Upper-ocean-to-atmosphere radiocarbon offsets imply fast deglacial carbon dioxide release

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Radiocarbon in the atmosphere is regulated largely by ocean circulation, which controls the sequestration of carbon dioxide (CO₂) in the deep sea through atmosphere–ocean exchange. During the last glaciation, lower atmospheric CO₂ levels were accompanied by increased atmospheric radiocarbon concentrations that have been attributed to greater storage of CO₂ in a poorly ventilated abyssal ocean. The end of the ice age was marked by a rapid increase in atmospheric CO₂ concentrations coinciding with reduced ¹⁴C/¹²C ratios (Δ¹⁴C) in the atmosphere, suggesting the release of ‘old’ (¹³C-depleted) CO₂ from the deep ocean to the atmosphere. Here we present radiocarbon records of surface and intermediate-depth waters from two sediment cores in the southwest Pacific and Southern Oceans. We find a steady 170 per mil decrease in Δ¹⁴C that precedes and roughly equals in magnitude the decrease in the atmospheric radiocarbon signal during the early stages of the glacial–interglacial climatic transition. The atmospheric decrease in the radiocarbon signal coincides with regionally intensified upwelling and marine biological productivity, suggesting that CO₂ released by means of deep water upwelling in the Southern Ocean lost most of its original depleted-¹⁴C imprint as a result of exchange and isotopic equilibration with the atmosphere. Our data imply that the deglacial ¹⁴C depletion previously identified in the eastern tropical North Pacific must have involved contributions from sources other than the previously suggested carbon release by way of a deep Southern Ocean pathway, and may reflect the expanded influence of the ¹³C-depleted North Pacific carbon reservoir across this interval. Accordingly, shallow water masses advecting north across the South Pacific in the early deglaciation had little or no residual ¹³C-depleted signals owing to degassing of CO₂ and biological uptake in the Southern Ocean.

Observations in the glacial atmosphere of higher radiocarbon activity (Δ¹⁴C) accompanied by lower CO₂ concentrations, relative to modern values, are believed to have been caused primarily by climate-induced changes in the ocean’s thermohaline circulation isolating abyssal ocean carbon from the atmosphere. Although independent estimates of ¹⁴C production indicate that changes in thermohaline circulation cannot fully account for atmospheric Δ¹⁴C changes since the last ice age, considerable evidence suggests that CO₂ with a ¹⁴C content significantly lower than it is today was sequestered in the deep glacial ocean below a depth of 2.5 km (refs 8–11). Whereas the full extent of the deep carbon reservoir remains to be constrained, it seems that a significant portion of this glacially sequestered CO₂ was reintroduced into the atmosphere by means of ocean outgassing during the last glacial–interglacial transition. This ¹³C-depleted carbon contributed to atmospheric CO₂ increase while reducing the atmospheric radiocarbon signal by ~20% (ref. 7), leaving a strong imprint on the Δ¹⁴C distribution in the ocean.

Because deep water masses outcrop in the Southern Ocean, surface processes there probably regulated carbon injection into the atmosphere and CO₂ partitioning among the several ocean reservoirs. Correlations among Antarctic temperature records, ¹³C-depleted atmospheric CO₂ increase, changes in Southern Ocean upwelling and model results support this explanation. However, there is still no direct verification of the pathways taken by ¹³C-depleted CO₂ from the deep ocean to the atmosphere.

Here we present early deglacial surface and intermediate-depth Δ¹⁴C profiles that were reconstructed using radiocarbon activities from coexisting planktonic and benthic foraminifera in two sediment cores in the sub-Antarctic and subtropical South Pacific near New Zealand. The region and shallow water depths of both core sites are ideal for assessing deglacial ¹⁴C changes in sub-Antarctic mode water (SAMW; core RR0503-JPC64) and Antarctic intermediate water (AAIW; core MD97-2120) that are derived from the entrainment of abyssal water masses into the upper-ocean thermocline by wind-driven upwelling in the Southern Ocean (Fig. 1). Subtropical mode water (STMW) has components of SAMW entrained from equatorial undercurrent waters or can be partially sourced through the Tasman Sea.

