ESSD-2024-437: Global and National CO_2 Uptake by Cement Carbonation from 1928 to 2024

General Comment

The manuscript, ESSD-2024-437, represents the fourth update of the Global Cement Carbon Uptake Database. Compared to previous versions, this update enhances the records to the country level, extends the temporal coverage, and reduces uncertainty by focusing on cement clinker production rather than apparent consumption. The manuscript is well-written, and I believe such an update is valuable to the community. Below, I have outlined several comments that may help improve this work.

Response: We are sincerely appreciated of the valuable comments and suggestions provided by the reviewer. Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have carefully revised the paper according to the reviewer' comments and provided comprehensive explanation of the revisions made to the manuscript and offered a point-by-point response.

Major Comments

1. **Forecasting for 2024**

   The ARIMA temporal forecasting model is commonly applied when data series exhibit high autocorrelation, such as seasonal cycles. However, the annual production and carbon uptake data in this study are strongly influenced by economic development and policy-making in one specific year, leading to high variability (as shown in Figure 4). How do the authors justify the use of ARIMA in this context?

   Response: Thank you very much for your comment. The ARIMA model is one of the valuable projecting techniques in forecasting to the upcoming events in time series analysis. The model is particularly effective for modeling temporal dependencies and forecasting when there is significant autocorrelation, even in the presence of high variability. This model has been widely used for forecasting production in various industries, including oil (Ning et al., 2022), gold (Mutele and Carranza, 2024), and fossil fuels (Ediger et al., 2006), and its applicability has been validated through comparisons with other forecasting models. In this study, as you are concerned, cement production is influenced by economic and policy, but we used a longer time series (1928-2023) to forecast production for 2024, which extended dataset allows the model to account for both short-term fluctuations and long-term trends, enhancing the robustness of the forecast. Furthermore, the differencing process ARIMA model reduces the impact of external factors by removing long-term trends from the data. To further validate the model, we conducted forward-chained cross-validation by progressively rolling the training set forward, evaluating the model's predictions at each step, and calculating the root mean square error (RMSE) for each forecast. We then compared the average of all the RMSE values with the standard deviation (SD) of the time series data. Generally, RMSE values exceedingly twice the SD suggest that the model's predictions are less reliable. Our results show that the ratio of RMSE to SD for each country ranges from 0.022 to 0.516, indicating that the models exhibit relatively small errors. The following Table 1 illustrates the validation of the model for 42 countries, with information on the remaining countries provided in SI data 1 in Supplementary table1.

Table 1: Summary of ARIMA models and their cross-validation results for 2024 cement production forecasts for 42 countries

Code	ISO	Countries	ARIMA Method	RMSE	SD	RMSE/SD
1	AUS	Australia	ARIMA (0,1,0)	244.10	3179.1	0.077
2	AUT	Austria	ARIMA (1,1,0)	208.79	1950.2	0.107
3	BEL	Belgium	ARIMA (2,1,3)	505.73	2198.4	0.230
4	BGR	Bulgaria	ARIMA (0,1,0)	214.04	1786.4	0.120
5	CAN	Canada	ARIMA (0,1,1)	636.66	4631.6	0.137
6	HRV	Croatia	ARIMA (0,1,0)	103.72	1331.4	0.078
7	CYP	Cyprus	ARIMA (1,1,0)	79.11	627.4	0.126
8	CZE	Czechia	ARIMA (0,1,0)	624.55	3299.5	0.189
9	DNK	Denmark	ARIMA (0,1,0)	177.17	741.1	0.239
10	FIN	Finland	ARIMA (0,1,0)	115.70	553.2	0.209
11	FRA	France	ARIMA (0,2,1)	995.70	8976.8	0.111
12	DEU	Germany	ARIMA (0,1,0)	2193.67	13967.9	0.157
13	GRC	Greece	ARIMA (0,1,0)	508.48	5753.4	0.088
14	HUN	Hungary	ARIMA (0,1,1)	561.58	1698.3	0.331
15	IRL	Ireland	ARIMA (1,0,0)	477.69	2250.5	0.212
16	ITA	Italy	ARIMA (0,1,0)	1978.69	15521.0	0.127
17	LUX	Luxembourg	ARIMA (2,1,2)	36.31	394.3	0.092
18	NLD	Netherlands	ARIMA (0,1,0)	173.91	1282.7	0.136
19	NOR	Norway	ARIMA (2,1,2)	126.16	726.9	0.174
20	POL	Poland	ARIMA (0,1,0)	869.71	6756.4	0.129
21	PRT	Portugal	ARIMA (1,1,0)	592.86	3514.8	0.169
22	ROU	Romania	ARIMA (0,1,0)	563.89	4762.6	0.118
23	SVK	Slovakia	ARIMA (0,1,1)	228.89	1575.7	0.145
24	SVN	Slovenia	ARIMA (1,1,0)	62.46	489.6	0.128
25	ESP	Spain	ARIMA (1,1,0)	1297.60	13601.9	0.095
26	SWE	Sweden	ARIMA (0,1,0)	167.96	927.1	0.181
27	CHE	Switzerland	ARIMA (1,1,0)	230.72	1650.4	0.140
28	GBR	United Kingdom	ARIMA (0,1,1)	784.47	3841.1	0.204
29	USA	USA	ARIMA (0,1,1)	3591.85	24688.3	0.145
30	MEX	Mexico	ARIMA (0,1,1)	1047.13	15544.8	0.067
31	BRA	Brazil	ARIMA (4,1,0)	1405.55	21850.8	0.064
32	EGY	Egypt	ARIMA (3,2,1)	1198.08	18044.4	0.066
33	TUR	Turkey	ARIMA (2,2,2)	1764.20	25147.4	0.070
34	IRN	Iran	ARIMA (1,1,0)	1319.29	21925.7	0.060
35	SAU	Saudi Arabia	ARIMA (2,1,3)	1137.73	18218.0	0.062
36	IND	India	ARIMA (0,2,1)	3749.43	105504.2	0.036

