Supplementary of

Global carbon uptake of cement carbonation accounts 1930-2021

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The detail calculation methods for uptake assessment of concrete, mortar, waste and CKD four types and service, demolishment and second use three life stages are described below.

S1 Concrete uptake assessments

In service stage, after carbonated coefficients in different environment and the correction factors was set (Lagerblad et al., 2005; Pade and Guimaraes, 2007; Zafeiropoulou et al., 2011; Andersson et al., 2013), the carbonation rate of the different strength class materials was set for further use as shown in equation:

$$k_{ci} = Co_{environment} \times \beta_{ad} \times \beta_{CO_2} \times \beta_{CC} \quad (1)$$

Where k_{ci} is the carbonation rate of class i. $Co_{environemnt}$ is the carbonated coefficients under different environments, usually under air or buried environments. β_{ad}, β_{CO_2} and β_{CC} are cement additives, CO_2 concentration, and coating and cover, respectively.

Based on the Fick's second law, then the concrete carbonation depth can be calculated by the following:

$$d_{ci} = k_{ci} \times \sqrt{tl} \quad (2)$$

Where d_{ci} is the depth which depended on carbonation rate and reaction time till the end of service stage. Furthermore, the carbonated amounts over a certain service time can be described as following:

$$Wc_{use_i} = C_{ci} \times \frac{d_{ci}}{Tw} \quad (3)$$

Where Wc_{use_i} is the mass of carbonated cement used in concrete over a certain period of time during the use stage. C_{ci} is the cement content in class i concrete. Tw is the average thickness of concrete structure.

Finally, the concrete uptake in service stage can be calculated through equation 5.

The concrete structures would move to demolishment stage when they were end of service as civil infrastructures. Usually, the end of use structure would be crashed into small size particles (Kikuchi et al., 2011). Thus, in this study, a simplified model of carbonation in demolishment stage is established based on the assumptions that the carbonation starts from the outer surface, moving inwards radially as Fig s1. In this model, the three distinct groups of distributions ($b \le D_{0i}$, $a \le D_{0i} < b$, $a > D_{0i}$) were defined according to the maximum diameter (D_{0i}) of a particle when undergo full carbonation in compressive strength class *i* in the respective range of minimum (a) and maximum diameters (b). Thus, the calculation can be expressed as follow:

$$F_{di} = \begin{cases} 1 - \int_a^b \frac{\pi}{6}\left(D - D_{0i}\right)^3 \Big/ \int_a^b \frac{\pi}{6} D^3 & (a > D_{0i}) \\ 1 - \int_{D_{0i}}^b \frac{\pi}{6}\left(D - D_{0i}\right)^3 \Big/ \int_a^b \frac{\pi}{6} D^3 & (a \le D_{0i} < b) \\ 1 & (b \le D_{0i}) \end{cases} \tag{4}$$

$$D_{0i} = 2d_{di} = 2k_{di}\sqrt{t_d} \tag{5}$$

Where k_{di} is the diffusion coefficient of compressive strength class *i* in demolishment stage under "exposed to air" condition. t_d is the subsequent dealing time after service life. To avoid double counting, the carbonated content in service stage should be excluded. Thus, the cement uptake in this stage can be calculated as:

$$Uc_{d_i} = (Wci - Wc_{use_i}) \times F_{di} \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma \times \frac{M_{CO_2}}{M_{CaO}} \tag{6}$$

