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A two-fold strategy towards low-carbon concrete

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Abstract

Concrete is by a substantial margin the most widely used construction material. Projections indicate that the demand for concrete it will continue to increase to sustain the development of emerging economies. This paper presents a new perspective of low-carbon concrete by refocusing on the actual final product, highlighting the tremendous CO_2 saving opportunities of reducing the total paste volume of concrete while simultaneously using high performance, low-clinker cements in the so-called two-fold strategy (low clinker content, low paste volume concrete formulations). Different aspects of low paste volume concrete formulations are discussed based on a combination of published and new concrete performance data, showing the potential for CO_2 savings of the strategy and the technical opportunities to retain the robustness and reliability that make concrete such a versatile and widely used material. Chemical admixtures play a crucial role in reaching those objectives, as they enable to reduce the cement content while retaining the needed workability (slump and slump retention) for each application. The key issues relating to using those admixtures in low carbon concrete are highlighted.

Keywords: Sustainability; Paste volume; Clinker factor; Superplasticizers; Packing density

1 Introduction

1.1 The climate emergency, cement and concrete

Portland cement (PC), a combination of clinker (95%) and gypsum (5%), is the binder used to produce concrete. Concrete is the second most used substance by mankind after water [1], and it is the largest manufactured product on a mass basis. Due to this enormous utilization, PC is responsible for about 8% of manmade CO_2 emissions [2]. Consequently, a key objective when addressing the environmental footprint of the construction sector boils down to addressing CO_2 emissions relating to PC, but simultaneously this endeavor has proven to not be trivial. In essence, the problem converges to manufacturing less PC, which can be achieved by increasing the life-service of concrete, producing PC lean concrete, or replacing concrete by other materials.

The latter option has only limited scope because concrete is the only material on Earth that can be produced in the volume required to meet the current and future demand of construction [3]. Alternative materials should be part of the global solution but can only replace a rather small fraction of the concrete needed. Moreover, concrete is not only widely available, but also economical, locally produced, easy and safe to use, as well as versatile and durable. It is also the most efficient choice from an embodied CO_2 and energy point of view [4]. Thus, going back to the above-mentioned options, the overall high environmental footprint of the industry is associated with the quantity of PC consumed.

Using less concrete, through better structural design targeting shape efficiency and avoiding unnecessary overdesign and/or increasing life service are important levers but are beyond the scope of this paper. Rather, the paper focuses on material scale solutions, first summarizing options to reduce PC (clinker) use at the cement manufacturing level and second by using less cement in concrete. The first option largely relies on blended cements that incorporate mineral additions (supplementary cementitious materials, SCMs) in combination with clinker, enable substantial CO_2 reductions compared to traditional PC [1,5]. The second involves decreasing the total cement paste content. It builds upon existing concepts but is often marred in misconceptions. A main objective of this article is to bring some clarity on this subject to highlight the potential of the concrete industry to reduce its carbon footprint using low paste volume concrete more extensively.

1.2 Blended cements and the pursuit to reduce the clinker factor

Concrete is essentially a multi-phase material in which solid inclusions (fine and coarse aggregates, about 70% of the total volume in typical formulations) are held together in the hardened state by an inorganic binder made of hydrated cement paste (comprised of hydration products, anhydrous

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cementitious materials, pore solution and pores, about 30% of the total volume, Figure 1a). Despite concrete being mostly aggregates from a volumetric point of view, the overwhelming contributor to its CO_2 footprint is PC, Figure 1b.

The Global Cement and Concrete Association (GCCA) established a roadmap to achieve a net-zero industry by 2050 [3]. The roadmap identifies the carbon reduction needed from different stakeholders within the value chain of concrete to materialize this goal, Figure 2. It also clearly establishes an important role of carbon capture, utilization and storage (CCUS) towards this goal. However, these technologies are still scarce, complex (hence not always feasible) and expensive [6]. Moreover, the urgency to address the climate crisis calls for measures that can be deployed in the short term to maximize the CO_2 net present value of such technological interventions [7,8]. Consequently, the most effective way to reduce the CO_2 footprint fast and effectively is to minimize the clinker content in cement [1].

During the last two decades, an increasing adoption of blended cements has been observed [9]. They are widely used nowadays, being more common than PC in many regions of the world. Unfortunately, the availability of commonly used SCMs such as fly ash (by-product of burning coal) and slag (by-product of iron production in blast furnaces) is limited and, especially in the case of fly ash [10] and suitable slags, expected to rapidly decrease in the forthcoming decades [11]. The limited access to traditional SCMs largely explains the stagnation of the replacement level of clinker in blended cements since 2010 at around 25-30% substitution [1,12]. However, several countries are still using very low substitution levels and have opportunities to start by simply using limestone that does not pose many challenges up to substitution levels of around 15%. This is for example happening currently in the USA, switching from PC (Type I/II in ASTM C150) to almost exclusively Portland limestone cements (Type IL in ASTM C595), roughly saving 10% of CO_2 relative to the previous situation where PC was dominant. Beyond this, the combined use of limestone and (kaolinitic) clays in the form of limestone calcined clay cements (LC^3) offers a breakthrough opportunity as these materials are available in virtually unlimited quantities, and can enable a significant worldwide reduction in PC content [1,5,13,14].

LC^3 are a family of blended cements that incorporate limestone and calcined kaolinitic clays replacing 50% (or more) of clinker. They achieve similar strength to conventional PC from 7 days onwards [15,16]. Overall, LC^3 can save between 30 to 40% of the CO_2 emissions compared to PC [17]. Substantial progress has been made in the characterization and optimization of LC^3 [5,13,15,18,19], including the understanding of raw material production [20–22], hydration and blend design [16,19,23–26], microstructural development [27–30] and mechanical properties [15,29,31].

