill sludge. This residue is generated during the treatment of wastewater in paper production and can potentially be used in a lime-based plaster manufacturing, replacing the extraction of calcite. The application of the substitution modelling to calculate the life-cycle inventory of the intermediate treatment is shown below.

3.3.3.1 Feasibility Analysis and Provider Data Collection

Figure 3.7 shows the system boundaries of the Life-cycle 1 (production of paper) and the upcycling Life-cycle 2 in which PMS is reinserted for the lime-based plaster production. In the wastewater treatment, the effluent should be cauterized by using CaO, which reacts with soda ash (Na_2CO_3) and produces caustic soda (NaOH) along with $CaCO_3$ (lime sludge) as a waste material [60], [61]. Once the waste is treated, the most common practice has always been the landfilling disposal of the sludge (around 70% of the total generated) [62], [63]. Therefore, it is fair to consider PMS as a waste.

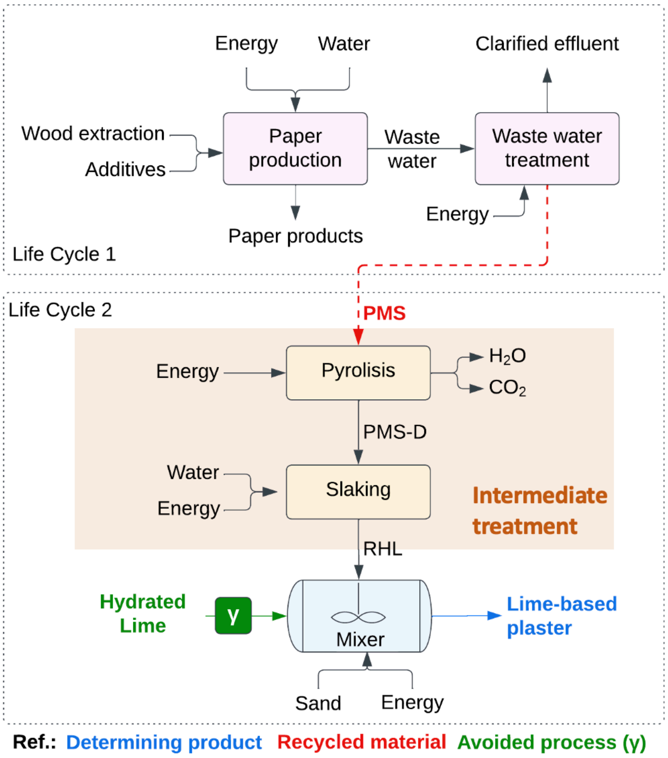


Figure 3.7. Simplified process of paper mill sludge generation (Life-cycle 1, secondary resource) and lime-based plaster manufacturing (Life-cycle 2, determining product)

In a real case, the cooperation between the paper industry and the plasters industry would be desirable to determine how much PMS could be provided. This collaboration between stakeholders is a key aspect to ensure the sustainable development of the construction materials. However, it is also possible to make some previous theoretical estimations. In terms of production of paper and pulp mill sludge, around 40 to 50 kg of sludge (70% primary sludge and 30% secondary sludge) is generated in the production of

1 ton of paper at a paper mill [64]. In 2020 in Europe around 8.5E7 t of paper and board were produced and the market is dominated by Germany (25.1%) [65] and the overall daily production of paper and board in Germany is 0.059 Mt. Assuming equal capacities of the plants, on average this value could be divided by the total amount of factories in Germany (160) to estimate a reference flow of the paper and board production for the Life-cycle 1 (372.6 t/d) [65]. Then considering the daily production and the ratio of sludge-to-paper, the maximum availability of the paper mill sludge of the Life-cycle 1 in Germany per reference flow of is 18.6 t_{sludge}/day.

3.3.3.2 Determination of the Relevant Properties of the Waste

The feasibility of the recycling depends on the physical and chemical properties of the PMS. With respect to the chemical composition, to have a snapshot of the ranges in which the mineralogical composition of the PMS can vary, literature was researched, and Table 3.4 is provided. Average moisture of the PMS, pH, and amount of organics are reported in the same Table and used for the LCI calculation. In general, the chemical composition of the sludge is relatively close to the one of Hydrated Lime and therefore the feasibility of its replacement is supported. The moisture and the organics content need to be considered for the process design, as they have to be removed and therefore energy will be consumed to power the intermediate treatment process for the conditioning.

Table 3.4. Chemical composition of Paper Mill Sludge, Hydrated Lime, and Portland Cement

	Component (wt.%)	**Paper mill sludge (PMS)** [66]–[71] [72] [69] [63] [73]	**PMS (assumed)**	**Hydrated Lime Commercial** [74]
Mineralogical composition (dry base)	SiO_2	2 – 20	15	0.13
	Al_2O_3	0.8 – 5	2.5	0.06
	Fe_2O_3	0.25 – 2.5	-	0.07
	CaO	16 – 90	75	98.53
	MgO	0.2 – 10	5	1.09
	Others	0 – 0.3	-	0.25
Physical parameters	Moisture (%)	28.4 ± 11.0	20	-
	pH	7.3 ± 1.8	7	-
	Organics	34.9 ± 21.1	40	-

3.3.3.3 Determination of the Functionality Performance

As the PMS is intended to be used as replacement of hydrated lime, the content of portlandite is used to account for the functionality performance. The maximum amount of portlandite that can be produced is calculated considering the assumed chemical composition (75% CaO) and the only reaction occurring is the lime slaking (Reaction 3.2).

$$CaO\ (s) + \mathrm{H_2O\ (l)} \rightarrow Ca(OH)_2(l) + \Delta H \qquad \Delta H_{rCaCO_3}{}^{\circ} = -65.3\ kJ/mol$$ **Reaction 3.2**

The percentages of portlandite can be considered equal to the percentages of CaO (Table 3.4) and the substitutability is calculated as follows

$$\alpha^{sr:disp} = \frac{[Ca(OH)_2]^{sr}}{[Ca(OH)_2]^{disp}} = \frac{0.75}{0.98} = 0.765$$

The substitutability would be equal to 1 in the case that the recycled material can provide as much hydrated lime as the substituted material.

3.3.3.4 Design of the intermediate treatment

After being transported to the plant, PMS must be conditioned to remove the initial humidity (20% of the total mass) and the organic matter (40% dry mass) according to Table 3.4. Previous studies have shown it can be removed by pyrolysis and that the main mass loss attributed to the decomposition of the organic matter in PMS takes place between 300-400 °C [75], [76]. The next step, after removing the humidity and the organic matter is the thermal activation of the PMS, which occurs at around 700-800°C. The water removal, organic material removal and calcium oxide synthesis process is modelled by a lime kiln. The energy requirements of the lime kiln were based on a previous work [16]. The main emissions associated to the raw materials during the calcination step are water vapor and carbon dioxide. The last one is divided in CO_2 from the calcite decomposition (Reaction 3.1) and organic matter decomposition (Reaction 3.3), and they were calculated as the stoichiometric amount resulting from the chemical reactions and the amounts defined in Table 3.4.

$$C + O_2 \rightarrow CO_2 + \Delta H \qquad \Delta H_{rC}{}^{\circ} = -393.5\ kJ/mol$$ **Reaction 3.3**

After the calcination, the calcined PMS is mostly CaO (PMS-D) and is hydrated using a lime hydrator, the energy requirements of which are also based on a previous work [16]. For simplification the UPMI and UPEI calculation are not shown, and Table 3.5 summarizes the LCI of the Intermediate Treatment of PMS, to produce a recycled hydrated lime (RHL).

Table 3.5. Life-cycle Inventory of the intermediate treatment processes of PMS to produce recycled hydrated lime (Life-cycle 2)

Operation/ process modelled		Processed amount		Inventory Amount		Sources & Notes
		Process Step: Pyrolysis				
Input	**PMS**	2.02	t	1	t	
	Transportation	338	tkm	200	km	Assumed
	Kiln fuel consumption	4461.6	MJ	2640	MJ/t PMS	Laveglia et al. [16]
Output	**PMS-D**	0.81	t	-	-	
	Water vapor (as emission)	0.40	t	0.2	t H_2O/t PMS	Calculated stoichiometrically (Humidity 20%)
	CO_2 **(as emission)**	2.46	t	1.46	t CO_2/t PMS	Assumed 40% organic matter (Reaction 3, do not count as emissions)
	CO_2 **(as emission)**	0.48	t	0.28	t CO_2/t PMS	Calculated stoichiometrically (Reaction 1)
	CO_2 **(as emission)**	0.26	t		55.9 t CO_2/TJ	Fuel combustion (Natural Gas)
		Process Step: Lime slaking				
Input	**PMS-D**	0.81	t			
	Water	0.19	t	0.32	t H_2O/t CaO	Calculated stoichiometrically
	Hydrator Electricity operation	0.35	kW	0.35	kW/t	Laveglia et al. [16]
Output	**RHL**	1	t			

3.3.3.5 Calculation of the Substitutability Amount

The technical substitution potential is calculated as follows according to Equation 3.2. The substitutability was calculated before. In the case of the Resource potential, the maximum amount to be inserted in the intermediate treatment is 18.6 t PMS/d and the effective amount to be used as replacement of the original resource is 1 t of RHL/2.02 t of PMS (Table 3.5). Therefore, the Resource potential is 9.20 t and the technical substitution potential is 7.04 t. From a practical point, the technical substitution implies that 9.20 t of the RHL are equivalent to 7.04 t of HL. Assuming that the functionality performance of the secondary resource is acceptable, this amount represents the maximum quantity of the transformed secondary resource that is available per reference flow of the Life-cycle 1 to be reinserted in the Life-cycle 2.

The reference flows of the Life-cycle 1 and 2 should be considered at the same time to determine the real influence of this value on the inventory of the determining product, in this case a lime plaster. If the amount that can be substituted is significant or not, depends on the process demand of Life-cycle 2 (production capacity of the plant). To illustrate this concept, consider the reference flow of Life-cycle 1 (368.75 t_{paper}/d; 18.63 t_{sludge}/d) and the LCI of Life-cycle 2 in Table 3.6 (based on Figure 3.7). Three alternatives of reference flows are evaluated in Table 3.6, keeping in all of them the mixing ratio 1:3 Hydrated Lime:Sand.

Table 3.6. Life-cycle Inventory of the lime-plaster production and effect substituting hydrated lime by RHL

Operation / process modelled Process step: Mixing		A. No substitution		B. Max substitution		C. Max substitution	
Input	Hydrated Lime	25	t	18	t	55.5	t
	Sand	75	t	75	t	187.5	t
	RHL	-	-	7	t	7	t
Output (Reference flow)	Lime Plaster	100	t	100	t	250	t

First, case A represents no substitution of hydrated lime (base case) with a reference flow of 100 t/d. Case B and C include a substitution of Hydrated Lime by the Recycled HL, with two different reference flows (100 t/d and 250 t/d). As the reference flow of the Life Cycle 2 increases, the potential benefit of the substitution diminishes from 28% (case B, 7 t_{sludge}/25 t_{binder}) to around 11% (case C, 7 t sludge/62.5 t binder), because even though the proportion binder:aggregate is fixed, more binder and aggregate is produced to satisfy the increased reference flow.

3.3.4 Decarbonizing the Lime-based Construction Materials Industry

The lime industry's carbon dioxide emissions are mainly concentrated in the calcination process, where $CaCO_3$ is converted to CaO. Approximately 0.79 tCO_2/t CaO emissions are inevitable due to the decomposition of calcium carbonate (65% of the total emissions). Additional CO_2 is generated from fuel combustion in the kiln, and the emissions vary depending on the fuel mix used [16].

Sections 3.3.1 to 3.3.3 demonstrate three strategies to reduce CO_2 emissions: carbon direct avoidance, carbon capture and circular economy. These strategies can be implemented in various ways and require a flexible and rigorous methodology. The process-oriented methodology developed in this paper enables the successful application of these strategies, considering different production contexts and scenarios. It allows for independent calculations based on scientific parameters, rather than relying solely on generic databases. The developed methodology offers freedom to LCA practitioners and facilitates the implementation of diverse decarbonization pathways. Figure 3.8 summarizes significant insights and lessons learned from the case studies conducted in this research.

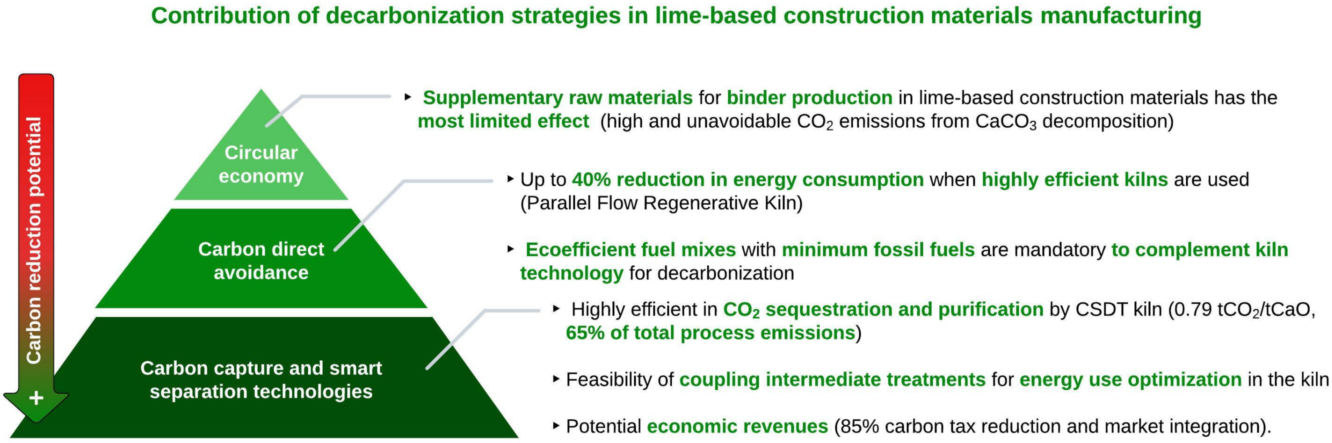


Figure 3.8. Effect of strategies on carbon reduction potential for decarbonizing lime-based construction materials industry.

3.3.4.1 Carbon Capture and Heat Recovery Through Smart Separation Technology

The design based on a smart separation of CO_2 aims at taking advantage of each possibility to reduce CO_2. In the best case, 0.79 tCO_2/tCaO from calcite decomposition can be captured. Furthermore, for the preparation of CaO, fuel has been burned to decompose the calcite, and the exhaust CO_2 brings along thermal energy that can be recovered. By implementing a heat exchanger in the intermediate treatment for the CO_2 stream, 533 MJ could be credited in the total energy consumption of the lime kiln. This represents 15% reduction in the total energy consumed (3367 – 3900 MJ/tCaO) and 13% (0.20 - 0.23 tCO_2/tCaO) reduction comparing scenarios with and without the technology.

Furthermore, given the further use of pure CO_2 as raw material in other industries, in the future and if this technology is applied, it could be considered a by-product and no longer an emission. As a result, part of the environmental load of CaO production can be allocated to CO_2 production, reducing the total environmental impact of the lime-based construction materials. The latter option can also have economic implications for the industry, because of the reduction in carbon taxes (up to 85%) and additional revenues could be obtained by the selling of the CO_2.

3.3.4.2 Carbon Direct Avoidance: More Efficient Kilns and Sustainable Energy Sources

In this paper the effect of the operational energy on the overall energy consumption for CaO manufacturing has been analysed. The preferred technology in Europe is a PFRK kiln because it requires 3900 MJ/tCaO vs around 6100 MJ/tCaO for the less efficient LRK technology (40% reduction in the total energy consumption). The technology of the kiln plays a significant role for the total CO_2 emitted, because by reducing the total energy consumption of the device, less fuel is required. In particular, the emissions of the fuel combustion depend on the fuel mix, because each has a specific carbon intensity factor. For the same energy consumption, if natural gas is used instead of lignite for CaO production, a 17% reduction in the total CO_2 emitted is obtained. This reduction can be up to 22% if a combination of eco-efficient fuels including biomass is used.

3.3.4.3 Circular Economy: Use of Alternative Raw Materials

Although the circular economy concept is useful to preserve the virgin materials from its extraction, it does not always lead to significant CO_2 reduction. In the case of lime-based materials, the main problem is that the emissions from $CaCO_3$ decomposition are a big share in the total CO_2 and they cannot be avoided. In order to be used as supplementary materials, the waste or by-products should have a similar chemical composition as hydrated lime (i.e., amount of CaO) and the same nature of emissions would be expected when they are calcined.

The type and number of intermediate treatments for the conditioning of the waste or by-product and the transportation required can limit the potential credits obtained. In the case of PMS, 60% of the material is organics and water, therefore more PMS needs to be processed to obtain the CaO required for Hydrated Lime preparation (1.44 t $CaCO_3$/t $Ca(OH)_2$ vs 2.02 t PMS/t $Ca(OH)_2$). As a result, the energy consumption in the kiln is around 12% more and the CO_2 of the fuel combustion is incremented by 23%. However, the specific emissions of the calcite decomposition in PMS are around 20% (0.60 and 0.48 t CO_2/t $Ca(OH)_2$) lower because of the reduced $CaCO_3$ content in the PMS ($\alpha^{sr:disp}=0.765$, Section 3.3.3). Focusing only on the calcination operation, the expected reduction by using PMS in comparison to the traditional manufacturing of hydrated lime (natural gas fuel) is around 9% [16].

3.4 Conclusions

This paper presents a new framework for calculating a parametrized life-cycle inventory (LCI) for lime-based materials manufacturing, incorporating process and materials design principles. Through three case studies, the successful implementation of three decarbonization strategies is demonstrated, leading to the following conclusions:

- CO_2 emissions in lime-based materials manufacturing are primary concentrated in the calcination operation, with 0.79 $tCO_2/tCaO$ being unavoidable due to calcite decomposition. Additional CO_2 emissions are influenced by fuel combustion in the kiln. Among the three decarbonization strategies discussed, carbon capture offers the highest potential for carbon reduction, followed by carbon direct avoidance and circular economy approaches.
- The choice of lime kiln technology significantly impacts total CO_2 emissions, as reducing energy consumption in the kiln reduces the need for fuel. The preferred technology, such as the Parallel Flow Regenerative Kiln consumes 40% less energy compared to less efficient options like the Long Rotary Kiln. Using natural gas instead of lignite as fuel can further reduce CO_2 emissions by up to 17%.
- Implementing a direct separation technology for CO_2 capture from calcite decomposition can capture 0.79 $tCO_2/tCaO$ in the best-case scenario. When combined with thermal energy recovery, it reduces kiln energy consumption by 15% and CO_2 emissions by 13%. The purity of captured CO_2 and its potential for use as a raw material in other markets make it a potential by-product in the future. The reduction in CO_2 emissions also has economic implications, as it can reduce carbon taxes by up to 85%, contributing to economic sustainability.
- The use of supplementary raw materials for binder production in lime-based construction materials has the most limited effect of the analyzed strategies, primarily due to the high and unavoidable CO_2 emissions from calcite decomposition. Using paper mill sludge as a replacement for hydrated lime in lime-based plaster manufacturing can reduce CO_2 emissions by up to 9%, depending on the intermediate treatment required.

The parametrized LCI modelling methodology presented in this paper provides greater autonomy to LCA practitioners by reducing reliance on generic databases. It incorporates scientific principles and transparent parameterization, enabling practitioners to calculate methodologically sound and well-grounded inventories. However, it requires a background in process engineering, detailed documentation of assumptions, and validation with industry data whenever possible.

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Chapter 4: Cradle-to-Gate Life-cycle Assessment of Current Lime-Based Rendering Materials

To make comparisons with future scenarios with enhanced sustainability, the baseline scenario of the current situation should be established for lime-based rendering materials.

In this chapter a real-case scenario is described in which a plant from one SUBLime industrial partner was studied. An unprecedented life cycle inventory analysing different binders, aggregates and additives was built and used for the life cycle assessment calculation. A life cycle cost calculation was also made to evaluate the effect of carbon pricing on the total cost of the material.

This environmental and economic analysis provides a robust scenario against which benchmarking of alternative manufacturing processes can be made.

Publication 2. How to Improve the Cradle-to-Gate Environmental and Economic Sustainability in Lime-Based Construction Materials? Answers from a Real-life Case-study

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4.1 Introduction

The present environmental crisis urgently calls for a shift towards a green and circular economy[1], [2] which requires a redesign of materials and industrial processes, along with in-depth technological and scientific knowledge. However, reaching the goal of a sustainable future requires much more than technological innovation alone. It requires a coordinated action of the scientific community, governments, stakeholders, and the society in general. One of the common needs among these actors, is to make rational decisions based on reliable information. In the field of sustainability, LCA has been considered as a widely preferred methodology to calculate the impact over the life-cycle of a product or service, through quantifying a set of indicators [3].

Sustainable development is a wide and complex concept that, in words of the Intergovernmental Panel on Climate Change (IPCC), refers to *“meeting the needs of today without compromising the possibility of future generations to meet their own needs”* [4]. It is based on three fundamental pillars that interact together: environment, economy, and society [5]. During the past decades, the use of Life Cycle Assessment (LCA), mostly in the first two pillars, has been triggered by the prevailing environmental crisis, energy consumption, materials scarcity, and a fast-changing economy [6]. All these concerns are pursued by the United Nation under the 2030 Sustainable Development Goals, more specifically "Good and Health well-being", "Affordable and Clean Energy", "Industry, Innovation and Infrastructure", "Sustainable Cities and Communities", "Responsible Consumption and Production" and "Climate Action" [7]. The magnitude of the problem demands a joint effort of all stakeholders.

The construction sector is among the biggest contributors that are responsible for the current environmental crisis. Main reason for this is that its materials are and will continue to be used in tremendous quantities all over the world. One of the worldwide most used materials is cement, the production of which during the last six decades has increased by a factor 34, while meanwhile the world population only grew by a factor 3 [8]. However, cement-based materials are not the solution for all our building needs. The wide variety of construction materials also includes lime, one of the oldest building materials, of which the use dates back to 7000 BC [9].

In the European market, around 20 Mt/y of Quicklime (main raw material) were produced with the construction sector holding around 18% of the sales [10] [11]. According to EULA [12], the construction and civil engineering industry are major users of lime-based products, ranging from aerated concrete and asphalt to bedding mortars, renders and plasters [12]. In particular, lime-based mortars are not only used in new construction but are also useful for restoration and preservation of cultural heritage [13], because their chemical composition, transport properties, etc. are compatible with the majority of Europe's built heritage [12]–[15]. It is relevant to point out the importance of restoration, since it enhances the cultural value of buildings, because by attracting millions of tourists every year they represent an important source of income for various European countries.

Despite its flexibility and use in many applications, very little is reported in literature about the environmental and economic performance of lime-based materials. While extensive research on environmental impact assessment of concrete and cementitious materials has been carried out over the last decade [14]–[18], little is known about a sustainable assessment of lime-based materials. As a matter of fact, only some research articles have addressed their environmental profile through a LCA ([19]–[22]) while no records of economic assessment have been found so far. Moreover, a detailed Cradle-to-Gate LCA has not yet been thoroughly researched, which is the stage in the life cycle of a material where most of the impact is generated. Having a detailed analysis of all stages from the cradle to the gate is the first step to improve the environmental indicators along with the economic indicators, when the prices of CO_2 emissions in the context of the European Trading System are considered. In particular, this assessment should consider the mix designs, type of coarse and lightweight aggregates, additives, etc.

Each component can contribute differently to the overall environmental and economic performance of a material, and it is relevant to know up to what extent. This is needed, in order to identify potential improved scenarios such as the selection of one component against other, ranges of variation of indicators, proportion of the total impact assigned to each component, etc. For instance, during the production of calcium oxide, being the precursor for hydrated lime production, up to 1.2 kg $CO_{2\ eq}$/kg lime is emitted [23], while in the case of 1 kg of Portland cement this is 0.9 kg $CO_{2\ eq}$ [24], [25]. As a result, an increased content of hydrated lime in a mix, would imply an increase of the Global Warming Potential [26]. However, the effect on other categories should also be considered when building an integral analysis of the overall environmental impact, beyond $CO_{2\ eq}$ emissions. The use of natural and polymeric lightweight aggregates could imply a significative difference.

Furthermore, a very critical aspect in terms of environmental sustainability is the types and proportions of the additives incorporated in the mix, the impact of which is usually reported as part of the binder production or, when reported separately, only $CO_{2\ eq}$ emissions are shown which may lead to a biased analysis of its true environmental impact [20], [27]. Considering the deficiency of comprehensive environmental studies in the area of lime-based materials, there is a clear lack of background data and inventory data required to model the component mixes, which in the end leads to the impossibility of comparing results in the field.

This research work aims at establishing a clear and robust reference for the environmental and economic impact assessment of producing lime-based mortars, renders and plasters. For doing so, careful attention was paid to the calculation of a reliable Cradle-to-Gate life-cycle inventory, based on a real plant in Germany. This research benefits from being carried out under the SUBLime network (a MSCA, ETN-ITN project) which unites not only six European Universities that are expert in lime-based materials, but also the most important producers of lime-based materials in Europe.

A great deal of effort has been put into analysing and selecting the most reliable existent datasets to model the background data, which has been validated under the network and can be used by interested parties for their own LCAs. For the first time in the literature, reliable average mix designs used in industrial production are disclosed and used for scenario analysis in life cycle assessment and life cycle costing. With respect to the economic assessment, the tax implications of CO_2 emissions in the context of the European Trading System were also included. In general, it is expected that the present research article contributes to the comprehension of two major aspects of sustainability, i.e., environmental, and economic, applied to lime-based construction materials manufacturing, while supporting producers and stakeholders to identify, through a real-life case study, potential hotspots for optimizing their own LCA scenarios.

4.2 Methodology

In this work two of the three main sustainability pillars are addressed, i.e., Environmental and Economic [32]. Regarding the first one, the LCA methodology (based on the ISO 14040/44 (2006)) is used to quantify and compare the environmental impact of manufacturing the Cradle-to-Gate mortars, renders and plasters along with a sensitivity analysis on various parameters. [33][34]. In addition, the economic aspect is addressed through LCC, and is carried out in parallel to the LCA using the same FU, system boundaries and inventory, including information about the variety of costs [35].

4.2.1Goal and Scope Definition

The aim of this study is to quantify the impact of the production of 1 ton of a dry mortar/render/plaster (considered as functional unit) on the environment, from the cradle (i.e., raw materials acquisition) to the gate of the factory, establishing the current baseline scenario of these materials. [36]. A specific set of parameters for each material is selected and used for the sensitivity analysis. The results of the LCA should help to identify the most critical parameters for the production stage and may serve as the most comprehensive analysis of lime-based materials, against which future technologies and developments in this area can be compared.

4.2.2Inventory Analysis

The inventory analysis is a critical phase of the environmental assessment, as the obtained results directly link to the quality of the data used in the LCA [25]. To build a LCI of the reference scenarios, the used raw materials, manufacturing plant (devices and streams) as well as the different considerations was based on the operation of a real plant, including the practical knowledge of the industrial partners within the EU SUBLime consortium. Where this was not possible, the field research was complemented by calculations, model estimations, and expert opinions to complete and validate the inventory data for materials, energy, and costs. The database used for modelling the processes is EcoInvent V3.6, which is the most complete one available for the construction sector [24], [37].

4.2.3Environmental Life-cycle and Life-cycle Cost Assessment

The OpenLCA software was used to run the proposed calculations. The approach followed is consistent with an attributional LCA, where the inputs and outputs were attributed to the FU of the system by linking the unit processes of a system under an allocation procedure [17], [25], [38]. With regards to the Environmental analysis, Impact 2002+ was selected as impact method since it addresses relevant impact categories of importance in the mining industry, such as Resources, Climate Change, Human Health, and Ecosystem quality [39]. The midpoint indicators analysed in this work are: Global Warming, Mineral Extraction, Non-renewable Energy, Terrestrial Ecotoxicity, Aquatic Ecotoxicity, Ozone Layer Depletion and Carcinogens.

Furthermore, with respect to the Economic assessment, the Life Cycle Cost methodology is not standardized as LCA and therefore, there is no unified procedure for calculating the costs[35]. To carry out this analysis, the starting point was the inventory of materials and energy considered for the environmental analysis (Section 4.2.2). The calculations are performed from the producer's perspective, as the main representative actor in the life-cycle, aiming at providing an estimate of the possible production costs and costs of pollutant and emissions. For production costs it is enough to consider the purchase price of materials, resources, and energy [35], [40]. In this research the data related to cost was determined in 2022 through market survey and company perspectives.

It is important to note that efforts have been made to make the data as representative as possible for reality. However, in addition to quantification, the main aim of the work is to provide information on the role played by the various components of the mortars, renders and plasters studied, to identify their effect on environmental and economic indicators. This is key to be able to propose more sustainable alternatives, both in production processes and in products incorporated in recipes.

4.3 Life-cycle Inventory of Lime-based Mortars, Renders, and Plasters

Figure 4.1 represents the flow diagram of the studied plant that produces lime-based renders and plasters (LBMRP).

To build the LCI, as a first step the average composition of the lime-based materials was investigated. Here, a distinction between basic compounds, and agents and additives are made. The materials in the first category are usually present, while the second group are sometimes incorporated to achieve a special property. Commonly, depending on the use of the product (e.g., render or plaster), the mix design will change.

Table 4.1 summarize ranges of variations and proportions, according to a survey performed among producers and different reviewed EPDs of lime-based renders and plasters [41]–[44]. The inventories and datasets used in this study have been thoroughly analysed and verified by the expert team of SUBLime, to ensure the representativeness of the results.

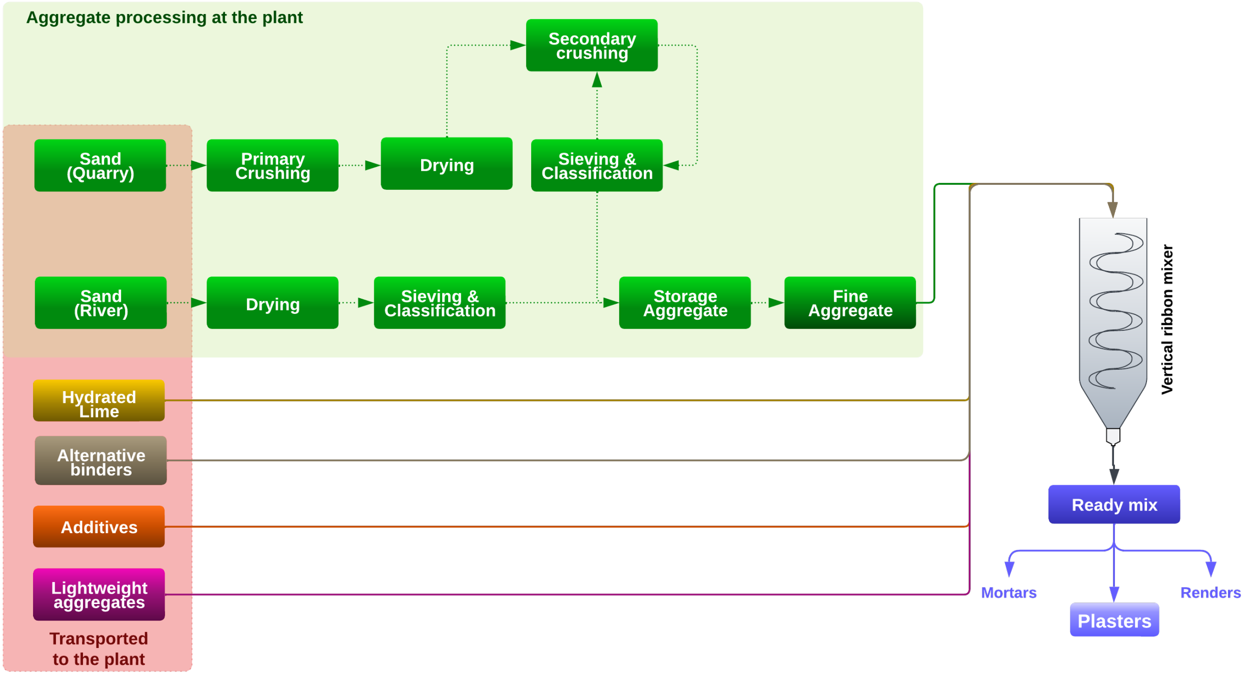

Figure 4.1. Flow diagram of the Cradle-to-Gate dry lime-based mortars, renders and plasters production

4.3.1 Main Consideration for Modelling Raw Materials Transported to Plant

The LCI to produce Hydrated Lime is based on a previous work [30]. With respect to cement, the production of this material is modelled by Ecoinvent 3.6 process “cement production, alternative constituents 6-20%, Europe without Switzerland” from Cradle-to-Gate. Moreover, for the sensitivity analysis, the use of Cement Type III (“cement production, blast furnace slag 66-80%, Europe without Switzerland”) will also be considered. The production of Metakaolin was modelled considering an extraction and calcination process based on [45], using the fuel mix in Germany 2020 as energy source, as reported in [30] (instead of charcoal as reported by the authors [45]).

With regard to the production of aggregates in Germany, for both the coarse aggregates and fine aggregates, it was considered that the sources are 53% from quarry (SQ) and 47% from the river (SR) [46]. Both sources undergo different processes (such as sorting and cleaning) before entering the sand treatment process at the mortar plant (drying, crushing, and sorting), therefore the LCI must address them separately at first [47]. To account for the production of sand from the quarry, the Ecoinvent process “gravel and sand quarry operation, CH” was used. This dataset corresponds to the production of a total of 1 kg of sand (35%) and gravel (65%), and a mass allocation was applied to account only for the sand production. With regards to the SR, the Ecoinvent 3.6 dataset “sand quarry operation, extraction from riverbed, GLO” was used to produce 1 kg of sand from the river.

Table 4.1. Components of typical lime-based mortar, render and plaster and ranges of variation

	Compound	Range (%wt.)	Comment
Aggregates	Coarse Aggregate	50-70	Natural sands, which contain natural minor and trace minerals along with the main minerals quartz (SiO_2) or calcite ($CaCO_3$)
	Fine aggregate	15-65	Limestone dusts which arise because of the preparation of natural sand to produce aggregates, as well as very fine sands.
	Mineral Lightweight aggregate	0-14	Natural or artificial inorganic lightweight aggregates to reduce the dry bulk density. Can be natural (pumice) or artificial (perlite).
	Polymer-based lightweight aggregate	0-1	Obtained by foaming manufactured organic, expanded polystyrene in spherical or particle form for reduction of the dry bulk density
Binders	Cement	0-22	As per EN 197-1 cement is used as a binder and is mainly made from limestone, marl or a mixture of limestone and clay.
	Hydrated Lime	5-20	As per EN 459 hydrated lime is used as binder and is made by treating at high temperature natural limestone, followed by a slaking process.
Additives*	Synthetic dispersion	<2.5	Polymer powder for improving the adhesive bond, elasticity, mechanical properties, etc. in thin-bed mortar.
	Water retention agent	<0.20	Cellulose ether, made from cellulose, which prevents dehydration from occurring in the fresh mortar too quickly.
	Air entraining admixture	<0.02	These reduce the bulk density of fresh mortar, improve workability, and reduce the tendency of contraction and stress cracking.
	Mineral pigments	<0.5	Mineral or synthetic powder-form colouring materials which are produced by mechanical processing of the relevant mineral substances such as chalk, clay, etc.
	Hydrophobic agent	<0.3	Water-soluble sodium oleate or zinc and calcium stearate for reducing the capillary absorption of the solid mortar.

**Ranges of concentration with respect to the total amount of binder*

Mineral Lightweight aggregates are broadly classified according to their origin as natural (pumice, diatomite, volcanic cinders, etc.) or artificial (perlite, clay, sintered fly ash, expanded shale, etc.)[48]. For the modelling of the first group, natural refers to the case in which only a grinding operation is used. Pumice was selected as reference material (Ecoinvent V3.6, “pumice quarry operation, GLO”). The second group comprehends industrially synthesized aggregates, that can even be produced from industrial by-products and wastes [49]. Expanded perlite aggregate (glassy form of rhyolitic or dacitic magma) has been used in constructional elements such as bricks, plaster, renders, etc. because of its low unit weight (driving economic benefits) and as a heat and sound insulator[50], [51]. The modelling of the artificial mineral lightweight aggregate production is done by using the Ecoinvent Process “expanded perlite production, GLO”. The second group of lightweight aggregates includes the polymer-based ones. To represent this case, expanded polystyrene [48] is used. Its production can be modelled by the same database with the process “polystyrene production, expandable, GLO”.

For the modelling of agents and additives, literature research was performed to determine the most common compounds that can give a specific property to the lime-based product. Given that the Ecoinvent database 3.6 has not developed datasets for all of them, the closest available process was selected as a proxy. As water retaining agent, hydroxypropyl methylcellulose and hydroxyethyl methylcellulose have been widely used [27], [52]. In this case the process is modelled by “carboxymethyl cellulose production, powder | APOS, S – RoW”. This material is also used to improve the flow resistance of the mortars, cellulose-based products have been used as thickening agent [53], [54]. As air entrainer, sodium lauryl ether sulphate or sodium alkyl-sulphate or hydroxyl-ethyl cellulose are usually encountered [27]. The AE in this case is modelled by “alkylbenzene sulfonate, linear, petrochemical | APOS, S – RoW”. Stearic and oleic acid, vegetable, and animal fats, as well as different anionic surfactants such as carboxylates, sulfates, sulfonates, and phosphates can be used as hydrophobic agents [55]. The dataset “polycarboxylates production, 40% active substance, RER” from Ecoinvent database 3.6 was used to model the hydrophobic agent production process. When it comes to dispersion agents, several materials can be used, among which ethylene vinyl acetate is a polymer used with great extent in construction materials [56] (Ecoinvent database 3.6, “ethylene vinyl acetate copolymer production, RER”). When the render/plaster requires a special colour, pigments are used. They are defined as colorant particles insoluble in the application medium where the colour, is a generic term for all colouring substances depending on the chemical composition and properties, according to ISO 18451-1:2019. In this case it is considered a white pigment (chalk, “calcium carbonate production, precipitated, GLO”) and a red pigment (Iron oxide, “portafer production, GLO”) [57].

4.3.2 Main Considerations for Modelling the Production Plant

The operations accounted for in modelling the production plant are shown in Figure 4.1. The main raw materials are transported to the factory, and no further processing besides the sand preparation and mixing of the materials is carried out at the plant. Neither storing operation in silos, nor packaging of the product are considered for the environmental impact.

The aggregates are treated separately, according to their origin. For the sand coming from the river, only transportation to the plant, drying and sorting is required. For the sand coming from the quarry, after being transported to the factory it is discharged on an endless screw conveyor, which feeds a jaw crusher (primary crushing step). This operation is modelled by a Jaw Crusher device selected from a catalogue (Power 45 kW). According to the manufacturer, a maximum feed size of 50 mm can be assumed, an output size of 5 mm and 5% of humidity. Then the quarry sand enters the drying process, which aims at reducing the humidity of the sand. The drying process is modelled by the Ecoinvent process “silica sand production, DE” which considers only the heat needed to dry the sand under 1% of humidity. After being dried, the sand goes to a dry air classifier which separates and recovers ultrafine, fine, and coarse materials (same system for both sand types). Given that it is needed to separate very fine fractions of sand, the device modelled is a centrifugal classifier (Power 10 kW) which can separate particles at cut off points between 20 and 100 microns. All particles above 29 mm are redirected to a secondary crushing operation, modelled by a vertical mill (Power 200 kW) which closes the cycle to the centrifugal classifier. All the required fractions of the sand can be obtained from the particle size classification operations. Each fraction is stored in different silos, which are not considered for the environmental assessment. In this case no allocation is attributed to the different fractions of the sand.

The Life-cycle Inventory to produce 1 ton of sand is shown in Appendix A1. At the plant, the components of the mortar, render or plaster are dry-mixed, using a vertical ribbon mixer device for solids (Power 22 kW). The packaging of the final product is not considered in this environmental analysis. An interesting remark, according to the discussion among the SUBLime network, is that even though in this case study the plant is fully powered by electricity, in other plants devices such as the sand drier use natural gas as fuel, which can drastically change the environmental profile, as demonstrated by the research to produce hydrated lime [30]. The modelling of the electricity production corresponds to the case of Germany in 2020 of a previous study [30].

4.3.3 Mortars, Renders, and Plasters: Mix Design and Sensitivity Analysis

The assessment of the environmental impact associated to the manufacturing of different products, has uncertainty on the results, because despite a specific mix design is studied, not all parameters are clearly defined. The goal of this section is to introduce a reference mix design according to the material and a set of relevant parameters of interest (proportion and type of binders, aggregates, transport distances, electricity consumption, etc.), which will serve to determine lower and upper limits for the impact indicators.

In the first place it is necessary to define the base reference cases. Table 4.2 to 4.4 summarizes LCI of the reference mortar, render and plaster. The compositions were decided based on the information provided in Table 4.1, surveys performed of the lime-based materials producers within the SUBLime project, reviews of EPDs and reference works from authors [13], [20], [28], [58]–[60]. In the case of cement-lime mortar, considering that the mechanical properties are important, it is primarily composed by aggregates and twice as much cement as lime (cement:lime 2:1) in mass. In addition to this, the render is generally used as an external coating of the wall. For renders the amount of cement is lower than for mortars because it is not subjected to the same service requirements, and the amount of HL increases (cement:lime 2:3), providing a smooth finishing and better breathability. Another difference with mortars is that part of the silica coarse aggregate is replaced by a 5% of lightweight aggregate. Finally, in the case of plaster, it is usually used as a protective or decorative coating of interior walls and ceilings. The binder is mostly composed of HL and usually no cement is present. Eventually, in some cases in which it is required that the plaster has a certain compressive strength, other supplementary materials are added. One of these can be metakaolin (up to 7% of the total binder), which due to its pozzolanic activity can provide additional hydraulic properties in an alkaline environment, without compromising the white aesthetic aspect of the plaster [61]. Because of its aesthetic purpose, generally the aggregates are composed of fine particles, from silica sand as well as lightweight aggregates (both natural and artificial).

The reference cases presented above, do not consider the incorporation of alternative aggregates, binders and additives and agents, which are evaluated in the case studies. For each material, key parameters have been identified and scenarios were developed. Tables 4.5 to 4.7 summarize the alternatives considered for mortars, renders, and plasters. Each row of the table corresponds to one case-scenario, and one or more parameters are modified with respect to the reference. A “minus (-)” in the table means that no change is carried out for this specific parameter in the base-case scenario.

Table 4.2. Life-cycle Inventory to produce a reference Mortar (M0)

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
			Mortar Manufacturing			
Input	**Sand aggregate**	0.75	t	0.75	t	LCI Appendix A1
	Cement	0.17	t	0.17	t	Modelled by Ecoinvent (cement production, alternative constituents 6-20%, Europe without Switzerland)
	Transport	17	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.00068	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Hydrated Lime	0.08	t	0.08	t	Modelled by SUBLime LCI [30]
	Transport	8	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.00032	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Dry mixer	4	kWh	4	kWh / t	Electricity mix (SUBLime designed) [30]
Output	**Mortar M0**	1.00	t	1.00	t	Output of the Render Manufacturing
	Emissions					Emissions along the production process of the Plaster Manufacturing

Table 4.3. Life-cycle Inventory to produce a reference Render (R0)

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
			Render Manufacturing			
Input	**Sand aggregate**	0.70	t	0.70	t	LCI Appendix A1
	Lightweight aggregate	0.05	t	0.05	t	Pumice quarry operation, GLO
	Transport	10	t*km	200	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.0002	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Cement	0.1	t	0.1	t	Modelled by Ecoinvent (cement production, alternative constituents 6-20%, Europe without Switzerland)
	Transport	10	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.0004	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Hydrated Lime	0.15	t	0.15	t	Modelled by SUBLime LCI [30]
	Transport	15	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.0006	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Dry mixer	4	kWh	4	kWh / t	Electricity mix (SUBLime designed) [30]
Output	**Render R0**	1.00	t	1.00	t	Output of the Render Manufacturing
	Emissions					Emissions along the production process of the Plaster Manufacturing

Table 4.4. Life-cycle Inventory to produce a reference Plaster (P0)

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
			Plaster Manufacturing			
Input	**Fine aggregate**	0.675	t	0.675	t	LCI Appendix A1
	Lightweight aggregate	0.07	t	0.07	t	Modelled by Ecoinvent (pumice quarry operation, GLO)
	Transport	14	t*km	200	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.0003	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Artificial lightweight aggregate	0.005	t	0.005	t	Modelled by Ecoinvent (expanded perlite production, GLO)
	Transport	1	t*km	200	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.00002	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Hydrated lime	0.25	t	0.25	t	Modelled by SUBLime LCI [30]
	Transport	25	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6| RoW)
	Endless screw conveyor	0.001	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [30]
	Dry mixer	4	kWh	4	kWh / t	Electricity mix (SUBLime designed) [30]
Output	**Plaster P0**	1.00	t	1.00	t	Output of the Plaster Manufacturing
	Emissions					Emissions along the production process of the Render Manufacturing

Table 4.5. Parameters considered in the sensitivity analysis of mortar's manufacturing

Parameters	C (%)	C_{Type} (-)	C_{TDis} (km)	HL_{TDis} (km)	A_{TDis} (km)	WRA (%)	AE (%)	Comments
M0	17	CEM II	100	100	-	-	-	Reference scenario (Table 4.2)
M1 (Values in % of total dry mortar mass composition)	25	-	-	-	-	-	-	Effect of replacing completely the HL by Cement.
M2 (Same amount of cement)	-	CEM III	-	-	-	-	-	Effect of changing the Cement Type.
M3 (Distance increase from 100 to 300 km)	-	-	300	-	-	-	-	Effect of an increase in the transport distance of the cement.
M4 (Distance decrease from 100 to 0 km)	-	-	-	0	-	-	-	Effect of no transport distance of HL (integrated plant).
M5 (Values in % of mass, with respect to the total binder content)	-	-	-	-	250	0.20	0.02	Effect of the maximum amounts of aids and additives in the mix.
M6 (Distance increase from 250 to 500 km)	-	-	-	-	500	0.20	0.02	M6 and increased transport distance of the Aids and Additives.

HL: Hydrated Lime, C: Cement, CType: Cement Type, CTDis: Transportation distance of cement, HLTDis Transportation distance of Hydrated Lime, ATDis Transportation distance of Additives, WRA: Water Retention Agent, AE: Air Entrainer.

Table 4.6. Parameters considered in the sensitivity analysis of render's manufacturing

Parameters	**HL (%)**	**LWA (%)**	**C_{TDis} (km)**	**HL_{TDis} (km)**	**A_{TDIS} (km)**	**DA (%)**	**WRA (%)**	**AE (%)**	**HA (%)**	**Comments**
R0	15	5	100	100	-	-	-	-	-	Reference scenario (Table 4.3)
R1 (Values in % of total dry mortar)	20	-	-	-	-	-	-	-	-	Effect of increasing the % of HL, replacing the cement.
R2 (Values in % of total dry mortar composition)	-	14	-	-	-	-	-	-	-	Effect of increasing % of LWA by replacing the sand .
R3 (Distance increase from 100 to 300 km)	-	-	300	-	-	-	-	-	-	Effect of an increase in the transport distance of the cement.
R4 (Distance decrease from 100 to 0 km)	-	-	-	0	-	-	-	-	-	Effect of no transport distance of HL (integrated plant).
R5 (Values in %, with respect to the total binder content)	-	-	-	-	250	2.5	0.20	0.02	0.3	Effect of the maximum amounts of aids and additives in the mix.
R6 (Distance increase from 250 to 500 km)	-	-	-	-	500	2.5	0.20	0.02	0.3	R5 and increased transport distance of the Aids and Additives.

HL: Hydrated Lime, LWA: Lightweight Aggregate, C_{TDis}: Transportation distance of cement, HL_{TDis} Transportation distance of Hydrated Lime, A_{TDis} Transportation distance of Additives, WRA: Water Retention Agent, AE: Air Entrainer, HA:Hydrophobic Agent.

Table 4.7. Parameters considered in the sensitivity analysis of plaster’s manufacturing

Parameters	**ALWA (%)**	**LWA (%)**	**MK (%)**	**HL$_{TDis}$ (km)**	**A$_{TDis}$ (km)**	**DA (%)**	**WRA (%)**	**AE (%)**	**P (%)**	**HA (%)**	**Comments**
P0	0.5	7	-	100	-	-	-	-	-	-	Reference scenario (Table 4.4)
P1 (HL replaced by MK)	-	-	7								Effect of replacing HL by MK as binder
P2 (Same % as P0, different ALA).	0.5	-	-	-	-	-	-	-	-	-	Effect of replacing perlite by polystyrene as ALWA
P3 (LWA from 7 to 14%).	-	14	-	-	-	-	-	-	-	-	Effect of increasing the LWA in replacement of the fine aggregate
P4 (Distance decrease from 100 to 0 km)	-	-	-	0	-	-	-	-	-	-	Effect of no transport distance of HL (integrated plant)
P5 (Values in %, with respect to the total binder content)	-	-	-	-	250	2.5	0.20	0.02	0.5	0.3	Effect of the maximum amounts of aids and additives in the mix, white pigment
P6 (Values in %, with respect to the total binder content)	-	-	-	-	250	2.5	0.20	0.02	0.5	0.3	P4 and red pigment instead of white pigment
P7 (Distance increase from 250 to 500 km)	-	-	-	-	500	2.5	0.20	0.02	0.5	0.3	P5 and increased transport distance of the Aids and Additives

HL: Hydrated Lime, ALWA: Artificial Light Weight Aggregate, LWA: Light Weight Aggregate, MK: Metakaolin, HL$_{TDis}$ Transportation distance of Hydrated Lime, A$_{Tdis}$ Transportation distance of Additives, DA: Dispersion Agent, WRA: Water Retention Agent, AE: Air Entrainer, P: Pigment, HA: Hydrophobic Agent.

4.3.4 Life-cycle Cost Inventory

With regard to the economic costs, a combined criterion on the recommendations proposed in the Life-cycle Cost manual published by NIST and by the SETAC Life Cycle Cost code of practice were followed [62], [63]. Three main categories of costs can be generalized:

(a) Direct production costs (raw materials, energy purchase, etc.).

(b) Indirect cost (treatment of industrial residues, etc.); and

(c) Externalities (associated to taxes for emitting a certain pollutant, such as CO_2).

The total costs are usually the sum of the initial costs, the operating costs, and the end-of-life costs. In this study, and also for the LCC calculation, direct production costs and externalities are considered.

With respect to the direct costs, in this study the focus is on variable costs (those which change with a variation of the production), excluding fixed costs which are constant regardless the level of production (e.g., initial costs, purchase of equipment, salaries, total taxes, etc.). The following variable production cost elements are included:

- Purchase of raw materials transported to the plant (for the reference mixes and the mixes employed for the scenario analysis).
- Transportation costs associated to the purchase of raw materials.
- Electricity consumption in the mixing plant (all devices included in the system boundaries).

Table 4.8 details the costs per unit of the aforementioned items, based on market survey in Germany in 2022. In the case of the hydrated lime and cement, only the production costs are reported (i.e., carbon taxes are not included).

For the calculation of externalities, in this study the allowance prices for EU emissions are considered, in the context of the Trading System [64]. Under this system, a price on carbon emissions is set, making CO_2 pollution less attractive for regulated firms. These prices are the result of a very complex interaction of policies which range from explicit pricing (carbon taxes, internal carbon fees, crediting mechanisms) to implicit pricing (fossil fuel energy taxes, renewable energy support measures, etc.) [65].

Table 4.8. Cost inventory of the mortars, renders and plasters

Group		Item	Unit of measure	Unit costs
Purchase of Materials	**Binders**	Hydrated Lime	€/kg	0.11
		Cement type II	€/kg	0.07
		Cement Type III	€/kg	0.06
		Metakaolin	€/kg	0.15
	Aggregates	Sand	€/kg	0.01
		Pumice (LWA)	€/kg	0.06
		Expanded perlite (PLA)	€/kg	1.77
		Polystyrene	€/kg	1.29
	Additives	CMC	€/kg	0.97
		ABS	€/kg	0.80
		PCX	€/kg	0.65
		EVA	€/kg	1.12
		CC	€/kg	0.45
		Iron oxide	€/kg	0.01
Transportation		Truck transportation	€/t.km	0.06
Electricity consumption		Electricity	€/kWh	0.11

In recent years the price of emission allowances has increased from below 10 € per metric ton of carbon, to above 90 € since the beginning of 2018 [65], [66]. This context shows a high fragility on the long-term robustness of the economic assessment results. However, it is still very relevant to incorporate this parameter, by not pursuing an exact quantification but rather an analysis of the relative contribution of the carbon pricing in the total cost.

As a mean of comparison and to evaluate the effect of the carbon pricing on the total cost assessment, the two values reported by the European Central Bank for 2018 (10 € per metric ton of carbon) and 2022 (90 € per metric ton of carbon) are considered in the present LCC calculation [66]. To include these costs, the specific emissions calculated through the LCA of producing the corresponding binder (cement and/or hydrated lime) were multiplied by the corresponding tax coefficient.

4.4 Life-cycle Environmental Impact Assessment

In this section it is presented the results of the LCIA of the reference scenarios and the case studies in two separate parts namely Reference scenario (Section 4.4.1) and Case studies (Section 4.4.2). In order to ease the readability, in Section 4.4.2 only the percentual difference referred to the corresponding reference scenario is shown. However, the reader is referred to the Appendixes A2 to A4 where the full set of results is provided.

With regards to the manufacturing plant, to have a better understanding of the sharing in the magnitude of the impact indicator, six categories have been created: Cement Production, Hydrated Lime Production, Sand Production, Additives & Aids Production, Total Transportation and Plant Operation. The first four categories refer to the production of the materials from the cradle to the gate of the corresponding production plant (e.g., cement plant). An important remark is that Sand Production involves the transportation of the sand to the plant and its preparation on-site. Furthermore, Total Transportation summarizes all the transportations of the rest of the raw materials to the plant and Plant Operation refers to the electricity required to operate the plant. For a better understanding, the analysis of the indicators is done by considering the areas of protection (endpoint damage categories) of the Impact 2002+ method [39].

4.4.1 Reference Scenario

Figures 4.2 and 4.3 show the results of the midpoint and endpoint indicators for the reference scenario of the lime-based mortar (M0), render (R0) and plaster (P0).

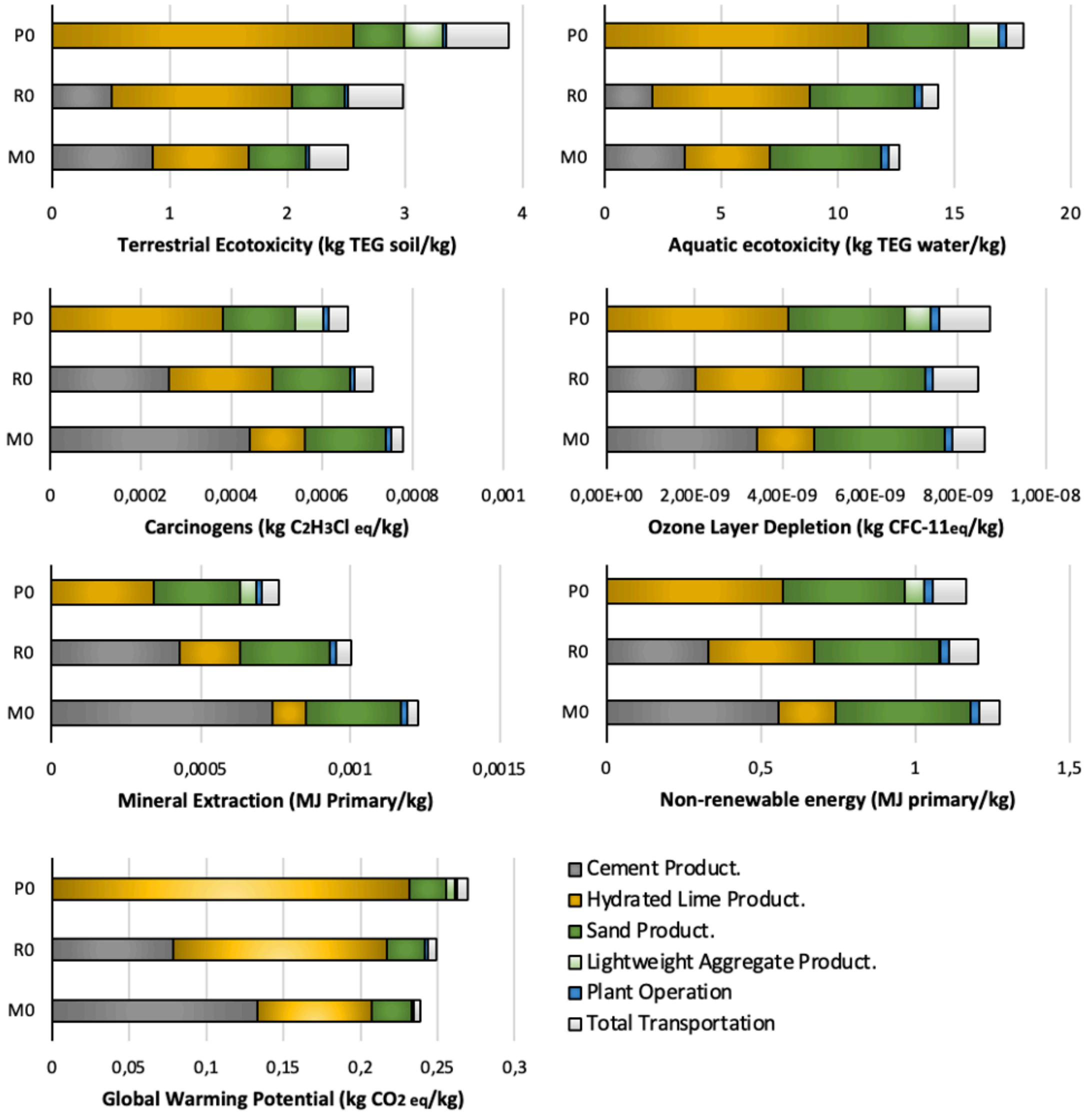

Figure 4.2. Midpoint indicators results for mortars, renders and plasters (total values per kg of material)

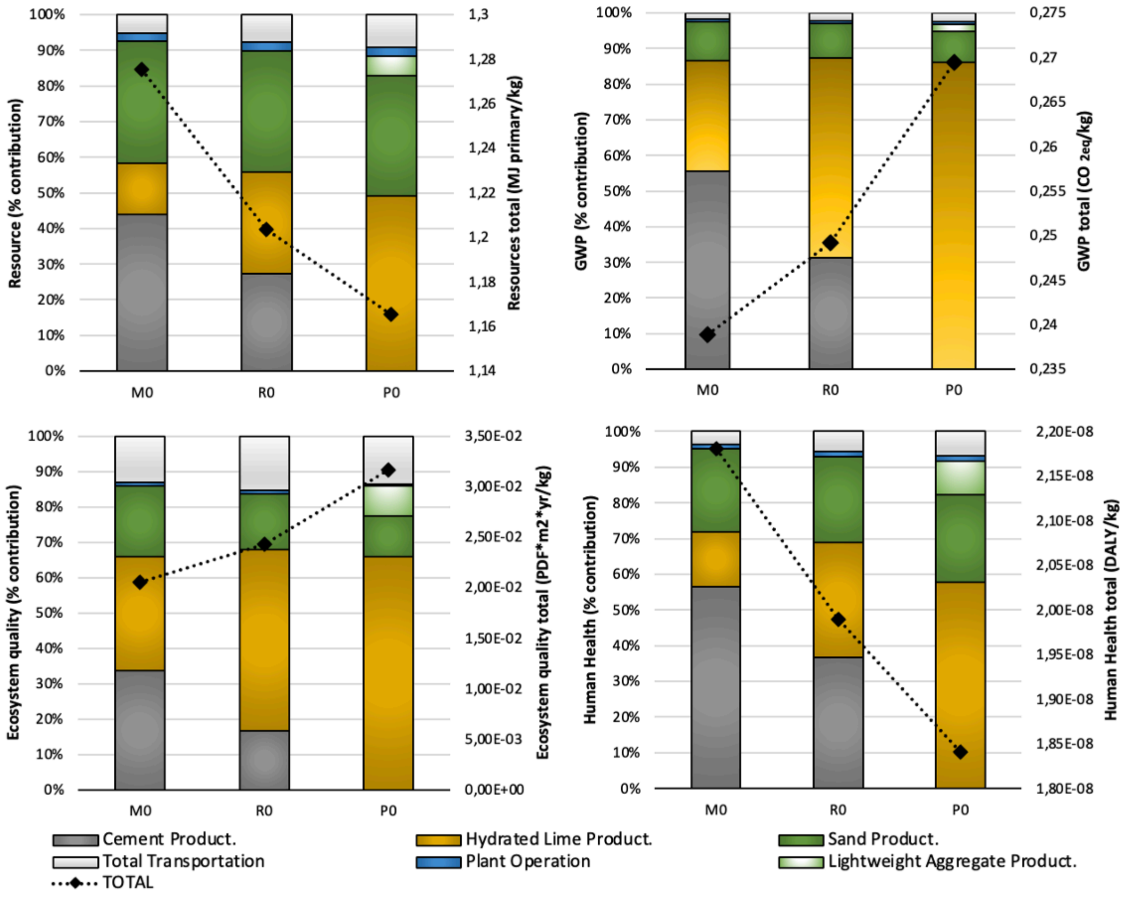

Figure 4.3. Endpoint indicators results for mortars, renders and plasters (% contribution and total values per kg of material)

4.4.1.1 Climate Change Category

To assess the potential contribution to Climate Change, the GWP indicator is often used. This midpoint indicator is measured by the Impact 2002+ method, considering the kg of CO_{2eq} emitted into air during a life cycle, that can be linked to the potential increase of 1.5°C above temperatures in the pre-industrial period [67]. In the Impact 2002+ method, the damage category “Climate change” is the same category as the midpoint indicator “Global Warming Potential”, therefore the last one is discussed hereafter.

According to the midpoint and endpoint indicators provided in Figure 4.2, between 0.17-0.26 kg CO_{2eq}. per kg of dry mortar are generated, depending on the analysed scenario. The order of magnitude of these results agrees with a recent study done by Diaz-Basteris et al. [20]. In all the cases, the binder production (i.e., cement and hydrated lime) dominates the indicator (around 80%). The mix-specific CO_2 contribution of HL (0.98 kgCO_2/kg HL) is higher than Cement Type II (0.76 kg CO_2/kg Cement) [68] [20]. Lime renders contain a lower amount of cement in the mix in comparison to mortars. As the proportion of HL in a mix increases, so does the GWP indicator for the renders (Figure 4.3). In this case the values range from 0.25 to 0.29 kg CO_{2eq} per kg of dry render, according to the specific case analysed. Due to its esthetical coating functionality, plasters do not contain cement as a binder (P0). In terms of binder production, HL dominates the GWP indicator and contributes up to 80%.

