



1 **Global and National CO₂ Emission from Lime Production Process and Carbonation sink**
2 **from 1930 to 2024**

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16 **Abstract**

17 Accurate quantification of both lime process emissions and carbonation sink is essential for the
18 Global Carbon Budget (GCB). By extending temporal coverage (1930–2024), refining spatial
19 resolution (11 major lime-producing countries), and expanding system boundaries (adding
20 Blast Furnace Slag, BFS), this study constructs the first standardized dataset of lime CO₂
21 process emissions and carbonation sink covering 81.09% of global lime production. We
22 estimate cumulative global lime process emissions are 15.29 Gt CO₂ (95% CI: 13.81–16.79 Gt
23 CO₂), with the construction and metallurgical sectors serving as primary sources, contributing
24 5.80 Gt CO₂ (37.92%) and 5.04 Gt CO₂ (32.95%), respectively. During the same period,
25 cumulative lime carbonation sink reached 7.33 Gt CO₂ (95% CI: 5.95–8.88 Gt CO₂), achieving
26 a carbon offset ratio (cement carbonation sink to process emission) of 47.65%, which is 8.32%
27 increase compared with Bing et al. (2023). The lime carbonation sink in 2024 accounted for
28 approximately 1.5%–2% of the global terrestrial carbon sink in 2023. China is the main
29 contributor, with cumulative emissions of 8.89 Gt CO₂ (58.13% of the global total) and
30 cumulative carbonation sink of 4.21 Gt CO₂ (57.45% globally) from 1930 to 2024.
31 Lime-stabilized soil (LSS, 36.53%), mortar (MOR, 18.66%), steel slag (SS, 17.73%), and
32 blast furnace slag (BFS, 12.83%) were the primary carbon uptake materials, collectively
33 accounting for 85.75% of the total carbonation sink. Significant regional disparities were



34 pronounced: developed countries (e.g., those in Europe, the United States, Japan, and Australia)
35 have already peaked in lime process carbon emissions, with net emissions gradually
36 approaching zero. In contrast, developing countries such as China and Brazil continue to exhibit
37 growth in both emissions and carbonation sink. Although their carbon-offset levels exceed 50%,
38 they face substantial pressure to reduce total emissions. This dataset provides critical data for
39 incorporating the lime carbonation sink into the Global Carbon Budget. It also contributes to
40 optimizing global carbon modelling and regional carbon-neutrality pathways. The dataset is
41 archived on Zendo <https://doi.org/10.5281/zenodo.18616060> (Bing et al., 2026)

42 1. Introduction

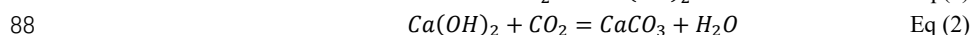
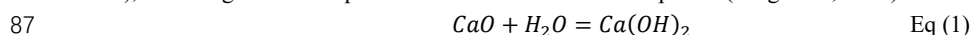
43 Lime serves as an indispensable feedstock in numerous industrial processes, playing a
44 pivotal role in sectors such as steelmaking, construction materials, chemical engineering, and
45 wastewater treatment (Revuelta, 2021). Over the past two decades, driven by urbanization,
46 industrialization, and the rapid development of large-scale infrastructure, global lime
47 production has surged by 3.4-fold, increasing from 121 Mt in 2004 to 420 Mt in 2024 (USGS,
48 2026a). Projections indicate that two-thirds of the global population will reside in urban areas
49 by 2050 (UN-Habitat, 2022). This ongoing urbanization process is expected to further drive the
50 demand for housing, transportation infrastructure, and environmental sanitation facilities,
51 thereby leading to sustained increase in the consumption of lime as a fundamental material.

52 Characterized by high carbon intensity, the lime industry is one of the major sources of
53 greenhouse gas emissions in the Industrial Processes and Product Use (IPPU) sector, ranking
54 third after the cement and steel industries (IPCC, 2006). Approximately 65% of global CO₂
55 emissions originates from the calcination of limestone (Han et al., 2022). While the remaining
56 35% of CO₂ emissions is attributed to fossil fuel combustion and electricity consumption
57 (Laveglia et al., 2024a). Unlike the energy sector, the CO₂ emissions in lime production process
58 are dictated by chemical reactions and cannot be fully eliminated through fuel switching or
59 energy efficiency improvements. Consequently, the lime industry is a quintessential "hard-to-
60 abate" industrial sector (Davis et al., 2018). Quantifying the CO₂ emissions from lime
61 production process is of great significance for CO₂ management and global carbon budget.

62 However, accurately quantifying process emissions from the global lime industry remains
63 a significant challenge, primarily due to data gaps and inconsistent statistical frameworks. First,
64 large volumes of lime are produced for internal use by steel, sugar, and chemical enterprises,
65 which does not enter commercial circulation. The prevalence of this "captive production" leads
66 to omit much of the output and severely underestimate the official activity data (USGS, 2026a).
67 Second, significant disparities in kiln technology exist across regions. The coexistence of
68 traditional vertical kilns in developing countries and high-efficiency rotary kilns in developed
69 countries leads to substantial fluctuations in emission factors (European Commission, 2013).
70 As Robbie Andrew noted in studies of global process emissions, the accuracy of emission
71 factors depends heavily on precise raw material composition (Andrew, 2019). Ignoring these
72 variations results in significant process emissions estimation biases. Furthermore, overlapping
73 statistical boundaries between lime and cement production often lead to misclassification or
74 omissions in historical data, which making it is difficult for existing databases to reflect the true



75 emission trajectory of lime industry (Andrew, 2019). According to the China Building Materials
76 Federation, total CO₂ emissions from China's building materials industry reached 1.48 billion
77 tons in 2020, representing 14.9% of the national total, with the lime sector contributing
78 approximately 8.1% of the industry's emissions (CBMEMA, 2026). Globally, the lime industry
79 contributes approximately 1% of total anthropogenic CO₂ emissions (Campo et al., 2021;
80 Bridlington et al., 2022). Notably, lime is extensively utilized in iron and steel, construction,
81 chemical engineering, agriculture, and environmental remediation (European Commission,
82 2013). During their lifecycle, lime-based materials undergo carbonation with atmospheric CO₂
83 via Eqs. (1) and (2), thereby sequestering a portion of the process CO₂ released by their
84 production (Simoni et al., 2022). Previous study revealed that between 1930 and 2020, global
85 lime materials cumulatively sequestered approximately 144.47 Mt C (95% CI: 101.62–196.11
86 Mt C), offsetting 38.83% of process emissions over the same period (Bing et al., 2023).



89 Carbonation represents a pivotal pathway for achieving net-zero emissions in the lime
90 industry. Although previous studies have indicated that CO₂ emissions from lime production
91 can be mitigated by substituting fossil fuels with residue-derived fuels (Zementwerke eV,
92 2021), replacing calcium carbonate calcination with electrochemical (Ellis et al., 2020), and
93 chemical decarbonization methods (Hanein et al., 2021), these approaches are currently
94 constrained by high costs and technical complexities and making challenges in large-scale
95 deployment (Simoni et al., 2022). Consequently, carbon sequestration via lime carbonation
96 plays an alternative role in the decarbonation of the sector, which creating an urgent need to
97 accurately quantify its contribution. Currently, China's Action Plan for Carbon Peaking by 2030
98 in the Building Materials Industry promotes utilization of industrial solid wastes, such as steel
99 slag and carbide slag (MIIT, 2026). Similarly, the European Lime Association explicitly
100 identifies Carbon Capture, Utilization, and Storage (CCUS) and natural carbonation as critical
101 measures, aiming to achieve CO₂ negative emissions by 2050 through technological upgrades
102 (EuLA, 2026a). The National Lime Association (NLA, 2023) in the United States also released
103 a roadmap targeting carbon neutrality by 2050, emphasizing the significance of the carbon sink
104 function of lime products. At present, the Global Carbon Budget (GCB) has incorporated
105 cement carbonation sink into the anthropogenic CO₂ sink (Friedlingstein et al., 2022). However,
106 the lime carbonation sink remains excluded. For context, carbon uptake by cement materials in
107 2021 accounted for approximately 8.23% of the global mean land carbon sink from 2010–2020
108 (Huang et al., 2023; Friedlingstein et al., 2020), whereas lime materials sequestered an amount
109 equivalent to approximately 1.09% of the mean land sink from 2012–2020 (Bing et al., 2023).
110 Incorporating the lime carbonation sink into the GCB would provide a more accurate reflection
111 of the global carbon cycle and optimize regional carbon budget allocations. Therefore, an
112 accurate accounting of lime carbonation sink is indispensable.

