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Review of concrete solutions for concrete's environmental impacts

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12 Abstract

13 The use of concrete is under scrutiny as it appears as one of the few human activities where the 14 transition toward a post-carbon society is not possible unless large investments in risky carbon capture 15 and storage are made. With current urbanization, it is also a sector that is expected to continuously 16 grow, leading to increased resource consumption and emissions. In this review, we aim to shed light 17 on the available solutions that can be implemented in the short and long term to reduce greenhouse 18 gas emissions. Rather than waiting for disruptive technologies that could transform a very slow moving

19 and risk-averse construction sector, this review focuses on the small improvements that every

20 stakeholder involved along the value chain of concrete production and use can achieve. We stress how

21 significant the combined effect of these marginal gains can be. By balancing societal needs,

22 environmental requirements, and technical feasibility, the intention of this review is to show credible

- 23 pathways for a transition to sustainable use of concrete.
- 24
- 25

26 **Key points**

27 • Cement usage is so massive, more than 4 billion tonnes per year worldwide, that large-scale 28 replacement by other materials within the next decade is not possible.

29 • Environmental impact of cement and concrete is low per unit of material, but the amount used makes the impact of the sector highly significant. 30

31 • Reductions in CO₂ emissions are possible through successive improvement all along the cement and 32 concrete value chain: less clinker in cement, less cement in concrete, less concrete in structures, and

33 less replacement of structures.

34 • By engaging all stakeholders of the construction sector, immediate savings of the order of 50% can 35 be reached without heavy investment in new industrial infrastructure or modification of standards.

36 Research and development need urgently to be conducted for post-2050 construction to meet future 37 emissions reduction targets. Alternative cement and faster carbonation of concrete should be 38 explored.

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Introduction 41 1

Concrete is the fundamental building block of our urbanizing world. It makes up the buildings and 42 43 infrastructure that enable businesses to operate and people to carry out their daily activities. Over the 44 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.

47 Concrete is a synthetic rock made of cement, sand, gravel and water, and is by far the most used man-48 made material. Cement, which is the mineral glue that sticks together sand and gravel in the concrete, 49 represents around 10% of concrete mass and is currently produced at around 4 Gt/year, almost the 50 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. 51 52 Among materials used for construction, cement accounted for 36% of the 7.7 GtCO₂ released globally in 2010 by construction activities ⁴, while steel accounts for 25% ⁵, plastics 8% ⁴, aluminum less than 53 54 4%, 6 and brick less than 1% 7,8 .

55 Concrete accumulates in the Earth's crust and is now considered to be one of the markers of the 56 Anthropocene ⁹ with an estimated 900 Gt added since the beginning of the industrial revolution ^{9,10}. 57 But it is important to remember than only about half of cement is used for concrete ¹¹, the rest being 58 used for blocks, mortars, and plasters. To grasp what volume these masses represent, one should 59 picture every person on Earth building every year the equivalent of a 20 cm thick concrete wall of 4.5 60 m² area, as well as plastering a wall surface of 35 m² with a 3 cm thick cement-based plaster 61 (considering 300 kg cement/m³ of concrete and 250 kg/m³ 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 world¹³ due to the achievement of urban transition and the stabilization of the population ¹³.

68 It is therefore crucial to act now and drastically reduce the environmental impact of construction 69 within the next decades during this urbanization peak. The new buildings are expected to consume 70 less energy during their operation, which should increase the focus on emissions related to concrete 71 ¹⁴. Actually, for a new typical masonry multifamily building type, steel-reinforced concrete represents 72 50% of the CO₂ emissions attributed to the building, followed by windows, insulation, ceramic tiles, 73 and paint¹⁵ (figure 1). Greenhouse gas (GHG) emissions ¹⁷, local scarcity of non-renewable resources 74 ¹⁸, energy consumption ¹¹, water use ¹⁹, dust and particulate matter emissions ²⁰, mercury emissions ²¹ 75 are known issues related with cement and concrete production. But no other known material has been 76 found to provide the same amount of service than reinforced concrete at such a low economic cost ¹¹. 77 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 78 79 environmental impacts of concrete production, while maintaining the favorable properties of concrete 80 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.

85

86 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 attention²², 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.

