

Thank you for your precious comments and suggestions. Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. The responds to the reviewer's comments are as following:

- 1. Comments:** Ln. 12. Needs to be rewritten. Suggestion: "However, during the life cycle of lime production, the alkaline components of lime will continuously absorb CO₂ from the atmosphere during use and waste disposal."

Response: Thank you for your valuable suggestions on how to improve this sentence.

Changes: The sentence in line 12 has been rewritten.

- 2. Comments:** It seems important to add standard deviation for all your result values. You mention the annual carbon sequestration of lime accounts for about 1.03±?? %, what would be the standard deviation associated to this estimate for example?

Response: The lime carbon sequestration model uses 115 input parameters, which follow statistical distributions such as normal, uniform, and triangular distributions. Due to the non-linear simulation process, the overall distribution is unclear. To provide more information about parameter estimation properties (Efron, 1982), we have utilized the percentile CI method to estimate the uncertainty range. This method determines the upper and lower limits of the interval based on the 2.5th and 97.5th percentiles. For further information, please refer to the following reference: <https://epubs.siam.org/doi/book/10.1137/1.9781611970319>.

Changes: None.

- 3. Comments:** Ln.330. The annual carbon uptake by lime represents 1.65% of the global forest sink. This will result in a very low global annual carbon sink then. The global carbon sink is the sum of carbon sink from ocean, land and cement. So, the 1.65% of the global forest sink cannot be equal to 1.65% of the global carbon sink. Consequently, lime sink cannot explain the 1.55% of the missing global carbon sink. This paragraph does not seem correct. The net emissions of lime production and impact on global carbon budget is an important point that needs to be developed in your study. Please develop.

Response: Thank you for your valuable suggestion. We appreciate your insight and have carefully considered it. Upon further review, we have decided to revise the section you mentioned. We now recognize that quantitative comparisons between different types of carbon sinks may have little practical significance. Therefore, we have chosen to remove this part from the updated version of our content.

Changes: Ln 349-352, 'Regarding the global carbon cycle, lime's annual carbon uptake is estimated to be approximately 1.09% of the average global land carbon sink from 2010 to 2020, which was approximately 3.18 Gt C yr⁻¹ (Global Carbon Budget, 2022). This indicates that lime's contribution to the global carbon cycle is significant and should be taken into account when considering strategies to mitigate carbon emissions.'

- 4. Comments:** You mentioned in your reply that you used auto-regressive models and other methods to predict the data from 1963 to 2000, but the method used in your study obtained the largest coefficient of determination. This seems important enough to be mentioned in your manuscript. I would suggest adding the different methods coefficient in supplement information to justify why you used the linear regression method for your study.

Response: Thank you for your valuable feedback, which we have incorporated into our manuscript. We have added the parameters statistics of various regression models to the following tables, as per your suggestion, and marked the changes in line 90.

Changes: See the following tables for details (SI-2 Data 4)
 Statistics of estimated parameters in regression models for predicting lime production in China 1963-1995.

Model	Regression method	Dependent Variable	Independent variable		Estimate Std.	Error	t value	Sig.	F-statistic	p-value	R-square	Adjusted R-square
1	Principal Component Regression	Lime production of China from China Construction Material Industry Yearbook	Production of calcium carbide, crude steel, cement and alumina, and completed area	(Intercept)	142.093	3.021	47.03	< 2e-16***	306.9	5.20E-14	0.9360	0.9329
				Z1 ¹	22.582	1.289	17.52	5.2e-14***				
2		Lime production of China from USGS	Production of calcium carbide, crude steel, cement and alumina, and completed area	(Intercept)	75.516	6.118	12.34	4.55e-13***	378.9	<2.2e-16	0.9289	0.9265
				Z1	45.674	2.346	19.46	<2e-16***				
3 ²	Stepwise Linear	Lime production	Production of calcium	(Intercept)	81.96	8.19	10.008	2.72e-08***	862.6	<2.2e-16	0.9954	0.9942

