

# RILEM TC CUC: Carbon dioxide uptake by concrete during and after service life – Opening Letter

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## Abstract

The ability of cement to chemically bind CO<sub>2</sub> in a carbonation reaction enables the material to act as a carbon sink, which could partly account for offsetting the CO<sub>2</sub> emissions associated with cement production. In response to the many open questions in this context, the RILEM Technical Committee CUC was created in May 2024. The focus of the TC will be to facilitate discussions regarding the current approaches to estimate the CO<sub>2</sub> uptake by cementitious materials (concrete and other construction products) during and after service life. This Opening Letter presents aspects of the motivation to establish the TC, a brief overview of the state of the art in the field, the scheduled work programme, and the anticipated impact of the TC outputs. Though it is not intended to be an exhaustive discussion of the challenges to be addressed, from what is summarized in this letter, it is clear that work is required to base estimates of CO<sub>2</sub> uptake by cementitious materials on a data set that is as extensive, accurate and forward-looking as possible. It is anticipated that the inclusion of interdisciplinary perspectives and data from academia and industry will enable progress in the field.

**Keywords:** Cement; Concrete; Recarbonation; CO<sub>2</sub> uptake; Carbonation rate.

## 1 Introduction

As the CO<sub>2</sub> concentration of the atmosphere is quickly nearing values that will lead to irreversible, dangerous changes of Earth's climate system [1-3], it is imperative to reduce anthropogenic CO<sub>2</sub> emissions as rapidly and as permanently as possible. Of the different industries facing this challenge, the cement industry has been identified as being one of the difficult-to-decarbonize sectors [4], meaning that efforts to achieve CO<sub>2</sub>-emission reductions in cement production and use should be even more serious and effective than in other sectors. Strategies implemented so far include significant improvements of the efficiency of the Portland cement production processes as well as a reduction of the clinker factor of cements; several potentially more impactful measures have been proposed and partly tested (e.g., carbon capture and storage, CCS) [5-10].

While it is clear that CO<sub>2</sub> emissions must be reduced as fast as possible, it has been argued that the extent to which such measures should be applied depends *inter alia* on the amount of CO<sub>2</sub> that is reabsorbed by cementitious materials during their service life [7, 8, 11]. This uptake of atmospheric CO<sub>2</sub> contributes to the long-term CO<sub>2</sub> mass balance of the atmosphere, and it may thus have a bearing on the optimal

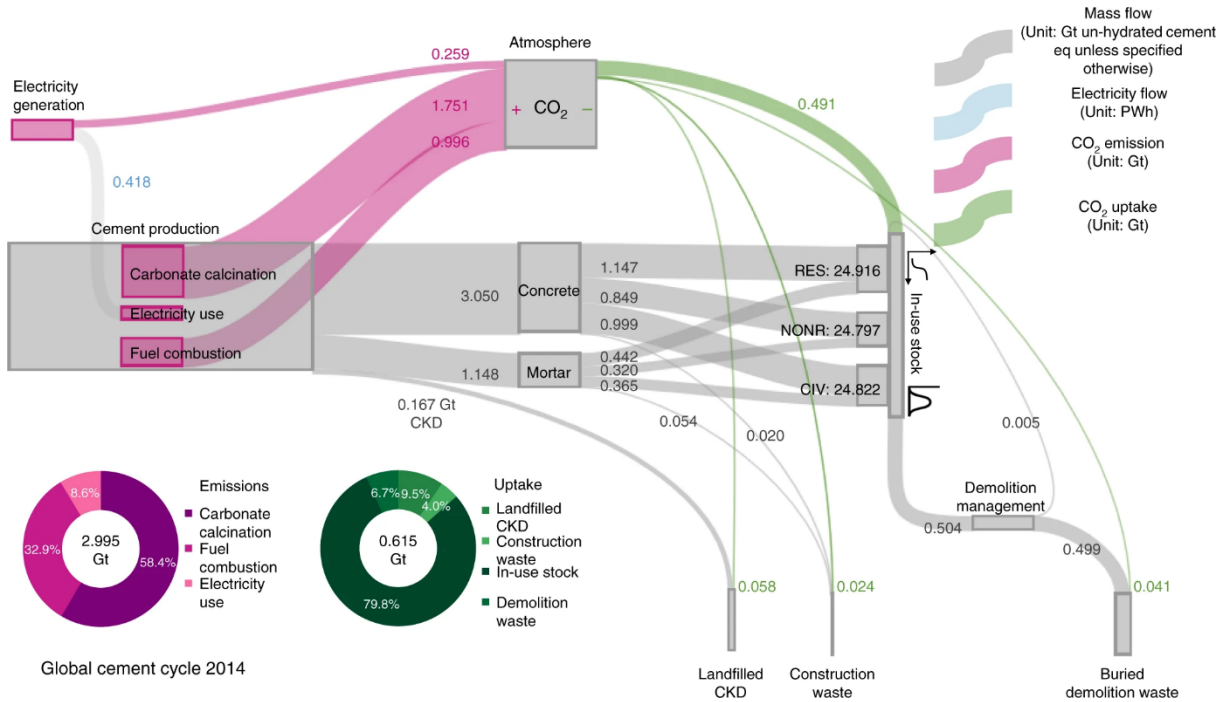
choice of construction materials from an environmental point of view. Moreover, not only newly built structures will take up CO<sub>2</sub>, but also the current stocks of concrete and mortar elements as well as other cement-based products (e.g., pavers, tiles, grouts), be they in service or demolished (*i.e.*, after service life), continue to absorb CO<sub>2</sub> from the atmosphere; thus, their contribution to national as well as the global CO<sub>2</sub> budget can be considered substantial [7, 12-14] (Figure 1). Recent research has demonstrated that it is important in this context to account for the fact that cement-related emissions and CO<sub>2</sub> absorption occur at different times, which has a significant effect on the resulting global warming potential of these fluxes; omission to consider this can lead to a significant overestimation of the climate benefits from cement carbonation [11].