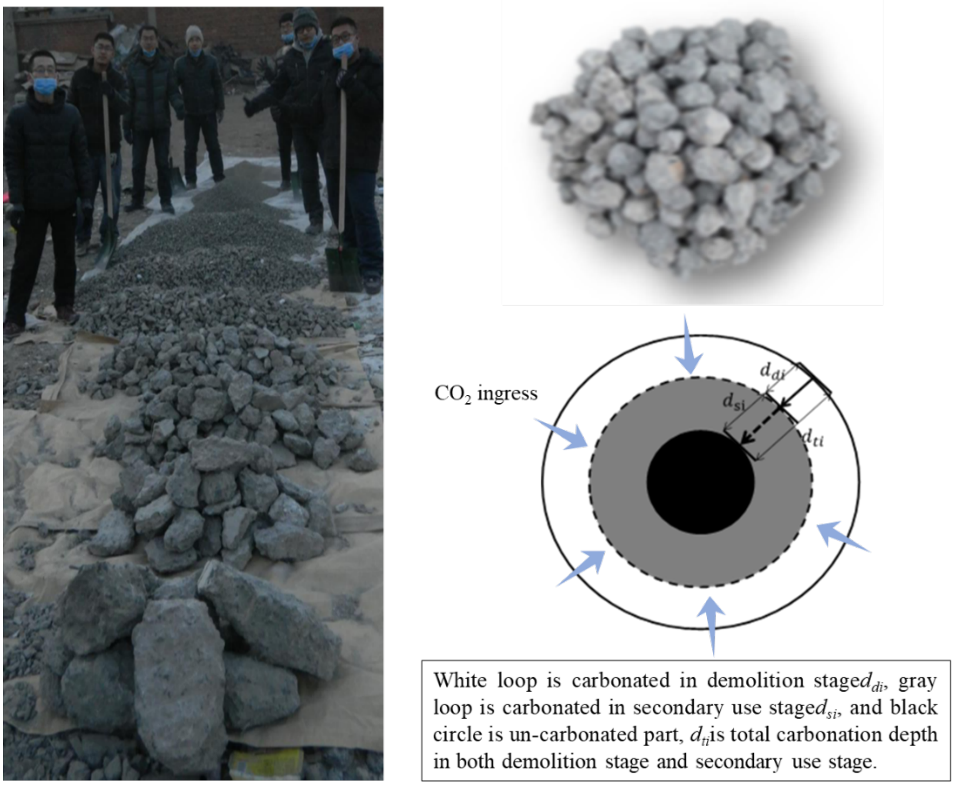

Fig s1. The on-site sampling and the spherical carbonation model of a concrete particle in the demolition stage and second-use stage. The left image is a photograph of on-site sampling; the right image is a schematic representation of the spherical carbonation model of a concrete particle in the demolition stage and second-use stage.

Usually, carbonation in the second-use stage is slower because a carbonated layer has formed out of the particle surface (Yoon et al., 2007; Papadakis et al., 2011). Thus, a time slag has been considered which was used to modify the equation 8. Then the carbonated depth in second-use stage is:

$$d_{s_{ci}} = \sqrt{k_{d_{ci}} \times \sqrt{t_d} + k_{si} \times \sqrt{t_s}} \quad (7)$$

Where $k_{d_{ci}}$ is the carbonation rate of class i concrete during second-use stage. t_d and t_s are total demolishment time and certain time in second-use stage. Then similar to demolishment stage, the particle size would affect the carbonation fraction (F_{si}) and could be calculated as follows:

$$F_{si}=\begin{cases}1-\int_{a}^{b}\frac{\pi}{6}\left(D-D_{ti}\right)^{3}\Big/\int_{a}^{b}\frac{\pi}{6}D^{3}-F_{di} & (a>D_{ti})\\ 1-\int_{D_{ti}}^{b}\frac{\pi}{6}\left(D-D_{ti}\right)^{3}\Big/\int_{a}^{b}\frac{\pi}{6}D^{3}-F_{di} & (a\le D_{ti}<b)\\ 1 & (b\le D_{ti})\end{cases} \tag{8}$$

Then, the total cement uptake amount in this stage can be expressed as follow:

$$Uc_{s_i}=(Wci-Wc_{use_i}-Wc_{d_i})\times F_{si}\times f_{cement}^{clinker}\times f_{clinker}^{CaO}\times\gamma\times\frac{M_{CO_2}}{M_{CaO}} \tag{9}$$

The factors and values mentioned before vary from different regions based on surveys.

2.3.2 Mortar uptake assessments

The mortar utilizations were separated into 3 subcomponents including: (1) rendering and plastering mortar, (2) masonry mortar, (3) maintenance and repairing mortar (Winter and Plank, 2007; Xi et al., 2016; Guo et al., 2021). Thus, the total carbon sequestering of mortar use can be described as below:

$$C_{\mathrm{mor}}=C_{\mathrm{rpt}}+C_{\mathrm{rmt}}+C_{\mathrm{rat}}$$

(16)

Where C_{rpt}, C_{rmt}, and C_{rmat} are the uptake of the corresponding component, respectively. Based on our previous experiment results of carbonation diffusion rates (k_m), in this study, k_m was used to replace k_c to establish a two-dimensional diffusion "slab" model, similar to that of concrete. Also, proportion of CaO conversion was updated to gamma 1(γ_1). In consequence, the carbonation of mortar used for rendering, plastering, and decorating is calculated as follows:

$$d_{rp}=k_m\times\sqrt{t} \tag{10}$$

$$f_{rpt}=\frac{d_{rpt}-d_{rp(t-1)}}{d_{Trp}}\times 100\% \tag{11}$$

$$C_{rpt}=W_m\times r_{rp}\times f_{rpt}\times f_{cement}^{clinker}\times f_{clinker}^{CaO}\times\gamma_1\times\frac{M_{CO_2}}{M_{CaO}} \tag{12}$$