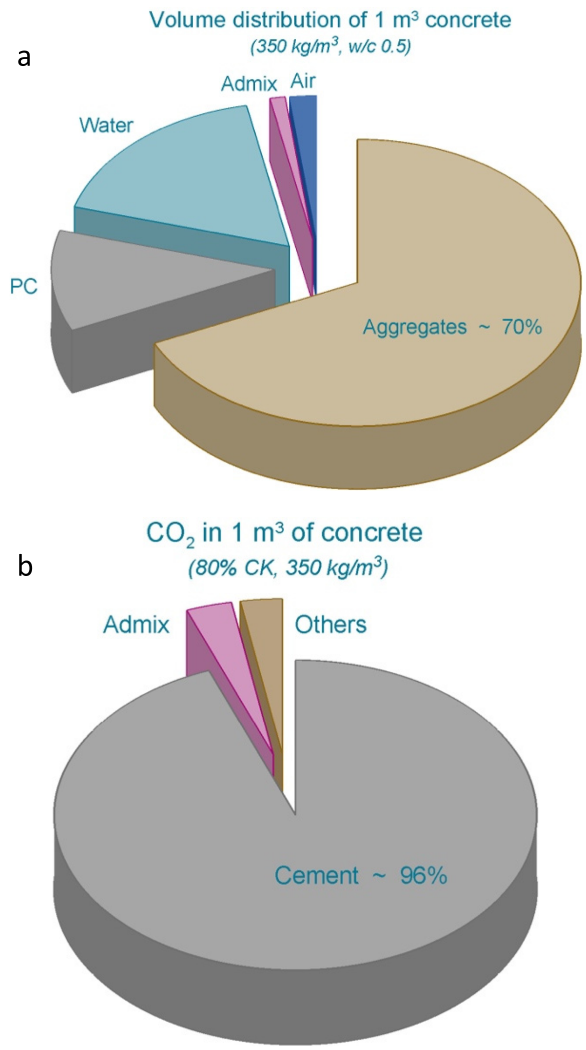

Figure 1. Typical volume distribution of 1 cubic meter of concrete (a) and mass distribution of CO2 footprint in 1 cubic meter of concrete (assuming 80% clinker factor) (b).

1.3 Moving the spotlight back to concrete

Low-carbon cements like LC^3 are steppingstones towards the widespread production of low-carbon concrete. However, they are only useful if they can deliver or improve the necessary performance of concrete in each service environment in which they are used. Therefore, while quantifying the CO_2 content per unit mass of cement is a useful optimization tool to formulate blended cements and assessing the efficiency of a given cement production process, it also masks relevant information, such as the actual amount of cement needed to produce concrete with a given set of properties. This is a more complex question that does not depend solely on the cement used, but also on the slump requirement (and use of admixtures), durability specifications, the aggregate grading and the desired use for the material (placing method, construction pace constraints, etc.). Furthermore, concrete construction is fundamentally a volumetric process, where designers define a set of measures that establish the volume of concrete required to materialize a given element.

Consequently, to maximize the CO_2 saving potential, the whole value chain of concrete construction should be considered [32]: cement, concrete and the structure, as shown schematically in Figure 2. Different strategies/technologies can be implemented depending on the stage of the value chain, implying the need for a variety of benchmarking metrics to quantify the environmental footprint according to the various stakeholders (cement producer, concrete producer, designer, contractor, owner and policy maker).

From a policy point of view, the trend in Europe so far has been to place thresholds at the structure level (CO_{2eq}/m^2 surface, e.g., the RE2020 in France). This in turn produced constraints at the material level in terms of specifications from the designers implementing that policy (i.e., stakeholders translated the policy thresholds into different benchmarking metrics, green arrow flow in Figure 2). This is a positive example and likely a future reference on where thresholds should be placed to effectively permeate through the value chain.

Despite the high potential for carbon reductions by optimizing structures, the pace at which it can be done is affected by codes and designer practice. For example, serviceability considerations (deformation limits, insulating capacity, confinement requirements, etc.) can often impose more stringent limitations on design than structural capacity does. While onboarding designers in the sustainable endeavor is necessary, addressing the material (concrete) is likely an intervention that can be deployed more quickly on a global scale. Both approaches should be seen as complimentary, and it certainly should be understood that reducing the carbon footprint of construction materials will sooner or later reach an asymptotic limit. However, the ability to adopt low-carbon concrete rapidly means that it has an essential role to play in the current climate emergency.

2 Low-carbon concrete

2.1 What is low-carbon concrete?

The terms low-carbon, sustainable, green or environmentally friendly concrete are commonly used today by producers, owners, specifiers, designers and regulators, often referring to materials with substantially different qualities in terms of environmental footprint [33]. There is no clear definition on what “low carbon” means (reference, thresholds) and often the discussion ends in a matter of perception based on a specific market, leading to the definition of different, local descriptions of low-carbon concrete [3,11,12,34]. As an example of the observed scatter in the current situation, the average CO_2 footprint in 2019 for ready-mix concrete dispatches in Switzerland was 216 kg CO_{2eq}/m^3 concrete for a typical strength of 30 MPa [35]. In contrast, the average concrete footprint in the Great Lakes area (USA) is reported to be within 280-340 kg CO_{2eq}/m^3 concrete for strengths between 20-30 MPa [36], thus between 30 and 57% higher.

Almost certainly, it will not be possible to establish a universal definition that effectively embraces the inherent local dependencies of the concrete industry (different availability of raw materials, industrialization degree of the concrete industry, possibility to access new technologies owing to costs, target properties/use conditions of the concrete). Rather, the definition of low-carbon concrete should consider the particularities of the individual markets and socio-economical aspects. Nevertheless, some strategies to design low-carbon concrete mixtures can be outlined and then applied locally, as discussed below.

2.2 Total paste volume in concrete

It is widely accepted that modern concrete contains more cement than is needed. The reasons behind this are diverse: some national standards prescribe the minimum binder content for different durability exposure classes such as the European standard EN-206 [37] (overall minimum cement content of 280 kg/m^3, however there is a large dispersion among member countries as the standard is not harmonized [38]), and they can also be relatively arbitrarily specified on a project basis. Importantly, there also is often a lack of availability of aggregates with sufficient size fractions to improve packing (i.e., minimize the volume of voids [39–41]) and finally there is tradition. The minimum paste contents in some standards such as EN-206 were originally specified to ensure workability when chemical admixtures, particularly modern superplasticizers such as PCEs, were not yet available. With such admixtures now broadly available and being used commercially, the inherited limitation on binder content is no longer justified, and standards must evolve to reflect the current state of technology.

The paste volume (cement plus water) should be sufficient to fill the voids between the aggregates, leading to values of around 180-200 lt./m^3 (18-20%) for well graded (highly packed) aggregate size distributions [42]. Considering that PC accounts for most of the embodied CO_2 in concrete (Figure 1b), reducing the total paste volume in concrete constitutes a second avenue (in addition to clinker factor reduction) to reduce the overall amount of clinker in concrete. This simultaneous consideration of these well-known approaches provides a *two-fold strategy* to approach the development of low-carbon concrete, which sustains the ultra-green concrete (UGC) initiative led by the author [a] and schematically presented in Figure 3. The essence, technical challenges and environmental benefit potential of such an approach are covered in the next sections of this article.

