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Author Lisiecki, LE

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A benthic δ^{13} C-based proxy for atmospheric pCO₂ over the last 1.5 Myr

L. E. Lisiecki¹

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[1] A high-resolution marine proxy for atmospheric pCO₂ is needed to clarify the phase lag between pCO₂ and marine climate proxies and to provide a record of orbital-scale pCO₂ variations before the oldest ice core measurement at 800 ka. Benthic δ^{13} C data should record deep ocean carbon storage and, thus, atmospheric pCO_2 . This study finds that a modified δ^{13} C gradient between the deep Pacific and intermediate North Atlantic $(\Delta \delta^{13} C_{P-M})$ correlates well with pCO₂. $\Delta \delta^{13}C_{P-\underline{M}}$ reproduces characteristic differences between pCO₂ and ice volume during Late Pleistocene glaciations and indicates that pCO2 usually leads terminations by 0.2-3.7 kyr but lags by 3-10 kyr during two "failed" terminations at 535 and 745 ka. $\Delta \delta^{13}C_{P-M}$ 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 pCO₂ of the last 1.5 Myr is estimated to be 155 ppm at ~920 ka. Citation: Lisiecki, L. E. (2010), A benthic δ^{13} Cbased proxy for atmospheric pCO₂ 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 pCO2 remains uncertain, preventing reconstruction of the sequence of climate responses associated with pCO2 change before 50 ka. A high-resolution marine proxy for pCO₂ could solve this problem and extend pCO₂ estimates beyond the oldest ice core measurement.

[3] Alkenone δ^{13} C and boron-based proxies reconstruct pCO₂ 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; *Tripati et al.*, 2009; *Pagani et al.*, 2009; *Seki et al.*, 2010]. Existing higher-resolution benthic δ^{13} C records also have the potential to record changes in atmospheric pCO₂ because glacial-

interglacial changes in pCO₂ are associated with changes in the Σ CO₂ and δ^{13} 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 δ^{13} C-based proxies and uses the one best correlated with ice core pCO₂ to evaluate the phase lag between pCO₂ and benthic δ^{18} O and to estimate pCO₂ from 1.5–0.8 Ma.

2. Background

[4] Decreased deep water ventilation and increased Southern Ocean productivity are thought to reduce glacial pCO₂ by removing carbon from the surface and sequestering it in the deep ocean [e.g., Toggweiler, 1999; Brovkin et al., 2007; Martínez-Garcia et al., 2009]. These processes also decrease the δ^{13} C value of deep waters as sinking low- δ^{13} 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 pCO₂ and benthic δ^{13} C changes are not positively correlated, increasing atmospheric pCO₂ but decreasing mean ocean δ^{13} 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 pCO₂ and deep Pacific δ^{13} C, but the observed correlation is much weaker (r = 0.5) due to low-frequency (~400-kyr) variations in δ^{13} C not observed in pCO₂ [Köhler et al., 2010].

[5] The $\delta^{13}C$ gradient between deep and intermediate waters ($\Delta\delta^{13}C_{D-I}$) has also been suggested as proxy for pCO₂ because it should reflect the Σ CO₂ 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]. $\Delta\delta^{13}C_{D-I}$ may also remove the mean-ocean $\delta^{13}C$ signal associated with changes in terrestrial carbon storage. Previous comparisons of $\Delta\delta^{13}C_{D-I}$ and pCO₂ 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 δ^{13} C, $\Delta \delta^{13}$ C_{*D*-*I*} and the average of the two as possible proxies for pCO₂. The signal-tonoise ratio of δ^{13} C signals is enhanced by averaging data from multiple cores within the same watermass to produce δ^{13} 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 ($\Delta \delta^{13}$ C_{*P*-*N*A})

¹Department of Earth Science, University of California, Santa Barbara, California, USA.

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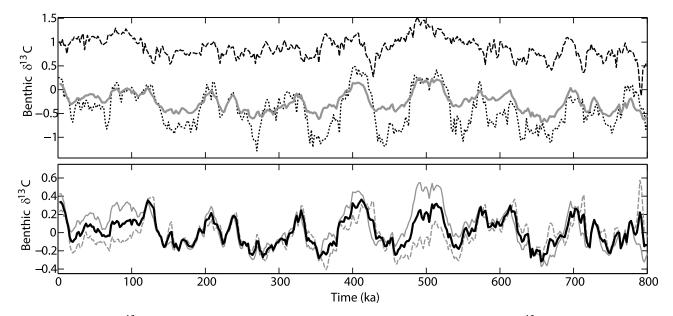


