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Review of concrete solutions for concrete's environmental impacts

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Abstract
The use of concrete is under scrutiny as it appears as one of the few human activities where the transition toward a post-carbon society is not possible unless large investments in risky carbon capture and storage are made. With current urbanization, it is also a sector that is expected to continuously grow, leading to increased resource consumption and emissions. In this review, we aim to shed light on the available solutions that can be implemented in the short and long term to reduce greenhouse gas emissions. Rather than waiting for disruptive technologies that could transform a very slow moving and risk-averse construction sector, this review focuses on the small improvements that every stakeholder involved along the value chain of concrete production and use can achieve. We stress how significant the combined effect of these marginal gains can be. By balancing societal needs, environmental requirements, and technical feasibility, the intention of this review is to show credible pathways for a transition to sustainable use of concrete.

Key points

• Cement usage is so massive, more than 4 billion tonnes per year worldwide, that large-scale replacement by other materials within the next decade is not possible.

• Environmental impact of cement and concrete is low per unit of material, but the amount used makes the impact of the sector highly significant.

• Reductions in CO_2 emissions are possible through successive improvement all along the cement and concrete value chain: less clinker in cement, less cement in concrete, less concrete in structures, and less replacement of structures.

• By engaging all stakeholders of the construction sector, immediate savings of the order of 50% can be reached without heavy investment in new industrial infrastructure or modification of standards.

• Research and development need urgently to be conducted for post-2050 construction to meet future emissions reduction targets. Alternative cement and faster carbonation of concrete should be explored.

1 Introduction

Concrete is the fundamental building block of our urbanizing world. It makes up the buildings and infrastructure that enable businesses to operate and people to carry out their daily activities. Over the past centuries, concrete has laid the foundation of the industrialized society [1]. Infrastructure, such as transportation, electric power systems, water and wastewater systems, buildings from single-floor houses to high-rise buildings – even those with steel or timber frames - all rely on concrete.

Concrete is a synthetic rock made of cement, sand, gravel and water, and is by far the most used man-made material. Cement, which is the mineral glue that sticks together sand and gravel in the concrete, represents around 10% of concrete mass and is currently produced at around 4 Gt/year, almost the same amount as food [2]. Over the past 65 years, its consumption increased ten-fold [3]. In comparison steel production has been increased by a factor 3 and timber construction stayed nearly constant [3]. Among materials used for construction, cement accounted for 36% of the 7.7 GtCO_2 released globally in 2010 by construction activities [4], while steel accounts for 25% [5], plastics 8% [4], aluminum less than 4%, [6] and brick less than 1% [7,8].

Concrete accumulates in the Earth's crust and is now considered to be one of the markers of the Anthropocene [9] with an estimated 900 Gt added since the beginning of the industrial revolution [9,10]. But it is important to remember than only about half of cement is used for concrete [11], the rest being used for blocks, mortars, and plasters. To grasp what volume these masses represent, one should picture every person on Earth building every year the equivalent of a 20 cm thick concrete wall of 4.5 m^2 area, as well as plastering a wall surface of 35 m^2 with a 3 cm thick cement-based plaster (considering 300 kg cement/m^3 of concrete and 250 kg/m^3 for cement plaster).

By 2050, urbanization is expected to add 2.5 billion people to the global urban population, mainly in Asia and Africa [12]. Together with the pressure to fill the already sizable housing deficit and lack of reliably functioning infrastructure, it is anticipated that this population growth will cause a surge in demand for building materials, including concrete. After 2050, one can expect a reduction of construction demand in most regions of the world[13] due to the achievement of urban transition and the stabilization of the population [13].

It is therefore crucial to act now and drastically reduce the environmental impact of construction within the next decades during this urbanization peak. The new buildings are expected to consume less energy during their operation, which should increase the focus on emissions related to concrete [14]. Actually, for a new typical masonry multifamily building type, steel-reinforced concrete represents 50% of the CO_2 emissions attributed to the building, followed by windows, insulation, ceramic tiles, and paint[15] **(figure 1).** Greenhouse gas (GHG) emissions [17], local scarcity of non-renewable resources [18], energy consumption [11], water use [19], dust and particulate matter emissions [20], mercury emissions [21] are known issues related with cement and concrete production. But no other known material has been found to provide the same amount of service than reinforced concrete at such a low economic cost [11]. Considering the tremendous volume used, unique properties and simplicity of use, its replacement seems not to be feasible in a decade [16]. As a result, it is vital to develop solutions to mitigate the environmental impacts of concrete production, while maintaining the favorable properties of concrete and in the face of increasing global demand.

In this paper, we first review the different environmental impacts of cement and concrete production, use, and disposal. We then look at potential routes for improvement pointing out what can be implemented within the next decade and what needs to be considered in a longer term. This leads to policy and stakeholder actions that could pave the way toward a decarbonized construction sector.

2 Cement and concrete environmental impact

Environmental impacts related to the production of a material used in such vast amounts are inevitable. Issues related to resource depletion and global change attract a large attention[22], but other issues related to local health aspects have also recently been pointed out [23]. In this section we will step through the different environmental issues related to cement and concrete production, and show that for most of them implementation of stringent and effective regulation would solve most of the problems, except for climate change, where technological breakthroughs are needed.

