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THE MYSTERIOUS ^{14}C DECLINE

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ABSTRACT. Fundamental to the field of radiocarbon dating is not only the establishment of the temporal record of the calendar age-radiocarbon age offsets but also the development of an understanding of their cause. Although part of the decline in the magnitude of this offset over the past 40,000 can be explained by a drop in ^{14}C production rate associated with a progressive increase in the strength of the Earth's magnetic shielding, it is clear that changes in the distribution of ^{14}C among the Earth's active carbon reservoirs are also required. In particular, the steep 15% decline in the ^{14}C to C ratio in atmospheric CO_2 and surface ocean ΣCO_2, which occurred in a 3 kyr-duration interval marking the onset of the last deglaciation, appears to require that a very large amount (at least 5000 gigatons) of ^{14}C-deficient carbon was transferred to or within the ocean during this time interval. As no chemical or stable isotope anomaly associated with this injection appears in either the marine sediment or polar ice records, this injection must involve a transfer within the ocean (i.e. a mixing of 2 ocean reservoirs, one depleted in ^{14}C and the other enriched in ^{14}C). Although evidence for the existence of a salt-stabilized glacial-age abyssal ocean reservoir exists, a search based on benthic-planktic age differences and ^{13}C measurements appears to place a limit on its size well below that required to account for the steep ^{14}C decline.

INTRODUCTION

Some of the most exciting discoveries in the field of radiocarbon have been those related to the establishment of the time history of the magnitude of the so-called calendar offset. de Vries (1958) was the first to clearly demonstrate that the ^{14}C years deviated from calendar years. The Holocene history of these deviations was then mapped out by Suess (1965), Damon et al. (1978), and Stuiver (1982) based on measurements of ring-dated tree wood. Kromer and Becker (1993) managed to push this record beyond the Holocene into late deglacial time. A major breakthrough came when Fairbanks et al. (2005) obtained and ^{14}C-dated pristine glacial-age corals from shallow borings off the coast of Barbados. Bard et al. (1990), using the atom-counting technique developed at Caltech (California Institute of Technology) (Edwards et al. 1987), obtained ^{230}Th–^{234}U ages on these corals and showed, to everyone's amazement, that the older the coral sample, the larger the age offset.

Although I followed this ongoing quest with considerable interest, I never joined it. I've often wondered why. I started working in Lamont's ^{14}C laboratory on 15 June 1952 as a summer intern. By the summer of 1954, as a young graduate student doing research under J Lawrence Kulp's direction, I was placed in charge of its conversion from solid carbon counting to CO_2 counting. And over the next decade, I threw myself into directing the operation of Lamont's ^{14}C lab. As far as I can remember during this period, I made only 1 measurement on a sample of known age (bread, i.e. from Pompeii) and its ^{14}C age came out right on its calendar age. Perhaps this is why, early on, I failed to join the quest to calibrate the ^{14}C timescale. But by 1962 when a ^{14}C conference was held in Cambridge, England, I was fully aware that sizeable offsets did exist. I vigorously debated a proposal to switch from the use of Willard Libby's 5568 yr half-life to Ingrid Olsson's newly determined value of 5730 yr. I sided with those favoring the retention of the Libby half-life. My primary reason was that, as we were already aware that ^{14}C ages differed from calendar ages, any error in the half-life would be automatically taken into account by the offset correction. Further, as the seemingly simple conversion of ages from the Libby scale to the BC-AD scale proved sufficiently confusing to some of our "customers," we were, at that time, forced to list both our ages on both scales in our ^{14}C date lists. This alerted me to the much greater confusion that would arise when it came to distinguishing those ^{14}C ages published using the Libby half-life from those to be published using the Olsson half-

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life. For better or worse, the Libby half-life advocates won the day, and we still use the 5568 half-life.

Come to think of it, I did make an abortive attempt to determine the magnitude of the offset in glacial age samples. In the early 1960s, graduate student Aaron Kaufman and I made ^{14}C and ^{230}Th age determinations on $CaCO_3$ samples (shell, tufa, chara, and ostracods) from shoreline deposits of closed-basin lakes Lahontan and Bonneville. Although broad agreement between the 2 sets of ages was achieved, the scatter in the offsets between them was sufficiently large (and sometimes of one sign and sometimes of the other) that we had to settle for the consolation that the uranium series ages confirmed that we were obtaining a ^{14}C chronology useful in our quest to establish the first-order history of the fluctuations in the sizes of these lakes (Broecker and Kaufman 1965).

