

## A 23 m.y. record of low atmospheric CO<sub>2</sub>

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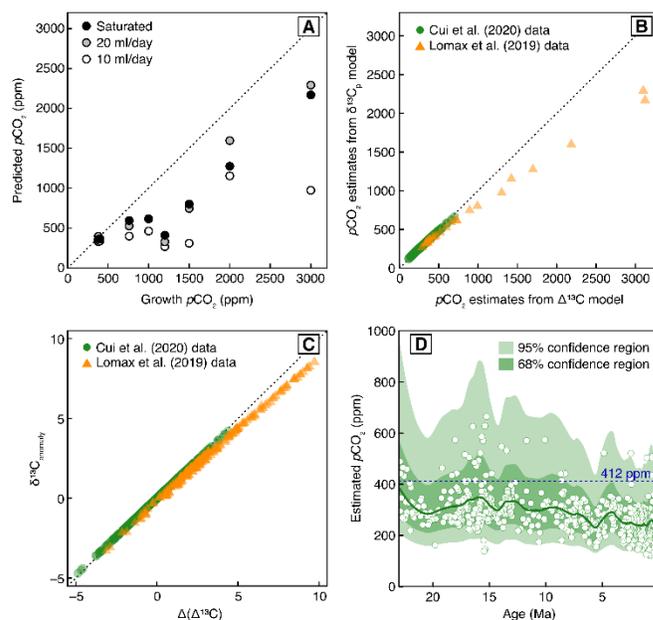
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In their recent paper, Cui et al. (2020) used a new iteration of their C<sub>3</sub> plant proxy to reconstruct *p*CO<sub>2</sub> over the past 23 m.y. The initial version of this proxy used carbon isotope discrimination ( $\Delta^{13}\text{C}$ , calculated as the offset between the  $\delta^{13}\text{C}$  of plant tissue [ $\delta^{13}\text{C}_p$ ] and atmospheric CO<sub>2</sub> [ $\delta^{13}\text{C}_{\text{atm}}$ ]) to estimate paleo-CO<sub>2</sub> (Schubert and Jahren, 2015), but recent work by different research groups has questioned the utility of this proxy (e.g., Kohn, 2016; Stein et al., 2020). Previously, we used  $\Delta^{13}\text{C}$  data from *Arabidopsis thaliana* plants grown experimentally under different moisture and *p*CO<sub>2</sub> conditions to show that this proxy is strongly impacted by variations in moisture availability and underpredicts *p*CO<sub>2</sub> (Lomax et al., 2019). Here, we argue that the new version of the C<sub>3</sub> proxy presented by Cui et al., which is centered on  $\delta^{13}\text{C}_p$  rather than  $\Delta^{13}\text{C}$ , suffers from the same shortcomings. Therefore, it is unsuitable for addressing the core question posed in their paper, that is, how *p*CO<sub>2</sub> levels in the geological past compare with those both in the present and predicted for the near future.

Using the new  $\delta^{13}\text{C}_p$  proxy to reconstruct *p*CO<sub>2</sub> from our existing *A. thaliana* data set (Lomax et al., 2019; Jardine and Lomax, 2021) shows that, like its predecessor, this proxy underestimates *p*CO<sub>2</sub> (Fig. 1A), although the effect is even more pronounced than previously (Fig. 1B). The proxy struggles to successfully predict *p*CO<sub>2</sub> for plants grown in >400 ppm conditions, which is particularly problematic because this is the core threshold for assessing whether past *p*CO<sub>2</sub> values exceed those of today. *p*CO<sub>2</sub> estimates are likely lower in this iteration of the proxy because rather than deriving a new relationship between  $\delta^{13}\text{C}_p$  and *p*CO<sub>2</sub>, Cui et al. used the model parameters (the A, B, and C terms) from their  $\Delta^{13}\text{C}:\textit{pCO}_2$  curve (Schubert and Jahren, 2015). However, the  $\delta^{13}\text{C}_{\text{anomaly}}$  term of Cui et al. (see their equations 1 and 2) does not equal the  $\Delta(\Delta^{13}\text{C})$  term of Schubert and Jahren (2015, see their equations 1 and 4) (Fig. 1C). The result is that *p*CO<sub>2</sub> predicted from  $\delta^{13}\text{C}_p$  is even lower than *p*CO<sub>2</sub> predicted from  $\Delta^{13}\text{C}$ , with the downward bias becoming particularly apparent at *p*CO<sub>2</sub> > 400 ppm (Fig. 1B).