2.1 Cement and concrete production

Portland cement is composed of four major oxides: CaO, SiO_2, Al_2O_3 and Fe_2O_3 coming from raw materials, usually limestone, clay, and small amounts of "corrective" materials such as iron ore, bauxite, and sand to reach the desired chemical composition. Raw materials are crushed, mixed and milled into a raw meal, which is then heated in the pre-heating system to dissociate carbonate into calcium oxide and carbon dioxide **(Figure 2)**. The meal is then calcined in a rotary kiln at up to 1500°C where reactions between calcium oxide and other elements produce calcium silicates and aluminates [24–26]. The melted material is then cooled rapidly to form an assemblage of C_3S, C_2S, C_3A and C_4AF, called clinker. This clinker is inter-ground with gypsum to a finer product called cement. Concrete is produced by mixing cement with sand, gravel (or crushed stone), water, and chemical admixtures. It is produced in a concrete plant and transported by concrete truck to the construction site, or directly mixed at the construction site. Concrete is also used to produce precast elements. Finally, cement can be used in plaster and mortar when mixed with water, sand, lime and chemical admixtures, both on site and in premix mortar factories.

2.2 Local health issues

GHG emissions from cement production have grown to prominence in environmental sustainability discussions. However, based on the economic valuation of damages caused by air pollutant emissions and GHG emissions, respectively, recent studies have indicated that the economic burden of resulting health damages could rival the climate damages from cement production [23], in particular when considering particulate matter.

The inhalation of small particles, $PM_{2.5}$ and PM_{10}, is recognized to have significant consequences on human health - notably linked to respiratory infection, pulmonary disease, lung cancer, heart attacks, among other diseases [27]. In the production of cement and cement-based materials, the emissions of PM come from various processes, primarily from material-derived particulates from acquisition, storage, and handling [23]. Additionally, secondary PM formed from nitrogen oxide and sulfur oxide emissions associated with the high thermal energy demand and fuels used in cement kilns can further contribute to health burdens[23].

Currently, appropriate filtering and capture of PM can mitigate many of these emissions from cement production. Modern electrostatic precipitators and baghouses significantly reduce PM emissions [28]. For example, cement kiln dust can be captured to a great extent and reused in the production of more clinker if it has appropriate alkali content, thus reducing emissions [29]. Further, use of scrubbers or alterations in energy mixes can drive down both PM and other air emissions. Yet regulations requiring use of such technologies vary by region. For instance, a recent study in Zambia showed that $PM_{2.5}$ concentrations were still 5 to 10 times higher within the vicinity of the cement plant than 1 km away and respiratory symptoms were 3 times higher [30].

The emissions of PM from cement and concrete production are clearly areas in which appropriate policy can drive down undesired environmental burdens [17]. Aggregate production, ready mix operation, construction and demolition generate additional dust which are more difficult to control.

2.3 Regional resource scarcity

To supply the equivalent of 4.5 m^2 of typical concrete wall and 35 m^2 of cement plaster per capita and per year, more than one ton of gravel, 2.5 tons of sand and 550 kg of cement are required. At the global level, this demand translates to the direct use of 5.4 Gt of limestone for clinker and filler production and 17.5 Gt of aggregate, to produce annually approximately 10 billion m^3 of concrete [31].

This tremendous demand for resources has led to a growing concern about potential contributions to regional resource scarcity for concrete production [32]. In the United States from 1900 to 2000, a period of significant infrastructure development in the nation, the demand for crushed stone, sand, and gravel grew from being ~60% greater to being ~290% times greater than the sum of all other raw material flows used by human activities in that same time period [33]. Although sand and gravel are widely abundant on Earth [34], they are usually not transported over long distances due to the economic cost of transporting such heavy materials[35,36]. Therefore, a construction boom increases the local pressure for natural sand and coarse aggregates close to urban areas [18]. But as quarries and sand mines become undesirable due to NIMBYism or too expensive to operate close to urban areas, transportation distances will increase. For example, aggregates for San Francisco Bay Area (California) come from British Columbia (Canada).

Uncontrolled aggregate extraction damages ecosystems, thus biodiversity, and has potential cascading affects that impact human wellbeing [37–39]. But these environmental damages are primarily a result of poor resource management [40] as natural sand extraction usually does not require complex operations and can be carried on as informal activity near large cities [41]. Thus, quantification of sustainable resource extraction possibilities around cities should focus on additional factors other than local availability, such as land use changes [42], resource accessibility [43] and consideration of political and economic actions [44]. While local pressures on resources have been noted, it is possible to reduce the impact of concrete production on ecosystems by using secondary or non-depleted bulk local resources. An untapped resource in urban context is the excavation material [45]. Each new construction generates excavation materials usually landfilled outside the city. In Switzerland it represents a similar amount as the primary material required to build [46]. In China, considering 0.9 t excavation per m^2 built [47], this material could currently supply half of the construction material requirement[48]. A better use of this material is possible through washing to extract sand and gravel from it[49] or directly through clay based concrete [50]. Finally, sustainable resource management can be achieved through promotion of strong policy and regulatory frameworks, such as certification systems to secure the good practice along the supply chain (e.g., CSC label [51]).

2.4 Global environmental issues

One of the most commonly discussed environmental impacts related to cement production is the high level of GHG emissions. Among materials used for construction, cement accounts for 36% of emissions related to construction activities[4] and 8% of total anthropogenic CO_2 emissions [52]. At least 70% of the GHG emissions from concrete production is due to cement production **(Figure 1)** [31]. Unlike regional resource scarcity and local health issues, for which in many cases there are mitigation strategies that can be implemented today, technological breakthroughs are needed globally to meet GHG emissions mitigation goals.

Within cement manufacturing, the predominant source of emissions is the kilning stage; grinding, sorting of raw materials, and packaging of cement bags have minor impacts [17,53]. Most of the emissions are from the decomposition of limestone (calcination) and associated with energy use [54]. Energy-derived emissions can be notably reduced through kiln efficiency and choosing lower-carbon fuels [55]. Current average cement production emits approximately 0.31 kg of CO_2 per kg of cement from energy-resource combustion [56]. Calcination represents around two-thirds of the total GHG emissions from cement produced using a state of the art dry process rotary kiln equipped with pre-calciner [57]. These are the main reasons why cement production is considered to be difficult to decarbonize [16] as decarbonization of the energy supply will not eliminate the material-related CO_2 emissions from calcination[16]. However, there have been many mitigation strategies for these emissions, several of which are discussed in the subsequent sections.

