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Carbonation as a method to improve climate performance for cement based material

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A B S T R A C T

Concrete is today considered to be a crucial material in the ongoing complex transformation of our society in a sustainable direction. One prioritized requirement in this transformation is to reduce the emissions of CO_2 that are associated with concrete production and use. One well-known factor which, so far, has not been considered is (re)carbonation of built concrete products over their life cycle. This paper presents research to quantify such uptake in reports to the United Nations Framework Convention on Climate Change (UNFCCC). It also presents the current status of considering carbonation in the EU Emissions Trading System (EU ETS) and in International Environmental Product Declarations (EPD). The paper also presents some possibilities for further research on how to improve climate performance by considering carbonation.

1. Introduction

The paper deals with cement-based materials (concrete and mortar). The word *concrete* is however frequently used as a generic term and also used in this paper for both concrete and mortar. The term *cement* is used solely for "common" cements with Portland clinker as one component. Special cements are not treated, with the exception of a short mentioning in Chapter 4.1.

Concrete is the most used construction material in the world. Its use dates back to before and during the Roman Empire, but its modern use started around 200 years back with the introduction of industrial production of Portland cement. Raw materials for cement are globally widely available and today, the properties of concrete are well proven both in diverse climates and over a long timeframe. These properties are very suitable for sustainable construction.

However, one important aspect to consider is that cement production is a source of CO_2 emissions, mainly due to its high temperature process. A global average estimate of the emissions per tonnes clinker can be as 36% from use of fuels and 64% from calcination of limestone (a primary raw material) at high temperatures [1].

Several research and development activities related to cement are under way including fossil-free energy, complete electrification of the process, Carbon Capture Use or Storage (CCU/CCS), development of existing binders and additions, as well as introduction of new binders, [2,3]. It is also a development work that needs involvement of other construction actors, depending on the influence from design, manufacture, construction etc. as well as involving society's efforts to create a sustainable society. Reference [2] argues that with engagement from different actors and stakeholders, significant reductions in CO_2 emissions can be achieved.

Carbonation is a chemical reaction by which CO_2 penetrates the concrete and reacts with the hydration products, forming mainly calcium carbonate. Knowledge of carbonation of existing concrete structures is well-established. Both CSH gel (short for Calcium-Silicate-Hydrate) and $Ca(OH)_2$ form a part of the cured concrete. CO_2 in the atmosphere, in contact with concrete, will primarily react with $Ca(OH)_2$ in the concrete according to the principle reaction (1). below, but will also react with the CSH gel. These reactions, which describe the uptake of CO_2 in concrete, are called carbonation, see 1. below.

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \quad (1)$$

The carbonation reaction takes place in several steps [4,23]. The actual uptake reaction between the calcium and carbonate ions takes place in the water phase of the pore solution in the concrete. Water and moisture are thus an important part of carbonation.

The table below, taken from FprCEN/TR 17310.2018 [18] (the figures are originally presented in [6]) provides the amount of CaO available for carbonation for the different reaction products for Portland cement (Table 1).

The table shows that also other reaction products, such as monosulphate and ettringite carbonate, but the quantity in common cements

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Table 1
Binding capacity of Portland cement in terms of total CaO.

Calculations.		Hydrate phases				
		CSH	CH	AFm	Aft	All
a)	Hydrate phase content, %	50	25	10	10	95
b)	CaO molar ratio	0,42	0,76	0,36	0,27	–
c)	CaO in cement hydrate phase, %	21	19	3,6	2,7	46*
d)	Assumed degree of carbonation in hydrated cement, %	0,5	1	0,75	0,5	–
e)	CaO available for carbonation in cement hydrate, %	11	19	2,7	1,3	33
	CaO available relative to the total CaO, %	23	41	5,8	2,9	72

is small compared to portlandite and CSH-gel.

Carbonated concrete is chemically stable and increases the strength of the concrete with cements based on Portland clinker. The carbonation is associated with a lowering of the pH in the concrete, thus reducing the chemical protection of built in reinforcement. Internationally, therefore, construction codes have design rules that the built in reinforcement needs to be protected by a concrete layer covering the steel rebar and must meet certain criteria based on, for example, climate exposure, material properties and the designed service life. Corrosion risk of steel rebar embedded in carbonated concrete requires also humidity. Indoor reinforced concrete protected from rain and exposed to RH < 75% have a negligible corrosion [35].

This CO_2 uptake has historically not been considered in the CO_2 discussion about climate change and calculation methods in this context are still today not generally considered.

Calculating the uptake of CO_2 in concrete and other cement-containing products as well as its impact on the global climate is a very complex task and there is also no unambiguous calculation method that can be used, but approximate and empirical models may constitute such a calculation base. The purpose of the present article is therefore not to present final calculation methods but to initiate a process in which the global uptake of CO_2 in cementitious products can be calculated and taken into account in the global climate work and in assessments of e.g. various concrete products. Consequently, this article presents a proposal for three different calculation methods with different calculation complexity and accuracy in accordance with the IPCC guidelines.

The first part in this paper presents international regulations covering calculation and reporting of CO_2 emissions related to concrete. The second part presents an approach to calculate the annual CO_2 uptake of existing concrete construction. It is based on the report [4], which in turn is based on the pioneering paper [5] and six other studies [6–13] (8 papers) resulting in three calculation methodologies, with different accuracy and complexity, to be in line with the emission inventory guidelines of the IPCC (Intergovernmental Panel on Climate Change). The third part discusses the importance of knowledge of CO_2 uptake to develop new sustainable concrete design processes as well as cement and concrete products. Finally, conclusions for further development and research are presented.

2. Different ways to monitor, quantify and report CO_2 emissions

The impact of greenhouse gas emissions on climate change can be measured and monitored in different ways, depending on the geographical boundaries and methodological approaches. When considering different knowledge-based systems, one must take into account that these systems have been developed for different goals and with different starting points. Thus, the outcome of the systems may differ considerably.

Below, three international systems for monitoring, quantifying and reporting CO_2 emissions and their consideration to carbonation of CO_2 in concrete are presented.