	Regression	n of China from China Construction Material Industry Yearbook	carbide, cement and alumina, and completed area	calcium carbide	4.096	0.67	6.113	1.50e-05***				
completed area				0.035	0.007	5.232	8.22e-05***					
cement				-0.054	0.01	-5.607	3.94e-05***					
alumina				0.013	0.001	9.254	7.99e-08***					
4		Lime production of China from USGS	Production of crude steel	(Intercept)	4.53887	7.72733	0.587	0.561	472.7	<2.2e-16	0.9422	0.9402
				calcium carbide	0.30844	0.01419	21.741	<2e-16***				

1, One principal component (Z1) is obtained in principal component analysis

2, Model 3 was selected as the best regression method in this study

*** indicates a significant difference (p<0.001)

Fitted coefficients and standard errors of ARIMA models for predicting lime production in China 1949-1962.

Model	Fitting method	external regressor variable		Coefficients	Standard error	R-square
5	ARIMA(0,1,0)	Production of calcium carbide, cement and crude steel	crude steel	0.1368	0.0467	0.9828
			cement	-0.0588	0.0163	
			calcium carbide	3.7173	0.9585	

Fitted coefficients and standard errors of ARIMA models for predicting lime production in China 1930-1948

Model	Fitting method	External regressor variable		Coefficients	Standard error	R-square
6	ARIMA(0,2,2)	Without	MA1	-1.1748	0.1497	0.9786
			MA2	0.2902	0.1604	

Fitted coefficients and standard errors of ARIMA models for predicting lime production in global 1930-1962

Model	Fitting method	External regressor variable		Coefficients	Standard error	R-square
7	ARIMA(1,0,0)	Production of global crude steel, cement, and alumina	AR1	0.7929	0.0787	0.9849
			intercept	131322.7	11798.17	
			crude steel	0.0871	0.0274	
			cement	-0.0082	0.0104	
			alumina	1.0343	0.4059	

5. Comments: Supplement information SI data 4 of “Lime material production and uses”, how are calculated current year, previous year and total? What do the values represent? Captions to the different tables are missing.

Response: Thank you for your suggestion. We have readjusted the table and added corresponding notes. See SI-1 Data 4 of “Lime carbon emission and uptake results” for details. (<https://doi.org/10.5281/zenodo.7759053>)

Changes: See SI-1 Data 4 of “Lime carbon emission and uptake results” for details.

‘1. From a vertical perspective, the sum of the vertical data in the table represents the annual carbon sequestration of lime-based materials, which is the total amount of carbon sequestered. The diagonal data indicates the carbon sequestration amount of lime-based materials produced in the current year, whereas the amount of carbon sequestration in previous years can be calculated by subtracting the current year's value from the annual carbon sequestration. (For example, in 1935, the annual carbon sequestration of lime-based materials was 8.11651 million tons (Mt), of which 7.83204 Mt was due to the carbonization of lime-based materials produced in the same year. The remaining carbon sequestration amount of lime-based materials produced in the years 1930-1934 were 0.00149, 0.00135, 0.00116, 0.00245, and 0.34222 Mt, respectively, adding up to a total of 0.33448 Mt in historical years.);

2. Horizontally, the horizontal data refers to the annual carbon sequestration of lime-based materials produced in a certain year over time. Taking 1935 as an example, in addition to the carbonation that occurred in that year, the lime-based materials produced in 1935 absorbed CO₂ from the atmosphere annually from 1936 to 2020, with the amount of CO₂ absorbed declining from 0.31732Mt in 1936 to 0.00023Mt in 2020.’

6. Comments: It would be useful to precise which supplementary information table need to be read in the manuscript instead of just mentioning “see the Supplementary Information”. Additionally, might be better to have the supplementary Information in PDF as well.

Response: Thank you for your suggestion. We have incorporated your feedback into our manuscript. Specifically, we have added statements about specific tables to the supplementary information as per your suggestion. Additionally, we have also included an introduction to the attachment information in the "Data availability" section. We appreciate your input and guidance, and thank you for taking the time to review our work.”

