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An investigation of the global uptake of CO2 by lime from 1963 to 2020

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Abstract. Lime is responsible for the continuous and stable absorption of CO_2 from the atmosphere during its cycle via the carbonation reaction. However, the magnitude of the global uptake of CO_2 by lime under natural conditions remains unclear. Here, existing data on materials associated with the production, utilisation, and disposal stages of lime-containing materials were analysed using a comprehensive model to obtain regional and global estimates for the sequestration of carbon from 1963 to 2020. The CO_2 emissions linked to the production of lime during the investigated period were also estimated. The results reveal that the global uptake of CO_2 by lime increased from 38.25 Mt (95 % confidence interval, CI:27.85-51.38 Mt) in 1963 to 134.33 Mt (95 % CI:90.37-139.29 Mt) in 2020. Cumulatively, approximately 4.05 Gt of CO_2 (95 % CI:3.02-5.25 Gt CO_2) were sequestered by lime produced between 1963 and 2020, and this amount corresponds to 38.79% of CO_2 emissions from the associated process during the period. Lime-containing materials in China accounted for 63.12 % of the total uptake, and among the three stages of the lime cycle, the utilisation stage accounted for the highest CO_2 sequestration (~74.05 %). The results also demonstrate that lime, which is usually omitted from emission inventories as a carbon sink, is very important to the carbon cycle. The present study indicates that the CO_2 uptake by lime can reduce the carbon footprint of lime production process and provide scientific proof for further research of the potential of lime-containing materials in carbon capture and storage. The ** data utilised in the present study can be accessed at https://doi.org/10.5281/zenodo.7112485 (Ma, 2022)

1 Introduction

According to the latest report (6th Assessment) of the Intergovernmental Panel on Climate Change (IPCC), anthropogenic activities are responsible for the unprecedented increase in the concentration of CO_2 in the atmosphere, which attained 415

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ppm in 2021 (NOAA. ESRL, 2022.). In 2019, approximately 24 % (14 Gt CO_2-eq) of the net global anthropogenic emissions originated from industrial sources, and lime production emerged as the second highest industrial source after cement production (IPCC, 2021; Shan et al., 2016). Similar to cement, lime is mainly produced via the heating of limestone ($CaCO_3$) in a kiln at temperatures of 900–1200 °C. The CO_2 generated during this process is commonly released into the atmosphere (Greco-Coppi et al., 2021). During limestone decomposition, fossil fuel combustion, which is used to provide energy for the process, is an indirect source of CO_2, but this is often accounted for in the energy sector (IPCC, 2021).

The enormous quantity of lime produced in the world (~430.0 Mt in 2020; USGS, 2022) is mainly employed in the following sectors: (1) chemical industry, such as for the production of precipitated calcium carbonate (PCC), manufacturing of paper, and refining of sugar; (2) environmental remediation/treatment, including water treatment, acid mine drainage, and flue gas desulphurisation; (3) metallurgical industry, for instance as a fluxing agent in the production of iron and steel; and (4) construction industry for building materials including lime mortar and lime-stabilised soil-asphalt mixtures (National Lime Association, 2020). Many lime-based materials, including wastes produced in different industries, re-absorb some of the CO_2 released, and thereby sequester CO_2 throughout the lime cycle (carbonation), owing to the unstable calcium oxide in these materials (Cizer et al., 2012a). According to Renforth (2019), approximately 34% of lime can directly or indirectly remove CO_2 from the atmosphere and absorb CO_2 during the utilisation stage. The carbonation process can be described using the following reactions:

$$CaO+H_2O=Ca(OH)_2 \tag{1}$$

$$Ca(OH)_2+CO_2=CaCO_3+H_2O \tag{2}$$

Carbonation proceeds progressively from the exterior to the interior of lime-containing materials via the diffusion of CO_2 into particles, followed by its reaction with hydration products of calcium oxide (Cizer et al., 2012b; Despotou et al., 2016). Therefore, carbonation can be considered as a mineralisation technology for carbon capture, utilisation, and storage (CCUS) (Lai et al., 2021; Snæbjörnsdóttir et al., 2020). Samari et al. (2020) indicated that lime-based materials have also been proposed as solid sorbents in direct air capture (DAC) technologies (extraction CO_2 directly from the atmosphere). In practice, however, because of material and environmental factors, only 70–80 % of the CaO in lime can be converted into $CaCO_3$ (Bhatia and Perlmutter, 1983). In previous studies, the carbonation process and factors influencing its rate (Ma et al., 2019), as well as strategies for improving the sequestration of carbon using lime-containing materials under controlled laboratory conditions (Pan et al., 2012; Baciocchi, 2017), have been examined. Pan et al. (2020), for instance, estimated the CO_2 reduction potential of lime-based solid wastes (e.g. lime mud, red mud and iron and steel slags) in mineralisation technologies, and highlighted a substantial potential for the storage of CO_2 in these wastes. The maximum achievable carbonation capacity of these solid wastes via direct mineralisation is approximately 310 Mt of CO_2 per year. Renforth (2019) estimated the global potential of CO_2 uptake through carbonation of lime and related alkaline materials up to the year 2100 (approximately 2.9–8.5 Gt of CO_2 per year), and indicated that this process can substantially mitigate CO_2 emissions during manufacturing of the associated materials. However, existing studies are limited to estimation of the carbon reduction potential via accelerated carbonation instead of carbon sequestration throughout the lime cycle under realistic conditions.

In the present study, a carbon sequestration analytical model was utilised to evaluate the global uptake of CO_2 by lime-containing materials during the three stages of the lime cycle from 1963 to 2020. The aims were to highlight the magnitude of the lime carbon sink on a global scale and to estimate the net CO_2 emission associated with the production of lime. In addition, characteristics of the uptake of CO_2 by lime and the contribution to the carbon cycle were examined. The present study significantly improves the global carbon uptake model and it provides theoretical support for the utilisation of lime-containing materials in carbon capture and storage (CCS).