[a] The Ultra-Green Concrete (UGC) project is supported by the Swiss National Science Foundation (SNSF) and is led by Dr. Zunino at ETH Zürich (https://ultragreenconcrete.com).

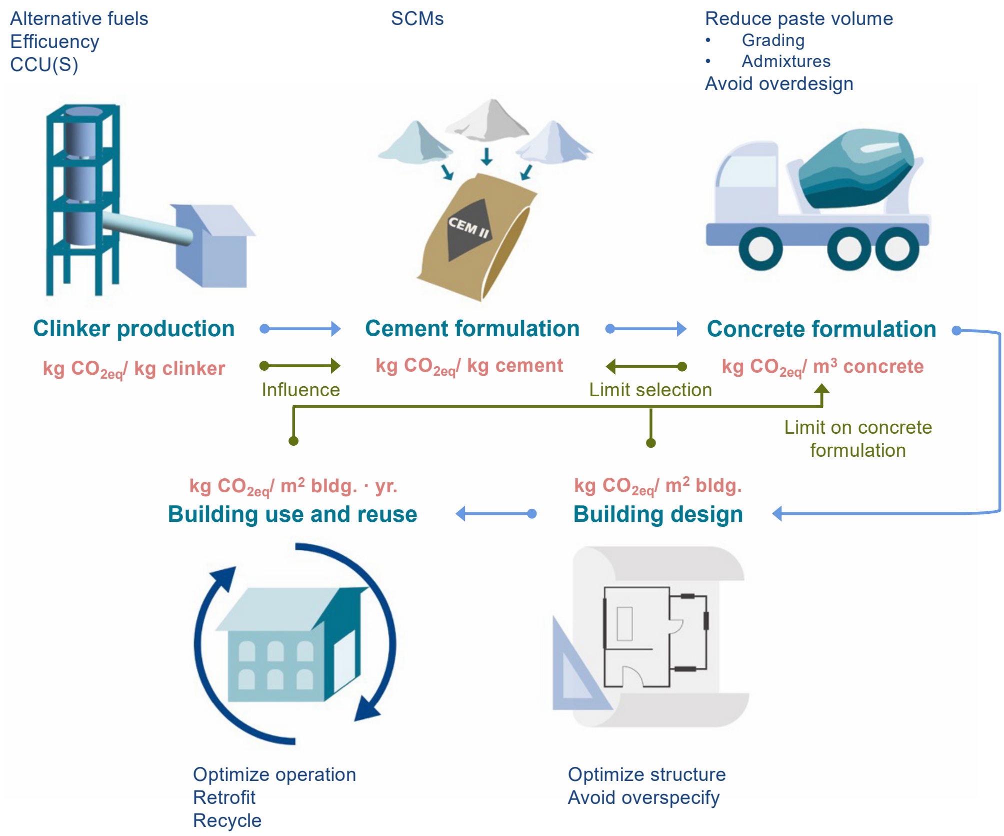

Figure 2. Concrete construction value chain, highlighting possible interventions at each level (in blue) to reduce embodied CO2. Different environmental metrics are shown below in red and are related to the stakeholders they address. The interaction between policy (at the structure level) and metrics is represented in green.

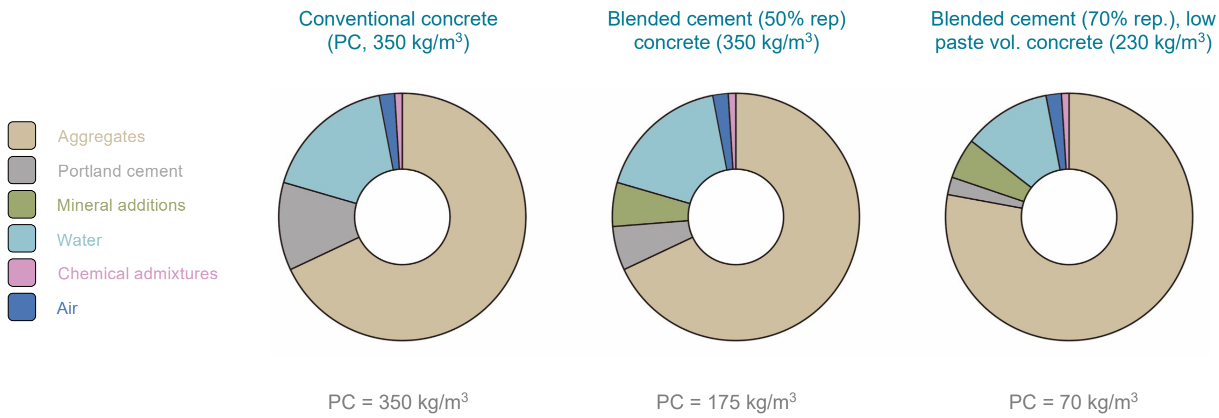

Figure 3. Scheme showing the volume composition of 1 cubic meter of concrete when the two-fold strategy is applied to the mixture design. The equivalent mass of PC per cubic meter is shown below for reference. The aggregate fraction considers sand plus coarse aggregates, while mineral additions could include SCMs and/or fillers.

2.3 Low paste volume concrete formulations: myths and realities

When a reduction of the total paste volume is considered for a given concrete formulation, a set of technical challenges arise that should be properly addressed to make this approach feasible. In addition, there is also a set of instilled misconceptions and myths surrounding concrete formulations with low paste volume that hinder practitioners from considering such paste volume cuts. The presumed proportionality between cement content (i.e., paste volume at fixed w/cm) and strength is likely the most common one. As shown in Figure 4 (adapted from the work of Hermida et al., [43]) compressive strength of concrete does not decrease, but rather increases (even if only slightly) with the reduction in total paste volume. With the exception of lightweight aggregates, cement paste is the weakest phase in concrete [44,45] and thus, failure occurs through the paste or the interfacial transition zone (ITZ) between paste and aggregates [46,47]. In the case of modern, low-carbon cements such as LC^3 the behavior follows the same trend (Figure 4, data from [48]). Another interesting observation is that two of the LC^3 systems (paste volume below 150 lt./m^3) show a significantly lower relative strength. In this case, the amount of paste was not sufficient to fill the voids between the aggregate matrix, compromising compactability and ultimately strength.

