

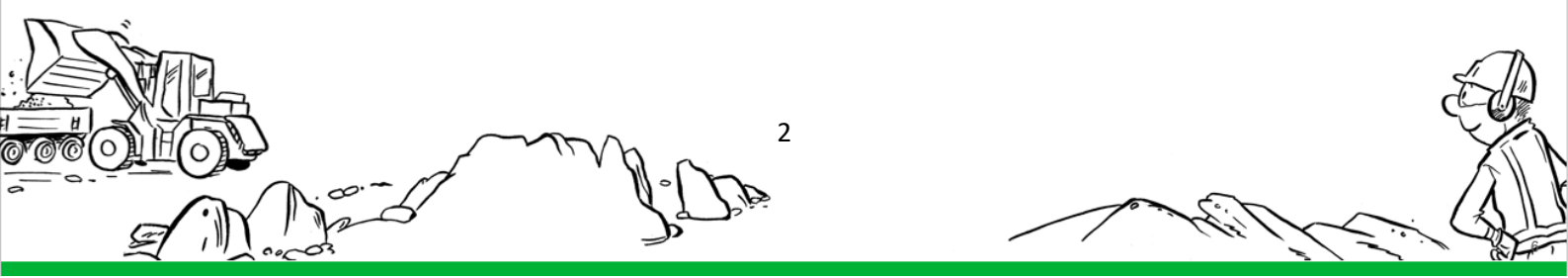
# CO<sub>2</sub>NCRETE SOLUTION

## – Report Q1/2020, Literature Summary



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## Environmental Impact of the Cement Industry

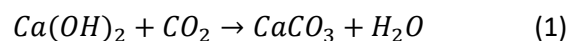
The concentration of carbon dioxide (CO<sub>2</sub>) in the atmosphere has increased at an exponential rate since the beginning of industrialization. It is said to have increased by 40 % from the preindustrial era; from about 280 ppm to about 406 ppm in the year 2017. Currently the increase is in the range of 2,2 ppm per year, which is expected to elevate the global average temperature 2-3 C° when the CO<sub>2</sub> concentration reaches 450 ppm by the year 2050.

Concrete has been used in different forms for over 2000 years and it is the most used construction material in the world, about 25 billion tons of concrete was used back in 2009 and the pace has been growing throughout time. The production of the primary binding agent in concrete, cement, has increased to 30-fold since 1950 and 4-fold since 1990 to date. Though concrete contains only 10-20 % of cement, it causes most of the emission in concrete production, as can be seen in table 1 below. Cement production contributes on estimation 5-8 % of all global greenhouse gas emissions, 50-70 % of which is during the calcination of limestone. From the stoichiometry of calcination, it can be seen that 510 kg of CO<sub>2</sub> is released per 1 ton of clinker, bringing the total emissions to about 0,7-1,0 tons CO<sub>2</sub> / ton of clinker.

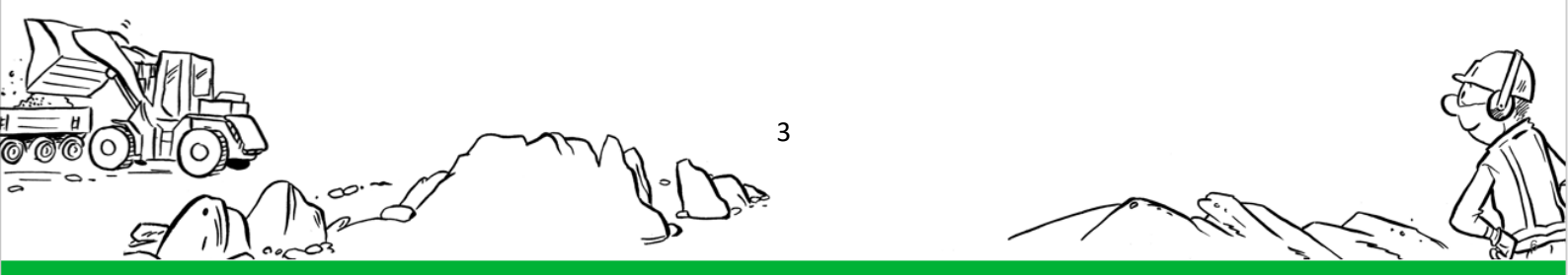
Table 1: Listed the emitted CO<sub>2</sub> and the need of energy of different parts on concrete production per 1 m<sup>3</sup> of concrete.

