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Global carbon uptake of cement carbonation accounts 1930-2021

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Abstract:

The main contributor to the GHG footprint of the cement industry is the decomposition of alkaline carbonates during clinker production. However, systematic accounts for the reverse of this process - namely carbonation of calcium oxide and other alkaline oxides/hydroxides within cement materials during cements' life cycle have only recently been undertaken. Here, adopting a comprehensive analytical model, we

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provide the most updated estimates of CO_2 uptake by cement carbonation. The accumulated amount of global CO_2 uptake by cements produced from 1930 to 2021 is estimated to be 22.9 Gt CO_2 (95% Confidence interval, CI: 19.6-26.6 Gt CO_2). This amount includes the CO_2 uptake by concrete, mortar, and construction waste and kiln dust. The cumulative carbon uptake by cement materials from 1930 to 2021 offsets 55.1% of the emissions from cement production (41.6 Gt CO_2, 95% CI: 38.7-47.2 Gt CO_2) over the same period, with the greater part coming from mortar (58.5% of the total uptake). China has the highest cement carbon uptake, with cumulative carbonation of 7.06 Gt CO_2 (95% CI: 5.22-9.44 Gt CO_2) since 1930. As a result of rapidly increased production in recent year, over three-quarters of the cement carbon uptake has occurred since 1990. Additionally, our results show little impact of the COVID-19 pandemic on cement production and use, with carbon uptake reaching about 0.92 Gt CO_2 (95% CI: 0.78-1.10 Gt CO_2) in 2020 and 0.96 Gt CO_2 (95% CI: 0.81-1.15 Gt CO_2) in 2021. Our uniformly formatted and most updated cement uptake inventories provide coherent data support for including cement carbon uptake into future carbon budgets from the local to global scale. The latest version contains the uptake data till 2021, showing the global uptake increasing pattern and offering more usable and relevant data for evaluating cement's carbon uptake capacity. All the data described in this study are accessible at https://doi.org/10.5281/zenodo.7516373 (Bing et al., 2023).

1 Introduction

With continued urbanization in the developing world and infrastructure projects worldwide, cement consumption has increased rapidly (Low, 2005). The cement production process is an energy-intensive and CO_2-emitting process, the total CO_2 emission of which amounts to 5–8 % of global CO_2 emissions (IEA, 2019; Xuan et al., 2019; Friedlingstein et al. 2022). The worldwide average CO_2 emission coefficient of ordinary Portland cement (OPC) is 0.86 kgCO_2/kg (Damtoft et al., 2008), which comprises the release of 0.53 kgCO_2 /kg of clinker owing to the decomposition of

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limestone during calcination. While in use, though, cement materials that are exposed to air naturally undergo carbonation (Pade and Guimaraes, 2007; Renforth et al., 2011; Huntzinger et al., 2009), a physicochemical process where atmospheric CO_2 gradually absorbs into concrete's structure and reacts with alkaline components such as CaO in a moist environment. The main carbonation mechanisms that are responsible for the carbon uptake can be attributed to the oxides, hydroxide and silicate constituents, as described by Reactions (R1) and (R2).

$$\mathrm{Ca(OH)_2} + \mathrm{CO_2} \rightarrow \mathrm{CaCO_3} + \mathrm{H_2O} \tag{R1}$$

$$\mathrm{Ca}_x\mathrm{Si}_y\mathrm{O}_{(x+2y)} + x\mathrm{CO_2} + z\mathrm{H_2O} \rightarrow x\mathrm{CaCO_3} + y\mathrm{SiO_2}\cdot z\mathrm{H_2O} \tag{R2}$$

Unfortunately, from the perspective of offsetting emissions in the production of cement, carbonation is a slow process that occurs over the entire life-cycle of cementitious materials, in contrast to the instantaneous CO_2 emissions during their production (Andersson et al., 2013). It has been shown that up to a quarter of the CO_2 emitted in cement production can be reabsorbed throughout a building's life and recovery phase (Xi et al., 2016). Quite a few procedures for evaluating the CO_2 footprint over cement's lifecycle have been suggested (Damineli et al., 2010; Renforth et al., 2011; Yang et al., 2013; Cao et al., 2020). Most procedures, however, consider only a case limited system boundary and material type such as concrete service stage, recycling phase of concrete after demolition (Andersson et al., 2013; Yang et al., 2014; Xi et al., 2016; Cao et al., 2020; Kaliyavaradhan et al., 2020), and do not take other types and stages of the lifecycle into systematic account. In our previous study (Guo et al., 2021), which incorporated the merits from other work (Andersson et al., 2013; Yang et al., 2014; Xi et al., 2016; Cao et al., 2020; Kaliyavaradhan et al., 2020) and the updated clinker ratio and/or cement production data, we constructed a comprehensive analytical model to estimate the time-series of cement CO_2 uptake inventories and estimated that 21.02 Gt CO_2 had been sequestered in cements produced between 1930 and 2019, which abated 55% of the corresponding process emission over the same

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period.

