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The CO_2 uptake of concrete in a 100 year perspective

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Abstract

More than 50% of the CO_2 emitted during cement production originates from the calcination of limestone. This CO_2 is reabsorbed during the life cycle of cement based product such as concrete and mortars in a process called carbonation.

The impact that concrete carbonation has in the assessment of CO_2 emissions from cement production has not been fully documented. Specifically, there is a lack of knowledge about the carbonation of demolished and crushed concrete. The existing models for calculating carbonation do not take into account what takes place after the concrete has been demolished. Consequently, the contribution of the cement and concrete industry to net CO_2 emissions may be significantly overestimated.

This paper encompasses theoretical work, laboratory studies, surveys and calculations based on the concrete production in the Nordic countries of Denmark, Iceland, Norway and Sweden. The estimated CO_2-uptake through carbonation of the concrete produced in the year 2003 seems during a 100 year period to amount to a significant proportion of the CO_2 emitted by calcination of the raw mix used to produce the Portland cement used in the concrete.

Keywords: Carbonation; Concrete; Transport properties; Life cycle inventory

1. Introduction

The major part of the CO_2 emission from the production of concrete is related to the production of the cement. In the cement kiln CO_2 is released from calcination of limestone (~ 50%) and from the combustion of fuels (~ 50%), and cement production accounts for approximately 5% of the global CO_2 emissions.

One important issue seldom considered in the environmental debate regarding cement based materials is the re-absorption, or uptake, of CO_2 from the atmosphere. Upon exposure to air concrete will over time absorb atmospheric CO_2. This process is termed carbonation, and is an intrinsic property of Portland cement based concrete. Carbonation will occur during the service life of a concrete structure, as well as after demolition.

Carbonation of concrete is traditionally viewed as a deterioration mechanism, because it lowers the pH of the concrete. The lower pH causes the reinforcement steel to lose its electrochemical protection, so that corrosion may initiate and progress. Indeed for many concrete structures not exposed to severe environment, such as de-icing salts, sulphate ion or freeze–thaw action, the concrete composition and reinforcement cover are designed to avoid carbonation-induced reinforcement corrosion during the service life of the concrete. However, in other types of concrete, carbonation is either negligible (high strength/performance

Table 1
Stable phases in Portland cement paste at different pH (from Lagerblad [14])

Intact concrete	·······································>			Carbonated concrete
Stage 1	Stage 2	Stage 3	Stage 4	Stage 5
$Ca(OH)_2$	–	–	–	–
CSH Ca/Si > 1.5	CSH, 1.5 > Ca/Si > 0.5			SiO_2 with some CaO, Ca/Si < 0.5
–	$Ca(OH)_3$	$Ca(OH)_3$	$Ca(OH)_3$	$Ca(OH)_3$
AFm	AFm	$Al(OH)_3$	$Al(OH)_3$	$Al(OH)_3$
AFt	AFt	AFt	$Fe(OH)_3$	$Fe(OH)_3$
pH > 12.5	pH < 12.5	pH < 11.6	pH < 10.5	pH < 10

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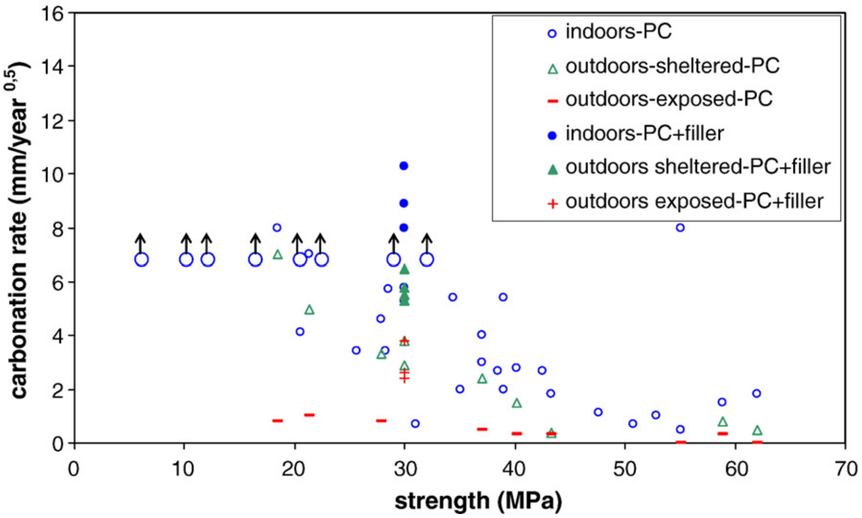

Fig. 1. Carbonation rate constants of concrete calculated based on data for carbonation depths. Note that the larger circles with an arrow represent carbonation rates higher than 6.8 (mm/year$^{0.5}$). Data from [15–17].

concrete), or of no concern at all (paving stone, etc.) because the concrete contains no reinforcement.

Studies on the carbonation of concrete have been mainly focused on durability issues. However, only few studies have focused on the balance between CO_2 emissions and CO_2 uptake in the life cycle of concrete.

