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?

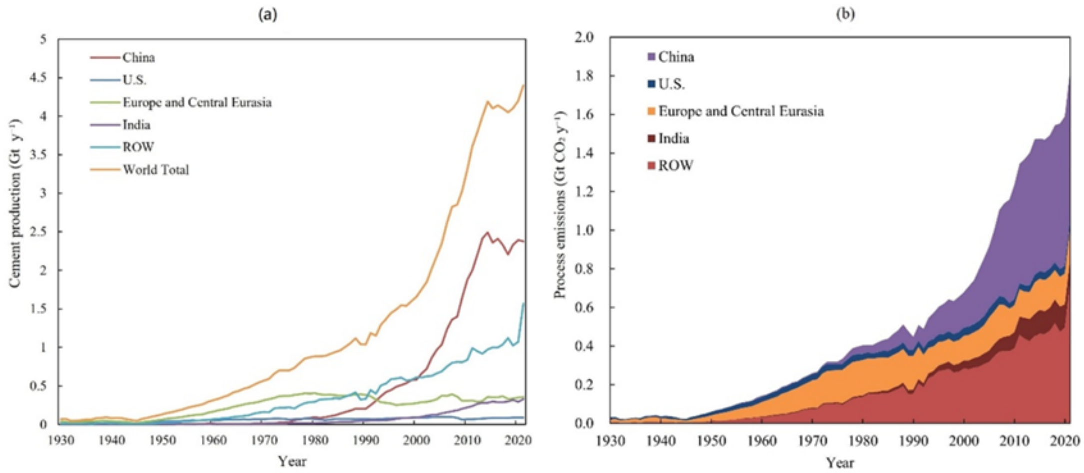
    Response: Sorry for making the confusing. 67% refers to in the 41.55Gt CO_2, 67% of it was emitted from 1930 to 1990. And, 71% of it was emitted from 1930 to 2019, which means the amount from 1990 to 2019 accounts for 4% of the totally global cumulative cement process CO_2 emissions till 2021.

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?

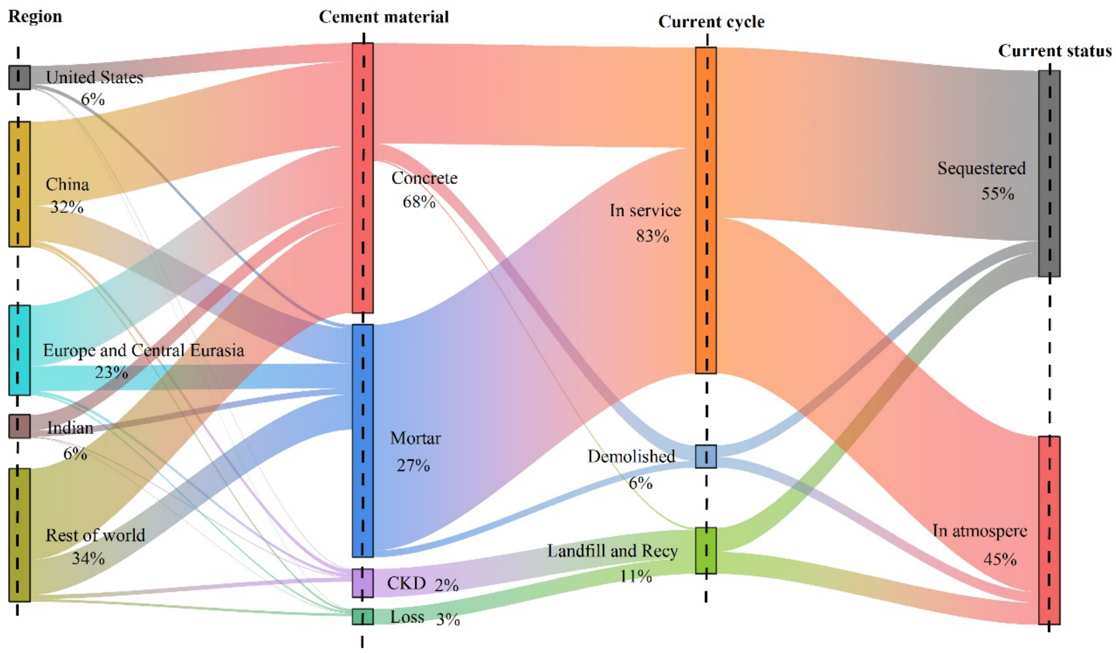
Response: In this work, to keep the consistency with the prior geographical division and data source, we continue to use the projected 2018 and 2019 cement production for Europe and Central Eurasia in the study of Guo et al., 2021. The 2020 and 2021 cement production for Europe and Central Eurasia in 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).

