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► To cite this version:

Hiam Dahanni, Anne Ventura, Laurédan Le Guen, Michel Dauvergne, A. Orcesi, et al.. Life cycle assessment of cement: Are existing data and models relevant to assess the cement industry's climate change mitigation strategies? A literature review. *Construction and Building Materials*, 2024, 411, pp.134415. 10.1016/j.conbuildmat.2023.134415 . hal-04387131

HAL Id: hal-04387131

<https://hal.science/hal-04387131v1>

Submitted on 11 Jan 2024

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Life cycle assessment of cement: are existing data and models relevant to assess the cement industry's climate change mitigation strategies? A literature review.

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ABSTRACT: The cement industry's strategies for achieving carbon neutrality are mainly based on different types of CO₂ mitigation strategies. In this article, we will explore three of these strategies (i) material efficiency strategy which consists in improving cement quality to reduce the quantities needed to maintain its performance, (ii) cement plant technologies which consist in improving the thermal efficiency in the kiln, improving grinding electrical efficiency, and low carbon fuel utilization, (ii) clinker-to-cement ratio reductions which consists in substituting clinker by less energy-intensive Supplementary Cementitious Materials (SCMs) derived from waste recovery. This paper investigates the existing literature published in the last decade about the Life Cycle Assessment (LCA) of cement production, in order to investigate if these strategies can actually be modeled. For the material efficiency strategy, we analyze how functional units of cement are defined in the literature. From a simplified system representing cement production, we represent energy and greenhouse gas (GHG) flows that can be affected by the mitigation strategies and we analyze if existing detailed data can be obtained and how they are modeled, considering especially allocation methods.

Our results show that heating energy is the main contributor to energy consumption compared to electric energy used for crushing and grinding materials. Concerning heating energy, some data clearly demonstrate the beneficial influence of energy efficiency measures. When specified, the allocation of the impacts of the production and combustion of secondary fuels is always in favor of cement (not accounted for in its life cycle). Thanks to allocation, the use of secondary fuels thus significantly reduces the energy and GHG emissions attributed to cement. However, better transparency practices are needed to clarify the origin and calculation of combustion-related emissions.

Concerning grinding energy, very little data are available, and none on the additional energy required to produce higher-quality cement, which would be necessary to take into account the material efficiency strategy. When (rarely) specified, the functional unit is defined by the cement's compressive

strength, but no link is made with the reference flow, which should distinguish different quantities according to this performance. We propose a method for performing this calculation.

The available data allowed us to analyze the effect of using Ground Granulated Blast Furnace Slag (GGBS) as a SCM. Different GGBS production process variations significantly affect the environmental effects, and these variations should be taken into account in LCA models. Unsurprisingly, allocation assumptions are also highly influential. The choice of allocation method significantly affects the results, with theoretical representations suggesting that the Partition method (both Mass Allocation - MA and Economic Allocation - EA) would yield the highest impacts, followed by Cut-off (CO) and System expansion with Substitution (SE). The specific system boundary adjustments in each method lead to distinct outcomes, thus providing valuable insights into waste recovery strategies. The implications of these methods for stakeholders show how environmental and political considerations are connected.

Whether for secondary fuels or for SCM, existing LCA methods for modeling substitution never take into account the actual availability of these recovered wastes in relation to the cement industry's needs. Geographical disparities exist and should be taken into account specifically for each cement production site.

1 INTRODUCTION

Cement manufacturing is reported to be one of the main contributors to global greenhouse gases emissions (GHGs) [1–3].

Different types of strategies have been identified to decrease the GHG emissions of the cement industry.

The first type of strategy is the material efficiency strategy, also considered as an important issue for climate change mitigation. In that line, using less cement should be considered. However, the global population growth and economic development suggest that the demand for concrete, and thus for cement, will globally increase rather than decrease. A reduction or stagnation in cement demand is still possible if cement quality is improved, requiring less cement for the same concrete performance.

The second type of strategy which is cement plant technologies concerns direct decrease of GHG emissions at the cement plant. For instance, [4] have presented various strategies such as: decreasing energy consumption by increasing energy efficiency of clinkerization kilns, reducing the temperature for calcination, shifting from fossil heating energies to renewable ones or to incineration of waste.

The third type is the clinker-to-cement reduction strategy which consists in decreasing the amount of clinker by substituting it with SCMs. The implementation of these strategies should provide cements with lower impacts, especially on global warming. The cement industry names the results of these strategies as producing “low carbon cement».

To assess the effectiveness of these strategies, LCA is an appropriate method. However, this requires suitable cement plant inventory models capable of producing results according to the different strategies implemented.

The material efficiency strategy raises the question of the functionality of cement for its use in concrete. Reducing the quantity of cement for a given use in concrete corresponds, in LCA, to calculating a Reference Flow (RF) corresponding to a desired functionality called Function Unit (FU). Assessing the effect of improving cement quality therefore comes down to defining its functionality.

Concerning the second and third type of strategies, i.e. direct decrease of GHGs emissions, Figure 1 depicts the different unitary operations involved in the cement production process, that are responsible for energy consumption and GHG emissions.

- In the raw material preparation operation (see system for cement in Figure 1), limestone and clay, and other selected materials are crushed, pre-blended, and homogenized to obtain the targeted particle size and chemical composition before the clinkerization process (calcination). During this operation, crushing and grinding require significant electricity consumption which contributes to GHG emissions[5,6] according to the electric mix.
- During the clinkerization operation (see system for cement in Figure 1), a mix of raw materials (commonly limestone and clay) is calcinated at a high temperature in a rotary kiln. This calcination process (the resulting product being called clinker) is an energy-intensive phase in cement production since it requires burning fuel to reach the target temperatures in the kiln. GHG emissions are thus due to both the combustion of fossil fuels to provide the heating energy, and the decarbonation of limestone that is a chemical reaction emitting CO₂ during calcination ($\text{CaCO}_3 + \text{clay} + \text{other materials} \rightarrow \text{CO}_2 + \text{cl} + \text{CaO}$) [2,5];
- The final operation consists in grinding the clinker (see system for cement in Figure 1) to produce a cement that achieves the desired fineness. This operation can either be conducted on:
 - o clinker mixed with a small addition of gypsum and/or limestone powder giving Ordinary Portland Cement (OPC),
 - o clinker mixed with SCMs that are mainly by-products from other industrial activities giving blended cement.

The grinding process requires electricity consumption [2] and thus GHG emissions will also depend on the electric mix.

Using less clinker by its substitution with SCMs mostly issued from recovery of industrial waste. The modeling of that action lever can consider different possible allocation methods shown in Figure 1. The allocation methods are also involved in the substitution of fossil fuels of the heating energy (energy 3 in Figure 1) issued from waste incineration although it is not represented in Figure 1 for clarity.

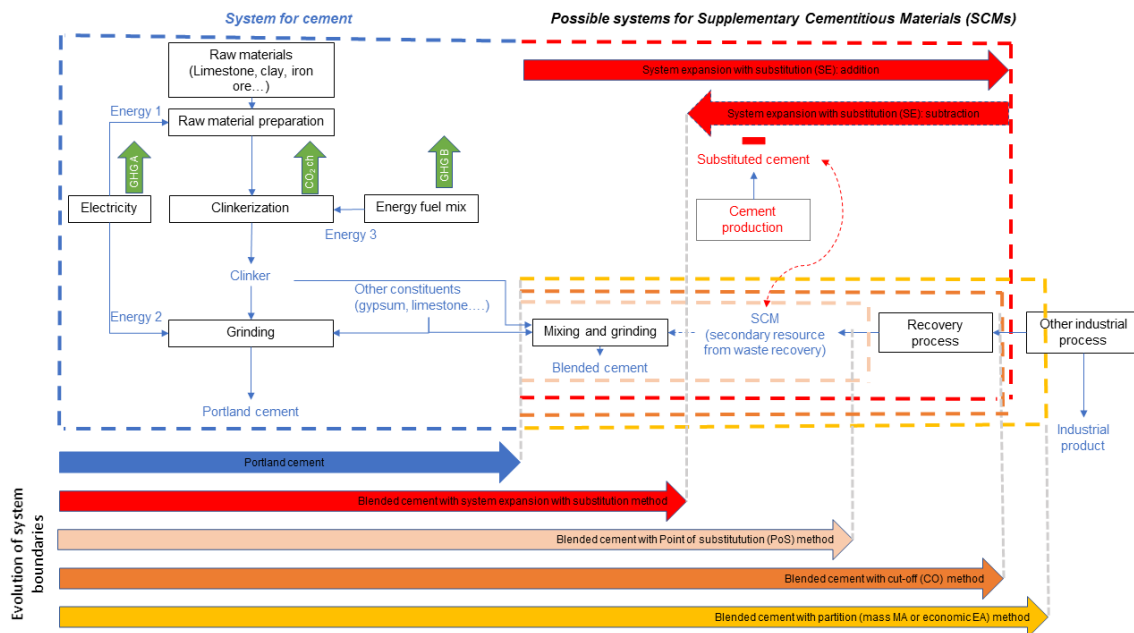


Figure 1: General scheme representing the main processes of cement manufacturing, energy consumptions and GHGs emissions and possible system boundaries according to the chosen allocation method for blended cement

The objective of this paper is to investigate if existing LCA models and data are suitable to assess the strategies presented in this paper. A literature review is conducted on LCAs of cement manufacturing.

