

Dear Editor and reviewer:

Thank you for your precious comments and advice. 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:

In response to 'Time period': We are very grateful to the reviewer for this comment. In this study, in order to establish the lime carbon sink database, we collected data on the production of lime, paper and paperboard, crude steel, alumina in different countries and different years, the proportion of lime usage in various fields, such as chemical (including PCC production, sugar, paper, calcium carbide, others), metallurgy (crude steel, alumina, others), construction (lime stabilized soil, lime mortar, others), environmental protection and others in different countries and different years, the proportion data of paper white sludge, calcium carbide slag, steel slag, red mud utilization and CaO content, etc.

Considering that the lime production data in the United States can be traced back to 1930, and the lime production data for earlier years in China and the world are not recorded. Therefore, we decided to fill this gap by fitting methods, thereby extending the time scale of the study to 1930.

First of all, we fitted China's lime production. The only source of China's lime production statistics is the "China Building Materials Industry Yearbook", which records the lime production data from 1996 to 2020, of which the data from 2015 to 2018 is missing; in addition, the statistical yearbook also introduces the use of lime in various industries. From this, we know that the production of lime in construction, steel, calcium carbide and alumina in the downstream sector of lime accounts for more than 90% of lime production, which is the main factor affecting lime production. Therefore, we collected China's cement production (1930-2020), the completed area of housing in the whole society (1963-2020), steel production (1949-2020), calcium carbide production (1949-2020), and alumina production (1954-2020) were fitted to the lime production data. Taking China's lime production as the dependent variable, the stepwise linear regression method was used to construct a regression model. The independent variables entering the model include calcium carbide production, the completed area of houses in the whole society, cement production and alumina production. Since the completed area data of houses in the whole society was only extended to 1963, the model only predicted the lime production data from 1963 to 1995. Then, through the ARIMA (0,1,0) model, with external control variables including the steel production, calcium carbide production, and cement production, we fit the lime production in China from 1949 to 1962 (the steel and calcium carbide production data were only extended to 1949). Finally, we use the ARIMA (2,2,0) model without external control variables to fit the lime production in China from 1930 to 1948. So far, we have obtained the fitted lime production data for China from 1930 to 2020.

After obtaining the Chinese lime production data, we corrected the global lime production data from the USGS from 1963 to 2020. The ARIMA (1,0,0) model was then used to fit the global lime production from 1930 to 1962 with global alumina production, steel production and cement production as external control variables.

As stated by the reviewer, the year 2020 as the termination year is related to the year of data update, and the starting year was set to 1963 for the following reason: In the report for cement from 1930 to 2019 in ESSD 2021 (Guo et al. <https://doi.org/10.5194/essd-13-1791-2021>), the data source for the cement production is United States Geological Survey (USGS), which

provides data since 1930. Similarly, in our study, global and U.S. lime, crude steel, and alumina production data were obtained from the USGS: *Bureau of Mines Minerals Yearbook* (<https://www.usgs.gov/centers/national-minerals-information-center/minerals-yearbook-metals-and-minerals>). However, in this database, lime production data are recorded from 2000 for China and from 1963 for the world. To maintain the consistency of data years and maximize the time scale, we took 1963 as the starting year, and fitted the missing lime production data in China from 1963 to 2000 to build the lime carbon sink database for 1963 to 2020.

In response to ‘Problem with single years’: Thanks very much for reviewer’s opinion.

a) According to our calculation, the global CO₂ emissions of lime production process in 2020 are 293.99 Mt, of which China, the United States and other countries emit 211.00, 71.72 and 11.73 Mt, respectively. Numerically, compared with 2019, except the United States, the CO₂ emissions of China, the rest of world and the world total have slightly increased, but at a small growth rate.

b) As mentioned in our methodology, the annual CO₂ emissions of the lime industry are closely related to its production, and in the case of China, the lime production was not seriously affected by COVID-19 pandemic in 2020, and USGS statistics show the same production in 2020 as in 2019 (310 million tons, as shown in the table 1 below), but as reviewer said, the statistics have considerable uncertainty, where Table 2 shows the results of CO₂ emission distribution of China’s lime production for 2018-2020 after 100,000 simulations, and it can be seen that although there is no difference between the production data of the two years, their carbon emission calculation results still have differences. The difference is about 0.02%. Considering the stochastic of the Monte Carlo simulation, this very small difference is acceptable.

Table 1 Lime production data for different regions 2018-2020

Lime production	2018	2019	2020
The United States (thousand tons)	18000	16900	15800
China (thousand tons)	300000	310000	310000
The rest of world (thousand tons)	106000	105100	101200
World total (thousand tons)	424000	432000	427000

Data from United States Geological Survey (USGS). Lime Statistics and Information. <https://www.usgs.gov/centers/national-minerals-information-center/lime-statistics-and-information>