2 Data and Methods

2.1 Lime production, resources usage proportion and treatment

In this study, China and the United States (U.S.) were considered individually, while all other producers were grouped together as "rest of the world" (ROW). The lime production data of China were obtained from China Statistical Yearbook (China Statistical Yearbook, 2022) and the data of U.S. and ROW were from (Lime Statistics and Information | USGS, 2022). However, data on lime production from 1963 to 2000 in China were not available in the existing databases. To ensure consistency with U.S. and ROW data, it was necessary to estimate the missing data in China. Since > 90 % of lime resources in China are used for construction and steel, calcium carbide, and alumina production (Almanac of the China Building Materials Industry, 2022), a regression model was thus built based on lime input and product output. The involving variables include completed floor areas (X1), crude steel production (X2), calcium carbide Production (X3), alumina production (X4) and lime production, which are provided in the China Statistical Yearbook (China Statistical Yearbook, 2022), China Chemical Industry Yearbook (China Chemical Industry Yearbook, 2022), and USGS (2022) (see the Supplementary Information). Due to the strong correlation between X1, X2, X3, and X4, we extracted xxx as the principal components through principal component analysis method. A linear regression model was then built with xx and lime production in known years (2002-2016). Through fitting analysis, we found that the principal component has a good fit with Chinese lime production, with correlation coefficient R of 0.987, and the corrected determination coefficient R2 of 0.972, which shows that the Chinese lime production obtained by the regression equation is reasonable.

Relatedly, according to data that were obtained from the USGS, approximately 15 %–42 % of lime resources in the U.S. are utilised in the chemical industry (mainly for petroleum refining and glass and rubber products production), whereas 30 %–51 % are employed in metallurgy (primarily in the production of crude steel), 5 %–14 % are used in the construction industry (principally for the production of lime stabilised soil (LSM) and LM), and approximately 8 %–43 % of the resources are applied in environmental protection and other fields. In the ROW, data on the usage of lime resources in different sectors including the industry were mostly obtained from publications (see the Supplementary Information).

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2.2 Estimation of emissions from processes

Regarding industrial processes, lime production is the second-highest source of carbon emissions after cement production, and thus, its contribution cannot be ignored (Shan et al., 2016a). CO_2 emissions from lime production are mainly linked to the calcination stage, during which calcium oxide (CaO or quicklime) is formed from the decomposition of limestone by heat (Despotou et al., 2016). This process is usually performed in a shaft or rotary kiln at high temperatures. The method recommended by the IPCC is widely used to estimate CO_2 emissions from industrial processes, and for the lime industry, three basic methodologies are utilised (IPCC, 2006). Considering the availability of lime production data, Method 1(multiplication of the regional lime production by the CO_2 emission factor) from the IPCC Guidelines for National Greenhouse Gas Inventories was utilised to calculate CO_2 emissions from lime production processes in the present, and this can be expressed as follows:

$$CE_{l,i} = m_{l,i} \times EF_l \tag{1}$$

where $CE_{l,i}$ is the annual CO_2 emissions, $m_{l,i}$ represents the production of the lime industry, and EF_l denotes the CO_2 emission factor associated with the lime production process.

Emission factors for the lime industry processes are determined using the composition of raw materials and the production technology. In the present study, 0.683-, 0.77-, and 0.75-ton CO_2 per ton of lime produced were adopted as emission factors for the US, China, and ROW, respectively (IPCC, 2006). Emission factors for the U.S. and ROW are those recommended for developed and developing countries according to the IPCC guidelines, whereas that for China was advanced by the National Development and Reform Commission of China.

2.3 Assessments of uptake during the lime cycle

Lime materials, which remove CO_2 from the atmosphere, belong to the following stages of the lime cycle: (1) production, (2) utilisation and (3) waste disposal (Fig. 1). Therefore, the CO_2 uptake by lime ($C_{l,total}$) was calculated using the following formula:

$$C_{l,total} = C_{l,pro} + C_{l,ser} + C_{l,wd} \tag{2}$$

where $C_{l,pro}$, $C_{l,ser}$, and $C_{l,wd}$ are the uptake components during the production, utilisation and waste disposal stages, respectively. The uptake of CO_2 in different stages of the lime cycle is examined subsequently.

2.3.1 Assessment of uptake during the production stage

The carbon sink of the lime production stage refers to the uptake of CO_2 by lime kiln ash, and this can be quantified using the following expression:

$$C_{l,pro} = m_{l,i} \times r_{lkd} \times f_{lkd}^{CaO} \times \gamma_{lkd} \times \frac{M_{CaO}}{M_{CO_2}} \tag{3}$$

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where $m_{l,i}$ is the quantity of lime produced, r_{lkd} represents the output rate of lime kiln ash, f_{lkd}^{CaO} denotes the concentration of CaO in dust, γ_{lkd} is the rate of conversion of CaO to $CaCO_3$ in dust, and M_{CaO} and M_{CO_2} are molar masses of CaO and CO_2, which in the present study are 56 and 44, respectively.

2.3.2 Assessment of uptake during the service stage

Processes that can absorb CO_2 in the lime utilisation stage principally comprise the production of Precipitated calcium carbonate (PCC, $C_{pcc,i}$), carbonation sugar (SUG, $C_{sug,i}$), lime-stabilised soil (LSS, $C_{lss,i}$) and lime mortar (MOR, $C_{mor,i}$). The uptake of CO_2 in this stage can be calculated as follows:

$$C_{l,ser} = C_{pcc,i} + C_{sug,i} + C_{lss,i} + C_{mor,i} \tag{4}$$

(1) Precipitated calcium carbonate and carbonation sugars

PCC is produced via the hydration of high-calcium quicklime, followed by a reaction of the resulting slurry and CO_2 (Wang *et al.*, 2002), and this reaction can be represented as follows: $Ca(OH)_2 + CO_2 = CaCO_3\downarrow + H_2O$. According to the law of conservation of mass, the uptake of CO_2 by lime in PCC can be calculated as follows:

$$C_{pcc,i} = m_{l,i} \times L_1 \times a_1 \times f_l^{CaO} \times \frac{M_{CaO}}{M_{CO_2}} \tag{5}$$

where L_1 is the proportion of lime that is used in the chemical industry, a_1 is the proportion of lime utilised in the chemical industry that is associated with PCC, and f_l^{CaO} is the concentration of CaO in lime. Similar to the principle of the carbon sink in the production of PCC, the uptake of CO_2 linked to the production of carbonation sugars (SUG) can be calculated using the following expression:

$$C_{sug,i} = m_{l,i} \times L_1 \times a_2 \times f_{sug} \times f_l^{CaO} \times \frac{M_{CaO}}{M_{CO_2}} \tag{6}$$

where a_2 is the proportion of lime used in the production of SUG in the industry and f_{sug} is the proportion of sugar produced using the lime-refining method.

