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Deglacial radiocarbon history of tropical Atlantic thermocline waters: absence of CO_2 reservoir purging signal

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Introductory paragraph

Atmospheric CO_2 increases during the Glacial to Holocene climate transition requires the outgassing of a deep and old oceanic CO_2 reservoir {Broecker, 2007 #639} thought to be located in the Southern Ocean {Anderson, 2009 #560;Broecker, Sikes 2000; 1998 #398;Skinner, 2010 #652}. The assumed and most readily applied purging mechanism involves subsurface Antarctic-sourced water circulating in the thermocline to the equatorial regions {Marchitto, 2007 #559; Stott, 2009 #577}, a view that has been met with conflicting results {De Pol-Holz, 2010 #635;Marchitto, 2007 #559;Rose, 2009 #656}. Here, we present evidence that deglacial radiocarbon ages of equatorial Atlantic thermocline waters were not anomalously old, contradicting the purging hypothesis. Using a novel approach (paired surface and deep-dwelling foraminifer ^{14}C analyses), we show that radiocarbon-depleted water characteristic of the isolated deep ocean CO_2 reservoir never reached the equatorial Atlantic thermocline. Together with other studies, these results require that the mechanism responsible for the deglacial CO_2 rise cannot invoke modern-like circulation and/or thermocline ventilation pathways.

Atmospheric CO_2 concentration recorded in ice cores show a two-step increase from Glacial to Holocene levels {Monnin, 2001 #653}. At these same times, radiocarbon component ($\Delta^{14}C$) measured in corals and planktonic foraminifera show a large decrease

{Fairbanks, 2005 #657;Hughen, 2006 #658}, which are unsupported by ^{14}C production changes alone {Hughen, 2006 #658}. These two steps in atmospheric CO_2 and radiocarbon history are associated with the climate events Heinrich 1 (ca. 16 ka BP) and the Younger Dryas (ca. 12 ka BP) {Broecker, 2000 #655;Petit, 1999 #654}. {Broecker, 2007 #639} proposed that CO_2 was stored in a deep oceanic reservoir during the glacial period, isolated from the atmosphere for several thousand years. {Adkins, 2002 #647} postulated that this old, CO_2-rich reservoir may have been near the Southern Ocean where pore water evidence indicates strongly stratified, high salinity bottom waters during the last glacial, a view supported by radiocarbon evidence {Sikes 2000; Skinner, 2010 #652}. The conventional purging hypothesis suggests that during the deglaciation, vigorous upwelling around Antarctica {Anderson, 2009 #560;Sachs, 2005 #646} opened up the reservoir, allowing the CO_2 and the ^{14}C-depleted water to spread in the ocean and the atmosphere.

This hypothesis remained unverified until two pulses of very low $\Delta^{14}C$ water were identified at intermediate depth off Baja (California) {Marchitto, 2007 #559} and in the eastern equatorial Pacific (EEP){Stott, 2009 #577}, coincident with the atmospheric CO_2 increases near Heinrich Event 1 and the Younger Dryas cooling events. The modern EEP is ventilated by intermediate Antarctic water (Antarctic Intermediate Water AAIW or SubAntarctic Mode Water SAMW) and deglacial AAIW/SAMW is believed to have extended much northward in the Atlantic and the Pacific {Anderson, 2009 #560;Bradtmiller, 2007 #650; Pahnke, 2008 #649} and up to Baja, California {Marchitto, 2007 #559}. Current views on the purging mechanism involve the ventilation of the deep oceanic CO_2 reservoir by Antarctic intermediate water masses circulating in the thermocline and outcropping in the low latitude upwelling regions; but incoherencies remain. This scenario is challenged firstly by the lack of ^{14}C anomaly in a deep water $\Delta^{14}C$ series in the production zone of AAIW/SAMW off Chile {De Pol-Holz, 2010 #635}. Secondly, the 3000 years lag between the last occurrence of the ^{14}C-depleted reservoir in the Southern Ocean {Skinner, 2010 #652} and the arrival of the ^{14}C anomaly in the equatorial Pacific {Marchitto, 2007 #559;Stott, 2009 #577} is not compatible with the modern 20-30 years ventilation age of the equatorial Pacific sub-surface water {Fine, 2001 #659}. This paper aims to test the hypothesis of a Southern Ocean reservoir ventilated by thermocline circulation during the deglaciation. AAIW/SAMW spreads in all three oceans {Talley, 1999 #641}, if Antarctic water drained the radiocarbon depleted CO_2 reservoir during the deglaciation, then the age difference between surface water and thermocline water in the equatorial Atlantic should increase tremendously during the evacuation of this reservoir.