Changes: Ln 380-401

‘SI-1 Lime carbon emission and uptake results, 1930-2020

Data 1. Annual carbon uptake by lime material and region

Data 2. Global carbon uptake by lime material and stage

Data 3. Global carbon uptake by region

Data 4. Annual global carbon uptake by lime material and relevant lag time, 1930 to 2020

Data 5. Cumulative process CO₂ emissions from lime production by region and category, 1930 to 2020

Data 6. Global process CO₂ emissions from lime production and carbon uptake by lime materials carbonation from 1930 to 2020

SI-2 Lime material production and uses, 1930-2020

Data 1. Lime production by region, 1930 to 2020

Data 2. Estimated production of lime in China, 1930 to 2020

Data 3. Estimated global lime production, 1930 to 2020

- Data 4. Parameters of lime production fitting model
- Data 5. Paper and paperboard production by region, 1930 to 2020
- Data 6. Steel production by region, 1930 to 2020
- Data 7. Alumina production by region, 1930 to 2020
- Data 8. Output rate by material
- Data 9. Estimates of lime used for different industries by region

SI-3 Uncertainty of lime carbon emission and uptake, 1930-2020

- Data 1. Variables considered in the uptake uncertainty analysis using a Monte Carlo method
- Data 2. The uncertainty of CO₂ emissions from lime production
- Data 3. The uncertainty of lime carbon uptake'

7. **Comments:** Figure 1. What are the meaning for the different colors? You mention in Figure caption “double solid lines”, I do not see any.

Response: The figure uses different colors to indicate various lime-based materials that are capable of absorbing CO₂ during different life cycles. Descriptions of different colors are supplemented in the notes to Figure 1. Additionally, the outermost border in the figure is indicated by 'double solid lines'

Changes: we have made modifications to Figure 1 and its annotations (Ln 122-127). Specifically, the changes we made are as follows:

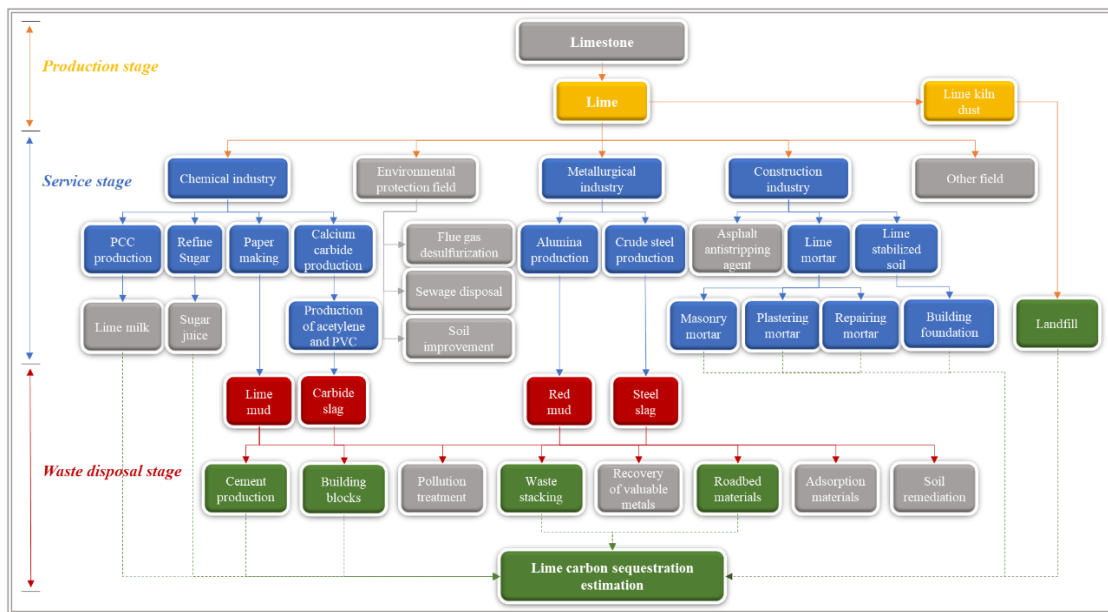


Figure 1: System boundary for the sequestration of carbon by lime. Solid arrows represent the material flow, dashed arrows indicate the carbon flow. (Yellow, blue, and red represent lime-based materials with carbon absorption capacity and their associated production processes, spanning from initial production through usage and waste disposal. Gray represents materials, production processes, or disposal methods with little carbon absorption capacity. Green represents the disposal method for lime-based waste that possesses carbon absorption potential.)