Because of these important implications, estimates of the CO<sub>2</sub> uptake by cementitious materials during their full life cycle should be as accurate as possible. Given that the assumptions underlying previous calculations of CO<sub>2</sub> uptake are based on decades-old data (see Section 2), that the climate and the composition of the atmosphere changes [15], that the compositions of cements have changed significantly in the last decades [5, 16], and that further changes of cement compositions and uses can be expected [5, 6], it is required

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that those previous assumptions are sometimes updated and extended. In this context, the RILEM Technical Committee (TC) CUC: “Carbon dioxide uptake by concrete during and after service life” has been set up in May 2024 at the RILEM Spring Convention & Conference in Milan, to inspect the material parameters that form the basis for such estimates, viz. the carbonation rate of concrete and mortars under

various conditions as well as the fraction of cement or CaO that is carbonated in the ‘fully’ carbonated materials. The present Opening Letter briefly describes the background and the foreseen work and impact of TC CUC.



**Figure 1.** Global mass flows in 2014 related to cement production and use, based on current estimates of stocks, carbonation rates, degree of carbonation etc. All stocks and flows of cement-based materials are expressed in un-hydrated cement equivalents, i.e., excluding aggregate in concrete and mortar. RES, residential buildings; NONR, non-residential buildings; CIV, civil engineering; CKD, cement kiln dust. Reproduced from Z. Cao *et al.*, Nat Commun (2020) 11: 3777 [7]; published under a Creative Commons license: <https://creativecommons.org/licenses/by/4.0/>.

## 2 State of the art and research needs

### 2.1 CO<sub>2</sub> uptake calculations

The carbonation rate (or the coefficient of carbonation,  $k_c$ ) of cementitious materials depends on many parameters. Some of the most important are intrinsic to the materials, such as the water/cement ( $w/c$ ) ratio and the type of cement, controlling the phase assemblage and pore structure evolution, while others are determined by the in-service environment, such as temperature, the relative humidity (*via* the degree of water saturation of the pore system), and the availability of atmospheric or dissolved CO<sub>2</sub>. Thus, the carbonation rate of cement-based construction materials varies widely between different materials and exposure conditions.

A seminal study by Lagerblad [17] collated  $k_c$  values that were suggested to be used for estimates of CO<sub>2</sub> uptake by concrete, including after service life (Table 1). According to this proposal, the carbonation rate of a concrete can be estimated based on its compressive strength and predefined exposure conditions. The values relate mainly to uncovered concrete made with ordinary Portland cement (OPC), but correction factors for surface treatment/cover and cements with

supplementary cementitious materials (SCMs) or limestone have been suggested (Table 2) [17]. As reported by Andersson *et al.* [13], the field data underlying the values in Tables 1 and 2 were collected in the 1980s.

