Environmental assessment of cement production with added graphene

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Nomenclature list

Bq	Becquerel
CE	Circular Economy
CFC	Chlorofluorocarbon
DB	Dichlorobenzene
FEP	Freshwater Eutrophication Potential (P eq.)
FETP	Freshwater Ecotoxicity Potential (1,4-DB eq.)
FPMFP	Fine Particulate Matter Formation Potential ($PM_{2.5}$ eq.)
FRSP	Fossil Resource Scarcity Potential (oil eq.)
GHG	Greenhouse Gas
Gr	Graphene
GWP	Global Warming Potential (CO_2 eq.)
HTP_c	Human Toxicity Potential, carcinogenic (1,4-DB eq.)
HTP_{nc}	Human Toxicity Potential, non-carcinogenic (1,4-DB eq.)
kWh	Kilowatt-hour
IRP	Ionizing Radiation Potential (Bq Co-60 eq.)
LCA	Life Cycle Assessment
LUP	Land Use Potential (m^2a eq.)
MEP	Marine Eutrophication Potential (N eq.)
METP	Marine Ecotoxicity Potential (1,4-DB eq.)
MJ	Megajoule
MRSP	Mineral Resource Scarcity Potential (Cu eq.)
NMVOC	Non-Methane Volatile Organic Compounds
OFP_{hh}	Ozone Formation Potential, human health (NO_x eq.)
OFP_{te}	Ozone Formation Potential, terrestrial ecosystems (NO_x eq.)
OPC	Ordinary Portland Cement
SODP	Stratospheric Ozone Depletion Potential (CFC-11 eq.)
t	tonne
TAP	Terrestrial Acidification Potential (SO_2 eq.)
TETP	Terrestrial Ecotoxicity Potential (1,4-DB eq.)
WCP	Water Consumption Potential (L)
yr	year

Abstract

Cement production significantly contributes to climate change, necessitating alternatives to mitigate the environmental impacts of this essential construction material. This study evaluates 18 environmental impacts of producing Ordinary Portland Cement (OPC) and Graphene (Gr) using life cycle assessment (LCA). Additionally, we explore whether mixing OPC and Gr can lower the life cycle environmental impacts of the final product (OPC_{Gr}). Our results show that OPC production in the United Kingdom generates 775 kg CO_2 eq./t, 57% only from geogenic CO_2 emissions. Gr production via electrochemical exfoliation in Australia results in 121,000-143,000 kg CO_2 eq./t, primarily due to electricity generation. Using hydro and nuclear power (e.g., in Brazil and France) can sharply reduce these impacts (global warming potential in the range of 11,000-35,000 kg CO_2 eq./t). Adding 0.02 wt% of Gr in powder form (Gr_{powder}) from Australia to the OPC and assuming a 16.5% reduction in its usage due to increased strength, results in 674 kg CO_2 eq./t OPC_{Gr} (a 13% reduction). However, some impact categories like marine eutrophication and freshwater ecotoxicity potentials increase sharply (> 28%). Using Gr_{powder} from Brazil and France further reduces the OPC_{Gr} global warming potential and the overall environmental footprint.

Keywords: construction materials; built environment; life cycle assessment (LCA); climate change mitigation; nanomaterials; composites.

1. Introduction

The construction sector is a major contributor to climate change. It has been estimated that, in 2009 alone, it was responsible for approximately 23% of the total CO_2 emissions (Huang et al., 2018). Among building materials, concrete stands out due to its versatility and strength. Its production involves the use of cement, water, gravel and admixtures. Of these inputs, cement has been identified as the most significant contributor to environmental impacts, being responsible for 5-8% of anthropogenic greenhouse gas (GHG) emissions (Gallego-Schmid et al., 2020). This mainly originates during the calcination and clinker formation steps since they require a large input of energy from fossil fuels and result in high geogenic CO_2 emission (Andersson et al., 2019; Petek Gursel et al., 2014). Recent studies suggest that every tonne (t) of cement produced in recent decades emitted 550-1,000 kg of CO_2 eq. (Dahanni et al., 2024; Georgiades et al., 2023). Although considerable reductions in energy consumption have been achieved in recent decades, producing one tonne of cement requires 20-200 kWh of electricity and 2,000-5,000 MJ of heat (Dahanni et al., 2024; Madlool et al., 2011). To evaluate impacts beyond CO_2 eq. emissions, life cycle assessment (LCA) can be used for estimating multiple categories from a “cradle-to-grave” perspective, providing a more comprehensive picture of the environmental burdens associated with cement production (Ige et al., 2021; Salas et al., 2016).

Among the standard options evaluated to reduce the environmental impacts of cement production are co-processing, waste heat recovery (Nidheesh and Kumar, 2019), and the addition of wastes such as used tyres, plastics, sewage sludge, and blast furnace slag (Dahanni et al., 2024; Georgiades et al., 2023; Hansted et al., 2022). More recently, the addition of graphene (Gr) and its derivates (e.g., graphene oxide, carbon nanotubes) to cement have been included in the list of promising routes to achieve more sustainable cement production (Makul, 2020; Zhao et al., 2020). This comes from the extraordinary physical attributes of Gr, including its large surface area (2,630 m^2/g) and tensile strength (130 GPa) (Lin and Du, 2020; Salami et al., 2023), with recent findings confirming significant enhancements in cement properties at low dosages (Chuah et al., 2014; Mukherjee et al., 2023), enabling lighter concrete structures. For instance, research on the addition of Gr at dosages of 0.05 wt% to Ordinary Portland Cement (OPC) increased its compressive strength by 79% and its tensile strength by 8%, (Krystek et al., 2019). The same dosage of graphene oxide was found to enhance the compressive strength of cement-waste concrete powder composite by over 19% (Sui et al., 2021). Furthermore, developments in the bulk production of Gr, especially electrochemical exfoliation (Achee et al., 2018; Liu et al., 2019; Yu et al., 2015), have led to a greater level of confidence in its production at industrial scales.

In this sense, studies have attempted to evaluate how graphene can improve cement/concrete properties while promoting environmental benefits. Long et al. (2018) indicated that mortars made of recycled fine aggregates containing 0.20 wt% of graphene oxide resulted in up to 6.7% GHG emissions reduction when compared to mortars with natural aggregates with equivalent strength. However, other environmental impacts have not been considered to fully attest to the environmental performance. Papanikolaou et al. (2019) performed the LCA of incorporating graphene nanoplatelets for self-sensing concrete structures, which indicated positive environmental results. However, these results focus on concrete production, graphene nanoplatelets produced in Italy, and focus on normalized person-equivalent results without detailing absolute values and trade-offs among the midpoint categories. Moreover, difficulties persist regarding the incorporation of Gr into cementitious materials due to poor dispersion and high associated costs (Huang et al., 2024; Yao et al., 2022).

Despite the above, it is important to acknowledge some gaps in the literature on these topics persist. First, studies on the life cycle environmental impacts of cement production in the Global North can now be considered to be outdated (most are previous 2014) due to older background databases and impact assessment methodologies (Bueno et al., 2016; Lu et al., 2017; Petek Gursel et al., 2014). Recent literature on cement production impacts comes from the Global South (more specifically Turkey, Brazil, Ecuador, China, Ethiopia and Myanmar) (Çankaya and Pekey, 2019; Petroche and Ramirez, 2022; Song et al., 2016; Stafford et al., 2016; Thwe et al., 2021; Wolde et al., 2024). In this sense, even though the work of Georgiades et al. (2023) provides an overview of potential routes for CO_2 eq. mitigation in cement production in Europe over the coming decades, it does not address other life cycle impact categories or the need for a better understanding of the initial data sources used for these estimations. Consequently, there is still a need to update initial assumptions and provide a more precise evaluation of the raw data used for building scenarios for future cement production in the UK and EU, especially given the complexities of cement production sustainability. Secondly, the environmental impacts of graphene production are more than often based on estimations from laboratory-scale studies, posing challenges in extrapolating findings to industrial scales (Cossutta et al., 2017; Munuera et al., 2022). Thirdly, thus far, a comprehensive assessment of the life cycle environmental advantages of graphene-enhanced cement has not been conducted. More specifically, the interaction between cement and graphene remains unexplored in terms of their detailed mid-point life cycle impacts. Research in this domain predominantly examines graphene-based nanomaterials, or concrete and loess (e.g., Yuan et al. (2023), Long et al. (2018) or Papanikolaou et al. (2019)), overlooking some of the dynamics and environmental trade-offs of graphene-cement mixtures demonstrated via updated and detailed industry data sources. These limitations hinder the ability to showcase the potential benefits that graphene could offer to the construction industry.

The main novelty of this paper relies on being the first robust, systematic and detailed LCA exploring the several life cycle environmental impacts of OPC_{Gr} production (i.e., Gr-enhanced OPC). As mentioned in the previous paragraph, the literature currently explores these topics separately, is based on laboratory scale studies, or explores similar materials (e.g., concrete, loess, graphene nanoplatelet, graphene oxide). In contrast, we considered detailed industry data for both OPC and Gr productions, along with our experimental results regarding OPC_{Gr} performance and mixing. We provided full results and tradeoffs among 18 midpoint life cycle impact categories. Additionally, our study addresses two further gaps in the literature. The first is the lack of up-to-date environmental impact results for cement production in the Global North, specifically on studies using recent background databases and environmental impact assessment methodologies. Lastly, it provides for the first time the life cycle environmental profile of industrial scale Gr production via electrochemical exfoliation of graphite (current estimates rely on laboratory scale information - e.g., Cossutta et al. (2017) and Munuera et al. (2022)). The results together with the life cycle inventories provided for both OPC and Gr production are expected to facilitate studies on these materials in the years to come. Finally, there is a discussion on how the results can aid in decreasing the environmental impacts of cement.