Figure 1. Benthic δ^{13} C stacks and potential pCO₂ proxies. (top) Regional stacks of benthic δ^{13} C from the intermediate North Atlantic (1145–2300 m depth, dashed line), South Atlantic (3700–4620 m, dotted), and equatorial Pacific (2520–3850 m, gray). (bottom) Pacific δ^{13} C stack (gray solid), $\Delta \delta^{13}$ C_{*P*-*NA*} (dashed), and the average of the two, $\Delta \delta^{13}$ C_{*P*-*NA*} (black solid). All three records are adjusted to have a mean of zero.

because watermass boundary movement provides an additional source of δ^{13} 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 δ^{13} C has approximately half the glacial-interglacial amplitude of South Atlantic δ^{13} C, the following analysis is not highly sensitive to which deep water stack is used because $\Delta \delta^{13}C_{P-NA}$ and $\Delta \delta^{13}C_{SA-NA}$ are well correlated from 800–0 ka (r=0.79).

sensitive to which deep water stack is used because $\Delta \delta^{13}C_{P-NA}$ and $\Delta \delta^{13}C_{SA-NA}$ are well correlated from 800–0 ka (r=0.79). [7] Pacific $\delta^{13}C$ and $\Delta \delta^{13}C_{P-NA}$ both correlate moderately well with pCO₂, but their average $\Delta \delta^{13}C_{P-M}$ produces the best correlation (Table 1 and Figure 1, bottom). Based on inter-core variability, $\Delta \delta^{13}C_{P-M}$ 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 $\Delta \delta^{13}C_{P-M}$ is scaled to the mean and standard deviation of ice core pCO₂, it has a root mean square error (RMSE) relative to pCO₂ of 17.5 ppm, whereas boron-based estimates have RMSE of 18.1 ppm [*Hönisch et al.*, 2009] and 24.9 ppm [*Tripati et al.*, 2009] (see auxiliary material).

[8] One possible physical explanation for the correlation between pCO₂ and $\Delta \delta^{13}C_{P-\underline{M}_2}$ is that $\delta^{13}C$ 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, $\Delta \delta^{13}C_{P-\underline{M}_2}$ corrects for differences in the amplitudes of variability between the Atlantic and Pacific. Alternatively, if Pacific $\delta^{13}C$ and $\Delta \delta^{13}C_{P-NA}$ are influenced differently by additional climatic processes, the average of the two should amplify the pCO₂ signal common to both.

[9] The correlation between marine proxies and pCO₂ is not a perfect evaluation metric because it depends on the particular marine and ice core chronologies used; therefore, $\Delta \delta^{13}$ C_{*P*-M/2} is also evaluated by whether it replicates features of the pCO₂ record that are independent of age model. Here

I focus on features that differ from the benthic δ^{18} O record [*Lisiecki and Raymo*, 2005] of deep water temperature and global ice volume. pCO₂ and northern hemisphere ice volume changes may differ if pCO₂ is controlled by southern hemisphere processes that are only weakly coupled to northern hemisphere climate [*Toggweiler*, 2008].

[10] One notable difference between δ^{18} O and pCO₂ (Figure 2, top) is that pCO₂ 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 δ^{18} 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 $\Delta \delta^{13}C_{P-\underline{M}}$ match those of pCO₂, with $\Delta \delta^{13}C_{P-\underline{M}}$ reaching a minimum 30–40 kyr before the δ^{18} O maximum during most glaciations (Figure 2, bottom). Additionally, $\Delta \delta^{13}C_{P-\underline{M}}$ correlates better with the magnitudes of glacial pCO₂ minima than δ^{18} O does. In $\Delta \delta^{13}C_{P-\underline{M}}$ and pCO₂, MIS 12 is less extreme than MIS 8, 10, and 16, whereas in δ^{18} O MIS 12 is similar to MIS 16 and more extreme than MIS 8 and 10. Thus, $\Delta \delta^{13}C_{P-\underline{M}}$ reproduces

Table 1. Proxy Correlation With pCO₂ for 800–0 ka

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

^aSee auxiliary material for component records.

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

^cHerbert et al. [2010].