The core RC24-08 is located in the Atlantic equatorial upwelling (1°20S, 11°54W, 3885m). In the Atlantic Ocean, AAIW/SAMW circulate in the interior south Atlantic, join the Western Boundary current along the Brazilian margin {Talley, 1999 #641} and is then upwelled and pushed eastward by the Equatorial Under Current (EUC) {Brandt, 2008 #616;Suga, 1995 #615}. Modeling experiment confirms observations that about 5 Sv (Sv ≡ 10^6 m^3 s^{-1}), out the about 19 Sv of southern-sourced water exported northward into the three oceans, reaches the equatorial Atlantic {Sen Gupta, 2007 #612}. Silicate content formed in the Southern ocean {Broecker, 1982 #382} traces the northward expansion of AAIW/SAMW (Figure 1). At the core location, AAIW/SAMW is found around 600 meters depth {Hanawa, 2001 #467;Sen Gupta, 2007 #612}. The bomb-corrected $\Delta^{14}C$ profile from the closest GLODAP station {Key, 2004 #643; see also Broecker and Olson, 1961} shows a steep gradient compared to mid-latitude profiles as ^{14}C-depleted water of southern origin are upwelled in the equatorial region (Figure 2). Model age of core RC24-08 was build on linear interpolation between ten ^{14}C dates on *G. ruber* calibrated into calendar age B.P. using the CALIB5 program and the standard marine correction reservoir {Stuiver, 1998 #67} (see supplementary material). Core RC24-08 has a high sedimentation rate of about 5.1cm/1000 years over the period covered by this study {McIntyre, 1996 #359} and high $CaCO_3$ content, reducing bioturbation and dissolution impacts on ^{14}C dates {Barker, 2007 #563}.

We measured oxygen and carbon stable isotope, trace element ratio and $\Delta^{14}C$ of three foraminifera species living at different water depth: *Globigerinoides ruber*, *Neogloboquadrina dutertrei* and *Globorotalia crassaformis*. Coretop oxygen isotopic composition ($\delta^{18}O$) and Mg/Ca ratio measurement for these species agree well with the expected values at the known habitat depth of these species (Figure 2). *G. ruber*

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Comment: It may be worth mentioning in the supplementary material that even if the surface age used to estimate calendrical/calibrated age model is wrong, that the gradient should be correct. Obviously, all bets are off if there a transient or slug of ‘funny’ water.

represents surface ocean condition, confirming its known calcification depth within the surface mixed layer (0-40m) (e.g. {Deuser, 1987 #17;Farmer, 2007 #623}). *N. dutertrei* calcifies at the base of the upper thermocline, around 100 meters depth {Anand, 2005 #199;Farmer, 2007 #623;Steph, 2009 #619; Fairbanks, Wiebe, and Be 1980}. *G. crassaformis* is a deep-dwelling foraminifera species, whose bulk shell $\delta^{18}O$ values indicate calcification around 500-600 meters depth {Bé, 1977 #205;Mücke, 1999 #126;Regenberg, 2009 #537;Steph, 2009 #619}. A series of $\delta^{18}O$ measurements on a transect of coretop samples between 35°N to 25°S in the Atlantic shows that *G. crassaformis* consistently calcifies over this depth range {Cléroux, 2009 #666}. In RC24-08, the $\delta^{18}O$ composition of *G. crassaformis* corrected for ice volume changes {Lambeck, 2001 #329} is very stable over the last 20 ka which give great confidence that this species kept its calcification depth (supplementary material). We therefore used *G. crassaformis* to monitor past variations in the radiocarbon content of basal thermoclines water (AAIW/SAMW) in the equatorial Atlantic.

