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

Reading this paper leaves me with the impression that they have probably done the analysis properly but it leaves me with many questions where the details are not sufficiently clear. Let me walk through the text and point our specific questions.

Response: Your expert review has been of great assistance in enhancing the scientific rigor and clarity of our work. We are truly thankful for your contribution. 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.

Line 36: I think we could use another sentence to make clear that there are 3 "types" of emissions discussed in this paper. Fossil fuel emissions, chemical process emissions, and the net emissions which are the sum of chemical process emissions and chemical uptake.

Response: We are grateful for your review. Your insight is correct, and it is necessary to provide a definition of net emissions to make it clearer for the readers. The emissions for the cement industry include both fossil fuel emissions and chemical process emissions. This study emphasizes the CO_2 sequestration by the cement. Therefore, we propose the definition of net emissions to reflect process emissions after deducting of cement carbon sequestration. This definition has been mentioned in previous studies (Xi et al., 2016). We have added it at the application of line 47 of the original text.

Changes: we have added the definition of "net emission" in line 47: "A substantial fraction of process CO_2 emissions from cement production is reabsorbed on a time scale of 100 years through natural carbonation of cement materials, the net cement emissions (namely industrial process of cement production minus the CO_2 sequestration from carbonation of cement materials) were only about 57% of the cumulative process emissions from 1930 to 2013 (Xi et al., 2016)."

Line 38: cites emissions for 2023 from a paper that was published in 2019?

Response: Thank you for your thoughtful comment. The article cited in here is "Andrew, R.M., 2019. Global CO_2 emissions from cement production, 1928–2018. Earth System Science Data 11, 1675–1710. https://doi.org/10.5194/essd-11-1675-2019." This article published in 2019 and presents global cement process carbon emissions from 1928 to 2018, the data is available at https://zenodo.org/records/11207133, The database is updated annually, with the latest version released in May 2024. The new data covers global cement process carbon emissions from 1880 to 2023. However, the citation for the data remains (Andrew, 2019).

Line 48: It seems to me that this uptake is discussed several times in the paper without recognizing that this is generally a deterioration of the cement and the properties for which we are producing the cement in the first place. Why would we want to ever

encourage the carbonation?

Response: We are extremely grateful for your thoughtful review. Carbonization of cement materials does significantly reduce the durability of buildings (Zhang et al., 2025). This paper emphasizes the significance of cement carbon absorption from the perspective of climate change. However, due to misstatements and other reasons, there has been a misunderstanding that we are blindly encouraging carbonation. Therefore, we have revised some of the descriptions. The cement carbon absorption discussed in this study refers to the natural carbonation process of cement, without accelerated carbonation. The focus of the study is to quantify the carbon absorbed in natural carbonation process. While this process contributes to sequestrate CO_2, carbonation of concrete reduces the material's strength and durability. Therefore, from a management perspective, we do not encourage promoting carbonation in concrete service life, except in the case of construction wastes management during the demolition and secondary use stages, which aligns with the management of construction wastes.

Changes: We have deleted the statement in lines 47-48 of the original text.

Line 49: We are a long way from net-zero emissions so long as fossil fuels are used to drive the process.

Response: We sincerely appreciate your insight. In this section, we referenced the cement roadmap to carbon neutrality proposed by cement association in Europe and United States, both of which mention cement carbonation as one of the zero-carbon pathways. This was intended to highlight the importance of accurately quantifying cement carbon uptake. However, the inappropriate phrasing in the original text may have led to the misunderstanding that we advocate for using carbonation as a way to drive the cement industry's net-zero emissions. Therefore, we have made revisions to this part. Thank you for pointing this out, it is of great assistance in enhancing the scientific rigor and clarity of our work.

Changes: We have modified the expression in line 49 of the original text to read: "The cement carbon uptake is helpful to achieve the cement net-zero ambitions for the cement industry"

Line 55: Somewhere in the text it needs to be clear whether this analysis supports or contradicts this 10%. It appears to me that this paper suggests values much larger than 10%.

Lines 363-364: I find this sentence hard to believe. It needs to be very clear. And the contrast with 10% in line 55 should be explicitly stated.

Response: Thanks for your thoughtful comment. 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.