2. Methodology

This section describes the methodological steps adopted in the study. A description of the OPC production facility in the UK and the Gr production facility in Australia can be found in section 2.1, and supplementary information (SI) section SI-1. Section 2.2 depicts the methodology adopted for the experiments adding 0.02 wt% of Gr into OPC to ascertain the potential effects on mechanical performance characteristics and environmental impacts of the resultant Gr-enhanced OPC (OPC_{Gr}). Lastly, section 2.3 presents the life cycle assessment

(LCA) methodology for estimating the environmental impacts associated with OPC, Gr, and OPC_{Gr} production.

2.1. OPC and Gr production facilities

The OPC production facility considered in this case study is one of the largest of its kind in the UK (output of 1,400,000 t/yr). The facility is integrated, with two similarly configured process lines. This implies that the major constituents (mainly limestone and shale) of the hydraulic binder (i.e., clinker) in the cement are all sourced from local quarries that are local to the plant, while other constituents are sourced from several locations across the UK. The raw materials from the quarries initially undergo size reduction in a crusher, so that the raw mixture can be fed to the raw mills, where they are further pulverised into fine powders (raw meal). The raw meal is then fed into a blending silo for homogenisation, to enhance the uniformity index at the pyroprocessing stage downstream. Pyroprocessing occurs at two stages - pre-calcination (in the pre-calciner for fuel optimisation and heat recovery) and calcination (in the rotary kilns). The output of the rotary kilns is the clinker, which is produced at a temperature of 1450 ^{o}C and later quenched to less than 100 ^{o}C in the grate coolers (Yunusa-Kaltungo et al., 2017; Yunusa-Kaltungo and Labib, 2021). A mixture of different proportions of pulverised coal, waste tyres, paper, and plastics are used to fuel the rotary kilns during clinkerisation.

The data for Gr production in this study was provided by a graphene manufacturer based in Australia. The method utilized in their production facility is the electrochemical exfoliation of graphite to produce paste (Gr_{paste}) and powder (Gr_{powder}) forms of graphene. This method is currently considered to be commercially viable because of its robustness, cost efficiency and scalability (Danial et al., 2021; Liu et al., 2019; Park et al., 2021). The plant is located near Perth (Western Australia) and produces around 100 t/yr.

2.2. Adding Gr to OPC (OPC_{Gr})

Experiments were conducted considering only Gr_{powder}, according to the recently published article by Yunusa-Kaltungo et al. (2024). Therefore, experiments using the Gr_{paste} have not been included in this work as its dispersion efficiency has not been evaluated. The rationale for the experimental design is to explore the equivalent water/cement ratio of the samples with and without graphene in terms of 28-day standard compressive strengths. The purpose of the experimental programme is not to prove the concept of graphene-enhanced concrete, which has been well-known and proved by a great number of researchers in the past decade (Dung et al., 2023; Lin and Du, 2020). Thus, laboratory experiments confirmed the performance benefits achievable by adding Gr_{powder} to OPC and allowed us to understand Gr's behaviour and the need to optimise the addition process (especially concerning cost-effectiveness and environmental friendliness) (Ghazizadeh et al., 2018; Yunusa-Kaltungo et al., 2024). A dosage of 0.02 wt% of Gr_{powder} (i.e., every tonne of OPC_{Gr} contains 20 kg of Gr) has been considered based on previous literature (Ho et al., 2020a, 2020b; Lin and Du, 2020). The experiments were made using mortar materials comprising cement, water, and sand to cast 50-mm cubes. Five mixing formulae were designed: one mixture contained 0.02 wt% Gr_{powder} and four mixtures were without graphene but the water/cement ratio varied (Table 1).

Table 1 – Materials weights for the mortar mixtures used in the experiments.

	Quantity (in grams)				
Mixture number	Graphene	Cement	Sand	Water	Total
1	0.00	1488	2475	612	4575.0
2	0.00	1424	2475	612	4511.0
3	0.00	1360	2475	612	4447.0
4	0.00	1276	2475	612	4363.0
5	0.25	1276	2475	612	4363.3

OPC CEM I 52.5N from Breedon Cement was used. The flow table tests were conducted before casting according to ASTM C230/C230M-21 (ASTM, 2021). After casting, the cubes were placed in a moisture room for 7-day and 28-day curing. A series of comprehensive tests were then conducted according to ASTM C109-2020 (ASTM, 2020). The dispersion method chosen was the dry addition (also known as the powder-to-powder dispersion method) due to its advantages such as simplicity and negligible requirements for further processing (Basquiroto de Souza et al., 2022; Lin and Du, 2020). The dispersion of Gr_{powder} was achieved through a purpose-designed and built fluidised bed that receives primary and secondary blending air from low-energy blowers. The electricity consumption of the fluidised bed used to disperse the Gr_{powder} into OPC was estimated by the authors during the experiments (resulting in 8.5 kWh/t) (Yunusa-Kaltungo et al., 2024), and this aspect is further commented on during the discussion section.

2.2.1. Composite properties

Table 2 compares the workability obtained from flow table tests of the samples. The results indicate that the incorporation of Gr_{powder} does not have clear effects on the flowability of the mortar paste since differences in flowability of all samples are within 15%. For the specimens without Gr_{powder} the strengths decrease when the water/cement ratio increases, indicating the smaller quantities of cement used. Moreover, even in the specimen with the largest water/cement ratio (i.e., when 0.02 wt% Gr_{powder} is added – mixture number 5), the compressive strength of the mortar samples improved and became higher than all the other samples.

Table 2 – Results for the compressive strengths and flowability for the 7-day and 28-day experiments with graphene in powder form (Gr_{powder}) added to Ordinary Portland Cement (OPC)

		7-day compressive strength (in Mpa)				28-day compressive strength (in Mpa)			
Mixture number	Flowability	Test 1	Test 2	Test 3	Average	Test 1	Test 2	Test 3	Average
1	165	56.01	56.04	56.44	56.16	67.77	65.10	65.58	66.15
2	170	54.33	56.35	54.26	54.98	65.25	66.89	64.27	65.47
3	175	53.71	53.51	53.37	53.53	63.61	64.52	61.35	63.16
4	180	50.1	48.97	48.76	49.28	61.23	58.37	61.88	60.49
5	183	55.2	55.18	61.14	57.17	68.02	75.40	62.98	68.80

By comparing values in samples 1 and 5, it can be observed that when as little as 0.02 wt% Gr_{powder} was utilized, more than 16.5 % cement could be saved while still achieving the same compressive strength. This could be attributed to the role of nucleation seeding of Gr to facilitate the hydration of cement particles and stimulate the formation of cement hydration products. Although the purpose-built fluidised bed used to homogenise the $G_{rpowder}$ and cement powders is laboratory scale with an approximately 50-litre capacity, its scale-up factor is very significant, when compared to other available laboratory equipment that can only handle 2-3 kg under extended residence times and energy consumption such as ultrasonicator, ultrasonic bath or high shear mixer (Dung et al., 2023).

2.3. Life cycle assessment

The LCA followed the attributional approach of the ISO 14040:2006 methodology (ISO, 2006a, 2006b). The four steps of the methodology are described in the next section.

2.3.1. Goal and scope definition

The first goal of this study is to evaluate the life cycle environmental impacts of the production of OPC in the UK and Gr in Australia; the second goal is to understand whether the addition of Gr can decrease the life cycle environmental impacts of OPC production in the UK. The functional unit considered in the study for OPC is "1 tonne (t) of product", and for Gr is "1 kilogram (kg) of graphene delivered". As already discussed in Dobbelaere et al., (2016), Sagastume Gutiérrez et al. (2017) and Ige et al. (2021), the different mechanical properties of the cement may impair the comparison with other LCAs. Therefore, eventual comparisons with the results found in this work are to be interpreted carefully and validated for OPC and OPC_{Gr} with similar characteristics to those described in section 2.2.

Figure 1 illustrates the life cycle stages of OPC production. Waste materials such as pulverized fly ash (PFA), papers & plastics, and tyre crumbs have been considered to generate impacts only from transport to the facility and onsite air emissions. Regarding the latter, air emissions other than carbon dioxide (CO_2), nitrogen oxides (NO_x), sulphur dioxide (SO_2), and non-methane volatile organic compounds (NMVOC) were not considered as they are assumed to be efficiently removed in the plant stack by bag filters. The water utilized for cooling purposes is sourced from local quarries. Figure 2 outlines the life cycle stages of the Gr production facility in Australia along with an additional stage for the transportation of the Gr product to the UK. It is worth noting again that Gr is produced in two forms: Gr_{paste} and Gr_{powder} (see section 2.3.2.2 for further information). Importantly, the infrastructure of the facilities (such as steel and other building materials) has not been considered due to their low significance to impacts from the equipment's long lifespan, recyclability and the significant impacts of the high energy consumption for these processes.