As a consequence of the annual demand for 4.5 m^2 of concrete and 35 m^2 of plaster for every inhabitant on the planet, there is an associated ~300 kg CO_2 of cement-related emissions per capita. For context, this would be approximately 3 times less than a return flight between London and New York city. While concrete's societal benefits are undeniably very important, GHG emissions are a real problem for the cement and concrete industries - and for the World.

3 Credible medium-term solutions to reduce cement demand

The peak in new construction will come in the next few decades, so it is most important to focus on reductions in environmental impact that can be achieved in the medium term, before 2050. The substantial reduction needed can only be achieved by considering efficiency at all stages of the value chain: clinker production, cement production, cement use in concrete or mortar, concrete use in construction, design of structures and use of structures. It is essential that all parts of this chain are considered as there is no point saving, for example, 30% CO_2 in cement production and then using twice as much cement as needed in the concrete. In this section, we explore the potential for further reductions in environmental impacts along this value chain **(Table 1)**. These solutions are of particular importance because they can often be implemented without new production technologies or infrastructure.

3.1 Efficiency of clinker production

As previously described the majority of CO_2 emissions in concrete, come from clinker production. Driven by the huge increase in energy costs associated with the oil crisis of the 1970s, there has been considerable progress in the energy efficiency of clinker production. Globally over 85% of cement kilns use energy-efficient dry methods, which do not need additional energy to evaporate water [58,59]. There has also been substantial progress in heat recovery and recycling **(Figure 2)**. State-of-the-art kilns achieve about 63% efficiency and through integrated approaches could reach 80% efficiency [60]. Such levels of efficiency make modern kilns among today's most efficient thermal machine in wide-scale industrial use. There is therefore limited scope for further improvement [55].

Modern cement kilns are also extremely flexible in terms of fuel source and many plants in Europe use various waste streams for more than 80% of the energy demand. In 2013 in Europe, around 1.3 million tyres (50% of total recycled tyres) were used as fuels for clinker production. Already in the 1990s in the United States, about 70% of all hazardous wastes were burnt in cement kilns. Waste materials derived from fossil fuels such as solvent, plastics, tyres are not regarded as carbon neutral. However, it is important to note that transferring waste fuels from incineration plants to cement kilns results in a significant net CO_2 reduction because cement kilns are more efficient [61]. Another advantage is that no toxic residues such as dioxins are generated since the ashes are completely incorporated in clinker[62]. The International Energy Agency Roadmap expected the worldwide use of "alternative fuels" to grow from 3% in 2006 to about 37% in 2050 and deliver around 15% of the targeted overall reduction in CO_2 emissions [55,63].

This increase in fuel efficiency means that fuel now accounts for only about one-third of the CO_2 emissions from clinker production. It is much more difficult to reduce the other two-thirds coming from the decomposition of limestone, which is related directly to the chemical composition of the clinker, namely the content of calcium oxide.

Given the difficulty of producing materials with substantially lower contents of CaO, it could be considered if there are sources of CaO other than limestone ($CaCO_3$). Unfortunately, practical sources of non-carbonate calcium are quite limited. The fine material left from crushing concrete for recycling aggregate is one potential source, which is just starting to be exploited. But in countries where most of construction will occur, the volume of new construction will far outstrip the volume of demolition.

3.2 Efficiency of cement production

By far the most promising route to large scale reduction in GHG emissions comes from substituting in the cement, as much clinker as possible by other materials, collectively known as SCMs (supplementary cementitious materials). This is a strategy already widely adopted. The most widely used SCM is fine limestone – the same as the raw material used to produce clinker, but as this is just ground, rather than being heated to high temperature it does not lose its CO_2 and has very low associated emissions [64]. Although this material is widely available, it has very limited reactivity and at levels of substitution above around 10-15% it is simply a filler [65].

The next two most used SCMs are fly ash and blast furnace slags, which are respectively by-products from coal power plants and iron industry [66]. While very valuable in decreasing environmental impact today [67], they amount to only 15% of current cement production and almost all sources are already used either in cement or later added to concrete. Furthermore, this amount is likely to decrease in the future, as we move away from using coal and more steel is recycled.

However, there is great potential for large scale CO_2 reductions through more extensive use of clays, which are very widely available worldwide and which when calcined (heated) to relatively modest temperatures can give a highly reactive SCM [68,69]. The substitution of clinker by a combination of calcined clay and limestone gives cements (so-called LC3) with good levels of performance, even at high substitution levels [64,70,71]. If clinker substitution is not limited by the availability of SCMs, as is the case for using calcined clays it can be estimated that overall CO_2 savings of 15-30% of current levels from cement, can be achieved worldwide.

3.3 Concrete efficiency

There is considerable scope to reduce CO_2 emissions by a more efficient use of cement in concrete through better mixture design. Studies show that for the same performance we can have a factor 3 of variation in cement content per cubic meter [72,73]. This variation is the result of different production technologies and lack of knowledge. Mixture proportions can be selected to meet necessary properties while reducing GHG emissions [74]. In general, manual mixing on site from cement in bags is the most inefficient. More efficient mixing in a concrete truck, or better still a ready mix plant, can reduce the cement content for the same properties by a factor of 2 [75]. The use of a proper mixture design in a concrete plant, with appropriate proportion of sand, gravel can lead to further reductions (up to 50%) without loss of strength or fluidity [76]. Such improvements are an untapped potential in emerging countries where most of the cement is sold in bags and used without proper technical control or mix design optimization [77]. Promoting industrialized concrete production as a replacement for site-mixing, especially in self-help housing schemes is a very effective way to reduce cement consumption in both concrete and mortar applications[78].