(2) Lime-stabilised soil

Under wet conditions, the carbonation rate of a LSS is approximately between 70 %–80 % over a duration of three months (Liu et al., 2018b). Therefore, it is assumed that LSS can be carbonated within a year, and the uptake of CO_2 by LSS is quantified using the following expression:

$$C_{lss,i} = m_{l,i} \times L_2 \times a_3 \times f_l^{CaO} \times \gamma_{lss} \times \frac{M_{CaO}}{M_{CO_2}} \tag{7}$$

where L_2 is the proportion of lime used in the construction sector, a_3 represents the proportion of lime employed in LSS in the construction sector, and γ_{lss} is the rate of conversion of CaO to $CaCO_3$ in LSS.

(3) Lime mortar

MOR is mostly used for the plastering of interior walls, with a typical thickness of 20 mm (Almanac of China building materials industry, 2022). Under natural conditions, the estimated carbonation rate of MOR is 1 mm $d^{-0.5}$ (Ventol et al., 2011)). Therefore,

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according to Fick's law of diffusion, a year is insufficient for the complete carbonation of a MOR layer. Consequently, the uptake of CO_2 by MOR is calculated using the following expressions:

$$C_{mor,i}=m_{l,i}\times L_2\times a_4\times f_{mor,i}\times f_l^{CaO}\times\gamma_{mor}\times\frac{M_{CaO}}{M_{CO_2}} \quad (8)$$

$$d_{mor}=k_{mor}\times\sqrt{t_{mor}} \quad (9)$$

$$f_{mor,i}=(d_{mor,i}-d_{mor,i-1})/d_T \quad (10)$$

where L_2 is the proportion of lime used in the construction sector, a_4 denotes the proportion of lime in MOR that is utilised in the construction sector, $f_{mor,i}$ represents the carbonation ratio of MOR in the i-th year, γ_{mor} is the rate of conversion of CaO to $CaCO_3$ in MOR, $d_{mor,i}$ represents the depth of carbonation of MOR in the i-th year; k_{mor} denotes the rate of carbonation of MOR, t_{mor} is the duration of carbonation of MOR and d_T is the thickness of MOR.

2.3.3 Assessment of uptake during the waste disposal stage

Lime employed in the production of paper, aluminium, calcium carbide, and steel generates by-products including lime mud, red mud, carbide slag (CS, $C_{cs,i}$), and steel slag (SS, $C_{ss,i}$), respectively. The alkaline component (CaO) in these wastes absorb CO_2 under natural conditions.

(1) Lime and red muds

Lime mud particles that are involved in the production of paper are usually fine and evenly distributed (Ma et al., 2021). In fact, particles < 40 μm account for 93 %, and the associated water contents range from 39 % to 60 % (Qin et al., 2015). However, as a paste, the penetration of CO_2 to react with the lime mud is limited. Consequently, a year is usually insufficient for the complete carbonation of lime mud.

Red mud is also characterised by fine particles as well as a porous structure, high specific surface area, and good stability in water (Wang et al., 2019). Similar to the principle of the carbon sink for lime mud, a year is insufficient for the complete carbonation of red mud (Liu et al., 2018b). The uptake of CO_2 by lime in lime and red muds is calculated using the following expression:

$$\varepsilon_{m,ij}=m_{p,ij}\times r_{m,ij}\times f_{m,j}^{CaO} \quad (11)$$

where $\varepsilon_{m,ij}$ denotes the mass of CaO in wastes (j=lime mud or red mud) that can be carbonated in year I, $m_{p,ij}$ is the quantity of paper and paperboard/alumina that are produced in the i-th year, $r_{m,ij}$ is the output rate of waste j and $f_{m,j}^{CaO}$ represents the concentration of CaO in waste j.

According to Fick's law of diffusion, the depth of carbonation of waste j ($d_{m,ij}$) can be obtained from the carbonation rate ($k_{m,j}$) and carbonation time (t_i) using the following expressions:

$$d_{m,ij}=k_{m,j}\times(\sqrt{t_i}-\sqrt{t_{i-1}}) \quad (12)$$

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$$R_{m,ij}=\begin{cases}\dfrac{k_{m,j}\times\sqrt{t_i}}{\dfrac{h_{m,j}}{t_{m,j}}\times t_i} & (t_i\leq t_{m,j})\\ \dfrac{d_{m,ij}}{h_{m,j}} & (t_{m,j}<t_i<100)\end{cases} \quad (13)$$

where $R_{m,ij}$ represents the fraction of waste j that is carbonated in the i-th year, $h_{m,j}$ is the height of the waste j pile and $t_{m,j}$ is the duration of the yard of the waste j. Accordingly,

$$C_{m,ij}=\varepsilon_{m,ij}\times(1-f_{m,ij}^{use})\times R_{m,ij}\times\gamma_{m,j}\times\frac{M_{CaO}}{M_{CO_2}} \quad (14)$$

where $C_{m,ij}$ is the uptake of CO_2 uptake of waste j during the i-th year, $f_{m,ij}^{use}$ denotes the utilisation rate of waste j and $\gamma_{m,j}$ is the rate of conversion of CaO to $CaCO_3$ in lime mud.

(2) Carbide slag

Carbide slag comprises particles that are dominantly between 10–50 μm, which usually contain moderate amounts of water (Lin et al., 2006). Stacking for approximately 15 d can reduce the concentration of CaO by approximately 50% (Hao et al., 2013). The uptake of CO_2 by CS can be calculated using the following expressions:

$$\varepsilon_{cs,i}=m_{l,i}\times L_1\times a_5\times p_l^{cs}\times r_{cs}\times f_{cs}^{CaO} \quad (15)$$

$$C_{cs,i}=\varepsilon_{cs,i}\times(1-f_{cs}^{use})\times\gamma_{cs}\times\frac{M_{CaO}}{M_{CO_2}} \quad (16)$$

where $\varepsilon_{cs,i}$ is the mass of CaO in CS in the i-th year, a_5 denotes the proportion of lime in calcium carbide that is utilised in the chemical industry, p_l^{cs} represents the output of calcium carbide per ton of lime input, r_{cs} is the output rate of CS, f_{cs}^{CaO} is the concentration of CaO in CS, f_{cs}^{use} is the utilisation rate of CS and γ_{cs} is the rate of conversion CaO to $CaCO_3$ in CS.