113 Previous efforts by our team provided a preliminary accounting of lime-based carbonation
114 sink in China and the United States (Bing et al., 2023). However, the study treated the rest of
115 the world as a homogeneous entity. It overlooks the significant divergence in process carbon
116 emissions and sequestration coefficients caused by variations in production technologies, raw
117 material compositions, and application industries across different nations. In particular, the



118 carbon sequestration of metallurgical wastes has been historically underestimated. For instance,
119 pig iron production requires the addition of quicklime and limestone as metallurgical fluxes
120 (Yang et al., 2024). The resulting slag, with calcium oxide (CaO) content ranging from 30% to
121 50% (Ren et al., 2021), presents substantial carbon sequestration (Gomari et al., 2024), which
122 is also one of key improvements of this study. Furthermore, global production rates of iron and
123 steel slag have generally declined due to optimized furnace lining materials and refined blowing
124 processes (Naito et al., 2015). Concurrently, the resource utilization of these materials has
125 continued to improve. On a global scale, blast furnace slag (BFS) and steel slag (SS) are widely
126 repurposed for cement production, road construction, building materials, and fertilizers (Heraiz
127 et al., 2025). However, significant disparities exist at the national level. The developed nations
128 have achieved comprehensive utilization rates of steel slag exceeding 90%, while the rate in
129 China remains approximately 20% (Gao et al., 2023). By accounting for these technological
130 and regional discrepancies, this study provides a more robust quantification of the global lime
131 process emissions and carbonation sink.

132 To address the challenges of global lime process emissions accounting and facilitate the
133 integration of lime carbonation sink into the GCB dataset, this study significantly expands the
134 data framework through the following improvements. (1) Spatiotemporal Reconstruction and
135 Verification of Activity Data. To bridge the statistical gaps caused by "captive production," the
136 accounting scope was extended to 11 major lime-producing countries, covering 87.62% of
137 global output. The United States Geological Survey (USGS) mineral statistics (USGS, 2026a)
138 complemented by national lime association reports and statistical yearbooks were used (SI-2
139 Data1). Cross-verification was performed to correct systemic omissions in official statistics.
140 Furthermore, the temporal scope was extended to 1930–2024 to synchronize with the Global
141 Carbon Budget timeframe. (2) Localized Refinement of Lime Emission Factors. Following the
142 "localized emission factor" methodology proposed by (Liu et al., 2015), emission factors was
143 adjusted based on specific calcium oxide (CaO) content requirements across downstream
144 sectors, such as the high-purity demands of the metallurgical industry. By integrating these
145 standards with sector-specific consumption ratios, lime emission factors were back-calculated,
146 a refinement that substantially reduces uncertainty in process emissions accounting. (3)
147 Integration of Lime Materials within Blast Furnace Slag (BFS). By recognizing that BFS
148 contains lime-based components with carbonation potential similar to steel slag, BFS was
149 formally incorporated into the accounting boundary. This inclusion ensures a more
150 comprehensive quantification of the global lime-related carbonation sink. (4) Dynamic
151 Parameterization of Technological Evolution. Through accounting for regional variations in
152 iron and steel industry and the global diversity of slag disposal environments, dynamic
153 accounting parameters were implemented. Specifically, a tiered temporal approach was applied
154 to reflect changes in slag production and utilization rates, which divided into 1930–1950
155 (wartime catalysts and industrial shifts), 1950–1970 (efficiency revolution and process
156 standardization), 1970–2000 (energy crises and technological optimization), and 2000–2024
157 (green development and intelligent manufacturing). Unlike previous studies that assumed static
158 values, this multi-period setting captures the 20-to-30-year generational cycles of metallurgical
159 technology, which could more accurately reflecting the impacts of technical progress on the
160 lime process emissions estimation and carbon sequestration potential of lime materials



161 embedded in slag. These improvements enhance the precision of global lime industry emission
162 inventories by correcting foundational data and refining emission parameters. Simultaneously,
163 the reduction in accounting uncertainties stemming from regional disparities and technological
164 transitions, providing a robust parametric foundation for independent national carbon
165 assessments.

166 **2. Data Sources and Methodology**

167 **2.1. Data Sources and Processing**

168 The dataset constructed in this study spans the period from 1930 to 2024, covering 11
169 major lime-producing nations (China, The United States of America, The United Kingdom,
170 France, Germany, Italy, Australia, Canada, Brazil, Japan, and Russia) as well as the "Rest of the
171 World" (ROW).

172 **2.1.1. Core Basic Data**

173 (1) Lime Production Data

174 1930–2020: Data for China and the USA were adopted from the verified dataset
175 established by (Bing et al., 2023), while production data for 2021–2024 were sourced from the
176 United States Geological Survey (USGS).

177 1930–1958: Lime production data for Japan (Shimanish, 2004), Brazil (IBGE, 2026),
178 France (ASF, 2026), and Canada (Dominion Bureau of Statistics, 2026) were derived from
179 historical literature and statistical yearbooks. For Italy and Germany, production figures were
180 estimated using multiple linear regression models; the detailed regression coefficients are
181 provided in Supplementary Table SI-2 Data 1. For Australia, lime production was calculated
182 based on the proportion of limestone allocated for lime manufacturing and the limestone-to-
183 lime conversion rate, with underlying limestone production data obtained from the *Year Book*
184 *Australia* (ABS, 2026). For the United Kingdom, lime production was estimated based on
185 limestone and dolomite production figures, utilizing specific allocation ratios for lime
186 production (0.161 and 0.049, respectively) and an average lime yield factor of 0.225 (Ordnance
187 Survey, 2026). Russian lime production data were sourced from the *Statistical Yearbook of the*
188 *USSR* (1961), with data for missing years filled using linear interpolation.

189 1959–2024: Data for the 11 countries were sourced from the United States Geological
190 Survey (USGS). Notably, data for Russia from 1959 to 1991 were derived by multiplying the
191 total lime production of the Soviet Union by Russia's average share (0.54); this proportion was
192 obtained from the Statistical Yearbook of the National Economy of the USSR (1961)
193 (Upravlenie S. U. T. statisticheskoe, 1961). Detailed datasets are provided in Supplementary
194 Table SI-2 Data 1.

195 (2) Industrial Product Output Data

196 For the period from 1930 to 2020, data on crude steel, alumina, and paper and paperboard
197 for China, the United States, and the global total were adopted from the verified dataset
198 constructed by (Bing et al., 2023). For 2021–2024, data on crude steel and alumina for China,
199 the United States, additional countries, and the global total were sourced from the USGS, while
200 data on paper and paperboard were obtained from the *FAO Yearbook of Forest Products*.
201 Specific data sources were organized as follows:

202 Crude steel and pig iron: Data for additional countries from 1959 to 2024 were retrieved
203 from the *USGS Iron and Steel Slag Database* (USGS, 2026b), while historical data from 1930



204 to 1958 were sourced from the British Geological Survey (BGS, 2026.). Detailed datasets are
205 provided in Supplementary Table SI-2 Data 2 and Data3.

206 Alumina: Data for additional countries covering 1959–2024 were obtained from the USGS
207 Bauxite and Alumina Database (USGS, 2026c). Data for the 1930–1958 period were derived
208 from the British Geological Survey (BGS, 2026) and the *Statistical Yearbook of France* (ASF,
209 2026). Detailed datasets are provided in Supplementary Table SI-2 Data 4.

210 Paper and paperboard: Data from 1946 to 2024 were sourced from the FAO Yearbook of
211 Forest Products (FAO, 2026), and data from 1930 to 1945 were obtained from the *United*
212 *Nations Statistical Yearbook* (UNSDW, 2026). Detailed datasets are provided in Supplementary
213 Table SI-2 Data 5.

214 **2.1.2. Key Calculation Parameters**

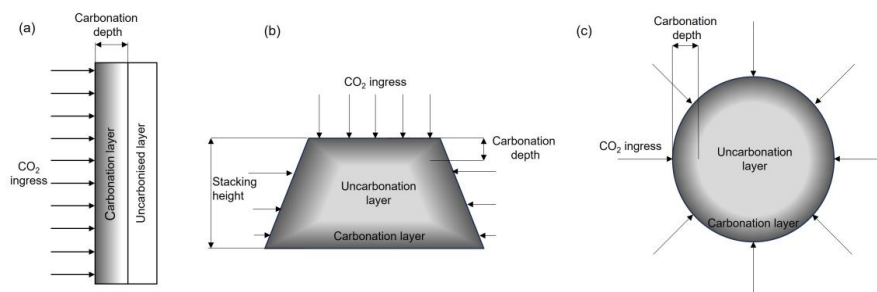
215 (1) Carbon Emission Factors

216 Carbon emission factors (CEF) were determined following the "Tier 2" approach outlined
217 in the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2026), based on
218 the CaO content of different lime types. Specifically, the CEFs for the United States, China, the
219 UK, France, Germany, Italy, Japan, Canada, Brazil, Russia, Australia, and other countries were
220 calculated as 0.742, 0.716, 0.732, 0.728, 0.733, 0.733, 0.728, 0.741, 0.737, 0.733, 0.729, and
221 0.737 t CO₂/t lime, respectively. Detailed literature sources are provided in Supplementary
222 Table SI-3 Data 1.

