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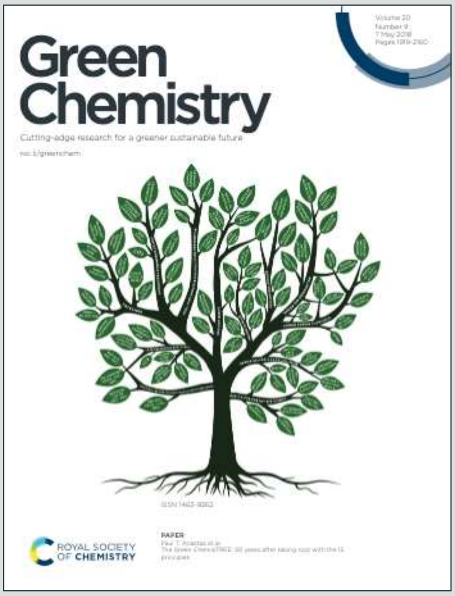

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From Quarry to Carbon Sink: Process-based LCA Modelling of Lime-based Construction Materials for Net-Zero and Carbon-Negative Transformation

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A comprehensive decarbonization approach is reported, involving Direct Separation Reactors (DSR) 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. Integrating a DSR kiln with carbon capture technologies (CCT) attained a remarkable 65% reduction of CO_2 emissions during hydrated lime production, with a minimum environmental impact from the CCT itself. Fully electrified DSR kilns, powered by renewable energy sources, achieve an astonishing 94% decrease in CO_2 emissions when compared to conventional reference scenarios, all without adverse environmental effects. In lime-based plasters, combining DSR kilns, natural carbonation, and eco-efficient energy sources, particularly with inclusion of natural gas, leads to carbon negativity. This efficiently offsets all production emissions and even cuts back an additional 30%. In the case of fully electrified DSR kilns, the results are a remarkable 149% CO_2 emissions reduction throughout the entire cradle-to-grave lifecycle. Carbon capture technologies reduce carbon tax costs by up to 26%, thereby enhancing the economic sustainability of these endeavours. 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.

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 1 (adapted from [10]). Supplementary Information 1 (SP1) provides a

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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.

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. 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].

Figure 1. Launched European/UK projects on decarbonization of the lime industry (adapted from [10]).

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. 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 Table SP1). 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 commodity* 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 potentially 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 was 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 was 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 was 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 Marie Skłodowska-Curie Actions ITN-ETN network (Innovative Training Network, European Training Network) [25]. This initiative has successfully brought together Europe's top-tier Universities, the European Lime Association, and major lime-based materials manufacturers to collaboratively devise strategies for achieving net-zero emissions and design the new generation of lime-based materials. 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

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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 (LBP). 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.

2. Research Methodology

2.1 Definition of the case studies

This study aims to assess the environmental impact 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 2, comprise the production of all LBP components (hydrated lime, different types of aggregates and additives), along with its use and end-of-life phase.

Figure 2. System boundaries for the study, with emphasis on various lime manufacturing scenarios during the cradle-to-gate 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 (specific datasets provided in Supplementary Table SP2) [11]. Kiln technology is on a comparison between the widely-used PFRK [26] and the Direct Separator Reactor designed by LEILAC [17]. To analyse the impact of these combinations on the overall environmental footprint of LBP production, four distinct scenarios are selected and presented (Figure 3).

Figure 3. Definition of the scenario analysis applied to the cradle-to-gate hydrated lime manufacturing.

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.

The environmental analysis of the LBP production is linked to a previously studied real plant, where aggregates and additives are transported to the site and dry mixed [8]. Power sources for the mixing plant align with the electricity sources used in each scenario. The plaster is then 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 3.1.2), 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.

2.2 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. 2) in the LCI. For more detailed information, reference is made to [3].

2.3 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 2.2). The calculations are performed from a producer's perspective. For production costs the purchase price

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of materials, resources, and energy were considered (See Section 3.1.2) [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 3.1.2.

3. Results and discussion

3.1 Life-cycle inventory

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 is available as Supplementary Information SP3. In this section though, focus is on the hydrated lime production, where Figure 3 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 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 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, moving also 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 1. Technical key parameters of PFRK and DST kiln technologies for scenario analysis

The technical data employed for the analysis of the novel DSR design (so-called LEILAC) is less disseminated. It is worth mentioning that two partners of the SUBLime network, the Belgian lime producer Lhoist and the British lime company Tarmac, worked also in the development of this technology. As shown in Figure 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 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 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 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 2), the inventory is based on a previous work by the authors [3].

In the case of 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 3). 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]-[33]. 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.

Table 2. Inventory of the calcination operation using PFRK kiln technology (based on[3]).

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.

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The DSR inventory is implemented in Table 3 and the design parameters of the carbon capture system can be found in the Supplementary Information (Table SP4). 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.

Table 3. Inventory of combined calcination and CO_2 recovery for lime production with DSR.

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 1).

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \qquad \text{(Reaction 1)}$$

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 1). Equation 2 is employed to calculate the kg of CO_2 sequestrated per functional unit at a given time.

$$x = k\sqrt{t} \qquad \text{Equation 1}$$

$$SC = 0.594 * FCH * (x/X_{total}) \qquad \text{Equation 2}$$

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 $CO2/Ca(OH)_2$), 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. 1) is reached. The adopted parameters as well as calculations performed are shown in Table 4. The coefficient k is an average value reported by [42]–[44].

Table 4. Parameters considered for the carbonation of the plasters during the use phase.