The current situation for concrete in terms of formulation and environmental footprint is better captured from a larger dataset. Hafez et al. [55] combined a database of concrete performance data (publicly available) which is extended here with unpublished original data (LC^3-based concrete formulations), reaching 1800 observations. The statistical metrics of the extended dataset are reported in the right side of Figure 5. Thereby, the CO_2 footprint (kg CO_{2eq}/m^3) of each concrete was computed based on the formulation data along with the CO_2 intensity factors shown in the left side of Figure 5. The average value in the dataset is 254 kg CO_{2eq}/m^3, with a standard deviation of 96.7 kg CO_{2eq}/m^3.

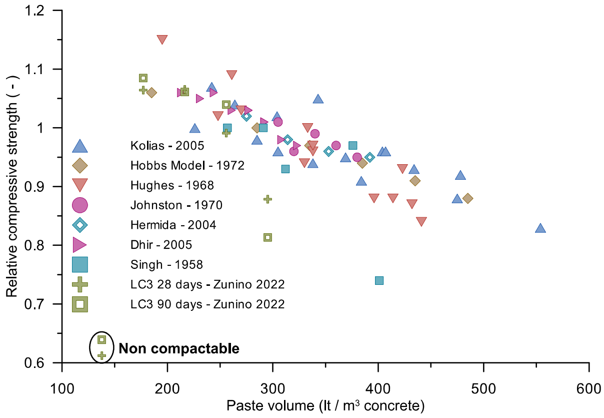

Figure 4. Relationship between relative compressive strength (normalized by linear regression to a paste content of 300 lt./m^3 for any given binder at fixed w/cm) and concrete paste volume. Adapted from [43], with data from [43,48–54].

In addition, the well-known dataset from Daminelli et al. [56] (1600 points) was also incorporated into the analysis, providing further support to the points discussed below. The two datasets (Hafez et al. + LC^3 and Daminelli et al.) were not combined as they have statistically different means (305 kg CO_{2eq}/m^3 in [56], at a confidence interval (α) of 5%). The difference is mainly explained by the lower average clinker factor in the dataset of Hafez et al. (0.68 vs 0.78 in [56], with the difference being statistically significant with $\alpha = 5\%$) rather than a lower average total paste volume (312 lt./m^3 vs 314 lt./m^3 [56], statistically equal with $\alpha = 5\%$).

	Concrete constituent	CO_{2eq} (A1-A3)	Ref.
Cementitious materials	Clinker	0.85	[17]
	Gypsum	0.0	[17]
	Limestone	0.00219	[17]
	Calcined clay	0.127	[17]
	Fly Ash	0.0	[17]
	Blast furnace slag	0.06021	[57]
	Natural pozzolan	0.00219	A
Concrete	Aggregates	0.00419	[17]
	Water	0.000658	[17]
	Superplasticizer	1.53	[58]

A: CO_2 intensity assumed equal to limestone (i.e., involving extraction and transport from quarry and grinding).

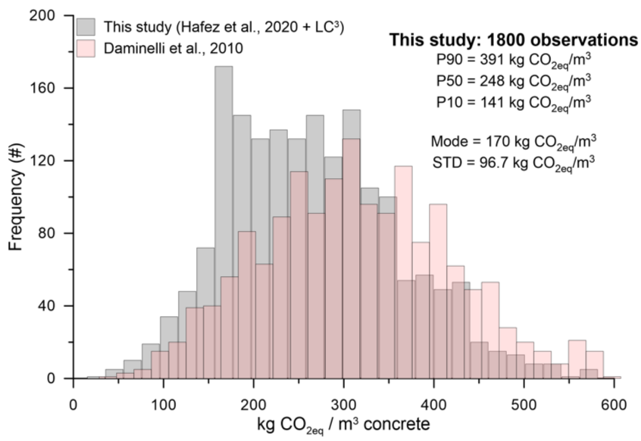

Figure 5. CO_2 intensity factors (left) used to compute the CO_2 footprint of the concrete formulations within the dataset, and statistical metrics (right) of the datasets considered.

Figure 6a shows the relationship between compressive strength at 28 days and the total cementitious material content per cubic meter. As seen, there is no clear correlation between these two variables, particularly for strengths below 60 MPa. Taking as an example 40 MPa of strength, there is more than 400 kg/m^3 of scatter difference in total cementitious material content within the dataset analyzed, highlighting the potential for optimization. Similarly, Figure 6b shows a comparison between strength at 28 days and CO_2 footprint of concrete (expressed as kg CO_{2eq}/m^3 concrete). For the same reference strength (40 MPa), the gap in CO_2 content is more than 300 kg CO_{2eq}/m^3 concrete.

It should be noted that this analysis does not consider possible differences in usability of these formulations (slump, compactability, durability class, etc.). Nevertheless, it provides an overview of the dispersion encountered within the literature and the huge opportunities for mixture design optimization and CO_2 savings. Another aspect that should be considered is that the data corresponds mainly to laboratory mixtures. Additional insights could certainly be gathered with a similar treatment of ready-mix concrete formulations.