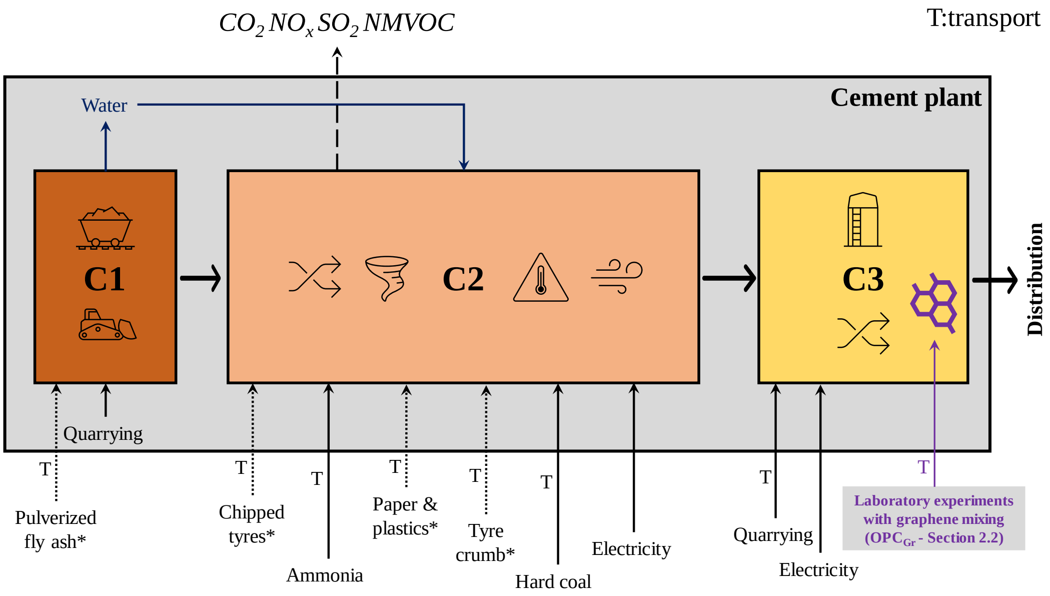

Figure 1 – Life cycle stages of Ordinary Portland Cement (OPC) production in the United Kingdom (see the addition of graphene at the end). Materials with an asterisk (*) are waste from other activities. For a detailed scheme showing the OPC production facility see SI section SI-1.

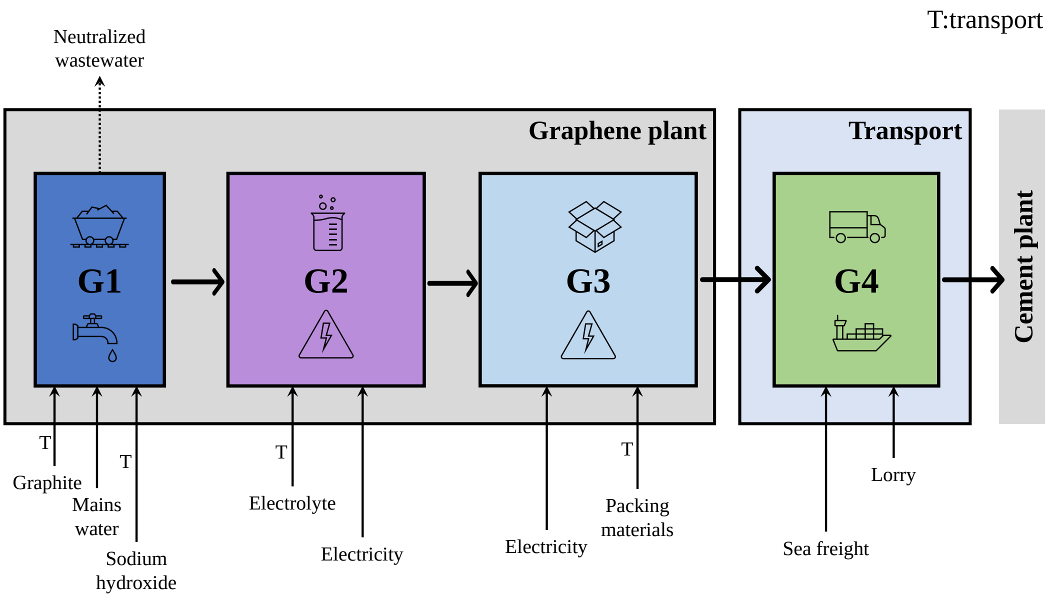

Figure 2 – Life cycle stages of graphene (Gr) production (for both paste and powder forms) and transportation to the cement production facility in the UK (see Figure 1). For a detailed scheme showing the Gr production facility see SI section SI-1.

2.3.2. Inventory analysis

The results of the life cycle inventory analysis of OPC and Gr production are presented in the next sections.

2.3.2.1. OPC production

Except for onsite air emissions constituents NO_x, SO_2 and NMVOC that were acquired from ecoinvent database, the information depicted in Table 3 was obtained from the aforementioned UK-based OPC manufacturing plant. Regarding onsite CO_2 emissions, it was reported to be 65% from geogenic origin and 35% from fuels combustion.

Table 3 - Life cycle inventory for the Ordinary Portland Cement (OPC) production in the United Kingdom. See Table S1 in SI-2 for ecoinvent v3.8 correspondence. Values per functional unit ("1 t of product").

Stage	**Value**	**Unit**	**Source**
C1 – Raw materials			
Limestone, milled	1,207	kg	OPC plant
Shale, milled	210.4	kg	OPC plant
Pulverized fly ash	70.51	kg	OPC plant
Transport, lorry	7.05	t.km	OPC plant
C2 – Calcination and clinker formation			
Chipped tyres	25.91 (710)	kg (MJ)	OPC plant
Ammonia	3.71	kg	OPC plant
CO_2	679	kg	OPC plant
NO_x	1.08	kg	ecoinvent
SO_2	0.35	kg	ecoinvent
NMVOC	0.06	kg	ecoinvent
Electricity	86.0	kWh	OPC plant
Paper & plastics	30.21 (426)	kg (MJ)	OPC plant
Tyre crumb	8.28 (284)	kg (MJ)	OPC plant
Hard coal	78.89 (2,130)	kg (MJ)	OPC plant
Transport, lorry	13.81	t.km	OPC plant
C3 – Cement milling			
Electricity	41.0	kWh	OPC plant
Limestone, milled	51.48	kg	OPC plant
Gypsum, milled	60.71	kg	OPC plant
Transport	4.86	t.km	OPC plant

Calorific values: chipped tyres 27.4 MJ/kg; paper & plastic 14.1 MJ/kg; tyre crumb: 34.3 MJ/kg. hard coal 27.0 MJ/kg.
The total heat required for clinker production is 3,550 MJ/t.

2.3.2.2. Gr production

Except for graphite production, energy for mains water supply, and transportation distances, the information found in Table 4 was obtained directly from the graphene manufacturer. As mentioned earlier the electrochemical exfoliation uses natural graphite, and for this study, we have considered generic sources obtained within Asia (Surovtseva et al., 2022). The transportation of the graphite to the facility, located near Perth in Western Australia, is carried out via sea freight. The production process involves multiple vessels containing electrolyte solutions, where the application of an electric current generates exfoliated Gr. More detailed information on the graphene production falls outside the scope of this LCA study as the process is still under development and information is currently unavailable/confidential.

The water for the facility is supplied by city mains and its energy intensity is estimated based on Perth's metropolitan area supply (35% desalinated, 36% groundwater, 26% surface water and 3% replenishment) (Water Coorporation, 2022), resulting in estimated electricity consumption of 1.45 kWh/m^3 - desalination 3.27 kWh/m^3, surface and groundwater 0.41 kWh/m^3, and replenishment 1.84 kWh/m^3 - based on values found in Tarpani et al. (2021). The facility consumes water at a rate of 0.89 m^3/kg of Gr and requires additional treatment through reverse osmosis to attain the desired quality before use in the process. The resulting wastewater is treated and then discharged under strict control into the local collection system. Since the resulting effluent contains a very low organic load and amounts to a negligible volume compared to the city's total volume, it was not included in the LCA system boundary.

The Gr production process achieves a yield of up to 90% for the graphite, with any residual graphite assumed to be either discarded in the wastewater without causing environmental harm or reused within the process. The solution containing Gr within the vessels undergoes filtration and drying. The drying process aims to achieve 80% water content to produce Gr_{paste} and 0% water content to produce Gr_{powder}. The Gr contains approximately 4% oxygen (C/O = 0.042), 5-10 layers, and contains less than 1% inorganic material. Afterwards, the Gr product is transported from Western Australia to Derbyshire in the UK. The transportation involves a sea freight journey spanning 17,000 km and a subsequent 100 km journey by road.

Table 4 - Life cycle inventory for graphene (Gr) production (in both paste and powder) by electrochemical exfoliation in Australia and transport to the United Kingdom. See Table S1 in SI-2 for ecoinvent v3.8 correspondence. Values per functional unit ("1 kg of graphene delivered").

Stage	Value	Unit	Source
G1 – Raw materials and wastewater treatment			
Graphite	1.10	kg	Gr plant
Transport, sea freight	8.11	t.km	Estimated
Electricity (water supply)	1.29	kWh	Estimated
Sodium hydroxide, liquid	0.32	kg	Gr plant
G2 – Electrochemical exfoliation			
Electricity	60	kWh	Gr plant
Sulphuric acid, liquid	0.16	kg	Gr plant
G3 – Reverse osmosis, mixing, heating, chilling and other			
Electricity	61* / 85**	kWh	Gr plant
Low-density polyethylene	25* / 50**	g	Gr plant
Cardboard	115* / 200**	g	Gr plant
G4 – Transportation to the United Kingdom			
Transport, sea freight	85* / 17**	t.km	Estimated
Transport, lorry	0.50* / 0.10**	t.km	Estimated

* Paste. **Powder.

2.3.3. Impact assessment

The SimaPro 9.3.0.2 software (PRé Sustainability Software, 2023) was used for process modelling. To provide and updated, representative and complete life cycle impact assessment (Bueno et al., 2016; Esnouf et al., 2018), the 18 midpoint impact categories from the latest impact assessment methodology available, ReCiPe 2016 midpoint (H) v1.06 (Huijbregts, 2016), are calculated and discussed in the following order: Global Warming Potential (GWP), Fossil Resource Scarcity Potential (FRSP), Mineral Resource Scarcity Potential (MRSP), Water Consumption Potential (WCP), Stratospheric Ozone Depletion Potential (SODP), Ozone Formation Potential - terrestrial ecosystems (OFP_{te}), Ozone Formation Potential - human health (OFP_{hh}), Particulate Matter Formation Potential (PMFP), Ionizing Radiation Potential (IRP), Terrestrial Acidification Potential (TAP), Freshwater Eutrophication Potential (FEP), Marine Eutrophication Potential (MEP), Terrestrial Ecotoxicity Potential (TETP), Freshwater Ecotoxicity Potential (FETP), Marine Ecotoxicity Potential (METP), Human Toxicity Potential - cancer (HTP_c), Human Toxicity Potential - non-cancer (HTP_{nc}), and Land Use Potential (LUP). To further increase the robustness and comprehensiveness of the study, the ecoinvent database v3.8 cut-off (Lu et al., 2017; Wernet et al., 2016) was used as background processes (see Table S1 in section SI-2 for correspondences).