THE CLIMATE CONNECTION

Many years passed before I reentered the age-offset arena. When I did, it was not to add to the documentation of the magnitude of the offsets, but rather to learn what they had to say about the ocean's role in the abrupt climate changes recorded in Greenland ice. In the mid-1980s, I proposed that the Younger Dryas cold snap was the result of a shutdown in what I had dubbed the "great ocean conveyor" (Broecker 1991). Because of this, a reconstruction of the temporal changes in the ^{14}C to C ratio in the surface waters of the Cariaco Basin grabbed my attention (Hughen et al. 1998). The ^{14}C measurements on foraminifera shells from calendar-dated varved sediments suggested that during the first 200 yr of the Younger Dryas, the ^{14}C content of atmospheric and surface oceanic carbon rose by about 5% and then during the remaining thousand years of the Younger Dryas coasted back down again. I viewed this finding as a confirmation of my hypothesis. As the production of new deep water in the northern Atlantic (i.e. the great ocean conveyor's descending limb) currently serves as the conduit for about three quarters of the ^{14}C supplied to the deep sea (Broecker and Peng 1982), its shutdown at the onset of the Younger Dryas would have led to the backlogging of newly produced ^{14}C in the atmosphere and upper ocean. By my calculations, in 2 centuries such a shutdown could have produced Hughen's 5% increase. Then, as the result of the reduction in density caused by the continuing downward mixing of heat into the deep sea, a new mode of deep sea ventilation kicked into action leading to a drawdown of the excess ^{14}C stored in the upper ocean. As the frigid conditions in the northern part of the planet generated by the shutdown were not abated by this resumption, I assumed that the new source of deep water must have been located in the Southern Ocean rather than in the northern Atlantic (Broecker 2003).

THE 190 PER MIL ^{14}C TO C DECLINE

Another decade was to pass before these comforting thoughts regarding the rise and fall of ^{14}C during the Younger Dryas were thrown into turmoil. As were most of us interested in ^{14}C geochemistry, I was puzzled by the seeming mismatch between the actual magnitude of the decline in the ^{14}C to C ratio in the atmospheric and surface ocean carbon over the last 25 kyr and that predicted by the drop in ^{14}C production resulting from a strengthening of Earth's magnetic shielding. It appeared that the only way this difference could be explained was to postulate that a sizeable volume of the deep sea water had been isolated from communication with the remainder of the ocean for a significant portion of glacial time and then during the interval of deglaciation this reservoir was mixed back into the whole. This idea was given an observational footing by chlorinity measurements on pore waters from sediment recovered by the Deep Sea Drilling Program (Adkins et al. 2002). The deconvolution of such a record from the southern Atlantic Ocean yielded a glacial age salinity 1 g/L larger than that based on the other profiles (and for that expected from the buildup of excess continental ice). The likely explanation was that dollops of brine rejected during the annual wintertime expansions of the sea ice surrounding Antarctica were able to penetrate to the abyss, thereby creating a water mass enriched in salt content. The density contrast created by this excess salt would have prevented this water mass from mixing with the remainder of the ocean. If so, its ^{14}C would have gradually decayed away and the equivalent amount produced by cosmic rays would have accumulated in the overlying waters.

My colleague, Bob Anderson, raised the ante by pointing out to me that the demise of this isolated reservoir appears to have occurred largely during the first phase of deglaciation (a 3-kyr-duration interval following Heinrich event 1 and often referred to as the Mystery Interval) (Denton et al. 2006). A drop of 190‰ in the $\Delta^{14}C$ of atmosphere and surface ocean carbon occurred (see Broecker and Barker 2007)! This drop has been documented in 3 records (see Figure 1): 1) for the surface ocean based on ^{14}C measurements on foraminifera shells from annually layered Cariaco Basin sediment (Hughen et al. 1998, 2004); 2) for the atmosphere based on ^{14}C measurements on a ^{230}Th-dated stalagmite (Beck et al. 2001); and 3) for the surface ocean based on ^{14}C measurements on pristine corals obtained from borings offshore from the island of Barbados (Fairbanks et al. 2005). The fact that the accumulation of ^{10}Be in Greenland ice did not undergo a dramatic decrease during the Mystery Interval appears to rule out a large drop in cosmic ray production as the cause for this ^{14}C decline (Muscheler et al. 2004). The only other viable explanation is that a large reservoir of ^{14}C-depleted ocean water was mixed into the remainder of the ocean during this time period.