|                           | CO <sub>2</sub> [kg] |      | Energy ([MJ]) |      |
|---------------------------|----------------------|------|---------------|------|
| <b>Cement</b>             | 92,3                 | 91 % | 551,2         | 83 % |
| <b>Aggregate (fine)</b>   | 2,5                  | 2 %  | 33            | 5 %  |
| <b>Aggregate (coarse)</b> | 2,1                  | 2 %  | 34,7          | 5 %  |
| <b>Admixtures</b>         | 1,5                  | 1 %  | 10,8          | 2 %  |
| <b>Water</b>              | 0                    | 0 %  | 0             | 0 %  |
| <b>Transport</b>          | 2,5                  | 2 %  | 34            | 5 %  |

The reaction product of calcination, cement clinker, however is not stable and hardened concrete will react with atmospheric CO<sub>2</sub> in a reverse reaction called carbonation (described in equation 1 below), where calcium hydroxide (Ca(OH)<sub>2</sub>) reacts with carbon dioxide to form calcium carbonate (CaCO<sub>3</sub>). This on a high level, simple reaction is in reality rather complex and very sensitive to outside conditions; however, it is clear that CO<sub>2</sub> is sequestered back to concrete with time.



In Finland the production of cement is about 1,5 million tons annually; which contains the production of 1,2 million tons of clinker. The related calcination emits about 0,6 million tons of carbon dioxide resulting in total CO<sub>2</sub>-emissions of about million 0,9 tons. This is around 1,3 % of all national greenhouse gas emissions.



## Concrete Chemistry and Carbonation

Concrete is a composite material consisting of cement, fine and coarse aggregate and water. Chemical admixtures are used in small portions to improve the mix to certain applications and various amounts of additives are used to replace cement. A chemical reaction between water and cement called hydration causes the hardening of concrete.

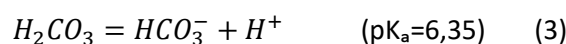
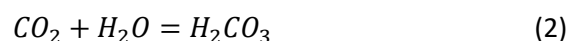
Limestone consists mainly of calcium carbonate (CaCO<sub>3</sub>) and accounts for about 95 % of raw materials in cement clinker. In production the raw materials are heated to near 1000 °C, which causes the limestone to calcinate (equation 1). Then the materials enter the clinker kiln where they reach a temperature of 1450 °C and turn into clinker. This phase, calcination and burning, is the major cause of emissions of the cement industry.

Clinker consists on average of about 65 % of CaO, a major part of which originates from CaCO<sub>3</sub>. With the assumption of all CaO originating from CaCO<sub>3</sub>, the production of 1 ton of clinker releases 510 kg of CO<sub>2</sub>. This sums up as total emissions in this phase to 670-770 kg of CO<sub>2</sub> per ton of clinker.

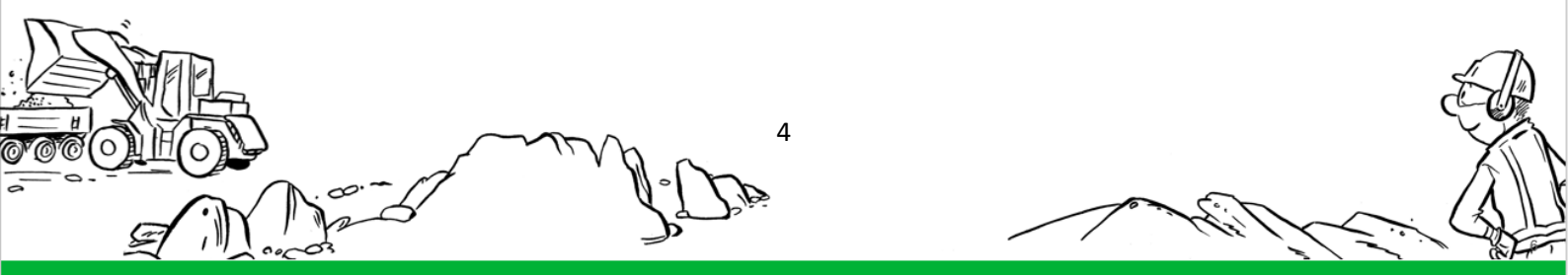
When hydrated cement is exposed to atmospheric conditions it starts to react with carbon dioxide, this phenomenon is called carbonation. The reaction lowers the pH of concrete which is why the phenomenon has been studied for a long time in the context of enforcement steel corrosion. The sequestration of CO<sub>2</sub>, though known for long, has only recently been identified as a carbon sink.

