

In response to 'SUMMARY and MAIN IMPRESSIONS': Thank you for recognizing the importance of this piece of research. Regarding the two issues you mentioned, on the former, we have presented the detailed information, including activity level data and carbon absorption factor data, as well as uncertainty analysis results by the Monte Carlo simulations, in the dataset at <https://doi.org/10.5281/zenodo.7112485>. And we will further improve the data information and description in the next revision. On the latter, according to the diffusion characteristics of CO₂ in different types of lime materials, we used the method of greenhouse gas inventory analysis, combined with Fick's second law, to build the mathematical model of lime carbon sink. See formula 2-21 for details. Furthermore, we will address your concerns in the following texts.

In response to 'GENERAL COMMENTS':

Abstract: Thank you for your suggestion. Lime is obtained by calcining limestone at high temperature, which will also produce by-product lime kiln ash. Globally, lime is mainly used in chemical industry, environmental protection, metallurgy, construction and other industries, and it can also be used to produce refractory materials. During use and waste disposal, lime will react with water and carbon dioxide in the air to produce calcium carbonate. According to the definition of "carbon sink" by IPCC, the carbon absorption of lime during use and waste disposal belongs to carbon sink, but has been ignored. We will add the clarification of lime carbon sink mechanism mentioned above in the revised abstract. As for another comment, the focus of this study is on the carbon sequestration accounting of lime. Therefore, more emphasis is placed on the carbon absorption of lime in this period. For clarification, we simply emphasized the proportion of lime carbon sink to lime carbon emission.

Ln 65. We are grateful for the suggestion. To be more clearly and in accordance with the reviewer concerns, we have added a more detailed interpretation as following: Due to the lack of accounting methods and the difficulty in obtaining data, the lime carbon sink has not been revealed. According to our calculation, the annual carbon sequestration of lime accounts for about 1.03% of the global terrestrial carbon sequestration. In addition to cement and alkaline solid waste, lime carbon absorption is also a carbon sink generated by human activities. The research results can explain the whereabouts of some missing carbon sinks in the global carbon sink, and then make up for the global carbon sink model.

Ln 79. We have re-fitted China's lime production. And relevant expressions have also been readjusted. The USGS records China's lime production from 1990 to 2020, but they are all estimates. In China, the only record of China's lime production is the "China Building Materials Industry Yearbook", whose values are provided by the China Lime Association. Its dataset is shown in Figure 1 for actual production, covering the years 1996-2014 and 2019-2020. Considering that the downstream sectors of lime in China mainly include construction, steel, calcium carbide, and alumina, we collected China's cement production (1930-2020), total housing area completed (1963-2020), steel production (1949-2020), calcium carbide production (1949-2020), and alumina production (1954-2020) as variables to fit the lime data. We have adopted the multivariate linear regression method you suggested. In order to avoid multicollinearity, a stepwise method is used to establish a model. The dependent variable in the model is lime yield, and the independent variable is calcium carbide yield, the completed area of houses in the whole society, cement yield and alumina yield. The determination coefficient of the model is 0.9954, and the adjusted determination coefficient is 0.9942. Given that data on

the completed area of housing in the whole society is limited to after 1963, the model only predicts lime production data from 1963-1995 and 2015-2018 (that is, the red line in Figure 1, the shaded part is the 95% confidence interval of the estimation result).

Ln 80. Thanks very much for reviewer's questions.

Data of lime production in 2001-2016 is compiled by the Chinese Lime Association, the only institution that records the production, and their figures are based on the demand of downstream consumer sectors, including crude steel, construction and chemical industry. Among them, the amount of lime used in crude steel, calcium carbide, construction and alumina accounts for more than 90% of the total lime production. Therefore, we select crude steel production, calcium carbide production, housing completion area and alumina production as the four variables for predicting lime production. Although there are no specific annual data records from 1963 to 2000, the use of lime in China has a long history. From the 7th century BC, lime became an important building material in China and began to be used, and according to the Chinese Statistical Yearbook, from 1963 to 2000, China was produced with calcium carbide, crude steel, alumina and other industrial products. Therefore, we assume that the data of China's lime production from 1963 to 2000 are still related to these four major industries. Your question is very reasonable. We also used autoregressive models and other methods to predict the data from 1963 to 2000, but the method mentioned in this article obtained the largest coefficient of determination.