To consider the material efficiency and clinker-to-cement ratio reduction strategies, the possible properties of cements will be analyzed from literature, to get a better understanding between various properties of cements. Indeed, increasing the quality of cement (actual strength of cement) or adding SCMs actually changes the chemical nature of cements and their properties. Thus, more or less cement can be required for a given usage inside concrete, according to its strength, the type of SCMs and their amounts. Reducing or increasing the amount of cement in concrete will of course change its environmental impacts. Thus, defining cement functionalities according to their properties, as well as calculating the corresponding RF according to these functionalities appears to be necessary to assess the environmental impacts of so-called “low carbon cements”. We will thus investigate how existing LCA studies consider this aspect.

To consider the second strategy, factors expected to have the greatest impacts in terms of Global Warming. This strategy will be investigated by analyzing either CO₂ emissions and CO₂ equivalent emissions (represented with green arrows in Figure 1) expressed according to The Global Warming Potential (GWP100) indicator provided by the investigated LCA studies. The different GHGs sources will be examined on OPC because that process always occurs whatever the type of cement produced (blended or not, i.e. with or without SCMs). The following data are extracted from our set of investigated references:

- the amount of electricity consumption for the crushing processes and the corresponding GHG emissions according to the electric production mix (GHG A in Figure 1),
- the amount of the heating energy for clinkerization and the corresponding GHG emissions according to the fuel mix used for combustion during calcination (GHG B in Figure 1),

- the amount of CO₂ emitted by the decarbonation of carbonates during the calcination process (CO₂ ch in Figure 1).

The influence of SCMs on the GWP indicator will be analyzed from our set of investigated references, according to their amounts as well as to the influence of the allocation methods used by their authors. The same analysis will also be conducted concerning the type of fuels used for heating energy.

2 METHODOLOGY

2.1 Selection of references

The literature review is focused on LCA studies published over the last decade (2012-2022), however data from Ecoinvent database 2007 [7] was also included. The selection of the references was based on the following keywords: (supplementary cementitious materials OR cement SCMs) AND (Life Cycle Assessment OR environmental impact), AND (cement). After conducting a comprehensive search using relevant keywords, a substantial number of references were identified. From this extensive list, the selection was narrowed down by focusing on references that specifically addressed OPC and blended cement with commonly employed SCMs, including GGBS, fly ash, and others.

A collection of 31 papers was obtained, from various sources: 17 journal papers, 1 conference paper, 11 Environmental declaration products (EPDs) and 2 reports.

For each reference, the following information was extracted:

- Description of the Reference Flow (RF) and the Functional Unit (FU) – these notions will be used in section 3.1;
- Types and amount of Portland cement and SCM in the cement mix design;
- Total value of Global Warming Potential (GWP100) and detailed values when possible (CO₂ ch, GHG A and GHG B as indicated in Figure 1);
- Allocation method used for the system boundary concerning SCMs from industrial by-products (cut-off, mass or economic allocation, system expansion with substitution, at point of substitution these methods will be recalled later in the article in section 3.3);
- Amount of operational energy flows: electricity for preparation (energy 1 in Figure 1) and grinding (energy 2 in Figure 1), energy for calcination (energy 3 in Figure 1);
- Considered life cycle phases (cradle to gate or cradle to grave);
- Country of cement production.

The 31 examined papers were filtered keeping those providing a result concerning GWP indicator, providing a total of 102 data for cement (each reference possibly providing several data corresponding to several case studies). We only considered data with no SCMs and with a single SCM and not with mixed SCMs, in order to better analyze the change of impacts for each possible SCM. We obtained 25 data for OPC (cement with no SCMs) and 77 data for blended cements [8–38]. Calculation of energy flows for cement production from reference data

2.2 Calculations of energy flows for cement production from reference data

This section explains how energy consumptions were calculated when authors did not directly provide the data.

The amount of energy 1 (see Figure 1), was obtained differently from the different references. In the Ecoinvent database 3.7 cut-off [39], the amount of electricity required for crushing gypsum, limestone powder and the constituents of clinker (crushed limestone, calcareous marl, clay) is considered. Each electricity quantity is multiplied by the amount of material needed to be produced. Thus, the total amount of these electricity quantities is summed up and is reported in Table 1. For other references, the energy amount for the primary grinding are provided by authors [10,14,18,25] and directly reported in Table 1.

The amount of energy 2 (see Figure 1) was obtained from the unit process of the cement plant in the Ecoinvent databases 2 and 3.7.1 cut-off [7,39] and directly collected from the other references [10,14,25]. The total electricity generated by the cement plant (energy 1+ energy 2) was given by [15].

The amount of energy 3 (see Figure 1) was obtained from the mass amounts of heating fuels for the production of 1 kg of clinker as provided by [39] and [25]. The total heating energy reported in Table 1 is thus calculated by multiplying the amount of each fuel by its heating value provided in the Ecoinvent report [7]. The amounts of fossil fuels and alternative fuels, expressed in energy units and reported in Table 1, were directly provided in the study by [10]. In the study of [15], the authors provided the coal combustion for incineration and the thermal energy, both were added to obtain the total heating energy as reported in Table 1. Energy 3 was also directly provided for other references [9,18]. The energy 3 amount that is provided corresponds to the total energy consumed at the cement plant. However, this total amount may not be attributed to the cement's life cycle, especially when secondary fuels are used. This aspect is fully discussed in sections 3.3.3 and 4.3.

2.3 Calculation of GHGs for cement production from reference data

This section explains how GHG emissions were calculated when authors did not directly provide the data.

The GHG A is obtained from the Ecoinvent 3.7 cut-off database [39] by calculating the electricity process contribution to the total GWP of cement. Some references only provided the GHG emissions resulting from the final grinding process [39], other references specified both from primary crushing and final grinding [10], while other references did not specify any emission [15,17,18].

GHGs B are estimated from the amounts of each energy intermediate input flow corresponding to the energy fuel mix that are multiplied by their CO₂ eq emissions resulting from their combustion. This is calculated from the carbon content of 1 kg of fuel provided in the database of [7], thus this method assumes that fossil fuels are burned in a complete combustion reaction. Similarly, to the calculation of energy 3, this amount corresponds to the total GHGs emitted from combustion at the cement plant. As highlighted earlier, it's important to note that the total emissions are not always solely attributed to the life cycle of cement, particularly when secondary fuels are employed. A detailed discussion of this aspect will be provided later in this article (see sections 3.3.3 and 4.3).

CO₂ ch emissions (from the decarbonation reaction of the calcination process in Figure 1) were obtained from the subtraction of GHG B from the total GWP of the clinker life cycle for the Ecoinvent 3.7 cut-off database [39] as shown in Table 1. For other references, CO₂ ch was directly provided and is reported in Table 1.

The values provided in these references were checked by calculating the amount of CO₂ ch per kg of cement, from the mass fraction of CaO present in the cement by Equations (1) and (2).

The amount of CO₂ emissions generated by the decarbonation of limestone is a key element of GHG emissions of cement production. As limestone is a primary raw material, it contains impurities that can have a significant influence on the CO₂ ch emissions of the final product. However, it is not possible to determine the exact amounts of CO₂ ch without knowing the exact composition of limestone as well as the exact composition of the input materials mix (i.e. limestone, and each other raw material used in clinker production). Thus, to estimate the range of variation in CO₂ ch emissions, we rely on the Tier 2 method given in the IPCC guidelines [41], based on the chemical reaction that provides CO₂ ch emissions from the calcination chemical reaction providing lime CaO (refer to the calcination equation in section 1). With this method, the value is calculated based on the lime fraction inside cement according to the following equations:

$$CO_{2\ ch} = f_{\frac{CaO}{clinker}} * f_{\frac{clinker}{cement}} * \frac{\text{Molar mass of } CO_2}{\text{Molar mass of } CaO} \quad (1)$$

$$CO_{2\ ch} = f_{\frac{CaO}{cement}} * \frac{\text{Molar mass of } CO_2}{\text{Molar mass of } CaO} \quad (2)$$

where $CO_{2\ ch}$ is the quantity of CO₂ emitted [kg/kg of cement], $f_{\frac{CaO}{clinker}}$ is the fraction of lime in clinker [-], $f_{\frac{CaO}{cement}}$ is the fraction of lime in cement [-] and $f_{\frac{clinker}{cement}}$ is the fraction of clinker in cement [-].

The average amount of lime CaO in clinker can vary depending on the specific raw materials used and the manufacturing process employed. The standard [42] for the classification of cement, CEM I, imposes two conditions:

- the total amount of SiO₂ and CaO must be at least 50% of the cement mass,
- the maximum possible amount of SiO₂ is limited to half of the amount of CaO.

By ensuring that these two conditions are met, it is possible to obtain a minimum CaO value of 33.3 %. This minimum amount results in a minimum CO₂ ch value of 0.26 kg CO₂/kg OPC. The maximum value is calculated assuming limestone without impurities and 95% clinker in the cement, which results in a CO₂ ch value of 0.60 CO₂ ch/kg OPC. The theoretical permissible range of variation of CO₂ ch is therefore between 0.26 and 0.60 kg CO₂ ch/kg OPC.

In practice, these extreme amounts of CaO are not commonly found in clinker. The mass ratio of CaO in clinker commonly ranges between 60 and 67% [7], which is consistent with the IPCC Tier 1 methodology [41] that gives a default emission factor of 0.51 kg CO₂/kg OPC, calculated based on an average lime CaO mass fraction of 64.6% in clinker. However, in our analysis of literature data, we assume that a CO₂ ch between 0.26 and 0.60 kg CO₂ ch/kg OPC is not an outlier value, and therefore is acceptable.

3 RESULTS

3.1 Reference Flow and Functional Unit

In LCA, defining a Functional Unit (FU) and a Reference Flow (RF) is an important step [21]. It is indeed crucial to ensure equivalent FUs among products while conducting comparative studies.

