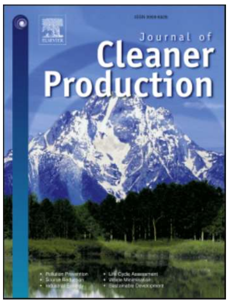

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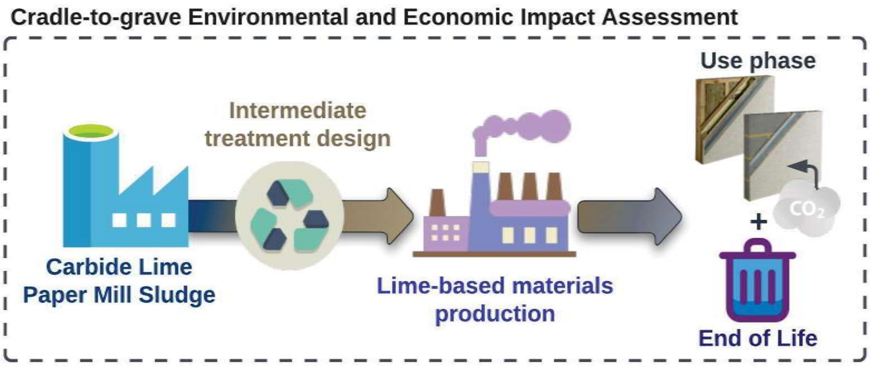
Cradle-to-grave Environmental and Economic Impact Assessment
Intermediate treatment design
Use phase
CO₂
+
Carbide Lime
Paper Mill Sludge
Lime-based materials production
End of Life

Cradle-to-Grave Environmental and Economic Sustainability of Lime-Based Plasters Manufactured with Upcycled Materials

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ABSTRACT

The production of CaO for lime-based plaster and render generates 1.2 t CO_2/t CaO, consumes 1.78 t $CaCO_3$/t CaO. This research paper examines the environmental and economic performance of upcycling paper mill sludge (PMS) and carbide lime (CL) as replacements for hydrated lime (HL) in lime-based plasters production. For this, a new Cradle-to-Gate industrial-scale inventory is designed, upscaling recent lab-scale innovations, investigating PMS and CL treatment processes, followed by a Cradle-to-Grave scenario analysis. The results show that incorporating CL in the plaster yields better environmental and economic outcomes compared to PMS. The intermediate treatment for CL is cost-effective and has low carbon emissions. The upcycling of CL eliminates 100% of CO_2 emissions, while PMS reduces emissions by 11%. The production of the traditional binder HL is more expensive than upcycling PMS (+69%) and CL (+65%), with carbon taxes accounting for 35%, 44% and 15% of production costs, respectively. The effect of an equilibrated carbon price to ensure fair market competition, considering the natural carbonation of lime (carbon credit) is discussed, and the cost assessment reveals a 47% and 54% reduction for upcycled plasters using PMS and CL, respectively, compared to traditional HL.

Keywords: circular economy; lime-based materials; carbide lime; paper mill sludge; life-cycle assessment; life-cycle cost

Highlights

- Availability, properties, and treatments of secondary lime resources
- Industrial-scale inventory for upcycling carbide lime (CL) & paper sludge (PS)
- Cradle-to-Grave assessment: CL reduces CO_2 most, trailed by PS and hydrated lime
- Key factors for Cradle-to-Grave impact: lime kiln heat consumption (PS) and drying operation (CL)
- Cost analysis of carbon taxes and credits across the material's life-cycle

Abbreviations

CE	Circular economy
CL	Carbide Lime
CMC	Carboxymethyl cellulose
DA	Dispersion Agent
ETS	European Trading System
EVA	Ethylene vinyl acetate
FU	Functional Unit
HL	Hydrated Lime
HLP	Hydrated Lime Plaster
LBM	Lime-based materials
LCA	Life-cycle Assessment
LCC	Life-cycle Cost
LCI	Life Cycle Inventory
LWA	Lightweight Aggregate
NIST	National Institute of Standards and Technology
PMS	Paper Mill Sludge
RCL	Recycled Carbide Lime
RCL (Ec.)	RCL with economic allocation
RCLBP	Recycled Carbide Lime Based Plaster
RPM	Recycled Paper Mill
RPMBP	Recycled Paper mill Based Plaster
SC	Sensitivity Coefficient
SR	Secondary resources
SETAC	Society of Environmental Toxicology and Chemistry
WRA	Water Retention Agent

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.

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 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].

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 intension 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 (min. 0.01 m^2/kW and 100 years). 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 1. In the case of secondary resources, the boundaries are covered from Cradle-to-Gate. Specific details on each case are provided in Section 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.

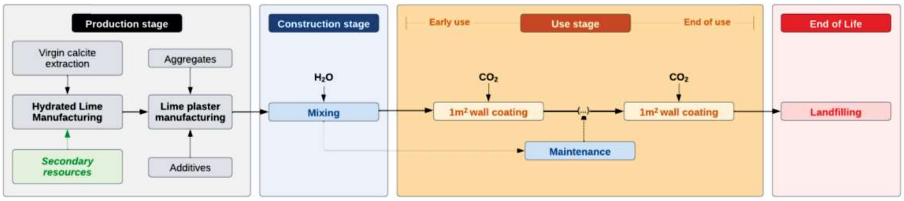


Figure 1. System boundaries considered in the Cradle-to-Grave environmental and economic assessment of lime-based plasters

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 present, 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 1 and 2 can be calculated correspondingly. In these equations, m and € represent the mass and price of main and by-products. In the identified Cradle-to-Gate system boundary of the secondary resource, the C_m and C_e coefficients are applied to the by-product of interest to assign them a portion of the environmental impact of the multifunctional process.

$$C_m = \frac{m_{by-product}}{m_{by-product} + m_{main\ product}}$$ **Equation 1**

$$C_e = \frac{(m \cdot \epsilon)_{by-product}}{(m \cdot \epsilon)_{by-product} + (m \cdot \epsilon)_{main\ product}}$$ **Equation 2**

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.