Gaseous CO<sub>2</sub> cannot react with hydrates in the cement directly as it must be dissolved in water in order to form carbonate ions. Carbonate ions will then react with the Ca-ions in the pore water, which makes the reaction sensitive to relative humidity. In neutral water CO<sub>2</sub> forms bicarbonate but as concrete is a very alkaline environment the bicarbonate dissociates and forms carbonate ions.

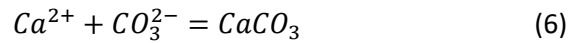
Carbonation includes chemical processes such as dissolution of portlandite, decalcification of calcium silicate hydrates (C-S-H) and dissolution of mono- and tri-substituted calcium aluminate hydrates (AFm and AFt). These reactions take place in the aqueous phase, which is why the presence of water is essential. The reaction is described in equations below:



In normal atmospheric conditions only a minor part of the dissolved CO<sub>2</sub> will hydrolyze to carbonic acid (H<sub>2</sub>CO<sub>3</sub>) but depending on the pH in the pore water, bicarbonate (HCO<sub>3</sub><sup>-</sup>) or carbonate ion (CO<sub>3</sub><sup>2-</sup>) will form.



HCO<sub>3</sub><sup>-</sup> ions form the majority in the pore water in the carbonated zone (intermediate pH) as HCO<sub>3</sub><sup>-</sup> ions will be present in the uncarbonated cement paste (high pH). The actual carbonization process, the phase transformations of which are presented in table 2, can be described:



*Table 2: Phase changes in the carbonation process. The CH, AFm and Aft will act as buffer phases and will be decomposed. The C-S-H will release CH, which carbonates resulting in a lower CA/Si ratio, which is linked to the lower pH.*

| Intact concrete | 1st stage | 2nd stage   | 3rd stage | Carbonated    |
|-----------------|-----------|-------------|-----------|---------------|
| CH              | -         | -           | -         | -             |
| C-S-H (1)       | C-S-H (1) | C-S-H (2)   | C-S-H (3) | SH (some CaO) |
|                 | CC        | CC          | CC        | CC            |
| Afm             | Afm       | Aft/Al(OH)3 | Al(OH)3   | Al(OH)3       |
| Aft             | Aft       | Aft         | Fe(OH)3   | Fe(OH)4       |
| pH>12,5         | pH<12,5   | pH<11,6     | pH<10,5   | pH<10         |



## CO<sub>2</sub>-binding Capacity of Concrete

Theoretically all CO<sub>2</sub> released in calcination can be absorbed back into the cement to form stable CaCO<sub>3</sub> and this can be the case in a geological time scale. However, as studies show, this is not the case in real life concrete structures. So, we must make distinction between theoretical and normal binding capacity of carbon dioxide into cement.

From the simplified equation of carbonation, CO<sub>2</sub>+CaO=CaCO<sub>3</sub>, it is evident that the stoichiometric ratio between all elements is 1:1; the formation of one CaCO<sub>3</sub> molecule, one CaO and one CO<sub>2</sub> molecule is consumed. From this it can be stated that CO<sub>2</sub> binding capacity is directly related to the amount of CaO in the clinker. OPC contains 95-100 % cement clinker, depending on the amount of gypsum added. On average the amount of CaO is 64 % of the clinker and the amount available for carbonation is about 60 %, due the addition of gypsum.