37	CHN	China	ARIMA (0,2,2)	29719.75	801120.9	0.037
38	KOR	South Korea	ARIMA (0,1,1)	1427.65	22713.0	0.063
39	JPN	Japan	ARIMA (1,2,2)	2550.74	31584.8	0.081
40	VNM	Vietnam	ARIMA (1,2,1)	1291.98	30389.8	0.043
41	IDN	Indonesia	ARIMA (2,2,3)	1177.49	21676.9	0.054
42	ZAF	South Africa	ARIMA (0,1,0)	436.09	4640.0	0.094

Changes: We performed cross-validation on the ARIMA forecasting model applied to 163 countries and included the results in the Supplementary tables (available from https://doi.org/10.5281/zenodo.14583866 Wu et al., 2024), with corresponding descriptions provided in the main text: "we use the ARIMA model to forecast cement production for the year 2024 and cross-validated the model (see SI data 1 in Supplementary table 1 for details)."

2. **CO_2 Uptake Characteristics**

The CO_2 uptake ability of concrete theoretically decreases significantly over time due to surface calcification. How does the CO_2 uptake model (Table 1) account for this characteristic? Including explicit figures to demonstrate this phenomenon would strengthen the analysis.

Response: Thank you very much for your valuable comments. The ability of concrete to absorb CO_2 does decrease significantly with surface calcification. Our carbon uptake model is primarily based on Fick' s second diffusion law, as proposed by civil engineering researchers (Andersson et al., 2013). According to this principle, the depth of carbonation (d) is proportional to the product of the carbonation rate (k) and the square root of the exposure time (t) (see Eq. (8) in the manuscript).

$$d = k \times \sqrt{t}$$

This model has been verified by many experimental studies (You et al., 2022), Figure 1 shows that the overall carbonation depth of the cement under different treatment is linearly related to the square root of time, with correlation coefficients above 97%.

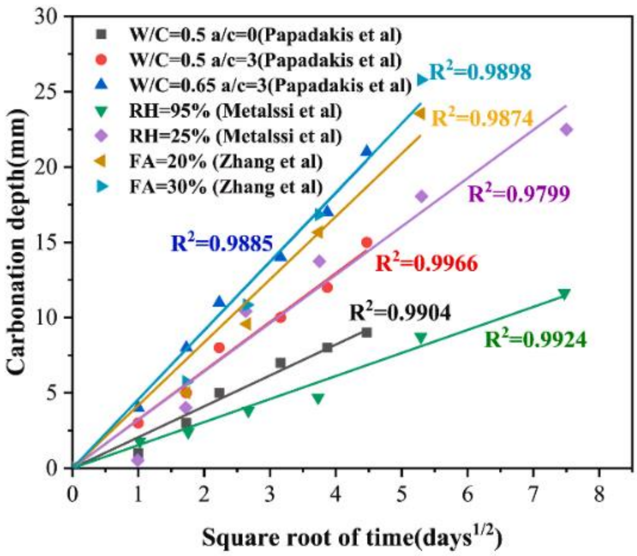


Figure 1. The relationship between carbonation depth and square root of time. W/C: water-cement ratio; a/e: aggregate-cement ratio; RH: relative humidity; FA: the dosage of fly ash (source: You et al., 2022. DOI: 10.1016/j.cemconcomp.2021.104315)