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. For the calculation of the endpoint categories, the midpoint indicators presented in Supplementary Information (Appendix 1) 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 3, 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.

$$SC = \frac{\frac{\Delta result}{initial\ result}}{\frac{\Delta parameter}{initial\ parameter}}$$ **Equation 3**

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].

Regarding economic impact assessment, the LCC methodology is not standardized as LCA and therefore, there is no unified procedure for calculating the costs [38]. To carry out this analysis, the starting point was the inventory of materials and energy considered for the environmental analysis (Section 2.2). 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 [29]. 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].

3. Results and discussion

3.1 Life Cycle Inventory

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 and energy sources are shown in Appendix 2 and 3 respectively. 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 Appendix 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 2. For the case studies, a full replacement of HL in the inventory of Appendix 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].

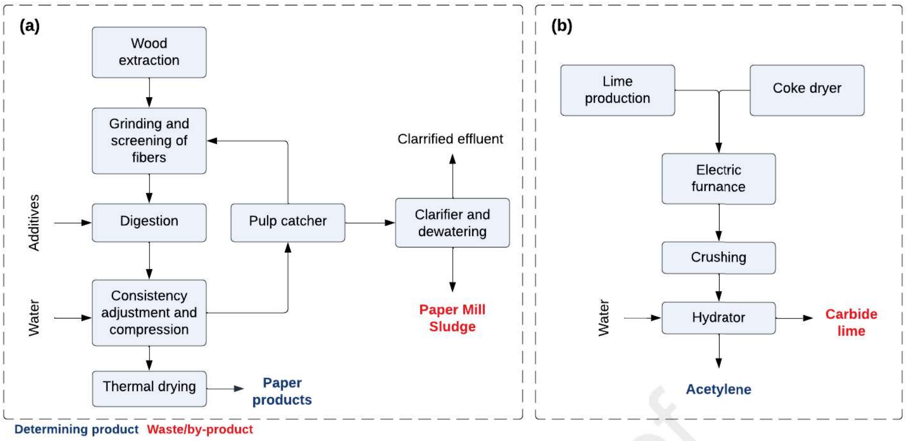


Figure 2. Production system of paper products and paper mill sludge (a) and production system of acetylene and carbide lime (b).

The production of paper (Figure 2a) involves a high consumption of energy, water, resources and generates a significant environmental impact [35]. 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 1b) [43, 44]. Calcium carbide is produced from calcium oxide and coke (Reaction 1) in an electric furnace at 2300°C. Then the calcium carbide is mixed with water to produce acetylene and CL, which is mainly composed of calcium hydroxide, as represented in Reaction 2 [41].

$$CaO\ (s) + 3C\ (s) \rightarrow CaC_2\ (s) + CO\ (g) \qquad \textbf{Reaction 1}$$

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

The CL by-product is generated as an aqueous slurry and is composed essentially 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. 1) and allocation by economic value (Eq. 2). The allocation coefficients considered for carbide lime production are shown in Table 1. Market prices are the result of averaged values collected in 2022 through market survey.

Table 1. 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

B. Use of secondary resources in lime-based plasters manufacturing

Table 2 summarizes a literature review of chemical composition ranges along physical properties and a comparison of different criteria for HL, PMS and CL.

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 largely of Ca(OH)2 (80%), could directly replace portlandite in HL production. The intermediate treatment strategies for PMS and CL are outlined below.

Table 2. 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. Mainly 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 mainly from the acetylene industry. The market is dominated by China (+26 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.	[37], [43]
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+})	-
Possible problems that can limit the application		Presence of organic compounds and variability of the chemical composition.	Presence of sulfides, organic compounds, and heavy metals.	-

- ***Intermediate treatment design of PMS***

Figure 2 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 2) 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 700-800°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].

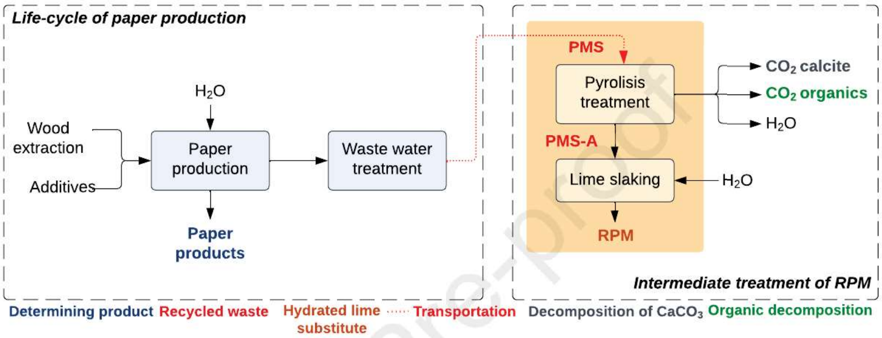


Figure 3. System boundaries to produce a recycled lime-based binder from paper mill sludge. *PMS: Paper Mill Sludge. PMS: Paper Mill Sludge; PMS-A: Paper Mill Sludge Activated; RPM: Recycled Paper Mill Sludge*

The main emissions in the lime kiln are water vapor and carbon dioxide from calcite decomposition and organic matter, calculated stoichiometrically (Reaction 3 and 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).

$$CaCO_3\ (s) \rightarrow CaO\ (s) + CO_2\ (g) \qquad \Delta H_{rC}{}^{\circ} = 177.3\ kJ/mol \tag*{\textbf{Reaction 3}}$$

$$C\ (s) + O_2(g) \rightarrow CO_2(g) \qquad \Delta H_{rC}{}^{\circ} = -393.5\ kJ/mol \tag*{\textbf{Reaction 4}}$$

The lime slaking is modelled following a previous work by the authors [25] and the LCI of the intermediate treatment is provided in Table 3. After the intermediate treatment, the final composition of the PMS is 80% $Ca(OH)_2$, 10% SiO_2, 5% Al_2O_3, 5% MgO.

- ***Intermediate treatment design of CL***

The intermediate treatment for carbide lime upcycling is shown in Figure 3, 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].

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).