The cement CO_2 uptake and emission dataset can be accounted annually. In this study, based on the previous data frameworks (Guo et al., 2021), we updated cement production and emission factors, and most up-to-date clinker ratio data of the year of 2020 and 2021. Adopting previous comprehensive analytical model (Guo et al., 2021), we updated the cement CO_2 uptake and emission dataset from 1930 to 2021. The inventories are constructed in a uniform format, which includes cement process-related emissions and cement uptake from four material types with three life stages burned in five countries or regions. The uniformly formatted time-series cement uptake inventories can be utilized widely. Using this consistent framework and models, we provide an updated annual cement carbon uptake to be used in the annual assessments of the global carbon budget (GCB) (Friedlingstein et al., 2022). These timely updated inventories can provide robust data support for further analysis of global or regional emissions reduction policy-making, especially for carbon-intensive industry like cement manufacturing industry. By accelerating carbon capture from existing cement materials and using waste concrete as a carbon storage material, cement could reduce its net carbon emission impact. The primary focus of this research is to update the cement carbon uptake data up to 2021 using a methodology consistent with our previous publication. By doing so, we aim to provide the most current and up-to-date data to accurately portray the impact of cement carbon uptake. The data can be downloaded freely from https://doi.org/10.5281/zenodo.7516373.

2 Data and Methods

The cement CO_2 uptake and process emission in this dataset were estimated in terms of the comprehensive analytical model and based on IPCC administrative territorial-based accounting scope. In addition, we also assessed the uncertainties in cement uptake and process emission estimates using the Monte Carlo method that IPCC recommended. The detail input data are in SI-Table 1 (available online only). Our inventories were constructed in two parts: process-related (cement) CO_2 emissions and

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cement material uptake. Figure 1 presents a diagram of the entire construction of our cement material carbonation uptake and cement emission inventories.

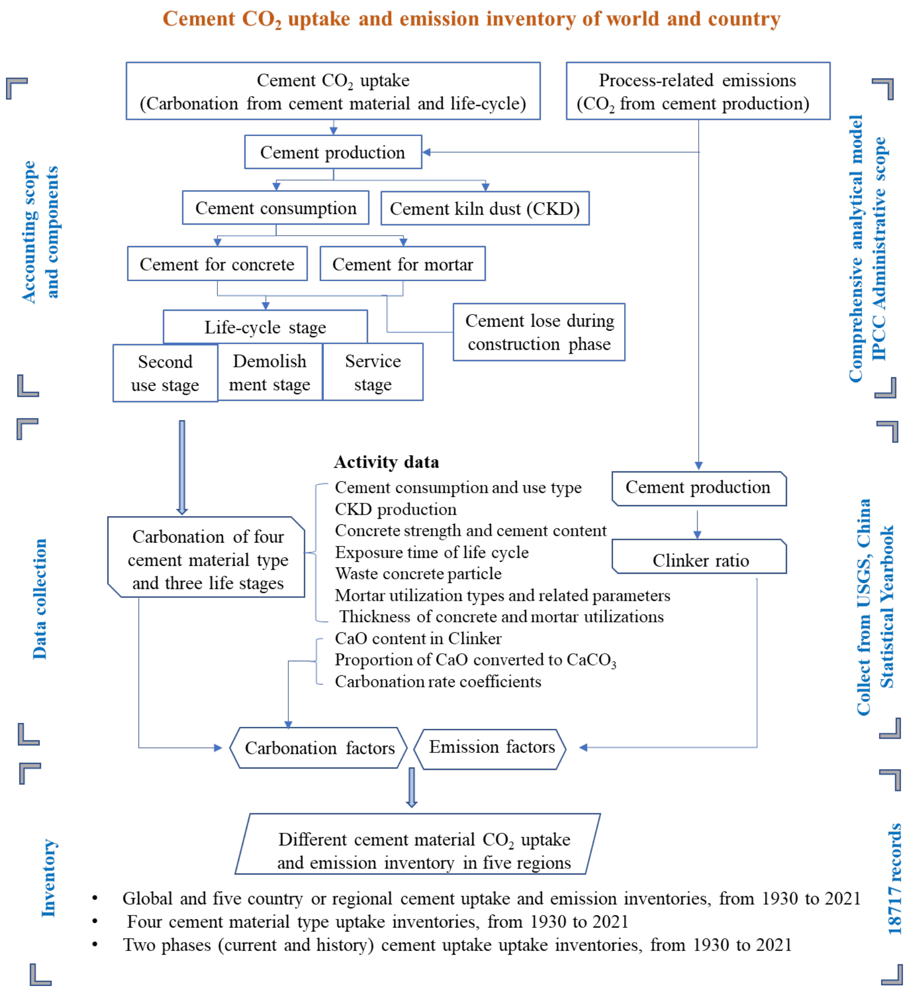

Figure 1. Diagram of cement CO_2 uptake and emission inventory construction.

2.1 Cement production data sources

To keep the consistency with the previous study (Xi et al., 2016; Guo et al., 2021), we still obtained the global cement production data from 1930 to 2021 from the United States Geological Survey (USGS) and geographically divided into five primary countries and aggregated regions, including China, the United States (US), Europe and central Eurasia (including Russia), India and the rest of the world (ROW). In this study,

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we updated cement production for year 2020 and 2021, and the global cement production was collected from USGS cement statistics and information annual report (USGS, 2022), regional cement productions were gained from China Statistical Yearbook (NBS, 2022), USGS cement annual publication (USGS, 2022), Trading Economics (2019) for China, the United States (US), Europe and Central Eurasia (including Russia) and India, respectively. The clinker ratio data was kept the same with the previous data sources (CCA et al., 2001-2005; Xu et al., 2012; Xu et al., 2014; Cao et al., 2017; MIIT, 2019) except the US which was collected from USGS annual cement report (USGS, 2022).