Concrete efficiency can be taken even further by engineering in such way that up to 60% of the cement can be replaced by fillers – simple ground material – in combination with dispersant admixture. This is an emerging technology that has been shown to be feasible in precast and ready-mix concretes in Germany [79] and Brazil [80]. It is also feasible in the dry set rendering mortar market [81]. The technology requires adequate supply chain of fillers and efficient dispersants, advanced knowledge and technical capability. Limitations are the cost of the dispersant admixture and existing concrete standards.

3.4 Construction efficiency

Waste on construction sites represents a largely underestimated amount of material. A large national study performed in Brazil showed wastage levels as high 50 to 100% [82]. The findings of this study are especially important and highlighted that waste rates were much higher for the use of cement sold in bags than for ready-mix. This issue is especially relevant in emerging countries, where the largest growth in concrete demand is expected and where quality control on construction site may be lower. Better design and site management practices were found to be important. Further, decisions taken during construction phase, such as the curing period before demolding concrete, can cause notable changes in the quantity of cement needed for concrete production [31]. Better control on water and aggregate humidity on construction site can also have a critical influence [83]. Since waste cost to builders, raising awareness was efficient on Brazilian market[82]. Education of construction workers is also a proven strategy [84].

3.5 Design efficiency

Research has shown that buildings use structural material inefficiently[82]. In structural systems, GHG reduction is complicated by the interplay of concrete performance (and hence mixture proportions) and the quantity of steel reinforcement, which are often highly constrained by codes. For reinforced concrete columns, an increase in concrete compressive strength typically leads to a reduction in GHG emissions, while for reinforced concrete beams, achieving same strength but with lower clinker is the target [85]. These combined optimization strategies of concrete strength, rebar content and clinker content can provide around 20% reductions in GHG emissions [85]. Similar reduction can be achieved for structures where the dead load is the key design parameter through the use of high performance concrete [86]. Orr and co-authors demonstrate that more efficient utilization of structural concrete had the potential to achieve material savings up to 30–40% through design optimization [87]. Although the magnitude of such savings is difficult to quantify, the works of De Wolf [88], Shank and co-authors [89] would also argue for 10%-20% reduction within conventional design constraints. Finally, savings can also be achieved by increasing the time to functional obsolescence of structure and avoiding the need for a structure to be demolished and rebuilt [90]. This is of particular importance for the existing infrastructure in Northern countries which have been mainly built in the period 1960-1980, with a planned service life of 50 years. Innovative solutions with ultra high performance concrete allow extension of the service life of infrastructure with less than 50% the GHG emissions required for conventional rehabilitation, and a fraction of what it would cost to rebuild them[91,92].

3.6 Reduction of GHG emissions all along the value chain

It is clear that working on marginal gains all through the value chain can lead to substantial savings in GHG emissions (**Table 2**). The savings are not necessarily additive and may not be appropriate in all applications, but Shanks and co-authors show that around 50% of clinker production, in the UK, could be reduced through combined application of existing technologies [89]. The substitution of cement with calcined clay and limestone has the biggest potential to reduce GHG emissions. Reducing the amount of cement in concrete has the next highest potential, followed by floor slab optimization through prefabrication and post-tensioning **(Table 2)**. The difficulty to implement these savings comes mainly from the fact that the construction sector is a fragmented industry with multiple stakeholders [93]. Outside the cement industry, which concentrates investment and production capacity, the other stakeholders from waste management companies to concrete producers or engineering office are often decentralized entities, relying on multiple independent offices [94] **(Table 2)**. Without strong enforcement policy implemented with a top down approach and efforts to integrate the value chain, the transformation of the construction sector will take time [95,96].

4 Necessary long-term development towards zero carbon concrete

After 2050, global society will continue to require infrastructure elements that can only realistically be constructed from concrete. Combining this with the need to move the sector to carbon neutrality, and considering the opportunities opened by a longer research and development timescale to demonstrate in-service performance of radically new material types and design strategy, there is significant interest in looking beyond established practices to investigate wholly different ways of producing and using concretes. This section explores the most promising options.

4.1 Breakthrough solutions, the reality behind the hype.

Abundant technical literature exists regarding possible disruptive technologies as alternative to cement production[97,98]. According to various authors, such technologies can play an essential role in the future of the construction sector by replacing cement in part or in full[99]. Several alternative cements have been shown to be able to contribute to reduced environmental impacts relative to conventional cements[100,101]. However, the pace of change in the construction industry, issues in materials availability or cost, and the technical limitations of some of these alternative technologies, mean that many proposed material alternatives are unrealistic from technical or resource standpoints and are unlikely to reach large-scale technical maturity before 2050 where a transition to net zero emissions is required. It is actually difficult for alternative cements to meet more than 5% of the projected future demand for cementitious materials [102,103].

Even though sufficiently mature alternative cements are already in use at commercial scale in many parts of the World, the production capacity expansion is limited. For example, calcium sulfo-aluminate (CSA) cements are well-known products, largely used in China. This technology is a real alternative compared to Portland cement as it is based on aluminum chemistry avoiding the decarbonation of limestone [104]. The main issue is lack of high-alumina raw materials, which limits its implementation to a few percent of cement production at most. Let's imagine that even if all current bauxite production was diverted from the production of aluminum it would not be sufficient to provide more than 10-15% of the current demand for cement.