Gajda and Miller [1] and Gajda [2] estimated the CO_2 absorbed by concrete in the US based on a survey on the volume of concrete produced, its uses, typical thicknesses, etc. This study did not consider the effect of demolition and subsequent crushing and assumed that only the CH phase in hydrated cements carbonates.

Jacobsen and Jahren [3] estimated the carbonation of concrete in Norway and the amount of CO_2 absorbed by that concrete. Their analysis did not include a survey on concrete uses, and a hypothetical concrete element with average properties was used for the calculations. They also assumed that only the CH phase in hydrated concrete carbonates.

The present paper attempts to estimate the CO_2 uptake in concrete produced in the year 2003 in the Nordic countries Denmark, Iceland, Norway and Sweden during a 100 year period of 70 years of service life followed by an additional 30 years after demolition. The necessary information to calculate the CO_2 uptake is obtained from a literature review of concrete carbonation with focus on the rate of carbonation and on the stability of the hydration products during carbonation, and a survey of the concrete production, demolition rates, recycling practices for the Nordic countries.

Table 2
Carbonation rate constants (CEM I) for various concrete cylinder strengths and concrete exposure conditions as used in the Nordic investigation of CO_2 uptake, Lagerblad [14]

Exposure condition	Compressive strength			
	<15 MPa (mm/(year)$^{0.5}$)	15–20 MPa (mm/(year)$^{0.5}$)	23–35 MPa (mm/(year)$^{0.5}$)	>35 MPa (mm/(year)$^{0.5}$)
Exposed	5	2.5	1.5	1
Sheltered	10	6	4	2.5
Indoors	15	9	6	3.5
Wet	2	1.0	0.75	0.5
Buried	3	1.5	1.0	0.75

2. The carbonation process — chemistry

Virtually all studies on rate of carbonation of concrete are based on the phenolphthalein test. This test is performed by spraying a solution of phenolphthalein onto a concrete section and measuring the distance between the concrete surface and the position of colour change (pH 8.2–10). Consequently, for the purpose of estimating CO_2 uptake in concrete it is necessary to know to what extent the different phases in the cement paste have carbonated at a pH of about 9.

The chemistry of the carbonation process of concrete is conceptually straightforward. It is a through-solution process where, usually, CO_2 from the atmosphere diffuses into the concrete. This diffusion results in a lowering of the pH, destabilizing all the cement hydration products (CSH, AFt, AFm, CH) [4–7]. These hydration products become unstable at different pH-values.

Calcium hydroxide has a solubility product of 9.95×10^{-4} at 20 °C, whereas calcium carbonate has a solubility product of 0.99×10^{-8} at 20 °C. As a result of 4-fold difference in the order of magnitude of the solubility products, calcium hydroxide will readily react with the carbonate ion to form calcium carbonate provided sufficient water is present. Therefore, calcium hydroxide

Table 3
Correction factors for different powder combinations to be multiplied by the rate constants provided for CEM I concrete in Table 2 [14]

Type of addition	Amount of addition (wt.%)					
	0–10	10–20	20–30	30–40	40–50	60–80
Limestone		1.05	1.10			
Fly ash		1.05		1.10		
Silica fume	1.05	1.10				
GGBF Slag	1.05	1.10	1.15	1.20	1.25	1.30

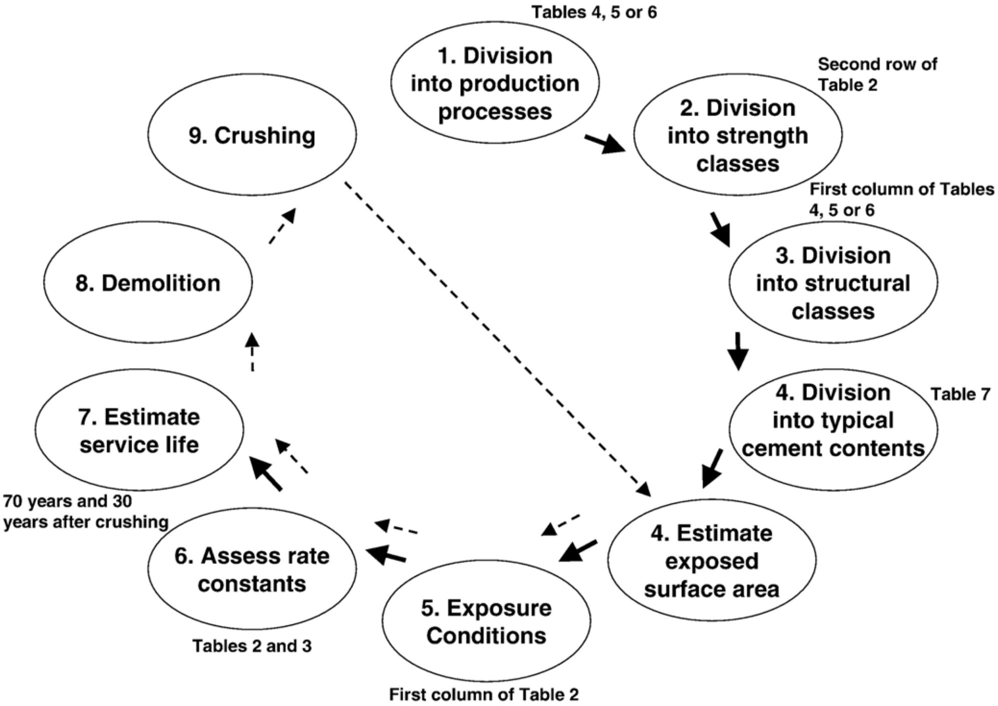

Fig. 2. Methodology for estimatimg the CO_2 uptake from Nordic concrete. Full arrows correspond to service life (70 years) and dotted arrows to secondary life (30 years).