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 4) and the service life (50 years) according to the diffusion model (Equation 1) [45]. After this time, the old (discarded) plaster is transported over 100 km to its final disposal site (see Table SP3). 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 Supplementary Information (Table SP5).

3.2. Environmental impact assessment

In this section first the cradle-to-gate scenario is analysed (Figure 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.

3.2.1 Manufacturing of hydrated lime

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 4 and 5 for electricity and fuel sources, respectively.

Figure 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 of energy. *DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction*

Figure 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*

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 3a-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

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low CO_2 emissions as provided in Figure 3d. 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 $CO_{2\ eq}$. [46].

The production of thermal energy is dominated by coal combustion with all impact indicators ranging between 50 to 85% of the endpoint categories (Figure 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 [47]. 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. 5c), the following reductions per MJ of energy are recorded: 20% for Resources, 10% for Ecosystem Quality and 60% for Climate Change (Fig 5a, 5b, 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 [48].

Retrospectively, when comparing the environmental impacts per MJ of energy, the Figures 3 and 4 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. 4d and 5d).

Figure 6 summarizes the results of the scenario analysis (Fig. 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 6a-c, the Ratio of each scenario (Si, i=1, 2, 3,…) compared to the `business as usual` reference case (S1) is shown. Figure 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.

Figure 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: Ecoefficient fuels, IT: Improved kiln technology, Hybrid: Operation by e-fuels and electricity. DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction*

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 3). As a decarbonization strategy, the use of eco-efficient fuel sources is a first step towards reduced CO_2 emissions (Figure 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 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 $tCO_2/tCa(OH)_2$, reducing the total emissions by 65% (S1 Vs. S3, Fig. 6d).

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. 3 and Fig. 4). The scenario with the highest decarbonization potential is S4C, in which around 52 $kgCO_2/tCa(OH)_2$ 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 lifecycle.

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3.2.2 Cradle-to-grave environmental impact assessment

With section 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 2).

Figure 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 [8]. It produces both the hydrated lime and aggregates and receives the remaining components through transportation from other locations (See Table SP3). 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 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. Endpoint impact indicators of the scenario analysis for the cradle-to-grave system boundaries (FU is 1 m^2 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. 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%) (See Supplementary Information, Table SP3). 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 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. 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. 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*yr/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. 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 several 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. 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 and shown in Figure 4c.

In the Climate Change endpoint category (Fig. 7d), the business-as-usual manufacturing scenario (S1) leads to the release of around 3 kg CO2eq/FU, being for around 85% dominated by hydrated lime production, because of the process emissions and fuel combustion (Fig. 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 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 5d explains these results in terms of a lower CO_2 intensity of natural gas compared to coal.

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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 (See Supplementary Information, Table SP1). The carbon capture system designed at the plant scale in this paper (Table SP4) offers a viable solution for managing CO_2 emissions. Both the inventory (Table 3) and impact assessment (Fig. 7) reveal that the additional devices integrated into the DSR kiln have a negligible environmental impact compared to the overall benefits they generate.

Figure 2 illustrates a series of scenarios, with S4 emerging as the most promising path toward achieving 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 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 3.4.

3.3 Economic impact assessment

Figure 8 shows the results of the total costs (€/FU) for each cradle-to-grave scenario provided in Figure 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 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.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 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 6d). In scenario S3, the specific contribution of carbon taxes is around 21%, leading to a 16% reduction in total costs, because of 25% CO2 reduction in hydrated lime manufacturing (compared to S1, Fig. 7d). A further reduction of emissions by electrifying the DSR leads also to a 23% cost reduction compared to the reference 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 $CaCO_3$ [49], 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 3.4.

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. 9). The most effective approach involves a coordinated action plan that can harmonize the needs of the private sector, environmental protection, and societal well-being, all within a regulatory framework that ensures equitable implementation of political decisions. In the following paragraphs, the current state, primary challenges, and the necessary actions across these different analytical levels are

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considered to achieve a fast decarbonization of the lime industry.

Figure 9. Scheme showing the interaction of all relevant stakeholders required to achieve a fast decarbonization of the lime and cement industry.

3.4.1 Manufacturing level

At the manufacturing level, producers hold a critical decision-making level: 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. 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 [50]. This shift also leads to a 16% reduction in production costs because of a reduced amount of carbon taxes paid (S3, Fig. 8). Future research should thoroughly compare current kiln technologies (refurbishment vs. new installation) and assess the potential effect of renewable energy technology costs.

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 [51]. However, realizing this potential requires legislative support and market development, as we will explore later in Sections 3.4.2 and 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 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 [52]. 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 [46].

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, 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. 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 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].

The recently introduced fit-for-55 package has incorporated updated carbon prices and incentives for carbon capture utilization, in the framework of the ETS [56]. Over the past five years, carbon prices have surged, rising nine-fold and exceeding 90 €/tCO_2 [57]. As discussed in Section 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 and protect thousands of jobs [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]. 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 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. 7d). The natural carbon sink offered by lime-based materials should be considered when developing policies and taxation coefficients for regulations.

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.

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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 10. This comprehensive approach is key to making significant steps forward in carbon reduction efforts.

Figure 10. CO_2 value chain starting from the emission source (i.e., the cement/lime plant). The Figure highlights the approach of CO2 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], [60], [61]. To achieve this, more research is essential in the field of construction materials, particularly in areas such as CO2 curing of concrete, industrial carbonation of recycled aggregates, and brick production[62] . 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.