But other alternatives could be able to be scaled up in the next 20 years. Alkali-activated cements have been discussed at some length as a potential alternative to Portland cement in many large-scale applications[103]. In regions where the supply of both suitable activators and precursors is plentiful, they have been shown to be economically and technically viable in precast and ready-mixed formats [105]. However, there remain supply-chain challenges related to availability of highly effective alkaline activators such as sodium silicate, which are not currently produced at sufficient scale to replace even a fraction of a percent of global Portland cement production. Work based on alkali-activation using more widely available salts such as sodium carbonate does show high potential for scalability of production[106] but is still in competition for the supply of aluminosilicate precursors, as the precursors are also used as SCMs in Portland cement-based concretes and already facing limited availability. Nonetheless, alkali-activated concretes have the capacity to integrate in their manufacture high alkali-content solid wastes which cannot normally be recovered[97,107].

One of the main challenges in the area of alkali-activated concretes, and other technologies based on industrial wastes, relates to the scale on which waste are needed to become a realistic input into large-scale construction. Waste which is generated at a rate of tens of tons per annum may be a major disposal challenge for many industries, but this is a scale which is far too small to be worth even considering for use in commercial-scale construction, unless the material has very specific technical characteristics that can improve performance of cementitious or concrete materials. Among the promising wastes available at the scales needed for realistic use in concretes are those which result from mining operations, biomass combustion, metallurgical recycling and/or modernized extractive metallurgy, and construction and demolition waste [108]. They all share the characteristic that they can be to some extent quality-controlled, which is essential to achieve the necessary consistency of construction products.

Magnesium-based cements can be produced based on magnesium carbonates or oxides, replacing limestone and using various alternatives to the conventional clinkerization process [109]. They can have a sustainability advantage if magnesium carbonate is obtained through carbonation of geologically-sourced magnesium silicate by uptake of CO_2 that would otherwise be emitted to the atmosphere [110]. However, past attempts to develop a scalable process have not succeeded, and the likely very high capital expenditure requirement makes implementation challenging even considering a 30-50 year perspective [111]. Furthermore, even if low-energy scalable processes become available for exploiting magnesium silicates, the availability of these materials is much more localized than limestone used to produce Portland cements, entailing significant transport costs if cements based on magnesium silicates are to be used on a global rather than local scale [112]. Moreover, magnesium silicates are less available near the Earth's surface, so deep mining operations would be required to recover the amounts needed to meet the demand for construction. Magnesium recovery from brines for use in cements has also been proposed [113], but is also probably geographically limited to regions in which large-scale seawater desalination is taking place or where salt lakes are accessible.

Many other suggested solutions are based on the idea of cement setting and hardening through carbonation of calcium oxide. This allows to capture the CO_2 emitted during cement production and to tend toward carbon-neutral cement. The problem is to find sources of calcium oxide that do not come from the decarbonation of limestone in the first place. If CaO is derived from limestone, then there can be no net gain as the CO_2 which can be reabsorbed can never be higher than the CO_2 emitted in the decarbonation step, and there would still be additional impacts from required process energy. Development of "Carbonatable Calcium Silicate Cement" (CCSC) technology has been developed thanks to recent development to accelerate and control carbonation industrially without excessive energy consumption[114]. Simple calcium silicate minerals such as wollastonite can carbonate very rapidly in relatively pure CO_2 gas (e.g., Solidia Cement[115]). These binders are well-suited for the fabrication of thin precast products, to allow CO_2 and water transfer during curing. They also involve some capital costs and non-negligible operating costs as well [102]. Finally, wollastonite is, as discussed above for magnesium silicate, not a well distributed resource in the Earth's crust and can thus only be a solution for some specific locations. As the current global wollastonite production amount is 500 000 tons per year, mainly in China [116], a transition towards this technology would require wollastonite extraction to increase by a factor of around 10 000.

There has been much focus on concrete made with alternative cements for over half a century, but the availability of raw materials, the confidence in long term performance, or the limitation to specific application in well-controlled environments make it unrealistic to consider any of these alternative as a direct one-for-one replacement for conventional cementitious materials within the next decades. However, when considering a more local context, and if it is proposed to move away from the idea that all locations in the World can (or even should) be using the same type of cement for all applications, there is a great deal that can be achieved by the production of fit-for-purpose local cement technologies and solutions specific to the areas where the desired resources do exist. The most critical issue here is cost; these alternative cements must be made scalable and cost-competitive, but can only occur at a local level.

4.2 Carbonation of cement and concrete

For classic Portland cement, further CO_2 savings can be achieved in the use phase and at end of life. Actually, when exposed to the atmosphere, cementitious materials can capture CO_2 through carbonation. The amount taken up is some fraction of the one released by limestone decomposition (calcination) during cement production.

Carbonation involves the calcium-containing phases from cement such as calcium-silicate-hydrates, calcium-aluminate-hydrates as well as portlandite ($Ca(OH)_2$) reacting with CO_2 to produce mainly calcium carbonate and other non-carbonated phases [117]. The reaction starts on the exposed surface and proceeds by CO_2 diffusing slowly inwards. This reaction has been extensively studied by engineers because, by reducing the pH of the concrete pore water below pH ~9.4, it may damage the electrochemical protection of mild steel reinforcement bars against corrosion [118] [123], which is deleterious for the durability of concrete structures exposed to high relative humidity or rain.

Carbonation depth is commonly described as a diffusion-limited process: $depth=k.t^{0.5}$ (where t is time and k a constant). The value of k for real concrete structures usually varies between 2 and 15 $mm/yr^{0.5}$ [119], meaning that a 200 mm thick concrete column can take 44 to 2500 years to reach pH 9.4. No systematic information exists on carbonation of other products such as mortars and renders, except for a mention [120] of unpublished results with k ranging from 6.1 to 36.9 $mm/yr^{0,5}$, the latter suggesting that a 30 mm layer of mortar will carbonate in merely 8 months!

