Final Author Comments

Manuscript ID: ESSD-2026-218

Title: Global and National CO_2 Emissions from the Lime Production Process and Carbonation Sink from 1930 to 2024

We sincerely thank the reviewers, the handling editor, and the scientific community for their valuable comments and constructive suggestions during the open discussion period. These inputs have greatly helped us improve the transparency of the dataset, the rigor of the methodology, and the robustness of the conclusions.

In response to all received referee comments (RCs) and community comments (CCs), we have systematically revised the manuscript. All substantive changes have been highlighted in red in the revised version. The key revisions and responses are summarized as follows:

Comment on essd-2026-218, Peiying Li

The study provides a comprehensive assessment of CO2emissions from lime production and carbonation sinks, establishing both global and national database covering the period 1930-2024. It offers valuable contributions to industrial carbon cycle research and data sharing. The study quantifies key national-level lime emissions at Tier 2 method, which improves the robustness of lime process carbon accounting. It further elevates lime carbonation to the level of global carbon balance and carbon budget discussions, supporting the integration of lime carbonation sink into broader carbon cycle. In addition, the inclusion of carbonation sink from blast furnace slag further strengthens the relevance of the study to industrial symbiosis and metallurgical carbon sink research. Overall, the dataset is comprehensive in temporal and spatial coverage, and the methodological framework is well developed, aligning well with the scope of ESSD as a high-quality data descriptor paper.

The following issues should be further clarified by the authors:

1. (L25-27) The statement "which is 8.32% increase compared with Bing et al. (2023)" could be clarified further. It is recommended to explicitly specify the compared time period and baseline value, rather than only citing the previous study, to improve interpretability for readers.

Author Response:

We sincerely thank the reviewer for this constructive suggestion. We agree that explicitly specifying the compared time period and baseline value improves the interpretability of this comparison. In response, we have revised the corresponding sentence in the Abstract to clearly state the 1930–2020 baseline value reported by Bing et al. (2023). We also corrected a minor terminology error in the parentheses, changing "cement carbonation sink" to "lime carbonation sink".

The revised text in the Abstract now reads:

"Cumulative carbonation uptake reached 4.8 Gt CO_2 (95% CI: 3.9–5.8 Gt CO_2), offsetting 42.2% of process emissions, 3.4% higher than the 38.8% estimate for 1930–2020 reported by Bing et al. (2023)."

Please see line31-33.

The corresponding modifications have been marked in red in the revised manuscript

2. (L618-625) The citation format "based on (Xi et al., 2016) and (Bing et al., 2023)" is not recommended. It should be revised to a standard academic form, e.g., "Xi et al. (2016)" and "Bing et al. (2023)" or a unified parenthetical citation.

Author Response: We appreciate the reviewer pointing out this formatting oversight. We have revised the in-text citation format to align with standard academic conventions. Specifically, the text in this section has been corrected

The revised text in the Abstract now reads:

" The uncertainty analysis in this study was designed to reflect data-source reliability, reconstruction intensity, and country-level heterogeneity. Building on the accounting model established by Xi et al. (2016) and the framework of Bing et al. (2023) "

Please see line886-888.

The corresponding modifications have been marked in red in the revised manuscript.

3. (L372-387) It is recommended to improve the referencing of supplementary datasets by clearly linking the updated data to the corresponding tables in this section.

Author Response: We appreciate this constructive recommendation. We fully agree that explicitly linking the updated data to the corresponding supplementary tables significantly improves the transparency and readability of the manuscript. In response, we have revised this section to include precise cross-references to the corresponding datasets (e.g., SI-2 Data 1, SI-

3 Data 1–15, SI-2 Data 3, and SI-3 Data 9) immediately following the description of each of the four key refinements. Additionally, we have corrected a few minor grammatical typos in this paragraph to improve the overall flow. The corresponding modifications have been marked in red in the revised manuscript.

The revised text in the Abstract now reads:

" This allowed us to establish, for the first time, a lime carbonation sink dataset covering 11 countries, which collectively accounted for an average of 78.9% of global lime production (see Supplementary Table SI-2, Data 1). (2) More detailed, country-specific parameterization. The parameters were updated for lime CO2 uptake in the newly added countries. The parameters include the proportion of lime usage across various sectors, the CaO content of different lime materials, and the production and utilization rates of lime materials. These improve the accuracy of the accounting compared to the CO2 uptake parameters used for the ROW region in our previous work (see Supplementary Table SI-3, Data 1–15). (3) Inclusion of BFS in the accounting system. BFS was incorporated to address the previous omission of its carbonation contribution from metallurgical slags (see Supplementary Table SI-2, Data 3). (4) Data-constrained temporal and regional parameterization of SS and BFS. Complete annual and country-specific data were unavailable for the SS and BFS generation coefficients and for the fractions directed to road-base applications or open-air stockpiles. We therefore used four non-overlapping accounting periods: 1930–1949, 1950–1972, 1973–1999, and 2000–2024. These periods represent broad stages in iron- and steelmaking technology and serve only as common accounting windows; they do not imply synchronous technological transitions across countries. Crude steel and pig-iron production were obtained from statistical data and were not determined by these assumptions. Country-specific slag generation coefficients and pathway-allocation fractions were used where available. Otherwise, separate values were assigned to developed and developing regions based on differences in industrialization, technology-adoption timing, and slag-management practices. The parameter values, assumptions, and supporting references are provided in Supplementary Table SI-3, Data 8–9."

Please see line584-604.

The corresponding modifications have been marked in red in the revised manuscript.

4. (L323-329) The manuscript includes lime mortar carbonation, which is also

commonly considered in cement carbonation sink studies. The authors are advised to further clarify the system boundary definition between lime mortar and cement mortar to avoid potential double counting across different mineral carbonation datasets, thereby ensuring more consistent integration into global carbon budget.

Author Response: We highly appreciate the reviewer's rigorous attention to system boundaries and the integration into the Global Carbon Budget. We confirm that there is absolutely no double counting, and we have updated the text to make this distinction explicitly clear.

To ensure precise accounting, we treat lime mortar and cement mortar as completely separate material flows based on the origin of their active CaO. Existing cement carbonation datasets strictly account for the carbonation of hydration products derived from cement clinker. In contrast, the lime mortar (MOR) quantified in our study originates exclusively from commercial lime production. We have revised the methodology section to explicitly define this system boundary, clarifying that our M_{mor} calculation relies solely on the proportion of commercial lime allocated to the construction sector, strictly excluding any CaO derived from cement clinker. The corresponding modifications have been marked in red in the revised manuscript.

The revised text in the Abstract now reads:

" To avoid double counting with cement-carbonation datasets, MOR includes only commercial lime used in construction and excludes CaO derived from cement-clinker hydration."

Please see line327-329.

The corresponding modifications have been marked in red in the revised manuscript.

5. A major highlight of this paper is the inclusion of blast furnace slag (BFS) into the lime carbonation sink accounting for the metallurgical industry for the first time. However, on a global scale, a significant amount of blast furnace slag is utilized in cement production. It is unclear whether the carbon uptake accounting for BFS and SS in this study overlaps with existing cement accounting. If there is no double-counting, please explicitly clarify this in the manuscript to avoid readers' confusion.

Author Response: We highly appreciate the reviewer's rigorous attention to system

boundaries. We confirm that there is absolutely no double-counting between our accounting of metallurgical slags and existing cement carbonation datasets, and we have explicitly addressed this from both a macro-boundary and a micro-parametric perspective in the revised manuscript. Macro System Boundary (Line 265-268): While Blast Furnace Slag (BFS) and Steel Slag (SS) are widely utilized as Supplementary Cementitious Materials (SCMs) in cement production, current global cement carbonation accounting strictly sets its boundary around the calcination and carbonation of cement clinker. To ensure maximum clarity, we have added an explicit boundary statement in Section 2.3 (Line 265-268), clarifying that our system boundary for metallurgical slags is strictly restricted to the carbonation of CaO originating exclusively from lime fluxes. Micro Parametric Isolation (Line 333–343): Mechanistically, as described in our BFS calculation formula, we introduce a specific localization parameter, l_{bfs}, which explicitly represents the proportion of CaO in the slag derived strictly from lime flux rather than other raw materials. By utilizing l_{bfs} to mathematically filter and isolate the lime-derived CaO fraction, our model eliminates the risk of boundary overlap with clinker-based cement carbonation accounting.

The revised text in the Abstract now reads: "To avoid double counting with existing cement-carbonation datasets, the system boundary for metallurgical slags (SS and BFS) is defined by the origin of CaO rather than by the subsequent use of the slag. Only CaO derived from lime fluxes and represented in the lime production activity data is attributed to the lime carbonation sink."

"By isolating the lime-derived CaO fraction, lbfs prevents boundary overlap with clinker-based cement-carbonation accounting, including when BFS is subsequently used as a supplementary cementitious material."

Please see line207-210、351-353

The corresponding modifications have been marked in red in the revised manuscript.

6. The lime production in Italy and Germany between 1930 and 1958 was estimated using a multiple linear regression model. It is recommended to briefly specify in the main text which key independent variables were used in this regression model. This will help readers quickly understand the rationality behind the early historical data reconstruction.

Author Response: We sincerely thank the reviewer for this valuable suggestion. We agree

that clearly describing the basis of the historical reconstruction in the main text is essential for improving the transparency and interpretability of the dataset.

Following a further review of the available historical sources, we revised the reconstruction methods for Germany and Italy. Therefore, multiple linear regression is no longer used to estimate lime production for these two countries. For Germany, historical lime production data were compiled directly from national statistical yearbooks published by the Statistisches Bundesamt. For Italy, the national statistical yearbooks published by the Istituto Centrale di Statistica reported the quantity of limestone used for lime manufacture. Based on years for which both limestone input and lime production were available, we derived an average limestone-to-lime conversion coefficient of 0.24. This coefficient was then applied to estimate lime production for years in which only the limestone input was reported.

Accordingly, Section 2.1.1 has been revised to describe these country-specific data sources and reconstruction methods explicitly. The revised text also summarizes the reconstruction approaches used for the other major lime-producing countries, while detailed activity data, source references, reconstruction parameters, and uncertainty assumptions are provided in Supplementary Table SI-2 Data1. The corresponding changes have been marked in red in the revised manuscript.

The revised text in the Abstract now reads:"Before continuous USGS coverage, country-specific historical sources and transparent proxy methods were used. Japanese limestone shipments to lime manufacture reported by Shimanishi (2004) were converted to lime output using 1.8 t limestone per tonne of lime and checked against overlapping industry statistics. Brazilian lime production for 1940–1964 was taken from the IBGE Physical Production Tables (Produção de cal; IBGE, 2026), while 1930–1939 values are explicitly identified as autoregressive estimates. French, Canadian, German, and former Soviet records were compiled from national statistical yearbooks, with Canadian publications cited by their historical publication years (Dominion Bureau of Statistics, various years). Australian lime production was converted from limestone allocated to lime manufacture using a limestone-to-lime conversion coefficient of 0.24 (ABS, 2026). For Italy, a mean limestone-to-lime coefficient of 0.24 was derived from years reporting both inputs and outputs. For the United Kingdom, fixed 1949-based allocation ratios of 0.161 for limestone and dolomite and 0.049 for chalk, together

with a lime-yield coefficient of 0.225, were used for years lacking direct observations (Ordnance Survey, 1957). Short gaps in Russian records were linearly interpolated; for 1959–1991, Russian output was estimated as 0.54 of reported Soviet lime production (Upravlenie S. U. T. statisticheskoe, 1961). All source classifications, equations, diagnostics, and uncertainty assumptions are documented in Supplementary Table SI-2, Data 1."

Please see line417-435

The corresponding modifications have been marked in red in the revised manuscript.

Comment on essd-2026-218, Xiaoqian Song

This study constructs a long-term (1930–2024) and high-resolution dataset of global lime production CO_2 emissions and carbonation sinks, covering 11 major lime-producing countries and including blast furnace slag (BFS) for the first time. The research topic is important, the method is rigorous, the data quality is high, and the results are reliable. It effectively fills the gap of lime carbonation sink in the global carbon budget and has important scientific significance and application value. However, minor revisions are needed regarding data description, parameter explanation, figure annotation, and partial expression. After minor revisions, the manuscript meets the publication standards of Earth System Science Data.

1.The manuscript involves many abbreviations and symbols in the equations; it is better to provide a list of abbreviations to let the readers easy to look up.

Author Response: Thank you for this helpful suggestion. We have added a new table entitled "Abbreviations and symbols" after the Abstract. The table lists the main abbreviations and equation symbols used in the manuscript, including BFS, CEF, CS, GCB, IPPU, LKD, LM, LSS, MOR, PCC, RM, ROW, SS, SUG.

The revised text in the Abstract now reads:

Abbreviations and symbols

Abbreviation / symbol	Definition
BFS	Blast furnace slag
CEF	Carbon emission factor
CI	Confidence interval
CS	Carbide slag
GCB	Global Carbon Budget
IPCC	Intergovernmental Panel on Climate Change
IPPU	Industrial Processes and Product Use
LKD	Lime kiln dust
LM	Lime mud
LSS	Lime-stabilized soil
MFA	Material Flow Analysis
MOR	Lime mortar
PCC	Precipitated calcium carbonate
RM	Red mud
ROW	Rest of the world
SS	Steel slag
SUG	Sugar-industry carbonation material
USGS	United States Geological Survey

Please see line 44-45

The corresponding modifications have been marked in red in the revised manuscript.

2.Line 262-264: "Beyond the production stage, nine industrial byproducts and materials are incorporated across three sectors: metallurgy (SS, BFS, RM), chemicals (PCC, CS, SUG, LM), and construction (LSS, MOR)". Why focusing on these nine industrial byproducts and materials? Do you have a reference to support the products selection?

Author Response: We thank the reviewer for this helpful comment. We have revised Sect. 2.3 to clarify the basis for selecting these materials. The material boundary of this study was developed based on the lime carbon sink framework proposed by Liu et al. (2018), who

reviewed lime carbon sinks from the perspective of material flow analysis and identified the main lime-related carbonation pathways in the chemical, metallurgical, construction, and lime kiln dust treatment sectors. Building on this framework, our study retained the major lime-based material flows and newly incorporated blast furnace slag (BFS) to address the previously omitted metallurgical carbonation sink. We have added Liu et al. (2018) as a supporting reference and clarified this rationale in the revised manuscript.

The revised text in the Abstract now reads:

The system boundary was established using material flow analysis (MFA; Fig. 1), based on the lime-carbonation framework of Liu et al. (2018). It includes ten lime-based materials classified by lifecycle stage. Lime kiln dust (LKD) is treated as a direct by-product of lime production. The remaining nine materials are grouped into three downstream sectors: metallurgy, including steel slag (SS), blast furnace slag (BFS), and red mud (RM); chemicals, including precipitated calcium carbonate (PCC), carbide slag (CS), carbonation sugar (SUG), and paper mill lime mud (LM); and construction, including lime-stabilized soil (LSS) and lime mortar (MOR). Most material flows follow Liu et al. (2018), while BFS is newly included to improve the accounting boundary.

Please see line 198-206

The corresponding modifications have been marked in red in the revised manuscript.

3.The format of equations should to be consistent. For example, Line 255 is (3), while others are labeled as "Eq (4)". Also, there are many equations appears in the text, and not numbers. For example, an equation in Line 300 and all equations in section 2.3.2. Please double check the formatting requirement of ESSD and revise them accordingly.

Author Response: We thank the reviewer for this helpful comment. We have carefully checked the equation formatting throughout the manuscript and revised it according to the ESSD formatting requirements. All displayed equations are now numbered consecutively using the format (n), and in-text references to equations are consistently written as Eq. (n). In addition, the equations that were previously embedded in the text, especially those in Sect. 2.2.1, have been converted into displayed and numbered equations to improve clarity and consistency.

Please see line 220-397

4. There are several typo errors, such as "CO2"should be "CO2".

Author Response: We thank the reviewer for pointing this out. We have carefully checked the manuscript and corrected the typographical and formatting errors throughout the text. In particular, CO2 has been revised to CO_2, and other chemical formulas and minor formatting issues have also been standardized as required.

5. Section 3.4: Strengthen the mechanism explanation of the time lag effect in the discussion to clarify why historical carbon sequestration keeps rising.

