# No signature of abyssal carbon in intermediate waters off Chile during deglaciation

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At the end of the Last Glacial Maximum (19,000 to 11,000 years ago), atmospheric carbon dioxide concentrations rose while the  $\Delta^{14}$ C of atmospheric carbon dioxide declined<sup>1,2</sup>. These changes have been attributed to an injection of carbon dioxide with low radiocarbon activity from an oceanic abyssal reservoir that was isolated from the atmosphere for several thousand years before deglaciation<sup>3</sup>. The current understanding points to the Southern Ocean as the main area of exchange between these reservoirs<sup>4</sup>. Intermediate water formed in the Southern Ocean surrounding Antarctica would have then carried the old carbon dioxide signature to the lower-latitude oceans<sup>5,6</sup>. Here we reconstruct the  $\Delta^{14}$ C signature of Antarctic Intermediate Water off the coast of Chile for the past 20,000 years, using paired <sup>14</sup>C ages of benthic and planktonic foraminifera. In contrast to the above scenario, we find that the  $\Delta^{14}C$ signature of the Antarctic Intermediate Water closely matches the modelled surface ocean  $\Delta^{14}$ C, precluding the influence of an old carbon source. We suggest that if the abyssal ocean is indeed the source of the radiocarbon-depleted carbon dioxide, an alternative path for the mixing and propagation of its carbon dioxide may be required to explain the observed changes in atmospheric carbon dioxide concentration and radiocarbon activity.

The fundamentals behind the atmospheric CO<sub>2</sub> increase and  $\Delta^{14}C$  decrease during the so-called mystery interval (17.5–14.5 kyr BP) and the contemporaneous deglaciation have remained elusive<sup>3</sup>. Ice-core records show that the overall deglacial increase in atmospheric CO<sub>2</sub> was  $\sim 100$  ppm (parts per million) (ref. 1), and records of atmospheric  $\Delta^{14}$ C based on absolutely dated surface corals<sup>7</sup>, speleothems<sup>8</sup> and planktonic foraminifera<sup>2</sup> indicate an overall deglacial decrease of  $\sim$  300%. Importantly, the reduction in atmospheric radiocarbon concentration occurred without an apparent concomitant decrease in the cosmogenic production rate<sup>2</sup>. If the radiocarbon and the CO<sub>2</sub> changes were linked through the injection of old carbon from the ocean, they suggest that the abyss remained isolated from the atmosphere for several thousand years during the last ice age and became progressively depleted in <sup>14</sup>C owing to radioactive decay<sup>3</sup>. This situation differs from the modern ocean where the oldest waters are 'only' 2,200 yr in the deep North Pacific Ocean as the result of a limited-but vet existent-communication between the abyssal ocean and the surface of the Southern Ocean.

The existence of an isolated deep-sea reservoir during the last ice age implies that the communication between the upper Southern Ocean and the ocean interior did not exist during the Last Glacial Maximum (LGM). This may have been caused by permanent sea-ice cover<sup>9</sup> or by upper-ocean stratification<sup>10</sup>. Yet, despite the fact that there is scattered evidence that an isolated abyssal reservoir may have existed during the LGM (refs 11, 12), its effect on the atmospheric compositional changes has remained questionable. An important turning point came when Marchitto et al.<sup>5</sup> found strong supporting evidence for this mechanism. By analysing the glacial to Holocene radiocarbon content of intermediate water benthic foraminifera in the eastern North Pacific Ocean, they produced a  $\Delta^{14}$ C time series that showed striking consistency with the deglacial atmospheric  $\Delta^{14}$ C and CO<sub>2</sub> changes. In particular, they documented two pulses of extremely depleted  $\Delta^{14}$ C water (>4,000 yr older than the contemporaneous atmosphere) coinciding with the mystery interval and the Younger Dryas (11,500–12,900 yr вр). Their interpretation is that these excursions represent the changes in the initial (preformed) radiocarbon content of Antarctic Intermediate Water (AAIW) acquired by mixing with the upwelled old abyssal water south of the subantarctic frontal zone. More recently, the finding of an extremely large <sup>14</sup>C age difference between benthic and planktonic foraminifera from intermediate depths off the Galapagos<sup>13</sup> seems to support this interpretation, which is also consistent with an apparent increase in Southern Ocean upwelling intensity interpreted from enhanced opal accumulation around Antarctica during the mystery interval<sup>14</sup>. If this hypothesis is correct, the effect of the deep ocean ventilation on atmospheric properties has been unveiled, paving the way for a robust theory of glacial-interglacial CO<sub>2</sub> changes<sup>15</sup>.