This empirical model is further adapted based on the type of concrete used in the CO_2 uptake model (Table 1 in Manuscript). Under this framework, the carbonation depth of cementitious materials is tied to the square root of their exposure time, meaning the annual rate of carbonation slows over time. The connotations of the model have been fully explained in our previous studies, and details can be found in the Methods part and Supplementary Figure 3 in Xi et al (2016). Considering that this study is mainly further update of the result of cement carbon uptake, the methods part is not repeated. However, as you mentioned in your comment, we have further refined the results to better demonstrate this feature. Specifically, we have included the time-lag data for cement carbon uptake in the supplementary data file (SI data 2 in SI table 3). The Figure 2 below highlights the time-lag effect on carbon uptake by cement from 1928 to 2024 (it is a refinement of the historical year carbon uptake in Figure 1e in the manuscript). As shown, the carbon uptake of the same batch of cement materials gradually decreases over time. For example, the carbon sequestration from global cement consumption in 1990 amounted to 121.0 Mt CO_2 during that year, whereas by 2023, the sequestration from the same cement has decreased to only 2.0 Mt CO_2.

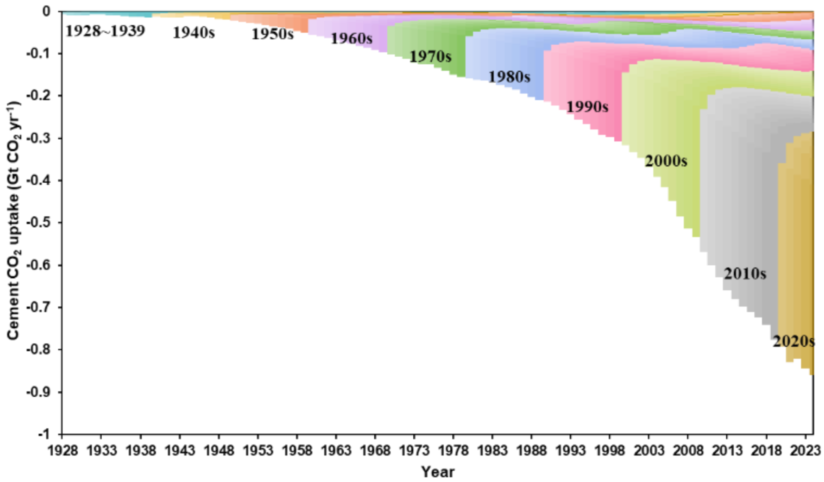


Figure 2. Time-lag effect on carbon uptake by cement from 1928 to 2024. Different colors represent changes in carbon sequestration over time for different years of consumption of cement.

Changes: We have added year-by-year data on carbon sequestration by cement materials to the annexed results file to demonstrate the time lag effect (SI data 2 in Supplementary table 3, available from https://doi.org/10.5281/zenodo.14583866 Wu et al., 2024).

In addition, we have added the reference to the method explanations in Section 2.3 and included Figure 2 along with its description in Section 3.1 of the revised draft. "Specifically, the annual carbonation rate of cementitious materials shows a steady decline (Figure 2). Cement materials can not fully carbonize in one year, they continue to absorb atmospheric CO_2 for the subsequent 100 years. Furthermore, the amount of carbon absorbed decreases as the depth of carbonation increases. For example, the carbon uptake of cement consumed in 1990 was 121 Mt, while the sequestration from the same cement has decreased to only 2.0 Mt in 2023."

3. **Input Data Summary**

It is recommended to summarize the metadata of input data (e.g., time span, resolution, references, and data links) in a table for ease of reference.

Response: We sincerely appreciate the detailed review and insightful suggestion. There are two input data tables in the Supplementary tables (SI table1 and SI table2), they are activity level data and cement uptake parameters, respectively. We've added the summary of the data to the front of these tables based on your suggestion.

Changes: We change the "Index" table in the Supplementary tables (available from https://doi.org/10.5281/zenodo.14583866 Wu et al., 2024) to a "Summary" table containing a summary of the input data information.

4. **Figure 1b**

The carbon offset levels in Figure 1b show a clear overall increasing, stable, trend (unit as percentage) over the past 100 years. Considering the construction substantially increased over the past century, does this indicate that the carbon uptake efficiency of materials is increasing over time? I did not follow. Additionally, uncertainty levels should be provided in this figure. The explanation of short-term disturbances, such as World War II, is reasonable, but the manuscript lacks interpretation for the long-term stable increase in carbon offset levels.

Response: Thank you very much for your helpful comment. Firstly, the steady increase in the offset level in Figure 1b does not imply that the rate of cement carbon uptake is necessarily increasing. In this figure, the offset level represents the ratio of global cement carbon absorption to cement carbon emissions. Since the activity level data of cement carbon uptake and carbon emission are all based on the consumption of cement clinker, the offset level seems to be related to the ratio of the carbon uptake parameters to the emission factor only. However, it is important to note that the accounting for cement carbon sequestration differs from that of cement carbon emissions (calculated as activity level data × emission factors). The key distinction lies in the fact that cement materials, particularly concrete, cannot fully carbonate within a single year. As a result, cement carbon sequestration exhibits a time-lag (Figure 2) relative to carbon emissions. In other words, cement carbon sequestration accumulates gradually over the years, unlike carbon emissions from cement production process which occur instantaneously. This is the primary reason for the steady increase in cement carbon offsets over time. Furthermore, since the carbon offset level in this figure is defined as the ratio of carbon sequestration to carbon emissions for cement, its uncertainty inherently stems from the uncertainties in both carbon sequestration and carbon emissions. However, determining the uncertainty of this ratio does not seem to have a significant or disproportionate impact on decision-making or practical applications. Therefore, this study prioritizes the quantification of carbon sequestration and the interpretation of its uncertainty, rather than overemphasizing the uncertainty of the offset ratio itself.