is not normally found in carbonated cement paste. A couple of studies have nevertheless reported that some calcium hydroxide is present in otherwise carbonated cement paste [2,8]. However, the presence of calcium hydroxide in carbonated concrete is not consistent with petrographic observations of real life concrete where the carbonation front is usually very sharp [6].

The CSH phase of non-carbonated concrete has a Ca/Si-ratio in the range of 1.5–1.8 [4,9–11]. Chen et al. [9] showed that the Ca/Si-ratio of CSH decreased with decreasing pH from values around 1.5 down to 0.11 at pH of 9.54. A pH of 9.54 is in the upper range of the phenolphthalein colour change interval suggesting that the Ca/Si-ratio of CSH in carbonated paste is even lower than 1.5. Stronach and Glasser [7], based on thermodynamic modelling of the $CaO–SiO_2–CaCO_3–H_2O$ system at 25 °C, found an invariant point at pH 10.17. At this pH, CSH, calcite and amorphous SiO_2 were found to coexist with the CSH having a Ca/Si ratio of 0.8. At a pH of 9.15, another invariant point was found with only amorphous silica and calcite present [7]. Consequently, it seems reasonable to assume that at least 50% of the CaO present in CSH of non-

Table 4
Ready-mix concrete in Nordic countries: production, exposure conditions and typical thicknesses of structural elements [22]

			Norway		Denmark			Sweden			Iceland	
			Volume (m^2)		Volume (m^2)			Volume (m^2)			Volume (m^2)	
Structural elements	Exposure conditions	Thickness (m)	25–35 Mpa	>35 Mpa	15–20 Mpa	25–35 Mpa	>35 Mpa	15–20 Mpa	25–35 Mpa	>35 Mpa	25–35 Mpa	>35 Mpa
Walls	Indoor	0.18	485,000		327,000	238,000		173,000	510,000		54,000	
	Outdoor sheltered		122,000		82,000	59,000		43,000	127,000		14,000	
	Outdoor		202,000		136,000	99,000		72,000	212,000		23,000	
Slabs	Indoor	0.20	504,000		339,000	245,000		179,000	530,000		55,000	
	Outdoor sheltered		31,000		22,000	15,000		11,000	33,000		4,000	
	Outdoor		95,000		64,000	46,000		34,000	99,000		10,000	
Foundation	Buried	0.24	180,000		121,000	88,000		64,000	189,000		20,000	
	Wet		180,000		121,000	88,000		64,000	189,000		20,000	
Structures	Outdoor sheltered	0.40		120,000			22,000			134,000		10,000
	Outdoor			240,000			44,000			269,000		20,000
	Buried			120,000			22,000			134,000		10,000
	Wet			120,000			22,000			134,000		10,000
Total volume of RMC			2,400,000		2,200,000			3,200,000			250	

Table 5
Pre-cast concrete products in Nordic countries: production, exposure conditions and typical thicknesses of different products [22]

		Norway		Denmark		Sweden		Iceland	
		Volume (m^3)	Thickness (m)	Volume (m^3	Thickness (m)	Volume (m^3	Thickness (m)	Volume (m^3	Thickness (m)
Paving	Outdoor	105,000	0.06	692,000	0.06	21,000	0.08	53,000	0.06
Blocks	Buried	90,000	0.16	154,000	0.08	33,000	0.08	52,000	0.08
Elements	Outdoor exposed	54,000	0.12	279,000	0.06	0	–	16,000	0.15
Pipes	Buried	154,000	0.06	62,000	0.03	113,000	0.03	26,000	0.04
Other	Outdoor exposed	89,000	0.3	90,000	0.03	97,000	0.03	0	–
Total volume (m^3)		492,000		1,277,000		264,000		147,000	

carbonated paste at pH 13–14 will have reacted to form $CaCO_3$ in carbonated concrete at pH around 9.

The stabilities of ettringite and monosulphate at different pH were studied by Gabrisova et al. [12]. Based on potentiometric measurements, they concluded that ettringite is unstable below pH 10.7 and that monosulphate is unstable below pH 11.6. Hemicarboaluminate and monocarboaluminate readily form when the AFm phase carbonates [4]. At pH below 11.92, monocarboaluminate dissolves and the calcium ions are released for carbonation. This was postulated by Damidot [13], who performed a theoretical study on the solubility of CAH phases involving the equilibrium phase diagram at 25 °C.