93

94 2.1 Cement and concrete production

95

Portland cement is composed of four major oxides: CaO, SiO₂, Al₂O₃ and Fe₂O₃ coming from raw 96 97 materials, usually limestone, clay, and small amounts of "corrective" materials such as iron ore, 98 bauxite, and sand to reach the desired chemical composition. Raw materials are crushed, mixed and 99 milled into a raw meal, which is then heated in the pre-heating system to dissociate carbonate into 100 calcium oxide and carbon dioxide (Figure 2). The meal is then calcined in a rotary kiln at up to 1500°C 101 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 C₃S, C₂S, C₃A and C₄AF, called 102 103 clinker. This clinker is inter-ground with gypsum to a finer product called cement. Concrete is produced 104 by mixing cement with sand, gravel (or crushed stone), water, and chemical admixtures. It is produced 105 in a concrete plant and transported by concrete truck to the construction site, or directly mixed at the 106 construction site. Concrete is also used to produce precast elements. Finally, cement can be used in 107 plaster and mortar when mixed with water, sand, lime and chemical admixtures, both on site and in 108 premix mortar factories.

109

110 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 PM₁₀, 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 burdens²³.

123 Currently, appropriate filtering and capture of PM can mitigate many of these emissions from cement 124 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 125 126 clinker if it has appropriate alkali content, thus reducing emissions²⁹. Further, use of scrubbers or 127 alterations in energy mixes can drive down both PM and other air emissions. Yet regulations requiring 128 use of such technologies vary by region. For instance, a recent study in Zambia showed that PM_{2.5} 129 concentrations were still 5 to 10 times higher within the vicinity of the cement plant than 1 km away 130 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.

135 2.3 Regional resource scarcity

To supply the equivalent of 4.5 m² of typical concrete wall and 35 m² 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 m³ of concrete ³¹.

140 This tremendous demand for resources has led to a growing concern about potential contributions to 141 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 142 143 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 144 abundant on Earth ³⁴, they are usually not transported over long distances due to the economic cost 145 of transporting such heavy materials^{35,36}. Therefore, a construction boom increases the local pressure 146 for natural sand and coarse aggregates close to urban areas ¹⁸. But as quarries and sand mines become 147 148 undesirable due to NIMBYism or too expensive to operate close to urban areas, transportation 149 distances will increase. For example, aggregates for San Francisco Bay Area (California) come from 150 British Columbia (Canada).

151 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 152 poor resource management ⁴⁰ as natural sand extraction usually does not require complex operations 153 and can be carried on as informal activity near large cities ⁴¹. Thus, quantification of sustainable 154 resource extraction possibilities around cities should focus on additional factors other than local 155 availability, such as land use changes ⁴², resource accessibility ⁴³ and consideration of political and 156 economic actions ⁴⁴. While local pressures on resources have been noted, it is possible to reduce the 157 158 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 159 excavation materials usually landfilled outside the city. In Switzerland it represents a similar amount 160 as the primary material required to build ⁴⁶. In China, considering 0.9 t excavation per m² built ⁴⁷, this 161 material could currently supply half of the construction material requirement⁴⁸. A better use of this 162 material is possible through washing to extract sand and gravel from it⁴⁹ or directly through clay based 163 concrete ⁵⁰. Finally, sustainable resource management can be achieved through promotion of strong 164 policy and regulatory frameworks, such as certification systems to secure the good practice along the 165 166 supply chain (e.g., CSC label ⁵¹).

167

168 2.4 Global environmental issues

169 One of the most commonly discussed environmental impacts related to cement production is the high 170 level of GHG emissions. Among materials used for construction, cement accounts for 36% of emissions 171 related to construction activities⁴ and 8% of total anthropogenic CO₂ emissions ⁵². At least 70% of the 172 GHG emissions from concrete production is due to cement production **(Figure 1)** ³¹. Unlike regional 173 resource scarcity and local health issues, for which in many cases there are mitigation strategies that 174 can be implemented today, technological breakthroughs are needed globally to meet GHG emissions 175 mitigation goals.