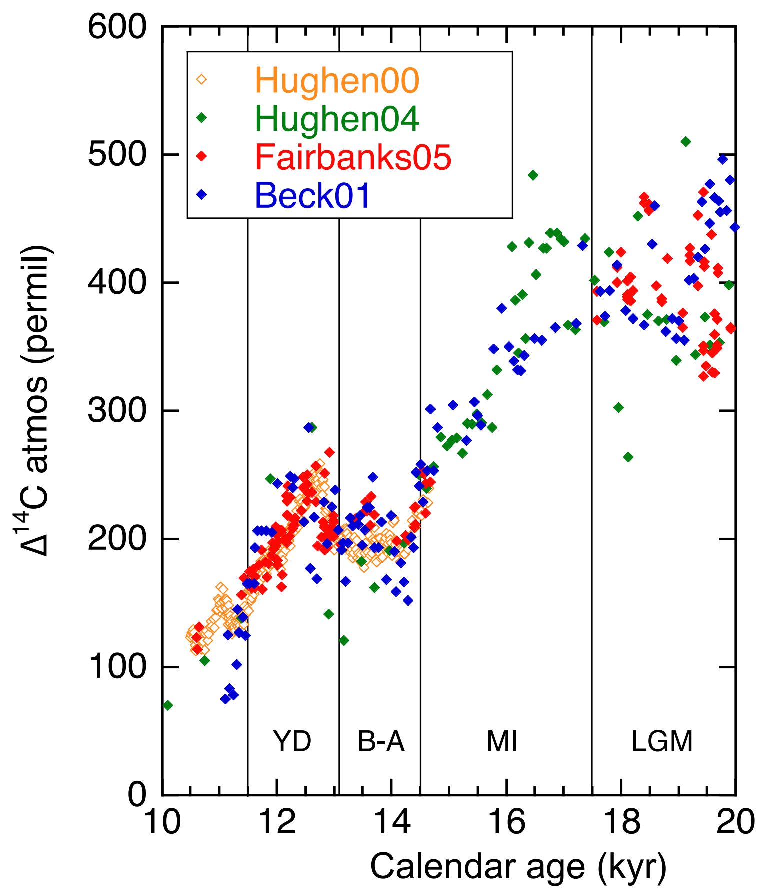

Figure 1 Reconstructions of the atmospheric ^{14}C for the period 10–20 thousand calendar yr ago. Hughen00 and Hughen04 represent data from Cariaco Basin sediments (Hughen et al. 2000, 2004), Fairbanks05 data come from Th-dated pristine submerged corals (Fairbanks et al. 2005), and Beck01 data are from a Bahamian stalagmite (Beck et al. 2001). As can be seen, in all records there is a decrease in ^{14}C from about +400‰ to about +200‰ during the 17.5- to 14.5-kyr time interval.

It is easy to rule out any other source of ^{14}C-depleted carbon. Even if the added carbon were free of ^{14}C, an enormous 5000 gigatons would be required. This corresponds to 15% of the inorganic carbon dissolved in today's ocean. Were such a huge amount of carbon introduced in any other form, its impact on ocean and atmosphere carbon chemistry and stable isotopic composition would stand out like a sore thumb in records from ice cores and ocean sediments. No hint of such a sore thumb has been found.

BENTHIC-PLANKTIC AGE DIFFERENCES

Working together with Stephen Barker and Elizabeth Clark, I have endeavored to confirm the existence of a glacial-age ^{14}C-depleted ocean reservoir. Our approach has been centered on the use of the age difference between coexisting benthic and planktic foraminifera shells. In today's ocean (or more specifically, the preindustrial ocean), the apparent ^{14}C age of equatorial deep water relative to that of overlying surface water ranges up to about 1600 yr. If, during peak glacial time, an isolated abyssal reservoir existed, then one would expect to find abyssal sediments with benthic-planktonic age differences far larger than this. It is my opinion that, despite considerable effort, no such sediment has been found. This comes as a surprise because the size of the required reservoir is so large that it should be easy to find. If it were isolated for only 1 ^{14}C half-life (i.e. for the duration of marine isotope stage 2), then its volume would have to have been about 30% that of the entire ocean. If salt stabilized, this large volume would have occupied the entire ocean below 2.9 km depth. Even if the reservoir had been isolated for several ^{14}C half-lives, it would have to have occupied the entire volume below about 3.9 km depth (see Figure 2).