An attempt to summarize the stability of the different cement paste hydration products with respect to carbonation has recently been made by Lagerblad [14]. He divided the carbonation process into five stages, each having progressively lower pH as shown in Table 1. In the first stage, termed “intact concrete”, the pH of the pore solution is greater than 12.5; and the solid phase assemblage consists of calcium hydroxide (CH), calcium silicate hydrate (CSH), tetra calcium mono-sulfoaluminate (AFm) and ettringite (AFt). When pH drops below 12.5, the CH will be transformed into calcium carbonate (stage 1). When pH drops below 11.6, the AFm will start decomposing (stage 2), and subsequently at pH below 10.5, the ettringite will decompose (stage 3). Finally, at pH below 10, the CSH which has already gradually decalcified in stages 2 and 3 will, given enough time, transform into silica gel and calcium carbonate, thus leaving a paste with essentially no un-carbonated CaO.

If it is assumed that 100% of the CaO found in CH, AFt and AFm, and 50% of the CaO found in CSH have been transformed into calcium carbonate in carbonated concrete, then around 75% of the CaO in the original clinker will have carbonated.

3. The carbonation process — rate of carbonation

Carbonation of concrete starts at the outer surface and moves progressively inwards. The process is controlled by the diffusion of CO_2 into the concrete. The depth of carbonation as a function of time can be described by Eq. (1) [15–17]

$$d = k \times t^{0.5} \tag{1}$$

where, d is the depth of carbonation, k is the rate constant, and t is time.

Although Eq. (1) is generally accepted as a good approximation, higher strength concrete or concrete under exposed outdoor conditions may not fit a square root time relationship. In such cases, the exponent seems to be less than 0.5, corresponding to a smaller increase in carbonation depth with time. This is especially true at later ages [17,18].

The carbonation rate constant has been determined by several authors for a variety of concrete composition and exposure conditions [4,15–17]. A summary of some of the reported rate constants are shown in Fig. 1. For concretes of identical exposure conditions and degree of hydration, the rate of carbonation is essentially determined by the residual or capillary porosity, which in turn largely determines the strength. The carbonation rate constant is normally found to be faster indoors compared to outdoor exposure for all types of concrete [15–17]. This is in agreement with the fact that carbonation rate is sensitive to relative humidity. The carbonation rate is a maximum at 40–80% relative humidity [19].

Recently, Lagerblad [14], based on a review of the literature, suggested that the carbonation rate constant of most common concrete could be chosen from Table 2 by categorizing the concrete based on strength and exposure conditions. As can be seen from Table 2, the rate constant varies from 0.5 to 15, which corresponds to carbonation depths between 4.2 mm and 83.7 mm after 70 years of service life. This reflects the importance of the differences in concrete quality and exposure conditions with respect to carbonation.

The estimated carbonation rate constants in Table 3 are valid for concrete based on the European CEM I[1] cement. This carbonation rate may be affected if the concrete contains supplementary cementitious materials either as part of the cement or added directly at concrete batch plant. In Europe, CEM II cement is abundantly used, with limestone often accounting for about 15% of the cement. Limestone reduces the clinker content per unit volume of concrete, which increases its rate of carbonation. According to Lagerblad [14], this is due to a decrease in the buffering capacity of the paste as there is less hydration product or CaO per unit volume that can be carbonated. Ali and Dunster [16] show data suggesting a slightly increased rate of carbonation after 9 years for C30 concrete made with 5% limestone dust in the powder.

For similar reasons, addition of fly ash, slag or silica fume also results in increased carbonation rates. For concretes with

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[1] Before addition of gypsum the European CEM I shall contain 95% clinker.

Table 6
Pre-cast elements in Nordic countries: production, exposure conditions and typical thicknesses of structural elements [22]

		Norway		Denmark		Sweden		Iceland	
		Volume (m^3)	Thickness (m)	Volume (m^3)	Thickness (m)	Volume (m^3)	Thickness (m)	Volume (m^3)	Thickness (m)
Hollow blocks	Indoor exposed	98,000	0.30	66,000	0.25	0	0.35	10,000	0.30
	Indoor painted	78,000		53,000		88,000		0	
	Indoor covered	20,000		13,000		88,000		0	
Other slabs	Indoor exposed	7000	0.12	12,000	0.25	0	0.20	0	–
	Indoor painted	5000		9000		16,000		0	
	Indoor covered	1000		3000		16,000		0	
Roof	Outdoor exposed	0	–	15,000	0.15	36,000	0.20	0	–
	Indoor painted	0		10,000		0		0	
Walls	Indoor painted	26,000	0.22	82,000	0.15	113,000	0.20	4,000	0.20
	Indoor exposed	9,000		27,000		38,000		0	
Facades	Outdoor exposed	36,000	0.20	56,000	0.20	78,000	0.20	4,000	0.20
	Outdoor painted	2000		3000		4000		0	
Columns/bemas	Indoor covered	7000	0.30	6000	0.30	32,000	0.30	500	0.30
Other	Outdoor exposed	8000	0.15	37,000	0.15	25,000	0.20	500	0.20
	Indoor painted	0	–	0	–	25,000		0	–
Total volume (m^3)		297,000		392,000		559,000		19,000	

fly ash and slag, Ali and Dunster [16] found significantly increased rates of carbonation, relative to concretes without these additives, particularly at indoor and outdoor sheltered exposure conditions (Fig. 1). In another series of experiments Ali and Dunster [16] found significantly increased rate of concrete carbonation with the addition of 5% silica fume. This trend was also reported by Maage and Skjölsvold [20] based on examination of actual concrete structures in Norway.