Changes: We have included an explanation of the long-term stable increase in carbon offset levels in the section 3.1 of the manuscript: "Unsurprisingly, the carbon offset level (uptake-to-emission ratio, Fig. 1b) show a clearly overall increasing trend over the past nearly 100 years. This trend is primarily due to the time-lag of cement carbonation, unlike the transient carbon process emissions from cement, the gradual accumulation of historical carbon sequestration results in a steady increase in carbon offset level. This effect becomes particularly evident during periods of declining cement production."

5. **Discussion on Cement Carbonation Risks**

Page 10, Line 237: The authors call for inter-industry collaboration to maximize CO_2 uptake from cement materials. While this is an important goal, it is worth noting that cement carbonation significantly reduces the durability of constructions. Reconstruction necessitated by reduced durability would lead to additional carbon emissions. Could the authors discuss the potential risks associated with relying on carbonation as a pathway to achieving carbon neutrality?

Response: We are especially thankful for your detailed feedback on cement carbonation risks, which has been very helpful. Carbonization of cement materials does significantly reduce the durability of buildings (Zhang et al., 2025). According to Huang et al. (2024), the carbon emissions from the production phase of building materials and the construction phase account for 15.6% and 1.6% of their full life cycle, respectively. Therefore, we can't reconstruct just to enhance the carbon absorption capacity of cement which exacerbate the risk of increased emissions. Instead, we should focus on extending the lifespan of buildings to align with advancements in engineering technology. For instance, the average building lifespan in some European countries is currently around 70 years, approximately 65 years in the United States, but only 35 years in China (SI data5 in Supplementary table 2, available from https://doi.org/10.5281/zenodo.14583866 Wu et al., 2024). Moreover, further research remains necessary to identify appropriate scenarios for departmental application. For example, Ostovari et al. (2021) analyzed the carbon footprint of the combined CO_2 mineralization and cement production based on life cycle assessment, and showed that integrating CO_2 mineralization with cement production has the potential to transform the cement industry from an unavoidable CO_2 source to a CO_2 sink. Given mineralization of cementitious materials as an important carbon neutral pathway, it is essential to evaluate its emission reduction potential and associated risks before implementing it in practice.

Changes: In order to avoid misunderstandings in the presentation, we have changed the presentation in the section 3.1: "The significant carbon sequestration of cement materials makes them one of important carbon sinks in the global carbon cycle. Moreover, the potential of carbon capture, utilization and storage (CCUS) can contribute 36 % of the reduction for the cement industry to achieve net-zero emissions (IEA, 2024). Many studies have explored the mechanisms and properties of accelerated carbonation in cement materials, including concrete (Alshalif et al., 2021, 2022), cement paste (Castellote et al., 2008; Morandeau et al., 2015), slag cement (Mo and Panesar, 2013), and CKD (Pu et al., 2023). Ostovari et al. (2021) demonstrated that integrating CO_2 mineralization with cement production has the potential to transform the cement industry from an unavoidable CO_2 source to a CO_2 sink. Certainly, carbon capture is widely regarded as the only viable solution for significantly reducing CO_2 emissions from cement production to meet the 2050 mitigation targets (Schneider, 2019), but further research is required to assess the economic costs and potential risks associated with their implementation."

6. **Comparison with Previous Studies**

As this study is an update of Huang et al. (2023) with some shared figures but updated results, it would be helpful to include an explicit comparison with previous reports. Are there any revised conclusions, corrections, or new insights presented in this update?

Response: Thank you for your valuable comments and suggestions. In Section 2.2 of the manuscript, we provide a detailed discussion of the improvements made in this study compared to the previous research (Huang et al., 2023). As mentioned in the manuscript, the key update of this study focuses on a more refined analysis of cement carbon sequestration across different countries worldwide. While the earlier version only covered the United States, China, India, and European countries (Xi et al., 2016; Guo et al., 2021; Huang et al., 2023), this study first expands the scope to include 163 countries. This refinement in country-level accounting for cement CO_2 uptake allows us to uncover trends in the global distribution of carbon sequestration by cement materials over nearly a century. Furthermore, we analyze the emission reduction characteristics of cement carbon sequestration in various countries, revealing that 21 countries have already achieved carbon neutrality in cement production. These findings represent one of the most significant new contributions of our study and is highlighted in Sections 3.2 and 3.3 of the manuscript.