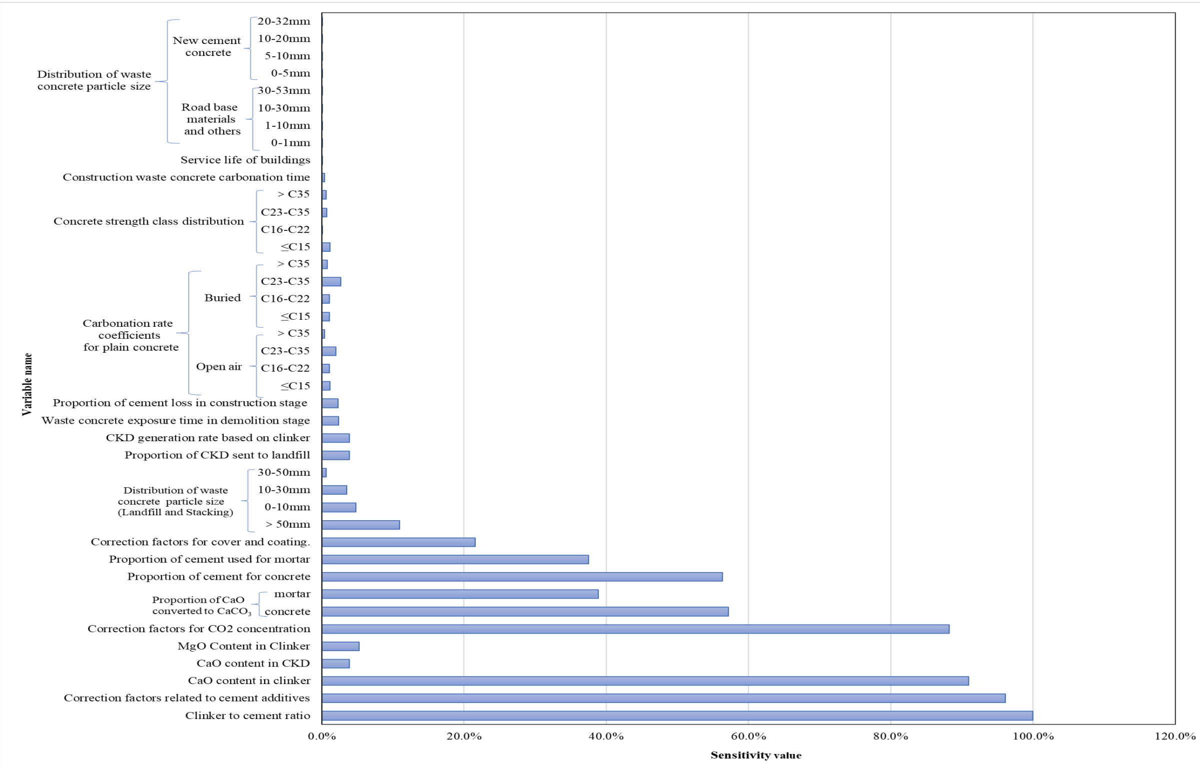
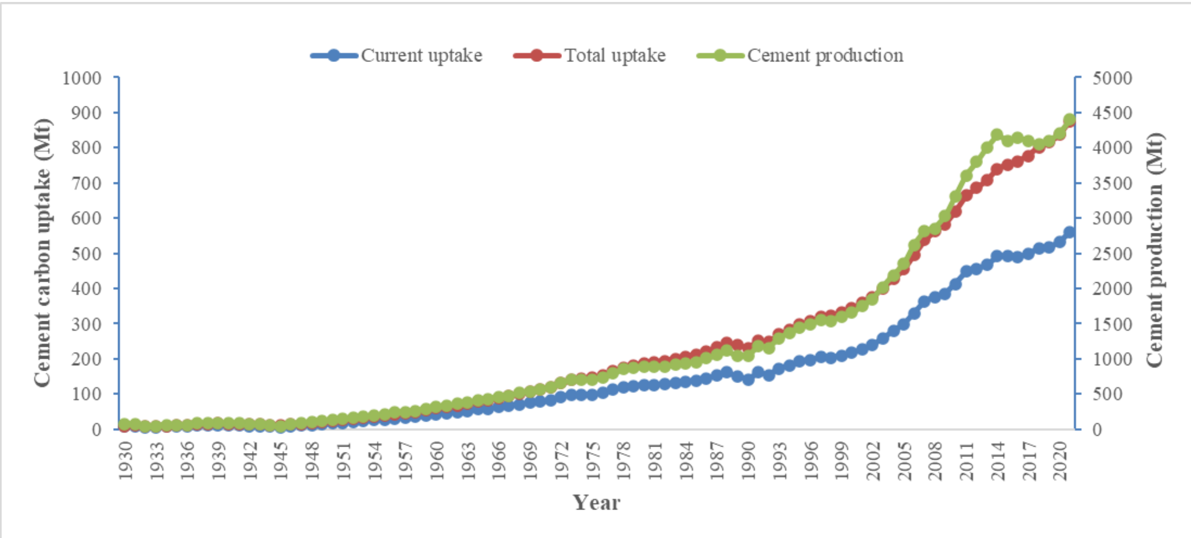


Fig.8 Sensitivity analysis of cement carbon uptake taking China's carbon uptake simulation as an illustrative case"

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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