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A benthic $\delta^{13}\text{C}$ -based proxy for atmospheric pCO_2 over the last 1.5 Myr

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[1] A high-resolution marine proxy for atmospheric pCO_2 is needed to clarify the phase lag between pCO_2 and marine climate proxies and to provide a record of orbital-scale pCO_2 variations before the oldest ice core measurement at 800 ka. Benthic $\delta^{13}\text{C}$ data should record deep ocean carbon storage and, thus, atmospheric pCO_2 . This study finds that a modified $\delta^{13}\text{C}$ gradient between the deep Pacific and intermediate North Atlantic ($\Delta\delta^{13}\text{C}_{P-NI}$) correlates well with pCO_2 . $\Delta\delta^{13}\text{C}_{P-NI}$ reproduces characteristic differences between pCO_2 and ice volume during Late Pleistocene glaciations and indicates that pCO_2 usually leads terminations by 0.2–3.7 kyr but lags by 3–10 kyr during two “failed” terminations at 535 and 745 ka. $\Delta\delta^{13}\text{C}_{P-NI}$ gradually transitions from 41- to 100-kyr cyclicity from 1.3–0.7 Ma but has no secular trend in mean or amplitude since 1.5 Ma. The minimum pCO_2 of the last 1.5 Myr is estimated to be 155 ppm at ~920 ka. **Citation:** Lisiecki, L. E. (2010), A benthic $\delta^{13}\text{C}$ -based proxy for atmospheric pCO_2 over the last 1.5 Myr, *Geophys. Res. Lett.*, 37, L21708, doi:10.1029/2010GL045109.

1. Introduction

[2] The 800-kyr record of atmospheric carbon dioxide concentration from Antarctic ice cores [Petit et al., 1999; Monnin et al., 2001; Siegenthaler et al., 2005; Lüthi et al., 2008] correlates well with Antarctic temperature [Jouzel et al., 2007] and many paleoclimate proxies from marine sediments (e.g., global ice volume [Hansen et al., 2007], sea surface temperatures [Lea, 2004]). However, age models for these two climate archives are developed independently of one another and have relative uncertainties of 5–10 kyr before 50 ka [Lisiecki and Raymo, 2005; Parrenin et al., 2007; Loulergue et al., 2007]. Therefore, the phase of marine climate proxies relative to atmospheric pCO_2 remains uncertain, preventing reconstruction of the sequence of climate responses associated with pCO_2 change before 50 ka. A high-resolution marine proxy for pCO_2 could solve this problem and extend pCO_2 estimates beyond the oldest ice core measurement.

[3] Alkenone $\delta^{13}\text{C}$ and boron-based proxies reconstruct pCO_2 concentrations in the surface ocean, but currently these records lack orbital-scale resolution and have error bars of at least ± 19 ppm [Hönisch et al., 2009; Tripathi et al., 2009; Pagani et al., 2009; Seki et al., 2010]. Existing higher-resolution benthic $\delta^{13}\text{C}$ records also have the potential to record changes in atmospheric pCO_2 because glacial-

interglacial changes in pCO_2 are associated with changes in the ΣCO_2 and $\delta^{13}\text{C}$ of the deep ocean [Oppo and Fairbanks, 1990; Flower et al., 2000; Hodell et al., 2003; Köhler et al., 2010]. This study empirically evaluates several possible benthic $\delta^{13}\text{C}$ -based proxies and uses the one best correlated with ice core pCO_2 to evaluate the phase lag between pCO_2 and benthic $\delta^{18}\text{O}$ and to estimate pCO_2 from 1.5–0.8 Ma.

2. Background

[4] Decreased deep water ventilation and increased Southern Ocean productivity are thought to reduce glacial pCO_2 by removing carbon from the surface and sequestering it in the deep ocean [e.g., Toggweiler, 1999; Brovkin et al., 2007; Martínez-García et al., 2009]. These processes also decrease the $\delta^{13}\text{C}$ value of deep waters as sinking low- $\delta^{13}\text{C}$ organic carbon remineralizes at depth and glacial overturning is decreased [Toggweiler et al., 2006; Köhler et al., 2010]. Reduced terrestrial carbon storage is the only glacial process for which pCO_2 and benthic $\delta^{13}\text{C}$ changes are not positively correlated, increasing atmospheric pCO_2 but decreasing mean ocean $\delta^{13}\text{C}$ [Shackleton, 1977; Brovkin et al., 2007; Köhler et al., 2010]. A carbon cycle box model that includes all of these processes predicts a strong, linear relationship ($r = 0.98$) between pCO_2 and deep Pacific $\delta^{13}\text{C}$, but the observed correlation is much weaker ($r = 0.5$) due to low-frequency (~400-kyr) variations in $\delta^{13}\text{C}$ not observed in pCO_2 [Köhler et al., 2010].