Author Response: We thank the reviewer for this helpful suggestion. We have revised Sect. 3.4 to strengthen the mechanistic explanation of the time-lag effect. Specifically, we clarified that annual CO_2 uptake is contributed by both newly produced lime-based materials and legacy materials produced in earlier years. Fast-carbonating materials mainly contribute to current-year uptake, whereas MOR, LM, RM, SS, and BFS follow multi-year carbonation processes. We further explained that the formation of a $CaCO_3$ product layer slows inward CO_2 diffusion, causing carbonation to shift from rapid initial uptake to a slower diffusion-controlled stage. Therefore, each production year adds a new cohort of slowly carbonating materials, while earlier cohorts continue to absorb CO_2 until their reactive Ca-bearing phases are progressively depleted. This explains why historical carbon sequestration keeps rising.

The revised text in the Abstract now reads:

Mechanistically, the time-lag effect reflects the accumulation of legacy material stocks and the progressive carbonation of reactive Ca-bearing phases. Within the accounting framework, annual uptake comprises uptake attributed to materials produced in the current year and continued uptake by incompletely carbonated materials inherited from previous years. For precipitated calcium carbonate (PCC) and carbonation sugar (SUG), carbonation is integral to the production process, and uptake is therefore assigned to the production year. Lime kiln dust (LKD), lime mud (LM), red mud (RM), and carbide slag (CS) are represented using literature-derived, one-year, pile-average CaO conversion parameters; this treatment does not assume complete conversion of CaO. By contrast, lime-stabilized soil (LSS) and lime mortar (MOR) follow the slab model, whereas steel slag (SS) and blast-furnace slag (BFS) follow the spherical-particle model and continue to carbonate over multiple years (Sect. 2.3.1–2.3.2).

As carbonation proceeds, the formation of a $CaCO_3$-rich product layer can impede inward CO_2 transport, shifting the reaction from relatively rapid initial uptake toward a slower,

diffusion-controlled stage (Xi et al., 2016; Niu et al., 2025). For metallurgical slags, carbonation is further controlled by particle size, reactive Ca-bearing mineral phases, humidity, temperature, and stockpiling or reuse conditions (Zhao et al., 2025). Each year introduces a new cohort of lime-based materials, while incompletely carbonated cohorts from previous years continue to absorb CO_2. Over most of the study period, additions to this historical reactive stock exceeded its depletion, explaining the sustained increase in historical uptake. This mechanism also explains why carbonation sinks can decline more slowly than process emissions in countries that have already reached their emission peaks.

Please see line 788-808

The corresponding modifications have been marked in red in the revised manuscript.

6.It is recommended to add "future work" to illustrate how to make the research better. For example, future scenario projections (2025–2050) will to studied to support global and national carbon neutrality goals.

Author Response: We thank the reviewer for this valuable suggestion. We have added a future-work paragraph to the Conclusion. The revised text states that future work should extend the historical dataset to 2025–2050 scenario projections and consider changes in lime demand, regional industrial structure, kiln technology, material recycling, slag utilization, CCUS deployment, and enhanced-carbonation pathways. This addition clarifies how the dataset can be further developed to support global and national carbon-neutrality assessments.

The revised text in the Abstract now reads:

Future work should extend this historical dataset to scenario projections for 2025–2050 to better support global and national carbon-neutrality assessments. Such projections should consider changes in lime demand, regional industrial structure, kiln technology, material recycling rates, slag utilization, CCUS deployment, and enhanced-carbonation pathways. In particular, linking the lime carbonation sink dataset with national mitigation scenarios would help quantify the potential contribution of lime-based carbon uptake to net-zero roadmaps and identify priority countries, sectors, and material streams for future emission reduction and carbon sink enhancement (Simoni et al., 2022; EuLA, 2026a; NLA, 2026).

Please see line 957-964

The corresponding modifications have been marked in red in the revised manuscript

Comment on essd-2026-218, Robbie Andrew

Review of "Global and National CO2 Emission from Lime Production Process and Carbonation sink from 1930 to 2024"

This article builds on Bing et al., 2023, adding further regional disaggregation and a longer period of analysis. It's good to see this work, but there are a lot of problems that need to be dealt with.

Main points

- The method is highly unclear and many assumptions are stated without any effort to justify them.
- The methods used by the same authors to estimate historical lime production data are highly suspect, but the result is labelled as "verified data".
- Some data sources have been misused.
- The apparent design of the uncertainty analysis results in an artificially low uncertainty of the overall carbonation rates.
- Since a model is the starting point of the work, it would make more sense to describe the model and then describe how the model's parameters were populated, the opposite of the current order. There is much confusion with variables being collected without any explanation of what they mean or what they are for.

Detailed comments

L38: Developing countries "face substantial pressure to reduce total emissions": What pressure? Internal or external? Many will disagree that developing countries "face substantial pressure". Either replace this with a more neutral statement ("must reduce total emissions") or support it with a citation that there is actual pressure from elsewhere.

Author response: We thank the reviewer for identifying this ambiguity. We agree that the phrase "face substantial pressure" could be interpreted as referring to external political or institutional pressure, which was not our intended meaning. We have therefore removed this wording and replaced it with a more neutral, technically supported explanation of the mitigation challenge associated with lime production. The revised text clarifies that calcination-derived process emissions cannot be eliminated solely through fuel switching or improvements in energy efficiency, rather than making an unsupported claim regarding external pressure.

The revised manuscript now reads:

"Lime production is carbon-intensive because CO_2 is released by carbonate calcination, with additional emissions from kiln fuels and electricity. Calcination generally accounts for about two-thirds of total lime-production-related CO_2 emissions, while energy use contributes much of the remainder (Han et al., 2022; Laveglia et al., 2024a). Consistent with IPCC inventory boundaries, this study quantifies process emissions from lime production and the subsequent carbonation uptake of lime-derived materials; energy-related emissions are excluded to avoid overlap with the energy sector. Because process emissions are governed by carbonate decomposition, they cannot be eliminated solely through fuel switching or energy-efficiency improvements, making lime a hard-to-abate industrial sector (Davis et al., 2018)."

Please see Lines 56–64 of the revised manuscript.

L54-55: The statement needs to make it clear that it is limited to the lime industry. Currently "global CO2 emissions" implies strongly that the context has changed.

Author response: We thank the reviewer for identifying this ambiguity. We agree that the original expression "global CO_2 emissions" could incorrectly imply a shift from the lime industry to total global emissions. We have therefore revised the text to specify explicitly that the stated proportion refers only to CO_2 emissions associated with lime production. We have

also clarified the distinction between process emissions and energy-related emissions and defined the accounting boundary of this study.

The revised manuscript now reads:

"Lime production is carbon intensive because CO_2 is released by carbonate calcination, with additional emissions from kiln fuels and electricity. Calcination generally accounts for about two-thirds of total lime-production-related CO_2 emissions, while energy use contributes much of the remainder (Han et al., 2022; Laveglia et al., 2024a). Following IPCC inventory boundaries, this study quantifies process emissions from lime production and the subsequent carbonation uptake of lime-derived materials; energy-related emissions are excluded to avoid overlap with the energy sector. Because process emissions are governed by carbonate decomposition, they cannot be eliminated solely through fuel switching or energy-efficiency improvements, making lime a hard-to-abate industrial sector (Davis et al., 2018)."

Please see Lines 56–64 of the revised manuscript.

L70: Naming one individual like this as a source of singular authority isn't appropriate. Just say "As has been noted". The citation you provide at the end of the sentence is sufficient to indicate who has noted this.

Author response: We thank the reviewer for this helpful comment. We agree that naming an individual here could inappropriately present the author as a singular authority. Following the reviewer's suggestion, we have removed the author's name and revised the sentence to: "As has been noted, accurate estimation of emission factors depends heavily on the precise characterization of raw-material composition (Andrew, 2019)." The citation has been retained at the end of the sentence to identify the source of this observation.

Please see Lines 97-98 of the revised manuscript

L96: "decarbonation" should be "decarbonization"

Author response: We thank the reviewer for pointing this out. We have corrected "decarbonation" to "decarbonization" in Line 123.

L127-129: The authors talk several times about the "utilization rate" of slags, but without explaining why this is important. Why is only utilized slag assumed to result in carbonation? What happens to anything that is not "utilized"? According to the equations, zero carbonation, but why? Why should slag that is not "utilized" not undergo

carbonation?

Author response: We thank the reviewer for highlighting this ambiguity. We agree that our previous use of the term "utilization rate" did not adequately explain its role in the carbonation accounting and could incorrectly suggest that only utilized slag carbonates, whereas unutilized slag is assumed to undergo zero carbonation.

We have therefore clarified the definitions and accounting treatment of these parameters. Specifically,U_{ss} and U_{bfs}) do not represent the overall utilization rates of steel slag (SS) and blast-furnace slag (BFS). Instead, they are pathway-allocation parameters representing the combined fractions of slag used in road-base applications or stored in open-air stockpiles. Both pathways permit reaction with atmospheric or pore-water CO_2 and are therefore included in the natural-carbonation accounting. Consequently, unutilized slag stored in open-air stockpiles is included rather than assigned zero carbonation.

Slag incorporated into other valorized products enters downstream product systems, where carbonation depends on product-specific service conditions, exposure environments, particle sizes, and transport limitations. Because sufficiently consistent data are unavailable to quantify these pathways and their inclusion could create boundary overlap with downstream product accounting, they are excluded from the present system boundary. This exclusion does not imply that their actual carbonation is zero; it means only that their carbonation is not quantified within the accounting boundary of this study.

We have revised the manuscript to explain these distinctions explicitly and to clarify that the included SS and BFS fractions are carbonated using the spherical-particle model. The revised text also describes the mass-balance approach used to isolate the lime-derived CaO fraction in BFS and directs readers to the country- and period-specific parameters in Supplementary Table SI-3, Data 15.

The revised manuscript now reads:

"The BFS mass included in natural-carbonation accounting is:

$$M_{bfs} = m_{bfs} \times r_{bfs} \times l_{bfs} \times U_{bfs} \quad (22)$$

where m_{bfs} represents blast-furnace iron production; r_{bfs} is the BFS generation rate, with time-dependent values provided in Supplementary Table SI-3, Data 1; l_{bfs} is the proportion of CaO in BFS derived specifically from lime input; and U_{bfs} is the combined fraction of total BFS directed to road-base applications or stored in open-air stockpiles and therefore included in the natural-carbonation accounting. By isolating the lime-derived CaO fraction, l_{bfs} prevents

boundary overlap with clinker-based cement-carbonation accounting, including when BFS is subsequently used as a supplementary cementitious material. U_{bfs} is not the ratio of open-air stockpiling to road-base use and does not affect the estimated total production of BFS. It is a pathway-allocation parameter used only to identify the fraction of total BFS included within the natural-carbonation boundary. Carbonation of this fraction is calculated using the spherical-particle model described in Eq. (11).

It should be noted that U_{ss} and U_{bfs} represent the fractions of SS and BFS, respectively, used in road-base applications or stored in open-air stockpiles and therefore included in the natural-carbonation accounting. These parameters differ from the overall utilization rates described earlier, which represent the proportions of slag recycled through all resource-utilization pathways. SS and BFS used in road-base applications or stored in open-air stockpiles can react with atmospheric or pore-water CO_2. However, the actual extent of carbonation depends strongly on slag mineralogy, particle size, gas and moisture transport conditions, and other factors (Pullin et al., 2019; Chukwuma et al., 2021; Elyasi Elyasi Gomari et al., 2024). Slag incorporated into other valorized products is considered to enter downstream product systems. Because its subsequent carbonation depends on product-specific service and exposure conditions, it is excluded from the present accounting (Li et al., 2022). Therefore, exclusion of this fraction from the equations indicates only that its carbonation lies outside the accounting boundary of this study, rather than that its actual carbonation is zero.

The lime-derived CaO fraction in BFS was determined using an ironmaking-burden mass-balance approach. First, quicklime consumption per tonne of pig iron was estimated from the total blast-furnace burden consumption per tonne of pig iron, the shares of sinter, pellets, and lump ore in the burden, and quicklime consumption per tonne of sinter. The resulting quicklime input was converted to lime-derived CaO and divided by the total CaO contained in the generated BFS, which was calculated from the BFS generation rate and BFS CaO content. Country- and period-specific parameters used in this calculation are provided in Supplementary Table SI-3, Data 15."

Please see Lines 345–378 of the revised manuscript.

L133: The authors make it sound like the main goal is being included in the GCB dataset, rather than producing best-possible estimates of global lime carbonation. The points set out in this paragraph are not requirements (stated or otherwise) of being included in the GCB dataset. Rather, they are about making the dataset more comprehensive. That ought to be the goal of the study. Please reword.

Author response: We thank the reviewer for this important clarification. We agree that the primary objective of this study should be to improve the accuracy, transparency, and comprehensiveness of global lime-carbon accounting, rather than to satisfy presumed

requirements for inclusion in the Global Carbon Budget (GCB). Potential integration into the GCB is an application of the resulting dataset, not the criterion used to design the study.

We have therefore rewritten this paragraph to remove the implication that the methodological improvements represent GCB requirements. The revised text now presents them as four advances intended to improve global estimates of lime process emissions and carbonation uptake:

"To improve the accuracy and transparency of global lime-carbon accounting, this study makes four principal advances. (1) Source-prioritized activity-data reconstruction: USGS statistics, national yearbooks, and industry records were harmonized for 11 major producing countries, representing 78.9% of global output, with proxy-derived and interpolated values explicitly identified (Supplementary Table SI-2, Data 1). (2) Composition-sensitive process-emission factors: following Liu et al. (2015), sector-specific CaO requirements and national lime-use structures were combined to derive country-level weighted emission factors (Supplementary Table SI-3, Data 1). (3) A consistent material-flow boundary: lime-derived CaO was traced across production, construction, metallurgical, and chemical pathways while excluding non-lime CaO inputs. (4) Data-constrained temporal and regional parameterization: SS and BFS generation and pathway-allocation parameters were assigned for 1930–1949, 1950–1972, 1973–1999, and 2000–2024, using country-specific data where available and documented regional assumptions otherwise. These intervals are common accounting windows and do not imply synchronized technological transitions among countries."

Please see Lines 159–172 of the revised manuscript..

L140-141: "to synchronize with the Global Carbon Budget timeframe": What does this mean? Which timeframe? The GCB is updated every year, and the only fixed time is the start year of 1750, which doesn't match anything in this study.

Author response: We thank the reviewer for pointing this out. We agree that the phrase "to synchronize with the Global Carbon Budget timeframe" was unclear. Our intention was simply to indicate that the dataset was updated through 2024 using the latest available data, rather than to suggest that the Global Carbon Budget has a fixed timeframe corresponding to that of our study. To avoid this ambiguity, we have removed the reference to synchronization with the Global Carbon Budget and clarified that the dataset was updated through 2024 to

provide a more current and comprehensive time series of global lime process emissions and carbonation sinks.

L150-151: "Dynamic Parameterization of Technological Evolution": It's unclear what this means. What is described here suggests that technology changes happened in all places at the same time, which seems very unlikely when we are discussing developed and developing countries through the course of the 20th century.

Author response: We thank the reviewer for pointing out this ambiguity. We agree that the phrase "Dynamic Parameterization of Technological Evolution" could incorrectly imply that technological changes occurred simultaneously across developed and developing countries. This was not our intention.

We have removed this phrase and replaced it with the more precise term "data-constrained temporal and regional parameterization." The revised text explains that the periods 1930–1949, 1950–1972, 1973–1999, and 2000–2024 are common accounting windows used to organize the available historical data. They do not represent synchronized technological transitions across countries. Within these windows, country-specific parameters were applied where data were available; otherwise, explicitly documented regional assumptions were used.

The revised text now reads:

"(4) Data-constrained temporal and regional parameterization: SS and BFS generation and pathway-allocation parameters were assigned for 1930–1949, 1950–1972, 1973–1999, and 2000–2024, using country-specific data where available and documented regional assumptions otherwise. These intervals are common accounting windows and do not imply synchronized technological transitions among countries."

Please see Lines 168–172 of the revised manuscript.

L174-175: "the verified dataset": Do the authors here simply mean "peer-reviewed"? When was the dataset of Bing et al 2023 "verified"? This statement misleads the reader into believing that the start point is already a gold standard and no more discussion is necessary. I am highly sceptical of the lime production "data" used by this study and the previous one before 1990. Please explain what this enormous amount of lime was used for during decades when most uses of lime were at very low scale and indeed please also explain how it was produced. The only plausible candidate I can conceive of is

construction, since most other uses were probably extremely small (steel, sugar, etc.). If it mostly went to construction, how many new buildings each year does that imply, and is that realistic? Further, have the authors considered how much energy would have been required to produce that much lime in those decades? Where did that energy come from, and how does it compare to estimates of China's total energy consumption at the time? These sorts of sense checks are critical when such methods have been used. Bing et al 2023 used ARIMA with no control variables to extrapolate the entire period 1930-1948. This sort of model has zero real-world constraints, and simply continues a trend as if the world were the same in the past, with the same construction needs, infrastructural capacity and energy availability, and assuming that the period 1949-1962 (already suspect) are robust. Adding wide uncertainty bounds to these estimates is insufficient. The estimates must be revisited, and the authors cannot just cite one of their own peer-reviewed articles as evidence of a "verified" dataset.