95 % of 1 ton of Portland cement is cement clinker; from which 617 kg of CaO can theoretically be converted to CaCO<sub>3</sub> through reaction with atmospheric CO<sub>2</sub>.

$$CO_2^{Theoretical} = 617 \text{ kg} \times \left(\frac{M_{CO_2}}{M_{CaO}}\right)^2 = 617 \text{ kg} \times \frac{44}{56} = 485 \frac{\text{kg}_{CO_2}}{\text{ton}_{cement}}$$

Defining the normal CO<sub>2</sub> binding capacity of cement, the quantity of formed hydrate phases needs to be considered as well as the chemistry of carbonation of each hydrate phase. The principle CaO-containing hydrate phases are assumed to make 95 % of the cement paste; shown in table below. In calculations, the carbonation degree is used as in table 3.

In the table it is shown that the available CaO content for carbonation is around 72 % of the total CaO.

Table 3: available CaO in different phases of cement; giving the normal maximum CO<sub>2</sub> uptake potential.

| Element  | CH   | C-S-H | AFm  | Aft  | Sum |
|--|------|-------|------|------|-----|
| Phase content                                    | 25   | 50    | 10   | 10   | 95  |
| CaO molar ratio                                  | 0,76 | 0,42  | 0,36 | 0,27 | -   |
| CaO content in hydrated cement (%)               | 19   | 21    | 3,6  | 2,7  | 46  |
| Assumed carbonation degree                       | 1    | 0,5   | 0,75 | 0,5  | -   |
| CaO available for carbonation in hydrated cement | 19   | 11    | 2,7  | 1,3  | 33  |
| CaO available relative to total CaO (%)          | 41   | 23    | 5,8  | 2,9  | 72  |

From the calculations and estimation mentioned, the theoretical binding capacity can be converted to normal binding capacity.

$$CO_2^{Normal} = CO_2^{Theoretical} \times CaO_{\% \text{ available}} = 485 \frac{\text{kg}_{CO_2}}{\text{t}_{cement}} \times 72 \% = 349 \frac{\text{kg}_{CO_2}}{\text{t}_{cement}}$$



## Carbonation Mechanism and Kinetics

The carbonation mechanism depends on the proportions of different phases in the cement paste, which correlate with the binder type, curing, and water/binder ratio of the paste. The reaction will occur in the water phase and depend on the solubility and speed of diffusion through the material. Concentration differences control the diffusion and so diffusion processes and the structure of the carbonated layer need to be considered. In the process carbon dioxide and carbonate ions diffuse inwards to the material, of which the gas diffusion is much faster. The carbonation speed is thus related to the humidity in the pore system. In concrete too dry, the gas diffuses fast to the material, but the actual reaction is unable to happen with no water. In fully water saturated concrete, only carbonate ions can move, which makes the carbonation very slow. The optimum humidity depends on the pore structure of concrete; in general, low water/binder-ratio gives a denser structure which makes carbonation slower.

In carbonation the transformation of CH to calcite gives a volume change of 11 % and to the metastable vaterite of 14 %. The volume changes will affect the porosity in the carbonate layer and thus the speed of carbonation. This is an indication that the rise in volume of calcite fills the empty space in the capillary system, densifying the cement paste.

Submerged concrete will also carbonate but at a very low pace and in another mode. Carbon dioxide has difficulties entering the pore system as the structure is filled with water. This means that the carbonate ions in water need to be taken to account, not the CO<sub>2</sub> gas. When the concentration of carbonate ions is low, the carbonation is controlled by outward diffusion of Ca-ions and the carbonate will form close to the concrete surface.

Carbonation in concrete will take place whenever there is carbon dioxide and water available. The speed is dependent on the pace that carbon dioxide and/or carbonate ions move in the concrete and react with the cement paste. In normal conditions a constant amount of CO<sub>2</sub> can be assumed to be in contact with the surface.