(3) Steel slag

Steel slag cannot be carbonated within a year because its hydration commonly requires more than 4 years (Wang and Yan, 2010). In the present study, the SS particle was approximated as a uniformly-densified sphere. The fraction ($R_{s,i}$) of SS that is carbonated can be estimated using the following expressions (Xi et al., 2016):

$$D_{ss,i}=2d_{ss,i}=2k_{ss}\times\sqrt{t_i} \quad (17)$$

$$R_{s,i}=\begin{cases}100\%-\int_a^b\frac{\pi}{6}(\mathrm{D}-\mathrm{D}_{ss,i})^3/\int_a^b\frac{\pi}{6}\mathrm{D}^3\times100\% & (\mathrm{a}>\mathrm{D}_s)\\ 100\%-\int_{\mathrm{D}_0}^b\frac{\pi}{6}(\mathrm{D}-\mathrm{D}_{ss,i})^3/\int_a^b\frac{\pi}{6}\mathrm{D}^3\times100\% & (\mathrm{a}\leq\mathrm{D}_{ss,i}\leq\mathrm{b})\\ 100\% & (\mathrm{b}<D_s)\end{cases} \quad (18)$$

$$\Delta R_{s,i}=R_{s,i}-R_{s,i-1} \quad (19)$$

where $D_{ss,i}$ is the maximum diameter of SS that complete carbonation in the i-th year, $d_{ss,i}$ represents the depth of carbonation of SS in the i-th year, k_{ss} is the rate of carbonation of SS, t_i is the carbonation duration, D is the diameter of SS,

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a and b are the minimum and maximum SS diameters, respectively, and *a* and *b* represent the corresponding minimum and maximum diameters of SS particles in a given size distribution. The annual carbonation of SS ($C_{ss,i}$) can then be calculated using the following expressions:

$$\varepsilon_{ss,i} = m_{s,i} \times r_{ss} \times f_{ss}^{CaO} \quad (20)$$

$$C_{ss,i} = \varepsilon_{ss,i} \times \Delta R_{s,i} \times f_{ss}^{use} \times \gamma_{ss} \times \frac{M_{CaO}}{M_{CO_2}} \quad (21)$$

where $\varepsilon_{ss,i}$ is the mass of CaO in SS in the i-th year, $m_{s,i}$ represents the mass of crude steel that was produced in the i-th year, r_{ss} is the output rate of steel slag, f_{ss}^{CaO} is the concentration of CaO content in steel slag, f_{ss}^{use} is the ratio of SS that is utilised as stacking and roadbed material and γ_{ss} is the rate of conversion of CaO to $CaCO_3$ in SS.

2.4 Calculation of annual and cumulative uptakes

Even though the uptake of carbon can be estimated using alkaline materials in different stages of the lime cycle, the global and regional CO_2 absorption values were obtained via the aggregation of all alkaline materials. In the global and regional carbon sink accounting, parameters such as the production of lime, proportion of lime utilised in different sectors, diffusion or carbonation coefficient, output rate, concentration of CaO, conversion ratio of CaO to $CaCO_3$, particle size distribution and height of lime or red mud pile among others, were utilised as inputs for the model (see the Supplementary Information). Basically, for the uptake of CO_2 in year t_i, the cumulative uptake of CO_2 in year t_i minus that for year t_{i-1} can be obtained from the following expression:

$$\Delta C_{l,total}^{t_i} = \sum C_{l,total}^{t_i} - \sum C_{l,total}^{t_{i-1}}$$

and thus, the contribution of the annual uptake of carbon to the total carbonation can also be calculated.

2.5 Uncertainty analysis

A Monte Carlo analysis was used to determine the uncertainty associated with the uptake of carbon by lime. This uncertainty originates from input parameters (e.g. activity level data and carbon absorption factors) of the carbon sink accounting for model and quality of the statistics infrastructure. In the present study, 53 causes of uncertainty (see the Supplementary Information) associated with the estimated uptake of CO_2 were identified after more than 100,000 simulations.

3 Results

3.1 Aggregated regional and global emissions from the production of lime

Figure 2 shows the estimated CO_2 emissions from lime production processes in China, the U.S. and ROW from 1963 to 2020. Globally (Fig. 2a), emissions of CO_2 from lime production processes doubled from 0.15 Gt yr^{-1} (95 % CI:0.14–0.17 Gt) in 2002 to 0.30 Gt yr^{-1} (95 % CI:0.27–0.32 Gt) in 2020. During this period (2002–2020), the average annual rate of increase was

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3.41%, which is significantly higher than the rate for 1963–2002 (0.73%). The cumulative emissions of CO_2 from 1963 to 2020 is 10.17 Gt (95 % CI:9.06–11.32 Gt), but emissions decreased in 2009. This decrease was likely caused by the global financial crisis in 2008, during which downstream lime industries experienced severe problems, such as excess produce, low production quantities, and stiff competition (Dong et al., 2010). Subtraction of the amount of CO_2 absorbed from CO_2 emissions up to 2020 based on estimates in the present study produced a cumulative net emission of 9.06 Gt.

Changes in emissions of CO_2 from 1963 to 2020 in China are displayed in Fig. 2b, and these accounts for approximately 50% of the global emission. Alternatively, China was primarily responsible for the increase in the global emission from lime production processes during the period studied. In China, from 1963 to 2020, the average annual emission of CO_2 was 98 Mt yr^{-1}, and the average annual rate of increase was 2.13%. Notably, the rapid global increase in CO_2 emissions started in 2002. This finding is consistent with estimates from studies on the uptake of carbon by cement carbon based on similar approaches (Cui et al., 2019). These results are closely linked to the development of downstream sectors of the lime industry in China, such as the iron and steel, light and chemical, construction and materials industries (Shan et al., 2016b). By 2020, emissions of CO_2 in China attained to 211 Mt yr^{-1} (95 % CI:187–236 Mt), whereas the cumulative emission was 5700 Mt (95 % CI:4849–6628 Mt), and this accounted for 56.03 % of global CO_2 emissions associated with lime production processes. The result that was obtained in the present study is higher than the 172 Mt yr^{-1} that was forecasted for the same period by Tong et al. (2019), and this difference is attributed mainly to the emission reduction scenarios that are considered in each study.

In the U.S., from 1963 to 2020, CO_2 emissions from lime production processes remained at around 12 Mt yr^{-1}, and the cumulative emissions by 2020 were approximately 753 Mt, and this represents 13.21% of the global emission. This relatively low value is because of a fairly stable production of lime in the U.S. and significant import of lime from Canada (Lime Statistics and Information | U.S. Geological Survey, 2022). Relatedly, for the ROW, the cumulative emission was 71 Mt, and this represents 24.4% of the global emission.