2.3.3.1. *Sensitivity analysis*

Due to timely concerns over the climate change potential of Gr production, this study firstly provides a sensitivity analysis for the results obtained for GWP (in kg CO_2 eq./kg) when the Gr is produced in different locations of the world (Australia, Brazil, China, France, the UK and the United States) (see Table 5). Note that it is assumed that the Gr manufacturing process in these locations is the same as in Australia and consumes the same amount of energy and materials to produce 1 kg of graphene (in both paste and powder forms). In addition to Australia and the UK, these locations were selected based on their significant global share of cement production (Brazil, China, and the United States collectively account for over 65% of global cement production) (Nidheesh and Kumar, 2019; Schneider et al., 2011) in addition to their diverse electricity generation sources (e.g., Brazil, China and France – see Table 5).

Table 5 - Electricity generation mix in 2018 by source for countries considered during the sensitivity analysis of graphene production (International Energy Agency, 2023).

Country	Coal	Natural gas	Oil	Nuclear	Hydro	Wind	Solar	Biofuel	Geothermal	Waste
Australia	61.2%	20.9%	1.9%	0.0%	6.2%	5.8%	3.9%	0.0%	0.0%	0.0%
Brazil	3.3%	9.9%	2.3%	2.7%	65.1%	6.9%	0.6%	9.2%	0.0%	0.0%
China	66.6%	2.9%	0.1%	4.2%	17.1%	5.1%	2.5%	1.2%	0.0%	0.2%
France	2.0%	5.5%	0.0%	73.2%	11.0%	4.4%	2.0%	1.1%	0.0%	0.8%
United Kingdom	5.4%	39.7%	0.5%	19.5%	2.4%	17.1%	3.9%	9.3%	0.0%	2.1%
United States	29.2%	33.6%	1.0%	19.1%	7.1%	6.2%	2.2%	1.3%	0.4%	0.0%

A second sensitivity analysis was made to analyse the life cycle environmental impacts of OPC_{Gr} for a Gr_{power} dosage of 0.02 wt% (as depicted in section 2.2). This was made by comparing samples 1 and 5, where it can be observed that when as little as 0.02 wt% of Gr_{powder} was utilized more than 16.5 % cement can be saved while still achieving the same compressive strength (Table 2). From this result, it was assumed that a proportionally lower consumption of OPC_{Gr} is required to achieve the same desired results in terms of mechanical strength compared to the OPC discussed in section 2.3.2.1 (i.e., an increase of 16.5% in compressive strength reduces in 16.5% the amount of OPC_{Gr} needed). This has also been analysed considering when Gr_{powder} is being produced in the countries found in Table 5. The results are shown and commented on in section 3.3.

3. Results

The results for the life cycle impacts of OPC production in the UK can be found in section 3.1. After, the life cycle impacts of Gr production in Australia are discussed in section 3.2, including the sensitivity analysis for GWP when its production occurs in different countries (as commented in section 2.3.3.1). Finally, the life cycle impacts of OPC_{Gr} are discussed in section 3.3.

3.1. Impacts of OPC production

The following subsections comment on the life cycle impacts results of OPC production and their respective environmental hotspots. The results, according to the life cycle stages defined in section 2.3.1, can be found in Figure 3; and the contributions to these impacts are outlined in Figure 4. It can be seen that calcination and clinker formation during stage C2 account for the majority of the environmental impacts associated with OPC. This particular stage alone is responsible for more than 70% of the environmental impacts, except in MRSP, SODP, and TETP. An overall analysis of the results reveals that the onsite release of CO_2, NO_x, and SO_2 during stage C2 is the primary cause of impacts in terms of GWP, OFP_{te}, OFP_{hh}, FPMFP, and TAP. Electricity consumption is the main driver of impacts in SODP, IRP, and LUP, and it also significantly contributes to eco and human toxicities. Furthermore, the extraction and preparation of hard coal as a fuel for stage C2 contributes to over 40% of the impact in FRSP, FEP, MEP, FETP, and METP, as well as human toxicities.

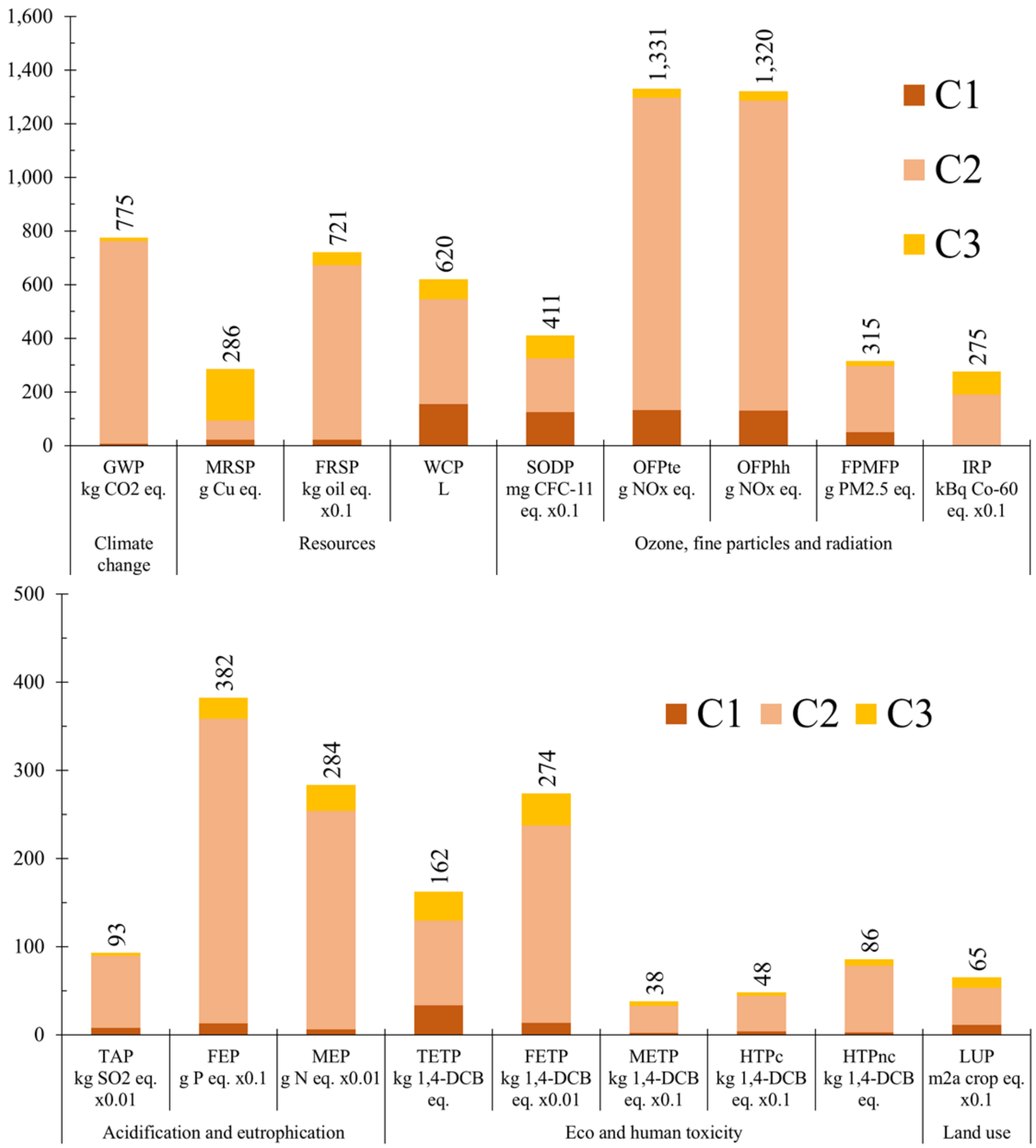

Figure 3 - Life cycle impacts for the Ordinary Portland Cement (OPC) production in the United Kingdom (for life cycle stages see Figure 1). Results per functional unit “1 t of product”. GWP: Global Warming Potential. FRSP: Fossil Resource Scarcity Potential. MRSP: Mineral Resource Scarcity Potential. WCP: Water Consumption Potential. SODP: Stratospheric Ozone Depletion Potential. OFP_{te}: Ozone Formation Potential - terrestrial ecosystems. OFP_{hh}: Ozone Formation Potential - human health. FPMFP: Fine Particulate Matter Formation Potential. IRP: Ionizing Radiation Potential. TAP: Terrestrial Acidification Potential. FEP: Freshwater Eutrophication Potential. MEP: Marine Eutrophication Potential. TETP: Terrestrial Ecotoxicity Potential. FETP: Freshwater Ecotoxicity Potential. METP: Marine Ecotoxicity Potential. HTP_c: Human Toxicity Potential – cancer. HTP_{nc}: Human Toxicity Potential – non-cancer. LUP: Land Use Potential.