2.1. Reporting of national CO_2 emissions to the UNFCCC

During the Rio Conference in 1992, the UNFCCC (United Nations Framework Convention on Climate Change) was established as a framework for international cooperation with the aim to combat climate change by limiting average global temperature increases.

Countries report annual greenhouse gas emissions and removals yearly to the UNFCCC. The annual emissions reported to the UNFCCC covers emissions from activities within the country's territorial boundaries. The country reporting is strengthened by legally binding agreements, such as the Kyoto Protocol and the Paris Agreement. The requirements and accounting procedures for emissions and removals were reached through extensive negotiations by the parties to the agreements.

The guidelines for estimating emissions and removals for national reporting are developed by the IPCC (Intergovernmental Panel on Climate Change) [21]. The carbonation process of CO_2 in cement products is mentioned in the latest version of the guidelines (from 2006) [21]. But, at that time the scientific consensus was that further work was needed before carbonation could be included into national inventories. Due to the potentially large amount of CO_2 that has been and will be removed from the atmosphere and stored in cement-based products, it is vital that the next update of the guidelines include methodologies for estimating such activities for country reporting. The guidelines also acknowledge that CO_2 capture and storage (CCS) is an option that could be used to reduce greenhouse gas emissions.

The forest industry, in its annual country-based CO_2 balance calculations, includes both annual emissions and uptake. In Chapter 3, a similar approach to also include CO_2 uptake from existing structures in concrete in the calculations is described.

2.2. EU climate and energy frameworks for lowering climate impact

The European Union has its own requirements on reduced climate impact [39,40], including EU/ETS (Emissions Trading System) for industries, like European cement and steel industries, and EU/ESR (Effort Sharing Regulation) which deals with emissions from other sectors like transport, agriculture and forestry. In order to meet the EU's new energy and climate targets for 2030, Member States are required to establish a 10-year National Energy and Climate Plan (NECP) for the period from 2021 to 2030 to ensure consistent reporting by the EU and its Member States under the UN Framework Convention on Climate Change and the Paris agreement. In Sweden, the emissions reported within EU/ESR in 2014 were 34 million tonnes CO_2 eq. [14] while Swedish industry emissions, EU/ETS, 2014 amounted to about 20 million tonnes CO_2 eq. out of which the cement used in Sweden accounts for about 2 million tonnes CO_2 eq.

The EU/ETS includes > 13,000 industry plants and covers around 45% of the EU's total greenhouse gas emissions. Both European steel and cement industries are encompassed by the requirements based on plant-specific emissions.

The main part of the greenhouse gas emissions in Europe comes from traffic, agriculture and forestry and is to be covered by EU/ESR. The EU/ESR, in contrast to EU/ETS, includes flexibility related to annual uptake both in the forestry and CO_2 bound in construction timber. However, the CO_2 uptake by concrete during the use stage is not regarded.

2.3. Environmental product declaration, EPD

In a sustainable society, buildings are constructed securing their function during the life cycle. To investigate the total environmental burden of a product or structure, Life Cycle Assessments (LCA) are used. In a LCA the environmental impact from emissions to air, water and soil are quantified and declared. For the construction sector, the EPD (Environmental Product Declaration) system has been developed based on the standards EN 15804:2012 [15] and ISO 21930:2017 [16]. EPD is increasingly used as a criterion in the various existing environmental certification systems.

The European standard EN 16757:2017 (Sustainability of construction works – Environmental product declarations - Product Category Rules for concrete and concrete elements) [17] provides additional rules specifically for concrete and concrete elements. Even if many mortar applications, with only fine aggregate, like rendering, and concrete products like paving stones and blocks, are not comprised by the definition of concrete in [17], they can from a carbonation and CO_2 uptake view, however be regarded as concrete products with fine aggregate. Concerning especially the part on CO_2 uptake, Annex BB in [17], the text is accordingly valid and can be applied also for mortar. The impact of the use and end-of-life stages includes carbonation of the concrete. When CO_2 uptake is not considered, this shall be stated in the EPD. Annex BB in this standard provides a possible method to assess CO_2– uptake through carbonation in different life cycle stages. Other calculation methods may be used if transparently documented. Further information can be found in the Technical Report FprCEN/TR 17310:2018 (Carbonation and CO_2 uptake in concrete) [18].

Steel products and constructions are treated in the same way as for concrete. For wood products and construction, there is not yet any established product system, instead the same forest system is used as for the UNFCCC. There have been several attempts to distribute the CO_2 balance from timber construction over time, but due to the lack of defined timber product systems, this is not possible [19,20].

3. Approach to calculate CO_2 uptake in existing concrete structures in line with IPCC guidelines

3.1. Introduction

With the aim to provide methodologies for estimating CO_2 uptake of concrete in line with the IPCC, IVL report No. 2309 [4] presents a background and calculation models. The report is briefly summarized in this chapter. The models of carbonation and CO_2 uptake in concrete are based on existing and well documented knowledge, expressed for instance in the European standard EN 16757:2017 [17]. This standard can be used for calculation of CO_2 uptake of a product during its life cycle, (known as the product approach).

For the reporting to UNFCCC according to IPCC's guidelines, it is however necessary to calculate the annual CO_2 uptake of all concrete structures in a region or country (known as the society approach). For this purpose, one normally needs information about the existing building stock, for instance, composition, amount and age of concrete, and type and exposure of structures. This can be a challenging work for many countries. The IVL report has derived information of the results from seven countries that have done this type of advanced national CO_2 uptake calculations to achieve both simplified and more advanced calculation methods.