The baseline age model for core MD97-2120 on the south Chatham Rise (east of New Zealand) is anchored with ¹⁴C dating and fine-tuned between 13 and 19 kyr BP by correlating the stable oxygen isotope (δ¹⁸O) record of its planktonic foraminifera with the δ¹⁸O record of the European Project for Ice Coring in Antarctica (EPICA) Dronning Maud Land (EDML) ice core (Figs 2a and Supplementary Fig. 1). This age model allows the use of ¹⁴C as a tracer that is not biased by using it as a stratigraphic tool. The age model for core RR0503-JPC64 in the Bay of Plenty (north of New Zealand) was developed by combining ¹⁴C-derived calendar-age tie points from the planktonic foraminifera Globorotalia inflata with tephra chronology from the core tied to established New Zealand chronologies and the δ¹⁸O record of the EDML ice core. Tephra stratigraphy suggests continuous sedimentation throughout the early deglaciation, before ~13,700 calendar years BP (the Waiohau tephra). However, above this tephra, there seems to be a 4.1-kyr hiatus. For both cores, we used the CALIB v5.0.1 program and MARINE04 database to convert the radiocarbon data to calendar ages. (See Supplementary Information for stratigraphic and methodological details.)
To assess partitioning of $^{14}$C among the atmosphere, subsurface and surface water masses, we compared age-corrected water-mass $^{14}$C values based on foraminifera with contemporary atmosphere values, IntCal04$^{30}$ and Cariaco$^{7}$, on the Hulu cave timescale$^{6}$ (Fig. 2). The two atmospheric $^{14}$C levels are offset through the early deglaciation (Fig. 3a), which can be attributed to IntCal04 pre-dating Cariaco on the Hulu cave timescale$^{6}$. Owing to higher sample density, we consider the Cariaco–Hulu record$^{7}$ better suited for comparison with our marine radiocarbon records. The planktonic species G. inflata in core RR0503-JPC64 is well suited to tracing STMW because it inhabits the upper thermocline, at the depth of STMW. SAMW is advected rapidly, on the order of decades$^{21}$, from the sub-Antarctic zone to the tropics, where its $^{14}$C signal is entrained upwards into STMW and recorded by G. inflata$^{1}$ (Fig. 1).

During the early deglaciation, from 18 to 15.8 kyr BP, the benthic and planktonic $^{14}$C values in our marine records decrease by $\sim$170%. In the subtropical core at 18 kyr BP, the difference between the atmospheric signal and the G. inflata planktonic signal is 20% (slightly less than the modern value; Fig. 2c). Initially, the foraminiferal $^{14}$C values decrease at a greater pace than atmospheric $^{14}$C, such that at 16.2 kyr BP the difference between atmospheric and planktonic $^{14}$C reaches 100–150% (Fig. 2b, c). After this, the planktonic $^{14}$C record decreases more slowly whereas atmospheric $^{14}$C decreases sharply, such that within the next 1.7 kyr (the period indicated by red shading on Fig. 2) the atmosphere–ocean difference is reduced almost to zero. The $^{14}$C in SAMW (indicated by benthic foraminiferal $^{14}$C in the Bay of Plenty core) largely mirrors the trend seen in planktonic $^{14}$C. At 18 kyr BP, SAMW is depleted by 150% relative to atmospheric values and decreases at a greater rate than the planktonics do until benthic $^{14}$C is about 200% below the contemporaneous atmospheric values at 16.2 kyr BP. After this, SAMW $^{14}$C plateaus until the ocean–atmosphere offset returns, between 15.0 and 14.5 kyr BP, to the value it had 18 kyr BP. AAIW, as recorded by benthic $^{14}$C in our Chatham Rise core, displays fundamentally similar trends to the subtropical. The planktonic $^{14}$C record is analogous to the others, but shows oscillations and changing offsets among species from 17.8 to 16.5 kyr BP (Fig. 2b). These probably reflect variations in subsurface-to-surface and ocean-to-atmosphere carbon transfer and interspecies habitat differences. (See Supplementary Information for a full discussion.)

Our marine $^{14}$C records support a southern locus as an exit route for CO$_2$ from a deep and isolated marine carbon reservoir during the early deglaciation, and help constrain the dynamics among the atmospheric CO$_2$ increase and $^{14}$C decrease and the deglacial Southern Ocean degassing. Rapid CO$_2$ injection into the atmosphere begins...
17.8 kyr BP², whereas the drastic drop in atmospheric Δ¹⁴C begins later, ~16.5 kyr BP (Fig. 3a). The marine Δ¹⁴C decrease starts 17.8 kyr BP, is concurrent with the initial rise in atmospheric CO₂ and leads the atmospheric Δ¹⁴C decrease. Consequently, between 17.8 and 16.5 kyr BP the atmosphere/Southern Ocean isotopic difference increases by ~50%. The change in offset across this time suggests that ¹⁴C-depleted carbon upwelled into thermocline and surface waters in the Southern Ocean, but that air–sea exchange in the Southern Ocean was inadequate for that ¹⁴C-depleted CO₂ to attain isotopic equilibrium with the atmosphere. With constant air–sea mixing dynamics, the increase in atmospheric CO₂ concentration throughout this period would decrease surface marine Δ¹⁴C relative to the atmospheric value (by ~10%)¹,²,³ whereas higher air–sea exchange rates would minimize this effect. Thus, about 40% of the marine decrease may be attributed to injection of waters greatly depleted in ¹⁴C relative to the atmosphere.