Accounting for global cement carbon sequestration has been a primary goal of our research. In this study, we have expanded the time span of the global cement carbon sequestration database from the previous period of 1930 to 2021 to cover 1928 to 2024. Compared to global cement carbon uptake in 2021, our findings show a decline in 2022 and 2023, with respectively reductions of 1.1% and 2.8% from the previous year. However, global cement carbon uptake in 2024 is expected to experience a slight rebound, driven by strong market activity in Southeast Asia and Africa (Cheng et al., 2023), with an estimated increase of around 2.0% compared to the previous year.

In addition, this study calibrates cement clinker consumption, therefore two variables were excluded from the original model—the proportion of cement clinker and the ratio of cement consumption to production. This adjustment helps to reduce the uncertainty associated with these variables. This point is mentioned in the newly added section on uncertainty analysis.

Changes: We have added the description to section 3.1 of the revised draft. “The results show that global cement carbon uptake in 2022 is 0.82 Gt CO_2 (95 % CI: 0.69-0.98 Gt CO_2 yr^{-1}), a decrease of 1.1 % from 2021. It mainly attributable to the decline in both global cement production and apparent cement consumption in 2022, which decrease by 5.6 % and 6.2 % from 2021, respectively. In particular, as the largest cement producer, China's cement production and apparent consumption decreased by 11.1%. In 2023, global cement carbon uptake shows a 2.8 % increase from 2022, in which the global cement production declined by 1.4 %, but the apparent consumption of cement clinker increased by 2.0 %. This suggests a strong correlation between cement carbon uptake and cement consumption. A modest recovery in global cement consumption is anticipated for 2024, primarily driven by rapidly growing markets in South-East Asia and Africa (Cheng et al., 2023). This recovery is expected to correspond with a continuation of growth in the global cement carbon uptake, which is forecasted to reach 0.86 Gt CO_2 (95 % CI: 0.73-10.23 Gt CO_2 yr^{-1}), marking an increase of 2.0% from the 2023 levels.”

7. **Uncertainty and Future Directions**

Adding a dedicated section or paragraph to discuss data uncertainty and propose potential research directions would enhance the manuscript.

Response: Thanks very much for your opinion. The accounting results in this study are based on 10,000 Monte Carlo simulations, providing 95% confidence intervals for cement carbon absorption estimates across various materials, individual countries, and on a global scale (see Supplementary table 4). According to your suggestion, we conducted sensitivity analyses on the model parameters to clarify the impact of each parameter, offering valuable references for improving the accuracy of future accounting results.

Changes: We have added section 3.4 "Uncertainty Analysis" to discuss data uncertainty: "This study uses Monte Carlo method to simulate carbon uptake from the cement for 100,000 times to evaluate the uncertainty. The results reveal that the 95 % confidence interval for cumulative carbon uptake spanning from 1928 to 2024 ranges from 17.93 to 25.17Gt CO_2. The uncertainties associated with carbon sequestration from cement for each country are detailed in Supplementary Table 4. Our accounting is based on the accounting model of previous research (Huang et al., 2023), where the variable entries and sensitivity value are basically consistent with it (Fig 7). A key difference in our approach is that we have removed the two variables of "the proportion of clinker in cement" and "the ratio of cement consumption to production", because we use a more accurate cement clinker consumption in this study. For specific parameters, "CaO content in clinker" (92.0%) has the greatest impact on the scale of carbon absorption, because it widely affects the carbon absorption in all stages of cement consumption; secondly, "the proportion of cement used for concrete/mortar" and "the proportion of CaO converted to $CaCO_3$ in concrete/mortar", their sensitivity values are 66.4%, 27.5%, 67.09% and 28.72% respectively, due to these two parameters each affect the whole stage carbon absorption of concrete and mortar, and concrete and mortar account for the largest proportion of the total carbon absorption of cement materials. Other parameters have lower sensitivity values, mostly below 10%, because they only cause a slight impact on the local accounting results of the model. Therefore, it is essential to prioritize the more sensitive parameters and ensure their accurate collection and measurement across different countries to further minimize the uncertainty in the model's accounting results."