Since the publication of these values about two decades ago, they have been used without substantial adjustments in several assessments of national or global CO<sub>2</sub> uptake by cement carbonation [7, 12, 14]. Andersson *et al.* [13] increased the original values of  $k_c$  by a constant factor to account for the increased CO<sub>2</sub> concentration of the atmosphere (from ~340 ppm in 1980 to ~390 ppm in 2011), and they compared the results thus obtained with values of CO<sub>2</sub> uptake based on a differently modified set of  $k_c$  values. A subsequent statistical analysis of carbonation rates obtained from several sources [18] found that carbonation rates of concrete exposed outdoors were on average higher, and those of concrete exposed indoors were generally significantly lower than the values proposed in Table 1. These latter approaches are in line with clear statements in the original report [17] and an associated publication [19] which call for a verification or a revision of the suggested  $k_c$  values “when more data are available”.

Importantly, since the collection of the data underlying the original reports, the average compositions of cement has changed significantly, mainly through the increased use of blended cements [5, 16], and further developments in cement technology are very likely to occur [5, 6]. Because the carbonation behaviour, including carbonation rates, of blended cements and new cement/binder types is different from that of OPC [20-22], these advances are likely to have a bearing on the accurate calculation of the CO<sub>2</sub> uptake by existing as well as future structures and material stocks, and an accurate reflection of this influence will likely be more complex than the compilation of values in Tables 1 and 2. This has been partly considered by Van Roijen *et al.* [11], who used correction factors for the  $k_c$  of concretes with SCMs that were derived from a recent review by RILEM TC 281-CCC [20]. Nevertheless, the values provided in the original report [17] have propagated into the most recent version of European standard EN 16757, which stipulates rules and recommendations for Environmental Product Declarations (EPDs) for concrete (though in the standard the original  $k_c$  are increased by a constant factor, the same as that used by Andersson *et al.* [13], apparently to account for an increased atmospheric CO<sub>2</sub> concentration). Thus, there is a need to review and, if necessary, revise and extend this compilation of values, to evaluate how  $k_c$  have been determined, and to assess how representative such values are when used to simulate CO<sub>2</sub> uptake by cementitious materials in service and after-demolition conditions.

As regards the carbonation of cementitious material after demolition, particular uncertainties seem to exist. At least in Europe and the United States, much of the demolished concrete is used for road bases or backfills [14, 19, 23]. In such uses, the demolished concrete might be exposed to very high

relative humidities (RH) or covered with liquid water, which will considerably decrease its carbonation rate, potentially to negligible values [24, 25]. The 2005 report [17] gives a single value of  $k_c$  under “wet/submerged” conditions per concrete strength class (Table 1), and EN 16757 extends these only by stipulating a value of  $k_c = 0.2$  mm/Va for concrete below groundwater level. Considering the significant influence of RH (through internal water saturation) on carbonation rate [24, 25] and also the complex relationship between the RH of the surrounding gas phase, the properties (particularly pore structure as it relates to transport characteristics and sorption isotherms) of a concrete, and its internal RH [26], this approach appears to be a stark oversimplification of the actual processes, *i.e.*, carbonation as it occurs when demolished concrete is used in applications such as road bases.

Another crucial parameter for the estimation of the CO<sub>2</sub> uptake by cementitious materials is the fraction of CaO in the cement in the carbonated layer of concrete or mortar, which is converted to CaCO<sub>3</sub>, *i.e.*, the type and abundance of carbonation products. The 2005 report [17] gave a single value as the estimate of the fraction of CaO that carbonates, based on simplified calculations for Portland cement and data obtained under elevated CO<sub>2</sub> concentrations [23]; however, the report indicated that the behaviour under field conditions might differ. Indeed, it is known that the amount of CO<sub>2</sub> bound in the carbonated layer varies considerably between different materials and environmental conditions (Figure 2) [27]. EN 16757 considers that different degrees of carbonation can occur for different exposure conditions, but the influence of cement type is ignored.

**Table 1.** Carbonation rates for uncovered OPC-concretes to be used for estimates of CO<sub>2</sub> uptake, according to CBI’s 2005 report; correction factors apply for SCMs in the binder (see Table 2) and for surface treatments or covers [17].