2.2 Cement process emission calculation

In producing cement clinker, the major constituent of cement (OPC), limestone together with other carbonates are decomposed into their corresponding oxides and gaseous CO_2 via calcination, resulting in the process emission of the cement industry. It is a so-called hard-to-abate CO_2 emission source (Antunes et al., 2021) because no clear avenue has yet been found to replace this chemical process. Therefore, the process emission intensity (factor) is related to the composition of the clinker and its content in the cements in question. The IPCC recommended default value of process emission factor is 0.507 kg CO_2 kg^{-1} clinker (EFDB, 2002), without the emissions associated with $MgCO_3$. In our work, the value of clinker ratio for China was taken to be 0.51966 kg CO_2 kg^{-1} clinker for dry with preheater without pre-calciner, dry with preheater and pre-calciner, and dry without preheater (long dry) kilns, and 0.49983 kg CO_2 kg^{-1} clinker for semi-wet or semi-dry and wet or shaft kilns since 2005, as adapted from Shen's study (Shen et al., 2016). For other countries, Andrew's recent work (Andrew, 2019) established a sound foundation for those who are in absence of survey data. Besides, the survey data was obtained from the World Business Council for Sustainable Development (WBCSD) and the Global Cement Directory 2019 (publicly named as the GCD-2019 dataset). Finally, the use of integrated global plant-level capacity and technology information was maintained and continued in this study for higher accuracy

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in contrast to regionally averaged cement emission factors (Guo et al., 2021).

In general, the process emission can be calculated by Equation 1:

$$E_{process,i} = P_{cement,i} \times f_{clinker,i} \times EF_{CO_2,i} \tag{1}$$

Where $E_{process,i}$ is the cement process emission of the different regions. $P_{cement,i}$ is the regional cement production. The $f_{clinker,i}$ and $EF_{CO_2,i}$ are clinker to cement ratios and cement (clinker) carbon emission factors of these five regions respectively.

2.3 Cement life-cycle uptake assessments

The cement utilization was categorized by four types: concrete, mortar, cement kiln dust and cementitious construction wastes, which included three life stages (Xi et al., 2016; Guo et al., 2021) named:(1) service, (2) demolishment, and (3) second use. Thus, the whole carbon uptake process can be designed as

$$C_{uptake} = C_{concrete} + C_{mortar} + C_{wastes} + C_{CKD} \tag{2}$$

$$C_{concrete} = C_{l,tl} + C_{d,td} + C_{s,ts} \tag{3}$$

$$C_{mortar} = C_{l,tl} + C_{d,td} + C_{s,ts} \tag{4}$$

Where C_{uptake}, $C_{concrete}$, C_{mortar}, and C_{waste} are the uptake amounts of every types. $C_{l,\,tl}$, $C_{d,td}$, and $C_{s,ts}$ are the uptake amounts during service, demolition and secondary-use stages, respectively. Following our previous study, 100 years were considered to be the total life-cycle time. During service stage, cement materials are mainly used for civil infrastructures' constructions. Based on Fick's second law, a simplified model was applied in this work which introduced a two-dimensional diffusion "slab" process shown in Fig. 2. Fick's second law determines the relationship of carbonization depths and reaction time(tl) linked by diffusion coefficient (k), which can be described as:

$$d = k\sqrt{tl} \tag{5}$$

Then, based on the reaction of cement carbonation and IPCC's report, the carbonation calculation can be expressed to be

$$C = f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma \times \frac{M_{CO_2}}{M_{CaO}} \tag{6}$$

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Where the $f_{cement}^{clinker}$ is clinker ratio, $f_{clinker}^{CaO}$ is the CaO content in the clinker, and γ is the fraction of CaO that could be converted to $CaCO_3$. M_{CO_2} is molar mass of CO_2. M_{CaO} is molar mass of CaO.

In order to simplify the calculation model, some assumptions were applied in this study. Firstly, the diffusion front was assumed regarded to be the same as the carbonation front with the area behind the front was fully carbonated; and then, in the slab model shown as Fig. 2, the carbonation amounts is determined as a function of exposed surface area, carbonation depth and the cement content of concrete. Due to the influence on the carbonation process of exposure condition and materials properties, in this study, for concrete, a compressive-strength-class breakdown was carried out based on the regional standards. For mortar, the different kinds of utilization – rendering, masonry and maintenance were considered most important. Two main exposure conditions (buried and in open air) were considered, with different carbonation coefficients. Specifically, carbon sequestration of these four types of cementitious materials was calculated separately according to the methods described in the following sections.