- A FU is a quantifiable description of the performance of the product. Some of the product's properties can be linked to the performances if they are an objective which serve the user's needs. The performances of the product are ultimately determined by the market demand in which the product will be introduced [43]. Some markets are submitted to product standards which specify product's performances and can serve identifying and defining functionalities of products.
- The RF is defined as the amount of product(s) required to provide the performances specified by the FU (ISO 14040, 2006). The RF considers the desired performance or functionality specified by the FU and translates the latter into a specific product flow [43,45].

Let us examine thereafter how authors define FU and RF for cement from the literature review.

In their study, [19] compared several blended cements using three different quantities of GGBS and defined the FU as one metric ton of cement. In their work, the cement types were defined according to the Brazilian Standards [46] with an identical strength grade of 32 MPa. From this description we can deduce that the cement strength grade is in fact the FU, and that the "one metric ton of cement" is the RF, taken identical for all cements because they have an identical functionality, i.e. 32 MPa cement strength class.

In another study [17] a geopolymer binder was compared with various other cements with different mix designs as defined from the Indian standards: OPC [47], Pozzolan Portland cement (PPC) [48], and Portland slag cement (PSC) [49]. The authors do not specify the strength grade of their cements, but they specify some properties of the geopolymer binder: its setting time, its soundness (shrinkage/expansion) and its strength corresponding to 43 MPa. Thus, we assume that the cements used for the comparison had the same strength of 43 MPa and that their comparison based on identical amount (i.e. all RFs = 1 kg) would be justified in that case.

In their study [24] compared the carbon footprints intensities of 1 kg of different OPCs but without defining the characteristics of these OPCs.

The work of [15] have considered a reference cement with a strength grade of 42.5 MPa, to which they compare other cements having different strength grades. They suggest using a so-called "strength ratio", provided in Equation (3), and multiplying the environmental impacts of one unit of mass of cement by this ratio. They justify their "strength ratio" by referring to a Chinese standard concerning the calculation of energy consumption per unit product of cement [50].

$$\text{Strength ratio} = \sqrt{\frac{\text{Studied cement strength grade [MPa]}}{\text{Reference strength grade [MPa]}}} \quad (3)$$

Although the authors do not mention the concept of FU and RF, the use of their "strength ratio" as a multiplicative coefficient applied to the indicators is equivalent to calculating a different quantity of

cement according to the strength grade and thus obtaining a RF according to the FU unit defined as the strength class. This approach seems interesting because it is the only reference we found, that provides a relationship between cement FU as strength grade and RF. However, the relationship in Equation (3) is not explained. We indeed analyzed the Chinese standard referred to by the authors [50] as a justification, and it reveals that this ratio only concerns the energy consumption of the final grinding operation (i.e. energy 2 in Figure 1) expressed as a function of cement fineness. Therefore, this strength ratio cannot represent the environmental impacts resulting from the entire cement plant because it is only proportional to the grinding energy. Furthermore, the standard [50] provides a slightly different equation: it expresses the relation by a fourth degree root and not by a square root. The standard [50] does not provide an explicit justification. Consequently, it leads to considering that this equation cannot be suitable for expressing a relationship between the FU (defined as cement's strength class) and RF.

It also appears that the concept of FU is not currently apprehended by most of the examined LCA studies. There is commonly a lack of information concerning the performance or functionality of cement. Some references do not even mention a FU [39]. Many others define a mass unit amount as a comparison basis and name it FU, whereas it actually is a RF, as the cement characteristics are not equivalent nor described.

Among authors that specify and base their comparison on cement's characteristics, the strength grade is clearly the one chosen to define a FU, even if authors do not always name it in this way. It is the most frequently cited performance [14,18,21]. From the papers reviewed so far, Equation (3) is the only relationship that has been provided to calculate RFs (i.e. comparable amounts of cements according to equivalent FU) with different cement strength classes).

The cement's strength is characterized by the measurement of its strength in MPa at 28 days in the (EN 197-1, 2011) standard [42]. To be categorized in a class means that the cement's strength at 28 days must be at least equal to the value of the corresponding grade (32.5 or 42.5 or 52.5 MPa). The cement's strength grade is also characterized by its hydration speed (low, normal or fast). The strength of cement influences the durability of concrete. Thus, from a desired concrete strength value, different quantities of cements will be needed if they have different strength classes, which corresponds to the RF value assuming the FU of the cement is its 28-day strength. In practice, different classes of cement are used for different purposes, grade 52.5 MPa is used for high performance concrete while the other classes are generally used in masonry. However, since the strength value of the cement is not equal to the strength class, different amounts of cement could be used within the same strength class. Thus, the 28-day strength of the cement, and not its strength class, could be a relevant FU. However, the strength of cement is not its only physical property. Other properties such as its heat of hydration, consistency, loss on ignition, bulk density, and specific gravity are important characteristics for some uses [51,52].

Specifying properly the FU of cement is therefore an important issue in LCAs of cement, as it is likely to change its constituents and the values of some flows in the manufacturing processes:

- the type and quantity of its constituents, such as tricalcium aluminate, tricalcium silicate, dicalcium silicate, ferrite, magnesia, sulfur trioxide, iron oxide, alkalis, free lime, silica fume, and alumina, influence the chemical properties [53]
- In the cement plant, along with the amount of gypsum, the type and duration of the grinding process (and thus its energy consumption, see Figure 1) influence the fineness of the cement, which is also highly dependent on the quality of the "clay/limestone" mixture.

In section 4.1, we will propose a method for quantifying the RF according to a specified cement strength.

3.2 Influencing factors on energy and GWP of Portland cement (OPC) without SCMs

The amounts of operation energy intermediate flows (named energy 1, 2 and 3 in Figure 1) as well as GWPs and CO₂ (named CO₂ ch, GHG A, and GHG B in Figure 1) from the cement manufacturing of cement without SCMs were collected and/or calculated from our collection data for different countries as explained in the method section. The results are provided in Table 1.

The amount of electricity for raw material preparation (energy 1 in Figure 1) ranges from 0.19 to 0.95 MJ/kg OPC (if we consider the 0.00016 value as an outlier). This lowest value of 0.00016 MJ/kg OPC from [18] may be dependent on the processes used in the raw material preparation stage. Indeed, [18] specified that they used an elutriation process which is based on separating particles according to their size, shape, density, etc., which may lead to a lower energy consumption.

The electric energy for cement grinding (energy 2 in Figure 1) ranges from 0.11 to 0.57 MJ/kg OPC.

- The study of [15] specified the cement's strength grade but not the value of energy 2. In the Ecoinvent database 3.7 cut-off [39], the value of energy 2 was different for each of the investigated references, and could thus possibly be related to different cement strength classes, or to different types of equipment, but no piece of information is given to check these possibilities.
- The Ecoinvent database 2 [7] provided two different values of energy 2 for two cements strength classes 42.5 and 52.5, with a slight increase of energy consumption without more detail.
- The work of [14,18] provided both cement's strength grade and a value for energy 2. The authors of [18] performed their case study in USA for a 50 MPa cement strength grade. This value is close to the one considered in the work of [14], with 52.5 MPa. The amount of energy 2 (0.57 MJ/kg of cement) obtained by [14] is slightly higher than the one (0.56 MJ/kg of cement) found by [18]. However, both values are similar.
- For the Environmental Product Declarations (EPDs) references specified in Table 1, the type of operation inside cement plant that is considered for the energy amount is not accurately detailed and we can thus not obtain the detailed values of different energy flows.

Figure 2 presents the data of energy 2 (reported in Table 1) as a function of the cement strength grade, although there are only 3 references providing both values. If we consider the data from [7] with a 52.5 MPa cement's strength grade as an outlier, it seems that the value of energy 2 increases with the cement strength grade. However, we cannot conclude with certainty given the small number of points.

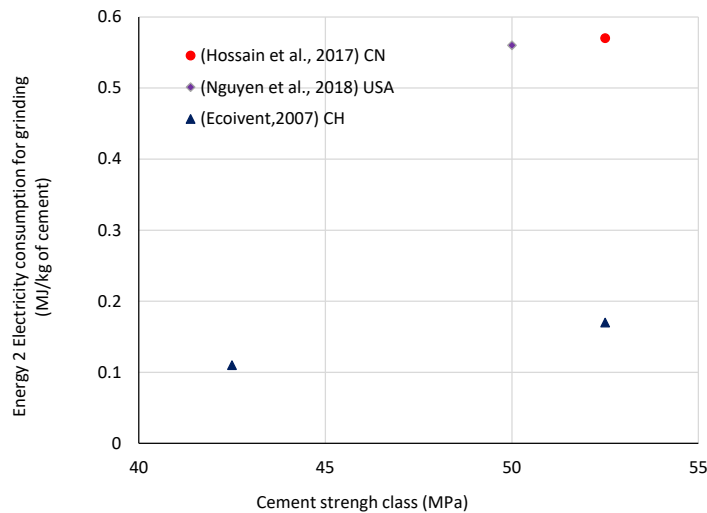


Figure 2: Electricity consumption in grinding (energy 2 in Figure 1) as a function of cement strength grade during cement manufacturing

The heating energy (energy 3 Figure 1) is also found quite variable between the collected data in Table 1, ranging from 1.34 to 5.31 MJ/kg OPC. This energy highly depends on the type of technology (wet or dry calcination): the amount of water inside raw meal will influence the amount of energy required to dry and calcinate these materials.