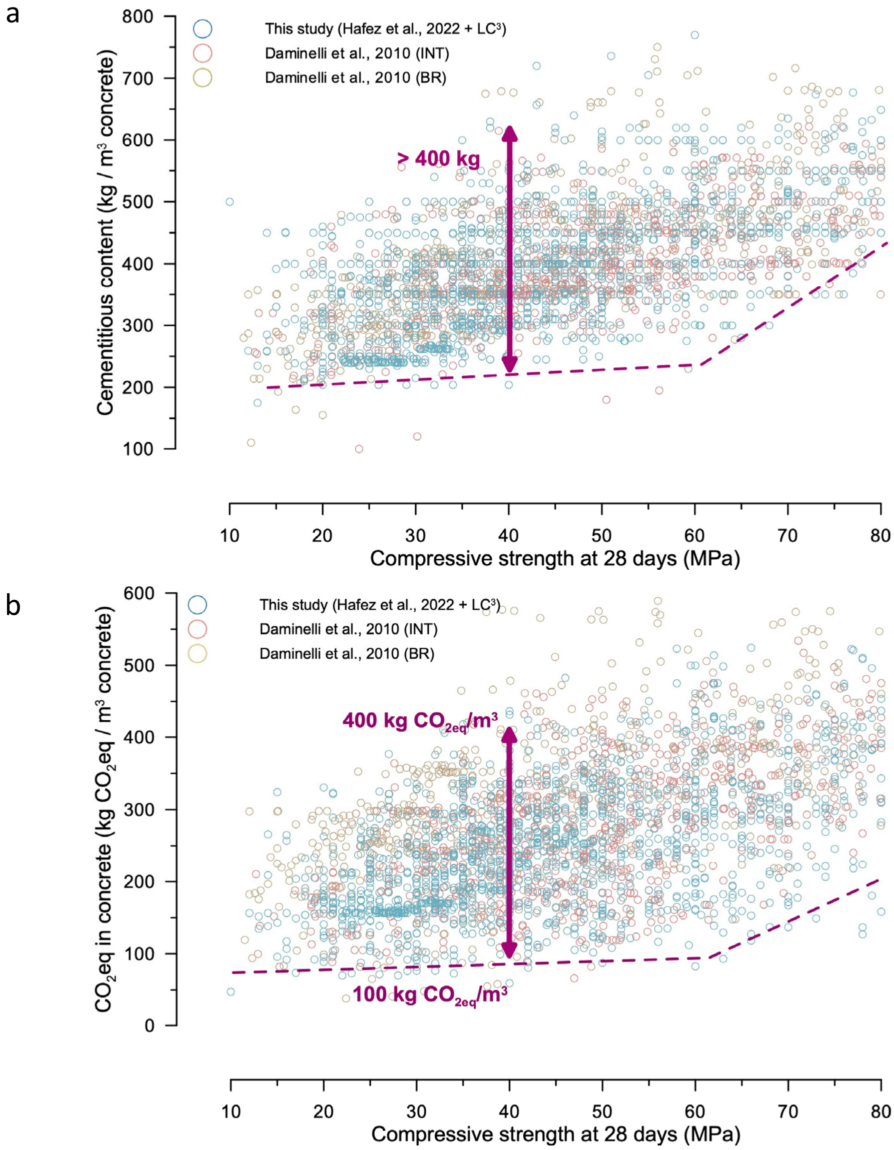

Figure 6. Compressive strength at 28 days versus binder (cementitious material) content per cubic meter (a) and (b) versus CO_2 footprint (expressed as kg CO_{2eq}/m^3 concrete) within the dataset described in Figure 5. The dataset from Daminelli et al. is shown as reference [56].

In terms of durability, the performance of low paste volume concrete formulations is debated, and further research is needed to clearly establish performance against different exposure conditions. The question is non-trivial for low paste volume concrete formulations, as on one hand transport (ions, liquid, gas) occurs through the cement paste matrix (volume is reduced) while on the other the amount of more porous ITZ (in general) increases [59–61]. Previous studies showed that a decrease in paste volume was associated with a decrease in creep, shrinkage and shrinkage-induced cracking [43,62], water absorption capacity and expansion by sulfate attack [43]. The carbonation depth (at constant w/cm) was observed to be independent of the paste volume. Leemann et al. observed a proportionality between the carbonation coefficient with the buffer capacity per unit volume of paste [63]. However, no systematic correlation between paste volume and carbonation coefficient for a given cement and w/cm was observed [64]. Regarding chloride attack, previous studies seem to indicate that resistance to chloride penetration reduces with a decrease in paste volume [65,66]. However, this change is not sufficient to worsen the classification of the concrete according to the predominantly employed RCPT test [67].

Other attributes often believed to be problematic with low paste volume concrete formulations are the additional technical challenges involved in their production at industrial scale and their robustness in terms of workability. Regarding industrial production, decades of experience in markets where lower cement contents are allowed show that, under an adequate ready-mix plant formulation control, these mixtures can be repeatability and reliability produced at industrial scale. As an example, in Chile the minimum cement content for reinforced concrete is 240 kg/m^3 [68]. Figure 7 shows a comparison of ready-mixed concrete from a single plant with different total cement contents (in all cases, the cement used contains natural pozzolans, with 73% clinker factor) and strengths. As observed, the variability between repeated batching of concrete with high or low cement content is comparable.

In terms of workability, reducing the paste volume brings the system closer to the maximum packing fraction of the granular skeleton (ϕ_m). While this may be beneficial in terms of segregation, it increases the sensitivity of flow to variations in water dosage and aggregate content/moisture content [69]. This can be mitigated by a better grading of the aggregate skeleton. Indeed, by raising the maximum packing fraction, the aggregate volume fraction at which workability diverges and robustness is lost also increases (Figure 8). In combination with adequate use of chemical admixtures, such strategies offer simple means of reducing paste volume without compromising workability. Additional work is needed to assess other properties of low paste volume concrete formulations such as pumpability and self-compacting applications, where the reduced quantity of fines might pose additional challenges. The incorporation of fine fillers could offer an effective strategy to tackle this issue while maintaining the clinker content low.

In many cases, particularly in emerging economies such as India and Latin America, bagged cement use dominates the concrete market [12]. In these cases, it is common to find unjustifiably high dosages of cement used to produce low to medium performance concrete formulations. Here, significant CO_2 savings can be achieved just by adopting well-known technologies like ready-mix concrete to enable better raw material control and batching accuracy/reproducibility.

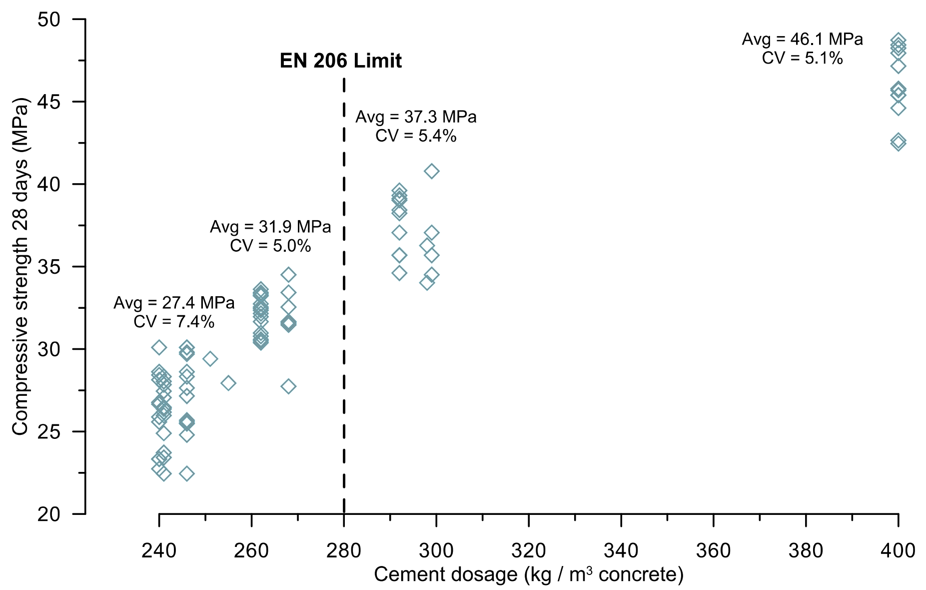

Figure 7. Variability of ready-mixed concrete from a single plant with different cement contents, conforming to the Chilean concrete standard NCh 170 [68]. In all cases, the same type of cement was used, target slump values 10-12 cm (except mixture with 400 kg/m^3, which is 20+ cm), considering a defective fraction of 10% and with a maximum aggregate size of 20 mm.

