Hydrated Lime Life-Cycle Assessment: Current and future scenarios in four EU countries

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Abstract

The environmental load associated to Hydrated Lime (HL) products is attributed to the limestone decomposition and the industrial production (combustion in the kiln, the electricity, the transports, etc.). Although the fuel and electricity mix used in the factory can be critical, no records of Life Cycle Assessment (LCA) have been found addressing this for HL. Considering the current environmental crisis, a shift to more sustainable sources of energy is expected. This paper studies, within the SUBLime EU network, the effect of the current fuel and electricity mix used in a HL plant, for Germany, Belgium, Portugal and Spain, as well as future scenarios. A theoretical Cradle-to-Gate Life Cycle Inventory for HL production was developed and used for scenario analysis, namely decarbonisation of the electricity matrix and replacement of hard coal by natural gas (NG) and biomass (B) in the fuel mix. The LCA for 2020 shows that, in 9 out of 15 indicators, the electricity consumption is significant. In terms of Global Warming Potential (GWP), 0.94 kg $CO_{2\,eq}$/kg HL are produced. Spain and Belgium have shown a better performance followed by Portugal and Germany. The results of future scenarios show that the shift to almost 100% renewable energies for electricity production reduce their sharing in almost all the indicators. As NG and B increase their proportion in the fuel mix, 9, 18 and 22% reductions in GWP in comparison to 2020 are achieved. However, 4 out of 15 indicators are higher than the reference due to the fuel mix.

Key words: Hydrated Lime, Renewable Energy Sources, Global Warming Potential, Sensitivity Analysis, Life Cycle Assessment

Highlights

- In 2020 in Europe an average of 0.94 kg $CO_{2\ eq}$/kg of Hydrated Lime are produced
- 9/15 indicators are sensitive to the electricity consumption in the plant
- In 2050, 4/15 indicators are higher than in 2020 due to the fuel mix
- The replacement of fossil solid fuel in the kiln allows up to 22% $CO_{2\ eq}$ savings.

Abbreviations

ASK	*Annular Shaft Kiln*
APREN	*Portuguese Renewable Energy Association*
AQEX	*Aquatic Eco toxicity*
AQA	*Aquatic Acidification*
AQE	*Aquatic Eutrophication*
BE	*Belgium*
BWE	*Federal Environmental Agency of Germany*
BAT	*Best Available Technologies document for the Production of Lime, Cement and Magnesium Oxide*
CG	*Carcinogens*
DALY	*Disability Adjusted Life Years*
DE	*Germany*
EuLA	*European Lime Association*
EU	*European Union*
EC	*European Commission*
ERs	*Annual Environmental Reports*
EPDs	*Environmental Product Declarations*
ETN	*European Training Network*
FSF	*Fossil Solid Fuels*
ITN	*Innovative Training Network*
FU	*Functional Unit*
GLO	*Global*
GWP	*Global Warming Potential*
GHGs	*Green House Emissions*
HL	*Hydrated Lime*
HC	*Hard Coal*
IR	*Ionizing Radiation*
IEA	*International Environmental Agency*
LCA	*Life Cycle Assessment*
LCI	*Life Cycle Inventory*
LO	*Land Occupation*
ML	*Milled Lime*
MoL	*Milk of Lime*
MLS	*Milled Limestone*
MFSK	*Mixed Feed Shaft Kiln*
ME	*Mineral Extraction*
NE	*Nuclear Energy*
NRE	*Non-renewable Energy*
NG	*Natural Gas*
NC	*Non-carcinogens*
OZLD	*Ozone Layer Depletion*
PL	*Pebble Lime*
PT	*Portugal*
PFRK	*Parallel Flow Regenerative Kiln*
PDF	*Potential Disappeared Fraction*
RO	*Respiratory Organics*
RoW	*Rest of the World*
RI	*Respiratory Inorganics*
RES	*Renewable Energy Sources*
SDG	*Sustainable Development Goals*
ES	*Spain*
SUBLime	*SUstainable Building Lime applications via Circular Economy and Biomimetic Approaches*
S1, S3, S3	*Scenario 1, 2 and 3 respectively*
TA	*Terrestrial Acidification*
TET	*Terrestrial Eco toxicity*
UN	*United Nations*

1. Introduction

Lime is one of the materials with the richest history and its use is as old as some of the most imponent civilizations that we have known. As an enabling material, lime renders and mortars had been found to be extensively applied in constructions all over the world of Israel (7000 BCE)[1], Syria (4250 BCE)[2], China (2000 BCE), Mayan, Inca and Aztec civilizations (500 BCE) [3]. Since then, it has played different roles, from structural functions to decorative techniques [4] and the knowledge for its preparation was transmitted from generation to generation. With the developing modern society and the increasing need of this material, the production of lime was completely industrialized. Nowadays the term "lime" (Calcium Oxide, CaO) is assigned to a product derived from limestone in an industrial process known as calcination [5] and not only its production but also uses have grown with time, becoming crucial in several applications. According to a report published by EuLA in 2018, the sales sector in 2016 was distributed among steel (39.2%), construction industry (18.1%), environmental protection (17.2%), other industrial consumers (13.8%), chemical industry (7.5%), export outside the EU (3.3%) and agriculture (1.9%) [6]. What is more, the Report for 'Competitiveness of the European Cement and Lime Sectors', issued in February 2018, states that in 2015 (latest available Eurostat), the lime and plaster industries represented more than 20 Mt/y [7]. It can be observed that the construction industry plays an important role in the scenario of lime consumption. Today, lime-based building products experience many challenges (e.g. raw material prices and environmental restrictions) that hinder the continuous development of innovative approaches in material design, process, product functionality and sustainability. This research is carried out within the framework of the SUBLime network [8], an EU ETN – ITN project that was born to provide answers to the former challenges, bringing together lime experts from the academia and the industry.

In terms of environmental performance in lime manufacturing, the production of 1 tonne of CaO generates around 1.2 tonnes of CO_2, contributing in about 1% to the global anthropogenic CO_2 ([9]–[11]). The environmental load associated to lime products can be accounted to two main causes. Firstly, almost two third of the emissions are linked to the chemical reactions during the calcination of calcium carbonate ($CaCO_3$) to produce CaO (0.78 kg CO_2/kg CaO) (reaction 1) [12]. The HL used in the construction industry is produced during a process called slaking (reaction 2).

$$CaCO_3(s) + \Delta H \rightarrow CaO\ (s) + CO_2(g) \qquad \Delta H^\circ = 177.8\ kJ/mol$$ **Reaction 1**

$$CaO\ (s) + H_2O\ (l) \rightarrow Ca(OH)_2\ (s) + \Delta H \qquad \Delta H^\circ = -65.2\ kJ/mol$$ **Reaction 2**

The second factor is the industrial production of quicklime itself, including the combustion of fuels in the kiln, the electricity needed to operate the plant, the transport of different materials, etc. Figure 1 shows the flow diagram of a plant that can produce four types of lime products: PL, ML, MoL and HL. The process starts from the extraction of limestone from a quarry, a series of particle size reduction operations, washing of the limestone to reduce the amount of fines, screening operations to separate different fractions of limestone and the calcination process itself where the material is heated in a kiln with temperatures above 1000°C [12]. As a result, quicklime in different particle sizes is obtained. The production of hydrated products (both MoL and HL) can happen in integrated plants (as shown in the Figure 1) or the ML can also be transported to a hydration plant at a different site. Some by-products might be generated during the process (such as MLS).

In terms of energy consumption, the calcination process in the kiln is the most energy intensive step in the lime production process; depending on the kiln it can vary from 3 to 9 MJ/kg CaO [11]. This operation accounts for around 90% of the total energy consumption and 99% of the specific impact of the whole process on global warming ([12]–[14]). Thus, many research articles mostly attribute the environmental impact of this process to the kiln operation. While it is true that calcination is a major contributor to global warming, there are other environmental problems not directly related to carbon dioxide equivalent emissions [15]. These include damage to ecosystem quality (Water Acidification or Terrestrial Eco toxicity) as well as damage to human health (ozone depletion or the generation of ionising radiation). These impacts can potentially be affected by the types of energy sources used as fuel for the furnace, as well as the electricity production matrix used to supply the plant, the impact of which may not be negligible. Likewise, being HL a product of high consumption in the world market, identifying in what proportion the energy sources have an impact would allow the design of strategies to comply with the SDG outlined by the UN [16]. In particular, 3 SDG are directly related to this research: Goal 7 "Affordable and Clean Energy", Goal 9 "Industry, Innovation and Infrastructure" and Goal 13 "Climate Action". The common line among them is the promotion of sustainable industries through energy efficiency and productivity, upgrading the technologies to provide clean energy from renewable sources (i.e. low-carbon development), contributing in turns to limit the increase in the Global Warming. However, there are very few records in the literature addressing this research line and, in particular for the construction industry and in lime-based mortars and plasters some studies have addressed a sensitivity analysis only on the content of HL in the dry mix and transport distances of cement, additives and sand [17], [18].

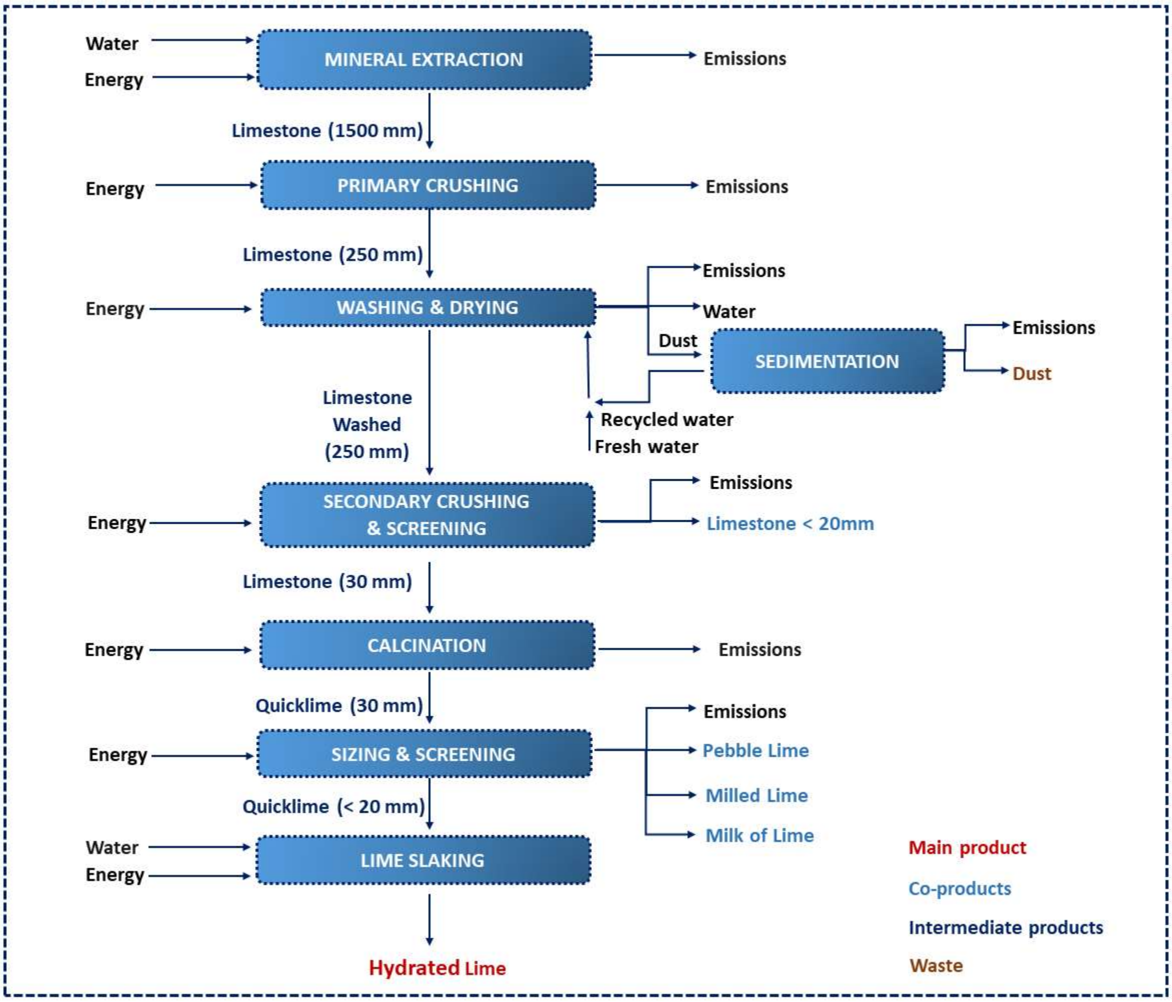

Figure 1. Industrial process for the production of Hydrated Lime – System boundaries considered in this study.