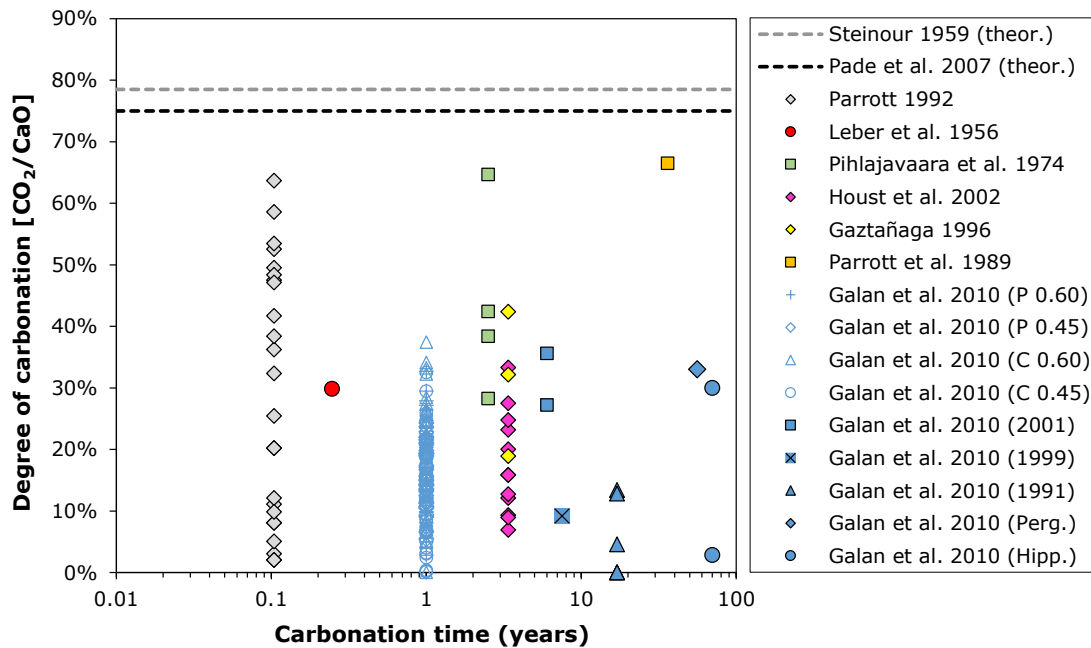
<b>Compressive strength:</b>	<b>&lt; 15 MPa</b>	<b>15–20 MPa</b>	<b>25–35 MPa</b>	<b>&gt; 35 MPa</b>
Wet/submerged	2.0 mm/Va	1.0 mm/Va	0.75 mm/Va	0.5 mm/Va
Buried	3.0 mm/Va	1.5 mm/Va	1.0 mm/Va	0.75 mm/Va
Exposed <sup>a</sup>	5.0 mm/Va	2.5 mm/Va	1.5 mm/Va	1.0 mm/Va
Sheltered <sup>b</sup>	10.0 mm/Va	6.0 mm/Va	4.0 mm/Va	2.5 mm/Va
Indoors	15.0 mm/Va	9.0 mm/Va	6.0 mm/Va	3.5 mm/Va

<sup>a</sup> Outdoors, exposed to rain.

<sup>b</sup> Outdoors, sheltered from rain.

**Table 2.** Correction factors for the carbonation rates in Table 1 depending on the type and fraction of SCMs in the binder [17].

<b>Amount:</b>	<b>&lt; 10 %</b>	<b>10–20 %</b>	<b>20–30 %</b>	<b>30–40 %</b>	<b>40–60 %</b>	<b>60–80 %</b>
Limestone	-	1.05	1.10	-	-	-
Silica fume	1.05	1.10	-	-	-	-
Fly ash	-	1.05	-	1.10	-	-
GGBFS	1.05	1.10	1.15	1.20	1.25	1.30



**Figure 2.** Degree of carbonation, *i.e.*, bound CO<sub>2</sub> referred to CaO in the cement (mol/mol), in the carbonated layers of different cementitious materials under a range of environmental conditions. The *grey* and the *black dashed lines* indicate theoretical estimates of the maximum CO<sub>2</sub> uptake. Data from [27].