3.2. Basic principles of calculation

When calculating the global environmental effects of the use of concrete, one must consider all concrete use at the same time. Cement and concrete are produced continuously and give rise to greenhouse gas emissions globally. At the same time, CO_2 is slowly absorbed in all concrete that has been produced that is still in service and can carbonate. It is this global net emission of CO_2 (emissions minus uptake) that gives the global impact of concrete use on the climate. This method of calculation is what is used in the annual national and global calculations of CO_2 for cement and concrete, except that the uptake of CO_2 by carbonation is not currently taken into account in any other way. This results in misleading emission values for greenhouse gases from concrete products. The international greenhouse gas reporting is done according to the UNFCCC (United Nations Framework Convention on Climate Change) protocols, while the methods for the CO_2 calculations are regulated by the IPCC (Intergovernmental Panel on Climate Change) through the document, 2006 IPCC Guidelines for National Greenhouse Gas Inventories [21]. The uptake of CO_2 is not new in the climate calculations, but occurs in different contexts, such as in forest growth or carbonation of minerals in soil and sea. Time aspects and durations for the binding of CO_2 can sometimes be a problem. When carbonating concrete, the binding of CO_2 is essentially permanent, as heating to the calcination temperature (~850 °C) would be required in order for CO_2 to re-enter the gas phase. Despite solid facts, the integration of CO_2 uptake and the use and communication of these values have been remarkably weak. Therefore, the study [4] mainly focuses on the methods to integrate the uptake of CO_2 in concrete into the national and global calculation models for CO_2.

The calculated amount of CO_2 driven off from the material can be considered as the maximum theoretical uptake of CO_2 due to carbonation of the different cement containing products. After the service life of a concrete structure, it will be demolished and typically crushed into finer pieces. This will increase the specific surface area and increase the carbonation rate. A complete CO_2 uptake model must therefore calculate the CO_2 uptake in the different cement containing products during their lifetime as well as in the end-of-life processes and when used as secondary products such as crushed concrete in a road base or as landfilling material. The emission and uptake models are illustrated in Fig. 1.

The CO_2 uptake in concrete is a relatively slow process that takes place over many years. The primary uptake is due to the hydrated Portland cement portion of concrete structures, such as bridges, house frames, concrete tiles, concrete roads, railway sleepers, cement mortar, etc. Also latent hydraulic concrete additions, such as blast-furnace slag and pozzolanic additions such as fly ash from coal combustion, which also can take up CO_2, can be important to include in the calculations.

3.3. Mathematical model for calculating CO_2 uptake in a concrete surface and a concrete structure

The depth of carbonation, d, can be calculated by the well-established formula.

$$d = k\sqrt{t}, \quad (1)$$

where t is time and k is the rate of carbonation. It depends on exposure and concrete quality. For covered surfaces with an initial resistance to penetration by air, the depth of carbonation has been shown to be better represented by polynomial equations [22].

To be able to calculate the CO_2 uptake, we also need the degree of carbonation, DOC, defined as the amount of CO_2 uptake by carbonation

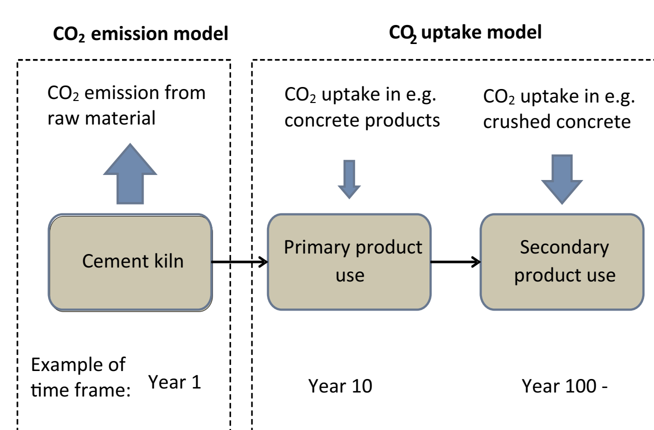

Fig. 1. Schematic figure showing the CO_2 balance in cement products over a certain period of time.

in relation to the theoretical maximum CO_2 uptake by carbonation (corresponding to 100% DOC). The maximum theoretical uptake of CO_2 in the clinker part can be equated with the CO_2 emission from calcination that is driven off from the material. The degree of carbonation is defined only within the area which has been considered carbonated and this area has been defined as the area exhibiting color change with phenolphthalein test. See for instance [17 and 23]. Other less distinct meanings do occur, but should not be used for the purpose of CO_2 uptake calculation.

The DOC depends on exposure of the concrete. References [5, 17, 18, 23]. Moist exposure, as in outdoor concrete, especially when exposed to rain, gives the highest values, while exposure in dry environment gives the lowest.

Values of k and DOC can be found for instance in [17] (EN 16757:2018, Annex BB). The k-values were original reported in [23], which presents evaluated and systematic compiled values, based on a number of studies. The values are reproduced in the standard [17] and in the IVL-report [4]. It is also reported in [41].

In [17] are also given corrections for the k-factor for cement with additional major constituents or concrete with mineral additions (fly ash, GGBS, lime stone and silica fume). Please note that although carbonation rate typically increases with increasing amount of addition, the CO_2 uptake normally decreases due to a reduced amount of carbonating compounds.

The CO_2 uptake in kg per m^2 concrete during t years for any application can according to [17] be calculated as:

$$CO_2 \text{ uptake at a surface} = (k \times DOC)\,(\sqrt{(t)}/1000) \times U_{tcc} \times C, \tag{2}$$

where

CO_2 uptake is the total uptake in kg CO_2/m^2 concrete surface.

k is the rate of carbonation for the surface in mm/√(t).

DOC is the degree of carbonation for the surface, and t is the number of years.

U_{tcc} is the maximum theoretical uptake in kg CO_2/kg cement. The value is ≈ 0.49 for Portland cement (CEM I). See [17] for explanation and values for cements with lower clinker content.

C is cement content in kg cement/m^3 of concrete.

For an application, structure or product the total CO_2 uptake in kg will then be calculated based on the sum of the uptake at all the different surfaces (i) according to;

$$\text{The total } CO_2 \text{ uptake} = (\Sigma(k_i \times DOC_i \times A_i))\,(\sqrt{(t)}/1000) \times U_{tcc} \times C \tag{3}$$

where A_i is the area of surface i in m^2.

The CO_2 uptake per m^3 can now be calculated by division with V, where V is the total concrete volume (m^3 for the application, structure or product).