223 (2) Carbonation-related parameters

224 CaO Content: Data on the CaO content of materials—including Steel Slag (SS), Blast
225 Furnace Slag (BFS), red mud (RM), and lime mud (LM)—were obtained through a systematic
226 review of the literatures and reports from industry associations (e.g., the European Lime
227 Association (EuLA,2026b), Detailed literature sources are provided in Supplementary Table
228 SI-3 Data 7.

229 The carbonation ratio (γ) is defined as the proportion of calcium oxide converted to
230 calcium carbonate. For Precipitated Calcium Carbonate (PCC) and Carbonation Sugar (SUG),
231 γ was set to 1, indicating complete carbonation during the production process (Wang and Shen,
232 2002). For Lime-Stabilized Soil (LSS), carbide slag (CS), and Lime Kiln Dust (LKD), the
233 annual carbonation ratio (R) was assumed to be 1, representing complete carbonation within
234 one year. Based on the specific disposal or stacking scenarios, different kinetic models were
235 applied. The slab model (Wu et al., 2024) was used to describe the carbonation process of lime
236 mortar (Fig. 1); the pile model (Xi et al., 2016) was adopted to simulate the carbonation of lime
237 mud and red mud; and the spherical model (Manocha and Ponchon, 2018) was employed for
238 SS and BFS.



239
 240 Fig. 1. Schematic diagrams of the carbonation models: (a) the slab model for lime mortar; (b) the
 241 pile model for paper lime mud and red mud; and (c) the spherical model for iron and steel slag.

242 (3) Industrial and Technological Parameters

243 Lime utilization share, the proportion of lime used across different sectors specifically
 244 construction (LSS + MOR), metallurgy (SS + BFS + RM), and the chemical industry (PCC +
 245 CS + SUG + LM) was determined for different countries based on the literatures. Detailed
 246 literature sources are provided in Supplementary Table SI-3 Data 3-6. For generation and
 247 utilization rates of SS and BFS, the parameters were established by reviewing country-specific
 248 literatures. Based on the technological evolution of the iron and steel industry (Naito et al.,
 249 2015), the timeline was divided into four distinct phases: 1930–1950, 1950–1970, 1970–2000,
 250 and 2000–2024. Detailed literature sources are provided in Supplementary Table SI-3 Data 9.

251 **2.2. Estimation of Process CO₂ Emissions from Lime Production**

252 Calculations were conducted using the Tier 2 method from the 2006 IPCC Guidelines for
 253 National Greenhouse Gas Inventories (IPCC, 2006), considering CO₂ emissions generated
 254 solely from limestone calcination. The calculation formula is as follows:

$$255 E_{progress,i} = P_{lime,i} \times EF_{lime,i} \quad (3)$$

256 where, $E_{progress,i}$ represents the industrial process CO₂ emissions from the lime industry in
 257 country i ; $P_{lime,i}$ denotes the annual lime production of country i ; $EF_{lime,i}$ is the CO₂ emission
 258 factor for lime production in country i .

259 **2.3. Estimation of CO₂ Uptake by Lime**

260 The system boundary, established via Material Flow Analysis (MFA), encompasses ten
 261 categories of lime-based materials classified by their lifecycle stages. Specifically, LKD is
 262 categorized as a direct by product of the lime production process. Beyond the production stage,
 263 nine industrial byproducts and materials are incorporated across three sectors: metallurgy (SS,
 264 BFS, RM), chemicals (PCC, CS, SUG, LM), and construction (LSS, MOR). Notably, BFS is
 265 newly integrated to rectify previous accounting omissions in the metallurgical carbonation sink.

266 **2.3.1. General Calculation Model**

267 (1) Carbon Sequestration

268 Based on the stoichiometry of the calcium oxide carbonation reaction, the governing
 269 equations for all materials were established by incorporating material production, CaO content,
 270 conversion efficiency, and carbonation ratio. The carbon sequestration (CO₂) is calculated as
 271 follows:

$$272 C = M \times f^{CaO} \times \gamma \times R \times \frac{M_{CO_2}}{M_{CaO}} \quad \text{Eq (4)}$$



273 where, M represents the production volume of lime materials; f^{CaO} denotes the mass
 274 fraction of CaO in the material (%); and γ signifies the conversion rate of CaO to CaCO₃,
 275 which is assumed to be 100% for PCC and SUG, while specific values for other materials are
 276 detailed in Supplementary Table SI-3 Data 1. The variable R indicates the annual carbonation
 277 rate (%); specifically, R is set to 1 for materials capable of complete carbonation within a single
 278 year (i.e., LKD, LSS, CS, PCC, and SUG), whereas for materials that do not fully carbonate
 279 within one year (i.e., MOR, LM, RM, SS, and BFS), R is calculated using distinct models
 280 tailored to each material type (see Supplementary Table SI-3 Data10). Finally, $\frac{M_{CO_2}}{M_{CaO}}$ represents
 281 the molar mass ratio of carbon to calcium oxide (44/56).

282 (2) Annual Carbonation Ratio

283 The carbonation process of lime-based materials was simulated using three geometric
 284 models, including the slab model, the pile model, and the spherical model.

285 For the slab model, the annual carbonation ratio R_{lime} is expressed as:

$$286 R_{lime} = (d_{lime,i} - d_{lime,i-1})/d_T \quad \text{Eq (5)}$$

287 where, $d_{lime,i}$ denotes the carbonation depth of lime-based materials (such as MOR) in
 288 year i , while d_T signifies the design thickness of lime.

289 In the pile model, the annual carbonation ratio R_{lime} is defined as:

$$290 R_{lime,i} = \begin{cases} \frac{k_{mud} \times \sqrt{t_i}}{h_{mud} \times t_i} (t_i \leq t_{lm}), \\ \frac{d_{mud,i}}{h_{mud}} (t_{mud} < t_i < 100), \end{cases} \quad \text{Eq (6)}$$

291 where, $R_{lime,i}$ signifies the carbonation ratio in the i -th year, d_{mud} is the carbonation
 292 depth calculated using the carbonation rate k_{mud} and time t_i , $d_{mud,i} = k_{mud} \times (\sqrt{t_i} - \sqrt{t_{i-1}})$,
 293 while h_{mud} and t_{mud} represent the stacking height and storage duration of the lime-based
 294 material, respectively.

295 In the spherical model, the annual carbon uptake ratio R_{lime} is expressed as:

$$296 R_{lime,i} = \begin{cases} 100\% - \frac{\int_a^b \pi (D - D_{slag})^3}{\int_a^b \pi D^3} \times 100\% & (a > D_{slag}) \\ 100\% - \frac{\int_{D_0}^b \pi (D - D_{slag})^3}{\int_a^b \pi D^3} \times 100\% & (a \leq D_{slag} \leq b) \\ 100\% & (b < D_{slag}) \end{cases} \quad \text{Eq (7)}$$

297 Where, D denotes the particle size of the slag; D_{slag} represents the maximum diameter
 298 of completely carbonated slag lumps (SS or BFS); a and b stand for the corresponding
 299 minimum and maximum particle sizes of slag particles in a given particle size distribution;
 300 $D_{slag} = 2d_{slag,i} = 2k_{slag} \times \sqrt{t_i}$, $d_{slag,i}$ is the carbonation depth of the slag in the i -th year,
 301 k_{slag} refers to the carbonation rate of the slag, and t_i denotes the carbonation duration.

302 (3) Calculation of Annual and Cumulative Uptake

303 The carbon uptake from the multi-year carbonation of BFS, SS, Red Mud, and Carbide
 304 Slag is calculated by multiplying the annual carbonation ratio by the solid waste mass. Regional
 305 totals are obtained by summing the uptake across all lime-based materials. Additionally, the
 306 annual CO₂ uptake for the t_i th year is defined as the increment in cumulative uptake from year
 307 t_{i-1} to t_i , calculated using Equation (8). This quantifies the annual contribution of lime materials



308 to the total carbon sink.

$$309 \quad \Delta C_{lime,total}^{t_i} = \sum C_{lime,total}^{t_i} - \sum C_{lime,total}^{t_{i-1}} \quad \text{Eq (8)}$$

310 Where, $\Delta C_{lime,total}^{t_i}$ denotes the carbon uptake in the i -th year, $\sum C_{lime,total}^{t_i}$ represents
 311 the cumulative carbon uptake to the i -th year, and $\sum C_{lime,total}^{t_{i-1}}$ is the cumulative carbon
 312 uptake to the $(i-1)$ -th year.

