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Carbon Isotope Constraints on the Deglacial CO_2 Rise from Ice Cores

Jochen Schmitt,[1,2]* Robert Schneider,[1] Joachim Elsig,[1] Daiana Leuenberger,[1] Anna Lourantou,[3]† Jérôme Chappellaz,[3] Peter Köhler,[2] Fortunat Joos,[1] Thomas F. Stocker,[1] Markus Leuenberger,[1] Hubertus Fischer[1,2]

[1]Climate and Environmental Physics, Physics Institute, and Oeschger Centre for Climate Change Research, University of Bern, CH-3012 Bern, Switzerland. [2]Alfred Wegener Institute for Polar and Marine Research, 27570 Bremerhaven, Germany. [3]UJF–Grenoble 1/CNRS, Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE) UMR 5183, 38041 Grenoble, France.

*To whom correspondence should be addressed. E-mail: schmitt@climate.unibe.ch

†Present address: Laboratoire d'Océanographie et du Climat (LOCEAN), Institut Pierre Simon Laplace, Université P. et M. Curie (UPMC), 75005 Paris, France.

The stable carbon isotope ratio of atmospheric CO_2 ($\delta^{13}C_{atm}$) is a key parameter to decipher past carbon cycle changes. Here we present $\delta^{13}C_{atm}$ data for the last 24,000 years derived from three Antarctic ice cores. We conclude that a pronounced 0.3‰ decrease in $\delta^{13}C_{atm}$ during the early deglaciation can be best explained by upwelling of old, carbon-enriched waters in the Southern Ocean. Later in the deglaciation, regrowth of the terrestrial biosphere, changes in sea surface temperature, and ocean circulation governed the $\delta^{13}C_{atm}$ evolution. During the Last Glacial Maximum, $\delta^{13}C_{atm}$ and CO_2 were essentially constant, suggesting that the carbon cycle was in dynamic equilibrium and that the net transfer of carbon to the deep ocean had occurred before then.

During the last 800,000 years (800 kyr), atmospheric CO_2 concentrations have varied in close relation to Antarctic temperatures (*1*, *2*) and the general waxing and waning of continental ice sheets. In particular, CO_2 rose from a stable level of 190 parts per million by volume (ppmv) during the Last Glacial Maximum to about 280 ppmv in preindustrial times, showing pronounced differences in atmospheric CO_2 rates of change in the course of the last glacial/interglacial transition (*3*). Many processes have been involved in attempts to explain these CO_2 variations, but it has become evident that none of these mechanisms alone can account for the 90 ppmv increase in atmospheric CO_2. A combination of processes must have been operating (*4*, *5*), with their exact timing being crucial. However, a unique solution to the deglacial carbon cycle changes has not been yet found.

In this respect, high-resolution and precise $\delta^{13}C_{atm}$ records from Antarctic ice cores are needed to better constrain the evolution of carbon cycle changes during the last deglaciation. On millennial time scales, $\delta^{13}C_{atm}$ is primarily influenced by the $\delta^{13}C$ of dissolved inorganic carbon (DIC) ($\delta^{13}C_{DIC}$) and sea surface temperature (SST), which controls the isotopic fractionation during air/sea gas exchange. The continuous rain of isotopically light organic material to the interior of the ocean draws down carbon from the surface layer to intermediate and deep waters, where the organic carbon is remineralized. Consequently, a vertical $\delta^{13}C_{DIC}$ gradient is established, controlled by the interplay of the ocean circulation with this so-called "biological pump". The more intense the circulation, the smaller the gradients are for $\delta^{13}C_{DIC}$, DIC, oxygen and nutrients. Superimposed on these marine carbon cycle processes are climate-induced changes in terrestrial biosphere carbon storage, which result in a net change in the carbon isotopic composition of the ocean/atmosphere system. On orbital time scales, weathering and sedimentation of $CaCO_3$ affect $\delta^{13}C_{DIC}$, $\delta^{13}C_{atm}$ and atmospheric CO_2 as well.