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

Because of the increase in use of more sustainable sources of energy, lower environmental impacts for the production of lime-based products are expected, as well as different environmental loads according to the geographic localization of the plants. Therefore, it is necessary to quantify the benefits of the changes in current and future scenarios for the production of HL in Europe, through LCA and sensitivity analysis. This paper deals with a case study for DE, BE, PT and ES and pursues the following objectives:

a) To present a Cradle-to-Gate LCI for the production of hydrated lime in a theoretical plant;
b) To quantify the environmental impact for regionalised production of HL, identifying the influence of energy sources used on the indicators considered;
c) To carry out a sensitivity analysis on the environmental impact indicators, considering the particular projections of each country in decarbonisation of the electricity matrix and the use of alternative fuels with potential CO_2 emission reductions.

2. Methodology

The LCA methodology was used to quantify and compare the potential environmental impact of the current and future scenarios for HL production in each of the analysed countries. This methodology is defined as "the compilation and evaluations of the inputs, outputs and potential environmental impact of a product system throughout its life cycle" [25]. The research carried out is based on the ISO 14040/44 (2006), and accordingly four main steps are to be performed: goal and scope, inventory analysis, life-cycle impact analysis and interpretation of the results [25], [26].

2.1. Goal and Scope Definition of the Case-Study

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

Table 1. Capacity of the theoretical plant

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

To perform a proper study regarding the environmental impact of products, it is critical to make an unambiguous definition of its scope. The studied plant is shown in Figure 1 and the process is based on the flowchart of one lime industry. It represents the production of different lime-based materials from the cradle (extraction of raw materials) to the gate of the factory. The operating process aims at producing the 4 products listed in Table 1. Even so, as a result of the unit process "Secondary Crushing & Screening" different fractions of ML are produced. Due to the operating conditions of the kiln, those fractions below 20 mm are not desirable. Instead of being disposed, they are sold as MLS to another industry.

2.2. Description of the scenarios

In order to analyse the effect of different energy sources and the effect of the geographic localization of the theoretical plant, the data of the current and future scenarios were obtained from different sources, among which are included the Webpage of the IEA, Energy department's/ministries and recognised Institutions in the field of Energy. The consideration of the effect of the energy source and its environmental implications is addressed to the best knowledge of the authors. Figure 2 summarises the current and potential future scenarios for the electricity mix in DE, BE, PT and ES.

2.2.1 Germany

A report by the International Energy Agency [27] states that until 2018 DE's energy system was still largely based on fossil fuels. Coal, oil and natural gas had the higher proportion in the total primary energy supply and total final consumption used for power generation. Nevertheless, renewable sources or energy from solar, wind power, biomass and other sources have increased their share in the German electricity mix. According to a Report by the BWE, the share of gross electricity consumption covered by renewables in 2019 (42.1%) rose by approx. 8% to nearly 243 billion kilowatt-hours compared to 2018 (37.8%) [28]. The increase was mostly due to favourable weather conditions and the further expansion of offshore wind-powered installations and of PV installations. In 2019, solar energy accounted for 19.1% of DE's electricity generation from renewables. Biomass contributed by 20.6%. Hydropower accounted for 8.3%, geothermal energy for 0.1%. More than half of the overall 242.5 billion kilowatt-hours generated came from wind power, with onshore wind power accounting for 41.7% and offshore wind power for 10.2%. In 2020 the amount of renewable energies has increased to around 45.5%, composed by the sources shown in Figure 2a. According to a very extensive study commissioned by the Federal Environmental Agency of DE and conducted by the Fraunhofer IWES, by 2050 DE has the technical and ecological feasibilities to base its electricity supply system completely on renewable energies [29]. Figure 2b shows the technologies that can be potentially used to fulfil the energy demand of the country. It is based on the projection that the electricity consumption will be around 10% lower than 2005 levels and for each technology, the area potentially available for its deployment was determined and reduced considering ecological considerations, competing land uses and settlement area [29].

2.2.2 Belgium

According to the "BE's Integrated National Energy and Climate Plan 2021-2030" [30], the country has made an effort to decrease the proportion of conventional sources in the electricity production matrix since 1990. In terms of the market share of total end consumption, petroleum products remain the principal source of energy (52%), followed by natural gas (24%) and electricity (17%). Natural gas is the dominant fuel in the industrial and residential sectors (35 % and 38 % respectively in 2015). In the transport sector, consumption is dominated by petroleum products (95%) [30]. As mentioned before, each country is subjected to its own reality for energy production. In the case of BE, due to their limited energy resources it is highly dependent on other countries for its energy supply. In 2015, its total primary energy production accounts for about 20 % of its total primary energy consumption and nuclear energy accounted for 73.9% of BE's energy production. The same year, the proportion for renewable fuels and waste was 19.5 %. In 2016, renewable energy accounted for 8.65 % of total final energy consumption [30]. In recent years, BE has made progress in developing renewable energy. In Figures 2c and 2d the transition from the current to a potential future scenario can be observed [31]. BE has proposed a series of scenarios for climate neutrality by 2050. The main climate neutral scenario is called the "CORE 95" scenario and leads to a reduction in GHGs emissions of about 95% in 2050 w.r.t. 1990 and to so-called negative emissions of about 5% of 1990 GHGs emissions, thereby leading to climate neutrality by 2050. Several changes in behaviour, lifestyle and marked societal changes in the way people move, house and feed themselves as well as a drastic decrease in energy demand are responsible for this change. Furthermore, the model implies several changes into the energy sources used, not only the reduction of conventional sources but the increment in the proportion of novel green sources such as Biofuels [32].

2.2.3 Portugal

PT is one of the EU countries that experienced a major financial crisis in 2008, and since then has been recovering. Furthermore, they were able to accelerate the structural changes required to shift from energy intensive activities and decoupling economic growth from energy demand. Nevertheless, until 2019 they remained reliant on imported fossil fuels, accounting for around 76% of primary energy supply (43% oil, 24% natural gas and 6% coal) [33]. PT has the resources to produce electricity from their rivers and wind, being almost 54% of the electricity generation covered by these sources with a high use of bioenergy in industries and buildings. In 2020 around 60% of the electricity supply in PT was based on RES (Fig. 2e). Compared to 2019, in 2020 the use of non-renewable sources decreased by 11.9% [33]–[35]. The country was also one of the first in the world to set 2050 carbon neutrality goals. The developed policies consider the key role of hydrogen for achieving carbon neutrality, but same as BE, a change in lifestyle and market is needed along with monetary incentives for green alternatives. The APREN, has recently (2018) published a report analysing the opportunities of the electricity sector, to achieve the required decarbonisation level. They have concluded that the contribution of renewable electricity should be around 94% in 2050. Saying that, a change in the energy mix supply must be carried out, reducing the proportion of conventional fuels such as coal and natural gas, and increasing greener sources such as wind offshore and onshore and solar energy. The last one is estimated to represent 30% of Portuguese electricity mix, while wind will reach 39% [36]. Figures 2e and 2f summarise the projected scenario by APREN, considering a reduction of GHGs emissions from the energy sector up to 75% in 2050 [34].

2.2.4 Spain

As in many countries of Europe, the regulation of the electricity sector in Spain is undergoing a profound reform. According to the Spanish Ministry of Energy the main objective of this reform is to ensure the economic and financial sustainability of the electricity system, while guaranteeing electricity supply with the necessary levels of quality and at the lowest possible cost, an effective level of competition in the sector and all of this framed within the principles of environmental protection of a modern society [37]. The demand for electricity in ES has consolidated its positive trend from 2015, and the demand in 2018 reached 268,808 GWh (0.4% up on the previous year). Moreover, the generation registered a fall of 0.5% with respect to 2017, affecting mainly coal-fired and combined-cycle generating stations, whose production decreased by 17.2% and 18.9% respectively. Wind power has increased by 1.5%. The rest of the electricity generation technologies showed minimal or insignificant variations [38]. This observed decrement in the use of energy demand was registered again in 2020 falling around 5.5% in comparison with 2019. The 2020 report of the Spanish Electric System shows that the proportion of renewable energies and non-renewable sources to produce electricity were 45.5% and 54.5% respectively [39], [40]. Among the energy sources for electricity generation, non-renewable nuclear energy can be highlighted, combined cycle and cogeneration and mainly wind and hydro sources for green technologies (Figure 2g). In the case of ES, it was not possible to establish a 2050 scenario in terms of used technologies for electricity production. Instead, the data for 2030 is taken from the Integrated National Energy and Climate Plan 2021-2030 [40]. In the future, the contribution of hydroelectricity is not expected to grow significantly given to the available resources already being used, whereas solar and wind are expected to grow in interesting proportions to contribute to the RES (Figure 2h) [20], [40].