313 2.3.2 Details of Material-Specific Calculations

314 (1) Lime materials from the production progress (LKD)

315 LKD is a byproduct of lime calcination. Its production is calculated as $M_{lkd} =$
 316 $m_{lime} \times r_{lkd}$, where r_{lkd} represents the industry default generation rate of kiln dust. Due to
 317 its fine particle size and high specific surface area, LKD undergoes rapid carbonation during
 318 stockpiling; therefore, the carbonation ratio is assumed to be $R_{lkd} = 100\%$.

319 (2) Lime Materials in the Construction Industry (LSS, MOR)

320 For LSS, $M_{lss} = m_{lime} \times L_1 \times a_1$, where L_1 represents the proportion of lime utilized
 321 in the construction sector, and a_1 denotes the share of lime allocated to LSS within the
 322 construction sector.

323 For MOR, $M_{mor} = m_{lime} \times L_1 \times a_2$, where a_2 denotes the proportion of lime allocated
 324 to MOR within the construction sector. The slab model was adopted to calculate the carbonation
 325 ratio R_{mor} , as expressed in the following equation: $R_{mor} = \frac{d_{mor,i} - d_{mor,i-1}}{d_T}$, where d_T
 326 represents the design thickness of the mortar, and $d_{mor,i}$ denotes the carbonation depth in the
 327 i -th year. Based on Fick's laws of diffusion, the carbonation depth is calculated as $d_{mor} =$
 328 $k_{mor} \times \sqrt{t_{mor}}$, where k_{mor} is the carbonation rate coefficient of MOR, and t_{mor} represents
 329 the duration of carbonation.

330 (3) Lime Materials in the Metallurgical Industry (SS, BFS, RM)

331 For SS, The production of SS is strongly correlated with crude steel output ($m_{crude\ steel}$).
 332 The mass is calculated as: $M_{ss} = m_{crude\ steel} \times r_{ss} \times U_{ss}$, where r_{ss} represents the steel
 333 slag generation rate (time-dependent values are provided in SI-3 Data 1), and U_{ss} denotes the
 334 ratio of stockpiling or roadbed utilization. The carbonation process was approximated using the
 335 uniform spherical model proposed by Bing et al. (2023), as shown in Equation (7).

336 For BFS, $M_{bfs} = m_{bfs} \times r_{bfs} \times l_{bfs} \times U_{bfs}$, where, r_{bfs} is the generation rate of blast
 337 furnace slag (time-dependent values, see SI-3 Data 1); l_{bfs} represents the proportion of CaO
 338 in the slag derived specifically from lime; and U_{bfs} denotes the stockpiling/utilization ratio,
 339 following the same logic as SS. The carbonation model is consistent with that of SS (spherical
 340 model, Equation 7).

341 For RM, $M_{rm} = m_{alumina} \times r_{rm} \times (1 - U_{rm})$, where, r_{rm} is the red mud generation rate,
 342 and U_{rm} is the comprehensive utilization rate of red mud. The carbonation ratio was
 343 calculated using the pile model (Equation 6). Based on the average storage duration in red mud
 344 yards and the laboratory-measured carbonation rate k_{rm} , the carbon sequestration was
 345 quantified cumulatively on an annual basis.

346 (4) Lime Materials in the Chemical Industry (PCC, SUG, CS, LM)

347 For PCC and SUG, $M_{pcc} = m_{lime} \times L_2 \times b$, where, L_2 represents the proportion of total



348 lime utilized in the chemical industry, and b denotes the share of lime allocated to PCC or SUG
349 production within this sector.

350 For LM, $M_{lm} = m_{pappre} \times r_{lm} \times (1 - U_{lm})$, where m_{pappre} is production of paper and
351 paperboard, r_{lm} is the lime mud generation rate, and U_{lm} is the recycling rate. Given the
352 similar physicochemical properties between lime mud and red mud, the carbonation rate of red
353 mud was used as a reference. Consequently, the pile model (Equation 6) was adopted to
354 calculate the carbonation ratio.

355 For CS, $M_{cs} = m_{lime} \times L_2 \times b_3 \times p_{lime}^{cs} \times r_{cs} \times (1 - U_{cs})$, where, L_2 is the proportion of
356 lime utilized in the chemical industry; b_3 denotes the share of chemical lime allocated to
357 calcium carbide production; p_{lime}^{cs} represents the yield of calcium carbide per ton of lime; r_{cs}
358 is the generation rate of carbide slag; and U_{cs} signifies the utilization rate of the slag.

359 **2.4 Uncertainty Assessment**

360 We identified 18 groups of influencing factors associated with the estimation of carbon
361 emissions and carbon uptake in the lime production process. These factors comprise 1,894
362 specific input parameters, each characterized by a distinct statistical distribution (see
363 Supplementary Table SI-3 Data1-15). Given the difficulty in obtaining the various values for
364 these parameters, Monte Carlo method recommended by the 2006 IPCC Guidelines for
365 National Greenhouse Gas Inventories was employed to assess the uncertainty associated with
366 the carbon emissions and uptake of lime materials. The statistical characteristics of these 1,894
367 variables were put into the model and Monte Carlo simulations with 10,000 iterations were
368 performed to generate simulated results for carbon emissions and uptake. Subsequently,
369 statistical analysis was conducted to determine the median values. The 90% confidence
370 intervals (CIs) were calculated using the percentile method; specifically, the 5th and 95th
371 percentiles of the modeled results served as the lower and upper limits of the 90% interval.

372 Compared with our previous research, this study incorporates refinements in four key
373 aspects. (1) Regional refinement to enhance reference value. We have separated the United
374 Kingdom, France, Germany, Italy, Russia, Japan, Brazil, Australia, and Canada from the ROW
375 category used in previous studies. This allowed to establish a lime carbonation sink dataset for
376 11 countries for the first time, covering an average of 81.09% of global lime production. (2)
377 More detailed, country-specific parameterization. The parameters were updated for lime CO₂
378 uptake in the newly added countries. The parameters include the proportion of lime usage across
379 various sectors, the CaO content of different lime materials, and the production and utilization
380 rates of lime materials. These improve the accuracy of the accounting compared to the CO₂
381 uptake parameters used for the ROW region in our previous work. (3) Inclusion of BFS in the
382 accounting system. the research gap in metallurgical slag carbonation sinks was filled by
383 incorporating BFS. (4) The SS and BFS parameters were updated based on technological
384 evolution across four distinct periods (1930–1950, 1950–1970, 1970–2000, and 2000–2024).
385 By updating the production and utilization rates of steel slag and BFS, the impact of
386 technological progress on carbon sequestration by metallurgical slags were reflected, thereby
387 enhancing the accuracy of historical data.

388 **3. Results and Discussion**

389 **3.1 Global Process CO₂ Emissions from Lime Production**

390 Figure 2 shows the global process carbon emissions from lime production. Between 1930



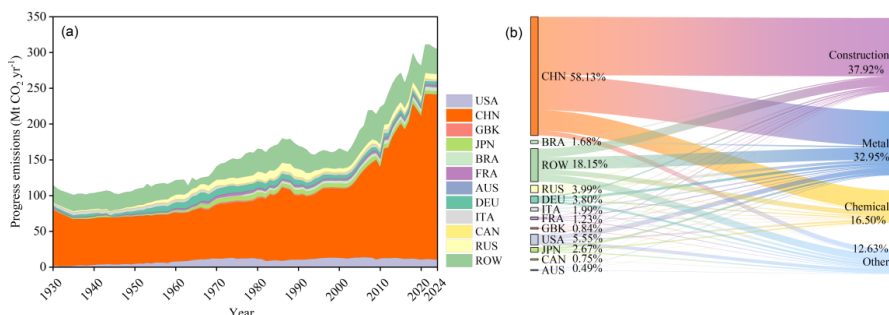
391 and 2024, global process CO₂ emissions from the lime industry exhibited a significant upward
392 trend. Annual emissions rose from 113.35 Mt CO₂ yr⁻¹ (95% CI: 46.43–180.72 Mt CO₂) in 1930
393 to 299.28 Mt CO₂ yr⁻¹ (95% CI: 288.74–310.02 Mt CO₂) in 2024. The cumulative emissions
394 over this period were 15.29 Gt CO₂ (95% CI: 13.81–16.79 Gt CO₂, Fig. 2a). It is driven by the
395 continuous expansion of global urbanization and industrialization. Particularly, the demand for
396 lime in sectors such as construction, metallurgy, chemical engineering, and environmental
397 protection (Wu et al., 2024), increased by approximately 200.8%, growing from 139.62 Mt in
398 1930 to 420 Mt in 2024 (Lime Statistics and Information | U.S. Geological Survey, 2026)
399 (Supplementary Table SI-2 Data 1).