Author response: Response:

We thank the reviewer for this important and constructive comment. We agree that our previous wording was inaccurate. By "verified dataset", we intended to refer to a dataset that had been published after peer review. However, we recognize that this wording could lead readers to believe that the dataset was already a gold-standard historical record requiring no further scrutiny. We have therefore replaced "verified dataset" with "a source-prioritized and internally cross-checked activity dataset" in the revised manuscript.

We also agree with the reviewer that the pre-1990 lime-production estimates require more careful treatment. In the revised manuscript, we no longer rely on the ARIMA fitting results used in Bing et al. (2023) for 1930–1995. We recognize that an unconstrained time-series extrapolation is not appropriate for historical periods with substantially different construction demand, industrial capacity, fuel availability, and statistical coverage. We have therefore reconstructed China's 1930–1995 lime-production series using historical source materials and sectoral activity constraints.

Specifically, as described in Section 2.3.1 of the revised manuscript,

"To illustrate the source harmonization procedure, China's 1930–1995 lime-production series was reconstructed by linking three consecutive source segments: a limestone-proxy

reconstruction for 1930–1949, a sectoral activity-based reconstruction for 1950–1985, and directly reported or association-based statistics for 1986–1995.

For 1930–1949, continuous national lime-production statistics were unavailable for China, particularly for 1930–1948. We therefore reconstructed annual lime production using limestone production as the primary proxy. Limestone production for mainland China and Northeast China was obtained from Guan (2007), which reports annual limestone production for 1912–1949. Observed lime-production records for Northeast China from Manshu Kojo Tokei Sokuho, Showa 15 (Kotoku 7) were used to calibrate the conversion relationship between limestone and lime production, yielding a limestone-to-lime conversion coefficient of 0.21. The calibrated relationship was then applied separately to mainland China and Northeast China, and the two regional estimates were combined to derive national lime production for 1930–1949.

For 1950–1985, we adopted a sectoral reconstruction approach constrained by activity data and historical evidence. Based on the sectoral lime-use shares reported by Liu (2018), total lime demand was allocated to the construction, iron and steel, calcium carbide, and alumina sectors. Steel, calcium carbide, and alumina production were used as activity indicators for their respective sectors, and through-origin proportional regression models were developed to estimate missing sectoral lime consumption. Construction lime demand was reconstructed using completed building floor area from Cao et al. (2019) as the activity proxy and constrained by historical information on lime production and application structure reported by Liao (1995), based on statistics from the China Lime Association.

For 1986–1995, lime production data were obtained from Liao (1995), based on China Lime Association statistics, and the China Building Materials Statistical Yearbook. Before merging the three source segments into a continuous historical series, statistical boundaries were harmonized through overlapping-year comparisons, consistency checks against sectoral activity indicators, and calibration of conversion coefficients where necessary to ensure temporal continuity and comparability. The harmonized series was subsequently linked with later statistical records to produce a continuous national dataset. All fitted parameters, reconstructed results, model diagnostics, and uncertainty distributions are documented in Supplementary Table SI-2 Data1.

This example illustrates how historical datasets with different statistical boundaries,

reporting practices, and data availability were harmonized into a temporally consistent activity dataset before subsequent emission estimation and uncertainty analysis. To reflect the additional uncertainty introduced by source integration, source-specific uncertainty ranges were assigned to reported statistics, regression-derived estimates, conversion-derived values, and interpolated data, and propagated through the Monte Carlo analysis (Section 2.4)"

Please see Lines 436-472 of the revised manuscript.

In addition, Liao and Yin (1995) provide two important historical constraints on the plausibility of the revised series: first, annual lime-production records for 1986–1992; and second, the historical statement that China's annual lime production increased from approximately 20–30 million tonnes in the late 1970s to 130 million tonnes in 1995. These historical ranges were used both to constrain the reconstruction and to assess whether the reconstructed results were consistent with the documented development of China's lime industry. Therefore, the revised estimates are based on historical sectoral activity and reported production ranges, rather than being extrapolated using an ARIMA model.

We also carefully considered the reviewer's suggestion to validate the estimates by comparing the energy required for lime production with China's total energy consumption in the corresponding years. However, after consulting the China Coal Industry Statistical Data Compilation and the China Coal Industry Yearbook, we did not find continuous historical records of coal consumption specifically allocated to lime production. Therefore, we cannot accurately calculate year-by-year historical energy consumption for lime production. Liao and Yin (1995) report that the average energy consumption of China's lime production around 1995 was approximately 182 kg ce t^{-1} lime. However, comparable annual fuel-consumption data are unavailable for earlier decades. We therefore did not use a highly uncertain reconstructed energy series as a formal validation constraint.

These revisions substantially change the treatment of the early-period activity data. The revised manuscript no longer refers to the earlier dataset as "verified", no longer relies on ARIMA extrapolation for 1930–1948, and instead provides a source-prioritized reconstruction that explicitly incorporates historical constraints and uncertainty treatment. The fitted parameters, reconstructed results, model diagnostics, and uncertainty distributions are documented in Supplementary Table SI-2 Data1.

L177: Shimanish 2004 appears to only present data on sales of limestone to the lime sector, which the authors have transcribed directly (SI-2.xlsx, sheet Data1) as production of lime in Japan. This means that the numbers used by the authors are about double what they should be. I don't have time to check all sources, but this points to a fundamental misunderstanding of the input data being used. Are the data for Brazil, France, etc. also limestone, or are they really lime? I advise the authors to check their data sources again.

Author response: We have now recalculated Japanese lime production for 1930–1958. Shimanishi (2004) indicates that approximately 1.8 t of limestone was required to produce 1 t of lime in Japan during this period. We therefore divided the limestone shipments to the lime sector by 1.8 to estimate lime production. The converted estimates were cross-checked against independent Japanese lime-industry statistics reported by Tokoro (1966) for overlapping years, yielding a mean relative error of approximately 7.7%. Given the limited availability of historical lime data, we consider this agreement acceptable. The conversion equation, source data, validation results, and additional reference have been added to SI-2, and the complete calculation procedure will be provided as a supplementary file. All results affected by this correction have been recalculated.

Following the reviewer's recommendation, we also re-examined the corresponding data sources for other countries. The Brazilian data were obtained from official tables of the Brazilian Institute of Geography and Statistics (IBGE) explicitly reporting lime production (Produção de cal) for 1940–1958 and 1950–1964, rather than limestone production. Values for 1930–1939 were extrapolated using an autoregressive model and are now clearly identified as model estimates in SI-2. The French data for 1930–1958 were obtained from the French Statistical Yearbook. We rechecked the original French entries and confirmed that they refer to chaux (lime), rather than calcaire (limestone). Therefore, no corrections were required for the original Brazilian or French data.

To improve transparency and prevent similar ambiguity, SI-2 now distinguishes among directly reported lime-production data, lime production converted from limestone data, and model-derived estimates.

Please see Lines 436-472 of the revised manuscript.

L177: IBGE, 2026: The reference is to a webpage that discusses briefly the role of

IBGE in teaching, and has nothing about lime, lime data or data of any sort. A reviewer is left to guess how this ended up as the reference for this dataset.

Author response: We thank the reviewer for identifying this incorrect reference. We agree that the previously cited IBGE webpage does not contain lime-production data and therefore cannot support the historical Brazilian series. The generic webpage was cited in error and has been replaced with the specific IBGE Physical Production Tables containing the series identified as Produção de cal (lime production).

We also clarified the temporal coverage and reconstruction status of the Brazilian data. The values for 1940–1964 are directly reported lime-production statistics from the IBGE tables, whereas the values for 1930–1939 are not reported observations and are explicitly identified as autoregressive estimates. The complete source classification and reconstruction details are now documented in Supplementary Table SI-2, Data 1, allowing readers to distinguish reported data from modeled values.

The revised manuscript now states:

"Brazilian lime production for 1940–1964 was taken from the IBGE Physical Production Tables (Produção de cal; IBGE, 2026), while 1930–1939 values are explicitly identified as autoregressive estimates."

Please see Lines 422 of the revised manuscript.

.We have replaced the incorrect link with the webpage containing the relevant data tables. We have also revised SI-2 to distinguish clearly between official observations and model-derived estimates and to provide the table titles, coverage periods, download link, and extrapolation method. Thus, the issue arose from an incorrect reference link and insufficient documentation, rather than confusion between Brazilian lime and limestone data. Correct Source: https://seculoxx.ibge.gov.br/economicas/atividades-economicas/tabelas-de-producao-fisica.html

L178: "Dominion Bureau of Statistics, 2026": This clearly refers to very old publications (Canada has not been a dominion for many decades), so the publication year is clearly not 2026. Replace 2026 with "various years" in this citation, since the data were presumably retrieved from many reports published over many years.

Author response: We thank the reviewer for identifying this bibliographic error. We agree

that "2026" represents neither the publication year nor an appropriate citation year for the historical reports issued by the Dominion Bureau of Statistics. These Canadian data were compiled from multiple historical publications issued in different years.

We have therefore replaced "Dominion Bureau of Statistics, 2026" with "Dominion Bureau of Statistics, various years." The individual reports are now cited according to their original publication years in the supplementary source documentation.

The revised manuscript states:

"French, Canadian, German, and former Soviet records were compiled from national statistical yearbooks, with Canadian publications cited by their historical publication years (Dominion Bureau of Statistics, various years)."

Please see Lines 418–435 of the revised manuscript and Supplementary Table SI-2, Data 1.

L179-180: It's unclear to me what the regression for Germany is supposed to be. The regression coefficients suggest a model of lime ~ steel + cement. This should be presented in the MS, not delegated to an SI file. The authors haven't explained why they use this model. In modern times, cement plants do not use lime, and any correlation between cement production and lime production is probably just a sign that both are needed in an economy, without indicating any direct link between the two. At certain points in the past German plants might have blended lime in their final product, but that would have changed dramatically over time. That a regression model produces a good fit to the training data does not mean that it is a good model for predicting earlier periods. The training period for Germany was from 1959, which is when new standards were introduced, curtailing the use of lime, but this training period has been used to predict earlier periods where very different behaviour exised. It is therefore very difficult to have any faith in these historical extrapolations.

Author response: We sincerely thank the reviewer for this important methodological criticism. We agree that the previous regression based on German steel and cement production did not provide a sufficiently robust basis for reconstructing earlier lime production. In particular, a statistical correlation between cement and lime production does not establish a

direct material relationship, and a model calibrated using post-1959 observations should not be assumed to represent the different industrial structures and standards of earlier decades.

We therefore removed the regression model for Germany entirely, including the steel- and cement-production predictors and the associated regression coefficients. After conducting an additional source search, we obtained historical German lime-production records from national statistical yearbooks accessed through the German National Library. These directly reported statistics now form the basis of the German historical series. Missing values between reported years were filled only by linear interpolation, rather than by extrapolation using steel or cement production. The German process-emission and carbonation-uptake estimates were subsequently recalculated using the revised activity data.

The revised manuscript now clarifies that:

"French, Canadian, German, and former Soviet records were compiled from national statistical yearbooks, with Canadian publications cited by their historical publication years (Dominion Bureau of Statistics, various years)."

The treatment of missing German observations and the original statistical-yearbook sources are documented in Supplementary Table SI-2, Data 1. No steel–cement regression is retained in either the revised manuscript or the supplementary dataset. Please see Lines 418–435 of the revised manuscript.

L185-186: "specific allocation ratios": Are these temporally constant? If not, is that appropriate?

Author response: Thank you for this comment. The allocation ratios used for the United Kingdom are temporally constant in our reconstruction. They were derived from Table IV of the Ordnance Survey explanatory text, which reports the principal uses of limestone, dolomite, and chalk in Great Britain in 1949. In this table, 16.1% of limestone and dolomite output and 4.9% of chalk output were classified as "burnt for lime at quarry". We therefore used 0.161 and 0.049 as fixed proxy allocation ratios to estimate the carbonate raw materials used for lime production in years for which direct lime production statistics were unavailable.

We agree that this assumption was not sufficiently clear in the original manuscript. We have revised the text to explicitly state that these ratios are fixed, 1949-based proxy allocation factors rather than time-varying annual parameters. We also acknowledge that fixed allocation

ratios may not fully capture temporal changes in the end-use structure of limestone, dolomite, and chalk. Nevertheless, in the absence of continuous direct lime production statistics for the early period, this approach provides a transparent and reproducible proxy for reconstructing UK lime production.

L216-208: How did the authors know the CaO contents for all these countries? Tier 2 means that data are available on the types of lime produced. Specifically, High-calcium lime (CaO + impurities), Dolomitic lime, and Hydraulic lime. I cannot see this in the MS nor in the SI data. Please clarify.

Author response: We thank the reviewer for this important comment. We agree that the original manuscript did not adequately explain the sources and resolution of the CaO-content data and did not clearly distinguish our parameterization from the activity data required for a complete IPCC Tier 2 approach.

Annual, country-specific production data disaggregated into high-calcium, dolomitic, and hydraulic lime were not available for all countries and historical periods. We therefore did not assume that the precise annual CaO content or production share of each lime type was known for every country. Instead, reported CaO-content measurements were compiled from peer-reviewed studies, technical standards, national statistics, and reports published by industrial associations. Where reported by the original sources, the observations were classified according to lime type, including high-calcium, dolomitic, and hydraulic lime, and according to their construction, metallurgical, or chemical end use. Where country-specific observations were insufficient, broader literature-derived ranges were used rather than assigning an unsupported precise value.

Probability distributions were constructed according to the available observations. Triangular distributions were defined using minimum, most probable, and maximum values, whereas normal distributions were defined using the reported or calculated mean and standard deviation. These distributions were sampled in the Monte Carlo analysis to propagate compositional uncertainty into the process-emission and carbonation-uptake estimates.

We agree with the reviewer that an IPCC Tier 2 approach requires activity data or production shares differentiated by lime type. Because such disaggregated production data were unavailable for many countries and historical periods, our method should not be interpreted as

a complete IPCC Tier 2 implementation. We have therefore removed or qualified the Tier 2 terminology and now describe the method as a literature-based probabilistic parameterization of CaO-content uncertainty. Supplementary Table SI-3, Data 7 has also been expanded to report the underlying CaO-content observations, lime and material classifications, end uses, references, and probability-distribution parameters.

L229: I think it would make more sense here to present the method before the data. The authors are planning to use a model and then need to populate that with data. Please restructure the paper. In some fields, such as social science, one has a dataset that one wishes to explore, so one first discusses the dataset and then explains the methods one uses for that purpose. In this article, the authors start with a model and need data to feed into that model, which is very different. It then makes much more sense to start with the model description then explain the data used. That would help the reader's understanding.

Author response: We thank the reviewer for this helpful structural suggestion. We agree that, because the study first establishes an accounting framework and then populates it with activity data and parameters, presenting the methods before the data sources provides a clearer logical sequence.

We have therefore reorganized Section 2 as follows:

Section 2.1, Estimation of Process CO_2 Emissions from Lime Production, introduces the process-emission model and country-specific emission-factor calculation.

Section 2.2, Estimation of CO_2 Uptake by Lime, defines the system boundary, general calculation model, carbonation models, and material-specific equations.

Section 2.3, Data Sources and Processing, subsequently describes the lime-production series, industrial activity data, historical reconstruction procedures, conversion coefficients, interpolation, and source harmonization.

Section 2.4, Uncertainty Assessment, explains how uncertainty in activity data and model parameters is propagated through the accounting framework.

This revised structure first explains the quantities to be calculated, the governing equations, and the required variables, and then presents the data used to populate the models. Cross-references, equation numbers, figures, and supplementary-table citations were updated accordingly. Please see Sections 2.1–2.4 of the revised manuscript.

L231: "gamma was set to 1, indicating complete carbonation during the production process": I'm not sure that "indicating" is the right word here. That gamma was set to 1 indicates only that all CaO is carbonated, not that it happens in the production process. Perhaps instead say why it is 1, i.e. that already during the production of these products, the lime is fully carbonated.