Table 3. LCI 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 [25]
	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 4, do not count as emissions)
	CO_2 (calcite decomposition)	0.47	t	0.58	t CO_2/tPMS-A	Calculated stoichiometrically (Reaction 1)
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

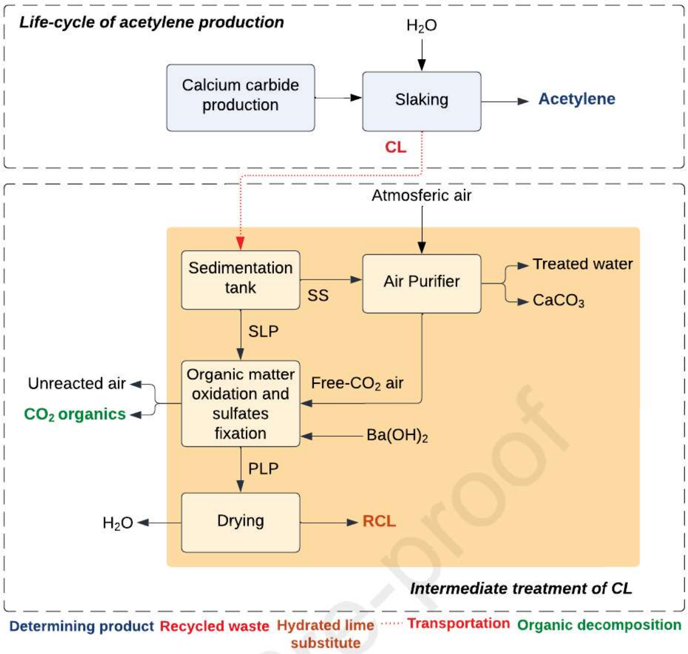


Figure 4. System boundaries to produce a recycled lime-based binder from carbide lime. CL: Carbide Lime; SS: Saturated Solution; SLP: Solid Lime Paste; PLP: Purified Lime Paste; RCL: Recycled Carbide Lime

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 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.

$$S^{2-} + 2O_2 \rightarrow SO_4^{2-}$$ **Reaction 5**

$$Ba^{2+} + SO_4^{2-} \rightarrow BaSO_4$$ **Reaction 6**

$$Ba^{2+} + 2(OH)^{-} \rightarrow Ba(OH)_2$$ **Reaction 7**

The final step of the treatment requires drying the paste. The LCI of the RCL production is shown in Table 4. After the intermediate treatment, the final composition of RCL is 89.5% $Ca(OH)_2$ and 10.5% $CaCO_3$.

Table 4. LCI 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 organic
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 S4). For the environmental and economic calculations, it is supposed that all plasters exhibit a comparable thermal performance according to the functional unit defined in Section 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% range in the dry mix of commercially available lime-based plasters [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 may increase the porosity in the matrix, which does not negatively affect the thermal insulation properties. 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 (Appendix 4) 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 needed in the future.

3.1.2 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 8).

$$Ca(OH)_2(aq) + CO_2(g) \rightarrow CaCO_3(s) + H_2O(l) \qquad \textbf{Reaction 8}$$

The diffusion of the CO_2 through the plaster (i.e., CO_2 sequestration) can be simplified by a diffusion-like process (Equation 4). Equation 5 is used to calculate the kg of CO_2 sequestrated per functional unit.

$$x = k\sqrt{t} \qquad \textbf{Equation 4}$$

$$SC = 0.594 * FCH * \frac{x}{X_{total}} \qquad \textbf{Equation 5}$$

where **x** (mm) is the carbonated thickness of the plaster at a given time ***t*** (days), ***k*** (mm/day0.5) 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. 4) is reached. The adopted parameters are shown in Table 5. The k coefficient is an average of values reported by [18], [20].

Table 5. 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)**	**SC (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

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.

3.1.3 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.
- Electricity consumption at the mixing plant.

Specific cost breakdowns per unit for these items are outlined in Table 6, 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 6. 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 6) by the same carbon price.

3.2 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.

3.2.1 Cradle-to-Gate of the factory

Figure 5 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. 4 (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]).

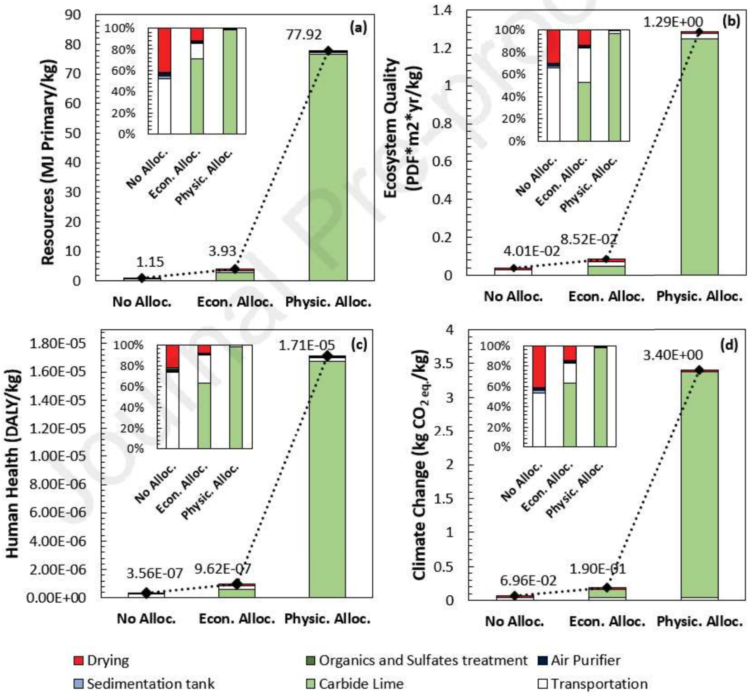


Figure 5. Cradle-to-Gate LCA results of producing 1 kg of RCL with and without application of allocation coefficients. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) endpoint indicator values and percentual contribution of each production step.

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 (Table 2). 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 6 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).