Table 2 Distribution of China's lime production process CO₂ emission results from Monte Carlo 100,000 simulations for 2018-2020 (unit: Mt)

percentile	2018	2019	2020
2.5	180.63	186.48	186.64
5	183.74	189.88	189.85
50	204.08	210.97	211.00
95	225.15	232.65	232.66
97.5	228.70	236.44	236.37

c) 2020 is a remarkable year, according to the report of Global carbon budget 2021 (<https://doi.org/10.5194/essd-14-1917-2022>), Global fossil CO₂ emissions were 5.4 % lower in 2020 than in 2019, because of the COVID-19 pandemic, with a decline of 0.5 GtC to reach 9.5 ± 0.5 GtC (9.3 ± 0.5 GtC when including the cement carbonation sink) in 2020. Nevertheless,

the carbon emission reductions in 2020 due to the COVID-19 are mainly in the transport sector, which is associated with the transport and logistics control measures to overcome the epidemic. However, lime industrial production had limited or no impact from the epidemic. From the production point of view, the COVID-19 pandemic does not affect the technological level, nor the supply capacity of lime industrial industry. In addition, the COVID-19 epidemic happened mainly during the gathering period of major festivals, such as the Spring Festival in China. This period is supposed to be the low season for industrial production, and enterprises will stop working, which further reduced the impact suffered by the lime industry. Therefore, the carbon emission during lime production in 2020 also showed a more stable trend.

In response to ‘Impact estimate’: Thank you for your comments on this part. The global industrial process carbon emissions accounted for 24% of all direct anthropogenic emissions in 2019 (IPCC AR6), and the lime production process belongs to the industrial sector in the IPCC accounting. In addition to the carbon emissions caused by the burning of fossil fuels used for calcining limestone and purchased electricity (the fuel burning and electricity emissions are accounted in the energy sector), about 70% of carbon emissions in the lime industry are from the emissions of lime industrial processes, that is, the emitted from the production of lime by heating and decomposition of limestone $\text{CaCO}_3 \xrightarrow{\text{heat}} \text{CaO} + \text{CO}_2 \uparrow$). The ‘associated process’ (line 20) implied the lime process that generates carbon emissions, i.e. the lime production process. We agree with the reviewer's comment and in the next revision we will clarify the relevant statement from the reader's perspective.

According to Shan's study (<https://doi.org/10.1016/j.apenergy.2015.04.091>), lime production is the second largest industrial process carbon emission after cement production. Like cement production, lime will produce a large number of lime-based materials during its production, use and waste disposal. These materials are prone to carbonation under natural conditions. Therefore, when we study its carbon emissions, it is also very necessary to explore how large the carbon sink of this alkaline material is. Like cement, our calculation of carbon sequestration process of lime-based materials can compensate for the partial IPCC methodological deficiency of lime carbon sequestration. We compared the lime carbon sequestration with the lime industrial process carbon emissions. The lime carbon sequestration was 134.33Mt, accounting for 38.79% of the lime industrial process carbon emissions. Obviously, the lime carbon sink is of great significance to the lime industry and the industrial carbon balance. We agree with the reviewer, and will explicitly clarify the importance of the lime carbon sink for the carbon balance of the industrial sector using our own calculation in the next revision. Furthermore, through our estimation that the annual carbon sequestration of lime in 2020 is comparable to the global annual averaged carbon sequestration of siliceous rocks, it can be indicated that the carbon sink of lime-based materials had an incremental impact on the global carbon cycle year by year under the influence of human activities. This is also equivalent to approximately 1.03% of the global terrestrial annual carbon sink from 2010 to 2020, which can interpret the whereabouts of some global missing carbon sinks, and is of significance for the global carbon cycle research.

In response to ‘Suggest’: Thank you for your suggestions.

a) We used the Monte Carlo method to estimate the CO₂ uptake of lime materials and the ultimate result was measured by the statistical indicator MEDIAN, and the 95% confidence

interval as a range of uncertainty. Therefore, the final result can only be considered as approaching the true value with a higher probability, but not the true value. This is the most probable approximation in the situation where the true value cannot be obtained.

b) It is correct that, as the reviewer suggested, that we can also use C to indicate the impact. However, to facilitate the harmonization with our previous research published in ESSD (<https://essd.copernicus.org/articles/13/1791/2021/>), here we also adopt CO₂ as the unit of denotation.

In response to the questions about 'Reference part', thank you for your comments. At present, the article is more about the reference of historical years in lime carbon sink accounting, and we will further add the content and literature related to the impact of climate change in the next revision.

In response to the questions about 'Posting uncertainties in separate file to actual data': We thank the reviewers for their careful and detailed comments, which prompted us to reconsider the presentation of the data from the reader's perspective. Putting the input parameters of the model, uncertainties, and simulation results in one file does seem to confuse the reader. In the next revision, we will make a change to post the uncertainties and Monte Carlo simulation results in separate files to more clearly show the sources of uncertainties, the processing and generation of the data, and the ultimate accounting results.

In response to the 'Questions validation': we thank the reviewer for the concern of data accuracy, and we fully agree with the importance of data accuracy. Our explanation is that in this paper, we employed the IPCC method, i.e. the inventory analysis method, to calculate the carbon sequestration of lime materials, i.e. the carbon sequestration of lime materials is equal to the activity level data multiplied by the sequestration factor, so it was not necessary to evaluate the accuracy of the model itself. The focus of our work was on how to minimize the range of uncertainties that exist in the parameters themselves, and improve the hit probability of the final results by reducing the parameters' uncertainties. As mentioned above, the reviewer presented a comparison of lime sinks with GHG emissions and with cement sinks; we also carried out a comparison of lime sinks with cement sinks and industrial process carbon emissions in our paper, but strictly speaking, these were not validations of the model. In the framework of Monte Carlo simulation, the modelled result is only the best possible approximation of the true value. It is a statistical probability, with stochastic characteristics, therefore normally no validation is required.