Until recently (*6*), analytical constraints represented the fundamental limitation on the utility of $\delta^{13}C_{atm}$ ice core records (*7*, *8*). Here we provide evidence (Fig. 1) about possible causes of carbon cycle changes with measurements of $\delta^{13}C_{atm}$ from two Antarctic ice cores (EPICA (European Project for Ice Coring in Antarctica) Dome C and Talos Dome), performed with three independent methods in two different labs (referred to as Bern sublimation, Bern cracker and Grenoble mill data) (*6*, *9*). One of our records is based on a sublimation method (*10*) that avoids the effects associated with incomplete gas extraction and thus yields more precise results (see Supporting Online Material (SOM)). A stringent residual analysis of the three data sets shows virtually no offset between the two Bern data sets and only a small systematic offset between the Bern and Grenoble data of 0.16‰, which can be explained by a method-dependent systematic fractionation. After correction of this offset, we combined the three $\delta^{13}C_{atm}$ records over the last 24 kyr using an error-weighted Monte Carlo bootstrap approach. This method showed that all three data sets are essentially compatible within their analytical uncertainties. To make full use of the resolution and precision of the data, the inclusion of all three data sets is required, although all our conclusions are also supported by the individual records. The final data set consists of 201 individual measurements, each reflecting typically 2 to 4 replicates and with an analytical 1σ error between 0.04 and 0.12‰. Since the resulting Monte Carlo Average (MCA) removes most of the analytical uncertainties, it contains less high-frequency variability compared to the raw data. This is in line with the centennial-scale low-pass filtering inherent to the bubble enclosure process at Dome C. Accordingly, the retained variability can be regarded as the signal most representative of millennial $\delta^{13}C_{atm}$ changes (see SOM for details regarding the MCA and its uncertainty).

Our $\delta^{13}C_{atm}$ data are in good agreement with previously published lower-resolution records (*6*, *9*). Our record shows a very stable level between 24 and ca. 19 kyr before present (BP, where present is defined as 1950), with an average $\delta^{13}C_{atm}$ of -6.45‰ (tables S1 and S2), similar to the -6.35‰ of the Late Holocene (Fig. 2B). Given the fact that a large set of environmental parameters such as atmospheric CO_2, global SST, terrestrial carbon storage, and ocean circulation have varied between the LGM and the Late Holocene, almost identical $\delta^{13}C_{atm}$ values indicate that opposing effects must have offset each other (*11*). This becomes clear if we look at three first-order effects on $\delta^{13}C_{atm}$: A SST rise of 1 K translates into a 0.1‰ increase in $\delta^{13}C_{atm}$, due to temperature-dependent fractionation between atmospheric CO_2 and marine DIC species (*12*). Assuming a global LGM-to-Holocene SST rise of 3 K would result in about 0.3‰ higher $\delta^{13}C_{atm}$ for the Holocene, provided that SST distribution and CO_2 gross flux exchange patterns remained constant. This effect is further augmented by the uptake of isotopically light carbon by the land biosphere and counterbalanced by the smaller vertical gradient in $\delta^{13}C_{DIC}$ in the Holocene ocean, supported by marine data (*13*). The fact that both $\delta^{13}C_{atm}$ and CO_2 show little variation from 24 to 19 kyr BP points to the carbon cycle being essentially in dynamic equilibrium at that time. As can also be seen in Fig. 2, the climate variations related to Heinrich stadial 2 (HS2) and Dansgaard-Oeschger event 2 (DO2) had little effect on the global carbon cycle during this time interval. Howev-