Within cement manufacturing, the predominant source of emissions is the kilning stage; grinding, 176 sorting of raw materials, and packaging of cement bags have minor impacts ^{17,53}. Most of the emissions 177 178 are from the decomposition of limestone (calcination) and associated with energy use ⁵⁴. Energy-179 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 CO₂ per kg of cement from energy-180 resource combustion ⁵⁶. Calcination represents around two-thirds of the total GHG emissions from 181 182 cement produced using a state of the art dry process rotary kiln equipped with pre-calciner ⁵⁷. These 183 are the main reasons why cement production is considered to be difficult to decarbonize ¹⁶ as 184 decarbonization of the energy supply will not eliminate the material-related CO_2 emissions from 185 calcination¹⁶. However, there have been many mitigation strategies for these emissions, several of 186 which are discussed in the subsequent sections.

187 As a consequence of the annual demand for 4.5 m^2 of concrete and 35 m^2 of plaster for every inhabitant 188 on the planet, there is an associated ~300 kg CO₂ 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 thecement and concrete industries - and for the World.

192

193 3 Credible medium-term solutions to reduce cement demand

194 The peak in new construction will come in the next few decades, so it is most important to focus on 195 reductions in environmental impact that can be achieved in the medium term, before 2050. The 196 substantial reduction needed can only be achieved by considering efficiency at all stages of the value 197 chain: clinker production, cement production, cement use in concrete or mortar, concrete use in 198 construction, design of structures and use of structures. It is essential that all parts of this chain are 199 considered as there is no point saving, for example, 30% CO₂ in cement production and then using 200 twice as much cement as needed in the concrete. In this section, we explore the potential for further 201 reductions in environmental impacts along this value chain (Table 1). These solutions are of particular 202 importance because they can often be implemented without new production technologies or 203 infrastructure.

204

205 3.1 Efficiency of clinker production

206 As previously described the majority of CO₂ emissions in concrete, come from clinker production. 207 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 208 209 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 210 achieve about 63% efficiency and through integrated approaches could reach 80% efficiency ⁶⁰. Such 211 levels of efficiency make modern kilns among today's most efficient thermal machine in wide-scale 212 213 industrial use. There is therefore limited scope for further improvement ⁵⁵.

214 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 215 216 tyres (50% of total recycled tyres) were used as fuels for clinker production. Already in the 1990s in 217 the United States, about 70% of all hazardous wastes were burnt in cement kilns. Waste materials 218 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 219 a significant net CO₂ reduction because cement kilns are more efficient ⁶¹. Another advantage is that 220 221 no toxic residues such as dioxins are generated since the ashes are completely incorporated in 222 clinker⁶². The International Energy Agency Roadmap expected the worldwide use of "alternative fuels" 223 to grow from 3% in 2006 to about 37% in 2050 and deliver around 15% of the targeted overall 224 reduction in CO₂ emissions ^{55,63}.

This increase in fuel efficiency means that fuel now accounts for only about one-third of the CO₂ 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 (CaCO₃). 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.

234

235 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.

255

256 3.3 Concrete efficiency

257 There is considerable scope to reduce CO_2 emissions by a more efficient use of cement in concrete 258 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 259 260 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 261 262 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 2⁷⁵. The use of a proper mixture design in a 263 concrete plant, with appropriate proportion of sand, gravel can lead to further reductions (up to 50%) 264 without loss of strength or fluidity ⁷⁶. Such improvements are an untapped potential in emerging 265 countries where most of the cement is sold in bags and used without proper technical control or mix 266 design optimization ⁷⁷. Promoting industrialized concrete production as a replacement for site-mixing, 267 268 especially in self-help housing schemes is a very effective way to reduce cement consumption in both 269 concrete and mortar applications⁷⁸.

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.

276

277 3.4 Construction efficiency

278 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 279 280 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 281 282 growth in concrete demand is expected and where quality control on construction site may be lower. 283 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 284 changes in the quantity of cement needed for concrete production ³¹. Better control on water and 285 aggregate humidity on construction site can also have a critical influence ⁸³. Since waste cost to 286 builders, raising awareness was efficient on Brazilian market⁸². Education of construction workers is 287 288 also a proven strategy ⁸⁴.