Table 1: Operational energy intermediate flows and GHG A, GHG B and CO₂ ch for the cement production processes for 1 kg of OPC

Unit	Country	Cements strength grade [MPa]	Electricity for raw material preparation	Electricity for finishing grinding [MJ/kg OPC]	Heating energy for calcination	GWP from electricity	Estimated CO ₂ from decarbonation reaction [kg CO ₂ eq/kg OPC]	Estimated GWP from clinkerization combustion	Total GWP for cement life cycle
Flow in Figure 1			Energy 1	Energy 2	Energy 3	GHG A	CO ₂ ch	GHG B	GWP total
Reference									
[39]	CA	NS	0.36	0.20	3.60	NS	0.37	0.41	0.84
[39]	RER	NS	0.28	0.14	2.10	0.047	0.56	0.20	0.86
[39]	CH	NS	0.45	0.16	1.34	NS	0.56	0.15	0.75
[39]	PE	NS	0.28	0.13	3.88	NS	0.44	0.38	0.88
[39]	BR	NS	0.25	0.18	2.2	NS	0.44	0.30	0.84
[39]	USA	NS	0.28	0.20	2.10	0.068	0.57	0.19	0.88

[39]	ZA	NS	0.46	0.134	3.84	0.18	0.35	0.41	1.00
[39]	IN	NS	0.19	0.11	3.09	0.11	0.36	0.36	0.89
[25]	AU	NS	0.22	0.14	4.40	NS	0.52	0.44	0.97
[9]	DE	42.5	NS	NS	3.52	NS	0.49	0.18	0.779
[15]	CN	42.5	0.14 ¹		5.31 ²	NS	0.51	0.24	0.80
[10]	ESP	NS	0.33	0.14	3.53	NS	0.52	0.39	0.80
[14]	CN	52.5	0.95	0.57	4.53	0.081	0.51	0.31	0.98
[24]	AU	NS	NS	NS	NS	0.10 ³	NS	NS	1.3
[18]	USA	50	0.00016	0.56	4.7	NS	NS	NS	0.99
[17]	IN	NS	NS	NS	NS	NS	NS	NS	NS
[7]	CH	42.5	0.19	0.11	3.32	NS	NS	0.32	NS
[7]	CH	52.5	0.19	0.17	3.32	NS	NS	0.32	NS
[33]	GR	52.5N	NS	NS	3.81	0.021	0.90		0.92
[31]	BY	42.5	NS	NS	NS	NS	NS	NS	0.92
[31]	BY	42.5	NS	NS	NS	NS	NS	NS	0.91
[36]	SK	52.5	NS	NS	NS	NS	1.02		1.09
[29]	CR	NS	NS	NS	NS	NS	NS	NS	0.78

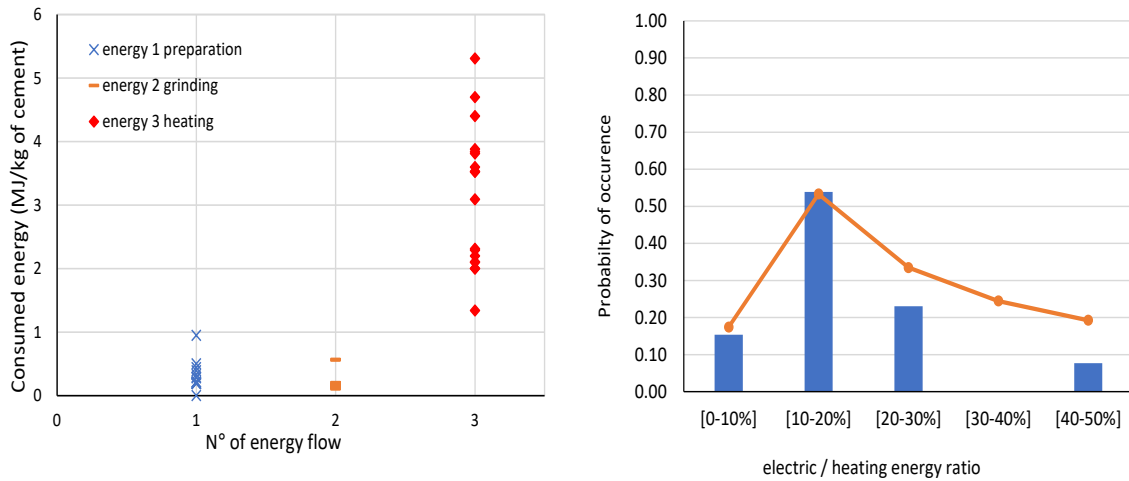
¹ It is the total electric energy consumption generated by the cement plant (energy 1 + energy 2)

² The energy provided is equal to the energy of the coal incineration and the thermal energy

³ The value is estimated from the bar graph provided in the reference

NS: Not specified

The electric energy consumed for both preparation and grinding are quite small compared to the heating energy as can be observed in Figure 3a.

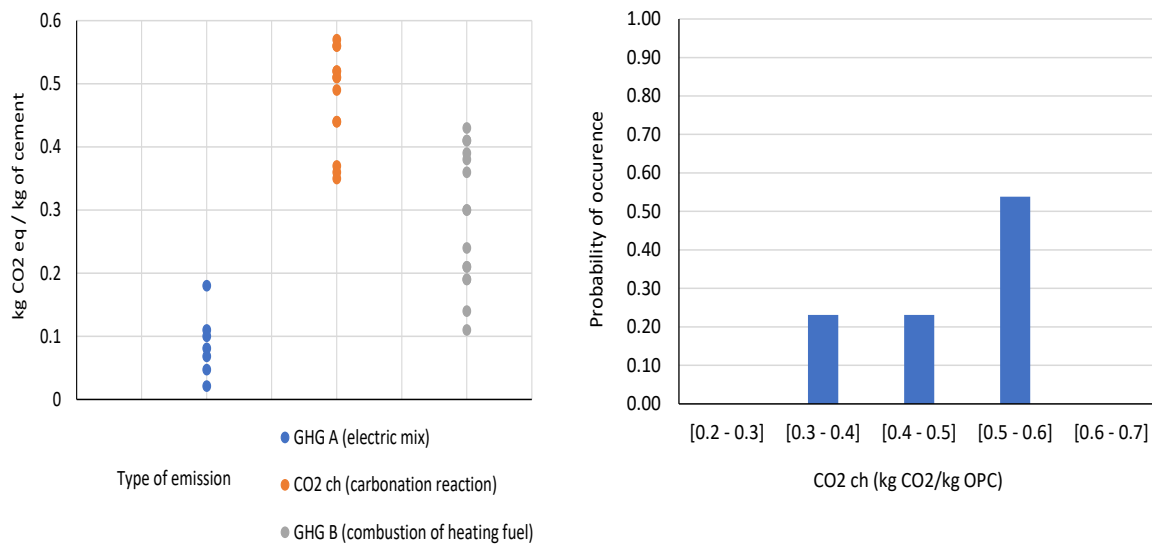


a) values of consumed energies

b) frequency of data by interval of grinding / heating energy ratio - i.e. (energy 1 + energy 2) / energy 3 and approximated probability distribution

Figure 3: Comparisons of energy flows between references

The ratio of electric energy (energy 1 + energy 2) divided by heating energy (energy 3) is represented in Figure 3b. It ranges between 8% as minimum [25] and 46% as a maximum (Ecoivent,2020 CH) [39]. Figure 3b shows that the interval [10-20%] and our 13 available data can be approximated by a lognormal distribution with mean value equal to 15% and standard deviation equal to 4.5%.



a) values of GHG emissions

b) frequency of CO₂ ch by interval of emission

Figure 4: comparison of GHG and CO₂ ch emissions between references

Figure 4a shows that other sources of GHGs (i.e. GHA A and B) are generally lower than CO₂ ch emissions. These emissions depend both on the amount of energy 1 and 2 (see Figure 1) and on the electric mix of the considered country. Thus, from the cement industry perspective, reducing these emissions means reducing energy consumption.

As shown in Table 1, the range of CO₂ ch emissions ranges from 0.35 to 0.57 kg CO₂/kg OPC. We know that they almost exclusively depend on the decarbonation of limestone and can be calculated from the CaO amount of cement (see section 2.3). Some references presented in Table 1 (Ecoinvent, 2020 CA,ZA, PE,BR &IN) [39] do not provide the exact amount of CaO, however, by applying Equation (2), the range of CaO is found between 45% and 56% that is lower than the range of 60-67% defined as usual by [7] but consistent with the [42] standard requirements for CEM I. For (Ecoinvent, 2020 RER, CH, USA) [39] the amount of CaO is found between 71 and 73%, which is generally higher than the usual range.

Studies conducted by [9], [10] and [14] obtain 0.54 kg CO₂ ch/kg OPC, 0.53 kg CO₂ ch/kg OPC and 0.55 kg CO₂ ch/kg clinker, respectively. These studies closely align with the default emission factor of the IPCC tier 1 method [41] that gives a default emission factor of 0.51 kg CO₂/kg OPC. Our results show that the CO₂ ch value ranges have around 50% probability to range between 0.5 and 0.6 kg CO₂ / kg OPC as shown in Figure 4b.