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We calculated upper thermocline and deep-thermocline deglacial $\Delta^{14}C$ using paired radiocarbon analyses of *G. ruber*, *N. dutertrei* and *G. crassaformis* (Figure 3, see methodology). The modern $\Delta^{14}C$ difference between the atmosphere and seawater at 100 m and 600 m depth in the Eastern equatorial Atlantic are -60‰ and -100 ‰ respectively {Key, 2004 #643}. Upper thermocline $\Delta^{14}C$ reconstructions from *N. dutertrei* measurements follow the atmospheric composition {Reimer, 2009 #644} with an average offset of 80‰. Considering the average error on $\Delta^{14}C$ calculation of about 35‰ for these data, we conclude that over the last 20 ka, the age offset between the upper thermocline and the atmosphere remained consistent with modern value.

Around 4ka, the $\Delta^{14}C$ difference between *G. crassaformis* and the atmosphere is about -140‰, i.e. larger than the modern value. -140‰ is the modern $\Delta^{14}C$ of intermediate water in the south Atlantic (Glodap {Key, 2004 #643}), this would suggest enhanced upwelling around 4ka in the equatorial Atlantic but interpretation would require further investigation. Between the glacial period and the early Holocene, the $\Delta^{14}C$ of the deep thermocline reconstructed from *G. crassaformis* dating is on average 125‰ lower

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than the $\Delta^{14}C$ of the atmosphere. Through the deglaciation and over the periods with very depleted $\Delta^{14}C$ recorded off Baja, the $\Delta^{14}C$ difference between *G. crassaformis* and *G. ruber* is remarkably constant and follows atmospheric $\Delta^{14}C$. We conclude that during the early deglaciation the deep-thermocline of the equatorial Atlantic doesn't show an incursion of "old water". Our Atlantic thermocline $\Delta^{14}C$ records are very similar to the benthic-planktonic $\Delta^{14}C$ reconstruction measured off Chile at 1000 meters depth, i.e. within the modern formation area of AAIW/SAMW {De Pol-Holz, 2010 #635}, suggesting that both sites were influenced by the same water mass.

Several lines of evidence indicate that southern-sourced intermediate water invaded the North Atlantic intermediate depth during the glacial and deglacial period. Neodymium isotope in the tropical Atlantic {Pahnke, 2008 #649}, Cd/Ca ratio and $\delta^{13}C$ measurement in the North Atlantic {Rickaby, 2005 #561} or opal fluxes reconstruction from $^{231}Pa/^{230}Th$ ratio in the equatorial Atlantic {Bradtmiller, 2007 #650} have all been interpreted as AAIW replacing a weak North Atlantic Deep Water over glacial period. Several studies also showed increased upwelling conditions in the equatorial region during glacial and cold event {Abrantes, 2000 #668;Bradtmiller, 2007 #650}. Despite some disagreement between periods of maximal/reduced upwelling {Farmer, 2005 #661;Kim, 2003 #662}, all studies suggest glacial upwelling was greater than that of the Holocene. RC24-08 downcore $\delta^{13}C$ measurement and opal concentration analyses confirm the influence of water coming from the Southern Ocean and active upwelling during the deglaciation (supplementary material). The $\delta^{13}C$ *G. crassaformis* from the core RC24-08 increases of about 0.5‰ between glacial and late Holocene periods (Figure 3) in agreement with glacial $\delta^{13}C$ value lighter than Holocene in the South Atlantic {Marchitto, 2006 #636}. We conclude that over the last 20ka, AAIW/SAMW, or its deglacial equivalent, was always influencing the Atlantic equatorial thermocline but was not carrying any depleted ^{14}C water from the old southern reservoir.

We brought evidence that deep-dwelling foraminifera in the equatorial Atlantic, representing thermocline water fed by subsurface Antarctic water, did not see any abnormally old water over the deglaciation. We argue that intermediate Antarctic water was not purging the old CO_2 reservoir.