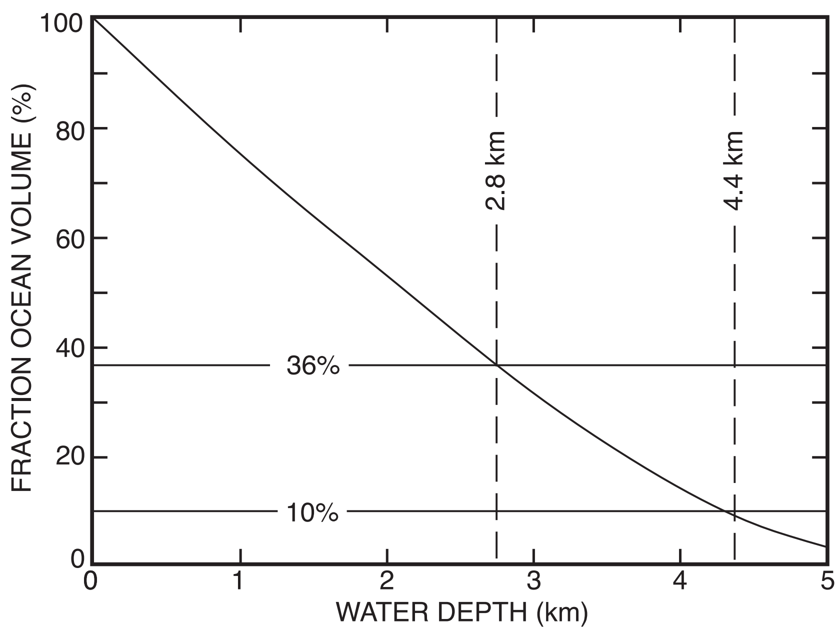

Figure 2 Plot of fraction of the ocean's volume lying below any given water depth. About 36% of the ocean's volume lies below 2.8 km (i.e. the depth of sediment core MD01-2386) and only about 10% below 4.4 km (i.e. the depth of sediment core TTN013-18).

This said, before outlining the results of this search, it is necessary to point out that even so, documenting the existence of this reservoir constitutes a difficult challenge. One aspect of the challenge has to do with the generally low sedimentation rates found at water depths below 3 km. They are for the most part so low that bioturbation mixes glacial-age foraminifera with Holocene-age foraminifera. Coupled with temporal changes in the relative abundance of benthic foraminifera and the planktonic foraminifera of choice, this can create large biases in the benthic-planktic age difference (Peng and Broecker 1984). Another problem has to do with dissolution. Sediments deposited at depths greater than 4 km in the Pacific Ocean are often devoid of the whole planktic shells required for picking because of dissolution-induced breakage. Finally, if the sediments are obtained close to steep continental margins where lithogenic fluxes are high enough to obviate the impact of bioturbation, deposition of previously deposited (i.e. pre-aged) foraminifera by slumping or by swift boundary currents becomes a problem (Broecker et al. 2004).

THE RECORD IN PACIFIC OCEAN SEDIMENT

As the deep Pacific accounts for roughly half of the ocean volume, it must have hosted a sizable portion of the sought-after isolated abyssal reservoir. Hence, we have focused our search on the sediments from this ocean. Our most reliable and most complete record comes from a core located at 2.8 km depth in the western equatorial Pacific. As in the age range of interest its sedimentation rate is about 50 cm per kyr, bioturbation is not an issue. The agreement among the ^{14}C ages for coexisting planktic species speaks to the absence of large biases due to either reworking or to secondary calcite. As shown in Figure 3, the benthic-planktic age differences obtained for 16 samples extending back to 20 kyr in calendar age show no significant differences from that based on the preindustrial ^{14}C

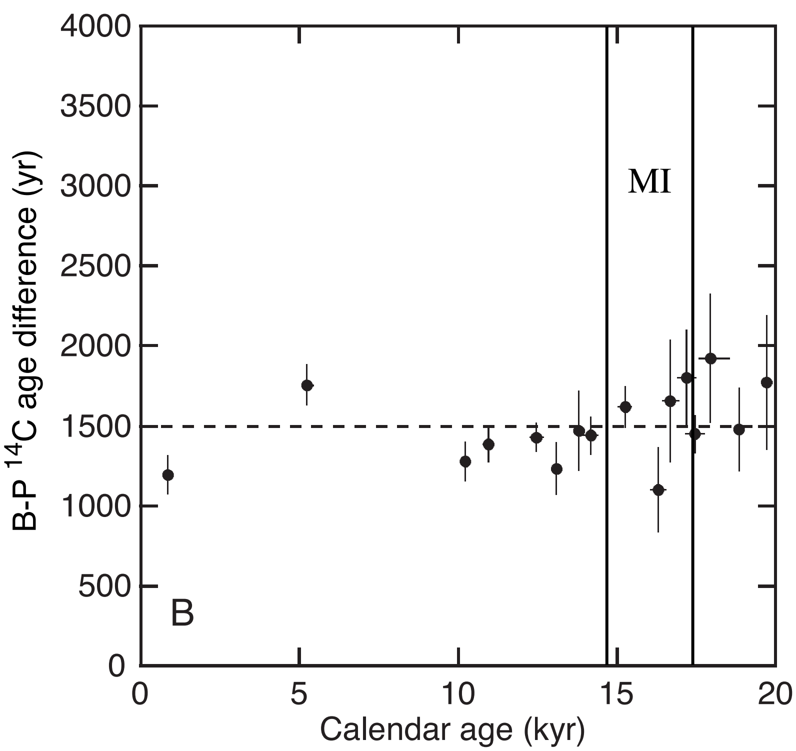

Figure 3 Record of the benthic-planktonic ^{14}C age offset from sediment core MD01-2386 (Broecker et al. 2007, 2008). The horizontal dashed line is today's age difference. The 2 vertical lines bracket the Mystery Interval.