3.3 *Influence of the use of recovered waste*

3.3.1 *Definition and choice of allocation methods*

Different allocation methods can be applied in LCA modelling to determine how the environmental burdens of the recovery and possibly avoided processes will be allocated between the main product and the by-product or secondary product (i.e. a product originated from a waste after recovery). As SCMs generally come from recovered waste, the LCA system model is subject to the choice of the allocation method as shown in Figure 1 and further described below. The choice of a system model for waste recovery aims at answering how to consider and assign the environmental impacts and benefits of recycling to different product sub-systems [54]. The question of “assignments” of the impacts is equivalent to the notion of responsibility given to the products produced by stakeholders. In other words, the choice of an allocation method has a political meaning [55]. This political meaning can be analyzed through the goal of an LCA study concerning recovery, either “waste treatment”, “waste-to-product transformation” or “secondary product production” [56]. A critical aspect of this analysis involves asserting a specific perspective in alignment with the stakeholder involved. Indeed, behind the allocation issue for waste recovery, stand three economic actors that can be defined as: (i) the “waste producer” producing both the main and waste; (ii) the “recycler” transforming the waste into a secondary product through different possible recovery ways such as reuse, refurbishing, or recycling, and finally (iii) the “user” that is the one choosing the secondary product to substitute another existing product on the market. In the case of blended cement using Ground Granulated Blast Furnace Slag (GGBS) as a by-product recovered from the steel industry: the “waste producer” is the steel industry producing the slag, the “recycler” is the industry transforming the slag into a material usable for cement, and the “user” is the cement industry. This example is used below to explain and illustrate the different allocation methods and their political meaning.

The point of substitution (PoS) is defined as the point of the system at which the secondary product can substitute a primary product into a given market of process. This concept comes from the European directive [57] that defines the “end of waste point”. Thus, the PoS allocation method attributes all the impacts before the point of substitution, i.e. all processes of recovery treatments, to the main product. In our example, the GGBS by-product is considered similarly to a waste: the steel industry, and thus the steel product endorses full responsibility of environmental impacts, and its system boundaries integrate all processes until the secondary product is provided, i.e. the recovery process of steel slag is attributed to the production of steel and not to GGBS production. Both the recycler and user benefit from a “zero impacts” secondary product. This method is in line with the “polluter pays principle” of the current European regulation. As a political consequence, unless incentivized by taxes or regulations,

the waste producer may not be encouraged to sort nor recover its waste, as landfilling may appear as a preferable solution with possibly lower environmental impacts than recovery processes: this is especially the case for inert waste for which landfilling processes do not have important environmental impacts.

The cut-off (CO) method allocates the environmental impacts of the recovery process to the secondary product. This method attributes all processes from by-product to secondary product as part of the system allocated to the secondary product. Concerning political consequences, the waste producer, the steel producer in our example, has reduced impacts compared to the PoS method, and both the recycler and user, although taking responsibility for the impacts of the recovery process, benefit from a valuable product with generally small environmental impacts. In our example, the cement industry can obviously notice a major environmental benefit from choosing GGBS as a partial replacement of clinker.

The partition method specifies that the secondary product endorses the impacts of the recovery processes as well as a part of the production system of the main product. This part of the production system of the main product is determined according to a partition rule. In our example, GGBS endorses a part of the steel production system. Two current rules (mass and economic) are generally found and described below [58].

- The mass allocation (MA) rule which is based on mass ratios as shown in Equation (4) [55].

$$C_m = \frac{m_{by-product}}{m_{by-product} + m_{main product}} \quad (4)$$

where C_m = mass allocation factor affected to the by-product (or secondary product); $m_{by-product}$ = mass of by-product (or secondary product) [kg]; $m_{main product}$ = mass of main product [kg].

- The economic allocation (EA) rule considers the allocation relatively to the selling prices of the main product and by-product. It is defined in Equation (5).

$$C_e = \frac{p * (m_{by-product})}{p_{by-product} * (m_{by-product}) + p_{main product} * (m_{main product})} \quad (5)$$

where C_e = economic allocation factor affected to the by-product (or secondary product); p = price per unit of the material; $m_{by-product}$ = mass of by-product [kg]; $m_{main product}$ = mass of main product [kg].

The partition method (either based on mass or economic ratios) shares environmental burdens of the production system between main product and its by-products. This means that the waste producer is “discharged” from a part of the environmental burden that is transferred to the recycler. In our example, the steel industry is “discharged” from a part of the impact affected to the steel product, that is transferred to the GGBS recycler’s product. This method is not in line with the “polluter pays principle” of current European regulation, and it does not encourage the development of recovery solutions because the user will not obviously notice if the secondary product has any environmental benefits. However, in the case of blended cement, GGBS have long been known as providing interesting binding properties. GGBS is thus a valuable product, and its recovery is a current practice. Thus, choosing a partition method instead of CO or PoS is not expected to change the behavior of the steel industry.

In all previous methods (MA, EA, PoS and CO), the potential benefits of recovering a waste are not included. Recovery takes place in order to both avoid waste elimination (especially landfill and space occupation) and preserve natural resources. By not including substitution to one (or more) products into the system model, these possible benefits cannot be estimated.

The system expansion with substitution (SE) method was proposed to assess those potential environmental benefits. It stipulates that the production processes of the studied product are subtracted by the processes associated with the production of a substituted and functionally equivalent product. Thus, for the secondary product, environmental loads from the recovery processes are added to the system and avoided processes are subtracted from the system [55]. In our example the use of GGBS will avoid the use of OPC, thus, the amount of avoided OPC that is functionally equivalent, is subtracted from the system. This is also called "avoided impacts". However, the rules for choosing the equivalent product, and to assign the avoided impacts are not trivial. In our example, avoided impacts could be assigned either to steel or to blended cement using GGBS. According to [59], the avoided impacts of a substituted product should be assigned to the waste producer (i.e. steel industry in our example). However, when establishing waste management or circular economy strategies, which is especially the case of SCMs in blended cement, the studied product is the recovered product (GGBS) and not the main product (steel). In order to assess the potential benefits of recovery, the avoided impacts are thus attributed to the secondary product (i.e. GGBS in our case) and the main product is excluded from the system. Finally, the problem of assignment of environmental burden is displaced but not resolved by the SE method. Furthermore, this means that LCAs resulting from an SE method are not additive. Indeed, if LCAs of GGBS and steel are used in another LCA study (for example LCA of a building using reinforced cement concrete with blended cement and steel), and if that both GGBS and steel are calculated with a SE method, environmental credits are overestimated because they are subtracted twice (once for GGBS and once for steel). For this reason, it is often recommended that avoided impacts are accounted separately and not integrated to LCAs of the concerned product, if this LCA is to be used as an input data in another LCA. The SE method is interesting when one wants to investigate and compare different recovery paths, but not for a database purpose. The European standard EN15804 [60] for Environmental Products Declarations (EPDs) in the Building and construction sector, recommends the use of partition (preferably economic allocation) because EPDs aim to serve as data provider for LCAs of buildings. However, avoided impact beyond the product's life cycle, i.e. due to recovery, and corresponding to module D in the EN 15804 standard [60] can be calculated as an additional information.

Finally, it should be noted that the allocation method is only a mathematical distribution of environmental impacts between several products, but that the total environmental impacts remain the same in reality.

In our corpus of references, authors use these different methods to consider either the addition of SCMs into OPC or the use of waste incineration for the heating energy (energy 3 in Figure 1). These different system boundary rules should thus lead to major differences in results. According to theoretical methods described and schemed in Figure 1, the partition method (either MA or EA) should provide the highest impacts followed by CO and then SE (PoS has not been found in our corpus of references). Indeed, with the MA or EA, the secondary product endorses a system with the largest system boundaries because it includes a part of steel production. In contrast, narrower boundaries are observed when applying the CO method. The SE method has larger boundaries, but a part of the system, corresponding to avoided OPC, is subtracted, thus it is expected to provide the smallest environmental loads to blended cement. The ranking between MA and EA cannot be predicted in

general: it depends on the mass proportions between main and recovered product from production process, and on their selling prices proportions respectively.

3.3.2 Substitution of clinker by Ground Granulated Blast Furnace Slag

In this paper only GGBS is considered for clinker substitution, as it is the only SCM for which a sufficient data sample could be collected (40 out of 102 data), and with several data available for each allocation method. We have compared GWP of various GGBS cement mix designs and compared them for 1kg of blended cement. GWP values as a function of GGBS ratio in blended cement as shown in Figure 5.

Relatively to the explanation of allocation methods in the previous section, we focus on the production of blended cement using GGBS, thus on the “waste user’s” side. This means that the more or less favorable allocation methods are observed in order to decrease the impacts assigned to blended cement, but this does not mean that total impacts of the total system (steel, slag and cement) are decreased. Furthermore, for this SCM, the selling price of the GGBS is relatively small compared to the selling price of steel, thus economic allocation should allocate lower environmental impacts to GGBS compared to mass allocation (the mass ratio of GGBS to steel is around 20%).

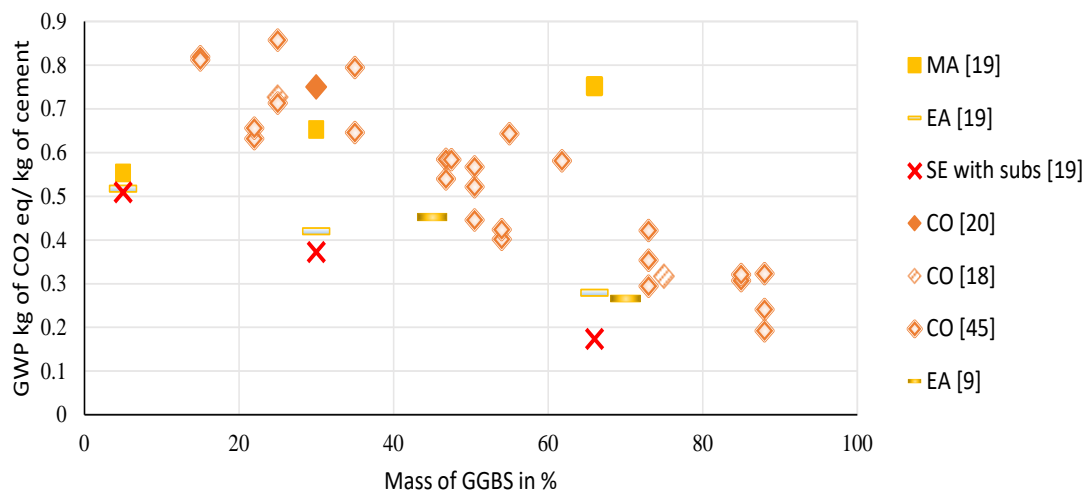


Figure 5: The influence of the mass amount of GGBS and the allocations methods on the global warming potential–mass allocation (MA) rule , economic allocation (EA) rule , cut-off (CO) method and system expansion with substitution (SE).