Furthermore, carbonation depth unfortunately does not translate immediately to carbon capture because reaching pH<9.4 requires only a fraction of the available CaO and MgO to be combined with CO_2[121]. It is known that this fraction – the degree of carbonation – is maximum at the surface and decreases inwards (Figure 3) [121]. Capture-focused studies have often assumed a simplified profile (Figure 3). The maximum carbonation degree, in terms of available CaO converted to $CaCO_3$, reported in these models varies but can be up to 100% [120]. Lower figures appear more realistic, e.g., 50% as a final carbonation extent for crushed materials [122] or 30-90% [123]. Factors such as porosity, chemical composition of the hydrates, presence of SCMs, cement paste volume, and environmental conditions influence the maximum carbonation degree achieved[117]. Therefore, with today's knowledge, there is large uncertainty in any estimation of the amount of CO_2 that can be captured by a single structure [122].

Nevertheless, a few estimates of CO_2 capture by the in-use stock of cementitious products and waste have been published[128,129], with global estimates varying between 0.9 Gt in 2013 [120] to 0.7 ±1.2 Gt for 2015 [124]. In these global studies, capture is about 25% of the total annual CO_2 emissions from cement production. Values of 14-19.6% of annual emissions have been published for Portugal [125] and 17% for Sweden [123]. Further data collection on carbon capture of cementitious materials in current structures is needed. So far systematic data are limited to a Swedish study [123]. Some initial international efforts are described in PD CEN/TR 17310:2019[126], but the methodology of that study requires further extension and refinement to capture the range of influential parameters described above.

However, it is important to understand that this carbon uptake cannot be used to reduce the attributed current environmental impact of concrete production as this capture has already been happening. Some companies have started to explore CO_2 mineralization for products used in concrete (e.g. [127]). To be able to claim for a carbon uptake to count as a carbon sink related to COP21 Paris agreement targets, one would need to intentionally increase and hasten the carbonation process. This is what we explore in the following sections.

Increased CO_2 uptake at end of life

At the end of concrete's lifetime when it may be crushed into smaller pieces for reuse as aggregate in new concrete, carbonation could be increased due to higher surface exposure. This is by far the most discussed possibility to increase CO_2 uptake during concrete's life cycle. The total potential uptake could be around 75% of the initial limestone decalcination emissions [122]. This represents about 110 kg CO_2/m^3 for average concrete [128]. However, currently crushed concrete is stockpiled into a construction and demolition heap, and due to limited porosity of the heap itself, the carbonation of the piled aggregates is actually limited [122]. Afterward, recycled aggregates are reused as road subbase or in new concrete, which again reduces the carbonation potential due to limited access to CO_2 [129]. An increased carbonation rate could be achieved by longer exposure of crushed aggregates to the air or through enhanced processing such as accelerated carbonation [130]. However, the volume of materials to handle, the need to bring back these materials from demolition sites to concentrated industrial treatment facilities (with associated CO_2, particulate matter, and noise emissions from both transport and crushing), as well as the very low price of aggregates in many regions, makes full-scale development and deployment challenging in the global context.

Increased CO_2 uptake in the use stage

Reinventing industrial practices to increase CO_2 uptake is conceptually feasible since carbonation only reduces the durability of steel reinforced concrete that is exposed to outdoors wet and dry cycles or high humidity [118,131], which is only a fraction of the ~40% of cement going to reinforced concrete [132]. For all other elements, carbonation is mostly beneficial, including sometimes increased strength and reduced porosity[117]. More than 80% of cement is used in applications where higher carbonation will not induce durability concerns [132]. Therefore, engineers could be educated to embrace carbonation under these circumstances and actually design for carbonation.

Cements with a high SCM fraction not only emit less CO_2 during production, but also carbonate significantly faster and to a higher degree than conventional Portland cement [117,133,134]. In one example, the carbonation rate is increased by a factor 3 and maximum carbon uptake by a factor 2[135]. As a consequence, the replacement of current cements with high-SCM cements would, as a first step, reduce the total amount of CO_2 released into the atmosphere during cement production (due to reduced clinker content). Then, as carbonation is also faster, it will reduce the time during which the emitted CO_2 is staying in the atmosphere and the associated additional radiative forcing.

Considering the need for CO_2 diffusion to enable carbonation, design changes in terms of geometry (thickness) and CO_2 permeable surface coverings are also possibilities to reduce the time for capture and reduce the amount of materials and cement. 3D printing technology can introduce a degree of freedom making possible not only new shapes – increasing surface/volume ratio[136] - but also to vary the composition of concrete inside a given component. However, this technology is still in its infancy[137].

Carbon Capture and Storage

No cement can be neutral in overall CO_2 emissions unless carbon capture and storage (CCS) is used. Different technologies are available [138] (absorption, membrane process, mineral carbonation, oxyfuel, and others). Investment cost ranges between 200 and 300 million Euros per kiln [59], inducing a possible increase of between 50 to 100% in the price of cement [139], which can increase social inequalities. Actually, the increase in total costs for the construction of a middle-class multifamily residential building is limited to 1%, even when the cement price is doubled [140]. On the contrary, for low cost housing, the cost of cement represent 5 to 10% of construction costs and a price increase would directly impact final costs. Finally, legal issues to define which stakeholder will have to carry the risk associated with CO2 storage is not solved [141,142]. These legal uncertainties are delaying large scale implementation, although all experts are urging the sector to act fast [143].