[5] The $\delta^{13}\text{C}$ gradient between deep and intermediate waters ($\Delta\delta^{13}\text{C}_{D-I}$) has also been suggested as proxy for pCO_2 because it should reflect the ΣCO_2 concentration gradient between the deep ocean and better-ventilated intermediate ocean [Oppo and Fairbanks, 1990; Flower et al., 2000; Hodell et al., 2003; Toggweiler et al., 2006]. $\Delta\delta^{13}\text{C}_{D-I}$ may also remove the mean-ocean $\delta^{13}\text{C}$ signal associated with changes in terrestrial carbon storage. Previous comparisons of $\Delta\delta^{13}\text{C}_{D-I}$ and pCO_2 yielded moderate correlations but did not extend beyond 400 ka [Oppo and Fairbanks, 1990; Flower et al., 2000; Hodell et al., 2003].

3. Proxy Evaluation

[6] Here I evaluate Pacific $\delta^{13}\text{C}$, $\Delta\delta^{13}\text{C}_{D-I}$ and the average of the two as possible proxies for pCO_2 . The signal-to-noise ratio of $\delta^{13}\text{C}$ signals is enhanced by averaging data from multiple cores within the same watermass to produce $\delta^{13}\text{C}$ stacks for the deep Pacific, deep South Atlantic, and intermediate North Atlantic (Figure 1 and Table S1 of Text S1 of the auxiliary material) [Lisiecki, 2010].¹ The deep Pacific stack is used for the deep-intermediate gradient ($\Delta\delta^{13}\text{C}_{P-NI}$)

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Figure 1. Benthic $\delta^{13}\text{C}$ stacks and potential pCO_2 proxies. (top) Regional stacks of benthic $\delta^{13}\text{C}$ from the intermediate North Atlantic (1145–2300 m depth, dashed line), South Atlantic (3700–4620 m, dotted), and equatorial Pacific (2520–3850 m, gray). (bottom) Pacific $\delta^{13}\text{C}$ stack (gray solid), $\Delta\delta^{13}\text{C}_{P-NA}$ (dashed), and the average of the two, $\Delta\delta^{13}\text{C}_{P-N/2}$ (black solid). All three records are adjusted to have a mean of zero.

because watermass boundary movement provides an additional source of $\delta^{13}\text{C}$ variability in the deep South Atlantic [Venz and Hodell, 2002] that does not affect the deep Pacific [Matsumoto *et al.*, 2002; Lisiecki, 2010]. For further discussion see the auxiliary material. Although Pacific $\delta^{13}\text{C}$ has approximately half the glacial-interglacial amplitude of South Atlantic $\delta^{13}\text{C}$, the following analysis is not highly sensitive to which deep water stack is used because $\Delta\delta^{13}\text{C}_{P-NA}$ and $\Delta\delta^{13}\text{C}_{SA-NA}$ are well correlated from 800–0 ka ($r=0.79$).

[7] Pacific $\delta^{13}\text{C}$ and $\Delta\delta^{13}\text{C}_{P-NA}$ both correlate moderately well with pCO_2 , but their average $\Delta\delta^{13}\text{C}_{P-N/2}$ produces the best correlation (Table 1 and Figure 1, bottom). Based on inter-core variability, $\Delta\delta^{13}\text{C}_{P-N/2}$ has a 1- σ uncertainty of 0.11‰ from 0.8–0 Ma and 0.13‰ from 1.5–0.8 Ma (equivalent to 17.5 ppm and 19.2 ppm, respectively). When $\Delta\delta^{13}\text{C}_{P-N/2}$ is scaled to the mean and standard deviation of ice core pCO_2 , it has a root mean square error (RMSE) relative to pCO_2 of 17.5 ppm, whereas boron-based estimates have RMSE of 18.1 ppm [Hönisch *et al.*, 2009] and 24.9 ppm [Tripathi *et al.*, 2009] (see auxiliary material).