As it can be seen in Figure 5, one reference [19] located in Brazil, compared different allocation methods for the same mass ratio of GGBS in cement (25%). The ranking of GWP according to the chosen method is found as expected: MA is found higher than EA and SE shows the lowest emissions. However, the CO was not considered in this study.

The EA method was applied to the production of cement with GGBS by [9] on a German case study with a 45% mass ratio of GGBS and authors obtained GWPs of 0.452 kg CO₂ eq/kg of cement. For similar GGBS ratios (45% and 50.5%) two other studies using CO method [9,39] find similar GWPs compared to the EA method [9], with values of 0.452 and 0.446 kg CO₂ eq/kg of cement respectively. If this the EA method provides similar results that CO, it means that, although it includes a part of steel production, the value of the coefficient, obtained from equation (5), is so small that it does not make a big difference compared to the CO method.

The work of [18,20] also applied the CO method for case studies located in the USA, with similar GGBS mass ratios of 25% [18] and 30% [20]. This resulted in similar GWPs of 0.75 and 0.727 kg CO₂ eq/kg of cement respectively.

Overall, increasing the amount of GGBS causes a decrease in impacts of blended cement for all allocation methods except MA. The MA conducts to include an important part of the impacts of steel production (around 20%) that are allocated to GGBS. Therefore, as the amount of the GGBS increases, the augmentation of impacts of the steel production are not compensated by the decrease in the amount of OPC in blended cement production.

Finally, compared to OPC with GWPs ranging from 0.75 to 1.3 kg CO₂ eq / kg of cement (see GWP total in Table 1), blended cements with GGBS range from 0.18 to 0.86 kg CO₂ eq / kg of cement (Figure 5). There is indeed an important reduction of impacts, due to the reduction of OPC for CO and EA methods, and due to both reduction of OPC and substitution of OPC for SE. For the SE method, as can be seen in Figure 1, the substituted OPC can be accounted twice: once by reducing the amount of OPC in blended cement, and a second time by applying the subtraction with the SE method. This should theoretically not happen, but it is often conducted this way because of a lack of sufficient information or of a good understanding of the method. We have no details from the authors [19] to check if the SE method was correctly applied.

3.3.3 *Substitution of fossil fuel by waste incineration for the heating energy*

The total energy needed for heating in the calcination process is found to be the highest amount of energy consumed by cement production (Figure 3a).

These fuels can be fossil fuels (primary fuels) or derived from waste (secondary fuels). Indeed, cement plants often serve as waste incineration facilities and recover the heating energy from incineration. Because the cement plant is considered as an elimination process for the incinerated waste, and not as a recovery process, the incineration process is allocated to the waste producer. In that sense, the energy provided by the incineration, as well as the emissions from that incineration are not integrated into the cement's life cycle.

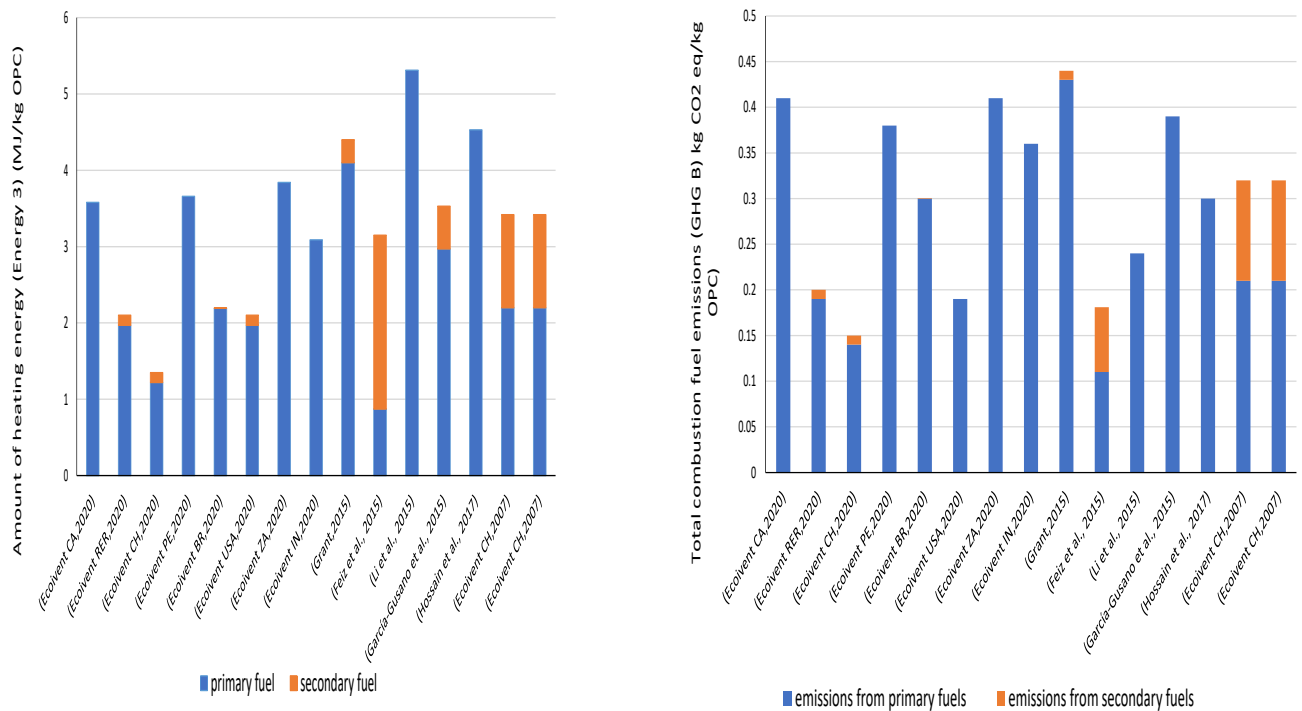
According to the investigated literature, data from [7] integrate information on secondary fuels but logically allocate no impacts to the cement production: all the flows of the combustion of the secondary fuel are considered as waste elimination and thus allocated to the waste producer. Similarly, in the work of [9] the environmental impacts of secondary fuels combustion have not been allocated to cement.

The rest of the examined literature did not explicitly state whether the environmental impacts were allocated to the secondary fuels or not.

The contribution of secondary fuels to the total values of energy and GHG B emissions are detailed in Figure 6a and Figure 6b respectively.

By observing the total amount of heating energy (energy 3 in Figure 1) one can detect effects of an improvement of the energy efficiency of combustion such as data from Ecoinvent CH, 2020 [39] in Figure 6a. One can also observe that the use of secondary fuels is considerably influent on the results attributed to the cement's life cycle. Substituting the primary fuels with secondary fuels as shown in Figure 6b, can significantly decrease the cement's life cycle GHGs because those from secondary fuels are not allocated to cement. Thus, there is a significant decrease in GHG B when the dominant fuels are secondary as in [9]. Also, we can note that Ecoinvent CH, 2007 [7] give lower GHG B emissions compared to Ecoinvent CA, 2020 [39] because there is a significant quantity of primary fuels substituted

by secondary fuels. It is surprising however, that the use of secondary fuels appears to decrease between 2007 and 2020.



- a) Amount of total heating energy, primary energy and secondary energy – only primary fuels are allocated to the cement’s life cycle
- b) Amount of total GHG B detailed from primary only primary fuels are allocated to the cement’s life cycle and secondary fuels - only primary fuels are allocated to the cement’s life cycle

Figure 6: Comparison of the influence of the primary and secondary fuel energy on the energy and GHG B emissions obtained from combustion

4 DISCUSSION

4.1 Calculation method for quantifying the RF according to the strengths of cement and concrete

Our literature review [16,24] showed that no clear relationship between RF and FU could be found. However, this is an important question, as adding SCMs or increasing cement’s fineness changes the properties of cement, and thus its amount required for a target performance in use. It also showed that cement strength grade is commonly identified as a FU for cement products, thus, the amount of RF for each type of cement could be quantified based on the cement’s strength.

The performance in use of cement depends in fact on the compressive strength which is desired for mortar or concrete. In the literature, different models can be used for calculating concrete compressive strength at 28 days [61–64] according to the cement’s strength. But these models are only applicable to concrete composed of OPC and natural aggregates.

Bolomey’s model [65], see equation (6), allows to calculate the amount of cement needed for a desired compressive strength of concrete and can be applied to concrete made with blended cements.

However, it neglects the geological type of the aggregates such as the shape regularity and ratio, chemical composition, and surface texture [66]. Bolomey's model takes into account the quality and the size of the aggregates, the dosage of effective water and binder, the actual cement strength and the amount of entrained air in concrete.

$$B_{eff}^{blended} = \left[\left(\frac{f_{ck,avg}}{G * F_c} \right) + 0.5 \right] (W_{eff} + V_a) \quad (6)$$

Where: $B_{eff}^{blended}$: effective binder [kg/m³] for blended cement]; $f_{ck,avg}$: average strength of concrete at 28 days [MPa]; G: granular coefficient [-]; F_c : actual strength of cement [MPa]; W_{eff} : dosage of effective water [kg/m³] and V_a : dosage of the air entraining agent [l/m³].