Cement types other than CEM I, or concrete with additions like slag and fly ash, are normally considered to have higher carbonation rates. Figures are given in [17]. Compare also the first paragraph under the k-factor Table 2 above. More information on carbonation rate for blended cements can be found in [18,23]. The CO_2 uptake is however insufficiently known for some other cement types, and the temporary solution is to consider uptake only in the clinker reaction products. See further discussion in Chapter 6. For cements other than CEM I, consideration of clinker content can be done by multiplying the uptake of a CEM I cement with a factor, < 1, equal to actual clinker content in % divided by 95 (the normal clinker content in % of CEM I).

For very accurate calculations, following Tier 3 methodology, see Chapter 3.6.1, a known CO_2 content for different exposures or with a general increased value, may also be taken into consideration. In [22], one example of a theoretical model that offers a possibility to consider actual and different CO_2 content in the air is presented. In the Swedish study [5] results are presented for a general CO_2 content value of 393 ppm (6,0 E-04 kg CO_2/m^3 air). When using [22] the increase of CO_2 uptake is about 10% higher compared to the uptake calculated on historical levels of CO_2 content (≈330 ppm).

3.4. Annual CO_2 uptake in a concrete product or structure

In a specific concrete product, such as roof tiles, or concrete structural application, such as apartment buildings, there is a dynamic yearly input of new construction as well as an outflow of construction that is at the end of its service life. Therefore, a yearly input has to be calculated for each year the products and structures are used, as well as when they are no longer in use. The annual uptake of CO_2 may be described by Eq. (4) from [5] where the annual uptake ΔCO_2 in year t is equal to the difference between the accumulated uptake at the end of year t and (t-1), respectively.

$$\Delta CO_2 = CO_2^{\text{uptake in year } t} = \sum CO_2^{\text{uptake, }(t)} - \sum CO_2^{\text{uptake, }(t-1)} \tag{4}$$

The yearly inputs can be calculated by formulas (2) and (3) based on either the cement used in the segment or by knowing the actual production facts. The yearly output (structures taken out of service) can be modelled either by an estimated service-life or by knowing the actual output.

A detailed description of the calculation steps can be found in [5] where an example of a numerical calculation for roof tiles is also given.

3.5. Annual CO_2 uptake and CO_2 balance in concrete structures – the Swedish example

In order to integrate all existing structures in a region or a country, one should have knowledge about the total building stock and preferably the historical concrete production. These rather laborious investigations have been carried out in the Swedish study [5]. Also, in the global study [11], a similar procedure is carried out.

The Swedish study uses data for 100 years of cement consumption, 60 years of distribution of that cement to different applications, and detailed knowledge of how concrete structures are distributed throughout the building sector, to estimate the stock of concrete applications. The k-values and the degree of carbonation are very similar to the ones in EN 16757 [17]. However, polynomial equations are used [22] instead of k-values to calculate the depth of carbonation for indoor covered surfaces. The uptake each year is calculated as the difference between two consecutive years and is summed up for all 7 of the applications/products. Material data for two examples, bridges and residential buildings, are shown in Table 3. The result of the calculation is shown in Fig. 2.

The paper reports that for the year 2011, the corresponding CO_2 balance was reduced by 17% due to the CO_2 uptake.

3.6. Synthesizing annual CO_2 uptake in a society – with an IPCC context

National greenhouse gas emission and removal inventories are reported to the UNFCCC yearly, based on IPCC methodologies, as described in Chapter 2.1 above. The IPCC has classified its methodological approaches in three different tiers, according to the quantity of information required, and calculation accuracy (IPCC, 2006). Generally, the higher tier, the more accurate but complex calculation methods.

Tier 1 represents a general and simplified approach based on national or international statistics together with standardized assumptions (factors and parameters) from the IPCC. Tier 1 can be used as a starting point to make an early emission or removal estimate of the level of magnitude. Tier 1 can also be used when national information is lacking or if a specific source is considered to be of minor importance/magnitude. For more significant sources, Tier 2 or Tier 3 should be used. Tier 2 generally uses the same methodological approach as Tier 1 but applies statistics, factors and parameters which are more detailed and specific to the country. Tier 3 generally refers to complex models or

Table 2
k-factors [mm/year^{½}] for calculation of depth of carbonation for different concrete strength classes (cylinder) and exposure conditions and degree of carbonation for different exposure conditions. (For Portland cement).

Concrete strength	≤ 15 MPa	15–20 MPa	25–35 MPa	≥ 35 MPa	Degree of carb
Parameters	Value of k-factor, in mm/year ^{0,5}				Percentage
Civil engin. structures					
Exposed to rain		2,7	1,6	1,1	85
Sheltered from rain		6,6	4,4	2,7	75
In ground [a]		1,1	0,8	0,5	85
Buildings					
Outdoor					
Exposed to rain	5,5	2,7	1,6	1,1	85
Sheltered from rain	11	6,6	4,4	2,7	75
Indoor in dry climate[c]					
With cover[b]	11,6	6,9	4,6	2,7	40
Without	16,5	9,9	6,6	3,8	40
In ground[a]		1,1	0,8	0,5	85

[a] Under groundwater level k = 0,2.
[b] Paint or wall paper. (Under tiles, parquet and laminate k is considered to be 0.)
[c] Indoor in dry climate means that the RH is normally between 45 and 65%.

Table 3
Swedish data used for bridge and residential building.
From [5][a].

	Bridge		Residential building	
Unit	per bridge		per apartment	
	m^2	MPa	m^2	MPa
Type of application/surfaces				
Indoor				
Without surface cover			93	20–25
Painted			247	20–25
With tiles/clinkers			20	20–25
With plastics/linoleum			0.3	20–25
With parquet/laminate			68	20–25
Slab on ground				
With mineral wool			0.1	30–45
With polystyrene			18	30–45
Without insulation, coarse gravel	170.8	> 45	12	30–45
With insulation, with sand/ gravel			22	30–45
Outdoors				
Exposed to rain	43.8	> 45	38	30–45
Sheltered from rain	422	> 45	8	30–45
Total area A (m^2/unit)	636.6		526.4	
Volume V (m^3/unit)	277		67.42	

[a] For the year 2010.