4.4.1.2 Resource Category

For the Non-renewable energy midpoint, it is interesting to note that even though the binder production usually leads to the highest consumption of non-renewable energy (because of the Kiln operation), the Sand Production has a significant influence as well (around 25%). Several reasons can be considered for this result. First, 75% of the material is composed of sand aggregate, which turns into a high demand of the material (Table 4.4). Second, the amount of energy required for the sand preparation is relatively high (around 70 kW/t), mainly due to the drying process (See Appendix A1). Last, it needs to be considered that the supply source of electricity in Germany is for around 60% composed of non-renewable sources including coal, natural gas, and nuclear energy [69]. However, the binder production (Cement + HL) has a higher specific contribution: 3 MJ primary/kg binder against 0.33 MJ primary/kg Sand.

In this case transportation operation is relevant, because of the solid fossil fuels burned in the lorry. As main contributors Crude Oil (45.8 MJ primary/kg Crude Oil), Natural Gas (38.30 MJ $_{\text{Primary}}$/m^3 Natural Gas) and Hard Coal (19.10 MJ Primary/kg Hard Coal) are highlighted. In the case of the mineral extraction indicator, for the mortars the relative contribution of HL was around 8% (cement:lime ratio of 2:1). As the amount of HL in the render's mix (cement:lime ratio 1:4) increased, the total contribution of the HL process increases around 3 times. In general, the values for plasters are lower than for mortars and renders, because they do not include cement and the cement production process has a higher specific intensity than the HL production.

In the endpoint Resources indicator (Fig. 4.3), the tendency previously described of the reduction of the indicator with the increase of HL in the mix is confirmed. In general, the binders can contribute up to 65%, followed by the sand production. This shows the importance of switching to alternative energy sources.

4.4.1.3 Human Health Category

Regarding the Carcinogens indicator, a reduction in the amount of lime leads to a reduction in the indicator, because the cement production has a higher process intensity (0.00258 kg $C_2H_3Cl_{eq}$/kg of cement) than the production of hydrated lime (0.00151 kg $C_2H_3Cl_{eq}$/kg of HL). The production of the lightweight aggregate has a significative contribution as well, despite the low quantities used (P0). Furthermore, in the Ozone Layer Depletion indicator, air emissions coming from the combustion of fossil fuels used for electricity and heat production within the system boundaries, are the main components.

The principal contributor is methane in different molecular configurations: as Bromo trifluoro bromochlorodifluoromethane and dichlorofluoromethane Natural gas utilization is a robust contributor to the release of these species along with NO_2 emissions [70] [71] [72]. This also explains why cement has a higher process contribution than hydrated lime (sintering temperature around 1400°C against around 1000°C for hydrated lime). There is an interesting influence of the sand production in all cases (around 40%), because of the high electricity consumption and technology used in the process. In the electricity production, emissions from natural gas in a power plant (mainly chlorodifluoromethane and Bromo difluoromethane) are the main agents.

When considering the endpoint Human Health damage category (Fig. 4.3), the binders contribution ranges from 70 to 60% and a reduction in the cement content leads also to a reduction in the indicator.

4.4.1.4 Ecosystem Quality Category

In the Terrestrial Ecotoxicity indicator, a significant share of the transportation is recorded, which can be assigned to the metals (mainly 65% Zinc, 17% Copper and 15% Aluminum) released during the truck transportation, contributing from 8 to 19% [73], [74]. An increase of 9% of HL in the mix (M0 to R0), leads to an increment of the impact for the reference case with around 20% (Figure 4.2). An analysis of the inventory reveals two main causes for the impact of HL production: one is the combustion of fuels (55% of the relative contribution) because of the use of hard coal, while the other is the blasting operation to obtain the limestone from the quarry (45% of the relative contribution). Because of this fact, for plasters (Figure 4.3) an increment of 37.5% and 25% is recorded in comparison to Mortars and Renders respectively.

In the aquatic ecotoxicity indicator, the contribution of the sand preparation is quite significant, and here again, the same reasons can be given as in the case of the non-renewable energy indicator. The energy matrix to produce electricity for the sand preparation plays a key role, where the main contributor are the emissions during coal combustion in the power plants [75]. The process intensity of hydrated lime is higher than cement. Consequently, increments of 33.3% and 22.2% are recorded for P0 versus M0 and R0 respectively. It is also relevant to mention that for both indicators the production of the lightweight aggregate in the plasters, can contribute almost to the same degree as the production of sand, despite the significant differences in the amount of each component in the mix. For the aquatic ecotoxicity the share of LWA production is even bigger than one of the transportations.

Finally, a rise in the Ecosystem quality indicator is recorded as the proportion of hydrated lime in the mix increases (Fig. 4.3). Transportation plays on this endpoint category the most significant role. Seeing the significant relative weight of the fuel and electricity mix on the binder production, the importance of switching to eco friendlier energy sources to obtain a balanced reduction of the indicators must be stressed once more [30].

4.4.2Case Studies

In this section, the results of the case studies described in Section 4.3.3 for mortars, renders and plasters is presented. The effect on the indicators is calculated as the relative percentual difference relative to their base case (i.e., (Mi-M0)*100%/M0, i= 1, 2, etc.) in Tables 4.9, 4.10 and 4.11.

In these Tables a colour code was adopted to ease the interpretation. If the absolute relative difference is equal to or lower than 5%, the indicator is considered not sensitive (green colour). For ranges between 5% to 50% it is considered sensitive (orange colour) and above 50% it is considered highly sensitive (red colour). The proportion of sensitive indicators is reported as well.

4.4.2.1 Lime-based Mortars

Following Table 4.9 and as discussed previously, the CO_2 intensity of the hydrated lime process is higher than cement type II production. Therefore, a total replacement of hydrated lime by cement in the mix decreases the magnitude of the indicator in comparison to (M1/M0). Furthermore, a reduction of 30% is obtained when Cement type II is replaced by Cement type III (M2/M0), due to the lower CO_2 emissions associated to the reduction of clinker by Blast Furnace Slag as supplementary cementitious material [68].

Table 4.9. Relative percentual difference of each case study for mortars referred to the base case

		M1/M0	M2/M0	M3/M0	M4/M0	M5/M0	M6/M0
Midpoint Indicators	ME	19%	260%	4%	-1%	39%	41%
	NRE	6%	-8%	11%	-2%	28%	31%
	CG	10%	1%	5%	-1%	61%	63%
	OLD	3%	-9%	12%	-3%	41%	42%
	TE	-17%	23%	18%	-4%	24%	30%
	AE	-16%	-14%	5%	-1%	29%	32%
End Point Indicators	GWP/CC	-5%	-29%	2%	-1%	9%	10%
	R	-6%	7%	6%	-1%	24%	26%
	HH	1%	1%	6%	-1%	22%	27%
	EQ	-16%	1%	18%	-4%	32%	37%
Proportion of sensitive indicators		7/10	7/10	6/10	0/10	10/10	10/10

Table 4.10. Relative percentual difference of each case study for renders referred to the base case

		R1/R0	R2/R0	R3/R0	R4/R0	R5/R0	R6/R0
Midpoint	ME	-14%	1%	3%	-2%	88%	89%
	NRE	-4%	1%	13%	-3%	69%	70%
	CG	-8%	1%	3%	-2%	203%	203%
	OLD	-2%	1%	18%	-5%	45%	45%
	TE	9%	1%	12%	-7%	37%	38%
	AE	9%	1%	3%	-2%	33%	33%
End Point	GWP/CC	3%	1%	1%	-1%	14%	14%
	R	-4%	1%	4%	-3%	68%	69%
	HH	-8%	1%	3%	-2%	203%	203%
	EQ	9%	0%	9%	-7%	37%	38%
Proportion of sensitive indicators		6/10	0/10	3/10	2/10	10/10	10/10

Table 4.11. Relative percentual difference of each case study for plasters referred to the base case

		P1/P0	P2/P0	P3/P0	P4/P0	P5/P0	P6/P0	P7/P0
Midpoint	ME	5%	-1%	-1%	-5%	111%	113%	112%
	NRE	3%	33%	0%	-6%	73%	71%	74%
	CG	5%	71%	-9%	-13%	193%	190%	194%
	OLD	1%	-3%	2%	-8%	45%	44%	47%
	TE	-2%	2%	-1%	-2%	26%	25%	27%
	AE	0%	2%	-1%	-3%	28%	27%	28%
End Point	GWP/CC	-5%	3%	0%	-2%	14%	13%	14%
	R	2%	33%	0%	-6%	73%	71%	74%
	HH	10%	71%	1%	-4%	224%	220%	224%
	EQ	-1%	2%	-1%	-2%	27%	26%	27%
Proportion of sensitive indicators		1/10	4/10	1/10	4/10	10/10	10/10	10/10

Moreover, additives added to a mix may cause an increase up to 9% of the indicator, even though their proportion in a mix is lower than 3% of the total amount of binder (i.e., 3% of 25%), showing that any particular addition should be treated carefully, even when this effect is disregarded in several articles [20]. It is shown that in general transportation does not produce significant changes to the GWP because the transportation mean is modelled by a Euro 6 lorry which is the most efficient way of transportation for lorries in terms of CO_2 emissions and the most up-to-date according to European regulation. However, significant contributions are found to Ecosystem Quality and Resources areas because they also considered the impact of obtaining the fuel required for transportation.

In the case of Mineral Extraction, most remarkable scenario is M2/M0, in which Cement Type II is replaced by Cement Type III. In this case, the indicator increases by 260% with respect to M0 (0,0043 MJ Surplus for Cem Type II against 0,0225 for Cem Type III). This is explained because the latter has between 60 to 80% cement replaced by GGBFS. Same has happened with other SCMs (e.g., Fly Ash), where GGBFS is, as such, no longer considered as waste, but more as a useful by-product, since it meets the requirements of the European Union directive 2008/98/EC that qualifies the by-product status [25], [76]. With the extraction of iron ore also other scarce metals are extracted such as Copper, Zinc and Lead. In both cases the main contributor to the indicator is Copper (Impact factor of 36,7 MJ $_{surplus}$/kg Copper) and the percentual relative difference in the amount of copper in each inventory is around 60%. The amount of mineral Zinc (Impact factor of 4.09 MJ $_{surplus}$/kg Zinc) in each inventory also holds responsibility, with a difference of around 90% between them. Finally, the third main contributor is Lead (Impact factor of 7.35 MJ $_{surplus}$/kg Zinc) and the percentual relative difference between both inventories is around 90%. Digging deeper into GGBFS used as SCM, it comes from the Ecoinvent V 3.6 “pig iron production – RoW” process, in which 0.3 kg of GGBFS as by-product per kg of Iron are generated. The mineral extraction indicator for this process is 0.021 MJ surplus/kg GGBFS which multiplied by 80% replacement in Cem Type III gives a contribution of Slag of 0.016 MJ Surplus/kg Cement. The GGBFS modelled in Ecoinvent and used later as raw material for Cem III production, applies a mass allocation of around 30%, which explains the obtained results.

For the Non-renewable energy midpoint, the transport distances appear to even have a higher significance on this impact indicator, mostly due to an increased cement transport distance (M3, up to 11%). However, the highest increment is accounted for M5/M0 and M6/M0, with the incorporation of additives in the mix (up to 28%) and a duplication in their transport distance respectively. With regard to the Resources endpoint, it is relevant to point out that in all analysed cases (M0 to M6) the share of sand processing is between 20-30% of the damage category (also observed in Section 4.4.1) and is almost equivalent to the share of cement. However, if the proportions in the mortar are considered, the specific contribution of each process is 0.26 MJ $_{Primary}$/kg Sand and 1.17 MJ Primary/kg Cement. Therefore, to reduce this impact category a change in the binder design would have higher impact than a change in the amount of aggregate used.

The carcinogenic indicator is highly sensitive to the additives production, that can contribute to up to 35% of the indicator (61-63% in total including the transport, M5/M0 and M6/M0). The 35% corresponding to the additives, is dominated by the water retention agent production, resulting in the release of aromatic hydrocarbons to the environment, being the main carcinogen agent (70% of the total CMC production). Now moving forward to the Ozone Layer Depletion indicator, air emissions coming from the combustion of fossil fuels used for electricity and heat production within the system boundaries, are the main components. The indicator is also sensitive to the emissions associated to transport (M3/M0) and additives production (M5/M0 and M6/M0) resulting in increments of 12%, 41% and 42% respectively (with respect to M0). When considering the endpoint Human Health damage category, the additives incorporated in M5/M0 and M6/M0 cases significantly increase the magnitude of the indicator (around 27%).

Regarding Terrestrial Ecotoxicity, an increment in the cement transport distance (M3/M0) has the highest impact compared to the rest of the indicators, which can be assigned to the metals (mainly 65% Zinc, 17% Copper and 15% Aluminium) released during the truck transportation [73], [74]. It is also interesting to note that around 17% reduction can be achieved by rising the amount of cement by 8% (M1/M0). The contribution of additives is not negligible (21%) especially when considering the transportation required, the total share can be up to 30%. In the aquatic ecotoxicity indicator around 32% increment is attributed to the additives (20 % additives production and 12% due to its transportation). With regards to the last statement, and from an endpoint level viewpoint (Ecosystem quality), it is important to remark that the transportation plays a significant role, as discussed in Section 4.4.1, along with the use of additives.

4.4.2.2 Lime-based Renders

With respect to the GWP indicator, in general the cases R1 to R4 do not significantly influence the indicator (Table 4.10). However, the GWP indicator increases with up to 14% due to the production of the additives (R5 vs R0). According to the inventory, the biggest contributor (10%) is the CMC, which emits around 3.86 kg $CO_{2\ eq}$/kg of CMC produced. Furthermore, the EVA production is responsible for the 4% extra $CO_{2\ eq}$ emissions (process intensity 2.08 kg $CO_{2\ eq}$/kg of EVA produced).

In terms of Mineral Extraction, as was explained before, cement has a higher process intensity than hydrated lime and therefore, a reduction of 14% is observed for R1/R0 (increment by 20% in HL content). Further, for the cases incorporating additives production (R5/R0 and R6/R0), the indicator increases up to 89% (45% assigned to the additives and the rest to the transportation). The process intensity of the CMC is around 0.11 MJ surplus/kg of CMC and of EVA 0.05 MJ $_{\text{surplus}}$/kg of EVA. The relative contribution of the first one to the Mineral Extraction indicator is up to 30% and for the second one 15%. Considering the Non-renewable energy category, transportation operation starts being more relevant, because of the solid fossil fuels burned in the lorry. As main contributors Crude Oil (45.8 MJ primary/kg Crude Oil), Natural Gas (38.30 MJ Primary/m^3 Natural Gas) and Hard Coal (19.10 MJ Primary/kg Hard Coal) are highlighted. In the extreme case R3/R0 where the distance of cement transportation changes from 100 to 300 km, the increment is around 13%. The tendency previously observed regarding the additives production is shown also in this indicator, where global increments of around 70% are obtained for R5-R6 when compared to the reference scenario R0. The Resources endpoint indicator has shown to be extremely sensitive to the production of the additives and increments of around 70% are expected when they are incorporated in the mix (R5/R0, R6/R0).

For the Carcinogenic indicator, it is only worth mentioning that in cases R5/R0 and R5/R0, with the production of additives, being a source of several organic compound emissions, incorporation of different types of them, even in small quantities, may lead to fundamental changes in the results. For R6/R0, the indicator increases more than 200%, caused by the share of additive production being around 70% for

R6. The influence of each additive is listed as follows (from higher to lower). EVA releases aromatic hydrocarbons during its production (3537.5 kg $C_2H_3Cl_{eq}$/kg of EVA), representing 43% of the carcinogens. It is followed by CMC (20% of the carcinogens), releasing besides aromatic hydrocarbons, arsenic (3.54 E4 kg $C_2H_3Cl_{eq}$ /kg of EVA) and dioxins (1.72 E9 kg $C_2H_3Cl_{eq}$/kg of EVA). Finally, polycarboxylates and ABS contribute in a similar proportion (around 2.5% each), with the release of similar emissions. In the ozone layer depletion indicator, the transportation distance remains being a significant factor which can amount to 18% of the indicator when the transportation distance of cement is duplicated (R3/R0). With respect to the additives production, increments up to 45% are recorded with CMC dominating by far the indicator (around 60% of the total share of the additives). In the endpoint damage category Human Health, a considerable increment in the quantification (more than 2 times the reference) when additives are incorporated in the mix in scenario, showing that this a variable that must be watched when pursuing a balanced reduction of the indicator.

In all analysed cases (R0-R6) the binder production is responsible for between 70 to 50% of the emissions associated to terrestrial ecotoxicity. Transport in total can amount to 12% of the whole indicator when cement transportation distance is increased (R4/R0). The second main contributor is the production of the additives plus its transportation (R5/R0, R6/R0) which can contribute up to 38%, CMC production being the most important (15%). The emissions to soil which contribute to the impact are Aluminium, Zinc and Copper. Similar analysis is valid for aquatic ecotoxicity. In general, for the endpoint indicator Ecosystem Quality, the main contributors remain the binders with special emphasis on hydrated lime production. CMC production followed by transportation process are also relevant variables that need to be considered.

4.4.2.3 Lime-based Plasters

In the Climate Change category, the previous comments for mortars and renders are valid. It is only worth mentioning that a reduction of around 5% is achieved when Hydrated Lime is replaced by Metakaolin, because its specific emissions are lower [30], [77] (Table 4.11). Even though more additives are added to the mix design, their average contribution is still in the order of the analysed case for renders (14 %). Once again, only CMC and EVC contribute significantly.

Regarding the Mineral Extraction indicator as main feature to highlight, plasters incorporate a wide variety of additives and together they can more than double the indicator (P5/P0,P6/P0, P7/P0). In the case of P7/P0, around 60% of the indicator is dominated by the additives production. In particular, the main contributors are CMC (69%), EVC (21%), followed by ABS (7% and process intensity 0.08 MJ primary/kg ABS). None of the pigments used in the plaster contribute significatively to the indicator. In the case of Non-renewable energy, what must be highlighted is the case P2, in which 5% of the perlite LWA is replaced by expanded polystyrene. The first one is a natural lightweight aggregate that does not need an intensive processing, meanwhile the second one is the result of an industrial process with a relevant environmental impact [78]. The results show that this indicator can increase up to 33% due to the use of polystyrene instead of perlite. The inventory reveals that the process intensity of PLYA is around 89 MJ primary/kg of PLYA and for perlite it is 13 $\text{MJ}_{\text{primary}}$/kg of Perlite, around 7 times smaller. In the case of the additives, a similar situation occurs as described for the mineral extraction indicator. For the endpoint indicator Resources, two main variables become important: the type of aggregate (i.e., natural, or artificial lightweight aggregate), where even small proportions of PLYA in the mix can generate significant changes (33%, P2/P0), and the proportion and type of additives, same as described previously.

Regarding the Carcinogens indicator there are two important cases to highlight: P2/P0 and P7/P0. In the first case, it is highly relevant to notice that even a small proportion (0.5%) of the used artificial lightweight aggregate PLYA in replacement of the perlite, has a huge impact on the indicator, representing around 71% of increment. The reason behind is found in the inventory analysis, where there is a high impact factor associated to the release of polycyclic aromatic hydrocarbons (3537 kg $C_2H_3Cl_{eq}$/kg of polystyrene) and dioxins to the air, during the production of expanded polystyrene. The second case (P7/P0) is highlighted because increments of around 200% with respect to the reference case P0 are recorded. In general, the additives production represents 70% of the total indicator (P7/P0). As described for the case of renders, the main contributor is once again EVA production, followed by CMC and lastly with a shared place of importance PLYC and ABS production. Even though additives (in particular, CMC) still play a significant role, their contribution is lower in ozone layer depletion than in the case of carcinogens (P7 shows 45% higher Ozone Layer Depletion than P0). There are two lessons that can be learned from the endpoint indicator Human Health. The first one is that whenever it is possible, the use of organic polymers as lightweight aggregate should be avoided because the indicator is highly sensitive to their addition even in very small quantities (71%, P2/P0). Second, the use of additives has an important impact, due to the emissions during the production process (224%, P7/P0). This shows the importance of moving towards bio-based products to obtain eco-friendlier additives for construction materials [79], [80]. It is fundamental to find alternatives for WRA and DA because the indicators considered have proven to be highly sensitive to them.

Finally, regarding the Ecosystem quality endpoint category, same as for mortars and renders, the binder production remains the most significant process. Seeing the significant relative weight of the fuel and electricity mix on the binder production, the importance of switching to eco friendlier energy sources to obtain a balanced reduction on the indicators must be stressed once more [30]. It is also interesting to see that even though the use of metakaolin can lead to better mechanical properties, it does not have a meaningful contribution to reduce the impacts in this category. However, special attention should be given to the PLYA. The indicator has proven to be sensitive to its proportion, because it has a higher relative process intensity than the sand and therefore, a replacement of the perlite by PLYA can lead to high increments of the indicators. Previous comments about the need for searching for new types of water retention agents, are still valid. To conclude the Life Cycle Impact Assessment section and aiming at summarizing most relevant findings of the case studies, Figure 4.4 shows the proportion of sensitive indicators for different categories according to Tables 4.9 to 4.11 with the relative position of mortars, renders and plasters indicated.

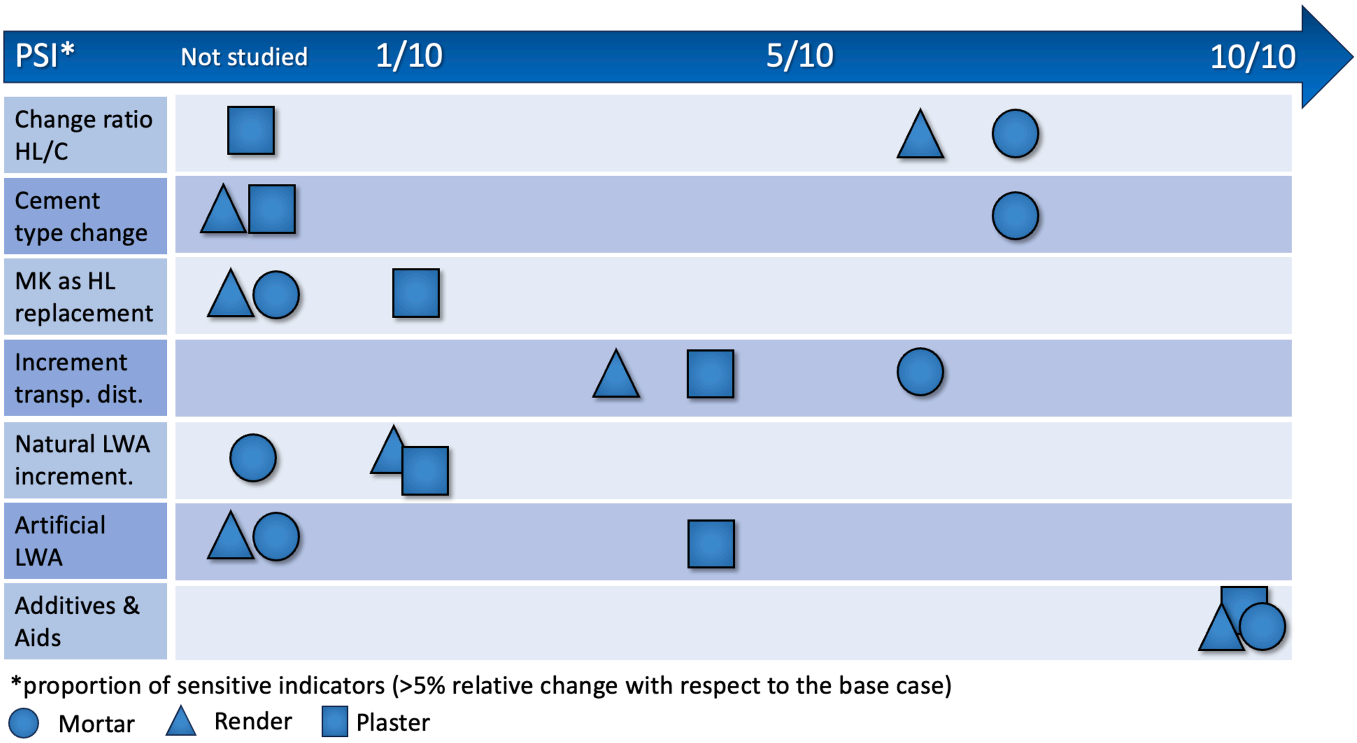

Figure 4.4. Proportion of sensitive indicators for mortars, renders and plasters (>5% relative change with respect to base case)

4.5 Life-cycle Cost Assessment

In Figure 4.5 the results of the economic assessment for mortars, renders and plasters in the scenario (A) with carbon pricing 2018 (10 € per metric ton of carbon) and scenario (B) with carbon pricing 2022 (90 € per metric ton of carbon) are presented. On the left axis the graphs show the relative contribution of the cost components in the overall cost and on the right axis the total cost. All items of the direct cost production associated to the raw material purchase, transportation, plant operation (electricity consumption) as well as the externalities (carbon pricing) have been disaggregated for a better analysis of the relative contribution.

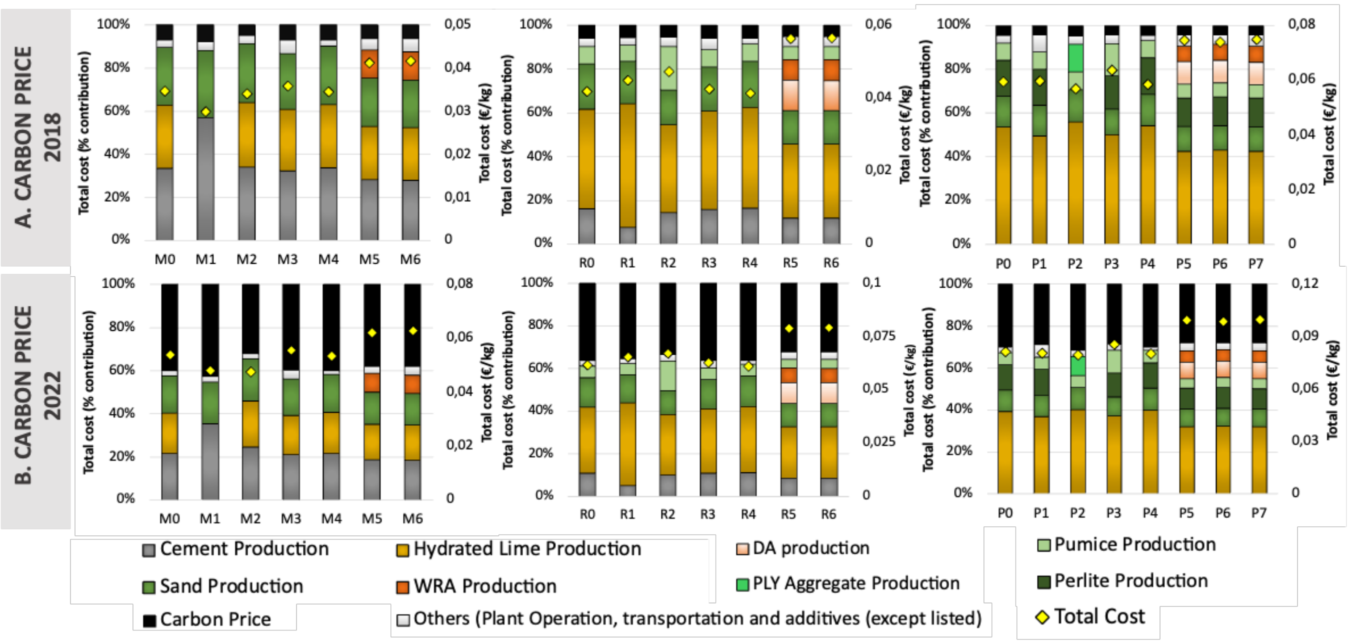

Figure 4.5. Life Cycle Cost Assessment Results for Mortars, Renders, and Plasters considering the effect of carbon pricing in 2018 (A) and in 2022 (B).

When the carbon pricing of 2018 is considered, in general it is observed that its contribution for all analysed cases is around 5-10% of the total cost. With respect to the binder's contribution, for mortars, renders, and plasters, they account for around 50 to 60%. However, as described in the Cost Inventory, the specific cost of the HL production is higher than the cement. Therefore, as the proportion of HL in the mix increases, so does the total cost. Total costs vary between 0.03-0.04 €/kg, 0.05-0.06 €/kg and 0.06-0.08 €/kg for mortars, renders and plasters respectively. The aggregate production process in mortars accounts for around 20-30% of the total costs, meanwhile the production of the additives for around 12%, almost entirely attributed to the WRA production. In the case of renders, it is worth mentioning that the total cost is sensitive to the Pumice production, when part of the sand aggregate is replaced (specific contribution of the total aggregate +13% for R2 compared to R0). The total cost of the additives production is almost the same as the total aggregate cost (20% of the relative contribution). It was found that the contribution of the DA (around 12%) contributes significatively and as much as the WRA, despite the higher concentration of the last one in the mix. Replacing the production of perlite by PLYA slightly lowers the total cost. However, the large environmental impact associated to its production has been discussed previously (Section 4.4).

In scenario B the effect of the sudden change on carbon pricing is evaluated (from 10 to 90€ per metric ton of carbon). The construction materials industry is a business that is highly affected by these taxes, because of the chemical processes taking place in the materials manufacturing. For lime-based materials the carbon pricing is highly relevant, because unlike most carbon intensive industries, most lime emissions are not generated by heat or power generation but by the chemical reaction that occurs when limestone is heated [30]. EuLA as the representative of the lime producers in Europe, in 2017 has published a position paper discussing the abovementioned issue and argued that policies should be developed in the framework of the EU ETS to maintain the integrity of the internal market and ensure a global level playing field[87]. As previously discussed in Section 4.4, from 80 to 90% of the emissions in mortars, renders and plasters are attributable to the binder's production, with a release of up to 0.26 kg $CO_{2\ eq}$/kg of mortar, 0.29 kg $CO_{2\ eq}$/kg of render and 0.32 kg $CO_{2\ eq}$/kg of plaster. Therefore, the total costs for mortars, renders and plasters experience roughly an increment of 24, 26 and 30% respectively. For mortars and renders, up to 40% of the total cost is related to the carbon price. For plasters the carbon pricing share is around 30%, because even though more $CO_{2\ eq}$ is released during its production, the manufacturing of plasters was most expensive of the three materials (almost 40% than for mortars and 20% more than renders).

Even though a more comprehensive and broad analysis of the economic parameters is a must, there are two main relevant outputs of this simple case study. On the one hand, in the building materials business, accounting for externalities is critical in the context of a very turbulent market. Sharp increments in carbon pricing over a very short period (such as 2018-2022) can have a meaningful impact on the overall costs, with the industries not being able to easily modify their production capabilities to quickly rising costs [66].

On the other hand, for a sustainable production it is decisive to consider the three pillars of sustainability: economic, environmental, and social [32]. The results of this study with respect to the first two support the idea that a closer look at the Cradle-to-Gate emissions is key to reduce the environmental impacts and costs. In this context, new technologies such as on-site carbon capture, fuel savings by using highly efficient kiln technologies, improving the use of waste heat, among others, are alternatives being considered in the lime industry [12]. However, as introduced before, the value chain of the construction materials is large and the achievement of these goals requires the commitment of all involved parties, from producers to policy makers and customers.

4.6 Conclusions

Based on the results of the cradle-to-(gate of the factory) environmental and economic life cycle assessment of a real case-study to produce lime-based mortars, renders and plasters, following conclusions could be drawn:

- A detailed Life Cycle Inventory to produce lime-based mortars, renders and plasters has been provided including a wide variety of additives and aids. The inventories and datasets from EcoInvent database V 3.6 used in this study, have been thoroughly analysed and verified by the expert team of SUBLime, to ensure the representativeness of the results and to facilitate the implementation of the inventory by other stakeholders, in their own specific cases.
- Regarding the environmental impact assessment, the results are divided into the following categories:
    - **Climate change.** In terms of Global Warming Potential (GWP) up to 0.26, 0.29 and 0.32 kg $CO_{2\ eq}$ per kg of material is released for mortars, renders, and plasters respectively. This increase from mortars over renders to plasters, is mainly attributed to the fact that the mix-specific contribution of HL (0.98 kg CO2/kg HL) is higher than for the Cement Type II (0.76 kg CO_2/kg Cement). Around 70 to 80% of the GWP is dominated by the binders' production, followed by the coarse aggregate (15-20%) and additives production (10%). Water retention agent (WRA) and synthetic dispersion agent (DA) were the highest contributors in the additive's sharing.
    - **Resources.** A key role in this indicator is played by the binders (25-50% of the indicator) and the coarse aggregate (15-50% of the indicator). The specific contribution of each process is 0.26 MJ Primary/kg Sand and 1.17 MJ Primary/kg Cement-Hydrated Lime. Therefore, a reduction in the specific load of the binder is a must to reduce the indicator. For this purpose, efforts must be put in switching to low carbon energy sources (both for fuels and electricity production). The type and proportion of additives (mainly WRA and DA) contribute significatively to the overall impact regarding the use of resources (around 40%). Transportation distances should be minimized to achieve around 10% reduction of the indicator. In the case of plasters, the type of aggregate becomes critical. Small proportions of polystyrene in replacement of perlite as artificial lightweight aggregate can generate increments up to 33% of the indicator.

  - **Human health.** Whenever possible, the use of organic polymers as lightweight aggregate should be avoided because the indicator is highly sensitive to it (around 50%) even in very small quantities. The use of additives can contribute up to 70% of the indicator and account for as much as three times the sum of the binder and aggregate production. The emissions of polycyclic aromatic hydrocarbons and dioxins during the production process have a very high impact factor. Therefore, it stresses the importance of moving towards bio-based products to obtain eco-friendlier additives in construction materials. It is fundamental to find alternatives for WRA and DA because the indicators are highly sensitive to them.
  - **Ecosystem quality.** For mortars, renders, and plasters the binder remains the most significant (60-80% of the indicator). A significant relative weight of the fuel and electricity mix on the binder production has been found. Therefore, the switch to eco-friendly fuels can also reduce the ecosystem quality indicator, by diminishing the process intensity of the binders and the sand production. Previous comments about the need for searching for new additives for water retention agents, are still valid here.
- The economic assessment accounted for direct costs (raw material purchase, transportation, and plant operation) and externalities (carbon pricing in 2018 and 2022).
  - In general, in 2018 and for mortars, renders and plasters, the total costs are distributed between binders (40-55%), aggregates (25-30%), additives (15%) and carbon pricing (10%). However, the results are severely affected by the sudden change in 2022 of the carbon pricing. From 80 to 90% of the emissions in mortars, renders and plasters are attributable to the binder's production, with a release of up to 0.26 kg $CO_{2\ eq}$/kg of mortar, 0.29 kg $CO_{2\ eq}$/kg of render and 0.32 kg $CO_{2\ eq}$/kg of plaster. Because of this, the total costs for mortars, renders and plasters exhibit an increment of about 24, 26 and 30% respectively.
  - The share of the carbon pricing on the 2022 total cost can be up to 40%. In the building materials business, accounting for externalities is critical in the context of a very turbulent market. The reduction of the emissions can have a direct impact on both environmental and economic sustainability.

Future research lines should explore the effect of the system boundary expansion, considering the use phase (specific properties of the lime-based materials) where potential improvements of the indicators can be achieved by carbon capture during the service life. The unavailability of specific datasets in Ecoinvent needed for this LCI modelling is considered a limitation of the study. Finally, a more comprehensive and broader analysis of the economic parameters would enable a better understanding of the economic sustainability of lime-based materials in a global sense.

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Chapter 5: Applying the Concepts of Circular Economy in Lime-based Rendering Materials

The circular economy strategy studied in this thesis has two approaches (i.e., two parts): closed-loop and open-loop upcycling.

The **closed-loop upcycling** is based on the philosophy of keeping materials in closed cycles (Cradle-to-Cradle) for as long as possible. As an application, the complete recyclability at the end of life of a lime-based render was experimentally addressed, evaluating relevant technical and durability properties of second life renders in comparison with first life renders. The decarbonization potential of the circular economy “Cradle-to-Cradle” against the linear economy “Cradle-to-Grave” is quantified through LCA. The results (Paper 3) are presented in the first part of this chapter.

When by-products from other markets or waste materials are introduced as secondary resources (SR) in a different manufacturing process to produce an added value, the strategy is called **open-loop upcycling**. A Cradle-to-Grave LCA was conducted for the case of lime-based renders manufactured with a by-product from the acetylene industry, carbide lime, and paper mill sludge, a residue from the paper industry. The intermediate treatments for the conditioning of the SR were designed based on the proposed methodology and the results were compared against the traditional scenario. The results (Paper 4) are presented in the second part of the chapter 5.

5.1 Closed-loop Upcycling

Publication 3. Circular Design, Materials Properties, Service Life and Cradle-to-Cradle LCA of Lime-based Materials

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5.1.1 Introduction

Lime, a historic building material, has been widely employed in constructions dating back to ancient times, including ancient Israel (7000 BCE)[1], Syria (4250 BCE)[2], China (2000 BCE), and civilisations like the Mayan, Inca, and Aztec (500 BCE) [3]. Presently, the construction industry, accounts for 18% of sales of lime in Europe (20 Mt/y in 2016 [4]) to manufacture lime-based products and, particularly, lime-based mortars/renders for heritage restoration and modern masonry systems [5]. However, these materials pose challenges to comply with the UN 2050 sustainable development goals [6] because of substantial CO_2 emissions due to lime calcination operation [7], extraction of virgin raw materials, and contributing to significant amounts of construction and demolition waste once the service life has passed [9]. The calcination process, generating 1.2 tonnes of CO_2 per tonne of CaO, is a primary emitter and constituting for about 1% of the global anthropogenic CO_2 ([10]–[12]). Moreover, it is energy-intensive, accounting for 90% of total energy consumption (calcination temperatures around 900 °C) and 99% of the specific impact on global warming in lime production ([13]–[15]). The production of Hydrated

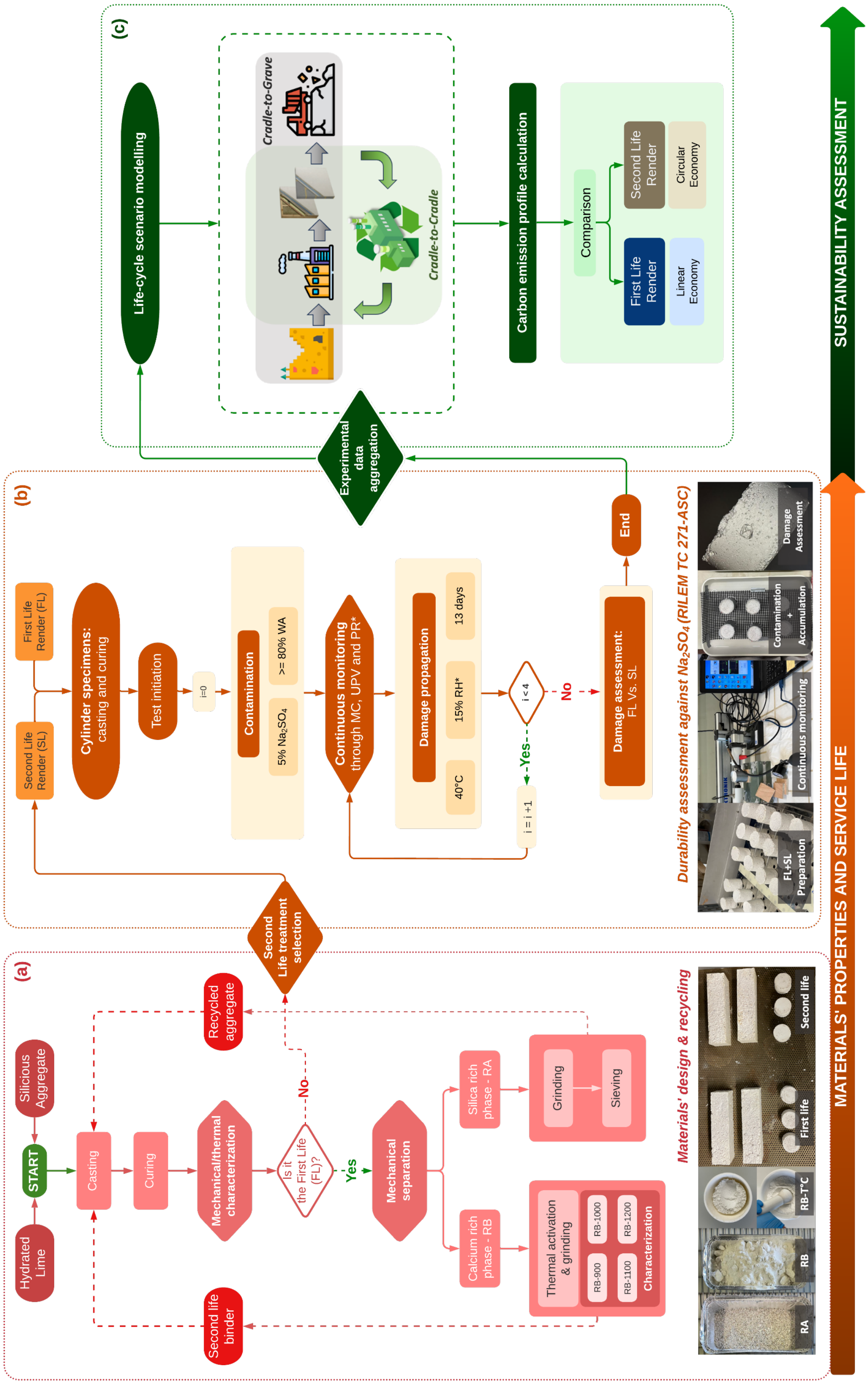
Lime (HL), used in ready mix mortars/renders, involves the slaking of lime. In particular, HL manufacturing can contribute up to 80% of the CO_2 emissions in those ready mixes [7].

In the conventional lifecycle of building materials, products are manufactured, used, and discarded, while contributing to environmental challenges. Embracing a reduce-reuse-recycle (3R) circular economy paradigm offers a solution by transforming waste into valuable resources [9], potentially reducing emissions. One prevalent strategy for minimizing the environmental impact of lime-based mortars and renders involves open upcycling - substituting hydrated lime with various by-products or wastes in the mix. These supplementary materials, including calcined clays, ceramic waste powder, crushed bricks and industrial by-products like fly ash [16]–[19] can reduce virgin raw material extraction and cradle-to-gate CO_2 emissions.

An alternative strategy is a closed-loop upcycling, aiming to maintain materials in closed cycles (Cradle-to-Cradle) for as long as possible [20]. While concrete has seen many recent successes in recycling construction and demolition waste [21]–[26], implementing a similar Cradle-to-Cradle approach for lime-based building materials is underexplored. The present research focuses on assessing the feasibility of producing a fully recycled, second-life lime render through a closed-loop upcycling process, eliminating the need for virgin hydrated lime and aggregate. In the study, the technical materials properties and environmental key-performance indicators of these recycled lime renders are compared, with first-life lime renders made solely from virgin raw materials. By quantifying the benefits of a closed-loop circular economy, this research contributes to understanding the environmental advantages over the traditional linear (cradle to grave) economy scenario.

5.1.2 Experimental Program and Environmental Calculations

Our research showcases a circular design for upcycling second-life lime-based renders, comparing material properties and environmental impact with first-life renders. The interdisciplinary approach integrates materials science to derive essential input parameters for Life Cycle Assessment (LCA) analysis. A general overview incorporating this integrated approach is presented in Figure 5.1. Thus, the methodology of the present research is divided into materials assessment and sustainability assessment. The specific details are addressed in the following sections.



*RA: Recycled Aggregate; RB: Recycled Binder; MC: Mass Change; UPV: Ultrasonic Pulse Velocity; PR: Photographic Record; WA: Water Absorption; RH: Relative Humidity

Figure 5.1. Research methodology to assess materials' properties, service life (a, b) and sustainability performance (c) of upcycled second-life versus first-life renders

5.1.2.1 Materials

The lime-based render investigated in this study is composed of commercial hydrated lime (EN 495) and standard silica sand (EN 196-1). Table 5.1 summarizes the characterization data of the HL: chemical composition obtained by X-ray fluorescence, density according to ASTM C124 and particle size distribution by a LA950V2 (Retzsch) laser granulometry. No additives nor artificial lightweight aggregates were used in this study, to avoid additional effects in the system.

Table 5.1. Chemical composition (mass %) and physical properties of the HL used in this study.

	CaO	SiO_2	Fe_2O_3	Al_2O_3	MgO	K_2O	K_2O	Total
Chemical composition (%)	98.53	0.13	0.07	0.06	1.09	0.01	0.01	99.89
Physical properties					Density (g/cm^3)		2.21	
					Particle size distribution (μm)	d_{10}	5.57	
						d_{50}	7.35	
						d_{90}	10.09	

5.1.2.2 Experimental Upcycling Design

In this study we distinguish between first-life (FL) and second-life (SL) render, the last one resulting from the complete upcycling (i.e., no extra raw material added except for water) of the first-life. The proportion of binder:aggregate was kept 1:3 (in mass) and the water:binder ratio was 1:1. The render was mixed according to EN196 and specimens of 4x4x16 cm^3 (Type A samples) as well as cylinders of 5cm diameter and 5 cm high (Type B samples) and cylinders of 4 cm diameter and 2 cm high (Type C samples) were casted. Aiming at full carbonation (i.e., end-of-life), both sample types were cured for 21 days in a climatic chamber (80% relative humidity, 20°C and 4% volume CO_2). After curing, the compressive strength of type A samples was determined as the average of 6 measurements using an INSTRON device. Type A samples were also used for open porosity tests by water absorption under vacuum in accordance with EN 1936:2007. Type C samples were dried at 40°C until constant mass (relative difference between 2 consecutive measurements $<0.1\%$) and the thermal conductivity determined using a TPS 1500 device (ISO 22007-2). Same sample types were scanned with X-rays at a resolution of 45 μm voxel size (120 kV, 45 W, exposure time 300 ms and 0.5 mm aluminum filter) using the CoreTOM scanner (TESCAN) at the Ghent University's UGCT facilities. A total of 1440 projections were obtained per sample. The scans were then reconstructed using the Panthera 1.4.4 software (TESCAN XRE), and the image analysis was performed on Avizo 2020.3.

In the closed-loop upcycling of the mortars, the resulting samples after the mechanical tests were carefully grinded by hand using a laboratory mortar until 25% of the resulting fines under pass-sieve 250 μm were collected (MFR) along with the rejected fractions (i.e., recycled aggregate, RA). Both materials were further processed to be used in the second life render.

The RA was grinded and sieved to comply with the grading of the standard silica sand used previously (EN 196-1) and was reused for the SL casting. The proportion of SiO_2 and $CaCO_3$ present in the RA is estimated by considering the initial mix design and the characterization of the MFR. To determine the proportion $CaCO_3/SiO_2$ in MFR, thermogravimetric analysis (DTA/TG) and X-ray diffraction (XRD) were carried out. A Netzsch STA449F5 apparatus was used for DTA/TG. The samples were heated from 30°C to 980°C in Al_2O_3 crucibles with a heating rate of 20K/min under N_2 environment. XRD data were collected using a PANalytical X'Pert Pro MPD diffractometer in a θ-2θ configuration using incident beam monochromator CuKα radiation (λ=1.54 Å). The samples were scanned between 5° and 70° 2θ with the X'celerator detector. A thermal activation of the MFR was carried out in a laboratory furnace at 900, 1000, 1100 and 1200°C for 2 hours, giving a recycled binder RB (RB-900, RB-1000, RB-1100, RB-1200). After the thermal activation, the samples were again grinded using a laboratory mortar and sieved again (#250). The characterization of the RB powders was carried out through XRD (range 10-70 2θ).

The second life mortars were cast exactly as the FL samples but using the RB and the RA as raw materials, resulting the following samples SL-900, SL-1000, SL-1100 and SL-1200. The curation of the samples and determination of properties was kept equal as for the first life mortars.

A compromise decision of the adequate calcination temperature was done considering the impact of energy consumption and the performance of the renders in terms compressive strength and thermal conductivity. The selected calcination temperature was used to cast the samples used to evaluate the resistance of the FL and SL renders against sulfate attack.

5.1.2.3 Durability Against Sulfate Attack

Sodium sulfate is a commonly used salt for durability assessment on concrete and other porous building materials. It has proven to have a high damage potential due to the induced crystallization pressures in the pore system during its transformation into its two stable polymorphs (Thenardite Na_2SO_4; and Mirabilite $Na_2SO_4\cdot10H_2O$) when exposed to changes in the surrounding environmental conditions [58].

A comparison between the behaviour of the first and second life render against sulfate attack was carried out based on the RILEM TC 271-ASC recommendation adapted for renders [27]–[29] (Fig 5.1b). It consists of an initial contamination step, followed by cycles of damage propagation (i.e., weathering cycles) consisting of rewetting, equilibration, and drying steps. Cylindrical specimens of 5 cm diameter and 5 cm height were cast and cured following the procedure indicated in Section 5.1.2.2. The samples were sealed on the laterals with a paraffin film and textile tape keeping the upper and lower faces uncovered. The initial contamination of the samples was done by submerging their bottom in 3 mm of Na_2SO_4 solution (5% w/w) until complete saturation, equal to the capillary moisture content, according to EN1925-1999. The mass gain of the samples by capillarity during contamination and subsequent rewetting steps was tracked and used to calculate the water absorption coefficients (WAC). After the contamination the specimens were sealed on the bottom and stored in an oven at 40±2°C and relative humidity of 15±5% until 80% of the mass of the initial water used for contamination, for an initial rehydration of the thenardite crystals. After drying, the bottom sealing was removed, and samples were rewetted with demineralized water by capillarity using 80% of the mass of the initial water used for contamination, for an initial rehydration of the thenardite crystals. Then, the bottom was sealed, and the samples were kept for 24 hours in room conditions, to allow equilibration of the system and growth of the mirabilite crystals. Finally, the samples were placed in an oven at 40°C and 15% RH for 14 days. The weathering cycle was performed 4 times.

At the end of the accelerated weathering protocol, the debris of the surface was carefully collected and rinsed with demineralized water to dissolve the salts and filtered and dried in an oven at 40±2°C and 15±5% RH for 24 h to collect the resulting render debris fraction. As an arbitrary criterion to establish the end of the service life of the render, 5% render debris achieved after the accelerated weathering protocol was considered as a critical value. A photographic record of the surface of the sample before and after each cycle was kept. The progress of the salt front was monitored by ultrasonic pulse velocity (UPV) at the end of every drying step. The determination of P wave propagation time is automatic. The frequency of the transducers was 50 kHz. Plasticine was used as a coupling agent to fill the gap between transducers and the sample. Changes in velocity may indicate microstructural differences [30], in which an increase would indicate pore filling, and a decrease would be an indication of salt mobilization or generation of damage in the form of cracks.

5.1.2.4 Environmental Impact Assessment

The environmental assessment was carried out through Life-cycle assessment (LCA) methodology (ISO 14040/44, 2006). The methodology consists of four steps: goal and scope definition, inventory analysis, impact calculation and interpretation.

This study aims at quantifying the potential enhanced sustainability of recycling a lime-based render at the end of life. Two scenarios are compared through LCA (Figure 5.2). Scenario 1 considers the impact from Cradle-to-Grave, which means from the extraction of raw materials until the final disposition at the end of life (Figure 5.2a). Scenario 2 (Fig. 5.2b) evaluates the full upcycling of the render, under the hypothesis of selective demolition, from Cradle-to-Cradle. This approach means that instead of being disposed of at the end of life, the first life render is processed as raw meal to produce the second life render (Figure 5.2b).

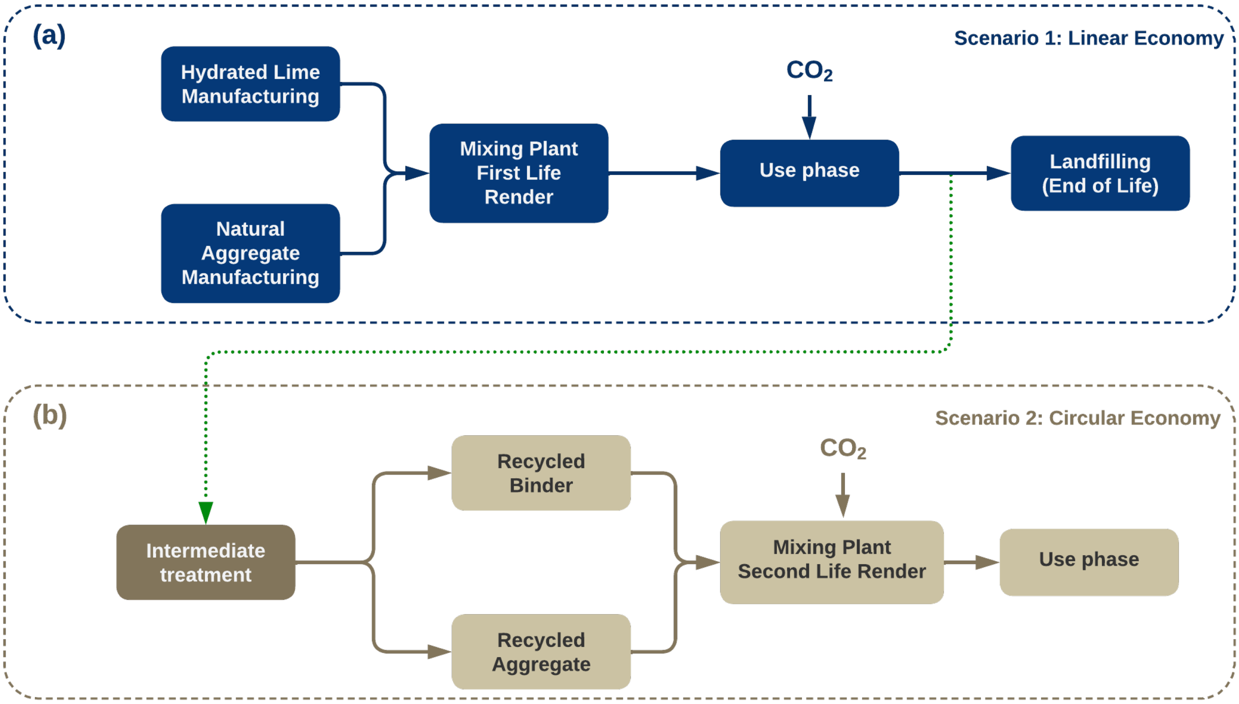


Figure 5.2. System boundaries considered in the study: Linear economy, Scenario 1 (a) and Circular economy, Scenario 2 (b) (closed loop at the end of life).

The evaluation of the CO_2 reduction is carried out through different functional units, increasing the complexity of each one. The CO_2C indicator is used as the first functional unit to carry out the comparison. This indicator stands for the kg of CO_2 emitted per unit kg of each render produced from Cradle-to-Gate of the factory, involving all required intermediate treatments for the conditioning of the upcycling of the render. This functional unit does not take the primary functions of the render into consideration.

The system boundaries are then expanded to include the whole life-cycle of the material, from Cradle-to-Grave for scenario A and from Cradle-to-Cradle for scenario B. The CO_2 associated to 1 kg of FL and SL render under such boundaries is calculated as shown in Equations 5.1 and 5.2.

$$CO_{2\,SL} = \sum CO_{2\,M} - CO_{2\,UP}$$ **Equation 5.1**

$$CO_{2\,FL} = \sum CO_{2\,M} - CO_{2\,UP} + CO_{2\,EoL}$$ **Equation 5.2**

where $CO_{2\,M}$ *represents the* CO_2 *emissions for the manufacturing of the respective render,* $CO_{2\,UP}$ *is the* CO_2 *sequestrated during the use phase, and* $CO_{2\,EoL}$ *are the emissions associated with the disposal of the FL render.*

Subsequently, to account for the specific performance of each render and compare them, a modified approach proposed by previous authors is used [35]. Under this approach, a ratio of the CO_2 emissions of each render including the compressive strength (Equation 5.3), the thermal conductivity (Equation 5.4) and the integration of both properties (Equation 5.5) is used to estimate the CO_2 reductions of Scenario B against Scenario A. When the ratio on these equations is equal or lower than 1, it means that a reduction target can be achieved by upcycling the render (Scenario 2) while keeping comparable technical performance.

$$FU_2 = \frac{CO_{2_{SL}}}{CO_{2_{FL}}} * \left(\frac{CS_{FL}}{CS_{SL}}\right)$$ **Equation 5.3**

$$FU_3 = \frac{CO_{2_{SL}}}{CO_{2_{FL}}} * \left(\frac{k_{SL}}{k_{FL}}\right)$$ **Equation 5.4**

$$FU_4 = \frac{CO_{2_{SL}}}{CO_{2_{FL}}} * \left(\frac{CS_{FL}}{CS_{SL}}\right) * \left(\frac{k_{SL}}{k_{FL}}\right)$$ **Equation 5.5**

where CS represents the compressive strength, and k represents the thermal conductivity of the FL and SL render.

The calculation of the life-cycle inventory is based on a process-based methodology developed by the authors, which allows the design and implementation of the experimental recycling cycle at a manufacturing scale, through mass and energy balances in the production plant [8].

The environmental impact is calculated using the software OpenLCA, the database EcoInvent V.3.6 and the Impact 2002+ method. For the calculation of the inventory amount of the CO_2 sequestrated during the use phase, it was considered a complete carbonation, considering the thickness of the rendering layer (usually <10 mm) and the standard service life (50 years). The Life-cycle Inventory used to carry out the comparisons between Scenario 1 and 2 is provided in Appendixes A5 and A6.

5.1.3Results and Discussion

5.1.3.1 First and second life binder

Characterization of the recycled render fines (MFR)

First life renders were mechanically separated and sieved as indicated in Figure 5.1a (Section 5.1.2.2). Figure 5.3 a and b show the XRD diffractogram pattern and the thermogravimetric analysis (DTA/TG) results of the MFR before the thermal activation, respectively.

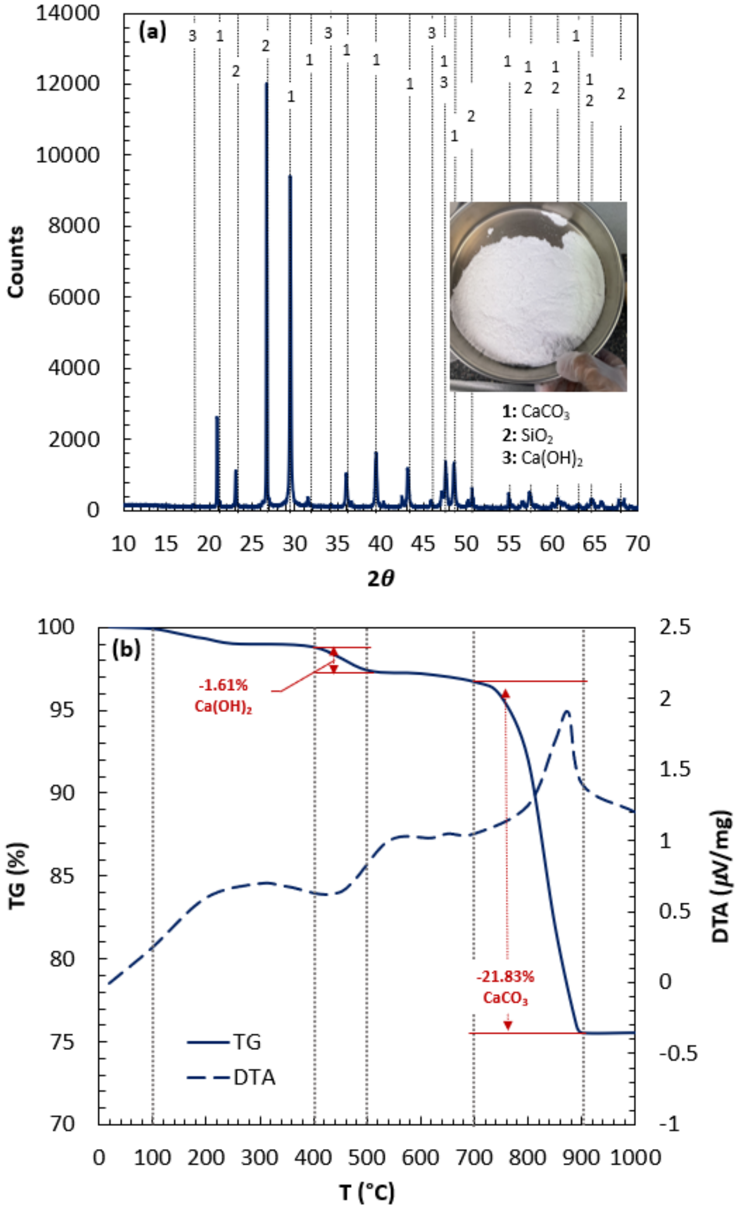


Figure 5.3. XRD diffractogram (a) and TG/DTA of the MFR sample (b)

The XRD shows that the main mineralogical phases present in MFR are calcite, specifically at 24.6, 29.4, 36, 39.5,47.5, 49.9° 2θ, quartz at 21, 27.5, 36.5, 46, 51° 2θ and with minor presence of portlandite (18, 34 and 46° 2θ) [36]–[39]. The presence of portlandite and calcite was confirmed through DTA/TG (Fig. 5.3b), registering a mass loss (related to the final dry mass of the sample) of 1.6% between 400-600°C and 21.83% between 700 and 900°C, attributable to their decomposition [40]. Considering that quartz cannot decompose at the studied range, the estimated proportion through stoichiometric calculations of SiO_2 in MFR is 43.82% (49.61% $CaCO_3$ and 6.57% $Ca(OH)_2$). From this result, the estimated proportion of SiO_2 and $CaCO_3$ in the RA is 85 and 15 % respectively. An important parameter to highlight, MFR the raw meal for the second life binder preparation, is the ratio effective CaO/SiO_2 of 0.67 for temperatures above 900°C. As discussed by the authors, the calcination temperature and the CaO/SiO_2 ratio have a major influence on the synthetized clinker phases [41].

Thermal activation of the MFR

MFR was subjected to a thermal activation at 900, 1000, 1100 and 1200°C. A photographic record of MFR and the obtained binders at 900 and 1200°C, along with the XRD patterns are shown in Figure 5.4. In Figure 5.4a a light colour changes of the RB-900 and RB-1200 compared to MFR was observed as the calcination temperature increases.

In Figure 5.4b it is observed that the peaks attributed to calcite (29.4, 36, 39.5,47.5, 49.9° 2θ) in Fig. 5.3a are no longer present at 900°C, confirming the complete decomposition of calcite observed through DTA/TG (Fig. 5.3b). As a result, CaO is formed and observed already at 900°C at 32.4, 37.5, 54.0 and 64.2° 2θ [42]. The increment of the firing temperature to 1200°C increases the intensity of the peaks assigned to CaO (specifically at 32.4 and 37.5° 2θ).

The cell parameter of the CaO calculated for 900°C and 1200°C is 0.4811 and 0.4805 nm. The decrement in the cell parameter is since CaO retains the typical rhombohedral structure of $CaCO_3$ after decomposition at 900°C and the crystal lattice is rather imperfect. With the rise in the temperature, the CaO crystals grow and the crystal system changes to cubic structure, and the array of atoms in the crystal lattice tend to be more regular [43]. This fact also explains the increment in the sharpness of the peak observed at 37.5° 2θ.