Recognizing the effect of powder additions on concrete carbonation, Lagerblad [14] suggested that the carbonation rate constant of these types of concretes could be estimated by multiplying the CEM I rate constant by a correction factor. The correction factors that Lagerblad proposed are found in Table 3 [14]. As can be seen, Lagerblad suggested that the higher the amount of powder addition, the higher the correction factor, and the faster the carbonation. He also suggested that the addition of silica fume and slag has a more pronounced influence on the carbonation rate than addition of limestone and fly ash.

The following sections of this paper calculate the CO_2 uptake of concrete in the Nordic countries for a 100 year period based on the rate constants in Table 2 and the correction factors for use of mineral additions in Table 3. Likewise, the assumption that 75% of the original CaO in the clinker of the cement has converted to calcium carbonate in carbonated concrete will be applied.

Table 7
Cement content in ready-mix concrete, pre-cast elements and pre-cast concrete products [22]

		Norway	Denmark	Sweden	Iceland
		kg cement / m^3 concrete			
Ready-mix concrete	15–20 MPa	–	165	220	–
	25–35 MPa	390	240	240	330
	> 35 MPa	290	240	240	330
Pre-cast concrete products		390	390	390	380
Pre-cast elements		350	350	400	350

4. Methodology for calculating CO_2 uptake in concrete

The methodology used for calculating the CO_2 uptake in concrete is illustrated in Fig. 2. After the required information on concrete production and use is gathered, three equations are used to determine the amount of CO_2 absorbed by the concrete:

a) Depth of carbonation. Input: concrete composition, duration of exposure, concrete strength class, exposure conditions. See Eq. (1), Tables 2 and 3.

b) Volume of carbonated concrete. Input: surface area (A) of structural elements, depth of carbonation (d). For example, in the case of ready-mix concrete, the carbonated volume can be calculated from Eq. (2) as:

$$\text{Carbonated concrete } (m^3) = \Sigma(A_{\text{slabs}} \times d) + (A_{\text{walls}} \times d) + (A_{\text{foundations}} \times d) + \ldots \qquad (2)$$

c) Amount of CO_2 absorbed per volume of carbonated concrete can be calculated from Eq. (3). Input: mass of Portland cement clinker per m^3 concrete (C), mass fraction of CaO in the cement clinker (CaO), molar mass of CO_2 and CaO (M), and fraction of CaO available for carbonation (0.75). The mass of Portland cement clinker per m^3 concrete (C) in Nordic countries is given in Table 7. An average CaO content of 65% is assumed

Table 8
Recycled concrete in Nordic countries [23]

	Norway	Denmark	Sweden	Iceland
Rubbled (tons)	812,500	1,000,000	1,140,000	52,266
Recycled concrete rubbled (%)	30	90	60	0
% of RCA				
Bound	1	0	3	0
Unbound-below ground	94	100	92	0
Unbound-above ground	5	0	5	0

Table 9
Particle size distribution of recycled concrete aggregate in Nordic countries [23]

Typical sizes of RCA	Norway (%)	Denmark (%)	Sweden (%)	Iceland (%)
<1 mm	10	21	10	0
1–10 mm	25	30	25	0
10–30 mm	20	44	40	0
>30 mm	45	5	25	0

based on typical values reported by Taylor [4] (avg. CaO=65.3%) and Lawrence [21] (avg. CaO=64.18%).

$$\mathrm{CO_2\ uptake\ (kg\ CO_2/m^3\ concrete)} = 0.75 \times C \times \mathrm{CaO} \times \frac{M_{\mathrm{CO_2}}}{M_{\mathrm{CaO}}}\,(\mathrm{kg/m^3}) \qquad (3)$$

A lifetime of 100 years was adopted for these calculations, where the service life of the concrete was estimated to be 70 years. The same methodology is used for concrete that is demolished, crushed and reused after service life, but with additional information regarding volume, exposure conditions and surface area of crushed concrete. The carbonation of demolished concrete was calculated for 30 years.

5. Concrete production, use and demolition in the Nordic countries

5.1. Survey on concrete production

In order to calculate the volume of concrete that carbonates during service life and after demolition, it is necessary to know the volume of concrete produced, the exposed area, the thickness of different concrete structures and their exposure conditions. Therefore, a comprehensive survey of concrete production, use and demolition in Nordic countries was performed. The main sources of information for this survey were the European Ready-Mix Concrete Organization (ERMCO) database on the statistics for ready-mixed concrete and local statistics from producers and different concrete associations. However, information on concrete usage, exposure conditions and structural dimensions (thicknesses) of ready-mix and pre-cast concrete was difficult to obtain or not available at all. In those cases, estimates based on personal judgment had to be performed.