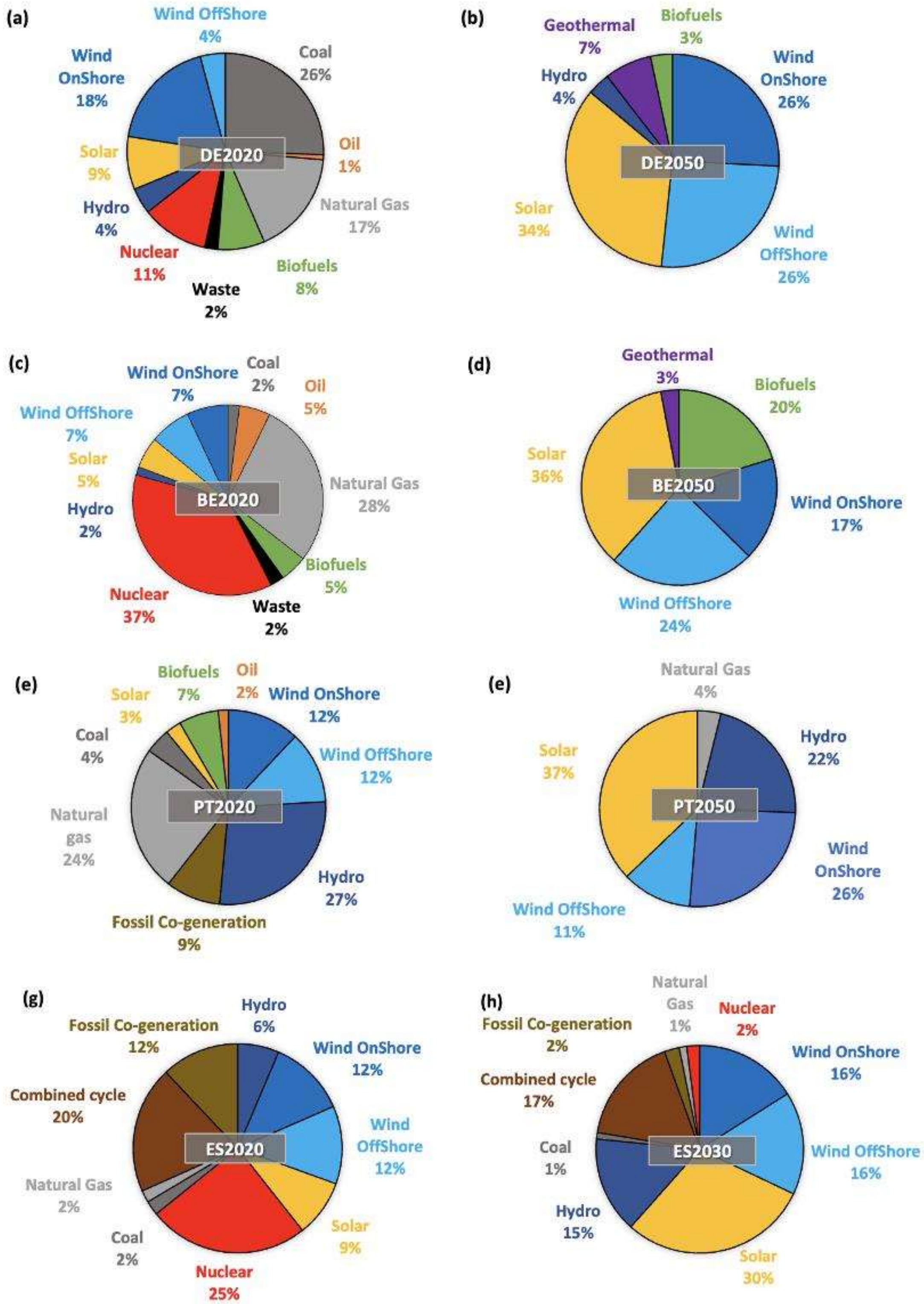

Figure 2. Electricity mix for the baseline scenario (2020) and potential future scenarios (2030/2050) in DE (a, b), BE (c, d), PE (e, f) and ES (g, h)

2.2.5 Sources of thermal energy for the lime kiln – Fuel Mix in Europe

As previously mentioned, a critical unit in the production of lime is the kiln of a lime factory. It is the most energy intensive step of the production line and highest the specific energy consumption depending on the used technology. This unit process also comprises the biggest share of emissions, coming from both raw material decomposition and the fuel combustion. The emissions associated to the limestone decomposition are actually well known, and can be assumed to be around 0.78 kg CO_2/kg CaO (Reaction 1). However, during the combustion of fuel, a wide range of gaseous products (i.e. emissions) are generated, along with thermal energy as a result of the exothermic reaction. The nature of these emissions and its potential impact on the environment, obviously depend on the fuel type used. According to the BAT document, except for mixed feed shaft kilns, all types of kilns can operate with all types of fuels (solid, liquid and gaseous) [11]. This opens doors for reducing the emissions, by considering that around 30% of the emissions during the production of lime are accounted to the fuel combustion [12]. Furthermore, up to until 2008 the most common used fuels in the EU were solid fuels like lignite, coal, petcoke and coke; in addition to natural gas, liquid and waste fuels and biomass [11].

The last EuLA report of 2019 [12], indicates the composition of the average fuel mix used by the European Lime industries in 2010 (Figure 3). In terms of the fuel mix, FSF are principally made of hard coal, lignite and petrol coke, whereas gaseous fuels are made of NG and liquid fuels of light fuels [41]. It can be observed that there is very limited use of biomass and waste in lime production. Some explanations for this context can be found considering that, the heterogeneity of these type of fuels, operating conditions in the kiln (including the mixing of volatiles and oxygen), pre-treatment needed (usually drying), as well as the complexity of the combustion reactions and emissions, make it less easy to work with other than traditional fossil fuels [42]. At the same time, in particular in lime manufacture, the quality of the product can be severely affected if the waste and biomass does not comply with the very precisely defined physical properties [11].

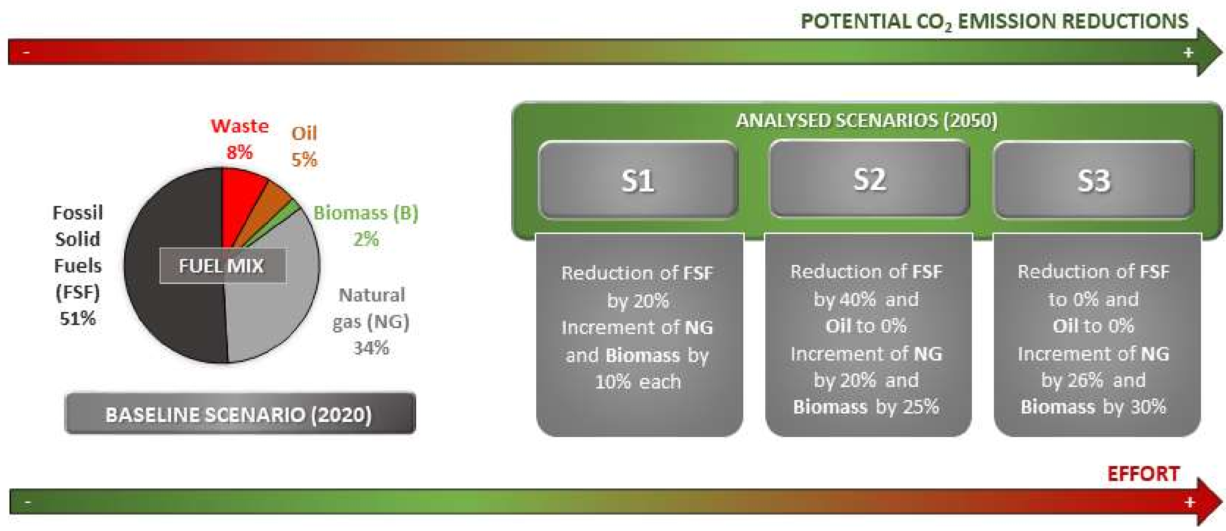

Figure 3. Average fuel mix (2010) based on EuLA report (baseline scenario)[12] and alternative fuel mix scenarios for potential CO_2 emission reduction.

When it comes to reducing the emissions, FSF need to be replaced as much as possible. To illustrate the problem, it is enough to consider the CO_2 emission factor (tCO_2/TJ) in DE of lignite (103.8), light fuel oil (74) and natural gas (55.9) [43]. In theory, around 50% of combustion emissions can be saved by using natural gas instead of lignite. On top of that, a recently published empirical assessment in 10 European countries has shown that natural gas is the main fossil fuel used to back up renewable energy sources [44]. Furthermore, an increase of the proportion of biomass can not only lead to reduce the CO_2 emissions, but also to achieve zero net CO_2 emission if they are grown in a sustainable way [45], [46] [47]. Considering the aforementioned facts, the scenarios proposed in Figure 3 are based on the chance of reducing the amount of FSF, shifting to eco-friendlier fuels by incrementing the proportion of NG and biomass. In the baseline scenario (2020), a minimum effort to follow current regulations is needed, meanwhile no significant environmental improvements are taking place. Nevertheless, moving to a more sustainable model implies making economic, technological, and socio-cultural sacrifices. This means that less efficient kilns need to be replaced by PFRK and it might be also possible that extra operation units are required to deal with the pre-treatment of the biomass. In addition to that, cultural changes and/or financial investments along with new policies will be needed to absorb part of the costs for the use of alternative sources of energy (such as NG or Biomass), otherwise the cost of a final product can be severely affected [41].

2.3. Life Cycle Inventory and Life Cycle Assessment

The inventory analysis is a critical phase of the environmental assessment, as the obtained results are directly linked to the quality of the data used in the LCA [48]. The ISO Standard 14044 establishes that the data should address: time-related coverage, technology coverage, precision, completeness, representativeness, consistency, reproducibility and (un)certainty of the information [26]. The data gathering poses a problem in itself because the sources from which they can be obtained are multiple and it is not always possible to obtain the same level of detail for every part of the system under study. The best-case scenario is to build up the LCI in partnership with the industries through the use of detailed questionnaires, which is rather unlikely to happen for research purposes due to the critical nature of the information required. Some industries communicate publicly the impact of their activities through ERs and EPDs. However, neither of the abovementioned documents are mandatory nowadays, despite the fact that the EC has been recommending since 2002 that EPDs should be compulsory [49]. This results in the unavailability of the data and when available, also holds the risk of misinterpretation or double counting. However, the larger part of the LCIs are mostly based on data from EPDs, ERs and literature (i.e. papers and theses) [48]. While a certain level of truthfulness is missed, generic databases are a powerful source for process modelling, among which EcoInvent [50] and Gabi [51] are considered the most complete available for the construction sector [52]. A less traditional approach to build the LCI of the system process would be to design, at a certain level of detail, the plant that will deal with the product of interest. This is not always easy to do, nor recommended to all the practitioners of the LCA methodology, because it requires an integral background of transport phenomena (mass, energy, and momentum) as well as the process engineering criteria to select the adequate devices to model the process accurately. Although literature shows that traditional data sources are widely used for research purposes, the advantage of the aforementioned methodology is that having the unit processes discretised allows to fully control the main parameters in each step of the production line (energy and mass requirements). Unlike the use of generic databases, this approach allows to easily perform the sensitivity analysis proposed in this paper.

2.3.1. Hydrated Lime Production

For the production of HL, the system process is defined in Figure 1 and the theoretical capacity of the plant is stated in Table 1. The system process is composed by 6-unit processes. From the aforementioned information, the quantities of all materials that enter and leave each unit based on the principle of law of conservation of mass was calculated. For the units dealing with chemical reaction, the mass of the reactants/products (i.e. in the Shaft Kiln) was determined through stoichiometry and the extent of the reactions was considered fully completed. A series of assumptions and simplifications were made, such as the humidity of the limestone as it enters to the Washing Process or its final humidity after the Drying Process. Some material flows such as calcite, water (as resource and emissions) or carbon dioxide (as emission) as well as the transports from the quarry to the plant were modelled by EcoInvent 3.6 database [50]. After having all the material flows specified, the energy requirements were determined. It is important to highlight that only the energy required to operate the devices is considered in the design, while other energy demands such as the electricity of the administrative offices or heating of the plant is not pondered. The devices were selected from catalogues of a variety of producers considering multiple criteria: type of material, feed and output size, capacity of the device, energy source (fuel, electricity), most used technologies in the lime production sector, among others. An overview of all considered devices is presented in Table 3. From the catalogues, the Power (kW) and the min. and max. capacity (t/h) of the devices was stored. For each device, the specific power (kWh/t of material) was calculated as the power divided by the average capacity. The kiln used in the process required special attention, given that is the central part of the process. According the BAT document 90% of all kilns used in Europe are Shaft kiln type [11]. From this amount 21% corresponds to MFSK, 29% to PFRK, 13% to ASK, and 37% comprises a variety of shaft kilns under the category "others" [11]. Among all the specified technologies, PFRK comprises the higher percentage and therefore was selected as the kiln of the factory.

Finally, the LCI comprises the material and energy requirements to produce 1 tonne of HL from cradle to gate, being this, the FU of the study, used to compare the environmental performance of the analysed scenarios.

Table 2. Overview of the main sources for characterization factors and impact categories according to the Impact 2002+ impact method. Obtained from Impact 2002+ User Guide [53].