CO_2 has the potential to be used as a resource in the production of polymers and energy storage, among others [63], [64]. 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 [51]. 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. Effective cooperation among stakeholders is crucial to achieving 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 [65]. 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?

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:

1. 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 lifecycle.
2. 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.
3. Environmental benefits throughout: Ecosystem quality, resources, and human health indicators remain largely unaffected by these decarbonization efforts.
4. Economic sustainability: Carbon capture technologies not only benefit the environment but also enhance economic sustainability, lowering carbon taxes by up to 23%.
5. 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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Author Contributions

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.

Conflicts of interest

We confirm that there are no conflicts to declare.

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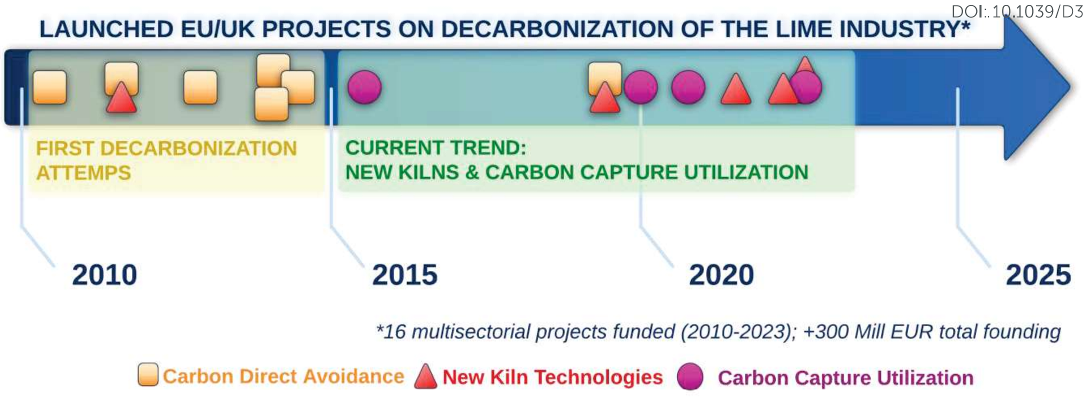

Figure 1. Launched European/UK projects on decarbonization of the lime industry (adapted from [10]).

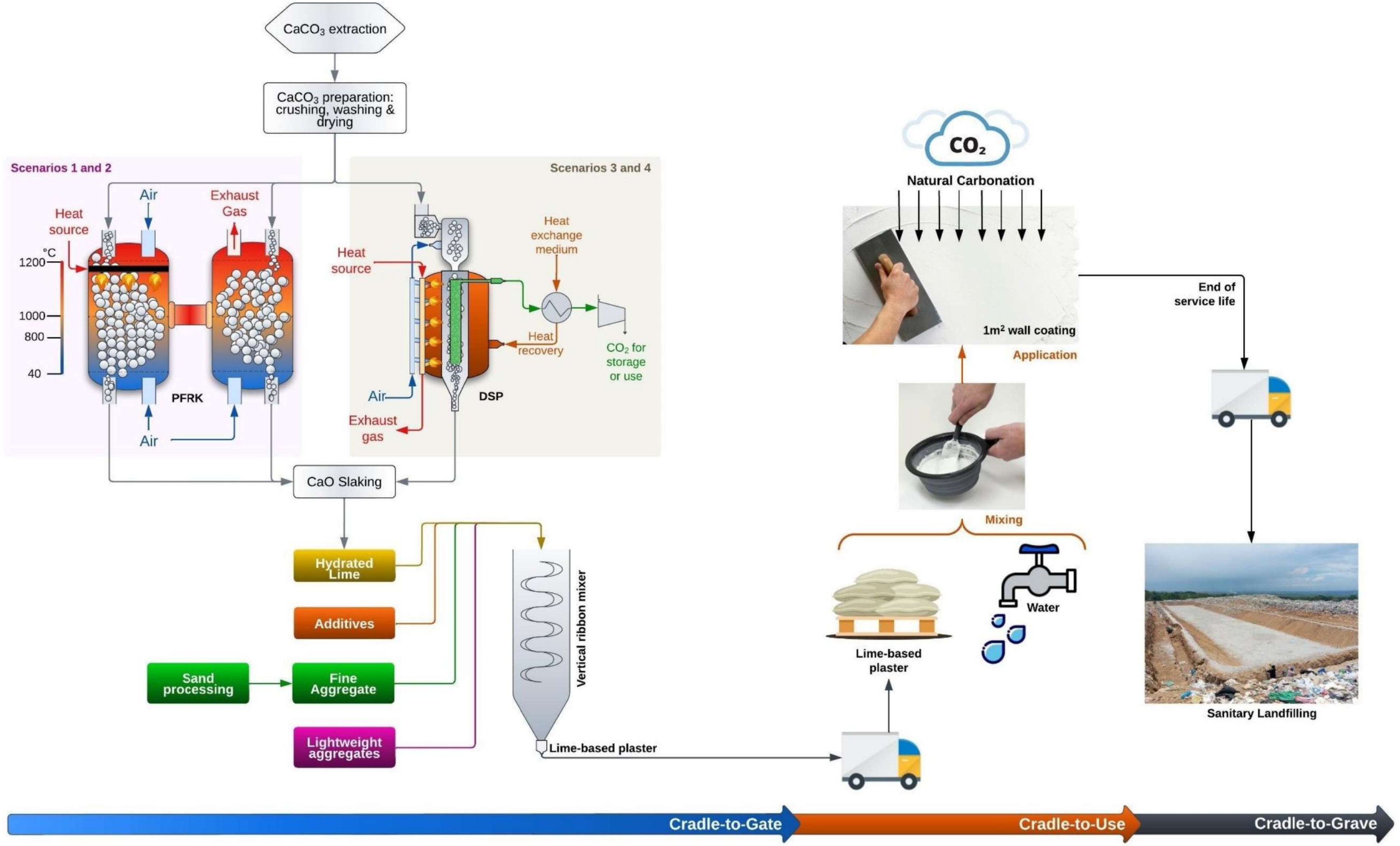

Figure 2. System boundaries for the study, with emphasis on various lime manufacturing scenarios during the cradle-to-gate phase.