For the other three regions, we have carried out the same analysis for the United States, the world and other countries. According to USGS records, the downstream use sectors of lime in the United States mainly include construction, chemical industry, metallurgy, environmental protection and refractory materials. We collected the crude steel production, alumina production and cement production in the United States from 1963 to 2020. The correlation between each variable is shown in the table below. It can be seen that there is a strong correlation between the lime production in the United States and each variable. In the construction of a general linear model, the determining coefficient of the model is 0.636.

Table 1 Correlation of various variables in the United States from 1963 to 2020

	crude steel production	alumina production	cement production	lime production
crude steel production	1			
alumina production	0.827**	1		
cement production	-0.017	-0.036	1	
lime production	0.335*	0.289*	0.716**	1

** . Correlation is significant at the 0.01 level (2-tailed).

* . Correlation is significant at the 0.05 level (2-tailed).

Likewise, for other countries, there is a strong positive correlation between lime yield and other variables. A general linear model is constructed with lime yield as the dependent variable and

other variables as the independent variable, and its determination coefficient is 0.749.

Table 2 Correlation of variables in ROW

	crude steel production	alumina production	cement production	lime production
crude steel production	1			
alumina production	0.942**	1		
cement production	0.903**	0.951**	1	
lime production	0.782**	0.613**	0.602**	1

** Correlation is significant at the 0.01 level (2-tailed).

From 1963 to 2020, the global lime production has a strong positive correlation with the global crude steel, alumina, and cement production. Taking the global lime production as the dependent variable and other variables as independent variables, the obtained model has strong collinearity. Therefore, the principal component analysis method is selected to obtain a principal component, and the coefficient of determination obtained by the model is 0.946.

Table 3 Correlation matrix between global lime production and various variables

	crude steel production	alumina production	cement production	lime production
crude steel production	1			
alumina production	0.984**	1		
cement production	0.985**	0.986**	1	
lime production	0.980**	0.964**	0.959**	1

** Correlation is significant at the 0.01 level (2-tailed).

Therefore, this linear relationship is also very obvious on a global scale, but this linear relationship is relatively weak in the United States and other countries.

Ln 96. According to the USGS, the data of global lime production began in 1963. And the "availability of lime production data" mentioned in this paper refers to the data currently available to statistical institutions or literature journals. There are many uncertain detailed data, which are included in the supplementary materials.

Ln 96. and Equation 1. We are very grateful to the reviewer for this comment. As we mentioned above, Tier 1 is based on the default emission factor of the national lime production process provided by the IPCC, and the proportion of usage of various lime uses; the emission factor in tier 2 is collected by national or plant level data; for Tier 3, the emission factor should be specific to the carbonate consumption process in each factory. The lime production data in this study belong to the national level, and the emission factor uses the default emission factor provided by the IPCC, which is not detailed to the factory level of each country, so Tier 1 method is selected; Thanks very much for reviewer's opinion. In Equation 1, l refers to different

types of lime use, including PCC, sugar making, lime-stabilized soil and lime mortar, and i refers to different years. We will add it in the revision.

Ln 101. I'm very sorry for the careless typo here. The emission factors used in our actual calculation are 0.683 for China, 0.77 for the United States, and 0.75 for other countries. Among them, we use IPCC2006 tier1 for the emission factors of the United States and other countries, and China's emission factor is based on China's provincial greenhouse gas emission guidelines, which is consistent with the research of Shan and is more in line with China's reality. In the IPCC guidelines, Equation 2.8 illustrates how to calculate the Tier 1 emission factor for lime production.