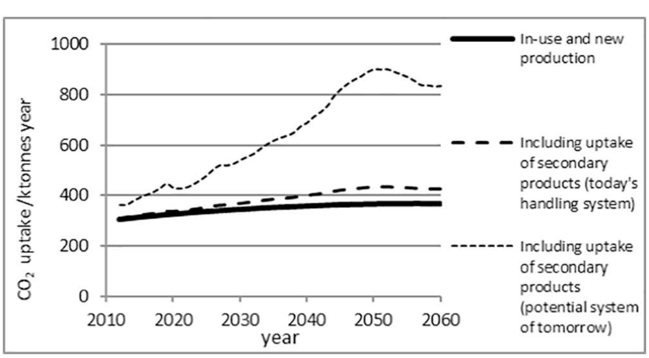

Fig. 2. Calculation of a future scenario for CO_2 uptake in Sweden. From [5].

monitored data (e.g. emission measurements).

The IPCC (2006) states that emission and removal inventories “should be accurate in the sense that they are neither over- nor underestimated as far as can be judged, and precise in the sense that uncertainties are reduced as far as practicable”. Progressing from Tier 1 to Tier 3 generally represents a reduction in the uncertainty of emission and removal estimates, though at a cost of an increase in complexity of data and analyses. Emissions and removals are reported to the UNFCCC together with information on estimated uncertainties, typically presented as a 95% confidence interval.

Seven studies from different countries are compiled in Table 4, (extract from [4]) representing Ireland, the Netherlands, Norway, Spain, Sweden, Switzerland, and “Global”.

Based on this, three different methods for calculating the annual CO_2 uptake in a country is suggested, Tier 1, 2 and 3. In the ongoing work, the above proposed calculation methods will be tested and evaluated in some selected countries. Based on the results of these tests, a broader international implementation with different calculation support is then planned.

3.6.1. Tier 3

The methodology and accuracy of the calculations in the Swedish paper [5] and the global paper [11] can serve as a model for Tier 3. Tier 3 is the most advanced and accurate model and these two studies estimate the annual uptake in existing building stock by using extensive knowledge of historical cement use and distribution among different applications. Main prerequisites and results from [5] are presented above in Chapter 3.5. For detailed information see [5].

In order to simplify and at the same time make more accurate calculations of CO_2 uptake, more advanced computer models are needed that take into account many of the different factors that affect CO_2 uptake. In Tier 3, it is also possible to use complex national calculation models requiring collaboration with various cement and concrete researchers. Such complex computer models should, of course, undergo a normal scientific review, which is a common procedure within the IPCC.

3.6.2. Tier 2

Five other national studies of annual CO_2 uptake in existing structures are also compiled in [4], see Table 3. These studies [6–10,12,13] are used to develop a Tier 2 method, where the requirement for historical cement and concrete data is less than for Tier 3.

The studies are based on a single year, or a few years, of data on cement and concrete use. They make use of the assumption that the uptake during one year in concrete that was produced during the previous (100) years, which is the desired information, can equal the uptake in the coming (100) years in concrete produced during the same year. This can be characterized as “onward” calculation, which is

Table 4
Short overview of existing models for calculation of CO_2 uptake and their background.

	Sweden	Norway	Netherlands	Ireland	Switzerland	"Global"	Spain
General framework							
Reference of study	[5]	[6]	[7]	[8,26]	([9,10] recent information)	[11]	[12,13]
Fly ash and slag	Not taken into account	Taken into account	Slag taken into account for both carbonation depth and uptake (0.14 kg CO_2/kg slag)	Not taken into account	Not relevant	Only clinker content accounted	YES included
Results							
Calculated as	Uptake in existing building stock, estimated with the previous 100 years cement consumption	Uptake in the 2011 production of concrete during coming 100 years	Uptake in 2015 production of concrete during coming 60 years.	Uptake in one year production of concrete during coming 100 years	Uptake in one year production of concrete during coming 50 years	Uptake in existing building stock, estimated with the previous years (1930–2013) cement consumption	Calculated as % of clinker fabrication INCLUDING THE FUEL (multiply by 1.7 the results for referring to decarbonation only
Corresponding to	17% of production emissions year 2011	15% of production emissions (18% including recovery)	19% of production (incl. import) emissions (23% including end of life stage)	16% of calcination emissions, corresponding to about 10% of total.	Declared to be 10% of the total emissions, corresponding to $(1/0.63) \times 10 = 16\%$ of the calcination emissions.	Uptake 1930–2013 corresponds to 43% of calcination emissions	2.7% of emitted CO_2 by decarbonation
Carbonation theory							
Exposure classes	11	6	5	Residential 6, Civil engineering 3, Commercial 7		5	Standard: interior, outdoors sheltered and non-sheltered from rain
Strength classes	4	4	7	5	3 (?)	4	2 concretes and paste
Carbonation degree	50–90%, specified for each exposure	70%	40–85%, specified for each exposure	Not specified	75%, changed to 50% in [10]	80% for concrete 91.5% for mortar	Main aim of the study
Concrete data							
No. of applications (type of product)	7	24	5	3	20	Depending on region, most detailed from China	Concrete elements in general
End-of-life and secondary use							
End-of-life processes	Demolished, crushed, stockpiled and reused as unbound material	Demolished, crushed, stockpiled and reused as unbound material	Demolished, crushed, stockpiled, and reused.	Only shortly treated	Demolished, crushed, stockpiled, and reused.	Demolished, crushed, and secondary use. Only small uptake	No recycling or secondary use
Comments	Uptake during one year (2011) in existing buildings is calculated from 100 years cement statistics and 60 years concrete application statistics		Concrete blocks are responsible for a significant proportion (24%) of the CO_2 uptake.	Open texture concrete blocks and roof tiles are responsible for a significant proportion of the CO_2 uptake	The uptake fig. 16% of the calcination emissions should according to the later recommendations be reduced by a factor 1.5, that is to 10.7%. In order to be consistent with the methods in the other countries a 100 years perspective should be applied, resulting in $1.414 \times 10.7 = 16\%$	Uptake during 1930–2013 as well as present (2013). Uptake in mortar is significantly contributing to the large figures	The low uptake figure of 2.7% has been adjusted by a recent paper: Andrade C, "experimental Evaluation of the Degree of Carbonation in three Environments" and in [4] (IVL-re-port).The new value being 15%

further explained in [4].