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

2.3.2. Electricity and fuel mix

Both the electricity and fuel mix were created as different processes for each country, and their components and proportions adjusted correspondingly to represent the analysed scenario. For the sake of reproducibility of this research, in the Complementary information section (Tables A1-5) the full package of providers used to model the electricity and fuel mix from the EcoInvent Database 3.6 can be found. The proportion of each source in the electricity mix of each country (current and future) is stated in in Section 2.2. For the modelling of their production the EcoInvent 3.6 database [50] was used, selecting the geographic location in the respective country whenever it was possible. For instance, for DE in 2020, 18% of the electricity mix is modelled by "Electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S – DE". In terms of the fuel mix, each contributing proportion was modelled through the heat production process of EcoInvent 3.6 database [50] (i.e. heat production, heavy fuel oil, at industrial furnace 1MW - Europe without Switzerland). Due to the lack of data in the used database, it was not possible to geographically localize the heat production for each country. On the contrary, the "Europe without Switzerland" process was used for all countries.

Table 3. Life Cycle Inventory of the Cradle-to-Gate HL production

OPERATION	PROCESS MODELLED	PROCESSED AMOUNT		INVENTORY AMOUNT		SOURCES & NOTES
		AMOUNT	UNIT	AMOUNT	UNIT	
Mineral extraction & Primary Crushing (I)						
Input	**$CaCO_3$**	7.00	t			Modelled by EcoInvent (Calcite, in ground)
	Water use	0.15	m^3	0.02	m^3 / t $CaCO_3$	Modelled by Ecoinvent limestone quarry operation
	Land occupation	0.59	m^2*year	0.08	m^2*year / t $CaCO_3$	Modelled by EcoInvent (Land Occupation - RoW)
	Blasting	1.13		0.16	kg / t $CaCO_3$	Modelled by EcoInvent (Blasting - RoW)
	Diesel consumption (Truck hauling, drilling machine and Loading machine)	161.00	MJ	23.00	MJ / t $CaCO_3$	Modelled by EcoInvent (Diesel, burned in building machine - GLO)
	Explosive	1.13	kg	0.16	kg / t $CaCO_3$	Modelled by EcoInvent (Explosive production Tovex)
	Transformation due to mineral extraction	0.05	m^2	0.01	m^2 / t $CaCO_3$	Modelled by Ecoinvent limestone quarry operation
	Recultivation (limestone mine)	0.05	m^2	0.01	m^2 / t $CaCO_3$	Modelled by Ecoinvent limestone quarry operation
	Jaw Crusher	1.87	kWh	0.27	kWh / t $CaCO_3$	Electricity mix (SUBLime designed)
	Conveyor belt	0.03	kWh	0.004	kWh / t $CaCO_3$	Electricity mix (SUBLime designed)
Output	**$CaCO_3$ Crushed[1]**	7.00	t			Main product as a result of (I)
Washing, drying and sedimentation (II)						
Input	**$CaCO_3$ Crushed[1]**	7.00	t			Input from (II)
	Water for washing	0.98	t	0.14	t / t $CaCO_3$ Crushed[1]	Modelled by SUBLime
	Sedimentary pool Operation	27.10	kWh	3.88	kWh / t $CaCO_3$ Crushed[1]	Electricity mix (SUBLime designed)
	Washing Machine Operation	5.47	kWh	0.78	kWh / t $CaCO_3$ Crushed[1]	Electricity mix (SUBLime designed)
	Drying Machine Operation	9.33	kWh	1.33	kWh / t $CaCO_3$ Crushed[1]	Electricity mix (SUBLime designed)
Output	**$CaCO_3$ Washed**	6.16	t	0.88	t $CaCO_3$ Washed / t $CaCO_3$ Crushed[1]	Dry $CaCO_3$, first crushing operation
	Fines washed	0.70	t	0.10	t / t $CaCO_3$ Crushed[1]	Modelled by Ecoinvent (disposal, ordinary industrial waste)
	Water	0.53	t	0.08	t / t $CaCO^3$ Crushed[1]	Humidity removed after the Washing Process
Secondary crushing and screening (III)						
Input	**$CaCO_3$ Washed**	6.16	t			Input from (II)
	Jaw Crusher Operation	4.10	kWh	0.67	kWh / t $CaCO_3$ washed	Electricity mix (SUBLime designed)
Output	**$CaCO_3$ Crushed[2]**	4.92	t	0.80	t $CaCO_3$ Crushed[2] / t $CaCO_3$ Washed	Main product as result of (III) - Allocation by mass (0.8)
	MLS	1.23	t	0.20	t MLS / t $CaCO_3$ Washed	Allocation by mass (0.2)
Calcination (IV)						
Input	**$CaCO_3$ Crushed[2]**	4.92	t			Input from (III)
	Shaft Kiln Operation	133.00	kWh	27.10	kWh / t $CaCO_3$ Crushed[2]	Electricity mix (SUBLime designed)
	Shaft Kiln fuel consumption	13000.00	MJ	2640.00	MJ / t $CaCO_3$ Crushed[2]	Fuel mix (SUBLime designed)
Output	**CaO**	2.76	t	0.56	t CaO / t $CaCO_3$ Crushed2	Product as a result of (IV)
	CO_2	2.17	t	0.44	t CaO / t $CaCO_3$ Crushed[2]	Stoichiometric CO_2 emission due to Limestone decomposition
Screening & Sizing (V)						
Input	**CaO**	2.76	t			Input from (IV)
	Vertical Mill Operation	92.40	kWh	33.50	kWh / t $CaCO_3$ Crushed[2]	Electricity mix (SUBLime designed)
Output	**CaO for Hydrated Lime**	0.77	t	0.28	t CaO for Hydrated Lime / t CaO	Main product as result of (IV) - Allocation by mass (0.28)
	PL	1.00	t	0.36	t Pebble Lime / t CaO	Co-product as result of (IV) to be sold as Pebble Lime - Allocation by mass (0.36)
	ML + CaO for MoL	1.00	t	0.36	t CaO for ML & MoL / t CaO	Co-product as result of (IV). Less than 0.4% of the produced CaO is used to produce MoL, therefore it is allocated altogether with ML- Allocation by mass (0.36)
Lime Hydration (VI)						
Input	**CaO for Hydrated Lime**	0.77	t			Input from (V)
	Water	0.25	t	0.32	t / t CaO for Hydrated Lime	Modelled by SUBLime (Ecoinvent tap water production, Europe without Switzerland)
	Lime Hydrator Operation	0.27	kWh	0.35	kWh / t CaO for Hydrated Lime	Electricity mix (SUBLime designed)
Output	**Hydrated Lime**	1.00	t			Main product as result of (III)
	Emissions, Waste					Emissions and waste along the production chain of Hydrated Lime

Following the analysis on the fuel mix, it is important to be aware about the limitation of the dataset selected to model the generation of thermal energy through the combustion taking place in a kiln. Even though the best attempt to represent the system under study was done by selecting the closest most representative process available in EcoInvent V3.6, the data detailed in Table A5 does not specifically represent the combustion process in a lime kiln. As a result, the process specific emissions may vary depending on the case specific type of the kiln technology. The results presented in this work should be interpreted as an approximation under these assumptions.

2.3.2. Life Cycle Assessment

The software OpenLCA was used for the impact assessment. The approach followed is consistent with an attributional LCA, where the inputs and outputs were attributed to the FU of the system by linking the unit processes of a system under an allocation procedure [48], [54], [55]. In this case, to divide the impacts arising from the same process between products and co-products was done through a mass allocation.

Regarding the impact method, and in particular in view of the production of HL, it is critical to include impact categories accounting for waste production and mineral resource depletion. These categories which are two of the major impacts of mineral industry sector, are something that the most used impact analysis methods (e.g. Eco-indicator 99 and CML 2002) do not include [52], [56]. On the contrary, Impact 2002+ addresses the damage categories of Resources, Climate Change and Ecosystem quality through the use of Midpoint categories such as Global warming, Land occupation, Terrestrial Ecotoxicity and Mineral Extraction among others [53]. An overview of the considered baseline impact categories and their characteristic factor is presented in Table 2.

3. Results and discussion

3.1. Life Cycle Inventory of the Hydrated Lime Production

For the development of the LCI of the HL, the theoretical plant of Figure 2 was divided into six unit processes, unifying them according to the related operations being performed. The parameters as well as the technical considerations for the selection of the devices in each unit operation are described below.

- Mineral extraction and Primary Crushing (I): The truck hauling carries the material around 1.4 km. This average distance is the result of the analysis of several lime factories. Afterwards, it is discharged on a conveyor belt that ends in the primary crushing, which is modelled by a Jaw Crusher designed and selected from a catalogue (Power 45 kW). It was assumed that the limestone comes with 10% of dust, 2% humidity, feed size 1500 m and output size 250 mm.
- Washing, drying and sedimentary pool (II): The washing machine was designed and a Log Washer was selected from a catalogue (Power 45 kW). During the washing it was assumed that 5% of the water leaves with the washed limestone (i.e. with no fines). The washing water is directed to a Sedimentary Pool, where the main devices considered are 3 equivalent centrifugal pumps (Design theoretical power 7.5 kWh). The dust leaves this device as waste and the recirculated water is 90% of the feed to the Washing machine. The device used in the process “dryer” is a Rotary Kiln Drier (Power 30 kW) and the material leaves the drier with depreciable humidity.
- Secondary crushing and screening (III): The main device of this process is a Jaw Crusher (feed size 250 mm, output size 30 mm, Power 30 kW) selected from a catalogue. The MLS screened to below 30 mm (around 20% of the crushed limestone) are sold as by-product.
- Calcination (IV): According to the literature, the average energy consumption of this device is 3.9 ± 0.5 MJ/kg CaO [11], [12], [57]–[60]. This value is depending on many factors among which are included the kiln efficiency, the type of fuel used, the capacity of the kiln, the amount of air in excess and the temperature of the air. The feed size is 30 mm. It was assumed that the CO_2 emissions associated to the limestone decomposition are equivalent to the stoichiometric amount resulting from Reaction 1 (0.44 $tCO_2/tCaCO_3$).

- Screening & Sizing (V): The main device is a Vertical Fine-powder Mill (feed size 30 mm, output size <20 mm, Power 335 kWh) designed and selected from a catalogue. The CaO produced is allocated in mass according to the amount required in the manufacture of each product (i.e 36% for PL, 36% for ML, 4% to MoL and 28% for HL).
- Slaking (VI): The device is a Multi Stages Hydrator (Power 20 kW) designed and selected from a catalogue to fulfil the requirements.

From the aforementioned considerations, the LCI for the Cradle-to-Gate production of HL is presented in Table 3. In this table it is presented the 6 Unit Operations described, the detail of the modelled process, the processed amount (i.e. the mass and energy requirements of each unit process to produce 1 t of HL) and the inventory amount (i.e. the normalized amount of the particular unit process per t of the reference unit).

As a means to analyse the quality of the LCI, a LCA under the conditions described in Section 2.3 was performed and the equivalent $CO_{2\,eq}$ emissions per kg of product (CaO and $Ca(OH)_2$) calculated. Table 4 shows the comparison of the results of this study (called SUBLime according to the running ITN EU project) to different other sources. The results reported by EEA [11] for CaO indicate that only the CO_2 emissions due to the limestone decomposition have been considered (stoichiometric). The EuLA LCI study has passed successfully the independent external critical review from Rina Consulting [61], being the most representative dataset Europe-wide. Both, EcoInvent and SUBLime results represent the production of Lime and Hydrated Lime in DE. In Table 4, it can be observed that the SUBLime results are in the same order of magnitude as results from the other mentioned sources for both considered products. For quicklime production, the greatest differences are with the reported values from EEA. Furthermore, the differences with the EcoInvent database [50] can be explained considering different system boundaries and/or technologies used for the production (types of kiln, fuels, etc.). However, the results of EuLA and SUBLime are very close (7% relative difference). Beyond the differences detected, the theoretical study correlates well with the sources used for comparison. Therefore, the SUBLime LCI for the cradle-to-gate production of HL (Table 3) is used in the following section for the case-studies.