400 Statistical analysis reveals a strong correlation between lime carbon emissions and lime
401 production. However, the growth rate of process emissions was lower than that of lime
402 production, suggesting that technological advancements have reduced the carbon emissions
403 intensity of lime production (Laveglia et al., 2024b). From 1930 to 2024, the construction sector
404 accounted for the highest lime-related carbon emissions (Fig. 2b). Emissions increased from
405 46.61 Mt CO₂ yr⁻¹ (95% CI: 11.63–86.62 Mt CO₂) in 1930 to 128.98 Mt CO₂ yr⁻¹ (95% CI:
406 96.26–161.46 Mt CO₂) in 2024. Cumulative emissions from the construction sector reached 5.8
407 Gt CO₂ (95% CI: 4.11–7.62 Gt CO₂), representing 37.92% of the total cumulative emissions
408 (Fig. 2b). The metallurgical industry followed closely with emissions rising from 36.69 Mt CO₂
409 yr⁻¹ (95% CI: 15.64–64.87 Mt CO₂) in 1930 to 96.51 Mt CO₂ yr⁻¹ (95% CI: 66.24–128.95 Mt
410 CO₂) in 2024. This sector contributed 5.04 Gt CO₂ (95% CI: 3.64–6.64 Gt CO₂) cumulatively,
411 accounting for 32.95% of the total (Fig. 2b). Carbon emissions from chemical industry were
412 increasing from 17.70 Mt CO₂ yr⁻¹ (95% CI: 8.04–29.16 Mt CO₂) in 1930 to 45.91 Mt CO₂ yr⁻¹
413 (95% CI: 34.64–58.41 Mt CO₂) in 2024. The cumulative emissions for this sector were 2.52 Gt
414 CO₂ (95% CI: 1.91–3.21 Gt CO₂), comprising 16.50% of the total (Fig. 2b). Cumulative carbon
415 emissions from other sectors amounted to 1.93 Gt CO₂ (95% CI: 1.32–2.65 Gt CO₂), accounting
416 for 12.63% of the total.

417 Between 1930 and 2024, distinct trend in process CO₂ emissions from lime production
418 was observed across different countries. As shown in Fig. 2a, developed nations such as the
419 USA, Germany, France, the UK, and Japan reached their carbon emissions peak around the
420 1970s, followed by a gradual decline. Notably, emissions in the USA resumed an upward trend
421 entering the 21st century. In contrast, developing countries such as China, Brazil, and Russia
422 generally exhibited an increasing trend in emissions. Specifically, Russia experienced a
423 significant reduction in the 1990s due to the dissolution of the Soviet Union. China emerged as
424 the primary contributor to both global lime production and associated carbon emissions. China's
425 annual emissions rose from 74.34 Mt CO₂ yr⁻¹ (95% CI: 7.27–141.63 Mt CO₂) in 1930 to
426 219.65 Mt CO₂ yr⁻¹ (95% CI: 209.36–230.22 Mt CO₂) in 2024, reflecting an average annual
427 growth rate of 1.16%. Cumulative emissions from China were 8.89 Gt CO₂ (95% CI: 7.42–
428 10.36 Gt CO₂), accounting for 58.13% of the global total (Fig. 2b). The USA ranked as the
429 largest emitter among developed nations, with cumulative emissions of 954.03 Mt CO₂ (95%
430 CI: 910.64–997.41 Mt CO₂), representing 6.24% of the global total. Russia (611.06 Mt CO₂),
431 Germany (581.75 Mt CO₂), Japan (408.58 Mt CO₂), Italy (305.00 Mt CO₂), Brazil (258.20 Mt
432 CO₂), and France (189.22 Mt CO₂) contributed 3.99%, 3.8%, 2.67%, 1.99%, 1.68%, and 1.23%
433 to the global total, respectively. The contributions of the United Kingdom (129.43 Mt CO₂),



434 Australia (76.38 Mt CO₂), and Canada (115.21 Mt CO₂) were relatively small, each accounting
 435 for less than 1% (Fig. 2b). Cumulative emissions from the ROW amounted to 2.78 Gt CO₂ (95%
 436 CI: 2.65–2.91 Gt CO₂), constituting 18.15% of the global total (Fig. 2b).



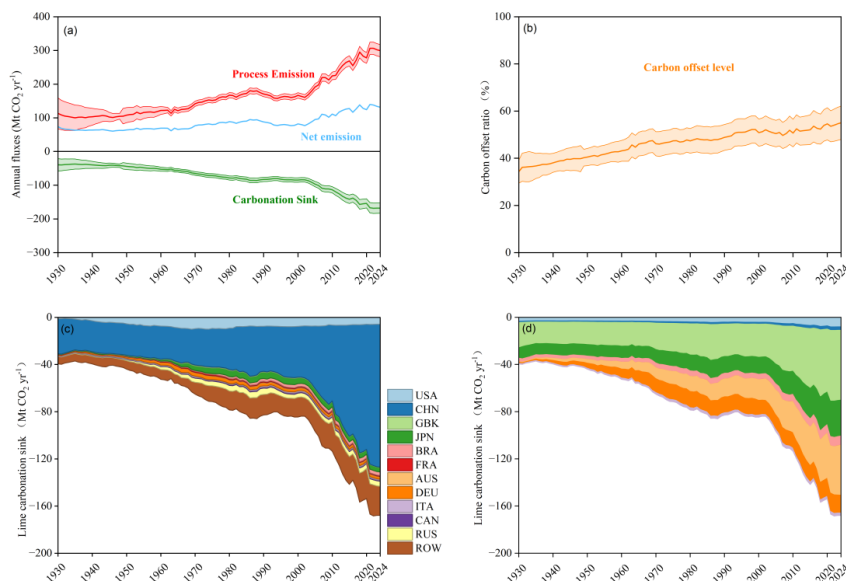
437
 438 Fig. 2 Global process carbon emissions from lime production. (a) Global annual CO₂ emissions;
 439 (b) Cumulative process CO₂ emissions from the lime industry by country and sector.

440 3.2 Global Carbon Uptake by Lime Carbonation

441 Global lime carbonation sink increased from 39.34 Mt CO₂ yr⁻¹ (95% CI: 12.41–69.76) in
 442 1930 to 167.80 Mt CO₂ yr⁻¹ (95% CI: 132.82–209.11) in 2024 (CAGR: 1.55%), with totally
 443 accumulative 7.33 Gt CO₂ sink (95% CI: 5.95–8.88) (Fig. 3a). Concurrently, net emissions rose
 444 from 74.83 Mt CO₂ yr⁻¹ in 1930 to 133.04 Mt CO₂ yr⁻¹ in 2024, resulting in cumulative net
 445 emissions of 8.00 Gt CO₂ (Fig. 3a). This indicates that lime materials reabsorbed approximately
 446 47.65% of their process emissions (Fig. 3b), about 8.32% increase over previous estimates. The
 447 11 nations contributed 6.03 Gt CO₂ (82.27% of the global total). By refining additional 9
 448 countries' data, the grouped ROW share decreased significantly from 34.35% (Bing et al., 2023)
 449 to 17.6% (Fig. 3a). The accounting boundary was improved by including BFS and applying
 450 phased parameterization for SS and BFS to reflect technological evolution.

451 The rising carbon offset level (Fig. 3b) is attributed to the carbonation time lag (Niu et al.,
 452 2024). Materials such as MOR, RM, SS, BFS, and LM carbonate over years rather than
 453 instantaneously, leading to the accumulative sequestration of historical lime materials legacy.
 454 The uncertainty of the updated lime carbonation sink is reduced by 16.58%. The lime
 455 carbonation sink in 2024 is equivalent to 1.5%–2% of the 2023 global terrestrial sink
 456 (Friedlingstein et al., 2025). This natural and stable "anthropogenic sink" supports IPCC AR6
 457 accounting, enhances GCB completeness, and facilitates global carbon neutrality.