The peaks attributed to SiO_2 (Fig. 5.3a) are also identified in samples RB-900 and RB-1200. However, from MFR to RB-1200 specific peaks of SiO_2 decrease their intensity at 21, 23 and 51° 2θ. During heating, from 600°C quartz transforms to different polymorphs (α-quartz, β-quartz, β-tridymite and β-cristobalite), contributing to a more amorphous structure[44]. This process is also influenced by mechanical handling (separation and co-grinding) of the first life render, enhancing the reactivity between CaO and SiO_2 during the thermal treatment. The formation of β-C_2S and C_3S at 34.2, 36.8 and 41.4° 2θ in coincidence with [37], [45], [46], explain the observed decrement in the mentioned SiO_2 peaks. Furthermore, the peak of uncalcined MFR at around 39.5° 2θ attributed to SiO_2 and shared with β-C_2S and α'-C_2S decreases its intensity as the temperature increases[45]. Although a decrement in the CaO peaks is expected, the double phenomena of lime consumption and crystal lattice refinement can potentially play a coupled role on the observed diffractogram. However, the peaks at 54.2 and 64.2° 2θ remain in comparable intensities at 900 and 1200, which could give an indication of the consumption of CaO to form C_2S and C_3S.

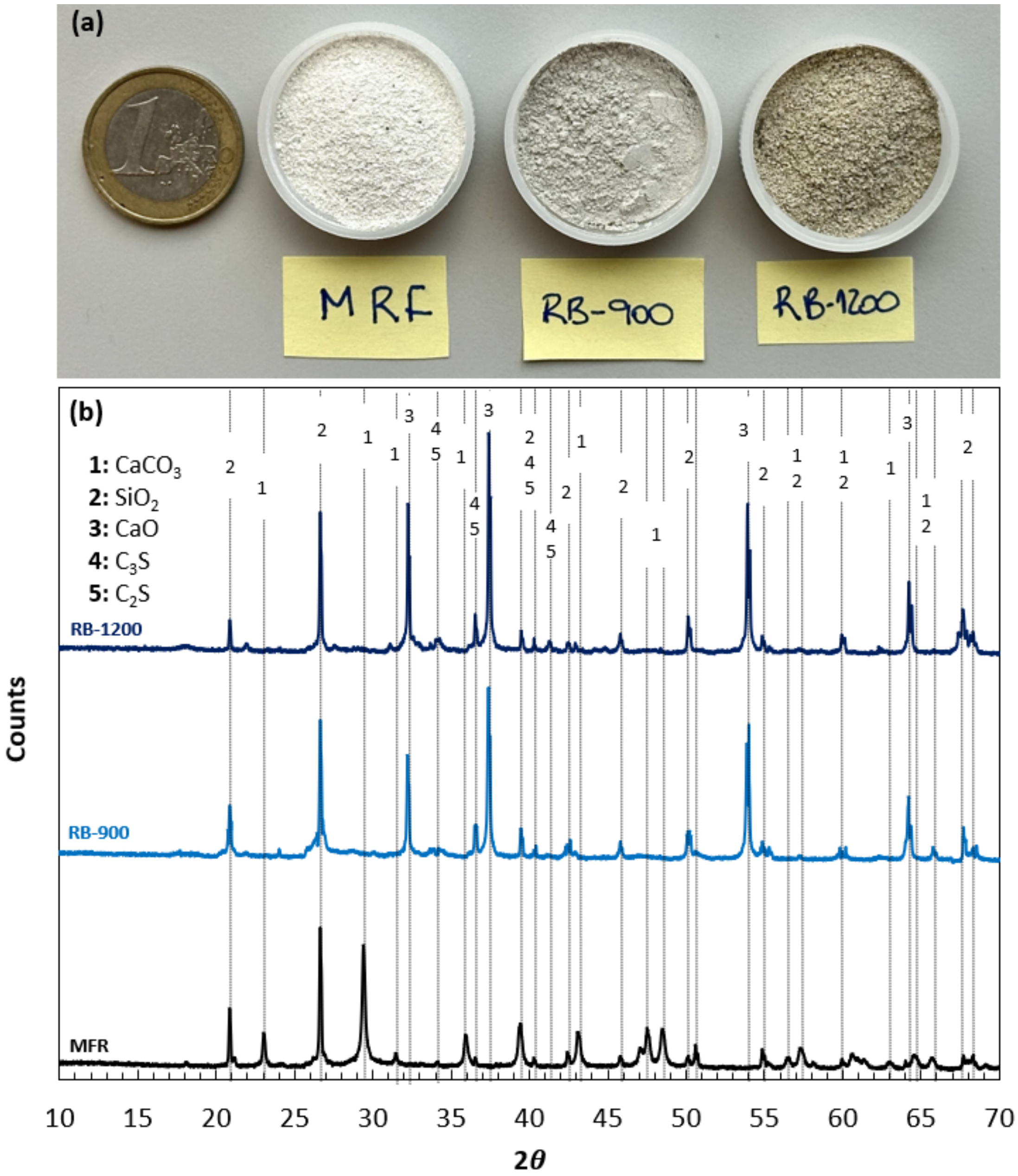


Figure 5.4. Comparison of the aspect (a) and XRD diffractogram (b) of the MRF, RB-900 and RB-1200 samples

5.1.3.2 First and Second Life Render

Microstructure characterization

Several materials' properties, ranging from mechanical and, thermal, and to durability characteristics are related to the internal microstructure of the specimens. The porosity of the samples was firstly determined by open porosity measurements on type A samples. The results were 29.1±0.5, 33.4±0.3, 32.5±0.5, 31.7±0.3, 30.9±0.4% for FL, SL-900, SL-1000, SL-1100 and SL-1200 samples, respectively. The values are in the order of previous studies performed on similar lime-based mortars [43], [44]. The lower porosity is registered for the FL sample, which makes sense considering the higher proportion of portlandite present in the binder compared to SL series, and consequently more carbonates precipitating in the pore system [45]. The dilution effect of the binder by the silica quartz is one of the reasons that explains the higher porosity of the SL series compared to FL (Section 5.1.3.1). The second reason could be associated with the use of recycled aggregate, which in general leads to lower compressive strength compared to natural aggregates [23], [46]. However, a decreasing trend in the open porosity is observed as the calcination temperature rises. As described in Section 5.1.3.1, increasing the firing temperature leads to a higher reactivity of the CaO phases combined with the formation of C_2S and C_3S phases (Fig. 5.4), which can contribute to a refined matrix, attenuating the dilution effect.

Although open porosity determination is a reliable, repeatable, and time-effective technique, a main disadvantage is that the method can only account for connected pores open to the surface of the sample [47]. As a complementary technique, X-ray micro-CT is used to visualize the pores inside the specimens FL and SL-1000 in 3D images [48]. The results of the scan and the pores frequency histogram are shown in Figures 5.5 and 5.3, respectively.

A region of interest comprised by the same volume of for each scanned sample was selected. The segmentation of the air voids above 1.8E+05 μm^3 was done using interactive threshold, and the same intensity range was chosen for both samples. Then, a label analysis operation was performed to calculate the voids' geometric properties. The individual voids are represented by distinct colors in Figure 5.5. All small spots below 1.8E+05 μm^3 (3 voxels) were not considered in the counts.

The volume analysis demonstrates a homogeneous air void distribution for both cylinders. Nevertheless, the SL-1000 sample shows a higher density of air voids with respect to FL, as displayed in Fig. 5.5. According to the frequency histogram (Fig. 5.6), both FL and SL mortars have a unimodal distribution with predominantly small pores below 1.0E+07 μm^3. In this case, SL-1000 shows a homogeneous increase in frequency of all pore fractions, which increments the total porosity volume. This is supported by the increase in open porosity percentage of SL renders, compared to FL.

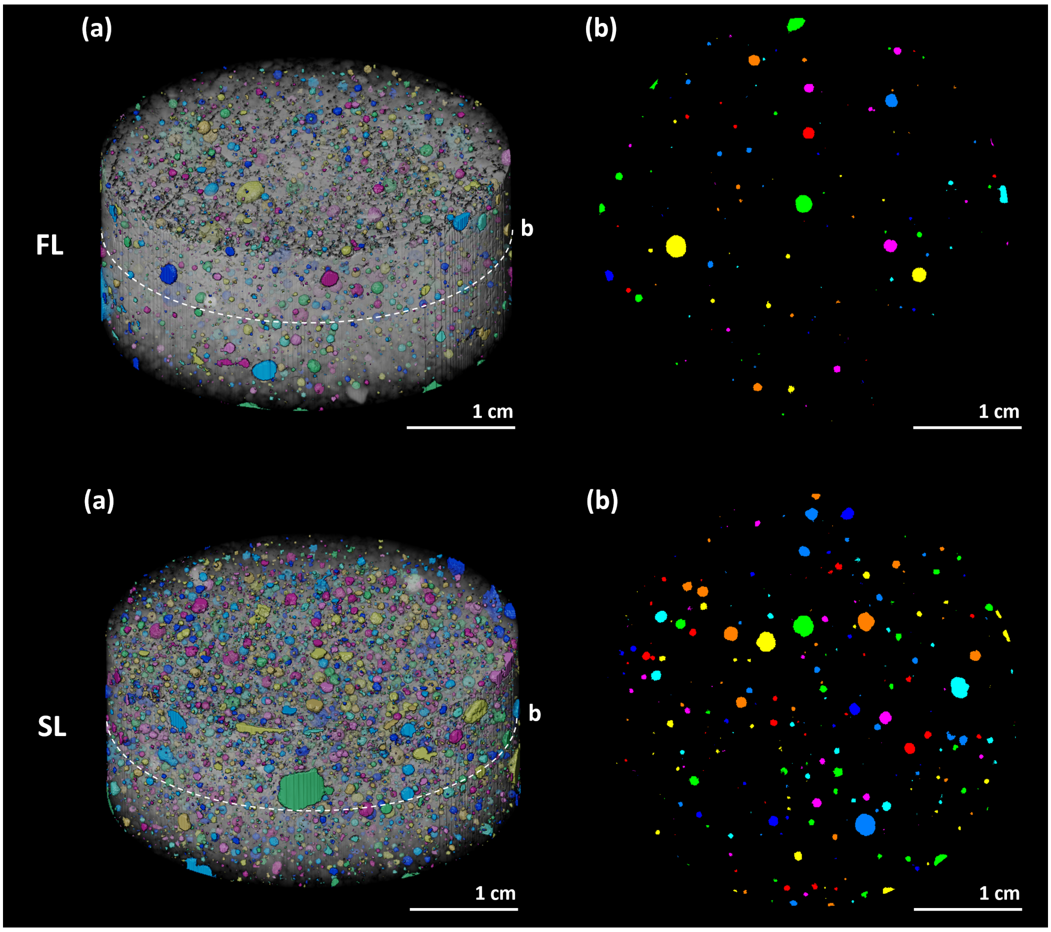


Figure 5.5. 3D volumes (a) and cross sections of FL (top) and SL-1000 (bottom) cylinders (b). The colours indicate individual segmented air voids distributed along the matrix. In (b) The dotted lines indicate the location of the cross section.

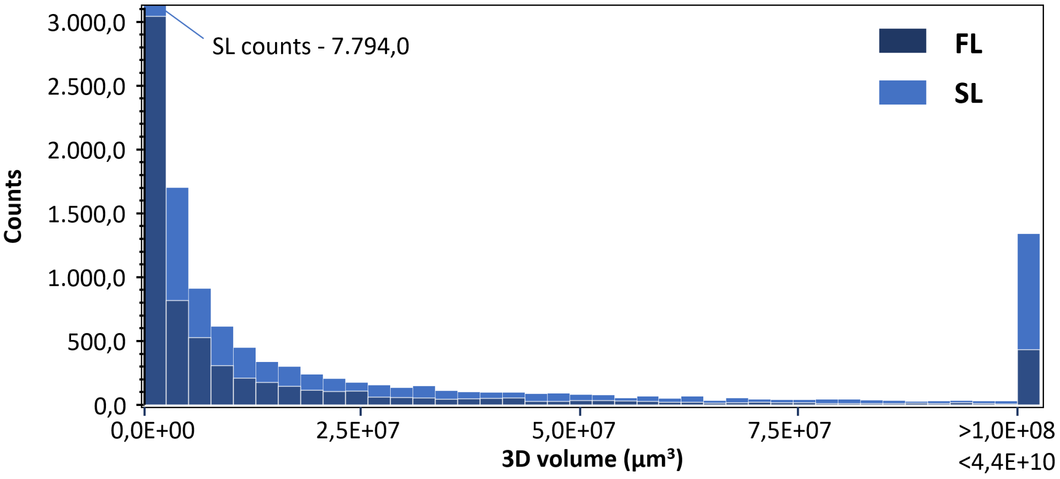


Figure 5.6. Frequency histogram of air voids distribution by 3D volume. An overflow bin was set at 1.0E+08 μm^3 to highlight the main differences in the factions below 2.5E+07 μm^3.

Compressive strength and thermal conductivity

The compressive strength measured on Type A samples for the first and second life is presented in Figure 5.7. The left axes show the absolute values, and the right axes show the ratio between the compressive strength of each sample SL_i (i = 900, 1000, etc.) subjected at a specific thermal treatment and FL.

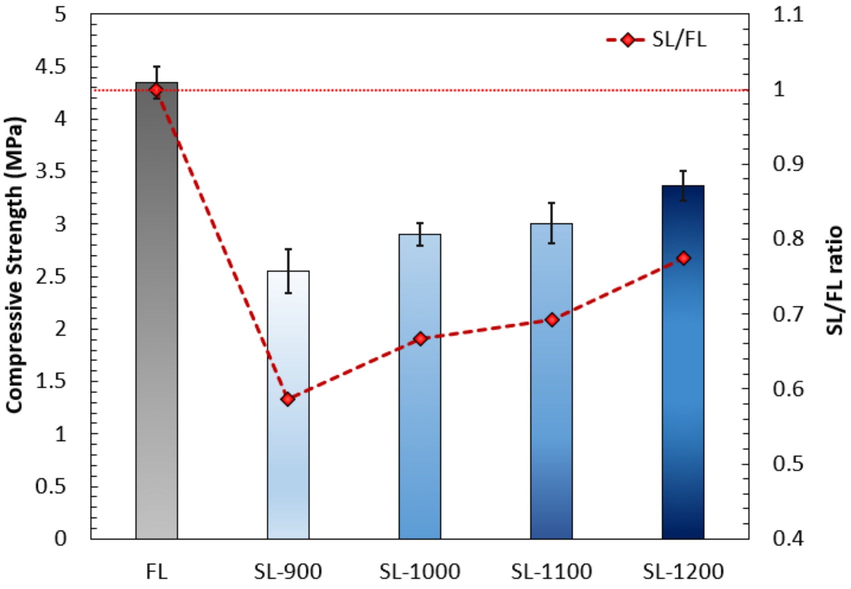


Figure 5.7. Compressive strength of First Life and Second Life mortars (calcined at 900, 1000, 1100 and 1200°C) after carbonation and ratio between Second Life and First Life (SL/FL) strength results

The highest compressive strength value is registered for the FL sample, which also had a denser matrix with lower porosity in comparison with to the SL-series as discussed in the microstructural characterization (Section 5.1.3.1). The mineral transformation of $Ca(OH)_2$ to $CaCO_3$ because of the carbonation reaction has a significant influence on the microstructure of the material [53]. The dilution effect of the SiO_2 in the RB-series is registered in the CS results of the second life mortars, with an increasing trend in the SL/FL ratio as the calcination temperature rises. This effect could be explained by considering the increased reactivity of the CaO and the additional contribution of hydrated products of the clinker phases analyzed in Section 5.1.3.1. The higher open porosity of SL-1200 compared to FL explains the lower CS and confirms that a lower proportion of calcium in the binder leads to a less connected matrix. The results agree with previous studies on lime-based mortars under accelerated carbonation [48], [54].

A critical design parameter of lime-based render/plaster is thermal conductivity. An important remark is that pure lime-plasters are not considered as insulators [55]. Yet a wide variety of additives can be used to improve the thermal properties of lime-based renders and plasters [56], [57]. However, this study aims at a comparison between first life and fully recycled second life renders. Given that the inclusion of additional components in the mix might cause perturbations in the system, no additives were added. The thermal conductivity results are 1.231, 1.002, 1.015, 1.099 and 1.107 W/mK for FL, SL-900, SL-1000, SL-1100 and SL-1200 respectively. The highest thermal conductivity is found for the FL sample, which is logical considering that a refined pore structure (Fig. 5.5 and 5.6) leads to a denser material with reduced insulation value [54], [55], [57]. The increment of the thermal conductivity in the RB series can be explained by considering the same principle, where the hydration and carbonation products accommodate in the internal structure of the mortar, producing a reduction in the porosity[48].

Treatment selection

To define an optimal calcination regime, the required final product needs to be defined. In this case, the material is used as external coating and according to the literature review, the compressive strength, and the thermal conductivity in the design of the render/plaster are relevant parameters [55]–[57]. As was previously observed, both characteristics are closely related to the microstructure of the material and have an inverse behaviour. A compromise decision was made considering the calcination temperature, the compressive strength, and the thermal conductivity, which led to the selection of 1000 °C for the calcination treatment. In the next section the durability against sulfate attack will be assessed for the FL and SL-1000 samples.

5.1.3.3 Durability against Na_2SO_4

Sodium sulfate is a commonly used salt for durability assessment on concrete and other porous building materials. It has proven to have a high damage potential due to the crystallization pressures induced in the pore matrix during its transformation into its two stable polymorphs when exposed to changes in the surrounding environmental conditions [61]. This protocol consisted of one contamination step to load the FL and SL mortars of a Na_2SO_4 solution (5%), followed by 4 weathering cycles to induce precipitation, dissolution, and rehydration of Na_2SO_4 to cause decay of the materials. At the end of the test, the debris was collected and separated in their respective mortar and salt fraction by dissolution with demineralized water. Figure 5.8 displays the relative mass differences before and after the weathering tests, as well as the respective debris fractions as a percentage of the initial dry mass. The final mass difference is attributed to the final amount of salts that the system absorbed during the initial contamination, and that will be used to induce damage through weathering cycles.

It can be observed in Fig. 5.8 that SL-1000 samples show higher mass gains than FL. This can be attributed to the higher space availability within the pore system of SL specimens for salts to precipitate in (Section 5.1.3.2). As the crystals are less constrained due to the space availability, they would tend to continue growing without damaging the pore walls until reaching a critical pore-filling that could lead to failure. This could also explain why SL mortar, having a higher concentration of salts in the system, resulted in less final debris. The fact that higher amount of salt was retained by the SL matrix could represent a problem should the weathering cycles continue. On the other hand, FL mortar gained less mass at the end of the test, but yielded double the mortar debris, and 3 times as many salts in the form of efflorescence with respect to SL mortar. This result could be attributed to the reduced porosity due to the packed microstructure observed in Section 5.1.3.2, constraining the space in which crystallization pressures occur. By this, the tensile strength of the mortar is surpassed [62], causing cracking and crumbling in the form of debris. Nevertheless, none of the samples have exceeded the 5% of render debris established as critical percentage, and therefore it is assumed that both would have an equivalent service life if exposed to the same degrading agent.

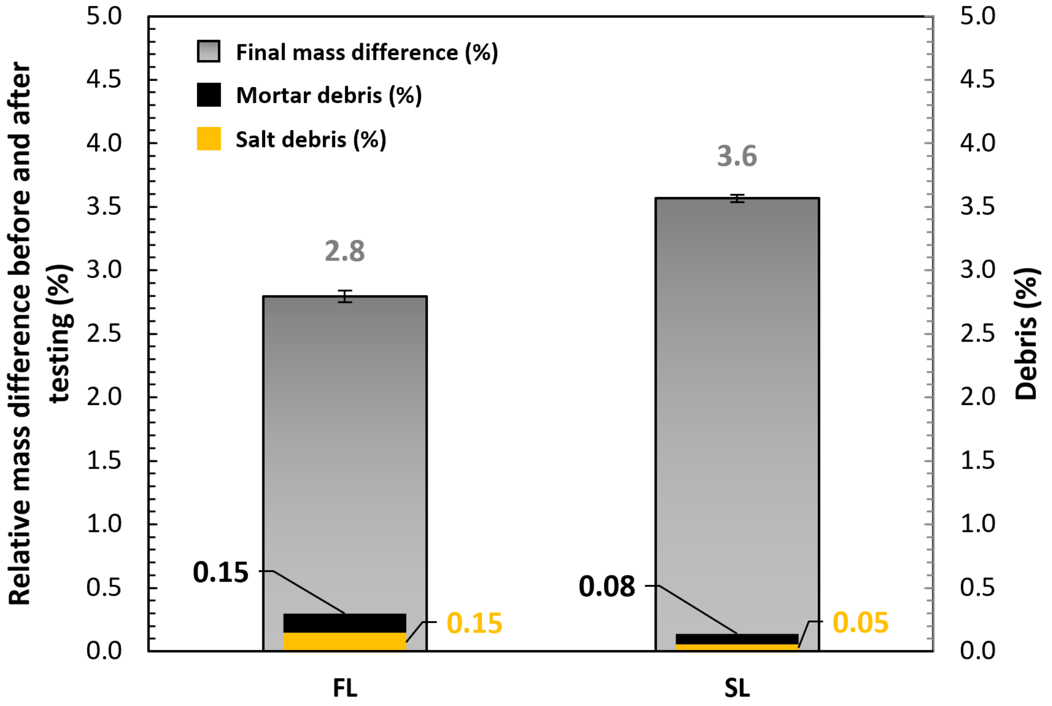


Figure 5.8. Final damage assessment after weathering cycles using Na_2SO_4 (5% concentration). The Gray colour indicates the final mass gain with respect to the initial weight. Black and yellow indicate the final debris quantification (%).

Continuous monitoring of the specimens was carried out during the durability test to understand the different physical and mechanical damage processes. Figure 5.9 shows the comparison between the initial and the final state of the specimens' drying surfaces and the effects of salt crystallization to their aesthetics. FL showed a salt crust and efflorescence formed by the wetting and drying cycles [63].

After brushing them off for debris quantification, most of the original surface of the mortar had been lost, and the aggregates were completely exposed. SL mortar had a similar effect but to a smaller extent. In this case, the efflorescence did not cover the entirety of the surface, and the drying surface was only partially affected. This agrees with the debris analysis in Figure 5.8.

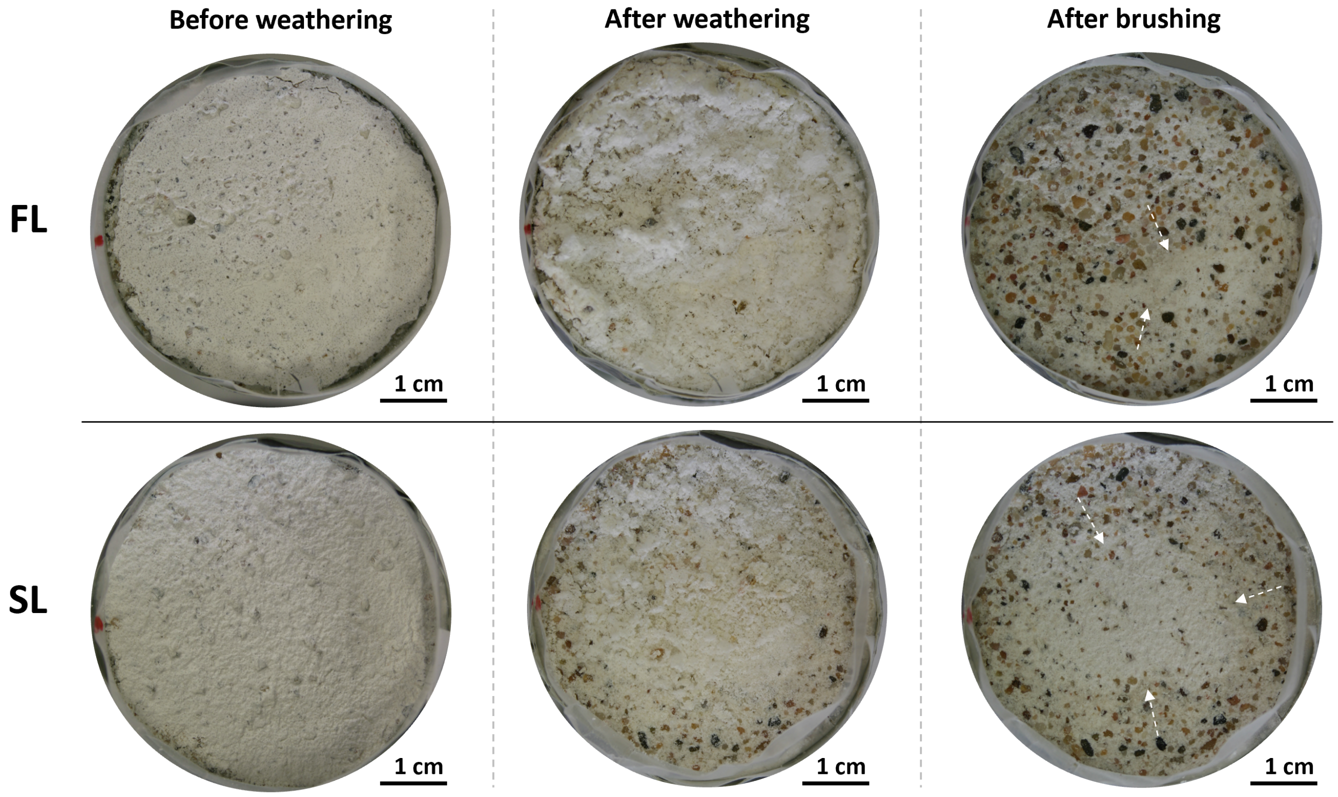


Figure 5.9. Photographic record of a representative sample of FL and SL-1000. The arrows point at the areas that remained unaltered after the weathering cycles.

In Figure 5.10, the UPV measurements taken every 10 mm from the bottom to the top of the cylinder after every drying step are reported.

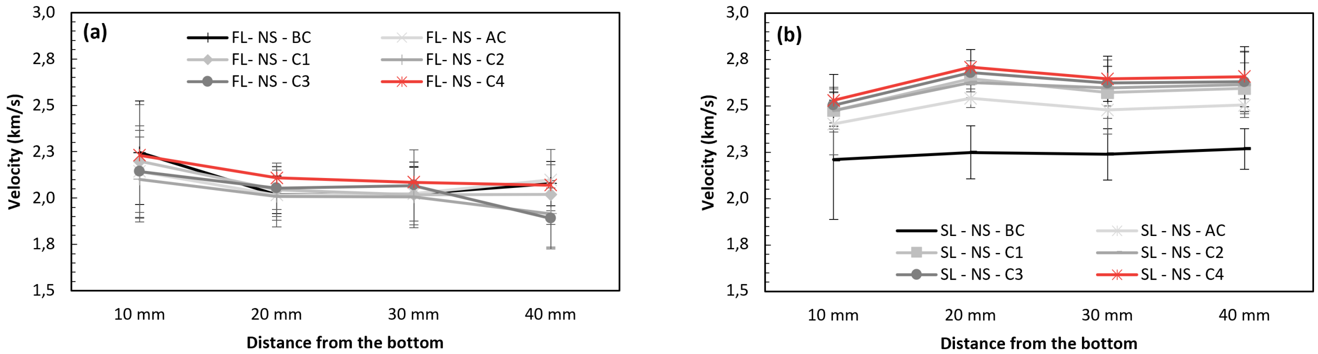


Figure 5.10. UPV measurements during continuous monitoring after every drying cycle of (a) FL and (b) SL-1000 samples. The red lines show the final state of the samples. BC-before contamination; AC-after contamination; C-number of cycles (1, 2, etc.).

Both mortars displayed a similar velocity before the initial contamination took place (~2,3 km/s). Nevertheless, the following measurements indicate a completely different behaviour as the cycles progressed. FL mortar remained relatively more stable than SL. The slight decrease in velocity towards the drying surface might be an indication of the creation of cracks or voids in the consecutive weathering cycles, and finally stabilizing near the starting point. On the other hand, SL mortar displayed a continuous increase in velocity after every weathering cycle. This could be a proof of the homogenous pore filling and progress of the salt front towards the surface aided by the continuous rewetting and drying steps. The consistent increase in density could be related to the continuous growth of the Na_2SO_4 crystals on the available space, forming a denser structure. It is important to highlight that this experimental method for continuous monitoring is not standard practice for this type of test, and it is used to prove its feasibility for continuous monitoring of small specimens.

The WAC of FL and SL-1000 samples is shown on Figure 5.11.

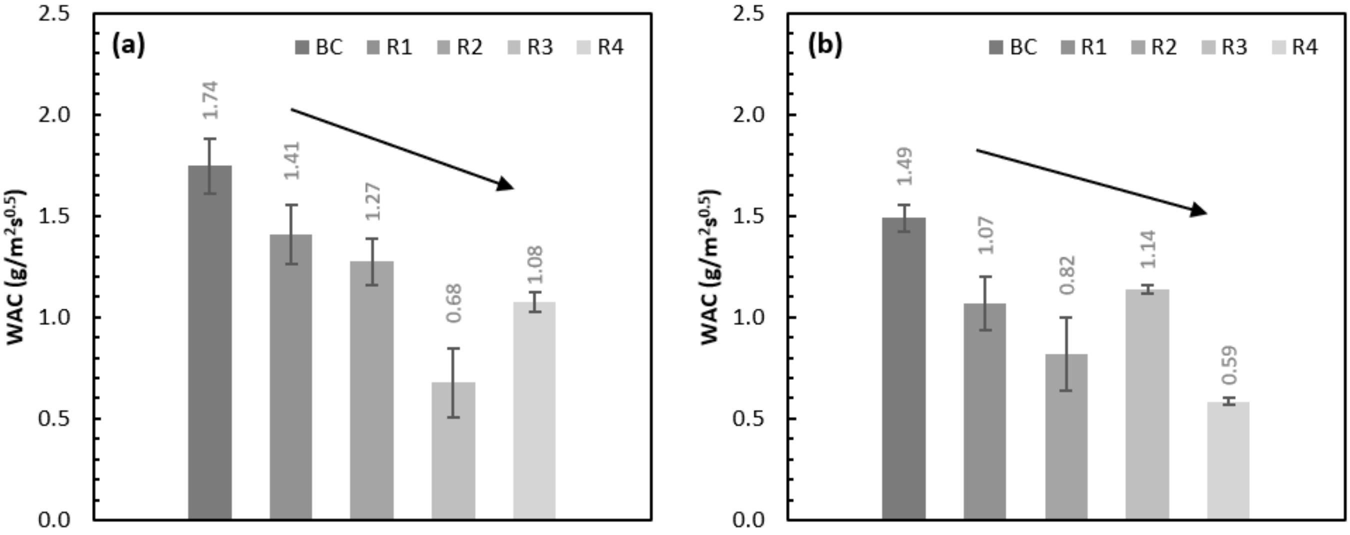


Figure 5.11. Water absorption coefficients of FL (a) and SL (b) mortars calculated during the contamination (BC) and rewetting steps (R1-4).

The initial WAC was calculated using the Na_2SO_4 solution, and the consecutive measurements were done during the rewetting steps using demineralized water. Both mortars expose a decrease in WAC at the end of the test. The difference relies on FL mortar having higher initial and final WAC, whereas SL mortar goes below half the initial value. In both cases, this reduction can be explained by the degree of pore filling by the end of the cycles, modifying the total permeability of the mortars. This was achieved by the progressive accumulation of salts, and their differences rely on the final mass gains of each mortar as shown on Figure 5.8.

5.1.3.4 Environmental impact assessment

This section presents the findings of the impact assessment, specifically focusing on CO_2 emissions in two key stages: the manufacturing phase (Cradle-to-Gate) and the cradle-to-use phase.

Cradle-to-Gate

Figure 5.12 displays the Global Warming Potential (GWP) indicator results for the cradle-to-gate production of the raw materials, first-life renders, and the second-life renders. For a better understanding of each material's specific contribution to the final mix, a comparison between hydrated lime and RB-1000 binder, as well as between natural aggregate and recycled aggregate, is illustrated in Figure 5.12a and 5.12b, respectively. The results are presented per kilogram of binder and aggregate produced. Finally, Figure 5.12c breaks down the total magnitude of the indicators to highlight the contribution of each considered item, presented per kilogram of first and second life render.

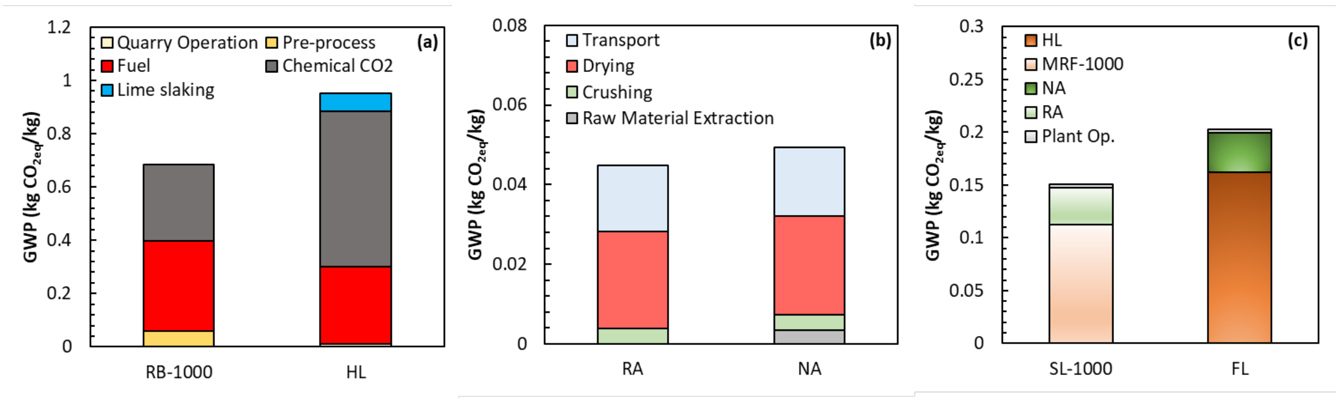


Figure 5.12. Global Warming Potential results from Cradle-to-Gate: Comparison of the MRF-1000 binder against hydrated lime (HL) production (a); Comparison of the recycled aggregate (RA) against natural aggregate (NA) (b); and Comparison of the render using fully recycled materials (SL-1000) and the First Life render (FL) (c)

In the manufacturing of binders (Fig. 11a), a notable 29% reduction in the total GWP indicator is observed for RB-1000 compared to HL. This reduction primarily stems from decreased chemical CO_2 emissions resulting from limestone decomposition. While hydrated lime production generates 0.59 kg CO_2/kg $Ca(OH)_2$ due to complete $CaCO_3$ decomposition [61], RB-1000 (Section 5.1.3.1), with 35% lower chemical CO_2 emissions from MFR calcination, experiences a rise of approx. 14% in fuel-specific contribution due to its higher calcination temperature (1000°C vs. 900°C). Furthermore, considering the co-production of MFR and RA during the initial separation process, 25 % of the emissions of this treatment (mass allocation) are allocated to MFR and consequently to RB-1000 manufacturing (Appendix 6).

In the case of aggregate manufacturing (Fig. 5.12b), similar processes occur in natural [56] and recycled aggregate production (Appendix 6), with comparable energy consumption for conditioning (drying, crushing and classification) [62], [63]. However, the avoided extraction of natural raw materials in recycled aggregates (RA) production reduces the indicator by approx. 10% (RA vs. NA). While the next section focuses on landfilling impacts, the avoided extraction of raw materials contributes to indicator reduction and supports efficient resource use in circular economy model [64], [65]. The emerging trend of accelerated carbonation of recycled aggregates holds promise for further reducing cradle-to-gate material footprints and enhancing recycled aggregate properties [25], [66]. A new design of kiln technologies with direct CO_2 separation and carbon capture systems from cement/hydrated manufacturing, can offer an environmentally friendly alternative for carbon use at the point of emission [8].

Finally, the reduction in GWP observed for the manufacturing of the second-life binder and recycled aggregate are mirrored in the GWP of the second-life render compared to the first-life render (Fig. 5.12c). The GWP indicator is dominated by the binder's production (75-79%), as observed in previous research by the authors [7]. Due to the lower CO_2 emissions of RB-1000 compared to HL (Fig. 5.12a), a 25% reduction is achieved for the second-life render compared to the first-life render (Fig. 5.12c).

Cradle-to-Grave vs. Cradle-to-Cradle

The findings in previous Section exclusively focus on the manufacturing phase, neglecting material performance. As outlined in Section 5.1.2.4, for a comprehensive comparison between the first and second life render, it is essential to broaden the system boundaries to encompass all life stages. Table 5.2 presents CO_2 equivalent calculations for the manufacturing phase, CO_2 sequestration during the use phase and emissions from the End-of-Life scenario for the FL render. Results for CO_2 FL and CO_2 SL, calculated using Equations 1 and 2, are juxtaposed in the same table. These results are utilized for the calculation of FU2, FU3 and FU4 according to Equations 5.3 to 5.

Table 5.2. Results of the CO_2 FL and CO_2 SL

Material	Phase						Total
	Manufacturing	kg CO_{2eq}	Use	kg CO_{2eq}	End of Life	kg CO_{2eq}	
First Life Render	Hydrated Lime	0.162	Carbonation	-0.149	Transport	0.082	CO_2 FL
	Natural Aggregate	0.038			Landfilling	0.0087	
	Plant Operation	0.003					
	$Total_{manufacturing}$	0.203	$Total_{use}$	-0.149	Total $_{End-of-Life}$	0.091	0.145
Second Life Render	RB-1000	0.113	Carbonation	-0.071			CO_2 SL
	Recycled Aggregate	0.034					
	Plant Operation	0.003					
	$Total_{manufacturing}$	0.0151	$Total_{use}$	-0.071	Total $_{End-of-Life}$	N/A	0.080

In the manufacturing phase, SL-1000 demonstrates a lower environmental footprint than FL render due to reduced calcite in the recycled binder, leading to lower chemical CO_2 emissions. However, this results in reduced carbon sequestration during the material's use phase [67], with FL sequestrating around 53% more CO_2 than SL-1000 (Table 5.2). Considering that both renders had a similar durability performance against sulfate attack (See Section 5.1.3.3), the maintenance operation was not accounted for at the use phase. The End-of-Life phase is only considered for FL render, in this Cradle-to-Grave scenario. The transportation of the waste to the landfilling centre is a major contributor to air emissions due to fuel consumptions[68], dominating the End-of-Life indicator (around 90%). The total emissions $CO_{2\ FL}$ and $CO_{2\ SL}$, excluding technical material properties, show SL-1000 render reduces CO_2 emissions by 45%.

When incorporating compressive strength (CS) (Section 5.1.3.2) FU2 value is 0.83, only a 17% reduction is observed due to lower mechanical performance of the second life render. Conversely, improved thermal behaviour of SL-1000 (Section 5.1.3.2) highlights its higher porosity and leads to a 55% CO_2 reduction in FU3 (0.45). A balanced approach as used in FU4 (0.68) considering both compressive strength and thermal behaviour results in 32% reduction.

For all considered scenarios, the recyclability of the lime render at the end of life holds promise for contributing to lime industry decarbonation and circular economy. However, results are sensitive to analysis scope and material's properties considered, necessitating careful consideration of application and well-documented assumptions for fair and transparent life cycle assessments.

5.1.4 Conclusions

This paper evaluated the feasibility of producing a fully recycled second-life lime render through a closed-loop upcycling process and has quantified the environmental benefits of a closed-loop circular economy compared to a traditional linear economy. The key conclusions are as follows:

- The fines from the mechanical separation of a first life render used as raw material for the second life binder exhibited a composition of 50% $CaCO_3$, 44% SiO_2 and 6% $Ca(OH)_2$. Thermal activation at 900 to 1200 °C resulted in the formation of CaO, consumption of SiO_2 and the formation of clinker phases.
- Microstructure characterization revealed higher porosity in second-life renders compared to the first life render, attributed to the dilution of the second-life binder by inert SiO_2. Porosity decreased with an increase in calcination temperature. X-ray micro-CT analysis confirmed the higher porosity of recycled renders. FL and SL renders show a unimodal pore size distribution with predominantly small pores below 1E+07 μm^3. SL renders show a homogeneous increase in frequency of all pore fractions, which increments the total porosity volume. This is supported by the increase in open porosity percentage of SL renders measured by water absorption under vacuum, compared to FL.
- Second-life renders exhibited 20 to 40% lower compressive strength (compared to the reference), with the best performance observed for calcination of the recycled binder at 1200°C. Conversely, thermal conductivity increased with reduced porosity and higher calcination temperature. The SL-1000 sample was identified as the treatment balancing the effect of processing temperature and technical properties.
- Debris collected after accelerated weathering was slightly higher for FL than for SL, attributed to lower space availability within their pore microstructure. This agreed with open porosity, micro-CT, water absorption coefficient and ultrasonic pulse velocity tests.
- The environmental assessment showed that from Cradle-to-Gate 25% CO_2 emissions can be cut for the SL-1000 compared to FL. The Cradle-to-Cradle scenario is more eco-efficient than the Cradle-to-Grave, leading to reductions ranging from 17 to 55% emissions when relevant material properties. However, results are contingent on analysis scope and materials properties, emphasizing the importance of case-specific inputs and comprehensive documentation in environmental impact assessment.

Finally, the findings encourage the adoption of circular economy principles to minimize construction and demolition waste, conserve natural resources and reduce CO_2 emissions. Future research should consider formulations closer to market-available options, incorporating lightweight aggregates and additives to assess their impact on the recyclability and properties of second-life renders.

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5.2 Open-loop Upcycling

Publication 4. Cradle-to-Grave Environmental and Economic Sustainability of Lime-based Plasters Manufactured with Upcycled Materials

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5.2.1 Introduction

One of the humanity's major challenges is to change production and consumption patterns to reduce the ecological footprint while achieving economic growth and sustainable development. Construction materials, particularly lime-based materials, play a significant role, because it is predicted that by 2050 two-thirds of all humanity will be living in cities[1]. This will lead to higher demand for construction materials and increased environmental impacts during production [2]–[4].

Lime based materials (LBM) have a significant share in the European construction market, including concrete blocks, bricks, mortars, renders and plasters [5]. In 2020, Europe used 4 million tons of quicklime in civil engineering and construction[6] [7], resulting in 4.6 million tons of CO_2 (1.2 t CO_2/t of CaO) and the consumption of 6.78 million tons of $CaCO_3$. Quicklime production emissions primarily come from the chemical decomposition of calcite (65%) and fuel combustions (35%). To create more sustainable materials, a combination of strategies is needed, including sustainable energy sources, efficient devices, carbon capture technologies and circular economy (CE) practices [8].CE is crucial due to the scarcity of natural resources and the issue of waste production associated with current production and consumption practices.

Circular economy is a production and consumption model focused on minimizing waste while preserving the value of products and materials. The European Union's action plan for circular economy emphasizes product design, production process, raw materials consumption, and the use of waste as secondary raw materials to address the complexities of the value chain. Lime-based construction materials require 1.78 t of $CaCO_3$ per ton of CaO produced. Therefore, substituting virgin raw materials with waste or by-products as secondary resources (SR) is highly desirable. Depending on the SR's properties and the required conditioning in intermediate treatment, a potential reduction in the CO_2 footprint can be achieved. Additionally, industries producing SR can benefit by reducing their own environmental impact and minimizing waste landfilling.

Compared to cement-based materials, the scientific knowledge regarding the use of alternative materials in the manufacturing of lime-based construction materials is currently limited. One probable reason is that in cement-based materials, various supplementary cementitious materials with different chemical compositions can replace clinker and produce hydration products with similar characteristics [9]. However, in LBM, especially in renders and plasters, CaO plays a central and unique role. It provides several advantageous properties such as weathering resistance, thermal resistance, reduced water penetration, enhanced breathability and moisture control, increased bond strength and reduced cracking, among others [10]–[12]. Therefore, the search for SR that can provide CaO and replace virgin calcite in LBM is limited. The limited existing research addresses waste utilization in lime-based production [13]–[17] with a focus on SR from paper and acetylene industries. However, a gap remains in quantifying environmental and economic sustainability through rigorous inventory assessment. This paper introduces a novel approach, calculating industrial-scale inventory through literature and patented lab-scale treatments, bridging theory and practical implications. This enhances sustainability evaluation, aiding decisions on resource allocation, process refinement, and improvements.

Another limitation is the insufficient environmental and economic assessments of lime-based renders and plasters throughout their entire life-cycle, including the use phase (Cradle-to-Grave). Most of the existing research focuses primarily on the production stage, neglecting the critical importance of the use phase [18]–[22]. For lime-based construction materials, the use phase is particularly crucial. During this phase, the natural carbonation of portlandite occurs, leading to the sequestration of carbon dioxide and the production of calcium carbonate. This carbonation process serves as the main binding agent, holding all the components in the matrix together. Theoretically, 0.59 kg CO_2/kg $Ca(OH)_2$ can be sequestrated during the material's use phase. The natural carbon sink provided by LBM should be considered in the environmental impact assessment since carbonation is an inherent and necessary reaction for the material to fulfil its intended function.

In the context of striving to fulfil the Paris Agreement, the European Trading System (ETS) has implemented a carbon tax of 90 €/t CO_2 [23]. This has created significant economic pressure for industries to enhance their environmental performance and reduce greenhouse gas emissions. However, the current application of ETS imposes the same tax principle on all industries, regardless of their ability to recapture CO_2 in other stages of their life-cycle. It is worth noting that only a few materials have been extensively documented for their potential to sequester CO_2 during their use phase, with lime being one of them [20]. To ensure fair market competition, the design of balanced carbon prices necessitates political and economic taxing policies at the European level [24]. These policies should integrate the role of products, their properties, and consider tracking the potential CO_2 profile of materials throughout their life-cycle.

This research work aims to assess and compare the environmental and economic performance of lime-based plasters through their entire life cycle, from Cradle-to-Grave. The plasters are manufactured using traditional hydrated lime as well as two secondary resources: paper mill sludge (PMS), a waste from the paper industry, and carbide lime (CL), a by-product from the acetylene industry. The study offers valuable insights into the Cradle-to-Gate/Grave framework, with a specific emphasis on the design of intermediate treatment for preparing secondary resources. It delves into the impact of allocation procedures for CL, as well as the effects of carbonation and maintenance throughout the use phase and the end-of-life scenario. Through an economic assessment the influence of various factors on the overall life-cycle of lime-based plaster is evaluated, including materials manufacturing, maintenance actions, carbon emissions, carbon taxes and potential carbon credits.

5.2.2 Methodology

To address the deficit in quantifying environmental and economic sustainability via thorough inventory assessment, a novel process-based methodology [8] for life-cycle inventory calculations previously proposed by the authors is employed. The approach is applied to implement energy transition scenarios [25] and compare kiln technologies with carbon capture systems[8] in hydrated lime manufacturing, as well as to establish a baseline scenario for the manufacturing of modern lime-based plasters [26]. This research work makes one step forward and calculates industrial-scale inventories by amalgamating insights from pertinent literature and patented lab-scale treatments for upcycling secondary resources in lime-based plasters manufacturing (See section 5.2.2.2). This harmonization bridges the chasm between theoretical constructs and real-world applications, amplifying sustainability evaluation and facilitating informed choices regarding resource allocation, process enhancement, and overall improvements.

The Life-cycle Assessment (LCA) methodology (ISO 14040/44, 2006) is used to calculate the environmental impact over the life cycle of the plasters. Four main steps are performed: 1) definition of goal and scope, 2) inventory analysis, 3) life-cycle impact analysis and 4) interpretation of results [27], [28]. In addition, the economic aspect is addressed through Life-cycle cost (LCC) analysis and is carried out in parallel to the LCA using the same Functional Unit (FU), system boundaries and inventory, including information about the raw materials costs, energy purchase, transportation, among others. [29].

5.2.2.1 Goal and Scope Definition

The goal of this study is to quantify the impact of maintaining covered for 100 years a wall of 1 m^2 with a minimum thermal insulation of 0.01 m^2/kW by using a lime-based plaster (thermal conductivity 0.2-0.56 W/mK) produced by traditional hydrated lime (HL) and two upcycled materials, CL from the acetylene and PMS from paper industry.

The study intents to quantify, assess, and discuss the potential environmental and economic implications of substituting hydrated lime as a binder in dry mixtures with alternative waste/by-products from secondary resources. At present, there is a significant knowledge gap regarding the environmental and economic performance of lime-based building materials, particularly within the context of circular economy, serving as the primary motivation for undertaking this study. The research work is targeting two main groups audience: i) the scientific community and ii) manufacturers of lime-based building materials. As for the scientific community, the research outcomes show the urgent need for further investigation of this particular field of interest. Along with this, it is the intention to communicate directly with manufacturers, urging them to recognize the benefits of integrating waste/by-products into their production processes.

The FU adopted in this study is the amount of plaster required to achieve the designed thermal insulation and service life. The study covers from cradle-to-grave, which means from the production of the raw materials to the use phase and the end of life. The system boundaries are shown in Figure 5.13. In the case of secondary resources, the boundaries are covered from Cradle-to-Gate. Specific details on each case are provided in Section 5.2.3.1.1. During the use phase, the carbon capture potential of the plaster and the maintenance and repair activities are also accounted for. Finally, an economic life cycle cost assessment is considered, related to the selected FU.

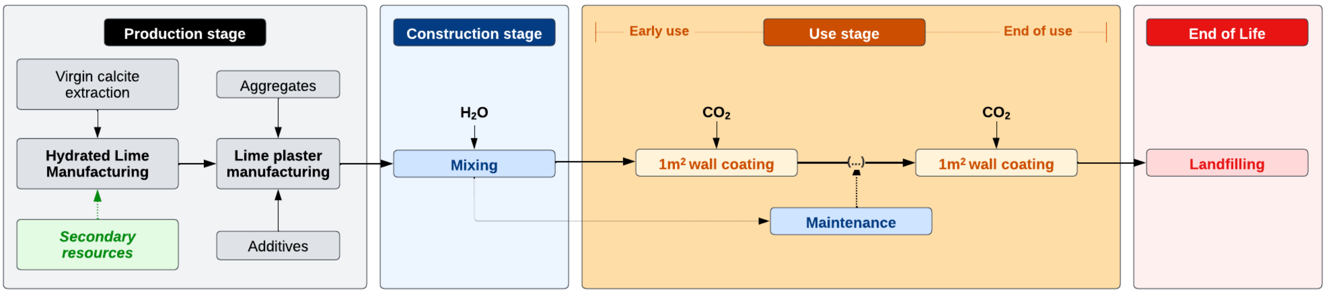


Figure 5.13. System boundaries considered in the Cradle-to-Grave environmental and economic

5.2.2.2 Inventory Analysis

The Life Cycle Inventory (LCI) analysis is a critical phase of the environmental assessment, as the obtained results directly link to the quality of the data used in the LCA [20]. In the cradle-to-gate production stages, all impacts arising from the extraction of raw materials and intermediate treatment of

SR until the gate of the factory (i.e., lime-based plaster ready for use) are accounted. In the upcycling of waste/by-products it is critical to design accurately the intermediate treatment processes and evaluate their impacts, to displace the above-mentioned impacts related to the original material (e.g., for the replacement of hydrated lime by paper mill sludge). At the moment, there are no available datasets to model the intermediate treatments introduced in this article. For the calculation of the inventory, a process-oriented methodology previously introduced by the authors is employed [8]. This methodology involves the calculation of the unit-process mass and energy inventory based on materials and energy balances, process-engineering design, and materials science. Additional validation of the materials flows, equipment capacities and devices were conducted within the context of the EU SUBLime project (https://sublime-etn.eu/), a MSCA network which encompasses the largest European lime producers.

During the inventory analysis, it is also important to determine whether allocation procedures are required for multifunctional processes. The allocations considered in this study are by mass and economic value. A mass allocation coefficient (C_m) and economic allocation coefficient (C_e) using Equation 5.6 and 5.7 can be calculated correspondingly. In these equations, m and € represent the mass and price of main and by-products. In the identified system boundaries of the secondary resource, the C_m and C_e coefficients are applied to the by-product of interest to assign a portion of the environmental impact of the multifunctional process.

$$C_m = \frac{m_{by-product}}{m_{by-product} + m_{main\ product}} \qquad \textbf{Equation 5.6}$$

$$C_e = \frac{(m \cdot \epsilon)_{by-product}}{(m \cdot \epsilon)_{by-product} + (m \cdot \epsilon)_{main\ product}} \qquad \textbf{Equation 5.7}$$

The Cradle-to-Gate system boundaries are expanded to the Cradle-to-Grave and the carbonation of the plaster as well as its durability are considered. Regarding the production costs, they were determined in 2022 through market survey and company perspectives [24].

5.2.2.3 Environmental Life-cycle and Life-cycle Cost Assessment

The software OpenLCA was used to run the environmental and economic calculations. The used database is EcoInvent V3.6 [30]. For the Environmental analysis, Impact 2002+ was selected as impact method since it addresses relevant impact categories of importance in the mining industry, such as Resources, Climate Change, Human Health, and Ecosystem quality [42]. For the calculation of the endpoint categories, the midpoint indicators presented in Appendix 7 were considered.

To evaluate the robustness of the results, a three-step analysis was conducted. Initially, a contribution analysis offered a swift overview, highlighting the most significant contributors to the impact indicators. This step facilitated the identification of critical processes that warranted further investigation.

Subsequently, a sensitivity analysis was conducted, involving diverse scenarios by varying the selected parameters in the inventory.

The sensitivity coefficient (SC), following the formulation proposed by [31] in Equation 5.8, was computed. The SC represents the ratio of two relative changes for the endpoint indicators. A SC of 1 indicates that a 5% increase in the parameter value results in a corresponding 10% increase in the final result. This metric serves as a valuable tool for evaluating the sensitivity of the results to changes in specific parameters, offering insights into the robustness of the findings.

$$SC = \frac{\frac{\Delta result}{initial\ result}}{\frac{\Delta parameter}{initial\ parameter}} \qquad \textbf{Equation 5.8}$$

In the final step, a Monte Carlo analysis was utilized for parameters with an SR equal to or greater than 1, to systematically propagate uncertainties within the LCA model arising from input variations [31].

The LCC methodology is not standardized as LCA and therefore, there is no unified procedure for calculating the costs [29]. To carry out this analysis, the starting point was the inventory of materials and energy considered for the environmental analysis (Section 5.2.3.1). The calculations are performed from the producer’s perspective. For production costs it is enough to consider the purchase price of materials, resources, and energy [38], [43]. The calculation of the carbon cost in 2022, considers the CO_2 emitted during the production of the binder in the mix (traditional or upcycled hydrated lime) multiplied by the carbon price. To discuss the influence of the sequestration of CO_2 during the use phase, a carbon credit (90 €/t CO_2) is introduced in the economic assessment [32].

5.2.3Results and Discussion

5.2.3.1 Life-cycle Inventory

5.2.3.1.1 Manufacturing Stage

The manufacturing stage includes the production of the lime-based plasters with traditional hydrated lime (reference scenario) and HL generated by upcycling PMS and CL (alternative scenarios). The datasets used to model the life-cycle inventory of the plasters is shown in Appendix A7. The specific inventory of the reference lime-based plaster with HL, is part of a previous work by the authors and can be consulted in Table 4.4. In this section we focus specifically on the inventory calculation of the SR upcycling.

Paper mill sludge and carbide lime are materials generated because of the production of paper and acetylene, respectively. A simplified flowchart of each production process is shown in Figure 5.14. For the case studies, a full replacement of HL in the inventory of Table 4.4 was assumed (i.e., 0.25t), either by recycled paper mill sludge (RPM) or by recycled carbide lime (RCL), generated after the intermediate treatment. As a result, the plasters RPMBP and RCLBP are obtained correspondingly.

Paper mill sludge

The pulp and paper industry plays an integral role in the global economy. Approximately 400 million tons of paper and paperboard are produced globally [33]. According to the last available report by the Renewable Recycled Responsible European Paper Association, in 2020 in Europe the amount of Paper and Pulp companies was 683, with a total production of 116933 MTonnes. Germany is the biggest producer (25.1%) followed by Sweden (11%), Italy (10%), Finland (9.6%), France (8.1%) and Spain (7.4%) [34]. The production of paper (Figure 5.14a) involves a high consumption of energy, water, resources and generates a significant environmental impact [35].

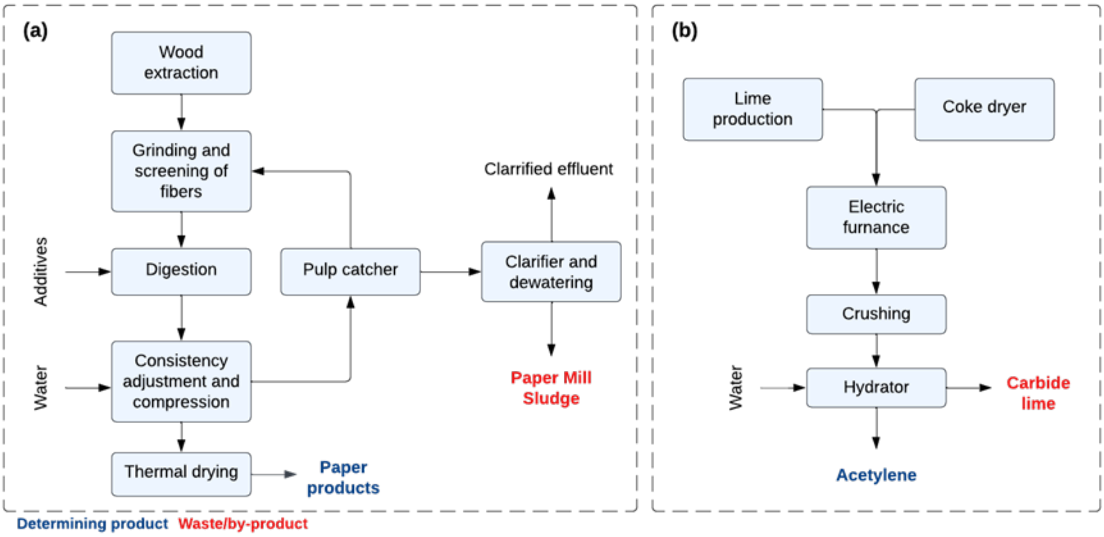


Figure 5.14. Production system of paper products and paper mill sludge (a) and production system of acetylene and carbide lime (b).

It starts from the extraction of the wood and grinding to produce wood fibers. The pulp fibers are pretreated to give them the properties required for the specific type of paper. During the process of paper production, $CaCO_3$ is added to the pulp, to give the paper a whiter color. Chemicals such as organic fillers (starch, latex), colors, aluminum sulfate, etc. are used to make paper with different properties. Afterwards the suspension is dewatered, pressed, and dried. Different paper shapes in form of rolls or packs of sheets are obtained [36].

PMS represents around 87% of the output of the wastewater treatment [37], [38]. During the chemical recovery process, smelt or chemicals are dissolved in water in their molten stage, to form green liquor, cauterized by reacting Na_2CO_3 with CaO and producing NaOH along with $CaCO_3$ (lime sludge). In terms of production of paper and pulp mill sludge, around 40 to 50 kg of sludge (dry) is generated in the production of 1 ton of paper [33]. The most common management practice for this residue has always been the landfilling disposal of the sludge (around 70% of the total generated) [39], [40]. Therefore, it is a reasonable assumption to treat PMS as a waste and no environmental burdens from the paper industry shall be allocated to PMS.

Carbide lime

Acetylene (C_2H_2) is typically used as welding heat source and fuel (calorific value of 56 kJ/m^3) and plays a crucial role in the manufacturing of several products such as vinyl chloride, 1,4-butanediol, acetaldehyde, many types of esters and ethers among other products [41], [42]. Although there are other methods, the calcium carbide process is universally and traditionally employed (Figure 5.14b) [43], [44]. Calcium carbide is produced from calcium oxide and coke (Reaction 5.1) in an electric furnace at 2300°C. Then the calcium carbide is mixed with water to produce acetylene and CL, which is composed by calcium hydroxide, as represented in Reaction 5.2 [41].

$$CaO\ (s) + 3C\ (s) \rightarrow CaC_2\ (s) + CO\ (g) \qquad \textbf{Reaction 5.1}$$

$$CaC_2\ (s) + 2H_2O\ (l) \rightarrow C_2H_2\ (g) + Ca(OH)_2\ (l) \qquad \textbf{Reaction 5.2}$$

The CL by-product is generated as an aqueous slurry and is composed by calcium hydroxide ($Ca(OH)_2$≈85–95%) with minor parts of calcium carbonate ($CaCO_3$≈1–10%), unreacted carbon and silicates (1–3%)[45]–[47].

The market for acetylene in Europe is quite small, China being the major producer of acetylene worldwide (7.62 million tons in 2018) [45]. Carbide lime has several uses, as raw material of polyvinyl chloride (PVC), a substitute of lime for agricultural purposes and several industrial processes [15], [48]-[51]. CL fulfills all the requirements of the EU directive 2008/98/EC28 to qualify for by-product status: (1) further use of the substance is certain; (2) the substance is produced as an integral part of a production process; (3) the substance can be used directly without any further processing other than normal industrial practice; and (4) further use is lawful [52]. Therefore, it is fair to allocate part of the environmental load of acetylene production to the lime-based plasters producer. In this case three procedures are applied: no allocation, allocation by mass (Eq. 5.6) and allocation by economic value (Eq. 5.7). The allocation coefficients considered for carbide lime production are shown in Table 5.3. Market prices are the result of averaged values collected in 2022 through market survey.

Table 5.3. Allocation coefficients for carbide lime

Product	Mass produced	Market price	Mass allocation	Economic allocation
Acetylene	1 kg	20.75 €/kg	26.04	0.99
Carbide Lime	2.84 kg	0.02 €/kg	73.96	0.01

5.2.3.1.2 Use of secondary resources in lime-based plasters manufacturing

Table 5.4 summarizes a literature review of chemical composition ranges along physical properties and a comparison of different criteria for HL, PMS and CL.

Table 5.4. Ranges of chemical composition for hydrated lime, paper mill sludge and carbide lime and comparison criteria of the waste or by-product to be used as hydrated lime replacement.