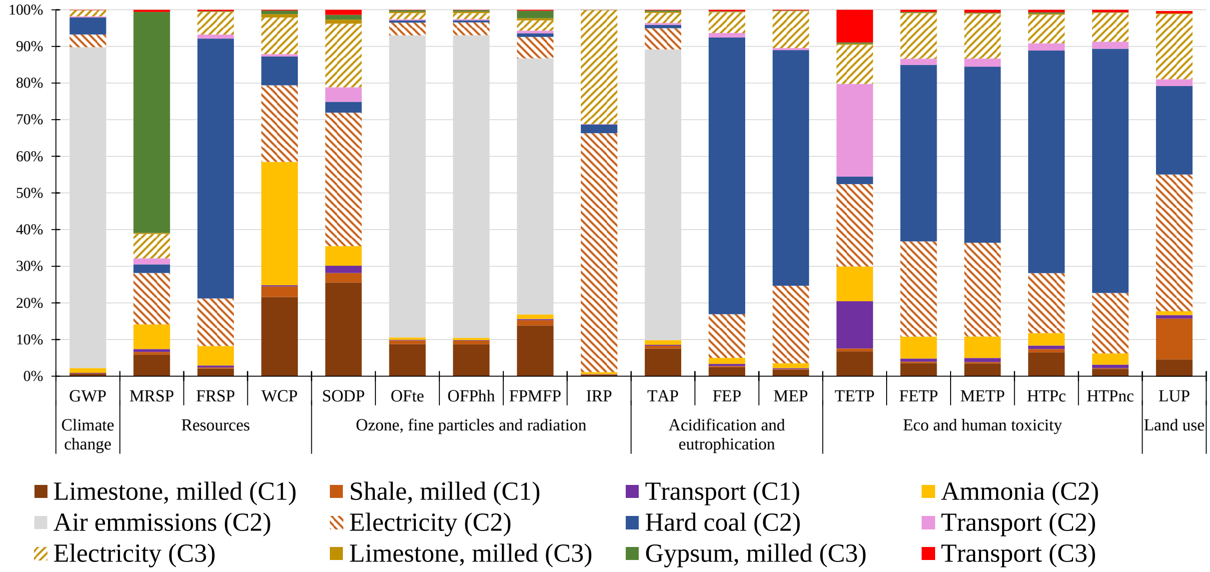

Figure 4 - Contributions (in % of the total impact) of the life cycle processes to the production of Ordinary Portland Cement (OPC) in the United Kingdom. See Figure 3 for impact categories nomenclature.

3.1.1. Climate change

The GWP of OPC production in the UK has been estimated to be 775 kg CO_2 eq./t, as depicted in Figure 3. Interestingly, 88% of the emissions contributing to this impact originate directly from the calcination and clinker formation – see Figure S1. Of the emissions during these processes, 65% are CO_2 emissions of geogenic origin (441.35 kg CO_2 eq./t) while 237.65 kg CO_2 eq./t is attributed to the combustion of hard coal and wastes being used as fuel in the kilns. Therefore, 57% of the total GWP of the OPC is solely from geogenic CO_2 emission. From the remaining 12%, hard coal mining and preparation, and electricity consumption contribute each to approximately 5% (or 39 kg CO_2 eq./t) each.

3.1.2. Resources

The MRSP is estimated to be 286 g Cu eq./t of OPC. It is worth noting that gypsum used during the cement milling stage (C3) is the primary contributor to this impact (approximately 60% of the total) and can be attributed to this mineral being considerably less abundant when compared to limestone or shale (United States Geological Survey, 2020). FRSP amounts to 72.1 kg oil eq./t. The main contributor, approximately 71% of the total impact, is attributed to the depletion of hard coal used as a fuel during calcination and clinker formation (stage C2). WCP is approximately 620 L/t, and ammonia production used during stage C2 and electricity consumption (in stages C2 and C3), contribute each for approximately 31% of the total impact. The first comes from steam reforming processes in ammonia production and the second is mostly from decarbonized water for cooling towers and flue gas scrubbers in nuclear and natural gas power plants.

3.1.3. Ozone, fine particles and radiation

The SODP associated with the production of OPC is estimated at 41.1 mg CFC-11 eq./t. Of this total, nearly 25% is attributed to the quarrying of limestone and shale (from N_2O to air from the use of explosives containing calcium and ammonium nitrate for blasting). Electricity consumption during stages C2 and C3 contributes to 50% of the total in this impact, from N_2O emissions from fossil fuel burning reaching the stratosphere. Regarding the impacts of OFP_{te} and OFP_{hh}, the results are similar, with values of 1.33 kg NO_x eq./t and 1.32 kg NO_x eq./t, respectively. Onsite emissions during stage C2 account for 82% of the total impact in both categories, while limestone quarrying in stage C1 makes a significant contribution of 9-10% to these two impacts due to NO_x emissions from diesel combustion in machinery. For FPMFP, the onsite emissions of SO_2 during calcination and clinker formation (stage C2) play a major role, contributing to 71% of an estimated total of 315 g $PM_{2.5}$/t. Lastly, the impact on IRP is estimated at 27.5 kBq Co-60 eq./t. Electricity consumption during stages C2 and C3 is responsible for nearly all the impact in this category, more specifically from Radon-22 emissions derived from Uranium milling residues associated with nuclear power generation.

3.1.4. Acidification and eutrophication

The TAP associated with the production of OPC is 0.93 kg SO_2 eq./t. Among the contributors, onsite air emissions of NO_x and SO_2 during stage C2 account for 79% of this total. Other significant contributions of 7-9% come from limestone quarrying in stage C1 and electricity consumption during stages C2 and C3. For FEP and MEP, the results are 38.2 g P eq. and 2.84 g N eq./t, respectively. The primary contributor to both categories is hard coal mining and preparation in stage C2, responsible for 75% and 65% of the impacts, respectively.

These two impacts are largely derived from mining activity spoils containing phosphate (PO_4^{3}) and nitrate (NO_3^-) in water. Electricity consumption during stages C2 and C3 is the next major contributor, accounting for 20% of the FEP impact and 35% of the MEP impact.

3.1.5. Eco and human toxicity

The TETP associated with the production of OPC is estimated at 162 kg 1,4-DCB eq./t. Various processes contribute significantly to this total, but approximately 80% of the overall impact is associated with the transportation of materials, more specifically hard coal and wastes (i.e., PFA, chipped tyres, paper & plastics, tyre crumbs) to the facility, emitting copper and antimony to air from brake wear. A smaller share comes from electricity consumption at stages C2 and C3, originating from copper smelting and refining activities for transformation and transmission networks, as well as heat and power co-generation by wood chips, both resulting in emitting copper and zinc to air. For FETP, the result is 2.74 kg 1,4-DCB eq./t, while for METP it is 3.8 kg 1,4-DCB eq./t. Both have similar contribution profiles, with hard coal mining and preparation for fuel during calcination and clinker formation in stage C2 contributing to 45% of this total. The electricity consumption in stages C2 and C3 account for nearly 40% across both categories. Regarding HTP_c, the impact is estimated at 4.8 kg 1,4-DCB eq./t, while HTP_{nc} is 86 kg 1,4-DCB eq./t. The primary contributors to these categories are hard coal mining and preparation in stage C2 (60-66%) (mostly chromium VI emissions from mining-related activities), electricity consumption during stage C2 (19%), and electricity consumption during stage C3 (9%).

3.1.6. Land use

The results for LUP show that electricity consumption is the primary contributor to this impact, accounting for 54% of the total (estimated at 6.5 m^2a crop eq./t). More specifically, electricity consumption during calcination and clinkerisation (stage C2) contributes 36%, while electricity consumption during stage C3 contributes 18%. This impact is associated with the consumption of wood pellets from sustainable forest management in Sweden that are utilized for heat and electricity cogeneration in the UK. Other significant contributors include hard coal mining and preparation in stage C2, which accounts for 23%, and limestone and shale quarrying in stage C1, which contributes 15%.

3.2. Impacts of Gr production

Figure 5 shows the results for both forms of graphene, Gr_{paste} and Gr_{powder}, produced in the Australian facility while Table 6 the contributions (in %) for each life cycle stage. The analysis shows that Gr_{paste} has usually 15-20% lower impacts than Gr_{powder}. Among all the impact categories considered, electricity is identified as the primary contributor, accounting for > 97% of the total impacts, with only a few exceptions. This is the case for instance in OFP_{te} and OFP_{hh}, when transportation of Gr_{paste} from Australia to the UK (stage G4) contributes to 6.7% of the impact. However, in general, the impacts from the transportation of Gr from Australia to the UK are relatively low, being on average 2.2% for Gr_{paste} and 0.4% for Gr_{powder} in most impact categories. Overall, stage G2 is responsible for 40-48% while stage G3 accounts for 49-58% of the total impacts.

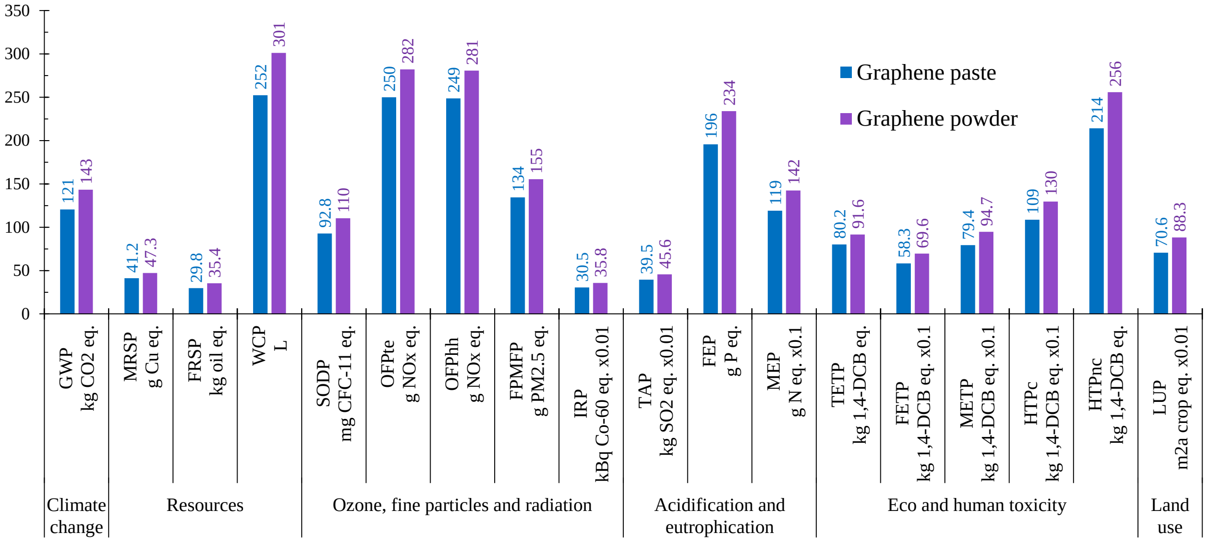

Figure 5 - Life cycle impacts for graphene (Gr) production in paste and powder forms. See Figure 3 for impact categories nomenclature and Table 6 for contributions from life cycle stages. Results per functional unit "1 kg of graphene delivered" produced in Australia and delivered to the United Kingdom.