289

290 3.5 Design efficiency

Research has shown that buildings use structural material inefficiently⁸². In structural systems, GHG 291 reduction is complicated by the interplay of concrete performance (and hence mixture proportions) 292 293 and the quantity of steel reinforcement, which are often highly constrained by codes. For reinforced 294 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 295 296 target⁸⁵. These combined optimization strategies of concrete strength, rebar content and clinker content can provide around 20% reductions in GHG emissions⁸⁵. Similar reduction can be achieved for 297 298 structures where the dead load is the key design parameter through the use of high performance concrete⁸⁶. Orr and co-authors demonstrate that more efficient utilization of structural concrete had the 299 potential to achieve material savings up to 30–40% through design optimization ⁸⁷. Although the mag-300 nitude of such savings is difficult to quantify, the works of De Wolf⁸⁸, Shank and co-authors⁸⁹ would 301 302 also argue for 10%-20% reduction within conventional design constraints. Finally, savings can also be 303 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 304 305 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 ser-306 307 vice life of infrastructure with less than 50% the GHG emissions required for conventional rehabilita-308 tion, and a fraction of what it would cost to rebuild them^{91,92}.

309

310 3.6 Reduction of GHG emissions all along the value chain

311 It is clear that working on marginal gains all through the value chain can lead to substantial savings in 312 GHG emissions (Table 2). The savings are not necessarily additive and may not be appropriate in all 313 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 314 315 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 316 317 prefabrication and post-tensioning (Table 2). The difficulty to implement these savings comes mainly 318 from the fact that the construction sector is a fragmented industry with multiple stakeholders ⁹³. 319 Outside the cement industry, which concentrates investment and production capacity, the other 320 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}.

324

325 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.

332

333 4.1 Breakthrough solutions, the reality behind the hype.

334 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 335 336 the future of the construction sector by replacing cement in part or in full⁹⁹. Several alternative cements have been shown to be able to contribute to reduced environmental impacts relative to 337 conventional cements^{100,101}. However, the pace of change in the construction industry, issues in 338 339 materials availability or cost, and the technical limitations of some of these alternative technologies, 340 mean that many proposed material alternatives are unrealistic from technical or resource standpoints 341 and are unlikely to reach large-scale technical maturity before 2050 where a transition to net zero 342 emissions is required. It is actually difficult for alternative cements to meet more than 5% of the 343 projected future demand for cementitious materials ^{102,103}.

344 Even though sufficiently mature alternative cements are already in use at commercial scale in many 345 parts of the World, the production capacity expansion is limited. For example, calcium sulfo-aluminate 346 (CSA) cements are well-known products, largely used in China. This technology is a real alternative 347 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 348 349 a few percent of cement production at most. Let's imagine that even if all current bauxite production 350 was diverted from the production of aluminum it would not be sufficient to provide more than 10-15% 351 of the current demand for cement.

352 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 353 applications¹⁰³. In regions where the supply of both suitable activators and precursors is plentiful, they 354 355 have been shown to be economically and technically viable in precast and ready-mixed formats ¹⁰⁵. 356 However, there remain supply-chain challenges related to availability of highly effective alkaline 357 activators such as sodium silicate, which are not currently produced at sufficient scale to replace even 358 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 359 production¹⁰⁶ but is still in competition for the supply of aluminosilicate precursors, as the precursors 360 361 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-362 content solid wastes which cannot normally be recovered^{97,107}. 363

364 One of the main challenges in the area of alkali-activated concretes, and other technologies based on 365 industrial wastes, relates to the scale on which waste are needed to become a realistic input into large366 scale construction. Waste which is generated at a rate of tens of tons per annum may be a major 367 disposal challenge for many industries, but this is a scale which is far too small to be worth even 368 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 369 370 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 371 metallurgy, and construction and demolition waste ¹⁰⁸. They all share the characteristic that they can 372 be to some extent quality-controlled, which is essential to achieve the necessary consistency of 373 construction products. 374

375 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 376 a sustainability advantage if magnesium carbonate is obtained through carbonation of geologically-377 sourced magnesium silicate by uptake of CO₂ that would otherwise be emitted to the atmosphere ¹¹⁰. 378 379 However, past attempts to develop a scalable process have not succeeded, and the likely very high 380 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 381 magnesium silicates, the availability of these materials is much more localized than limestone used to 382 produce Portland cements, entailing significant transport costs if cements based on magnesium 383 silicates are to be used on a global rather than local scale ¹¹². Moreover, magnesium silicates are less 384 available near the Earth's surface, so deep mining operations would be required to recover the 385 amounts needed to meet the demand for construction. Magnesium recovery from brines for use in 386 cements has also been proposed ¹¹³, but is also probably geographically limited to regions in which 387 388 large-scale seawater desalination is taking place or where salt lakes are accessible.