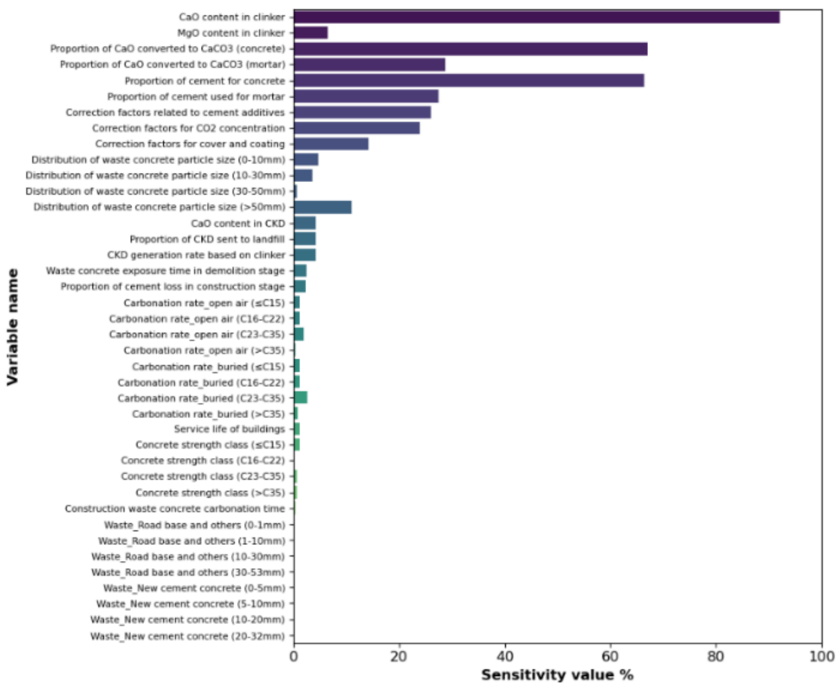


Figure 7: Sensitivity analysis of cement carbon uptake.

Minor Comments

1. **Page 2, Line 48**: The manuscript refers to cement carbonation as a "permanent CO_2 uptake method." Given that the carbon uptake ability changes over time, why is it characterized as permanent?

   Response: Thank you for your valuable comment. The term "permanent" in the manuscript refers to the long-term sequestration of CO_2, meaning that once CO_2 is fixed in a substance or geological stratum through mineralization or geological sequestration, it remains stable over time and is not released back into the atmosphere.

   Changes: We believe there may have been a misunderstanding. To clarify, we have replaced the term "permanent" with "long-term sequestration".

2. **Page 2, Line 55**: This report suggests a nearly 50% uptake from cement carbonation, which differs significantly from the 10% uptake reported by PCA. Could the authors explain this discrepancy?

   Response: Thanks very for your careful review. The significantly larger of cement CO_2 uptake offsets level in this study (46%) compared to the value (10%) in Portland Cement Association (PCA) of the United States can be attributed to the difference in accounting scope. While the PCA report only includes the carbon sequestration of concrete, our analysis covers a broader range of cement materials, including four types: concrete, mortar, construction waste materials, and cement kiln dust (CKD). In our result, the global carbon uptake from concrete materials accounts for approximately 15.4% of the total emissions from cement production (averaged over the past 100 years), which aligns with the findings in the PCA report. Notably, mortar emerges as the largest contributor to carbon sequestration, accounting for 48% of the total. However, due to limited data and model constraints, few studies have focused on mortar, representing an area for future optimization of our model, as discussed in the Outlook section of the manuscript.

Changes: We have emphasized in the manuscript where we introduced the PCA report that he was referring only to concrete materials: "…, highlights that concrete buildings can reabsorb up to 10% of the CO2 emitted during the cement and concrete production process".

3. **Page 3, Line 79**: Citing the previous three updates of the Global Cement Carbon Uptake Database in this section would help readers better understand the evolution of the dataset.