5 Stakeholder actions for future implementation of sustainable cement and concrete

Concrete: the most destructive material on Earth. This is how this material is presented by some general media [22]. But housing and infrastructure needs for growing urban population make its use unavoidable, and its environmental impacts come more from the scale of use rather than its per-unit contribution. It is hard to blame the material or the technology while the cause is mainly the urbanization and the massive use of such material. Certainly, concrete allows us to handle the social challenges of housing and infrastructure demand with a minimum environmental impact per product delivered. We pointed out that stringent regulation and control can push the widespread implementation of already-used technologies to reduce environmental and health impacts associated with material extraction, water consumption, particulate matter, and heavy metals emissions. GHG emissions and contributions to climate change are the urgent remaining challenges to focus on. It is accepted by industry as well as public actors that cement and concrete have an environmental burden. However, there is far less consensus when dealing with action to remediate to this situation.

Depending on the efficiency of the cement plant and the amount of waste co-processed, a same product coming from two different cement plants will have very different environmental impacts [144]. More transparency and better measurement could help the various stakeholders to make informed choices. In this perspective, the Concrete Sustainability Council (CSC) is a recent initiative from the cement and concrete industry to follow the Forest Stewardship Council (FSC), which awards certification that a finished concrete product can fulfil sustainability criteria along the upstream value chain up to the cement plant. This includes environmental and social issues such as land use, air quality, water, biodiversity, health and safety, labor practices[51].

Concrete is also typically seen a single material, but the diversity of cement types and concrete mixes makes it such that for similar strength and durability performance one can triple the carbon footprint of the product[72,145]. As long as specifications are based on material formulations or recipes (which is currently the most popular approach in standards worldwide), or even on technical performance (strength, fluidity) and not on environmental performance, there will be no incentive from the concrete producer to propose an environmentally friendly mix design. Some concrete producers have started to guarantee to their customers that they will provide a given class of low carbon concrete (15, 25 and 40% lower CO_2 than average standard) at a construction site (e.g., Vertua[146]). This is a clear step forward and shows a change taking place in the profession. Moving standards from a recipe (prescriptive) basis to a performance basis is essential, but demands that "performance" is defined holistically and including environmental considerations if it is to have the necessary effect on emissions across the sector.

At the structural level, one can observe the same misunderstanding. It is possible to design materially efficient structures, but clients usually do not ask for it [147] and without a request from the client (or a national or regional policy requiring that this be done), the design team has no incentive to optimize their structure and will go for very regular 20 cm thick slabs.

Efforts from all stakeholders, from policymakers downwards, are therefore required to accumulate all marginal gains available **(Table 2)**. However, time constraints, fragmented supply chains, and lack of awareness are some of the many barriers for implementation. In order to motivate all the different actors involved in cement use, a set of benchmarks can be proposed [111]. In Europe, it was proposed to use for the cement producers, the $tCO_2/t_{clinker}$ metric, which should be lower than 0.7 [55]. Concrete producers should achieve less than 3.5 kg clinker/m^3/MPa for a standard concrete mix (30-50 MPa) [72].

Engineering offices that design concrete structures should achieve less than 250 kg CO_2/m^2 floor area for the concrete allocated to the structure [88] and prescribe exposure class with no corrosion risk when concrete is used indoors. For construction companies, less than 500 kg CO_2/m^2 floor area for the whole building is a good benchmark [148]. These are European benchmark propositions and they need to be tailored to the local context. In particular, they become highly irrelevant when looking at the informal concrete production sector [83], which represents a non-negligible part of cement consumption.

Carbonation should be taken as an opportunity. Thanks to the current movement toward using cements with high amounts of clinker substitution, we can design for faster carbonation and shorten considerably the carbon overshoot due to urbanization. As long as concrete is not directly exposed to 90% relative humidity and construction details are finished with high quality, there will be no durability issue. Innovative corrosion resistant steel alloy may also solve this problem

No single "silver bullet" innovation will achieve sustainable cement use and cement industry will not solve all problems acting in isolation. It is part of a loosely coupled and complex network of actors that collaborate to produce buildings and infrastructure [93], from the material producer, the engineering office, the architect, the construction manager, the policy maker and the owner of the future building. And it is the collaboration between actors that produces significant differences[149]. Like in sport, it is the combination of marginal gains which actually makes the difference [150,151].

Box 1: Heavy metals and hazardous substances emissions

Emissions during cement production

The fuels and raw materials used in cement kilns can be sources of hazardous air pollutant emissions such as polychlorinated dibenzo-_p_-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) [21]. This can be a matter of concern in some countries, such as China [152]. However, when regulation and controls are implemented, no increased level of pollutants can be measured in the vicinity of cement plants [153–155]. As heavy metals [156] and other hazardous compounds are incorporated in the clinker and cement kiln dust [60,157], appropriate control devices and exhaust filters can mitigate heavy metal and hazardous air emissions [60,157]. Furthermore, the high temperatures and the alkaline conditions in cement plants allow for the full decomposition of the fuel's organic part [154,158,159].

Emissions at End of life. Leaching from SCMs

Several industrial wastes can cause leaching of heavy metals when stockpiled. Yet their use as partial cement replacement can often stabilize them due to the high pH of interstitial pore solution which precipitates heavy metals complexes[160]. Such benefits have been shown to be less effective in the case of poorly cured concrete [161]. While currently used industrial wastes as partial replacement of cement do not appear to have leaching issues in appropriately cured concrete, alternatives such as municipal solid waste incineration (MSWI) bottom ash and non-ferrous slags are anticipated to have some chloride and metal leaching issues[162]. However, even in these cases, it is thought that leaching of undesirable compounds can be mitigated through the use of pre-treatments to remove or convert potentially harmful compounds [66]. For instance, pre-hydration or carbonation can be used to reduce metal leaching from MSWI bottom ashes [163,164].