Table 6 - Contributions (in % according to life cycle stages) to the environmental impact of graphene (Gr) produced in Australia and delivered to the United Kingdom. See the life cycle stages in Figure 2 and the total impacts in Figure 5.

	G1		G2		G3		G4	
Impact category	Paste	Powder	Paste	Powder	Paste	Powder	Paste	Powder
GWP	1.69%	1.42%	48.3%	40.6%	49.3%	57.9%	0.73%	0.12%
FRSP	5.00%	4.36%	46.7%	40.7%	43.9%	54.1%	4.44%	0.78%
MRSP	1.70%	1.43%	48.1%	40.5%	49.3%	57.9%	0.89%	0.15%
WCP	5.63%	4.72%	46.2%	38.7%	47.9%	56.5%	0.29%	0.05%
SODP	1.80%	1.52%	48.3%	40.6%	49.3%	57.8%	0.66%	0.11%
OFP_{te}	2.29%	2.03%	45.0%	39.8%	46.0%	56.9%	6.74%	1.19%
OFP_{hh}	2.29%	2.03%	45.0%	39.8%	46.0%	56.9%	6.73%	1.19%
FPMFP	2.33%	2.01%	46.3%	40.1%	47.4%	57.2%	3.99%	0.69%
IRP	17.0%	14.5%	36.4%	31.0%	43.4%	54.0%	3.30%	0.56%
TAP	2.04%	1.77%	46.5%	40.3%	47.3%	57.2%	4.20%	0.73%
FEP	1.38%	1.16%	48.8%	40.8%	49.7%	58.0%	0.07%	0.01%
MEP	1.44%	1.21%	48.8%	40.8%	49.8%	58.0%	0.03%	0.00%
TETP	3.72%	3.25%	47.6%	41.6%	44.3%	54.3%	4.47%	0.78%
FETP	1.67%	1.40%	48.9%	41.0%	49.3%	57.6%	0.14%	0.02%
METP	1.65%	1.39%	48.9%	41.0%	49.3%	57.6%	0.16%	0.03%
HTP_c	1.56%	1.31%	48.6%	40.8%	49.5%	57.9%	0.36%	0.06%
HTP_{nc}	1.51%	1.27%	48.9%	40.9%	49.5%	57.8%	0.08%	0.01%
LUP	2.88%	2.31%	40.0%	32.0%	54.9%	65.3%	2.21%	0.35%

GWP: Global Warming Potential. FRSP: Fossil Resource Scarcity Potential. MRSP: Mineral Resource Scarcity Potential. WCP: Water Consumption Potential. SODP: Stratospheric Ozone Depletion Potential. $OFPt_{te}$: Ozone Formation Potential - terrestrial ecosystems. $OFPh_{hh}$: Ozone Formation Potential - human health. FPMFP: Fine Particulate Matter Formation Potential. IRP: Ionizing Radiation Potential. TAP: Terrestrial Acidification Potential. FEP: Freshwater Eutrophication Potential. MEP: Marine Eutrophication Potential. TETP: Terrestrial Ecotoxicity Potential. FETP: Freshwater Ecotoxicity Potential. METP: Marine Ecotoxicity Potential. HTP_c: Human Toxicity Potential – cancer. HTP_{nc}: Human Toxicity Potential – non-cancer. LUP: Land Use Potential.

3.2.1. Climate change

The GWP of Gr_{paste} is 16% more than Gr_{powder} (121 kg and 143 kg CO_2 eq./kg, respectively). The primary contributor to the GWP for both forms of graphene is electricity consumption (from lignite, hard coal and natural gas burning for electricity generation in power plants). In the case of Gr_{paste}, nearly 48.3% of the impact is attributed to electricity consumption during the electrochemical exfoliation process (stage G2). In comparison, nearly 49.3% is attributed to electricity consumption during the processes involved in stage G3. For Gr_{powder}, stage G2 contributes 40.6% of the impact, and stage G3 contributes 57.9% due to the higher electricity consumption required for extra mixing, heating, and chilling to remove water content. Note the GWP results from the Australian electricity grid can be considered high since it is predominantly generated from fossil fuels (84%, see Table 5). Therefore, the next subsection depicts the results for the GWP of graphene production in other locations (see section 2.3.3.1).

3.2.1.1. Sensitivity analysis

The findings presented in Figure 6 show the results for the GWP for Gr production in the countries in Table 5. As can be seen, the results for the Australian electricity grid are relatively high, comparable to its production in China (120-143 kg and 130-156 kg CO_2 eq./kg respectively). This is largely due to the electricity sources of these two countries being mostly derived from fossil fuels (84% for Australia and 70% for China). The lowest results in this category were achieved for Brazil and France, as they are based on hydro (Brazil, 65%) or nuclear (France, 73%) power generation. Hence, the results for Gr production for electrochemical exfoliation in these countries are estimated at 29-35 kg CO_2 eq./kg in Brazil and 11-13 kg CO_2 eq./kg in France. The production of Gr in the UK and the United States showed intermediate results among the countries evaluated. For the latter, the results are lower since it employs more wind and biofuel energy sources, and less coal for electricity generation than the former. The results are 39-47 kg CO_2 eq./kg for production in the UK and 63-75 kg CO_2 eq./kg for production in the United States (or approximately 38% greater than the UK and about half of the results obtained for Australia or China).

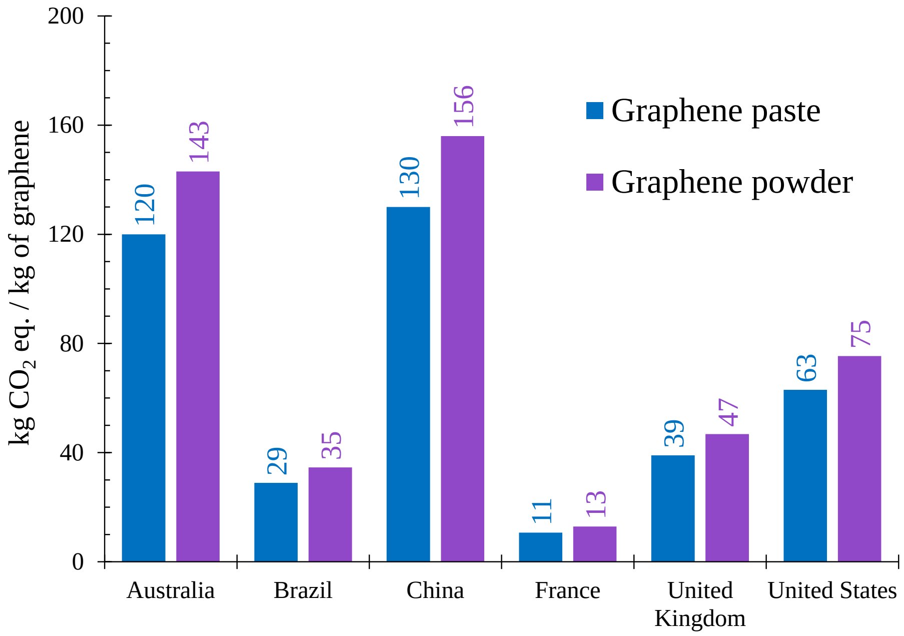

Figure 6 - Global Warming Potential (GWP, in kg CO_2 eq./kg of graphene) considering the electricity mix of different countries (see Table 5 in section 2.3.3.1). The results are for system boundary "cradle to gate" (stages G1, G2 and G3 only, see Figure 2). For the results of other impact categories for Gr_{powder} consult Table S2 in SI section SI-3.

3.2.2. Resources

The paste form of graphene has an MRSP of 41g Cu eq./kg, while the powder form has a higher value of 47g Cu eq./kg. The main hotspot contributing to this category is the electricity consumption, from coal-fired power plants making use of large quantities of steel in their infrastructure. In terms of FRSP, the difference between the two product forms favours Gr_{paste}, with a result of 30 kg oil eq./kg, while the Gr_{powder} resulted in 35 kg oil eq./kg. The hotspots in this category are coal and natural gas depletion for power plants used in electricity generation. When it comes to WCP, Gr_{paste} results of 252 L/kg and Gr_{powder} 301 L/kg, mostly from decarbonized water for cooling towers and flue gas scrubbers in power plants.

3.2.3. Ozone, fine particles and radiation

In SODP Gr_{powder} has an impact of 110 mg CFC-11 eq./kg, while Gr_{paste} has a lower impact of 93 mg CFC-11 eq./kg, a difference of 15% in favour of the paste form. Like the other impact categories, the hotspot for SODP is again electricity consumption (N_2O to the stratosphere from emissions from hard coal, natural gas and lignite burning in power plants).