Table 4. CO_{2eq} emissions per ton of product.

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

3.2. Life Cycle Impact Assessment

Figure 4-8 shows the LCA results for the production of 1 kg of HL, under the conditions described in Section 2.3. The results comprise the environmental impact of current and potential future scenarios for DE, BE, PT and ES. Four categories have been created, to account for environmental impact assigned to each indicator and scenario analysis, namely: Raw material emissions (Limestone decomposition), Quarry operation (including the drilling, blasting at the quarry and the transport by truck to the primary crushing), Electricity (used to operate the devices of the plant, including all the operations mentioned in Table 3) and Kiln Operation (fuel consumption to provide the thermal energy required for the limestone decomposition).

3.2.1 The 2020 scenario

Figure 4 shows the environmental impact for the production of 1kg of HL in 2020 in DE, BE, PT and ES. In these results, the only variable is the electricity mix considered in each country for the year 2020, while the share of quarry and kiln operation are the same for each country. In general, it is noted that the environmental impact of HL production depends on the country.

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

In terms of the Ecosystem quality, two midpoint categories are highlighted: Aquatic Eutrophication and Land Occupation. The first one, quantifies a major water quality issue, related to excessively high environmental levels of macronutrients (nitrogen and phosphorus), which provoke an increased growth of algae [63]. The higher effects in this category are registered for DE and PT, mostly due to the proportion of hard coal and hydroelectric production of energy. The Land Occupation indicator takes into account the area occupied, the duration of occupation and the damaging potential for ecosystem quality of a specific land use type ($m^2_{organic\ arable\ land}$*year) [64]. It gives an insight of the damaging potential for the ecosystem quality of using a specific area, for a certain amount of time and for a specific activity. The mining industry is well known for having a significant impact on the use of natural resources, and usually intuitively the highest impact is assigned to the lime quarry that is being exploited. However, it is very interesting to find out that the production of energy (both heat and electricity and mainly from hard coal and natural gas) was dominating the indicator, even though a land occupation for the extraction of limestone was considered in the Life Cycle Inventory (Table 3 of the manuscript). While common sense would suggest that the Quarry operation would have a significant contribution to the magnitude of the indicator, this is not the case. The reasons are various. As mentioned before the Land Occupation indicator is much more complex than the mere use of a specific area, because it takes into account in which way the land is used, to assign the impact factors that characterize the elementary flows, that in the end are aggregated in the indicator. Digging deeper into the Impact2002+ method, the specific factor assigned to the Land Occupation of the Mineral Extraction site is around 35% less than the impact factor assigned to the production of heat and electricity (on average). This impact factor is multiplied by the inventory result, which is the second explanation for the results that were found. For instance, taking a look at the scenario 2020 for Germany (Figure 4), when analyzing the inventory results, the Mineral Extraction and Primary Crushing operation contribute for around 4% to the impact category. The remaining 96% of the inventory is distributed to the land occupation for the production of energy (around 65% heat production, 31% electricity production). Therefore, the contribution of the land use of the quarry is negligible in comparison to the one assigned to the energy production.

Concerning the Human health damage category, the Ozone Layer Depletion and Ionizing Radiation stand out. To start with, the ozone layer is a band of gasses, mostly ozone (O_3), located 15-30 km above the Earth (stratosphere) that absorbs most of the Sun's ultraviolet radiation. After the discovery of a dangerous 'ozone hole' in the stratosphere, all nations in the world agreed in 1987 to take action under the Montreal Protocol on substances that deplete the ozone layer (ODS) [65]. In 2009 the EU released a regulation on highly detrimental substances, including chlorofluorocarbons, hydrochlorofluorocarbons, hydrobromofluorocarbons [66], although climate change and greenhouse gasses such as methane and nitrous oxide may also have an effect [67]. Looking at the results of the baseline scenario, the electricity (DE, BE and PT) is responsible for around for around 50% of the ozone depletion indicator (kg CFC-11 $_{eq}$). This is mainly related to the electricity production from fossil fuels (primarily natural gas and hard coal), as well as biofuels because of the associated NO_2 emissions of the combustion [68]. In ES the impact of the electricity is smaller, and therefore, the overall value of the indicator, because the proportion of electricity production by natural gas and hard coal is also smaller in comparison to the rest of the countries. Secondly, Ionizing Radiation comprises wavelengths between 10^{-8}-10^{-15} m (UV, X-ray and Gamma rays, for instance) which are very high in energy. It can cause biological effects, particularly change of molecules within the cell and is proven to have a carcinogenic effect, malformation, growth retardation and impaired brain function [69], [70]. The radiation is emitted by radioactive materials (such as Uranium, Plutonium, etc.) called radionuclides and taken into account in the LCA method, measured in units of Bq Carbon-14 $_{eq}$. The extraction, processing and disposal of radionuclides for nuclear energy production are a major source of ionizing radiation [71]. Consequently, countries dependent on electricity production by NE are most likely to have higher values on the IR indicator. In the case of BE (37% nuclear power share, Fig. 2), around 70% of the indicator corresponds to the electricity consumption in the plant. This is the case of BE that with 37% of the electricity mix composed by NE, and around 70% of the indicator is attributed to the electricity consumption in the plant. The trend is followed by ES (25% of NE), DE (11% of NE) and PT (0% of NE).

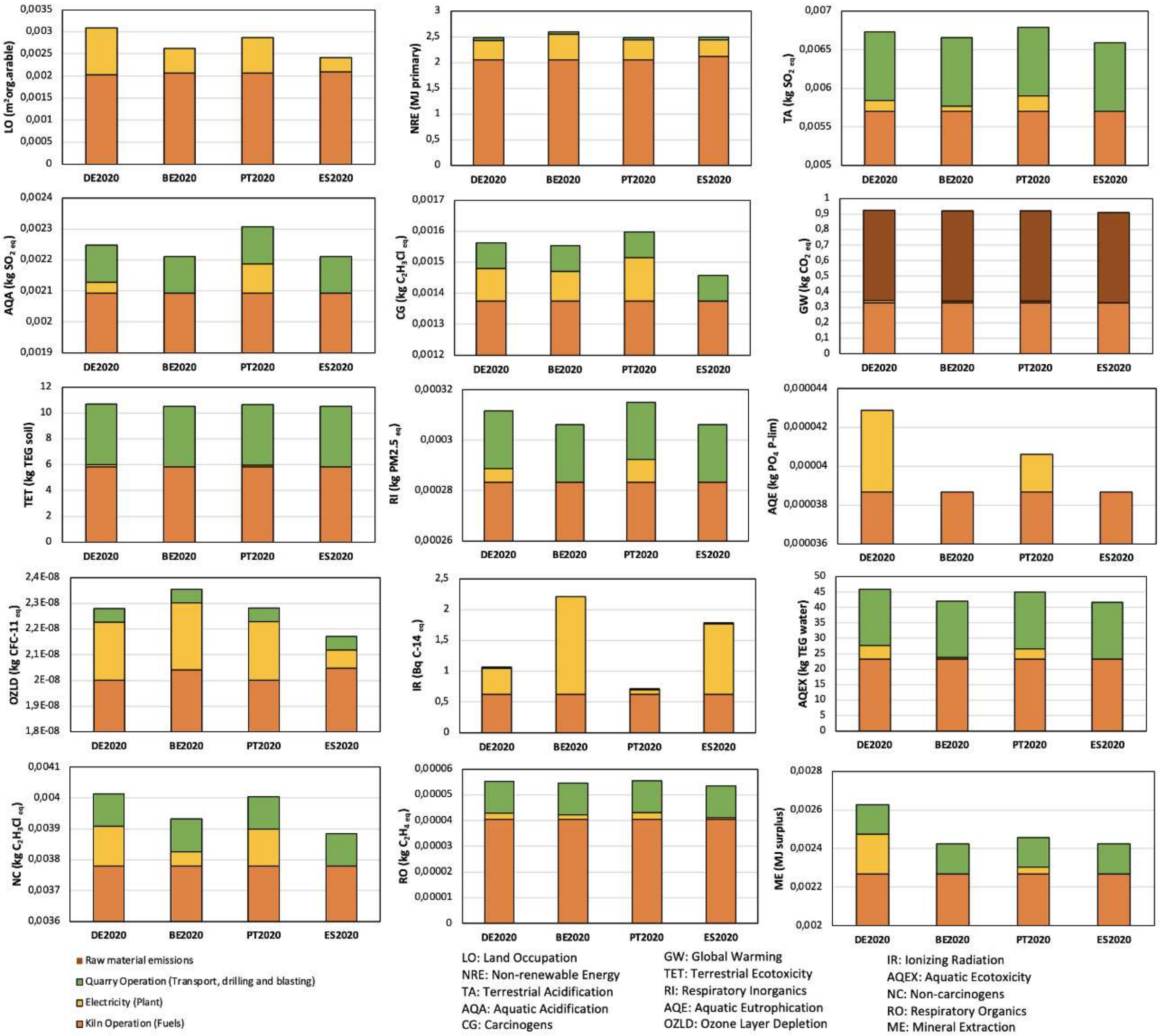

Figure 4. Life Cycle Impact Assessment of 1 kg Hydrated Lime in 2020 for DE, BE, PT and ES.

3.2.2 The current vs. potential future scenarios

The scenario analysis involves the simultaneous effect of switching to a decarbonized electricity matrix and the potential CO_2 savings due to a lower proportion of solid fossil fuels used in the kiln. Figures 5-8 show the LCA results for the 2030/2050 electricity mix matrix and the S1, S2 and S3 fuel mix scenarios along with the baseline results shown in the previous section, for comparison of the evolution.

Regarding the fuel mix used in the kiln, the shift to alternative sources have a positive effect to reduce the $CO_{2\ eq}$ emissions. Specifically, the GWP indicator is reduced by around 9, 18 and 22% for S1, S2 and S3 respectively in comparison to the current scenario (DE, BE, PT and ES). This effect is produced mainly due to the reduction in the use of FSF and the increment in the proportion of NG and Biomass in the mix. For the best-case (S3), the proportion of NG (60%) was almost doubled and Biomass (32%) increased sixteen times compared to the baseline scenario. Nowadays, it is believed that as the power sector undergoes a low-carbon transformation, natural gas is the only fossil fuel technology likely to remain an important source of flexibility for the power systems in the future [72]. The previous results corroborate the prediction of potential CO_2 emissions savings for the analysed scenarios in Figure 3. Nevertheless, despite the fact that the GWP improves under these circumstances, the performed LCA reveals that other impact indicators are negatively affected. As shown in the previous section, is interesting to note that for some of these indicators, the sources of the electricity mix used in each country contribute to a significant extent, and cannot be disregarded.