8. **Comments:** Ln. 257. “This figure is higher” do you mean the results of the figure are higher? How can your results be higher than Tong et al., 2019 if you considered an emission reduction scenario?

Response: Thanks very much for the opinion. Ln. 257 of the article compares our calculated data

on lime carbon emissions in 2020 (49.93Mt C yr⁻¹) with the Tong et al., 2019, in which China's lime carbon emissions in 2020 are predicted to be 46.91Mt C yr⁻¹, but this forecast considered an emission reduction scenario, they assumed the technology penetration rate of CCU would reach 5% by 2020 in China. However, as of 2020, the CCU technology was rarely utilized in China's lime industry. Consequently, the amount of carbon emissions produced by lime manufacturing is likely higher than in a scenario where CCU technology is used. Therefore, it is reasonable to assume that our calculations may have overestimated the carbon emissions generated by this industry. Our statement may not be clear enough, and we have revised it in Ln. 259.

Changes: Ln 260-264, 'The current figure exceeds the 46.91 Mt C yr⁻¹ forecasted for 2020 by Tong et al. (2019), which can be attributed primarily to the emission reduction scenarios they considered, assuming a technology penetration rate of 5% for CCU in China by 2020. However, it is important to note that as of 2020, CCU technology was seldom employed in China's lime industry. Therefore, the actual amount of carbon emissions produced by lime manufacturing is likely to be higher than in the scenario considered by Tong et al. Thus, our calculations are reasonable.'

9. Comments: The carbon sink increases with time but because the production has increase.

This increase for both the sink and the emission seems to be proportional to each other.

This should be mentioned in the discussion.

Response: Thank you for your suggestion. We have added a description of this section to the discussion of the new revised manuscript.

Changes: Ln 335-336, 'The carbon sink increases over time, but this increase is due to an increase in production. It seems that both the increase in the sink and the emissions are proportional to each other.'

10. Comments: Ln. 360. It is not correct to say that lime should be considered as a carbon sink.

The net emissions show a carbon source.

Response: Thank you for your opinion. We have revised this expression to make it more scientific

Changes: Ln 404-405, 'The national greenhouse gas inventories guideline and global carbon budgets could be improved by accounting for lime uptake, which can offset approximately 38% of emissions from industrial lime processes.'

11. Comments: One of your conclusions should be that the sink associated with lime life cycle should not be neglected and should be considered for future carbon cycle studies. However, there are still some questions not answered in your study about the emission inventories used here which could overestimate or underestimate lime production or lime sink and make your results biases. More development should appear in your discussion regarding these aspects.

Response: Thank you very much for your suggestion. It will be extremely valuable in guiding the revision of our paper.

Changes: Ln 335-348, 'The carbon sink increases over time, but this increase is due to an increase in production. It seems that both the increase in the sink and the emissions are proportional to each other. Our research results on carbon emissions and carbon absorption are significantly impacted by lime production. However, due to the lack of available data on annual lime production in China and worldwide during the early years, we used fitting methods to fill the gap of lime production and estimate it up to 1930. The statistically inferred 95% confidence interval was then used as the uncertainty range for lime production. To incorporate this uncertainty range into the accounting model for carbon sequestration and carbon emissions, we

used Monte Carlo simulations, and after 10,000 iterations, we obtained the final accounting results for carbon sequestration and carbon emissions. Therefore, from the interpolation of production data to the final accounting of carbon sinks and carbon emissions, all potential sources of uncertainty have been fully considered in the accounting process. Thus, this is a crucial way to obtain lime carbon sink and carbon emissions data from 1930 to 2020 under current data conditions. However, as our understanding of basic data and the mechanisms of lime production, carbon sequestration, and carbon emissions deepens, and as we improve our activity level data, such as lime-based material utilization, waste stacking, and recycling rates, and optimize carbonization parameters under different exposure conditions, there is still considerable potential for improving the accuracy of long time series lime material carbon sequestration and carbon emission accounting.’