So far, the chase to locate the CO_2 reservoir and/or its evacuation vector led conflicting results (Figure 1, Table 1). Old water have been found in the Pacific Ocean at 23°N off Baja {Marchitto, 2007 #559} and in the equatorial region {Stott, 2009 #577}, in the Arabian Sea {Bryan, 2009 #669} and possibly off Brazil in the Atlantic {Mangini, 2010 #670}. In the North Atlantic, very old ventilation ages have been found both in the West and the East {Robinson, 2005 #681;Skinner, 2004 #680} at intermediate depth. Mostly interpreted as southern-sourced water invasion versus NADW during the deglaciation, the $\Delta^{14}C$ recorded in both studies are compatible with the very-depleted ^{14}C water found off Baja. Old water has not been found off Chile {De Pol-Holz, 2010 #635}, off New Zealand {Rose, 2010 #656} and in the equatorial Atlantic (this study). All these sites reside today in AAIW/SAMW pathway except the ones in the northern hemisphere mid-latitude. If AAIW/SAMW was not the vector for the purging of the deep reservoir then two possibilities remain. The deglacial intermediate circulation was fairly different from today and the model of a more northward extension in response to weak North Atlantic Deep water formation is over simplistic. If we assume that the northern sites where ^{14}C-water was found were bathed by southern water, then there must have been a very strong gradient in the ^{14}C content at some depth in the Atlantic and the Pacific. Future work to determine the depth of this gradient will aid in reconstructing the nature of intermediate depth circulation. The second option to reconsider is an isolated reservoir in the North Pacific {Broecker, 2004 #671;Galbraith, 2007 #672;Shackleton, 1988 #645}. With ^{14}C-depleted water found in the Atlantic sector of the Southern Ocean, in the Arabian Sea and eastern Pacific, can we imagine multiple reservoirs responding to different incentives at different times?

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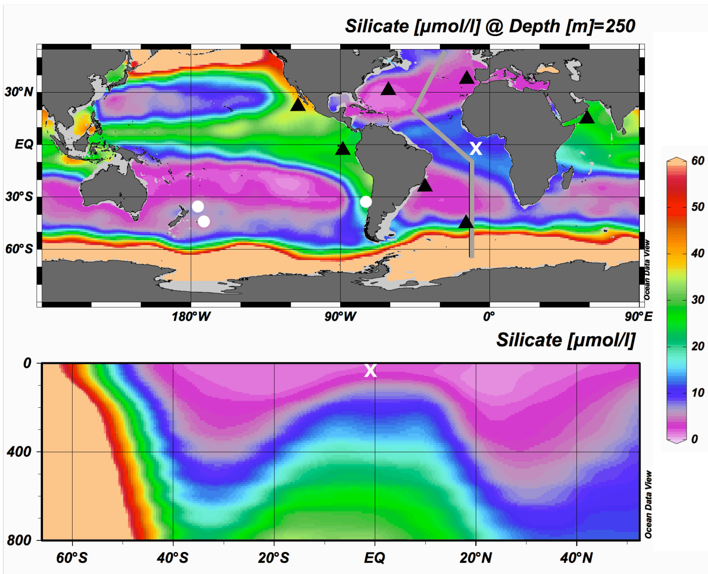


Figure 1: Silicate content at 250 meters depth (top) and along a meridional Atlantic profil drawn in black (bottom). The production zone, around Antarctica is showed in clay color, the silicate is then exported with AAIW / SAWM and upwelled in the equatorial region. The cross marks the location of core RC24-08. Studies with evidence for (black dots) or against (white dots) the presence of ^{14}C-depleted water are shown, see table 1 for references.