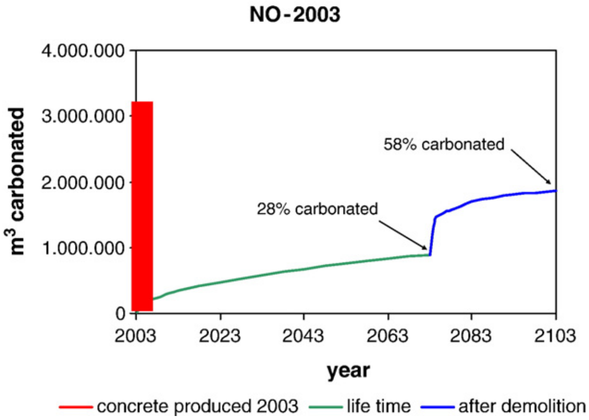

Fig. 4. Carbonation of concrete produced in Norway during service life and after demolition [24].

The concrete produced in Nordic countries during 2003 was divided into three production types: ready-mix concrete, pre-cast concrete elements, and pre-cast concrete products. Within each production type, strength classes (C-15, C-35, etc.) and structural elements (walls, foundations, paving stones, etc.) were identified. A typical mixed design was assigned to each strength class, and typical thicknesses and exposure conditions were assigned to each structural element. Tables 4–7 show these results.

5.2. Survey on concrete demolition

A survey on the demolition of concrete in Nordic countries was performed. The aim of the survey was to quantify the amount of demolished concrete today, and to find out what percentage of demolished concrete is recycled for secondary use. Additionally, data on the uses of recycled concrete and the sizes of recycled concrete aggregate (RCA) were gathered.

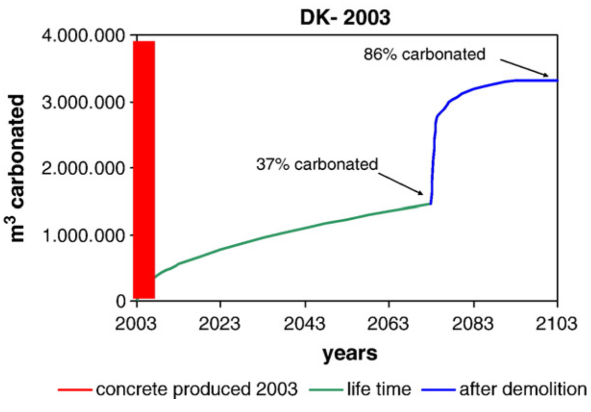

Fig. 3. Carbonation of concrete produced in Denmark during service life and after demolition [24].

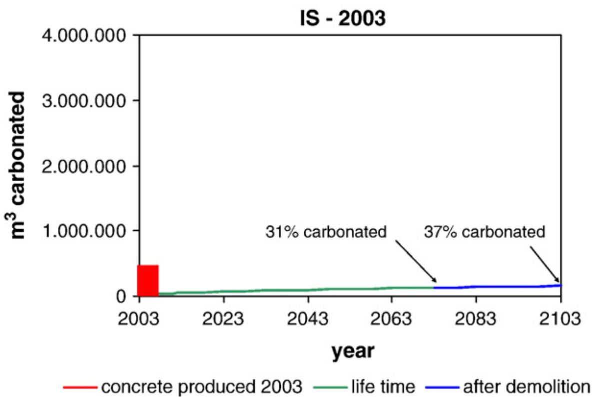

Fig. 5. Carbonation of concrete produced in Iceland during service life and after demolition.[24].

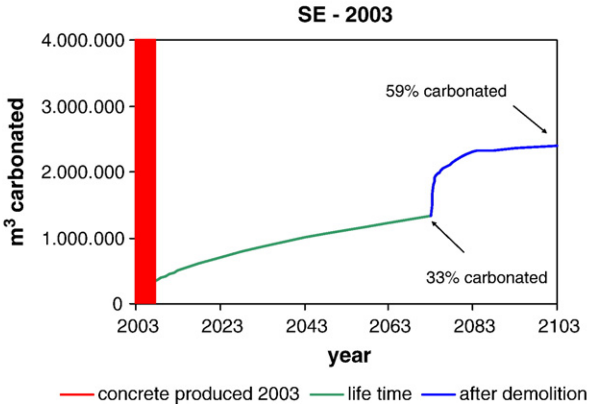

Fig. 6. Carbonation of concrete produced in Sweden during service life and after demolition [24].

The volume of concrete rubble (i.e. demolished concrete) generated in Denmark, Sweden and Norway in 2003 amounts to approximately 15% of the concrete produced per year (7% in the case of Iceland). The percentage of the concrete produced in 2003 that will be demolished after its service life was estimated to be between 72 and 87%. All concrete that is not wet (submerged) or buried is assumed to be demolished.