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distribution in the deep Pacific (Broecker et al. 2007, 2008). This tells us 3 important things: 1) if an isolated reservoir existed prior to the Mystery Interval, its upper boundary was deeper than 2.8 km and hence its volume could be no more than 36% that of the ocean; 2) if the water in a ^{14}C-depleted abyssal reservoir were stirred into the remainder of the ocean during the Mystery Interval, this mixing event did not perturb the ambient surface to 2.8 km ^{14}C to C difference in the western equatorial Pacific Ocean; 3) the constancy of the difference of ^{14}C to C ratio between the surface and 2.8 km in the equatorial Pacific is consistent with a nearly constant ventilation rate of the deep Pacific over the last 20 kyr (i.e. from the peak of the last glaciation to present).

A number of additional less complete records of benthic-planktic age differences have been obtained at shallower depths in the deep Pacific (see Figure 4). These include 3 from the western North Pacific (Ohkushi et al. 2004), 4 results from the South China Sea (Broecker et al. 1990), 3 from the Sea of Okhotsk (Keigwin 1998), and 5 from the western equatorial Pacific (Broecker et al. 2004). In a recent paper, Stott et al. (2007) present 8 results from western equatorial Pacific sediment core MD98-2181 ranging from 9 to 27 ^{14}C kyr. The average benthic-planktic ^{14}C age difference is 1493 yr. Again, no age trend is seen. This strengthens our conclusion that if a glacial-age ^{14}C-depleted reservoir existed, it must have been located deeper than 2.8 km.

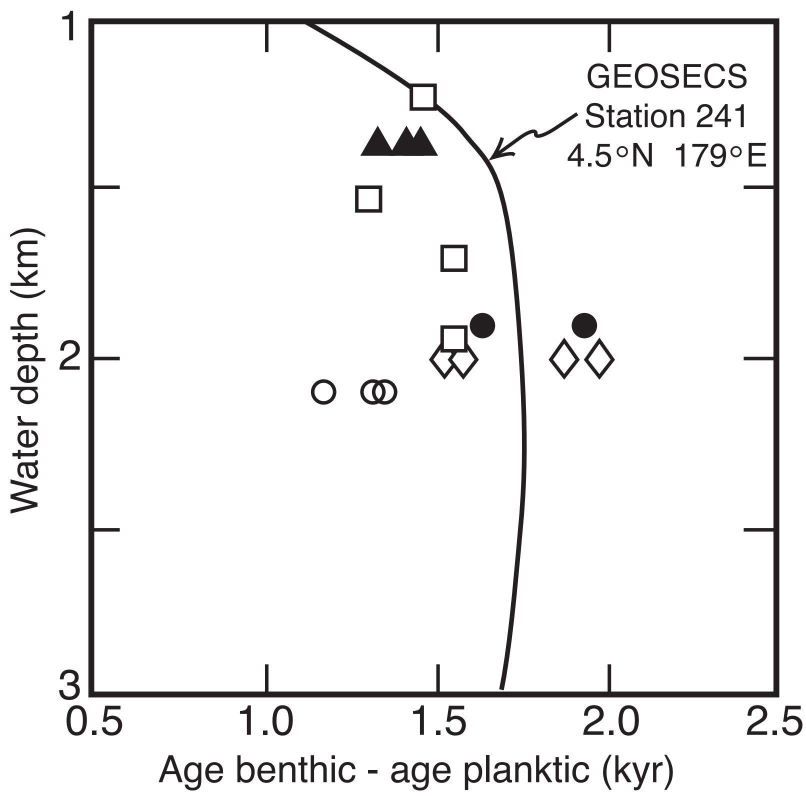

Figure 4 Summary of LGM ventilation times based on benthic-planktic age differences for cores from the western Pacific. The open squares are for cores from the Sea of Okhotsk (Keigwin 1998), the solid triangles are for a core from off of Japan (Ohkushi et al. 2004), the open diamonds are from a core at a depth of 2.7 km in the South China Sea (sill depth 2.0 km) (Broecker et al. 1990), the solid circles are for the Admiralty Island core and the open circles are for Morotai Basin core (Broecker et al. 2004). The solid line is based on measurements on water samples collected as part of the GEOSECS survey.