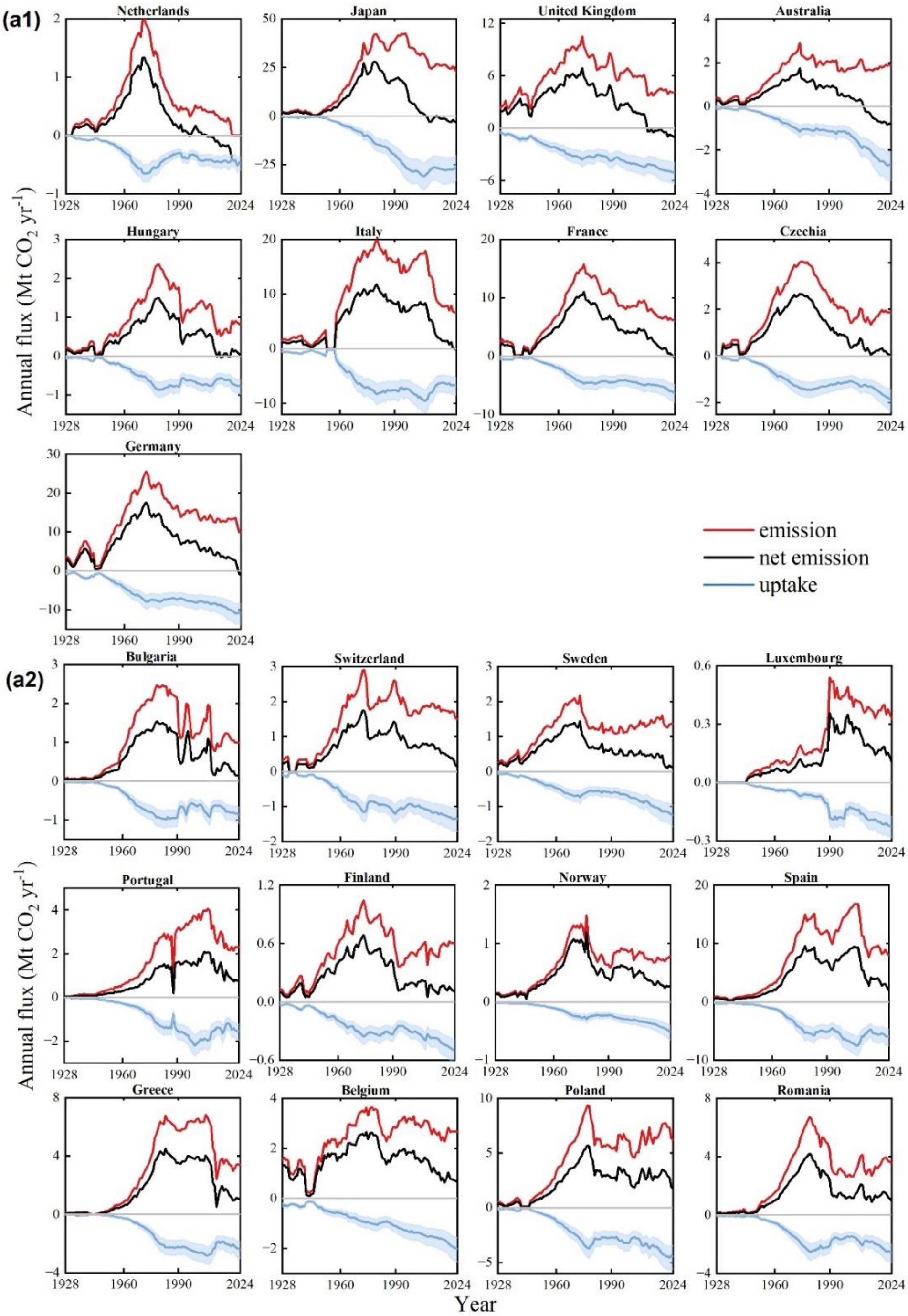
   Response: Thanks very for your careful review. The previous three database are Xi et al., 2016; Guo et al., 2021; Huang et al., 2023, respectively. We have cited them in the manuscript as you suggested.

   Changes: We have added the citation of the previous database in the manuscript.

4. **Figure 4 & 5**: provide full spells of the countries in Appendix or supplement would be helpful. Any possibility to include the uncertainty range?

   Response: Thank you very much for your suggestion, we have changed the abbreviation of the countries name in the Figure 4 to the full spelling and increased the uncertainty of the carbon uptake, the detailed data can be found in the Supplementary table 4 (available from https://doi.org/10.5281/zenodo.14583866 Wu et al., 2024).

   Changes: We have changed the figures as you suggested to the following:



(a1)
Netherlands
Japan
United Kingdom
Australia
Hungary
Italy
France
Czechia
Germany
emission
net emission
uptake
(a2)
Bulgaria
Switzerland
Sweden
Luxembourg
Portugal
Finland
Norway
Spain
Greece
Belgium
Poland
Romania
Annual flux (Mt CO_2 yr^{-1})
Year