Speaking of the Ecosystem quality category, the Land Occupation midpoint category is still of interest. For all the considered countries of production, it is observed that the contribution of the kiln operation to the indicator decreases along with the replacement of the fossil solid fuel (Fig. 5). In the potential future scenarios, for DE, PT and ES the contribution of the electricity mix decreases with the increment of renewable sources. However, the opposite is true for BE, where the indicator appears to be highly sensitive to the increment of the biogas co-generation by 15% (Fig 5). An opposite behaviour is observed for the Terrestrial Ecotoxicity indicator, with an increasing trend as new scenarios with potential CO_2 emission savings are proposed. This midpoint category represents the environmental impact of metals released into the soil. The main idea behind the indicator is that it distinguishes between Lewis acids and Lewis basis and considers the strength of the metal complexation and toxicity, with Cr(VI), Sb(III), Sb(V), As(III) and As(V) being most toxic in soil because of their ability to bind with organic matter [53], [73]. Not surprisingly, the Quarry Operation emerges as a very important supplement, because the drilling and blasting step that uses explosives in the quarry to break the limestone are additional sources of heavy metals [74]. The other predominant step is the Kiln Operation. In the baseline scenario, around 40% of the TE is due to the effect of the hard coal in the fuel mix, given that the coal mining activity can affect the ecosystem by the release of Lead and Cadmium (high mobility in the soil-plant system)[75], [76]. The TE indicator increases with the increment of the biomass proportion in the mix (from 2% to 32%), which might be attributed to the effect of the forestry activity [77].

In connection to the Human health damage category, for the Ozone Layer Depletion, around 45% of the indicator corresponds to the Kiln Operation, again, because of the combustion of the fossil fuels. The indicator also appears to be sensitive to the diesel consumption in the Quarry Operation. For the subsequent scenario analysis, an increase in the use of renewable sources of electricity production has a positive impact, diminishing its fraction of the OZLD category. Nevertheless, the indicator increases globally and this is essentially because of the increment of natural gas as fuel for the kiln. Moreover, in section 3.2.1 the implications of using nuclear energy in terms of ionizing radiation were discussed. In general, all the analysed countries are planning to completely replace nuclear energy in the future (Figure 2) and, as a result, the influence of the electricity in the scenario analysis is depreciable. However, fuels also hold responsibility on this regard. In particular, the use of hard coal (and other fossil sources) also contributes to IR, because during the burning process, wastes containing small amount of naturally-occurring radioactive materials are generated [78]. Therefore, as expected, the Kiln Operation also contributes to the indicator, although in a lower proportion (reduction of the sharing between 5-30%).

Finally, covering the Resources-related damage category, the Non-renewable energy indicator ($MJ_{total\ primary\ non\text{-}renewable\ energy}$/mass or volume) is featured. The midpoint category considers for the calculations, the consumption in terms of the total primary energy extracted for energy carriers and the upper heating values of the energy source [53]. The category is almost entirely dominated by the Kiln operation, where the highest consumption of Non-renewable energy is produced. It may be interesting to note that as the shift to less carbon-intensive sources is achieved (replacing fossil solid sources by natural gas); the indicator does not improve but on the contrary. To understand the reason behind, a detailed analysis of the inventory to produce 1 MJ of thermal energy from NG and HC was performed (Ecoinvent V3.6, see Table A5 for reference of the used providers). For NG, the Impact2002+ method assigns an impact factor of 38.3 $MJ_{primary}/m^3$ and 0.033 m^3 of natural gas are required as the only flow contributing to the indicator (aprox. 1.2 $MJ_{primary}$/MJ delivered). Furthermore, in the case of HC, 0.0021 kg of crude oil and 0.0013 kg brown coal are part of the inventory with an assigned impact factor of 45.8 $MJ_{primary}$/kg and 9.9 $MJ_{primary}$/kg respectively (0.12 $MJ_{primary}$/MJ delivered). In all cases, the contribution of the Electricity and the Quarry Operation is not significant. In retrospect, it may be important to mention the Mineral Extraction indicator, as it complements with the Non-renewable energy indicator to assess the resource depletion. It is measured in $MJ_{surplus}/kg_{extracted}$ and expresses the expected increase in extraction energy needed to extract 5 times the cumulative extracted amount, considering that the resources become scarcer with the time [64]. It may be worth mentioning that even though it is related to the extraction process of limestone (because of the specific energy consumption in the LCI, Table 3), this indicator is referring to the energy used in the process and not the depletion of the limestone mineral itself. In all cases, three main components of the indicator can be distinguished: Quarry Operation, Electricity and Kiln Operation. For the Quarry Operation, the contributing sub-processes are the blasting and explosive production, accounting for around 6-8% of the total indicator depending on the analysed country. However, the indicator is mostly dominated by the fuels providing heat to the kiln in the first place, and the electricity to power the plant on the second place. In the current scenario (Figure 4), for DE2020 the indicator is around 8% higher than the average, because of the current electricity matrix and in particular, due to the hard coal, natural gas and biogas sources. Hard coal and natural gas used as fuel in the kiln are also the main components of the Kiln Operation share. In the future scenarios, the switch to a lower $CO_{2\,eq}$ fuel matrix actually does not improve the indicator, because the reduction in the impact associated to the hard coal is compensated by the increased effect of the natural gas and the biomass production (which also includes the energy required to dry the wood chips, as explained in Section 3.2.1).