	Hydrated Lime (HL)	Paper mill sludge (PMS) from paper industry	Carbide lime (CL) from acetylene industry	Sources
		Chemical composition (wt.%) and physical properties		
SiO_2	0.13	2 – 20	3.0 – 5.0	[47], [48], [53]-[57]
Al_2O_3	0.06	0.8 – 5	0.4 – 1.5	
Fe_2O_3	0.07	0.25 – 2.5	0.59 – 1	
CaO	98.53	60 – 90	55.0 – 92.0	
MgO	1.09	0.2 – 10	0.14 – 1.3	
Na_2O	-	-	0 – 0.1	
K_2O	0.01	-	-	
TiO_2	-	-	0 – 0.7	
SO_3	-	-	0.5 – 0.7	
Ignition loss	0.1	14 ± 56	10 - 40	
Humidity (%wt.)	-	28.4 ± 11.0	30.0 ± 5.0	
Organics (%wt.)	-	34.9 ± 21.1	9.0 ± 5.0	
		Comparison criteria for the upcycling of PMS and CL		
Origin and potential availability of the resource in Europe		The origin is from the paper mill industry. Available in Germany, with a share of around 25% of the European market. Around 880000 – 1100000 tons of dry sludge per year in Europe.	The origin is from the acetylene industry. The market is dominated by China (+21 Million Tons) and no significant production is carried out in Europe.	[47], [58], [59]
Current final disposition of the waste		Mainly landfilled	By-product utilized in polymeric industries among others.	[33], [43], [44]
Allocation procedure required		No (it is a waste)	Yes (it is a by-product)	-
Market competition of the construction industry with other industries		Agriculture and heat or power generating plants	PVC industries, agriculture, and civil constructions.	[39], [43]
Potential role in the lime-based mortars/plasters mix		As a source for hydrated lime production, potential pozzolanic activity due to the presence of amorphous silica phases.	As complete replacement of hydrated lime in the dry mix.	[31], [54]
Potential pretreatment required to be upcycled		Removal of organics, humidity, thermal activation of calcite and lime slaking.	Removal of organics, humidity and traces of sulfides and heavy metals (Pb^{+2}, Hg^{2+})	[37], [43]
Potential problems that can limit the application		Presence of organic compounds and variability of the chemical composition.	Presence of sulfides, organic compounds, and heavy metals.	-

Both PMS and CL share compatible chemical compositions with traditional HL, indicating they could replace the conventional binder. However, their distinct physical properties influence how they should be conditioned for SR and the energy needed for processing. PMS is mostly $CaCO_3$, potentially substituting virgin calcite, while CL, consisting of $Ca(OH)_2$ (80%), could directly replace portlandite in HL production. The intermediate treatment strategies for PMS and CL are outlined below.

Intermediate treatment design of PMS

Figure 5.15 shows the proposed intermediate treatment for PMS preparation, considering an average transportation distance of 200 km to the LBM manufacturing plant. To calculate the LCI, the initial PMS composition (Table 5.4) was averaged: 75% CaO, 15% SiO_2, 5% Al_2O_3, and 5% MgO. Initial humidity (20% of total mass) and organics (40% of dry mass) were removed through pyrolysis, with the main mass loss taking place within the temperature range of 300-400°C. Subsequently, thermal activation of PMS at 800-900°C follows [60]-[62]. These processes take place in a lime kiln, used to model the water removal, organic material removal, and calcium oxide synthesis [25].

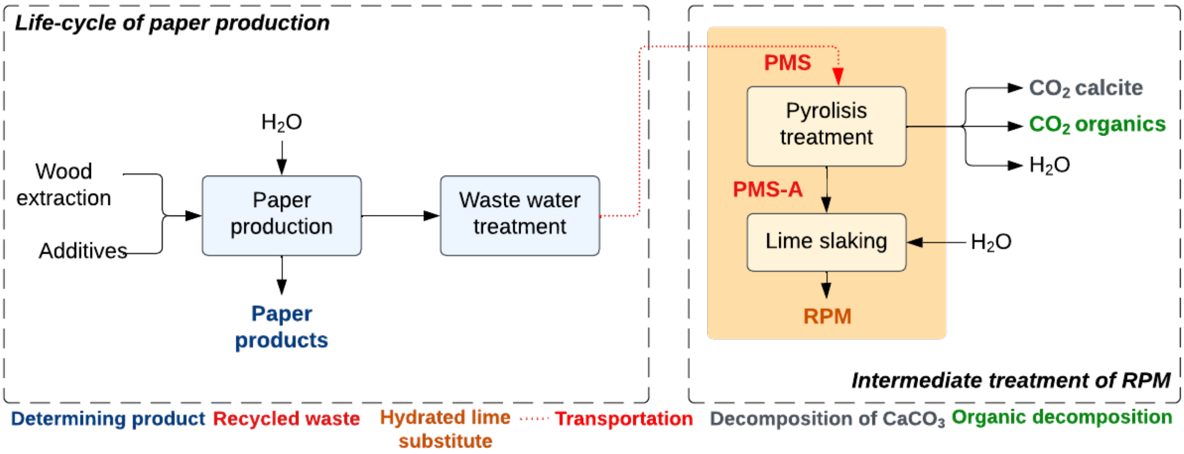


Figure 5.15. System boundaries to produce a recycled lime-based binder from paper mill sludge. *PMS: Paper Mill Sludge. PMS: Paper Mill Sludge; PMS-A: Paper Mill Sludge Activated; RPM: Recycled Paper Mill Sludge*

The main emissions in the lime kiln are water vapor and carbon dioxide from calcite decomposition and organic matter, calculated stoichiometrically (Reaction 5.3 and 5.4 respectively). The heat released from combustion of organics is considered as an energy credit for the lime kiln, which reduces the total energy demand per mass of processed material, in comparison to regular hydrated lime production (2640 MJ/t vs 2040 MJ/t).

$$CaCO_3\ (s) \rightarrow CaO\ (s) + CO_2\ (g) \qquad \Delta H_{rc}{}^{\circ} = 177.3\ kJ/mol \qquad \textbf{Reaction 5.3}$$

$$C\ (s) + O_2(g) \rightarrow CO_2(g) \qquad \Delta H_{rc}{}^{\circ} = -393.5\ kJ/mol \qquad \textbf{Reaction 5.4}$$

The lime slaking is modelled following a previous work by the authors [25] and the LCI of the intermediate treatment is provided in Table 5.5. After the intermediate treatment, the final composition of the PMS is 80% $Ca(OH)_2$, 10% SiO_2, 5% Al_2O_3, 5% MgO.

Table 5.5. Life-cycle Inventory to produce 1 t of recycled lime-based binder from paper mill sludge

Operation/ process modelled		Processed amount		Specific Inventory Amount		Sources & Notes
Process Step: Pyrolysis						
Input	**PMS**	2.02	t	1	t	
	Transportation	404	tkm	200	km	Assumed
	Kiln fuel consumption	2100.8	MJ	1040	MJ/t PMS	2640 – 4MJ/kg * 400 kg (cellulose). Fuel mix Germany 2020
	Kiln Electricity operation	54.74	kWh	27.10	kWh/t PMS	Electricity mix Germany 2020 [25]
Output	**PMS-A**	0.81	t	-	-	Activated PMS
	Water vapor (humidity removal)	0.33	t	0.2	t H_2O/t PMS	Calculated stoichiometrically (Humidity 20%)
	CO_2 (organic decomposition)	2.46	t	1.46	t CO_2/t PMS	Assumed 40% organic matter (Reaction 5.4, do not count as emissions)
	CO_2 (calcite decomposition)	0.47	t	0.58	t CO_2/t PMS-A	Calculated stoichiometrically (Reaction 5.3)
Process Step: Lime slaking						
Input	**PMS-D**	0.81	t			
	Water	0.19	t	0.32	t H_2O/t CaO	Calculated stoichiometrically (0.32 t H_2O/tCaO)
	Hydrator Electricity operation	0.35	kW	0.35	kW/t	Electricity mix Germany 2020[25]
Output	**RPM**	1	t	-	-	0.8 t $Ca(OH)_2$/tRPM

Intermediate treatment design of CL

The intermediate treatment for carbide lime upcycling is shown in Figure 5.16, based on a patented laboratory purification process [63]. Calcium carbide is hydrated with excess water to produce acetylene, yielding a paste with approx. 65% water content, and dispersed solid components. To obtain 1 ton of RCL, 3.14 tons of carbide lime are required. The initial purification occurs in a sedimentation tank, where the gravity-driven decantation separates solids, yielding a calcium hydroxide-saturated solution. The solid composition averages 85% portlandite, 10% calcite, and 5% impurities (sulfides and organic matter), as per [63].

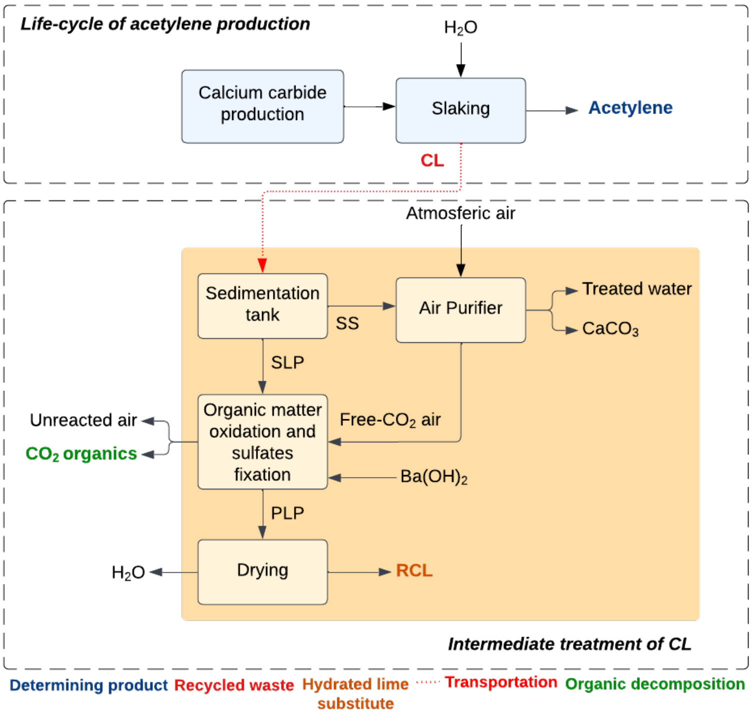


Figure 5.16. System boundaries to produce a recycled lime-based binder from carbide lime. CL: Carbide Lime; SS: Saturated Solution; SLP: Solid Lime Paste; PLP: Purified Lime Paste; RCL: Recycled Carbide Lime

Atmospheric air is purified by bubbling it through a hydroxide-saturated solution within a lime precipitator, to treat the impurities in the solids (i.e., removing organics and sulphates). Carbon dioxide (CO_2) from the air reacts with the portlandite in solution, yielding calcium carbonate and a gas mixture of nitrogen (N_2) and oxygen (O_2). Through recirculation via a diaphragm pump, the gas undergoes multiple cycles, effectively eliminating CO_2. The resulting calcium carbonate can be separated via sedimentation for subsequent reuse as an aggregate/filler (e.g., in lime-based plasters).

The CO_2-free air stream is then introduced (bubbled) into an agitated reactor, which promotes oxidation of sulfides to sulphates and degradation of organic matter, avoiding carbonation of the CL paste. Organic oxidation aligns with Reaction 5.4. To counteract potential sulfate precipitation during usage and its detrimental impact on plaster, barium hydroxide is employed. This compound induces the formation of highly insoluble barium sulfate, also capable of incorporating heavy metals into its structure. Calculations for barium hydroxide usage consider sulfide content in the solids according to Reaction 5.5 to 5.7.

$$S^{2-} + 2O_2 \rightarrow SO_4^{2-}$$ **Reaction 5.5**

$$Ba^{2+} + SO_4^{2-} \rightarrow BaSO_4$$ **Reaction 5.6**

$$Ba^{2+} + 2(OH)^{-} \rightarrow Ba(OH)_2$$ **Reaction 5.7**

The final step of the treatment requires drying the paste. The LCI of the RCL production is shown in Table 5.6. After the intermediate treatment, the final composition of RCL is 89.5% $Ca(OH)_2$ and 10.5% $CaCO_3$.

Table 5.6. Life-cycle Inventories to produce 1 t of recycled lime-based binder from Carbide Lime.

Operation/process modelled		Processed amount		Specific Inventory Amount		Sources & Notes
Sedimentation tank						
Input	**Carbide Lime (CL)**	3.14	t	1	t	-
	Stirrer	0.59	kWh	0.19	kWh/t	Calculated
	Transportation	628	tkm	200	km	Assumed
Output	**Solid Lime paste (SLP)**	1.1	t	0.35	tSLP/tCL	
	Saturated Solution (SS)	2.04	t	0.65	tSS/tCL	Solubility of $Ca(OH)_2$ 1 g / 630 g
Air purifier						
Input	**SS**	2.04	t	-	-	Calculated
	Atmosferic air (AA)	6	t	2.94	t AA/tSS	0,03% CO_2, 21% O_2, 79% N_2
	Air pump	4.5	kWh	0.75	kWh/t	Laveglia et al. [25]
Output	**Purified air (PA)**	5.99	t	-	-	CO_2 removal
	Calcite	0.004	t	-	-	Precipitated $CaCO_3$
	Treated Water	1.89	t	-	-	
Organic matter oxidation and sulfates fixation						
Input	**SLP**	1,1	t	-	-	
	PA	5.99	t	5.44	t PA/tSLP	21% O_2, 79% N_2
	Stirrer	0.21	kW	0.19	kWh/t	Laveglia et al. [25]
	$Ba(OH)_2$	1.83E-6	t	-	-	Calculated as the amount of S present (6239 PPM* 21 kg)
Output	**Purified Lime paste (PLP)**	1.1	t	-	-	10% humidity
	CO_2	0,08	t	-	-	Organic decomposition
	Air	5.93	t	-	-	After reaction with oganic
Drying						
Input	**PLP**	1,10	t	-	-	
	Heating	220	MJ	200	MJ/t	-
Output	**RCL**	1	t	-	-	(0.89 t $Ca(OH)_2$/tRCL)
	H_2O	0,10	t	-	-	Water Vapor

Functionality performance of the plasters

HL is completely replaced by the upcycled materials, RPM and RCL in the mix design (Table 4.4, P6). For the environmental and economic calculations, it is supposed that all the plasters exhibit a comparable thermal performance according to the functional unit defined (Section 5.2.2.1). The first reason is that for the reference and upcycled plaster formulations, the proportion of hydrated lime falls within the typical 15-25% in the dry mix of commercially available materials [26]. Hydrated lime itself does not significantly contribute to insulation, but provides matrix cohesion, building breathability and moisture transport [64], [65]. Lowering the effective hydrated lime content can increase the porosity in the matrix, which does not negatively affect the thermal insulation properties. The second reason is that insulation properties are predominantly determined by the introduction of natural and artificial lightweight aggregates (highly porous) and additives such as air entrainers (Table 4.4, P6) that introduce air bubbles in the system, rather than the binder concentration [66], [67]. Although these assumptions were validated within the scope of the SUBLime network, further data refinement through experimental characterization of the upcycled plasters is required.

5.2.3.2 Use phase and End of Life

A density of 1.1 kg/L plaster and a durability of 50 years was assumed according to declarations from producers [68]–[71]. During the service life, CO_2 is absorbed reacting with portlandite to generate calcium carbonate, which is the main responsible of the hardening of the plaster (Reaction 5.8).

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$ **Reaction 5.8**

The diffusion of the CO_2 through the plaster (i.e., CO_2 sequestration) can be simplified by a diffusion-like process (Equation 5.9). Equation 5.10 is used to calculate the kg of CO_2 sequestrated per functional unit.

$$x = k\sqrt{t}$$ **Equation 5.9**

$$SqC = 0.594 * FCH * \frac{x}{X_{total}}$$ **Equation 5.10**

where x (mm) is the carbonated thickness of the plaster at a given time t (days), k (mm/day$^{0.5}$) is the diffusion coefficient of CO_2, SC (kg CO_2/m^2) is the mass of CO_2 sequestrated per area of coated wall, 0.594 is a conversion factor (molecular weight ratio $CO_2/Ca(OH)_2$), FCH (kg $Ca(OH)_2/m^2$) is the amount of hydrated lime per area of coated wall, X_{total} (mm) is the total thickness of the plaster.

The adopted parameters are shown in Table 5.7. The k coefficient is an average of values reported by [18], [20]. With a wall's expected service life spanning 100 years, and a full plaster replacement after 50 years, the old (discarded) plaster underwent transportation over 100 km to its final disposal site. In the End-of-Life phase, HLP, RPMP and RCLP plasters are considered inert because of their chemical composition (mostly composed of SiO_2 and $CaCO_3$). To model the 'grave' scenario, the landfilling of the plasters is considered and the closest available dataset (treatment of waste concrete, inert material landfill | waste concrete | APOS, S) from EcoInvent Database V3.6 was selected.

Table 5.7. Parameters considered for the carbonation of the plasters during the use phase.

Plaster	kg plaster/ m^2	FCH (kg $Ca(OH)_2/m^2$)	D (mm)	k ($mm/day^{0.5}$)	Full carbonation time (days)	SqC (kg CO_2/m^2)
HLP	9.66	2.41	8.70	1.00	76	1.43
RPMP	9.66	1.93	8.70	1.00	76	1.15
RCLP	9.66	2.14	8.70	1.00	76	1.27

5.2.3.3 Life-cycle Cost Inventory

Regarding costs, a unified approach was followed, the incorporating recommendations from both the NIST Life Cycle Cost manual and the SETAC Life Cycle Cost code of practice [72]. Costs generally fall into three categories: (a) Direct production costs (e.g., raw materials, energy procurement); (b) Indirect cost (e.g., treatment of industrial residues); and (c) Externalities (linked to pollutant-related taxes, such as CO_2 emissions). Direct costs in this study focus on variable costs, those fluctuating with production changes, excluding fixed costs that remain constant regardless of production level (e.g., equipment purchase, salaries, total taxes). Notably, variable production cost components include purchase of materials transported to the plant, transportation costs related to raw material purchase and electricity consumption at the mixing plant.

Specific cost breakdowns per unit for these items are outlined in Table 5.8, based on a 2022 market survey in Germany [26]. While production costs for hydrated lime are detailed, carbon taxes are excluded. Carbide Lime is assigned a by-product cost, whereas Paper Mill Sludge acquisition bears no cost.

Table 5.8. Cost inventory of the plasters' life cycle [26].

Group		Item	Unit of measure	Unit costs
Purchase of Materials	Binders	Hydrated Lime	€/kg	0.11
		Carbide Lime	€/kg	0.02
	Aggregates	Sand	€/kg	0.01
		Pumice	€/kg	0.06
		Polystyrene	€/kg	1.29
	Additives	Carboxymethyl celullose	€/kg	0.97
		Alkylbenzene sulfonate	€/kg	0.80
		Polycarboxyllate	€/kg	0.65
		Ethylene vinyl acetate	€/kg	1.12
	Others	Barium Hydroxide	€/kg	36.00
Transportation		Truck transportation	€/t.km	0.06
Electricity consumption		Electricity	€/kWh	0.11
End of Life		Sanitary landfilling	€/kg	0.02

The research also evaluates the costs associated to CO_2 emissions within the EU emission system [32]. Prior research by the authors found that the current carbon price (90 €/t) can contribute up to 30% of total lime-based plaster production costs [26].

During the use phase of the material, the spontaneous carbonation of plaster's portlandite (sequestration) is essential for hardening, gaining strength and functional performance. The European Lime Association underscores the significant impact of re-carbonation of CO_2 emissions from manufacturing stage on environmental performance of lime-based materials [74], though its economic implications remain unaddressed. A balanced ETS might integrate material's CO_2 sequestration capacity over their lifespan as a credit, encouraging investments in cleaner production systems. In this study, *Carbon cost* is initially calculated by multiplying CO_2 emissions from binder production (derived from LCA results) by the 2022 carbon price (90 €/t). *Carbon credit* is assessed by multiplying sequestrated CO_2 (as shown in Table 5.7) by the same carbon price.

5.2.3.4 Environmental Life-cycle Impact Assessment

The environmental life-cycle impact assessment section is divided in two parts: a Cradle-to-Gate analysis and a Cradle-to-Grave analysis. The first part focuses on the production process of the binders (HL, RPM and RCL) to compare the impacts of the traditional HL scenario with the intermediate treatments of the SRs. In the second part the boundaries are expanded to consider the environmental performance when coating 1 m^2 of wall with the plasters from the cradle to the end of life.

5.2.3.4.1 Cradle-to-Gate of the Factory

Figure 5.17 presents impact assessment results for RCL production and various allocation procedures applied to carbide lime production in the acetylene industry. In the case of the CL intermediate treatment itself, shown in Fig. 5.16 (no allocation applied), transportation is a primary contributor in each indicator, accounting for 50-70% (due to lorry fossil fuels combustion). The second significant contributor is the electricity-consuming drying process to achieve desired humidity in treated RCL. Notably, CL treatment avoids kiln operations (unlike PMS), leading to significant emission reduction by sidestepping calcite decomposition and fuel combustion. Specifically, RCL achieves a remarkable 94% reduction in Climate Change impact compared to HL production (≈0.94 kg CO_2/kg HL, [25]).

The acetylene production process is widely recognized for its significant environmental impact [53]. Application of economic and mass allocation methods to carbide lime production yields notable effects on environmental impact, as evident in Figure 5.17 (Table 5.3). Specifically, the economic allocation approach brings about a moderate increase, while mass allocation imposes substantial environmental impact on the industrial by-product. This is primarily due to the energy-intensive and CO_2-emitting nature of the main precursor CaC_2, synthesized from coal in acetylene production [43]. Mass allocation could discourage CL usage as a hydrated lime replacement, disproportionately attributing impacts to the by-product rather than the main product (2.84 kg CL/kg acetylene). Thus, allocating the environmental impact of acetylene production to CL by mass is deemed inequitable. The economic allocation approach emerges as the preferable choice, acknowledging carbide lime's by-product status while achieving balanced impact allocation to RCL production. Hence, this approach is further applied to carbide lime production for RCL manufacturing.

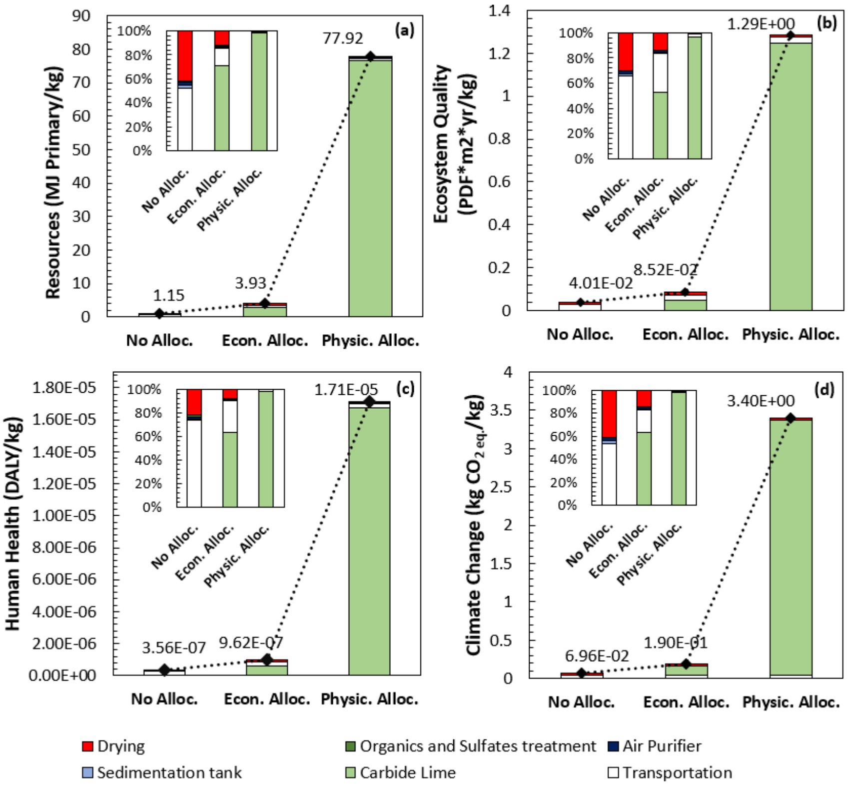


Figure 5.17. Cradle-to-Gate LCA results of producing 1 kg of RCL with and without application of allocation coefficients. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) endpoint indicator values and percentual contribution of each production step.

Figure 5.18 presents the results of the endpoint impact indicators to produce RPM compared to HL and RCL (economic allocation). A percentage contribution of different processes is also provided in each impact category. For simplification, in the case of RCL with economic allocation (RCL (Ec.)), the impact corresponding to economic allocation of the acetylene process and the intermediate treatment of carbide lime itself have been aggregated (for details refer to Fig. 5.17).

Regarding Resources, Ecosystem Quality and Human Health indicators (Fig. 5.18a-c), these metrics consider non-renewable energy, particularly sensitive to fossil fuels. Kiln energy consumption is modelled as the current fuel in Germany (85% non-renewable) and electricity mix (55% non-renewable) [25]. For RPM and HL, in 3 out of 4 impact categories, pyrolysis operations dominate due to this factor, evident in both HL and RPM production. For RPM, transportation of PMS also significantly contributes to these indicators due to lorry fossil fuels combustion (30-40%) [67]. However, the overall impact magnitude for RPM is lower compared to HL production, primarily due to credit from organic matter combustion in the kiln (Table 3). In the case of RCL (Ec.), the Resources impact category is around 7% higher than RPM

(13% lower than HL), because of the allocated impact from acetylene production which consumes high quantities of fuel in the kiln, as explained before. In the Climate Change category (Fig. 5.18d), RPM production results in a 14% reduction in CO_2 emissions compared to HL. The main reason behind is that in the RPM treatment, 19% less CO_2 due to calcite decomposition is generated compared to HL (0.47 $kgCO_2/kg$ RPM Vs. 0.58 $kgCO_2/kg$ HL, Table 5.5). Further reduction is achieved due to the lower energy consumption for RPM, due to the credits of the organic matter (Table 5.5). Nevertheless, the CO_2 from the transportation operation diminishes the overall CO_2 reduction (Fig. 5.18d). Further information on the midpoint indicators employed to calculate the endpoint categories for RCL (Ec.) and RPM is provided in Appendixes 8 and 9 respectively.

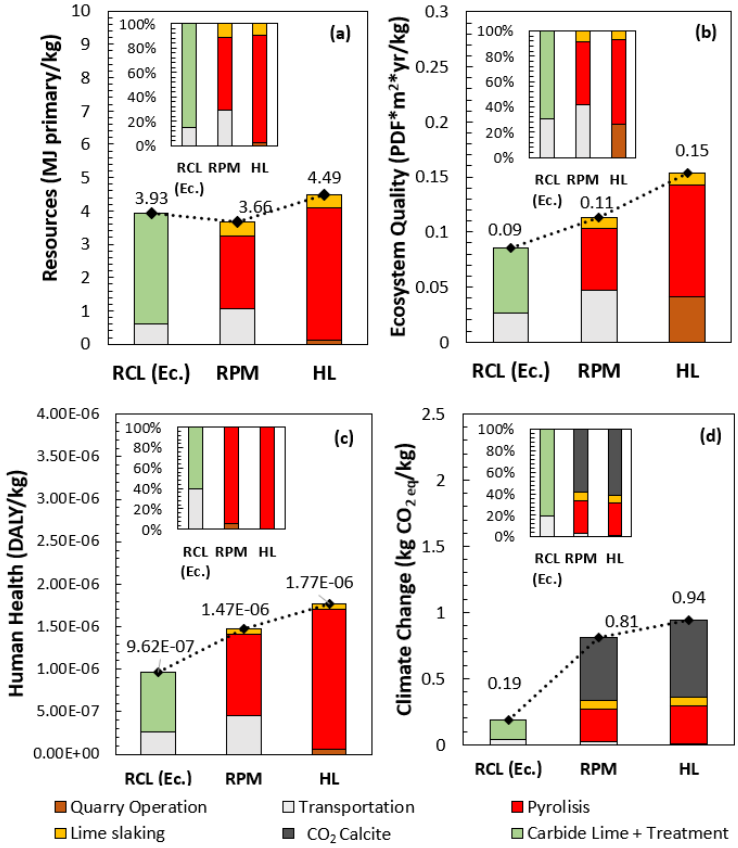


Figure 5.18. Cradle-to-Gate LCA results of producing 1 kg of RCL (Ec.), RPM and HL. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) endpoint indicator values and percentual contribution of each production step.

All in all, from Cradle-to-Gate it is observed that the upcycling of the secondary resources can contribute to reduce the environmental impact in all endpoint categories, in comparison to HL. The climate change category deserves special attention, as 80% CO_2 reduction relative to HL can be achieved by upcycling carbide lime (Fig 5.18d). Even after applying an economic allocation in the acetylene production process (Fig. 5.17d), the absence of a kiln operation (in which calcite is decomposed) for the intermediate treatment of CL, has a significant positive effect. However, it is important to highlight that the availability of secondary resources as close as possible to the production plant play a critical role, in terms of logistics but also costs and environmental impact due to the transportation required.

5.2.3.4.2 Cradle-to-Grave

Figures 5.19 and 5.20 present the Cradle-to-Grave environmental impact assessment results for RPMBP and HLP, and RCLBP and HL lime-based plasters, respectively. The midpoint indicators used for the calculations can be consulted in Appendix 10. The temporal environmental impact commences at year 0, as the time in which the plasters are produced. The assessment integrates main life cycle stages, including manufacturing (initial plaster production), use (complete plaster replacement, i.e., maintenance, and landfilling of the used plasters, after 50 years of service) and end-of-life (plaster landfilling). For the Climate Change category, CO_2 sequestration by portlandite during the use phase is also considered (Table 5.7). The left axis showcases specific environmental impact at each action point (e.g., material production, CO_2 sequestration by carbonation of the plaster, or maintenance), while the right axis illustrates cumulative environmental load across the life cycle, spanning from time 0 to end-of-life (100 years).

Scenario 1: Upcycling of paper mill sludge

Resources category (Fig. 5.19a)

In the manufacturing phase, the dominant contributor to the indicator (40% of total) is the production of the binder (RPM and HL) mainly due to non-renewable energy consumption during calcination. The process intensity ($MJ_{Primary}$/kg material) of RPM is 11% lower than HL. The second significant influence (35%) arises from additive production, notably Water Retention Agent (WRA) and Dispersion Agent (DA) (0.11 $MJ_{surplus}$/kg of WRA and 0.05 $MJ_{surplus}$/kg of DA). Fine aggregate production accounts for approx. 16% of the total, primarily attributed to sand drying (Appendix A1), which demands relatively high energy input (around 70 kW/t).

After 50 years, plaster maintenance replacement, elevates the total indicator by 47% and 49% for RPMBP and HLP, respectively. Landfilling the demolished plaster (after the maintenance action) contributes 10% to the indicator. Its impact predominantly stems from transportation and landfill. Ultimately, at the end of life (100 years), no significant differences in the endpoint indicator are observed between RPMBP and HLP (with RPMBP demonstrating a 4% reduction).

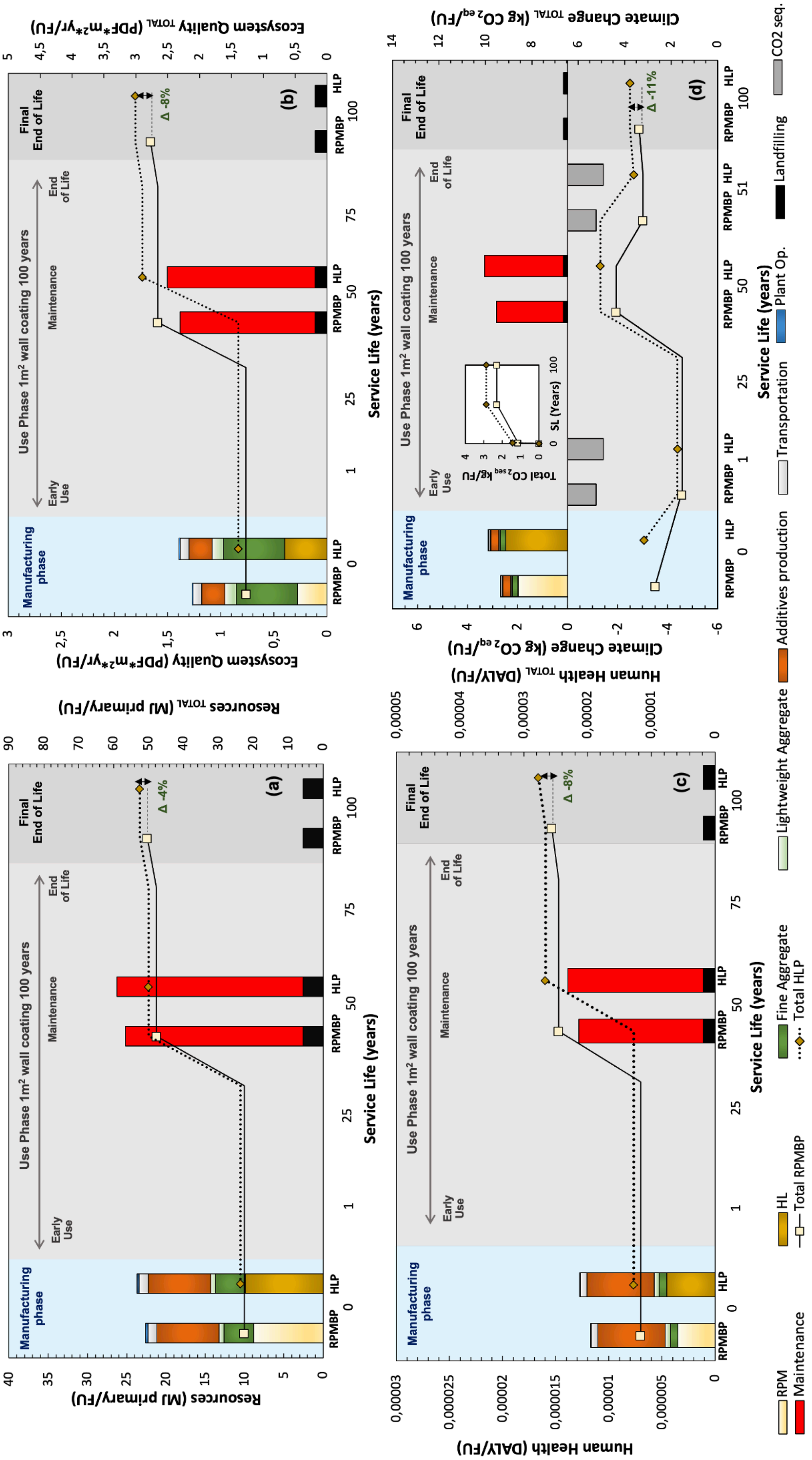


Figure 5.19. Results of the Cradle-to-Grave LCA of RPMBP against HLP. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) impact indicators

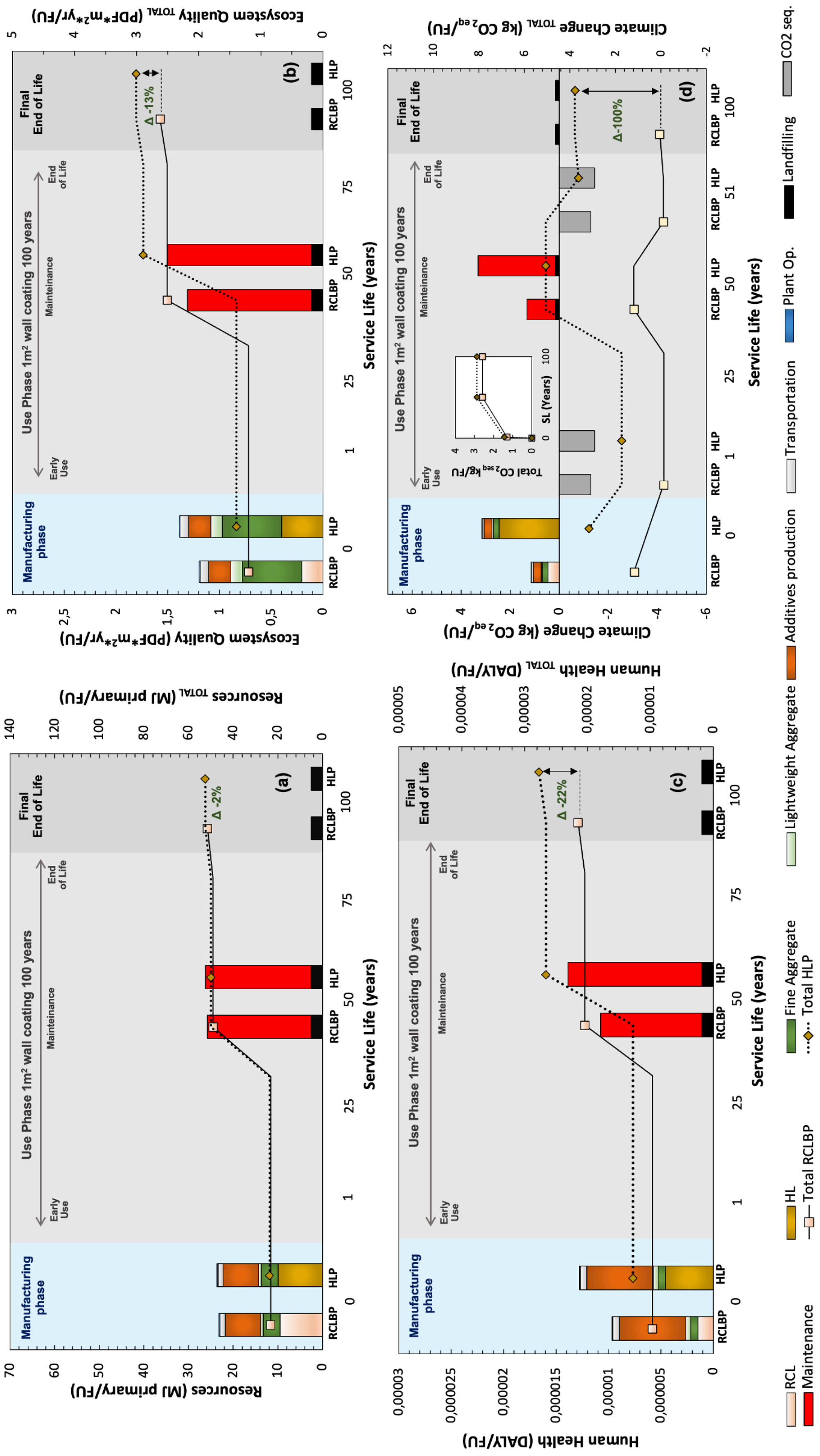


Figure 5.20. Results of the Cradle-to-Grave LCA of RCLBP against HLP. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) impact indicators

Ecosystem Quality category (Fig. 5.19b)

The production of fine aggregate in the manufacturing stage of both plasters, has the greatest share in the ecosystem quality indicator (45% for RPMBP and 42% for HLP). The reason is the high amount of sand used in the inventory (75% in mass) and the significant consumption of electricity, of which 26% is produced by burning coal according to the used electricity mix for Germany [26]. Second main contributor is the production of the binders (21% for RPMBP, 29% for HLP), again because of the emissions during the calcination operation, and particularly because of the hard cold combustion in the kiln (56% of the share). Among the additives used, despite the small quantities in the inventory, the most contributing one is the WRA, followed by the DA (76% and 17% of the additives' contribution, respectively). Lightweight aggregate (LWA) production and transportation contributes to a similar extent in both materials. For LWA, emissions of aluminium to soil during the manufacturing are mostly responsible for the impact. For transportation, the combustion of the fuel is the main contributor. After the first maintenance, the total indicator for both cases increase with around 49%. When the end of life is reached, the indicator ecosystem quality indicator is 8% lower for RPMBP compared to the traditional plaster HLP.

Human Health category (Fig. 5.19c)

Among the contributors to this indicator, it is noted that the additives take a large share in both cases, being 54 and 49% of the total for RPMBP and HL respectively. The explanation is the high release of polycyclic aromatics to the environment during the production of DA and WRA[25]. The binders' production ranks second regarding its share in the overall impact indicator, the fuel combustion in the kiln again being the main contributor, specifically the combustion of coal and of natural gas [25]. The maintenance action shows a similar increment for both materials compared to the manufacturing stage (49%). Same as in the Ecosystem Quality indicator, the overall impact can be reduced by 8% when HL is replaced by RPM.

Climate Change category (Fig. 5.19d)

The binders dominate the emissions of CO_2, being 74 and 77% of the total for RPMBP and HL respectively. As observed in the Cradle-to-Gate analysis (Section 5.2.3.4.1), the lower CO_2 emissions from calcite decomposition and fuel combustion lead to 15% reduction when HL is replaced by RPM in the production of plasters. Additives added to a mix may cause an increase up to 9% of the indicator, even though their proportion in a mix is lower than 3% of the total amount of binder (i.e., 3% of 25%) [26].

During the use phase, the portlandite present in the plaster sequestrates CO_2, leading to the hardening of the material by $CaCO_3$ formation. As shown in Table 5.7, during the first year of use, both plasters carbonate for 100%, removing 1.43 and 1.15 $kgCO_2/m^2$ for RPMBP and HL respectively. This is considered as a credit in the total environmental impact during the first year, and a reduction of 59% and 54% is obtained for RPMBP and HLP respectively (Fig. 5.19d). The maintenance stage and the disposal of the plaster at 50 years increases the Climate Change indicator by 40% for RPMBP and 29% for HLP, relative to the initial manufacturing stage. If no carbonation would have been considered, the increments would have been 52% and 43% respectively. After the replacement of the plasters takes place, the new material carbonates, sequestrating the same amount of CO_2 as in the early use.

All in all, when the final end of life is achieved, the use of RPM as HL replacement leads to around 11% reduction in the CO_{2eq} emissions. The total CO_2 sequestrated during the service life of the wall coating, considering the maintenance period, is 2.30 (RPMBP) and 2.87 (HLP) kg CO_2/FU.

Scenario 2: Upcycling of carbide lime

Resources category (Fig. 5.20a)

As was previously observed in the Cradle-to-Gate analysis of the RCL (Section 5.2.3.4.1), the production of acetylene demands a high consumption of energy and when applying the economic allocation procedure, the use of recycled carbide lime as HL replacement does not produce significative improvements. Allocating 1% of the Resources impact from acetylene industry to CL production generates almost the same burdens as the extraction of virgin calcite (1.79 t $CaCO_3$/t CaO) and the calcination operation, the 2 more resources impactful stages in the manufacturing phase of HL. As a result, the production of RCLBP and HLP generate no significative differences for the resources impact category. Therefore, also considering the similar impact for the maintenance, the accumulated impact when the life cycle is completed (i.e., Final End of Life), shows only 2% reduction compared with HLP.

Ecosystem Quality category (Fig. 5.20b)

With regards to the Ecosystem Quality indicator, in the case of RCLBP, the binder's production represents 17% of the total impact at the manufacturing stage and for HLP 29%. It is relevant to highlight that even though the total emissions at initial production are reduced, the areas of damage are different for RCLBP compared to HLP. For the first one, emissions coming from the partial allocation of the impacts of acetylene to RCL production are the main responsible and emissions to air are more significant (Respiratory organics +32% increment), because of the higher emissions of coal combustion. Furthermore, for HL, emissions to the earth and water because of virgin calcite extraction (use of explosives) are of greater importance (Terrestrial Ecotoxicity +68%, Aquatic ecotoxicity +66%). Yet emissions arising from the combustion process play a significant role, because 51% of the used fuels in the kiln are solid fossil[26]. When the final end of life is reached (100 years), an overall reduction of 13% is obtained for RCLBP against HL, regarding the ecosystem quality.

Human Health category (Fig. 5.20c)

At the manufacturing stage and as observed in the Scenario 1 (RPMBP), the production of additives dominates the indicator, sharing 65% and 50% of the total for RCLBP and HLP. In the case of the binders, the main midpoint indicators contributing to Human Health category are Carcinogens and Ozone Layer Depletion. For HL production, these indicators were 55% and 44% higher than for RCL, explaining therefore the lower impact of RCLBP. Moreover, because of the lower specific emissions of RCL preparation (Section 5.2.3.4.1), 24% reduction is obtained in the manufacturing of RCLBP against HLP. The maintenance stage at 50 years increases the total indicator for both plasters around 52%. When the end of life is reached, the accumulated impact shows a very significative reduction of 22% for RCLBP versus HLP.

Climate Change category (Fig. 5.20d)

Among all the endpoint indicators analysed, Climate Change suffers the most significant changes. As was previously highlighted, the manufacturing of RCL has a very low carbon intensity, because the intermediate treatment is rather simple and requires no calcination operation (Figure 5.16). At the manufacturing stage of the plasters, a 64% reduction in the total CO_2 emissions is noticeable for RCLBP compared to HLP (1.15 and 3.16 kg CO_2/m^2 respectively). During the first year of application, the carbon sequestration responsible for the hardening of the plaster takes place. In the case of the RCLBP, 1.27 kg CO_2/m^2 are sequestrated, meaning that until the maintenance action, the RCLBP plaster is carbon negative (-0.13 kg CO_2/FU). Meanwhile, the carbonation of the HLP reduces by 45% the climate change indicator, as already pointed out during the comparison against PMS.

By considering the maintenance action and the landfilling during the use phase, HLP suffers the highest increment compared to RCLBP (65% relative to the initial production stage). The re-carbonation of the plaster after the maintenance reduces again the accumulated $CO_{2\ eq}$ by 28% for HLP and in the case of RCLBP, it leads again to carbon negative values (-0.11 kgCO_2/FU). At the End of Life, even though the plaster manufactured with carbide lime is not carbon negative anymore (0.04 kg CO_{2eq}/FU) due to the impacts of landfilling, a 100% reduction compared to HLP in the accumulated impact over the life cycle is achieved.

5.2.3.4.3 Sensitivity and Uncertainty Analysis

This section specifically focuses on conducting a sensitivity analysis for critical parameters within the life-cycle inventory of PMS and CL when utilized as a HL replacement in lime-based plasters. For a sensitivity analysis concerning traditional lime-based plasters (used here as a reference), readers are directed to a prior publication by the authors [26], where extensive studies have already been conducted.

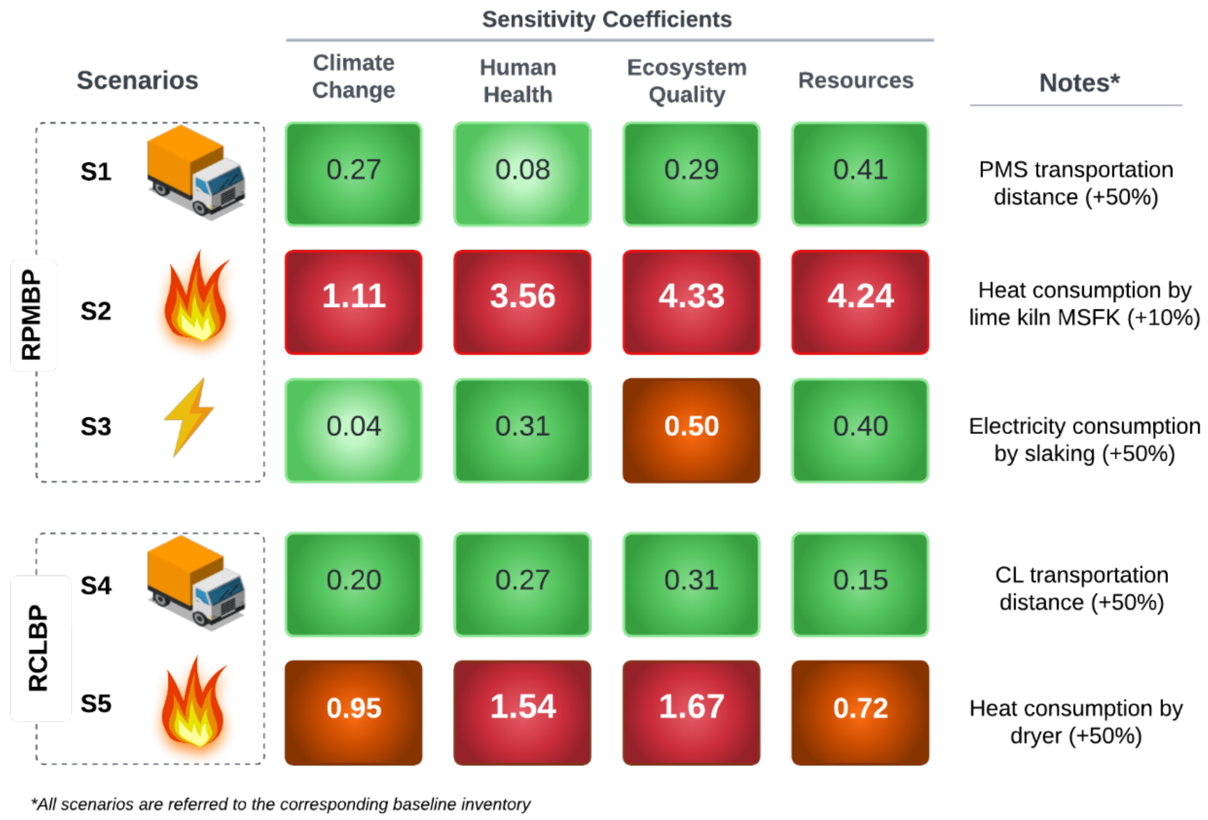
In the inventory modelling process, meticulous calculations have been performed on the data using a comprehensive process-based methodology developed by the authors [8]. This methodology facilitates the integration of mass and energy balances with current technologies employed during the manufacturing stage. To enhance reliability, both the data and assumptions have undergone verification within the SUBLime network. Additionally, expert judgment from manufacturers has been employed to ensure that equipment types, energy consumptions, and other factors closely align with real-world conditions.

Nevertheless, it has been observed that the intermediate treatment of secondary resources significantly impacts environmental indicators in both the Cradle-to-Gate and Cradle-to-Grave phases (as discussed in Sections 3.2.1 and 3.2.2). Therefore, there is a particular interest in assessing how changes in inventory parameters of the secondary resources can influence the output results from Cradle-to-Grave. Figure 9 summarises the results of the Sensitivity Coefficients (Eq. 3) for different scenarios, in comparison to baseline results (Figures 5 and 6). An arbitrary classification through colour code was adopted, and non-sensitive parameters were marked in green ($SC<0.5$), moderated sensitive in orange ($0.5 \leq SC \leq 1$) and highly sensitive in red ($SC > 1$). This study limits the uncertainty analysis to parameters with $SC > 1$.

Concerning the scenarios, for PMS, an increased transportation distance of the resource to the plant by 50% has been evaluated (S1). S2 considers calcination taking place in a Mixed Feed Shaft Kiln (MFSK), the second most used kiln in Europe after Parallel Flow Regenerative Kiln (PFRK), as utilized in Table 3. An increment in the consumption of electricity during the slaking procedure is evaluated in S3. Moreover, for RCL, two inventory parameters were of interest: an increment by 50% in the transport distance of CL to the processing plant (S4) and an increment by 50% in the energy consumption during the drying operation (S5).

The analysis of the sensitivity coefficients in Figure 9 reveals that for RPM, the endpoint indicators are highly sensitive to a change in kiln technology, even for small increments (+10%) in heat consumption. This result is not surprising, considering that the pyrolysis operation dominates midpoint and endpoint indicators (see Figure 6 and Appendix 6). In the case of RCL, the heat consumption of the dryer is much more sensitive in all endpoint indicators than the transport distance, especially for the Human Health and Ecosystem Quality indicator.

The sensitivity analysis facilitated the identification of parameters for inclusion in the uncertainty analysis, specifically focusing on the heat consumption in the lime kiln for RPM (S2) and the heat consumption during the drying operation for RCL (Ec.) (S5). For stochastic modelling, log-normal distributions were adopted due to their exclusion of negative values and better representation of mass and energy consumptions [31], [75]. The parameters for modelling the distribution are based on the "Best Available Technologies for Cement and Lime in Europe" document [76], combined with previous research by the authors [8] and supplemented by expert manufacturer judgment within SUBLime for devices operating under similar conditions.



		Sensitivity Coefficients				
	Scenarios	Climate Change	Human Health	Ecosystem Quality	Resources	Notes*
RPMBP	S1	0.27	0.08	0.29	0.41	PMS transportation distance (+50%)
	S2	1.11	3.56	4.33	4.24	Heat consumption by lime kiln MSFK (+10%)
	S3	0.04	0.31	0.50	0.40	Electricity consumption by slaking (+50%)
RCLBP	S4	0.20	0.27	0.31	0.15	CL transportation distance (+50%)
	S5	0.95	1.54	1.67	0.72	Heat consumption by dryer (+50%)

**All scenarios are referred to the corresponding baseline inventory*

Figure 5.21. Sensitivity coefficients of endpoint indicators for RPMP and RCLBP from Cradle-to-Grave

Table 5.9 introduces the mean values and standard deviation (95% confidence interval) for the midpoint impact indicators from Cradle-to-Grave (see Appendix 7) after conducting 10000 iterations. Acceptable standard deviations are obtained for the midpoint indicators, indicating robustness in the results presented in Section 3.2.2. However, it is important to note a limitation of the study: the uncertainty analysis was performed for the most sensitive parameters (i.e., SR>1). Additional research and practical experience in processing these resources for the production of lime-based plasters are necessary to identify potential critical parameters in real-world operations. This extends beyond the scope of the current study.

Table 5.9. Uncertainty of Cradle-to-Grave midpoint impact indicators for RPMBP and RCLBP

Parameter	Mean	Unit	Distribution	GSD^{2*}
Heat consumption in the lime kiln during RPM production	2640	MJ	Log-normal	7
Heat consumption in the dryer during RCL production	220	MJ	Log-normal	4

Midpoint indicator	Unit per FU	RPMBP		RCLBP	
		Mean	Standard deviation	Mean	Standard deviation
Respiratory organics	kg C2H4 eq	1.79E-3	4.82E-4	9.76E-4	8.62E-6
Ozone layer depletion	kg CFC-11 eq	3.40E-7	3.69E-8	2.79E-7	6.50E-8
Respiratory inorganics	kg PM 2.5 eq	3.17E-3	6.57E-4	2.37E-3	2.52E-4
Non-carcinogens	kg $C_2H_4Cl_{eq}$	9.36E-2	4.27E-3	8.16E-2	5.05E-3
Ionizing radiation	Bq C-14 $_{eq}$	20.97	0.67	53.21	5.21
Terrestrial acid/nutri	kg $SO_{2\ eq}$	4.50E-2	8.01E-3	3.89E-2	6.60E-3
Terrestrial ecotoxicity	kg TEG soil	220.21	14.64	190.98	11.24
Land occupation	m2org.arable	0.22	8.02E-3	0.31	1.70E-2
Aquatic ecotoxicity	kg TEG water	428.90	60.46	354.18	22.49
Global warming	kg $CO_{2\ eq}$	3.64	0.51	0.06	0.04
Mineral Extraction	MJ $_{surplus}$	3.71 E-2	3.18E-3	4.16E-2	7.74E-4
Non-renewable energy	MJ $_{primary}$	48.30	5.14	49.42	1.06

5.2.3.4.4 Environmental Sustainability of Lime-based Plasters Manufactured with Upcycled Materials

For plaster produced using PMS, while a reduction in life cycle impact is evident for RPMBP compared to HLP across all four endpoint categories, the overall effect is not highly significant. This can be attributed, to the intermediate treatment of PMS, which involves operations like traditional HL manufacturing, as discussed in the Cradle-to-Gate analysis. The kiln's usage in this treatment significantly influences the overall impact due to its energy intensity and specific emissions during calcite decomposition. This emphasizes that the upcycling of waste materials may not always be the most efficient route to enhance material environmental sustainability.

In contrast, manufacturing lime-based plaster with carbide lime demonstrates superior environmental performance across all indicators compared to paper mill sludge. This stems from the properties of the supplementary lime materials. Although both materials are rich in CaO, their conditioning for hydrated lime replacement involves vastly different intermediate treatments. Prior research by the authors [8] underscores that achieving similar functional performance to hydrated lime through calcination operations often yields limited reduction in impacts and CO_2 emissions due to high energy demand (fuel consumption) and inevitable emissions from calcite decomposition (0.79 kg CO_2/kg CaO). Despite the need for allocation coefficients corresponding to carbide lime's by-product status, the simplicity of its intermediate treatment significantly reduces the environmental impact of RCL manufacturing.

Sections 5.2.3.4.1 and 5.2.3.4.2 underscore the pivotal role of intermediate treatment for RPM in influencing both midpoint and endpoint environmental indicators. To enhance Cradle-to-Cradle sustainability, focused efforts are required, particularly in addressing environmental concerns associated with the lime kiln. Mitigating stationary combustion emissions necessitates reducing coal in the fuel mix while increasing the use of low-energy carbon sources like biomass, supported by natural gas, as elucidated in a previous study [25]. In making well-informed decisions, it is crucial to conduct a comprehensive analysis considering various impact indicators to strike a balance between damage and benefits. Furthermore, addressing process emissions from calcite decomposition can be achieved cost-effectively by adopting kiln technology with direct CO_2 separation, exemplified by the innovative approach developed by LEILAC [8], [76], [77]. This kiln not only offers efficient emissions control but also aligns with Europe's net-zero industry future plan, as it can operate entirely in an electrified mode [78].

In the case of RCL, despite the need for allocation coefficients related to carbide lime's by-product status, the simplicity of its intermediate treatment significantly diminishes environmental impact compared to HL. Exploring the feasibility of achieving net-zero and even carbon-negative scenarios becomes paramount when RCL is utilized in lime-based construction materials. With the initial manufacturing emissions at remarkably low levels, the possibility of CO_2 sequestration during the utilization phase creates opportunities for optimistic scenarios. Electrification of transport and advancements in energy efficiency in the drying operation, as revealed in the sensitivity analysis (refer to Fig. 5.21), emerge as critical optimization hotspots that warrant further exploration and study.

When comparing supplementary material alternatives, environmental criteria are not the sole consideration. As analysed in Table 5.4, the feasibility of waste/by-product reuse depends on resource availability at specific production plant’s locations and logistics required for transportation. In particular, integrating secondary resources into the construction industry necessitates a multifaceted approach, encompassing scientific research, developmental initiatives, and a coordinated action plan across the entire value chain. This involves fostering collaboration among building material producers, vertical and horizontal markets, while aligning with the 2050 Circular Economy Plan of the European Commission [78]. Breaking barriers between regional markets is crucial for establishing effective upcycling pathways and minimizing landfilling in waste management. Offering taxation incentives to collaborative markets can further promote cooperation. When selecting by-products or wastes for upcycling lime-based material manufacturing, a critical assessment of regional availability is imperative. For instance, PMS appears more viable in the European region, while in China Carbide Lime (CL) is preferable based on production sources. An important consideration is that the equipment needed for manufacturing PMS is commonly available in the construction industry. In contrast, producing CL would necessitate additional investment in equipment or the outsourcing of by-product treatment, an aspect that requires further analysis. Comprehensive environmental impact studies are also essential for CL, especially given its by-product status and dual use in polymer manufacturing. The most effective pathway for reducing overall environmental impact, whether using CL as a mineral binder or as a polymer precursor, is yet to be determined. Finally, economic considerations are pivotal, leading us to delve into the subsequent section, which addresses the economic assessment of plasters manufactured using PMS and RCL in comparison to the traditional HLP scenario.

5.2.3.5 Economic Sustainability of Lime-based Plasters Manufactured with Upcycled Materials

Figure 5.22 shows the results of the LCC (€/FU) from Cradle-to-Grave for the case of the plaster manufactured with traditional hydrated lime and upcycled RPM and RCL. In the Figure also the stages of the life-cycle in which the cost takes place are indicated and the total cost of the upcycled alternatives (RPMBP and RCLBP) compared to the reference HLP. All items of the direct production cost associated to the raw material purchase, transportation, plant operation (electricity consumption) as well as maintenance action, landfilling and the externalities (carbon pricing) have been disaggregated for a better analysis of the relative contribution. The cost of the CO_2 emissions is distributed between the initial manufacturing phase and the use phase (i.e., CO_2 related to the maintenance action).

With regards to materials manufacturing, the production cost of the RPM and RCL also includes the transportation of the materials to the plant. At first sight it can be observed that producing the traditional binder HL has a higher cost than the upcycling of RPM (+ 69%) and RCL (+ 65%). Therefore, the replacement of HL in the mix is likely to decrease the total production costs. Without including the CO_2 taxes, the contribution of the binders to the manufacturing cost of HLP, RPMBP and RCLBP ranks as follows: HL (63%), RPM (31%) and RCL (37%). From the materials perspective, the second and third important contributors are the production of the fine aggregate ranging from 15 to 29%, and LWA from 10 to 17%.

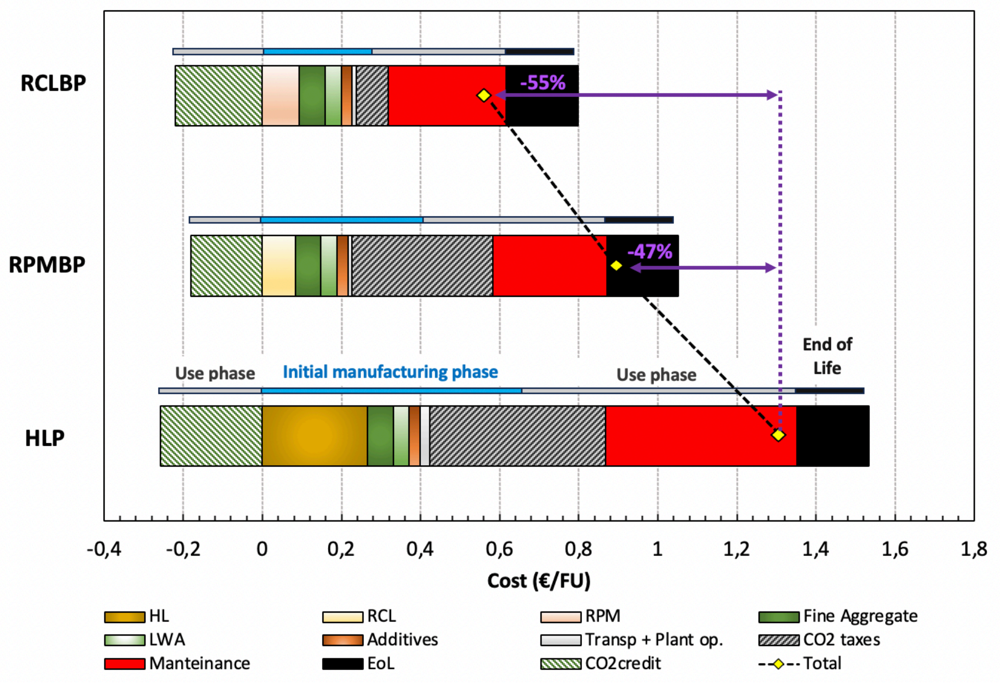


Figure 5.22. Life Cycle Cost Assessment results from Cradle-to-Grave of the plasters manufactured with traditional HL and upcycled RPM and RCL

Under the European Trading System, a fixed price per kg of CO_2 (in 2022, 90€/t CO_2) is levied on the industries. The emissions associated to the initial binder's production in HLP, RPMBP and RCLBP are 2.45, 1.98 and 0.45 kg CO_2 respectively. As indicated in previous research by the authors, in the case of HLP the share of CO_2 in the total cost of the plasters manufacturing is around 30% [26]. Due to the lower emissions and production costs of the upcycling alternatives, the share of CO_2 taxes is 44% for RPMBP and 15% for RCLBP.

In the use phase of the wall as functional unit, there are two components to consider. On the one hand the cost of the maintenance action which includes the production of the new materials and the costs of the related carbon emissions. If no credits are considered (current legislation scenario), the use phase (maintenance and CO_2 taxes) accounts for 44, 41 and 38% of the total costs for HLP, RPMBP and RCLBP respectively. The current tax framework does not distinguish between industries that are assured of sequestration of CO_2 emitted during the production stage, at other stages of the product life-cycle. As shown in Section 5.2.3.4.2, the re-carbonation capacity of lime-based materials is well documented, proven, and necessary for the material to perform its function. To ensure a fair market competition, the design of equilibrated carbon prices requires taxing policies at a political and economic European level [24], integrating the role of products and their properties, along with tracking of a potential CO_2 uptake profile of the materials over their life-cycle (Figure 5.19d and 5.20d).