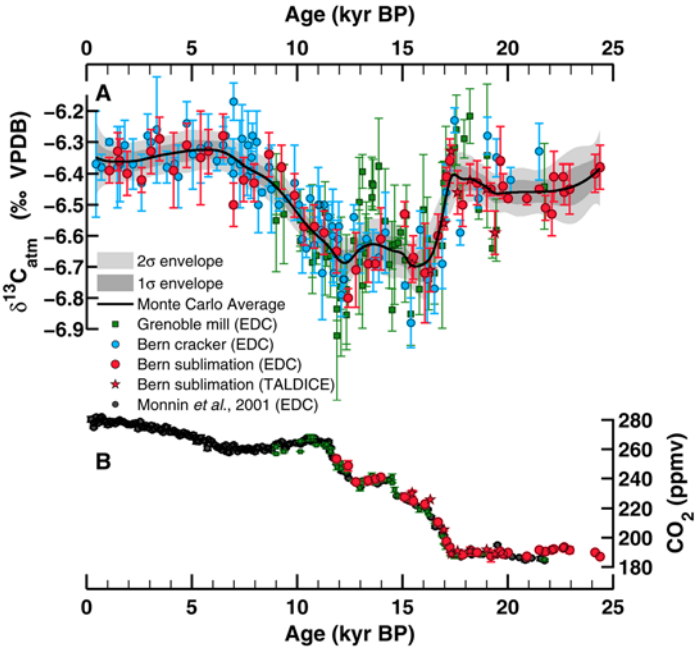

Fig. 1. Ice core reconstructions of atmospheric $\delta^{13}C$ and CO_2 concentration covering the last 24 kyr. (**A**) $\delta^{13}C_{atm}$ of atmospheric CO_2 measured with three different methods on two different ice core drill sites. Blue circles: Bern cracker data, green squares: Grenoble mill data (*9*) after offset correction, red circles: Bern sublimation data. Red stars indicate values from the sublimation method but measured on Talos Dome Ice Core (TALDICE). Error bars represent the standard deviation of replicate measurements. The black line is the result of 4000 Monte Carlo simulations representing an error-weighted average of the different $\delta^{13}C_{atm}$ data sets. The light and dark shaded areas represent the 2σ and 1σ error envelope around the Monte Carlo Average (see SI). (**B**) CO_2 concentration. Black circles represent earlier measurements on EDC (*3*), other symbols are the same as in panel **A**. Note: All ice core records are plotted on a synchronized age scale (*32*).

er, given the opposing trends for reconstructed atmospheric $\Delta^{14}C$ ($\Delta^{14}C_{atm}$) (*14*, *15*) and the expected $\Delta^{14}C_{atm}$ evolution (*16*) based on variations in ^{14}C production rate (*17*, *18*), the global ^{14}C budget was not balanced (Fig. 2A).

After a very small increase in $\delta^{13}C_{atm}$ at the very end of the glacial, a sharp drop in $\delta^{13}C_{atm}$ starting at 17.5 kyr parallels the onset of increasing atmospheric CO_2. Taken at face value, this would point to an early SST rise that preceded the onset of the CO_2 increase. When we apply a crude SST correction to our $\delta^{13}C_{atm}$ databased on a global estimate of SST temperature changes during the transition (see SOM), this $\delta^{13}C_{atm}$ increase vanishes (Fig. 2B). Note, however, that this 0.06‰ excursion is within the uncertainties of our data and that other effects could also lead to this small enrichment in $\delta^{13}C_{atm}$. The 0.3‰ drop in $\delta^{13}C_{atm}$ after the onset of the transition at 17.5 kyr BP is accompanied by a CO_2 increase of about 35 ppmv and a 190‰ drop in $\Delta^{14}C_{atm}$ (*19*), which has been attributed to a release of old carbon from the deep ocean. This coeval drop in $\delta^{13}C_{atm}$ and $\Delta^{14}C_{atm}$ during the so-called "mystery interval", 17.5 – 14 kyr BP (*19*), is arguably the most enigmatic carbon cycle change in the course of the transition and will be discussed in more detail below.

After the broad $\delta^{13}C_{atm}$ minimum is reached at about 16 kyr BP, $\delta^{13}C_{atm}$ increases slightly by 0.1‰ during the pronounced Bølling-Allerød (BA) warming. Other than circulation changes in the Southern Ocean (*20*), the regrowth of the terrestrial biosphere in the northern hemisphere could contribute to this increase in $\delta^{13}C_{atm}$ (*4*). However, since the SST-corrected $\delta^{13}C$ evolution (Fig. 2B) does not show any increase, a robust process attribution requires precisely dated SST reconstructions and transient carbon cycle modeling.

An almost linear rise by 0.06‰ per kyr follows the second $\delta^{13}C_{atm}$ minimum at 12.2 kyr BP, leading to maximum values of -6.33‰ at around 6 kyr BP. This rise might be largely explained by the continuing regrowth of the terrestrial biosphere (*21*), in concert with smaller contributions from SST warming and changes in circulation and export production (*9*, *22*). From this mid-Holocene maximum, $\delta^{13}C_{atm}$ values decline slightly to reach values of -6.35‰ at 0.5 kyr BP, as previously reported (*6*).