In the categories OFP_{te} and OFP_{hh}, transportation contributes more to these impacts. For Gr_{paste}, the results are similar in these two categories, with total impacts of 250 g and 249 g NO_x eq./kg, respectively. Transportation of Gr_{paste} from Australia to the UK contributes around 6.7% to the total impact (Table 6). On the other hand, Gr_{powder} requires less transportation due to its lower water content, resulting in a smaller contribution of transportation (around 1.2% of the total in these two impact categories) but a greater impact in these two categories (282 g and 281 g NO_x eq./kg respectively), representing an increase of about 12% when compared to graphene paste. This is due to higher electricity consumption during stage G3 (61 kWh and 85 kWh/kg for paste and powder forms respectively – see Table 4). In FPMFP, the contribution of sea freight to the UK is small for Gr_{paste} (less than 4%) and negligible for Gr_{powder} (0.69%), with total impacts of 134 g and 155 g $PM_{2.5}$ eq./kg respectively. Lastly, for IRP the results were 0.31 kBq Co-60 eq./kg for Gr_{paste} and 0.36 kBq Co-60 eq./kg for Gr_{powder}, mainly associated with Uranium milling residues associated with nuclear power generation.

3.2.4. Acidification and eutrophication

Gr_{paste} exhibits a 15% lower impact in TAP compared to Gr_{powder} (0.40 kg SO_2 eq./kg and 0.46 kg SO_2 eq./kg, respectively). The primary contributors to this impact category are air emissions of SO_2 and NO_x to air from hard coal and lignite power plants for electricity generation. In the category of FEP, Gr_{paste} has an impact of 196 g P eq./kg, while Gr_{powder} has a higher impact of 234 g P eq./kg. The hotspot for this impact category is phosphate emission to water during the mining and treatment of lignite, as well as the mining of hard coal spoils used for electricity generation. Similarly, for MEP, the impacts are 11.9 g N eq./kg for Gr_{paste} and 14.2 g N eq./kg for Gr_{powder}. Once again, coal mining for electricity generation is the hotspot contributing the most to this category.

3.2.5. Eco and human toxicity

The results for TETP indicate that Gr_{paste} has an impact of 80 kg 1,4-DB eq./kg, while Gr_{powder} has a higher impact of 92 kg 1,4-DB eq./kg, a 15% difference between the two forms. In this category, there is a small contribution from sea freight during transportation from Australia to the UK, of 4.47% of the total impact for Gr_{paste} and 0.78% for Gr_{powder}. In FETP,

Gr_{paste} has a lower impact with a result of 5.8 kg 1,4-DB eq./kg, while Gr_{powder} has a higher impact of 7.0 kg 1,4-DB eq./kg. This is due to the higher consumption of electricity which results in higher emissions of zinc, copper and nickel from the mining and treatment of lignite and hard coal. In METP, the results show that Gr_{paste} has an impact of 7.9 kg 1,4-DB eq./kg and Gr_{powder} has a higher impact of 9.5 kg 1,4-DB eq./kg (both from zinc emission to water during mining activities). The impacts in the HTP_c and HTP_{nc} are dominated by emissions of zinc and chromium VI to water from fossil fuels mining activities for electricity generation. In HTP_c, the impact for Gr_{paste} is 10.9 kg 1,4-DCB eq./kg, while for Gr_{powder} is 13 kg 1,4-DCB eq./kg. For HTP_{nc} the results are 214 kg and 256 kg 1,4-DCB eq./kg respectively.

3.2.6. Land use

In LUP, the Gr_{paste} is a result of 0.71 m^2a crop eq./kg, while Gr_{powder} of 0.88 m^2a crop eq./kg, representing a difference of over 20% in favour of the former. The impact is nearly entirely from electricity consumption that comes from the occupation of forests and land for mining in Australia.

3.3. Impacts of OPC_{Gr} production

This section discusses the environmental impact of the experiments on the addition of 0.02 wt% of Gr_{powder} to the OPC (i.e., OPC_{Gr}) which results in saving 16.5% of OPC, as explained in section 2.2 and section 2.3.3.1. The results for the 18 life cycle environmental impacts of the OPC and OPC_{Gr} production in the UK when the Gr_{powder} is being produced in Australia, Brazil, China, France, the UK and in the United States are compared in Figure 7 (see SI-3 Table S2 for the life cycle impacts of Gr_{powder} production in these countries).

	OPC	Graphene origin					
		Australia	Brazil	China	France	United Kingdom	United States
GWP [kg CO_2 eq.]	775.2	673.8	655.6	675.9	652.0	657.6	662.4
MRSP [g Cu eq.]	286.1	250.7	249.9	253.4	261.2	255.2	253.9
FRSP [kg oil eq.]	72.05	66.98	62.49	66.16	61.67	63.80	64.48
WCP [L]	620.3	581.0	1,015.7	598.2	614.8	571.2	608.6
SODP [mg CFC-11 eq.]	41.11	54.23	50.25	41.33	38.17	40.20	40.80
OFPte [kg NO_x eq. x0.01]	133.1	116.3	112.7	118.8	112.1	113.0	113.0
OFPhh [kg NO_x eq. x0.01]	132.0	115.4	111.7	117.9	111.2	112.1	112.1
FPMFP [g $PM_{2.5}$ eq.]	315.1	290.8	274.3	304.8	267.9	270.7	296.6
IRP [kBq Co-60 eq. x0.1]	275.3	248.1	253.0	251.6	383.0	298.7	284.8
TAP [kg SO_2 eq. x0.01]	93.03	85.82	80.66	87.11	78.95	79.85	81.04
FEP [g P eq.]	38.24	71.45	33.19	37.31	33.06	33.81	42.06
MEP [g N eq.]	2.835	4.805	2.940	2.741	2.712	2.608	3.087
TETP [kg 1,4-DCB eq.]	162.1	154.3	152.1	157.6	150.5	150.6	149.2
FETP [kg 1,4-DCB eq. x0.01]	274.0	352.0	252.6	273.3	253.1	257.4	276.9
METP [kg 1,4-DCB eq. x0.01]	379.3	484.5	348.4	376.3	348.4	355.7	380.8
HTPc [kg 1,4-DCB eq. x0.1]	48.28	62.73	42.58	49.23	42.58	43.45	47.31
HTPnc [kg 1,4-DCB eq.]	85.77	115.73	75.61	85.23	76.25	77.33	85.13
LUP [m^2a crop eq. x0.1]	65.17	58.29	62.59	60.24	57.88	64.11	59.80

Figure 7 - Life cycle impacts comparison of Ordinary Portland Cement (OPC) with OPC$_{Gr}$ (OPC with Gr$_{powder}$ dosage of 0.02 wt% and being produced in Australia, Brazil, China, France, United Kingdom and the United States) assuming 16.5% improvement in its compressive strength (see details in section 2.2 and in section 2.3.3.1). Legend: red represents the worst and green the best result, with the other shades representing the values in between. Both OPC and OPC$_{Gr}$ are assumed produced in the United Kingdom. The system boundary for Gr$_{powder}$ is "cradle to gate" (stages G1, G2 and G3 only, see Figure 2). For nomenclature on environmental impact categories see Figure 3.

As can be seen, according to the assumptions adopted in this study, the GWP can indeed be decreased by adding Gr_{powder} onto OPC. On average, it can decrease GWP by 14.5% using Gr_{powder} produced in these countries. The greatest reduction is achieved by using Gr_{powder} produced in France (see section 3.2.1.1 and Figure 6), with a reduction of 15.9%. This was followed by Gr_{powder} is produced in Brazil (15.4%) and in the UK (15.2%). To contextualize, taking only the OPC facility evaluated in this study (output of 1,400,000 t/yr) and the result for Gr_{powder} being produced in the UK, the dosage of 0.02 wt% would ultimately result in reducing emissions from OPC production in the UK in 164,963 t CO_2 eq./yr - equivalent to removing 50,914 small petrol cars from the roads assuming 0.27 kg CO_2 eq./km and travelling around 12,000 km/yr each (Huijbregts, 2016; Wernet et al., 2016). For the Gr_{powder} produced in Australia however, this reduction is smaller, of 142,172 t CO_2 eq./yr.

When considering the 18 environmental impact categories simultaneously, the results require a more complex analysis. This is because in some categories OPC_{Gr} have either equivalent and, in some cases, even greater impacts than the OPC. This can be seen in the spider chart in Figure 8 depicting a visual comparison between OPC and the OPC_{Gr} when Gr_{powder} is being produced in different countries (see Table 5) - it does not include Gr_{powder} transportation to the OPC production facility in the UK (which does not significantly impair interpretation due to its low contribution to impacts, see Table 6). As can be seen, if the Gr_{powder} is produced in Australia, the results for SOD, FEP, MEP, FETP, METP and human toxicities would significantly increase when compared to the other alternatives. Even though showing a reduction in the other eleven impact categories (including GWP), if considering the 18 impact categories together the Australian Gr_{powder} would increase the life cycle impacts of the OPC_{Gr} by 11.4% on average when compared to the OPC. The UK and French-produced Gr_{powder} are the ones showing the best overall results, with a significant reduction in most environmental impacts (8.8% and 8.0% on average, respectively) – both only increasing IRP.