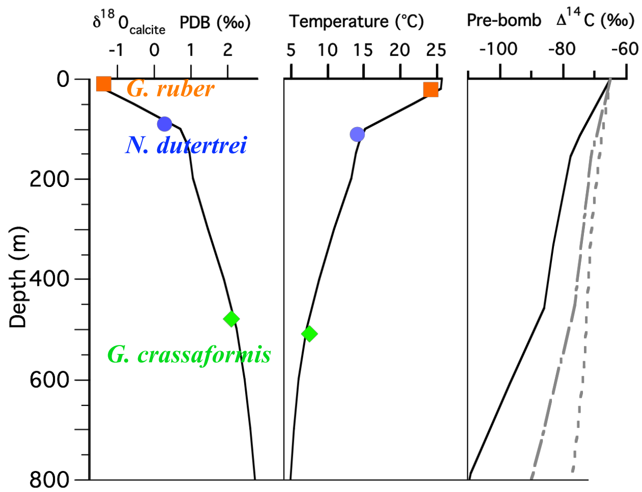


Figure 2: Modern oceanographic data at the core site and comparison with coretop foraminifera geochemical data (*G. ruber* in orange, *N. dutertrei* in bleu and *G. crassaformis* in green). From left to right: $\delta^{18}O$ calcite in equilibrium with $\delta^{18}O$ water composition {LeGrande, 2006 #436;Shackleton, 1974 #242}; temperature of seawater extracted from WOA {Conkright, 2002 #233} and calculated calcification temperature from Mg/Ca ratio; Pre-bomb $\Delta^{14}C$ ‰ extracted from the closest to RC24-08 core site GLODAP station {Key, 2004 #643} (black line), in the mid-latitude North Atlantic (short dotted line) and in the mid-latitude South Atlantic (long dotted line).

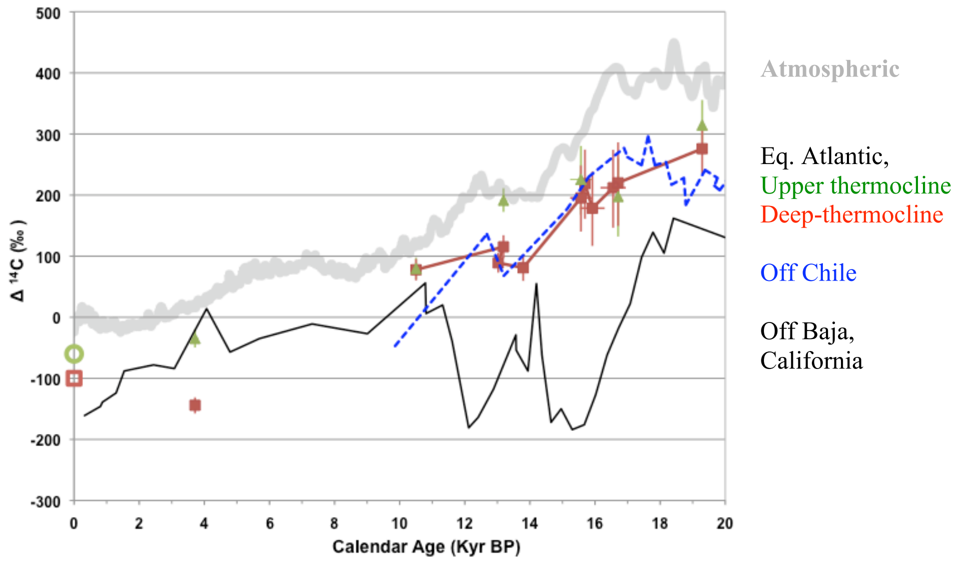
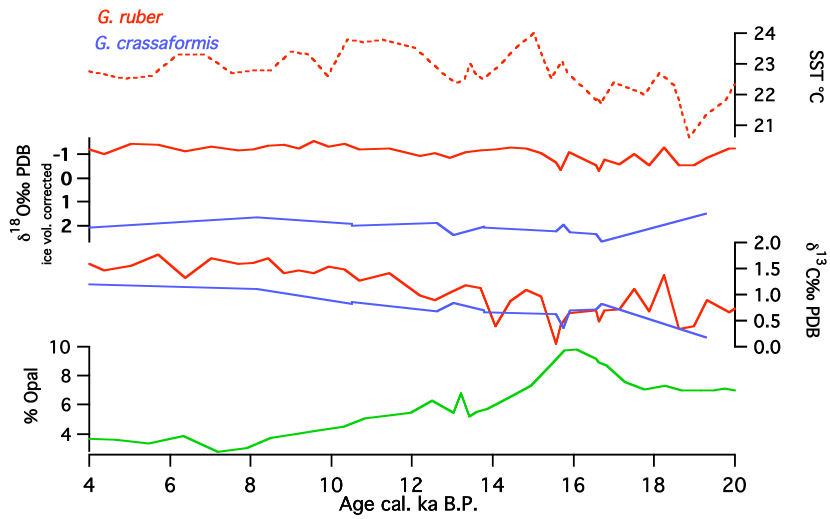