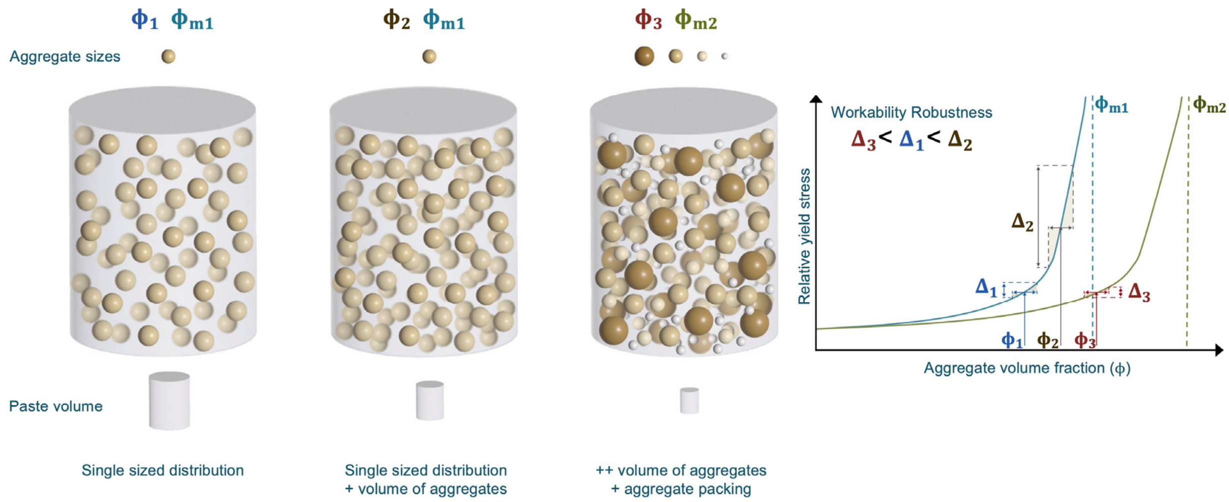

Figure 8. Influence of an increase in solid volume fraction of aggregates (ϕ_i) and maximum packing fraction (ϕ_{mi}) on the relative yield stress, represented using the Chateau and Ovarlez model [70]. The same variation in ϕ_i (emulating variability expected within the proportioning process) is imposed on all systems, and the change on relative yield stress (Δ_i, representing workability robustness) is shown.

2.4 Chemical admixtures: the enablers of low-carbon concrete

Arguably, the biggest challenge associated with reducing the paste volume in concrete is to retain sufficient workability as less paste between aggregates is available. If in addition, highly substituted blended cements are used (which normally exhibit higher specific surface areas than PC [30,71–73]) the situation becomes particularly challenging. Superplasticizers (SP) are the most used rheology modifiers to control rheology of concrete, both in terms of flow and flow retention. They are polymeric dispersants that reduce yield stress of pastes and particulate suspensions at a constant solids content [74,75], and in general modern SPs are based on polycarboxylate-ethers (PCEs) [76,77]. Their effectiveness in concrete operates through their impact on the paste rheology.

The underlying mechanisms of action of such admixtures are complex, involving specific interactions between admixtures and cement [78–82] as well as among admixtures themselves [83,84]. In highly substituted blended cements, the interactions of these compounds with the surface of the SCMs are of particular interest as they become dominant in highly substituted systems (i.e., where most of the specific surface area corresponds to SCMs), Figure 9. In addition, PCEs can delay cement hydration by modifying the rate of alite dissolution [85], an undesired effect that can compromise their effectiveness in low-carbon concrete applications. The technical feasibility of the *two-fold strategy* requires new formulations of SPs that provide sufficient flow and flow retention without excessive retardation of hydration. Preliminary trials at laboratory scale with LC^3 (230 kg/m^3, w/cm 0.43) showed that sufficient rheology can nevertheless be obtained with current commercial admixtures [48]. With tailor-made formulations, under development by all major admixture producers, the situation most likely will improve, opening the gates for cements with even higher substitution levels.

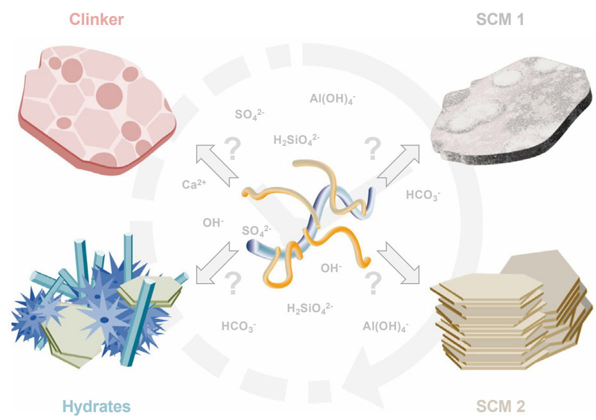

Figure 9. Schematic representation of the complex adsorption (and competitive adsorption) problem that new SP formulations need to tackle to provide enough flow/flow retention without excessive retardation. The adsorption of admixtures on surfaces and the hydration reactions are dynamic processes. Adapted from [86].

2.5 Environmental benefits of the two-fold strategy

Table 1 shows the environmental footprint of 1 cubic meter of concrete (kg CO_{2eq}/m^3 concrete) as a function of clinker content and total cementitious material content (proportional to paste volume at constant w/cm of 0.5). Clinker is replaced by a combination of calcined clay and limestone in a 2-to-1 mass proportion (CO_2 intensities in Figure 5). To approach a realistic situation, the reduction of clinker or/and paste volume are assumed to yield an increase in water demand, and thus the dosage of SP increases (values interpolated from the experimental observations shown in [48,87]). As seen, both elements of the *two-fold strategy* offer a substantial potential to reduce the carbon footprint of concrete. For the conditions analyzed, reducing clinker and/or binder content always yields a reduction in CO_2, despite the increase in SP dosage. Furthermore, this highlights that in regions where highly substituted blended cements are still not available, a substantial move towards low-carbon concrete can be achieved solely by optimizing the concrete mixture design. Certainly, the best outcome is seen when both components of the strategy are applied simultaneously. Nevertheless, they are independent and could be implemented in a decoupled manner to adapt to each market scenario and to the intended application of the material.