3.2 Lime uptake of carbon by regions

According to the carbon sequestration model utilised in the present study to estimate the annual carbon in sinks in different regions, the global uptake of CO_2 by lime-containing materials increased from 38.25 Mt (95 % CI:27.85–51.38 Mt) in 1963 to 134.33 Mt (95 % CI:90.37–193.29 Mt) in 2020, and this represents an average annual rate of increase of 2.29%. Figure 3 shows the annual uptake of CO_2 in different regions and stages of the lime cycle between 1963 and 2020, whereas the area represents the cumulative uptake in each region under natural conditions. Cumulatively, 4053.61 Mt of CO_2 (95 % CI:3016.63–5251.90 Mt) were sequestered by lime-containing materials between 1963 and 2020. The highest sequestration was in China (~63.12 %, 2542.94 Mt) because of the associated high production of lime materials (China Statistical Yearbook, 2022), followed by the ROW (~33.71 %, 1358.23 Mt) and the US (~3.16 %, 127.41 Mt). Obviously, variation trends in the three regions from 1963 to 2020 are similar (see the Supplementary Information).

The cumulative uptakes of CO_2 by lime materials in different regions are displayed in Fig. 4. Notably, the top three lime-containing materials (LSS, MOR and SS) account for 81.75 % of the global uptake of CO_2 by such materials. Regarding China,

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the cumulative uptake of CO_2 by all lime materials was 2542.94 Mt, and the amount of CO_2 that was removed by LLS (1360.96 Mt) exceeds the sum removed by all other materials (Fig. 4a). In the U.S., the uptake was dominated by carbonating SS (Fig. 4b), LSS and SUG, and the cumulative amounts were 42.64, 22.57 and 21.06 Mt, respectively. In the ROW, SS (558.97 Mt), LSS (369.23 Mt) and MOR (188.37 Mt) constituted the top materials (Fig. 4c).

3.3 Uptake of CO_2 in different stages of the lime cycle

Among the stages of the lime cycle, the utilisation stage accounted for the highest uptake of CO_2 (2922.66 Mt) from 1963 to 2020, and this represents 74.05 % of the total. Relatedly, the uptake of CO_2 during the production and waste disposal stages were 101.66 and 933.61 Mt, respectively (Fig. 2b).

Since 1963, the production stage is associated with a significant amount of dust, which is a by-product of the production of lime, and the uptake of CO_2 by this dust in 2020 attained 2.95 Mt. This contribution is attributed to the development of the lime industry and the increase in the disposal of lime kiln dust (LKD) in landfills (Latif et al., 2015). The concentration of CaO in the ash of lime kilns is approximately 54.88%, and thus, this continuously absorbs CO_2 in landfills (Bobicki et al., 2012). The annual and cumulative uptake of carbon by lime materials during the utilisation stage varied significantly, but these produced the following trend: LSS > MOR > PCC > SUG (Table 1). As commonly used building materials, LSS and MOR correspondingly removed 1729.04 and 837.77 Mt of CO_2. Considering the consumption of lime in the construction sector over the past five decades and its increasing utilisation worldwide, especially in China and other developing countries, its uptake of CO_2 will certainly increase in the future (Renforth, 2019). The carbon fixation amounts of PCC and SUG of 233.30 and 125.54 Mt, respectively, accounted for < 10 % of the total uptake during the utilisation stage.

Regarding the waste disposal stage, the CO_2 absorption is mainly associated with carbonation of SS (Table 1). The cumulative uptake estimate in the present study is 721.33 Mt. The iron and steel industry, which is a basic industry in industrialised countries, produces approximately 180–270 Mt of SS annually (Iron and Steel Slag, 2022). However, the alkaline content of SS is due to the high amount of lime used in the iron and steel making process. Therefore, SS sequesters a high amount of CO_2 in stockpiles and as roadbed material (Bobicki et al., 2012). Owing to its elevated concentration of $Ca(OH)_2$, high specific surface area and efficient carbonation process, CS is linked to the sequestration of approximately 200.84 Mt of CO_2 (Huang et al., 2004; Hao et al., 2013). The uptake of red and lime muds is approximately 0.45 Mt of CO_2 (Table 1). This low uptake is assigned to the high content of water in these wastes, which hinders the diffusion of CO_2 into their particles under exposure.

4. Discussion

Although the national greenhouse gas inventories guideline involves methods for quantifying CO_2 emissions that are linked to lime production processes, the IPCC neglected carbon sequestration by lime (IPCC, 2006). According to the analysis conducted in the present study, the uptake by lime-containing materials rapidly increased from 1963 to 2020 in all stages of

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the lime cycle. In 2020, the global uptake of CO_2 by lime was equivalent to 2.15% of the global industrial emissions of CO_2; therefore, neglecting this sink caused an overestimation of the global carbon emission from industrial processes. Regarding the global carbon cycle, the annual carbon uptake by lime was approximately 1.65% of the average global forest ecosystem sink from 2001 to 2010, and this can explain approximately 1.55% of the missing global carbon sink (Data supplement to the Global Carbon Budget 2021, 2022). Therefore, if the lime sink is incorporated, the global carbon budget, which already includes data for carbon sinks of the ocean, land, and cement can be improved.

To further illustrate the function of lime as a carbon sink, results obtained in the present study were compared with data for the uptake of CO_2 by materials containing different minerals (Table 2). Rocks containing silicate and carbonate minerals are abundant in nature and are continuously extracting CO_2 from the atmosphere. According to recent studies, the annual average amounts of carbon sequestered by natural carbonate and silicate minerals are 3.26 and 0.13 Gt yr^{-1} (Li et al., 2018; Zhang et al., 2021). However, the weathering of these minerals resulting in sequestration of CO_2 from the atmosphere occurs over a timescale of at least 104 years (Berner et al., 1983).

Obviously, compared to natural carbonate and silicate minerals, the carbonation process involving alkaline materials produced by human activities, such as cement, SS and other solid wastes, is relatively faster under natural conditions (Berner et al., 1983). Lime materials, such as MOR and SS, similar to cement and natural materials, also serve for the removal of CO_2 from the atmosphere for several years or decades (Fig. 5). Alternatively, the uptake of CO_2 in each year involves lime materials that were generated or consumed in previous and current years: the former accounts for 17 % of the total uptake, whereas the latter represents 83 %. These results are inconsistent with those obtained for the cement carbon sink, where most of the carbon absorption is linked to previous years. This difference is attributed to the higher calcium content, smaller particle size, and more active chemical properties of lime materials. These characteristics suggest that lime-containing materials, especially LKD and SS, are suitable for carbon capture and storage via mineralisation. Therefore, promoting the carbonation process can mitigate impacts of CO_2 emissions (Pan et al., 2020).