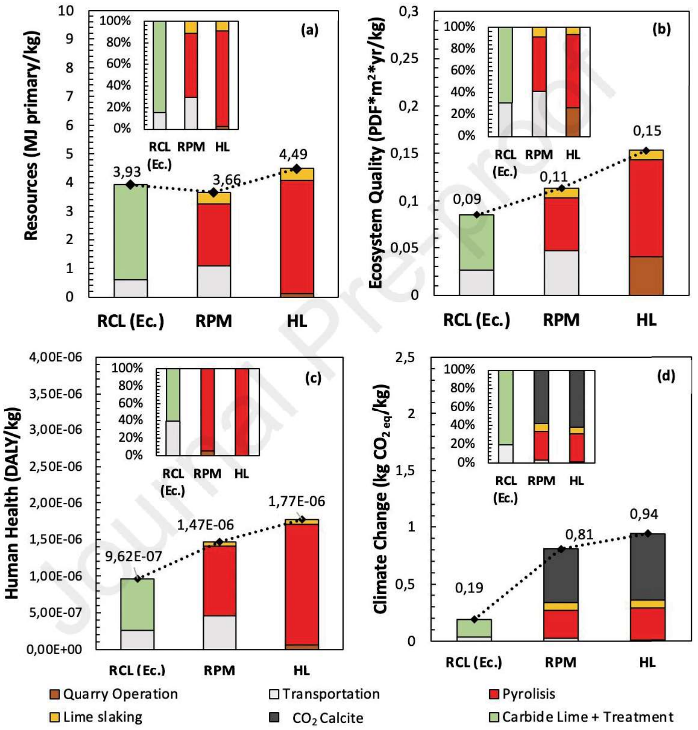


Figure 6. Cradle-to-Gate LCA results of producing 1 kg of RCL (Ec.), RPM and HL. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) endpoint indicator values and percentual contribution of each production step.

Regarding Resources, Ecosystem Quality and Human Health indicators (Fig.5a-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 for RPM (13% lower than for 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. 5d), RPM production results in a 14% reduction in CO_2 emissions compared to HL. The main reason is that in the RPM treatment, due to calcite decomposition 19% less CO_2 is generated compared to HL (0.47 kg CO_2/kg RPM vs. 0.58 kg CO_2/kg HL, Table 3). Further reduction is achieved due to the lower energy consumption for RPM, because of the credits of the organic matter (Table 3). Nevertheless, the CO_2 from the transportation operation diminishes the overall CO_2 reduction (Fig. 6d). Further information on the midpoint indicators employed to calculate the endpoint categories for RCL (Ec.) and RPM is provided in Appendixes 5 and 6 respectively.

All in all, for the Cradle-to-Gate situation, 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 6d). Even after applying an economic allocation in the acetylene production process (Fig. 5d), 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 plays a critical role, in terms of logistics but also costs and environmental impact due to the required transportation.

3.2.2 Cradle-to-Grave

Figures 7 and 8 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 7**.** 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). 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).

A. Scenario 1: Upcycling of paper mill sludge

Resources category (Fig. 7a)

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 4.1), 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).

Ecosystem Quality category (Fig. 7b)

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 [25]. 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 increases 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. 7c)

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[67]. 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. 7d)

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 3.2.1), 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].Click or tap here to enter text.

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, 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. 7d). 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

B. Scenario 2: Upcycling of carbide lime

Resources category (Fig. 8a)

As observed in the Cradle-to-Gate analysis of the RCL (Section 3.2.1), acetylene production demands a high energy consumption. When applying the economic allocation procedure, the use of recycled carbide lime (CL) as replacement for HL does not result in significant improvements. Allocating 1% of the Resources impact from the 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, which are the two most resource-impacting stages in the manufacturing phase of HL. Consequently, the production of CL and HL based plaster shows no significant differences in the resources impact category. Considering the similar impact for maintenance, the accumulated impact at the end of the life cycle (i.e., Final End of Life) shows only 2% reduction compared to HL plaster.

Ecosystem Quality category (Fig. 8b)

Regarding the RCLBP, binder production represents 17% of the total impact at the manufacturing stage, while for HLP, it is 29%. Although total emissions at initial production are reduced, the areas of damage are different for RCLBP compared to HLP. For RCLBP, emissions from the partial allocation of the impacts of acetylene to RCL production are the main contributors, and emissions to air are more significant (Respiratory organics +32% increment) due to higher coal combustion. In contrast, for HL, emissions to the earth and water due to virgin calcite extraction (use of explosives) are of greater importance (Terrestrial Ecotoxicity +68%, Aquatic ecotoxicity +66%). Combustion process emissions also play a significant role for HL, as 51% of the fuels used in the kiln are solid fossil [25]. When the final end of life is reached (100 years), an overall reduction of 13% is obtained for RCLBP compared to HL in terms of ecosystem quality.

Human Health category (Fig. 8c)

During the manufacturing stage, as observed in the Scenario 1 (RPMBP), the production of additives dominates the indicator, constituting 65% and 50% of the total for RCLBP and HLP, respectively. In terms of binders, the main midpoint indicators contributing to Human Health category are Carcinogens and Ozone Layer Depletion. In HL production, these indicators are 55% and 44% higher than in RCL, explaining the lower impact of RCLBP. Additionally, owing to the lower specific emissions from RCL preparation (Section 3.2.1), a 24% reduction is achieved in the manufacturing of RCLBP compared to HLP. As the maintenance stage at 50 years unfolds, the total indicator for both plasters increases around 52%. Upon reaching the end of life, there is a highly significative reduction of 22% for RCLBP compared to HLP in terms of accumulated impact.