The modelled annual CO_2 uptake depends on material and design of the existing building structures. Previous work [5], has by way of sensitivity studies, showed that the knowledge and quality of data for material and applications for recent years is important for this aspect, since the uptake is proportional to the square root of time. The recent years are especially important for slender and low strength products like mortar and masonry, which have high rate of carbonation and might be carbonated all through in one or a few years. Since this historical information normally is not available nationally, IPCC recommends that some type of splicing technique is used to resolve the data gap, such as expert estimations [21]. This is mainly relevant when either the application solutions are drastically changed or when there ia a large change in the amount of concrete use in civil engineering and housebuilding or masonry construction occur.

Another important factor for the results is knowledge of historical cement use over a long period. In case this information is not available nationally, in line with IPCC recommends, expert estimations can be used [21].

Both Tier 2 and 3 require knowledge of cement consumption statistics and concrete production and use, but on a different level. The calculation as such is simpler with the Tier 2 methodology.

A suggested general description of the required steps in Tier 2 is presented for the use stage. The calculations are based on actual historical cement consumption, at least 20–30 years back, and may be used together with knowledge of one recent year of concrete use. Normally, at least 5 applications are needed, corresponding to at least 65% of the cement consumption, for instance: bridges, residential buildings, office buildings, roof tiles, pavement, shotcrete, sleepers, and mortar. Applications outside the chosen ones are treated as the ones most similar to the defined ones. An example of a typical calculation scheme can be as follows:

- ➢ Estimate the historical annual cement consumption and estimate recent annual concrete production and the distribution among the different applications.
- ➢ Calculate the CO_2 uptake for each application during 100 years. (normally).
- ➢ Calculate the sum of the annual CO_2 uptake of all concrete applications.
- ➢ The sum of the cement content in the produced concrete should always be checked against the cement consumption.

The CO_2 uptake in kg per m^3 concrete for each application during t years can be calculated as shown above, Chapter 3.3.

Recommendation for calculating the CO_2 uptake at end-of-life stage and in secondary use is given in [4]. The result should be presented as annual amount of CO_2 uptake for a country or region.

3.6.3. Tier 1

Tier 1 provides a simplified calculation method for estimating the annual uptake of CO_2 in existing concrete structures on a national basis. The model should be used primarily in cases where resources are missing to perform more accurate calculations according to the calculation methods described for Tier 2 and Tier 3, since the uncertainty is relatively high. The national annual CO_2 uptake in concrete in the use stage (existing structures) can however be estimated according to this methodology. The uptake values are related to the reported calcination emissions from the consumed clinker (produced – export + import) in the corresponding country.

The seven studies from different countries are compiled in Table 4. The methods are all based on the well-accepted and documented carbonation rate model of square root of time dependency. The inventories of existing concrete structures comprise different applications, exposure, and concrete quality. The age distribution of concrete in place is estimated by cement consumption statistics over time. This makes it possible to calculate a good estimate of the annual CO_2 uptake in the existing structures.

The annual uptake figures, in these studies, presented as CO_2 uptake in percent of the corresponding calcination emissions, have been used for establishing this simplified, general method, Tier 1, for estimation of the annual uptake in a country.

To get comparable figures from the studies, some adjustments (normalization) have been carried out. The high figure in the Global study (43%) is adjusted in [4] due to the very large amount of thin mortar applications included in the study. The low Spanish value (3%) is also adjusted in [4] due to low chosen values of DOC, the uptake in all interior surfaces and surfaces in contact with other materials, as well as buried concrete were assumed to be zero and the specific surface values (m^2/m^3) for the structures are low.

Numbers that are given as percentage of total production emissions have also been multiplied by 1.6 to get the uptake as percentage of calcination emissions.

After this normalization the series of numbers of the CO_2 uptake related to the calcination emissions are 16, 25, 24, 27 15, 15 and 19%. With the help of these seven studies it has been possible to establish a relation (Tier 1) for a country or region between the annual concrete CO_2 uptake and the annual CO_2 emissions at clinker calcination.

To be in line with the recommendation of the IPCC Guidelines, i.e. to not overestimate, nor underestimate in the calculations, the mean value should be chosen, and uncertainties presented as 95% confidence interval [21].

The population of all countries' annual figures of CO_2 uptake in cement products, related to the calcination emissions, could be considered normally distributed. A simple statistical analysis of the results based on the seven studies, assuming normal distribution, ends up with a mean value of 20% of the reported emission from calcination of consumed cement clinker and a standard deviation of 5.1%. The mean value could then be presented as 0.20 ± 0.038 or $0.20 \pm 19\%$. However, even if the seven studies are independent of each other, they cannot be considered as randomly selected or a statistically representative sample of an average country's annual concrete CO_2 uptake. The true mean value could thus deviate from 0.20 and the 95% confidence interval is thus likely higher than ± 19%. The area of uncertainties thus needs further investigation and research. The calculated CO_2 uptake in the use stage is given below.

The annual uptake in the use stage can accordingly be estimated as $0.20 \times$ (the reported emission from calcination of consumed cement clinker).

If the mortar for rendering applications, in total, amounts to > 10% but < 30% of the cement consumption, the annual uptake factor in the use stage can be estimated at

$0.20 + 0.0115(MR - 10)$, where MR is the mortar percentage for rendering.

For countries with recent highly increased use of concrete we suggest that, just as for Tier 2, a correction could be made. This mean that instead of the actual cement use a specific year, the mean cement used 20 years backwards could be used. This correction should be validated before final decision.