Author response: We thank the reviewer for this precise observation. We agree that the original wording incorrectly suggested that setting $\gamma = 1$ demonstrated when carbonation occurred, whereas it only specified the assumed extent of CaO conversion.

We have revised the formulation to state the process-based reason for assigning complete conversion. For precipitated calcium carbonate (PCC) and carbonation sugar (SUG), lime reacts with CO_2 as an integral part of the production process. Therefore, the effective CaO conversion fraction,Φ_m, is set to 1 for these pathways. This wording distinguishes the physical timing of carbonation from the numerical value assigned to the conversion parameter.

The revised text now reads:

"For PCC and SUG, carbonation is integral to production andΦ_m is set to 1 (Wang and Shen, 2002)."

The corresponding equations now defineΦ_m specifically for PCC and SUG, while the ultimate conversion fraction γ_m and time-dependent function $R_m(a)$ are retained for materials whose carbonation progresses after production. Please see Lines 220–238 of the revised manuscript..

L232-235: No explanation of why R=1 was used for these categories. Since this is a critical parameter, and LSS is the largest share of lime carbonation in the study's results, this assumption must be justified. For example, without being an expert in these things, I struggle to see why LSS would have R=1 when much CaO is locked up more tightly in C-A-H and C-S-H, and even the CaO that is free as Ca(OH)2 has very little access to CO2 because it is highly compressed and generally under layers of asphalt or concrete. This is engineered soil (a term that the authors could add to aid understanding), and the goal is maximum stability, which means high compression. So how is R=1? Further, R "was assumed to be 1, representing complete carbonation within one year" seems incorrect. R=1 doesn't mean complete carbonation within one year; it means that maximum

carbonation is reached within one year. It has to be combined with gamma to say how much that maximum is. The use of "complete" here is incorrect.

Author response: We sincerely thank the reviewer for identifying both the physical and terminological problems with the previous treatment. We agree that assigning $R = 1$ to lime-stabilized soil (LSS) was insufficiently justified. LSS is a highly compacted engineered soil, and its carbonation can be restricted by low gas permeability, overlying pavement layers, and the incorporation of Ca into C–A–H and C–S–H phases. It should therefore not be assumed to reach its maximum carbonation extent within one year.

We have removed LSS from the $R = 1$ category. In the revised framework, LSS and lime mortar (MOR) are represented using a time-dependent slab model (Fig. 2a). Their effective conversion fraction is calculated as

$$\Phi_m = \gamma_m R_m(a)$$

where γ_m is the material-specific ultimate CaO conversion fraction and $R_m(a)$ is the cumulative fraction of that ultimate extent reached at material age a. LSS carbonation therefore progresses according to its material-specific carbonation-rate coefficient and design thickness rather than being completed in the production year. The LSS-specific parameters and uncertainty ranges are reported in Supplementary Table SI-3, Data 10, Data 14, and Data 16.

We also agree that "complete carbonation" was technically incorrect. The revised manuscript now states explicitly that $R_m(a)$=1 means only that the pathway-specific ultimate carbonation extent defined by γ_m has been reached. It does not mean that all CaO has been converted unless $\gamma_m = 1$. Thus, the cumulative proportion of CaO converted at age a is $\gamma_m R_m(a)$, not $R_m(a)$ alone.

The revised text reads:

> "For LSS, MOR, SS, and BFS, γ_m is the material-specific ultimate CaO conversion fraction and $R_m(a)$ is the cumulative fraction of that ultimate extent reached at material age a. Thus, $\gamma_m R_m(a)$ represents the cumulative proportion of CaO converted by that age. $R_m(a)$=1 indicates that the pathway-specific ultimate extent has been reached, but does not imply conversion of all CaO unless $\gamma_m = 1$."

and:

> "LSS and MOR use the slab model (Fig. 2a)."

The description of LSS has also been clarified to identify it as a compacted engineered soil. Please see Lines 220–247 and the material-specific LSS equations in the revised manuscript

L234-235: "different kinetic models were applied": This is the only mention in the MS of the term "kinetic model", with no explanation. Why were different kinetic models applied? Some explanation to the reader would be helpful. Are the "kinetic models" those stylised assumptions depicted in Figure 1?

Author response: We thank the reviewer for this important comment. We agree that the term "different kinetic models" was imprecise and that the physical basis for selecting the different representations was not sufficiently explained. The models referred to are the geometry-based carbonation representations shown in the revised Fig. 2, rather than unexplained empirical kinetic models.

We have replaced the original terminology and clarified that model selection is based on the characteristic material geometry and CO_2-exposure pathway:

Lime-stabilized soil (LSS) and lime mortar (MOR) are represented by the slab model (Fig. 2a), because carbonation progresses inward from an exposed planar surface.

Fine-particle materials stored in stockpiles, including lime kiln dust (LKD), paper mill lime mud (LM), red mud (RM), and carbide slag (CS), are represented by the layered-stockpile mechanism (Fig. 2b). This representation explains rapid carbonation near the exposed surface and lower conversion at greater depths because of restricted CO_2 transport and successive burial by newly deposited material.

Steel slag (SS) and blast-furnace slag (BFS) are represented by the spherical-particle model (Fig. 2c), in which carbonation progresses inward from the particle surface.

For the slab and spherical-particle models, time-dependent carbonation depth follows the square-root-of-time relationship and is converted into a carbonated fraction according to material geometry. By contrast, the layered-stockpile diagram provides the physical basis for using a literature-reported, depth-averaged one-year CaO conversion fraction; it is not treated as an independently calibrated time-dependent kinetic model. These simplified representations capture the principal geometry and exposure conditions of each material pathway but do not attempt to simulate all field-scale transport and microstructural processes.

The revised manuscript now states:

" The conversion factor γ and temporal function \(R\) therefore describe different processes. The former constrains the ultimate chemically available CaO fraction, whereas the

latter represents progression toward that extent. Model selection follows the order shown in Fig. 2. LSS and MOR use the slab model (Fig. 2a). The layered-stockpile representation for LKD, LM, RM, and CS (Fig. 2b) explains rapid carbonation near the exposed surface and lower conversion at greater depths because of restricted CO_2 transport and successive covering by newly deposited material. SS and BFS use the spherical-particle model (Fig. 2c)."

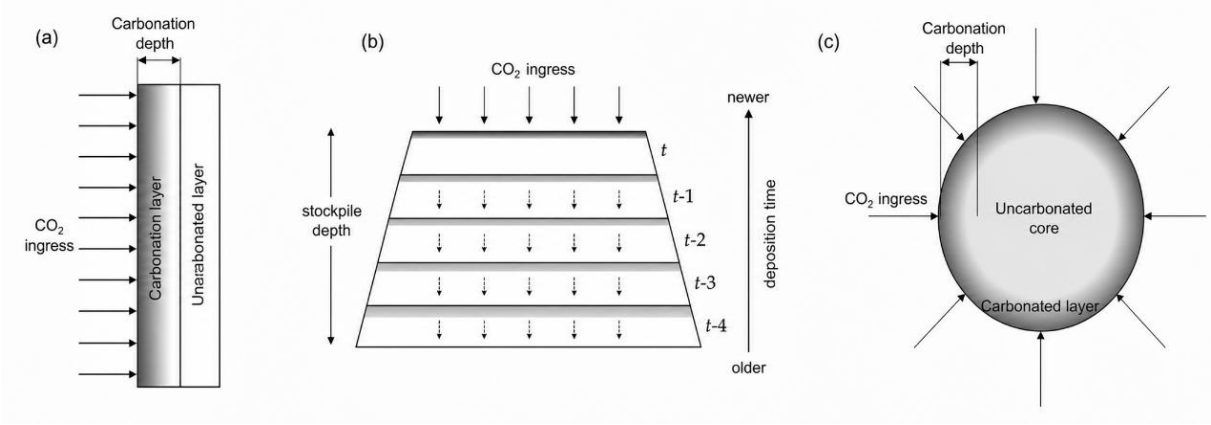

Fig. 2. Schematic carbonation representations: (a) slab model for LSS and MOR; (b) layered stockpile mechanism for LKD, LM, RM, and CS; and (c) spherical-particle model for SS and BFS.

Please see Lines 239–250 and the annual-carbonation-ratio equations in the revised manuscript.

Fig 1: The diagram for the pile model suggests CO2 ingress only from the top and sides, yet also suggests just as much carbonation of the bottom layer. How is this consistent?

Author response: We thank the reviewer for identifying this inconsistency in the original schematic. We agree that the previous pile diagram could be interpreted as assigning similar carbonation to all layers, despite showing CO_2 ingress primarily from the exposed surfaces.

We have revised the diagram and accompanying explanation to represent the sequential deposition and burial of stockpiled material more clearly. Each newly deposited layer is initially exposed at the stockpile surface and can undergo relatively rapid carbonation. As additional material is deposited, the older layer is buried, CO_2 transport becomes increasingly restricted, and its subsequent carbonation proceeds more slowly. The bottom layers can therefore exhibit some carbonation because they were previously exposed surface layers, but they are not assumed to carbonate to the same extent as the currently exposed upper layer.

The revised Fig. 2b indicates the direction of deposition from older bottom layers to newer

upper layers and the progressive reduction in CO_2 penetration with depth. The figure is a conceptual representation of the physical mechanism and does not assign identical conversion fractions to individual layers. Because continuous depth-dependent measurements are unavailable for all materials and countries, the quantitative calculation uses the literature-reported one-year, depth-averaged CaO conversion fraction and its uncertainty range rather than assuming uniform carbonation throughout the stockpile.

The corresponding text now explains that:

"The layered-stockpile representation for LKD, LM, RM, and CS (Fig. 2b) explains rapid carbonation near the exposed surface and lower conversion at greater depths because of restricted CO_2 transport and successive covering by newly deposited material."

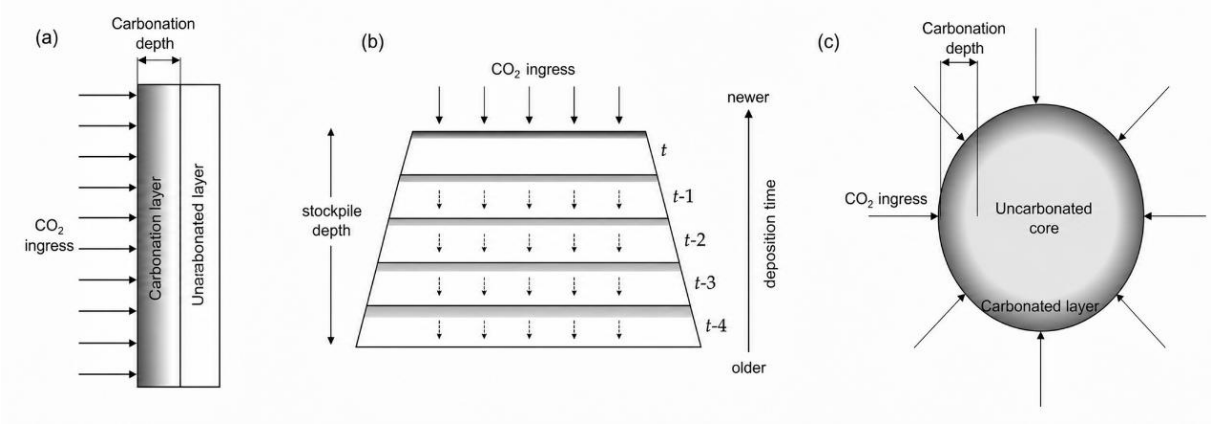

Fig. 2. Schematic carbonation representations: (a) slab model for LSS and MOR; (b) layered stockpile mechanism for LKD, LM, RM, and CS; and (c) spherical-particle model for SS and BFS.

Please see the revised Fig. 2b and the associated description and equations in Section 2.2.1.

L249: Again, it's highly unclear what this timeline division actually means in practice. Which parameters were changed for each period? This is fundamental and should not be left to the reader trying to understand the method by studying the accompanying data files.

Author response: We thank the reviewer for this important comment. We agree that the original manuscript did not explain clearly which parameters were controlled by the historical-period division or how the division was implemented in the calculations.

We have revised the manuscript to state explicitly that the four periods—1930–1949, 1950–1972, 1973–1999, and 2000–2024—are used to assign two types of parameters for steel slag (SS) and blast-furnace slag (BFS):

The SS and BFS generation coefficients, r_{ss} andr_{bfs}, which convert crude-steel or pig-iron production into total slag generation.

The SS and BFS pathway-allocation fractions, U_{ss} and U_{bfs}, which identify the fractions used in road-base applications or stored in open-air stockpiles and therefore included in the natural-carbonation accounting.

These periods do not control crude-steel or pig-iron production, sectoral lime-use shares, CaO contents, or other material parameters. In particular, U_{ss} and U_{bfs} are not overall slag-utilization rates; they are pathway-allocation parameters specific to the natural-carbonation boundary.

The four periods are data-constrained accounting windows representing broad historical stages in iron- and steelmaking technology and slag-management practices. They do not imply that technological transitions occurred simultaneously in all countries. Country-specific coefficients and pathway-allocation fractions were applied where data were available. Otherwise, separate documented assumptions were assigned to developed and developing regions. The corresponding numerical values, assumptions, and references are reported in Supplementary Table SI-3, Data 8–9.

The revised manuscript now states:

"Complete annual and country-specific data were unavailable for the SS and BFS generation coefficients and for the fractions directed to road-base applications or open-air stockpiles. We therefore used four non-overlapping accounting periods: 1930–1949, 1950–1972, 1973–1999, and 2000–2024. These periods represent broad stages in iron- and steelmaking technology and serve only as common accounting windows; they do not imply synchronous technological transitions across countries. Crude-steel and pig-iron production were obtained from statistical data and were not determined by these assumptions. Country-specific slag-generation coefficients and pathway-allocation fractions were used where available. Otherwise, separate values were assigned to developed and developing regions based on differences in industrialization, technology-adoption timing, and slag-management practices."

Please see Lines 168–172, Section 2.4, and Supplementary Table SI-3, Data 8–9 of the revised manuscript.

L255:256: The subscript clearly should not be "progress" but rather "process".

Author response: We thank the reviewer for identifying this typographical error. The subscript “progress” has been corrected to “process”.

L274: It's now ten years since Xi et al 2016. Perhaps we should not expect readers to trawl back through literature to understand the process here. What does gamma actually represent physically? Is it actually meant to represent that the diffusion process in fact does not proceed according to Fick's law because the resulting CaCO3 reduces the ability of CO2 to enter the substance? I think many readers would appreciate having such extra insight.

Author response: We thank the reviewer for this helpful comment. We agree that the physical meaning of γ should be explained directly in the manuscript rather than requiring readers to consult Xi et al. (2016).

In the revised framework, γ_m represents the material-specific ultimate CaO conversion fraction: the maximum fraction of the total CaO in material m that is chemically and mineralogically available for conversion to $CaCO_3$ under the corresponding pathway conditions. It accounts for the fact that some Ca may occur in phases that do not carbonate, or carbonate only to a limited extent, under natural exposure conditions. It is therefore a constraint on the ultimate conversion extent rather than a carbonation-rate parameter.

The temporal function $R_m(a)$ describes the progression toward this material-specific ultimate extent at material age a. Accordingly,

$$\Phi_m = \gamma_m R_m(a)$$

and $\gamma_m R_m(a)$ is the cumulative fraction of the total CaO converted by that age. An $R_m(a)$ value of 1 means that the pathway-specific ultimate extent defined by γ_m has been reached; it does not mean that all CaO has been converted unless $\gamma_m = 1$.

We have also clarified that γ_m does not represent deviation from Fickian diffusion or the transport resistance caused by the formation of a $CaCO_3$ product layer. For the slab and spherical-particle pathways, time-dependent CO_2 transport and inward carbonation are represented by $R_m(a)$, the carbonation-rate coefficient, and the corresponding geometry-based square-root-of-time relationships. The formation of a carbonated surface layer and the associated reduction in CO_2 ingress therefore affect the temporal progression represented by $R_m(a)$, rather than the ultimate chemical-availability parameter γ_m.

For PCC and SUG, carbonation is integral to production, and the effective conversion fraction Φ_m is set directly to 1. For LKD, LM, RM, and CS, the model uses the literature-reported one-year, pile-average effective conversion fraction $\underline{\gamma}_{m,1}$, rather than separating γ_m and $R_m(a)$.