Only a few benthic planktic age measurements exist for Pacific Ocean glacial-age sediments from water depths greater than 2.8 km. Further, those that do, do not pass the reliability tests we require. This said, none show the very large age differences we seek. One of these results is of particular importance. It comes from a core located at 4.4 km depth in the central equatorial Pacific. As shown in Figure 2, only 10% of the ocean lies beneath this depth. A single glacial-age sample yielded a benthic-*G. sacculifer* age difference of 1600 yr (Figure 5). While the validity of this result is marred by the fact that the ages for 3 other planktic species came out within the error of that for the benthics, it is difficult to imagine that downward bioturbation of young shells or secondary ^{14}C reduced the age of the benthics by several thousands of years. As can be seen in Figure 5, the *G. sacculifer* age falls on a line joining those for much younger and much older samples. But, I must admit it would be preferable if sediment with an accumulation rate much higher than 3 cm/kyr were to yield such a result.

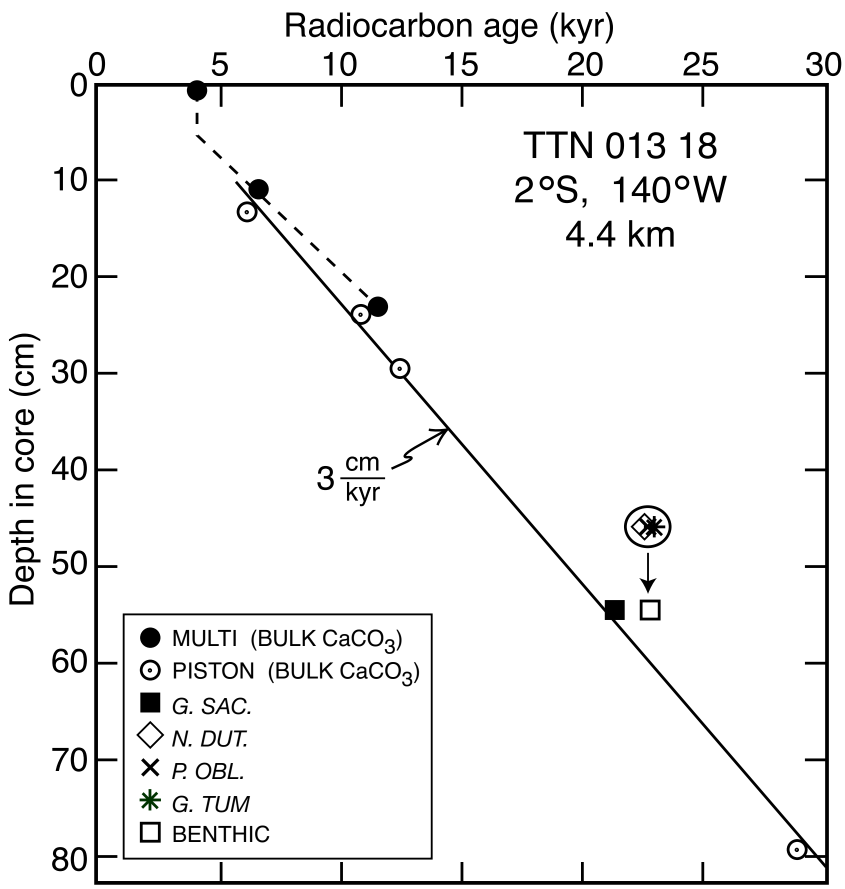

Figure 5 ^{14}C results on a low-accumulation-rate abyssal core from the central equatorial Pacific (Broecker and Barker 2007).

Another set of results of importance comes from a core located far north in the Pacific (54.6°N, 145.8°W, 3.65 km). Four LGM age samples yielded benthic-planktic age differences between 1600 and 1900 yr (Gailbraith et al. 2007). These results place a firm upper limit of the upper boundary of the isolated reservoir. It must be deeper than 3.6 km and hence cannot occupy any more than 20% of the ocean's volume.