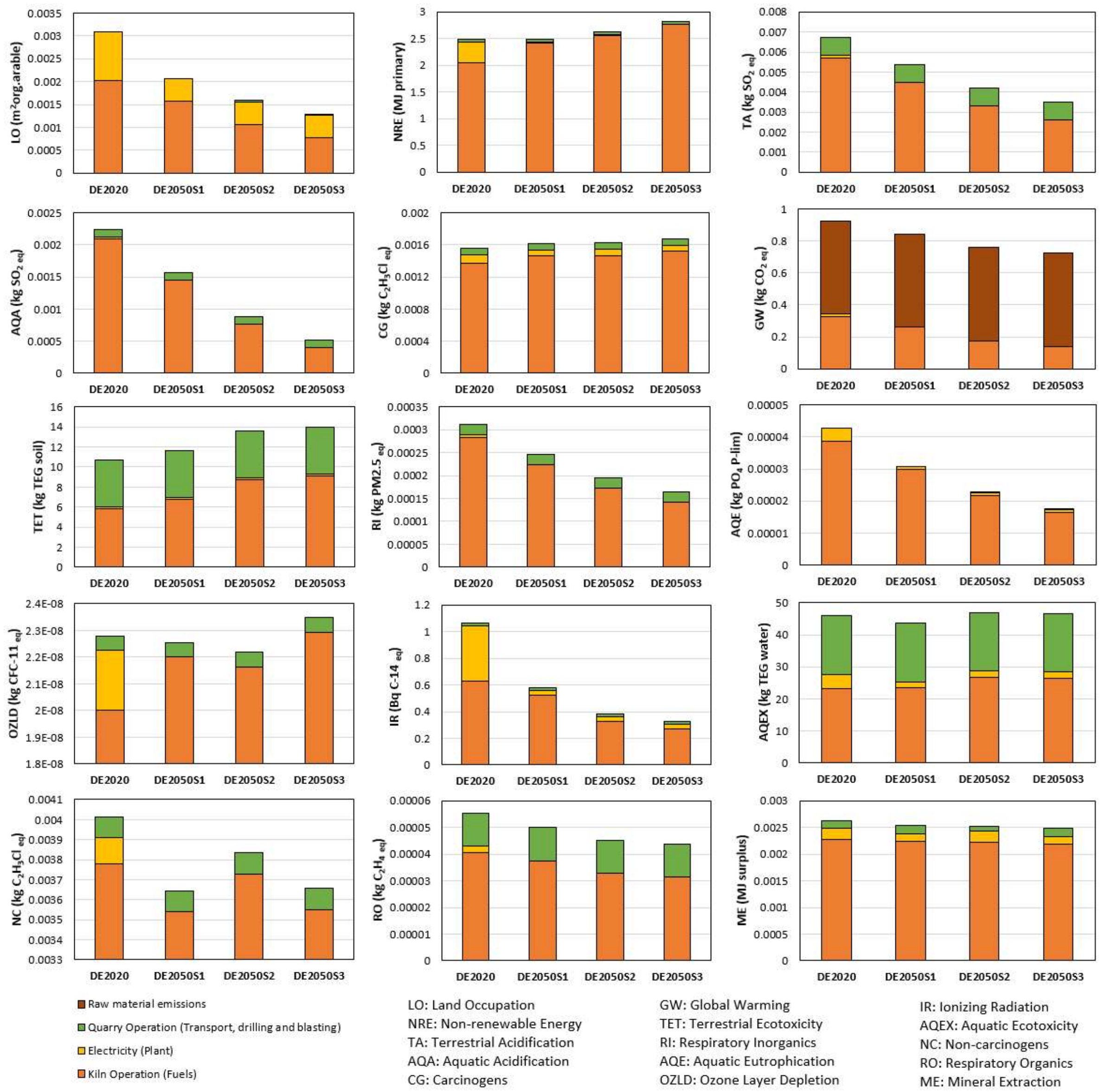

Figure 5. Life Cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2050) in DE

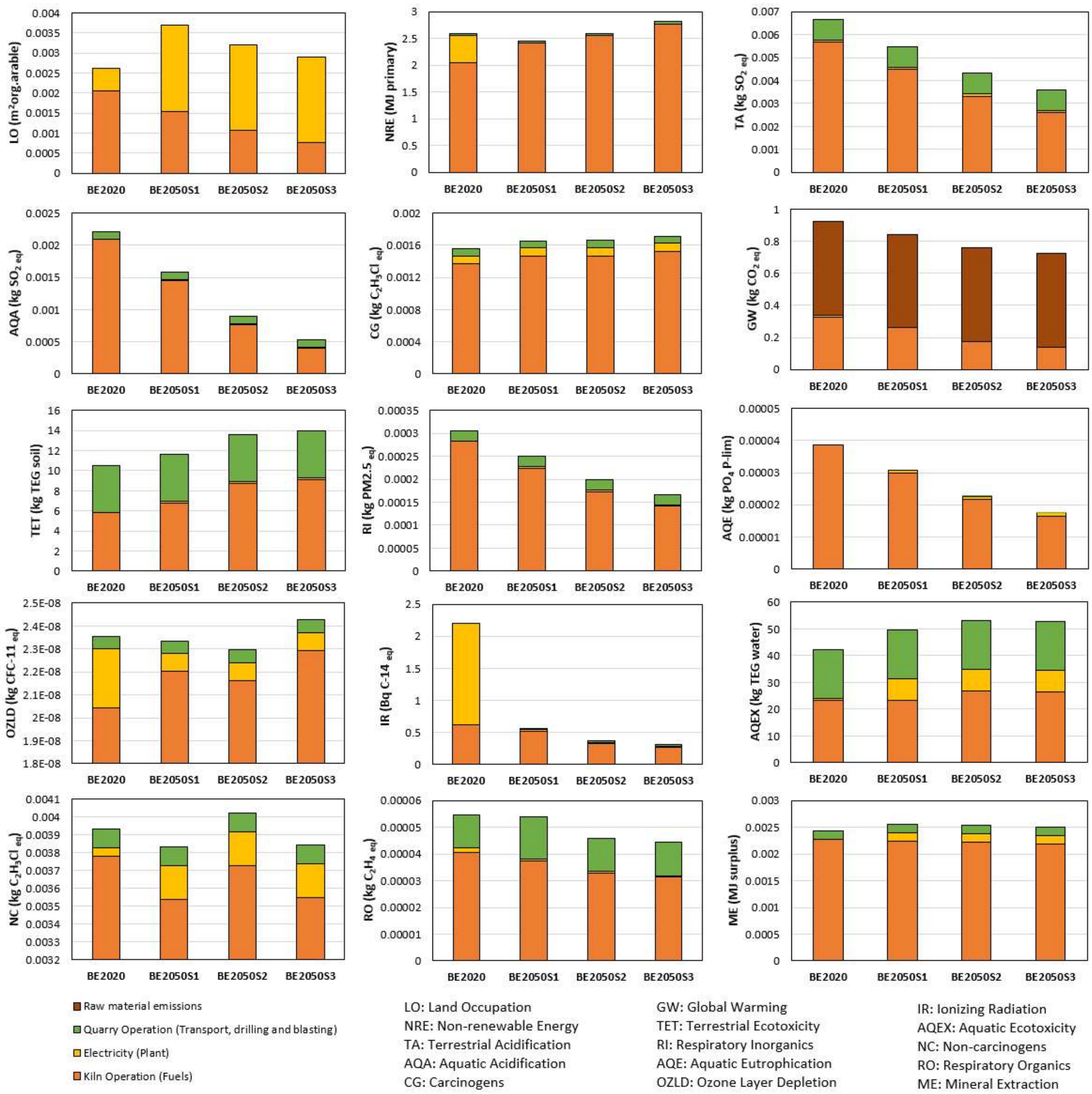

Figure 6. Life Cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2050) in BE

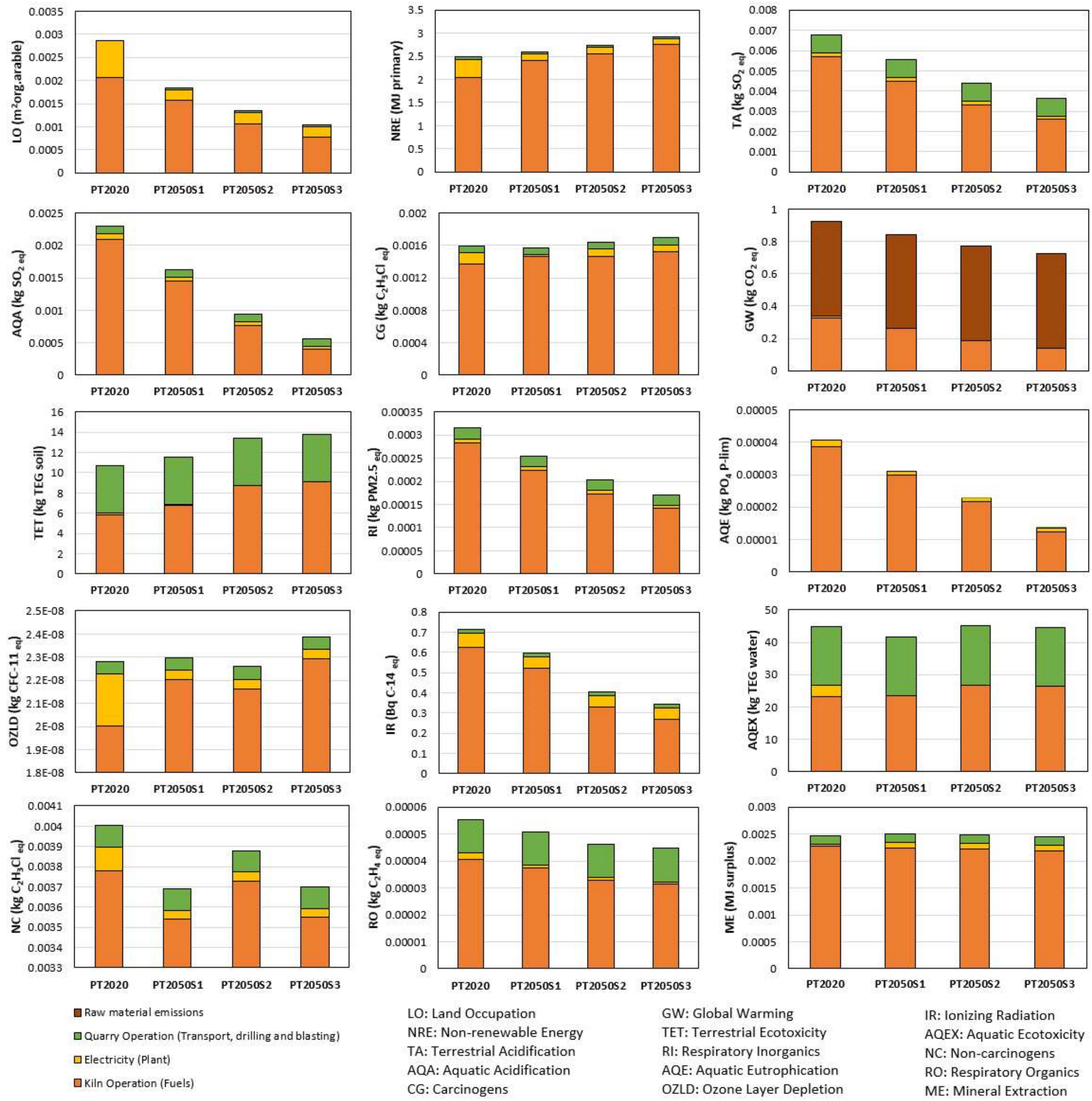

Figure 7. Life Cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2050) in PT

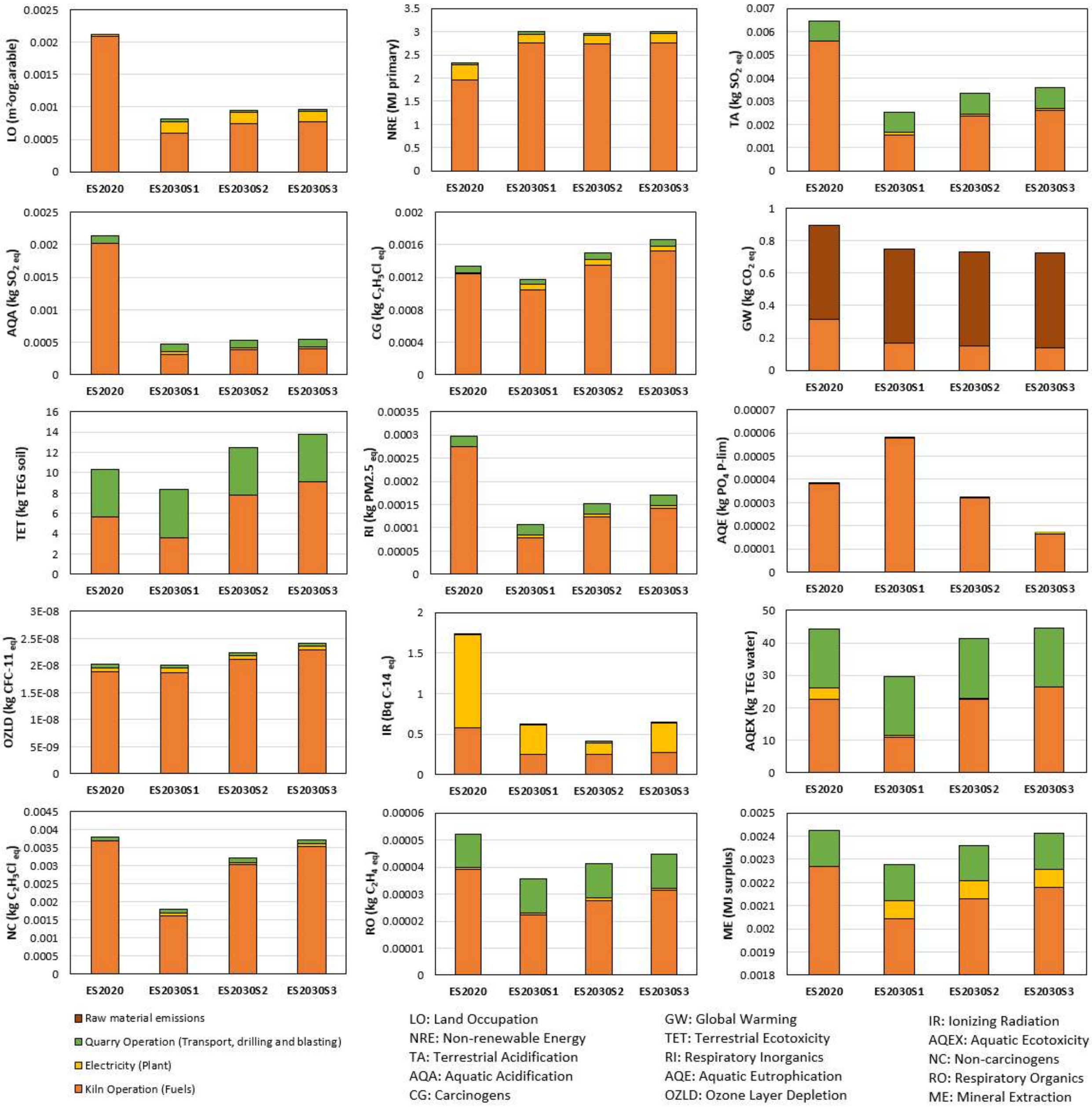

Figure 8. Life Cycle Impact Assessment of 1 kg Hydrated Lime – Current (2020) and potential future scenarios (2030) in ES

3.2.3 Final comments

First a few words about the inventory as such. Along the study the contribution of the fuel mix to the environmental impact was highlighted. During the Life Cycle Inventory analysis, it is relevant to highlight that the results are subjected to the approximation that the selected most representative kiln process from EcoInvent 3.6 database also represent the combustion taking place in the lime kiln. However, as the process specific emissions related to the kiln technology could play a relevant role as well, this needs to be re-examined. This study is aimed at providing a reference document that serves as a first approximation to the listed scenarios and is open for potential improvement in the future whenever more detailed information on life cycle inventories becomes available.

Moving forward to the analysis of the results, Figure 9 aims at summarizing the workflow of the scenario analysis development, as well as emphasizing the main findings of the research carried out. As revealed in the literature review, for the building of the scenario analysis, Europe has a great potential to produce energy with a lower carbon footprint. Currently (baseline scenario 2020), the share of the renewable energy for electricity production for the analysed countries is around 30, 39, 45 and 60% for BE, ES, DE and PT respectively (Figure 2). Nevertheless, the results show that not only the amount of RES feeding the mix is important, but also what type of non-renewable sources are being used. In particular, the indicators have proven to be highly sensitive to the proportion of hard coal and natural gas. Therefore, for the current scenario (Figure 4), ES and BE have shown the lower environmental impact, namely in 7 out of 15 indictors and 5 out of 15, respectively, followed by PT (1 out of 15) and DE (1 out 15). As a key remark, in 12 out of 15 indicators, the electricity mix contributes to a meaningful extent to their magnitude. Naturally, the fuel used in the kiln is also significant in all the impact categories analysed and dominates around 13 of the indicators. This fact is, once again, attributed to the FSF.