The revised manuscript now states:

> "For LSS, MOR, SS, and BFS, γ_m is the material-specific ultimate CaO conversion fraction and $R_m(a)$ is the cumulative fraction of that ultimate extent reached at material age a. Thus, $\gamma_m R_m(a)$ represents the cumulative proportion of CaO converted by that age. $R_m(a) = 1$ indicates that the pathway-specific ultimate extent has been reached, but does not imply conversion of all CaO unless $\gamma_m = 1$."

and:

> "The conversion factor γ and temporal function R therefore describe different processes. The former constrains the ultimate chemically available CaO fraction, whereas the latter represents progression toward that extent."

Please see Lines 224–247 and the annual-carbonation-ratio equations in Section 2.2.1 of the revised manuscript

L284: Not "including", since the authors present all of the options, not only some. Use a colon to introduce this complete list. And add "(depicted in Figure 1)" at end of sentence.

Author response: We thank the reviewer for this correction. We agree that "including" was inappropriate because the sentence presents the complete list of model representations rather than selected examples. We have replaced it with a colon and added an explicit reference to the corresponding figure. Because the figures were renumbered during manuscript restructuring, the schematic is now Fig. 2 rather than Fig. 1.

The revised sentence reads:

"Three geometry-based representations were used to describe the temporal development of carbonation: a slab model, a layered-stockpile model, and a spherical-particle model, as depicted in Fig. 2."

Please see the Annual Carbonation Ratio subsection in Section 2.2.1 of the revised manuscript.

L316-318: This explanation of why the authors choose R=1 for LKD comes after they've already said twice that R=1. Please re-organise.

Author response: We thank the reviewer for this helpful comment. We agree that the original organization introduced the assumption before explaining its basis. During revision, we not only reorganized the text but also reconsidered the underlying treatment. LKD is no longer assigned $R = 1$ or assumed to reach 100% carbonation within one year.

The general calculation model now introduces the LKD treatment when the pathway-specific effective conversion fraction is first defined. For LKD, LM, RM, and CS, the evaluation age is fixed at one year and the effective conversion fraction is expressed as:

$$\Phi_m = \underline{\gamma}_{m,1}$$

where the barred parameter is the literature-reported, one-year pile-average CaO conversion fraction. Its reported average and range are sampled directly in the Monte Carlo analysis. The subsequent LKD-specific subsection now only identifies the LKD generation pathway and refers readers to Supplementary Table SI-3, Data 14 and Data 17 for the carbonation parameters. The repeated statements assigning $R = 1$ have therefore been removed. Please see the general calculation model in Section 2.2.1 and the LKD subsection in Section 2.2.2.

L327: Method 2 here says it uses Fick's law, but Method 3 doesn't mention it, even though it clearly uses it. Readers will be very confused by this point. Saying you use Fick's law in only one subsection implies you don't use it in the others.

Author response: We thank the reviewer for identifying this inconsistency. We agree that mentioning the diffusion assumption only in the slab-model subsection could imply that the spherical-particle model used a different basis. We have therefore moved the common assumption to the introduction of the quantitative time-dependent models.

The revised text now states that the slab and spherical-particle pathways both use the square-root-of-time relationship derived from the common Fickian diffusion representation:

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

where d is carbonation depth, k is the material-specific carbonation-rate coefficient, and t is material age. The two models differ in how carbonation depth is converted into the carbonated fraction according to planar or spherical geometry.

The layered-stockpile pathway is now distinguished explicitly from these quantitative time-dependent models. Because continuous depth profiles were unavailable, LKD, LM, RM, and CS use the literature-reported one-year, depth-averaged conversion fraction rather than an

independently calibrated Fickian depth function. This revised organization makes clear which pathways use the square-root-of-time relationship. Please see the Annual Carbonation Ratio subsection and Eqs. (8)– (11) in Section 2.2.1.

L333-334: "U_ss denotes the ratio of stockpiling or roadbed utilization": Unclear. The ratio of stockpiling to roadbed utilization? The ratio of stockpiling utilization or the ratio of roadbed utilization? None of these interpretations are easy to understand. Do the authors mean the share of stockpiling in total? Or do they mean the share of stockpiling vs the share used in roadbeds? What differentiates these two uses? Is the mass of SS produced a function of the share that is stockpiled? Please reword for clarity.

Author response: We thank the reviewer for identifying this ambiguity. We have replaced the phrase "ratio of stockpiling or roadbed utilization" and now define U_ss directly as the combined fraction of total steel slag directed to road-base applications or stored in open-air stockpiles and therefore included in the natural-carbonation accounting. It is not the ratio of stockpiling to road-base use.

We have also separated total SS generation from the mass selected for carbonation accounting. Total SS production is calculated independently as:

$$M_{ss,total} = m_{crude\ steel} \times r_{ss}$$

The mass included within the natural-carbonation boundary is then calculated as:

$$M_{ss,carb} = M_{ss,total} \times U_{ss}$$

Thus, U_ss does not affect estimated SS production; it only allocates a fraction of the produced slag to pathways included in the present accounting boundary. Road-base use and open-air stockpiling are different management pathways, but both permit contact with atmospheric or pore-water CO_2. Slag incorporated into other valorized products enters downstream product systems and is excluded because its carbonation depends on product-specific service and exposure conditions (Pullin et al., 2019; Chukwuma et al., 2021; Elyasi Elyasi Gomari et al., 2024; Li et al., 2022). This exclusion means that its carbonation lies outside the present accounting boundary, not that its actual carbonation is zero. Please see the SS calculation and boundary explanation in Section 2.2.2.

L339: "following the same logic as SS", but the logic for SS was not explained either.

Author response: We thank the reviewer for identifying this unclear cross-reference. We agree that "following the same logic as SS" required readers to infer the meaning of U_bfs and was therefore inadequate. We have removed this phrase and now define every BFS parameter directly.

Total BFS generation is determined independently from blast-furnace iron production and the BFS generation rate:

$$M_{bfs,total} = m_{bfs} \times r_{bfs}$$

The lime-derived BFS fraction included within the natural-carbonation boundary is calculated as:

$$M_{bfs,included} = m_{bfs} \times r_{bfs} \times l_{bfs} \times U_{bfs}$$

Here, l_bfs isolates the proportion of CaO in BFS originating specifically from lime input, while U_bfs is the combined fraction of total BFS directed to road-base applications or stored in open-air stockpiles. U_bfs is not the ratio of stockpiling to road-base use and does not affect total BFS generation; it only identifies the pathway fraction included within the natural-carbonation boundary. The revised manuscript also explains that excluded BFS enters downstream product systems and is outside the present accounting boundary rather than being assumed to undergo zero carbonation. Please see the BFS calculation and boundary explanation in Section 2.2.2 and Supplementary Table SI-3, Data 15

L359: Uncertainty assessment: I see no mention of whether any variables are assumed to be correlated. I take it that all variables are assumed to be uncorrelated, i.e. their errors are entirely independent of each other? In that case the authors use an uncertainty method (Montecarlo) that benefits the authors greatly in that the more they subdivide the total into separate variables, the smaller their final overall uncertainty assessment will be, by definition. That's how addition in quadrature works. If we start with a global estimate without subdividing by region, then we might say total lime production is 200 Mt (say) and relative uncertainty is 10% then our absolute uncertainty is ±20 Mt. If we then subdivide into four equal regions, each with 10% uncertainty, and we assume the errors are independent, then we add the errors in quadrature, so the absolute uncertainty would be sqrt(4x5^2)=10Mt and the relative uncertainty would be 10/200=5%. We have halved our uncertainty simply by subdividing the global into four regions, but only because we have assumed that the errors in these four regions are independent. The same problem arises when the use of lime is subdivided and errors are assumed uncorrelated, and so for any subdivision.

Author response: We thank the reviewer for this important and technically insightful

comment. We agree that the treatment of correlation assumptions in Monte Carlo uncertainty propagation must be stated explicitly. The reviewer is correct that, if subdivided components are assumed to have independent errors, aggregation can reduce the relative uncertainty of the total estimate, even when the uncertainty assigned to each component has not changed.

In response, we revised Section 2.4 to clarify the correlation assumptions used in our Monte Carlo framework. In the baseline implementation, no explicit cross-country or cross-parameter correlation matrix was imposed; therefore, sampled variables were treated as independent within their assigned distributions, except for deterministic accounting relationships applied after sampling. We now explicitly state that the reported 95% uncertainty intervals should be interpreted as conditional on this independence assumption.

We also acknowledge in the revised manuscript that positive correlations may exist among some parameters, for example among countries relying on similar statistical sources or among material parameters governed by common industrial technologies. If these correlations were explicitly modeled, the aggregated uncertainty intervals could be wider than those reported here. To avoid overstating the benefit of subdivision, we have also revised the discussion and no longer interpret reduced aggregate uncertainty as evidence that subdivision alone improves data quality. Instead, the revised text emphasizes that the reported uncertainty intervals represent propagated uncertainty under the stated assumptions, and that unmodeled correlations remain an important limitation and a direction for future methodological improvement.

The revised text in the Abstract now reads:

We identified 18 groups of factors affecting the estimation of process CO_2 emissions from lime production and carbonation uptake by lime-based materials, comprising 1,894 input parameters with specified statistical distributions (Supplementary Table SI-3, Data 1–15). Given the substantial uncertainty in these parameters, we applied the Monte Carlo method recommended by the 2006 IPCC Guidelines for National Greenhouse Gas Inventories. The statistical distributions of these variables were incorporated into the accounting model, and 10,000 Monte Carlo iterations were performed. Final estimates are reported as the median of the simulated distributions, and the 95% uncertainty intervals (UIs) were calculated using the percentile method, with the 2.5th and 97.5th percentiles used as the lower and upper bounds, respectively.

Uncertainty introduced by historical activity-data reconstruction and source harmonization was explicitly included in the Monte Carlo framework. Source-specific uncertainty ranges were assigned according to data reliability, record completeness, and reconstruction method. Directly reported statistics were assigned narrower ranges than

reconstructed data. For regression-derived estimates, uncertainty was characterized using model fitting errors, calibration diagnostics, and goodness-of-fit information reported in Supplementary Table SI-2 Data1. For conversion-derived and interpolated estimates, uncertainty ranges were assigned based on the corresponding activity-data and conversion-parameter uncertainties in Supplementary Table SI-3. Rather than introducing an additional standalone "source-integration" parameter that would be difficult to constrain independently, we incorporated discrepancies among overlapping datasets, uncertainty associated with limestone-to-lime conversion, differences in statistical boundaries, and regression-based reconstruction uncertainty into the uncertainty ranges assigned to historical activity data. This treatment differentiates complete statistical records from reconstructed, converted, and interpolated values, and allows uncertainty levels to vary among countries and time periods according to source quality.

During each Monte Carlo iteration, these activity-data uncertainty terms were jointly sampled with emission factors, CaO contents, material-use shares, carbonation ratios, and other model parameters, and then propagated through the process-emission and carbonation-uptake accounting models to generate distributions of annual and cumulative emissions, uptake, and net emissions.

No explicit cross-country or cross-parameter correlation matrix was imposed in the baseline Monte Carlo implementation; therefore, sampled variables were treated as independent within their assigned distributions, except for deterministic accounting relationships applied after sampling. We acknowledge that positive correlations may exist among some parameters, for example among countries using similar statistical sources or among material parameters governed by common industrial technologies. Accordingly, the reported 95% UIs should be interpreted as conditional on this independence assumption and may be narrower than intervals obtained under a fully correlated error structure. This limitation is now explicitly stated, and the reported aggregate uncertainty is not interpreted as evidence that subdivision alone improves data quality.

L363: Looking at sheet Data1, it is difficult to understand why in every case the minimum of the uncertainty range is higher than the maximum. This must surely be an error?

Author response: We thank the reviewer for identifying this error. During the preparation of Supplementary Table SI-3, Data 1, the minimum and maximum values were inadvertently reversed because of a data-compilation error. We have corrected all affected entries and systematically rechecked the uncertainty ranges to ensure that each minimum value is lower than its corresponding maximum value. The corrected file, together with the other revised data files, will be uploaded to Zenodo upon completion of all revisions, and the dataset associated with https://doi.org/10.5281/zenodo.18616060 will be updated accordingly.

L369-371: "The 90% confidence intervals (CIs) were calculated using the percentile method; specifically, the 5th and 95th percentiles of the modeled results served as the

lower and upper limits of the 90% interval." This is not a particular method; it is the definition of the 90% CI.

Author response: We thank the reviewer for this clarification. We agree that the selected percentiles directly define the bounds of the reported interval and should not have been described as a separate "percentile method." We have therefore removed this wording.

We have also changed the reported interval from 90% to 95% to ensure consistency throughout the manuscript. Because the interval represents uncertainty propagated from the assigned input distributions through the Monte Carlo simulation, we refer to it in the uncertainty-assessment section as a 95% uncertainty interval. Final estimates are reported as the medians of the simulated distributions, and the interval bounds are defined directly by the corresponding quantiles.

The revised text now reads:

"Final estimates are reported as the median of the simulated distributions. The 2.5th and 97.5th percentiles define the lower and upper bounds, respectively, of the 95% uncertainty interval."

Please see Section 2.4, "Uncertainty Assessment," of the revised manuscript.

L375: "allowed to establish": replace with "allowed us to establish" or "allowed the establishment of".

Author response: We thank the reviewer for identifying this grammatical error. We have replaced "This allowed to establish" with "This allowed us to establish" and revised the surrounding sentence for clarity.

L383-385: Now, after perhaps 3-4 times visiting the idea of "technological evolution", the authors provide a clue as to what that means. It is the utilization rates of steel slag and BFS that are assumed to change for each period? Presumably not both "production and utilization rates", since production rate is based on "data", not on assumptions of technological change? Why would utilization rates of slag change in the same way in developed countries as in developing countries during the 20th century? This set of assumptions is described without prefacing to say that data are not available so the authors make some assumptions to fill the data gap. Calling it "Dynamic Parameterization of Technological Evolution" it very flowery language that makes it

sound much fancier than it really is. It's an assumption because data aren't available. I still struggle to see how the "industrial shifts" occurring in Europe and the USA in the 1930s would also have occurred in China of the 1930s, ditto "efficiency revolution" in the 1950s and 1960s.

Author response: We thank the reviewer for this important comment. We agree that the original phrase "Dynamic Parameterization of Technological Evolution" was unnecessarily elaborate and obscured the fact that assumptions were introduced because complete annual, country-specific data were unavailable. We have replaced it with the more transparent description "data-constrained temporal and regional parameterization of SS and BFS."

We have also clarified the distinction between statistical activity data and the parameter assumptions. Crude-steel and pig-iron production are obtained from historical statistics and are not determined by the four-period division. The parameters assigned by period are:

The SS and BFS generation coefficients, which convert reported crude-steel and pig-iron production into estimated slag generation. These coefficients are not steel or iron production rates.

The pathway-allocation fractions representing the portions of SS and BFS directed to road-base applications or open-air stockpiles and consequently included in the natural-carbonation accounting. These parameters are not the overall slag-utilization rates.

These parameters were temporally assigned because complete annual and country-specific series are unavailable. The four periods—1930–1949, 1950–1972, 1973–1999, and 2000–2024—are therefore common accounting windows used to organize the available evidence and fill data gaps. They do not imply that China, Europe, the United States, or other regions experienced identical technological or industrial transitions at the same time.

Country-specific coefficients and pathway-allocation fractions were used where available. When country-level information was unavailable, separate documented values were assigned to developed and developing regions to reflect differences in industrialization, technology-adoption timing, and slag-management practices. Developed and developing countries were therefore not assumed to follow the same parameter trajectory.

The revised manuscript now states:

"Complete annual and country-specific data were unavailable for the SS and BFS

generation coefficients and for the fractions directed to road-base applications or open-air stockpiles. We therefore used four non-overlapping accounting periods: 1930–1949, 1950–1972, 1973 – 1999, and 2000 – 2024. These periods represent broad stages in iron- and steelmaking technology and serve only as common accounting windows; they do not imply synchronous technological transitions across countries. Crude-steel and pig-iron production were obtained from statistical data and were not determined by these assumptions. Country-specific slag-generation coefficients and pathway-allocation fractions were used where available. Otherwise, separate values were assigned to developed and developing regions based on differences in industrialization, technology-adoption timing, and slag-management practices."

The parameter values, assumptions, data availability, and supporting references are documented in Supplementary Table SI-3, Data 8–9. Please see Section 2.4 of the revised manuscript.