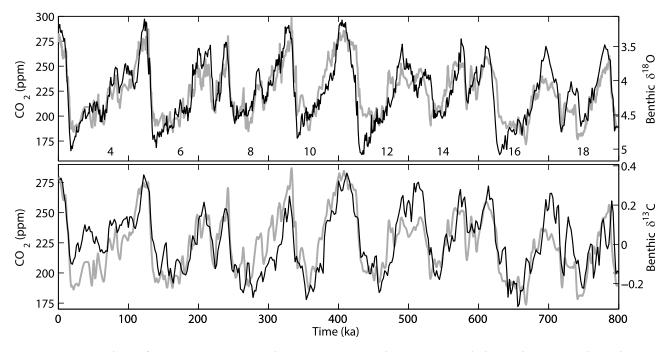


Figure 2. Comparison of pCO₂ (gray) [*Petit et al.*, 1999; *Monnin et al.*, 2001; *Siegenthaler et al.*, 2005; *Lüthi et al.*, 2008] with (top) benthic δ^{18} O (black) [*Lisiecki and Raymo*, 2005] and (bottom) $\Delta \delta^{13}$ C_{*P*-M} (black). Glacial stages are labeled by MIS number. In Figure 2 (bottom), pCO₂ has been smoothed with a 2-kyr boxcar² filter.

many pCO₂ responses that are independent of age model uncertainty and differ from ice volume change.

4. Termination Lags Between pCO₂ and δ^{18} O

[11] Comparison of $\Delta \delta^{13}C_{P-\frac{M}{2}}$ and the ice core pCO₂ record provides an opportunity to link marine and ice core age models. Abrupt increases in pCO₂ and $\Delta \delta^{13}C_{P-\frac{M}{2}}$ 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 pCO₂ and ice volume during terminations are evaluated by comparing $\Delta \delta^{13}C_{P-\underline{N4}}$ and benthic δ^{18} O changes within marine sediments. During most terminations $\Delta \delta^{13}C_{P-\frac{M}{2}}$ leads δ^{18} O by 0.2–3.7 kyr, but $\Delta \delta^{13}C_{P-\frac{M}{2}}$ lags δ^{18} O by 9.8 and 3.5 kyr during Termination 6 (535 k_a^2) and MIS 18 (745 ka), respectively (Table S3). These lags are also found between benthic δ^{18} O and δ^{13} C within individual Pacific cores (Figure S2). An anomalous phase relationship between ice volume and pCO2 may explain why these two warming events are weaker than most Late Pleistocene terminations. During both "failed" terminations, the initial δ^{18} O change is approximately half the amplitude of most Late Pleistocene terminations; δ^{18} 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 $\Delta \delta^{13} C_{P-M}$ lag during these two failed terminations suggests that full deglaciation requires an early pCO₂ 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 $\delta^{18}O$ and $\Delta\delta^{13}C_{P-\frac{M}{2}}$ supports the hypothesis of *Toggweiler* [2008] that glacial changes in pCO₂ 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 $\Delta\delta^{13}C_{P-\frac{M}{2}}$ and $\delta^{18}O$ (Table S3), an increase in pCO₂ amplitude, and the phase lock between Antarctic temperature and northern hemisphere insolation during the last five terminations [*Kawamura et al.*, 2007].

5. Estimates of pCO₂ for 1.5–0.8 Ma

[14] Here $\Delta \delta^{13}C_{P-\underline{M}}$ -based estimates of pCO₂ from 1.5-0.8 Ma are compared with several other paleoclimate records that may correlate with pCO₂. A proxy for South Atlantic surface productivity (the logarithm of alkenone concentration at ODP Site 1090) [Martínez-Garcia et al., 2009] reproduces of the same glacial trends observed in pCO₂ and $\Delta \delta^{13}$ C_{P-M} (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 pCO₂ fluctuation, the proxy's correlation with pCO2 may be enhanced by sensitivity to climate changes correlated with pCO₂, such as South American aridity and westerly wind strength [Martínez-Garcia et al., 2009]. Similarity between $\Delta \delta^{13} C_{P-\frac{M}{2}}$ 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