However, it is likely that the cement composition affects the amount of CO<sub>2</sub> that is taken up on carbonation not only *via* the CaO fraction of the cement, but also *via* more complex material parameters such as the relative abundance of reaction products (*cf.* [20, 27, 28]) and the spatial arrangement of these hydrate phases, including the pore structure of the materials. It seems that this issue has not yet been explored in detail, with the notable exception of the work of Steiner *et al.* [28]. However, it might be anticipated that more data concerning this topic will appear during the lifetime of TCUC. In this context it is also important to note that the determination of the thickness of the carbonated layer, *i.e.*, the carbonation depth, is not always unequivocal. These difficulties are illustrated by a recent study, which has shown that the pH in the carbonated zone of mortars can differ by more than one pH unit between different materials [29, 30], potentially affecting the carbonation depth measured with a pH indicator solution such as phenolphthalein as well as the fraction of 'carbonated CaO' in the carbonated layer (see also [31]). Thus, as for  $k_c$ , a refinement of the current value for the bound CO<sub>2</sub> on which estimates of CO<sub>2</sub> uptake are based is required to obtain more accurate results. This should include not only a consideration of blended cements, but also potential new binder types such as alkali-activated materials and hybrid alkaline cements in which CO<sub>2</sub> binding occurs to a larger extent through reactions of alkali metals rather than calcium.

## 2.2 CO<sub>2</sub> uptake capacity of blended and non-Portland cementitious materials

The use of SCMs has increased over the past decades, as it is a rapidly implementable strategy for reducing the clinker factor and also to improve the durability of cement-based products [32]. Both of these aspects are contributing significantly to decarbonising the construction sector. However, the use of some SCMs clearly increases the carbonation susceptibility of cementitious materials [20, 22]. From the perspective of CO<sub>2</sub> uptake capacity, it can be argued this is an additional advantage or incentive for using blended cements, but from a materials performance perspective, the increased carbonation experienced by blended Portland cements can lead to reductions in compressive strength and internal microstructure alterations (e.g., increased porosity/permeability) making the material more susceptible to developing other durability issues. This implies that CO<sub>2</sub> uptake calculations cannot be evaluated alone to determine the sustainability credentials of cement-based materials, if such CO<sub>2</sub> uptake comes at the cost of additional maintenance, repair, or replacement of such products in the medium to longer term.

A similar argument applies to non-Portland cementitious systems, with ultra-high contents of SCMs (e.g., alkali-activated and/or hybrid alkaline cements), which, depending on their mix design and conditions for evaluation of the carbonation performance, can exhibit comparable or higher carbonation rates than blended Portland cements [21]. It has also been found that pH measurements using an indicator solution, which is the common practice for evaluating

carbonation of concrete products, and whose results are used being used for CO<sub>2</sub> uptake capacity estimation, are not suitable for estimating the formation of carbonates in some systems [29, 30].

The increasing use of new blended cements and non-Portland binders, for which pertinent field data can be obtained only in some cases, necessitates that assessments of CO<sub>2</sub> uptake by these materials are partly based on the results of laboratory tests, including accelerated tests performed at elevated CO<sub>2</sub> concentrations. This in turn requires examination of the relationships that have been used to convert carbonation rates obtained under accelerated conditions to rates under natural conditions. In addition, relationships between CO<sub>2</sub> concentration, RH and carbonation rate are needed for these new cement/binder types to estimate  $k_c$  in environments in which CO<sub>2</sub> concentrations and RH can differ considerably from standard conditions (e.g., urban and industrial areas *versus* rural areas; demolished concrete/recycled aggregates below ground).

### 2.3 Potential implications in whole-life-carbon assessment of cementitious products

There is great interest to account for the CO<sub>2</sub> uptake potential of different cement-based products in such calculations, as it has been reported by the Intergovernmental Panel on Climate Change (IPCC) that cement-based built environment elements are important carbon sinks [33]. For determining the sustainability credentials of different products, life cycle analysis (LCA) is often conducted. In the case of cementitious products, the global warming potential (GWP) is calculated considering the allocated CO<sub>2</sub> emissions of each of its components, as reported in different databases. Sacchi and Bauer [34] evaluated the potential impact of including carbonation potential in background inventories. It was identified that although it is difficult to determine whether accounting for carbonation will change the LCA results, neglecting it might be including a mild bias in favour of cement-poor products, and recommend including cement carbonation in the development of life cycle inventories. Given that the inclusion of recarbonation in such inventories requires estimations of the carbonation potential during service life, emerging approaches are being proposed for conducting cradle-to-cradle LCA of cement-based products for example those proposed by Kumar *et al.* [35] and Sacchi and Bauer [34], with the aim to integrate CO<sub>2</sub> uptake calculation in EPDs. It is worth noting that it is often opaque which assumptions are being adopted for estimating the extent and progression over the time of recarbonation, e.g., when such calculations are integrated in EPDs of some commercial products. Therefore, developing a common practice to identify when and how CO<sub>2</sub> uptake can be accounted for is urgently needed.