Table 1. Total CO_{2eq} in kg per cubic meter of concrete as function of clinker factor and binder content (constant w/cm of 0.5). Clinker reduction is assuming a replacement by a combination of calcined clay and limestone in a 2-to-1 mass proportion. Reduction in clinker and/or binder content presumes an increase in SP dosage, leading to a similar initial slump value in all cases.

SP (%wt. cementitious)			**Binder content (kg cementitious/m³)**					
			0.0	*0.15*	*0.30*	*0.45*	*0.60*	*0.75*
			350	**325**	**300**	**275**	**250**	**230**
0.0	**Clinker Factor (% weight of cementitious)**	**95**	291	271	252	233	213	197
0.1		**90**	278	259	241	223	204	189
0.2		**85**	265	248	230	212	195	181
0.3		**80**	252	236	219	202	186	172
0.4		**75**	239	224	208	192	176	164
0.5		**70**	226	212	197	182	167	155
0.6		**65**	214	200	186	172	158	147
0.7		**50**	174	163	152	141	130	121
0.8		**45**	161	151	141	131	120	112
0.9		**40**	148	139	130	121	111	104
1.0		**35**	135	127	119	111	102	95
1.1		**30**	123	115	108	101	93	87
1.2		**25**	110	103	97	90	84	78
1.3		**20**	97	91	86	80	75	70
1.4		**15**	84	80	75	71	66	62

Figure 10 shows the CO_2 footprint of the concrete formulations contained in the datasets normalized by their compressive strength at 28 days. This approach to data representation introduced in [56] enables one to better account for the higher embodied CO_2 commonly seen in higher strength mixtures, but that can in turn yield to optimized (lower material volume) structural elements for the same application. Concrete formulations based on LC^3 technology are shown in green. In addition, the ready-mix concretes shown in Figure 7 are plotted in blue. Finally, the environmental footprints from ECOPact[b] formulations from Holcim Germany are shown (purple and teal) to highlight a recent industrial move towards sustainability.

In general, the data obtained from industrial producers is encouraging (ECOPact formulations and the ready-mix concretes shown in Figure 7), as they are below the average of the dataset considered (254 kg CO_{2eq}/m^3 concrete). LC^3-based concrete formulation seem to break through the general trend, as they allow one to produce concrete with a significantly higher strength and with a lower embodied CO_2 compared to other technologies, given the ability of LC^3 to match PC strength with 50% clinker substitution [13,16,26], which is not the case of other common SCMs [88]. As CEM II/C – M (Q-LL) (LC^3 in the European EN 197-5 standard) becomes readily available in Europe and other regions, it is highly likely that the environmental performance of ready-mix concrete will improve. Still, and as shown in Table 1, minimizing the paste volume offers an additional, substantial opportunity to lower CO_2 independent of the cement type(s) available.

3 Challenges and opportunities for low-carbon concrete

3.1 Standards and specifications: from deemed-to-satisfy approaches to performance-based validation.

As previously discussed, some standards such as the EN 206 set prescriptive limits for the minimum cement content of concrete depending on durability exposure classes [37]. As shown in this paper, there is a tremendous opportunity to reduce CO_2 (and likely cost) of concrete mixtures by shifting towards lower paste volumes. The issue with prescriptive standards is that performance is verified based on deemed-to-satisfy criteria that rely heavily on past experience, sometimes with different materials from the ones concerned that exhibit a significantly different performance and additionally not reflecting the possibilities offered by modern commercial chemical admixtures. It is therefore imperative to shift towards performance-based standards and specifications (or at least, allow an alternative performance verification) to unlock the full potential of the *two-fold strategy*. In this regard, the adoption of exposure resistance classes (ERC) in Europe will enable one to qualify new concrete formulations based on objective metrics established around well-known testing methods. Attention should be placed on the requirements of (often long-lasting and expensive) experimental testing required to establish compliance of products with ERCs, which in this case fall within the scope of the concrete producer. The need for additional testing to certify a product might create an incentive to reduce the number of formulations offered and shift towards a *one size fits all* approach, leading to overdesign and an unnecessary increase in CO_2.

[b] ECOPact and ECOPact Prime are trademarks of HOLCIM. Data was compiled from publicly available Environmental Product Declarations (EPDs) that can be accessed at https://ibu-epd.com/en/published-epds/.

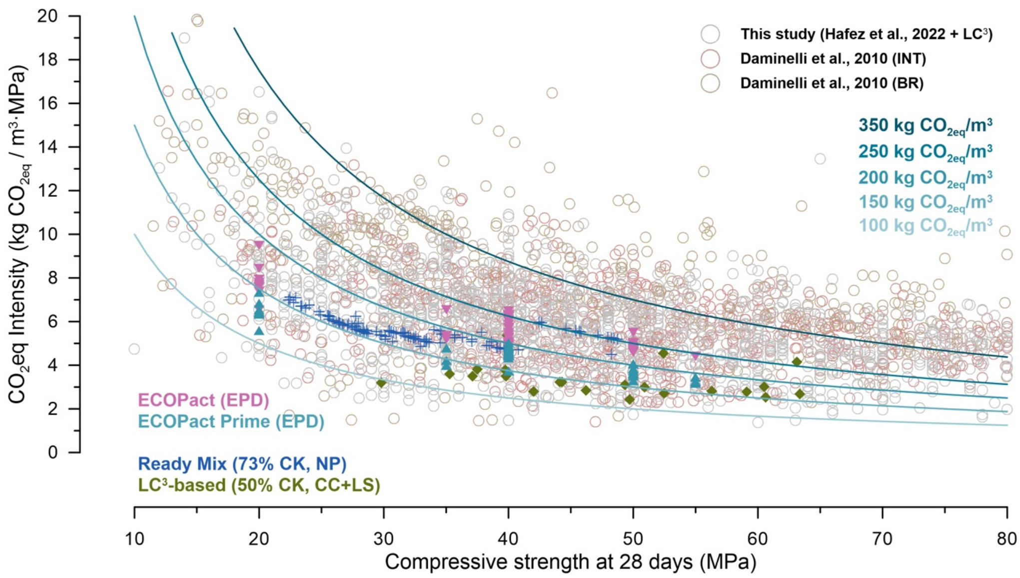

Figure 10. CO_{2eq} intensity (CO_{2eq} per unit volume normalized by compressive strength) of the mixtures contained in the datasets analyzed, based on the factors shown in Figure 5. All values correspond to real strength tests. Data for ECOPact are taken from EPDs (characteristic strength and CO_2 footprint reported).