L400-403: "Statistical analysis reveals a strong correlation between lime carbon emissions and lime production. However, the growth rate of process emissions was lower than that of lime production, suggesting that technological advancements have reduced the carbon emissions intensity of lime production": I struggle to understand this. The authors only have process emissions, so any change in emissions isn't about tech development, but just about types of inputs used. Since the numbers are entirely based on the data used, there should be no need to guess the cause of change ("suggesting"), and the authors should be able to explain definitively. The reason there is a strong correlation between lime carbon emissions and lime production is simply that the parameters in the model for deriving emissions from production vary to a very small degree, and one does not need after-the-fact statistical analysis to show this.

Author response: We thank the reviewer for this important clarification. We agree that the original text incorrectly interpreted the slower increase in process emissions as evidence of technological advancement. This study quantifies calcination-related process emissions and excludes emissions from kiln fuels and electricity. It therefore does not provide a basis for inferring improvements in kiln technology, fuel efficiency, or energy-related emission intensity.

We also agree that a post hoc correlation analysis is unnecessary because lime production

is the principal activity variable in the process-emission equation. A close relationship between lime production and estimated process emissions is consequently inherent in the accounting model rather than an independent empirical finding. We have removed both the correlation statement and the unsupported technological interpretation.

The revised text now explains the difference directly from the model structure:

"Global lime process emissions broadly followed the trend in lime production because lime production is the principal activity variable in the emission-accounting model. However, emissions increased more slowly than production because the production-weighted emission factor decreased over time. In the model, this emission factor varies with the allocation of lime among downstream sectors and the corresponding CaO contents of lime products. Therefore, the divergence between the growth rates of production and process emissions reflects changes in the modeled composition and sectoral use of lime rather than improvements in production technology."

The citation to Laveglia et al. (2024b), which was previously used to support the technological interpretation, has also been removed from this passage. Please see Section 3.1 of the revised manuscript.

Fig 3: The uncertainty in China's process emissions here clearly indicates a problem. The method used before 1949 is based on the estimates from 1949 (ARIMA) but the value in 1948 has LOWER uncertainty here than the value in 1949. This cannot be. The uncertainties must be properly combined. The uncertainties in 1948 are a combination of the uncertainty from the ARIMA method and the uncertainty from the estimates they are trained on.

Author response: We thank the reviewer for identifying this inconsistency in the original figure. We agree that, under the previous ARIMA backcasting approach, the uncertainty of the reconstructed pre-1949 estimates should have included both the uncertainty of the observations used for model calibration and the additional uncertainty introduced by backcasting. Consequently, the 1948 estimate should not have displayed a narrower uncertainty interval than the 1949 value on which the reconstruction depended.

This issue has been resolved through a substantive revision of the Chinese historical activity data. The ARIMA reconstruction for 1930–1948 has been removed entirely. China's

lime-production series for 1930 – 1949 is now reconstructed using historical limestone production as a proxy. Limestone-production records for mainland China and Northeast China were obtained from Guan (2007), and observed lime-production records for Northeast China were used to calibrate a limestone-to-lime conversion coefficient. The calibrated relationship was applied separately to mainland China and Northeast China, after which the two regional estimates were combined to obtain national lime production.

The associated uncertainty has also been recalculated. For the pre-1950 conversion-derived series, the Monte Carlo analysis jointly propagates uncertainty in the historical limestone activity data, the calibrated limestone-to-lime conversion coefficient, differences in statistical boundaries, and source harmonization. These activity-data uncertainties are then sampled together with the country-specific emission factor and other model parameters. The revised pre-1949 estimates therefore no longer depend on an ARIMA prediction trained from the 1949 observation, and the anomalous uncertainty pattern identified by the reviewer is no longer present.

All Chinese process-emission estimates and their 95% uncertainty intervals were recalculated, and the corresponding figure and supplementary data were updated. Please see the revised Fig. 4, Sections 2.3.1 and 2.4, and Supplementary Table SI-2, Data 1.

L688: "these regions face acute pressure": From whom? The implication here is that developed countries are exerting "acute pressure" on developing countries. Is that the authors' position?

Author response: We thank the reviewer for identifying this ambiguity. We did not intend to imply that developed countries or any other external actors were exerting pressure on developing regions. We have therefore removed the phrase “face acute pressure” and replaced it with a neutral statement based directly on the results: lime process emissions in developing regions continued to increase despite relatively high carbonation-offset levels, indicating the need for further reductions in total emissions.

L872: The author's name is rendered "Shimanishi" not "Shimnishi".

Author response: We thank the reviewer for identifying this typographical error. We have corrected the author’s name from “Shimnishi” to “Shimanishi” throughout the manuscript, supplementary materials, and reference list.

Comment on essd-2026-218, Lei Li

This manuscript presents a potentially valuable dataset on global and national CO2 process emissions from lime production and associated carbonation sinks from 1930 to 2024. The topic is timely and relevant to Earth system carbon accounting, industrial decarbonization, and the possible inclusion of lime carbonation in global carbon budget assessments. The attempt to extend earlier work by increasing country-level resolution, incorporating blast furnace slag, and updating material-specific parameters is worthwhile. However, the manuscript requires revision before it can be considered for publication.

1. **Historical lime production data are reconstructed using a mixture of USGS data, statistical yearbooks, regression estimates, conversion ratios, and interpolation. The authors should provide much more detail on regression models, predictors, goodness-of-fit, uncertainty propagation, and validation.**

Author Response: We thank the reviewer for this helpful comment. We agree that the previous description did not provide sufficient detail on how historical lime production was reconstructed, particularly where different data sources and reconstruction methods were combined.

To improve clarity, we have restructured the manuscript so that the methodological framework is now presented before the activity data description. The reconstruction method is described in Section 2.3.1, followed by the corresponding data sources and country-specific implementation details. This change helps readers first understand the logic of the model and then see how the required input data were compiled.

We have also substantially revised the lime-production data description. In the revised manuscript, historical lime production is reconstructed using a source-prioritized approach. Directly reported statistics from USGS, national statistical yearbooks, historical literature, and industry reports are used wherever available. For Japan, Brazil, France, Canada, Germany, and

the former Soviet Union, historical lime production data were compiled directly from national statistical yearbooks or historical statistical sources. Therefore, the previous regression-based extrapolation for countries such as Germany is no longer used. For Australia, Italy, and the United Kingdom, lime production was reconstructed from limestone or limestone/dolomite data using country-specific allocation ratios and lime-yield conversion coefficients. Missing Russian records were filled by linear interpolation, and Russian production during 1959–1991 was estimated from Soviet Union lime production using Russia's historical production share.

Regression models are now only used where direct records were unavailable and sectoral activity constraints were necessary, particularly in the reconstruction of China's 1950–1985 lime production. In this case, steel, calcium carbide, alumina production, and completed building floor area were used as activity indicators for sectoral lime demand, with historical sectoral-use information used as constraints. The fitted parameters, reconstructed results, goodness-of-fit diagnostics, uncertainty assumptions, and uncertainty distributions are now provided in Supplementary Table SI-2 Data1.

We have further clarified how uncertainty was propagated. Source-specific uncertainty ranges were assigned to reported statistics, conversion-derived estimates, regression-derived estimates, and interpolated values, and these uncertainties were propagated through the Monte Carlo framework described in Section 2.4. Validation and consistency checks were performed using overlapping-year comparisons, consistency with independent sectoral activity indicators, and comparison with reported historical production ranges where available. These revisions provide a more transparent and reproducible basis for the historical lime-production dataset.

The revised text in the Abstract now reads:

"2.3 Data Sources and Processing

Based on the published 1930-2020 lime carbon uptake dataset established by (Bing et al., 2023), we recalibrated China's lime production data for 1930-1995, expanded the lime-related basic activity data to nine additional countries, including the United Kingdom, France, Germany, Italy, Japan, Australia, Brazil, Russia, and Canada, and updated the dataset to 2024.

For the lime-related basic activity data, USGS statistics were used as the primary data backbone where available, followed by national statistical yearbooks, industry association reports, regression-based estimates, conversion-coefficient estimates, and linear interpolation

for short data gaps. Where multiple sources overlapped, we systematically compared statistical units, product definitions, accounting boundaries, and year-to-year changes to identify missing observations, source inconsistencies, and implausible data changes. We acknowledge that combining activity data from different sources increases the complexity of uncertainty, especially because statistical boundaries, reporting practices, and data quality may differ among sources and periods.

2.3.1. Lime Production Data

Before continuous USGS coverage, country-specific historical sources and transparent proxy methods were used. Japanese limestone shipments to lime manufacture reported by Shimanishi (2004) were converted to lime output using 1.8 t limestone per tonne of lime and checked against overlapping industry statistics. Brazilian lime production for 1940–1964 was taken from the IBGE Physical Production Tables (Produção de cal; IBGE, 2026), while 1930–1939 values are explicitly identified as autoregressive estimates. French, Canadian, German, and former Soviet records were compiled from national statistical yearbooks, with Canadian publications cited by their historical publication years (Dominion Bureau of Statistics, various years). Australian lime production was converted from limestone allocated to lime manufacture using a limestone-to-lime conversion coefficient of 0.24 (ABS, 2026). For Italy, a mean limestone-to-lime coefficient of 0.24 was derived from years reporting both inputs and outputs. For the United Kingdom, fixed 1949-based allocation ratios of 0.161 for limestone and dolomite and 0.049 for chalk, together with a lime-yield coefficient of 0.225, were used for years lacking direct observations (Ordnance Survey, 1957). Short gaps in Russian records were linearly interpolated; for 1959–1991, Russian output was estimated as 0.54 of reported Soviet lime production (Upravlenie S. U. T. statisticheskoe, 1961). All source classifications, equations, diagnostics, and uncertainty assumptions are documented in Supplementary Table SI-2, Data 1.

To illustrate the source harmonization procedure, China's 1930–1995 lime-production series was reconstructed by linking three consecutive source segments: a limestone-proxy reconstruction for 1930–1949, a sectoral activity-based reconstruction for 1950–1985, and directly reported or association-based statistics for 1986–1995.

For 1930–1949, continuous national lime-production statistics were unavailable for China,

particularly for 1930–1948. We therefore reconstructed annual lime production using limestone production as the primary proxy. Limestone production for mainland China and Northeast China was obtained from Guan (2007), which reports annual limestone production for 1912–1949. Observed lime-production records for Northeast China from Manshu Kojo Tokei Sokuho, Showa 15 (Kotoku 7) were used to calibrate the conversion relationship between limestone and lime production, yielding a limestone-to-lime conversion coefficient of 0.21. The calibrated relationship was then applied separately to mainland China and Northeast China, and the two regional estimates were combined to derive national lime production for 1930–1949.

For 1950–1985, we adopted a sectoral reconstruction approach constrained by activity data and historical evidence. Based on the sectoral lime-use shares reported by Liu (2018), total lime demand was allocated to the construction, iron and steel, calcium carbide, and alumina sectors. Steel, calcium carbide, and alumina production were used as activity indicators for their respective sectors, and through-origin proportional regression models were developed to estimate missing sectoral lime consumption. Construction lime demand was reconstructed using completed building floor area from Cao et al. (2019) as the activity proxy and constrained by historical information on lime production and application structure reported by Liao (1995), based on statistics from the China Lime Association.

For 1986–1995, lime production data were obtained from Liao (1995), based on China Lime Association statistics, and the China Building Materials Statistical Yearbook. Before merging the three source segments into a continuous historical series, statistical boundaries were harmonized through overlapping-year comparisons, consistency checks against sectoral activity indicators, and calibration of conversion coefficients where necessary to ensure temporal continuity and comparability. The harmonized series was subsequently linked with later statistical records to produce a continuous national dataset. All fitted parameters, reconstructed results, model diagnostics, and uncertainty distributions are documented in Supplementary Table SI-2 Data1.

This example illustrates how historical datasets with different statistical boundaries, reporting practices, and data availability were harmonized into a temporally consistent activity dataset before subsequent emission estimation and uncertainty analysis. To reflect the additional uncertainty introduced by source integration, source-specific uncertainty ranges

were assigned to reported statistics, regression-derived estimates, conversion-derived values, and interpolated data, and propagated through the Monte Carlo analysis (Section 2.4)."

"2.4 Uncertainty Assessment

Uncertainty from historical activity-data reconstruction was also considered in the Monte Carlo analysis. For directly reported statistics, uncertainty ranges were assigned according to source reliability and data completeness. For regression-based estimates, the standard errors reported by the calibrated regression models were used to characterize the uncertainty of reconstructed lime production. For conversion-ratio and interpolated estimates, uncertainty ranges were assigned based on the uncertainty settings of the corresponding activity data and conversion parameters in Supplementary Table SI-3. These activity-data uncertainty terms were sampled together with emission factors, CaO contents, material-use shares, carbonation ratios, and other parameters in the 10,000-iteration Monte Carlo simulation."

2. Including BFS is an important contribution, according to the authors, but the system boundary requires clearer justification. The authors should explain how the lime-derived CaO fraction in BFS is determined.

Author Response: We thank the reviewer for this important comment. We have revised Sects. 2.2 and 2.2.2 to clarify both the system boundary and the calculation of the lime-derived CaO fraction in BFS.

Building on the Material Flow Analysis framework of Liu et al. (2018a), the system boundary is defined according to the origin of CaO rather than the subsequent use of the slag. For SS and BFS, only CaO originating from lime fluxes represented in the lime production activity data is attributed to the lime carbonation sink. CaO derived from iron ore, gangue minerals, limestone, dolomite, and other non-lime inputs is excluded.

Although some SS and BFS are subsequently used in cement or clinker production, the existing global cement carbonation datasets considered in this study do not account for the carbonation of lime-derived CaO contained in these metallurgical slags, particularly BFS. Therefore, including this fraction in the present study does not result in double counting.

We have also explained the determination of the lime-derived CaO fraction in BFS. An ironmaking burden mass-balance approach was applied. Quicklime consumption per tonne of pig iron was estimated using the total blast-furnace burden consumption, the proportions of

sinter, pellets, and lump ore in the burden, and quicklime consumption per tonne of sinter. The resulting quicklime input was converted to lime-derived CaO and divided by the total CaO contained in the generated BFS, calculated from the BFS generation rate and its CaO content. The country- and period-specific parameters are provided in Supplementary Table SI-3 Data15.

The revised text in the Abstract now reads:

2.2 Line 207-215

"The system boundary is defined according to the origin of CaO rather than the subsequent use of the slag. For SS and BFS, only the CaO originating from lime fluxes represented in the lime production activity data is attributed to the lime carbonation sink. CaO derived from iron ore, gangue minerals, limestone, dolomite, and other non-lime inputs is excluded. Although some SS and BFS are subsequently used in cement or clinker production, existing cement carbonation inventories do not account for the carbonation of lime-derived CaO contained in these metallurgical slags, particularly BFS. Therefore, including this lime-derived CaO in the present study does not overlap with existing cement carbonation estimates."

2.2.2 Line 371-378

"The lime-derived CaO fraction in BFS was determined using an ironmaking burden mass-balance approach. First, quicklime consumption per tonne of pig iron was estimated from the total blast-furnace burden consumption per tonne of pig iron, the shares of sinter, pellets, and lump ore in the burden, and quicklime consumption per tonne of sinter. The resulting quicklime input was converted to lime-derived CaO and divided by the total CaO contained in generated BFS, which was calculated from the BFS generation rate and BFS CaO content. Country- and period-specific parameters used for this calculation are provided in Supplementary Table SI-3 Data15."

3. The manuscript assumes that materials such as LSS, CS, and LKD can fully carbonate within one year. This assumption directly affects the estimated annual CO_2uptake and may lead to overestimation if carbonation is actually slower.

Author Response: We thank the reviewer for this important comment. We agree that the original treatment and the phrase "complete carbonation within one year" could overstate the carbonation of these materials. The revised framework no longer assumes that LSS, CS, or LKD fully carbonates within one year.

LSS is now treated as a compacted engineered soil and represented by a time-dependent slab model. Its effective CaO conversion fraction is calculated as $\gamma_m R_m(a)$, where γ_m is the material-specific ultimate CaO conversion fraction and $R_m(a)$ describes progression toward that extent at material age a. Therefore, $R_m(a) = 1$ means only that the pathway-specific ultimate extent has been reached; it does not mean that all CaO has been converted unless $\gamma_m = 1$.

For LKD and CS, together with LM and RM, the evaluation age is fixed at one year, but the effective conversion fraction is not set to 1. Instead, the model uses the literature-reported, one-year pile-average CaO conversion fraction and its reported range, which are sampled directly in the Monte Carlo analysis. Only PCC and SUG are assigned an effective conversion fraction of 1 because carbonation is integral to their production processes.