Concrete rubble is generally crushed at crushing plants and stockpiled for a period between 2 weeks and 4 months. This aggregate is mainly used in road construction, either as part of the sub-base or as a top layer in minor roads. Currently, from 0% (Iceland) to 90% (Denmark) of demolished concrete is recycled in Nordic countries as shown in Table 8. Norway and Sweden expect that by the year 2010 approximately 70% of their demolished concrete will be recycled, and therefore a 70% recycling rate was used in the calculations for concrete produced in 2003 by these two countries. Consequently, out of the concrete produced in 2003 in Denmark, Norway, Sweden and Iceland 78%, 51%, 43% and 0% become RCA, respectively.

Data on the uses of recycled concrete aggregate in Nordic countries were gathered as part of the survey and are also presented in Table 8. Note that recycled concrete aggregate is primarily used in below ground applications, which results in low rates of carbonation. Additionally, data on the typical size distribution of recycled concrete aggregate were gathered and are presented in Table 9.

6. Concrete carbonation and CO$_2$ uptake within 100 years

The volume of carbonated concrete after service life and demolition is shown in Figs. 3–6. The differences in percentage of carbonated concrete between countries are mainly due to the use of concrete of different strength classes in ready-mix concrete and to the recycling practices in each country.

Calculations on concrete that is exposed to aggressive environments show that only 4.8% of this type of concrete is carbonated after 70 years (data from Denmark). This translates into an average carbonation depth of 9.6 mm after 70 years. The low volume of carbonated concrete is primarily due to the fact that infrastructures are built with concrete of the highest strength class (>35 Mpa), and therefore have a very low carbonation rate (see Table 2). In addition, infrastructure concrete is thicker than other types of concrete, i.e. it has a small exposed surface area.

After 100 years, between 58% and 86% of concrete in Sweden, Norway and Denmark will be carbonated based on these calculations. Iceland places demolished concrete in a landfill rather than crushing it, and for this reason only 37% of Icelandic concrete is carbonated after 100 years. The large volume of carbonated concrete after demolition in Denmark is due to the fact that 90% of demolished concrete is crushed and recycled.

The effect of crushing on the volume of carbonated concrete can be seen in the evolution of the surface area of a 10 m long wall when it is demolished into concrete rubble and crushed as RCA (Fig. 7). The volume of carbonated concrete increases exponentially when concrete is demolished, since carbonation is directly related to the exposed surface.

The CO$_2$ uptake of concrete produced in Denmark, Sweden, Norway and Iceland after 100 years is presented in Table 10

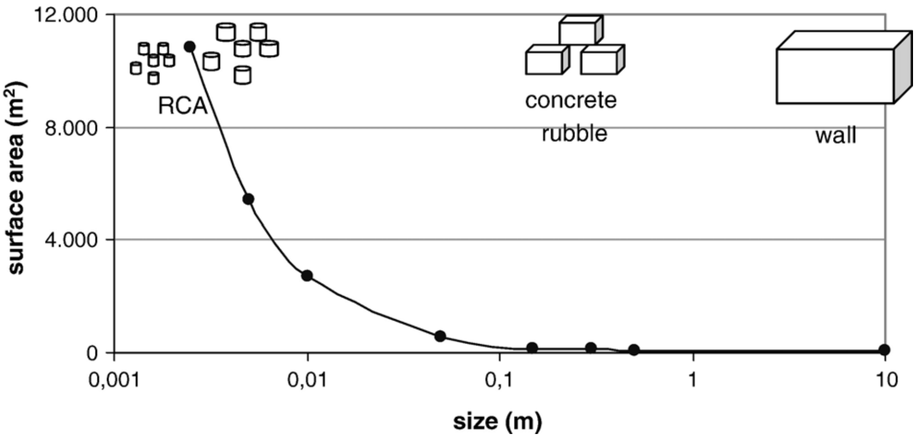

Fig. 7. Evolution of surface area from a wall to RCA (length=10 m; thickness=0.18 m; height=2.5 m).

Table 10
CO_2 balance of concrete produced in Nordic countries

	Denmark (2003)	Norway (2003)	Sweden (2003)	Iceland (2003)
CO_2 uptake after 100 years (metric tons)	340,000	220,000	240,000	21,000
CO_2 emission from calcination (metric tons)[a]	600,000	675,000	730,000	58,000

[a] CO_2 emission from the production of the cement used in Nordic countries. Source: IBRI; Aalborg Portland; Norcem and Cementa.

along with the CO_2 emitted during calcination of the cement used in that concrete.

7. Discussion

The CO_2 absorbed by all concrete present in the USA was estimated by Gajda and Miller [1] and Gajda [2] based on a survey on the volume of concrete produced, its uses, typical thicknesses, etc. The carbonation rate constants used in these studies are taken from experimental data from a set of 500 individual samples tested in their laboratory. These studies consider only the carbonation of structures during their service life (100 years) without considering concrete demolition and recycling. Gajda and Miller [1] conclude that 38% of concrete produced in the US in the year 2000 will be carbonated after its service life. This agrees reasonably well with the percentages calculated in this study that range from 28% in Norway to 37% in Denmark. Note that the percentages in Nordic countries are slightly lower since they were calculated over a period of 70 years rather than 100 years.