389 Many other suggested solutions are based on the idea of cement setting and hardening through 390 carbonation of calcium oxide. This allows to capture the CO₂ emitted during cement production and to 391 tend toward carbon-neutral cement. The problem is to find sources of calcium oxide that do not come 392 from the decarbonation of limestone in the first place. If CaO is derived from limestone, then there 393 can be no net gain as the CO₂ which can be reabsorbed can never be higher than the CO₂ emitted in the 394 decarbonation step, and there would still be additional impacts from required process energy. Development of "Carbonatable Calcium Silicate Cement" (CCSC) technology has been developed 395 thanks to recent development to accelerate and control carbonation industrially without excessive 396 energy consumption¹¹⁴. Simple calcium silicate minerals such as wollastonite can carbonate very 397 rapidly in relatively pure CO₂ gas (e.g., Solidia Cement¹¹⁵). These binders are well-suited for the 398 fabrication of thin precast products, to allow CO₂ and water transfer during curing. They also involve 399 400 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 401 a solution for some specific locations. As the current global wollastonite production amount is 500 000 402 tons per year, mainly in China ¹¹⁶, a transition towards this technology would require wollastonite 403 extraction to increase by a factor of around 10 000. 404

405 There has been much focus on concrete made with alternative cements for over half a century, but 406 the availability of raw materials, the confidence in long term performance, or the limitation to specific 407 application in well-controlled environments make it unrealistic to consider any of these alternative as 408 a direct one-for-one replacement for conventional cementitious materials within the next decades. 409 However, when considering a more local context, and if it is proposed to move away from the idea 410 that all locations in the World can (or even should) be using the same type of cement for all 411 applications, there is a great deal that can be achieved by the production of fit-for-purpose local 412 cement technologies and solutions specific to the areas where the desired resources do exist. The most 413 critical issue here is cost; these alternative cements must be made scalable and cost-competitive, but 414 can only occur at a local level.

416 4.2 Carbonation of cement and concrete

For classic Portland cement, further CO₂ savings can be achieved in the use phase and at end of life. Actually, when exposed to the atmosphere, cementitious materials can capture CO₂ through carbonation. The amount taken up is some fraction of the one released by limestone decomposition (calcination) during cement production.

421 Carbonation involves the calcium-containing phases from cement such as calcium-silicate-hydrates, 422 calcium-aluminate-hydrates as well as portlandite $(Ca(OH)_2)$ reacting with CO_2 to produce mainly 423 calcium carbonate and other non-carbonated phases ¹¹⁷. The reaction starts on the exposed surface 424 and proceeds by CO_2 diffusing slowly inwards. This reaction has been extensively studied by engineers 425 because, by reducing the pH of the concrete pore water below pH ~9.4, it may damage the 426 electrochemical protection of mild steel reinforcement bars against corrosion ¹¹⁸ ¹²³, which is 427 deleterious for the durability of concrete structures exposed to high relative humidity or rain.