[8] One possible physical explanation for the correlation between pCO_2 and $\Delta\delta^{13}\text{C}_{P-N/2}$ is that $\delta^{13}\text{C}$ variability in the deep and intermediate Atlantic may be amplified relative to their Pacific and global mean counterparts, e.g., due to changes in temperature and/or deepwater formation processes in the Atlantic. Thus, $\Delta\delta^{13}\text{C}_{P-N/2}$ corrects for differences in the amplitudes of variability between the Atlantic and Pacific. Alternatively, if Pacific $\delta^{13}\text{C}$ and $\Delta\delta^{13}\text{C}_{P-NA}$ are influenced differently by additional climatic processes, the average of the two should amplify the pCO_2 signal common to both.

[9] The correlation between marine proxies and pCO_2 is not a perfect evaluation metric because it depends on the particular marine and ice core chronologies used; therefore, $\Delta\delta^{13}\text{C}_{P-N/2}$ is also evaluated by whether it replicates features of the pCO_2 record that are independent of age model. Here

I focus on features that differ from the benthic $\delta^{18}\text{O}$ record [Lisiecki and Raymo, 2005] of deep water temperature and global ice volume. pCO_2 and northern hemisphere ice volume changes may differ if pCO_2 is controlled by southern hemisphere processes that are only weakly coupled to northern hemisphere climate [Toggweiler, 2008].

[10] One notable difference between $\delta^{18}\text{O}$ and pCO_2 (Figure 2, top) is that pCO_2 generally reaches its minimum early in each glaciation and then remains constant (e.g., Marine Isotope Stage (MIS) 6 and 12) or increases slightly (e.g., MIS 16) whereas benthic $\delta^{18}\text{O}$ does not reach its glacial maximum until immediately before each termination, due to continuing ice sheet growth [Thompson and Goldstein, 2006; Lea *et al.*, 2002]. Glacial trends in $\Delta\delta^{13}\text{C}_{P-N/2}$ match those of pCO_2 , with $\Delta\delta^{13}\text{C}_{P-N/2}$ reaching a minimum 30–40 kyr before the $\delta^{18}\text{O}$ maximum during most glaciations (Figure 2, bottom). Additionally, $\Delta\delta^{13}\text{C}_{P-N/2}$ correlates better with the magnitudes of glacial pCO_2 minima than $\delta^{18}\text{O}$ does. In $\Delta\delta^{13}\text{C}_{P-N/2}$ and pCO_2 , MIS 12 is less extreme than MIS 8, 10, and 16, whereas in $\delta^{18}\text{O}$ MIS 12 is similar to MIS 16 and more extreme than MIS 8 and 10. Thus, $\Delta\delta^{13}\text{C}_{P-N/2}$ reproduces

Table 1. Proxy Correlation With pCO_2 for 800–0 ka

Proxy	Correlation
N. Atlantic $\delta^{13}\text{C}^a$	0.19
Deep S. Atl $\delta^{13}\text{C}^a$	0.69
Deep Pacific $\delta^{13}\text{C}^a$	0.66
$\Delta\delta^{13}\text{C}_{P-NA}^a$	0.58
$\Delta\delta^{13}\text{C}_{P-N/2}^a$	0.75
$\log(\text{alkenone conc.})^b$	−0.71
Tropical SST stack ^c	0.64

^aSee auxiliary material for component records.

^bSite ODP 1090 [Martínez-García *et al.*, 2009] (see auxiliary material for age model).

^cHerbert *et al.* [2010].

Figure 2. Comparison of pCO_2 (gray) [Petit et al., 1999; Monnin et al., 2001; Siegenthaler et al., 2005; Lüthi et al., 2008] with (top) benthic $\delta^{18}\text{O}$ (black) [Lisiecki and Raymo, 2005] and (bottom) $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ (black). Glacial stages are labeled by MIS number. In Figure 2 (bottom), pCO_2 has been smoothed with a 2-kyr boxcar filter.

many pCO_2 responses that are independent of age model uncertainty and differ from ice volume change.

4. Termination Lags Between pCO_2 and $\delta^{18}\text{O}$

[11] Comparison of $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ and the ice core pCO_2 record provides an opportunity to link marine and ice core age models. Abrupt increases in pCO_2 and $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ have similar ages on their respective age models (Table S2), suggesting that the marine and ice core age models [Lisiecki and Raymo, 2005; Parrenin et al., 2007; Loulergue et al., 2007] are consistent to within 2.7 kyr during terminations. However, age model evaluation away from terminations is hampered by weaker correlation of the records' suborbital-scale variability.