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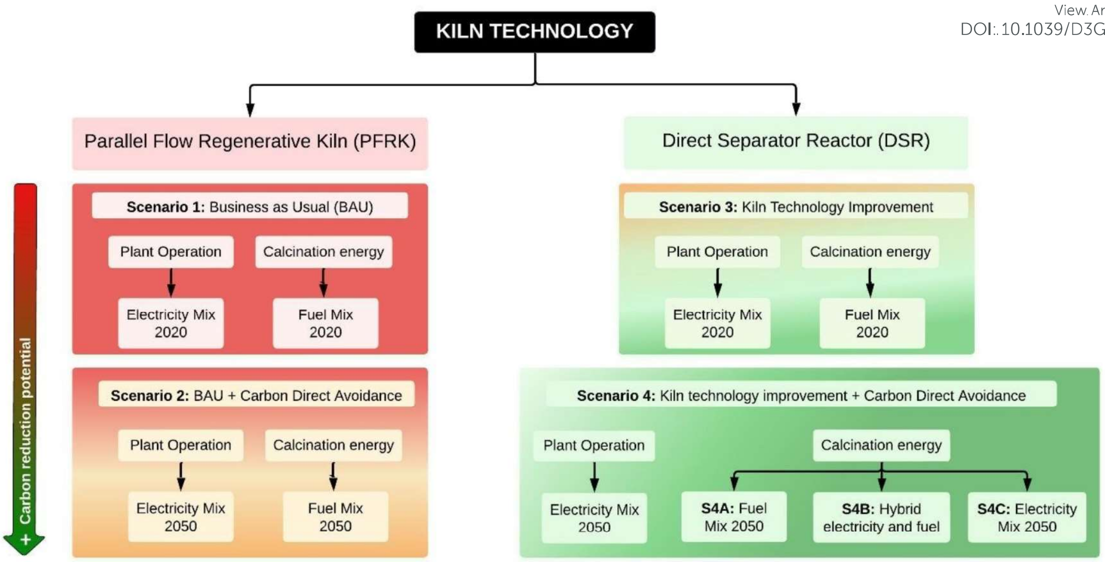

Figure 3. Definition of the scenario analysis applied to the cradle-to-gate hydrated lime manufacturing.

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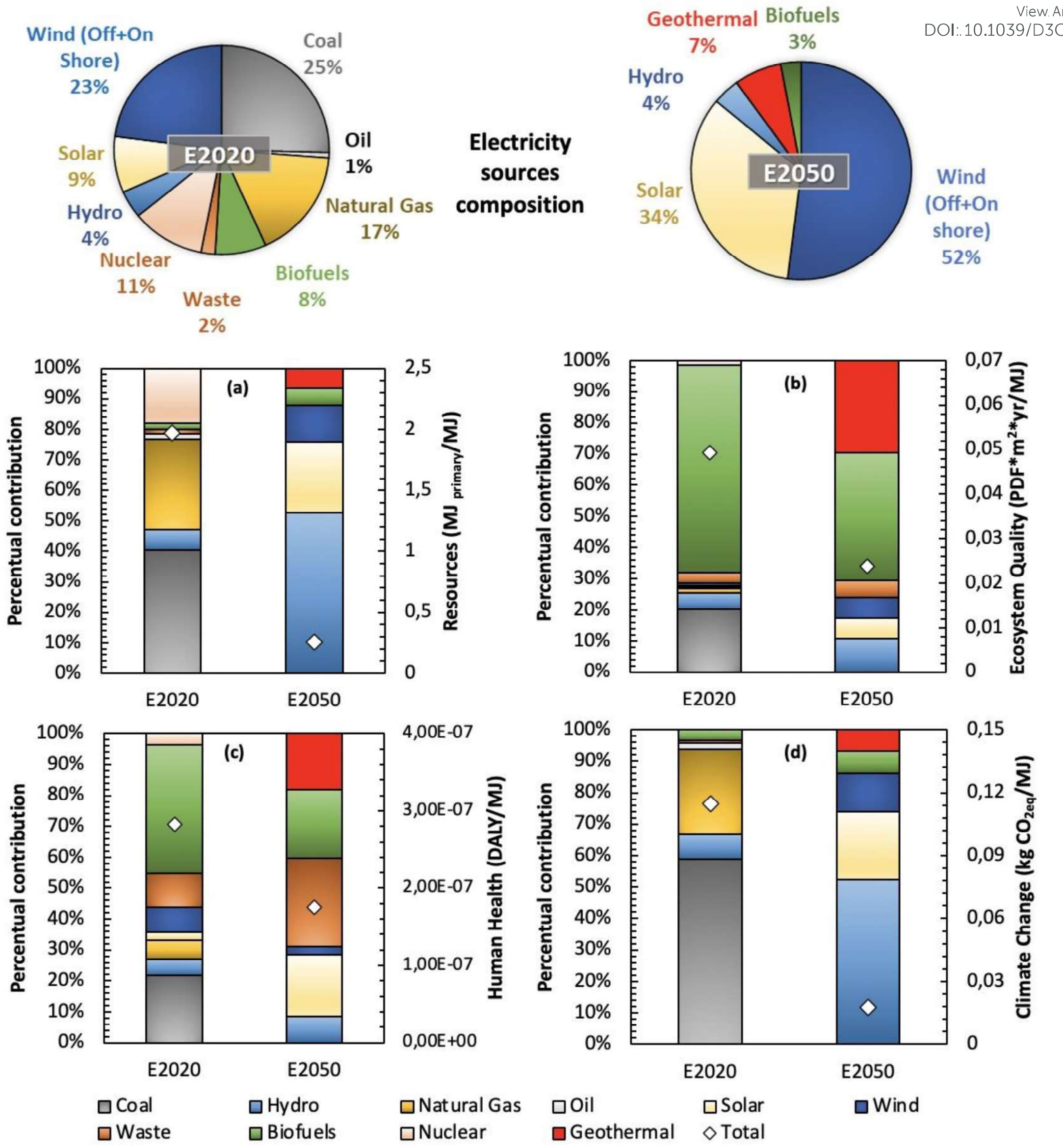