OTHER INDICATORS

An additional source of evidence is available in the search for the glacial-age isolated abyssal reservoir. Besides losing a major portion of its ^{14}C, this reservoir must have lost a major portion or even all of its dissolved oxygen. Going hand in hand with decreases in O_2 are decreases in the ^{13}C to ^{12}C ratio in the reservoir's dissolved inorganic carbon. For each 10 µmol/kg drop in O_2, there would have been a 0.1‰ drop in $\delta^{13}C$. Such a decrease would be recorded by benthic foraminifera. The ^{13}C record kept in benthic foraminifera has a big advantage in that single shell can be measured and its ^{18}O to ^{16}O ratio can be used to verify it formed during the last glacial maximum time interval. This being the case, bioturbation is not an issue. Although this technique has not been widely employed in the search for the isolated reservoir, one very telling record is available. Alan Mix obtained complete benthic ^{13}C and ^{18}O records for 2 sediment cores from 4.4 km depth in the central equatorial Pacific. The glacial ^{13}C values he found are no different than those for cores in the 2 km depth range (see Figure 6). Taken at face value, this suggests that the glacial O_2 content at 4.4 km depth was no more than 20 µmol/kg lower than that at 2 km. In today's ocean, the correlation between $\delta^{13}C$ and $\Delta^{14}C$ suggests that the rate of O_2 consumption is on the order of 150 µmoles per millennium. Hence, if the water at 4.4 km had been isolated for 5 or so millennia, one would expect that *all* its O_2 would have been utilized. Yet, the $\delta^{13}C$ for benthic foraminifera appear to constrain the residence time to have been similar to that at 2 km. Indeed, this is what the benthic-*G. sacculifer* age difference obtained in one of these cores appears to be telling us (see above).

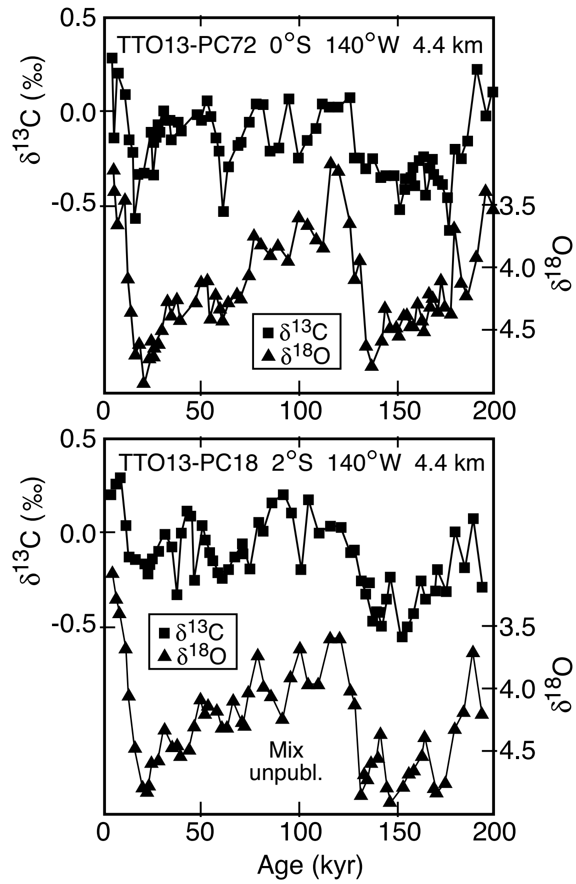

Figure 6 Stable isotope records for benthic foraminifera from 2 neighboring central Pacific abyssal sediment cores (results obtained by Alan Mix, published by Broecker and Barker 2007). As indicated by the ^{18}O to ^{16}O ratios, each record covers the last two 100,000-yr cycles. The important point is that the ^{13}C to ^{12}C ratios for samples from the full glacial intervals average 0.3 to 0.4‰ lower than those for times of peak interglaciation. Hence, they suggest that during peak glacials, oxygen utilization at 4.4 km was not significantly greater than that for the 2- to 3-km range.

THE SMOKING GUN?

Dramatic evidence for the demise of a ^{14}C-depleted ocean reservoir comes from a sediment core raised from intermediate water depth on the eastern margin of the North Pacific Ocean (23.5°N, 111.6°W, 705 m). ^{14}C measurements on benthic foraminifera contained in this core (well dated by correlation with the record in Greenland GISP2 ice core) reveal 2 time intervals when the site appears to have been bathed in water highly depleted in ^{14}C. One of these corresponds to the Mystery Interval and the other to the Younger Dryas (see Figure 7). Marchitto et al. (2007) envision this ^{14}C-depleted water to have been that from the sought-after glacial-age isolated ocean reservoir as it was being mixed into the remainder of the ocean. In order to explain why this event is not recorded in the 2- to 3-km water depth range in the western Pacific Ocean, these authors postulate that during its demise the isolated reservoir water upwelled in the Southern Ocean and part of it was promptly exported northward as intermediate depth water. Considering that during the Mystery Interval the apparent age of waters off Central America dropped by more than 3 kyr and that considerable entrainment of surrounding water must have occurred during its long passage from the abyss, if this scenario is correct, then the original isolated reservoir water must have been even more depleted in ^{14}C.