- 428 Carbonation depth is commonly described as a diffusion-limited process: depth=k.t^{0.5} (where t is time 429 and k a constant). The value of k for real concrete structures usually varies between 2 and 15 mm/yr^{0.5} 430 ¹¹⁹, meaning that a 200 mm thick concrete column can take 44 to 2500 years to reach pH 9.4. No 431 systematic information exists on carbonation of other products such as mortars and renders, except 432 for a mention ¹²⁰ of unpublished results with k ranging from 6.1 to 36.9 mm/yr^{0,5}, the latter suggesting 433 that a 30 mm layer of mortar will carbonate in merely 8 months!
- 434 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 435 CO_2^{121} . It is known that this fraction – the degree of carbonation – is maximum at the surface and 436 decreases inwards (Figure 3) ¹²¹. Capture-focused studies have often assumed a simplified profile 437 (Figure 3). The maximum carbonation degree, in terms of available CaO converted to CaCO₃, reported 438 in these models varies but can be up to 100% ¹²⁰. Lower figures appear more realistic, e.g., 50% as a 439 final carbonation extent for crushed materials ¹²² or 30-90% ¹²³. Factors such as porosity, chemical 440 composition of the hydrates, presence of SCMs, cement paste volume, and environmental conditions 441 influence the maximum carbonation degree achieved¹¹⁷. Therefore, with today's knowledge, there is 442 large uncertainty in any estimation of the amount of CO_2 that can be captured by a single structure ¹²². 443
- Nevertheless, a few estimates of CO₂ capture by the in-use stock of cementitious products and waste 444 have been published^{128,129}, with global estimates varying between 0.9 Gt in 2013 ¹²⁰ to 0.7 ±1.2 Gt for 445 2015 124 . In these global studies, capture is about 25% of the total annual CO₂ emissions from cement 446 production. Values of 14-19.6% of annual emissions have been published for Portugal ¹²⁵ and 17% for 447 Sweden ¹²³. Further data collection on carbon capture of cementitious materials in current structures 448 is needed. So far systematic data are limited to a Swedish study ¹²³. Some initial international efforts 449 are described in PD CEN/TR 17310:2019¹²⁶, but the methodology of that study requires further 450 extension and refinement to capture the range of influential parameters described above. 451
- 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 CO₂ 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.
- 458
- 459 Increased CO₂ uptake at end of life

460 At the end of concrete's lifetime when it may be crushed into smaller pieces for reuse as aggregate in 461 new concrete, carbonation could be increased due to higher surface exposure. This is by far the most 462 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 463 CO₂/m³ for average concrete ¹²⁸. However, currently crushed concrete is stockpiled into a construction 464 and demolition heap, and due to limited porosity of the heap itself, the carbonation of the piled 465 aggregates is actually limited ¹²². Afterward, recycled aggregates are reused as road subbase or in new 466 concrete, which again reduces the carbonation potential due to limited access to CO₂¹²⁹. An increased 467 468 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, 469 470 the need to bring back these materials from demolition sites to concentrated industrial treatment 471 facilities (with associated CO₂, particulate matter, and noise emissions from both transport and 472 crushing), as well as the very low price of aggregates in many regions, makes full-scale development 473 and deployment challenging in the global context.

474

475 *Increased CO₂ uptake in the use stage*

Reinventing industrial practices to increase CO₂ 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 porosity¹¹⁷. 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.

483 Cements with a high SCM fraction not only emit less CO_2 during production, but also carbonate 484 significantly faster and to a higher degree than conventional Portland cement ^{117,133,134}. In one example, 485 the carbonation rate is increased by a factor 3 and maximum carbon uptake by a factor 2¹³⁵. As a 486 consequence, the replacement of current cements with high-SCM cements would, as a first step, 487 reduce the total amount of CO_2 released into the atmosphere during cement production (due to 488 reduced clinker content). Then, as carbonation is also faster, it will reduce the time during which the 489 emitted CO_2 is staying in the atmosphere and the associated additional radiative forcing.

490 Considering the need for CO₂ diffusion to enable carbonation, design changes in terms of geometry 491 (thickness) and CO₂ permeable surface coverings are also possibilities to reduce the time for capture 492 and reduce the amount of materials and cement. 3D printing technology can introduce a degree of 493 freedom making possible not only new shapes – increasing surface/volume ratio¹³⁶ - but also to vary 494 the composition of concrete inside a given component. However, this technology is still in its infancy¹³⁷.

495

496 Carbon Capture and Storage

No cement can be neutral in overall CO₂ emissions unless carbon capture and storage (CCS) is used. 497 Different technologies are available ¹³⁸ (absorption, membrane process, mineral carbonation, oxyfuel, 498 and others). Investment cost ranges between 200 and 300 million Euros per kiln ⁵⁹, inducing a possible 499 increase of between 50 to 100% in the price of cement ¹³⁹, which can increase social inequalities. 500 Actually, the increase in total costs for the construction of a middle-class multifamily residential 501 building is limited to 1%, even when the cement price is doubled ¹⁴⁰. On the contrary, for low cost 502 503 housing, the cost of cement represent 5 to 10% of construction costs and a price increase would 504 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.