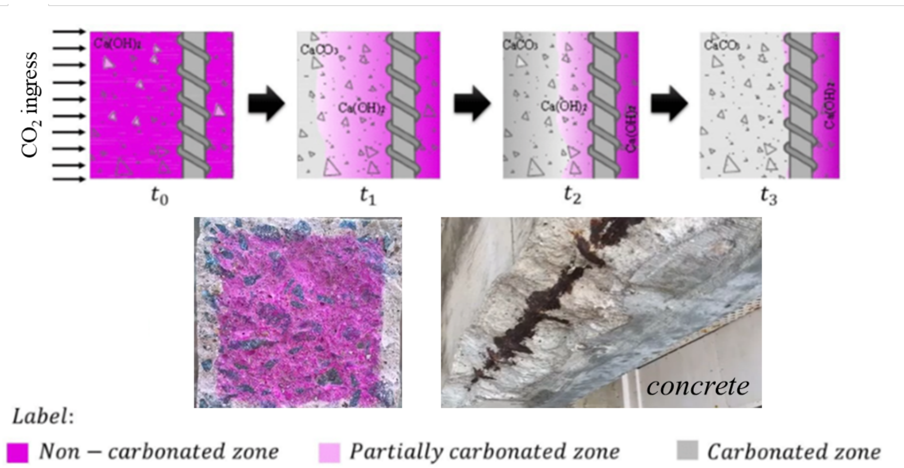

Fig 2. A schematic representation of carbonation model of concretes.

2.3.1 Concrete uptake assessments

In service stage, after carbonated coefficients in different environment and the

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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:

$$k_{ci} = Co_{environment} \times \beta_{ad} \times \beta_{CO_2} \times \beta_{CC} \tag{7}$$

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:

$$d_{ci} = k_{ci} \times \sqrt{tl} \tag{8}$$

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:

$$Wc_{use_i} = C_{ci} \times \frac{d_{ci}}{Tw} \tag{9}$$

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. Since usually the end of use structure would be crashed into small size particles (Engelsen et al., 2005; Kikuchi et al., 2011). 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 3. In this model, the three distinct groups of distributions (b≤D_{0i}, a≤D_{0i} <b, a>D_{0i}) were defined according to the maximum diameter (D_{0i}) of a particle when undergo full

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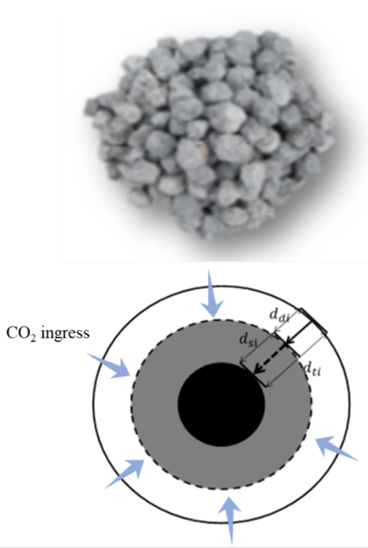
carbonation in compressive strength class *i* in the respective range of minimum (a) and maximum diameters (b). Thus, the calculation can be expressed as follow:

$$F_{di} = \begin{cases} 1-\int_a^b \frac{\pi}{6}\left(D-D_{0i}\right)^3 \Big/ \int_a^b \frac{\pi}{6} D^3 & (a > D_{0i}) \\ 1-\int_{D_{0i}}^b \frac{\pi}{6}\left(D-D_{0i}\right)^3 \Big/ \int_a^b \frac{\pi}{6} D^3 & (a \le D_{0i} < b) \\ 1 & (b \le D_{0i}) \end{cases} \tag{10}$$

$$D_{0i} = 2d_{di} = 2k_{di}\sqrt{t_d} \tag{11}$$

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:

$$Uc_{d_i} = (Wci - Wc_{use_i}) \times F_{di} \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma \times \frac{M_{CO_2}}{M_{CaO}} \tag{12}$$



White loop is carbonated in demolition staged_{di}, gray loop is carbonated in secondary use staged_{si}, and black circle is un-carbonated part, $d_{ti}$is total carbonation depth in both demolition stage and secondary use stage.

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Fig 3. 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:

$$d_{s_{ci}} = \sqrt{k_{d_{ci}} \times \sqrt{t_d} + k_{si} \times \sqrt{t_s}} \tag{13}$$

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:

$$F_{si} = \begin{cases} 1 - \int_a^b \frac{\pi}{6}\left(D - D_{ti}\right)^3 \Big/ \int_a^b \frac{\pi}{6} D^3 - F_{di} & (a > D_{ti}) \\ 1 - \int_{D_{ti}}^b \frac{\pi}{6}\left(D - D_{ti}\right)^3 \Big/ \int_a^b \frac{\pi}{6} D^3 - F_{di} & (a \le D_{ti} < b) \\ 1 & (b \le D_{ti}) \end{cases} \tag{14}$$

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

$$Uc_{s_i} = (Wci - Wc_{use_i} - Wc_{d_i}) \times F_{si} \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma \times \frac{M_{CO_2}}{M_{CaO}} \tag{15}$$

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:

$$C_{mor} = C_{rpt} + C_{rmt} + C_{rat} \tag{16}$$

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

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(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:

$$d_{rp} = k_m \times \sqrt{t} \tag{17}$$

$$f_{rpt} = \frac{d_{rpt} - d_{rp(t-1)}}{d_{Trp}} \times 100\% \tag{18}$$

$$C_{rpt} = W_m \times r_{rp} \times f_{rpt} \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma_1 \times \frac{M_{CO_2}}{M_{CaO}} \tag{19}$$

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 Fig. 4. Thus, the calculation could be as follows.