As mentioned above, the carbon cycle changes during the mystery interval have been a matter of intense debate (*19*, *20*, *23*). Our high-resolution $\delta^{13}C_{atm}$ record together with other records of carbon cycle changes and insights from models may help to constrain hypotheses put forward to explain the mystery interval. The rise in CO_2 and the decline in $\delta^{13}C_{atm}$ and $\Delta^{14}C_{atm}$ between 17 and 15 kyr BP fit the concept of bringing DIC-rich waters with old carbon into exchange with the atmosphere. Indicative ^{14}C signals of upwelling of old, CO_2-enriched deep water were found in Pacific intermediate waters (*24*), but others (*23*) ruled out such old water in the northeast Pacific, and evidence for a ^{14}C-depleted glacial deep ocean remains elusive (*19*, *23*, *25*). These $\Delta^{14}C$ studies were usually confronted with variable reservoir age between benthic and planktonic foraminifera. A study using deep sea corals now circumvents this problem by applying absolute U-Th dating and shows that the deep glacial Southern Ocean indeed ventilated its ^{14}C-depleted reservoir during the mystery interval (*26*).

The constant $\delta^{13}C_{atm}$ values during the late glacial indicate that the build-up of such an old, DIC-rich reservoir must have occurred before 24 kyr BP. A large number of records mark the start of the deglaciation around 17 kyr BP (Fig. 2). Within the uncertainty in marine and ice core age scales, the CO_2 increase, the pronounced $\Delta^{14}C_{atm}$ drop (*15*), the resumption of vigorous Southern Ocean upwelling as recorded in intense deposition of biogenic opal (*20*), and the launch of ice-rafted debris layers at the beginning of the Heinrich 1 stadial (*27*) all occurred simultaneously. Interestingly, our $\delta^{13}C_{atm}$ record shows its largest deviation of 0.3‰, i.e., the entire $\delta^{13}C_{atm}$ decrease from the LGM to the Preboreal (PB), within the first 2 kyr after the start of the deglaciation. Within the same interval, CO_2 rose by 30 ppmv from 190 ppmv to 220 ppmv, i.e., only 35% of the LGM-PB rise. Together with the trend reversal in $\delta^{13}C_{atm}$ toward the end of the mystery interval, this indicates that only a fraction of the glacial/interglacial CO_2 increase can be explained by an intensification of deep ocean ventilation bringing isotopically depleted and carbon-rich water to the surface of the Southern Ocean. Our new, high-resolution $\delta^{13}C_{atm}$ data constrain the period of this release of isotopically depleted carbon from the deep ocean to the atmosphere to between 17.4 kyr BP and 15 kyr BP. This interpretation of the proxy records is quantitatively in line with dynamical ocean model results that link deep ocean ventilation, atmospheric CO_2, $\delta^{13}C_{atm}$, $\delta^{13}C_{DIC}$, opal burial, and radiocarbon (*28*).

Alternative hypotheses (*29*, *30*) invoking the release of old carbon from permafrost or carbon locked under continental ice sheets are unlikely to explain the carbon cycle changes in the mystery interval because the amount of terrestrial carbon needed to account for the ^{14}C drop is very large, at 5000 Gt (*25*), and would conflict with the moderate 30 ppmv rise in atmospheric CO_2. Moreover, it would lead to an overall decline in $\delta^{13}C_{DIC}$, which is not observed in benthic foraminifera in the deep ocean (*13*, *22*). Also, a carbonate dissolution event at the sea floor that would have to accompany such a large terrestrial carbon release into the atmosphere/ocean system is not imprinted in the deglacial marine