Fick's laws of diffusion can be used to describe the advancement of carbonation in concrete. The carbonation coefficient, often called the k-value, takes all crucial parameters of concrete properties and exposure environment into account, carbonation can be described in a simple equation.

$$d_c = k\sqrt{t} \quad (7)$$

, where  $d_c$  is depth of carbonation, k is the carbonation coefficient (k-value) and t is time in years. With k-values for the most important types of concrete and conditions it is possible to get a good estimate of CO<sub>2</sub> uptake

Very dry concrete doesn't carbonate as water is needed in the pore system to form ions that react and form calcite. Carbonation is very slow in opposite conditions, in wet environments as water fills the porous system disabling the diffusion of gas. Thus, the optimal relative humidity is in between these extreme conditions and dependent of water/binder-ratio, degree of hydration and type of binder. The maximum speed of carbonation is achieved when the RH is in the range of 60-80 % (inside the concrete).



Diffusion rate and so the carbonation rate increases in elevated temperature, which means that indoor climate and exposure in warmer regions will lead to faster carbonation.

Diffusion takes place in the cement paste and not the aggregate, assuming the aggregate is dense. The amount of cement paste does not affect the speed of carbonation, but it must be taken into account when calculating CO<sub>2</sub> uptake.

Lower water/cement-ratio or water/binder-ratio and high level of hydration give denser concrete because of less porosity in the structure. It will also result in denser carbonate products and consequently slow down carbonation in all environments. A denser concrete will result in denser carbonation layer which both will hinder the diffusion of CO<sub>2</sub> into the concrete. A lower degree of hydration will give a more porous paste structure. This is presumably the case in indoor environment with a relatively low RH. In outdoor structures, prolonged hydration will make the paste denser which leads to a denser carbonation layer, resulting in slower carbonation.

A generalization can be made and the relationship between concrete strength and porosity can be used to enable calculation for different cement types in most cases.

The exposed area of a concrete surface is important as carbonation is a reaction from the surface inwards. Gas will diffuse through the porous structure but cracks and the interface between paste and aggregate must be taken into account as well. The interface is more porous than the plain paste and faster carbonation can be observed in these thin areas. Cracks help the gas to mitigate deeper to begin with which enables even more surface for carbonation.

A higher concentration of carbon dioxide in the atmosphere will increase the rate of carbonation. This means that carbonation rate is higher in suburban areas with more CO<sub>2</sub> emissions as well as indoors. The effect of higher carbon concentration is considerable and this needs to be taken into account in calculations of carbon sinks.

Carbonation rate is dependent of the diffusion rate of gas through the surface into the material. Naturally coverings, such as paint or wallpaper, will affect the diffusion. Application to protect outside surfaces will control the relative humidity in the structure and so they will speed up the carbonation. In inside structures surface treatments work the opposite in respect to carbonation.

Carbonation is a surface phenomenon so the area of the exposed surface must be known in order to calculate the CO<sub>2</sub> uptake of a structure. Concrete type and the environment that the concrete is exposed to must be identified. Parameters mentioned, such as additives, different binders, surface coverings and CO<sub>2</sub> concentration give additional correction values for the k-value. A matrix for k-values for different strength concrete in different environments is presented in table below.

*Table 4: k-values for different compression strengths and environments*

| Strength         | <15 Mpa | 15-20 Mpa | 25-35 Mpa | >35 Mpa |                        |
|------------------|---------|-----------|-----------|---------|------------------------|
| <b>Exposed</b>   | 5       | 2,5       | 1,5       | 1       | [mm/a <sup>0,5</sup> ] |
| <b>Sheltered</b> | 10      | 6         | 4         | 2,5     | [mm/a <sup>0,5</sup> ] |
| <b>Indoors</b>   | 15      | 9         | 6         | 3,5     | [mm/a <sup>0,5</sup> ] |
| <b>Wet</b>       | 2       | 1         | 0,75      | 0,5     | [mm/a <sup>0,5</sup> ] |
| <b>Buried</b>    | 3       | 1,5       | 1         | 0,75    | [mm/a <sup>0,5</sup> ] |





## Carbonation of Recycled Concrete

In Finland about 1 million tons of concrete is recycled yearly. Practically all recycled concrete aggregate (RCA) is sold to use as road base and to other construction applications without additional financing from third parties. Today the use of RCA always requires an environmental permit.