Changes: We have emphasized in the manuscript where we introduced the PCA report that he was referring only to concrete materials: "highlights approximately 10% of the CO_2 generated during the manufacture of cement and concrete can ultimately be absorbed over the life of a concrete structure (not including cement mortar),…"

Line 62: I think this should be accuracy rather than precision? Likewise in line 68.

Response: Thank you very much for your suggestion. The optimization of the accounting model in this study is to make the estimation further reflect the true carbon uptake, so using accuracy is a more appropriate word. Your careful review has greatly helped us to improve the quality of our manuscripts.

Changes: We have reviewed the entire text and corrected several instances where 'precise' was used in place of 'accurate', the details are as follows:

Line 30: "This study provides an accurate bottom-up quantification to cement carbonation sinks at national and global levels."

Line 62: "Therefore, it is imperative that these uptake estimates are as accurate as possible."

Line 63: "However, due to the lack of detailed activity data and accurate carbonation parameters for various countries, …"

Line 68: "…is imperative to collect more accurate activity data on cement consumption with improved spatial resolution."

Line 104: "To provide a more accurate national-level database of carbon uptake in cement, …"

Line 361: "… and provide a more accurate bottom-up quantification."

Line 90: source = sources

Response: Thank you for your suggestion.

Changes: We have changed the source in Line 90 to sources.

Line 94: cement clinker data estimated how?

Response: Thank you for pointing this out. Cement clinker consumption data for each country were mainly obtained by multiplying the clinker-to-cement ratio with cement production figures. Cement production data for 163 countries and regions were primarily sourced from the USGS, while clinker-to-cement ratio data for China and India were obtained from statistical data and surveys in prior research (Xi et al., 2016; Guo et al., 2021; Huang et al., 2023). For other countries, cement-to-clinker ratio data were referenced from Andrew's estimates (Andrew, 2020).

Changes: We have changed the line 94 and 95 to: "Estimated cement clinker consumption data were derived by multiplying clinker-to-cement ratio (the ratio of cement clinker consumption to cement production) with cement production. Cement production data for 163 countries and regions from 1928 to 2022 were accessed from the United States Geological Survey (USGS)."

Line 95: spell out USGS one time and provide the reference.

Response: Thank you for your suggestion.

Changes: We have added the full spelling of USGS and the reference in Line 95: "Cement production data for 163 countries and regions from 1928 to 2022 obtained from United States Geological Survey (USGS)."

Line 98: define "cement clinker ratio"

Response: Thank you for pointing this out. The clinker-to-cement ratio is the ratio of cement clinker production to cement production.

Changes: We have added the define of clinker-to-cement ratio and changed the "cement clinker ratio" to "clinker-to-cement ratio".

Line 106: source of import - export data.

Response: Thank you for pointing this out. The imported and exported data accessed from UN Comtrade Database. https://comtradeplus.un.org/.

Changes: we have added the source of the import and export data of clinker.

Line 107: what is meant by "cement utilization proportion"?

Response: Thank you for pointing this out. The parameters presented here are the proportion of cement used for concrete and mortar in each country. In the accounting model for cement CO_2 uptake, the cement material is categorized into concrete and mortar at the construction service stage (Table 1 in the manuscript), and the estimation for carbon sequestration is constructed according to the respective utilization of these cement materials. Consequently, the amount of cement consumed for each material serves as important activity-level data, determined by multiplying their respective proportion by the total cement consumption.

Changes: We have modified the expression of the parameter here and added the relevant explanation: “The proportion of cement used for concrete and mortar in 42 countries, which is the share of concrete and mortar in total cement consumption respectively”

Line 111: why do we care about “strength class”?

Response: Thank you for your comment. Concrete strength is one of the fundamental performance indicators of concrete and serves as a comprehensive parameter for assessing its quality, which showed large impacts on carbonation rate. It is closely related to the water-cement ratio of the concrete, and it reflects the combined influence of factors on concrete quality such as cement type, cement content, aggregate type, admixtures, as well as construction quality and curing methods. The study shows that concrete with higher strength tends to have greater density, increased resistance to CO_2 diffusion, and a lower carbonation rate. The following table lists the carbonation rates for different concrete strengths, (see in Supplementary Table 2, can be accessed on Zenodo at https://doi.org/10.5281/zenodo.14583866). Therefore, in this study in order to account for the carbon sequestration of cement materials in different countries and regions, we further refined the distribution of concrete strength grades in 42 countries based on the previous study (see in data 2 in Supplementary Table 2)