EQUATION2.8

$$EF_{lime} = 0.85 \times EF_{high\ calcium\ lime} + 0.15 \times EF_{dolomitic\ lime}$$

the $EF_{high\ calcium\ lime}$ is 0.75, and the default EF for dolomitic may be 0.86 or 0.77 for developed countries and developing ones. Substituting it into the formula, the emission factors of developed and developing countries are obtained as 0.77 and 0.75, respectively.

When setting the uncertainty of emission factors for different countries, we considered the emission factors for all lime types mentioned in the IPCC guidelines. The minimum value of the emission factor is 0.59, that is, the hydraulic lime type, and the maximum emission factor is 0.86 for developed countries. For developing countries, it is 0.77. Therefore, in our model, the maximum and minimum emission factors for China and ROW are 0.77 and 0.59, respectively, and 0.86 and 0.59 for the United States.

Equation 3. The ash yield of different lime kilns is mainly obtained through literature collection (the ash yield of lime kilns is between 0.09 and 0.1). As an important calculation parameter of lime kiln ash, it participates in the simulation of 100000 times by Monte Carlo method.

Section 2.3. Thank you for your suggestion. Because there are many parameters involved in this step, we put the relevant reference sources in the SI file, and the link is <https://doi.org/10.5281/zenodo.7112485>. We are happy to add this part in the next revision

Ln193. We can agree on this. The lime carbon sink model here is constructed according to the diffusion law of CO₂ in different lime materials. For the carbon sink model of each material, see the public trial 2-21. The Ln 193 part represents the accounting method of the annual lime carbon sink. The description of the carbon sink will also be added in the next revision.

Ln195. Because different lime materials have different carbon absorption rates, some alkaline solid wastes with large particle size cannot be completely carbonized within one year. Therefore, to calculate the carbon sink of lime materials in the current year, the accumulated carbon sink of lime in t_i year should be subtracted from the carbon sink of lime in t_{i-1} year. In the next version, we will also add a description of this part.

Section 2.5. The input variables and parameter distribution of uncertainty are shown in the annex. For your suggestions, we will also add relevant descriptions of uncertainty in the next version

Section 2.5. Uncertainty analysis description and carbon sequestration analytical model will be further improved in the next manuscript. In response to the question about

Ln.207-211. (1) Reasons for the decline in other years: In addition to 2008, other significant carbon emissions decline years include 1982, 1990, 1993, 2002 and 2016.

(2) China's lime carbon emissions have risen sharply since 2002, with an average annual growth rate of 4.86% from 2002 to 2020, compared with an average annual growth rate of 0.65% from 1963 to 2002. This is mainly due to the steady growth of China's macro economy after 2002.

(3) The results in Ln 204-211 of the article are based on our model's calculations, which we will describe in a revised draft thanks to the expert's reminder.

(4) Thank the experts for their suggestions on net emissions, and we will add this part to the revised version.

Ln.214. Thanks to the advice of reviewer, the average annual growth rate of China's lime carbon emissions from 2002 to 2020 was 4.86%, which was higher than the global average annual growth rate of 3.69% during the same period.

Ln 231. According to bp 'bp Statistical Review of World Energy', The total global carbon dioxide emissions in 2020 is 32.284 billion tons. In 2020, the total global carbon dioxide emissions will be 32.284 billion tons. In the same year, the carbon emission of lime industry process was about 293.98 million tons, accounting for 0.91% of the global total carbon emissions. For different regions, China, the United States and other countries account for 2.13%, 20% and 30% of the total carbon emissions of the lime industry respectively

Figure 3.b. mainly shows the carbon sequestration of lime in different life cycle stages. We have revised 'Figure 2.b' to 'Figure 3.b' in Ln 247.

Ln. 248. Well, the carbon absorption of dust is increasing. We will introduce the carbon absorption in 1963 to further improve the description.