It is worth noting that the radiocarbon records off Baja California and Galapagos are only circumstantially related to the Southern Ocean overturning strength because neither site is in the direct path of Southern-Ocean-derived water masses in the modern ocean. If the extremely  $\Delta^{14}$ C-depleted waters documented at these sites are indeed related to the efflux of CO<sub>2</sub> from the deep sea through the Southern Ocean, then the presumed transport of the signal to the north must have been in the form of intermediate water masses that formed in contact with the surface of the Southern Ocean, namely Subantarctic Mode Water/AAIW (or glacial analogues)<sup>5</sup>. To more directly evaluate the preformed <sup>14</sup>C content of AAIW during deglaciation, we have produced a  $\Delta^{14}C$ time series based on paired planktonic and benthic foraminifera <sup>14</sup>C ages in sediment core SO161-SL22 from the Chile margin in the eastern South Pacific Ocean (Fig. 1). The core was raised from 1,000 m water depth off central Chile (36°13' S, 73°40' W) and it constitutes a 894 cm sedimentary sequence of high-deposition terrigenous, hemipelagic sediments spanning the past  $\sim$ 22,000 yr. Synoptic and climatological observations show that the core site is bathed by low-salinity, high-oxygen AAIW as expected from its proximity to the main Subantarctic Mode Water/AAIW formation region west of Drake Passage, and its subsequent northward flow within the eastern branch of the South Pacific subtropical gyre<sup>16-18</sup> (Supplementary Information). A potential caveat for our method is

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# Salinity 35.0 34.8 34.6 34.6 34.4 34.4 34.2 34.2 34.0

**Figure 1 | The study area and core locations.** Map of ocean salinity at 1 km depth showing the location of cores SO161-SL22 (36° S, eastern South Pacific Ocean, this study), VM21-30 (1° S, equatorial Pacific Ocean<sup>13</sup>) and the sedimentary composite MV99-MC19/GC31/PC08 (23.5° N, eastern North Pacific Ocean<sup>5</sup>) discussed in the text. The intermediate-depth low-salinity signatures of AAIW and North Pacific Intermediate Water can be distinguished by the purple-pink colours.

that our study area is an active upwelling centre today that could eventually corrupt the planktonic–benthic <sup>14</sup>C age difference by bringing old deep waters into the surface. However, the similarity between our planktonic <sup>14</sup>C-anchored  $\delta^{18}$ O stratigraphy and the Antarctic ice-core air-temperature proxy history suggests that our planktonic foraminifera were not affected substantially by this process (see Methods and Supplementary Information).

Today, AAIW hydrographic characteristics (including  $\Delta^{14}$ C) are set by the mixing of Southern Ocean surface water with upwelled circumpolar deep water, with subsequent entrainment into the thermocline of the South Pacific Ocean by means of subduction. This process mainly takes place during the austral winter where a significant deepening of the mixed layer occurs in combination with downwelling forcing imposed by the regional wind stress curl<sup>18</sup>. The Southern Ocean preset surface properties are in turn, a function of the upwelling intensity south of the polar front, which brings deep, old (low  $\Delta^{14}$ C) circumpolar waters in contact with the atmosphere, effectively increasing the Southern Ocean surface water <sup>14</sup>C-reservoir age to >1,000 yr instead of the  $\sim$ 400 ± 100 yr of the temperate ocean<sup>19</sup>. Thus, more intense upwelling south of the Drake Passage is accompanied by lower  $\Delta^{14}C$  (>age) in the surface of the Southern Ocean and consequently in AAIW. A similar argument applies to the stable isotope ratio  $\delta^{13}$ C as exploited previously by Spero and Lea<sup>6</sup> to explain the pervasive appearance of a deglacial planktonic  $\delta^{13}$ C minimum in tropical records. We assume that the transition time of AAIW from its formation area to our site is almost instantaneous with respect to the <sup>14</sup>C half life, so that our age-corrected benthic radiocarbon measurements should reflect the preformed <sup>14</sup>C of AAIW, unaffected by <sup>14</sup>C decay.