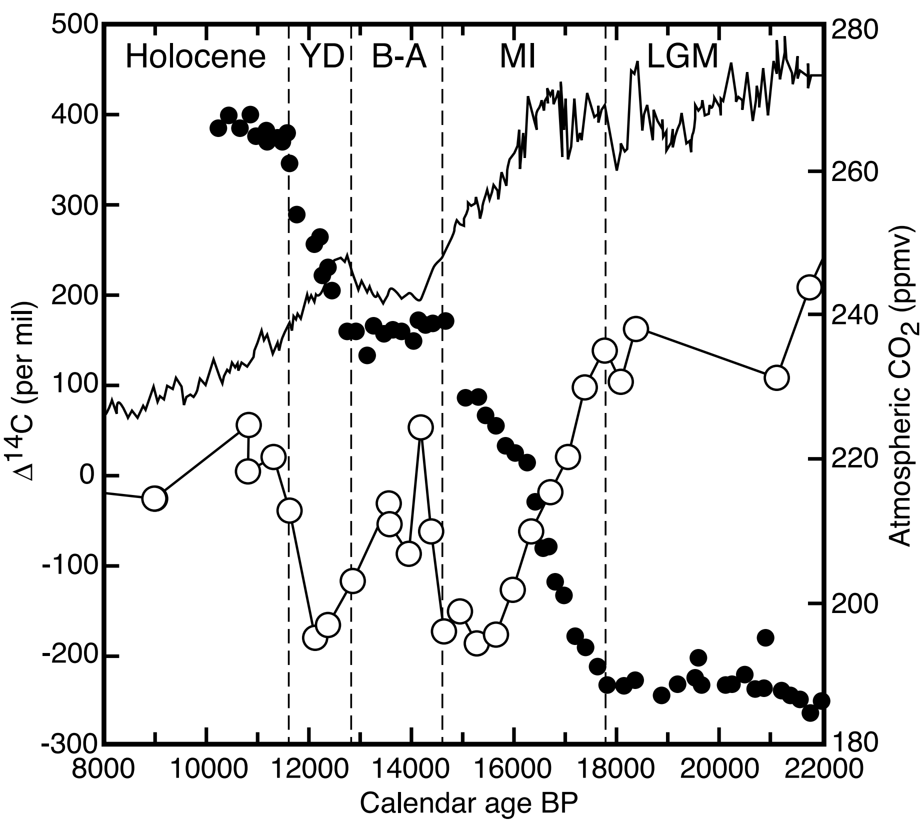

Figure 7 The ^{14}C record for benthic foraminifera from an intermediate-water-depth core (720 m) raised in the eastern Pacific Ocean off Central America (open circles). The timescale is set by correlation with the GISP2 ice core. The ^{14}C record reconstructed for the atmosphere is shown by the solid curve. Also shown is the CO_2 record from the Vostok Antarctica ice core (solid circles). The point of this diagram prepared by Marchitto et al. (2007) is that the large dips in $\Delta^{14}C$ for the benthics during the Mystery Interval (MI) and Younger Dryas (YD) reflect the demise of the ^{14}C-depleted abyssal reservoir. Reprinted with permission.

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I can think of 2 other ways in which to explain the low $\Delta^{14}C$ value for benthic foraminifera from the Mystery Interval and Younger Dryas. One is that the shells are reworked by bottom currents during the Mystery Interval and Younger Dryas. If so, similar ^{14}C anomalies should be present in the planktic shells (which have yet to be measured). The other is that the shells could have calcified in ^{14}C-depleted pore waters, in which case they should have distinctly lower ^{13}C to ^{12}C ratios than those from the adjacent sediment sections (again, as yet to be measured). But I must admit, neither of these explanations seems likely.

Were it not for the fact that the steep drop in upper ocean ^{14}C during the Mystery Interval appears to require the existence of a highly ^{14}C-depleted reservoir, I would declare the Marchitto et al. (2007) scenario to be highly improbable. But, as such a reservoir must have existed, I suspect that they may have found the smoking gun. However, as outlined above, when it comes to locating the actual reservoir, we seem to have painted ourselves into a corner.

CONCLUSIONS

Clearly the quest to explain the sharp drop in the ^{14}C to C ratio that occurred during the Mystery Interval is an ongoing one. In that sense, this paper is a progress report. The implications extend beyond the Mystery Interval. For, if the benthic foraminifera results obtained by Marchitto et al. (2007) record the demise of an isolated abyssal reservoir, then this demise must have occurred in 2 steps, one during the Mystery Interval and the other during the Younger Dryas. If so, my earlier thought that during the Younger Dryas a shutdown in the delivery of ^{14}C to the deep sea was followed by a rejuvenation of ventilation requires rethinking!

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