Figure 4. Electricity sources composition used to model current (2020) and future scenarios (2050) according to[11]. Figures a to d show the endpoint indicators with percentual contribution (left axis) and total impact (right axis) of components to produce 1 MJ of energy. *DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction*

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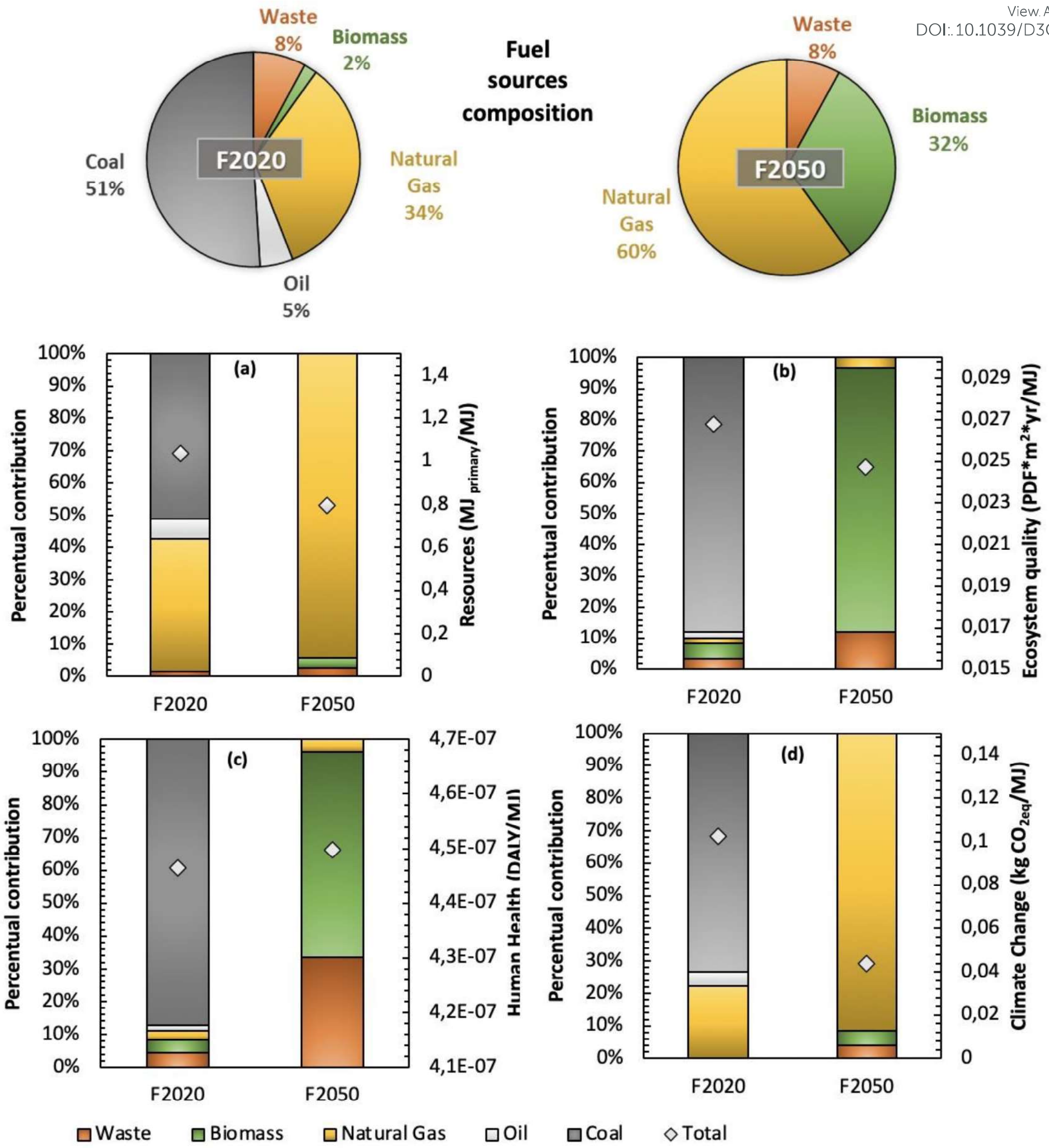