Between the LGM and the start of the mystery interval (21,000-18,000 yr BP) the apparent ventilation age of intermediate waters off Chile decreased from ~800 to 300 uncorrected <sup>14</sup>C years (Fig. 2). Throughout the mystery interval, the age difference between the co-occurring planktonic and benthic foraminifera fluctuated around 400 yr. It rose at the beginning of the Bølling/Allerød to decrease again during the Younger Dryas and finally increase during the early Holocene (although our Holocene data point has large errors associated with small sample size). For reference, the modern pre-industrial AAIW <sup>14</sup>C age difference with the surface was ~550 years near our site<sup>20</sup>. We note the complete



Figure 2 | Apparent age difference between paired benthic and planktonic foraminifera in core SO161-SL22. Benthic minus planktonic foraminifera raw <sup>14</sup>C ages plotted against the planktonic calibrated calendar age. The vertical error bars are the sum of the  $\pm 1\sigma$  error of the planktonic and benthic foraminifera. The horizontal error bars are the  $\pm 1\sigma$ error of the calibration to calendar years with zero uncertainty in the reservoir correction. The yellow shading indicates the 'mystery interval'. Heinrich event 1 (H1), Younger Dryas (YD) and the Bølling/Allerød (BA) are indicated. The orange arrow shows the estimated preindustrial, prebomb value for our site<sup>20</sup>.

lack of an incursion of any 'old water' during the mystery interval as seen in the Galapagos paired benthic–planktonic data where the age difference reached more than  $\sim$ 6,000 yr, more than ten times larger than at the Chile margin during the same time period. This comes as a surprise because AAIW is the hypothetical signal carrier during the deglaciation to both Galapagos and Baja California.

To isolate our results from the effect of atmospheric  $\Delta^{14}$ C transients, we calculated the age-corrected  $\Delta^{14}C$  of our Chile margin benthic foraminifera using the co-occurring calibrated planktonic foraminifera calendar age and compared it with the results from off Baja California and the Galapagos (Fig. 3). Throughout the deglaciation our  $\Delta^{14}$ C record followed very closely the evolution of the upper temperate ocean with no significant deviations larger than the estimated intermediate depth oceanatmosphere  $\Delta^{14}$ C difference of -120% (ref. 20). Figure 3 shows that the consistency of our data with the assumed surface ocean record is in clear contrast with the benthic results off Baja California and the Galapagos. Our core site lies close to the modern-day AAIW formation region and therefore should contain the most direct evidence of the initial AAIW  $\Delta^{14}$ C values exported from the Southern Ocean to lower latitudes (that is, overturning strength). At face value, our data suggest no mixing between AAIW and any hypothetical isolated abyssal reservoir during the mystery interval or the Younger Drvas despite the fact that Atlantic Nd isotope data show that vigorous AAIW export was apparently taking place during those events<sup>21</sup>. Second, from a radiocarbon perspective, it is clear that very different water masses bathed the eastern South and the equatorial to North Pacific Ocean intermediate depths during the mystery interval and the early Younger Dryas. Although we do not have data for the late Younger Dryas, the overall evolution of our  $\Delta^{14}$ C suggests a close connection with the surface ocean (Fig. 3).

The fact that our  $\Delta^{14}$ C record does not deviate substantially from the projected temperate mixed layer throughout the deglaciation suggests that AAIW's formation was at a location where the reservoir effect was at times only 200–300 yr older than the temperate ocean, that is ~300 yr younger than the Southern Ocean surface today. If the upwelling–reservoir age relationship was valid during the last deglaciation, we would expect that the apparent re-invigorated Southern Ocean upwelling during the mystery

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#### Figure 3 | Deglacial radiocarbon activity of Pacific intermediate waters.

The radiocarbon activity ( $\Delta^{14}$ C) of intermediate waters off Chile (orange circles, 1 $\sigma$  combined AMS-<sup>14</sup>C and calendar age error lines), Baja California<sup>5</sup> (red, errors as published) and off the Galapagos<sup>13</sup> (blue, no errors reported) compared with the calculated mixed-layer values of CalPal\_2007\_Hulu calibration curve (grey, 1 $\sigma$  errors). The YD, BA, Antarctic Cold Reversal (ACR) and H1 climate events are indicated.

interval<sup>14</sup>—supposedly the result of the poleward migration of the Southern Westerlies<sup>22</sup>—would have increased the surface Southern Ocean reservoir age owing to the mixing with the old respired CO<sub>2</sub> present in the abyss during the LGM. An alternative could be that the water mass ventilating our site during the deglaciation was subducted north of the subantarctic front and hence, completely free of any Southern Ocean imprint. Increasing evidence from the Atlantic, both in observations<sup>21</sup> and in models<sup>23</sup>, and from the West Pacific<sup>24,25</sup> indicates a re-invigoration of AAIW production during the deglaciation. We therefore believe that the idea of a deglacial AAIW being formed elsewhere and routed in a completely different way so as to not affect the intermediate waters off Chile is unlikely. Unless we accept an alternative formation mechanism for the young intermediate waters observed in our record, the opal/upwelling intensity proxy for the Southern Ocean may need further revision.