Figure 3: Atmospheric and oceanic radiocarbon activity over the last 20ka. Atlantic equatorial $\Delta^{14}C$ of the upper thermocline (green) and the deep-thermocline (red) follows the atmospheric signal (light thick grey) with an offset similar to the modern offsets, represented by the green circle and the red square for 100m and 600m depth respectively. $\Delta^{14}C$ signal reconstructed off Chile (dotted orange blue) {De Pol-Holz, 2010 #635}. These two AAIW/SAMW records contrast with $\Delta^{14}C$ reconstruction off Baja, California {Marchitto, 2007 #559} (orange). Around 16 ka, large error bars associated with deep-thermocline $\Delta^{14}C$ reconstruction results from large uncertainties on the ^{14}C age – calendar age calibration curve {Reimer, 2009 #644}.

Table 1

Location	latitude	longitude	depth dated	14C-depleted water found?	Reference
Baja California	23°5 N	111°6 W	705m	Yes	Marchitto at al., 2007
Arabian Sea	18°15 N	57°39 E	596m	Yes	Bryan et al., submitted
	17°59 N	57°35 E	820m		
Pacific equatorial	1°13 S	89°41 W	617m	Yes	Stott et al., 2009
Southern Atlantic ocean	44°46 S	14°12W	3770m	Yes	Skinner et al., 2010
Western North Atlantic	~35 °N	~60°W	1100-1700m	Yes	Robinson et al., 2005
East North Atlantic	37°48 N	10°10W	3146m	Yes	Skinner and Schackleton, 2004
Off Brazil	~ 22° S	~ 40° W	621m	Possibly	Mangini et al., 2010
	~ 24° S	~ 43° W	781m		
Chile	36°13 S	73°40 W	1000m	No	De Pol-Holz et al., 2010
New Zealand	37°22 S	177°00 E	651m	No	Rose et al., ACCEPTED
	43°32 S	174°55 E	1210m		
Equatorial atlantic	1°20 S	11°54W	~ 600m	No	This study



Supplementary material: RC24-08 downcore reconstruction from *G. ruber* (red) and *G. crassaformis* (blue). From top to bottom: SST from Mg/Ca measurement, ice volume corrected oxygen isotopic composition, carbon isotopic composition and opal percent. SST and $\delta^{18}O$ data show very little change, we infer that temperature and salinity water column structure remained very stable over the last 20ka. $\delta^{13}C$ and opal data both indicates enhanced influence of southern-sourced water and/or enhanced upwelling.

METHOD

Stable isotope measurement.

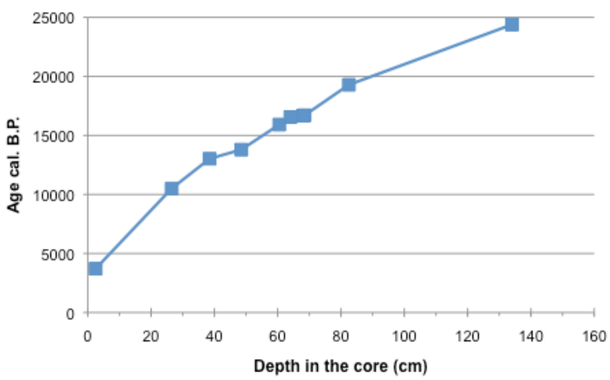
G. ruber was selected in the 250-355 μm size fraction, *G. crassaformis* and *N. dutertrei* were selected in the 355-425 μm size fraction. Samples were ultrasonically cleaned with ultra pure water and analyzed with Optima VG instruments at The Sate University of New York at Albany or at LDEO. Both labs use NBS19 as reference standard with a long term external reproducibility of 0.05‰ on $\delta^{18}O$.