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$$C_{\mathrm{rmat}} = C_{\mathrm{mbt}} + C_{\mathrm{mot}} + C_{\mathrm{mnt}} \tag{20}$$

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

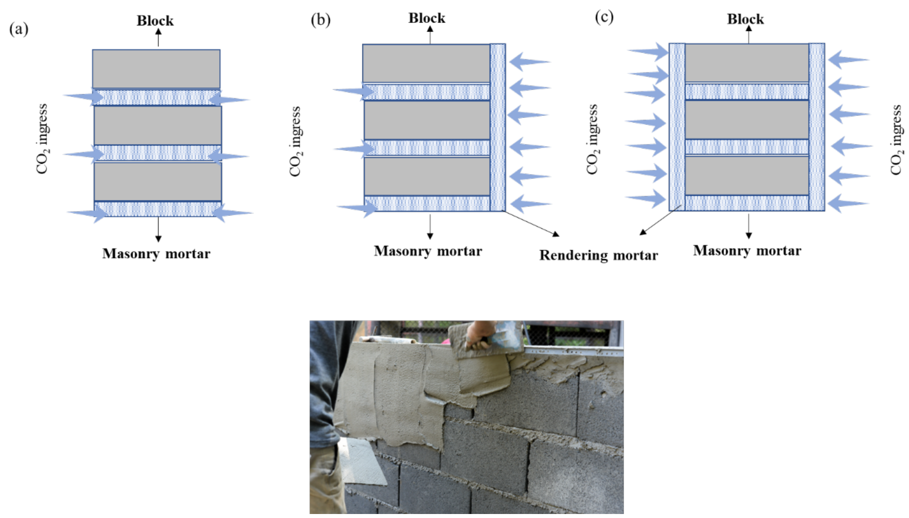

Fig. 4. A schematic representation of the carbonation model for masonry mortar. (a) masonry mortar without rendering; (b) masonry mortar with one-side rendering; (c) masonry mortar with two-side rendering.

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.

$$d_{mb} = \begin{cases} 0 & (t \le t_r) \\ 2\left(K_m \times \sqrt{t} - d_{Trp}\right) & (t > t_r) \end{cases} \tag{21}$$

$$f_{mbt} = \begin{cases} 0 & (t \le t_r) \\ \left(d_{mbt} - d_{mb(t-1)}\right) / d_w \times 100\% & (t_r < t \le t_{sl}) \\ 100\% - d_{mbt_{sl}} / d_w \times 100\% & (t = t_{sl} + 1) \end{cases} \tag{22}$$

$$C_{mbt} = W_m \times r_{rm} \times r_b \times f_{mbt} \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma_1 \times \frac{M_{CO_2}}{M_{CaO}} \tag{23}$$

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

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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.

$$C_{waste} = C_{wastecon} + C_{wastemor} \tag{24}$$

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:

$$C_{wastecon} = (\sum_1^n W_{ci} \times f_{con} \times r_{con}) \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma \times \frac{M_{CO_2}}{M_{CaO}} \tag{25}$$

$$C_{wastemor} = (\sum_1^n W_{mi} \times f_{mor} \times r_{mor}) \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma_1 \times \frac{M_{CO_2}}{M_{CaO}} \tag{26}$$

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

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as landfilled waste (USEPA, 1993; Khanna, 2003). In this work, its carbonation can be calculated as below.

$$C_{CKD} = W_{cem} \times r_{CKD} \times r_{landfill} \times f_{cement}^{clinker} \times f_{CKD}^{CaO} \times \gamma_2 \times \frac{M_{CO_2}}{M_{CaO}} \tag{27}$$

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.

2.3.5 Uncertainty analysis

Based on Monte Carlo simulation, in this study, the uncertainty estimations are applied in cement process carbon emission and cement carbon uptake models separately. The uncertainty ranges of cement process emission and carbon uptake are in SI-Table 4 (Bing et al., 2023). The previous works (Xi et al., 2016) have illustrated the sources of uncertainties. Coherently to previous studies (Xi et al., 2016; Guo et al., 2021), the annual global cement carbon uptake and emission was obtained from regional or material use aggregation, which include 26 variables and factors, shown as SI-Table 2 (Bing et al., 2022). Notably, the annual median at a higher level is not equal to the sum of its sublevel components when evaluate the carbon uptake at each level due to the different statistics based on the Monte Carlo simulation results (see https://doi.org/10.5281/zenodo.7516373; Bing et al., 2023; Guo et al., 2021). In this work for our model used for 2020 and 2021, most of the distributions and their features of variables remain and refer to the previous estimation. But, the clinker to cement ratio of US is updated based on USGS cement annual report of 2021, leading to a change that the random errors are within the range of ±5 % (a uniform distribution).

3 Results and discussions

3.1 Global and reginal CO_2 emissions from cement process

Although, carbon reduction policies have become more stringent and technologies

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more effective since 2019 and accompanied by uncertainties factors that the Covid-19 occurred, global CO_2 emissions from cement processes have been increasing rapidly over the recent past decades due to the continuous growth in the production of cement and related clinker as well, but showing a slightly lower average annual growth rate of 2019 (8.57%) than that of recent past decades (8.68%). According to our calculations and estimates, the global cement process CO_2 emissions have increased from 0.03 Gt yr^{-1} in 1930 to 1.81 Gt yr^{-1} in 2021. Over the period 1930-2021, global cumulative cement process CO_2 emissions amounted to 41.55Gt (95% CI: 38.74-47.19 Gt CO_2), of which ~67% was since 1990, little fewer than that of 2019 (71%). This illustrates that the rapid increase in cement process emissions is mainly driven by industrialization and urbanization accompanied by the development of the global economy. From 1930 to 2021, global cement production increased over 6000%, while the growth rate of CO_2 emissions (5547.31%) was slightly lower than that of cement production, partly due to the relative decreases in average clinker ratios from ∼89 % in 1930 to ∼70 % in 2019. (Wang et al., 2021).

The regional contribution of CO_2 emissions from the cement process has been altered over the period 1930-2021. As shown in Fig. 5, the CO_2 emissions from the cement process in each region show an overall growth trend, while the growth rate varies by country and region. Among all regions, China experienced the most dramatic increasing emission trend with an annual growth rate of 7.7% and reached 0.76Gt CO_2 (95% CI:0.73-0.80Gt CO_2) in 2021. China contributed 33.5% of cumulative process emissions (13.91Gt CO_2, 95% CI:12.44-17.00 Gt CO_2) during the period 1930-2021. Meanwhile, ROW (mainly developing countries/regions), Europe, and the US were responsible for about 35.6% (14.78Gt CO_2, 95% CI:13.17-17.87 Gt CO_2), 23.98% (9.96 Gt CO_2, 95% CI:8.71-12.46 Gt CO_2), and 6.3% (2.62Gt CO_2, 95% CI:2.29-3.27 Gt CO_2) of total cumulative emissions, respectively. India has experienced an incremental growth trend in recent years, totally emitting 2.56 Gt CO_2 (95% CI:2.33-3.02 Gt CO_2), accounting for around 6.2% of process emissions. China and ROW kept their absolute

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leader role in cement CO_2 emissions till 2021, but the share of India has decreased significantly from ~10% to 6.2% in recent 2 years, partly because of shrink of the cement market during Covid pandemic (Schlorke et al., 2020).

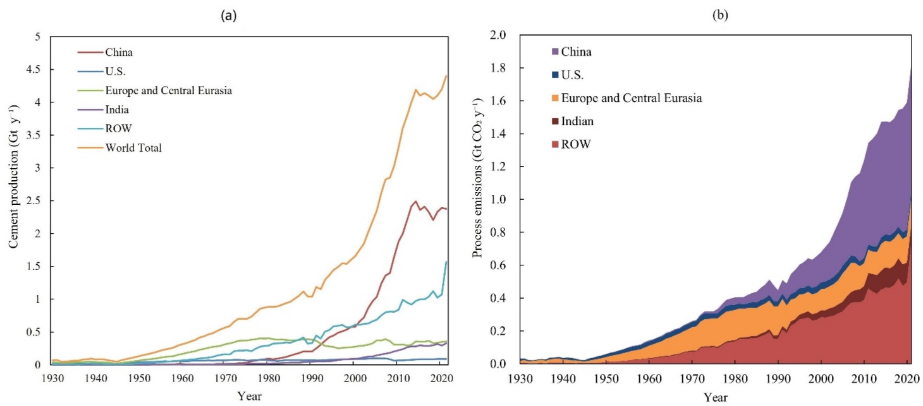

Fig. 5 Regional and global cement production (a) and process emissions (b) from 1930 to 2021

3.2 Cement carbon uptake by region and material type

According to our estimates, the total global CO_2 uptake by cement reached 0.96 Gt CO_2 (95% CI: 0.81-1.15 Gt CO_2) in 2021, with an average annual growth rate of 7.9%. This means that 30.8% of CO_2 emission from the cement process in 2021 was offset by cement carbon uptake in that year. Global cumulative CO_2 uptake by cement was estimated to be 22.90 Gt CO_2 (95% CI: 19.64-26.64 Gt CO_2), equivalent to ~55% of the cumulative emissions over the same period. As we can see in Fig. 6, in China, cement carbon uptake has increased from 0.05 Mt in 1930 to 426.77 Mt in 2021; its cumulative uptake has reached 7.06 Gt CO_2 (95% CI: 5.22-9.44 Gt CO_2), accounting for 30.8% of global cumulative uptake. The cement carbon uptake in China was growing exponentially, while the growth curves in the US and European countries were relatively smooth. This is mainly because the cement demand in China has observed a rapid growth in recent decades, while developed countries have been close to saturation after the 1980s. Moreover, concrete structures in developed countries have a longer service life (estimated 70 years). As for the rest of world, the total carbon uptake by

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cement has also increased significantly (from 0.74 Mt in 1930 to 328.23 Mt in 2021), and the growth trend in these countries was smoother than in China but more dramatic than in the US and Europe.

In addition, the amount of cement carbon uptake varies depending on the type of cement material. Mortar contributes the largest portion of cement carbon uptake although its application scale is much less than concrete (~73% for concrete use and ~24% for mortar use). This is because mortar, as a building decoration material, has the characteristics of small thickness, large exposed surface area, and therefore fast carbonation kinetics. According to Fig.6, in 2021, the carbon uptake by mortar and concrete were 536.85 Mt and 325.95 Mt, accounting for 55.6% and 33.8% of the total cement carbon uptake, respectively. Meanwhile, CKD and loss waste absorbed 62.60 Mt (6.5%) and 34.97 Mt (3.6%) CO_2, respectively.

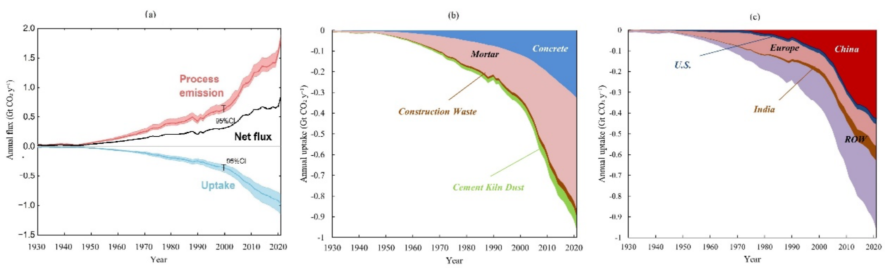

Fig. 6 Annual cement carbon uptake induced net emission (a) and cement CO_2 uptake by different cement materials (b) and by different country or region (c) from 1930 to 2021

3.3 Features of cement carbon uptake

The natural carbonation of cement materials is a slowly dynamic process and thus the carbon uptake by cement has obvious time lag effects. As shown in Fig.7, part of carbon uptake in a given period was contributed by cement materials in previous periods. This is because the cementitious materials carbon uptake is very slow process, leading to a long time to accumulate to manifest and during the demolishment period of cement materials, crushing increases its newly exposed surface area and carbonation

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rate, allowing the carbon uptake capacity of cement materials to persist for a long time. With this feature, the cement carbon uptake capacity can be affected by the service life of cement buildings, and the average lifetime in China (40 years) is less than in the US and Europe (65~75 years). Therefore, countries such as China with a higher speed of cement carbonation cycle can make relatively greater contributions to cement carbon uptake. However, the majority of cement carbon uptake was still attributed to the consumption use stage, providing ~64% share in 2021.

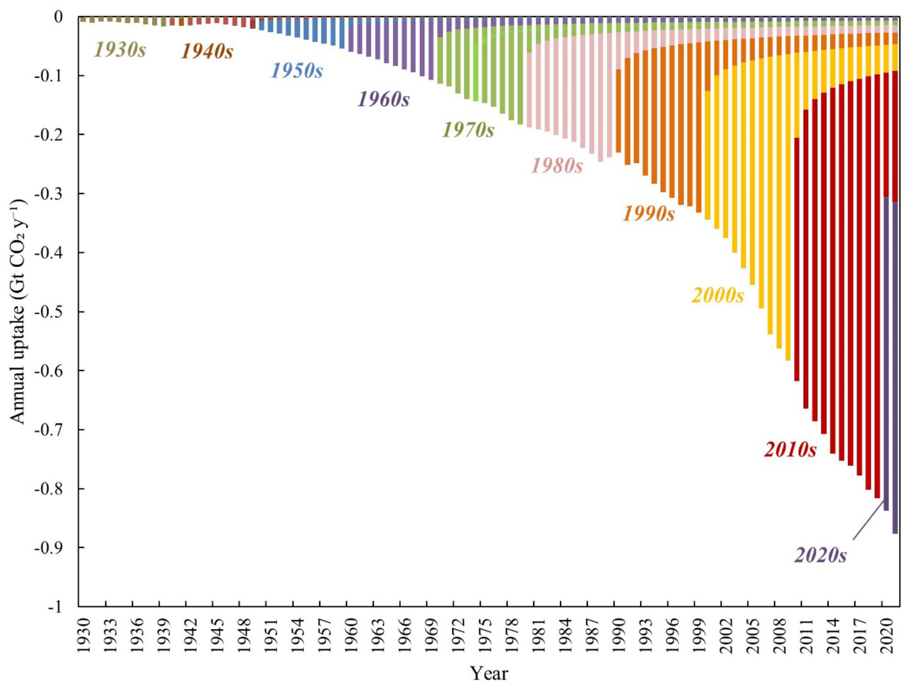

Fig. 7 The cumulative characteristic of cement carbon uptake. The colour-coded bar areas represent the amount of uptake by the cement produced/consumed in each decade from 1930 to 2021. The fractions of uptake that occurred in each decade post-1990 are annotated. The “tails” indicate that cement produced in a certain time will keep absorbing CO_2 beyond its consumption use stage, and the annual uptakes are composed of current and historical contributions.

We can also learn from Fig.8 that the growth rate of historical carbon uptake spiked after the 1990s. It is noteworthy that 75.4% of the cement carbon uptake has occurred

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since the 1990s, larger than that of 2019 (71%). This surge can be explained by the surplus absorption in the demolition phase due to the historically produced cement in European countries during the 1930s and 1940s, on the one hand, and by the considerably increased demand for cement materials in China after the implementation of the reform and opening-up policy, on the other hand.

Besides, the offset level (55.1%) is slightly higher than our previous estimate for 1930-2019 (~52%) (Guo et al., 2021), mainly due to the rapid increase demands from ROW during covid pandemic (Schlorke et al., 2020).