507

508 5 Stakeholder actions for future implementation of sustainable cement and concrete

509 Concrete: the most destructive material on Earth. This is how this material is presented by some 510 general media²². 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 511 512 contribution. It is hard to blame the material or the technology while the cause is mainly the 513 urbanization and the massive use of such material. Certainly, concrete allows us to handle the social 514 challenges of housing and infrastructure demand with a minimum environmental impact per product 515 delivered. We pointed out that stringent regulation and control can push the widespread 516 implementation of already-used technologies to reduce environmental and health impacts associated 517 with material extraction, water consumption, particulate matter, and heavy metals emissions. GHG 518 emissions and contributions to climate change are the urgent remaining challenges to focus on. It is 519 accepted by industry as well as public actors that cement and concrete have an environmental burden. 520 However, there is far less consensus when dealing with action to remediate to this situation.

521 Depending on the efficiency of the cement plant and the amount of waste co-processed, a same 522 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 523 524 choices. In this perspective, the Concrete Sustainability Council (CSC) is a recent initiative from the 525 cement and concrete industry to follow the Forest Stewardship Council (FSC), which awards 526 certification that a finished concrete product can fulfil sustainability criteria along the upstream value 527 chain up to the cement plant. This includes environmental and social issues such as land use, air quality, 528 water, biodiversity, health and safety, labor practices⁵¹.

529 Concrete is also typically seen a single material, but the diversity of cement types and concrete mixes 530 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 531 532 currently the most popular approach in standards worldwide), or even on technical performance 533 (strength, fluidity) and not on environmental performance, there will be no incentive from the 534 concrete producer to propose an environmentally friendly mix design. Some concrete producers have 535 started to guarantee to their customers that they will provide a given class of low carbon concrete (15, 536 25 and 40% lower CO₂ than average standard) at a construction site (e.g., Vertua¹⁴⁶). This is a clear step forward and shows a change taking place in the profession. Moving standards from a recipe 537 538 (prescriptive) basis to a performance basis is essential, but demands that "performance" is defined 539 holistically and including environmental considerations if it is to have the necessary effect on emissions 540 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 tCO₂/t_{clinker} metric, which should be lower than 0.7 ⁵⁵. Concrete producers should achieve less than 3.5 kg clinker/m³/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.
- 557 Carbonation should be taken as an opportunity. Thanks to the current movement toward using 558 cements with high amounts of clinker substitution, we can design for faster carbonation and shorten 559 considerably the carbon overshoot due to urbanization. As long as concrete is not directly exposed to 560 90% relative humidity and construction details are finished with high quality, there will be no durability
- 561 issue. Innovative corrosion resistant steel alloy may also solve this problem
- 562 No single "silver bullet" innovation will achieve sustainable cement use and cement industry will not 563 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.
- 566 And it is the collaboration between actors that produces significant differences¹⁴⁹. Like in sport, it is
- the combination of marginal gains which actually makes the difference ^{150,151}.
- 568

570

572 Box 1: Heavy metals and hazardous substances emissions

573 *Emissions during cement production*

574 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 575 576 can be a matter of concern in some countries, such as China¹⁵². However, when regulation and controls are implemented, no increased level of pollutants can be measured in the vicinity of cement plants ^{153–} 577 ¹⁵⁵. As heavy metals ¹⁵⁶ and other hazardous compounds are incorporated in the clinker and cement 578 kiln dust ^{60,157}, appropriate control devices and exhaust filters can mitigate heavy metal and hazardous 579 air emissions ^{60,157}. Furthermore, the high temperatures and the alkaline conditions in cement plants 580 allow for the full decomposition of the fuel's organic part ^{154,158,159}. 581

582

583 *Emissions at End of life. Leaching from SCMs*

Several industrial wastes can cause leaching of heavy metals when stockpiled. Yet their use as partial 584 585 cement replacement can often stabilize them due to the high pH of interstitial pore solution which precipitates heavy metals complexes¹⁶⁰. Such benefits have been shown to be less effective in the case 586 of poorly cured concrete ¹⁶¹. While currently used industrial wastes as partial replacement of cement 587 588 do not appear to have leaching issues in appropriately cured concrete, alternatives such as municipal 589 solid waste incineration (MSWI) bottom ash and non-ferrous slags are anticipated to have some 590 chloride and metal leaching issues¹⁶². 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 591 potentially harmful compounds ⁶⁶. For instance, pre-hydration or carbonation can be used to reduce 592 metal leaching from MSWI bottom ashes ^{163,164}. 593