3.6.4. End-of-life stage and secondary use

In addition to the CO_2 uptake over the use stage of structures, there is uptake in the end-of-life stage and the secondary use, normally as crushed material. Detailed information on the amount of this material and its uptake is rare, so it is currently difficult to give a reliable estimate. The percentage of concrete recycling, given in some countries is unfortunately not sufficient, since it is mainly the age at demolition, the processing of the demolished material and the applications for the crushed concrete that determines the actual uptake.

The end-of-life stage (demolishing, crushing and storage) normally includes storage in large unsheltered piles, during a rather short period of time. Moreover, the recycling rate (the annual amount of demolished and crushed concrete in relation to the annual production) is normally low in most countries. The volumes can however be anticipated to increase in the future, since more concrete structures will reach the end of their service lives. It is therefore important to base the uptake calculations on the real amounts of concrete in end-of-life handling.

For Tier 1, the following is proposed in [4]:

Annual uptake in the end-of-life stage (2%) and secondary use (1%) can be estimated at (0.02 + 0.01) × (the reported emission from calcination of consumed cement clinker). Alternatively, the following estimation can be done in the end-of-life stage and the secondary use.

➢ If the annual amount of concrete being taken out of service and processed on a recycling plant is known, the CO_2 uptake in the end-of-life stage can be calculated as 10 kg CO_2/m^3 concrete.

➢ If the annual amount of crushed concrete, entering the secondary use as unbound material, is known, the uptake can be calculated to 10 kg CO_2/m^3 concrete.

For Tier 2 the following is proposed in [4]:

For normal handling procedure or recycling rate < 5%:

Same as for Tier 1.

For improved handling procedure:

A preliminary suggestion is that the uptake could be set to 20 kg CO_2/m^3 of concrete if an enhanced procedure with air access in the fractions and at least 4 months' storage in at least three fractions is applied. In this case, the amount of concrete needs to be known.

Today, most used concrete ends up in crushed form to be recycled as new products i.e. in secondary use. It is therefore important to base the uptake calculations on the real amounts of concrete in secondary use. The CO_2 uptake in secondary use is quite similar to the uptake in primary use, so similar calculation methods could be used. However, even more factors are unknown for secondary use, so it can be difficult to create general but accurate methods. The more exact methods are often quite specific and depend on the type of secondary use (country specific) and may be treated under Tier 3. In Tier 1 a more general, conservative method is proposed.

Under favourable conditions, the total uptake (primary use + end-of-life + secondary use) can amount to about 75% [18,23] of the maximum theoretical potential (equal to the calcination emission), corresponding to about 110 kg CO_2/m^3 for an average concrete. Favourable conditions in the end-of-life stage comprise for instance sheltered storage of crushed graded material. See also Chapter 4.2.

3.7. Uncertainties

In the IPCC context, uncertainties in various components of a national emissions inventory are used to describe the precision of annual emissions at national level, but also to guide decisions on which tier level to use for specific sources of emissions and sinks [21]. It is therefore important to use pragmatic approaches when determining the uncertainty values. In many cases, statistical random samples are not available, and uncertainties may thus often be a combination of measured data, published information, model outputs, and expert judgement [21].

As mentioned above, using higher tier methods should typically reduce uncertainties in estimates, as they should reduce bias and better represent the complexity of a system.

Model input data, which consists of cement production and cement use in various products, as well as cement types and concrete qualities, has both high availability and good reliability in most countries, see [5]. A dynamic flow macro-model [24] is used to describe the accumulation of used cement in the products and structures. The model has been designed to be robust to historical variations. The quality of the results is fully influenced by the quality of the input data and its statistical distribution for the most recent year.

A reference for the uncertainties can be the timber industry inclusion of temporally bound CO_2 in carbon stocks as harvested wood products (HWP) [25]. This is accepted to be done with simple assumptions, for example wood panel and round-wood used in construction in Sweden is said to on an average have a service life of 25 and 35 years respectively.

4. Quantitative knowledge of CO_2 uptake brings new opportunities to apply sustainability concepts

This paper shows that we now have possibilities to both quantify the CO_2 uptake in concrete products and structures during their lifetimes (ref [17], EN 16757:2017), but also to quantify the annual uptake in the existing building stock in a country or region (ref [4], IVL-report B 2309, 2018).

These new calculation methods present a reasonable CO_2 balance, based on scientific methods, for concrete, including the uptake of CO_2. This knowledge of carbonation can also provide us with new tools to further reduce the climate impact of concrete products and structures, by choosing solutions that increase the CO_2 uptake, where it can be made without any harm to concrete durability. Some examples of this are discussed below.

A life cycle approach is essential when calculating the CO_2 impact of concrete, i.e. to estimate the CO_2 uptake in concrete from cradle to grave. Further information on how to do this for the production stage, use stage, and end-of-life stage, can be found in the Technical Report FprCEN/TR 17310:2018 [18].

4.1. Increased CO_2 uptake in the use stage

The effect of cement content, cement type and additions, as well as structural design, on CO_2 emissions is familiar knowledge, which is frequently used for designing concrete with low CO_2 impact in the production stage. The CO_2 uptake conditions for different concretes are now also to be considered. It involves both the rate of carbonation and degree of carbonation in the carbonated zone of the concrete product or structure.

Limiting the delivered concrete to the design strength (not too high) and allowing using a later age than 28 days for fulfilling the requirement on concrete strength (enabling lower W/C-ratio), will normally give a faster CO_2 uptake.

The CO_2 uptake is well known from the perspective of protecting reinforcement bars for reinforced concrete and indirect result is that it is well known that unprotected concrete products (that are not re-inforced) can be allowed to carbonate relatively quickly.

Paint, wallpaper and other coatings decrease the carbonation rate and thus the CO_2 uptake. The report [22] is presenting a method for using diffusion resistance for the coatings for calculation of CO_2 uptake. More differentiated calculation possibilities give the opportunity to use suitable covers for enhanced carbonation, when accepted for corrosion reasons.

Apart from slag and fly ash there is a growing interest in calcined clay and other fillers [36].

Clay suitable for thermal processing is available in many parts of the world. Data on CO_2 uptake in binders with calcined clay has not been found. Limestone fillers are shown to increase carbonation rate, while also contributing to strength, especially with aluminous compounds in the cement.