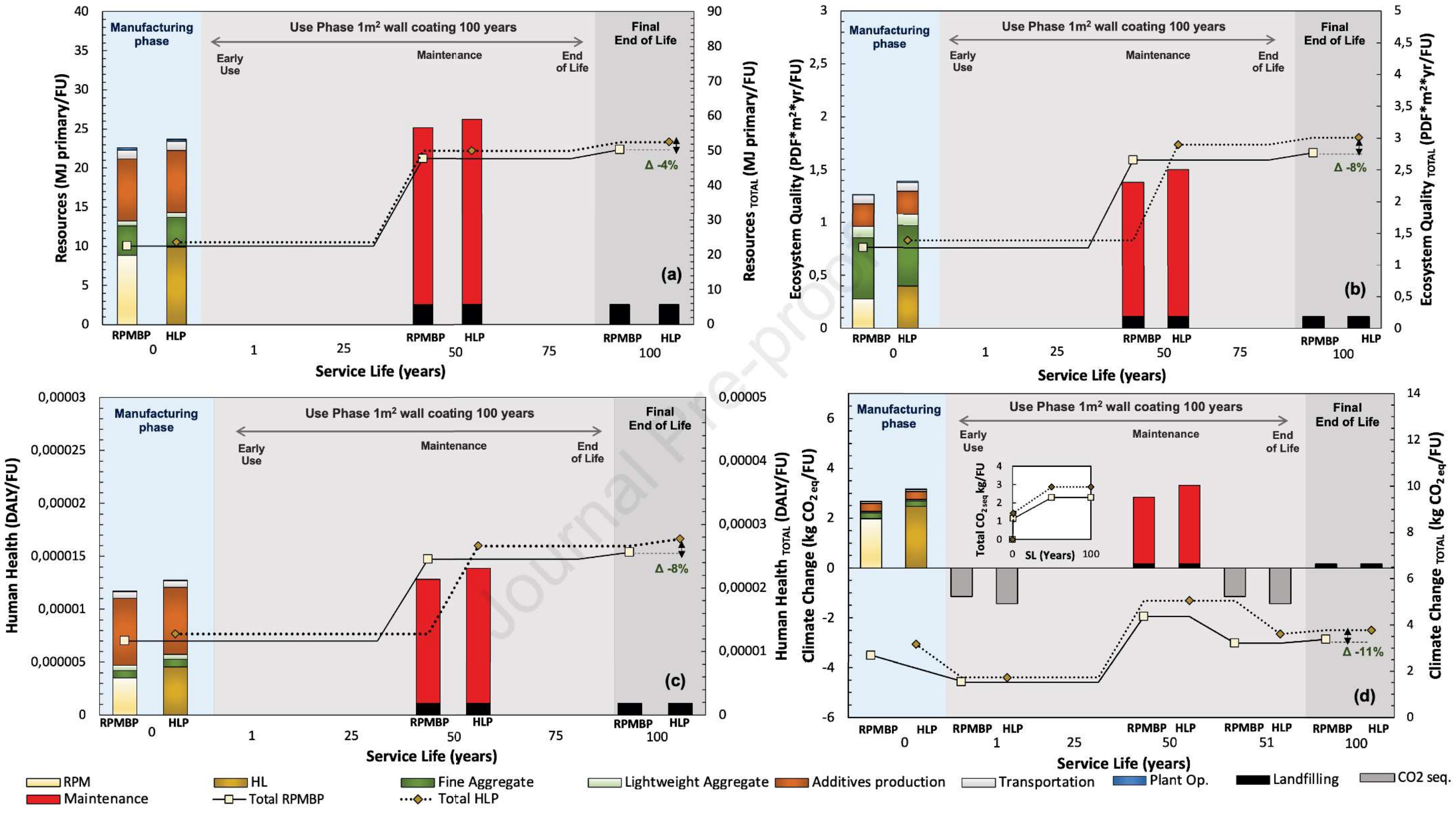


Figure 7. Results of the Cradle-to-Grave LCA of RPMBP against HLP. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) endpoint impact indicators.