Table: Concrete carbonation rate coefficients (K) for various concrete strengths and exposure conditions

Region	Exposure condition	Compressive strength (mm/(year)$^{0.5}$)			
Europe (Plain concrete)	k	≤15 MPa	16–20 Mpa	23–35 Mpa	>35MPa
	Exposed outdoor	5	2.5	1.5	1
	Sheltered	10	6	4	2.5
	Indoors	15	9	6	3.5
	Wet	2	1	0.75	0.5
	Buried	3	1.5	1	0.75
China (Plain concrete)	Exposed outdoor	6.1	3.9	2.4	1.3
	Sheltered	9.9	7.1	4.8	2.5
	Indoors	13.9	9.8	7.0	4
	Buried	3.8	1.9	1.0	0.5
	Wet	1.9	1.0	0.7	0.3
USA	uncoated	7.1	6.9	3.8-5.4	2.5
	Coated	n/a	3.5	1.9-2.7	n/a

Changes: We have added reasons for collecting data on concrete grades in different countries: “Concrete strength is a comprehensive parameter for assessing its quality and the carbonization rate generally decreases with increasing concrete strength class (Pade and Guimaraes, 2007), so we collected data on concrete strength classes for 42 countries."

Line 113: what is CEIC? Spell out once

Response: Thank you for your suggestion. CEIC stands for "China Economic Information Center."

Changes: We have added the full spelling of CEIC: "the concrete categories were estimated based on building types from China Economic Information Center Data (CEIC, 2024)."

Line 113: what fraction of cement is used for roads as opposed to buildings?

Response: Thank you for your comment. The utilization of concrete primarily includes construction and infrastructure (such as road and hydraulic engineer). Due to different exposure conditions, particularly variations in CO_2 concentration, the carbonation rates also differ in these projects. In the accounting model for cement CO_2 uptake of this study, both construction and infrastructure are considered as the service stage for concrete. Given data availability, we did not further refine the activity-level data for this stage, such as the fraction of cement for different types of buildings and infrastructure. However, concrete carbonation rate coefficient in the model is adjusted based on environmental factors, according to Eq. 9 in the manuscript:

$$k = \beta_{csec} \times \beta_{ad} \times \beta_{CO_2} \times \beta_{cc} \quad (9)$$

k represents carbonation rates, which is calibrated by exposure conditions (β_{csec}), cement additives (β_{ad}), CO_2 concentration (β_{CO_2}) and coating and cover (β_{cc})

The correction value for CO_2 concentrations in different environments are shown in the following table (see in Supplementary Table 2, which can be accessed on Zenodo at https://doi.org/10.5281/zenodo.14583866). In summary, although the proportion of cementitious materials used in different engineering facilities, such as roads, is not considered, their effect on the carbonation rate is taken into account in the Monte Carlo simulations.

Table: CO_2 concentration and correction value under different environment

Location	CO_2 concentration(ppm)	modified parameter*
Urban	625	1.2
Rual	300	1
Seaside	225	0.93
Industrial area	1200	1.41
Road	1200	1.41
Buried	3000	1

Line 114: "this factor", which factor? Lines 114-118 are not very clear to me.

Response: Thank you for your comment. The factor in here is "Building lifespan". The building lifespan determines the exposure time of concrete during its service phase. In the accounting model for cement CO_2 uptake, we divide the concrete carbon sink into three stages based on principle of life cycle assessment: service, demolition, and secondary-use (including both disposal in a landfill and recycling, with a total lifespan of 100 years in the model. Given the differences in building styles across countries, the varying building lifespans result in differences in concrete exposure duration within the accounting model. Therefore, we collected data on the building lifespan for 42 countries. For the collection of this parameter, we prioritized the use of statistical data and research data (e.g., China), followed by the use of engineering design data and model simulation data instead, such as for Vietnam and India.

Changes: We have changed the introduction in Lines 114-118 of manuscript: "Building lifespan determines the exposure time of concrete during the service stage, which is crucial for setting up the concrete lifecycle in the accounting model. Therefore, we collected data on the distribution of building lifespans for 42 countries. Building lifespan data were primarily referenced from statistical and survey data(Xi et al., 2016), for countries with limited statistical data, such as Vietnam and India, engineering design and model data were used (Bhyan et al., 2023， Ji et al., 2021)."