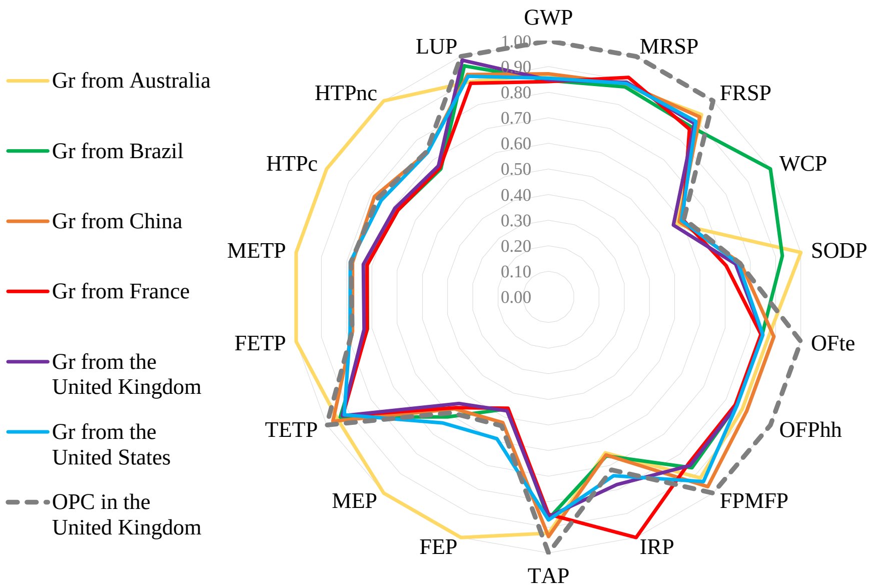

Figure 8 - Spider chart comparing the life cycle impacts of Ordinary Portland Cement (OPC) with OPC_{Gr} (OPC with Gr_{powder} dosage of 0.02 wt% and being produced in Australia, Brazil, China, France, United Kingdom and the United States) according to improvements in its compressive strength (see details in section 2.2 and in section 2.3.3.1). Both OPC and OPC_{Gr} are assumed produced in the United Kingdom. The system boundary for Gr_{powder} is "cradle to gate" (stages G1, G2 and G3 only, see Figure 2). For nomenclature on environmental impact categories see Figure 3.

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The intermediate results stem from Gr_{powder} produced in Brazil, China and the United States, leading to higher results in only a few categories (see SI-3 Table S3). The results suggest the following potential average reduction in life cycle impacts of OPC_{Gr} when Gr_{powder} is produced in: Brazil 4.4% (but with increases in WDP, SODP and MEP); China 5.1% (but with increases in SODP and HTP_c); United States 4.6% (but with increases in IRP, FEP, MEP and FETP). Considering the low significance of Gr_{powder} transportation (see section 3.2), these results appear promising for the widespread and global adoption of Gr_{powder} in OPC production.

4. Discussion

As mentioned in the introduction, cement production is a major contributor to global GHG emissions. To mitigate this problem and address related challenges, several methods have been identified to improve the environmental sustainability of cement production, evaluated through LCA methodology (Dahanni et al., 2024; Georgiades et al., 2023). According to the descriptions provided in section 3.1, shifting to alternative greener fuel combustion for calcination and clinkerisation like oxy-fuel, biomass and waste-derived fuels can reduce GHG emissions considerably (and possibly further reduce impacts from hard coal production like FRSP, FEP, MEP and HTC), and can be achieved by incorporating new concepts and designs to the processes (Hanein et al., 2020; Schneider et al., 2011). More recently, and as demonstrated in the present study, the use of nanomaterials (e.g., graphene, graphene oxide, graphene nanoplatelets and carbon nanotubes) has shown to be capable of promoting more sustainable cement and concrete productions (Long et al., 2018; Papanikolaou et al., 2019).

Related to this, our experiments for OPC_{Gr} did not include the use of paste graphene. Consequently, we did not assess the environmental impacts of OPC_{Gr} utilizing this form of graphene. However, there is literature available on the dispersion of Gr within the cement-graphene matrix (Dung et al., 2023; Lin and Du, 2020; Wang and Zhong, 2022). This presents an intriguing area for future research, as achieving equivalent or superior results in cement properties with paste graphene could potentially yield even better environmental impact outcomes for OPC_{Gr} (see Figure 5 and Figure 6). Exploring this avenue could significantly enhance our understanding and application of Gr in construction materials, driving further advancements in sustainability and performance. The parameters adopted for the fluidisation experiments in this work (especially energy consumption, geometry of fluidised vessel, residence time, and orientation of the mixing air from the low-energy blowers) are crucial when designing industry-scale homogenisers for cement and Gr_{powder} where there might be requirements to handle tonnes of bulk solids per production batch (Wei et al., 2024; Yao et al., 2022).

The main driver of the life cycle environmental impacts of graphene production via electrochemical exfoliation is electricity consumption (see section 3.2). The GWP of its production is estimated at 121-143 kg CO_2 eq./kg when done in Australia, and the sensitivity analysis revealed that this impact can be decreased to 11-35 kg CO_2 eq./kg if in Brazil or France. These results are aligned with previous findings in the relevant literature. For instance, the values estimated by Cossutta et al. (2017) are in the 57-332 kg CO_2 eq./kg range with more than 80% coming from electricity consumption. This indicates that utilizing hydro and nuclear power sources for electricity generation represents a straightforward approach to reducing the environmental impacts of Gr production by electrochemical exfoliation. In terms of electricity consumption, this process currently requires 60-147 kWh/kg, in falls within the minimum found for other Gr production methods evaluated in Munuera et al. (2022) - which ranged from 58-522 kWh/kg. This large variation can be explained by some of the methods being at the initial stages of development (e.g., experimental designs) and relying more heavily on the use of chemical reagents (e.g., Hummer's method) (Arvidsson, 2017; Arvidsson et al., 2014; Arvidsson and Molander, 2017). Furthermore, advancements in graphene production methods are ongoing, with forecasts indicating potential reductions in power consumption through process optimization and the realization of economies of scale.

Cement and concrete, as primary and highly consumed construction materials, have many significant life cycle environmental impacts. The implementation of circular economy (CE) strategies has great potential to foment the reduction of impacts coming from this important industry. One approach involves the reduction of natural resources during production and the use of these materials by the construction industry (both topics covered in the present study). Further strategies to extend the lifespan of concrete structures through thoughtful design and effective maintenance practices emerge as other important aspects (Marsh et al., 2022; Norouzi et al., 2021). In this regard, the adoption of digital technologies for embracing CE approaches can accelerate the transition towards a more sustainable construction sector (Çetin et al., 2021; Wangler et al., 2019). As an example, deep learning techniques offer a valuable approach to simultaneously consider the physicochemical information and mechanical properties during cement and concrete production. This enables the strategic exploration of chemical reactions involved in the calcination and clinkerisation processes, thereby facilitating the optimization of energy performance, better material utilization and the use of different inputs in the process (Hanein et al., 2020; Mahjoubi et al., 2023).

5. Conclusion

Cement production constitutes a substantial source of global CO_2 emissions, necessitating immediate action to mitigate its adverse effects on climate change and other environmental consequences. This study focuses on performing an updated and robust life cycle assessment (LCA) of Ordinary Portland Cement (OPC) and Graphene (Gr) productions - and the combination of both (OPC_{Gr}). The data for OPC production is derived from one of the largest cement production facilities in the United Kingdom (UK), while the data for Gr production is obtained from an electrochemical exfoliation plant located in Western Australia. To evaluate the potential benefits of incorporating Gr in powder form (Gr_{powder}) into OPC, laboratory experiments were conducted to examine the effect of its addition on the compressive strength of OPC at a dosage of 0.02 wt%.

The LCA results indicate that OPC production in the UK generates 775 kg CO_2 eq./t, with geogenic CO_2 emissions during calcination and clinkerisation being the bulk contributor (57%). This particular stage alone is responsible for more than two-thirds of most of the other environmental impacts, except mineral resource scarcity, stratospheric ozone depletion, ionizing radiation, and terrestrial ecotoxicity potentials. The onsite release of NO_x and SO_2 is the primary factor contributing to impacts related to ozone formation, terrestrial acidification, and the formation of fine particles. Electricity consumption plays a crucial role in stratospheric ozone depletion, ionizing radiation, and land use potentials, besides making a significant contribution to eco and human toxicities. The extraction and preparation of hard coal contribute to over 40% of the impacts in six categories. Concerning Gr, its production in paste and powder forms in Australia results in 121 kg and 143 kg CO_2 eq./kg, respectively. Electricity consumption is mainly responsible for the environmental impacts, and a sensitivity analysis indicated that the use of hydro and nuclear sources can easily reduce the results in this category by over 50%.

Experiments on the incorporation of Gr_{powder} into OPC showed an increase in the compressive strength of the resulting OPC_{Gr} by 16.5%. By assuming that this increase in mechanical property would result in consuming a proportionally lower quantity of OPC_{Gr} for the same purpose, estimations of how this would impact the 18 environmental impacts compared to OPC production in the UK with Gr_{powder} produced in different countries were conducted. Even though the global warming potential decreases for all cases (in 12.8-15.9%), it can result in increased impact in some categories. Particularly in freshwater and marine eutrophication potentials, the impacts increase by 1.09-1.87x using the Australian and United States-produced Gr_{powder}. The OPC_{Gr} with UK and French-produced Gr_{powder} demonstrated the best overall results, with average reductions in environmental impacts of 8.8% and 8.0% respectively - increasing only ionizing radiation potential. When Gr_{powder} is produced in Brazil, China and the United States, the average reductions in OPC_{Gr} impacts amount to 4.4%, 5.1% and 4.6%, respectively. Consequently, this indicates that utilizing cleaner energy sources in graphene production can significantly contribute to reducing the environmental impacts of OPC_{Gr} production.

In conclusion, the use of graphene in the construction industry offers significant opportunities for enhancing material properties and sustainability. Our experiments using graphene powder show it is capable of improving cement's mechanical performance. However, this approach also presents challenges, such as ensuring consistent quality and managing the cost and scalability of production. Given the superior environmental results of the paste form, further experiments and estimates should prioritize understanding the dispersion of this form and its impact on cement strength. Additionally, our estimates face limitations in understanding the impacts of graphene production and applications, including the rapid decarbonization of electricity grids and the varying amounts of graphene needed to achieve optimal cement strength for different building uses. These factors must be considered in future research to fully understand and maximize the benefits of graphene-enhanced construction materials.

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