The  $\Delta^{14}$ C record produced by Marchitto *et al.*<sup>5</sup> continues to be an important palaeoceanographic landmark given its accurate dating and time resolution. Our results show that, despite the initial encouraging supporting evidence provided by some observations<sup>13,14</sup> and theory<sup>22</sup>, the cautionary quantitative remarks expressed by Broecker and co-workers<sup>3,26</sup> towards the existence of an abyssal <sup>14</sup>C-depleted reservoir still hold, including the extent to which this mechanism can account for the observed changes in atmospheric radiocarbon and CO<sub>2</sub> during the deglaciation. The increasing number of sedimentary records showing anomalous low radiocarbon at intermediate depths around the world demands further investigation on its causes. Our work shows that their connection with the Southern Ocean overturning strength is not entirely straightforward and alternative explanations for their presence should be explored. Understanding glacial-interglacial CO<sub>2</sub> cycles still remains an elusive test for our proficiency in the study of the Earth as a system.

## Methods

Sediments from core SO161-SL22 were washed and sieved to  $63 \,\mu\text{m}$  before drying and weighing. Paired benthic and planktonic foraminifera were picked from the >150  $\mu$ m fraction. To minimize the effect of bioturbation, we mostly (three exceptions) dated benthic *Uvigerina peregrina* at horizons with local maxima in its abundance (individuals/dry weight) (Supplementary ref. S1).

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Radiocarbon analyses were measured at the National Ocean Science Accelerator Mass Spectrometer Facility, Woods Hole Oceanographic Institution and the Keck Carbon Cycle Accelerator Mass Spectrometer facility of the University of California, Irvine.

The age model for the core was derived from the calibrated planktonic ages using the CalPal\_Hulu\_2007 calibration curve. Previous work<sup>27</sup> in our study area has shown no substantial deviation of the local surface reservoir age ( $\Delta R$ ) with the mean 400 yr assumed for the temperate ocean, despite it being an active upwelling centre today. Compelling evidence for this is the similarity between the planktonic  $\delta^{18}$ O stratigraphy in our core and the Antarctic Byrd Ice-core  $\delta^{18}$ O record (Supplementary Fig. S1) when a constant reservoir correction of 400 yr is used for the planktonic <sup>14</sup>C age. The only way that the AAIW  $\Delta^{14}$ C values off Chile would be as low as in Baja California is by applying a reservoir correction of the order of 3000 yr (even larger to reproduce the Galapagos data set). Although plausible in terms of the hypothetical upwelling of the old reservoir corrupting our planktonic ages, the age model derived from it would fix the deglacial  $\delta^{18}$ O shift in our core at unrealistically young ages (see Supplementary Information).