From a different perspective, with the transition towards a performance-based approach to design and specification of concretes and mortars, and the adoption of the Circular Economy, the integration of concepts such as exposure classes and design service life should also be considered in sustainability evaluations to determine the best strategies for

end-of life management of cementitious products [36]. In this scenario, it is imperative to determine the CO<sub>2</sub> uptake capacity during service life *versus* reclaiming/repairing materials for making decisions regarding end-of-life management for cement-based products.

### 3 Work of the TC and anticipated relevance

The scheduled work programme of TC CUC includes mainly data mining research. New data on the carbonation behaviour of cementitious materials, particularly for new cement/binder types, becomes available at an increasing rate, which will be utilised by the TC. In addition, the TC work will involve exchange of good-practices information and laboratory protocols, and possibly experimental data. Depending on the capacities of the participating labs, limited experimental work (e.g., on the fraction of carbonated CaO and carbonation rates under wet/submerged conditions) may be performed to obtain additional data. The TC will be active for five years; this period includes a launch phase (year 1), the collation and processing of literature and additional data (years 2–4), and the finalisation of the outcomes (year 5).

Currently, the research/literature review of TC CUC is scheduled to focus on:

1. Definition of 'carbonation depth' and related concepts; parameters influencing the reaction products and 'degree of carbonation' in the carbonated layer.
2. Overview of the parameters (carbonation rate, fraction of carbonated CaO) employed in previous assessments of (global or regional) CO<sub>2</sub> uptake by cement carbonation.
3. Influence of humidity, CO<sub>2</sub> concentration, cement type, and concrete compressive strength on the carbonation rate and CO<sub>2</sub> uptake during lifetime (focus on *structural/reinforced concrete*; inclusion of below-ground concrete, e.g., foundations).
4. Influence of the humidity, including wet/submerged conditions, and CO<sub>2</sub> concentration on the carbonation rate and CO<sub>2</sub> uptake during *concrete recycling* (incl. storage after crushing) and *secondary life*.
5. Influence of humidity, CO<sub>2</sub> concentration, and concrete element type on the carbonation rate and CO<sub>2</sub> uptake during lifetime (focus on *non-reinforced concrete products*, e.g. concrete blocks).

The above assessments are expected to facilitate an update, including a more detailed classification according to cement type etc., of crucial parameters that are used to estimate the CO<sub>2</sub> uptake during and after the service life of structures made from cementitious materials as well as the service lifetime of reinforced concrete structures.

The TC will report its outcomes as reviews and, if a consensus can be reached, recommendations regarding the influence of the CO<sub>2</sub> concentration on the carbonation rate of concretes and mortars, including quantitative relationships to describe this influence; the influence of RH on the carbonation rate of concretes and mortars, including wet/submerged materials; and the type and abundance of the reaction products in the

carbonated layer of cementitious materials. This will include recommendations regarding future research ('research roadmap').

The outcomes of the TC will be of relevance for the construction industry and beyond. As outlined above, mitigating climate change caused by anthropogenic CO<sub>2</sub> emissions is the key challenge of our societies, and recent assessments of global CO<sub>2</sub> uptake by cement carbonation have indicated that the latter can play a significant role in this regard, implying also a significant economic and political relevance of this process. As demonstrated in Section 2, an update of parameters that are fed into estimates of service lifetime and CO<sub>2</sub> uptake of concrete and mortar is required. The results of the TC work will thus be particularly relevant for academics and practitioners dealing with service lifetime and CO<sub>2</sub> uptake estimates for cementitious materials. It is anticipated that the results will facilitate the further development of best practices for issuing EPDs for cementitious materials, and the evaluation of the environmental merits of construction materials in general. In addition, the work of the TC will likely have a bearing on methods for the calculation and reporting of national greenhouse gas (GHG) emissions/removals, such as that undertaken by the Task Force on National Greenhouse Gas Inventories (TFI) of the Intergovernmental Panel on Climate Change (IPCC) [37]. As the implications of such estimates are far-reaching, the results of the TC work are expected to be of interest for the general scientific community, policy makers, industry, and the general public as well.

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## Authorship statement (CRedit)

**G. J.G. Gluth:** Conceptualization, Writing – Original Draft, Writing – Review & Editing. **S. A. Bernal:** Conceptualization, Writing – Original Draft, Writing – Review & Editing.

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