Figure 5. Fuel sources composition used to model current (2020) and future scenarios (2050) according to [11]. Figures a to d show the endpoint indicators and percentual contribution of each fuel source to produce 1 MJ of energy. *DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction*

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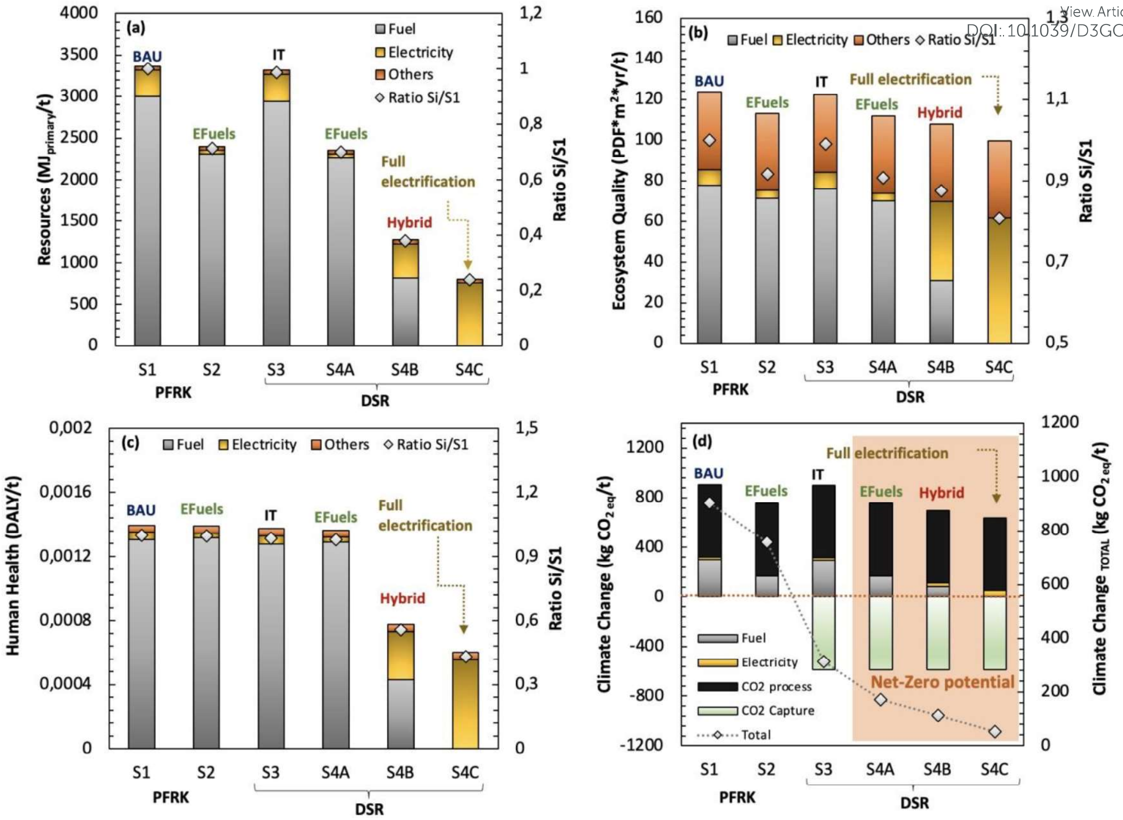

Figure 6. Endpoint impact indicators of the scenario analysis for the cradle-to-gate system boundaries to produce 1 ton of hydrated lime. Figures a-c show the ratio of each scenario with respect to the reference S1. Figure d shows the total effect on the climate change indicator according to each technology and energy sources. *BAU: business-as-usual, E-Fuels: Ecoefficient fuels, IT: Improved kiln technology, Hybrid: Operation by e-fuels and electricity. DALY: Disability Adjusted Life Years, PDF: Potentially Disappeared Fraction*