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#### References

- 1. Monnin, E. *et al.* Atmospheric CO<sub>2</sub> concentrations over the last glacial termination. *Science* **291**, 112–114 (2001).
- Hughen, K., Southon, J., Lehman, S., Bertrand, C. & Turnbull, J. Marine-derived <sup>14</sup>C calibration and activity record for the past 50,000 years updated from the Cariaco Basin. *Quat. Sci. Rev.* 25, 3216–3227 (2006).
- Broecker, W. & Barker, S. A 190‰ drop in atmosphere's Δ<sup>14</sup>C during the 'Mystery Interval' (17.5–14.5 kyr). *Earth Planet. Sci. Lett.* 256, 90–99 (2007).
- Fischer, H. *et al.* The role of Southern Ocean processes in orbital and millennial CO<sub>2</sub> variations—A synthesis. *Quat. Sci. Rev.* doi:10.1016/j.quascirev.2009.06.007 (2009).
- Marchitto, T., Lehman, S., Ortiz, J., Fluckiger, J. & van Geen, A. Marine radiocarbon evidence for the mechanism of deglacial atmospheric CO<sub>2</sub> rise. *Science* 316, 1456–1459 (2007).
- Spero, H. & Lea, D. The cause of carbon isotope minimum events on glacial terminations. *Science* 296, 522–525 (2002).
- Fairbanks, R. *et al.* Radiocarbon calibration curve spanning 0–50,000 years BP based on paired Th-230/U-234/U-238 and C-14 dates on pristine corals. *Quat. Sci. Rev.* 24, 1781–1796 (2005).
- Beck, J. *et al.* Extremely large variations of atmospheric <sup>14</sup>C concentration during the last glacial period. *Science* 292, 2453–2458 (2001).
- Stephens, B. & Keeling, R. The influence of Antarctic sea ice on glacial–interglacial CO<sub>2</sub> variations. *Nature* 404, 171–174 (2000).
- Francois, R. *et al.* Contribution of Southern Ocean surface-water stratification to low atmospheric CO<sub>2</sub> concentrations during the last glacial period. *Nature* 389, 929–935 (1997).
- 11. Adkins, J., McIntyre, K. & Schrag, D. The salinity, temperature, and  $\delta^{18}$ O of the glacial deep ocean. *Science* **298**, 1769–1773 (2002).
- Sikes, E. L., Samson, C. R., Guilderson, T. P. & Howard, W. R. Old radiocarbon ages in the southwest Pacific Ocean during the last glacial period and deglaciation. *Nature* 405, 555–559 (2000).
- Stott, L., Southon, J., Timmermann, A. & Koutavas, A. Radiocarbon age anomaly at intermediate water depth in the Pacific Ocean during the last deglaciation. *Paleoceanography* 24, PA2223 (2009).
- Anderson, R. *et al.* Wind-driven upwelling in the southern ocean and the deglacial rise in atmospheric CO<sub>2</sub>. *Science* **323**, 1443–1448 (2009).
- 15. Toggweiler, J. Shifting westerlies. Science 323, 1434–1435 (2009).
- Sloyan, B. & Rintoul, S. Circulation, renewal, and modification of Antarctic mode and intermediate water. J. Phys. Oceanogr. 31, 1005–1030 (2001).
- Iudicone, D., Rodgers, K., Schopp, R. & Madec, G. An exchange window for the injection of Antarctic Intermediate Water into the South Pacific. *J. Phys. Oceanogr.* 37, 31–49 (2007).
- Schneider, W. & Bravo, L. Argo profiling floats document Subantarctic Mode Water formation west of Drake Passage. *Geophys. Res. Lett.* 33, L16609 (2006).
- Butzin, M., Prange, M. & Lohmann, G. Radiocarbon simulations for the glacial ocean: The effects of wind stress, Southern Ocean sea ice and Heinrich events. *Earth Planet. Sci. Lett.* 235, 45–61 (2005).
- Key, R. *et al.* A global ocean carbon climatology: Results from Global Data Analysis Project (GLODAP). *Glob. Biogeochem. Cycles* 18, GB4031 (2004).
- Pahnke, K., Goldstein, S. & Hemming, S. Abrupt changes in Antarctic Intermediate water circulation over the past 25,000 years. *Nature Geosci.* 1, 870–874 (2008).
- Toggweiler, J., Russell, J. & Carson, S. Midlatitude westerlies, atmospheric CO<sub>2</sub>, and climate change during the ice ages. *Paleoceanography* 21, PA2005 (2006).
- Knorr, G. & Lohmann, G. Southern Ocean origin for the resumption of Atlantic thermohaline circulation during deglaciation. *Nature* 424, 532–536 (2003).

## NATURE GEOSCIENCE DOI: 10.1038/NGEO745



- Pahnke, K. & Zahn, R. Southern Hemisphere water mass conversion linked with North Atlantic climate variability. *Science* 307, 1741–1746 (2005).
- Bostock, H., Opdyke, B., Gagan, M. & Fifield, L. Carbon isotope evidence for changes in Antarctic Intermediate Water circulation and ocean ventilation in the southwest Pacific during the last deglaciation. *Paleoceanography* 19, PA4013 (2004).
- 26. Broecker, W. The mysterious C-14 decline. Radiocarbon 51, 109–119 (2009).
- Mohtadi, M. *et al.* Deglacial pattern of circulation and marine productivity in the upwelling region off central-south Chile. *Earth Planet. Sci. Lett.* 272, 221–230 (2008).

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#### Author contributions

R.D.P.-H. and L.K. conceived the study. R.D.P.-H. collected the foraminifera and wrote the paper with the help of all the co-authors. J.S. supplied ideas that shaped the final version. D.H. and M.M. collected the core material and provided the samples. Core SO161-SL22 was retrieved for M.M. doctoral thesis work. All authors contributed to the writing of this manuscript.

### Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturegeoscience. Reprints and permissions information is available online at http://npg.nature.com/reprintsandpermissions. Correspondence and requests for materials should be addressed to R.D.P.-H.