The total sequestration potential per FU of the analysed scenarios over the life-cycle was shown in Table 5.7 and implemented in Figure 5.19d and 5.20d. Figure 5.21 includes the credits of the carbon sequestrated during the use phase, considering the same carbon price as for the emissions (in 2022, 90€/kg). The contribution of the carbon credit to the total cost varies depending on portlandite content of the binder. Therefore, for HLP the highest carbon dioxide credits are observed (0.26 €/FU), followed by RCLBP (0.22 €/FU) and RPBMP (0.18 €/FU). Moreover, it is interesting to observe that even though in several studies the landfilling of the materials is disregarded [80]–[82], it can contribute from 12 to 32% of the total costs over the life-cycle (carbon credit included).

To conclude, the cost assessment shows a 47% and 54% reduction for RPMBP and RCLBP, compared to the traditional HLP. The case of the plaster with the incorporation of RCL has shown both, in environmental and economic assessment the best performance, because the intermediate treatment to prepare the binder is cost-effective and low carbon. Nevertheless, it is important to consider that this application is not feasible in Europe, due to the limited availability in the region (Table 5.4). This fact points out the relevance of making a thorough analysis and selection of the potential materials to be used as supplementary lime materials, along with the intermediate treatment required for the preparation of the secondary resource.

5.2.4 Conclusions

This paper compares the environmental and economic performance of lime-based plasters using traditional hydrated lime, paper mill sludge (PMS), and carbide lime (CL). Methodological novelty encompasses:

i) Developing a Cradle-to-Gate industrial-scale inventory through process design upscaling of recent lab-scale innovations,

ii) Investigating the PMS and CL treatment processes,

iii) Examining a Cradle-to-Grave scenario including maintenance and CO_2 sequestration analysis.

The assessments findings are summarized as follows:

Environmental Impact Assessment

- The Cradle-to-Gate analysis shows that using PMS in plaster production leads to 18% resource savings and 20% reduced CO_2 emissions compared to hydrated lime. Transportation and pyrolysis treatment (decomposition of $CaCO_3$ and fuel emissions) are identified as main contributors. CL has even lower environmental impact than PMS due to the absence of pyrolysis. This results in an 80% reduction in CO_2 emissions and a 13% reduction in resources usage compared to HL. In case of economic allocation, the impact from acetylene production contributes 63% and 71% to the total CO_2 emissions and resource usage indicators, respectively.
- Despite the lower availability in Europe, the Cradle-to-Grave analysis reveals that upcycling CL achieves superior environmental performance, with 100% reduction in global warming potential compared to traditional plaster with HL. The reduction for upcycled PMS is 11%.
- Sensitivity analysis reveals heat consumption in the lime kiln (PMS treatment) and drying operation (CL treatment) as critical variables significantly affecting the environmental impact from Cradle-to-Grave.

Economic Impact Assessment:

- Manufacturing traditional hydrated lime has higher costs compared to upcycling PMS (+69%) and CL (+65%).
- The emissions associated with the binder's production in plasters, considering CO2 taxes, account for a significant portion of total costs. The use phase, including maintenance and considering current CO_2 taxes, accounts for 44%, 41% and 38% of the total costs for HL, PMS and CL plasters, respectively.
- To ensure fair market competition, carbon pricing policies at a European level should integrate product properties and consider the potential CO_2 uptake profile over the life-cycle.

- When considering a carbon credit for CO_2 sequestration by lime during the plaster's life-cycle, the cost assessment shows a 47% and 54% cost reduction for upcycled PMS and CL plasters compared to traditional HL.

In summary, using upcycled materials like PMS and CL in lime-based plasters can lead to environmental benefits and cost reductions, with CL demonstrating superior performance. Policy interventions are necessary to ensure fair carbon pricing and incentivize the adoption of sustainable alternatives.

One prominent limitation of this study pertains to the reliance on calculated input data for modelling the life-cycle inventories of the intermediate treatments for PMS and CL. Despite the detailed and expert-screened modelling approach, the incorporation of experimental data is essential to refine and enhance the accuracy of the inventories. Another constraint arises from the assumption that the upcycled plasters exhibit equivalent functionality and long-term performance compared to the reference material, a conjecture that warrants experimental validation.

Addressing these limitations requires further research efforts, particularly emphasizing the optimization of intermediate treatments. Special attention should be directed towards the lime kiln for PMS and the drying operation for CL, as highlighted in the sensitivity analysis. This emphasis on optimization will contribute to a more robust and reliable understanding of the sustainability of the studied processes.

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Chapter 6: The Role of Energy Sources in Hydrated Lime Production

The environmental impact of producing hydrated lime, a crucial precursor for lime-based construction materials, is markedly affected by the energy sources employed in the manufacturing plant. The selection of an energy source is intrinsically linked to the geographic location of the facility.

In this chapter, a comprehensive scenario analysis and quantitative impact assessment are undertaken, utilizing projected trends in sustainable energy production for both energy and fuel up to the year 2050 in Germany, Belgium, Portugal, and Spain. The objective was to rigorously quantify the repercussions of this transition on the Cradle-to-Gate environmental impact. Subsequently, the potential decarbonization scenario for energy is juxtaposed with the existing scenario as of 2020.

Publication 5. Hydrated Lime Life-cycle Assessment: Current and Future Scenarios in Four EU Countries

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6.1 Introduction

Lime is one of the materials with the richest history and its use is as old as some of the most imponent civilizations that we have known. As an enabling material, lime renders and mortars had been found to be extensively applied in constructions all over the world of Israel (7000 BCE)[1], Syria (4250 BCE)[2], China (2000 BCE), Mayan, Inca and Aztec civilizations (500 BCE) [3]. Since then, it has played different roles, from structural functions to decorative techniques [4] and the knowledge for its preparation was transmitted from generation to generation. With the developing modern society and the increasing need of this material, the production of lime was completely industrialized. Nowadays the term “lime” (Calcium Oxide, CaO) is assigned to a product derived from limestone in an industrial process known as calcination [5] and not only its production but also uses have grown with time, becoming crucial in several applications.

According to a report published by EuLA in 2018, the sales sector in 2016 was distributed among steel (39.2%), construction industry (18.1%), environmental protection (17.2%), other industrial consumers (13.8%), chemical industry (7.5%), export outside the EU (3.3%) and agriculture (1.9%) [6]. What is more, the Report for ‘Competitiveness of the European Cement and Lime Sectors’, issued in February 2018, states that in 2015 (latest available Eurostat), the lime and plaster industries represented more than 20 Mt/y [7]. It can be observed that the construction industry plays an important role in the scenario of lime consumption. Today, lime-based building products experience many challenges (e.g., raw material prices and environmental restrictions) that hinder the continuous development of innovative approaches in material design, process, product functionality and sustainability. This research is carried out within the framework of the SUBLime network [8], an EU ETN – ITN project that was born to provide answers to the former challenges, bringing together lime experts from the academia and the industry.

In terms of environmental performance in lime manufacturing, the production of 1 tonne of CaO generates around 1.2 tonnes of CO_2, contributing in about 1% to the global anthropogenic CO_2 ([9]–[11]). The environmental load associated to lime products can be accounted to two main causes. Firstly, almost two third of the emissions are linked to the chemical reactions during the calcination of calcium carbonate ($CaCO_3$) to produce CaO (0.78 kg CO_2/kg CaO) (Reaction 6.1) [12]. The Hydrated Lime (HL) used in the construction industry is produced during a process called slaking (Reaction 6.2).

$$CaCO_3(s) + \Delta H \rightarrow CaO\ (s) + CO_2(g) \qquad \Delta H° = 177.8\ kJ/mol \qquad \textbf{Reaction 6.1}$$

$$CaO\ (s) + H_2O\ (l) \rightarrow Ca(OH)_2\ (s) + \Delta H \qquad \Delta H° = -65.2\ kJ/mol \qquad \textbf{Reaction 6.2}$$

The second factor is the industrial production of quicklime itself, including the combustion of fuels in the kiln, the electricity needed to operate the plant, the transport of different materials, etc. Figure 6.1 shows the flow diagram of a plant that can produce four types of lime products: Pebble Lime (PL), Milled Limestone (ML), Milk of Lime (MoL) and HL. The process starts from the extraction of limestone from a quarry, a series of particle size reduction operations, washing of the limestone to reduce the amount of fines, screening operations to separate different fractions of limestone and the calcination process itself where the material is heated in a kiln with temperatures above 1000°C [12]. As a result, quicklime in different particle sizes is obtained.

The production of hydrated products (MoL and HL) can happen in integrated plants (as shown in the Figure 6.1) or the ML can also be transported to a hydration plant at a different site. Some by-products might be generated during the process (such as MLS).

In terms of energy consumption, the calcination process in the kiln is the most energy intensive step in the lime production process; depending on the kiln it can vary from 3 to 9 MJ/kg CaO [11]. This operation accounts for around 90% of the total energy consumption and 99% of the specific impact of the whole process on global warming ([12]–[14]). Thus, many research articles mostly attribute the environmental impact of this process to the kiln operation. While it is true that calcination is a major contributor to global warming, there are other environmental problems not directly related to carbon dioxide equivalent emissions [15]. These include damage to ecosystem quality (Water Acidification or Terrestrial Eco toxicity) as well as damage to human health (ozone depletion or the generation of ionising radiation).

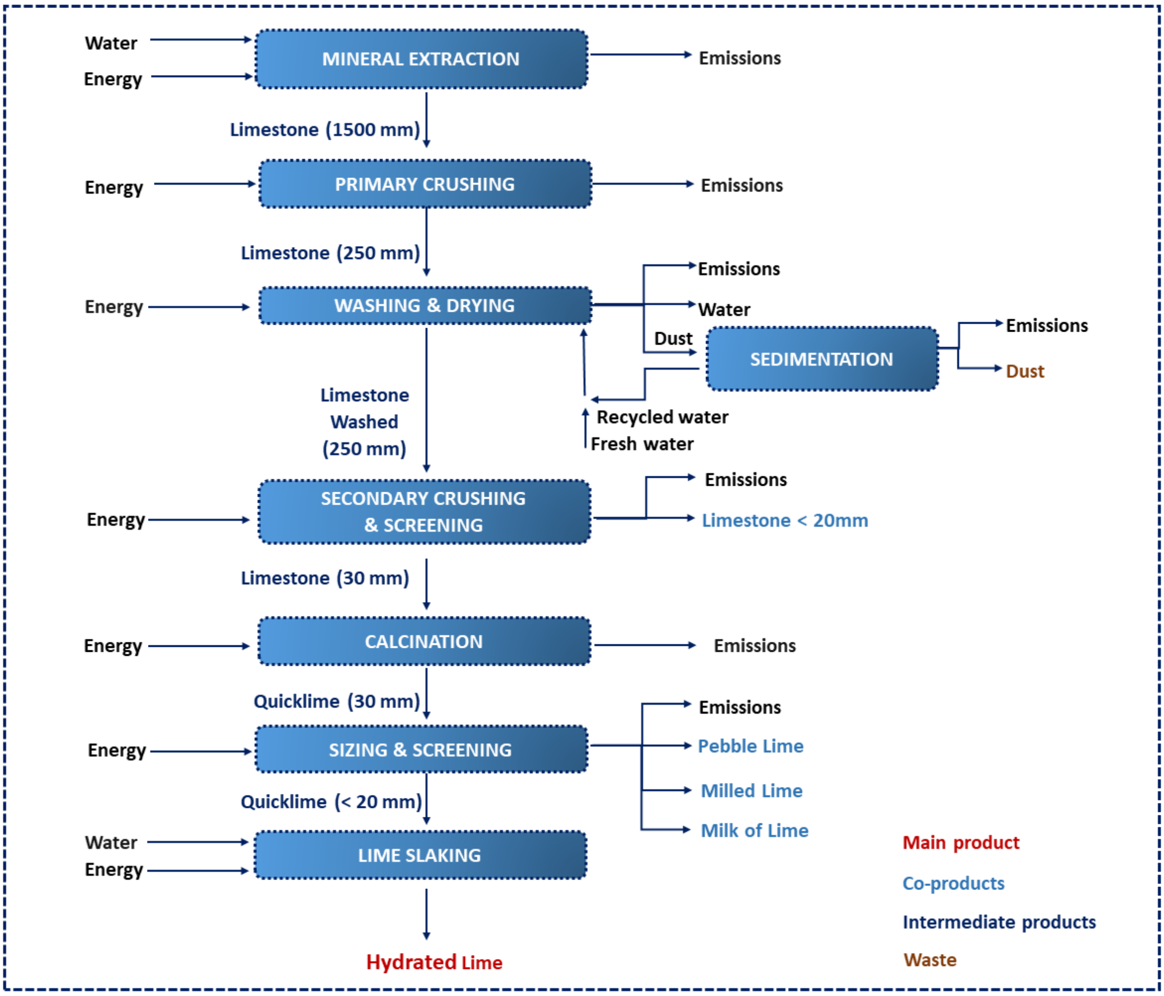


Figure 6.1. Industrial process for the production of Hydrated Lime – System boundaries considered in this study

These impacts can potentially be affected by the types of energy sources used as fuel for the furnace, as well as the electricity production matrix used to supply the plant, the impact of which may not be negligible. Likewise, being HL a product of high consumption in the world market, identifying in what proportion the energy sources have an impact would allow the design of strategies to comply with the SDG outlined by the UN [16]. In particular, 3 SDG are directly related to this research: Goal 7 “Affordable and Clean Energy”, Goal 9 “Industry, Innovation and Infrastructure” and Goal 13 “Climate Action”. The common line among them is the promotion of sustainable industries through energy efficiency and productivity, upgrading the technologies to provide clean energy from renewable sources (i.e., low-carbon development), contributing in turns to limit the increase in the Global Warming. However, there are very few records in the literature addressing this research line and, in particular for the construction industry and in lime-based mortars and plasters some studies have addressed a sensitivity analysis only on the content of HL in the dry mix and transport distances of cement, additives and sand [17], [18].

Furthermore, within the framework of an environmental crisis, a growing world population, the need to reduce greenhouse emissions and the limited sources of fossil fuels, it is imperative to shift to more sustainable management of natural resources and sources of energy supply [19]–[22]. Therefore, all over the world several actions and policies are being taken, to speed up actions towards a "green future" and ensure a significant penetration of RES in power generation [23]. In 2016 a Reference Scenario for the EU in terms of energy, transport and GHGs emissions has been published, expressing that by 2020 the use of RES would increase to 37.2% of net electricity generation, composed for about 52% by variable amounts of solar and wind RES [24]. By 2050, the same report also states that wind (offshore and onshore) will provide the largest contribution (25%), followed by nuclear (18%), solar and hydro (11% each), and biomass (6%). Meanwhile, gaseous fuels and oil will account for around 26%. This circumstance challenges the capability of the European countries to adapt their current technologies, considering the resources and environmental conditions in each country (availability of rivers, wind potential, etc.) which tighten the boundaries of what can actually be achieved. This heterogeneous scheme indicates that different sources of RES in different percentages, will be used to supply energy in the future for each particular country of the EU. It is therefore of interest to anticipate the potential impact that this may have on the environment.

Because of the increase in use of more sustainable sources of energy, lower environmental impacts for the production of lime-based products are expected, as well as different environmental loads according to the geographic localization of the plants. Therefore, it is necessary to quantify the benefits of the changes in current and future scenarios for the production of HL in Europe, through LCA and sensitivity analysis.

This paper deals with a case study for DE, BE, PT and ES and pursues the following objectives:

(a) To present a Cradle-to-Gate LCI for the production of hydrated lime in a theoretical plant.

(b) To quantify the environmental impact for regionalised production of HL, identifying the influence of energy sources used on the indicators considered.

(c) To carry out a sensitivity analysis on the environmental impact indicators, considering the particular projections of each country in decarbonisation of the electricity matrix and the use of alternative fuels with potential CO_2 emission reductions.

6.2 Methodology and Scenarios Description

The LCA methodology was used to quantify and compare the potential environmental impact of the current and future scenarios for HL production in each of the analysed countries. This methodology is defined as "the compilation and evaluations of the inputs, outputs and potential environmental impact of a product system throughout its life-cycle" [25]. The research carried out is based on the ISO

14040/44 (2006), and accordingly four main steps are to be performed: goal and scope, inventory analysis, life-cycle impact analysis and interpretation of the results [25], [26].

6.2.1Goal and Scope Definition of the Case-Study

The aim of this study is to quantify the impact of the production of HL on the environment, considering a theoretical plant installed in four different geographic locations: DE, BE, PT and ES, operating with a specific energy and fuel mix matrix according to the future projection of each country by 2050. The impact will be evaluated in comparison with the current scenario (2020). The daily capacity of the plant as well as the amount of production is shown in Table 6.1. The production 1 kg of HL from cradle (i.e., from the extraction of raw materials) to the gate of the factory is used as FU of this study.

Table 6.1. Capacity of the theoretical plant

	Pebble Lime	**Milled Lime**	**Milk of Lime**	**Hydrated Lime**	**Total**
Production (t/d)	75	75	75	75	300

To perform a proper study regarding the environmental impact of products, it is critical to make an unambiguous definition of its scope. The studied plant is shown in Figure 6.1 and the process are based on the flowchart of one lime industry. It represents the production of different lime-based materials from the cradle (extraction of raw materials) to the gate of the factory. The operating process aims at producing the 4 products listed in Table 6.1. Even so, as a result of the unit process "Secondary Crushing & Screening" different fractions of ML are produced. Due to the operating conditions of the kiln, those fractions below 20 mm are not desirable. Instead of being disposed, they are sold as MLS to another industry.

6.2.1.1 Description of the Scenarios

In order to analyse the effect of different energy sources and the effect of the geographic localization of the theoretical plant, the data of the current and future scenarios were obtained from different sources, among which are included the Webpage of the IEA, Energy department's/ministries and recognised Institutions in the field of Energy. The consideration of the effect of the energy source and its environmental implications is addressed to the best knowledge of the authors. Figure 6.2 summarises the current and potential future scenarios for the electricity mix in DE, BE, PT and ES.

6.2.1.1.1 Germany

A report by the International Energy Agency [27] states that until 2018 DE's energy system was still largely based on fossil fuels. Coal, oil and natural gas had the higher proportion in the total primary energy supply and total final consumption used for power generation. Nevertheless, renewable sources or energy from solar, wind power, biomass and other sources have increased their share in the German electricity mix. According to a Report by the BWE, the share of gross electricity consumption covered by renewables in 2019 (42.1%) rose by approx. 8% to nearly 243 billion kilowatt-hours compared to 2018 (37.8%) [28]. The increase was mostly due to favourable weather conditions and the further expansion of offshore wind-powered installations and of PV installations. In 2019, solar energy accounted for 19.1% of DE's electricity generation from renewables. Biomass contributed by 20.6%. Hydropower accounted for 8.3%, geothermal energy for 0.1%. More than half of the overall 242.5 billion kilowatt-hours generated came from wind power, with onshore wind power accounting for 41.7% and offshore wind power for 10.2%. In 2020 the amount of renewable energies has increased to around 45.5%, composed by the sources shown in Figure 6.2a. According to a very extensive study commissioned by the Federal Environmental Agency of DE and conducted by the Fraunhofer IWES, by 2050 DE has the technical and ecological feasibilities to base its electricity supply system completely on renewable energies [29].

Figure 6.2b shows the technologies that can be potentially used to fulfil the energy demand of the country. It is based on the projection that the electricity consumption will be around 10% lower than 2005 levels and for each technology, the area potentially available for its deployment was determined and reduced considering ecological considerations, competing land uses and settlement area [29].

6.2.1.1.1 Belgium

According to the “BE’s Integrated National Energy and Climate Plan 2021-2030” [30], the country has tried to decrease the proportion of conventional sources in the electricity production matrix since 1990. In terms of the market share of total end consumption, petroleum products remain the principal source of energy (52%), followed by natural gas (24%) and electricity (17%). Natural gas is the dominant fuel in the industrial and residential sectors (35 % and 38 % respectively in 2015). In the transport sector, consumption is dominated by petroleum products (95%) [30]. As mentioned before, each country is subjected to its own reality for energy production. In the case of BE, due to their limited energy resources it is highly dependent on other countries for its energy supply. In 2015, its total primary energy production accounts for about 20 % of its total primary energy consumption and nuclear energy accounted for 73.9% of BE’s energy production. The same year, the proportion for renewable fuels and waste was 19.5 %. In 2016, renewable energy accounted for 8.65 % of total final energy consumption [30]. In recent years, BE has made progress in developing renewable energy. In Figures 6.2c and 6.2d the transition from the current to a potential future scenario can be observed [31]. BE has proposed a series of scenarios for climate neutrality by 2050. The main climate neutral scenario is called the “CORE 95” scenario and leads to a reduction in GHGs emissions of about 95% in 2050 w.r.t. 1990 and to so-called negative emissions of about 5% of 1990 GHGs emissions, thereby leading to climate neutrality by 2050. Several changes in behaviour, lifestyle and marked societal changes in the way people move, house, and feed themselves as well as a drastic decrease in energy demand are responsible for this change. Furthermore, the model implies several changes into the energy sources used, not only the reduction of conventional sources but the increment in the proportion of novel green sources such as Biofuels [32].

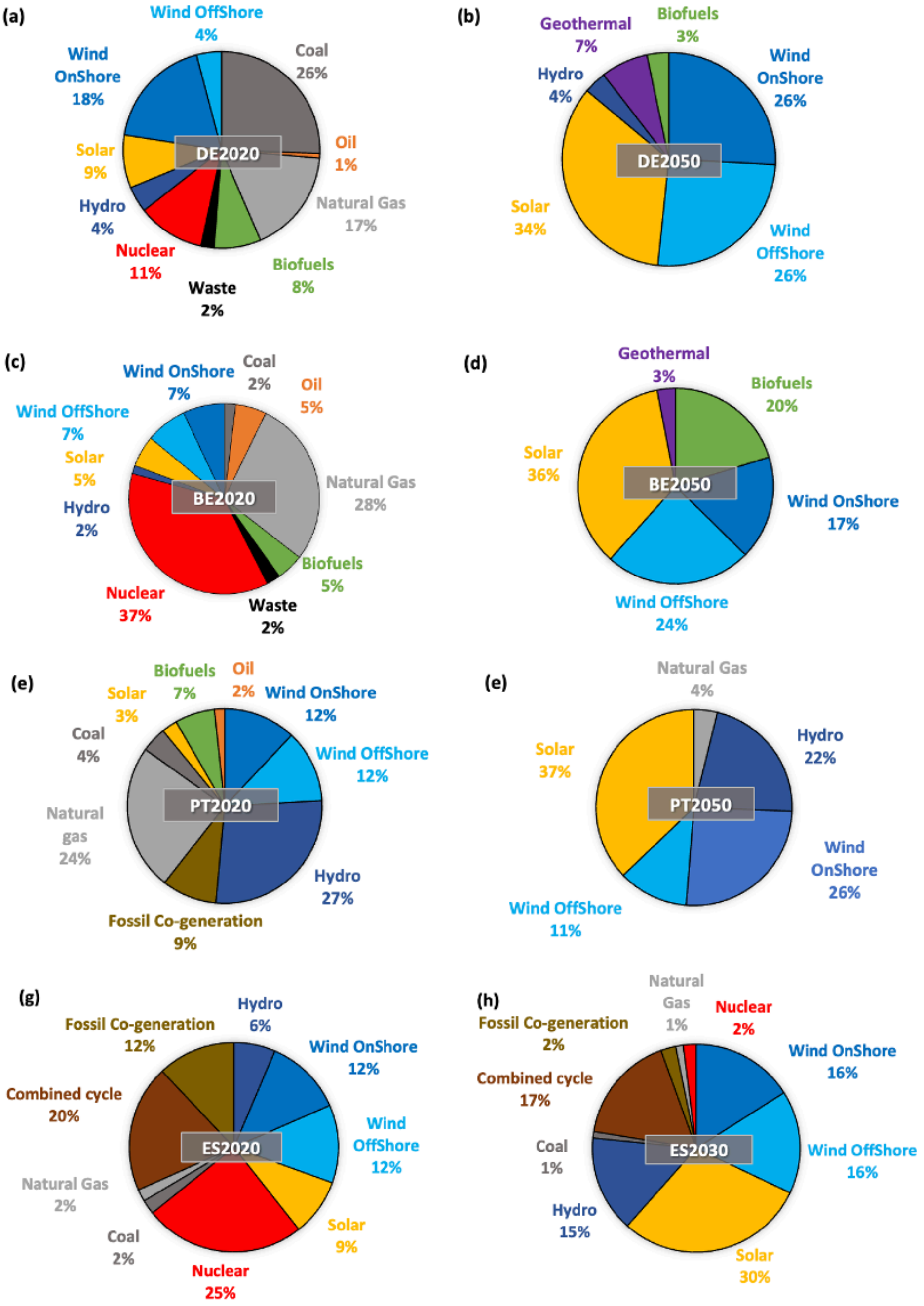


Figure 6.2. Electricity mix for the baseline scenario (2020) and potential future scenarios (2030/2050) in DE (a, b), BE (c, d), PE (e, f) and ES (g, h)

6.2.1.1.2 Portugal

PT is one of the EU countries that experienced a major financial crisis in 2008, and since then has been recovering. Furthermore, they were able to accelerate the structural changes required to shift from energy intensive activities and decoupling economic growth from energy demand. Nevertheless, until 2019 they remained reliant on imported fossil fuels, accounting for around 76% of primary energy supply (43% oil, 24% natural gas and 6% coal) [33]. PT has the resources to produce electricity from their rivers and wind, being almost 54% of the electricity generation covered by these sources with a high use of bioenergy in industries and buildings.

In 2020 around 60% of the electricity supply in PT was based on RES (Fig. 6.2e). Compared to 2019, in 2020 the use of non-renewable sources decreased by 11.9% [33]–[35]. The country was also one of the first in the world to set 2050 carbon neutrality goals. The developed policies consider the key role of hydrogen for achieving carbon neutrality, but same as BE, a change in lifestyle and market is needed along with monetary incentives for green alternatives. The APREN, has recently (2018) published a report analysing the opportunities of the electricity sector, to achieve the required decarbonisation level. They have concluded that the contribution of renewable electricity should be around 94% in 2050. Saying that, a change in the energy mix supply must be carried out, reducing the proportion of conventional fuels such as coal and natural gas, and increasing greener sources such as wind offshore and onshore and solar energy. The last one is estimated to represent 30% of Portuguese electricity mix, while wind will reach 39% [36]. Figures 6.2e and 6.2f summarise the projected scenario by APREN, considering a reduction of GHGs emissions from the energy sector up to 75% in 2050 [34].

6.2.1.1.3 Spain

As in many countries of Europe, the regulation of the electricity sector in Spain is undergoing a profound reform. According to the Spanish Ministry of Energy the main objective of this reform is to ensure the economic and financial sustainability of the electricity system, while guaranteeing electricity supply with the necessary levels of quality and at the lowest possible cost, an effective level of competition in the sector and all of this framed within the principles of environmental protection of a modern society [37]. The demand for electricity in ES has consolidated its positive trend from 2015, and the demand in 2018 reached 268,808 GWh (0.4% up on the previous year). Moreover, the generation registered a fall of 0.5% with respect to 2017, affecting mainly coal-fired and combined-cycle generating stations, whose production decreased by 17.2% and 18.9% respectively. Wind power has increased by 1.5%. The rest of the electricity generation technologies showed minimal or insignificant variations [38]. This observed decrement in the use of energy demand was registered again in 2020 falling around 5.5% in comparison with 2019.

The 2020 report of the Spanish Electric System shows that the proportion of renewable energies and non-renewable sources to produce electricity were 45.5% and 54.5% respectively [39], [40]. Among the energy sources for electricity generation, non-renewable nuclear energy can be highlighted, combined cycle and cogeneration and mainly wind and hydro sources for green technologies (Figure 6.2g). In the case of ES, it was not possible to establish a 2050 scenario in terms of used technologies for electricity production. Instead, the data for 2030 is taken from the Integrated National Energy and Climate Plan 2021-2030 [40]. In the future, the contribution of hydroelectricity is not expected to grow significantly given to the available resources already being used, whereas solar and wind are expected to grow in interesting proportions to contribute to the RES (Figure 6.2h) [20], [40].

6.2.1.1.4 Sources of Thermal Energy for the Lime Kiln: Fuel Mix in Europe

As previously mentioned, a critical unit in the production of lime is the kiln of a lime factory. It is the most energy intensive step of the production line and causes the highest specific energy consumption depending on the used technology. This unit process also comprises the biggest share of emissions, coming from both raw material decomposition and the fuel combustion. The emissions associated to the limestone decomposition are actually well known and can be assumed to be around 0.78 kg CO_2/kg CaO (Reaction 6.1).

During the combustion of fuel, a wide range of gaseous products (i.e., emissions) are generated, along with thermal energy as a result of the exothermic reaction. The nature of these emissions and its potential impact on the environment, obviously depend on the fuel type used. According to the BAT document, except for mixed feed shaft kilns, all types of kilns can operate with all types of fuels (solid, liquid, and gaseous) [11]. This opens doors for reducing the emissions, by considering that around 30% of the emissions during the production of lime are accounted to the fuel combustion [12]. Furthermore, up to until 2008 the most common used fuels in the EU were solid fuels like lignite, coal, pet coke and coke; in addition to natural gas, liquid and waste fuels and biomass [11].

The last EuLA report of 2019 [12], indicates the composition of the average fuel mix used by the European Lime industries in 2010 (Figure 6.3). In terms of the fuel mix, FSF are principally made of hard coal, lignite and petrol coke, whereas gaseous fuels are made of NG and liquid fuels of light fuels [41]. It can be observed that there is very limited use of biomass and waste in lime production. Some explanations for this context can be found considering that, the heterogeneity of these type of fuels, operating conditions in the kiln (including the mixing of volatiles and oxygen), pre-treatment needed (usually drying), as well as the complexity of the combustion reactions and emissions, make it less easy to work with other than traditional fossil fuels [42]. At the same time, in particular in lime manufacture, the quality of the product can be severely affected if the waste and biomass does not comply with the very precisely defined physical properties [11].

When it comes to reducing the emissions, FSF need to be replaced as much as possible. To illustrate the problem, it is enough to consider the CO_2 emission factor (tCO_2/TJ) in DE of lignite (103.8), light fuel oil (74) and natural gas (55.9) [43]. In theory, around 50% of combustion emissions can be saved by using natural gas instead of lignite. On top of that, a recently published empirical assessment in 10 European countries has shown that natural gas is the main fossil fuel used to back up renewable energy sources [44]. Furthermore, an increase of the proportion of biomass can not only lead to reduce the CO_2 emissions, but also to achieve zero net CO_2 emission if they are grown in a sustainable way [45], [46] [47].

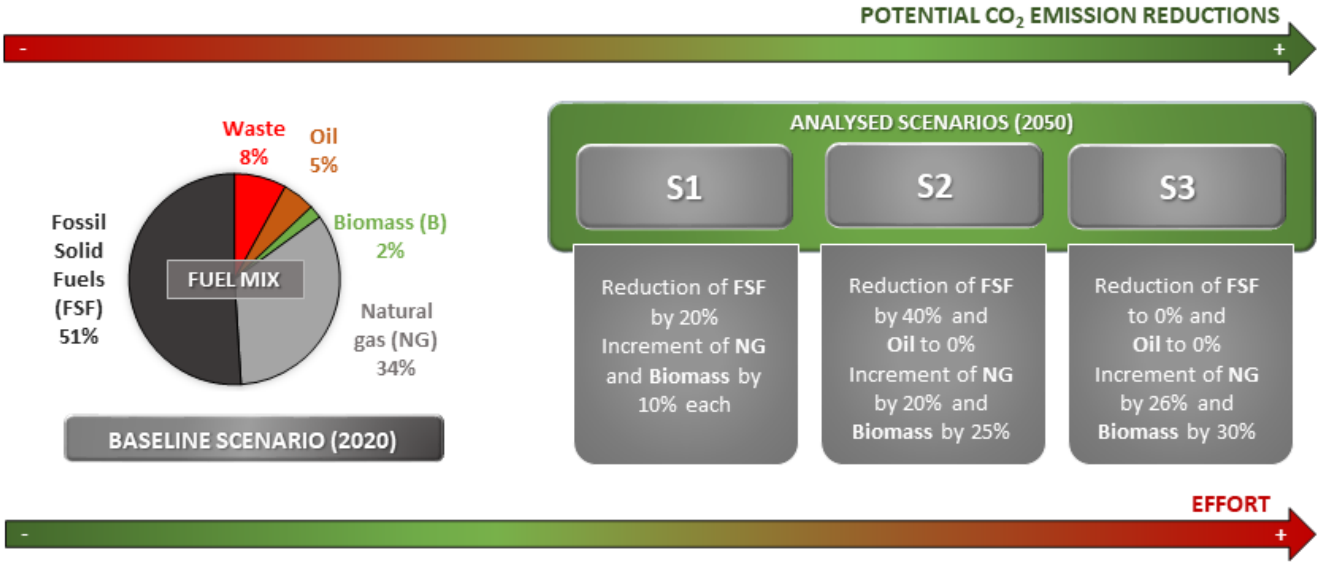


Figure 6.3. Average fuel mix (2010) based on EuLA report (baseline scenario)[12] and alternative fuel mix scenarios for potential CO_2 emission reduction.

Considering the aforementioned facts, the scenarios proposed in Figure 6.3 are based on the chance of reducing the amount of FSF, shifting to eco-friendlier fuels by incrementing the proportion of NG and biomass. In the baseline scenario (2020), a minimum effort to follow current regulations is needed, meanwhile no significant environmental improvements are taking place. Nevertheless, moving to a more sustainable model implies making economic, technological, and socio-cultural sacrifices. This means that less efficient kilns need to be replaced by PFRK and it might be also possible that extra operation units are required to deal with the pre-treatment of the biomass. In addition to that, cultural changes and/or financial investments along with new policies will be needed to absorb part of the costs for the use of alternative sources of energy (such as NG or Biomass), otherwise the cost of a final product can be severely affected [41].

6.2.2 Life-cycle Inventory and Life-cycle Assessment

The inventory analysis is a critical phase of the environmental assessment, as the obtained results are directly linked to the quality of the data used in the LCA [48]. The ISO Standard 14044 establishes that the data should address: time-related coverage, technology coverage, precision, completeness, representativeness, consistency, reproducibility and (un)certainty of the information [26]. The data gathering poses a problem in itself because the sources from which they can be obtained are multiple and it is not always possible to obtain the same level of detail for every part of the system under study. The best-case scenario is to build up the LCI in partnership with the industries through the use of detailed questionnaires, which is rather unlikely to happen for research purposes due to the critical nature of the information required. Some industries communicate publicly the impact of their activities through ERs and EPDs. However, neither of the abovementioned documents are mandatory nowadays, despite the fact that the EC has been recommending since 2002 that EPDs should be compulsory [49]. This results in the unavailability of the data and when available, also holds the risk of misinterpretation or double counting.

The larger part of the LCIs are mostly based on data from EPDs, ERs and literature (i.e., papers and theses) [48]. While a certain level of truthfulness is missed, generic databases are a powerful source for process modelling, among which EcoInvent [50] and Gabi [51] are considered the most complete available for the construction sector [52]. A less traditional approach to build the LCI of the system process would be to design, at a certain level of detail, the plant that will deal with the product of interest. This is not always easy to do, nor recommended to all the practitioners of the LCA methodology, because it requires an integral background of transport phenomena (mass, energy and momentum) as well as the process engineering criteria to select the adequate devices to model the process accurately. Although literature shows that traditional data sources are widely used for research purposes, the advantage of the aforementioned methodology is that having the unit processes discretised allows to fully control the main parameters in each step of the production line (energy and mass requirements). Unlike the use of generic databases, this approach allows to easily perform the sensitivity analysis proposed in this paper.

6.2.2.1 Hydrated Lime Production

For the production of HL, the system process is defined in Figure 6.1 and the theoretical capacity of the plant is stated in Table 6.1. The system process is composed by 6-unit processes. From the aforementioned information, the quantities of all materials that enter and leave each unit based on the principle of law of conservation of mass was calculated. For the units dealing with chemical reaction, the mass of the reactants/products (i.e., in the Shaft Kiln) was determined through stoichiometry and the extent of the reactions was considered fully completed. A series of assumptions and simplifications were made, such as the humidity of the limestone as it enters to the Washing Process or its final humidity after the Drying Process. Some material flows such as calcite, water (as resource and emissions) or carbon dioxide (as emission) as well as the transports from the quarry to the plant were modelled by EcoInvent 3.6 database [50].

After having all the material flows specified, the energy requirements were determined. It is important to highlight that only the energy required to operate the devices is considered in the design, while other energy demands such as the electricity of the administrative offices or heating of the plant is not pondered. The devices were selected from catalogues of a variety of producers considering multiple criteria: type of material, feed and output size, capacity of the device, energy source (fuel, electricity), most used technologies in the lime production sector, among others. From the catalogues, the Power (kW) and the min. and max. capacity (t/h) of the devices was stored. For each device, the specific power (kWh/t of material) was calculated as the power divided by the average capacity. The kiln used in the process required special attention, given that is the central part of the process. According to the BAT document 90% of all kilns used in Europe are Shaft kiln type [11]. From this amount 21% corresponds to MFSK, 29% to PFRK, 13% to ASK, and 37% comprises a variety of shaft kilns under the category "others" [11]. Among all the specified technologies, PFRK comprises the higher percentage and therefore was selected as the kiln of the factory. Finally, the LCI comprises the material and energy requirements to produce 1 tonne of HL from cradle to gate, being this, the FU of the study, used to compare the environmental performance of the analysed scenarios.

Electricity and Fuel Mix

Both the electricity and fuel mix were created as different processes for each country, and their components and proportions adjusted correspondingly to represent the analysed scenario. The proportion of each source in the electricity mix of each country (current and future) is stated in in Section 6.2.1. For the modelling of their production the EcoInvent 3.6 database [50] was used, selecting the geographic location in the respective country whenever it was possible. For instance, for DE in 2020, 18% of the electricity mix is modelled by "Electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S – DE".

In terms of the fuel mix, each contributing proportion was modelled through the heat production process of EcoInvent 3.6 database [50] (i.e., heat production, heavy fuel oil, at industrial furnace 1MW - Europe without Switzerland). Due to the lack of data in the used database, it was not possible to geographically localize the heat production for each country. On the contrary, the "Europe without Switzerland" process was used for all countries. For the sake of reproducibility of this research, in the Appendix Section (Appendix A11 to A16) the full package of providers used to model the electricity and fuel mix from the EcoInvent Database V3.6 can be found.

6.2.2.2 Life-Cycle Assessment

The software OpenLCA was used for the impact assessment. The approach followed is consistent with an attributional LCA, where the inputs and outputs were attributed to the FU of the system by linking the unit processes of a system under an allocation procedure [48], [54], [55]. In this case, to divide the impacts arising from the same process between products and co-products was done through a mass allocation. Regarding the impact method, and in particular in view of the production of HL, it is critical to include impact categories accounting for waste production and mineral resource depletion. These categories which are two of the major impacts of mineral industry sector, are something that the most used impact analysis methods (e.g., Eco-indicator 99 and CML 2002) do not include [52], [56]. On the contrary, Impact 2002+ addresses the damage categories of Resources, Climate Change and Ecosystem quality through the use of Midpoint categories such as Global warming, Land occupation, Terrestrial Ecotoxicity and Mineral Extraction among others [53]. An overview of the considered baseline impact categories and their characteristic factor is presented in Table 6.2.

Table 6.2. Overview of the main sources for characterization factors and impact categories according to the Impact 2002+ impact method. Obtained from Impact 2002+ User Guide [53].

Midpoint category	Midpoint reference substance	Damage category	Damage unit
Human toxicity (carcinogens + non-carcinogens)	kg Chloroethylene into air$_{-eq}$	Human health	DALY
Respiratory (inorganics)	kg $PM_{2.5}$ into air$_{-eq}$		
Ionic radiations	Bq Carbon-14 into air$_{-eq}$		
Ozone layer depletion	kg CFC-11 into air$_{-eq}$		
Photochemical oxidation [= Respiratory (organics) for human health]	kg Ethylene into air$_{-eq}$		n/a
Aquatic ecotoxicity	kg Triethyleneglycol into water$_{-eq}$	Ecosystem quality	PDF*m^2*y
Terrestrial ecotoxicity	kg Triethyleneglycol into soil$_{-eq}$		
Terrestrial acidification/nitrification	kg SO_2 into air$_{-eq}$		
Aquatic acidification	kg SO_2 into air$_{-eq}$		
Aquatic eutrophication	kg PO_4^{3-} into water$_{-eq}$		
Land occupation	m^2 Organic arable land$_{-eq}$*y		
Water turbined	Inventory in m^3		
Global warming	kg CO_2 into air$_{-eq}$	Climate change (life support system)	kg CO_2 into air$_{-eq}$
Non-renewable energy	MJ or kg Crude oil$_{-eq}$ (860kg/m^3)	Resources	MJ
Mineral extraction	MJ or kg Iron$_{-eq}$ (in ore)		

6.3 Results and Discussion

6.3.1Life-cycle Inventory of the Hydrated Lime Production

For the development of the LCI of the HL, the theoretical plant of Figure 6.1 was divided into six-unit processes, unifying them according to the related operations being performed. The parameters as well as the technical considerations for the selection of the devices in each unit operation are described below.

- Mineral extraction and Primary Crushing (I): The truck hauling carries the material around 1.4 km. This average distance is the result of the analysis of several lime factories. Afterwards, it is discharged on a conveyor belt that ends in the primary crushing, which is modelled by a Jaw Crusher designed and selected from a catalogue (Power 45 kW). It was assumed that the limestone comes with 10% of dust, 2% humidity, feed size 1500 m and output size 250 mm.

- Washing, drying and sedimentary pool (II): The washing machine was designed, and a Washer was selected from a catalogue (Power 45 kW). During the washing it was assumed that 5% of the water leaves with the washed limestone (i.e., with no fines). The washing water is directed to a Sedimentary Pool, where the main devices considered are 3 equivalent centrifugal pumps (Design theoretical power 7.5 kWh). The dust leaves this device as waste and the recirculated water is 90% of the feed to the washing machine. The device used in the process "dryer" is a Rotary Kiln Drier (Power 30 kW) and the material leaves the drier with depreciable humidity.
- Secondary crushing and screening (III): The main device of this process is a Jaw Crusher (feed size 250 mm, output size 30 mm, Power 30 kW) selected from a catalogue. The MLS screened to below 30 mm (around 20% of the crushed limestone) are sold as by-product.
- Calcination (IV): According to the literature, the average energy consumption of this device is 3.9 ± 0.5 MJ/kg CaO [11], [12], [57]–[60]. This value is depending on many factors among which are included the kiln efficiency, the type of fuel used, the capacity of the kiln, the amount of air in excess and the temperature of the air. The feed size is 30 mm. It was assumed that the CO_2 emissions associated to the limestone decomposition are equivalent to the stoichiometric amount resulting from Reaction C3.1 (0.44 $tCO_2/t\ CaCO_3$).
- Screening & Sizing (V): The main device is a Vertical Fine-powder Mill (feed size 30 mm, output size <20 mm, Power 335 kWh) designed and selected from a catalogue. The CaO produced is allocated in mass according to the amount required in the manufacture of each product (i.e., 36% for PL, 36% for ML, 4% to MoL and 28% for HL).
- Slaking (VI): The device is a Multi Stages Hydrator (Power 20 kW) designed and selected from a catalogue to fulfil the requirements.

From the aforementioned considerations, the LCI for the Cradle-to-Gate production of HL is presented in Table 6.3. In this table it is presented the 6 Unit Operations described, the detail of the modelled process, the processed amount (i.e., the mass and energy requirements of each unit process to produce 1 t of HL) and the inventory amount (i.e., the normalized amount of the particular unit process per t of the reference unit).

Table 6.3. Life-cycle Inventory of the Cradle-to-Gate HL production

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
Mineral extraction & Primary Crushing (I)						
Input	**$CaCO_3$**	7.00	t			Modelled by EcoInvent (Calcite, in ground)
	Water use	0.15	m^3	0.02	m^3 / t $CaCO_3$	Modelled by Ecoinvent limestone quarry operation
	Land occupation	0.59	m^2*year	0.08	m^2*year / t $CaCO_3$	Modelled by EcoInvent (Land Occupation - RoW)
	Blasting	1.13		0.16	kg / t $CaCO_3$	Modelled by EcoInvent (Blasting - RoW)
	Diesel consumption (Truck hauling, drilling machine and Loading machine)	161.00	MJ	23.00	MJ / t $CaCO_3$	Modelled by EcoInvent (Diesel, burned in building machine - GLO)
	Explosive	1.13	kg	0.16	kg / t $CaCO_3$	Modelled by EcoInvent (Explosive production Tovex)
	Transformation due to mineral extraction	0.05	m^2	0.01	m^2 / t $CaCO_3$	Modelled by Ecoinvent limestone quarry operation
	Recultivation (limestone mine)	0.05	m^2	0.01	m^2 / t $CaCO_3$	Modelled by Ecoinvent limestone quarry operation
	Jaw Crusher	1.87	kWh	0.27	kWh / t $CaCO_3$	Electricity mix (SUBLime designed)
	Conveyor belt	0.03	kWh	0.004	kWh / t $CaCO_3$	Electricity mix (SUBLime designed)
Output	**$CaCO_3$ Crushed[1]**	7.00	t			Main product as a result of (I)
Washing, drying and sedimentation (II)						
Input	**$CaCO_3$ Crushed[1]**	7.00	t			Input from (II)
	Water for washing	0.98	t	0.14	t / t $CaCO_3$ Crushed[1]	Modelled by SUBLime
	Sedimentary pool Operation	27.10	kWh	3.88	kWh / t $CaCO_3$ Crushed[1]	Electricity mix (SUBLime designed)
	Washing Machine Operation	5.47	kWh	0.78	kWh / t $CaCO_3$ Crushed[1]	Electricity mix (SUBLime designed)
	Drying Machine Operation	9.33	kWh	1.33	kWh / t $CaCO_3$ Crushed[1]	Electricity mix (SUBLime designed)
Output	**$CaCO_3$ Washed**	6.16	t	0.88	t $CaCO_3$ Washed / t $CaCO_3$ Crushed[1]	Dry $CaCO_3$, first crushing operation
	Fines washed	0.70	t	0.10	t / t $CaCO_3$ Crushed[1]	Modelled by Ecoinvent (disposal, ordinary industrial waste)
	Water	0.53	t	0.08	t / t $CaCO^3$ Crushed[1]	Humidity removed after the Washing Process

Table 6.3. Life-cycle Inventory of the Cradle-to-Gate HL production *(continued)*

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
Secondary crushing and screening (III)						
Input	**$CaCO_3$ Washed**	6.16	t			Input from (II)
	Jaw Crusher Operation	4.10	kWh	0.67	kWh / t $CaCO_3$ washed	Electricity mix (SUBLime designed)
Output	**$CaCO_3$ Crushed[2]**	4.92	t	0.80	t $CaCO_3$ Crushed[2] / t $CaCO_3$ Washed	Main product as result of (III) - Allocation by mass (0.8)
	MLS	1.23	t	0.20	t MLS / t $CaCO_3$ Washed	Allocation by mass (0.2)
Calcination (IV)						
Input	**$CaCO_3$ Crushed[2]**	4.92	t			Input from (III)
	Shaft Kiln Operation	133.00	kWh	27.10	kWh / t $CaCO_3$ Crushed[2]	Electricity mix (SUBLime designed)
	Shaft Kiln fuel consumption	13000.00	MJ	2640.00	MJ / t $CaCO_3$ Crushed[2]	Fuel mix (SUBLime designed)
Output	**CaO**	2.76	t	0.56	t CaO / t $CaCO_3$ Crushed[2]	Product as a result of (IV)
	CO_2	2.17	t	0.44	t CaO / t $CaCO_3$ Crushed[2]	Stoichiometric CO_2 emission due to Limestone decomposition
Screening & Sizing (V)						
Input	**CaO**	2.76	t			Input from (IV)
	Vertical Mill Operation	92.40	kWh	33.50	kWh / t $CaCO_3$ Crushed[2]	Electricity mix (SUBLime designed)
Output	**CaO for Hydrated Lime**	0.77	t	0.28	t CaO for Hydrated Lime / t CaO	Main product as result of (IV) - Allocation by mass (0.28)
	PL	1.00	t	0.36	t Pebble Lime / t CaO	Co-product as result of (IV) to be sold as Pebble Lime - Allocation by mass (0.36)
	ML + CaO for MoL	1.00	t	0.36	t CaO for ML & MoL / t CaO	Co-product as result of (IV). Less than 0.4% of the produced CaO is used to produce MoL, therefore it is allocated altogether with ML- Allocation by mass (0.36)

Table 6.3. Life-cycle Inventory of the Cradle-to-Gate HL production *(continued)*

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
			Lime Hydration (VI)			
Input	**CaO for Hydrated Lime**	0.77	t			Input from (V)
	Water	0.25	t	0.32	t / t CaO for Hydrated Lime	Modelled by SUBLime (Ecoinvent tap water production, Europe without Switzerland)
	Lime Hydrator Operation	0.27	kWh	0.35	kWh / t CaO for Hydrated Lime	Electricity mix (SUBLime designed)
Output	**Hydrated Lime**	1.00	t			Main product as result of (III)
	Emissions, Waste					Emissions and waste along the production chain of Hydrated Lime

As a means to analyse the quality of the LCI, a LCA under the conditions described in Section 6.2.2.1 was performed and the equivalent $CO_{2\ eq}$ emissions per kg of product (CaO and $Ca(OH)_2$) calculated. Table 6.4 shows the comparison of the results of this study (called SUBLime according to the running ITN EU project) to different other sources. The results reported by EEA [11] for CaO indicate that only the CO_2 emissions due to the limestone decomposition have been considered (stoichiometric). The EuLA LCI study has passed successfully the independent external critical review from Rina Consulting [61], being the most representative dataset Europe-wide. Both, EcoInvent and SUBLime results represent the production of Lime and Hydrated Lime in DE.

In Table 6.4, it can be observed that the SUBLime results are in the same order of magnitude as results from the other mentioned sources for both considered products. For quicklime production, the greatest differences are with the reported values from EEA. Furthermore, the differences with the EcoInvent database [50] can be explained considering different system boundaries and/or technologies used for the production (types of kilns, fuels, etc.). However, the results of EuLA and SUBLime are very close (7% relative difference). Beyond the differences detected, the theoretical study correlates well with the sources used for comparison. Therefore, the SUBLime LCI for the Cradle-to-Gate production of HL (Table 6.3) is used in the following section for the case-studies.

Table 6.4. $CO_{2\ eq}$ emissions per ton of product.

Source	Stoichiometric	EEA	EuLA	Ecoinvent V3.6	SUBLime
kg $CO_{2\ eq}$ / kg CaO	0.75	0.75	1.17	0.95	1.26
kg $CO_{2\ eq}$ / kg $Ca(OH)_2$	0.59	-	0.92	0.85	0.94

6.3.2 Life-cycle Impact Assessment

Figure 6.4 to 6.8 shows the LCA results for the production of 1 kg of HL, under the conditions described in Section 6.2.1. The results comprise the environmental impact of current and potential future scenarios for DE, BE, PT and ES. Four categories have been created, to account for environmental impact assigned to each indicator and scenario analysis, namely: Raw material emissions (Limestone decomposition), Quarry operation (including the drilling, blasting at the quarry and the transport by truck to the primary crushing), Electricity (used to operate the devices of the plant, including all the operations mentioned in Table 6.3) and Kiln Operation (fuel consumption to provide the thermal energy required for the limestone decomposition).

6.3.2.1 The 2020 Scenario

Figure 6.4 shows the environmental impact for the production of 1kg of HL in 2020 in DE, BE, PT and ES. In these results, the only variable is the electricity mix considered in each country for the year 2020, while the share of quarry and kiln operation are the same for each country. In general, it is noted that the environmental impact of HL production depends on the country.

The Climate change damage category is measured by the Impact 2002+ method considering the kg of CO_2 into air-eq emitted during the life cycle, that can be linked to the potential increase of 1.5°C above temperatures in the pre-industrial period [45]. As for the GW indicator (kg$CO_{2\ eq}$/kg HL), for all the analysed countries the effect of the electricity mix used is negligible in comparison to the effect of the fuel mix and the inherent CO_2 emissions of the limestone decomposition. Around 60% of the total $CO_{2\ eq}$ emissions can be attributed to the chemical reaction of decomposition [12], [62], 39% are assigned to the fuel combustion and 1% to the electricity consumption at the plant. Even though, the chemical emissions are inevitable, there is room for improvements with respect to the fuel combustion emissions. In terms of global warming, the kg CO_{2eq} per kg HL produced can be considered equal to 0.94. However, the calcination does not dominate all categories. In 9 out of 15 indicators, the share of electrical energy consumption in the plant is not negligible and the most important four are discussed below.

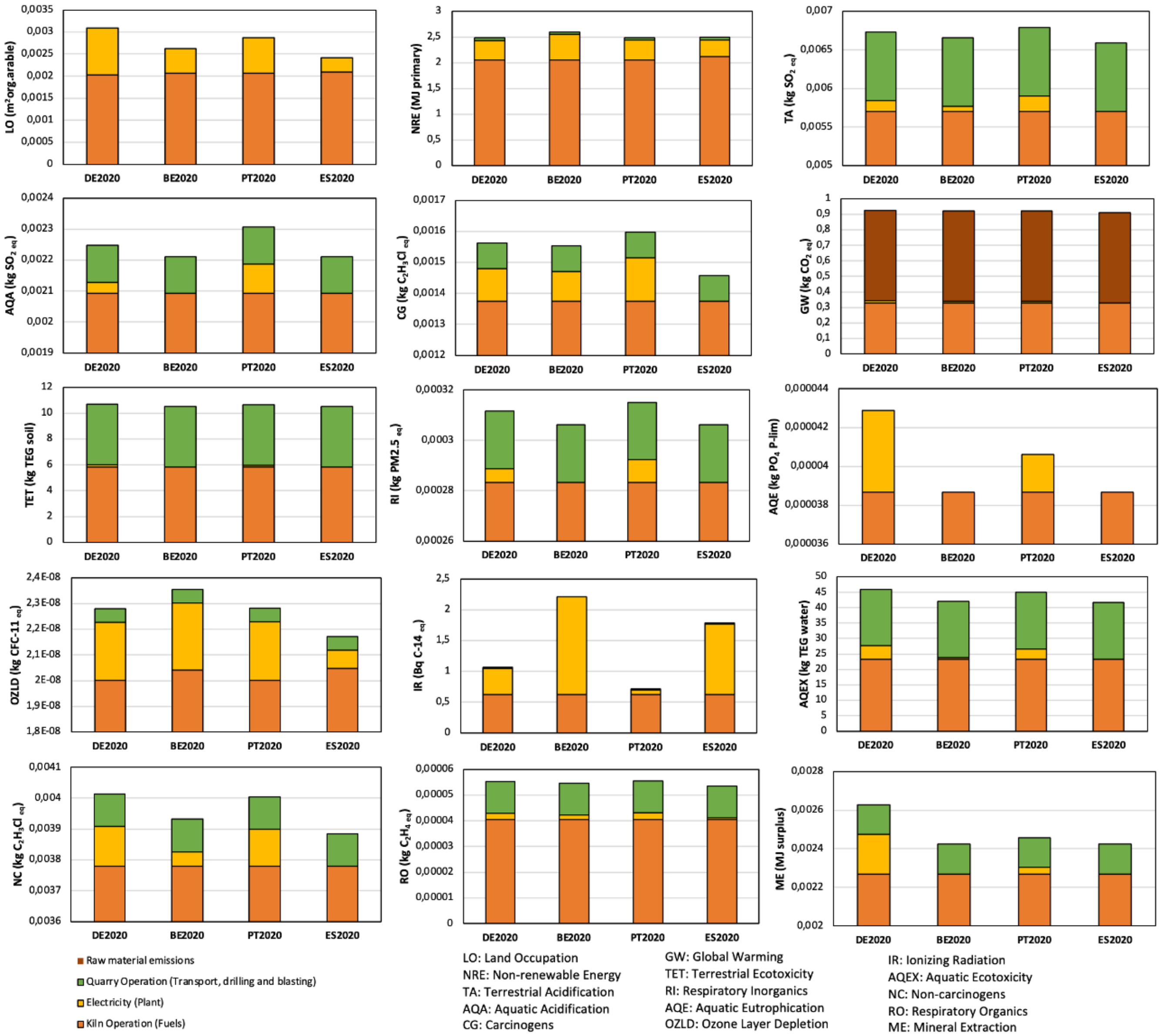


Figure 6.4. Life Cycle Impact Assessment of 1 kg Hydrated Lime in 2020 for DE, BE, PT and ES.

In terms of the Ecosystem quality, two midpoint categories are highlighted: Aquatic Eutrophication and Land Occupation. The first one, quantifies a major water quality issue, related to excessively high environmental levels of macronutrients (nitrogen and phosphorus), which provoke an increased growth of algae [63]. The higher effects in this category are registered for DE and PT, mostly due to the proportion of hard coal and hydroelectric production of energy. The Land Occupation indicator considers the area occupied, the duration of occupation and the damaging potential for ecosystem quality of a specific land use type (m^{2}organic arable land*year) [64]. It gives an insight of the damaging potential for the ecosystem quality of using a specific area, for a certain amount of time and for a specific activity.

The mining industry is well known for having a significant impact on the use of natural resources, and usually intuitively the highest impact is assigned to the lime quarry that is being exploited. However, it is very interesting to find out that the production of energy (both heat and electricity and mainly from hard coal and natural gas) was dominating the indicator, even though a land occupation for the extraction of limestone was considered in the Life Cycle Inventory (Table 6.4 of the manuscript). While common sense would suggest that the Quarry operation would have a significant contribution to the magnitude of the indicator, this is not the case. The reasons are various. As mentioned before the Land Occupation indicator is much more complex than the mere use of a specific area because it takes into account in which way the land is used, to assign the impact factors that characterize the elementary flows, that in the end are aggregated in the indicator. Digging deeper into the Impact2002+ method, the specific factor assigned to the Land Occupation of the Mineral Extraction site is around 35% less than the impact factor assigned to the production of heat and electricity (on average). This impact factor is multiplied by the inventory result, which is the second explanation for the results that were found. For instance, taking a look at the scenario 2020 for Germany (Figure 6.4), when analysing the inventory results, the Mineral Extraction and Primary Crushing operation contribute for around 4% to the impact category. The remaining 96% of the inventory is distributed to the land occupation for the production of energy (around 65% heat production, 31% electricity production). Therefore, the contribution of the land use of the quarry is negligible in comparison to the one assigned to the energy production.

Concerning the Human health damage category, the Ozone Layer Depletion and Ionizing Radiation stand out. To start with, the ozone layer is a band of gasses, mostly ozone (O_3), located 15-30 km above the Earth (stratosphere) that absorbs most of the Sun's ultraviolet radiation. After the discovery of a dangerous 'ozone hole' in the stratosphere, all nations in the world agreed in 1987 to act under the Montreal Protocol on substances that deplete the ozone layer (ODS) [65]. In 2009 the EU released a regulation on highly detrimental substances, including chlorofluorocarbons, hydrochlorofluorocarbons, hydrobromofluorocarbons [66], although climate change and greenhouse gasses such as methane and nitrous oxide may also have an effect [67]. Looking at the results of the baseline scenario, the electricity (DE, BE and PT) is responsible for around 50% of the ozone depletion indicator (kg $CFC\text{-}11_{eq}$). This is mainly related to the electricity production from fossil fuels (primarily natural gas and hard coal), as well as biofuels because of the associated NO_2 emissions of the combustion [68]. In ES the impact of the electricity is smaller, and therefore, the overall value of the indicator, because the proportion of electricity production by natural gas and hard coal is also smaller in comparison to the rest of the countries. Secondly, Ionizing Radiation comprises wavelengths between 10-8-10-15 m (UV, X-ray, and Gamma rays, for instance) which are very high in energy. It can cause biological effects, particularly change of molecules within the cell and is proven to have a carcinogenic effect, malformation, growth retardation and impaired brain function [69], [70]. The radiation is emitted by radioactive materials (such as Uranium, Plutonium, etc.) called radionuclides and considered in the LCA method, measured in units of Bq Carbon-14 eq.

The extraction, processing, and disposal of radionuclides for nuclear energy production are a major source of ionizing radiation [71]. Consequently, countries dependent on electricity production by NE are most likely to have higher values on the IR indicator. In the case of BE (37% nuclear power share, Fig. 6.2), around 70% of the indicator corresponds to the electricity consumption in the plant. This is the case of BE that with 37% of the electricity mix composed by NE, and around 70% of the indicator is attributed to the electricity consumption in the plant. The trend is followed by ES (25% of NE), DE (11% of NE) and PT (0% of NE).

6.3.2.2 The Current vs. Potential Future Scenarios

The scenario analysis involves the simultaneous effect of switching to a decarbonized electricity matrix and the potential CO_2 savings due to a lower proportion of solid fossil fuels used in the kiln. Figures 6.5 to 6.8 show the LCA results for the 2030/2050 electricity mix matrix and the S1, S2 and S3 fuel mix scenarios along with the baseline results shown in the previous section, for comparison of the evolution.

Regarding the fuel mix used in the kiln, the shift to alternative sources has a positive effect to reduce the $CO_{2\ eq}$ emissions. Specifically, the GWP indicator is reduced by around 9, 18 and 22% for S1, S2 and S3 respectively in comparison to the current scenario (DE, BE, PT and ES). This effect is produced mainly due to the reduction in the use of FSF and the increment in the proportion of NG and Biomass in the mix. For the best-case (S3), the proportion of NG (60%) was almost doubled, and Biomass (32%) increased sixteen times compared to the baseline scenario. Nowadays, it is believed that as the power sector undergoes a low-carbon transformation, natural gas is the only fossil fuel technology likely to remain an important source of flexibility for the power systems in the future [72]. The previous results corroborate the prediction of potential CO_2 emissions savings for the analysed scenarios in Figure 6.3. Nevertheless, despite the fact that the GWP improves under these circumstances, the performed LCA reveals that other impact indicators are negatively affected. As shown in the previous section, is interesting to note that for some of these indicators, the sources of the electricity mix used in each country contribute to a significant extent and cannot be disregarded.