Box 2: Water consumption

Water is one of the main constituents in concrete and its use can be as high as cement consumption by mass [31]. The direct water consumption used in cement products is equivalent to 400 L per capita each year. However, this water used as a constituent in concrete represents only about 20% of the total water consumed in its production [19,165]. The remaining water is energy-related or process-related [19]. Much of the process-related water is consumed during the quarrying, crushing, and washing of the raw materials used in the production of cement and concrete, e.g., as a dust suppression method [19,165]. The energy-related water consumption depends on cement kiln type [157] and the energy mixes which can vary significantly depending on location [166]. On average, less than 50% of water consumption associated with concrete production is linked to the cement [19] and water management strategies should thus be implemented all along the supply chain.

The cement and concrete sector plays a minor role in water scarcity discussions, contributing less than 5% of total water withdrawal [167] and in most countries less than 1% of total renewable water resources [19]. However, water is a complex interwoven environmental issue. For example, a transition from river aggregate to crushed aggregate in order to have sustainable management of mineral resources induces an increase in water consumption due to the need for washing crushed aggregates. Conversely, in emerging countries crushed stone are rarely washed which increase dust problems and health related issues as well as reducing the strength performance of concrete. There is therefore a water-mineral resources nexus, and development of crushed gravel has to be combined with closed-loop water treatment.

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7 Glossary

Aggregate: granular material, such as sand, gravel, crushed stone, or iron blast-furnace slag, used with a cementing medium to form hydraulic-cement concrete or mortar. Aggregate may be natural, manufactured or recycled. Aggregates make up some 60 -80% of the concrete mix. (ASTM C125 R2008)

Binder: Any material with binding property. It generally consists of cementitious material and water.

Biomass: substance wholly comprised of living or recently living (non-fossil) material. (ASTM E1705) —When considered as an energy source, biomass may be further subdivided into: (1) primary biomass—rapidly growing plant material that may be used directly or after a conversion process for the production of energy, and (2) secondary biomass —biomass residues remaining after the production of fibre, food, or other products of agriculture, or biomass by-products from animal husbandry or food preparation that are modified physically rather than chemically. Examples include waste materials from agriculture, forestry industries, and some municipal operations (manure, saw dust, sewage, etc.) from which energy may be produced

C_3S, C_2S, C_3A and C_4AF: clinker mineral phases noted with cement chemical notation. C stands for CaO, S for SiO_2, A Al_2O_3 and F for Fe_2O_3.

Cement: a cement sets and hardens by chemical reaction with water and is capable of doing so under water. (ASTM C125 R2015)

Cement Kiln Dust (CKD): CKD are collected during the firing of raw materials during the clinker manufacturing process. CKD consists of four major components: unreacted raw feed, partially calcined feed and clinker dust, free lime, and enriched salts of alkali sulfates, halides, and other volatile compounds.

Clinker: the active part of portland cement . It is a dark grey nodular material made by heating ground limestone and clay at a temperature of about 1400 °C - 1500 °C.

Concrete: a composite material that consists essentially of a binding medium within which are embedded particles or fragments of aggregate; in hydraulic-cement concrete, the binder is formed from a mixture of hydraulic cement and water. (ASTM C125 R2015)

Filler: mineral filler, a finely divided mineral product at least 65 % of which passes the 75-µm sieve. (ASTM C1777)

Gravel: coarse aggregate resulting from natural disintegration and abrasion of rock or processing of weakly bound conglomerate. (see aggregate) (ASTM C125 R2016)

Mortar cement: a mixture of finely divided hydraulic cementitious material, fine aggregate, and water in either the unhardened or hardened state; hydraulic mortar. (ASTM C219)

Plaster: hydraulic cement, a mixture of hydraulic cement, fine aggregate and water that hardens; used for coating surfaces, such as ceilings, walls and partitions. (ASTM C219)

Ready Mix Concrete: concrete manufactured and delivered to a purchaser in a fresh state. (ASTM C94)

Sand: fine aggregate resulting from natural disintegration and abrasion of rock or processing of completely friable sandstone. (ASTM C125 R2018)

Supplementary cementitious materials (SCM): an inorganic material that contributes to the properties of a cementitious mixture through hydraulic or pozzolanic activity, or both. Some examples of supplementary cementitious materials are fly ash, silica fume, slag cement, rice husk ash, and natural pozzolans. In practice, these materials are used in combination with portland cement. (ASTM C125 R2015).

Figures and tables caption

Figure 1: Examples of cement and concrete contribution to the global warming potential. *For European building stock, the contribution of building materials is reduced for existing buildings as their low energy performance induce large contribution of energy for heating. New buildings have much lower emissions during their operation and contribution of embodied emission is higher (Values are average value from 230 buildings mainly from Europe (75%) and Asia (25%)* [14]*)*. At the building level, the embodied emissions from a typical multifamily masonry building come mainly from reinforced concrete followed by contribution of windows *(Values are average of 35 buildings from France and Switzerland built between 2010 and 2015* [15,168]*)*. For the production of one cubic meter of concrete the main CO_2 emissions come from cement production followed by transport of raw materials *(Values are the average of main concrete type made with 25% SCMs in Australia* [169] *and Switzerland* [170]*)*. Finally, considering current clinker production efficiency and the replacement of 30% SCM in the final cement, main emissions are due to decarbonation of limestone and burning fuels, both processes involved in clinker production *(Values are average French values* [57,61,67]*)*.

Figure 2: Cement value chain. *From raw material extraction until the demolition of the building, numerous stakeholder are involved, but very seldom integrated (adapted from* [24,111]*).*

Figure 3: Carbonation profile through concrete. *Measures and model adapted from* [120,171]*. Carbon uptake range adapted from* [122,135]*.*

Table 1: Available technologies along the cement and concrete value chain, their improvement potentials and the stakeholder that should be encourage to take action. (data from [55] [172] [173] [61] [174,175] [89,147,176])

Table 2: Stakeholder description