595

594

596 Box 2: Water consumption

Water is one of the main constituents in concrete and its use can be as high as cement consumption 597 598 by mass ³¹. The direct water consumption used in cement products is equivalent to 400 L per capita 599 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 600 601 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}. 602 The energy-related water consumption depends on cement kiln type ¹⁵⁷ and the energy mixes which 603 can vary significantly depending on location ¹⁶⁶. On average, less than 50% of water consumption 604 associated with concrete production is linked to the cement ¹⁹ and water management strategies 605 606 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 607 5% of total water withdrawal ¹⁶⁷ and in most countries less than 1% of total renewable water resources 608 ¹⁹. However, water is a complex interwoven environmental issue. For example, a transition from river 609 aggregate to crushed aggregate in order to have sustainable management of mineral resources 610 611 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 612 health related issues as well as reducing the strength performance of concrete. There is therefore a 613 614 water-mineral resources nexus, and development of crushed gravel has to be combined with closed-615 loop water treatment.

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1004 **7 Glossary**

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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)

1009 Binder: Any material with binding property. It generally consists of cementitious material and water.1010

1011 **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
- 1020 C₃S, C₂S, C₃A and C₄AF: clinker mineral phases noted with cement chemical notation. C stands for CaO,
 1021 S for SiO₂, A Al₂O₃ and F for Fe₂O₃.
- 1023 Cement: a cement sets and hardens by chemical reaction with water and is capable of doing so under1024 water. (ASTM C125 R2015)
- 1026 **Cement Kiln Dust (CKD):** CKD are collected during the firing of raw materials during the clinker 1027 manufacturing process. CKD consists of four major components: unreacted raw feed, partially calcined 1028 feed and clinker dust, free lime, and enriched salts of alkali sulfates, halides, and other volatile 1029 compounds.
- 1031 Clinker: the active part of portland cement . It is a dark grey nodular material made by heating ground
 1032 limestone and clay at a temperature of about 1400 °C 1500 °C.
- 1034 **Concrete:** a composite material that consists essentially of a binding medium within which are 1035 embedded particles or fragments of aggregate; in hydraulic-cement concrete, the binder is formed 1036 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)
- 1040
 1041 Gravel: coarse aggregate resulting from natural disintegration and abrasion of rock or processing of
 1042 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)
- 1049
 1050 Ready Mix Concrete: concrete manufactured and delivered to a purchaser in a fresh state. (ASTM C94)
 1051
- 1052 Sand: fine aggregate resulting from natural disintegration and abrasion of rock or processing of1053 completely friable sandstone. (ASTM C125 R2018)
- 1054

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 1059 C125 R2015).

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1061 Figures and tables caption

1062 Figure 1: Examples of cement and concrete contribution to the global warming potential. For 1063 European building stock, the contribution of building materials is reduced for existing buildings as their 1064 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 1065 1066 average value from 230 buildings mainly from Europe (75%) and Asia (25%)¹⁴). At the building level, 1067 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 1068 Switzerland built between 2010 and 2015^{15,168}). For the production of one cubic meter of concrete the 1069 1070 main CO₂ emissions come from cement production followed by transport of raw materials (Values are the average of main concrete type made with 25% SCMs in Australia¹⁶⁹ and Switzerland¹⁷⁰). Finally, 1071 considering current clinker production efficiency and the replacement of 30% SCM in the final cement, 1072 1073 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). 1074

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1076 *Figure 2: Cement value chain.* From raw material extraction until the demolition of the building, 1077 numerous stakeholder are involved, but very seldom integrated (adapted from ^{24,111}).

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Figure 3: Carbonation profile through concrete. Measures and model adapted from ^{120,171}. Carbon
 uptake range adapted from ^{122,135}.

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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})
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 1086

1087 Table 2: Stakeholder description

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