Natural carbonation takes place from the outer layer of concrete. As the process slows with time, eventually approaching zero, the demolition and recycling enables significantly more exposed and uncarbonated surface to react with CO<sub>2</sub>. A large portion of cement paste will form powder so the quantity of exposed cement should be considered greater than just the larger particles of cement paste. This will increase CO<sub>2</sub> uptake considerably. In demolished concrete, the normal rules of carbonation will apply, meaning that the key factors affecting CO<sub>2</sub> uptake are the environment and the properties of the original paste. Several studies report that the smaller the particle size, the more extensive the carbonation.

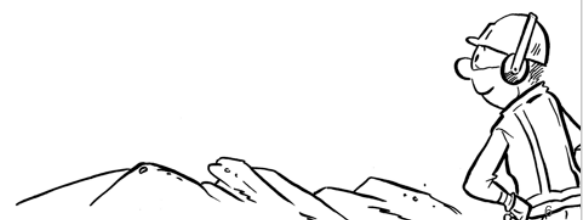
The amount of concrete recycled, the particle size, the exposure conditions of the crushed concrete and the time that the aggregate is allowed to be exposed are key factors on the CO<sub>2</sub> uptake in the post demolition phase. Several studies show the great importance of this phase, when carbon sequestration is considered; in a study by Piqueras and Gonzales the total uptake increased from 22 % of the total emissions to 47 %. Also, worth noting is that carbonation degrees in the range of 60-90 % were achieved in accelerated tests by Engelsen & al. which indicates a possible uptake of 60-80 % of calcination emissions for the life cycle of concrete.

In addition to more sequestered CO<sub>2</sub>, it is possible to improve the quality and strength of the concrete aggregate with carbonation. It should be noted that it is mandatory to remove steel reinforcements from RCA, which means that concrete needs to be demolished into rather small particles in order to achieve convenient removal. This means that plenty of fresh, uncarbonated surface is exposed during demolition, even without considering the carbon dioxide uptake.

Concrete aggregate has not been widely studied when it comes to CO<sub>2</sub> sequestration but as described above this phase in concrete life cycle presents great potential as a carbon sink. The methods and policies to concrete recycling and the use of recycled concrete should be considered to enhance the carbon sequestration.



*Picture 1: A sound barrier made of recycled concrete rocks at a recycling plant of Rudus.*



## Studies on CO<sub>2</sub> uptake

Great variation in carbon dioxide sequestration related studies can be observed which is clear indication of the complexity of the actual phenomenon of carbonation. The reaction is highly influenced by the properties of the actual carbonating concrete and the environment it is exposed to.

Some disagreement can be found in the amount of CaO available for carbonation; the relation often called carbonation degree. Though statements like in Felix & Possan, 2018, that 100 % of the CaO in the cement could carbonate as opposed to that of Galan & al. where it was stated that about 20 % of the calcium oxide could react with atmospheric CO<sub>2</sub> make variation on the topic, most studies agree that the realistic carbonation degree is in the range of 70-75 % of the CaO in the cement paste.

Attention must be paid when comparing these different studies as there are many different methods of analyzing the CO<sub>2</sub> uptake. Some calculations describe the status today, how much the current concrete structures absorb carbon dioxide, which can be compared to the emissions of the very same concrete or perhaps to the calcination emissions of that very year. Xi & al. calculated the total global cement emissions from 1930 to 2013 and analyzed the uptake characteristic to different regions and concluded that 43 % of the calcination related emissions would be absorbed in the existing concrete structures. Andersson & al. calculated the cement emissions generated in 2011 and compared it to the absorption of existing concrete in that year; they concluded that 27 % of the calcination emissions and 17 % of the total emissions were absorbed.