All affected annual uptake, cumulative uptake, material contributions, and uncertainty estimates were recalculated using these revised treatments. This removes the assumption of complete one-year carbonation and reduces the risk of overestimating uptake from LSS, CS, and LKD.

Revised manuscript text (Lines 230–247 and 310–316):

"For PCC and SUG, carbonation is integral to production and Φ_m is set to 1 (Wang and Shen, 2002). For LSS, MOR, SS, and BFS, γ_m is the material-specific ultimate CaO conversion fraction and $R_m(a)$ is the cumulative fraction of that ultimate extent reached at material age a. Thus, $\gamma_m R_m(a)$ represents the cumulative proportion of CaO converted by that age. $R_m(a) = 1$ indicates that the pathway-specific ultimate extent has been reached, but does not imply conversion of all CaO unless $\gamma_m = 1$. For LKD, LM, RM, and CS, the evaluation age is fixed at one year and Φ_m is represented by the literature-reported pile-average CaO conversion fraction reached after one year. The reported average values and ranges are used directly in the Monte Carlo analysis."

"LSS and MOR use the slab model (Fig. 2a). The layered-stockpile representation for LKD, LM, RM, and CS (Fig. 2b) explains rapid carbonation near the exposed surface and lower conversion at greater depths because of restricted CO_2 transport and successive covering by newly deposited material."

"Due to its fine particle size and high specific surface area, LKD can undergo rapid carbonation during stockpiling, but its carbonation ratio was not assumed to be 100%; the parameter values are provided in Supplementary Table SI-3, Data 14 and Data 17."

4. The confidence interval terminology is inconsistent. The uncertainty section says that the authors used the 5th and 95th percentiles from the Monte Carlo simulations. This gives a 90% interval, not a 95% confidence interval. However, many results in the manuscript are reported as 95% CI.

Author Response: We thank the reviewer for identifying this inconsistency. The reference to the 5th and 95th percentiles in the original uncertainty section was incorrect. All reported 95%

confidence intervals were recalculated and verified using the 2.5th and 97.5th percentiles of the 10,000 Monte Carlo simulation results.

We have also removed the phrase "percentile method," because the selected percentiles directly define the interval bounds rather than constituting a separate method. Final estimates are reported as the medians of the simulated distributions, and the 2.5th and 97.5th percentiles define the lower and upper bounds of the central 95% confidence interval. The terminology and numerical intervals have been checked for consistency throughout the Abstract, Methods, Results, figures, and supplementary data.

Revised manuscript text (Lines 583–594):

"We identified 18 groups of factors affecting the estimation of process CO_2 emissions from lime production and carbonation uptake by lime-based materials, comprising 1,894 input parameters with specified statistical distributions (Supplementary Table SI-3, Data 1–17). Given the substantial uncertainty in these parameters, we applied the Monte Carlo method recommended by the 2006 IPCC Guidelines for National Greenhouse Gas Inventories. The statistical distributions of these variables were incorporated into the accounting model, and 10,000 Monte Carlo iterations were performed. Final estimates are reported as the median of the simulated distributions. The 2.5th and 97.5th percentiles define the lower and upper bounds, respectively, of the 95% confidence interval (CI).".

Comment on essd-2026-218, Anonymous Referee

This manuscript addresses an important gap in long-term carbon budget assessments and emission inventories by providing a relatively detailed quantification of process emissions and carbonation sinks associated with lime production. The study is potentially valuable and relevant to the ESSD community, as it contributes to improving our understanding of an often overlooked source of anthropogenic carbon emissions.

However, before publication, a number of important issues need to be addressed. In particular, the methodological framework is difficult to follow, and several key assumptions and parameters require much clearer justification and validation. My major concerns are summarized below.

Major Comments

1. Positioning relative to previous carbon inventories

In the Introduction, the authors should more clearly describe how lime production has been treated in previous carbon emission inventories and carbon budget assessments. Some inventories may only account for cement-related emissions, while others at least report emissions from lime production. The authors should provide a more comprehensive review and discussion of existing treatments in major inventories, including but not limited to

Global Carbon Budget (GCB): https://essd.copernicus.org/articles/18/3211/2026/

EDGAR: https://essd.copernicus.org/articles/16/2811/2024/

MEIC: https://link.springer.com/article/10.1007/s11430-023-1230-3

CDIAC: https://essd.copernicus.org/articles/13/1667/2021/essd-13-1667-2021.html

, and clearly explain what gap remains to be filled by the present study.

Author Response: We thank the reviewer for this helpful suggestion. We agree that the Introduction

should more clearly position the present study relative to existing carbon emission inventories and carbon budget assessments. We have revised the Introduction to explicitly discuss how major inventories treat lime production and related carbonate-process emissions, including CDIAC-FF, MEIC-global-CO_2, EDGAR, and the Global Carbon Budget. Specifically, we now clarify that CDIAC-FF and MEIC-global-CO2 mainly focus on fossil fuel combustion and cement production, whereas the Global Carbon Budget includes cement carbonation and only partial lime-production emissions, with non-global coverage. We also note that EDGAR provides a broader gridded greenhouse gas emission inventory, but does not provide a life-cycle accounting of lime-product carbonation uptake. Based on this comparison, we have clarified the remaining gap addressed by this study: the lack of a standardized, country-resolved, long-term dataset that couples lime process emissions with lime carbonation sink. This revision has been added to the Introduction.

The revised text in the Abstract now reads:

"Existing carbon emission inventories and global carbon budget assessments treat lime production and subsequent carbonation uptake inconsistently. CDIAC-FF provides long-term global and national CO_2 emission series for fossil fuel combustion, gas flaring, and cement manufacture, but does not include process emissions from lime production (Gilfillan and Marland, 2021). MEIC-global-CO_2 improves source-category resolution and incorporates subnational information for major emitters, but mainly focuses on fossil-fuel combustion and cement-related CO_2 emissions and does not provide a dedicated, globally consistent representation of lime-production emissions and lime-product carbonation uptake (Xu et al., 2024). EDGAR provides a comprehensive national and gridded greenhouse gas emission inventory covering multiple sectors and industrial processes. However, its framework is emissions-oriented and does not quantify CO_2 uptake resulting from the carbonation of lime products during their use and disposal stages (Crippa et al., 2024). The Global Carbon Budget has incorporated cement carbonation uptake as a negative flux within the fossil CO_2 component, but explicitly notes that lime-production emissions are not yet globally covered. Current estimates include lime-production emissions for China and the United States, whereas most non-Annex I countries and emissions from other Annex I countries before 1990 remain unaccounted for (Friedlingstein et al., 2025). Consequently, existing datasets generally cover

only part of the lime carbon cycle: some omit lime production, some focus primarily on carbonate decomposition associated with cement manufacture, and others include lime process emissions but do not couple them with the subsequent carbonation uptake of lime products. To our knowledge, a standardized, country-resolved, long-term dataset with a consistent system boundary that jointly quantifies lime process emissions and carbonation uptake is still lacking, limiting the comprehensive integration of lime-related carbon sources and sinks into global carbon budget assessments."

Please see line 65-88

2. Clarification of the "verified dataset" terminology

The manuscript repeatedly refers to a "verified dataset" and cites the authors' previous work. However, it remains unclear what "verified" specifically means in this context. What metrics were used for verification? What independent data sources were used? Why should readers consider these datasets to be more reliable than alternative sources? Based on my understanding, true verification of historical lime production statistics is challenging. Therefore, the authors should provide a much more explicit explanation of the verification procedure and the associated limitations.

Author Response: We thank the reviewer for this important comment. We agree that the term "verified dataset" was imprecise. In the original manuscript, we used this term because part of the dataset had been published previously; however, we recognize that publication itself does not constitute independent verification. We have therefore revised the terminology and now describe the dataset as a "source-prioritized and internally cross-checked activity dataset."

In the revised Section 2.3, we clarified that, "Based on the published 1930-2020 lime carbon uptake dataset established by Bing et al. (2023), we recalibrated China's lime production data for 1930-1995, expanded the lime-related basic activity data to nine additional countries, and updated the dataset to 2024." We also clarified the data-priority rule: "USGS statistics were used as the primary data backbone where available, followed by national statistical yearbooks, industry association reports, regression-based estimates, conversion-coefficient estimates, and linear interpolation for short data gaps. Where multiple sources overlapped, we systematically compared statistical units, product definitions, accounting boundaries, and year-to-year changes to identify missing observations, source inconsistencies, and implausible data changes."

We also explicitly acknowledge the limitation that "combining activity data from different sources increases uncertainty complexity, particularly because statistical boundaries, reporting practices, and data quality may differ among sources and periods."

3. Integration of multiple activity data sources

The activity data are compiled from numerous sources. I understand that no single data source exists for such a long historical period, making data integration necessary. However, different sources often have substantial differences in statistical boundaries, collection methodologies, reporting practices, and data quality. Ensuring temporal consistency and comparability is therefore a major challenge. Even within a single source such as USGS, data quality may vary considerably over time.

The manuscript currently does not adequately explain how these issues were addressed. I strongly recommend that the authors provide a concrete example illustrating the process of harmonizing and merging different data sources. In addition, uncertainties arising from data integration should be explicitly reflected in the uncertainty analysis.

Author Response: We thank the reviewer for this helpful and constructive suggestion. We agree that integrating multiple activity-data sources over a long historical period is a major methodological challenge, because different sources may differ in statistical boundaries, product definitions, reporting practices, collection methods, and data quality.

To address this concern, we revised Section 2.1 to clarify the general data-prioritization and harmonization procedure. The revised manuscript now states that USGS statistics were used as the primary data backbone where available, followed by national statistical yearbooks, industry association reports, regression-based estimates, conversion-coefficient estimates, and linear interpolation for short data gaps. Where multiple sources overlapped, we systematically compared statistical units, product definitions, accounting boundaries, and year-to-year changes to identify missing observations, source inconsistencies, and implausible data changes.

We also added a concrete example in Section 2.1.1 using the recalibration of China's lime production data for 1930-1995. The revised manuscript explains that this series was reconstructed by linking three consecutive source segments: a limestone-proxy reconstruction for 1930-1949, a sectoral activity-based reconstruction for 1950-1985, and directly reported or association-based

statistics for 1986-1995. All fitted parameters, reconstructed results, model diagnostics, and uncertainty distributions are reported in SI-2 Data1.

The added text is: "To illustrate the source harmonization procedure, China's 1930–1995 lime-production series was reconstructed by linking three consecutive source segments: a limestone-proxy reconstruction for 1930–1949, a sectoral activity-based reconstruction for 1950–1985, and directly reported or association-based statistics for 1986–1995.

For 1930–1949, continuous national lime-production statistics were unavailable for China, particularly for 1930–1948. We therefore reconstructed annual lime production using limestone production as the primary proxy. Limestone production for mainland China and Northeast China was obtained from Guan (2007), which reports annual limestone production for 1912–1949. Observed lime-production records for Northeast China from *Manshu Kojo Tokei Sokuho, Showa 15 (Kotoku 7)* were used to calibrate the conversion relationship between limestone and lime production. The calibrated relationship was then applied separately to mainland China and Northeast China, and the two regional estimates were combined to derive national lime production for 1930–1949.

For 1950–1985, we adopted a sectoral reconstruction approach constrained by activity data and historical evidence. Based on the sectoral lime-use shares reported by Liu (2018), total lime demand was allocated to the construction, iron and steel, calcium carbide, and alumina sectors. Steel, calcium carbide, and alumina production were used as activity indicators for their respective sectors, and through-origin proportional regression models were developed to estimate missing sectoral lime consumption. Construction lime demand was reconstructed using completed building floor area from Cao et al. (2019) as the activity proxy and constrained by historical information on lime production and application structure reported by Liao (1995), based on statistics from the China Lime Association.

For 1986–1995, lime production data were obtained from Liao (1995), based on China Lime Association statistics, and the *China Building Materials Statistical Yearbook*. Before merging the three source segments into a continuous historical series, statistical boundaries were harmonized through overlapping-year comparisons, consistency checks against sectoral activity indicators, and calibration of conversion coefficients where necessary to ensure temporal continuity and comparability. The harmonized series was subsequently linked with

later statistical records to produce a continuous national dataset. All fitted parameters, reconstructed results, model diagnostics, and uncertainty distributions are documented in Supplementary Table SI-2 Data1.

This example illustrates how historical datasets with different statistical boundaries, reporting practices, and data availability were harmonized into a temporally consistent activity dataset before subsequent emission estimation and uncertainty analysis. To reflect the additional uncertainty introduced by source integration, source-specific uncertainty ranges were assigned to reported statistics, regression-derived estimates, conversion-derived values, and interpolated data, and propagated through the Monte Carlo analysis (Section 2.4)."

Finally, we revised Section 2.4 to make explicit how uncertainty from data integration was incorporated into the Monte Carlo framework.

The added text is: "In the harmonization of multiple activity-data sources, differences among overlapping datasets, uncertainties associated with limestone-to-lime conversion, differences in statistical boundaries, and uncertainties arising from regression-based reconstruction were explicitly incorporated into the uncertainty ranges assigned to historical activity data. These activity-data uncertainties were then propagated together with emission factors, CaO contents, material-use shares, carbonation ratios, and other model parameters in the 10,000-iteration Monte Carlo simulation."

4. Improving the transparency of the methodological framework

The current methodology is difficult to follow because it combines multiple dimensions, including country, industrial sector, and material type. I suggest adding a summary table that clearly documents which data sources and models are used for each country, sector, and material category. Such a table could also include a qualitative uncertainty rating (e.g. A, B, C, and D). This would substantially improve the transparency and reproducibility of the dataset.

Author Response: We thank the reviewer for this constructive suggestion. We agree that the methodology spans multiple dimensions, including country, sector, and material type, and that a compact methodological summary would improve transparency and reproducibility.

In the revised Data Sources and Methodology section, we added a methodological framework

summary table (Table 1), which documents the main activity-data sources, model or harmonization approach, material category, sectoral coverage, and qualitative uncertainty rating for each accounting module. To further improve readability, we also added a material-flow-analysis framework diagram that visually links input activity data, source harmonization, lime production and allocation, sector-specific material pathways, process CO2 emissions, carbonation uptake, annual and cumulative outputs, and Monte Carlo uncertainty propagation.

We also define the qualitative uncertainty ratings in the text: A indicates directly reported and temporally consistent data with low parameter uncertainty; B indicates direct statistics supplemented by literature parameters or limited harmonization; C indicates mixed direct and reconstructed data requiring regression, conversion factors, or boundary adjustment; and D indicates sparse historical or residual estimates with high integration uncertainty. Together, the table and framework diagram provide a compact and transparent guide to the full data-source and parameter documentation provided in SI-2 and SI-3. please see line 514

514 Table 1. Summary of activity-data sources, accounting models, and qualitative uncertainty ratings used in the lime process-emission and carbonation-sink
515 framework.