5. Data availability

All the original datasets of CO2 uptake by lime are available at https://doi.org/10.5281/zenodo.7112485 (Ma et al.,2022).This dataset contains three data files, including lime material production and uses, lime carbon emission and uptake results, and the uncertainty of lime carbon emission and uptake.

6. Conclusion

In the present study, a carbon sequestration model was utilised to quantify the global uptake of CO_2 by lime-containing materials from 1963 to 2020. The national greenhouse gas inventories guideline methods and carbon budgets can be improved by considering lime as a carbon sink. The main findings of the present study are summarised below.

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Global CO_2 emissions from lime production processes increased from 38.25 Mt yr^{-1} in 1963 to 134.33 Mt yr^{-1} in 2020. However, the cumulative uptake of CO_2 by lime-containing materials (4053.61 Mt) did offset approximately 25.82 % of these emissions. The uptake was highest in China (2542.94 Mt) because of the associated elevated production and consumption of lime in recent decades, and this accounted for > 63.12 % of the global uptake. Uptakes in the ROW and U.S. were 1358.23 and 127.41 Mt, respectively.

The uptake of CO_2 by lime-containing materials at different stages of its cycle varied significantly. In the utilisation stage, lime-containing materials, especially lime-stabilised soil and lime mortar, contributed the most to the total lime carbon sink (2922.66 Mt). This was followed by sequestration in lime materials (mainly steel slag and carbide slag) in the waste disposal stage (933.61 Mt), whereas the production stage was associated with 101.66 Mt.

Historically, weathering of lime-containing materials was thought to occur over a large timescale. In the present study, it was revealed that approximately 17 % of the annual uptake of CO_2 originated from lime that was produced in previous decades; therefore, this absorption potential cannot be ignored. In the future, carbon capture and storage can be improved via the use of lime-containing materials (e.g. steel slag and lime kiln dust).

Author contributions. LB and MM designed the study and prepared the manuscript with assistance from FX, JW, and LL. LL and MM performed the analyses, with the help of FX and LB on the analytical approaches. MM, LN, and FC performed the post-processing and analysis of the data as well as the review of the paper. LL and LB established the lime carbon sink accounting database, whereas LB and FX wrote the code and performed simulations of the datasets, with assistance from LL, MM, and LN. FX conceptualised and supervised the study.

Competing interests. The authors declare that they have no conflict of interest.

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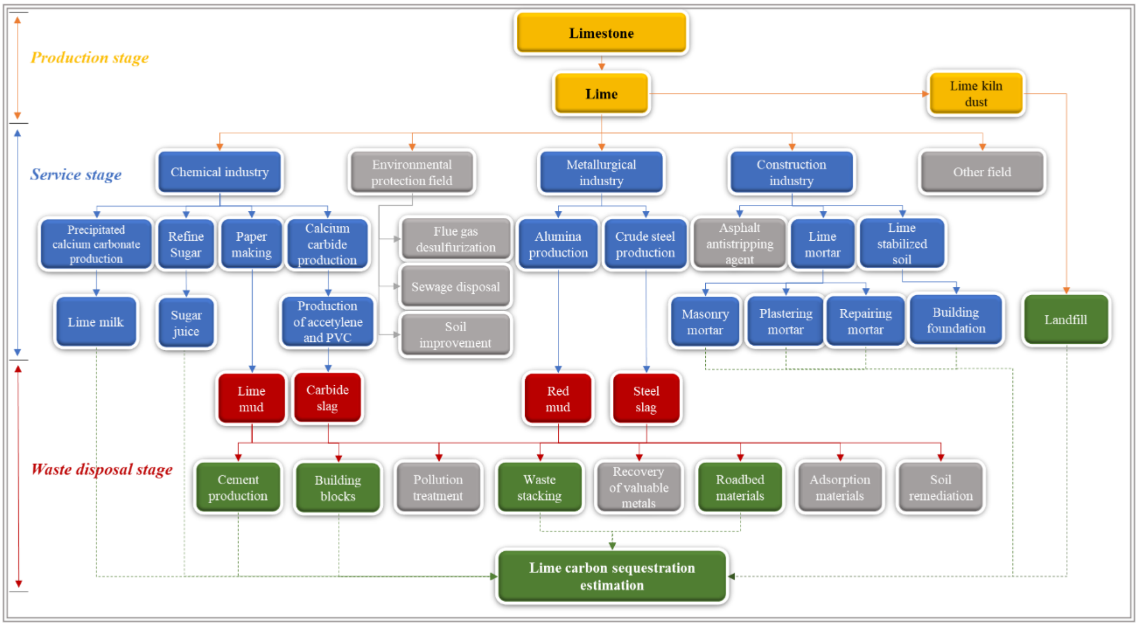
Figure captions



440 **Figure 1: System boundary for the sequestration of carbon by lime. Solid arrows represent the material flow, whereas dashed arrows indicate the carbon flow, and the double solid lines are accounting boundaries.**