The life cycle of concrete can be analyzed, when a certain amount of cement is followed from cradle to grave and the CO<sub>2</sub> balance is considered. These calculations give good, overall descriptions that can be used in various regions and circumstances and give comprehension on what could be done to enhance the carbon uptake. According to Kjellsen & al. 57 % of the calcination related emissions can be absorbed to concrete during a lifecycle of 70 years in service and a 30 years post demo phase. Possan & al. stated that 40-90 % of the emissions could be absorbed to concrete during a life cycle of 100 years, dependent on the concrete types used.

The crushing of demolished concrete makes the recycling phase a crucial part of the life cycle of concrete when it comes to CO<sub>2</sub> sequestration. When concrete is crushed, plenty of uncarbonated surface is exposed that can react with atmospheric carbon dioxide; with proper particle size and exposure time, as much as 80 % of the calcination emissions could be absorbed back into the concrete. Optimization of this recycling and second phase can more than double the CO<sub>2</sub> uptake during the lifecycle of concrete.

In table 5 are summarized conclusions of CO<sub>2</sub> uptake in different studies. As said, direct comparison is impossible to make as variations are great and different parameters are used. However, it can be concluded that concrete forms a potentially very large carbon sink that can be further enhanced with proper recycling and second phase usage.



| Reference                   | Absorption  |           | Time         |           | Point of View               | Details  |
|-----------------------------|-------------|-----------|--------------|-----------|-----------------------------|--|
|                             | Calcination | Total     | Service life | Post demo |                             |  |
| Andersson & al., 2013       | 27 %        | 17 %      | x            | -         | Life Cycle                  | 70 % carbonation degree  |
| Byrne & Nolan, 2016         | 15 %        | 9 %       | 8 weeks      | -         | -                           |  |
| Collins, 2013               | 55-65 %     | 34-41 %   | x            | x         | Life Cycle                  | 75 % carbonation degree  |
| Engelsen & al., 2005        | 60-80 %     | 38-50 %   | x            | x         | Life Cycle                  | carbonation degree in RCA 60-90 %  |
| Engelsen & al., 2016        | 24 %        | 15 %      | 100 a        | 100 a     | Uptake by existing concrete | 70 % carbonation degree  |
| Engelsen & Justnes, 2014    | 24 %        | 15 %      | 100 a        | 100 a     | Life Cycle                  |  |
| Felix & Possan, 2018        | 74 %        | 46 %      | 70 a         | 30 a      | Life Cycle                  | 100 % carbonation degree   |
| Fitzpatrick & al., 2015     | 16 %        | 10 %      | 100 a        | -         | Life Cycle                  |  |
| Jacobsen, 2001              | 11 %        | 7 %       | x            | x         | Uptake by existing concrete | 60 % carbonation degree  |
| Kaliyavaradhan & Ling, 2017 | 53 %        | 33 %      | x            | x         | Life Cycle                  | 11 kg CO <sub>2</sub> / 1 ton RCA ; 270 kg CO <sub>2</sub> / 1 ton cement uptake |
| Kikuchi & Kuroda, 2010      | 38 %        | 24 %      | x            | 3 months  | Life Cycle                  | 11 kg CO <sub>2</sub> / 1 ton RCA uptake   |
| Kim & Chae, 2016            | 16,5 %      | 11,2 %    | 40 a         | -         | Life Cycle                  |  |
| Kjellsen & al., 2005        | 57 %        | 30 %      | 70 a         | 30 a      | Life Cycle                  | 75 % carbonation degree  |
| Leemann & Hunkeler, 2016    | 8-21 %      | 8-21 %    | x            | x         | Life Cycle                  | 50 % carbonation degree  |
| Piqueras & Gonzales, 2014   | 35 / 75 %   | 22 / 47 % | x            | x         | Life Cycle                  |  |
| Possan & al., 2016          | 40-90 %     | 40-90 %   | 70 a         | 30 a      | Life Cycle                  |  |
| Xi & al., 2016              | 43 %        | 27 %      | 35-70 a      | x         | Uptake by existing concrete | 75 % carbonation degree  |
| Zhang & Wang, 2014          | 27-50 %     | 17-31 %   | 85-115 a     | -         | Life Cycle                  |  |



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