Line 135: it would be helpful to define carbonation ration and molar ratio

Response: Thank you for your comment. Carbonation ratio is the percentage of carbonation depth (d) compared to the theoretical maximum (D), it varies considerably with the exposure conditions. Molar ratio is the molar mass ratio of CO_2 to CaO (44/56≈0.786).

Changes: We have changed the explanation of calculation methods in Lines 134-139 of manuscript: "Where C is the carbon uptake by cement materials, W is the clinker consumption, which is adjusted by clinker production ($P_{clinker}$) with its exports (Ex) and imports (Im) (Eq. 6). F is annual carbonation ratio, which is the percentage of carbonation depth in accounting year (d) compared to the theoretical maximum carbonation depth (D) (Eq. 7). Based on Fick's diffusion law (Eq. 8, You et al., 2022), the carbonation depth of cement is the product of the carbonation rate (k) and the square root of time. The carbonation rate in the model is calculated by considering the impact of exposure conditions (β_{csec}), cement additives (β_{ad}), CO_2 concentration (β_{CO_2}) and coating and cover (β_{cc}) (Eq. 9). M is the molar mass ratio of CO_2 to CaO (0.786)."

Line 170: CKD could be defined in the table

Response: Thank you for your comment. Cement kiln dust (CKD) is a by-product of cement manufacturing. It is composed of micron-sized particles collected from electrostatic precipitators during the production of cement clinker.(Siddique, 2006). CKD will absorb CO_2 during landfill/waste treatment.

Changes: We have added the cement kiln dust (CKD) in the table.

Line 181: USGS defined and reference provided?

Response: Thank you for your comment. USGS is United States Geological Survey, they annually publishes data on cement production in different countries around the world, which can be accessed at https://www.usgs.gov/centers/national-minerals-information-center/cement-statistics-and-information.

Changes: We have added the reference of USGS, and its full spelling has been added in one time.

Line 186: IPCC reference?

Response: Thank you for your comment. Here we cited the 2006 IPCC Guidelines for National Greenhouse Gas Inventories, which can be accessed at https://www.ipcc-nggip.iges.or.jp/public/2006gl/index.html

Changes: We have added the reference Intergovernmental Panel on Climate Change (IPCC)

Line 236: "within the first year" Is this not true for all years?

Response: Thank you for your comment. Here, the author intends to explain that concrete materials cannot be completely carbonized within one year (Qiu, 2020), it leads to a time-lag effect in cement carbon uptake. The depth of carbonation reflects the diffusion range of CO_2 (He et al., 2025). Based on Fick's second law, the carbonation depth of concrete is the product of the carbonation rate factor and the square root of time (You et al., 2022). Studies have shown that the natural carbonation age of concrete ranges from 5 to 60 years (Pan et al., 2016) and that the increase in carbonation depth slows significantly over longer periods of carbonation curing (Qiu, 2020). Therefore, the results of accounting for carbon sequestration in concrete exhibit a time-lag effect, the carbon sequestration of concrete in the current year does not come exclusively from the cement consumed in that year.

Changes: Since the time-lag effect of carbon uptake by cement is in lines 255-259 in the original text, to avoid misunderstandings, we have deleted the original statement in line 236 and added that explanation to the time-lag effect section: "Specifically, the annual carbonation rate of cementitious materials shows a steady decline (Figure 2). Mortar and CKD, with their faster carbonation rates, are the primary cement materials contributing to current-year uptake. While concrete is the main material result in the time-lag effect of cement carbon sinks (Figure 2), it is because natural carbonation of concrete cannot be completed in one year (Pan et al., 2016) and the rate of carbonation gradually slows down (Qiu, 2020). 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."

Line 244: Again, is this not contrary to the reason that one makes cement in the first place?

Response: Thank you very much for your comment. We are sorry that our previous statement created a misconception that carbonization was being encouraged in order to achieve carbon neutrality. Carbonization of cement materials does significantly reduce the durability of buildings (Zhang et al., 2025). From a management perspective, we advocate enhanced carbonization of cement materials at the waste disposal and recycling stages, such as waste concrete (Mo and Panesar, 2013), and CKD (Pu et al., 2023).

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. It is necessary to strengthen the carbonation management of cement materials during the waste disposal and recycling stage. For example, many studies have explored the mechanisms and properties of accelerated carbonation in cement materials, such as waste concrete (Mo and Panesar, 2013), and CKD (Pu et al., 2023). 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."