As with the  $\Delta^{13}\text{C}$  version of the C<sub>3</sub> proxy, the new  $\delta^{13}\text{C}$ -based proxy is impacted by moisture availability, especially at higher *p*CO<sub>2</sub> levels (Fig. 1A). This is a critical issue in the time series presented by Cui et al., because hydrological changes are likely to have accompanied *p*CO<sub>2</sub>-driven temperature changes, for instance, across the mid-Miocene Climatic Optimum, ca. 17–14 Ma (Loughney et al., 2020). The extent to which the increase in *p*CO<sub>2</sub> reconstructed for this time by Cui et al. (Fig. 1D) is due to increases in moisture availability cannot be evaluated with this proxy, nor can the impact of long-term continental drying through the late Neogene on the overall downward *p*CO<sub>2</sub> trend.

Cui et al. used Monte Carlo resampling to quantify uncertainty in their *p*CO<sub>2</sub> reconstruction, and presented these uncertainties via a LOWESS smoother with a 68% confidence interval. A 68% confidence interval represents an abnormally low level of statistical confidence, and is too narrow to robustly determine whether *p*CO<sub>2</sub> values in the past exclude today's levels or those of the future. Plotting 95% confidence intervals (and therefore utilizing the usual  $\alpha = 0.05$  level for statistical inference) shows that *p*CO<sub>2</sub> values of >500 ppm are entirely consistent with Cui et al.'s reconstruction for much of the past 23 m.y., including in the Pliocene and Pleistocene. The C<sub>3</sub> proxy therefore fails to reject elevated *p*CO<sub>2</sub> conditions for the late Neogene and Quaternary, despite the downward biasing in the *p*CO<sub>2</sub> estimates themselves (Fig. 1D).



**Figure 1.** Reanalysis of the C<sub>3</sub> proxy. (A) *p*CO<sub>2</sub> predicted using the  $\delta^{13}\text{C}_p$ -based C<sub>3</sub> proxy of Cui et al. (2020) versus actual (growth) *p*CO<sub>2</sub>, from the data set of Lomax et al. (2019). Points are colored by water treatment. (B) Comparison of estimated *p*CO<sub>2</sub> using the  $\Delta^{13}\text{C}$ -based C<sub>3</sub> proxy of Schubert and Jahren (2015) and the  $\delta^{13}\text{C}_p$ -based C<sub>3</sub> proxy of Cui et al. (2020). (C) Comparison of the  $\Delta(\Delta^{13}\text{C})$  term of Schubert and Jahren (2015) and the  $\delta^{13}\text{C}_{\text{anomaly}}$  term of Cui et al. (2020). (D) The time series presented by Cui et al. (2020), based on their  $\delta^{13}\text{C}_p$ -based C<sub>3</sub> proxy, with a LOESS smoother and both 68% and 95% confidence intervals.

Understanding the relationship between *p*CO<sub>2</sub> and global climate is vital for forecasting the response of the climate system to anthropogenic CO<sub>2</sub> emissions. As such, *p*CO<sub>2</sub> proxies are essential, but they need to be robust and thoroughly validated. Terrestrial fossil organic carbon may be ubiquitous in sediments, but because of the impact of moisture availability on  $\delta^{13}\text{C}_p$ , and the inadequately derived relationship between  $\delta^{13}\text{C}_p$  and *p*CO<sub>2</sub> used by Cui et al., we maintain (Lomax et al., 2019) that the C<sub>3</sub> proxy is not suitable for reconstructing *p*CO<sub>2</sub> in the geological past.

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