3.2 Reduce and reuse with an environmental performance perspective

Reduce is a key goal within the low-carbon concrete construction discussion: reduce clinker in cement, reduce paste volume in concrete, reduce the amount of concrete in structures. In addition, reuse (retrofit, repurpose, recycle) is another concern that poses additional challenges in terms of design and durability. These concepts require a case-by-case examination and their influence on the desired output, particularly the environmental footprint, should be quantified and critically analyzed. For example, reducing the clinker factor in cement is commonly deemed as a positive strategy to reduce CO_2 *per se*. However, this only accomplishes its goal if the performance of the blended cement is sufficient to produce concrete with a target set of properties without the need to increase the total cementitious material content because of a reduction in w/cm. In some cases, this may lead to a higher CO_2 content per cubic meter of concrete as compared to formulations using higher clinker cements but with better performance, Table 1.

Design optimization and better technical specifications allowing the use of low-carbon materials is another highly needed step within the construction value chain. Even within current design codes, there is a tremendous variability in the CO_2/m^2 of building for different materiality of buildings [89,90]. Additive manufacturing (often referred as 3D concrete printing, 3DCP) has emerged as a new technology that could enable shape efficient elements with a substantial reduction in material use. As pointed out by Flatt and Wangler [91], shape efficiency is the only pathway through which 3DCP could contribute to sustainability, as the materials commonly used in extrusion-based applications have significantly higher (450-650 lt./m^3) cement paste volumes and consequently a higher CO_2 content due to the lack of coarse aggregates.

In perspective, Figure 11 shows the CO_2 content distribution of the dataset compiled in this paper and the CO_2 footprint of 20 3DCP formulations from the literature [91–99]. The average value for 3DCP, the dataset and the LC^3-based concretes are shown as a reference. The average CO_2 footprint of 3DCP mixtures is 1.75 times higher than the average of the dataset of conventional concretes, and 2.89 times higher than the average of the LC^3-based concretes. While limited to the extent of the data available for analysis, these values provide a guideline on the amount of material savings (e.g., through shape efficiency) that would be required to match or surpass the environmental efficiency of conventional concrete. This is a technology that is still in its infancy and improvements may be expected in the future, as suggested by some of the 3DCP mixtures that exhibit CO_2 footprints significantly lower [100] than the average.

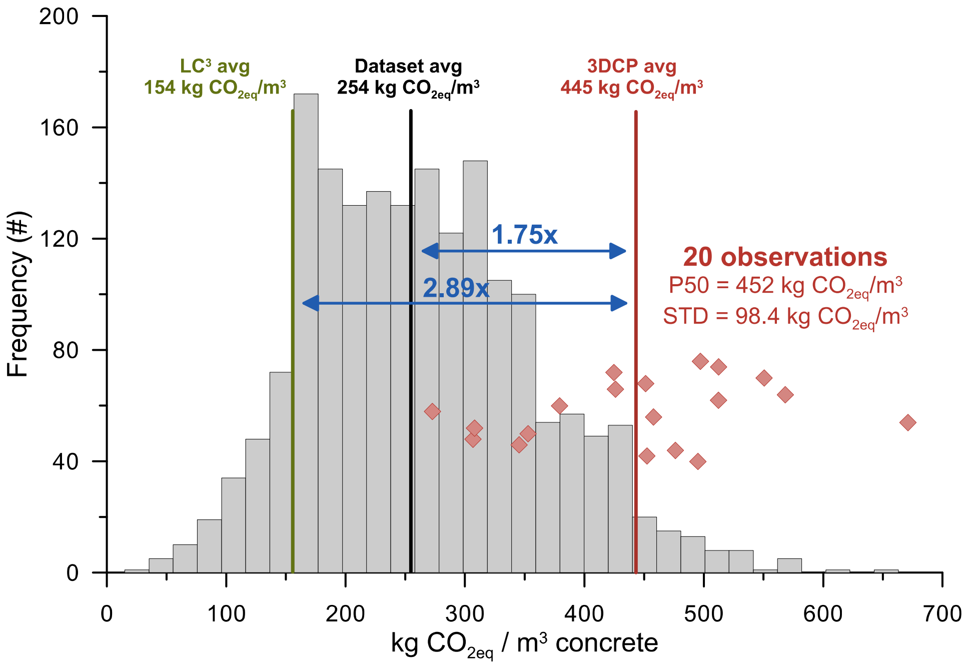

Figure 11. Comparison of CO_{2eq} footprint of conventional concrete and mixtures designed for 3DCP by extrusion (only the material contribution is accounted for). The averages of 3DCP, the main concrete dataset and LC³-based concretes are shown.

4 Concluding remarks and perspectives

In this article, the potential of a combined reduction in clinker content and paste volume (*two-fold strategy*) to drastically reduce the embodied CO_2 content of concrete was presented. The pros, cons and open challenges surrounding the concept of paste volume reduction were critically discussed. The possibility of implementing each component of the two-fold strategy independently provides flexibility to adapt low-carbon concrete formulations to each individual market. Availability of high-performance blended cements and the transition of standards towards performance-based specifications are critical steps that the industry and policy makers should address urgently. The reduction of total paste volume to reduce the carbon footprint of concrete is yet to be explored systematically with formulations tailored for different applications (open time, compactability, pumpability, different aggregate types and shapes among others). The perspectives are encouraging and the potential for CO_2 savings is enormous.

By placing the benchmarking for environmental footprint at the concrete level, the performance of blended cements, the contribution of mixture design optimization and the critical role of chemical admixtures can be properly accounted for. Moreover, a CO_2 per unit volume indicator could be easily incorporated by designers to estimate the CO_2 footprint per unit surface of a specific structure or determine compliance with regulations. Ultimately, the focus should progress higher up the value chain to the structure level. While the environmental footprint of concrete is only part of the puzzle within the life cycle of a building, it is probably the one where a decisive shift can be achieved faster and more effectively deployed at the industrial level, provided that the relevant standards can swiftly catchup. Simultaneously and looking at the mid-to-long term, strategies that might require a longer timescale for realization (design optimization, shape efficiency, CCUS among others) should continue to be developed to ultimately arrive to a balanced portfolio of measures that can be combined and adapted to fulfill the environmental and technical requirements of construction.

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