458 LSS exhibited the highest carbon uptake, with a cumulative absorption of 2.66 Gt CO₂
 459 (95% CI: 1.81–3.67), accounting for 36.53% of the total (Fig. 3d). Its annual uptake reached
 460 59.20 Mt CO₂ yr⁻¹ (95% CI: 41.89–78.94) in 2024. MOR followed in second place, contributing
 461 a cumulative carbon sink of 1.36 Gt CO₂ (18.66%) (Fig. 3d). SS and BFS ranked third and
 462 fourth, respectively: SS accumulated 1.29 Gt CO₂ (17.73%), while BFS—notably a material
 463 newly incorporated into this study—achieved a substantial cumulative uptake of 934.83 Mt
 464 CO₂ (95% CI: 678.30–1247.10), accounting for 12.83%. Collectively, these four materials
 465 contributed 87.47% (7.25 Gt CO₂) of the global lime carbonation sink. In contrast, other
 466 materials such as CS, LKD, and PCC made smaller contributions due to their limited production
 467 volumes.



468

469 Fig3. (a) Global annual lime process CO₂ emission and carbonation sink;(b) Carbon offset level
 470 (ratio of carbonation sink to process CO₂ emission); (c) Annual carbonation sink of lime materials by
 471 country and region; (d) Annual carbonation sink of different lime materials

472 China's lime carbonation sink was increasing from 29.6 Mt CO₂ yr⁻¹ (95% CI: 2.82–59.84)
 473 in 1930 to 121.27 Mt CO₂ yr⁻¹ (95% CI: 88.13–160.49) in 2024. Cumulative uptake reached
 474 4.21 Gt CO₂ (95% CI: 2.75–6.01 Gt), accounting for 57.45% of the global total (Fig. 3c), which
 475 is driven by sustained demand from large-scale construction and iron & steel industries. The
 476 United States is the largest contributor among developed nations, with annual lime carbonation
 477 sink rising from 1.22 Mt CO₂ yr⁻¹ (95% CI: 0.83–1.74) to 5.79 Mt CO₂ yr⁻¹ (95% CI: 4.29–
 478 7.53). Its cumulative uptake of 625.16 Mt CO₂ (95% CI: 451.77–839.45) represents 8.52% of
 479 the global total. The contributions from Russia (285.67 Mt CO₂), Germany (193.82 Mt CO₂),
 480 Japan (296.69 Mt CO₂), Brazil (132.20 Mt CO₂), and Italy (87.97 Mt CO₂) accounted for 3.89%,
 481 2.64%, 4.05%, 1.80%, and 1.19%, respectively (Fig. 3c). In contrast, France, the United
 482 Kingdom, Australia, and Canada each contributed less than 1%, directly reflecting their limited
 483 lime production.

484 3.3 Global Lime Carbonation Sink in Different Industries

485 Industry-based analysis indicates that global carbon uptake by lime-based materials
 486 exhibited a persistent increasing trend across all major industrial sectors from 1930 to 2024.
 487 However, substantial inter-sectoral differences are observed in terms of carbonation sink
 488 magnitude, dominant material types, and evolutionary pathways (Fig. 4). Overall, the
 489 construction sector represents the core contributor to the global lime carbonation sink, followed
 490 by the metallurgical sector, while the chemical industry provides a small yet stable contribution.

491 Carbon uptake in the construction sector increased from 30.14 Mt CO₂ yr⁻¹ in 1930 (95%
 492 CI: 7.42–59.34) to 89.63 Mt CO₂ yr⁻¹ in 2024 (95% CI: 55.72–133.23, Fig. 3a). The cumulative
 493 carbonation sink over the study period reached 4.02 Gt CO₂ (95% CI: 2.59–5.79), accounting



494 for 57.45% of the global lime carbonation sink (Fig. 4). This dominant contribution is primarily
495 attributable to the high penetration of lime-based materials in the construction sector and the
496 sustained mineral carbonation occurring throughout their long service life. As a key pillar of
497 global infrastructure development and urban expansion, the construction sector has maintained
498 the largest share of lime-related carbon sequestration throughout the entire study period due to
499 its substantial demand for lime materials. In particular, since the latter half of the twentieth
500 century, accelerated global urbanization and infrastructure investment have markedly expanded
501 the use of lime-based materials, thereby driving a rapid increase in carbon uptake within this
502 sector (Manzoor and Yousuf, 2020). The synergistic effects among different lime materials
503 within the construction sector have continuously amplified its overall carbonation sink capacity
504 at the global scale, underscoring its central role in the lime-based carbon sequestration.

505 The metallurgical sector constitutes the second-largest global lime carbonation sink, with
506 a cumulative carbon uptake of 2.23 Gt CO₂ (95% CI: 1.76–2.77), accounting for 30.56% of the
507 total carbonation sink. Lime-based materials generated during metallurgical processes,
508 particularly in iron and steel production, provide a substantial material basis for mineral
509 carbonation. With the sustained growth of global iron and steel output over the past century, the
510 carbon sequestration potential of slags in the metallurgical sector has increased significantly.
511 Notably, by incorporating refined historical-stage parameters, this study systematically
512 accounts for variations in the generation rates and utilization efficiencies of metallurgical by-
513 products across countries, thereby substantially improving the reliability of carbonation sink
514 estimates for this sector. Previous studies have demonstrated pronounced cross-country
515 differences in slag and blast furnace residue management strategies (Guo et al., 2018; USGS,
516 2026b; O'Connor et al., 2021; Horii et al., 2015), which showed developed countries
517 established relatively mature resource utilization systems at an early stage, whereas developing
518 countries have gradually transitioned from long-term stockpiling toward more efficient and
519 comprehensive utilization ways. The combined effects of technological advancement and
520 policy intervention have played a decisive role in shaping the long-term accumulation of the
521 metallurgical lime carbonation sink.

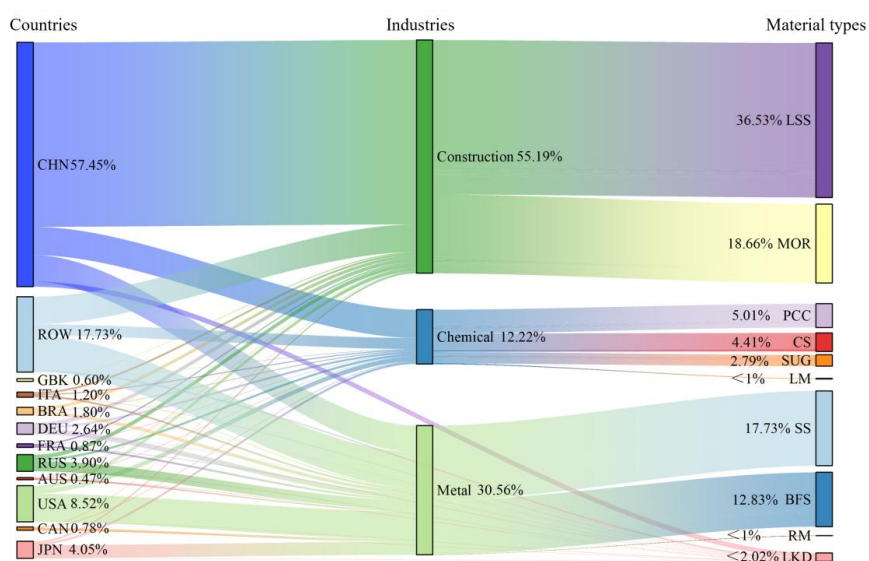
522 The chemical industry contributes a relatively smaller share to the global lime carbonation
523 sink, with a cumulative carbon uptake of 1.79 Gt CO₂ (95% CI: 1.05–2.78), accounting for
524 12.22% of the global total. In the chemical sector, lime-based materials sequester CO₂ mainly
525 through industrial carbonation reactions, and their contribution has increased steadily over the
526 study period, providing an important component to the global lime carbonation sink.

527 From spatial perspective, industry-dominated patterns of lime carbon sequestration exhibit
528 pronounced heterogeneity across countries and regions (Fig. 4). Developed countries (e.g., the
529 United States, Japan, and major European nations), characterized by long histories of iron and
530 steel production and well-established circular economy systems, have long been dominated by
531 metallurgical-sector lime carbonation sink, with cumulative effects that are particularly evident
532 on historical timescales (Spring and Cirella, 2022). In contrast, developing countries
533 undergoing rapid industrialization and urbanization (e.g., China, Brazil, and parts of the rest-
534 of-world regions) have experienced surging demand for lime-based materials in the
535 construction sector, which has gradually become the primary regional source of lime-related
536 carbon sequestration. In China, carbon uptake by lime materials in the construction sector



537 accelerated markedly in the early twenty-first century, closely coinciding with the timing of
 538 rapid urbanization (Cai et al., 2020).