Starting around 16.5 kyr BP, increased opal fluxes at latitude 53°S (Fig. 2d) suggest substantial strengthening of Southern Ocean upwelling south of the Antarctic polar front that increased marine biological productivity (also in evidence at our subpolar site; Fig. 3c). Coincident with the initiation of enhanced upwelling, the atmospheric Δ¹⁴C decrease accelerated but radiocarbon levels in STMW, SAMW and possibly AAIW levelled out (Figs 2 and 3). Vigorous mixing and deeper convection are plausible mechanisms to accelerate ¹³C–¹⁴C exchange and reduce the atmosphere–ocean Δ¹⁴C difference. A concurrent release of highly ¹⁴C-depleted CO₂ from the deep ocean would reduce the Δ¹⁴C in both the atmosphere and the upper ocean. These interpretations make an implicit assumption that Pacific glacial and deglacial circulation was analogous to that today. Alternatively, it is possible that water upwelling in the Southern Ocean at this time was not significantly depleted in ¹⁴C. Nonetheless, we suggest that deeper waters substantially depleted in ¹⁴C were drawn to the upper layers in the Southern Ocean after 16.5 kyr BP (perhaps breaching a deep hydrologic front), and that vigorous exchange transferred the ¹⁴C signal to the atmosphere. This caused the atmospheric Δ¹⁴C value to drop rapidly and the Southern Ocean/atmosphere Δ¹⁴C levels to become more closely aligned. At the start of the Antarctic cold reversal (~14.5 kyr BP), the atmosphere–ocean Δ¹⁴C offset stabilized at the modern value (Fig. 2). This is attributed to Southern Ocean cooling that caused westerly winds to shift northwards, weakening upwelling. This would have substantially damped ocean–atmosphere CO₂ and ¹⁴C exchange in the region.

The relatively small maximum Δ¹⁴C offset among the atmosphere, surface waters and subthermocline waters in the southwest Pacific (this study) and southeast Pacific indicates there was substantial isotopic exchange with the atmosphere in the course of wind-driven upwelling and outgassing in the Southern Ocean. These results depart significantly from the large Δ¹⁴C depletion reported from the eastern tropical North Pacific (ETNP; ETNP; Fig. 3b). In the ETNP, Δ¹⁴C depletion began ~17.8 kyr BP, which precedes by ~1.7 kyr the onset of enhanced upwelling in the Southern Ocean that is invoked to transport this signal to the ETNP. However, the depletion there is coincident with the onset of Heinrich event 1 and the collapse of NADW formation. The position of the ETNP site in an eastern

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**Figure 2** | Records of radiocarbon activities, Antarctic temperatures and Southern Ocean upwelling across the last deglaciation. a, Antarctic ice-core δ¹⁸O \(((¹⁸O/¹⁶O)_{\text{sample}}/(¹⁸O/¹⁶O)_{\text{standard}} - 1) \times 1000\) from EDCML (green) and the hydrogen isotope deuterium (D) from EPICA Dome C (light blue), each of which is a temperature proxy, placed on the Greenland GISP2 timescale. b, Atmospheric radiocarbon activities from the Cariaco basin (black), placed on the Hulu cave timescale, and south Chatham Rise marine Δ¹⁴C from planktonic and benthic foraminifera (blue): Globigerina bulloides (planktonic; up-triangles show new data (this study); down-triangles show data from ref. 17), Globorotalia inflata (planktonic; squares), Neogloboquadrina pachyderma (planktonic; diamonds), mixed G. bulloides and N. pachyderma (planktonic; half-open square), and mixed benthics (filled circles). c, Atmospheric radiocarbon activities from the Cariaco basin (black), placed on the Hulu cave timescale, and Bay of Plenty Δ¹⁴C from core Rтоп03-JPC64 (red): G. inflata (planktonic; open squares) and mixed benthic foraminifera (closed circles). d, Opal fluxes, a proxy for upwelling south of the Antarctic polar front, from sediment core TN057-13-4PC (53.2°S, 5.1°E) in the Southern Ocean (SO). The vertical dashed lines denote climatic intervals of the deglaciation as indicated at top: Heinrich event 1 (H1; timing as determined by reduced deep-water flux in the North Atlantic), the Bolling–Allerod/Antarctic cold reversal (BA/ACR) and the Younger Dryas (YD). The shaded area corresponds to the period of enhanced upwelling in the Southern Ocean and the rapid drop in atmospheric Δ¹⁴C.