[12] Climatic lags between pCO_2 and ice volume during terminations are evaluated by comparing $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ and benthic $\delta^{18}\text{O}$ changes within marine sediments. During most terminations $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ leads $\delta^{18}\text{O}$ by 0.2–3.7 kyr, but $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ lags $\delta^{18}\text{O}$ by 9.8 and 3.5 kyr during Termination 6 (535 ka) and MIS 18 (745 ka), respectively (Table S3). These lags are also found between benthic $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ within individual Pacific cores (Figure S2). An anomalous phase relationship between ice volume and pCO_2 may explain why these two warming events are weaker than most Late Pleistocene terminations. During both “failed” terminations, the initial $\delta^{18}\text{O}$ change is approximately half the amplitude of most Late Pleistocene terminations; $\delta^{18}\text{O}$ spends ~20 kyr at intermediate values of 3.8–4.2‰ and then briefly returns to more glacial values before achieving full interglacial conditions ~40 kyr after the initial warming. The $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ lag during these two failed terminations suggests that full deglaciation requires an early pCO_2 response.

[13] The initial trigger for terminations and the mechanistic link between pCO_2 and northern hemisphere ice volume

remain controversial [e.g., Huybers, 2009; Denton et al., 2010]. Variability in the phase between $\delta^{18}\text{O}$ and $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ supports the hypothesis of Toggweiler [2008] that glacial changes in pCO_2 are controlled by southern hemisphere processes only weakly linked to northern hemisphere insolation and ice volume. However, tighter coupling between the hemispheres appears to develop at ~500 ka, as suggested by smaller phase differences between $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ and $\delta^{18}\text{O}$ (Table S3), an increase in pCO_2 amplitude, and the phase lock between Antarctic temperature and northern hemisphere insolation during the last five terminations [Kawamura et al., 2007].

5. Estimates of pCO_2 for 1.5–0.8 Ma

[14] Here $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ -based estimates of pCO_2 from 1.5–0.8 Ma are compared with several other paleoclimate records that may correlate with pCO_2 . A proxy for South Atlantic surface productivity (the logarithm of alkenone concentration at ODP Site 1090) [Martínez-García et al., 2009] reproduces of the same glacial trends observed in pCO_2 and $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ (Figure 3, top) and has a similar correlation with pCO_2 (Table 1). Although South Atlantic productivity change appears to explain only 40–50 ppm of Late Pleistocene pCO_2 fluctuation, the proxy's correlation with pCO_2 may be enhanced by sensitivity to climate changes correlated with pCO_2 , such as South American aridity and westerly wind strength [Martínez-García et al., 2009]. Similarity between $\Delta\delta^{13}\text{C}_{\text{P-N/2}}$ and the alkenone record from 1.1–0.8 Ma provides additional support for the reliability of both proxies, particularly because they are linked to pCO_2 by different mechanisms.

[15] Both empirical proxies indicate that pCO_2 generally varies between 180–260 ppm from 1.1–0.8 Ma, except for a large oscillation at 950–900 ka (Figure 3, top). Both suggest a pCO_2 minimum at 920 ka of ~155 ppm, i.e., less than the

Figure 3. Proxy comparison. (top) pCO_2 (red) [Petit et al., 1999; Monnin et al., 2001; Siegenthaler et al., 2005; Lüthi et al., 2008], $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ (blue), alkenone concentration (green dashed) [Martinez-Garcia et al., 2009], boron-based estimates with error bars (black dots [Hönisch et al., 2009]; gray circles [Tripathi et al., 2009]; triangles [Seki et al., 2010]), and alkenone $\delta^{13}\text{C}$ estimates (squares) [Seki et al., 2010]. $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ and alkenone proxies are scaled to ppm using the mean and standard deviation of pCO_2 from 800–0 ka. (See auxiliary material for ODP 1090 age model.) (bottom) Changes in $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ (blue), WEP SST [Medina-Elizalde and Lea, 2005], and a tropical SST stack (purple) [Herbert et al., 2010] with trend reduced by $0.29^\circ\text{C}/\text{Myr}$ to match the WEP. $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ is scaled to $^\circ\text{C}$ using the standard deviation of the SST stack from 500–100 ka.

ice core pCO_2 minimum of 172 ppm at 668 ka. Many other paleoclimate records also contain evidence for extreme climatic conditions at ~ 900 ka, including anomalously low sea surface temperatures (SST), ocean circulation change, and increased Asian aridity [Clark et al., 2006].