539 Overall, the differentiated global distribution of lime carbonation sink across industries
 540 and regions fundamentally reflects the combined influences of industrial structure, economic
 541 development pathways, resource utilization efficiency, and policy frameworks. Clarifying the
 542 relative contributions of different sectors and their underlying evolutionary mechanisms is of
 543 critical scientific importance for designing sector-specific carbon management strategies for
 544 enhancing the carbon sequestration potential of lime-based materials.



545 Fig 4. Global Cumulative Lime Carbonation Sink in different countries, industries, and material
 546 types, 1930–2024
 547

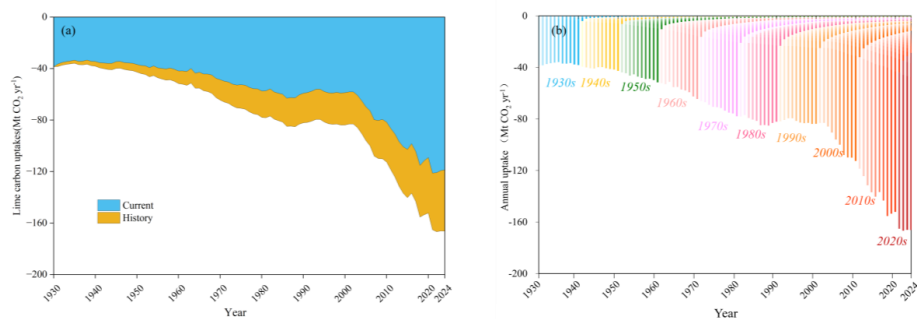
548 3.4 Time-Lag Effect of Lime Carbonation Sink

549 Based on estimates, both current-year and historical CO₂ uptake by global lime materials
 550 exhibited continuous growth trends (Fig. 5a). "Current-year uptake" refers to CO₂ absorbed by
 551 materials produced within the same year, whereas "historical uptake" denotes CO₂ sequestered
 552 in subsequent years by previously produced but uncarbonated materials.

553 Current-year uptake rose from 38.52 Mt CO₂ yr⁻¹ in 1930 to 118.65 Mt CO₂ yr⁻¹ in 2024
 554 (CAGR: 0.96%), with a cumulative total of 5.48 Gt CO₂, representing 75.24% of the total
 555 uptake. Conversely, historical uptake increased significantly from 1.99 Mt CO₂ yr⁻¹ in 1931 to
 556 47.59 Mt CO₂ yr⁻¹ in 2024 (CAGR: 2.12%), accumulating 1.77 Gt CO₂ (24.35% of the total)
 557 (Fig. 5a). LSS is the primary contributor to current-year uptake, accumulating 2.62 Gt CO₂
 558 (35.94% of the current cumulative total), followed by MOR with 1.04 Gt CO₂ (14.23%). In
 559 contrast, SS dominates historical uptake, accumulating 949.65 Mt CO₂ (53.78% of the historical
 560 cumulative total), followed by BFS with 570.33 Mt CO₂ (32.23%) (Supplementary Table SI-1
 561 Data 6). These results highlight the distinct temporal mechanisms of carbon uptake across
 562 different lime materials (Fig. 5b). Historical uptake for SS and BFS exceeds their current-year
 563 uptake because their carbonation is not instantaneous (Liu et al., 2018), and the majority occurs



564 in subsequent years. Additionally, environmental conditions such as temperature and humidity
565 influence the natural carbonation rates of SS and BFS (Zhao et al., 2025). Future efforts should
566 leverage Carbon Capture, Utilization, and Storage (CCUS) technologies to achieve efficient
567 sequestration (Yang et al., 2022).



568

569 Fig5. Delayed Effects of Lime Carbon Absorption, 1930–2024. (a) global lime annual CO₂
570 uptake in current and historical year; (b) global annual uptake of atmospheric CO₂ by lime,
571 disaggregated by years of production.

572 3.5 Peaking Trajectories and Regional Disparities in Lime Carbon Balance

573 Based on lime process emissions and carbonation sink data, this study reveals significant
574 regional heterogeneity in global lime carbon balance: developed nations (e.g., in Europe and
575 North America) have achieved carbon peaking, whereas most developing countries (e.g., China
576 and Brazil) remain in a growth trend.

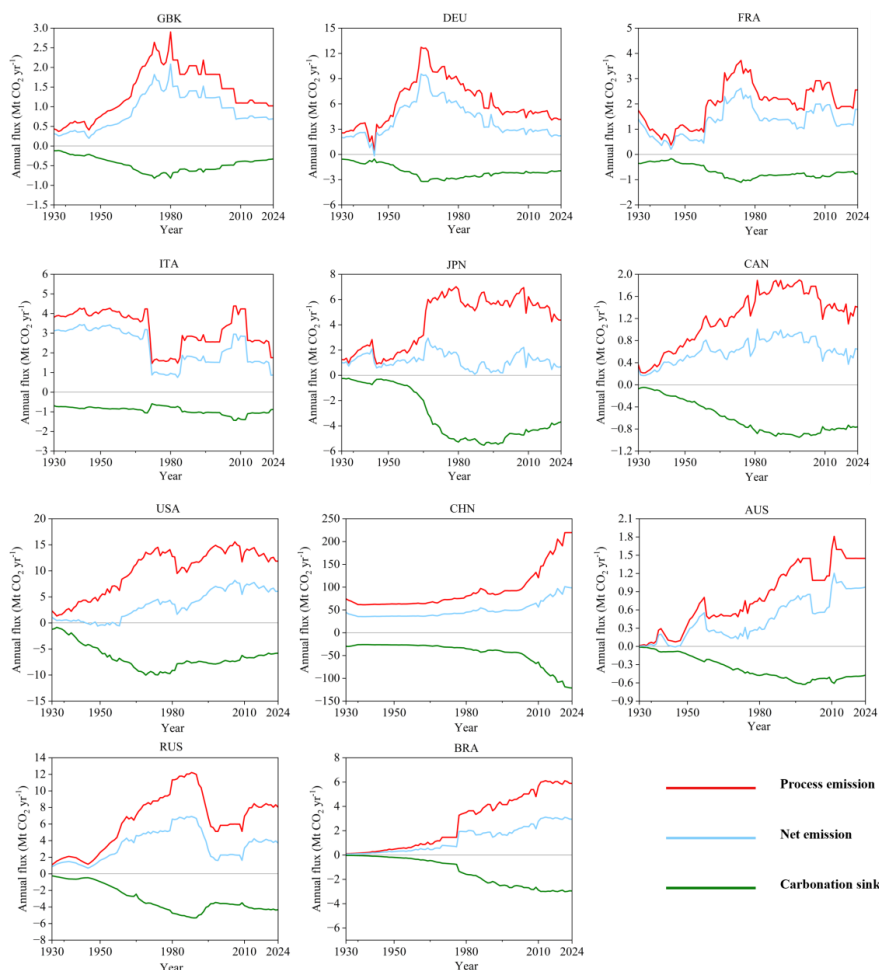
577 In developed countries, CO₂ emissions peaked between the 1960s and early 2000s.
578 Germany (1964, 12.73 Mt CO₂), France (1974, 3.72 Mt CO₂), Japan (1979, 7.01 Mt CO₂), and
579 the UK (1980, 2.90 Mt CO₂) peaked earlier, followed by Canada (1999, 1.89 Mt CO₂), the USA
580 (2006, 15.57 Mt CO₂), Italy (2008, 4.39 Mt CO₂), and Australia (2011, 1.81 Mt CO₂). The
581 carbon offset levels at their peaks were: 74.09% (Japan), 50.05% (Canada), 47.54% (USA),
582 33.55% (Australia), 28.15% (UK), 29.67% (France), and 25.00% (Germany), respectively.
583 Following the peak year, carbon emissions from lime production in these nations have exhibited
584 downward trend, with annual decline rates ranging from 0.8% to 5.2%. Carbonation sink has
585 also gradually decreased with lime production reduction. However, due to the time-lag effect
586 inherent in the carbonation of certain lime-based materials, the decline in carbon uptake has
587 been less pronounced than that of emissions. Consequently, the carbon offset level has
588 progressively increased, suggesting that future net emissions may approach zero or even
589 achieve negative values, thereby facilitating the realization of carbon neutrality within the lime
590 production sector. This characteristic is fundamentally attributed to the establishment of mature
591 secondary resource recycling systems and stringent climate policy constraints in developed
592 nations (Dolphin et al., 2023), particularly the high recovery and comprehensive utilization of
593 metallurgical by-products such as BFS and SS (Yi et al., 2012). In the fields of construction and
594 civil engineering, the substitution of primary lime materials with these alkaline solid wastes not
595 only substantially mitigates process-related carbon emissions during production but also
596 effectively sustains the carbonation sink potential through continuous mineral carbonation



597 during the full life cycle of their secondary application (Pan et al., 2017).