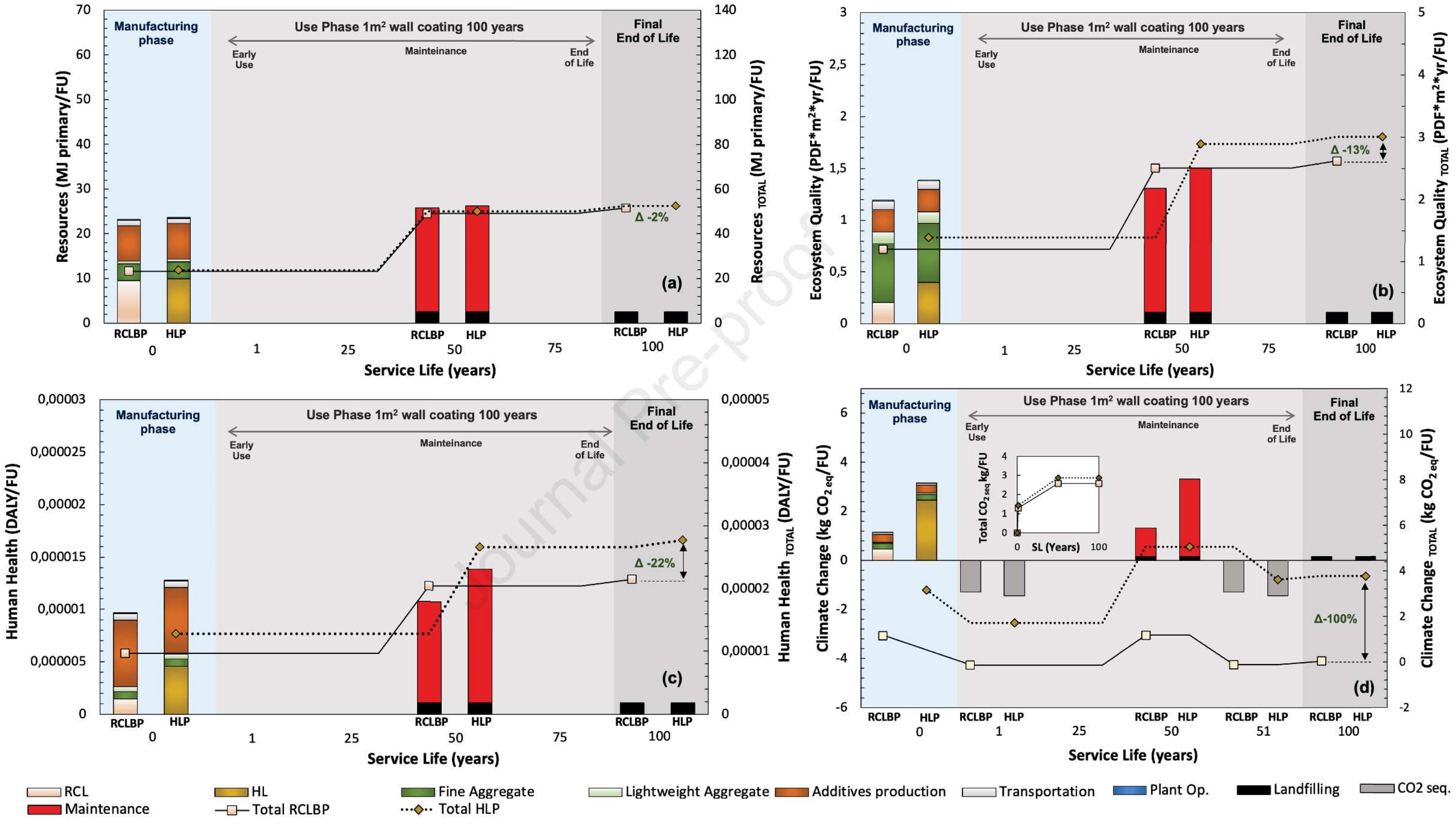


Figure 8. Results of the Cradle-to-Grave LCA of RCLBP against HLP. Resources (a), Ecosystem Quality (b), Human Health (c) and Climate Change (d) impact indicators

Climate Change category (Fig. 8d)

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 4). 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, 1.22 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_{2eq}/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.

3.2.3. Sensitivity and uncertainty analysis

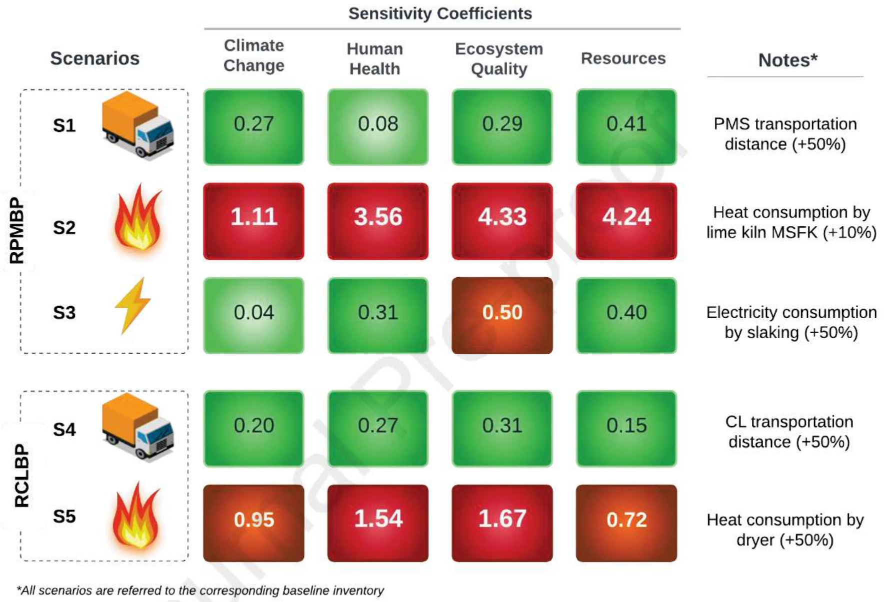
This section specifically focuses on conducting a sensitivity analysis for critical parameters within the life-cycle inventory of PMS and CL when utilized as a HL replacement in lime-based plasters. For a sensitivity analysis concerning traditional lime-based plasters (used here as a reference), readers 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.



	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 9. Sensitivity ratios of endpoint indicators for RPMP and RCLBP from Cradle-to-Grave

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. Table 7 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 7. 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

* GSD^2 = Geometric Standard Deviation Square

3.2.4. 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).

Sections 3.2.1 and 3.2.2 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. 9), 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 2, 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.

3.3. Economic sustainability of lime-based plasters manufactured with upcycled materials

Figure 10 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 an 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%.

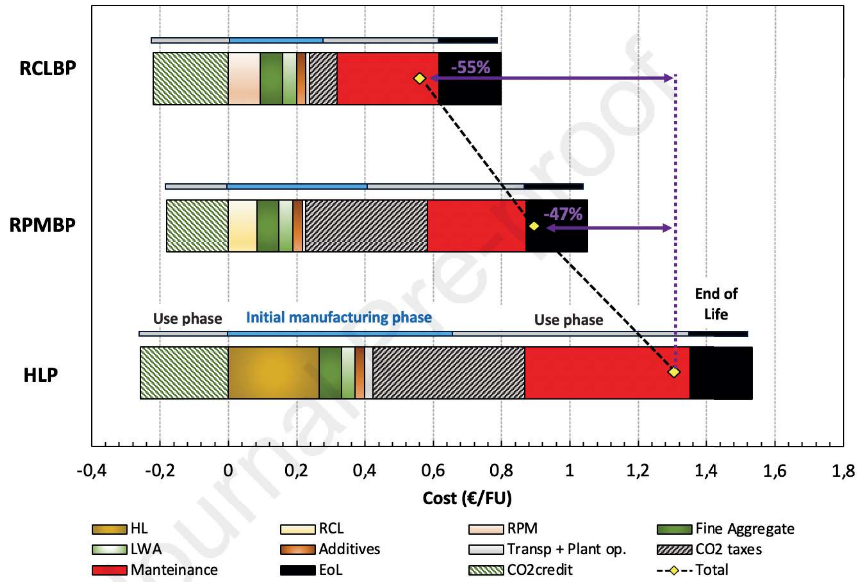


Figure 10. Life Cycle Cost Assessment results from Cradle-to-Grave of the plasters manufactured with traditional HL and upcycled RPM and RCL

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, 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 3.2, 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 7d and 8d).

The total sequestration potential per FU of the analysed scenarios over the life-cycle was shown in Table 5 and implemented in Figure 7d and 8d. Figure 9 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 2). 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.

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 CO_2 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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CRediT authorship 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.

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Appendix

Appendix 1. Impact2002+ Midpoint and Endpoint Indicators used in this study.