Ln. 270. Lime carbon sink and industrial process emissions show an upward trend from the trend. We show the change trend of emissions and absorption in different regions in Figure 2b and Figure 3a. The carbon emission of lime refers to the emission of limestone calcination production, which is the carbon emission of industrial process. In Figure 3b, we show the carbon absorption in the global life cycle. The carbon sink results calculated by us increase with time, which can explain this situation.

Ln. 285 and Figure 5. the cumulative sources of CO₂ as well as the cumulative net CO₂ emissions will be showed in the next revised draft. And we will add further discussion about the net emissions.

Ln. 300. In fact, our logic is: In our study lime carbon sink has been revealed, which is one of the important destinations of carbon emissions in the lime industry process, in other words, some industrial process carbon emissions are absorbed back by lime. If carbon sink is considered in the carbon emission accounting of industrial process, we can determine the net

carbon emission, then we have solved the problem that the lime industrial process is overestimated.

In response to ‘SPECIFIC COMMENTS’:

Ln. 14. The existing data refer to the data available in the national statistical yearbook, literature journals, international databases, etc. This part of data is included in the annex, and we will display it in the text according to your suggestions. We rephrased Ln14-16: Here, existing data on materials associated with the production, utilisation, and disposal stages of lime-containing materials were analysed using lime carbon sink accounting model to obtain regional and global estimates for the sequestration of carbon from 1963 to 2020.

Ln 15. The model name is ‘lime carbon sink accounting model’.

Ln 20. Yes. This process refers to the industrial process of lime carbon emission. We will improve the description of this part.

Ln. Yes. Total global uptake.

Ln 62. Agree with your opinion. These three parts will be supplemented.

Ln. 72. Yes, your statement is more precise. ‘There was no data during this period in the China Statistical Yearbook.’

Equation 2. ‘l’ in Equation 2. refers to lime

Ln 79. We have revised the estimation of lime production simulation. We have adopted the multivariate linear regression method. In order to avoid multicollinearity, a stepwise method is used to establish a model.

Ln 94-96. Rewritten sentence: Lime comes from the decomposition of limestone in shaft kiln or rotary kiln, and the carbon emission of this industrial process is estimated from the IPCC method (IPCC, 2006).

Ln 98. It refers to the current stage, or today.

Ln 105. Thanks to the advice of reviewer, The references here is “PR China National Development and Reform Commission. Guidelines for provincial greenhouse gas inventories; 2011. [Chinese Document].” We will add it to the revised draft.

Ln. 117. The problem has been corrected. Unit: g/mol

Ln. 155. It usually takes several years or even decades.

Ln 204 Thanks very much for reviewer’s reminder, we will add explanations on a global scale.

Ln. 445. This part is changed to: Annual CO₂ emissions from industrial processes and the associated uptake by lime from 1963 445 to 2020 at a global scale.

Equation 11. Indexes ‘m’ means red mud or lime mud; ‘p’ means production.

Ln. 182. In Equation 18.

Ln. 193. Change ‘utilised’ to ‘utilized’

Figure 1. mainly shows the situation of different life cycle stages of lime

Ln. 445. OK.

Figure 2. In fact, the global annual carbon emissions of lime come from our calculations. And we will make this part easier to understand.

Figure 2.a. Shadows represent uncertainty ranges.

Ln. 232 The areas with different colors represent the accumulated value of lime carbon sink in different areas.

Ln. 205. Thanks very much for reviewer’s reminder, we will add the meaning of CI (Confidence

Interval).

Ln. 233. 4053.61 Mt of CO₂ is for global scale.

Ln. 273-275. Specific values in Gt/yr will be mentioned in the next revision.

Ln. 288. Thank you for your modification. Your statement is more scientific and rigorous

Ln. 294. Chang 'CO2' to 'CO₂'.

Ln. 301-302. Change 'emission' to 'absorption' in line 301.