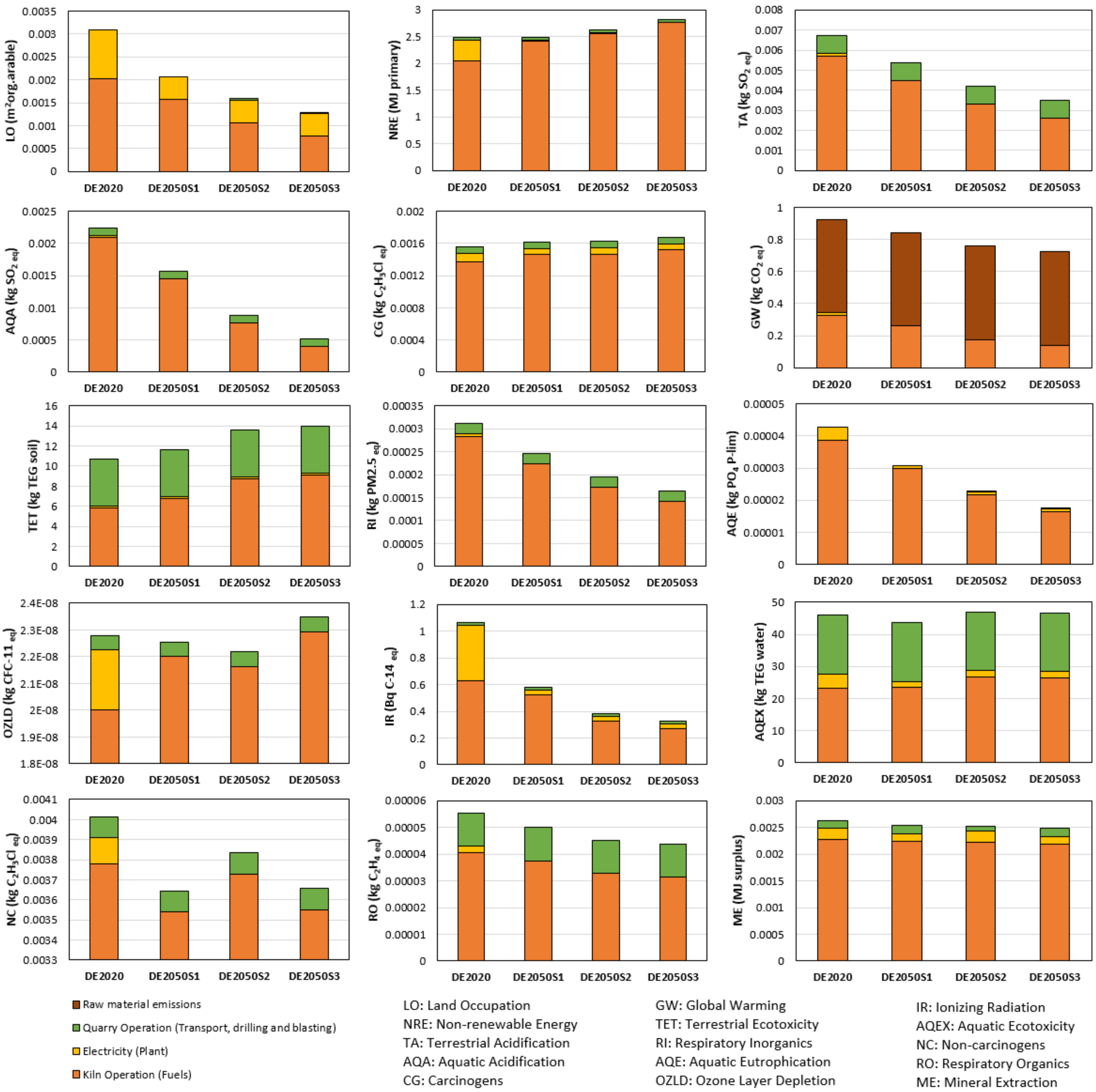
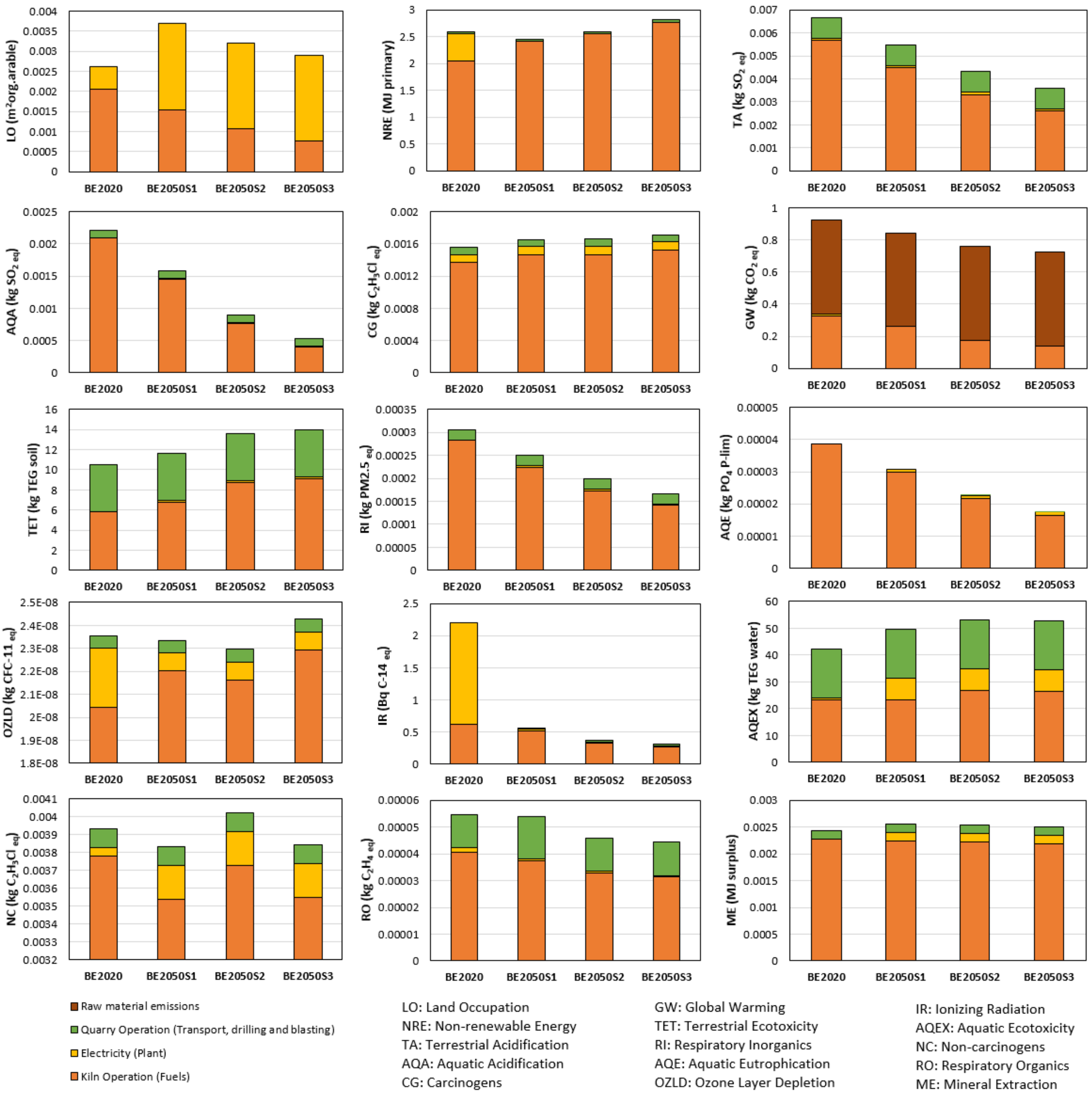


Figure 6.5. Life-cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2050) in DE



LO: Land Occupation
NRE: Non-renewable Energy
TA: Terrestrial Acidification
AQA: Aquatic Acidification
CG: Carcinogens

GW: Global Warming
TET: Terrestrial Ecotoxicity
RI: Respiratory Inorganics
AQE: Aquatic Eutrophication
OZLD: Ozone Layer Depletion

IR: Ionizing Radiation
AQEX: Aquatic Ecotoxicity
NC: Non-carcinogens
RO: Respiratory Organics
ME: Mineral Extraction

Figure 6.6. Life-cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2050) in BE

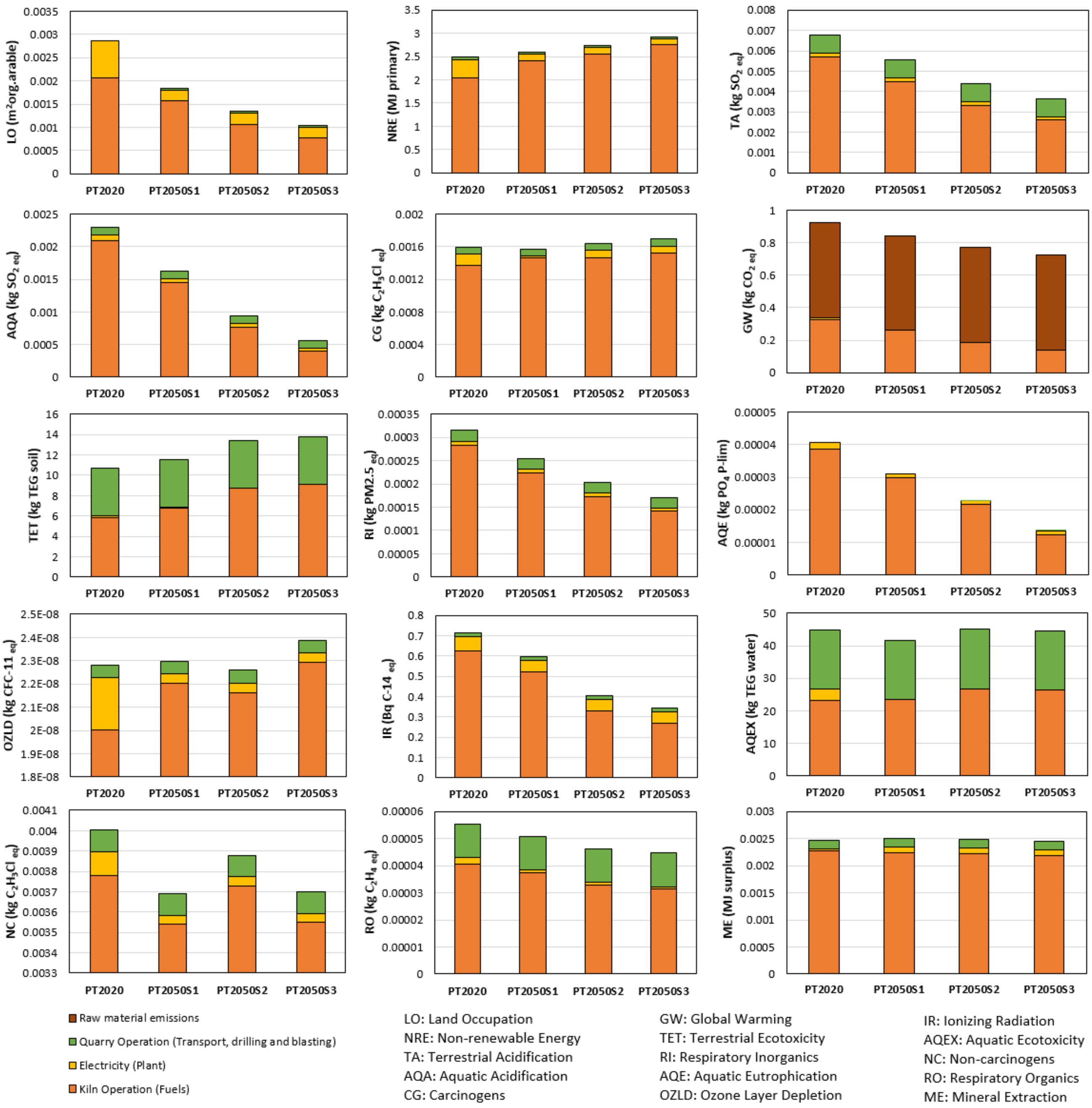


Figure 6.7. Life-cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2050) in PT

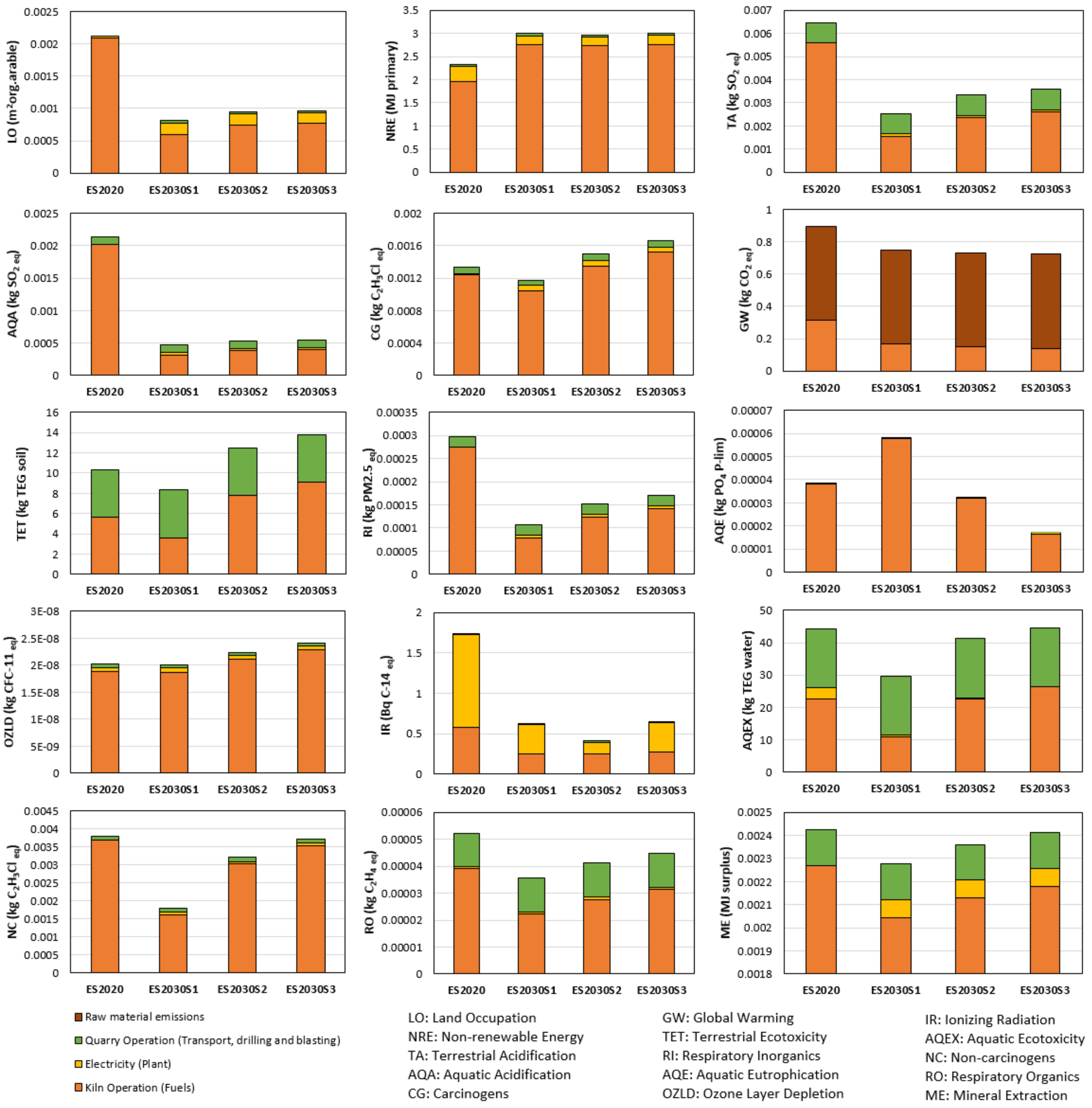


Figure 6.8. Life-cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2030) in ES

Speaking of the Ecosystem quality category, the Land Occupation midpoint category is still of interest. For all the considered countries of production, it is observed that the contribution of the kiln operation to the indicator decreases along with the replacement of the fossil solid fuel (Fig. 6.5). In the potential future scenarios, for DE, PT and ES the contribution of the electricity mix decreases with the increment of renewable sources. However, the opposite is true for BE, where the indicator appears to be highly sensitive to the increment of the biogas co-generation by 15% (Fig. 6.5). An opposite behaviour is observed for the Terrestrial Ecotoxicity indicator, with an increasing trend as new scenarios with potential CO_2 emission savings are proposed. This midpoint category represents the environmental impact of metals released into the soil. The main idea behind the indicator is that it distinguishes between Lewis's acids and Lewis's basis and considers the strength of the metal complexation and toxicity, with Cr(VI), Sb(III), Sb(V), As(III) and As(V) being most toxic in soil because of their ability to bind with organic matter [53], [73]. Not surprisingly, the Quarry Operation emerges as a very important supplement, because the drilling and blasting step that uses explosives in the quarry to break the limestone are additional sources of heavy metals [74]. The other predominant step is the Kiln Operation. In the baseline scenario, around 40% of the TE is due to the effect of the hard coal in the fuel mix, given that the coal mining activity can affect the ecosystem by the release of Lead and Cadmium (high mobility in the soil-plant system)[75], [76]. The TE indicator increases with the increment of the biomass proportion in the mix (from 2% to 32%), which might be attributed to the effect of the forestry activity [77].

In connection to the Human health damage category, for the Ozone Layer Depletion, around 45% of the indicator corresponds to the Kiln Operation, again, because of the combustion of the fossil fuels. The indicator also appears to be sensitive to the diesel consumption in the Quarry Operation. For the subsequent scenario analysis, an increase in the use of renewable sources of electricity production has a positive impact, diminishing its fraction of the OZLD category. Nevertheless, the indicator increases globally, and this is essentially because of the increment of natural gas as fuel for the kiln. Moreover, in section 6.3.1 the implications of using nuclear energy in terms of ionizing radiation were discussed. In general, all the analysed countries are planning to completely replace nuclear energy in the future (Figure 6.2) and, as a result, the influence of the electricity in the scenario analysis is depreciable. However, fuels also hold responsibility on this regard. In particular, the use of hard coal (and other fossil sources) also contributes to IR, because during the burning process, wastes containing small amount of naturally-occurring radioactive materials are generated [78]. Therefore, as expected, the Kiln Operation also contributes to the indicator, although in a lower proportion (reduction of the sharing between 5-30%).

Finally, covering the Resources-related damage category, the Non-renewable energy indicator ($MJ_{\text{total primary non-renewable energy}}$/mass or volume) is featured. The midpoint category considers for the calculations, the consumption in terms of the total primary energy extracted for energy carriers and the upper heating values of the energy source [53]. The category is almost entirely dominated by the Kiln operation, where the highest consumption of Non-renewable energy is produced. It may be interesting to note that as the shift to less carbon-intensive sources is achieved (replacing fossil solid sources by natural gas); the indicator does not improve but on the contrary. To understand the reason behind, a detailed analysis of the inventory to produce 1 MJ of thermal energy from NG and HC was performed (Ecoinvent V3.0, see Table A5 for reference of the used providers). For NG, the Impact2002+ method assigns an impact factor of 38.3 $MJ_{primary}/m^3$ and 0.033 m^3 of natural gas are required as the only flow contributing to the indicator (approx. 1.2 $MJ_{primary}$/MJ delivered). Furthermore, in the case of HC, 0.0021 kg of crude oil and 0.0013 kg brown coal are part of the inventory with an assigned impact factor of 45.8 MJprimary/kg and 9.9 $MJ_{primary}$/kg respectively (0.12 $MJ_{primary}$/MJ delivered).

In all cases, the contribution of the Electricity and the Quarry Operation is not significant. In retrospect, it may be important to mention the Mineral Extraction indicator, as it complements with the Non-renewable energy indicator to assess the resource depletion. It is measured in $MJ_{surplus}/kg_{extracted}$ and expresses the expected increase in extraction energy needed to extract 5 times the cumulative extracted amount, considering that the resources become scarcer with the time [64]. It may be worth mentioning that even though it is related to the extraction process of limestone (because of the specific energy consumption in the LCI, Table 6.4), this indicator is referring to the energy used in the process and not the depletion of the limestone mineral itself. In all cases, three main components of the indicator can be distinguished: Quarry Operation, Electricity and Kiln Operation. For the Quarry Operation, the contributing sub-processes are the blasting and explosive production, accounting for around 6-8% of the total indicator depending on the analysed country. However, the indicator is mostly dominated by the fuels providing heat to the kiln in the first place, and the electricity to power the plant on the second place. In the current scenario (Figure 6.4), for DE2020 the indicator is around 8% higher than the average, because of the current electricity matrix and in particular, due to the hard coal, natural gas and biogas sources. Hard coal and natural gas used as fuel in the kiln are also the main components of the Kiln Operation share. In the future scenarios, the switch to a lower $CO_{2\ eq}$ fuel matrix actually does not improve the indicator, because the reduction in the impact associated to the hard coal is compensated by the increased effect of the natural gas and the biomass production (which also includes the energy required to dry the wood chips, as explained in Section 6.3.1).

6.3.2.3 Final Comments

Figure 6.9 aims at summarizing the workflow of the scenario analysis development, as well as emphasizing the main findings of the research carried out.

As revealed in the literature review, for the building of the scenario analysis, Europe has a great potential to produce energy with a lower carbon footprint. Currently (baseline scenario 2020), the share of the renewable energy for electricity production for the analysed countries is around 30, 39, 45 and 60% for BE, ES, DE and PT respectively (Figure 6.2). Nevertheless, the results show that not only the amount of RES feeding the mix is important, but also what type of non-renewable sources are being used. In particular, the indicators have proven to be highly sensitive to the proportion of hard coal and natural gas. Therefore, for the current scenario (Figure 6.4), ES and BE have shown the lower environmental impact, namely in 7 out of 15 indictors and 5 out of 15, respectively, followed by PT (1 out of 15) and DE (1 out 15).

As a key remark, in 12 out of 15 indicators, the electricity mix contributes to a meaningful extent to their magnitude. Naturally, the fuel used in the kiln is also significant in all the impact categories analysed and dominates around 13 of the indicators. This fact is, once again, attributed to the FSF.

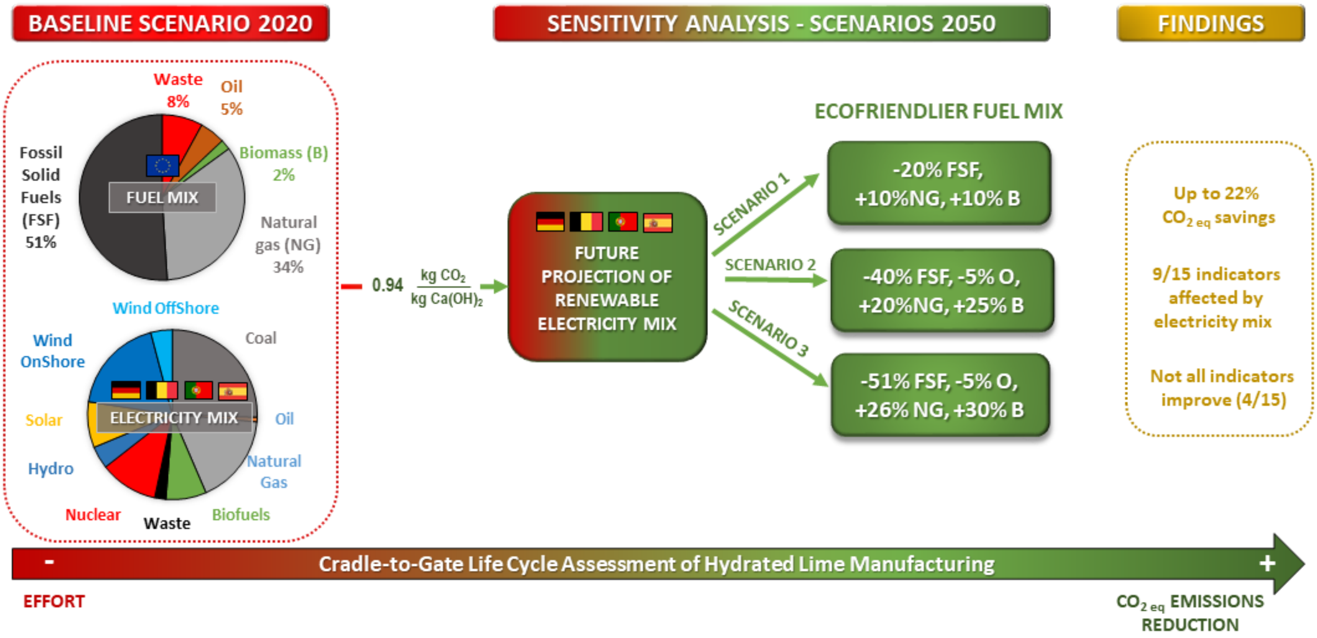


Figure 6.9. Workflow of the scenario analysis development and main findings of the research

In the future (sensitivity analysis), all the analysed countries are planning to base their electricity production on the use of two main sources: Wind power along with solar energy. These sources would dominate the sharing, amounting to around 70% of the market (Figure 6.2). In a lower proportion, other sources such as hydropower, biofuels and combined cycles appear as an alternative. The shift to almost 100% renewable energies for electricity production, certainly has a positive effect on the impact indicators, reducing their shared proportion in around 12 out of 15 indicators. Nevertheless, the opposite holds true for the different fuel mix scenarios.

Whereas significant reductions of $CO_{2\,eq}$ emissions are obtained by reducing the amount of FSF, for the analysed countries, the indicators NRE, CG, TET and OZLD increases up to 20, 8, 25 and 9% respectively (in comparison to 2020 scenario). While common sense would suggest that replacing current energy sources with those with a lower carbon footprint would lead to an improvement in all environmental indicators, the results show that this is not trivial. Therefore, it supports the idea that the issue of environmental sustainability cannot be limited to GWP alone, but requires a holistic analysis of the situation, using all available indicators and tools to interpret the reality reliably.

Furthermore, it is important to understand that sustainability is not simply a matter of reducing environmental impacts. Instead, the appeal of low-carbon and green-growth development paths stems from the desire to avoid 'locking in' to development paths that may become increasingly costly as resources become scarcer and carbon pricing becomes more important over time [79]. As mentioned during the analysis of potential fuel mix scenarios, achieving sustainability in the lime industry and therefore, reaching the goals of clean and eco-friendly production proposed by the UN, requires the cooperative collaboration of scientists, policymakers, the private sector, and society as a whole.

6.4 Conclusion

This paper reports a case study for the production of hydrated lime, located in Germany, Belgium, Portugal, and Spain. The focus is on the effect of the fuel and electricity mix used in the plant, in the current scenario and in potential future scenarios. The following conclusions can be drawn:

- A theoretical Cradle-to-Gate LCI for Hydrated Lime production was developed and a LCA was performed to compare the results with databases (EcoInvent V3.6) and reports by specific institutions (European Lime Association and European Environmental Agency). The environmental impact analysis of the theoretical study is in good agreement with results of the European Lime Association (7% relative difference for the Global Warming Potential indicator)
- For the 2020 scenario, in 9 out of 15 indicators, the share of electrical energy consumption in the plant is not negligible (from 5 to 50% relatively). The most affected indicators were Land Occupation, Aquatic Eutrophication, Ozone layer depletion and Ionizing Radiation. Among the first three the presence of fossil fuels dominates, while for Ionizing Radiation dominates the use of nuclear energy. In terms of Global Warming Potential, 0.94 kg CO_{2eq}/kg HL are produced. The emissions are due for 60% to limestone decomposition, 39% is attributed to the combustion of the fuel and 1% to the electricity consumption. Spain and Belgium have shown lower environmental impact, namely in 7 out of 15 indictors for Spain and 5 out of 15 for Belgium, followed by Portugal (1 out of 15) and Germany (1 out 15).
- Regarding the potential future scenarios, on the one hand, the shift to almost 100% renewable energies for electricity production, has a positive effect on the impact indicators, reducing their shared proportion in around 12 out of 15 indicators. Furthermore, as the proportion of fossil solid fuel decreases and NG and biomass increases in the fuel mix, a reduction of 9, 18 and 22% in the Global Warming Potential is achieved (compared to the 2020 scenario). Nevertheless, the results reveal that although Global Warming Potential is reduced, the indicators Non-renewable energy, Carcinogens, Terrestrial Eco-toxicity and Ozone Layer Depletion increase up to 20, 8, 25 and 9% respectively (in comparison to the 2020 scenario) due to the future fuel mix designs.

Finally, the result of this research implies that a change from fossil solid fuel to other sources (or a change to renewable energy only) will not necessarily lead to reduction for all impact categories and that a well-informed choice for a combination of energy sources should be made to obtain a balanced reduction for most impacts. Special attention should be paid to low-carbon and green-growth development paths to consider that resources become scarcer, and carbon pricing becomes more important over time.

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Chapter 7: Towards Net-zero and Carbon-negative Transformations in Lime-Based Rendering Materials

This chapter reports a comprehensive decarbonization approach, involving Direct Separation Reactors (DSR) with different operation modes and eco-efficient energy sources in the production of hydrated lime. Environmental and economic impacts are calculated through an in-depth life-cycle cradle-to-grave assessment for a lime-based render.

The feasibility of net-zero and carbon-negative scenarios in lime-based plasters, combining the decarbonization strategies at the manufacturing plant with the natural carbonation are proven.

To realize a swift and effective decarbonization of the lime industry, a harmonized effort is imperative and involves balancing interests of the private sector, environmental protection, and promoting societal well-being, all within a supportive regulatory framework. The challenges and coordinated actions across the value chain are discussed.

Publication 6: From Quarry to Carbon Sink: Process-based LCA Modelling of Lime-based Construction Materials for Net-zero and Carbon-Negative Transformation

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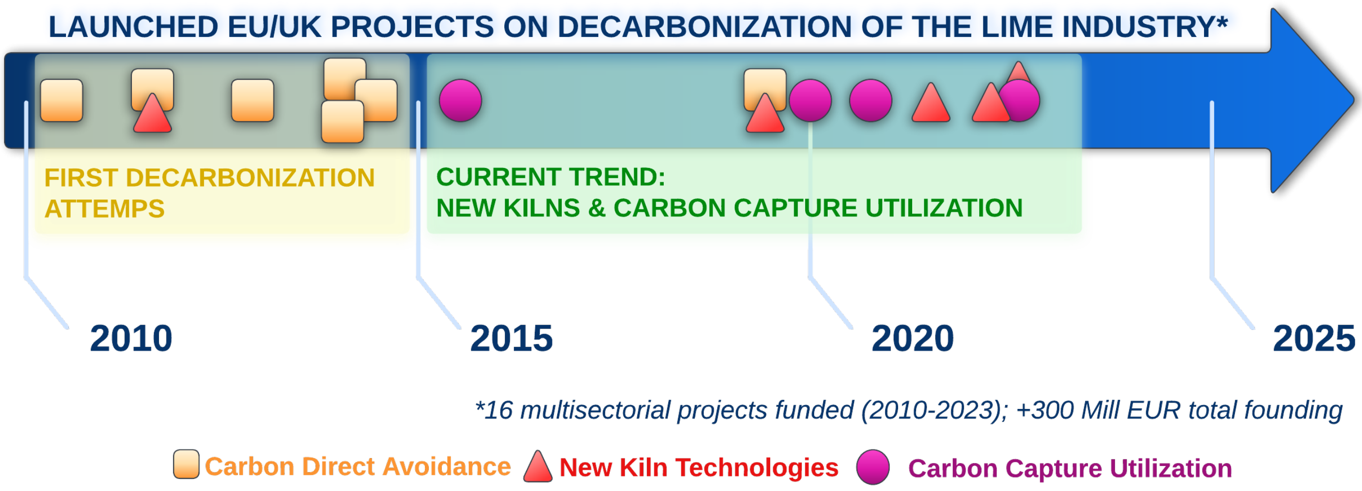
7.1 Towards Net-zero and Carbon-negative Transformations in the Lime Sector

Lime stands as one of the world's oldest and most vital construction materials. In 2020, the lime industry boasted a global market value of approximately 42 billion USD, accompanied by a total lime production of 396 million metric tons (Mt). Forecasts predicting a 21% market expansion and a 25% rise in lime production by 2028 represent formidable challenges to environmental sustainability [1]. Despite Europe's 2020 output of 20 Mt/y in 2020 [2], it lags Asia, the leading producer, boosted by a rapidly expanding construction sector [1]. However, the energy-intensive nature of lime production, regardless of factory location, yields an average of 1.2 t CO_2/t CaO [3]. When combined with the 90 €/tCO_2 carbon pricing under the European Trading System (ETS) in 2022 [4], these factors represent significant obstacles in achieving both environmental sustainability objectives and maintaining its market competitiveness.

Calcium Oxide (CaO) and Calcium Hydroxide ($Ca(OH)_2$) serve as the main precursors for lime-based construction materials, comprehending aeriated concrete, bricks, mortars, renders, and plasters used in both historical and modern structures[5]. Lime plays an indispensable role in imparting essential properties to these products [6]. Consequently, the projected sustained high production of lime threatens to escalate environmental impacts, endangering the realization of the United Nations' goal to limit global warming to 1.5°C [7]. In 2020 alone, industrial CaO production contributed 475.2 Mt of CO_2 globally, with 24 Mt allocated to the European market. These emissions are categorized as either unavoidable emissions - UE (generated during limestone decomposition into CaO and CO_2 in the lime kiln) or avoidable emissions - AE (associated with electricity consumption and fuel combustion during the manufacturing processes) [3].

In the realm of lime-based mortar, render, and plaster (LBMRP) production, an astounding 80% of the Cradle-to-Gate CO_2 emissions can be attributed to the manufacturing of $Ca(OH)_2$ within the dry mix [8]. It is therefore imperative that the decarbonization efforts focus primarily on lime manufacturing to effectively oppose this challenge. As a noteworthy milestone, the European Union has set ambitions goals, aiming for a net emissions reduction of at least 55% by 2030 when compared to 1990 levels [9]. To meet this target, various EU-funded projects have been initiated, working closely in collaboration with the industrial sector to pursue process emissions and possible decarbonation strategies, as depicted in Figure 7.1 (adapted from [10]). Appendix 17 provides a comprehensive list of key projects in the lime sector. Remarkably, out of 15 projects listed, 12 of them boast a combined budget nearing 290 million EUR, with funding distributed as follows: a substantial 79% coming from public institutions (EU/UK) and the remaining 21% sourced from private capital.



*16 multisectorial projects funded (2010-2023); +300 Mill EUR total founding

Figure 7.1. Launched European/UK projects on decarbonization of the lime industry (adapted from [10]).

During the period spanning 2010 to 2015, a significant portion of these projects focused on harnessing the potential of renewable energy sources like biomass, biogas, solar panels, and minor kiln adjustments to enhance overall thermal efficiency (Fig. 7.1). However, it's important to note that even with these efforts, approximately 30% of the total CO_2 emissions [11] still originate from AE, necessitating the pursuit of additional solutions to address the remaining UE. Regrettably, there is limited headway in the literature concerning UE from lime production from 2010 to 2020. Only a few articles explored strategies involving solvents, sorbents, mineralization, and alkalinization of waste materials, resulting in relatively modest technological advancements [12]–[15]. Nevertheless, since 2020, we have witnessed ground-breaking innovations in traditional lime manufacturing, with particular focus on kiln designs incorporating direct separation technology (DST), enabling the direct capture of CO_2 during the calcination of $CaCO_3$ [16].

Noteworthy EU initiatives, such as LEILAC1 and LEILAC2 (https://www.leilac.com/), boasting a combined budget of 55 million euros, have been dedicated to the development of the CALIX kiln equipped with DST technology. Their ambitious goal is to scale up production to 400 t/d [17].

Unlike any other binder material used in the construction industry, lime production has the distinctive competitive advantage of generating pure CO_2. Nevertheless, an effective management and utilization of this separated CO_2 becomes paramount. Consequently, alongside the development of new kiln technologies, projects specifically targeting the CO_2 value chain were initiated (Fig. 7.1). Captured CO_2 can either be sequestrated in storage sites (Carbon Capture and Storage, CCS) or, alternatively, harnessed to produce value-added products (Carbon Capture and Use, CCU). CCS faces several challenges, including high costs, limited financial incentives, and the challenge of securing adequate storage space [18], [19]. On the other hand, CCU, whenever combined with renewable energy sources, holds immense potential as a component for a sustainable circular economy [20], [21]. Several projects are actively pursuing this direction (see Appendix 14). One of the most recent and cutting-edge endeavours is COLUMBUS (https://columbus-project.com/), which secured a substantial budget of 150 million Euros in 2022 to produce e-methane from hydrogen and CO_2. Progress, however, remains ongoing. In the EU ETS Innovation Fund's 2023 call, a staggering 1.4 billion euros have been allocated for 8 decarbonisation projects, with 5 of them dedicated to CCU in the lime and cement industry [22].

The EU has taken significant steps to address climate change through the introduction of the Fit-for-55 package, which became EU law in July 2023. This package is specifically designed to provide the necessary legal framework to support the EU's 2030 Climate Target Plan [9], [23], [24]. These legislations encompass the establishment of deadlines, carbon pricing mechanisms, and targeted incentives for some CCU pathways. This paradigm shift allows the lime industry to consider CO_2 as a valuable resource that can be leveraged to produce value-added products through CCU technologies. The lime industry is now at a pivotal juncture, presenting a unique opportunity to play a decisive role in the sustainable transformation of multiple markets while simultaneously decarbonizing its own production processes. Embracing CCU technologies and aligning with the EU's sustainability goals can position the lime industry as a frontrunner in fostering environmental stewardship and economic viability, although specific solutions for each application of lime must be researched.

The present scientific paper stems from comprehensive research work grounded in the hypothesis that achieving carbon neutrality throughout the life-cycle of lime-based materials is possible by combining process improvements and natural carbonation. An all-encompassing sustainability strategy is formulated, centred around the principles of circular economy, carbon direct avoidance, and CO_2 capture. Within this framework, a process-oriented parametric methodology is introduced to rigorously calculate the Life-cycle Inventory (LCI) of these strategies, reducing reliance on generic databases while ensuring transparent inventories for upscaling novel technologies [3]. Subsequently, an environmental and economic assessment is conducted of LBMRP production at an actual plant, providing a first detailed study of present business-as-usual scenarios [8]. Lastly, the environmental impact of current hydrated lime production in four European countries is quantified and the effects of transitioning to eco-efficient energy sources are assessed [11].

All prior scientific advancements, including those outlined in this paper, have been realized within the framework of the Horizon 2020-funded SUBLime MSCA ITN ETN project [25]. This initiative has successfully brought together Europe's top-tier universities and major lime-based materials manufacturers to collaboratively devise strategies for achieving net-zero emissions. As a result, we have had the opportunity to engage in in-depth discussions, assess and amalgamate insights from both academia and the private sector, and encapsulate the outcomes of this collaborative effort within this paper.

To advance the frontier of knowledge previous findings are integrated by intricately modelling and upscaling the inventory of a direct separation reactor with carbon capture technology employed in the production of hydrated lime. The combined impact of this technology and the transition to sustainable energy is evaluated in alignment with the European Union’s 2050 net-zero carbon goal, while encompassing the entire Cradle-to-Grave life-cycle of lime-based plaster. Furthermore, a life-cycle cost analysis, incorporating the cost of carbon taxes, is employed to assess the influence of CO_2 reduction during the manufacturing stage. Through a thorough Life-cycle Assessment (LCA), a decarbonization strategy at the emission source is applied, in conjunction with carbonation of the material during its use phase, demonstrating that a carbon-negative transformation over the entire Cradle-to-Grave scope is achievable. The paper also discusses the primary challenges and coordinated actions required across the manufacturing sector, energy sector, regulatory bodies, and the market to expedite a full decarbonization of the lime industry.

7.2 Methodology

7.2.1Definition of the Case Studies

This study aims to assess the environmental impact of lime-based plaster (LBP) used as an external coating on a masonry wall throughout its Cradle-to-Grave life-cycle, considering various manufacturing scenarios for hydrated lime. The functional unit (FU) is represented by the amount of LBP required to cover a 1 m^2 size wall for 50 years with a minimum thermal insulation of 0.01 m^2/kW. The system boundaries, shown in Figure 7.2, comprise the production of all LBP components (hydrated lime, diverse types of aggregates and additives), along with its use and end-of-life phase.

Regarding the different hydrated lime manufacturing scenarios, the study focuses on two key aspects, i.e., energy sources and kiln technology. Previous work already investigated electricity and various fuel sources employed for hydrated lime manufacturing in Germany in 2020 and 2050 (Appendix 11 and 16-FMES3)

[11]. Kiln technology is on a comparison between the widely-used PFRK [26] and the Direct Separator Reactor designed by LEILAC [17].

To analyze the impact of these combinations on the overall environmental footprint of LBP production, four distinct scenarios are selected and presented (Figure 7.3). Scenario 1 represents the 'Business-as-Usual', employing PFRK technology with a notable reliance on fossil fuels for both the fuel and electricity source. Scenario 2 advances decarbonization efforts by optimizing the fuel and electricity sources with a focus on eco-efficient energy sources, following a strategy of carbon direct avoidance. Scenario 3 introduces a change in kiln technology, transitioning from PFRK to a Direct Separator Reactor, while maintaining current energy sources. The main benefit here is the evaluation of the capture of unavoidable emissions. Finally, Scenario 4 holds the most promising configuration, combining eco-efficient energy sources with direct separation technology. This last scenario is subdivided in three categories: S4A uses the 2050 fuel sources for calcination energy, S4B leverages the DSR's hybrid mode with 2050 electricity and fuel sources sharing the energy demand equally, and S4C as the most radical one, relies solely on the 2050 electricity sources for kiln energy. Note that in the case of DSR, the assessment considers additional equipment for CO_2 capture (i.e., heat exchangers and compressor), although specific CCU or CCS applications are beyond the scope of this study.

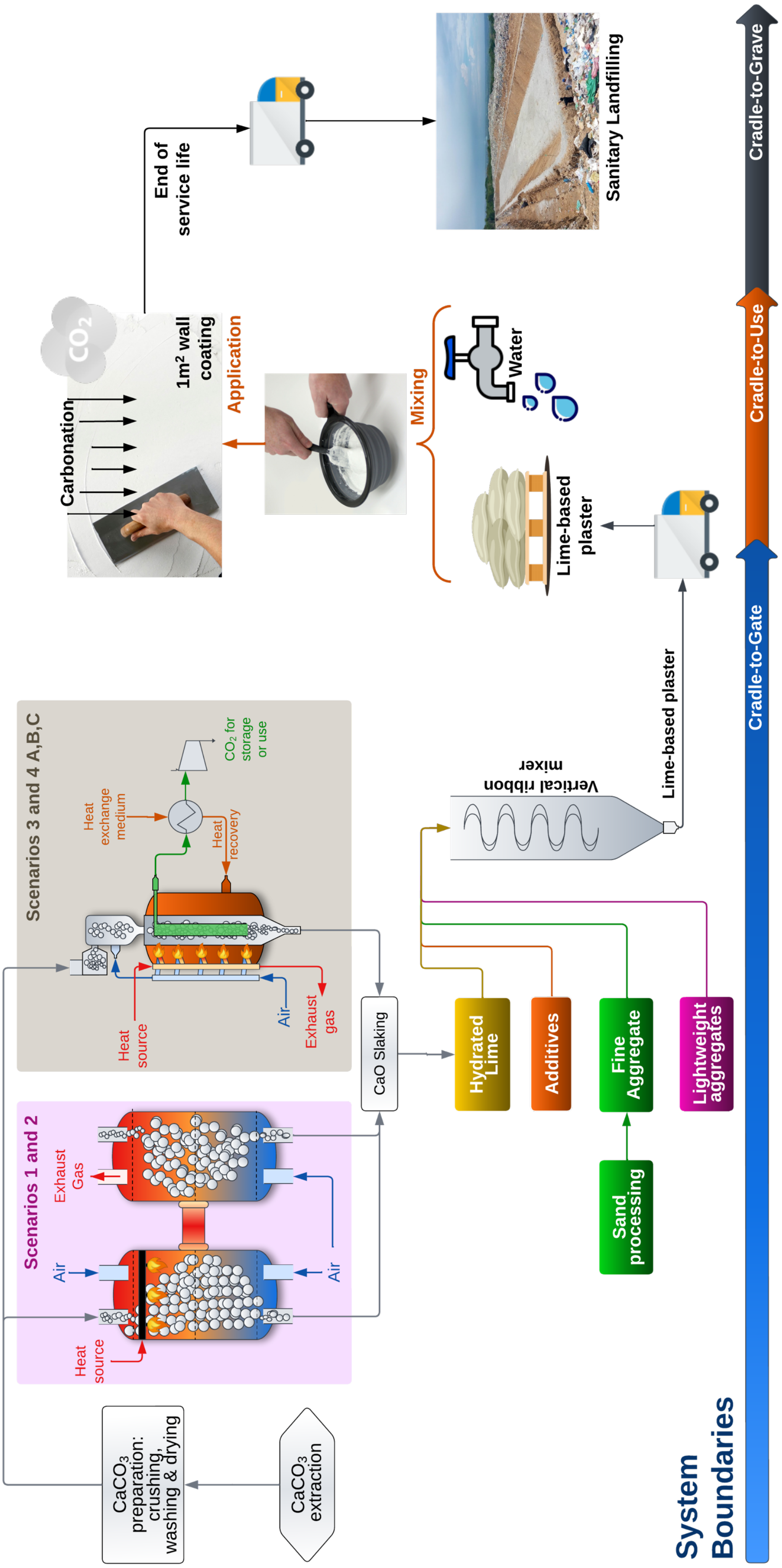


Figure 7.2. System boundaries for the study, with emphasis on various lime manufacturing scenarios during the Cradle-to-Gate phase

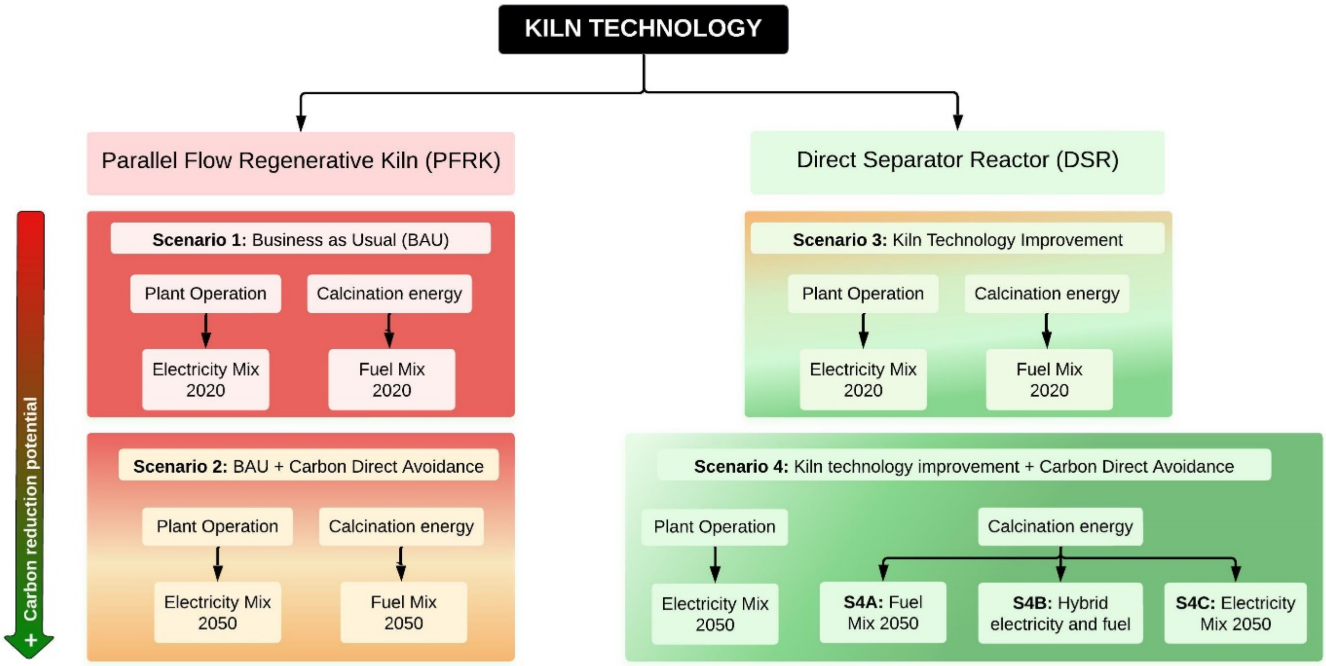


Figure 7.3. Definition of the scenario analysis applied to the Cradle-to-Gate hydrated lime manufacturing.

The environmental analysis of the lime-based plaster production is linked to a previously studied real plant, where aggregates and additives are transported to the site and dry mixed [5]. Power sources for the mixing plant align with the electricity sources used in each scenario. In every scenario it is considered that the plaster is transported to the construction site, mixed with water, and enters the use phase. During this phase, the LBP absorbs CO_2 from the environment (see Section 7.3.1), which is accounted for in the LCA. A predefined service life of 50 years is considered, followed by the traditional removal, transportation, and landfilling of the plaster. At this stage, the LCA and Life Cycle Costing (LCC) account only for transportation and landfilling.

7.2.1.1 Life-cycle inventory

The result of the environmental impact relies heavily on the quality of the life-cycle inventory (LCI). Therefore, a significant effort is put into calculating a rigorous inventory for the Cradle-to-Gate of the factory phase, with specific focus on the LCI modelling of the two kiln technologies used to produce hydrated lime. Given that there are no substantial reference LCIs for the case of a DSR kiln and current literature and generic databases do not address it, a process-oriented methodology developed by the authors is employed to calculate the inventory [3]. The same methodology provides a parametric framework that allows scaling up the DSR and carbon capture technologies (i.e., considering different production capacities), as well as the implementation of the scenario analysis (Fig. 7.2) in the LCI. For more detailed information, reference is made to [3].

7.2.1.2 Life-cycle Impact Assessment and Life-cycle Cost

The software OpenLCA with EcoInvent V3.6 was employed to run the environmental and economic calculations[27]. The impact assessment method is Impact 2002+, which addresses the relevant impact categories that are of importance for the mining industry, such as Resources, Climate Change, Human Health, and Ecosystem quality [28].

Contrarily to LCA, the LCC methodology is not standardized and therefore, no unified procedure exists for calculating life-cycle costs. To conduct such LCC, the starting point was the inventory of materials and energy considered for the environmental analysis (Section 7.3.1). The calculations are performed from a producer's perspective. For production costs the purchase price of materials, resources, and energy were considered [29]. Calculating the costs of carbon (price basis 2022), is based on the European Trading System, which considers the amount of CO_2 emitted during production of a binder in a lime-based mixture multiplied by the carbon price (90 €/t CO_2) [4]. More detailed information can be found in Section 7.3.1.

7.3 Results and Discussion

7.3.1 Life-cycle Inventory

7.3.1.1 Cradle-to-Gate of the Factory

A detailed overview LCI of a production plant for lime-based plaster, along with a specific recipe and datasets can be found in Appendix 18. In this section though, focus is on the hydrated lime production, where Figure 7.2 shows the unit operations included in the manufacturing process, highlighting the two lime kiln technologies considered in this study. A comparison of the key-parameters for both technologies, as well as the values taken for the analysis of the different scenarios is presented in Table 7.1.

The PFRK is a kiln technology that has been successfully employed in the lime industry for around 2 decades [26]. It is composed of two interconnected units represented by a pre-heater, burning and cooling zones (Figure 7.2). The burners are placed at the top of the first calcination unit, and the combustion gases move counter-directional to the limestone feed. The limestone will be decomposed in lime and carbon dioxide, at a temperature ranging from 900 to 1100°C. Through the cross-over channel, the combustions gases and the CO_2 from calcite decomposition are injected to the second unit, also moving counter-directional to the limestone introduced at the top, providing a very efficient calcination. The lime is received at the bottom of both units [16].

Table 7.1. Technical key parameters of PFRK and DST kiln technologies for scenario analysis

Key Parameter	**PFRK** [2], [16], [26]	**DSR** [16], [17], [30]–[32]
Energy efficiency (%)	80-90	< 80
Heat consumption (GJ/tCaO)	3,7-4,2	4,0 - 4,5
Electricity consumption (kWh/tCaO)	20-40	70-80
Fuel types	Solid, liquid, and gaseous, including biofuels	Solid, liquid, and gaseous, including biofuels and electricity
Production capacity (t CaO/d)	100-600	<200

The technical data employed for the analysis of the novel DSR design (so-called CALIX kiln) is less disseminated. It is worth mentioning that two partners of the SUBLime project, the Belgian lime producer Lhoist and the British lime company Tarmac, worked also in the development of this technology. As shown in Figure 7.2, this kiln consists of a pre-calciner, and an inner and an outer tubular body. In the outer tube the burners are located, that indirectly heat the inner tube, through which the hot gases calcine the limestone. The produced lime by the DSR has the same characteristics as the one produced in the conventional PFRK technology [31]. This system avoids mixing-up the process-CO_2 with the combustion gases, thus obtaining a high purity of CO_2 as a by-product that can be easily extracted from the reactor. This so-called process-CO_2 needs to be cooled-down and compressed to be handled for both CCS and/or CCU applications. Despite this major advantage compared to PFRK, the energy efficiency and capacity of DST systems is still lower (Table 7.1). An optimization of the energy efficiency can be achieved by effectively employing the residual energy of process-CO_2 while this DST system also enables the possibility to be electrified, and, as such, avoid the use of solid fossil fuels, as will be demanded by the EU in 2050 [33].

The LCI of each kiln technology, for producing 1 ton of CaO, is shown in Tables C4.2 and 3. In both cases the input is “Prepared $CaCO_3$” while the output is CaO, which is further-on slaked to produce hydrated lime (See Figure 7.2). As can be observed, the entire manufacturing process is considered in the impact assessment, but for sake of clarity a detailed view on the inventory modelling of the calcination process will be provided by considering the PFRK and DSR kilns and the sources of CO_2 emissions. In the case of the current PFRK technology (Table 7.2), the inventory is based on a previous work by the authors [3].

For the DSR, not only the energetic requirements to operate the reactor are included but also the extra operational units associated with carbon capture of the unavoidable process emissions (Table 7.3).

Table 7.2. Inventory of the calcination operation using PFRK kiln technology (based on [3])

			PROCESSED AMOUNT		COMMENTS
	OPERATION	**FLOW (mass/energy)**	**AMOUNT**	**UNIT**	
INPUT	**Calcination** (PFRK Technology)	$CaCO_3$ from “$CaCO_3$ preparation”	1.79	t	[10]
		Thermal Energy	3900	MJ	Table 7.1
		Electricity consumption	30	kW	Table 7.1
OUTPUT	**OPERATION**	**FLOW (mass/energy)**	**AMOUNT**	**UNIT**	
	CaO Production (PFRK Technology)	CaO (determining product)	1	t	[10]
		CO_2 ($CaCO_3$ decarbonation)	0.79	t	Unavoidable process emission (0.79 $tCO_2/tCaO$)
		CO_2 (fuel combustion)	Depending on the fuel source		
		CO_2 (electricity consumption)	Depending on the fuel source		

Table 7.3. Inventory of combined calcination and CO_2 recovery for lime production with DSR.

			PROCESSED AMOUNT		COMMENTS
	OPERATION	**FLOW (mass/energy)**	**AMOUNT**	**UNIT**	
INPUT	**Calcination** (DSR Technology)	$CaCO_3$ from “$CaCO_3$ preparation”	1.79	t	[10]
		Thermal Energy	3819.00	MJ	4250 MJ -431 MJ credit heat exchanger
		Electricity Consumption	75.00	kW	Table 7.1
	Carbon capture system (Heat exchanger)	Electricity Consumption	1.75	kW	Total electricity consumption for two heat exchangers
	(Compressor)	Electricity Consumption	1.03	kWh	[3]
OUTPUTS	**OPERATION**	**FLOW**	**AMOUNT**	**UNIT**	
	CaO Production (DSR coupled with carbon capture system Technology)	CaO (determining product)	1	t	[10]
		CO_2 (as co-product)	0.79	t	Process-emission (Economic allocation, Mass allocation)
		CO_2 (fuel combustion)	Depending on the fuel source		
		CO_2 (electricity consumption)	Depending on the electricity source		

Although there are several configurations for post-combustion carbon sequestration, in this study it was tried to remain as close as possible to the specific design of the DSR [30]. To attain an accurate calculation of the mass and energy balances, the designs of the devices were complemented by a simulation of the chemical process using Aspen HYSYS V12.1 software. The produced CO_2, resulting from calcite decomposition, is assumed to leave the reactor at 1000°C and 385 kPa. The final thermodynamical condition of this CO_2 depends on the transportation means leading to a possible modification of the equipment. For instance, trucks require the CO_2 in liquid form at 22 bar and -35°C while when transported through pipelines, usually the CO_2 will be in supercritical state (above 30°C and 22 bar) [34]–[37]. This latter option has been claimed to be the most economically and environmentally viable alternative, due to the possible larger amounts that can be transported and is therefore considered here for modelling of the carbon capture system.

For modelling the CO_2 treatment, the following strategy was applied. First, theoretical calculations of heat exchanges and minimum flow requirements were employed to achieve the final thermodynamic conditions desired for CO_2 transportation. Second, heat exchangers and compressors were selected from the software while the calculated data were implemented being the starting point for the simulation to determine the operational parameters and technical requirements for the equipment. Finally, different configurations were evaluated (such as size of devices, type of heat exchangers, intermediate temperatures) where the flow sheet of the simulation was adapted to select the configuration that minimizes the energy requirement of the process. Thus, with this not only the demand of energy but also the energy credits were accurately calculated.

The DSR inventory is implemented in Table 7.3 and the design parameters of the carbon capture system can be found in the Appendix 19. It may be relevant to mention that although this LCI is designed for transportation of CO_2 through pipelines, extra unit processes can easily be added to simulate other configurations without significant effort.

7.3.1.2 Use Phase, End-of-Life and Life-cycle Cost Inventory

A plaster density of 1.1 kg/L and a durability of 50 years were assumed according to declarations from producers [38]–[41]. During service life, CO_2 initiates a carbonation reaction with portlandite to generate calcium carbonate and is responsible for the plaster hardening (Reaction 7.1).

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$ **Reaction 7.1**

The diffusion of CO_2 through the plaster (i.e., CO_2 sequestration) can be described by a simplified equation representing a diffusion-like process (Equation 7.1). Equation 7.2 is employed to calculate the kg of CO_2 sequestrated per functional unit at a given time.

$$x = k\sqrt{t}$$ **Equation 7.1**

$$SC = 0.594 * FCH * \frac{x}{X_{total}}$$ **Equation 7.2**

where x (mm) is the carbonated thickness of the plaster at a given time t, k (mm/day$^{0.5}$) is the diffusion coefficient of CO$_2$, SC (kg CO$_2$/m^2) is the mass of CO$_2$ sequestrated per area of coated wall, 0.594 is a conversion factor (molecular weight ratio CO$_2$/Ca(OH)$_2$), FCH (kg Ca(OH)$_2$/m^2) is the amount of hydrated lime per area of coated wall, X_{total} (mm) is the total thickness of the plaster. SC is calculated until the time of maximum carbonation (Eq. 7.1) is reached.

The adopted parameters as well as calculations performed are shown in Table 7.4. The coefficient k is an average value reported by [42]–[44].

Table 7.4. Parameters considered for the carbonation of the plasters during the use phase.

Plaster	kg plaster/ m^2	FCH (kg Ca(OH)$_2$/m^2)	X_{total} (mm)	k (mm/day$^{0.5}$)	Full carbonation time (days)	SC (kg CO$_2$/m^2)
HLP	9.66	2.41	8.70	1.00	76	1.43

A service life of 50 years is assumed for the plaster. At the end of the service life, complete carbonation of the portlandite in the plaster is assumed (0.59 kg CO$_2$/kg Ca(OH)$_2$), based on the thinnest of the layer (<9 mm, Table 7.4) and the service life (50 years) according to the diffusion model (Equation 7.2) [45]. After this time, the old (discarded) plaster is transported over 100 km to its final disposal site (see Appendix 18). The EcoInvent V3.6 dataset "treatment of waste concrete, inert material landfill | waste concrete | APOS, S" was employed to model the landfilling of the plaster at the end of life. Specific data regarding costs at different stages of the life cycle are provided in Appendix 20.

7.3.2Environmental Impact Assessment

In this section first the Cradle-to-Gate scenario is analysed (Figure 7.2) to evaluate the effect of various energy sources and the kiln technologies regarding their environmental impact of the production of hydrated lime and lime-based plaster. Secondly, the expansion of the boundaries to the Cradle-to-Grave are included. Finally, the business-as-usual scenario is thoroughly compared with the enhanced sustainable scenarios, not only in terms of Climate Change but also in the damage areas of Human Health, Resources and Ecosystem quality.

7.3.2.1 Manufacturing Scenario

A carbon direct avoidance strategy imposes a switch to low-CO$_2$-emission energy sources used for electricity production and as fuels to provide thermal energy. In the calcination operation, fuels can contribute between 35-40% to the Global Warming Potential, while also causing potential damage to other environmental areas, depending on the selected energy source. The electricity consumption in the business-as-usual production of hydrated lime, doesn't significantly influence the GWP [11]. However, in a hybrid or even full electrification scenario of the lime kiln, a different situation might be expected.

Incorporating the effect of kiln technology in the environmental impact leads to a significant increase of the system's complexity. Therefore, at first it is required to have a clear understanding of the contribution electricity and fuel production have on the environmental impact for current and future scenarios. The environmental impact per MJ of energy in the four selected areas of damage is shown in Figures 7.4 and 5 for electricity and fuel sources, respectively.

Regarding electricity production in 2050 it is noticeable that all endpoint indicators project a significantly reduced impact compared to the current scenario, by 88% for Resources, 48% for Ecosystem Quality, 38% for Human Health and 84% for Climate Change, (Figure 7.4a-d). In E2020 the combustion of Natural Gas and Coal dominates the Resources (70%) and Climate Change (85%) indicators. Meanwhile, although the proportion of biofuels is only 8%, significant impacts in the areas of Ecosystem Quality (65%) and Human Health (41%) are recorded, which is mainly related to the treatment of biowaste and sewage sludge to be combusted in a cogeneration unit. This type of electricity production is still highly efficient and showing low CO_2 emissions as provided in Figure 7.4d. With respect to E2050, it is highlighted that the production of electricity through Wind and Solar technologies (86% of the electricity sources) contributes only moderately to all categories. The highest contribution is found by hydropower, being 4% in the electricity sources and accounting for 50% of the $CO_{2\ eq}$. [45].

The production of thermal energy is dominated by coal combustion with all impact indicators ranging between 50 to 85% of the endpoint categories (Figure 7.5). Human Health and Ecosystem quality are specifically affected since during coal combustion significant amounts of CO_x, SO_x, and NO_x are emitted, affecting the air quality and being the origin of several illnesses [46]. This is one of the main reasons why coal plants are being phased-out in Europe. In the 2050 scenario, significant improvements are observed for the key environmental indicators, as the proportion of coal is completely replaced by an enhanced share of natural gas and biomass. With Human Health F2020 and F2050 remaining in the same order (Fig. 7.5c), the following reductions per MJ of energy are recorded: 20% for Resources, 10% for Ecosystem Quality and 60% for Climate Change (Fig 7.5a, 7.5b, 7.5d). It is assumed that Natural Gas will be the main fossil fuel to back up renewable energy sources on the path to decarbonization, as it generates 44% less CO_2 compared to coal [47].