Where d_{rp} is the carbonation depth of rendering mortar. k_m is the carbonation rate coefficient of cement mortar. t is a certain exposure time of rendering mortar after construction. f_{rpt} is the annual carbonation percentage of rendering mortar in year t. $d_{rp,t}$ and $d_{rp,t-1}$ are the carbonation depths of rendering mortar in year t and last year (t − 1), respectively. $d_{T_{rp}}$ is the thickness for rendering mortar utilization. C_{rpt} is the annual carbon uptake of rendering mortar. W_m is the amount of cement use for mortar. r_{rp} is the use ratio of rendering mortar cement in total mortar cement. $\gamma 1$ is the proportion of CaO in mortar cement that fully carbonated to $CaCO_3$.

Calculation for carbon uptake of repairing and maintaining cement mortar is similar to rendering, plastering, and decorating mortar, with differences in the utilization thickness and the percentage of mortar for repairing and maintaining.

Differences were appeared on the calculation of mortar carbon uptake for masonry due to the difference of the partially exposed condition, thicker utilization layers, and their covering by rendering mortar on masonry wall surfaces. Based on surveys, here, the masonry walls were regarded to be three types: walls with both sides rendered (C_{mbt}), walls with one side rendered (C_{mot}), and walls without rendering (C_{mnt}). The main difference is the place of retendering layers on the wall upon the masonry as shown in the transformation previous picture of Fig. s2 (Guo et al., 2021). Thus, the calculation could be as follows.

$$C_{rmat} = C_{mbt} + C_{mot} + C_{mnt} \tag{13}$$

Where C_{mbt}, C_{mot} and C_{mnt} are the uptakes of the above classification, respectively.

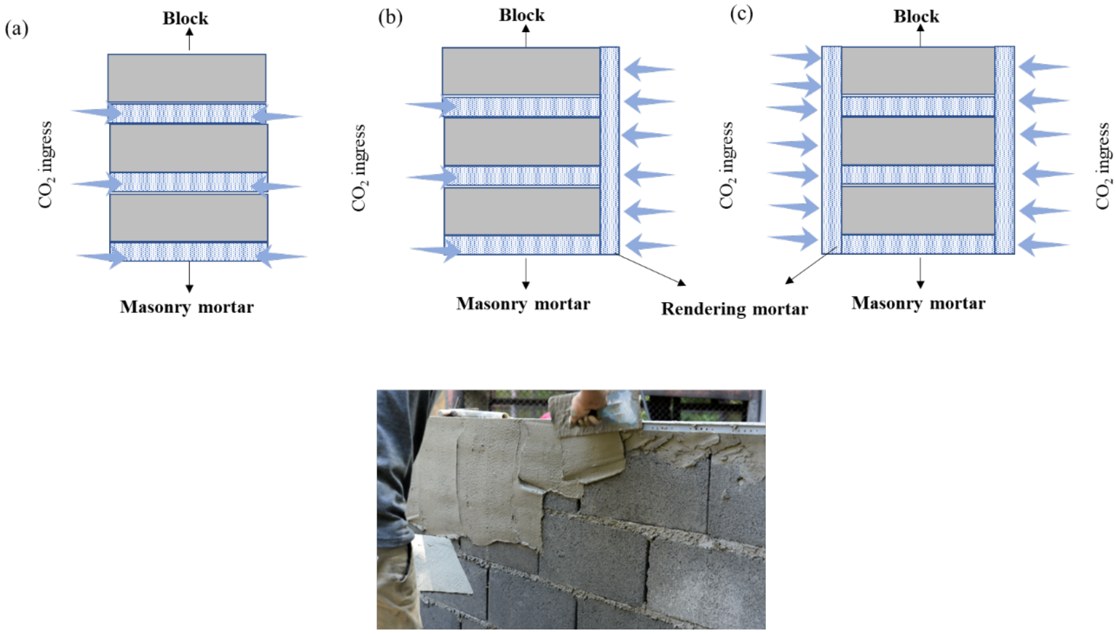

Fig. s2. The carbonation model for masonry mortar and masonry mortar actual use in real life. The top image is a schematic representation of the carbonation model for masonry mortar. (a) masonry mortar without rendering; (b) masonry mortar with one-side rendering; (c) masonry mortar with two-side rendering; the bottom image is a schematic photo for actual use in real life