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

Appendix 2. Datasets from Ecoinvent V3.6 used to model the production of different components of the life-cycle inventory

	Process	**Ecoinvent 3.6 Dataset**
Binder	Hydrated Lime	Laveglia et al. [25]
	Metakaolin	kaolin production, operation and transformation, CH
Coarse Aggregate	Silica Sand	gravel and sand quarry operation, CH
Lightweight Aggregate	Perlite	expanded perlite production, GLO
(artificial)	Polystyrene	polystyrene production, expandable, GLO
Additives	Water Retention Agent	carboxymethyl cellulose production, powder–RoW
	Air Entrainer	alkylbenzene sulfonate, petrochemical RoW
	Hydrophobic Agent	polycarboxylates production, 40% active substance, RER
	Dispersion Agent	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 3. Providers of Energy Source for Electricity and Fuel mixes – Germany 2020 (Based on [25])

	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 {Europe without Switzerland}| heat production, natural gas, at industrial furnace >100kW | Cut-off	34	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	Heat, district or industrial, other than natural gas {Europe without Switzerland}| heat production, at hard coal industrial furnace 1-10MW | Cut-off	51	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland

Appendix 4. Life Cycle Inventory for the production of a lime-based plaster, adapted from [26]

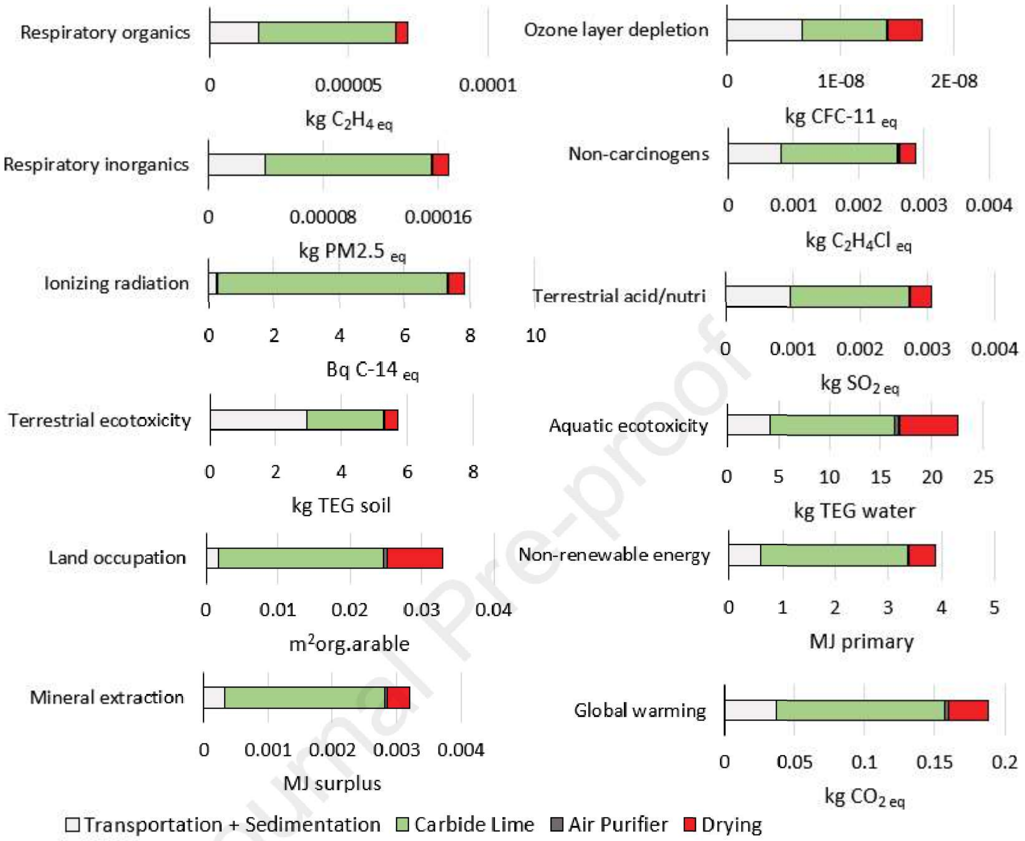
OPERATION	**PROCESS MODELLED**	**PROCESSED AMOUNT**		**INVENTORY AMOUNT**		**SOURCES & NOTES**
		AMOUNT	**UNIT**	**AMOUNT**	**UNIT**	
Plaster Manufacturing						
Input	**Fine aggregate**	0.675	t	0.675	t	See below Appendix 4.1
	Lightweight aggregate	0.07	t	0.07	t	Modelled by Ecoinvent (pumice quarry operation, 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 Germany 2020[25]
	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 Germany 2020[25]
	Hydrated lime (HL)	0.25	t	0.25	t	Laveglia et al, 2022 [25]
	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 Germany 2020[25]
	Dry mixer	4	kWh	4	kWh / t	Electricity mix Germany 2020[25]
Output	**Hydrated Lime Plaster (HLP)**	1.00	t	1.00	t	Output of the Plaster Manufacturing

In the reference scenario of Appendix 4, HL is used as binder and the lime-based plaster HLP is obtained. 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. The datasets to model their production are provided in Appendix 2. The electricity and fuel mixes used in the manufacturing stage are based on a previous work of the authors and available in Appendix 3.