The granular coefficient can be determined according to the dimension of aggregates which can vary from 16 mm to 63 mm and can be classified as thin, average or large. The quality of aggregates also influences the value of the granular coefficient which can be fair, average or excellent [67]. Bolomey's model requires the use of the actual strength of cement and not its strength grade, however this value is currently provided by cement producer in the products' technical data sheets.

To consider SCMs, one can use the effective water to binder ratio W_{eff}/B_{eff} as defined in NF EN 206+A2/CN [68]. However, this concept of SCMs covers the use of a single addition as a partial replacement of cement. The effective binder is described as follows:

$$B_{eff}^{blended} = C_{OPC} + k * SCMs \quad (7)$$

Where: $B_{eff}^{blended}$: effective binder [kg/m³] for blended cement, k : is the coefficient of the considered SCMs [-], C_{OPC} : is the dosage of Ordinary Portland Cement per cubic meter of concrete [kg/m³], SCMs: is the amount of supplementary cementitious materials per cubic meter [kg/m³].

The k -value concept enables the SCMs of FA and GGBS to be considered, that have a k -value of 0.40 and 0.60 respectively. The maximum amount of SCMs must meet the requirement specified in the standard NF EN 206+A2/CN [68].

The maximum value of SCMs must be attained in accordance with the following ratio NF EN 206+A2/CN and any excess should not be included in the calculation of the water/ ($C_{OPC} + k * SCMs$) ratio, nor in the minimum equivalent binder dosage:

$$SCMs_{max} = \frac{SCMs}{SCMs + cement} \quad (8)$$

Where $SCMs_{max}$: is the maximum quantity of SCMs that can be attained per cubic meter of concrete [kg/m³], SCMs : is the quantity of SCM added per cubic meter of concrete [kg/m³], cement: is the dosage of Ordinary Portland Cement per cubic meter of concrete [kg/m³]

The entrained air in concrete can vary from 15 l/m³ to 25 l/m³ [69].

We apply this method to obtain RFs for cements with strength of 32.5 MPa and 42.5 MPa used to achieve a 50 MPa compressive strength concrete, for different exposure. We considered an average value for the air entraining agent in concrete to be 20 l/m³. The water to cement ratio (w/c) is set to 0.4. In order to calculate the amount of cement required to achieve a targeted concrete strength, we calculate the granular coefficient based on the work of [67]. We set the dimension of aggregate to be equal to 20 mm because the properties and composition of concrete are designed for an aggregate dimension of 20 mm according to the standard NF EN 206+A2/CN [68]. This gives a granular coefficient of a value of 0.53. Results are shown in Table 2.

We clearly notice that for a given target compressive strength of concrete, the amount of cement needed for 1 m³ of concrete considerably varies according to the cement's strength. Less amount of cement is required for cement with a higher strength (42.5 MPa) for all exposure classes. The ratio between the quantities of calculated cement at 42.5 MPa and 52.5 MPa is 0.80 keeping all the other parameters constant. Thus, the GWP will vary accordingly.

Practically, cements are defined by their compressive strength class, defined as 32.5, 42.5 and 52.5 MPa classes in Europe. The class determines the further use of cement: for example, higher 52.5 MPa class is reserved for structural concrete. The class corresponds to a minimum required value of compressive strength, but it is not the actual value. For a given usage, the actual cement compressive strength can vary from the minimum value to the value of the higher class: for example, the actual compressive strength of a cement with a 32.5 MPa class can vary from 32.5 MPa to 42 MPa. The value of RF will thus vary accordingly as shown in Table 2. The real cement strength value is easily obtained from technical sheets provided by cement producers.

Table 2 shows important variations of RF for an identical exposure and cement strength classes. Thus, a calculation of RF according to the actual cement strength is very important to compare cements on a fair basis. The quantities assigned to the reference flows in the table correspond to the lower and upper limits of the cement strength class intervals, respectively.

Table 2: Variations of the Reference Flows [kg cement / m³ of concrete] of cement for a targeted 50 MPa concrete compressive strength, as a function of cement's strength taken as a Functional Unit (with a water to cement ratio of 0.4, an interpolated quality of aggregates of 0.53 and a volume of air entraining agent of 20 l/m³).

Exposure class	Cement strength class [68]					
	32.5 MPa		42.5 MPa		52.5 MPa	
	Min	Max	Min	Max	Min	Max
[68]	32.5 MPa	42.5 MPa	42.5 MPa	52.5 MPa	52.5 MPa	62.5 MPa
XS1	406.39	310.77	310.77	251.57	251.57	211.32
XS2	503.61	402.52	402.52	339.95	339.95	297.40
XS3	530.83	424.28	424.28	358.32	358.32	313.47
XD1	476.39	380.77	380.77	321.57	321.57	281.32

XD2	476.39	380.77	380.77	321.57	321.57	281.32
XD3	503.81	402.52	402.52	339.95	251.57	211.32

4.2 Grinding energy

As shown in the previous section, the cement's strength is an important parameter that influences the amount of cement required in a given concrete with a target compressive strength. Using a higher cement's strength can considerably reduce its amount in concrete, however, it can change the amount of energy required for grinding (energy 2 in Figure 1). Our data analysis in section 3.2 suggests that an increase of the cement strength is likely to increase energy 2.

In the context of slag use in blended cement, the quantity of grinding energy plays a significant role in achieving the desired particle size similarly to the cement production. The fineness of the slag particles directly impacts the specific surface area contributing to the enhancement of cement strength characteristics: [70] showed that :

- for a constant slag fineness with specific surface area at 4,000 cm²/g, and an increase of clinker fineness from 3,000 cm²/g to 4,000 cm²/g results in a greater 3 N/mm² increase in 2-day strength of the mix;
- whereas when keeping clinker fineness constant at 4,000 cm²/g and increasing slag fineness from 3,000 cm²/g to 4,000 cm²/g, the strength only increases by 1.4 N/mm².

This shows that clinker fineness is approximately 2 times more efficient than slag fineness on 2-day strength. Also, it is important to note that slag requires more time to be grinded than clinker, which has implications for cost-effectiveness [70] and thus on the value of energy 2 as well. Therefore, prioritizing finer grinding of clinker is essential in mitigating the emissions associated with finer grinding of slag.

According to laboratory tests on the grinding efficiency of minerals, the type of the grinding mill (ball or stirred mill) and the target fineness of minerals, influences the amount of energy required for grinding [71]. Table 3 shows the electricity consumption required at different grinding stages obtained from [71]. For each type of mill, increasing the fineness of mineral increase the energy consumption. For a given fineness, the stirred seems to consume less energy than the ball mill.

Table 3: Electricity required at different grinding stages for minerals – source: [71] with conversion from kWh/ton to MJ/kg.

Product 80% passing size (µm)	Energy (MJ/kg)	
	Ball mill	Stirred mill
Conventional grinding [70-1000 µm]	0.01-0.11	-
Regrinding [30-90 µm]	0.11-0.29	0.06-0.14
Fine grinding [8-30 µm]	0.29-0.68	0.14-0.29
Very fine grinding [1-9 µm]	-	0.29-0.58

In the case of the cement production process, the first step of grinding produces a raw meal with a mean particle diameter of approximately 200 µm. And for the second step of grinding, this diameter is

between 20 and 25 μm . According to data obtained from [71], these grinding raw meal would require an amount energy 1 between 0.01 and 0.11 MJ/kg and grinding cement would require an amount of energy 2 around between 0.14 and 0.29 MJ/kg. Values for energy 1 obtained in LCA studies (see Table 1) are much higher than observations from [71]. Values for energy 2 obtained in LCA studies (Table 1) are consistent with [71] for the minimum value of 0.11 MJ/kg, but not for the maximum value that is higher in Table 1 (0.57 MJ/kg) than in Table 3 (0.29 MJ/kg). However, tests from [71] were conducted at lab scale and cannot fully represent the industrial scale.

On the industrial scale, some data can be found on recommended setpoints of production rate according to target fineness. Production rate setpoints decrease with the increase of target fineness, thus showing an increase of the residence time of the material inside the grinder and thus increasing energy consumption. However, this energy increase is quite specific from the type of grinder [72]. Modeling this energy would require developing a database dedicated to industrial grinders.

4.3 *The use of secondary fuels*

Secondary fuels are always beneficial to the LCA of cement because their impacts are allocated to the waste producer. For cement, these benefits are reflected mainly by the decrease in the amounts of fossil fuels and GHGs emissions from combustion that are allocated to cement.

The Ecoinvent database calculated CO₂ emissions from various fuels (based on their average carbon content), but this assumes complete combustion, which is never reached in industrial combustion. Furthermore, it also seems that the heating value used for combustion of natural gas by the Ecoinvent database report [7] is outdated and particularly underestimated (20.33 MJ/m³). Indeed, the calculation methods of the impact factors provided by the database of Ecoinvent 3.9.1 cut-off [73] provides higher value (36 MJ/m³), so the calculation should be updated.

Also, it is difficult to validate the calculated CO₂ emissions with the effective measured emissions: total emissions measured at the stack of the cement plant cannot easily be attributed either to primary or secondary fuels, because of complex chemical reactions due to both burners' settings and chemical composition of fuels which are subject to variations within type of secondary fuels, various cements plants and various periods of time. For references that directly provide GHG emissions, it is neither possible to know if they are obtained from calculation or measurements, nor to check if they are reliable.

To make these data consistent and reliable, it would be advisable for each data supplier to provide all the following information: average composition of the heating energy mix over several years, specifying in particular the proportion of secondary fuels, and average quantities of CO₂ emitted by stacks based on measurements over the same period.