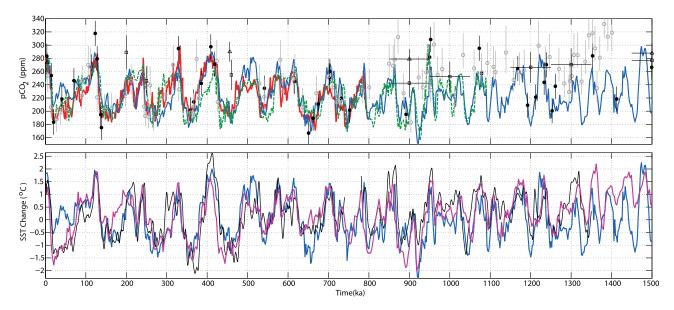


Figure 3. Proxy comparison. (top) pCO₂ (red) [*Petit et al.*, 1999; *Monnin et al.*, 2001; *Siegenthaler et al.*, 2005; *Lüthi et al.*, 2008], $\Delta \delta^{13}C_{P-\underline{M}}$ (blue), alkenone concentration (green dashed) [*Martinez-Garcia et al.*, 2009], boron-based estimates with error bars (black dots [*Hönisch et al.*, 2009]; gray circles [*Tripati et al.*, 2009]; triangles [*Seki et al.*, 2010]), and alkenone $\delta^{13}C$ estimates (squares) [*Seki et al.*, 2010]. $\Delta \delta^{13}C_{P-\underline{M}}$ and alkenone proxies are scaled to ppm using the mean and standard deviation of pCO₂ from 800–0 ka. (See auxiliary material for ODP 1090 age model.) (bottom) Changes in $\Delta \delta^{13}C_{P-\underline{M}}$ (blue), WEP SST [*Medina-Elizalde and Lea*, 2005], and a tropical SST stack (purple) [*Herbert et al.*, 2010] with trend reduced by 0.29°C/Myr to match the WEP. $\Delta \delta^{13}C_{P-\underline{M}}$ is scaled to °C using the standard deviation of the SST stack from 500–100 ka.

ice core pCO₂ 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 δ^{18} O values across the mid-Pleistocene transition (MPT) from 1.3-0.6 Ma is often attributed to decreasing glacial pCO2 values [e.g., Raymo, 1997; Herbert et al., 2010]. Although $\Delta \delta^{13} C_{P-M}$ 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 δ^{18} O from 1.3–0.6 Ma. Boronbased measurements suggest that glacial pCO2 minima decrease at ~800 ka [Hönisch et al., 2009; Tripati et al., 2009], but these sparse measurements may not reliably sample glacial minima (Figure 3, top). The $\Delta \delta^{13}C_{P-M}$ and alkenone proxies, which show no change in glacial ²pCO₂ minima, actually agree with the low-resolution pCO₂ 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 pCO₂ minima during the MPT cannot be reconciled with the amplitude of Pacific $\delta^{13}C$ 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 $\Delta \delta^{13}C_{P-\frac{N4}{2}}$ 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 $\Delta \delta^{13}C_{P-\underline{M}}$ from 1.25–0.2 Ma (Figure 3 (bottom) and auxiliary material). Thus, only the pCO₂ estimates of *Tripati et al.* [2009] are inconsistent with steady glacial pCO₂ minima since 1.25 Ma.

[18] However, before 1.25 Ma glacial temperatures in the trend-adjusted stack are $\geq 1^{\circ}$ C warmer than would be expected based on $\Delta \delta^{13}$ C_{*P*-<u>M</u>}. 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 $\Delta \delta^{13}$ C_{*P*-<u>M</u>} and pCO₂ is also possible, perhaps as the result of circulation or whole-ocean Σ CO₂ change. Additional high-resolution proxies are needed to improve confidence in glacial pCO₂ estimates, especially before 1.25 Ma.

6. Conclusions

[19] In conclusion, $\Delta \delta^{13}C_{P-\underline{M}}$ correlates well with ice core pCO₂ from 800–0 ka and reproduces many features of the pCO₂ record. Comparison of $\Delta \delta^{13}C_{P-\underline{M}}$ and pCO₂ suggests that marine and ice core age models [*Lisiecki and Raymo*, 2005; *Parrenin et al.*, 2007; *Loulergue et al.*, 2007] differ by ≤ 2.7 kyr at terminations. Within the marine sedimentary record $\Delta \delta^{13}C_{P-\underline{M}}$ usually leads δ^{18} 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 pCO₂ response appears necessary for complete deglaciation, and pCO₂ appears less tightly coupled to northern hemisphere ice volume before 500 ka.

[20] Several proxies that correlate with pCO₂ ($\Delta \delta^{13}C_{P-\frac{M}{2}}$, South Atlantic productivity [*Martínez-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 pCO₂ minima do not decrease during the MPT. Moreover, the minimum pCO₂ concentration of the last 1.5 Myr is estimated to occur at 920 ka. $\Delta \delta^{13}$ C_{*P*- $\frac{N4}{2}$} 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 pCO₂ before 1.3 Ma; thus, additional highresolution proxies are needed.

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L. E. Lisiecki, Department of Earth Science, University of California, Santa Barbara, CA 93106, USA. (lisiecki@geol.ucsb.edu)