Retrospectively, when comparing the environmental impacts per MJ of energy, the Figures 7.4 and 7.5 indicate that electrification of the lime kiln would lead to a reduced environmental impact compared to fossil fuel sources in both, current (2020) and potential future scenarios (2050). If the thermal energy in the kiln would be generated by the 2050 electricity sources instead of the 2050 fuel sources, around 60% CO_2 per MJ of energy will be saved (Fig. 7.4d and 7.5d).

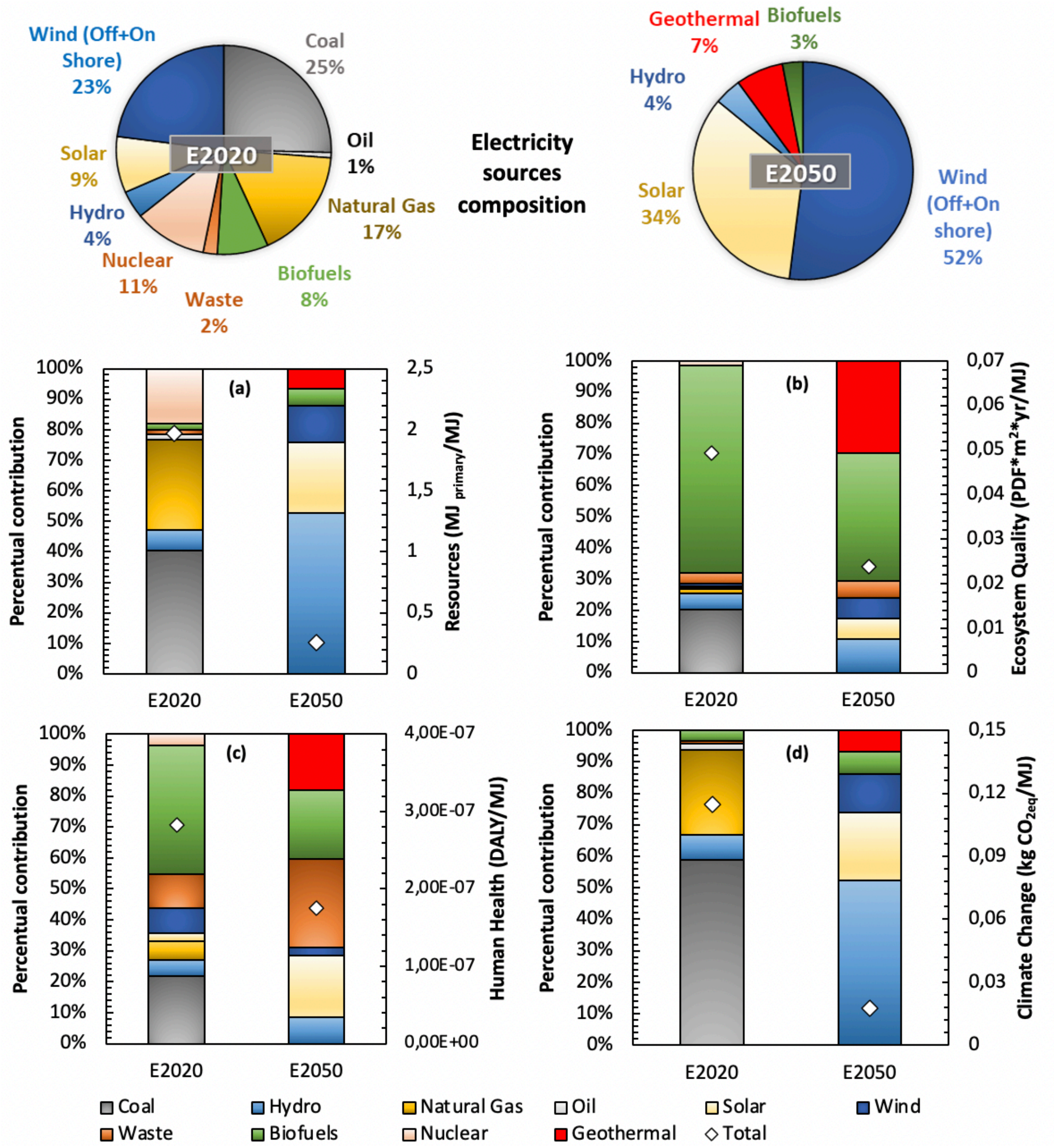


Figure 7.4. Electricity sources composition used to model current (2020) and future scenarios (2050) according to[11]. Figures a to d show the endpoint indicators with percentual contribution (left axis) and total impact (right axis) of components to produce 1 MJ. *DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction*

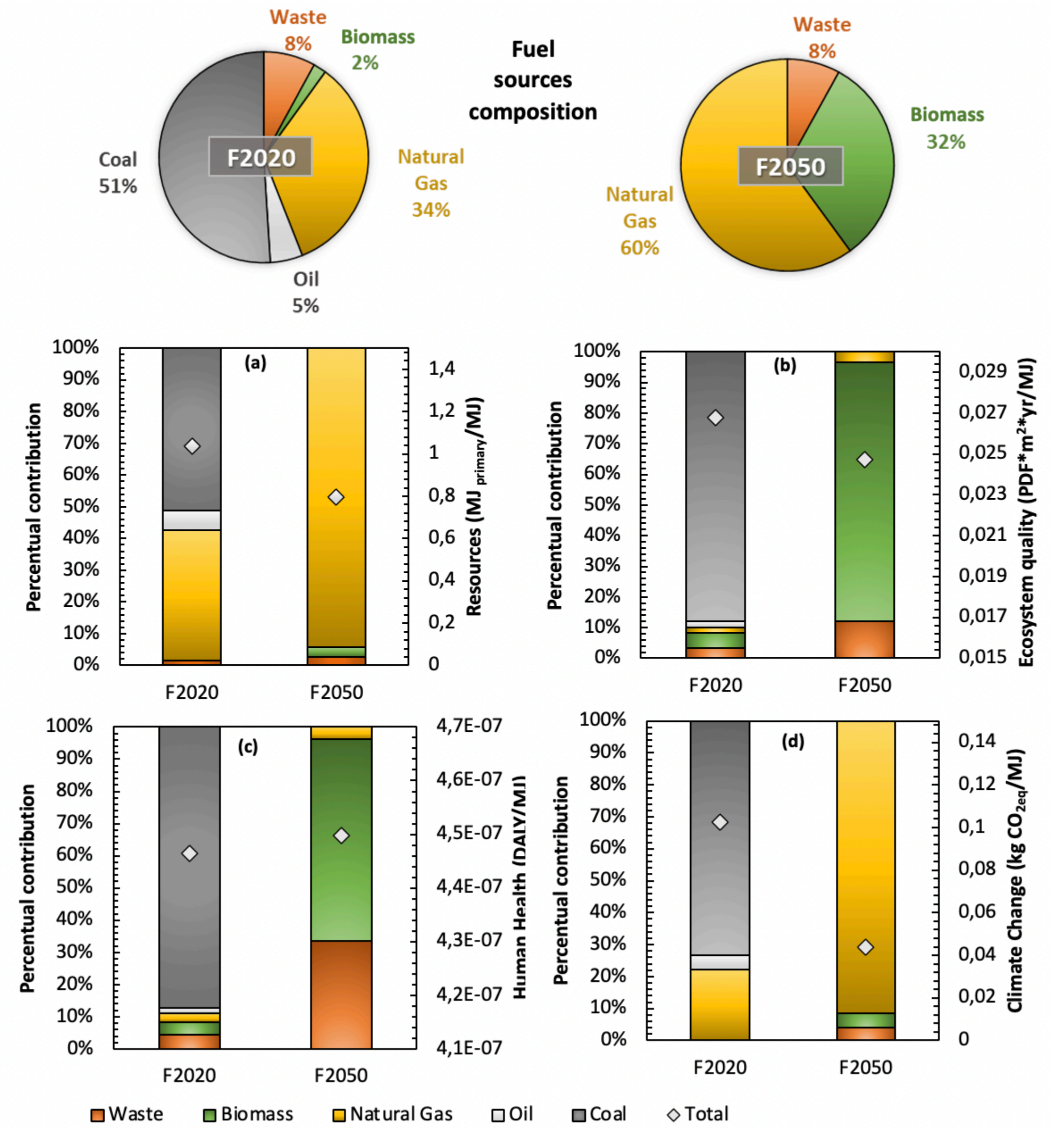


Figure 7.5. Fuel sources composition used to model current (2020) and future scenarios (2050) according to [11]. Figures a to d show the endpoint indicators and percentual contribution of each fuel source to produce 1 MJ of energy. DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction

Figure 7.6 summarizes the results of the scenario analysis (Fig. 7.3), showing progressive integration of decarbonization strategies at the plant scale. The impacts show the contribution of fuels and electricity in the kiln, as well as ´Others´ including impacts of other unit operations besides the kiln. On the right axis of Figures 7.6a-c, the Ratio of each scenario (S_i, i=2, 3…) compared to the \`business as usual\` reference case (S1) is shown. Figure 7.6d shows the CO_2 emissions associated to calcite decomposition, fuel combustion, electricity and carbon captured, which are depicted as negative emissions for scenarios S3 and S4. The total climate change indicator is tracked on the right axis.

As previously anticipated, a change in the fuel sources from F2020 to F2050, leads to a moderated reduction of the impact indicators of S2 compared to the business-as-usual scenario S1. In this case, only the sources of the thermal energy were modified, while keeping the same technology (PFRK kiln). The main responsible of these improvements is the elimination of coal as a fuel source (Figure 7.3). As a decarbonization strategy, the use of eco-efficient fuel sources is a first step towards reduced CO_2 emissions (Figure 7.5d), leading to 17% savings (S2 vs S1). The change in kiln technology for the impact categories Resources, Ecosystem Quality and Human Health does not produce any improvements, and the slight reduction in the total energy consumption shown in Table 7.2 and 3 (3819 and 3900 MJ/t CaO, for DSR vs. PFRK) is compensated by the increased requirement of electricity to operate the kiln and the carbon capture system. With respect to the latter carbon capture system, it is highlighted that the simplicity of the post-combustion treatment in the DSR kiln allows to obtain pure CO_2 without a significant increase of the energy consumption, thus balancing the environmental impact and costs. Furthermore, CO_2 captured by the DSR kiln leads to 0.56 tCO_2/t $Ca(OH)_2$, reducing the total emissions by 65% (S1 Vs. S3, Fig. 7.6d).

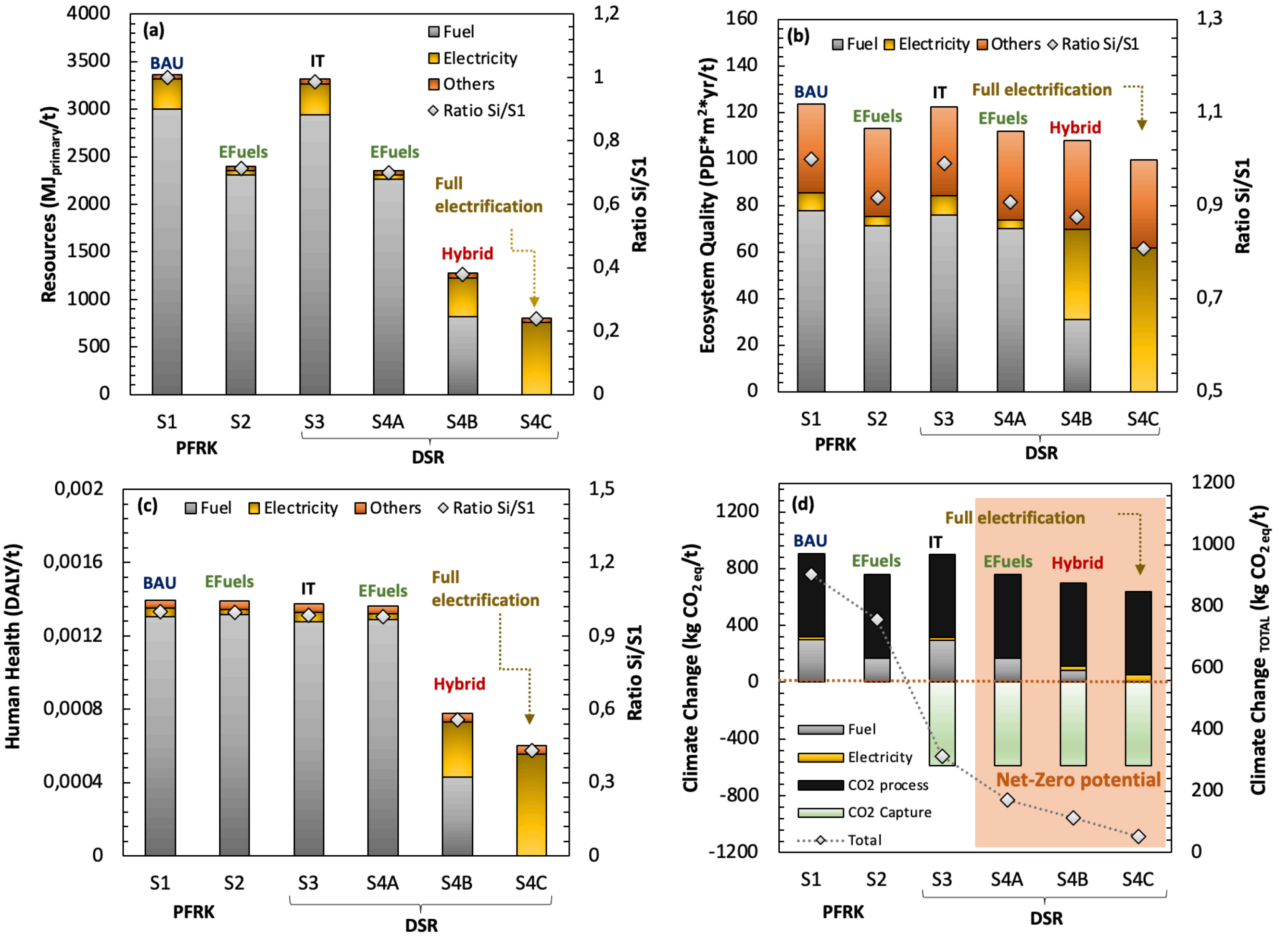


Figure 7.6. Endpoint impact indicators of the scenario analysis for the Cradle-to-Gate system boundaries to produce 1 ton of hydrated lime. Figures a-c show the ratio of each scenario with respect to the reference S1. Figure d shows the total effect on the climate change indicator according to each technology and energy sources. *BAU: business-as-usual, E-Fuels: Eco-efficient fuels, IT: Improved kiln technology, Hybrid: Operation by e-fuels and electricity. DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction*

The Series S4 combines a change of the PFRK kiln by DSR technology, including the replacement of fuels by electricity, along with the use of eco-efficient energy sources to produce thermal energy and electricity. The use of sustainable energy sources to produce thermal energy in the DSR kiln (S4A) has significant influence in 3 out of 4 endpoint categories, leading to reductions of 10%, 30%, and 80% for Ecosystem Quality, Resources and Climate Change categories compared to S1. Around 132 t CO_2/t $Ca(OH)_2$ are saved in S4A versus S3, indicating the potential synergy of the improved kiln technology and the reduction of solid fossil fuels in the energy sources. A progressive reduction of the impacts is observed as the replacement of fuels for electricity goes from 0% (S4A) to 100% (S4C) (see Fig. 7.4 and Fig. 7.5). The scenario with the highest decarbonization potential is S4C, in which around 52 kgCO_2/t$Ca(OH)_2$ are emitted, leading to reduced emissions by 94% compared to the reference scenario S1, making it the closest one to carbon neutrality.

The in-depth quantitative assessment of the present scenarios shows the positive impact the kiln equipped with carbon capture technologies and carbon direct avoidance. The assessment turned out to be an essential instrument for decarbonizing the manufacturing process of hydrated lime, which is the pivotal product not only for construction materials, but also for various secondary applications, i.e., from steel production to waste water treatment[2]. Moreover, as these results are derived from well-defined and scientifically rigorous inventory simulations, they may serve as a robust benchmark against which numerous processes related European mitigation projects on cement and lime can be compared. The following section explores the manufacturing of lime-based plasters and broadens the system boundaries to incorporate considerations for carbonation during the use phase and its final disposition at the end of the product's life-cycle.

7.3.2.2 Cradle-to-Grave

With section 7.3.2.1 highlighting possible improvements in the manufacturing stage of hydrated lime production across various technologies and energy scenarios, assessing any environmental enhancement should comply with the context of an appropriate FU. This assessment should encompass not only all components of a plaster as such, but also its performance during the use and end-of-life phases (depicted in Figure 7.2). Figure 7.7 shows the environmental impact associated with a 1 m^2 wall covered with a lime-based plaster specified for a 50-year period. The dry-mix manufacturing facility has been previously studied by the authors[5]. It produces both the hydrated lime and aggregates and receives the remaining components through transportation from other locations (Appendix A18). The various energy scenarios represent the production plant and are also applied to the sand production process. It's important to note that in Figure 7.7d, the CO_2 sequestration during the use phase is accounted for as a negative emission. Finally, the end-of-life phase encompasses the transportation of the material to the landfill as well as the environmental impact of the landfill itself.

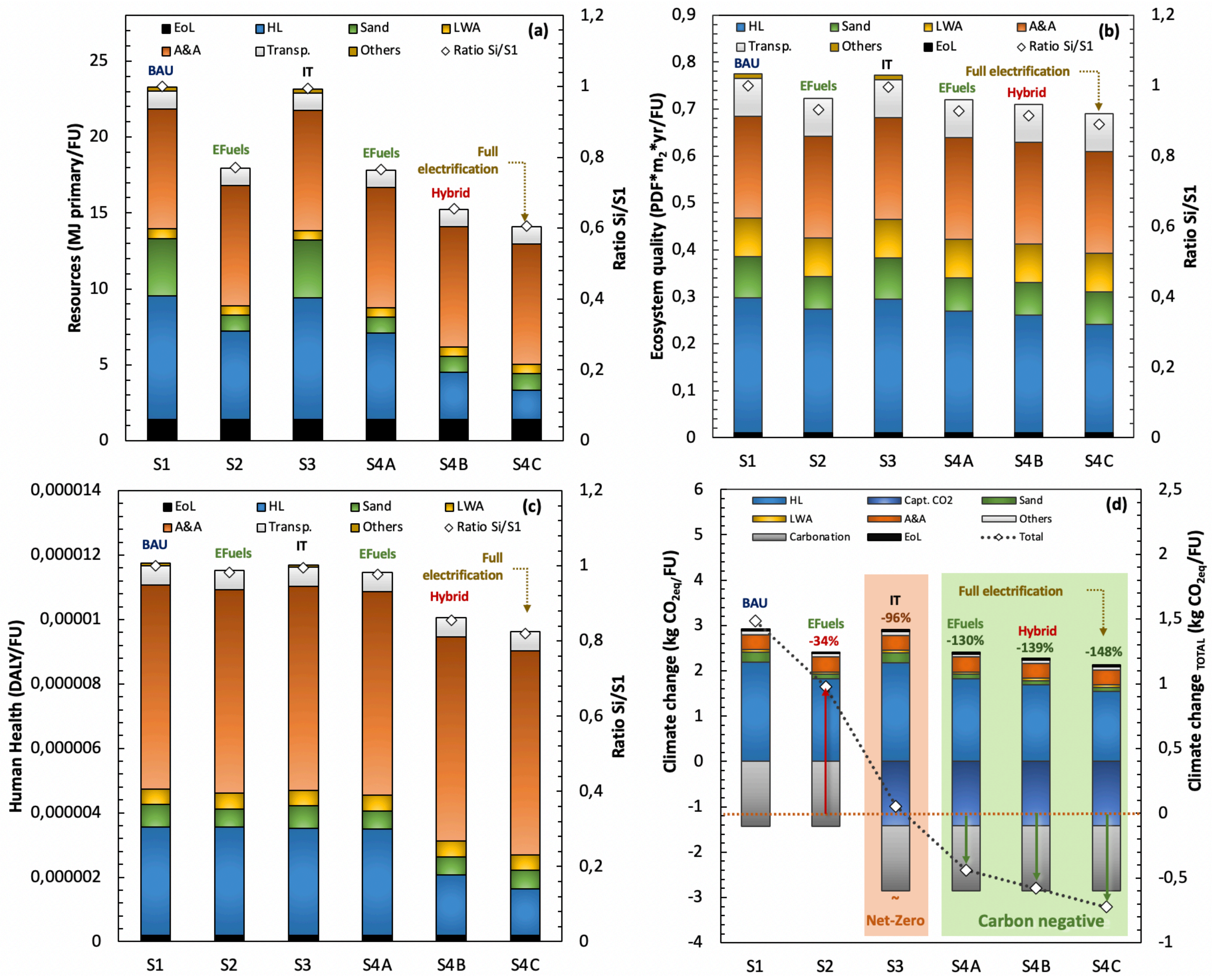


Figure 7.7. Endpoint impact indicators of the scenario analysis for the Cradle-to-Grave system boundaries (FU is 1 m² wall coating). Figures a-c show the ratio of each scenario with respect to the reference S1. Figure d shows the total effect on the climate change indicator according to each technology and energy source. BAU: business-as-usual, E-Fuels: Ecoefficient fuels, IT: Improved kiln technology, Hybrid: Operation by e-fuels and electricity. HL: Hydrated lime, LWA: Lightweight aggregate, A&A: Additives and Aids, EoL: End of Life. DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction

The additives have a significant impact on the Resources Category (Fig. 7.7a), with the dispersion and water retention agents, based on polymers, being particularly important. Although they are minor components in the mix composition, in S1 their share is comparable to the production of hydrated lime (35%) and in the scenario S4C, it dominates the indicator by around 60%. The production of sand in S1 and S3 also has a significant share (around 15%), mostly associated with the high energy consumption needed for sand preparation as observed in [5], and the high proportion used in the mix (around 75%) (Appendix A18). In the scenarios with eco-efficient energy sources, the specific contribution of sand to the indicator is reduced by two-thirds (S2 vs. S1 and S4A, B and C vs S3). As observed previously in

Figure 7.4a, the elimination of coal as an energy source leads to a substantial reduction of the indicator. Due to the fuel consumed to transport the materials to the plant, this operation can contribute between 5-7% depending on the scenario. The end-of-life contribution is mostly related to the impact of transporting the waste plaster to the landfill facilities rather than to the impact of the landfilling itself. The progressive electrification of the manufacturing process from Cradle-to-Gate with sustainable energy sources (S4A, B and C) leads to a 25-40% reduction compared to S1.

In the Ecosystem Quality (Fig. 7.7b), moderated decrements are observed when introducing sustainable energy sources and a change of kiln technology, as also priorly remarked in the analysis of hydrated lime production (Fig. 7.6b). The additives production emerged once again as significant contributor (between 20-30%). An analysis of the inventory results shows that during the production of the polymers used as water retention agent, aluminium, zinc, and copper as well as nitrogen and sulphur oxides are released to the soil and the air, contributing to terrestrial acidification and ecotoxicity. The effect of sustainable energy sources on hydrated lime and sand production is rather limited and no significant reductions on these processes are achieved through this strategy. The contribution of the artificial lightweight aggregate production is in the same order of magnitude as the sand, where the release of NO_x and SO_x during the production of the polymer are the main responsible for its high impact (12.64 $PDF*m^2*y/kg$). Because of the fuel combustion by trucks and their exhaust gases freely released to the air, transportation took a significant share in Ecosystem quality (9-11%) in all indicators. The End-of-Life contribution is rather negligible.

In the case of Human Health category (Fig. 7.7c), the indicator is absolutely dominated by the production of water retention agent, synthetic dispersion agent, and air entrainer (in descending degree of contribution) used as additives, with a share ranging between 60-75% of the total impact [5]. The main reason behind this, is the release during their production of aromatic compounds, arsenic, and dioxin compounds, with carcinogenic properties and high impact intensity (between 3.5E4-1.5E9 kg $C_2H_3Cl_{eq}$/kg). It is essential to emphasize that achieving sustainable development involves addressing not only climate change but also other factors affecting the quality of life for populations and responsible resource use. The findings in Fig. 7.7a-c show that it is of paramount importance to focus research not only on binders' production but also on the development of eco-friendlier additives with a lower environmental impact. The electrification of the lime kiln leads to around 20% reduction (S4C vs S1), as previously discussed in Section 7.3.2.1.

In the Climate Change endpoint category (Fig. 7.7d), the business-as-usual manufacturing scenario (S1) leads to the release of around 3 kg $CO_{2\ eq}$/FU, being for around 85% dominated by hydrated lime production, because of the process emissions and fuel combustion (Fig. 7.4d). Minor contributors are the additives and sand production (together amounting to around 14%). During the use phase, the lime-based plaster absorbs 0.59 kg CO_2/kg $Ca(OH)_2$ in a process known as carbonation, particularly, 1.43 kg CO_2/FU are sequestrated from the environment (Table 7.4). Consequently, during the use phase, around 50%

reduction in the climate change indicator is obtained. Yet relying on carbonation to accomplish the goal of climate neutrality is far from realistic, as the results show. Scenario S2, which includes the use of eco-efficient energy sources displays around 34% CO_2 reduction potential compared to S1. The analysis of Figure 7.5d explains these results in terms of a lower CO_2 intensity of natural gas compared to coal.

The transition in kiln technology, shifting from PFRK to DSR (S1 vs. S3), represents a significant and immediate reduction in CO_2 emissions over the entire product's life-cycle. In this case, not only the process emissions are captured (1.41 kg CO_2/FU), but also the sequestered CO_2 is accounted for. Together, they result in a remarkable 96% reduction in Climate Change, when compared to the reference S1. This quantitatively demonstrates that altering from the traditional PFRK to an innovative DSR system with carbon capture technology is the most effective approach to achieving carbon neutrality in lime-based construction materials. Moreover, it may be worth noting that implementing this change does not necessarily require the installation of an entirely new kiln, but rather can existing ones be retrofitted with a relatively low investment [16]. Several European projects are already exploring this alternative, and their results support this direction (Appendix A17). The carbon capture system designed at the plant scale in this paper (Appendix A19) offers a viable solution for managing CO_2 emissions. Both the inventory (Table 7.3) and impact assessment (Fig. 7.7) reveal that the additional devices integrated into the DSR kiln have a negligible environmental impact compared to the overall benefits they generate.

Figure 7.2 illustrates a series of scenarios, with S4 emerging as the most promising path to achieve carbon neutrality and potentially carbon negativity throughout a plaster's life cycle. However, to reach this goal, it is crucial to enhance the manufacturing process since relying solely on plaster carbonation will not be sufficient to offset Cradle-to-Gate emissions. While carbon capture technologies (S3) are highly effective, a shift in kiln technology alone will not suffice. Instead, an integrated strategy involving improved kiln technology powered by low CO_2 energy sources is essential to reach climate neutrality. Scenario S4A represents the initial step in this integration, demonstrating the feasibility of achieving carbon negativity (-130% compared to S1), assuming that all process CO_2 can be captured and stored/used (See challenges and opportunities in Section 7.3.4). This is accomplished by predominantly employing a mixture of natural gas and biomass for thermal energy, while eliminating coal usage. This scenario aligns with the European Union's pursuit towards greener energy sources, making a probable scenario to take place in the future [33]. Carbon negativity can be further extended when the kiln operates in hybrid mode (S4B, -139%) and/or complete electrification (S4C, -148%). Although sustainable electricity sources are being explored for various applications, their widespread adoption in massive construction materials production may face challenges as well, which will be discussed in Section 7.3.4.

7.3.3 Economic Impact Assessment

Figure 7.8 shows the results of the total costs (€/FU) for each Cradle-to-Grave scenario provided in Figure 7.2. Furthermore, on the left axis, all items representing the direct production cost associated to the raw material purchase, transportation, plant operation (electricity consumption) as well as maintenance action, landfilling and the externalities (carbon pricing) have been explicitly considered for a better analysis of their relative contribution. The main variable between the scenarios is the cost of CO_2 emissions (i.e., carbon tax).

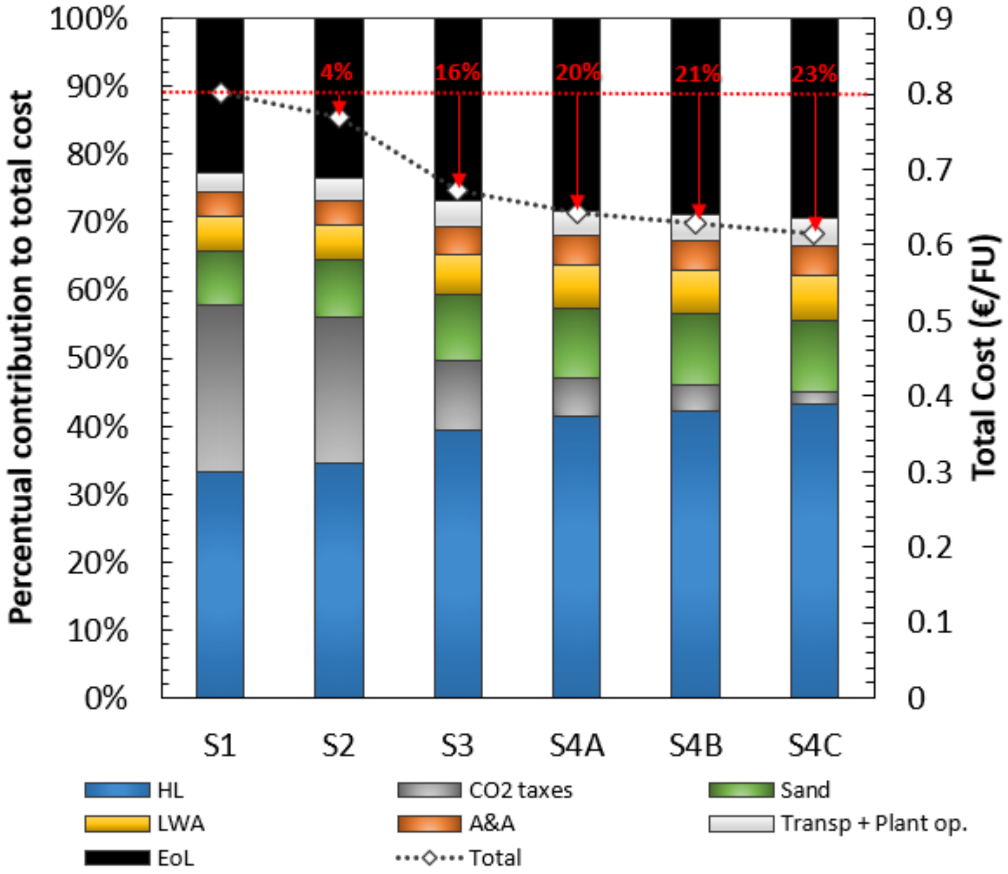


Figure 7.8. Life-cycle cost assessment results from Cradle-to-Grave of the decarbonization strategies, considering the effect of carbon taxes. HL: Hydrated lime, LWA: Lightweight aggregate, A&A: Additives and aids, EoL: End of Life.

In the business-as-usual scenario, around 58% of the cost is assigned to hydrated lime, including its production from Cradle-to-Gate (32%) and the cost of CO_2 emission as carbon taxes (26%) (S1, Fig. 7.8). The cost of emissions, within the current European Trading System (90 €/t CO_2) has almost the same influence as the production costs including raw materials, fuels, and energy. Therefore, it makes sense not only from an environmental point of view, but also from an economic one, to reduce emissions at the manufacturing stage. Several decarbonization strategies have been discussed in Section 6.3.2. The extent to which a switch to sustainable energy sources can be accomplished, depends on the region, as some countries might not be able to phase out coal as easily as others. However, investing in retrofitting current kilns or installation of DSR kilns, can be directly controlled by the companies and has demonstrated to be one of the most effective strategies to mitigate process emissions (0.56 $tCO_2/tCa(OH)_2$, Figure 7.6d). In scenario S3, the specific contribution of carbon taxes is around 21%, leading to a 16% reduction in total costs, because of 25% CO_2 reduction in hydrated lime manufacturing (compared to S1, Fig. 7.7d). A further reduction of emissions by electrifying the DSR leads also to a 23% cost reduction compared to the reference scenario.

In the business-as-usual scenario, approximately 58% of the cost is attributed to hydrated lime, encompassing its Cradle-to-Gate production (32%) and the cost of carbon emissions in the form of carbon taxes (26%) (S1, Fig. 7.8). Notably, the cost of emissions, within the current European Trading System (at 90 €/t CO_2), yields nearly the same influence as production expenses, including raw materials, fuels, and energy. Consequently, reducing emissions at the manufacturing stage not only aligns with environmental goals but also makes sound economic sense.

As previously discussed, (Section 7.3.2), the feasibility of various decarbonization strategies is regional dependent, as some countries may face more challenges in phasing out coal as an energy source. Nevertheless, investing in retrofitting existing kilns or installing DSR kilns offers a direct and highly effective means for mitigating process emissions (0.56 $tCO_2/tCa(OH)_2$, Figure 7.6d). In scenario S3, carbon taxes contribute around 21% to the costs, resulting in a 16% reduction in total cost, due to a 25% reduction of CO_2 emissions associated with hydrated lime manufacturing (compared to S1, Fig. 7.7d). A further reduction of the emissions through DSR electrification results in a 23% cost reduction compared to the reference scenario (See Figure 7.4, S4C scenario).

Another critical aspect regarding taxes on CO_2 emissions is that they often fail to account for certain products' capacity to act as carbon sinks during their use phase or at the end of their life cycle. Lime-based materials, for example, are capable to absorb CO_2 during their use, forming $CaCO_3$ [48], which is essential for fulfilling their intended function. A more equitable taxation system should distinguish between materials that sequester carbon and those that do not. This differentiation is crucial to ensure fair competition among sectors. The specific case of lime-based materials will be explored in more detail in Section 7.3.4.

7.3.4 Main Challenges and Actions Required Towards a Fast Decarbonization of the Lime Industry

The journey towards decarbonizing the lime sector is not solely the responsibility of the sector itself but is rather a multifaceted interplay involving various stakeholders with distinct decision-making roles and responsibilities at regulatory, market, and economic-political levels (see Fig. 7.9).

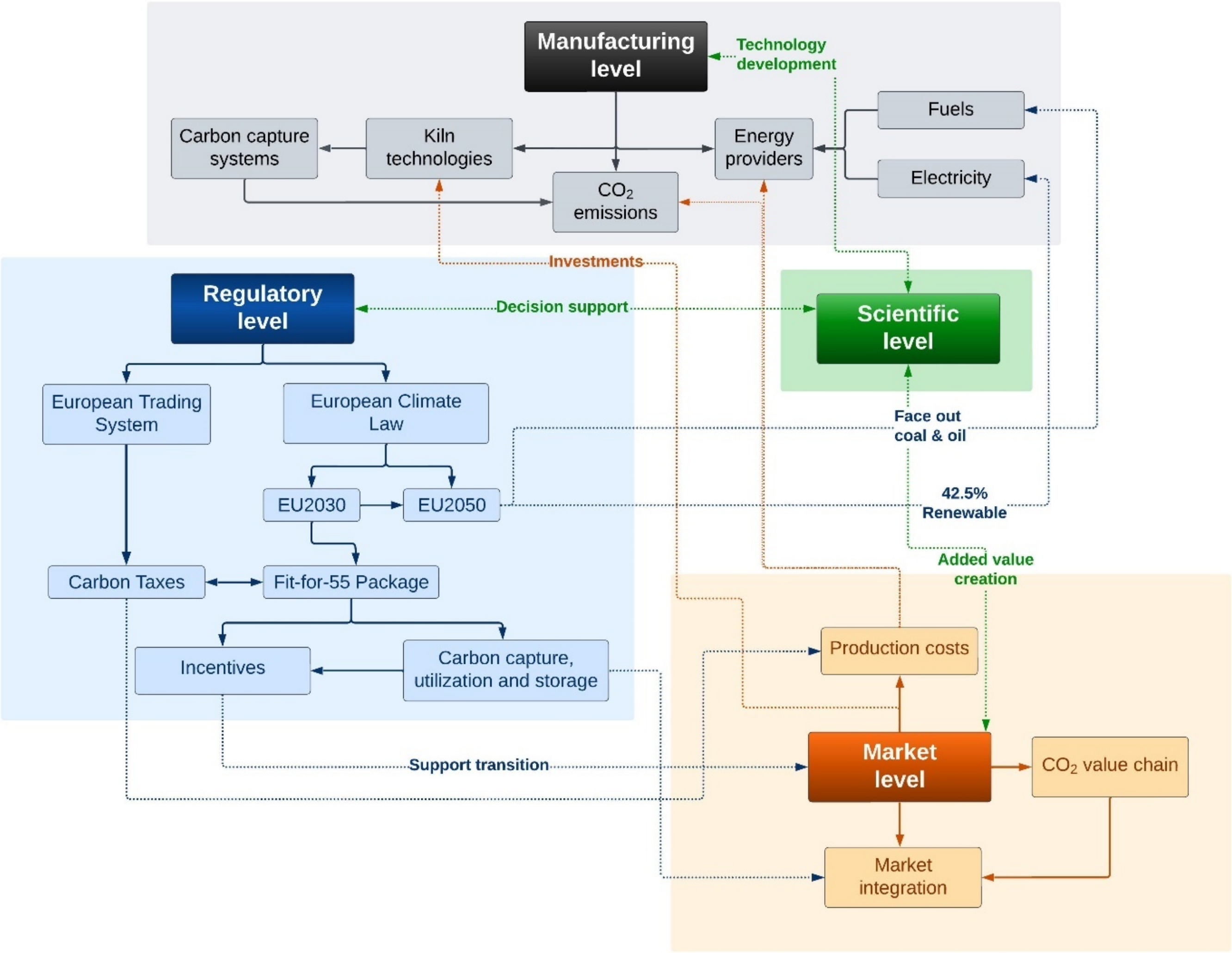


Figure 7.9. Scheme showing the interaction of all relevant stakeholders required to achieve a fast decarbonization of the lime and cement industry.

7.3.4.1 Manufacturing Level

At the manufacturing level, producers hold a critical decision-making lever: the choice of technology. A switch or retrofit from a PFRK to DSR kiln technology can result in the avoidance of 0.56 tons of CO_2 per ton of $Ca(OH)_2$ produced (S3, Fig. 7.6d), equating to a reduction of 65% in emissions, as previously mentioned. Remarkably, it may be worth noting that retrofitting kilns is often a straightforward process with low investment and adjustment requirements [49]. This shift also leads to a 16% reduction in production costs because of a reduced amount of carbon taxes paid (S3, Fig. 7.8). As emphasized in earlier sections, CO_2 emissions not only represent a global challenge but also provide a business opportunity. The pure CO_2 generated through the DSR process holds potential for use as a raw material in various industrial processes, transforming it from an emission into a valuable by-product [50].

However, realizing this potential requires legislative support and market development, as we will explore later in Sections 7.3.4.2 and 7.3.4.3.

Another crucial aspect at the manufacturing level pertains to the energy sources utilized in the kiln [11]. The availability of local resources, logistical considerations (e.g., gas pipeline infrastructure), and, fundamentally, the cost of energy itself all play pivotal roles. Manufacturers often have limited control over these factors, and the preferred choice usually revolves around the most cost-effective option. This is the primary reason why coal, despite its environmental impact, is still commonly used in European kilns, accounting for more than 50% of the energy sources [2].

Carbon taxes serve as an instrument to balance costs and encourage a shift towards lower CO_2 emission fuels, such as natural gas (See Section 7.3.4.3). However, a change to natural gas often necessitates significant investments in infrastructure, including special terminals for (intermediate) storage and/or processing, and the development of supply pipelines to serve end customers [51]. Moreover, when considering scenarios involving renewable electricity production, the resources required to build new low-carbon plants mainly comprise construction materials like concrete, steel, and glass. This can potentially double the current need for such materials over the next two decades, leading to additional emissions [45]. Therefore, to achieve the EU's carbon neutrality goals, implementation of carbon capture technologies in cement and lime production facilities becomes crucial for curbing CO_2 emissions, which will be accompanied by building new renewable power plants.

Beyond CO_2 emissions, it is also relevant to examine the consumption of other critical (rare earth) raw materials necessary for emerging technologies (e.g., Boron in wind turbines). Pursuing these materials' extraction may increase energy consumption in the quest for their procurement [52].

7.3.4.2 Regulatory Level

At the regulatory level, the European Commission has implemented a series of measures to facilitate progress toward climate neutrality [9], [23], [24]. First, the European Climate Law, issued in 2021, established a legally binding short-term EU2030 target aimed at reducing net greenhouse gas emissions by 55% compared to 1990 levels, with a long-term objective of achieving net-zero emissions by 2050 [53]. The recently released "fit-for-55" package in July 2023 outlines specific actions to attain the 2030 goal [54]. Among these measures, the reduction of methane emissions is of paramount importance, while one of the main sources of methane emissions is associated with the extraction, transport, distribution, and use of coal and oil. A mitigation plan for the use of these sources is being devised, with 2049 set as the deadline for long-term fossil energy source contracts. Furthermore, there is a goal to establish a market for hydrogen as a renewable energy source, supported by targets such as having 42.5% of energy consumed in the EU come from renewable sources by 2030, as outlined in Figure 7.4. These regulations are poised to necessitate adaptations in kiln technologies used in lime manufacturing, making the evaluation of the environmental impact of hybrid and fully electrified scenarios, as explored in this paper (see Section 7.3.2), particularly relevant. Furthermore, as discussed by a recent study, electrification of cement production processes would be economically competitive only for conservative prices, for which a support by the regulatory bodies will be key[55].

In 2005, the European Union introduced its primary tool for addressing emission reduction: the European Trading System (ETS) [56]. The recently introduced fit-for-55 package has incorporated updated carbon prices and incentives for carbon capture utilization [54]. Carbon prices are the outcome of a complex interplay of policies, encompassing, among others, internal carbon fees, crediting mechanisms, fossil fuel taxes, and support measures for renewable energy. Over the past five years, carbon prices have surged, rising nine-fold, and exceeding 90 €/tCO_2 [57]. As discussed in Section 7.3.3, carbon taxes currently pose a significant challenge in the lime sector, accounting for approximately 26% of production costs due to emissions originating from existing fuel sources and unavoidable emissions from calcite decomposition. It is imperative to offer support to the cement and lime industry through staged or segmented taxation policies during adaptation of their production processes to ensure their competitiveness in the market. This support is not only vital for survival of European industries but also for the well-being of over 50 thousand employees in the sector at large [58], [59].

The current application of the ETS employs the same tax approach for all industries, irrespective of their capacity to recapture CO_2 during other stages of their life cycle. It is essential to recognize that only a handful of materials have been extensively studied for their potential to sequester CO_2 during their use phase, with lime being one of the notable examples [43], [44], [58]. To ensure equitable competition in the market, the formulation of well-balanced carbon pricing requires coordinated political and economic taxing policies at the European level [4]. These policies should account for the properties and potential CO_2 profiles of products throughout their entire life cycle. For lime-based renders and plasters, the use phase holds particular importance, as illustrated in Section 7.3.2.2. Incorporating carbonation into the Cradle-to-Grave life cycle assessment reveals that changing to DSR kiln technology and reducing coal usage in the fuel sources can lead to a carbon-negative scenario (S4A, B and C, Fig. 7.7d). The natural carbon sink offered by lime-based materials should be considered when developing policies and taxation coefficients for regulations.

Advancing toward the implementation of new kiln technologies also represents challenges related to carbon utilization and storage. First, there is a need to develop both knowledge and practical experience to assess the scalability of this emerging technology[60]. The European Union, in collaboration with the private sector, has embarked on funding for developing numerous projects in the cement and lime sector (Appendix A17). However, many of these projects are in their initial stage, and substantial results may not be expected until the end of 2030. Moreover, it is crucial not only to establish clear incentives through legislation but also to make progress in consolidating a European carbon market with a primary focus on the carbon value chain. This specific aspect will be further explored in the next section.

7.3.4.3 Market level

At the market level, energy cost is a critical factor, especially in energy-intensive industries like lime and cement production. Collaborative efforts with regulatory authorities are essential to assist industries in transitioning to lower carbon emission energy sources. Additionally, dealing with unavoidable carbon emissions from limestone decomposition requires the adoption of carbon capture technologies and strategies for carbon utilization and storage. These solutions extend far beyond the current level of knowledge of the construction sector and necessitate the development of a strong carbon value chain, as shown in Figure 7.10. This comprehensive approach is key to making significant steps forward in carbon reduction efforts.

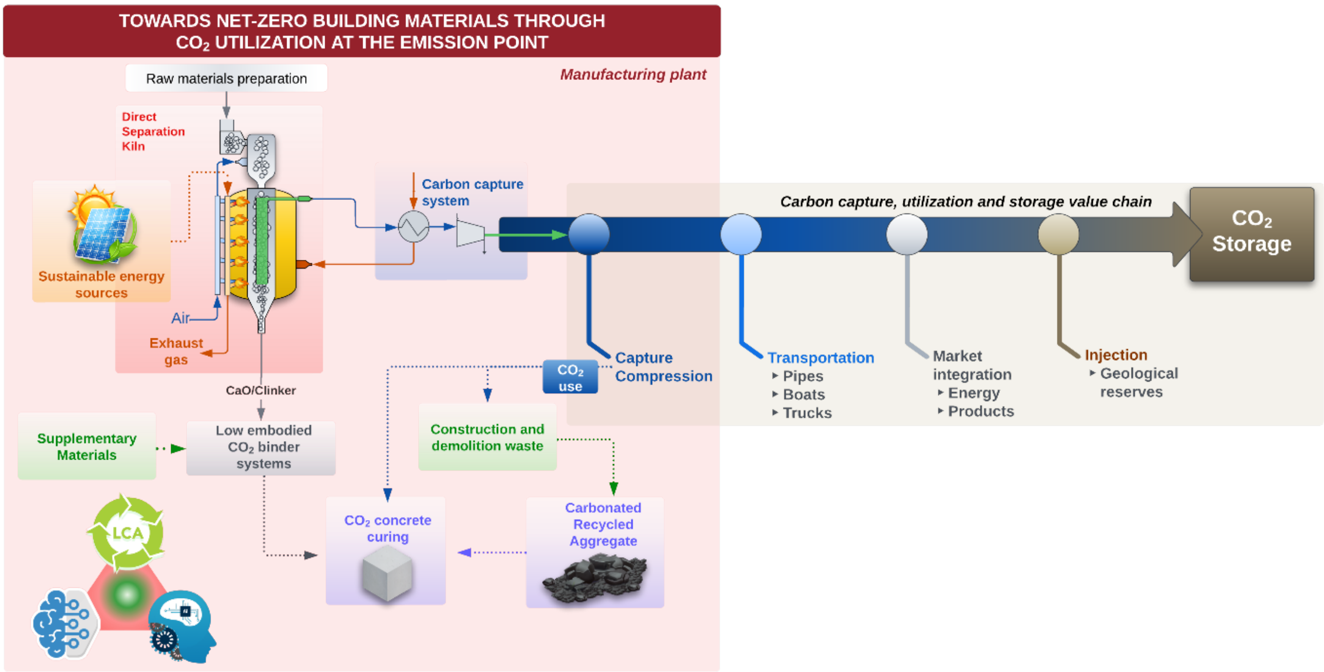


Figure 7.10. CO_2 value chain starting from the emission source (i.e., the cement/lime plant). The Figure highlights the approach of CO_2 use at the emission point to produce net-zero building materials

It may be obvious that the scale of the challenge necessitates a close collaboration among producers, researchers, policy makers, and the carbon market to effectively transform captured CO_2 into valuable products. This collective effort is fundamental for addressing the issue at hand. A pioneering initiative, CO_2 Value Europe (https://co2value.eu/), goes in this direction, uniting industries, start-ups, regional clusters, research organizations, and universities.

An alternative approach to reducing logistics costs and generating added value involves utilizing CO_2 at the emission source. The lime and cement sectors combined, contribute to approximately 10% of anthropogenic CO_2 emissions, and as such, can play a leading role in this transformation endeavour [3], [16], [61], [62]. To achieve this, more research is essential in the field of construction materials, particularly in areas such as CO_2 curing of concrete, industrial carbonation of recycled aggregates, and brick production [63]. As demonstrated in this paper, laboratory-scale research must be upscaled, modelled, and guided by a thorough life-cycle analysis, to a climate neutral pilot and industrial scales production.

However, it is important to acknowledge that not all captured CO_2 from the mineral industry can be (re)used on-site. Therefore, specific attention should be devoted to the logistics of CO_2 transportation. Most cost-effective alternative may involve storage facilities or pipelines, which would also necessitate substantial investments to establish connections between the European carbon market and cooperation with the energy sector to leverage their infrastructure [35].

CO_2 has the potential to be used as a resource in the production of polymers and energy storage, among others [64], [65]. However, logistics of CO_2 transportation and economies of scale play a decisive role in the choice of the right product and value chain set-up [50]. A highly viable option with substantial sustainability potential involves the CO_2 capture and utilization directly at its source of emission, as highlighted in Figure 7.10. This approach aims to generate sustainable construction materials through mineralization, thereby mitigating the logistical complexities associated with CO_2 transportation while simultaneously sequestering CO_2 within the material and creating additional value[66]. Some of these applications, such as the accelerated carbonation of recycled aggregates derived from construction and demolition waste or the curing of concrete under high CO_2 concentrations, have very recently garnered significant attention in the scientific community [67], [68]. Nevertheless, a deeper exploration of their chemical and durability properties is imperative at laboratory scale. Furthermore, the successful scaling-up of these alternatives necessitates a comprehensive performance design that encompasses not only technical attributes and cost considerations but also the outcomes of environmental impact assessments. Addressing this formidable challenge can be only accomplished by integrating data science, machine learning, and artificial intelligence tools in the design of net-zero processes and materials, as depicted in Figure 7.10. This approach would enable the quantitative and precise comparison of diverse CO_2 mineralization applications at the emission source to select the most eco-efficient one [69]. When such framework is coupled with the kiln technology and energy source transition strategy outlined in this paper, they hold the potential to yield construction materials with a net-zero carbon footprint. It is important to recognize that costs and benefits may not always align perfectly along the value chain. Effective cooperation among stakeholders is crucial to achieve market readiness for these alternatives.

Additionally, the storage of CO_2 in geological reserves is one of the latest options, but it is still considered costly and has the drawback of missing the opportunity to create added value from emissions [70]. This highlights the importance of carefully evaluating the trade-offs between cost and value creation in the pursuit of sustainable solutions.

A final crucial question in life-cycle assessment that remains open and will necessitate further discussion and agreements, especially as these technologies are implemented at large scales, pertains to the ownership of the benefits derived from captured CO_2: *should it belong to the emitter or the entity that utilizes it, or something in between?*

7.4 Conclusions

This paper demonstrates a “quarry to carbon sink” approach of lime-based construction materials through process-based LCA modelling for Net-zero and Carbon-negative Transformation. The following conclusions can be drawn:

- Integrated decarbonization strategy: This study rigorously assesses the combined impact of a decarbonization strategy involving a Direct Separator Reactor (DSR) and eco-efficient energy sources across the entire product life-cycle.
- Impressive emission reductions: The DSR technology, in various operational modes, has the potential to significantly reduce emissions, with the fully electrified, renewable-powered kiln coming closest to carbon neutrality from Cradle-to-Gate and carbon negative from Cradle-to-Grave.
- Environmental benefits throughout: Ecosystem quality, resources, and human health indicators remain largely unaffected by these decarbonization efforts.
- Economic sustainability: Carbon capture technologies not only benefit the environment but also enhance economic sustainability, lowering carbon taxes by up to 23%.
- Achieving rapid decarbonization of the lime industry requires a coordinated action plan that balances the needs of the private sector, environmental protection, and societal well-being within a regulatory framework that ensures equitable implementation of political decisions.

Future research should explore digitalization, machine learning and artificial intelligence tools to support the manufacturing of net-zero construction materials through mineralization using the captured CO_2 at the emission source, as well as other alternatives throughout the entire CO_2 value chain. A critical question to address regards to the ownership of benefits derived from captured CO_2.

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Chapter 8: Conclusions and Future Perspectives

The findings of this thesis are synthesized to provide comprehensive insights into the environmental implications of the analysed decarbonization strategies. This section aims to highlight the key outcomes and their significance in advancing sustainability goals within the lime sector and, particularly, lime-based construction materials industry.

Additionally, it delves into the broader implications of the research, discussing potential avenues for further investigation and exploring future perspectives on the continued decarbonization of manufacturing processes.

The section serves as a critical reflection on the study's contributions while paving the way for future research directions and practical applications in the pursuit of more sustainable construction practices.

8.1 Conclusions

8.1.1 General Overview

In this thesis, the feasibility of achieving net-zero and carbon-negative transformations throughout the life-cycle of lime-based rendering materials was rigorously evaluated and quantified. In the introduction (Chapter 2) it is clarified that hydrated lime, responsible for approximately 80% of the CO_2 emissions of the Cradle-to-Gate manufacturing of lime-based rendering materials, necessitated sustainable transformations to decarbonize its manufacturing process. An integral decarbonization strategy was implemented in life-cycle assessment calculations at the manufacturing stage, featuring principles of circular economy, direct separation reactors coupled with carbon capture technologies, and utilization of sustainable energy sources. Another crucial factor to incorporate into the environmental and economic impact assessments of lime-based materials is the carbonation occurring during the application phase of a render. Limited materials exhibit such well-documented potential for carbon uptake, constituting a spontaneous reaction essential to fulfil their role as building materials.

To ensure a robust quantification of net-zero and carbon-negative scenarios, two primary constraints were identified. First, is the challenge to obtain background data for modelling life-cycle inventories, and they are commonly sourced from generic, inflexible, and often unreliable databases. The quality of input data can significantly influence the results and their reliability in any model. The second challenge was the lack of comprehensive sustainability studies for modern lime-based construction materials, a critical factor in establishing a baseline for envisioning carbon reductions and benchmarking alternatives with enhanced sustainability.

The Methodology section (Chapter 3) introduced a parametrized life-cycle inventory modelling methodology, combining concepts from process engineering, materials science, and environmental engineering. This methodology, consistently applied throughout the thesis, incorporates scientific principles and transparent parameterization, enabling practitioners to calculate methodologically sound and well-grounded inventories. The second phase involved collaboration with Sievert, one of the largest ready-mix manufacturers in Germany and a key partner in the SUBLime network. This collaboration facilitated the integration of the developed methodology with first-hand data from the industry. In Chapter 4, critical variables in the production process were determined, alongside a comprehensive quantification of the impact of various components in modern mortars, renders, and plasters, establishing the most thorough analysis of current lime-based masonry mortars to date.

To assess the potential contribution of each decarbonization strategy, individual aspects were addressed in separate chapters (Chapter 5 to 7), identifying challenges and opportunities towards net-zero and carbon-negative transformations.

Chapter 5 delved into circular economy practices in lime-based renders manufacturing, emphasizing the role of closed and open-loop upcycling strategies for their potential to reduce CO_2 emissions and minimize raw materials extraction and waste landfilling.

Chapter 6 provided in-depth insights into the environmental impact of current energy scenarios in the manufacturing of hydrated lime, as well as improvements achievable through the use of low-carbon energy sources.

Finally, Chapter 7 presented a comprehensive comparison of current kiln technologies and energy sources with an enhanced direct separator reactor coupled with carbon capture systems and decarbonized energy sources. Under this improved production system, pure CO_2 can be entirely recovered and utilized in a diverse range of products and industries, transforming it from an emission to a valuable by-product of the lime industry. This chapter integrates the current knowledge generated in the thesis, covering from Cradle-to-Grave, and underscores that net-zero and even carbon-negative scenarios for lime-based renders are feasible. This approach contributes not only to environmental sustainability but also to positive economic impacts. The discussions within the SUBLime network, including the European Lime Association and major lime-based construction materials industries, are synthesized to conclude the chapter by addressing the actions and challenges required for a rapid decarbonization of the lime industry.

8.1.2 Conclusions Related to Net-Zero and Carbon-Negative Transformations in Lime-Based Construction Materials

As stated in Chapter 1, the main goal of this thesis was to propose an integral decarbonization strategy (Figure 1.1) encompassing the Cradle-to-Grave life-cycle of lime-based rendering materials and to quantify these scenarios on their feasibility to achieve net-zero and carbon negative transformations. The expected carbon profile (through Global Warming Potential indicator) of two scenarios, namely Business-as-Usual (BAU) and Decarbonized Process (DP) were described in Figure 1.2, being the core concept of the doctoral thesis. Each scenario was thoroughly addressed in each chapter and below a brief description of the main conclusions is provided.

► **Cradle-to-Gate Business-as-Usual Manufacturing of Lime-based Masonry Mortars, Renders and Plasters (Chapter 4)**

In order to benchmark potential improvements in the manufacturing process, it is firstly required to have a robust quantification of the environmental and economic impact of producing lime-based construction materials in the business-as-usual scenario. This was addressed in Chapter 4 and the following conclusions are drawn:

- In terms of Global Warming Potential (GWP) up to 0.26, 0.29 and 0.32 kg $CO_{2\ eq}$ per kg of material is released for lime-based mortars, renders, and plasters respectively. The HL:CEMII proportion (in weight) in the mixes was 8:17 for mortars, 3:2 for renders and 1:0 for plasters. The GWP increment from mortars over renders to plasters, is mainly attributed to the fact that the mix-specific contribution of HL (0.94 kg $CO_{2\ eq}$/kg HL) is higher than for the Cement Type II (0.76 kg $CO_{2\ eq}$/kg Cement). Around 70-80% of the CO_{2eq} emissions correspond to the binder's production in lime-based mortars, renders and plasters. Aggregates contribute 15 to 20% and additives production around 10% (Section 4.4.1).
- Artificial lightweight aggregates and additives incorporated in the mix have a huge damaging effect on resources depletion and human health, highlighting the importance of developing bio-based replacements (Section 4.4.2).
- Carbon taxes can have a share up to 40% of the total cost, showing that reducing CO_2 emissions would lead not only to environmental sustainability but also is economically advantageous (Section 4.3.4).

The BAU scenario developed in Chapter 4 for modern masonry renders is the reference Cradle-to-Gate scenario used in the next chapters to benchmark the decarbonized process strategies.

► Decarbonized Process Scenarios: Circular Economy, Renewable Energy Sources and Novel Kiln Technologies with Carbon Capture Systems (Chapters 5 to 7)

As outlined in Section 1.1 (Figures 1.1 and 1.2) the integrated decarbonization strategy involves circular economy principles (Chapter 5), renewable energy sources to power the manufacturing plant (Chapter 6), novel kiln technologies with carbon capture systems (Chapter 7) and carbon sequestration of the lime-based render during the use phase (addressed in Chapter 5 and 7). Below the main conclusions of each chapter on their contribution to net-zero and carbon-negative scenarios are stated.

Circular economy principles in lime-based rendering materials (Chapter 5)

Chapter 5 includes two upcycling strategies from the circular economy model: closed-loop (Section 5.1) and open-loop (Section 5.2) upcycling of waste and by-products for the manufacturing of lime-based renders. The main conclusions of these strategies in net-zero and carbon-negative scenarios are drawn below.

- **Closed-loop upcycling (Section 5.1)**
  - This research showcased a circular design for upcycling second-life lime-based renders (SLR), comparing material properties and environmental impact with first-life renders (FLR). The interdisciplinary approach integrates materials science to derive essential input parameters for

Life Cycle Assessment (LCA) analysis. A mineralogical, microstructural, mechanical, and thermal characterization for SLR and FLR were performed (Section 5.1.2).

- SLR showed improved thermal insulation, but lower compressive strength compared to FLR, which is mainly attributed to the denser matrix and lower porosity observed for FLR. Both materials show comparable durability against sulfate attack (Sections 5.1.3.1 to 5.1.3.3).
- The environmental impact assessment showed that from Cradle-to-Gate 25% CO_2 emissions can be cut for the SLR compared to FLR. The Cradle-to-Cradle scenario is more eco-efficient than the Cradle-to-Grave (considering the carbonation during the use phase), leading to CO_2 reductions ranging from 17 to 55% when relevant material properties are incorporated (Section 5.1.3.4).
- Although significant reductions can be obtained, the horizon of net-zero CO_2 emissions is still far away for the closed-loop upcycling. This is mostly attributed to the fact that in the kiln, significant CO_2 emissions from calcite decomposition and fuel combustion is being generated.

- **Open-loop upcycling (Section 5.2)**
  - Two secondary resources (SR), Paper Mill Sludge - PMS (residue from paper production) and Carbide Lime – CL (by-product from acetylene production) were evaluated on their capacity to replace HL in the manufacture of renders.
  - The intermediate treatment required for the conditioning of the secondary resources plays a pivotal role on the environmental impact (Section 5.2.3.1). From Cradle-to-Gate in comparison to hydrated lime-based plasters, PMS allows saving 18% resources and 14% CO_2 emissions, while CL 13% and 80% respectively (Section 5.2.3.4.1). The reason of the improved capability of CL to reduce CO_2 (even after allocation procedures) is that, unlike in PMS, no calcination takes place.
  - From Cradle-to-Grave (considering the carbon uptake during the use phase), CL can reach around 100% reduction compared to HL (close to net-zero scenario), while PMS only 11% (Section 5.2.3.4.2).
  - Both secondary resources reduce also the total costs compared to the traditional scenario, by diminishing purchase costs and costs associated to carbon taxes (Section 5.2.3.5). However, the availability of CL in the European region is limited because acetylene production has been phased out.

Closed and open-loop circular economy strategies show potential to reduce CO_2 emissions, although logistics and availability of the secondary resources in the region of the production plants can present difficulties to secure the supply chain and effectively account for the net-zero transformations.

The role of energy sources in hydrated lime production (Chapter 6)

The CO_2 emissions from fuel combustion in the kiln can represent around one third of the emissions to produce hydrated lime, the main binder in lime-based construction materials. Chapter 6 addresses thoroughly current energy sources of electricity and fuels used in the business-as-usual hydrated lime manufacturing in Europe. A scenario analysis of different decarbonization pathways is carried out (Decarbonized Process), contemplating the regional availability of resources in four European countries (Germany, Belgium, Portugal, and Spain). The BAU and DP scenarios in terms of energy sources used in the lime kiln are quantitatively compared with the main conclusions described below.

- A detailed life-cycle inventory for hydrated lime manufacturing was developed and used to parametrize and implement the BAU and DP scenarios (Section 6.3.1).
- In terms of Global Warming Potential, 0.94 kg $CO_{2\ eq}$/kg HL are produced on average. The emissions are due for 60% to limestone decomposition, 39% is attributed to the combustion of the fuel and 1% to the electricity consumption. Spain and Belgium have shown lower environmental impact, namely in 7 out of 15 indictors for Spain and 5 out of 15 for Belgium, followed by Portugal (1 out of 15) and Germany (1 out 15) (Section 6.3.2.1).
  A shift to almost 100% renewable energies for electricity production reduces the majority of the impacts. A decrement in the proportion of fossil solid fuels, while incrementing natural gas and biomass, can lead to around 22% CO_2 emission reduction (Section 6.3.2.2). However, while CO_2 emissions can be reduced, not all impact indicators follow the same trend when a change from fossil sources to decarbonized fuel/renewable energy is made (Section 6.3.2.3).