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boundary shadow zone makes subthermocline waters there sensitive to changes in steric height, thermocline depth\(^2\) and internal mixing dynamics\(^3\), which vary with subsurface water mass flux\(^7\). These would be sensitive to Northern Hemisphere thermohaline forcing. Also significant, the timing of the benthic \(^{14}C\) decrease in the ETNP coincides with an oceanic \(^{14}C\) plateau visible in marine fossils globally (~16.7–15.3 kyr BP\(^3\)), which is attributed to global thermohaline reorganization. Expansion of a deep, poorly ventilated, \(^{14}C\)-depleted North Pacific carbon reservoir with a different exit point, possibly the equatorial Pacific, could explain the ETNP \(^{14}C\) value (Fig. 3b). An alternative interpretation for the \(^{14}C\) difference between the ETNP and the South Pacific could be that \(CO_2\) exiting through the Southern Ocean did not fully tap into an old carbon pool.

Notably, the initial \(^{14}C\) decrease in marine records at ~18 kyr BP also coincides with the initial decrease in foraminifera \(^{13}C\) of the carbon isotope minimum event across the Southern and equatorial Pacific\(^4\). This event is generally interpreted as the entrainment of poorly ventilated, nutrient-rich waters from a glacial abyssal carbon pool. However, the absolute \(^{13}C\) minima at these widely dispersed sites are not contemporaneous, varying by 5 kyr (refs 4, 16, 29, 30), which points to local modification of any formation signal. The carbon isotope minimum event at our Southern Ocean (south Chatham Rise) site lasts from ~18 to 14 kyr BP (Fig. 3c).

Marine data from the southwest Pacific suggest a two-step ventilation of deep waters contributing \(CO_2\) to the atmosphere. The initiation of the atmospheric \(CO_2\) increase coincided with a nearly total collapse of NADW formation in the subpolar North Atlantic\(^4\) at the onset of Heinrich event 1, which affected density balances in waters outcropping in the Southern Ocean. We suggest that this brought \(^{14}C\)-rich, \(^{14}C\)-depleted water to the surface and that this water did not fully exchange isotopically with the atmosphere. Subsequent to this far-field forcing from the Northern Hemisphere, the ocean–atmosphere exchange in the Southern Ocean also responded to local forcing. Increasing Southern Ocean warmth\(^5\) caused changes in wind placement,\(^6\) altering shallow-water formation and lateral pumping dynamics\(^4,7\). Models indicate that entrainment of waters sourced from mid-depths such as upper CPDW fundamentally affect surface ocean productivity and \(^{14}C\) levels\(^6\). Although our data do not address the absence of a significant change in the Pacific deep-water \(^{14}C\) signal during the interval from 16.4 to 15.2 kyr BP\(^12\), intensified pumping from moderate depths is consistent with the lack of a significant change in Pacific deep-water \(^{14}C\) (refs 10, 12) through the early part of the deglaciation (17.8–16.2 kyr BP). The progressive shift of westerly winds to the south may have initiated a polar loop of shallow-water formation\(^4\) that successively stripped \(CO_2\) from mid-ocean depths before penetrating deeper into the ocean to release \(^{14}C\)-depleted \(CO_2\) from an older carbon reservoir. With moderate-deep waters as the main \(CO_2\) source before the Antarctic cold reversal, the \(^{14}C\)-depleted \(CO_2\) released after the Antarctic cold reversal must have been from a smaller, more depleted reservoir that remains to be volumetrically constrained and chemically defined\(^12\).

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Author Contributions K.A.R. participated in the RR0503 cruise, sampled cores, prepared sediments, speciated foraminifera for isotopic and radiocarbon analyses, performed all stable isotopic analyses and prepared figures. E.L.S. led the RR0503 cruise, sampled cores, speciated foraminifera for isotopic and radiocarbon analyses, prepared figures and wrote the paper. T.P.G. participated in the RR0503 cruise and performed all radiocarbon analyses. P.S. participated in the RR0503 cruise and identified all tephas. H.J.S. designed the study and, with T.M.H., supervised KAR during her MSc. R.Z. provided the MD core samples and shipboard colleagues during the Zheng leg 3 (RR0503) cruise funded by the National Science Foundation (NSF), which collected the RR core. Core MD97–2120 was collected through the International Marine Past Global Change Study (IMAGES) program and with the technical support of the Institut Polaire Français Paul Emile Victor (IPEV) who made the research vessel Marion Dufresne available for core retrieval. H.J.S., E.L.S. and T.P.G., and the shore analyses, were supported by NSF awards and the Evolving Earth Foundation, and the Geological Society of America provided support for K.A.R. during her MSc. R.Z. acknowledges support from the MICINN, Spain. A portion of this work was performed under the auspices of the US Department of Energy by Lawrence Livermore National Laboratory. We especially thank M. Cook for discussions, continuing input and suggestions throughout this study.

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