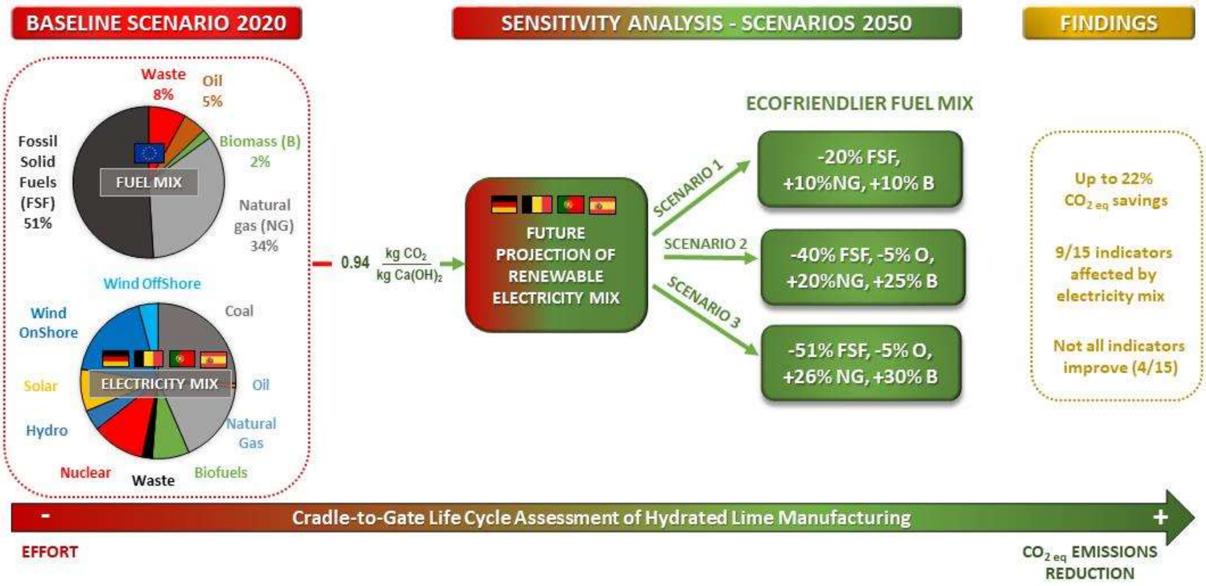

Figure 9. Workflow of the scenario analysis development and main findings of the research

In the future (sensitivity analysis), all the analysed countries are planning to base their electricity production on the use of two main sources: Wind power along with solar energy. These sources would dominate the sharing, amounting to around 70% of the market (Figure 2). In a lower proportion, other sources such as hydropower, biofuels and combined cycles appear as an alternative. The shift to almost 100% renewable energies for electricity production, certainly has a positive effect on the impact indicators, reducing their shared proportion in around 12 out of 15 indicators. Nevertheless, the opposite holds true for the different fuel mix scenarios. Whereas significant reductions of $CO_{2\ eq}$ emissions are obtained by reducing the amount of FSF, for the analysed countries, the indicators NRE, CG, TET and OZLD increases up to 20, 8, 25 and 9% respectively (in comparison to 2020 scenario). While common sense would suggest that replacing current energy sources with those with a lower carbon footprint would lead to an improvement in all environmental indicators, the results show that this is not trivial. Therefore, it supports the idea that the issue of environmental sustainability cannot be limited to GWP alone, but requires a holistic analysis of the situation, using all available indicators and tools to interpret the reality reliably. Furthermore, it is important to understand that sustainability is not simply a matter of reducing environmental impacts. Instead, the appeal of low-carbon and green-growth development paths stems from the desire to avoid 'locking in' to development paths that may become increasingly costly as resources become scarcer and carbon pricing becomes more important over time [79]. As mentioned during the analysis of potential fuel mix scenarios, achieving sustainability in the lime industry and therefore, reaching the goals of clean and eco-friendly production proposed by the UN, requires the cooperative collaboration of scientists, policymakers, the private sector, and society as a whole.

4. Conclusion

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

- A theoretical Cradle-to-Gate LCI for Hydrated Lime production was developed and a LCA was performed to compare the results with databases (EcoInvent V3.6) and reports by specific institutions (European Lime Association and European Environmental Agency). The environmental impact analysis of the theoretical study is in good agreement with results of the European Lime Association (7% relative difference for the Global Warming Potential indicator)
- For the 2020 scenario, in 9 out of 15 indicators, the share of electrical energy consumption in the plant is not negligible (from 5 to 50% relatively). The most affected indicators were Land Occupation, Aquatic Eutrophication, Ozone layer depletion and Ionizing Radiation. Among the first three the presence of fossil fuels dominates, while for Ionizing Radiation dominates the use of nuclear energy. In terms of Global Warming Potential, 0.94 kg CO_{2eq}/kg HL are produced. The emissions are due for 60% to limestone decomposition, 39% is attributed to the combustion of the fuel and 1% to the electricity consumption. Spain and Belgium have shown lower environmental impact, namely in 7 out of 15 indictors for Spain and 5 out of 15 for Belgium, followed by Portugal (1 out of 15) and Germany (1 out 15).
- Regarding the potential future scenarios, on the one hand, the shift to almost 100% renewable energies for electricity production, has a positive effect on the impact indicators, reducing their shared proportion in around 12 out of 15 indicators. Furthermore, as the proportion of fossil solid fuel decreases and NG and biomass increases in the fuel mix, a reduction of 9, 18 and 22% in the Global Warming Potential is achieved (compared to the 2020 scenario). Nevertheless, the results reveal that although Global Warming Potential is reduced, the indicators Non-renewable energy, Carcinogens, Terrestrial Eco-toxicity and Ozone Layer Depletion increase up to 20, 8, 25 and 9% respectively (in comparison to the 2020 scenario) due to the future fuel mix designs.
- Finally, the result of this research implies that a change from fossil solid fuel to other sources (or a change to renewable energy only) will not necessarily lead to reduction for all impact categories and that a well-informed choice for a combination of energy sources should be made to obtain a balanced reduction for most impacts. Special attention should be paid to low-carbon and green-growth development paths to consider that resources become scarcer and carbon pricing becomes more important over time.

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A. Complementary information: EcoInvent V3.6 processes used to model the Energy Mix

Table A1. Providers of Energy Source for Electricity Production – Germany 2020 and 2050

	Flow	Amount (MJ)	Description	Provider
DE 2020	electricity, high voltage	9	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - DE
	electricity, high voltage	4	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - DE
	electricity, high voltage	4	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - DE
	electricity, high voltage	18	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - DE
	electricity, high voltage	25	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - DE
	electricity, high voltage	17	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - DE
	electricity, high voltage	11	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - DE
	electricity, high voltage	1	Oil	electricity production, oil | electricity, high voltage | APOS, S - DE
	electricity, high voltage	9	Solar	electricity production, solar thermal parabolic trough, 50 MW | electricity, high voltage | APOS, S - RoW
	electricity, medium voltage	2	Waste	electricity, from municipal waste incineration to generic market for electricity, medium voltage | electricity, medium voltage | APOS, S - DE
DE 2050	electricity, high voltage	3	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - DE
	electricity, high voltage	7	Geothermal	electricity production, deep geothermal | electricity, high voltage | APOS, S - DE
	electricity, high voltage	26	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - DE
	electricity, high voltage	34	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	4	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - DE
	electricity, high voltage	26	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - DE

Table A2. Providers of Energy Source for Electricity Production – Belgium 2020 and 2050

	Flow	Amount (MJ)	Description	Provider
BE 2020	electricity, high voltage	28	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - BE
	electricity, high voltage	5	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - BE
	electricity, high voltage	7	Wind OffShore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	7	Wind OnShore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	37	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - BE
	electricity, high voltage	2	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - BE
	electricity, high voltage	5	Oil	electricity production, oil | electricity, high voltage | APOS, S - BE
	electricity, high voltage	5	Solar	electricity production, solar thermal parabolic trough, 50 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	2	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - BE
	electricity, medium voltage	2	Waste	electricity, from municipal waste incineration to generic market for electricity, medium voltage | electricity, medium voltage | APOS, S - BE
BE 2050	electricity, high voltage	24	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	17	Wind Onshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - BE
	electricity, high voltage	3	Geothermal	electricity production, deep geothermal | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	36	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	20	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - BE

Table A3. Providers of Energy Source for Electricity Production – Portugal 2020 and 2050

	Flow	Amount (MJ)	Description	Provider
PT 2020	electricity, high voltage	3	Solar	electricity production, solar thermal parabolic trough, 50 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	7	Biofuels	heat and power co-generation, biogas, gas engine | electricity, high voltage | APOS, S - PT
	electricity, high voltage	2	Oil	electricity production, oil | electricity, high voltage | APOS, S - PT
	electricity, high voltage	4	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - PT
	electricity, high voltage	12	Wind OffShore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - PT
	electricity, high voltage	12	Wind OnShore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - PT
	electricity, high voltage	24	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - PT
	electricity, high voltage	27	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - PT
	electricity, high voltage	9	Fossil Co-generation	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - PT
PT 2050	electricity, high voltage	26	Wind OnShore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - PT
	electricity, high voltage	37	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	22	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - PT
	electricity, high voltage	4	Natural gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - PT
	electricity, high voltage	11	Wind Offshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - PT

Table A4. Providers of Energy Source for Electricity Production – Spain 2020 and 2030

	Flow	Amount (MJ)	Description	Provider
ES 2020	electricity, high voltage	9	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	2	Natural Gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	12	Wind OffShore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - ES
	electricity, high voltage	20	Combined cycle	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	12	Fossil Co-generation	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	6	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - ES
	electricity, high voltage	2	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - ES
	electricity, high voltage	25	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - ES
	electricity, high voltage	12	Wind OnShore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - ES
ES 2030	electricity, high voltage	1	Coal	electricity production, hard coal | electricity, high voltage | APOS, S - ES
	electricity, high voltage	15	Hydro	electricity production, hydro, pumped storage | electricity, high voltage | APOS, S - ES
	electricity, high voltage	2	Fossil Co-generation	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	2	Nuclear	electricity production, nuclear, pressure water reactor | electricity, high voltage | APOS, S - ES
	electricity, high voltage	16	Wind Offshore	electricity production, wind, 1-3MW turbine, onshore | electricity, high voltage | APOS, S - ES
	electricity, high voltage	17	Combined cycle	electricity production, natural gas, combined cycle power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	1	Natural Gas	electricity production, natural gas, conventional power plant | electricity, high voltage | APOS, S - ES
	electricity, high voltage	30	Solar	electricity production, solar tower power plant, 20 MW | electricity, high voltage | APOS, S - RoW
	electricity, high voltage	16	Wind Onshore	electricity production, wind, 1-3MW turbine, offshore | electricity, high voltage | APOS, S - ES

Table A5. Providers of Energy Source for Kiln Fuel Feeding – Europe 2020 and Future Scenarios S1, S2, S3 in 2050

	Flow	Amount (MJ)	Description	Provider
2020	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 industri al furnace 1-10MW | Cut-off	51	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland
S1	Heat, district or industrial, natural gas {Europe without Switzerland}| heat production, natural gas, at industrial furnace >100kW | Cut-off, S	44	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	heat, district or industrial, other than natural gas	5	Oil	heat production, heavy fuel oil, at industrial furnace 1MW | heat, district or industrial, other than natural gas | 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, other than natural gas	12	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	31	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland
S2	Heat, district or industrial, natural gas {Europe without Switzerland}| heat production, natural gas, at industrial furnace >100kW | Cut-off	54	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	heat, district or industrial, other than natural gas	8	Waste	heat, from municipal waste incineration to generic market for heat district or industrial, other than natural gas | heat, district or industrial, other than natural gas | APOS, S - DE
	heat, district or industrial, other than natural gas	11	Fossil Solid Fuels	heat production, at hard coal industrial furnace 1-10MW - Europe without Switzerland
	heat, district or industrial, other than natural gas	27	Biomass	heat production, wood chips from industry, at furnace 1000kW | heat, district or industrial, other than natural gas | APOS, S - DE
S3	Heat, district or industrial, natural gas {Europe without Switzerland}| heat production, natural gas, at industrial furnace >100kW | Cut-off, S - Copied from Ecoinvent	60	Natural gas	heat production, natural gas, at industrial furnace >100kW - Europe without Switzerland
	heat, district or industrial, other than natural gas	32	Biomass	heat production, wood chips from industry, at furnace 1000kW | heat, district or industrial, other than natural gas | APOS, S - DE
	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