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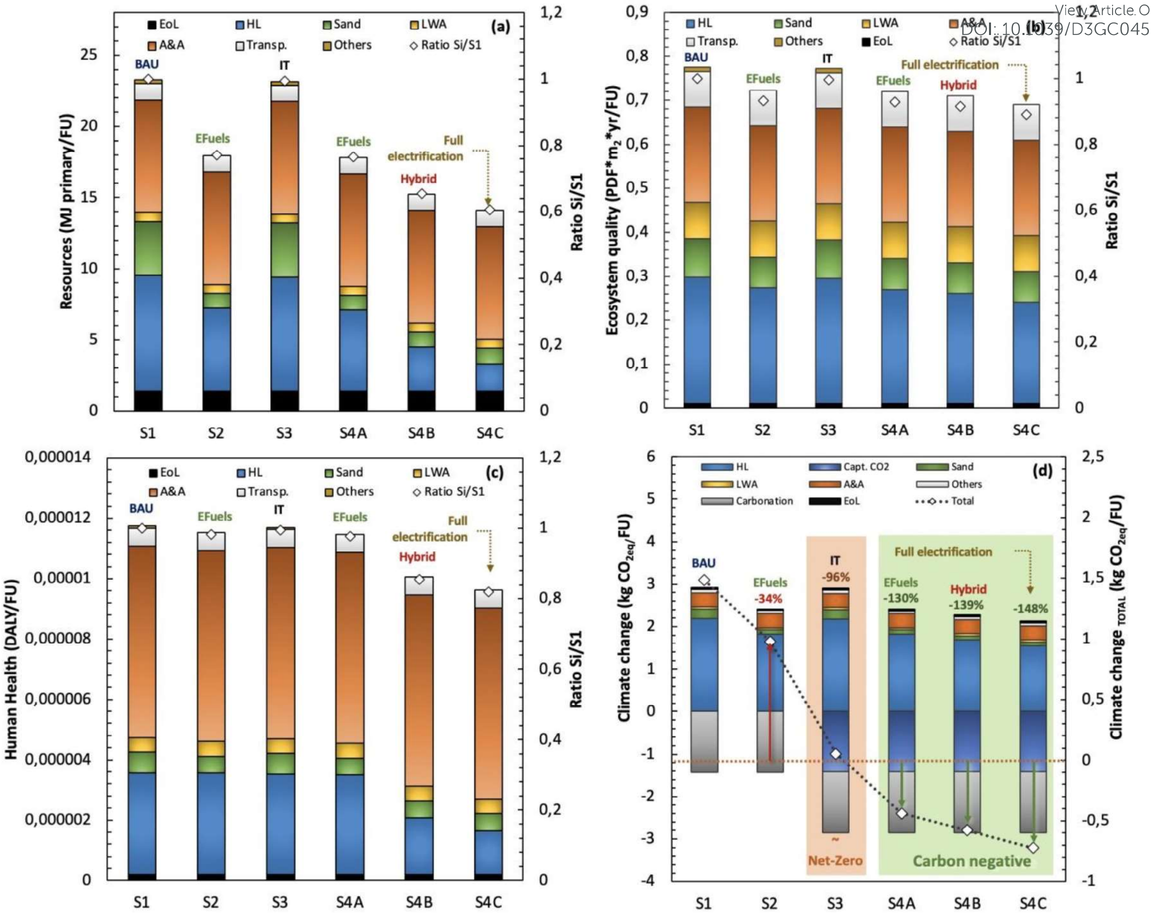

Figure 7. Endpoint impact indicators of the scenario analysis for the cradle-to-grave system boundaries (FU is 1 m² wall coating). Figures a-c show the ratio of each scenario with respect to the reference S1. Figure d shows the total effect on the climate change indicator according to each technology 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*

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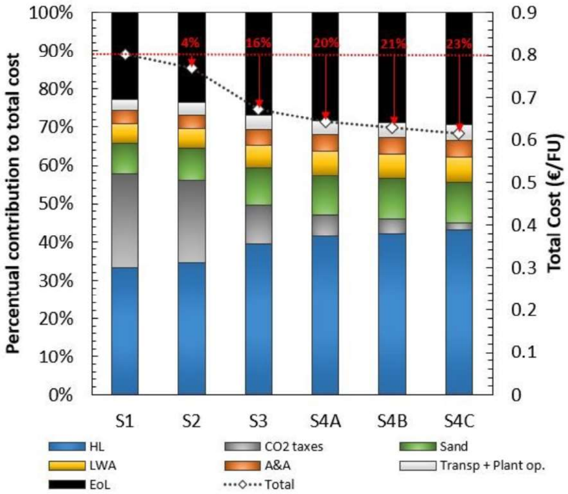

Figure 8. Life-cycle cost assessment results from cradle-to-grave of the decarbonization strategies, considering the effect of carbon taxes. *HL: Hydrated lime, LWA: Lightweight aggregate, A&A: Additives and aids, EoL: End of Life.*