4.4 *Blended cement with Ground Granulated Blast Furnace Slag (GGBS)*

Among our investigated references concerning LCAs of GGBS blended cements, none of them provided details concerning LCA of GGBS.

Beside the choice of the allocation method that influences the impacts associated to GGBS (as previously discussed in section 3.1.1), it is also interesting to investigate if changes in the slag production process itself can lead to significant changes in environmental impacts.

Several unitary operations (presented in Figure 7) are required for the slag recovery process. The recovery process of slag during the iron manufacturing involves the separation of liquid metal and slag

at the exit of the blast furnace. The slag is then cooled through a water quenching process and dried using an energy source before undergoing grinding and storage in silos. Subsequently, the GGBS is transported to other industrial sites, such as cement industries, as per the context of our paper.

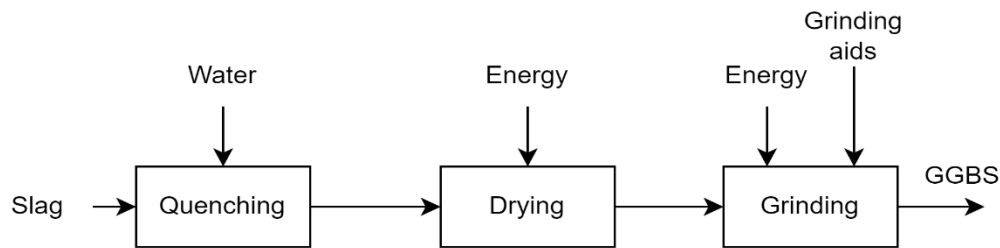


Figure 7: Scheme showing the different stages of slag recovery

According to this description, we assume that the following aspects can have a significant influence on the GHGs emissions:

- The moisture content of slag after the quenching operation and before entering the drying operation: the higher moisture content, the higher the amount of energy required to dry them and thus the higher GHGs emissions (according to the energy source used to dry them).
- The energy source used for the drying process can totally or partially be obtained from heat recovery of the blast furnace and thus reduce impacts linked to the drying process.
- the grinding energy operation can also consume electric energy but this aspect has been discussed in section 4.2.

Table 4 gathers climate change indicator values and information obtained from a panel of international data coming either from producers or from the Ecoinvent database. For the latter, a variant with the French electricity mix is presented, as well as a variant integrating the allocation value. This allocation was set at 1.4% in accordance with French authorities validated by the INIES program [74] that decided in favor of economic allocation between cast iron and slag. This allocation rule transfers climate change impact from melt to slag at 83 kg eq CO₂ [75]. Nevertheless, one must be careful to take into account the respective economic values of the cast iron and the slag, which may cause this percentage to vary, and the reality of the impacts of the production of cast iron, which may differ from those represented by the data in the Ecoinvent database.

Table 4: Global warming potential [kg CO₂ eq / ton GGBS] from various literature data

	Type of allocation with iron	Electric mix	Intermediate transportations	Drying	Grinding energy	Total GWP
ECOCEM 2014 [76]	CO	France	Yes	NA	NA	20.4
GWP	0		?	?	?	

				Humidity10%		
ECOCEM 2019 [77]	CO	Ireland	Yes	Dry natural gas	NA	32.0
GWP	0		14.1	?	?	
SCA [78]	CO	US	Yes	NA	NA	147.0
GWP	0		62.7	?	?	
JSW [79]	CO	India	Yes	NA	47 kWh	60.2
GWP	0		?	?		
ArcelorMittal [80]		5 Eur. countries (SP, PL, D, B, F)	Yes	NA	NA	82.8
GWP	?		?	?	?	
Ecoinvent [39]	CO	US	Yes (Train + truck)	Natural gas	91.1 kWh	81.2
GWP	0		3.2	24.5	49.8	
Ecoinvent with French mix [39]	CO	France	Yes (Train + truck)	Natural gas	91.1 kWh	41.7
GWP	0		3.2	24.5	10.2	
Ecoinvent with French mix + economic allocation [39]	EA	France	Yes (Train + truck)	Natural gas	91.1 kWh	124.7
GWP	83		3.2	24.5	10.2	

The GWP of GGBS can vary from 0.0204 to 0.147 kg CO₂ eq / kg GGBS according to existing LCA studies. It clearly appears that the allocation method has a significant impact on the result. Besides, very few information can be found concerning the contribution of unitary operations. According to collected data, transports, grinding and drying can all be very influent on the total GWP, depending on the electric mix, the transport distances, as well as on the source of heat for the drying operation.

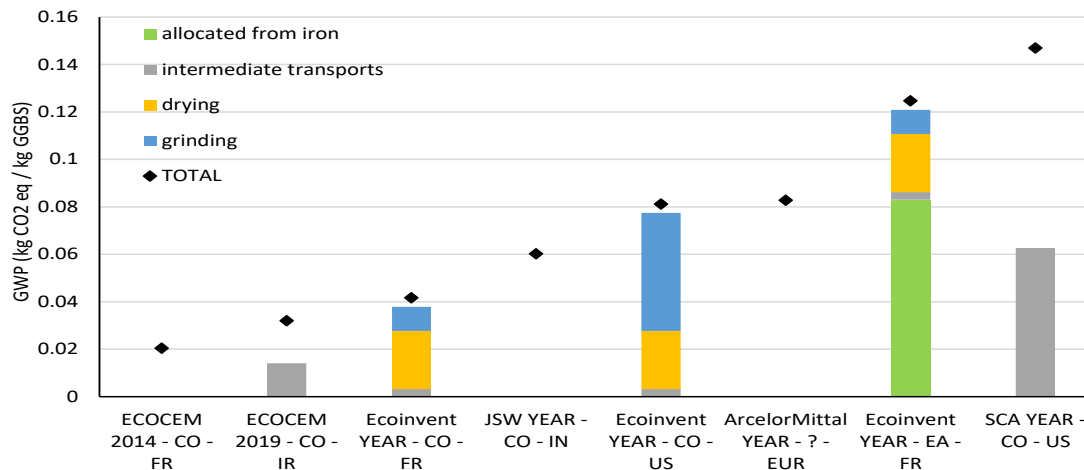


Figure 8: GWP of GGBS production obtained from the literature and contributions of allocation and unitary operations – Code of x-axis: Abbreviated reference – Year – allocation method – country (FR France, IR Ireland, IN India, EUR Europe, US United States of America)

5 CONCLUSION

This literature review showed that the question of functionalities of cements is not sufficiently addressed and that there is no scientifically explained relationship provided in literature that is capable of calculating the reference flow according to different strength classes. This aspect is very important since the material efficiency strategy can only be assessed if LCAs of cements can account for a decrease of required amount of cement according to an increase of its quality. According to existing studies, the main function of cement can be defined by its average compressive strength at 28 days measured on mortar according to NF EN 206+A2/CN standard. With this functional unit, we suggest to use Bolomeys' model [65] to calculate the reference flow. We show that for the same intended use in concrete of known compressive strength, the variability of the reference flow is significant for these cements of different qualities in the same strength class.

At the same time, we have explored how the effect of increasing cement quality on energy consumption is taken into account in the literature, as this requires a longer grinding time in the production process. A direct and scientifically based relationship between the increase of the fineness of cement and the electricity energy for grinding is however not found in the existing literature. The few values of energy consumption that were found are dispersed. A quick review of this aspect leads to the belief that this energy consumption is more related to the equipment than to the material itself, and we would need to have reference energy consumption data for each type of crusher. However, our results also show that the amount of electric energy required for grinding is generally smaller than that required for calcination by the combustion of fossil fuels in the clinkerization process. Producing a cement with higher quality, should not lead to significantly higher impacts on the environment, but could lead to fewer amounts of cement for identical mechanical performances of concrete. Beyond mechanical properties alone, it was previously shown that increasing the cement fineness allowed to reduce concrete porosity and thus allows using less concrete for an identical target service life performance [81], or allowing an increased durability. Material efficiency strategy is thus a promising strategy that could be more visible if LCAs of cements directly considered their mechanical performances in the functional unit.

Waste incineration can be considered as a good strategy to decrease the total CO₂ emissions resulting from the combustion process since the emissions associated with waste incineration are often not allocated or attributed to a specific fuel category. Moreover, the use of secondary fuels, can yield to very influential results depending on the allocation method employed.

Concerning the use of the recovered waste as Supplementary Cementitious Materials, the literature is very abundant concerning Granulated Ground Blast Furnace Slag, but not on other ones. Results are very dependent on the chosen allocation method that can considerably change the results of LCAs of blended cements. Allocation methods cannot be defined on a scientific basis, as they are political [55]. However, they should be harmonized between industrial sectors so as to avoid each sector choosing the method that most reduces its own impact. This could lead to data being less consistent and reliable when comparing different sectors. This is not the case today in Europe and it can conduct to omissions of impacts when LCA data from several sectors are used in the same LCA, precisely in the case of slag concrete [82]. A recent PhD report [83] has shown that mass allocation could be the most appropriate method in terms of mineral resource availability. Besides, we also showed important variabilities of climate change impacts on the recovery process of GGBS itself. These are due to several aspects: the amount and source of energy used for drying the slag after its quenching, the grinding energy and the transportation. Therefore, further LCA models should better account for variabilities of recovery processes that produce SCMs.

Finally, forthcoming research should focus on developing an LCA cement model taking into account the existing gaps analyzed herein. Such a model would help identify best and most efficient pathways towards low carbon cements and concrete.

ACKNOWLEDGMENTS

This research was funded in whole by the French National Research Agency (ANR) under the project ANR-20-CE22-0008-01 DEMCOM.

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