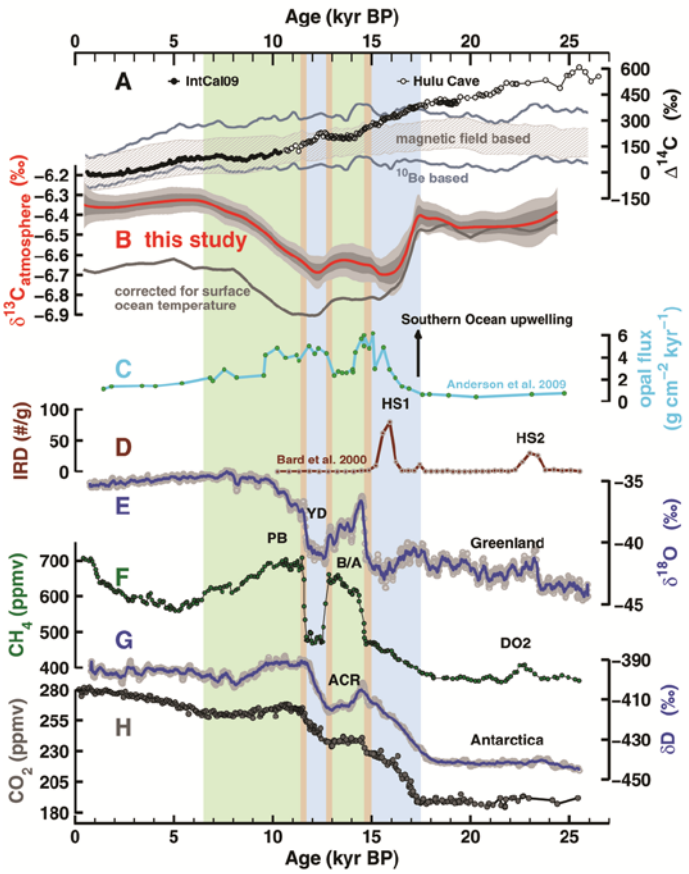

Fig. 2. Ice core reconstructions and marine records illustrating the evolution of major components of the Earth climate system over the last 24 kyr. (**A**) Reconstructed $\Delta^{14}C_{atm}$ from IntCal09 (*14*) and the ^{230}Th-dated Hulu Cave $\Delta^{14}C_{atm}$ record (*15*) compared with modeled (*16*) $\Delta^{14}C_{atm}$ assuming a constant carbon cycle under pre-industrial conditions but considering temporal changes in ^{14}C production (either based on ^{10}Be (*18*), upper and lower estimates enveloped in gray lines, or on paleomagnetic field intensity (*17*), hatched area). (**B**) Monte Carlo simulations (this study) of the evolution of $\delta^{13}C_{atm}$ before (red line represents the MCA, 2σ and 1σ envelopes are in gray) and after SST correction (gray line; see SI) (**C**) Opal flux in the Southern Ocean as a proxy for local upwelling (*20*). (**D**) Record of ice rafted debris (IRD) in the North Atlantic associated with Heinrich stadials (HS1 and HS2) (*27*). (**E**) Greenland temperature proxy $\delta^{18}O$ (*33*). (**F**) Reconstructed atmospheric CH_4 concentration (*34*) (**G**) Antarctic temperature proxy δD from the EDC ice core (*35*). (**H**) Compilation of reconstructed CO_2 shown in Fig. 1B. Green bars indicate intervals with a strong net terrestrial carbon build-up, blue bars indicate intervals where sequestered deep ocean CO_2 was released back to the atmosphere. Note: Ice core records are plotted on a synchronized age scale (*32*), other records are plotted on their individual age scales.

$CaCO_3$ record (*31*)

Consequently, despite the fact that the search for an extremely ^{14}C-depleted deep water mass in marine records has thus far not been successful (*23*) and might not even essential to explain the $\Delta^{14}C_{atm}$ anomaly (*26*), the release of carbon from the deep ocean remains the most plausible scenario to explain the early deglacial drop in our new $\delta^{13}C_{atm}$ record. Furthermore, model results suggest that a $\delta^{13}C_{atm}$ decrease of 0.3‰ and a CO_2 increase of about 30 ppmv can be accommodated by relatively small (about 20‰) and spatially complex changes in deep ocean $\Delta^{14}C$ (*28*). These changes may remain undetected in the search for the old abyssal water using benthic foraminifera (*19*, *25*). However, they are also too small to explain the reconstructed $\Delta^{14}C_{atm}$ decline in the mystery interval. Based on these considerations, the currently available marine and ice core information cannot be reconciled with the atmospheric radiocarbon record in a straightforward manner. One possibility to resolve this issue is to also reconsider a larger change in ^{14}C production between the Holocene and the glacial, and to work toward independent verification of the $\Delta^{14}C_{atm}$ history.

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Supplementary Materials
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Materials and Methods
Figs. S1 to S7
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