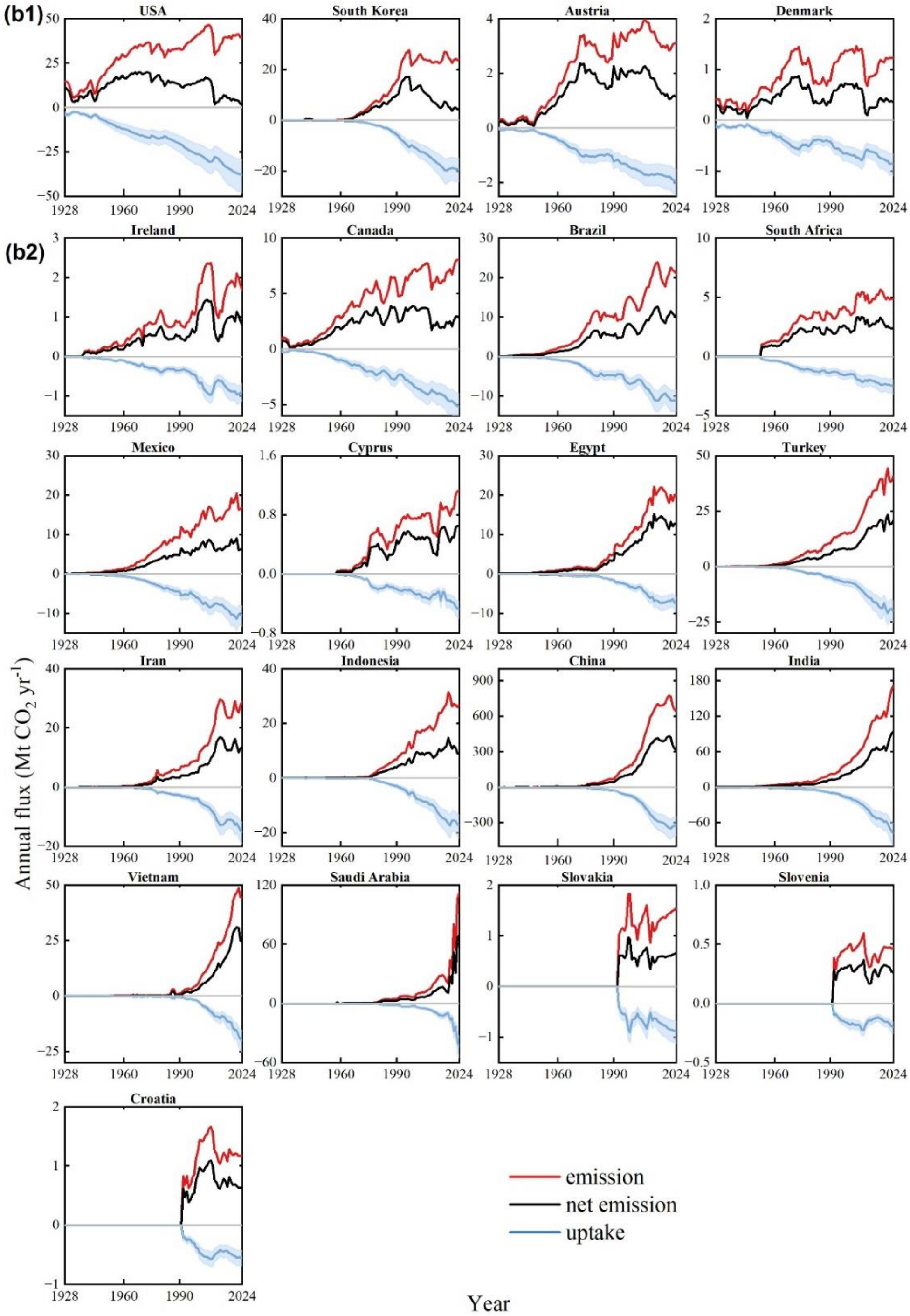


Figure 4: Cement carbon process emission and uptake in 42 countries during 1928-2024. (a) 21 countries in Group 1 that the cement process emissions have reached peaking. (b) 21 countries in Group 2 with process carbon emissions non-peaked.

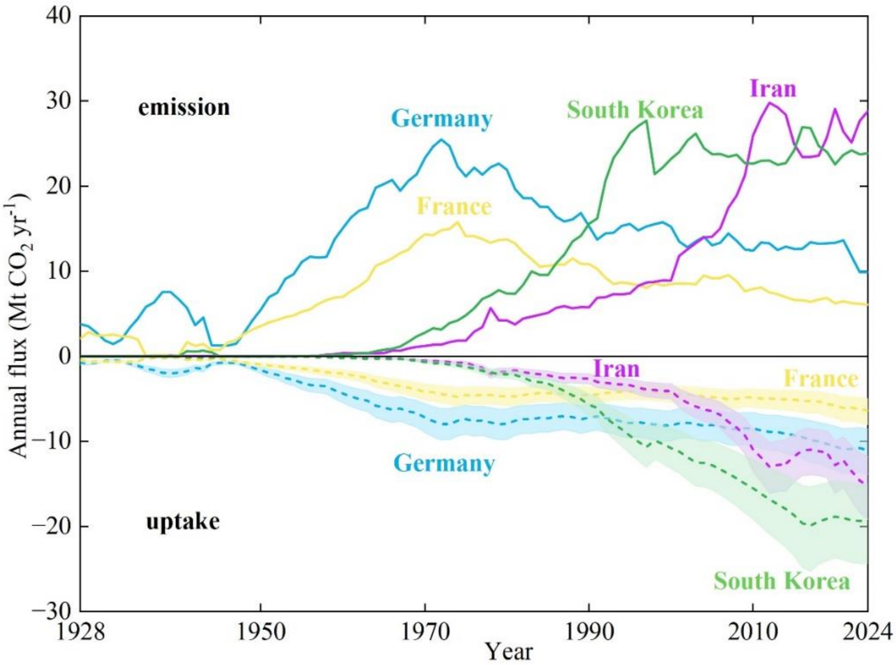


Figure 5: Comparison of trends of process carbon emission and uptake in peaked and non-peaked countries.

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