[16] The increase in glacial benthic $\delta^{18}\text{O}$ values across the mid-Pleistocene transition (MPT) from 1.3–0.6 Ma is often attributed to decreasing glacial pCO_2 values [e.g., Raymo, 1997; Herbert et al., 2010]. Although $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ gradually shifts from 41-kyr to 100-kyr cyclicity from 1.3–0.7 Ma (Figure S3), it does not match the secular trend or amplitude increase observed in benthic $\delta^{18}\text{O}$ from 1.3–0.6 Ma. Boron-based measurements suggest that glacial pCO_2 minima decrease at ~ 800 ka [Hönisch et al., 2009; Tripathi et al., 2009], but these sparse measurements may not reliably sample glacial minima (Figure 3, top). The $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ and alkenone proxies, which show no change in glacial pCO_2 minima, actually agree with the low-resolution pCO_2 estimates of Hönisch et al. [2009] and Seki et al. [2010] from 1.5–0.8 Ma to within uncertainty (including age uncertainty). Also, the results of a carbon cycle box model suggest that a change in glacial pCO_2 minima during the MPT cannot be reconciled with the amplitude of Pacific $\delta^{13}\text{C}$ variability [Köhler and Bintanja, 2008].

[17] Additionally, SST change at some tropical sites unaffected by upwelling is thought to be driven by changes in radiative forcing [Medina-Elizalde and Lea, 2005; Herbert et al., 2010]. An SST record from the Western Equatorial Pacific (WEP) warm pool shows no significant trend from 1.35–0.5 Ma [Medina-Elizalde and Lea, 2005], consistent with the results of the $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ and alkenone proxies. A recent tropical SST stack that includes both upwelling and non-upwelling sites shows a slight cooling trend [Herbert et al., 2010], but if its long-term trend is adjusted to match

the SST trend of the WEP and other non-upwelling sites, the SST stack agrees well with $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ from 1.25–0.2 Ma (Figure 3 (bottom) and auxiliary material). Thus, only the pCO_2 estimates of Tripathi et al. [2009] are inconsistent with steady glacial pCO_2 minima since 1.25 Ma.

[18] However, before 1.25 Ma glacial temperatures in the trend-adjusted stack are $\geq 1^\circ\text{C}$ warmer than would be expected based on $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$. The SST stack could be affected by possible upwelling change at 1.25 Ma, such as thermocline shoaling or cooling at source water formation sites. However, a change in the relationship between $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ and pCO_2 is also possible, perhaps as the result of circulation or whole-ocean ΣCO_2 change. Additional high-resolution proxies are needed to improve confidence in glacial pCO_2 estimates, especially before 1.25 Ma.

6. Conclusions

[19] In conclusion, $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ correlates well with ice core pCO_2 from 800–0 ka and reproduces many features of the pCO_2 record. Comparison of $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ and pCO_2 suggests that marine and ice core age models [Lisiecki and Raymo, 2005; Parrenin et al., 2007; Louergue et al., 2007] differ by ≤ 2.7 kyr at terminations. Within the marine sedimentary record $\Delta\delta^{13}\text{C}_{\text{P-N}_4}$ usually leads $\delta^{18}\text{O}$ by 0.2–3.7 kyr at terminations but lags by 3–10 kyr during “failed” terminations at 535 and 745 ka. Thus, an early pCO_2 response appears necessary for complete deglaciation, and pCO_2 appears less tightly coupled to northern hemisphere ice volume before 500 ka.

[20] Several proxies that correlate with pCO_2 ($\Delta\delta^{13}\text{C}_{\text{P-N}_4}$, South Atlantic productivity [Martinez-Garcia et al., 2009], and WEP SST [Medina-Elizalde and Lea, 2005]) and a carbon cycle box model [Köhler and Bintanja, 2008] suggest

that glacial pCO_2 minima do not decrease during the MPT. Moreover, the minimum pCO_2 concentration of the last 1.5 Myr is estimated to occur at 920 ka. $\Delta\delta^{13}\text{C}_{\text{P-M}}$ gradually shifts from 41-kyr cycles to 100-kyr cycles from 1.3–0.7 Ma but shows no secular trend in mean or amplitude over the last 1.5 Myr, whereas tropical SST records suggest warmer glacial maxima before 1.3 Ma [Herbert *et al.*, 2010]. This likely indicates that at least one of these proxies is affected by factors other than pCO_2 before 1.3 Ma; thus, additional high-resolution proxies are needed.

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