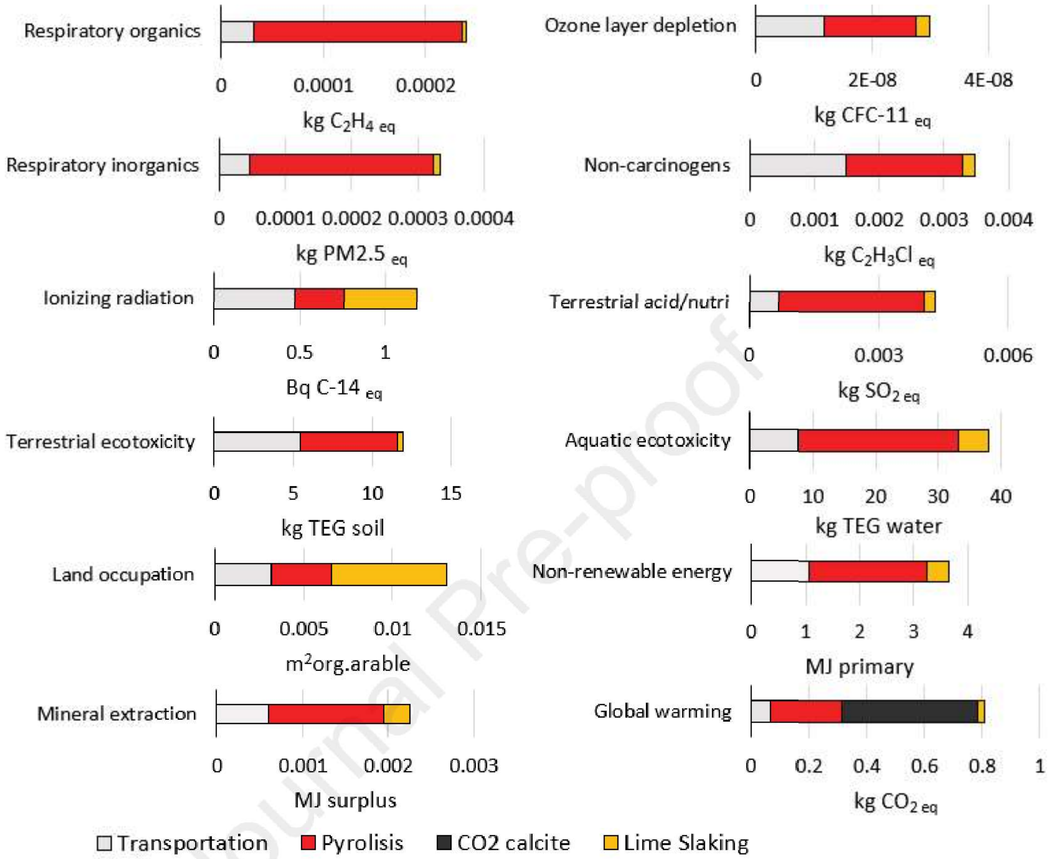
Appendix 4.1. Life Cycle Inventory to produce the fine aggregate for the plasters [26]

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 river bed, 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

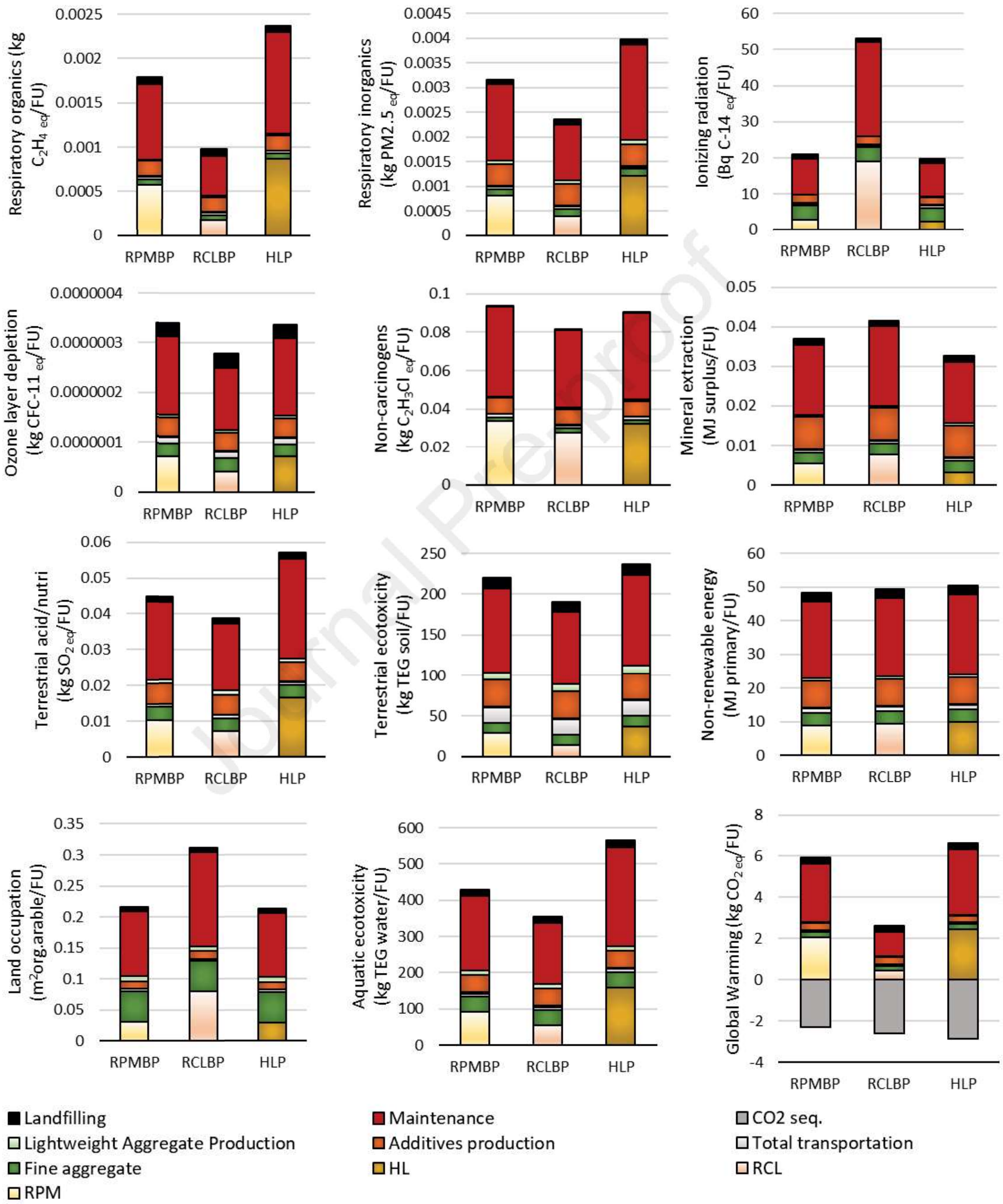
Appendix 5. Complete set of midpoint indicators per kg of RCL (Ec.) utilized in the calculation of the endpoint categories



Appendix 6. Complete set of midpoint indicators per kg of RPM utilized in the calculation of the endpoint categories



Appendix 7. 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



Declaration of interests

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

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: