RC 1

Comment: Huang et al. present updated data on global carbon uptake of cement carbonation for 2020 and 2021 based on their previous study for 1930-2019 (Guo et al., 2021). Although cement carbon emissions and uptake only account for a small fraction of the global carbon budget, it is not well studied and helpful to accurately understand the global carbon cycle. The manuscript is well written, I recommend publication after some revisions.

Response: Thank you for your precious comments and suggestions. Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. The responds to the reviewer's comments are as following:

1. **General comments**: Methods: Should be more specific and detailed on the different settings between this study and the previous one (Guo et al., 2021). Also, more details on the uncertainty analysis are needed.

   Response: Thanks for your suggestion. In fact, this study is consistent with previous one (Guo et al., 2021), using the same comprehensive analytical model and the same parameters for a common time period. The only difference is that the cement carbon uptake and emissions in this study is updated up to 1930-2021 from original study of 1930-2019 (Guo et al., 2021). Correspondingly, the parameters of cement production, clinker ratio and emission factors of the year of 2020 and 2021 is updated. In the Introduction part of line 82-101 (original line 80-99) and Data and Methods part of line 119-128 (original line 116-125), we have actually expressed the different settings between this study and the previous one (Guo et al., 2021). In addition, based on expert opinion, more details on the uncertainty analysis are added.

   Changes: In the revised version of line 188-195 and line 208-216, more details on the uncertainty analysis "Based on the kinetic models described in previous sections, in this study, the uncertainty estimations through Monte Carlo simulation are applied in cement process emission and cement carbon uptake separately. The term "uncertainty" in this study refers to the lower and upper bounds of a 95 % confidence interval (CI) around our central estimate, i.e. median. All of the input parameters of activity levels and emission and uptake factors, with corresponding statistical distributions, were fed into a Monte Carlo framework, and 10 000 simulations were performed to analyse the uncertainties in estimated carbon emissions and uptake." in line 188-195 and "Specially, the clinker ratio was set to range from 75 % to 97 % in a Weibull distribution with shape and scale parameters of 91.0 % and 25 for regional aggregation of the years of 1930–2021. For China and India, the clinker ratio distribution was unchanged for 1930–1989. For China, the range of coefficient values of the clinker ratio was set to 10%–20% for 1990–2004 with a Normal distribution; for 2004–2021, the random errors were calculated within the range of ±5% of the mean values with a uniform distribution. For India, the random errors were calculated within the range of ±10% for 1990–2001 and ±5% for 2002–2021 of the mean values with a uniform distribution." in line 208-216 are added.

2. **Specific comment**: Line 30: please specify the amount of carbon uptake for each type of cement use.

   Response: We are very grateful for your comment. Due to this is abstract section, we didn't intend to put too specific details to avoid main results being ignored. Thus, we would like to put the present of carbon uptake amounts of every category here.

   Changes: Line 29-31, change 'This amount includes the CO_2 uptake by concrete, mortar, and construction waste and kiln dust.' to 'This amount includes the CO_2 uptake by concrete, mortar, and construction waste and kiln dust, accounting for 30.1%, 58.5%, 4.0% and 7.1% respectively.'

3. **Specific comment:** Line 34: Add values for other regions.

   Response: Thanks for your comment. For here, we want to highlight the contribution of China due to its significant role. Similarly, we would like to use percentage here for amounts of other areas.

   Changes: line 36-37, add 'In addition, the carbon uptake amounts of USA, EU, India and rest of the world took 5.0%, 23.2%, 5.6% and 34.8% separately.'

4. **Specific comment**: Line 105: Should specify where online for SI-Table 1, otherwise the

readers will look for it on the ESSD webpage.

Response: Thanks for your reminder. It's actually from the Data storage and sharing platform of **Zenodo** which you can get from the link: https://doi.org/10.5281/zenodo.7516373. And the SI-Table 1 in the webpage that can be downloaded is our input data set.

Changes: In revised version of line 107-108, replace" (available online only)" to "(available from: https://doi.org/10.5281/zenodo.7516373)".

5. **Specific comment:** Line 116: How was India separated from ROW?

   Response: Thanks for your questions. The data of India was directly collected from United States Geological Survey (USGS). In our previous study (Xi et al., 2016), the world cement production was geographically divided into four primary countries and aggregated regions, including China, the United States (US), Europe and central Eurasia (including Russia), and the rest of the world (ROW). The cement production in ROW is obtained by subtracting China, the United States, and Europe and central Eurasia from global cement data. In our subsequent study (Guo et al., 2021), we noticed that India has now become the second-largest cement producer after China, with approximately 8 % of the world total in 2014 (IEA and WBCSD, 2018), then it divided geography 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) (Guo et al., 2021). The data of India was directly collected from United States Geological Survey (USGS). The cement production in ROW is obtained by subtracting China, the United States, Europe and central Eurasia, and India from global cement data. To keep the consistency with the prior geographical division (Guo et al., 2021), thus, we also use this division for our study.

6. **Specific comment**: Line 139: "For other countries"- specify the values used for other countries.

   Response: Thanks for your suggestion. This can be found from SI-Table 1 – SI date 3 (https://doi.org/10.5281/zenodo.7516373)). Generally, 1930-1950, cement production process CO_2 emission factors for all other counties are 0.5. After 1950, there was an increase in the factor, but the data variations across different regions remained consistent in every year.

Changes: In the revised line of 143-145, we add' (data can be accessed from SI-Table 1 – SI data 3 from https://doi.org/10.5281/zenodo.7516373))'.

7. **Specific comment**: Line 204-205: I'm not sure what it means here, try to clarify it in another way.

   Response: Sorry for making confusing. Here, we introduced the way that end of use cement could be usually treated. Most of them will be crushed into small particles for further use such burying.

   Changes: In the revised version of line 28-31 (in the Supplement document), we replace it with 'Usually, the end of use structure would be crashed into small size particles (Engelsen et al., 2005; 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.'

8. **Specific comment:** Figure 3: Need to explain in the figure legend what the left figure shows. Same for the bottom figure of Figure 4.

   Response: Thanks for your comment. For figure 3, the left image is a photograph taken on-site; the right image is the spherical carbonation model schematic diagram of a concrete particle in the demolition stage and second-use stage. For figure 4, the top image is the carbonation model schematic diagram for masonry mortar in different usages; the bottom image is for schematic photo for actual use in real life. In addition, original figure 3 and figure 4 have moved to Supplement document, accordingly, the original figure 3 and figure 4 is changed to figure s1 and figure s2.

   Changes: In the revised version of line 44-47 (in the Supplement document), change it with '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.' Line 95-99, correct it to ‘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’.

9. **Specific comment:** Figure 4: (a)(b)(c) look pretty like Figure 3 of Guo et al., 2021, consider removing them if they are not very important.

   Response: Yes, the image of (a)(b)(c) in original Figure 4 look pretty like Figure 3 in the study of Guo et al., 2021. In fact, the carbonization forms of masonry mortar in these two studies are the same, namely masonry mortar without rendering, masonry mortar with one-side rendering and masonry mortar with two-side rendering. To ensure the readability and completeness of the article, we decide to keep them and indicate that this figure is a transformation of previous figures (Guo et al., 2021). In addition, the detail method section has been moved to the supplement document. Accordingly, the original figure 3 changed to figure s2

   Changes: In the revised version of line 89-91 in the Supplement document, change it with “The main difference is the place of retendering layers on the wall upon the masonry as shown in the transformation previous picture of Fig. 4 (Guo et al., 2021).”

10. **Specific comment:** Line 316: 2023 instead of 2022.

    Response: Thanks for your reminding.

    Change: In the revised version of line 196, replace’ (Bing et al., 2022)’ to ‘(Bing et al., 2023)’.

11. **Specific comment:** Line 335: I’m confused with 67% and 71%, what are they referring to?

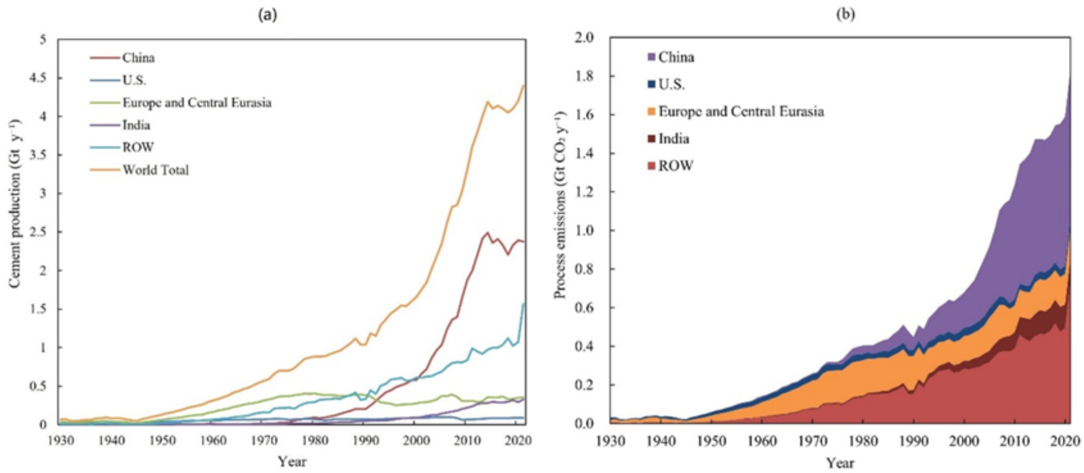
    Response: Sorry for making the confusing. 67% refers to in the 41.55Gt CO_2, 67% of it was emitted from 1930 to 1990. And, 71% of it was emitted from 1930 to 2019, which means the amount from 1990 to 2019 accounts for 4% of the totally global cumulative cement process CO_2 emissions till 2021.

Change: In the revised version of line 226 to 229, correct '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%).' to '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). Specifically, around 67% was accumulated from 1930 to 1990, little fewer than that from 1930 to 2019 (71%).'

12. Specific comment: Figure 5(b): "Indian" should be "India"

Response: Thanks for your reminding. In the revised version, we have changed "Indian" to "India".

Changes: In the revised version of line 252, the modified figure is as follows. Meanwhile, the original Figure 5 changed to Figure 3 due to other part changes.



13. Specific comment: Line 413: Should explain in the main text what 'current and historical contributions are referring to, this is also helpful for understanding Figure 8.

Response: Thanks for your comment. The cement uptake in certain year actually consists of two parts, namely the current uptake and historical uptake. The 'current' refers to the cement carbon uptake in certain year due to the use of newly produced cement. The 'historical' refers to the cement carbon uptake in certain year due to the cement use in previous years. For example, the cement carbon uptake in 2021 consists of two parts. Current uptake comes from the use of cement that produced in 2021; historical uptake comes from the accumulated carbon uptake in 2021 due to the application of cement in the historical period from 1930 to 2020. Thus, for here, we want to state that the cement carbon uptake created from a certain year's cement production will have long-term effects, not only influencing the current year but also offer accumulating impacts to the future.

Changes: In the revised version of line 290-293, we have added the expression of "The cement uptake in certain year actually consists of two parts, namely the current uptake and historical uptake. The current uptake refers to the uptake from the year cement is produced, and the historical uptake refers to the uptake accumulated from year before."

14. Specific comment: In Guo et al., 2021, 2018 and 2019 cement production for Europe and Central Eurasia were projected. How are they being treated in this study? Are there any other values projected?

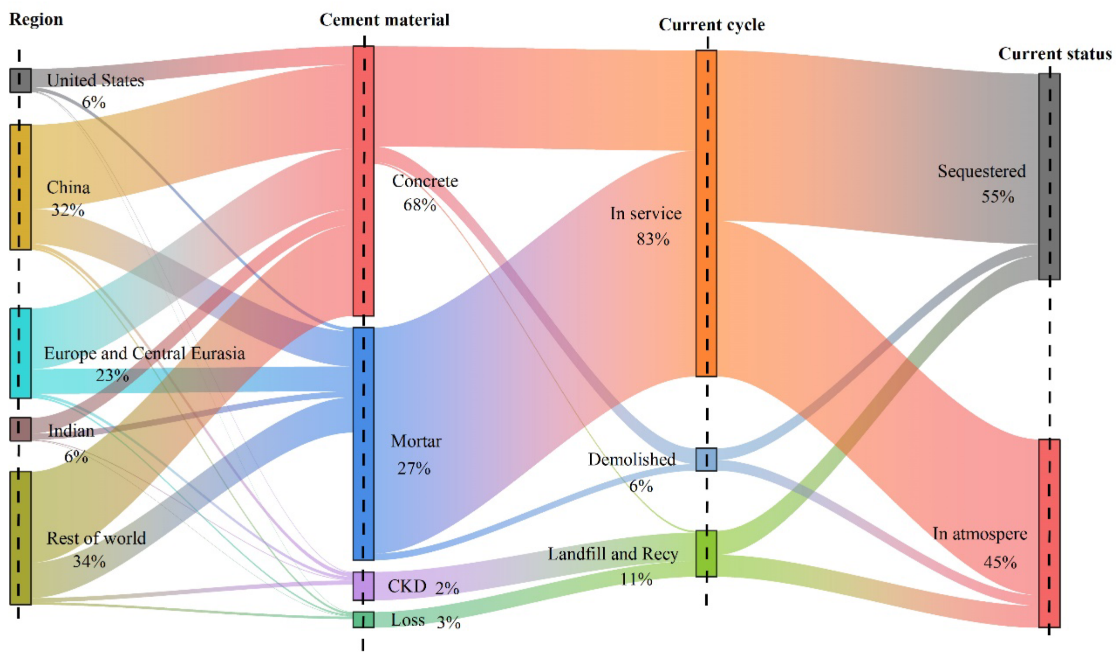
Response: In this work, to keep the consistency with the prior geographical division and data source, we continue to use the projected 2018 and 2019 cement production for Europe and Central Eurasia in the study of Guo et al., 2021. The 2020 and 2021 cement production for Europe and Central Eurasia in this study are also the projected values that use the same projected method, which has expressed in the SI-Table 1 (data can be accessed from SI-Table 1 from https://doi.org/10.5281/zenodo.7516373).

15. Specific comment: It is recommended to include a figure in the main text or supplementary similar to Figure 3 of Xi et al., Nature Geoscience, 2016, which provides a good general overview of the flow of global cement emissions.

Response: Thanks for your comment. In the main text, we have added one figure to express the flow of global cement process emissions 1930–2021.

Changes: In the revised version of line 326 to 335, the added some expression "Figure 7 traces the cumulative cement process CO2 emissions between 1930 and 2021 according to regional production and use of cement in different materials, and to the life cycle of each type of materials. From regional perspective, between 1930 and 2021, 6%, 32%, 23%, 6% and 34% CO2 emissions from cement production are from United States, China, Europe, India and rest of world, respectively. For cement material, the CO2 emissions are 68% from concrete, 27%

from mortar, 2% from loss cement in construction stage and 3% from CKD generation. The CO2 emissions are 83% in service life cement, 6% attributed to demolished cement, and 11% attributed to demolition cement landfill and recycling. Overall, the emissions during 1930 - 2021, are sequestered by cement materials and 43% are remaining in atmosphere." and one figure as following:



16. **Specific comment:** Any change in cement process and major constituents in recent years, considering the industry is moving towards more sustainable?

Response: According to the IEA, the main levers for cement producers are the increase in energy efficiency and the use of alternative materials, be it as fuel or raw materials. Accordingly, cement manufacturing technology has been upgraded rapidly and the use of alternative fuels has already increased significantly in recent years (Xu et al., 2022). In cement, the reduction of the clinker factor remains a key priority, and tremendous progress has already been made. The substitution of clinker in cement is the most effective way to reduce the carbon emissions. Now, cements with several main constituents were produced by replacing parts of the clinker content by supplementary cementitious materials. As such, fly ash, blast furnace slag as well as natural pozzolans were used in increasing amounts. (Schneider et al., 2015; Xu et al., 2022). Nevertheless, appropriate materials are limited in their regional availability. It remains to be seen to what extent they could substitute Portland cement clinker to a significant degree, and Portland cement is still the major cement today (Schneider et al., 2015). This also means the cement constituents has a significant impact on the cement process emissions, using clinker production is more accurate than using cement production when calculation cement process emissions (Andrew, 2019). Like other study (Andrew, 2019), we try to use cement production and variant clinker ratio that transform from clinker production to accurately calculate cement process emission in this study; while there is no cement clinker statistics, we use the cement clinker ratio parameter recommended by IPCC to calculate the cement process emissions (Andrew, 2019). For cement carbonation uptake, certainly, the cement additives will also affect the carbonation of cement due to the alkaline minerals such as CaO in the cement additives. In this study, we have considered the effect of additives on cement carbonization through the correction coefficient of additives, which has expressed in the SI-Table 1 (data can be accessed from SI-Table 1 of sheet 10 of from https://doi.org/10.5281/zenodo.7516373).

17. **Specific comment:** 100 years life-cycle time is assumed in the analysis. However, this can be very different on the regional scale, how the uncertainty from this is addressed?

Response: Like other studies (Pommer et al., 2006; Kapur et al., 2008; Mequignon et al., 2013; Yang et al., 2014), we use 100 years life-cycle time to study carbon uptake in cement. Certainly, the service life in different countries and world regions are different. Through data collection and analysis, the average service life in USA, China, Europe, India, and rest of world is found to be 65, 35, 70, 40 and 40; the average demolition stage is around 0.4; and the corresponding average secondary use stage is 43.6, 64.6, 29.6 and 59.6 for USA, China, Europe, India, and rest of world. In the uncertainty analysis, exposure times of cement materials in life cycle by region is an influencing factor, with Weibull distribution, which has expressed in the SI-Table 1 and SI-Table 2 (data can be accessed from SI-Table 1 of SI data 11 sheet and SI-Table 2 from https://doi.org/10.5281/zenodo.7516373).

18. **Specific comment:** I recommend adding a Results and Discussion subsection for uncertainty results and showing some comparison for the uncertainty contribution caused by different variables used in the analysis.

Response: Thanks for your constructive suggestion. In the main text, we have added a new subsection "3.4 Uncertainty analysis ", focusing on the results of the uncertainty analysis and the contribution of different variables to the overall uncertainty. Specifically, we have 1) presented the uncertainty ranges for the estimations of carbon uptake; 2) compare with other studies; 3) discussed the different contributions of key variables like clinker to cement ratio, correction factors related to cement additives, and CaO content in clinker et. al. to the overall uncertainty; 4) emphasized the significance of the uncertainty analysis and avenues to reduce uncertainty in future. We believe this new results and discussion section has well presented and discussed the key results of the uncertainty analysis. This not only makes the paper more complete but also allows readers to better understand the influence of different variables on the estimation results. We sincerely appreciate your valuable comments again.

Changes: In the revised version of line 347-382, the subsection of uncertainty analysis is as follows:

"**3.4 Uncertainty analysis**

The estimates of cement carbon uptake and emissions underwent through uncertainty analysis utilizing Monte Carlo simulation. The findings reveal that the 95% confidence interval for cumulative carbon uptake spanning from 1930 to 2021 ranges from 19.6 to 26.6 Gt CO_2, while the cumulative emissions exhibit a range of 38.7 to 47.2 Gt CO_2, as presented in SI-Table 4.

Through executing an OAT sensitivity analysis that use China's carbon uptake simulation as an illustrative case (Fig. 8), Overall, the main influential parameters can be categorized as cement material properties, carbonation efficiency parameters, and environmental factors three parts. Notably, cement material properties encompassing factors such as clinker to cement ratio (100%), correction factors related to cement additives (96.1%), and CaO content in clinker (90.9%) exerted the most substantial impact, given their direct influence on the scale of carbon uptake. Carbonation efficiency parameters encompassing the proportions of CaO converted to $CaCO_3$ for concrete and mortar, introduced significant uncertainty at levels of 57.2% and

38.9%, respectively. This underscores the pivotal role that carbonation efficiency uncertainty plays in determining outcomes. Environmental factors primarily encapsulated by the CO_2 concentration correction factor, took responsible for 88.2% of the uncertainty in predictions. Consequently, ambient CO_2 levels exercise a notable sway over the degree of result uncertainty. The uncertainty analysis provides a quantitative basis for assessing the influence of different factors on carbon uptake. Further collecting measured data and improving certainty of key parameters in the future will help reduce result uncertainty and improve estimation accuracy.

Furthermore, in order to establish the validity of this study, we attempted cross-validation. Generally, the coverage of the global cement carbonation uptake within the existing research is limited, with only a handful of studies (Xi et al., 2016; Guo et al., 2021; Cao et al., 2020) delving into this area. The majority of research focuses solely on specific regions, like Spain (Sanjuán, et al., 2020), Nordic countries (Pade and Guimaraes, 2007) or particular structures, such as The Itaipu Dam (Possan et al., 2017). Moreover, there is a notable discrepancy in the methodologies employed among studies that share similar scopes. Notably, the iterative updating approach is utilized in various studies but with distinct variations. For instance, Guo's research method builds upon the foundation established by Xi's work, a progression that Guo elaborates on in their paper (Guo et al., 2021).

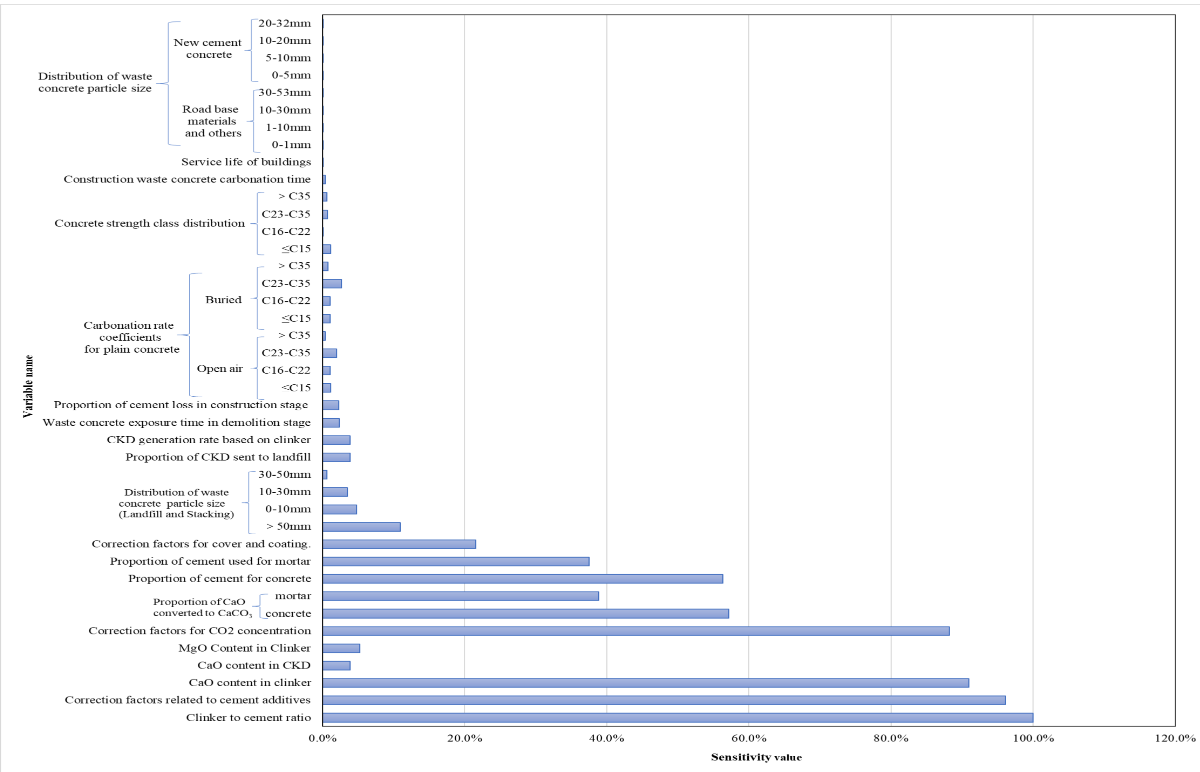
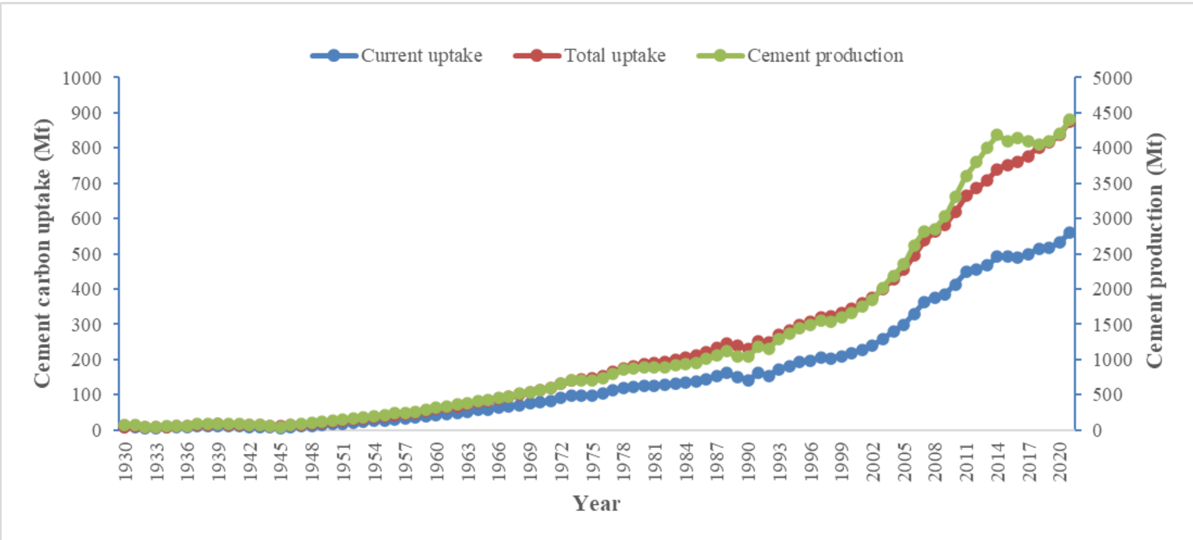


Fig.8 Sensitivity analysis of cement carbon uptake taking China's carbon uptake simulation as an illustrative case"

19. Specific comment: Figure 6(a): it shows the uptake was increasing fast during ~2000-2013, then the increase rate slowed down. Any explanation for this? I understand it is not ESSD guidelines to include data interpretation, but it is good to discuss what the reasons are, and maybe it is caused by some errors in the model.

Response: The annual cement uptake consists of two parts, namely current uptake and historical uptake. Overall, the current uptake plays a leading role, taking around 69% of total cement uptake. Meanwhile, the current uptake depends on the cement production of that year. So the cement uptake with fast increase rate during ~2000-2013 then with slowed down increase rate is due to the changes in cement production (See the following figure).



Changes: In the revised version, we add expression of "It shows that the cement uptake increasing fast during around 2000-2013, then the increase rate slowed down due to the changes in cement production." in line 258-260, and "The current uptake refers to the uptake from the year cement is produced, and have close relationship with the current cement production." in line 291-292.

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Pommer, K., Pade, C., Institut, D. T. & Centre, N. I. Guidelines: Uptake of Carbon Dioxide in the Life Cycle Inventory of Concrete. (Nordic Innovation Centre, 2006)

Kapur, A., Keoleian, G., Kendall, A. & Kesler, S. E. Dynamic Modeling of In-Use Cement Stocks in the United States. Journal of Industrial Ecology 12, 539-556 (2008).

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Xue X.Z., Huang B.J., Liu L.T., et al. Modernizing cement manufacturing in China leads to substantial environmental gains. Communications Earth and Environment, 2022, 3, 276.

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RC2

Comment: This study presents a dataset of global carbon uptake through cement carbonation, which holds significant importance in achieving the goal of Net-Zero emissions. This paper is written well. However, there are some comments that need to be addressed.

Response: Thank you for your valuable comments and suggestions. Those comments are very helpful for revising and improving our paper, as well as the important guiding significance to our researches. The responds to the reviewer’s comments are as following:

1. **Specific Comment:** First, the method section contains excessive details. It is suggested to move some of the text and figures to the Supporting Information, retaining only the essential parts for your calculations.

   Response: Thank you for your suggestion. The ESSD journal is a journal that focuses on original data or data collection. A detailed method description is important for data collection, so in the original article, we put all the details in the main text. In the revised version, in order to improve the readability of the article, we summarized the section 2 Data and Methods, and moved the detailed methods with revision (red part) according to suggestions from another expert into the supplement document.

   Changes: Generally, we move section 2.3.1 to 2.3.4 (original line 188 to 314) to Supplement document. Meanwhile, in the revised line 183-184, we changed it with "Specifically, carbon sequestration of these four types of cementitious materials was in the Supplement document" Accordingly, change original line 315 "2.3.5 Uncertainty analysis" to "2.4 Uncertainty assessment".

In supplement section, it will be display as below:

Supplementary of

Global carbon uptake of cement carbonation accounts 1930-2021

Zi Huang, Jiaoyue Wang and et al.

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:

$$d_{ci} = k_{ci} \times \sqrt{tl} \tag{2}$$

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:

$$Wc_{use_i} = C_{ci} \times \frac{d_{ci}}{Tw} \tag{3}$$

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 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 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:

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

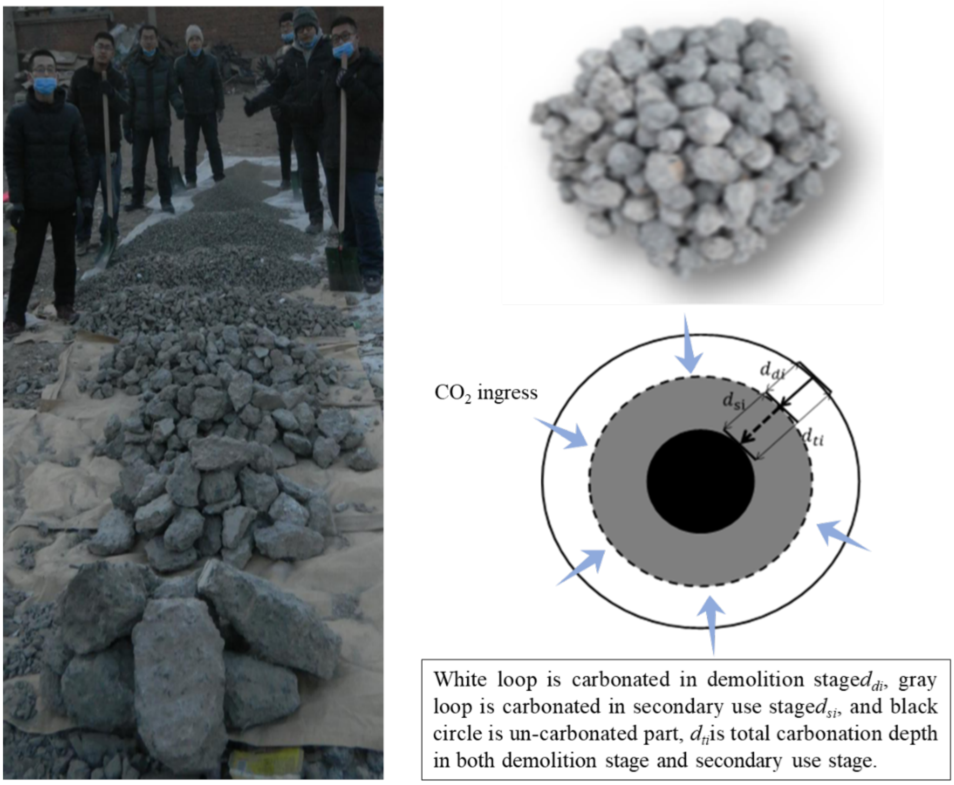

Fig 3. The on-site sampling and the spherical carbonation model of a concrete particle in the demolition stage and second-use stage. The left image is a photograph of on-site sampling; the right image is a schematic representation of the spherical carbonation model of a concrete particle in the demolition 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:

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

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

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

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:

$$d_{rp}=k_m\times\sqrt{t} \quad (10)$$

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

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

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. γ1 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. 4 (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.

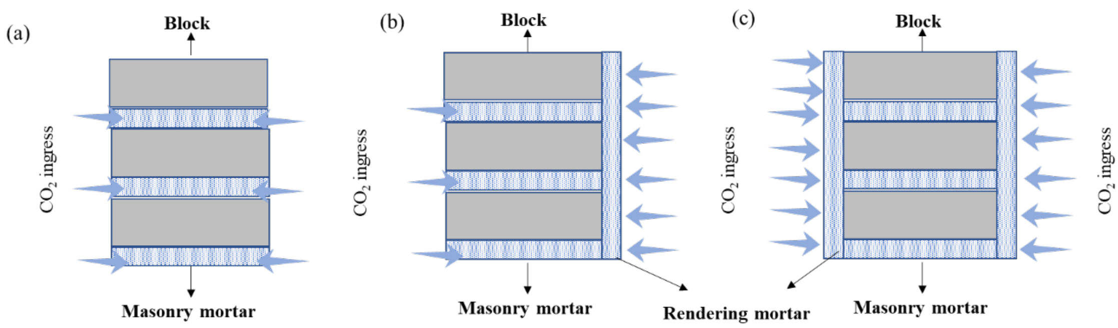

Fig. 4. The carbonation model for masonry mortar and masonry mortar actual use in real life. The top image is a schematic representation of the carbonation model for masonry mortar. (a) masonry mortar without rendering; (b) masonry mortar with one-side rendering; (c) masonry mortar with two-side rendering; 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.

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

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

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

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.

$$C_{waste} = C_{wastecon} + C_{wastemor} \quad (17)$$

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:

$$C_{wastecon} = (\sum_1^n W_{ci} \times f_{con} \times r_{con}) \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma \times \frac{M_{CO_2}}{M_{CaO}} \quad (18)$$

$$C_{wastemor} = \sum_1^n W_{mi} \times f_{mor} \times r_{mor} \times f_{cement}^{clinker} \times f_{clinker}^{CaO} \times \gamma_1 \times \frac{M_{CO_2}}{M_{CaO}} \quad (19)$$

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.

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

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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2. **Specific comment:** Second, since this is a data paper, it would be beneficial to include a table of your sample data in the main text. This will aid readers in understanding your data and variables better.

   Response: Thank you for your comment. Indeed, tables will aid readers in understanding data and variables better. We originally planned to include a table in the main text. However, it involves 92 annual data and multiple indicators over the period of 1930-2021, which are not aesthetically presented in a tabular form in the main text. In addition, the collected and resulted data involved in the article have been detailed in the dataset (https://doi.org/10.5281/zenodo.7516373) and presented in a better way as figures in the main text. So, there is no need to add table or change figure to table with same data. Finally, we decided to maintain the original figure format of the main text without addition of cumbersome tables.

3. **Specific comment:** Third, the uncertainty is calculated using the Monte Carlo method. It is essential to compare your results with those of previous studies to validate your estimates.

   Response: Thanks for your suggestion. We have added comparative explanation in the main text to validated our estimates. Generally, there are only a few researches (Xi et al., 2016; Guo et al., 2021; Cao et al., 2020) covered the global cement carbonation uptake, others only focusing on a specific area such as Spain (Sanjuán et al., 2020), Nordic countries (Pade and Guimaraes, 2007), The taipu Dam (Possan et al., 2017). In addition, those reaches that have the same boundary use different methods, specifically, iterative updating. Method in Guo's research was updated from Xi's study, which has been specified in Guo's paper. (Guo et al.,2021).

Changes: In the revised version of line 367-377, adding 'Furthermore, in order to establish the validity of this study, we attempted cross-validation. Generally, the coverage of the global cement carbonation uptake within the existing research is limited, with only a handful of studies (Xi et al., 2016; Guo et al., 2021; Cao et al., 2020) delving into this area. The majority of research focuses solely on specific regions, like Spain (Sanjuán, et al., 2020), Nordic countries (Pade and Guimaraes, 2007) or particular structures, such as The Itaipu Dam (Possan et al., 2017). Moreover, there is a notable discrepancy in the methodologies employed among studies that share similar scopes. Notably, the iterative updating approach is utilized in various studies but with distinct variations. For instance, Guo's research method builds upon the foundation established by Xi's work, a progression that Guo elaborates on in their paper (Guo et al., 2021).'

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CC 3

Comment: This study analyzes the global and regional uptake of CO_2 by cement material through carbonation from 1930 to 2021. This study is of interest for the global carbon community, as it is important to more accurately account for sources and sinks of CO_2 by cement-containing materials for better estimation of its impact on the carbon cycle. However, the manuscript is not clear for certain aspects of the study. Please, find my comments below.

Response: Thank you for your precious comments and suggestions. Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. The responds to the reviewer's comments are as following:

1. **Comment:** What is your contribution compared to the previous study (Such as Guo et al., 20201; Xi et al., 2016; Cao et al., 2020)? Please justify the importance and advancement of this dataset.

   Response: Thanks for your questions. We noticed this when we developed our works. Basically, our work is the extension of Guo's work. We calculated the carbon uptake from cement since 1930 till 2021 which is 3 years further than his study. But we keep the methodology same for a more systematic and accurate dataset generation and updating. Thus, difference between Xi's work and us is the same as that between Xi's and Guo's which has been discussed in Guo's paper (Guo et al., 20201). Cao's also make an improvement under this theme especially establish the future estimation system but the focus of this study is the amount of 2016. Thus, as we described in the texture, our work aims at updating the data within the same framework, enhancing the completeness of our database, thereby providing a reliable data foundation for our future forecasting endeavors. Plus, as you mentioned below, we included the data during the pandemic, which is also our spark.

2. **Comments:** Please provide reasons for regional division.

   Response: Thanks for your question. In our previous study (Xi et al., 2016), the world cement production was geographically divided into four primary countries and aggregated regions, including China, the United States (US), Europe and central Eurasia (including Russia), and the rest of the world (ROW). The cement production in ROW is obtained by subtracting China, the United States, and Europe and central Eurasia from global cement data. In our subsequent study (Guo et al., 2021), we noticed that India has now become the second-largest cement producer after China, with approximately 8 % of the world total in 2014 (IEA and WBCSD, 2018), then it divided geography 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) (Guo et al., 2021). The data of India was directly collected from United States Geological Survey (USGS). The cement production in ROW is obtained by subtracting China, the United States, Europe and central Eurasia, and India from global cement data. To keep the consistency with the prior geographical division (Guo et al., 2021), thus, we also use this division for our study. Meanwhile we followed USGS's geographical category of the cement production (US, China, EU, India, rest of world) to make our data source more convincing. USGS is one of the most completable databases of cement production which has the same statistics standard and criteria for each area. When we collected these data, we also considered to create a database manually by using other data source such as national statistics year books. But it is hard to combine these data with different statistics standards and criteria. Finally, we divided the world into these 5 areas.

3. **Comments:** The cement production process is an energy-intensive and CO_2-emitting process. I find you only focused on the CO_2 generated by the decomposition of calcium carbonate. What about the carbon emissions generated by energy consumption?

   Response: Appreciate for your comment. Generally, according to the definition of IPCC's carbon emission method (IPCC, 2006), emissions in cement production arise from fuel combustion (to heat limestone, clay, and sand to 1450 °C) and from the calcination reaction. Obviously, this kind of CO_2 in fuel combustion can be regarded as unnatural process in cement producing. There is a big potential to replace the current energy source to the renewable one and increase energy efficiency to reduce the CO_2 emission. (IPCC Fourth Assessment Report: Climate Change 2007: https://archive.ipcc.ch/publications_and_data/ar4/wg3/en/ch7s7-4-5.html) However, the processing emission that we defined in our study is a natural one which means it is hard to change the fact via a technical way. We noticed there are a lot of researches focusing on improving materials'(clinker) structure and characteristic to reduce the embodied carbon. However, they are not mature for industries currently. Thus, we decided to compare this kind of emission amounts to our uptake amount to show the potential of carbon reduction, which can solve the real issues in the real production and industry. This is what we do think having more practical value.

4. **Specific comment:** There are many types of cement, including fly ash cement, steel slag cement, etc. Sometimes, cement production does not originate from the decomposition of calcium carbonate directly, instead it is the mixing of purchased cement clinker. Will it affect the evaluation of carbon emissions and cement carbonization absorption in the cement industry process?

   Response: Thanks for your question. Now, with the development of technology, the addition of alternative materials such as steel slag, fly ash, natural pozzolans in cement has already increased in recent years (Schneider et al., 2015; Xu et al., 2022). The addition of clinker substitutes reduces the use of clinker, thereby reducing the process carbon emissions from limestone calcination (Xu et al., 2022). Indeed, the cement constituents has a significant impact on the cement process emissions. This means that using clinker production is more accurate than using cement production when calculating cement process emissions (Andrew, 2019). Like other study (Andrew, 2019), we try to use clinker production to accurately calculate cement process emission in this study, while there is no cement clinker statistics, we use the cement clinker ratio parameter recommended by IPCC to calculate the cement process emissions (Andrew, 2019). In this study, to maintain data homology with the cement carbon absorption formula, we use cement production and variant clinker ratio to calculate cement process emissions. Certainly, the variant clinker ratio is transformed from clinker production and cement production, and the clinker production has been corrected by import and export.

   The theme of the article is to calculate the carbon absorption of cement. There are many types of cement, and using cement production to calculate cement carbon absorption is correct. If only clinker is used to calculate cement carbon absorption, the carbonization of additives in other types of cement will be excluded, which will underestimate the amount of cement carbon uptake. Certainly, the cement additives will also affect the carbonation of cement due to the alkaline minerals such as CaO in the cement additives. In this study, we have considered the effect of additives on cement carbonization through the correction coefficient of additives, which has expressed in the SI-Table 1 (data can be accessed from SI-Table 1 of sheet 10 of from https://doi.org/10.5281/zenodo.7516373) and method of formula (1) in Supplementary document.

Changes: In the revised Data and Methods part, we further indicate the impacts of cement addition on carbon emission, for example "Given the current types of cement additives, if statistical data on cement clinker production is available, it is recommended that cement clinker production data be used directly to accurately estimate process emissions (Andrew, 2019)." in line 151-154.

5. **Comments:** The updated data is during the period of the Covid-19. Please add the detail elaboration on the impact of the Covid-19 on cement carbon emissions and uptake.