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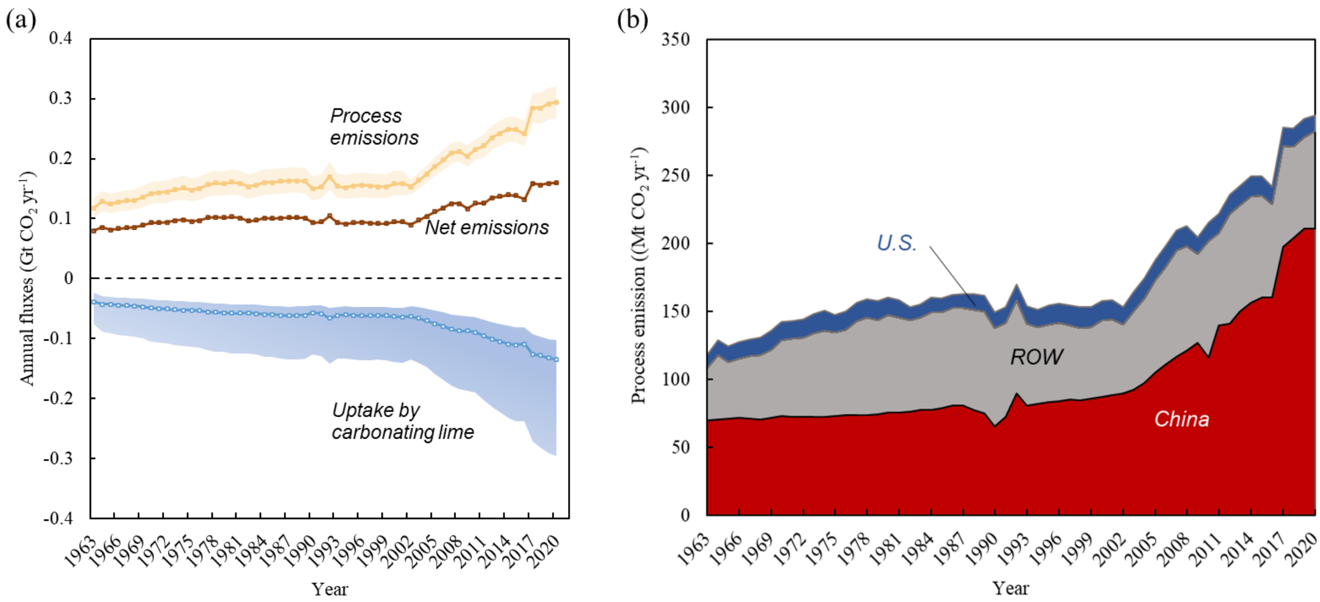

445 **Figure 2: (a) Annual CO_2 emissions from industrial processes and the associated uptake by lime from 1963 to 2020. (b) Country- and region-wise CO_2 emissions (median) generated by industrial processes from 1963 to 2020. ROW (Rest of World)**

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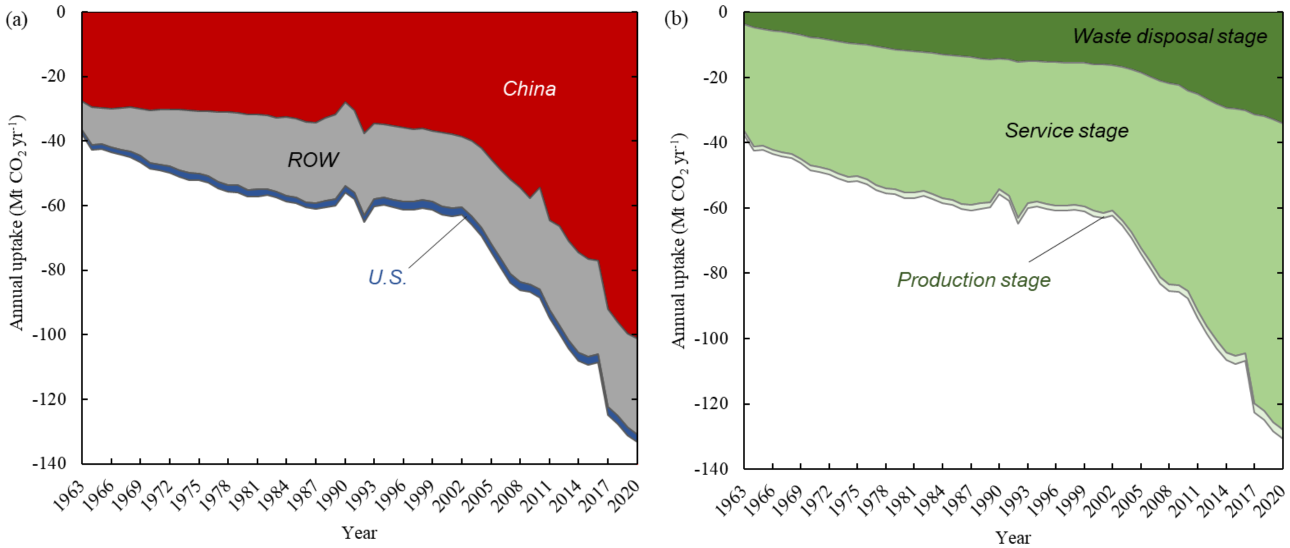

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Figure 3: Annual uptake of carbon dioxide by lime (a) in different regions and (b) during different stages of its cycle. ROW (Rest of World)

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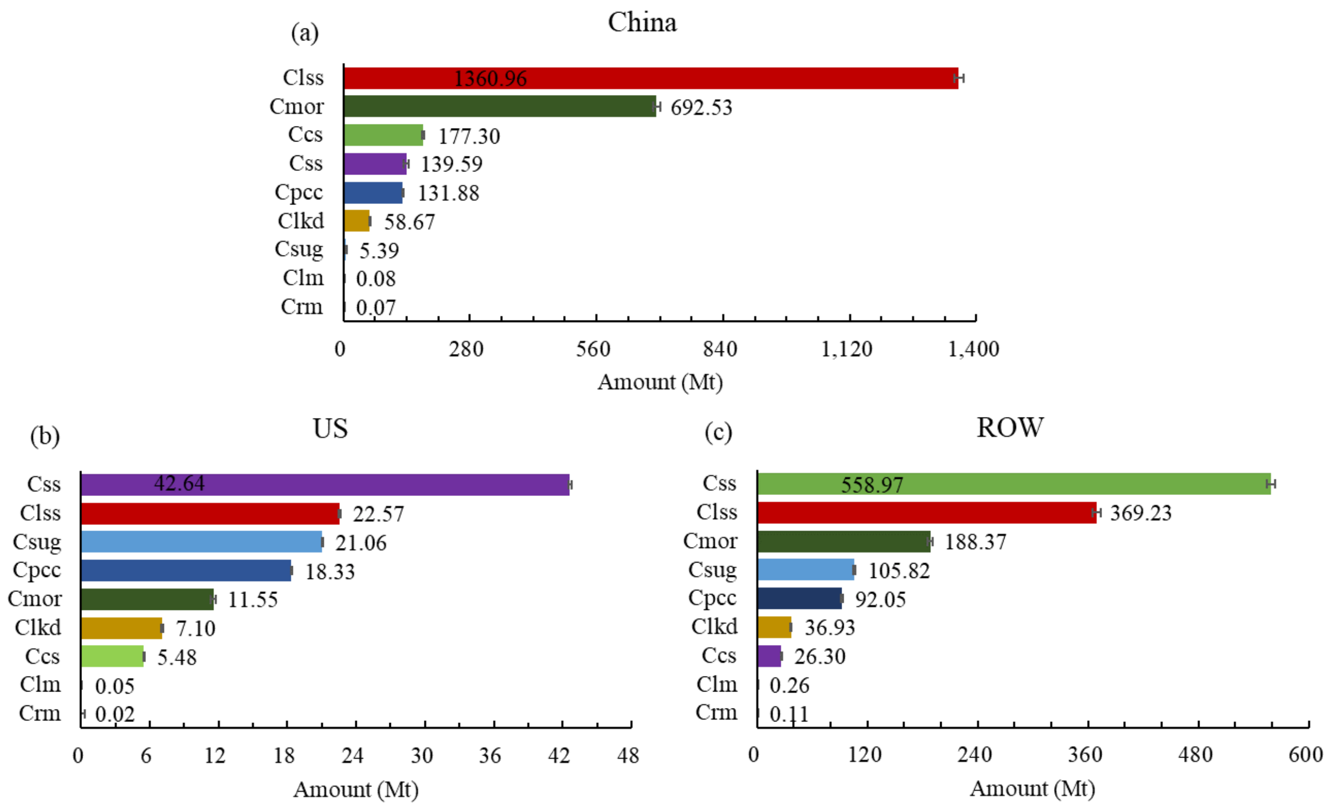