Quantitative demonstration of net-zero and carbon-negative transformations for lime-based rendering materials (Chapter 7)

In Chapters 1 to 6 it was quantitatively demonstrated that the CO_2 emissions concentrated in the lime kiln (due to limestone decomposition and fuel combustion) are the main barrier to achieve net-zero and eventually carbon-negative scenarios in lime-based construction materials. Therefore, Chapter 7 integrates in a Cradle-to-Grave impact quantification the effect of kiln technology, energy sources and natural carbonation in a systematic scenario analysis for a lime-based render. The most used kiln technology in Europe, Parallel Flow Regenerative Kiln (PFRK) is compared against a novel kiln technology (Direct Separation Reactor, DSR) that allows a direct separation and capture of the unavoidable CO_2 from calcite decomposition. BAU and DP energy sources (evaluated in Chapter 6) are implemented in the scenario analysis, which also incorporates the feature of the DSR kiln to operate in full electrified mode. The following conclusions are drawn:

- The synergistic impact of an integrated decarbonization strategy combining a DSR and eco-efficient energy sources in hydrated lime manufacturing shows the feasibility of scenarios close to net-zero CO_2 emissions and minimal adverse effects observed across ecosystem quality, resources and human health areas of damage (Section 7.3.2.1).

- At a render level and from Cradle-to-Grave (Section 7.3.2.2), scenarios close to net-zero are obtained only by combining the DSR at the manufacturing phase with the natural carbonation of the plaster. The DSR technology in various operational modes with decarbonized energy sources presents a remarkable potential for carbon-negative scenarios, meaning that the render behaves as a carbon sink during its use phase (i.e., removing CO_2 from the atmosphere). The economic viability by reducing carbon taxes of these innovations and the opportunities of additional circularity of the captured CO_2 is also highlighted (Section 7.3.3).
- Achieving rapid decarbonization of the lime industry demands a coordinated action plan, balancing the interests of the private sector, environmental protection, and societal well-being within a regulatory framework designed to ensure the equitable implementation of political decisions. Such a concerted effort is essential to navigate the complexities of rapid industry transformation while fostering sustainability across economic, environmental, and social dimensions (Section 7.3.4).

8.1.3 Wider implications of this research

Decarbonizing the lime process, being the precursor for hydrated lime as the main binder in lime-based construction materials, certainly holds promise to achieve net-zero and carbon-negative transformations over its complete life-cycle, as discussed in Section 8.1.2.

Lime is a critical material in a wide variety of applications and production processes but emits large quantities of CO_2 (around 1.1 kg CO_2/kg CaO, Chapter 6). Globally around 400 million tons of CaO are generated and used for lime-based construction materials, steel production, sugar refining and water purification. As a result, around 480 million tons of CO_2 are yearly produced in the world. While some extreme positions argue for halting lime production to reduce pollution, it's crucial to envision the consequences of a modern world without this material.

In this thesis, the effects of energy sources (Chapter 6) and kiln technologies (Chapter 7) on the environmental impact and, precisely, on CO_2 emissions of lime/hydrated lime Cradle-to-Gate manufacturing were quantitatively assessed. The direct separation reactor coupled with carbon capture technologies has the ability to separate all unavoidable emissions (around two thirds of the total) from limestone decomposition (0.78 kg CO_2/kg CaO) (Section 7.3.2.1). Not all kilns must be completely replaced, as many of them have the potential to be retrofitted. Further reduction must be achieved by changing the energy sources, particularly moving to low carbon fuels and renewable energy sources in the electricity grid (Section 6.3.2.2 and 7.3.2.1). These strategies can be globally applied and will have a direct influence on the manufacturing stage, improve the sustainability of lime in all its uses and applications.

An additional contribution of the research is to support through quantitative assessment the transformation of the mindset to turn CO_2 from an emission to a valuable by-product (Section 7.3.4). Over the last decades, CO_2 has been positioned as a great challenge for the industrial sector. However, the lime sector holds the advantage of producing pure CO_2 if a decarbonization framework such as mentioned before is applied. Under this philosophy, the large amounts of CO_2 generated by the industry are not considered a threat anymore but rather a competitive opportunity. Great advances are being made to secure a robust CO_2 value chain, integrating all stakeholders, from manufacturers, to governments and policy makers, market, and scientific community. CO_2 is expected to be in the nearby future the precursor of green energy sources and sustainable products, contributing to turn net-zero emissions from dreams to a concrete reality.

8.2 Future Perspectives

The thesis aimed at conducting a comprehensive and quantitative environmental and economic impact assessment of an integral decarbonization strategy, covering Cradle-to-Grave in lime-based rendering materials. The research was focused on a detailed Cradle-to-Gate scope of hydrated lime and renders manufacturing. The scope also included a Cradle-to-Grave analysis, considering a single render layer applied to a masonry wall. However, there are still several challenges in the field that deserve further research and attention. The following potential future steps are identified:

1. **From regional to global scale**

   The thesis, in general, has been focused on a regional scale in Germany. Even inside the European continent, the resources available in each country differ and so will the impact of energy use in the lime kiln. Net-zero emissions require a global unification of efforts. It is essential to conduct a thorough analysis of the challenges and opportunities, as representative as possible of the country-specific situation in terms of energy sources and potentiality for renewable energy development. The implementation of these scenarios at the manufacturing level will allow for a more rigorous quantification of CO_2 savings at a global scale. From a technology perspective, further economic assessment of investments in kiln retrofitting or complete replacement needs to be carried out.

2. **Masonry systems and buildings**

   In the present research only one component of masonry systems was addressed: the external rendering materials. However, masonry systems are more complex and include bricks, bedding mortars and insulation materials. In many of them, lime intervenes as raw material. Even though from a single-component point of view carbon-negative emissions can be achieved, at a masonry level this needs to be investigated. In particular, it can be assumed a complete carbonation of the render within a reasonable period of time, because it is applied as a thin layer exposed to the atmosphere. However, this will not be the case for a mortar, for instance. Furthermore, when operational energy at a building scale is included in the assessment, the results can be potentially different. Therefore, it is relevant to continue this research line assessing increasing scales and a variety of functional units.

3. **Carbon use at the emission point**

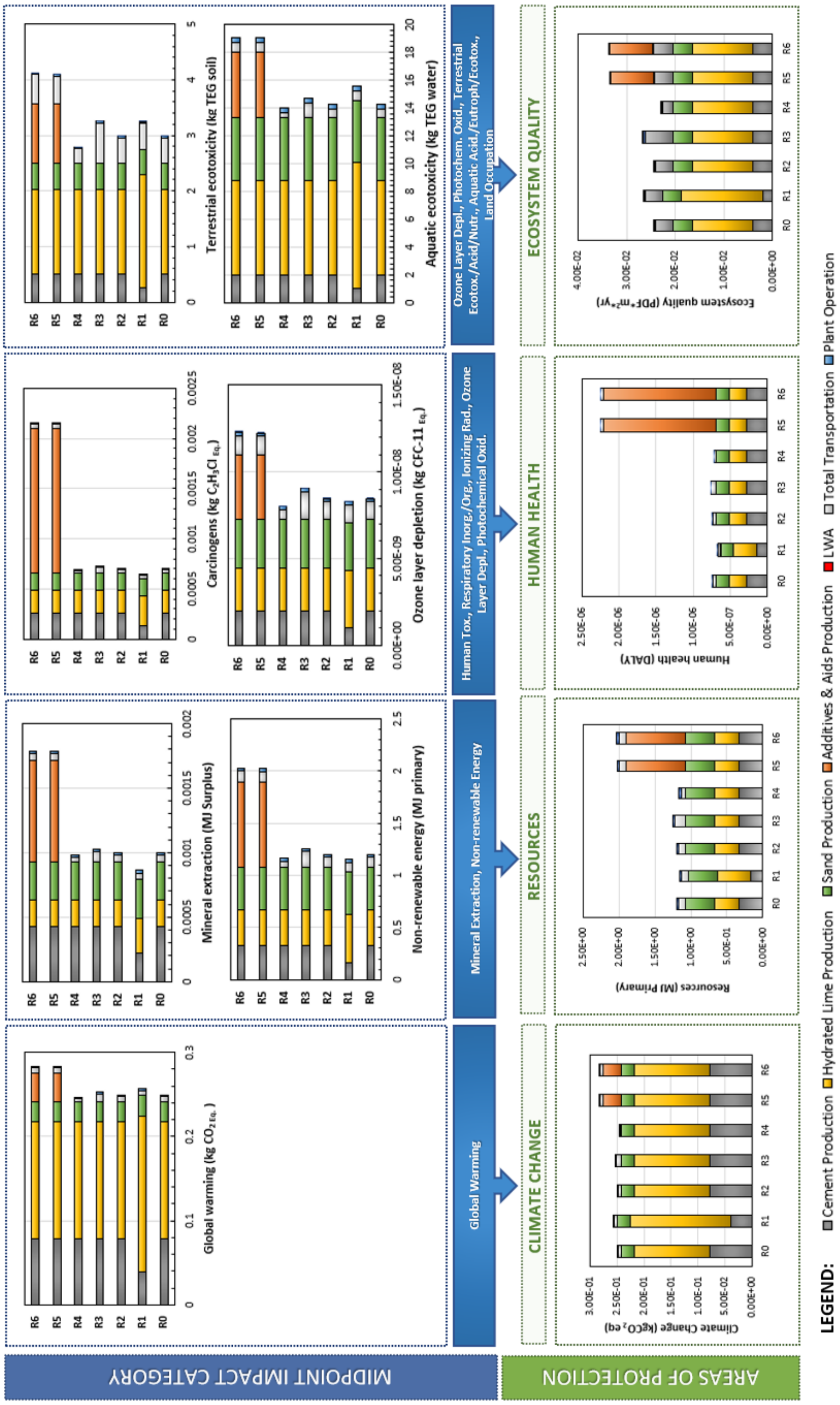
   After being captured, CO_2 must be used in a certain application to be effectively removed from manufacturing emissions. Several alternatives are being studied to produce green energy and a wide variety of products from CO_2. However, the logistics of CO_2 transportation are one of the most critical challenges in the CO_2 value chain. One potential alternative would be the use of CO_2 at the emission point, i.e., the manufacturing plant, for the production of sustainable building materials. Further research could explore CO_2 use as an admixture in concrete manufacturing, concrete precast curing, production of sand-lime bricks and improvement of recycled aggregates from construction and demolition waste by accelerated carbonation.

Appendix

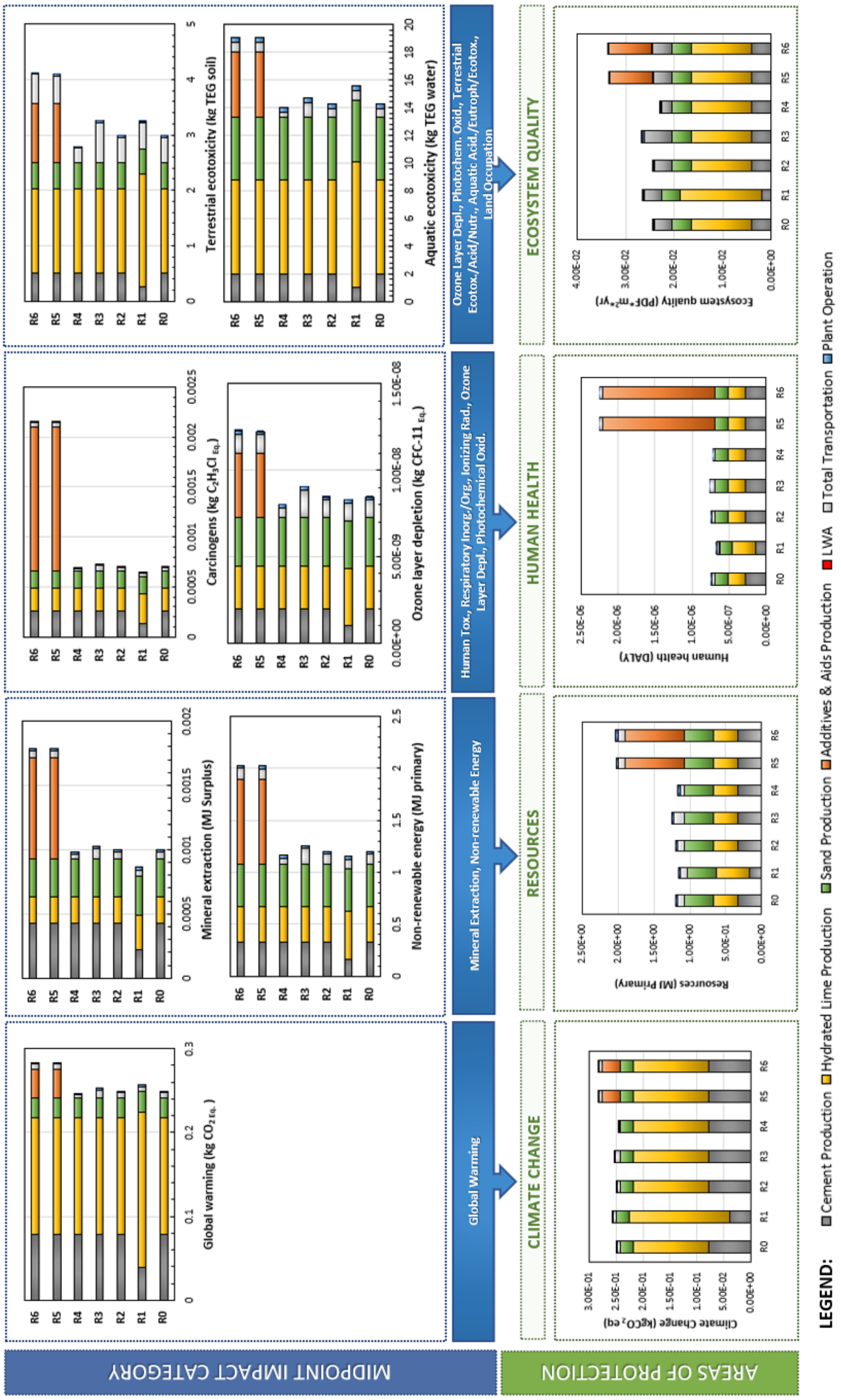
Appendix 1. Life-cycle Inventory to produce the sand for the mortars, renders and plasters.

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
Raw Materials Reception						
Input	**Sand from the Quarry**	0.56	t		t	Modelled by EcoInvent (gravel and sand quarry operation, CH)
	Transport to the factory	5.6	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO4 | RoW)
	Sand from the River	0.49	t	0.08	t	Modelled by EcoInvent (sand quarry operation, extraction from riverbed, GLO)
	Transport to the factory	4.9	t*km	100	km	Transport, lorry 16-32 metric ton, EURO4 | RoW)
	Conveyor belt	0.0042	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
Output	**Sand**	1.05	t	1.05	t	Output of the Raw Materials Reception
Primary crushing – Sand from Quarry						
Input	**Sand from Quarry**	0.56	t			Input from Raw Materials Reception
	Primary crushing	0.28	kW	0.5	kWh / t	Electricity mix (SUBLime designed)
	Conveyor belt	0.000112	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
Output	**Sand crushed**	0.56	t	1.05	t	Output of the Primary Crushing operation
Drying						
Input	**Sand crushed**	0.56	t	0.56	t	Input from Primary Crushing
	Sand from the River	0.49	t	0.56	t	Input from Raw Materials Reception
	Sand Drier	210	MJ	200	MJ / t	Modelled by EcoInvent (silica sand production, DE)
	Conveyor belt	0.004	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
Output	**Sand dried**	1.00	t	1.00	t	Output of Drying operation
	Water vapor	0.50	t	0.50	t	Modelled by EcoInvent (Emission to air, low population)
Classification and secondary crushing						
Input	**Sand dried**	1.00	t			Input from the Drying operation
	Centrifugal classification	2.00	kWh	2.00	kWh / t	Electricity mix (SUBLime designed)
	Secondary crushing	7.14	kWh	7.14	kWh / t	Electricity mix (SUBLime designed)
	Conveyor belt	0.004	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
Output	**R&P Sand**	1	t	1	t	Product of the sand production process
	Emissions					Emissions along the production process of Render/Plaster sand

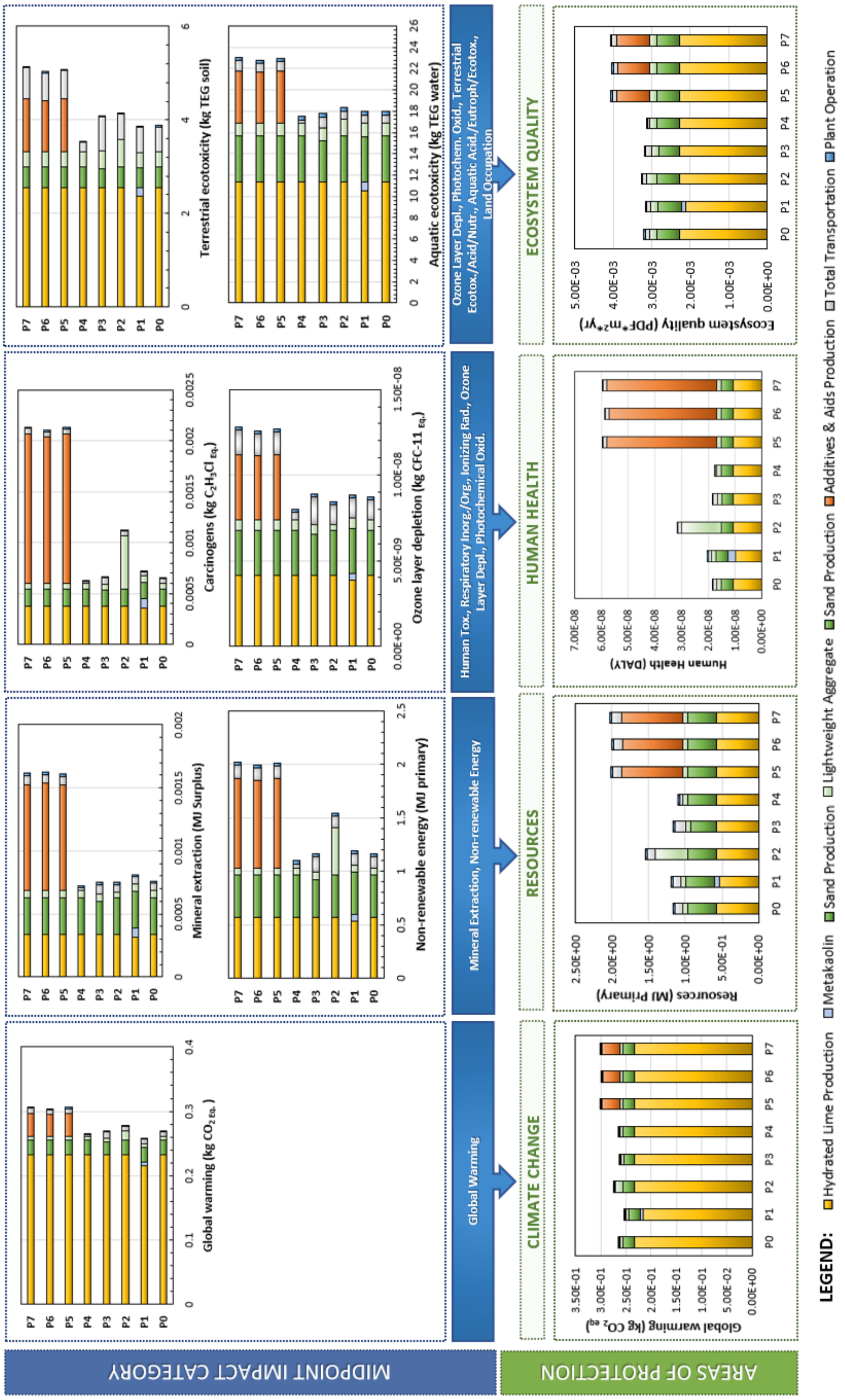
Appendix 2. Cradle-to-Gate Life-Cycle Assessment results per kg of mortar (reference and case studies, midpoint and endpoint indicators)



Appendix 3. Cradle-to-Gate Life-Cycle Assessment results per kg of render (reference and case studies, midpoint and endpoint indicators)



Appendix 4. Cradle-to-Gate Life-Cycle Assessment results per kg of plaster (reference and case studies, midpoint and endpoint indicators)



Appendix 5. Manufacturing process of a lime-based render

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
				Render Manufacturing		
Input	**Sand aggregate**	0.75	t	0.75	t	LCI available in [56]
	Transport	75	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.0003	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
	Hydrated Lime	0.25	t	0.25	t	Modelled by SUBLime LCI [41]
	Transport	25	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.0001	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
	Dry mixer	4	kWh	4	kWh / t	Electricity mix (SUBLime designed)
Output	**Render R0**	1.00	t	1.00	t	Output of the Render Manufacturing

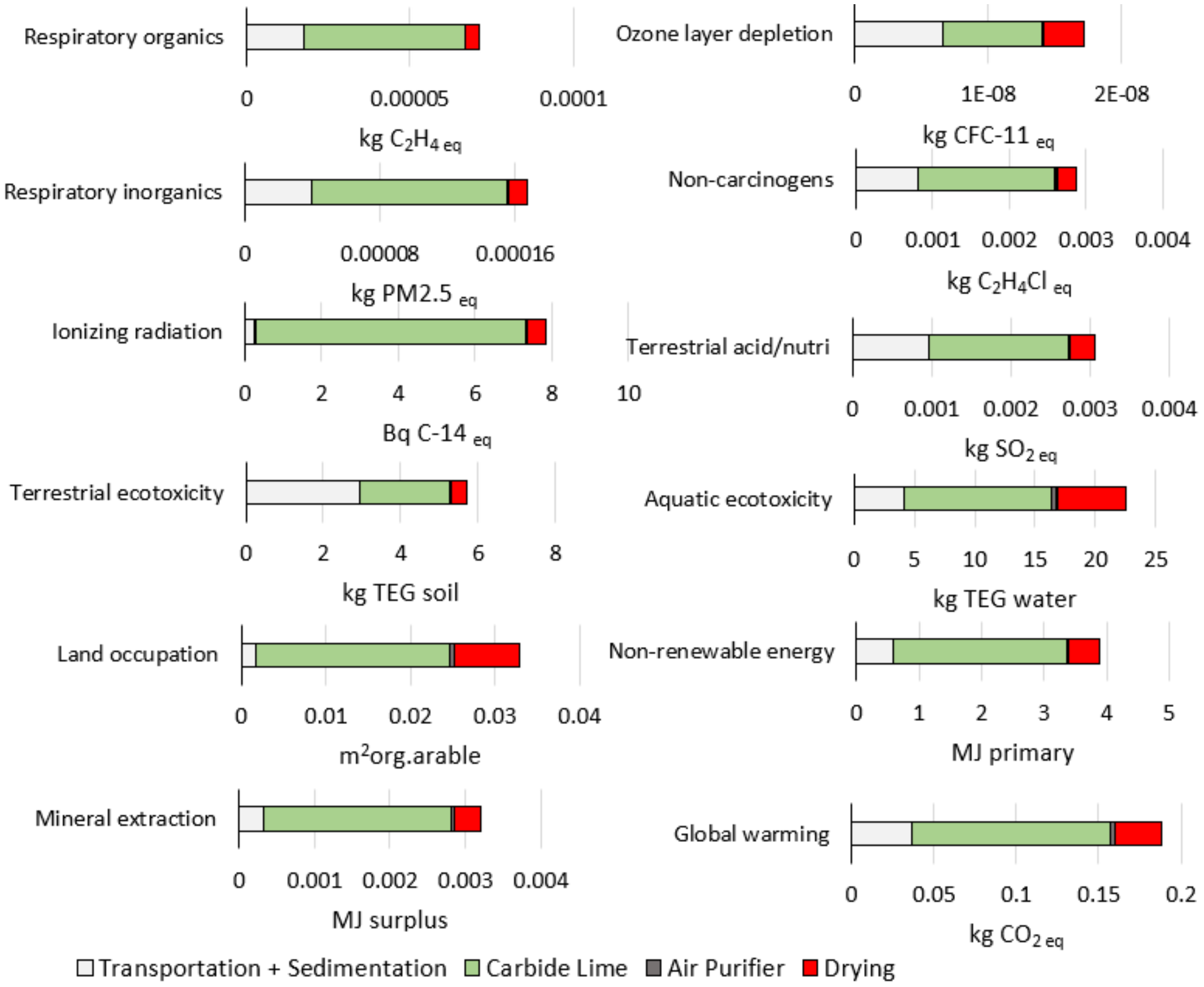
Appendix 6. Recycling of render at the end of life and production of recycled aggregate and second life binder

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
		Recycled treatment of 1 ton of CLM				
Input	**Render at the end of life**	1.05	t			
	Transport	100	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Primary Crushing	0.5	kWh	0.5	kWh/t	Electricity mix (SUBLime designed)
	Centrifugal classificator	2	kWh	2	kWh/t	Separation of the coarse fractions for recycling aggregate and fine fractions for binder production
	Secondary Crushing	7.14	kWh	7.14	kWh/t	Electricity mix (SUBLime designed)
	Dryier	220.5	MJ	210	MJ/t	Electricity mix (SUBLime designed)
Output	**MFR**	0.25	t			25% allocation
	RA	0.75	t			75% allocation
	Water Vapor	0.05	t			
		MFR Calcination and grinding				
Input	**MFR**	0.25	t			
	Endless screw conveyor	0.0003	kWh	0.004	kWh / t	Electricity mix (SUBLime designed) [41]
	Kiln Fuel consumption	660	MJ	2640	MJ/t	Fuel mix (SUBLime designed) [41]
	Kiln Electricity consumption	6.775	kWh	27.10	kWh/t	Electricity mix (SUBLime designed) [41]
	Vertical mill	8.25	kWh	33	kWh/t	Electricity mix (SUBLime designed) [41]
Output	SL-1000	0.1914				Stoichiometrically calculated (DTA/TG)
	CO_2	0.0546				Stoichiometrically calculated (DTA/TG)
	H_2O	0.0004				Stoichiometrically calculated (DTA/TG)

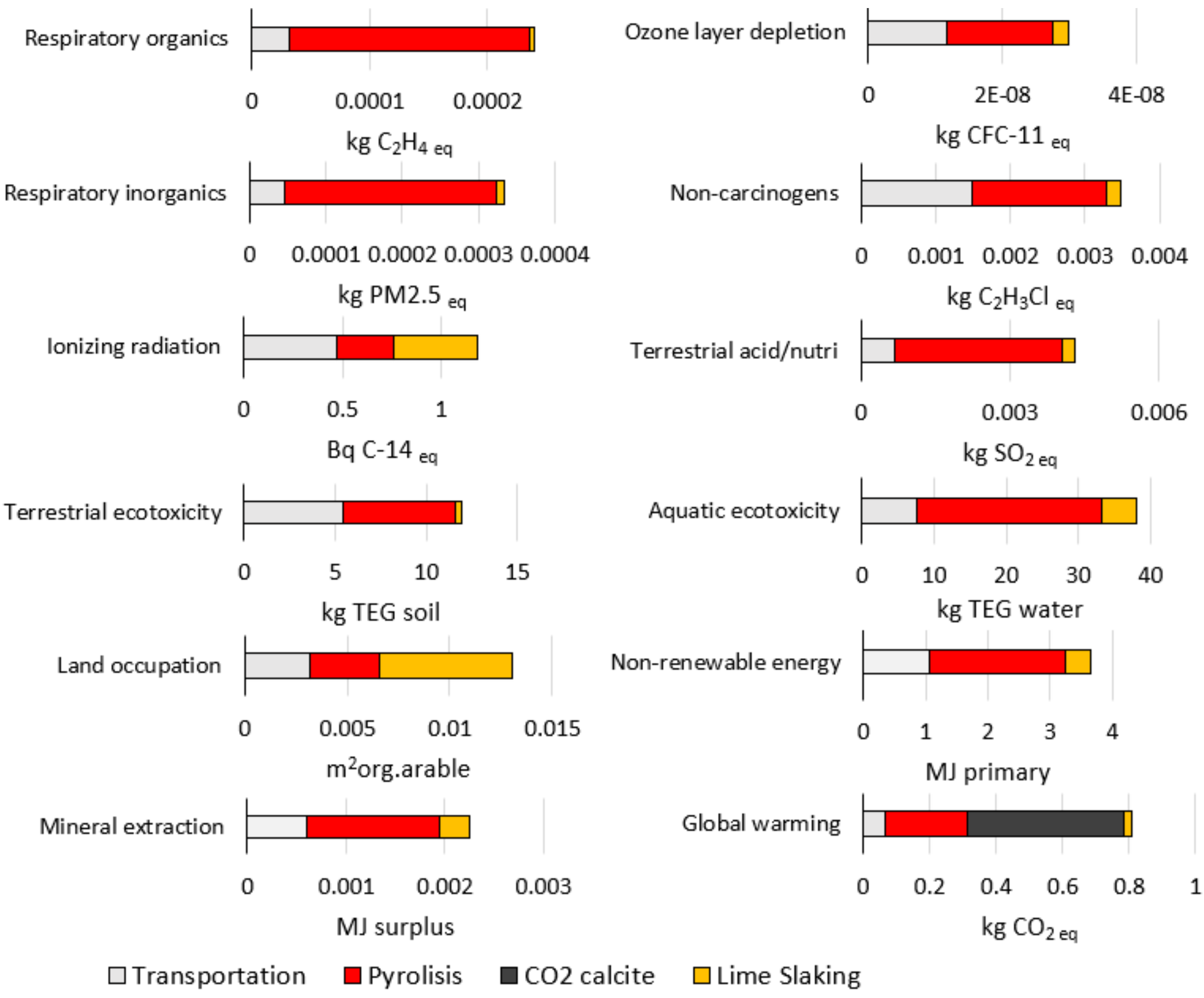
Appendix 7. Datasets from Ecoinvent V3.6 used to model the production of different components of the life-cycle inventory

Item	Process	Ecoinvent 3.6 Dataset
Binder	Hydrated Lime	SUBLime LCI
	Metakaolin	kaolin production, operation and transformation, CH
Coarse Aggregate	Silica Sand	gravel and sand quarry operation, CH
Lightweight Aggregate (LWA)	Pumice Aggregate	pumice quarry operation, GLO
	Perlite	expanded perlite production, GLO
(artificial)	Polystyrene	polystyrene production, expandable, GLO
Additives	Water Retention Agent (WRA)	carboxymethyl cellulose production, powder–RoW
	Air Entrainer (AE)	alkylbenzene sulfonate, petrochemical RoW
	Hydrophobic Agent (HA)	polycarboxylates production, 40% active substance, RER
	Dispersion Agent (DA)	ethylene vinyl acetate copolymer production, RER
Pigments	White Pigment	calcium carbonate production, precipitated, GLO
	Red Pigment	portafer production, GLO
Transportation	Lorry	transport, lorry 16-32 metric ton, EURO6 | RoW

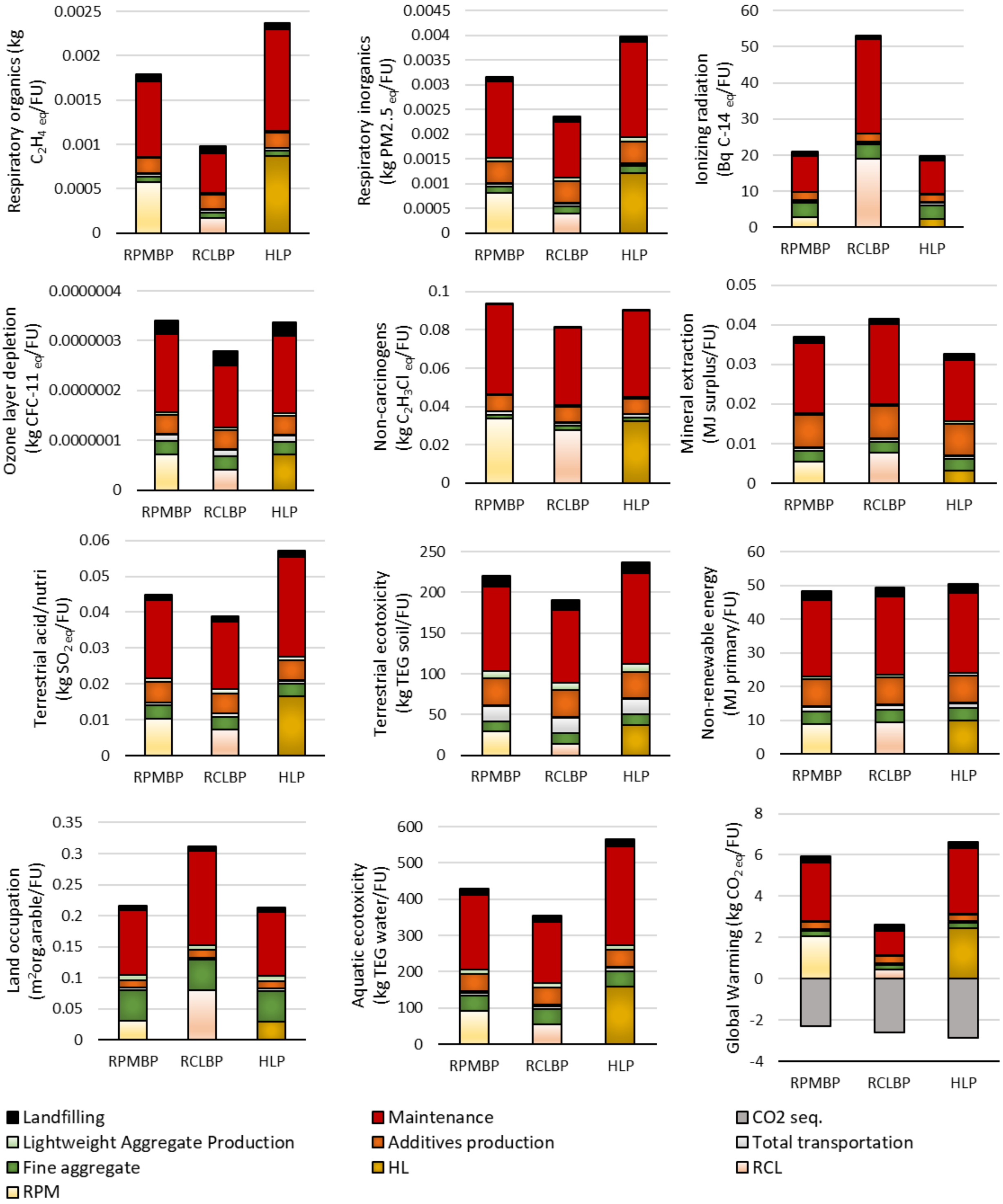
Appendix 8. Complete set of midpoint indicators per kg of RCL (Ec.) utilized in the calculation of the endpoint categories



Appendix 9. Complete set of midpoint indicators per kg of RPM utilized in the calculation of the endpoint categories



Appendix 10. Complete set of midpoint indicators per Functional Unit from Cradle-to-Grave for RPMBP, RCLBP and HLP utilized in the calculation of the endpoint categories



Appendix 11. Providers of Energy Source for Electricity and Fuel mixes – Germany 2020

	Flow	Amount (MJ)	Description	Provider
Electricity	electricity, high voltage	9	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S – DE
	electricity, high voltage	4	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S – DE
	electricity, high voltage	4	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S – DE
	electricity, high voltage	18	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S – DE
	electricity, high voltage	25	Coal	electricity production, hard coal | electricity, high voltage | APOS, S – DE
	electricity, high voltage	17	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S – DE
	electricity, high voltage	11	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S – DE
	electricity, high voltage	1	Oil	electricity production, oil | electricity, high voltage | APOS, S – DE
	electricity, high voltage	9	Solar	electricity production, solar thermal parabolic trough, 50 MW | electricity, high voltage | APOS, S – RoW
	electricity, medium voltage	2	Waste	electricity, from municipal waste incineration to generic market for electricity, medium voltage | electricity, medium voltage | APOS, S – DE
Fuel mix	heat, district or industrial, other than natural gas	2	Biomass	heat production, wood chips from industry, at furnace 1000kW | heat, district or industrial, other than natural gas | APOS, S - DE
	heat, district or industrial, other than natural gas	5	Oil	heat production, heavy fuel oil, at industrial furnace 1MW | heat, district or industrial, other than natural gas | APOS, S - Europe without Switzerland
	heat, district or industrial, other than natural gas	8	Waste	heat, from municipal waste incineration to generic market for heat district or industrial, other than natural gas | heat, district or industrial, other than natural gas | APOS, S - DE
	Heat, district or industrial, natural gas	34	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	Heat, district or industrial, other than natural gas	51	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland

Appendix 12. Providers of Energy Source for Electricity Production – Germany 2020 and 2050

	Flow	Amount (MJ)	Description	Provider
DE 2020	electricity, high voltage	9	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - DE
	electricity, high voltage	4	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - DE
	electricity, high voltage	4	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - DE
	electricity, high voltage	18	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - DE
	electricity, high voltage	25	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - DE
	electricity, high voltage	17	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - DE
	electricity, high voltage	11	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - DE
	electricity, high voltage	1	Oil	electricity production, oil | electricity, high voltage | APOS, S - DE
	electricity, high voltage	9	Solar	electricity production, solar thermal parabolic trough, 50 MW | electricity, high voltage | APOS, S - RoW
	electricity, medium voltage	2	Waste	electricity, from municipal waste incineration to generic market for electricity, medium voltage | electricity, medium voltage | APOS, S - DE

DE 2050	electricity, high voltage	3	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - DE
	electricity, high voltage	7	Geothermal	electricity production, deep geothermal | electricity, high voltage | APOS, S - DE
	electricity, high voltage	26	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - DE
	electricity, high voltage	34	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	4	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - DE
	electricity, high voltage	26	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - DE

Appendix 13. Providers of Energy Source for Electricity Production – Belgium 2020 and 2050

	Flow	Amount (MJ)	Description	Provider
BE 2020	electricity, high voltage	28	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - BE
	electricity, high voltage	5	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - BE
	electricity, high voltage	7	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	7	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	37	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - BE
	electricity, high voltage	2	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - BE
	electricity, high voltage	5	Oil	electricity production, oil | electricity, high voltage | APOS, S - BE
	electricity, high voltage	5	Solar	electricity production, solar thermal parabolic trough, 50 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	2	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - BE
	electricity, medium voltage	2	Waste	electricity, from municipal waste incineration to generic market for electricity, medium voltage | electricity, medium voltage | APOS, S - BE

BE 2050	electricity, high voltage	24	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	17	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	3	Geothermal	electricity production, deep geothermal | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	36	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	20	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - BE

Appendix 14. Providers of Energy Source for Electricity Production – Portugal 2020 and 2050

	Flow	Amount (MJ)	Description	Provider
PT 2020	electricity, high voltage	3	Solar	electricity production, solar thermal parabolic trough, 50 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	7	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - PT
	electricity, high voltage	2	Oil	electricity production, oil | electricity, high voltage | APOS, S - PT
	electricity, high voltage	4	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - PT
	electricity, high voltage	12	Wind OffShore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - PT
	electricity, high voltage	12	Wind OnShore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - PT
	electricity, high voltage	24	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - PT
	electricity, high voltage	27	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - PT
	electricity, high voltage	9	Fossil Co-generation	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - PT
PT 2050	electricity, high voltage	26	Wind OnShore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - PT
	electricity, high voltage	37	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	22	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - PT
	electricity, high voltage	4	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - PT
	electricity, high voltage	11	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - PT

Appendix 15. Providers of Energy Source for Electricity Production – Spain 2020 and 2030

	Flow	**Amount (MJ)**	**Description**	**Provider**
SP 2020	electricity, high voltage	9	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	2	Natural Gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	12	Wind OffShore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - ES
	electricity, high voltage	20	Combined cycle	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	12	Fossil Co-generation	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	6	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - ES
	electricity, high voltage	2	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - ES
	electricity, high voltage	25	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - ES
	electricity, high voltage	12	Wind OnShore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - ES

SP 2030	electricity, high voltage	1	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - ES
	electricity, high voltage	15	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - ES
	electricity, high voltage	2	Fossil Co-generation	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	2	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - ES
	electricity, high voltage	16	Wind Offshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - ES
	electricity, high voltage	17	Combined cycle	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	1	Natural Gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	30	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	16	Wind Onshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - ES

Appendix 16. Providers of Energy Source for Kiln Fuel Feeding – Europe 2010 and Future Scenarios 2050

	Flow	Amount (MJ)	Description	Provider
2010	heat, district or industrial, other than natural gas	2	Biomass	heat production, wood chips from industry, at furnace 1000kW |
	heat, district or industrial, other than natural gas	5	Oil	heat production, heavy fuel oil, at industrial furnace 1MW
	heat, district or industrial, other than natural gas	8	Waste	heat, from municipal waste incineration to generic market for heat district or industrial, other than natural gas
	Heat, district or industrial, natural gas	34	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	Heat, district or industrial, other than natural gas	51	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland
S1 2050	Heat, district or industrial, natural gas	44	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	heat, district or industrial, other than natural gas	5	Oil	heat production, heavy fuel oil, at industrial furnace 1MW
	heat, district or industrial, other than natural gas	8	Waste	heat, from municipal waste incineration to generic market for heat district or industrial, other than natural gas
	heat, district or industrial, other than natural gas	12	Biomass	heat production, wood chips from industry, at furnace 1000kW
	heat, district or industrial, other than natural gas	31	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland
S2 2050	Heat, district or industrial, natural gas	54	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	heat, district or industrial, other than natural gas	8	Waste	heat, from municipal waste incineration to generic market for heat district or industrial, other than natural gas
	heat, district or industrial, other than natural gas	11	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland
	heat, district or industrial, other than natural gas	27	Biomass	heat production, wood chips from industry, at furnace 1000kW
S3 2050	Heat, district or industrial, natural gas	60	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	heat, district or industrial, other than natural gas	32	Biomass	heat production, wood chips from industry, at furnace 1000kW | heat, district or industrial, other than natural gas
	heat, district or industrial, other than natural gas	8	Waste	heat, from municipal waste incineration to generic market for heat district or industrial, other than natural gas

Appendix 17. European/UK projects oriented to the decarbonization of the lime industry

Strategy	Area	Name of the project	Main goal	Period
Carbon direct avoidance	Energy efficiency	WHEATREC4PG: Waste Heat Recovery for Power Generation *Lime: Lhoist*	Recovers 4 MW of thermal power from a rotary kiln exhaust gas, and converts it to 0.5 MWe of low carbon electrical power	2012-2013
	Energy efficiency	CHP GENERATION: Combined Heat and Power (CHP) for Limestone Milling *Lime: Carmeuse*	Energy recovery from drying / heating off gases: Instead using 40% of natural gas' thermal energy for producing electrical power, in the CHP more than 70% of thermal energy is utilized	2014-2017
	Renewable energy sources	ENERGY OPTIMIZATION: Reduced energy consumption through optimized processes and capacity use *Lime: Nordkalk*	Biogas production from by-products of the food industry slaughterhouses and livestock-breeding as well as wastewater sludge	2011-2016
	Renewable energy sources	ADIREN4LIME: Anaerobic Digestion as Renewable Energy for the Lime Industry *Lime: Singleton Birch*	Production of biogas from biomass in anaerobic conditions and used as fuel for the lime kiln. Use in CHP	2014-2015
	Renewable energy sources	HYDROGEN FUEL ENERGY INNOVATION: Alternatives to natural gas for high calcium lime manufacturing *Lime: Mineral Products Association / British Lime Association*	Use of hydrogen as an alternative fuel for high calcium lime manufacturing. Effect on PFR and VSK.	2019-2022
	Renewable energy sources	BIOXYSORB: Biomass co-combustion under both air- and oxy-fuel conditions *Lime: Lhoist*	Emissions of 1st and 2nd generation biomasses, evaluation of sorbents and plant modifications for high thermal share biomass co-combustion	2013-2016
	Renewable energy sources	ENERGY GENERATION: Largest Solar panel farm in Wallonia by a mining company *Lime: Carmeuse*	13.200 solar panels will supply annually over 3.6 GWh of electricity to power lime production	2014-2017

Appendix 17. European/UK projects oriented to the decarbonization of the lime industry *(Continued)*

Carbon direct separation	Kiln technology development	LEILAC1: Low Emissions Intensity Lime and Cement *Lime: Lhoist & Tarmac*	Calix's DSR technology is an in-situ CO_2 capture technique that requires no additional chemicals or equipment	2016-2021
	Kiln technology scale up	LEILAC2: Low Emissions Intensity Lime and Cement *Lime: Lhoist*	X4 scale-up of LEILAC1 and upgrade into dual mode electricity/natural gas potentially balancing services to the grid	2020-2025
	Kiln technology development	ZERCAL: Zero-Carbon Lime pilot plant *Lime: Singleton Birch*	New oxy-fuel flash calcination process able to capture all CO_2 from lime production (lime decomposition and fuel combustion)	2021-2024
	Kiln technology development	Butterfly Project *Lime: Carmeus*	A new type of kiln designed to effectively capture and concentrate CO_2 released during lime production	2023 -
	Kiln technology development	EVEREST project *Lime: Lhoist*	Improved calcination and carbon capture for the largest lime plant in Europe	2023-
CO_2 value chain	Carbon use	COLUMBUS: Power to Methane *Lime: Carmeuse*	Transforming CO_2 generated during the lime production process and hydrogen into e-methane, a renewable gas that can be injected into the gas network or used to power vehicles and industry. Pure CO_2 is required.	2022-2025
	Carbon use	$LOWCO_2$ Lime: Calcinor	Incorporating CO_2 to alkaline residues (slags of energy plants, slags of steel mills, residues of RCDs construction and demolition and to produce methane/methanol	2019-2022
	Carbon use	NKL: Neutral Kero Lime Lime: Lhoist	innovative e-Kerosene process using CO_2 from a lime kiln and hydrogen produced from green electricity via the Fischer Tropsch process	2022-2026
	Carbon storage	CSM: Carbon Storage by Mineralization Lime: Nordkalk	A rock composed mainly of magnesium silicate mineral serpentine reacts with the CO_2 to form a stable compound, thus fixing the CO_2 permanently. Direct mineralisation of flue gas instead of separated and compressed CO_2.	2011-2016

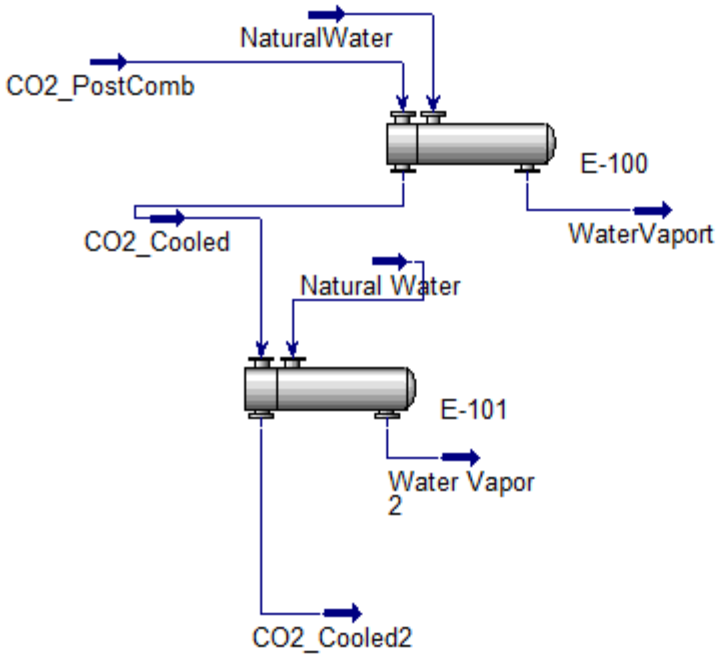
Appendix 18. Life-cycle inventory for the production of the lime-based plaster

The LCI for the production of the lime-based plaster is based on Section 4.4.1. The following additives are incorporated in the mix (referred to the binder in mass proportions): 2.5% Dispersion Agent, 0.20% Water Retention Agent, 0.02% Air Entrainer, 0.3% Hydrophobic Agent. The average transportation distance per additive is 250 km.

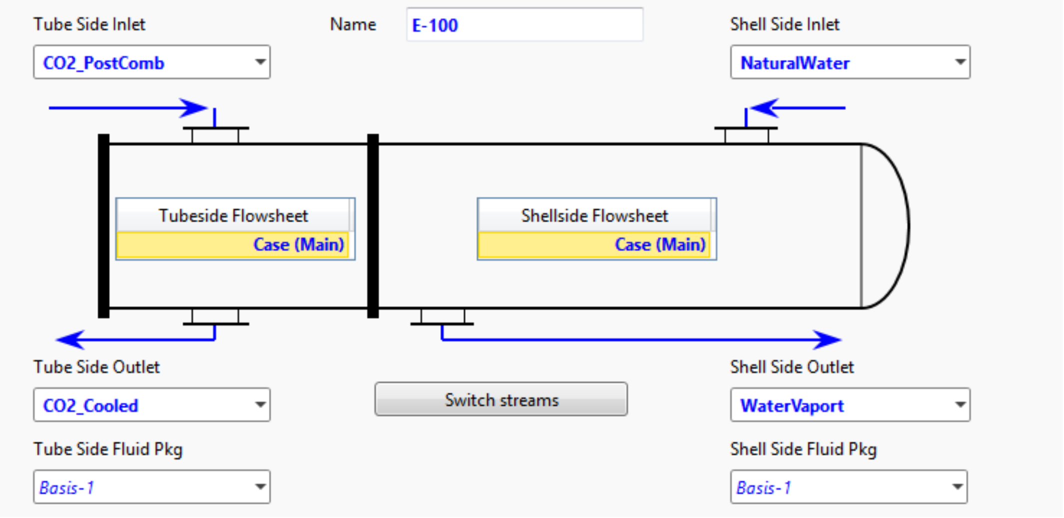
OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
			Plaster Manufacturing			
Input	**Fine aggregate**	0.675	t	0.675	t	Laveglia et al, 2023
	Lightweight aggregate	0.07	t	0.07	t	Modelled by Ecoinvent (pumice quarry operation, GLO)
	Transport	14	t*km	200	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.0003	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
	Artificial lightweight aggregate	0.005	t	0.005	t	Modelled by Ecoinvent (expanded perlite production, GLO)
	Transport	1	t*km	200	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6 | RoW)
	Endless screw conveyor	0.00002	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
	Hydrated lime (HL)	0.25	t	0.25	t	Laveglia et al, 2022 [10]
	Transport	25	t*km	100	km	Modelled by Ecoinvent (transport, lorry 16-32 metric ton, EURO6| RoW)
	Endless screw conveyor	0.001	kWh	0.004	kWh / t	Electricity mix (SUBLime designed)
	Dry mixer	4	kWh	4	kWh / t	Electricity mix (SUBLime designed)
Output	**Hydrated Lime Plaster (HLP)**	1.00	t	1.00	t	Output of the Plaster Manufacturing

Appendix 19. Design parameters of the carbon capture system (Aspen Hysys Simulation)

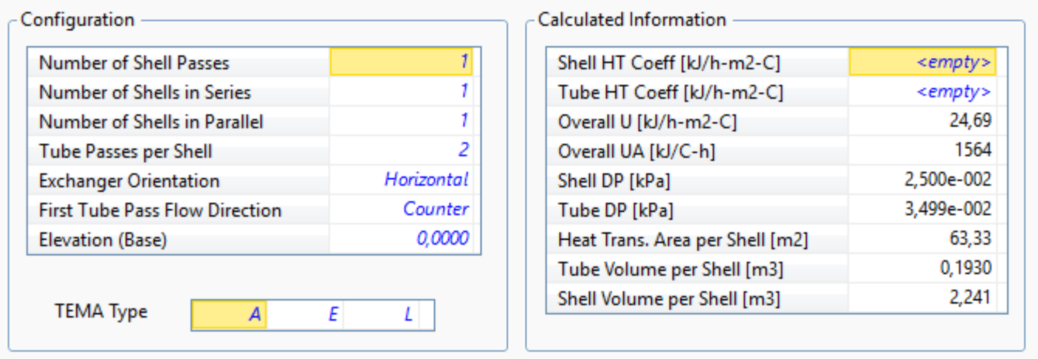
Appendix 19.1. CO_2 treatment design: Arrangement of heat exchangers



Appendix 19.2. Heat exchanger design



Tube Side Inlet: CO2_PostComb
Name: E-100
Shell Side Inlet: NaturalWater
Tube Side Outlet: CO2_Cooled
Shell Side Outlet: WaterVaport
Tube Side Fluid Pkg: Basis-1
Shell Side Fluid Pkg: Basis-1



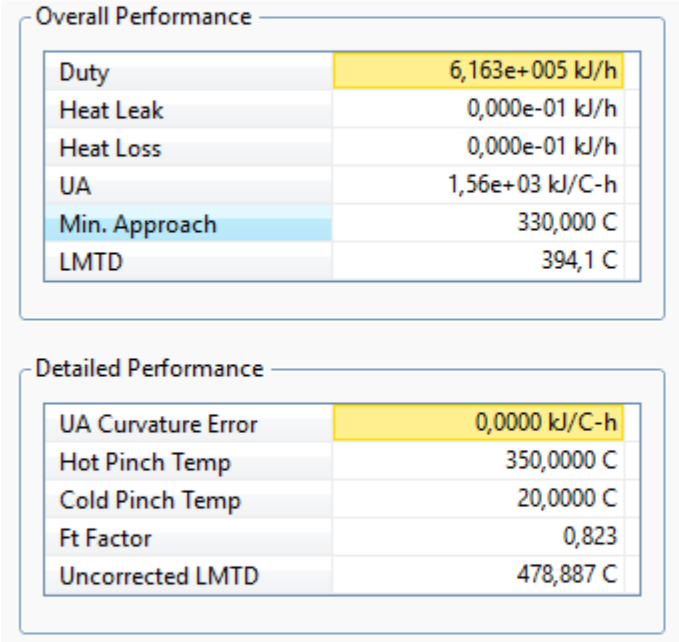
Configuration

Number of Shell Passes	1
Number of Shells in Series	1
Number of Shells in Parallel	1
Tube Passes per Shell	2
Exchanger Orientation	Horizontal
First Tube Pass Flow Direction	Counter
Elevation (Base)	0,0000

TEMA Type: A E L

Calculated Information

Shell HT Coeff [kJ/h-m2-C]	<empty>
Tube HT Coeff [kJ/h-m2-C]	<empty>
Overall U [kJ/h-m2-C]	24,69
Overall UA [kJ/C-h]	1564
Shell DP [kPa]	2,500e-002
Tube DP [kPa]	3,499e-002
Heat Trans. Area per Shell [m2]	63,33
Tube Volume per Shell [m3]	0,1930
Shell Volume per Shell [m3]	2,241



Overall Performance

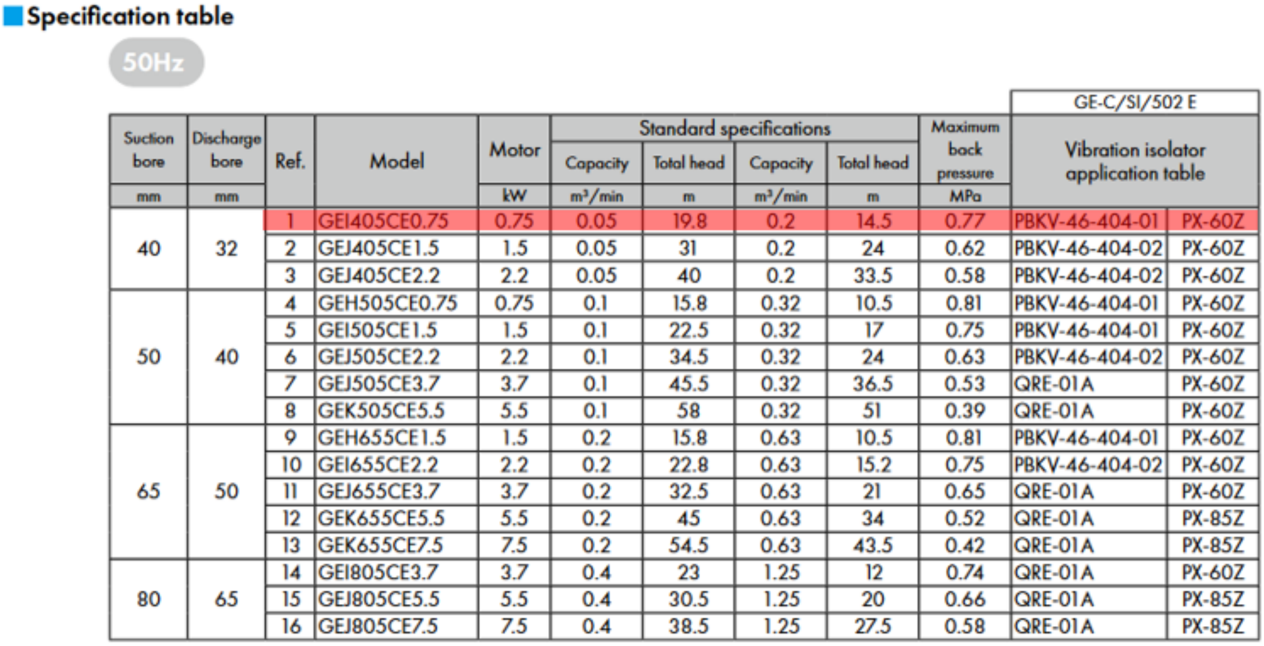
Duty	6,163e+005 kJ/h
Heat Leak	0,000e-01 kJ/h
Heat Loss	0,000e-01 kJ/h
UA	1,56e+03 kJ/C-h
Min. Approach	330,000 C
LMTD	394,1 C

Detailed Performance

UA Curvature Error	0,0000 kJ/C-h
Hot Pinch Temp	350,0000 C
Cold Pinch Temp	20,0000 C
Ft Factor	0,823
Uncorrected LMTD	478,887 C

Appendix 19.3. Pump requirement for the operation of the heat exchangers.

The electricity consumption of the heat exchanger is based on the pumping power required for the shell side. The pump is designed considering the pumping of natural water, at 20°C for 200 kg/h (3.3 L/m). https://www.kawamoto-global.com/web/data/ecatalog_u_pdf/2.pdf



Specification table

50Hz

GE-C/SI/502 E

Suction bore	Discharge bore	Ref.	Model	Motor	Standard specifications				Maximum back pressure	Vibration isolator application table	
					Capacity	Total head	Capacity	Total head			
mm	mm			kW	m³/min	m	m³/min	m	MPa		
40	32	1	GEI405CE0.75	0.75	0.05	19.8	0.2	14.5	0.77	PBKV-46-404-01	PX-60Z
		2	GEJ405CE1.5	1.5	0.05	31	0.2	24	0.62	PBKV-46-404-02	PX-60Z
		3	GEJ405CE2.2	2.2	0.05	40	0.2	33.5	0.58	PBKV-46-404-02	PX-60Z
50	40	4	GEH505CE0.75	0.75	0.1	15.8	0.32	10.5	0.81	PBKV-46-404-01	PX-60Z
		5	GEI505CE1.5	1.5	0.1	22.5	0.32	17	0.75	PBKV-46-404-01	PX-60Z
		6	GEJ505CE2.2	2.2	0.1	34.5	0.32	24	0.63	PBKV-46-404-02	PX-60Z
		7	GEJ505CE3.7	3.7	0.1	45.5	0.32	36.5	0.53	QRE-01A	PX-60Z
		8	GEK505CE5.5	5.5	0.1	58	0.32	51	0.39	QRE-01A	PX-60Z
65	50	9	GEH655CE1.5	1.5	0.2	15.8	0.63	10.5	0.81	PBKV-46-404-01	PX-60Z
		10	GEI655CE2.2	2.2	0.2	22.8	0.63	15.2	0.75	PBKV-46-404-02	PX-60Z
		11	GEJ655CE3.7	3.7	0.2	32.5	0.63	21	0.65	QRE-01A	PX-60Z
		12	GEK655CE5.5	5.5	0.2	45	0.63	34	0.52	QRE-01A	PX-85Z
		13	GEK655CE7.5	7.5	0.2	54.5	0.63	43.5	0.42	QRE-01A	PX-85Z
80	65	14	GEI805CE3.7	3.7	0.4	23	1.25	12	0.74	QRE-01A	PX-60Z
		15	GEJ805CE5.5	5.5	0.4	30.5	1.25	20	0.66	QRE-01A	PX-85Z
		16	GEJ805CE7.5	7.5	0.4	38.5	1.25	27.5	0.58	QRE-01A	PX-85Z

Appendix 20. Life-cycle cost inventory

Group		Item	Unit of measure	Unit costs
Purchase of Materials	**Binders**	Hydrated Lime	€/kg	0.11
	Aggregates	Sand	€/kg	0.01
		Pumice	€/kg	0.06
		Polystyrene	€/kg	1.29
	Additives	Carboxymethyl celullose	€/kg	0.97
		Alkylbenzene sulfonate	€/kg	0.80
		Polycarboxyllate	€/kg	0.65
		Ethylene vinyl acetate	€/kg	1.12
Others		Carbon tax	€/kg	0.09
Transportation		Truck transportation	€/t.km	0.06
Electricity consumption		Electricity	€/kWh	0.11
End of Life		Sanitary landfilling	€/kg	0.02

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Ghent University, Faculty of Engineering and Architecture (Belgium)

Thesis: "Towards Net-zero and Carbon-negative Transformations in Lime-based Construction Materials"

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Thesis: "Waste Glass Upcycling for the Manufacturing of Low CO_2 Cements"

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Marie Skłodowska-Curie Research Fellow - Early-Stage Researcher. European MSCA (ETN – ITN) SUBLime Consortium (Sustainable Building Lime Applications Via Circular Economy and Biomimetic Approaches). Technical University of Darmstadt and Ghent University. **Research Project:** "Cradle-to-Grave Sustainability of Novel Lime-based Materials"

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Scientific Dissemination

Research articles published in Q1 journals

Etcheverry, J.M., **Laveglia, A.**, Villagran-Zaccardi, Y.A., De Belie, N. (2024). A technical-environmental comparison of hybrid and blended slag cement-based recycled aggregate concrete tailored for optimal field performance. Developments in the Built Environment. https://doi.org/10.1016/j.dibe.2024.100370

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Laveglia, A., Trezza, M.A., Irassar, E.F. y Rahhal, V.F. (2022). Natural and artificial glassess as pozzolans in cementitious systems. AATH 2022 - X International Congress y 24° Technical Meeting of the Argentinean Association of Concrete Technology. Congress Proceedings. Buenos Aires, Argentina.

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Patent registration

Sambataro, L., **Laveglia, A.**, Ukrainczyk, N., Koenders, E. Carbon-Looping for Sand-Lime Brick manufacturing by Carbonation Curing in Autoclave. (26.09.2023). German Patent Office. Number: 102023126103.7

Scholarships and Prizes

Fully-founded scholarship to spend one month at Universities and Research Centers in Germany. German Academic Exchange Service (DAAD). September 2018.

Best scientific paper award to “Pozzolanic activity of waste glasses”. 6th Meeting of Young Researchers in Materials Science. Argentinean Society of Materials. Congress Proceedings. September 2017.