Category	Scope	Main data/model approach	Qualitative uncertainty	Detailed source
Lime production activity data	China, 1930-1995	Segmented reconstruction linking limestone-proxy estimates (1930-1949), sectoral activity-based reconstruction (1950-1985), and reported/association statistics (1986-1995).	C-D before 1985; B after 1986	SI-2 Data1
	Nine additional countries before continuous USGS coverage	Priority use of national statistical yearbooks and historical sources; limestone/dolomite conversion ratios or interpolation used only where direct records are incomplete.	B-C for reported series; C-D for converted/interpolated series	SI-2 Data1
	All countries, 1959-2024 where available	USGS statistics harmonized with historical national records and checked for temporal continuity.	B	SI-2 Data1
Process-emission accounting	All countries and ROW	Country lime production allocated by sector and combined with lime-use ratios, localized CaO contents, and process-emission factors.	B-C	SI-3 Data1-7
Carbonation sink accounting	Construction materials (LSS, MOR)	Slab/pile carbonation models using material allocation, CaO content, thickness, conversion fraction, and carbonation-rate parameters.	B-C	SI-3 Data8-11
	Chemical materials (PCC, CS, SUG, LM)	Output/use rates, utilization ratios, CaO contents, conversion fractions, and carbonation-rate parameters.	B-C	SI-3 Data8-13
	Metallurgical materials (SS, BFS, RM)	Industrial output, slag/red mud generation rates, utilization ratios, lime-origin allocation, and time-varying carbonation parameters.	C-D	SI-3 Data8-14

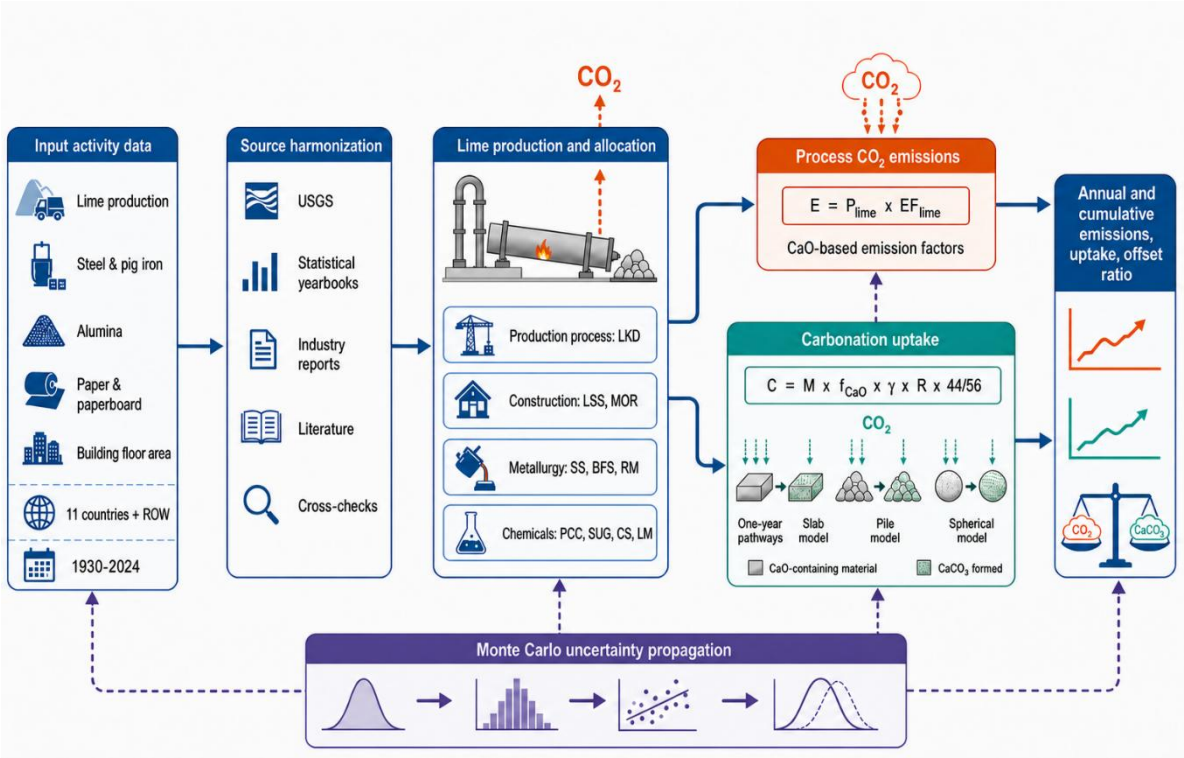

Fig. 1. Methodological framework for lime process-emission and carbonation-uptake accounting.

5. Potential underestimation of uncertainty

The uncertainty ranges shown in Figure 5 appear unrealistically small given the complexity of the underlying data and assumptions. This may partly result from overly narrow probability distributions assigned to key parameters, but it also appears that several important sources of uncertainty have not been considered.

For example, different data sources likely have different levels of reliability; original data and gap-filled data should not be assigned identical uncertainties; and uncertainty levels are expected to vary substantially across countries. These factors do not appear to be adequately represented in the current uncertainty framework. The authors should revisit the uncertainty analysis and provide stronger justification for the reported confidence intervals. It would be good if country-level uncertainties are shown in Fig. 6.

Author Response: We thank the reviewer for this constructive comment. We first wish to clarify a figure-numbering issue. Figure 5 in the original manuscript, now Fig. 6 in the revised manuscript, presents the delayed effects of lime carbonation uptake, including current-year uptake,

historical uptake, and annual uptake disaggregated by production year. This figure does not contain confidence intervals, uncertainty bands, or any other representation of uncertainty. Therefore, the reviewer's specific concern that the uncertainty ranges in the original Fig. 5 appear unrealistically small does not apply to that figure.

Nevertheless, we agree with the reviewer's broader concern that the uncertainty assessment should distinguish among data sources, reconstruction methods, countries, and model variables. We have therefore strengthened the uncertainty framework in Sect. 2.4. Source-specific uncertainty ranges were assigned according to data reliability, record completeness, and reconstruction method. Directly reported statistics were assigned narrower uncertainty ranges than reconstructed data. Regression-derived estimates were characterized using model-fitting errors, calibration diagnostics, and goodness-of-fit information. Conversion-derived and interpolated estimates were assigned ranges based on the uncertainties of the corresponding activity data and conversion parameters. Differences among overlapping datasets, limestone-to-lime conversion uncertainty, statistical-boundary inconsistencies, and uncertainty introduced by historical reconstruction were incorporated into the uncertainty ranges of the relevant activity data and propagated through the 10,000-iteration Monte Carlo analysis. This treatment ensures that directly reported and gap-filled data are not assigned identical uncertainties and allows uncertainty to vary among countries and historical periods.

Following the reviewer's recommendation, we also revised the original Fig. 6, now Fig. 7, to explicitly present country-level uncertainty. The revised Fig. 7 shows the median estimates and 95% confidence intervals for process emissions, carbonation uptake, and net emissions for each of the 11 countries. The corresponding results are discussed in Sect. 3.6. In 2024, the relative half-widths of the process-emission confidence intervals ranged from 4.5% to 4.8%, reflecting the greater availability of directly reported recent activity data. Carbonation-uptake uncertainty was substantially larger, with relative half-widths ranging from 19.2% in Russia to 36.2% in the United Kingdom, because these estimates additionally depend on material allocation, CaO content, carbonation behaviour, utilization pathways, and time-lag parameters. Net-emission uncertainty becomes particularly pronounced when process emissions and carbonation uptake are similar in magnitude.

At the global cumulative level, process emissions during 1930–2024 were estimated at 11.3 Gt

CO_2 (95% CI: 10.7–11.9 Gt CO_2), whereas cumulative carbonation uptake was 4.8 Gt CO_2 (95% CI: 3.9–5.8 Gt CO_2). The relative half-widths of these intervals were 5.1% and 19.5%, respectively, demonstrating that carbonation uptake is considerably more uncertain than process emissions. These revisions provide a more transparent justification for the reported confidence intervals and explicitly represent differences between directly reported and reconstructed data, among countries, and between emission and uptake processes.

Please see Sect. 2.4, Sect. 3.6, and revised Fig. 7.

6. Suspicious temporal signals potentially caused by data inconsistencies

Related to the previous concern, Figure 6 exhibits several unusual temporal features that appear difficult to explain physically. Examples include the sharp decline in Italy around 1980, the sharp decline in Australia around 2010, and the abrupt increase in Brazil around 1980. These features appear unnatural and may be artifacts introduced by inconsistencies among data sources.

The authors should carefully investigate these anomalies, improve the harmonization of different data sources where necessary, or provide convincing explanations for the observed patterns.

Author Response: We thank the reviewer for identifying these unusual temporal signals. We agree that abrupt changes in country-level trajectories require careful examination because they may reflect actual changes in lime production, changes in statistical coverage or reporting practices, or inconsistencies introduced by combining different data sources.

We first clarify that Fig. 6 in the original manuscript corresponds to Fig. 7 in the revised manuscript. We re-examined the underlying activity data and calculations for Italy, Australia, and Brazil. The identified discontinuities originate from variations already present in the reported lime-production data rather than from the carbonation model, interpolation, or transitions between different data sources. Lime-production data for all three countries during the periods in question were obtained from continuous USGS statistical series.

More specifically, the pronounced decline in Italy occurred mainly between 1970 and 1972 rather than around 1980. Modeled process emissions decreased from 4.2 Mt CO_2 yr^{-1} in 1970 to 1.5 Mt CO_2 yr^{-1} in 1972, following the corresponding decline in reported lime production. For Australia, process emissions increased from 1.2 Mt CO_2 yr^{-1} in 2009 to 1.8 Mt CO_2 yr^{-1} in 2011 and subsequently decreased to 1.6 Mt CO_2 yr^{-1} in 2012. For Brazil, the apparent increase occurred

mainly between 1976 and 1977, when process emissions increased from 1.5 to 3.3 Mt CO_2 yr^{-1}. This step is also present in the underlying USGS lime-production series.

Because no change in data source or reconstruction method occurred at these transition points, we retained the reported observations rather than applying interpolation or artificial smoothing. Such smoothing could replace reported values and conceal genuine short-term variability. Nevertheless, we acknowledge that a continuous statistical source does not completely exclude possible changes in survey coverage, statistical boundaries, or reporting practices. These short-term fluctuations should therefore be interpreted cautiously and should not be attributed to specific technological, economic, or policy drivers without independent supporting evidence. The uncertainty associated with the reported activity data has been propagated through the Monte Carlo analysis.

Minor Comments

1. Lines 54–57: The statement appears to be incorrect. It is unlikely that 65% of global emissions originate from limestone calcination. Please verify the calculation and revise the text accordingly.

Author Response: We thank the reviewer for pointing this out. We agree that the original wording was imprecise and could be incorrectly interpreted as stating that 65% of total global CO_2 emissions originate from limestone calcination. This was not our intended meaning. The percentage refers to the emission structure within lime production, where carbonate calcination generally accounts for about two-thirds of lime-production-related CO_2 emissions, while fuel combustion and electricity use account for much of the remaining share. We have therefore revised the sentence to explicitly specify the lime-production boundary.

We also clarified the inventory boundary to avoid mixing IPPU process emissions with energy-related emissions. According to the IPCC Guidelines, process emissions from carbonate calcination are reported under IPPU, whereas emissions from fuel combustion should be reported under the Energy sector to avoid double counting. Accordingly, the revised text now states that this study focuses on process emissions from lime production and the subsequent carbonation uptake of lime products, while energy-related emissions are not included in the accounting boundary.

Revised text:

"Characterized by high carbon intensity, lime production is an important source of process CO_2 emissions in the Industrial Processes and Product Use (IPPU) sector, particularly within the mineral industry category (IPCC, 2006). In lime production, CO_2 emissions arise mainly from carbonate calcination, with additional contributions from kiln fuel combustion and electricity use. Previous studies indicate that calcination generally accounts for about two-thirds of the total CO_2 emissions associated with lime production, while energy-related emissions account for much of the remaining share (Han et al., 2022; Laveglia et al., 2024a). Following IPCC inventory boundaries, the present study focuses on process emissions from lime production and the subsequent carbonation uptake of lime products, whereas energy-related emissions are not included to avoid overlap with the energy sector."

2. Line 180: The manuscript states that a linear regression model was used. However, the rationale for selecting this model, the data used for calibration, and the validation procedure are not adequately described.

Author Response: We thank the reviewer for this helpful comment. We agree that the original manuscript did not provide sufficient detail on the regression-based reconstruction. In the revised manuscript, we have clarified the rationale for using multiple linear regression, the calibration data, the predictor variables, and the validation procedure.

Specifically, multiple linear regression was selected because historical lime demand is closely linked to several major lime-consuming industrial sectors, while the available reported lime-production records for the early period are limited. This model structure therefore provides an interpretable relationship between lime production and relevant industrial drivers. The models for Italy and Germany were calibrated using overlapping years for which both reported lime production and predictor variables were available. For Germany, national crude steel and cement production were used as key independent variables. For Italy, crude steel and alumina production were used. These predictors were selected because metallurgy, cement-related construction activity, and alumina production are important lime-consuming sectors.

We also added a clearer description of the validation procedure. Model reliability was evaluated using coefficient signs, standard errors, p values, goodness-of-fit statistics (R^2), and residual diagnostics. In addition, the reconstructed series were checked for temporal continuity and consistency with adjacent reported records and with long-term trends in downstream industrial

indicators. The regression coefficients and diagnostic outputs are provided in Supplementary Table SI-2 Data1.

Revised text:

"For Italy and Germany, production figures were reconstructed using multiple linear regression models. Multiple linear regression was selected because historical lime demand is closely associated with major lime-consuming industrial sectors, while the available calibration records are limited and require an interpretable model structure. The models were calibrated over overlapping years for which both reported lime production and predictor variables were available. Specifically, the German model used national crude steel and cement production as key independent variables, whereas the Italian model used crude steel and alumina production. These predictors were selected because metallurgy, cement-related construction activity, and alumina production are major lime-consuming sectors. Model reliability was evaluated using coefficient signs, standard errors, p values, goodness-of-fit statistics (R^2), and residual diagnostics. The reconstructed series were further checked for temporal continuity and consistency with adjacent reported records and long-term trends in relevant downstream industrial indicators. The corresponding regression coefficients and diagnostic outputs are provided in Supplementary Table SI-2 Data1."

3. Line 220: The authors state that they "calculated" emission factors. Additional information should be provided regarding the methodology and underlying assumptions.

Author Response: We thank the reviewer for this helpful comment. We agree that the original description of how the carbon emission factors (CEFs) were calculated was too brief. In the revised manuscript, we have added a more explicit explanation of the calculation procedure and underlying assumptions.

The CEFs were calculated following the principle of the Tier 2 approach for lime production in the 2006 IPCC Guidelines, in which process CO_2 emissions are derived from the stoichiometric relationship between CO_2 and carbonate-derived CaO or CaO·MgO in lime products. In our study, sector-specific CaO contents were compiled for lime used in major end-use sectors, including metallurgy, construction, and the chemical industry. Because lime quality and CaO content differ among end-use sectors, these sectors were used as practical proxies for differences in lime product composition. Sector-specific emission factors were first calculated by multiplying the CaO or CaO·MgO content of lime used in each sector by the corresponding IPCC stoichiometric ratio.

Country-specific average CEFs were then calculated as weighted averages of sector-specific emission factors according to the lime-use shares of each sector.

This sector-weighted approach is consistent with the IPCC Tier 2 principle of deriving emission factors from lime composition, while allowing country-specific differences in lime-use structure and product quality to be reflected. We have also clarified that these CEFs refer only to process emissions from lime production and do not include fuel-combustion or electricity-related emissions. Detailed CaO contents, sectoral lime-use shares, assumptions, and literature sources are provided in Supplementary Table SI-3 Data 1.

Revised text:

"Carbon emission factors (CEFs) were calculated following the Tier 2 approach for lime production in the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006). Under this approach, process CO_2 emissions are derived from the stoichiometric relationship between CO_2 and carbonate-derived CaO or CaO·MgO in lime products. In this study, sector-specific CaO contents were compiled for lime used in major end-use sectors, including metallurgy, construction, and the chemical industry. Because lime quality and CaO content differ among end-use sectors, these sectoral categories were used as practical proxies for product composition. Sector-specific emission factors were first derived by multiplying the CaO or CaO·MgO content of lime used in each sector by the corresponding IPCC stoichiometric ratio. Country-specific average CEFs were then calculated as weighted averages of sector-specific emission factors according to the lime-use shares of each sector. This approach is consistent with the IPCC Tier 2 principle of deriving emission factors from lime composition, while allowing national differences in lime-use structure and product quality to be reflected. Detailed CaO contents, sectoral lime-use shares, assumptions, and literature sources are provided in Supplementary Table SI-3 Data 1." L182-196

4. The manuscript frequently reports values to two decimal places. Given the substantial uncertainties involved, one decimal place would likely be sufficient in most cases.

Author Response: We thank the reviewer for this helpful suggestion. We agree that reporting most results to two decimal places may imply a level of precision that is not warranted given the uncertainties associated with historical activity data, emission factors, material-use shares, and carbonation parameters. Following the reviewer's recommendation, we have revised the manuscript and rounded most reported values to one decimal place where appropriate. Values that require higher

precision for methodological clarity, such as key ratios or parameters listed in the Supplement, have been retained with sufficient significant digits.

5. Figure 2: The legend appears to contain a typo. “Progress emissions” may not be the intended term.

Author Response: We thank the reviewer for noting this typo. The intended term was “Process emissions”, referring to CO_2 emissions from carbonate decomposition during lime production. We have corrected “Progress emissions” to “Process emissions” in the legend of Fig. 2.

6. Please maintain a consistent color scheme throughout the manuscript. For example, Figures 2 and 3 appear to represent the same regional classification but use different colors, which makes it difficult for readers to follow the discussion.

Author Response: We thank the reviewer for this useful suggestion. We agree that consistent color coding is important for improving the readability and comparability of figures. We have revised the figures to ensure that the same regional categories are represented by the same colors throughout the manuscript, including Figs. 2 and 3. The figure legends have also been checked and updated accordingly to maintain consistency.