References of industrial accelerated carbonation of concrete made with existing cement for high early strength are presented in [27–30]. New cements are under development to reduce CO_2-emissions during production [2]. As an example, *Carbonatable calcium silicate cement* and *Magnesium cement* harden by carbonation and have accordingly a large uptake potential. For the former product CO_2 uptake during curing is estimated to be 300 kg CO_2 per tonne of cement [30], corresponding to about 75–120 kg CO_2/m^3 of concrete. The products are reported to require CO_2 to penetrate the structure at hardening, meaning either thin structures or curing chambers with supply of concentrated CO_2

4.2. Increased CO_2 uptake in end-of-life stages – demolishing, crushing, and storage

This is by far the most discussed potential possibility to increase the CO_2 uptake in the concrete life cycle. The reason is mainly that demolishing and crushing substantially increase the specific surface (m^2/m^3) of the concrete. Uptake possibilities are discussed in the following reports [4,5] and [31–34].

It is estimated and established in laboratory tests [32] that the total potential uptake is in the order of 75% of the calcination emissions, corresponding to about 110 kg CO_2/m^3 for an average concrete. The value 75% is a general long time figure for DOC of carbonated concrete (mean practical maximum uptake) The present normal handling procedures of demolished and crushed concrete render the practically achieved uptake to be much lower [4].

Some parts of the hardened cement paste will separate during the crushing process as very fine particles. These very fine particles will rapidly carbonate, possibly in days but up to a few weeks, [37]. The amount of these very fine particles depends on crushing and sieving equipment and process, and has not yet been accurately estimated. Report [4] is presenting a case with slightly > 20% of the cement paste is in this very fine fraction.

The crushed and carbonated material is suitable for replacing virgin rock material, in for instance new concrete or as road bases or ground filling (in-fill). Enhanced industrial carbonation of crushed concrete and the use of the carbonated material is studied in the French project FastCarb [34].

The recycling rate (the annual amount of demolished and crushed concrete in relation to the annual production) is normally low in most countries. The volumes can however be anticipated to increase in the future, since more concrete structures will reach the end of service life. This emphasizes the importance of finding end-of-life procedures that allow for a large uptake.

5. Conclusions

Design for corrosion durability of concrete structures presupposes a calculation of depth of carbonation, which is made according to long-standing, well-documented, tested and evaluated models. The same models can be used for calculation of the CO_2 uptake that occurs. The calculation requires however also knowledge of the Degree of Carbonation (DOC). (See Chapter 3.3 for definition).

Earlier published papers and reports have shown that the CO_2 uptake during use stage and end-of-life stage can amount to 10–15% of corresponding annual total emissions (fuel + calcination) from production of cement.

For products and structures, it is accepted in the EPD (Environmental Product Declarations) to declare environmental impact with consideration of CO_2 uptake. Detailed examples are provided in EN-standards.

The European Trading System (ETS) does not currently allow consideration of CO_2 uptake for the cement and concrete industry. This could be questioned since it is accepted for the forestry industry in their corresponding system, EU Effort Sharing Regulation (ESR).

The paper describes newly developed methods for calculation of annual national CO_2 uptake, and discusses their accuracy. This knowledge is set in line with the IPCC system view, which generally opens up for future inclusion and reporting to UNFCCC. A total CO_2 uptake in the use stage and end-of life stage of 23% of the national calcination emission, is presented, as a value for use in Tier 1. For Tier 2 and 3 methodologies are recommended for more advanced and accurate calculations.

The knowledge of CO_2 uptake and the availability of an accounting tool makes it possible to have concrete recognized as a CO_2 sink which more accurately represents the life cycle perspective. At the same time the knowledge makes it possible to use uptake as a new tool to reduce climate impact from concrete in a life cycle perspective. Examples are given in Chapter 4.

6. Suggestion for further research

This paper presents three levels of methods for calculating annual CO_2 uptake in cement products. This is the first step towards including such estimations in the international reporting to the UNFCCC. Due to the lack of representative data on a global scale, we have not been able to quantify the uncertainties of the methods. This is an area where further research should be accomplished.

To develop and increase the CO_2 uptake in built concrete structures and in reuse of crushed concrete is a new incremental way to reduce the CO_2 level in the air. Initially, there is a need to further determine mechanisms, suitable test methods and amount of CO_2 uptake in blended cements or concrete with additions in different exposures, as well as different handling procedures for demolished and crushed concrete in order to increase CO_2 uptake during end-of-life stage.

Development of deepened knowledge and use of early reactions in calcium silicate phases, in existing cements, by added CO_2 gas is also interesting. At the same time, further research and focus on CO_2 uptake of new cements, not based on calcium carbonate as raw material, is needed.

Development of concrete use for instance indoor concrete structures with fast CO_2 uptake and rendered hollow blocks with quick carbonation should be researched. Also surface coatings (such as renderings, paint, and wallpaper) for indoor use with low CO_2 diffusion resistance should be developed. Studies of indoor CO_2 uptake as a function of average RH should be conducted.

Increased empirical knowledge of k-factors and DOC for different concrete recipes and exposures should be provided.

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Supplementary information

As we hope is clear for the reader, we do not intend to present any final model but instead hope to give the reader an insight in the need to clearly and scientifically define also the environmental aspects when carrying out research in this complex subject. By this we have not focused on the large existing and reported material and structural knowledge about carbonation. Instead we strive to present the existing knowledge in line of the IPCC, which require complementary information rather than existing material data from carbonation. With our paper we hope further research will improve the other inputs needed to improve the climate models.

In many studies and peer-reviewed articles, environmental impacts are emphasized as important driving forces. However, it is common for these impacts not to be quantified instead considered more qualitatively. This can often be a weakness in assessments and comparisons of impact. Our paper deal with the impact on the climate in line with the IPCC system, which is important to know about and rely to for all scientists, just as with in Europe the complementary systems EPD, ETS and ESR.

In our paper we have specifically included information about how uncertainties are treated scientifically by IPCC.

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