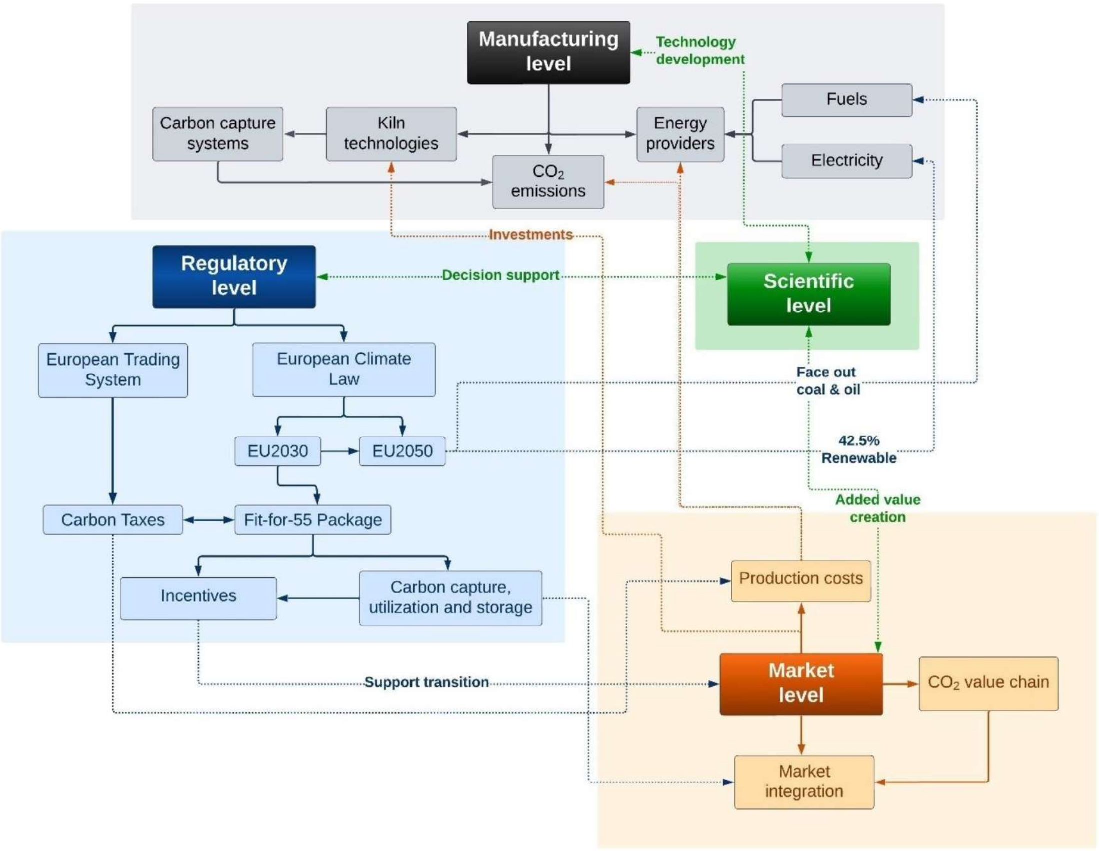

Figure 9. Scheme showing the interaction of all relevant stakeholders required to achieve a fast decarbonization of the lime and cement industry.

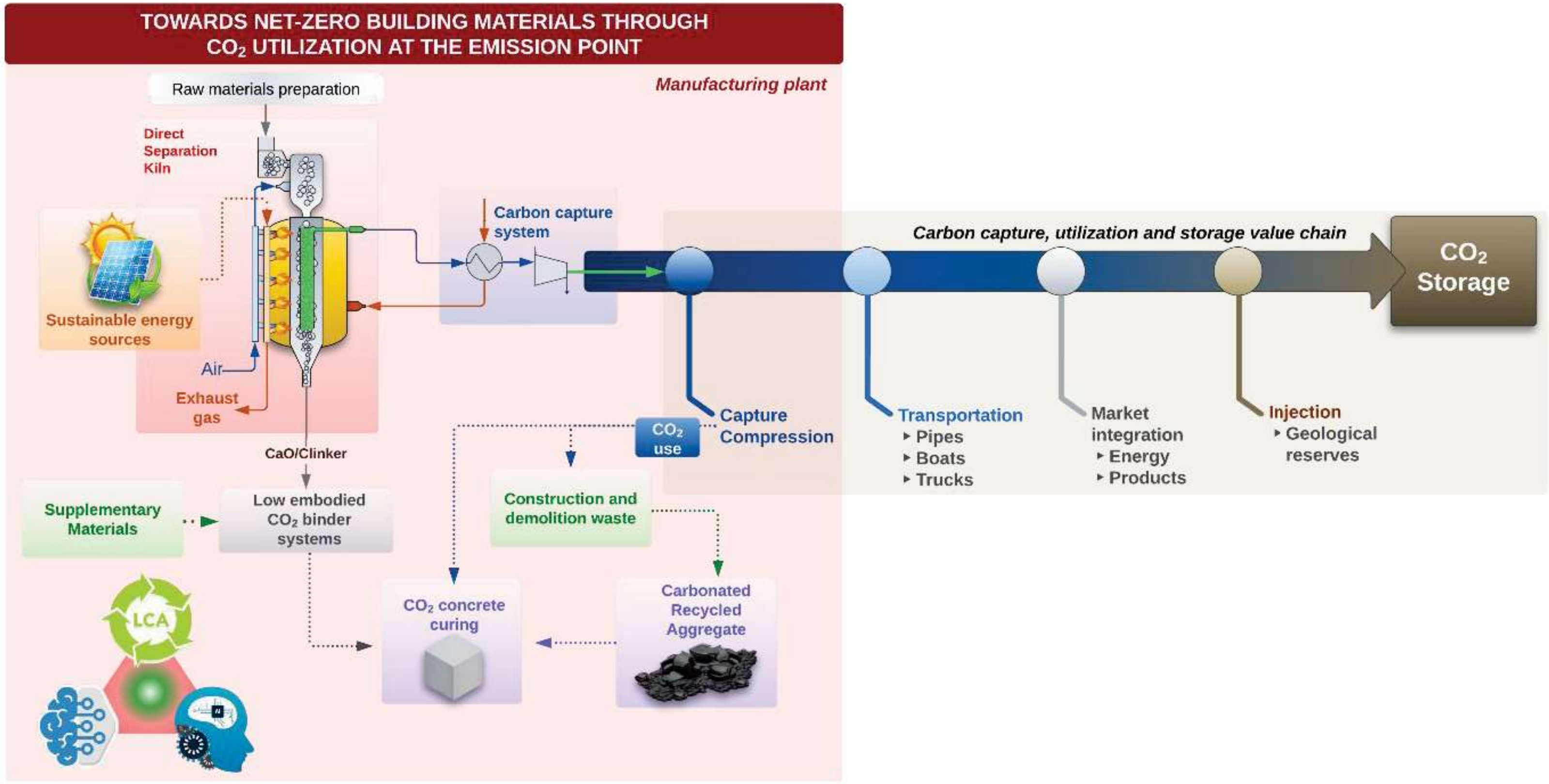

Figure 10. CO_2 value chain starting from the emission source (i.e., the cement/lime plant). The Figure highlights the approach of CO2 use at the emission point to produce net-zero building materials

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Table 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

Table 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 1
		Electricity consumption	30	kW	Table 1
	OPERATION	**FLOW (mass/energy)**	**AMOUNT**	**UNIT**	
OUTPUT	**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		

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Table 3. Inventory of combined calcination and CO_2 recovery for lime production with DSR.

	OPERATION	FLOW (mass/energy)	PROCESSED AMOUNT		COMMENTS
			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 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		

Table 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