Text beginning on line 246: use of the term "historical year" is not very clear. Does this refer to each historical year? Line 249: again, is this referring to each historical year?

Response: Thank you very much for your comment. As an example, for the total carbon uptake from cement in 2023, the current-year uptake is the carbon sequestration resulting from the carbonization of the cement consumed in 2023, and the historical-year uptake is the carbon sequestration in 2023 due to the incomplete carbonization of the cement consumed before 2023.

Changes: We changed the interpretation of historical-year uptake: "While the historical-year uptake refers to carbon uptake due to incomplete carbonization of cement materials consumed in the historical years and continues to carbonize in the current year, increasing to 0.42 Gt yr^{-1} in 2023."

Line 252: Does this really say that most C uptake takes place within current year production?

Response: Thank you very much for your comment. Your observations are correct. The carbonation rate of concrete materials during the building service phase decreases gradually, and although they cannot be fully carbonized in one year, most of the carbonation occurs in the first year of service. Therefore, carbon uptake in the current year is predominant. Additionally, the gradual increase in the share of carbon uptake in historical years is a result of accumulation.

Changes: we have added Figure 2 to improve the clarity and readability of this section:

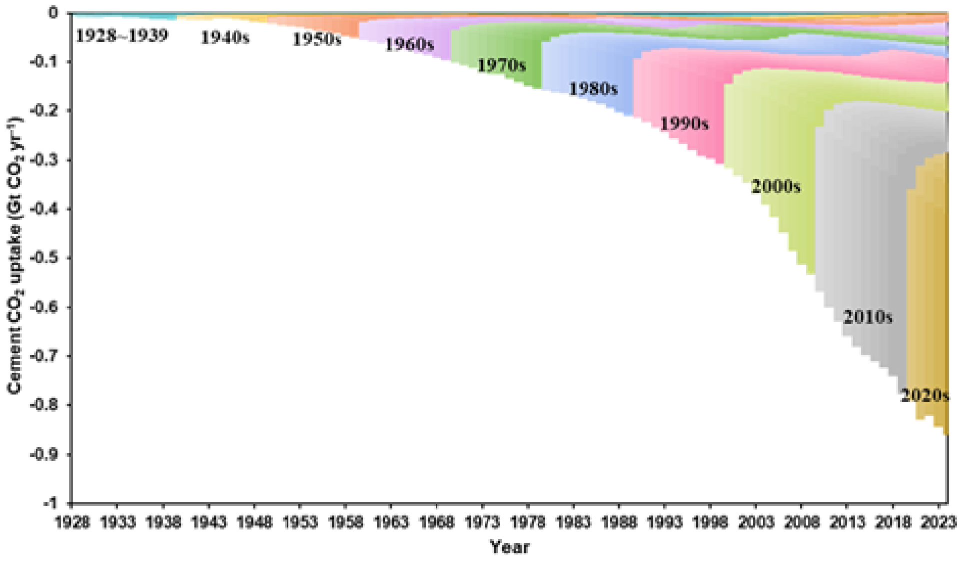

Figure 2: Time-lag effect on carbon uptake by cement from 1928 to 2024. Different colours represent changes in carbon sequestration over time for different years of consumption of cement.

Lines 305-306: Here it is important to distinguish clearly between emissions and net emissions.

Response: We are grateful for your review. The emissions for the cement industry include both fossil fuel emissions and chemical process emissions. Therefore, the emissions in this study refer to process emissions, and the net emissions reflect process emissions after deducting of cement carbon sequestration. We have added it at the application of line 47 of the original text.

Changes: We have added the definition of "net emission" in line 47: "A substantial fraction of process CO_2 emissions from cement production is reabsorbed on a time scale of 100 years through natural carbonation of cement materials, the net cement emissions (namely industrial process of cement production minus the CO_2 sequestration from carbonation of cement materials) were only about 57% of the cumulative process emissions from 1930 to 2013 (Xi et al., 2016)."

Many of these points may seem like nit picking of minor issues, but collectively they are very important to a clear understanding of what was done in this paper and how we should treat the results. I encourage complete and comprehensive discussion of data sources and computations.

Response: We are very appreciated of your meticulous review. Your comprehensive comments and suggestions have not only contributed to improve the quality of this manuscript but also broadened our perspective on this research topic.

Reference:

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