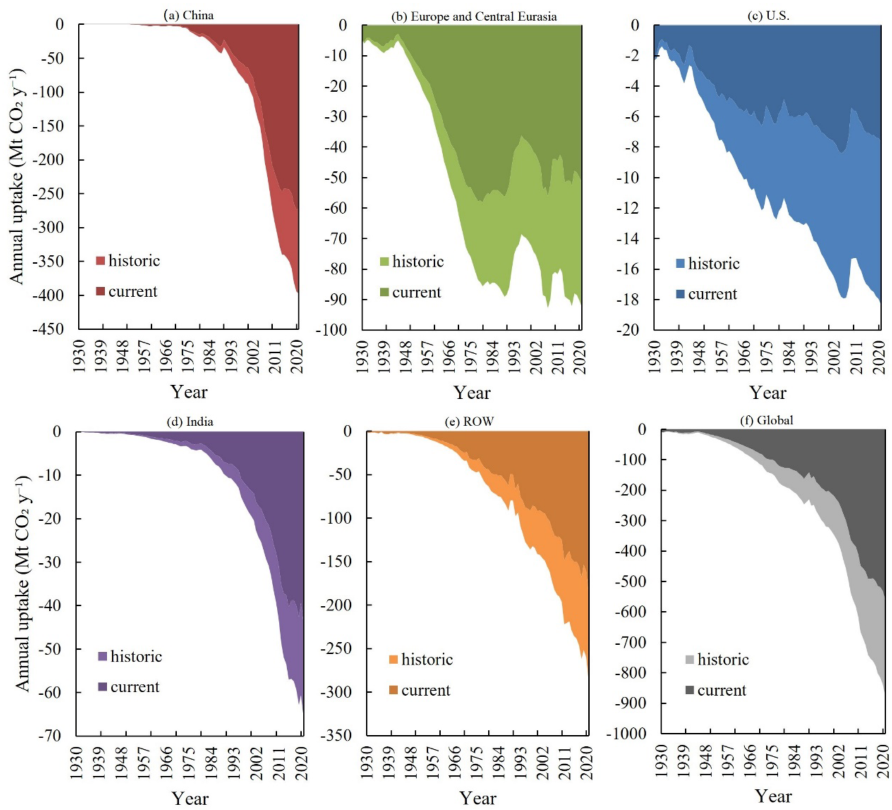

Fig. 8 Annual cement carbon uptake by cement material and region

4. Data availability

All the original datasets used for estimating the emission and uptake in this study and the resulting datasets themselves from the simulation as well as the associated

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uncertainties are made available by Zenodo at https://doi.org/10.5281/zenodo.7516373 (Bing et al., 2023).

5. Conclusions

Due to the unique characteristics of carbon uptake by cement, it is imperative to conduct a scientific and comprehensive estimation of cement carbon uptake. This is crucial for accurately assessing the environmental impact of the cement industry and supporting global carbon neutrality goals. From a kinetic standpoint, cement carbon uptake is a dynamic process that occurs during various stages, including production/consumption, demolition, and reuse. Therefore, it is highly significant to incorporate historical cement legacy sequestration and utilize dynamic clinker ratios to enhance the comprehensiveness and accuracy of estimation. Our objective in this study is to update our data in the temporal dimension, while maintaining consistency with our previous work in terms of methodology. Updating the data within the same framework will enhance the completeness of our database, thereby providing a reliable data foundation for our future forecasting endeavours.

Based on our estimations, the cumulative carbon uptake by cement materials from 1930 to 2021 amounts to 22.90 Gt CO_2 (with a 95% Confidence Interval, CI: 19.64-26.64 Gt CO_2). Mortar contributes approximately 58.5% of the total uptake, effectively offsetting 55.1% of the cumulative process emissions.

This dataset and estimation methodology can be employed as a valuable set of tools for evaluating cement carbon emissions and uptake throughout the dynamic processes encompassing the entire cement life cycle. While per capita cement stocks in Europe and the United States are reaching saturation levels, China has emerged as the dominant region in cement production and consumption following the implementation of China's reform and opening-up policy. Considering that cement demand in China and other developing countries is expected to continue increasing, it becomes evident that this trend will impact the assessment of global carbon neutrality. Therefore, it is crucial to make further efforts to improve the accuracy of cement carbon uptake estimation by incorporating direct clinker production data and experimentally derived spatially resolved conversion factors.

Author contributions

Zi Huang prepared, reviewed, and edited the manuscript with assistance from Jiaoyue Wang, Yijiao Qiu, Longfei Bing, Ying Yu, Rui Guo, Mingjing Ma, Le Niu, Zhu Liu and Fengming Xi. Zi Huang performed the analyses with support from Jiaoyue Wang, Mingjing Ma, Le Niu and Ying Yu on analytical approaches and figure making. Zi Huang, Jiaoyue Wang, and Longfei Bing, Yijiao Qiu curated the datasets. Longfei Bing, Fengming Xi and Zi Huang developed the code and performed the simulations with support from Yijiao Qiu. Dan Tong, Robbie M. Andrew, Pierre Friedlingstein and Josep G. Canadell reviewed, and edited the manuscript. Zhu Liu and Fengming Xi conceptualised and supervised the study.

Competing interests

The authors declare that they have no conflict of interest.

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