Here, similar to previous model of carbon uptake in concrete, considering the carbonation of front rendering, the calculation of carbon uptake of mortar for masonry is shown below.

$$d_{mb}=\begin{cases}0 & (t\le t_r)\\ 2\left(K_m\times\sqrt{t}-d_{Trp}\right) & (t>t_r)\end{cases} \tag{14}$$

$$f_{mbt}=\begin{cases}0 & (t\le t_r)\\ \left(d_{mbt}-d_{mb(t-1)}\right)/d_w\times100\% & (t_r<t\le t_{sl})\\ 100\%-d_{mbt_{sl}}/d_w\times100\% & (t=t_{sl}+1)\end{cases} \tag{15}$$

$$C_{mbt}=W_m\times r_{rm}\times r_b\times f_{mbt}\times f_{cement}^{clinker}\times f_{clinker}^{CaO}\times\gamma_1\times\frac{M_{CO_2}}{M_{CaO}} \tag{16}$$

Where d_{mb} is the total carbonation depth of masonry wall with both sides rendered. t is the exposure time of masonry mortar after construction. t_r is the time used when rendering mortar full carbonation. d_{Trp} is the thickness of rendering mortar on masonry wall. f_{mbt} is the annual carbonation percentage of masonry mortar with both sides rendered in year t. d_{mbt} and $d_{mb(t-1)}$ are carbonation depth of masonry mortar with both sides rendered in year t and (t − 1), respectively. d_w is the thickness of masonry wall. t_{sl} is the service life of construction. $d_{mbt_{sl}}$ is the carbonation depth of a masonry mortar with both sides rendered during service life. C_{mbt} is the annual carbon uptake of masonry mortar with both sides rendered in year t. r_{rm} is the ratio of cement use for masonry mortar in total mortar cement. r_b is the ratio of masonry mortar with both sides rendered in total masonry mortar.

2.3.3 Construction wastes uptake assessments

Cement wastes account for 1~3% of total cement consumption based on construction budget standards and survey data (Zhou, 2003; Lu et al., 2011). The main componence is concrete waste (45%) and mortar waste (55%) separately (Bossink et al., 1996; Huang et al., 2013). Thus, in this calculation, they would be considered individually, as shown below.

$$C_{waste} = C_{wastecon} + C_{wastemor} \tag{17}$$

Where $C_{wastecon}$ and $C_{wastemor}$ are the uptakes of concrete waste and mortar waste, respectively. Then, the construction wastes carbonation can be calculated as follow:

$$C_{wastecon} = \left(\sum\nolimits_1^n W_{ci} \times f_{con} \times r_{con}\right) \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma \times \frac{M_{CO_2}}{M_{CaO}} \tag{18}$$

$$C_{wastemor} = \sum\nolimits_1^n W_{mi} \times f_{mor} \times r_{mor} \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma_1 \times \frac{M_{CO_2}}{M_{CaO}} \tag{19}$$

Where W_{ci} is the cement used for concrete in strength class i. f_{con} is the loss rate of concrete cement during construction stage. r_{con} is the annual carbon uptake of waste concrete during construction stage. W_{mi} is the cement used for mortar in strength class i, f_{mor} is the loss rate of mortar cement. r_{mor} is the annual carbon uptake of waste mortar during construction stage.

2.3.4 Cement kiln dust (CKD) uptake assessments

CKD as the main by-product in cement manufacturing industry was mainly treated as landfilled waste (USEPA, 1993; Khanna, 2003). In this work, its carbonation can be calculated as below.

$$C_{CKD} = W_{cem} \times r_{CKD} \times r_{landfill} \times f_{cement}^{clinker} \times f_{CKD}^{CaO} \times \gamma_2 \times \frac{M_{CO_2}}{M_{CaO}} \tag{20}$$

Where W_{cem} is the cement production. r_{CKD} is the CKD generation rate when clinker production. $r_{landfill}$ is the ratio of CKD treated to landfill. f_{CKD}^{CaO} is the proportion of CaO in CKD (Siriwardena et al., 2015). γ_2 is the percentage of CaO in CKD that fully carbonated to $CaCO_3$. Additionally, due to its rapid carbonation, this equation is single year calculation.

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