   Response: Thanks for your suggestion. In our work, we mentioned this in the text (line 39-42, 361-364, 429-432). According to our calculation and estimation, the pandemic showed little impact on global cement industry. It is a fact during these years, the global carbon emission increased, but this can be explained by 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%). We noticed there are many reports mentioned it is indeed affected by the pandemic but it is from perspectives of supply chain, consumption and labor and also showing the imbalance of demand and production. (Schlorke et al., 2020). This can also be a proof of our results.

Changes: In the revised version of line 266-278, we have added more expression on the impact of pandemic on cement uptake, for example "Meanwhile, based on our calculation, during the pandemic (2020-2021), the global cement producing amount shows a continuous increasing trend since 2019, leading the CO_2 emission rising. Globally, the producing amounts for 2020 and 2021 are 1590.38 and 1819.48 Mt respectively, ROW's contribution ranked first, from 495.75 in 2020 to 725.83 Mt in 2021. It is believed that in 2021, with the recovery of pandemic, The demand for cement increases alongside the resumption of delayed construction projects during the pandemic. (Schlorke et al., 2020). But China is an exception, showing a slight drop on the cement production during 2019 to 2021 with 752.40, 774.45 and 748.64 Mt separately. This can be explained by the stick restriction policy and property crisis in China in 2020 and 2021. (Hale et al., 2022)"

6. **Specific comment**: I suggested that the authors could provide a clearer explanation of the importance of their research in achieving the goal of global carbon neutrality. They could further elaborate on why this issue is important and how their research can contribute to addressing it. Additionally, they could explore the practical application of carbon capture technology, as well as the cost and feasibility of this technology.

   Response: Thanks for your suggestion. The importance of our series of research were in building cement carbon uptake accounting methods and quantitative calculation of its carbon absorption, which has made up for the lack of methods in the IPCC national greenhouse gas inventories guideline (IPCC, 2006; Xi et al., 2016), and provided data and technical support for precise calculation of global carbon balance and carbon neutrality. For example, in the global carbon budget report, it has begun to consider the impact of cement carbon sequestration on global carbon balance (Friedlingstein et al., 2022). According to the analysis conducted in the present study, the cement materials' annual carbon uptake in 2021 is equivalent to 7.67% of the global industrial process emissions of CO_2 (Friedlingstein et al., 2022), approximately 8.23 % of the average global land carbon sink from 2010 to 2020 (Friedlingstein et al., 2022), approximately 23.80% of the average net global forest sink from 1990 to 2007 (Pan, et al., 2011). The cement carbon sink of China alone in 2021 was about 0.43 Gt CO_2 yr^{-1}, which accounts for 48% to 60% of the terrestrial carbon sink in China during the past decades (Yang et al., 2022). The substantial cement carbon sequestration making it one of the important carbon sinks that cannot be ignored in the national and global carbon cycle and carbon neutrality evaluation. Meanwhile, the carbonization of cement materials is considered as one of the most promising carbon dioxide capture and storage technology. Scientists and engineers are inspired by the carbonization effect of cement to develop carbon capture and storage technologies by using construction waste (Skocek et al., 2020; Hargis et al., 2021).

   According to IPCC special report on carbon capture, and storage (CCS) (Rubin and Coninck, 2005; Kheshgi et al., 2012), in principle, CCS is technically feasible and plays a major role in long-term scenarios where there is significant reduction in greenhouse gas emissions. However, CCS through geological storage is also facing questions due to its cost-effective in reducing emissions, uncertain potential storage capacity, uncertain long-term impacts and stability of the storage sites. A potentially suitable alternative to the geological storage is the mineral carbonation (Sanna, et al., 2014), also called mineralization, i.e. the concept of storing CO_2 in the form of calcium and magnesium carbonates and to use. Now, expert community proposed the mineralization of concrete waste and their utilization in cement can be realized within the construction sector since the carbonatable materials come from demolished concrete and the carbonated paste comprise a part of cement used in new concrete, which is in line with the concept of circular economy and the conservation of natural resources (Skocek et al., 2020). Certainly, the carbon capture, utilization and storage (CCUS) technology of mineralization is technically feasible, but further research is still needed to reduce economic costs and identify suitable application department scenarios. In the future, use of alkaline mineral carbon sequestration to achieve emission reduction will play an important role in achieving carbon neutrality goals (Chiang and Pan, 2017; Hargis et al., 2021).

Changes: In the revised version, we added some expression in the Result and Discussions part to identify the importance of cement carbon sequestration. For example, the sentences "Our series of research in building cement carbon uptake accounting methods and quantitative calculation of its carbon absorption has made up for the lack of methods in the IPCC national greenhouse gas inventories guideline (IPCC, 2006; Xi et al., 2016), and provided data and technical support for precise calculation of global carbon balance and carbon neutrality. In the global carbon budget report, it has begun to consider the impact of cement carbon sequestration on global carbon balance (Friedlingstein et al., 2022). According to the analysis conducted in the present study, the cement materials' annual carbon uptake in 2021 is equivalent to 7.67% of the global industrial process emissions of CO2 (Friedlingstein et al., 2022), approximately 8.23 % of the average global land carbon sink from 2010 to 2020 (Friedlingstein et al., 2022), approximately 23.80% of the average net global forest sink from 1990 to 2007 (Pan et al., 2011). The cement carbon sink of China alone in 2021 was about 0.43 Gt CO2 yr−1, which accounts for 48% to 60% of the terrestrial carbon sink in China during the past decades (Yang et al., 2022). The substantial cement carbon sequestration making it one of the important carbon sinks that cannot be ignored in the national and global carbon cycle and carbon neutrality evaluation. Meanwhile, the carbonization of cement materials is considered as one of the most promising carbon dioxide capture and storage technology. Scientists and engineers are inspired by the carbonization effect of cement to develop carbon capture, utilization and storage technologies (CCUS) by using construction waste (Skocek et al., 2020; Hargis et al., 2021). Certainly, the CCUS technology of mineralization is technically feasible, but further research is still needed to reduce economic costs and identify suitable application department scenarios. In the future, use of alkaline mineral carbon sequestration to achieve emission reduction will play an important role in achieving carbon neutrality goals (Chiang and Pan, 2017; Hargis et al., 2021)." in the lines 365-389.

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