Jacobsen and Jahren [3] estimated the carbonation of concrete in Norway and the amount of CO_2 absorbed by that concrete. Their study estimated that 16% of the CO_2 emitted due to calcination is reabsorbed by concrete during its service life. However, the analysis of Jacobsen and Jahren does not include a survey on concrete uses, and a hypothetical concrete element with average properties was used for the calculations. In addition, average values for the depth of carbonation of old structures and new structures were used as maximum carbonation depth, and the effect of concrete recycling on carbonation was taken into account by assuming that only 10% of concrete was demolished and recycled.

Table 11 compares the approaches and assumption made in this with those of Gajda [2] and Jacobsen and Jahren [3]. Note that [2] is a follow up paper from [1], and that Table 11 shows results only from the later study.

There are two main differences between the present study and previous studies. The first difference is the percentage of calcium available for carbonation. This study assumed that 75% of CaO will be available for carbonation, while the other two studies assumed that only the calcium present in CH carbonates (32 to 37% of CaO). In fact, Gajda and Miller [1] acknowledge that CSH carbonates but they only include the carbonation of CH in their calculations. If, as previously discussed, the change in pH to approximately 9 defines the carbonation depth, the assumption made in this study is probably more accurate.

The percentage of CaO available for carbonation was further reduced to 21% in the study by Gajda [2] in order to account for the presence of pozzolanic material in concrete. In general, CH decreases, the amount of CSH increases, and the CaO present in the silica gel increases after the pH drops in these types of concrete. Because [2] only calculates the carbonation of CH, the available calcium for carbonation decreases substantially in their study when CH decreases due to pozzalanic reaction. This study considers that 100% of the calcium present in aluminate phases and 50% of the calcium present in CSH carbonates. Hence, although the effect of pozzolans on carbonation was not considered, the decrease in the amount of available calcium for

Table 11
Comparison of different studies on CO_2 uptake in concrete

	This study	Carbonation of concrete in USA (Gajda [2])	Carbonation of concrete in Norway (Jacobsen and Jahren [3])
Carbonation factors mm/(yr)$^{0.5}$	See Table 1	6.9–5.4–3.8 (for strength classes 21, 28 and 35 MPa, respectively)	Assumes a maximum carbonation possible after the service life of the structure
% of concrete carbonated after service life	28% (NO)–37% (DK)–33% (SE)–31% (IS) (service life calculated for 70 years)	38% (service life calculated for 100 years)	Estimates that 19% of concrete carbonates after service life.
% of concrete demolished	73% (NO)–88% (DK)–72% (SE)–79% (IS)	0%	10%
% of concrete carbonated after demolition	58% (NO)–86% (DK)–59% (SE)–37% (IS)	Demolition is not accounted for	Estimates that 100% of demolished concrete carbonates.
% CaO available for carbonation	100% (CH+AFm+AFt) +50% (CSH)=75% CaO	100% CH+0% (CSH+AFm+AFt) =~38% CaO	100% CH+0% (CSH+AFm+AFt) =~32% CaO
% CaO available for carbonation due to effect of pozzolans	Effect of pozzolans is not accounted for	CaO available for carbonation is reduced to ~21% due to presence of pozzolans.	Effect of pozzolans is not accounted for
Wet and buried concrete	Carbonates at a very slow rate	Does not carbonate	Assumes that 10% of concrete does not carbonate, but does not do a detailed analysis of uses of concrete.
CO_2 uptake compared to the CO_2 emitted during calcination (%)	57% (DK)–33% (NO–SE)–34% (IS) The CO_2 absorption without demolition after 70 years in Denmark is 2.4 times lower than considering demolition, i.e. 24% of the CO_2 emitted (instead of 57%)	7.6%	16%

carbonation will be small. Nevertheless, this effect should be addressed in further studies.

The second difference between these studies is the effect of demolition and concrete recycling on carbonation. Concrete demolition and recycling can have a large impact in the degree of carbonation and the amount of CO_2 uptake by concrete. This effect is largely dependant on the recycling practices in each country. For instance, only the effect of demolition and crushing results in a CO_2 uptake that is 2.4 times larger than before demolition (data from Denmark). Recycling practices differ considerably even among Nordic countries. The usage and exposure conditions of crushed concrete will determine whether carbonation is possible. Many countries reuse crushed concrete as aggregate in new concrete. In those cases, and unless concrete is exposed to air for a significant period of time, carbonation will be negligible.

8. Conclusions

The effect of carbonation on the net CO_2 emissions related to concrete can be significant. This effect is highly influenced by the way concrete is handled after demolition. Demolition and subsequent crushing of concrete after service life increases the amount of carbonation significantly.

According to these results, demolition and crushing of concrete should be promoted, for example by implementing handling procedures for crushed concrete that maximize CO_2 uptake such as exposing crushed concrete prior to using it in embankments (low carbonation rate).

The effect of concrete carbonation on the net CO_2 emissions of cement-based materials should be introduced in life cycle screenings of these types of products, which can have a significant influence on the criteria for environmental labelling and on the selection of materials based on principles of environmentally correct design.

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