598 Conversely, developing countries exhibit continuous process emissions growth. China's
599 lime process emissions have risen since 1930 (CAGR: 1.16%), accelerating to 3.69% annually
600 post-2002 due to urbanization and infrastructure demands. Lime process emissions in China
601 reached 219.64 Mt CO₂ yr⁻¹ in 2024. Despite a high offset level of 55.21% (121.27 Mt CO₂
602 yr⁻¹), abatement pressure remains high. Lime process emissions in Brazil reached 5.89 Mt CO₂
603 yr⁻¹ in 2024 (offset: 50.11%). Although Russia peaked during the Soviet era, emissions
604 rebounded after 2000 (CAGR: 1.34%), reaching 8.04 Mt CO₂ yr⁻¹ in 2024 (offset: 53.87%).
605 Lime process emissions growth in these regions is driven by infrastructure demands amidst
606 industrialization and urbanization (Chen et al., 2022), while Russia's rebound correlates with
607 economic recovery and industrial capacity restoration post-2000.

608 In summary, the regional differentiation in process emissions and carbonation sink
609 trajectories reflect distinct development stages. Developed countries achieved inflection points
610 through industrial transformation and policy intervention, whereas developing countries face
611 the dual challenge of economic growth and carbon emissions reduction. Furthermore, the
612 divergence in carbon offset levels further reflects the disparities in the resource recycling
613 efficiency of lime materials across nations.



614
 615 Fig 6. Lime process emissions, carbonation sink, and net emissions in 11 Countries from 1930 to
 616
 617 2024

617 3.6 Uncertainty Analysis

618 Based on the accounting model established by (Xi et al., 2016) and the research framework
 619 of (Bing et al., 2023), this study incorporates new data on lime process emissions and
 620 carbonation sink for nine additional countries. Particular emphasis was placed on the CaO
 621 content of lime in different application industries, which is identified as the most significant
 622 factor influencing carbonation sink (Niu et al., 2024). Following IPCC recommendations, a
 623 Monte Carlo analysis with 10,000 iterations was conducted to quantify the uncertainties lime
 624 process emissions and carbonation sink. (Detailed uncertainty coefficients for lime material
 625 carbon sequestration by country are presented in Supplementary Table SI-3 Data1-15.)

626 The results indicate that the global cumulative process emissions in this study amount to
 627 15.29 Gt CO₂ (95% CI: 13.81–16.79 Gt CO₂), while the global cumulative lime carbonation
 628 sink is 7.33 Gt CO₂ (95% CI: 5.95–8.88 Gt CO₂). Compared to the previous estimates by Bing



629 et al. (2023) that reported 13.64 Gt CO₂ (95% CI: 11.61–15.72 Gt CO₂) and 5.29 Gt CO₂ (95%
630 CI: 3.73–7.19 Gt CO₂), they increase 12.1% and 38.43%, respectively. The annual lime
631 carbonation sink in 2020 reached 153.67 Mt CO₂ (95% CI: 122.63–190.35 Mt CO₂), which
632 showed 20.31% increase over the 127.73 Mt CO₂ (95% CI: 86.17–182.72 Mt CO₂) reported in
633 the previous study. Consequently, the cumulative carbon offset level (carbonation sink divide
634 process emissions) rose from 38.83% to 47.65%, with an increase of 8.32%. Furthermore, the
635 contribution of metallurgical slags to total lime carbonation sink increased from 15.62% to
636 26.9%, reflecting a growth of 11.23% impacted by BSF inclusion. Meanwhile, China's share of
637 cumulative carbonation sink was adjusted from 63.95% to 57.45%.

638 These discrepancies stem from quantifiable methodological optimizations rather than data
639 contradictions. The increase of global lime carbonation sink is jointly driven by the temporal
640 extension, the expansion of accounting boundary, and country-specific data refinement. The
641 inclusion of data for four additional years (2021–2024) based on USGS global lime production
642 statistics (reaching 420 million tonnes in 2024, with China accounting for 73.81%) contributed
643 an additional 746.79 Mt CO₂ in carbon sequestration, accounting for 9.01% of the total.
644 Moreover, the incorporation of BFS into the accounting boundary combined with solid waste
645 statistics from China's steel industry, where BFS constitutes over 50% of total iron and steel
646 solid waste (Pan et al., 2017), quantified a contribution of 934.83 Mt CO₂. This accounts for
647 11.27% of the total, effectively filling a critical gap in the carbonation sink accounting for
648 metallurgical slags. Additionally, by refining 11 core parameters on a country-specific data (e.g.,
649 correcting China's steel slag generation rate to 0.12–0.15 t/t crude steel for specific time
650 periods), the width of the 95% CI in this study was reduced by 12.47% compared to Bing et al.
651 (Bing et al., 2023), indicating a significant reduction in uncertainty.

652 **4. Data Availability**

653 The datasets generated and analyzed in this study are available in the Zenodo repository:
654 <https://doi.org/10.5281/zenodo.18616060> (Bing et al., 2026). The dataset comprises three data
655 files, including the results of lime-related carbon emissions and CO₂ uptake (Supplementary
656 Table SI-1), basic activity data for lime-based materials (Supplementary Table SI-2), and
657 uncertainty parameters associated with lime carbon emissions and CO₂ uptake (Supplementary
658 Table SI-3).

659 **5. Conclusion**

660 In this study, we further advanced the high-resolution accounting of global carbon
661 emissions and uptake within the lime industry. For emission accounting, we extended the
662 temporal scope to the period of 1930–2024 and refined the spatial resolution to 11 major
663 producing countries, capturing 87.62% of total global production. Furthermore, accounting
664 uncertainties were significantly mitigated by deriving localized emission factors based on the
665 specific calcium oxide requirements and consumption structures of downstream industries. Our
666 results demonstrate that cumulative emissions reached 15.33 Gt CO₂ (95% CI: 13.15–17.58 Gt
667 CO₂) between 1930 and 2024. The construction sector emerged as the primary emission source,
668 cumulatively releasing 5.8 Gt CO₂, followed by the metallurgical (5.04 Gt CO₂) and chemical
669 (2.55 Gt CO₂) sectors.

670 Regarding carbon uptake, we expanded the system boundary by incorporating blast
671 furnace slag (BFS) into the accounting framework for the first time, and dynamically updated



672 the output and utilization parameters of steel slag (SS) and BFS in distinct phases to reflect the
673 technological evolution of the iron and steel industry. During the same period, global lime
674 materials cumulatively sequestered 7.33 Gt CO₂ (95% CI: 5.66–9.32 Gt CO₂). This carbonation
675 sink offsets approximately 47.65% of historical process emissions and represents roughly
676 1.5%–2% of the 2023 global terrestrial carbon sink. China remains the dominant contributor,
677 with a cumulative uptake of 4.21 Gt CO₂ (95% CI: 2.75–6.01 Gt CO₂), representing 57.45% of
678 the global total. LSS, MOR, SS, and BFS were identified as the primary sequestering materials,
679 contributing 36.53%, 18.66%, 17.73%, and 12.83% to the total sink, respectively (collectively
680 85.75%). Historically, SS and BFS played the most significant roles in sequestration,
681 accounting for 53.78% and 32.23% of the cumulative historical uptake.

682 Regional divergence of lime carbonation sink is pronounced. Carbon sequestration is
683 primarily from LSS, MOR, and SS in developing nations such as China and Brazil, whereas
684 dominant from SS and BFS in developed regions like the United States. Developed economies,
685 including Europe, the United States Japan, and Australia, have already achieved carbon peaks,
686 characterized by declining gross emissions and net emissions approaching zero. In contrast,
687 developing nations continue to show simultaneous growth in both emissions and uptake.
688 Despite maintaining high offset levels, these regions face acute pressure from rising absolute
689 emission volumes. These findings underscore the critical carbon sink value of the lime industry
690 and provide a new lens through which to view global carbon neutrality pathways. Furthermore,
691 the enhanced accounting precision provided by this study offers the necessary empirical support
692 for incorporating lime carbon uptake into the Global Carbon Budget.

693
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695 from L.N., carried out data collection, organization, data analysis, and database construction.
696 L.B. was responsible for developing and parameterizing the accounting model, writing the
697 computational code, and generating the integrated accounting results. The manuscript was
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700
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