455 **Figure 4: Cumulative uptake of CO_2 uptake by lime-containing materials in different regions. ROW (Rest of World), Ccs (CO_2 uptake by carbide slag), Clkd (CO_2 uptake by lime kiln dust), Clss (CO_2 uptake by lime-stabilised soil), Cmor (CO_2 uptake by lime mortar), Cpcc (CO_2 uptake by Precipitated calcium carbonate), Crm (CO_2 uptake by red mud), Css (CO_2 uptake by steel slag), Csug (CO_2 uptake by carbonation sugar), Clm (CO_2 uptake by lime mud).**

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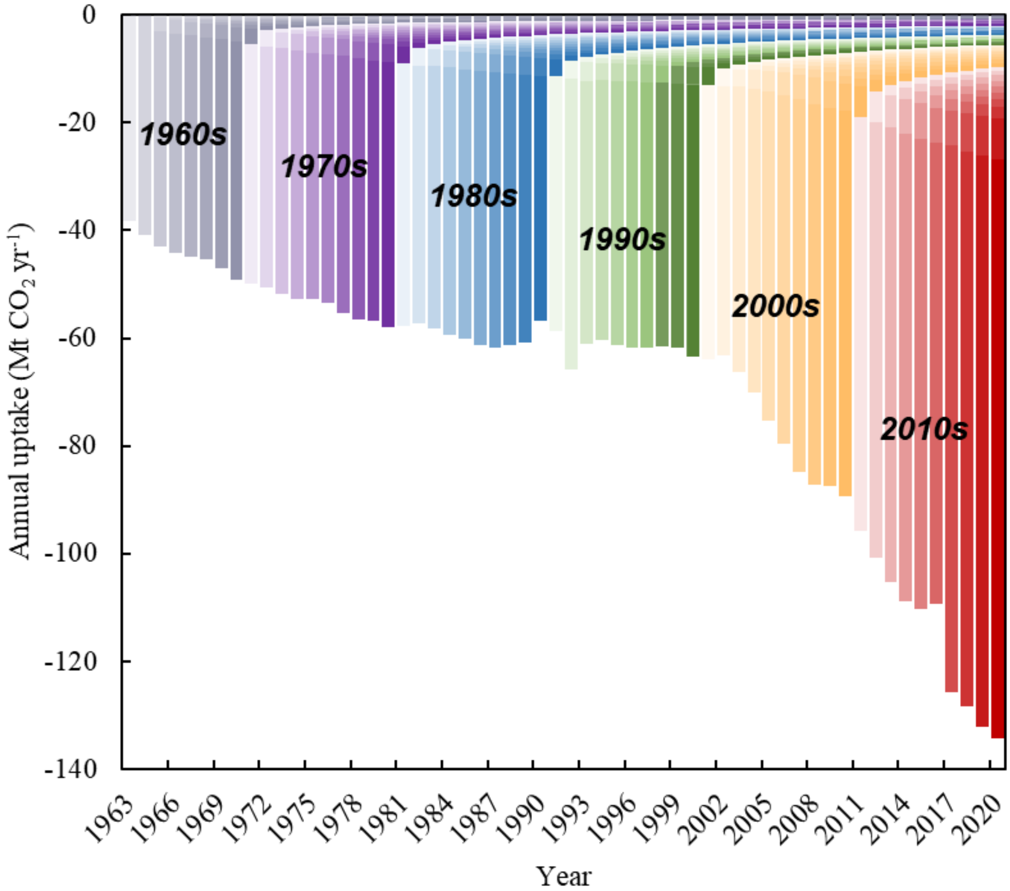

460 **Figure 5: Cumulative uptake of CO_2 by lime from 1963 to 2020.**

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465 **Table 1. Summary of the global uptake of CO_2 by lime-containing materials in different stages of its cycle**

Stage	Types of lime materials	CO_2 uptake in 2020 (Mt)	Cumulative CO_2 uptake from 1963 to 2020 (Mt)
Production	LKD	2.95	101.66
Service	LSS	57.03	1729.04
	MOR	27.51	834.77
	PCC	6.70	233.30
	SUG	2.43	125.54
Waste disposal	RM	0.01	0.15
	SS	27.34	721.33
	CS	6.84	200.84
	LM	0.01	0.29

LKD(Lime Kiln Dust), LSS (Lime-Stabilised Soil), PCC (Precipitated Calcium Carbonate), SUG (Carbonation Sugar), RM (Red Mud), SS (Steel Slag), CS (Carbide Slag), LM (Lime Mud)

470

Table 2 Comparison of CO_2 uptake by different types of materials

Region	Carbon sink type	Annual CO_2 uptake (Gt/yr)	Source
Global	Carbonate	2.42-4.11	(Li et al., 2018)
Global	Silicate	0.13	(Zhang et al., 2021)
Global	Lime	0.09-0.19	this study
Global	Cement	0.89	(Guo et al., 2021)
China	Steel slag	0.005	(Liu et al., 2018a)
China	Alkaline solid wastes	0.04-0.11	(Ma et al., 2022)