Mg/Ca ratio measurement.

Coretop data for *G. ruber*, *G. crassaformis* and *N. dutertrei* are from RC24-07 (1.3°S, 11.9°W, 3899m) located very close to RC24-08 (1°20S, 11°54W, 3885m); downcore data for *G. ruber* are from RC24-08. Selected size fractions are 250-355 μm for *G. ruber* and 355-425 μm for *G. crassaformis* and *N. dutertrei*. Samples were crushed and cleaned following the full reductive and oxidative trace metal protocol {Boyle, 1987 #620} and analyzed with an Ultima-C Jobin Yvon at LDEO. Long-term measurement of international standard routinely measured is within 1% of the reported value with standard deviation of 0.039 mmol/mol. *G. ruber* Mg/Ca ratio were converted to temperature using {Dekens, 2002 #60} calibration, coretop Mg/Ca values of *G. crassaformis* and *N. dutertrei* were calculated with the general deep-dwelling equation of {Cléroux, 2008 #451} and applying a 10% correction on Mg/Ca ratio to take into account the different cleaning protocol {Barker, 2003 #28}.

Model Age

^{14}C dates were calibrated into calendar age B.P. using the CALIB5 program and the standard marine correction reservoir {Stuiver, 1998 #67}. The model age was calculated by linear interpolation between 15 radiocarbon dates. Data acquired at LLNL and data from McIntyre and Molfino, 1996 with asterisk * were performed on monospecies samples of *G. ruber*, other data from McIntyre and Molfino, 1996 are from monospecies sample of *N. dutertei*.



				CALIB5, Marine curve				
lab no.	Depth (cm)	14C age (years)	± Error	-2 sigma	+2 sigma	mean	± Error	Reference
144051	2.5	3770	30	3612	3817	3715	0.10	LLNL, july 2009, this study
144054	26.5	9655	50	10397	10608	10503	0.11	LLNL, july 2009, this study
144060	38.5	11535	35	12939	13115	13027	0.09	LLNL, july 2009, this study
144062	48.5	12325	50	13672	13916	13794	0.12	LLNL, july 2009, this study
144067	60.5	13790	50	15539	16297	15918	0.38	LLNL, july 2009, this study
OS-3935*	64	14300	70	16167	16946	16557	0.39	McIntyre and Molfino, 1996.
OS-3930*	68	14400	70	16276	17056	16666	0.39	McIntyre and Molfino, 1996.
144070	68.5	14440	70	16316	17102	16709	0.39	LLNL, july 2009, this study
144073	82.5	16570	100	19087	19495	19291	0.20	LLNL, july 2009, this study
OS-3929	134	20800	80	24101	24670	24386	0.28	McIntyre and Molfino, 1996.

^{14}C measurement and $\Delta^{14}C$ calculation

We selected *G. ruber* larger than 250 µm, *G. crassaformis* and *N. dutertei* larger than 355 µm to avoid any juvenile specimens that might calcify at shallower depth. Prior graphitization about

5mg of foraminifera were gently crushed and cleaned by ultrasonication with methanol and weak acid leaching (0.001 N HNO3). Samples were graphitized and analyzed at Lawrence Livermore National Laboratory. Age-corrected Δ14C were calculated using the following equation {Adkins, 1997 #664}:

$$\Delta^{14}C = \left[\left(\frac{e^{-^{14}Cage/8033}}{e^{-cal.age/8266}}\right) - 1\right] \times 1000$$

where cal.age is the calendar age derived from *G. ruber* ^{14}C age and calibrated with IntCal 2009{Reimer, 2009 #644}.