

# Cosmogenic isotope $^{14}\text{C}$ : Production and carbon cycle

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## Global $\Delta^{14}\text{C}$ cycle

Radiocarbon is produced by galactic cosmic radiation interacting with atmospheric nitrogen, and enters the global carbon cycle as  $^{14}\text{CO}_2$ , which is well mixed within the atmosphere. Photosynthesis fixes  $\text{CO}_2$ , and hence  $^{14}\text{CO}_2$ , into plant organic matter and the global food chain. In addition,  $\text{CO}_2$  dissolves in water (primarily seawater) to form dissolved inorganic carbon (DIC) that can then be incorporated into marine carbonates. Conventional  $^{14}\text{C}$  dating assumes that initial  $^{14}\text{C}$  concentration has remained constant. However, atmospheric and surface ocean  $^{14}\text{C}$  concentrations have changed notably through time. This is due to changes in either the rate of  $^{14}\text{C}$  production in the atmosphere (a function of geomagnetic field intensity and solar variability), or the distribution of  $^{14}\text{C}$  between different reservoirs in the global carbon cycle (primarily deep ocean ventilation).

The Earth's geomagnetic field serves to shield the atmosphere from incoming cosmic rays, and when the magnetic field strength increases,  $^{14}\text{C}$  production decreases (and vice versa). Similarly, solar wind distorts the Earth's geomagnetic field in a way that reduces  $^{14}\text{C}$  production, and a rise in solar activity will cause a decline in  $^{14}\text{C}$  production. Records of  $^{14}\text{C}$  production variability in the past have been constructed using two primary methods: (1) as a function of past changes in geomagnetic field intensity (Laj et al., 2004) and, (2) by comparison to other cosmogenic nuclides (e.g.  $^{10}\text{Be}$  and  $^{36}\text{Cl}$ ; Muscheler et al., 2005). Over the past 50 kyr, the pattern of changes in  $^{14}\text{C}$  production reconstructed using the two methods agree very well (Fig. 1), although the absolute magnitude of  $^{14}\text{C}$  production rate is still largely uncertain. The global carbon cycle contains several reservoirs that exchange carbon on timescales relevant to the lifetime of  $^{14}\text{C}$  (101-104 years). Within the deep ocean in particular,  $^{14}\text{C}$  is sequestered from atmospheric exchange long enough for decay to reduce the deep ocean  $^{14}\text{C}$  activity significantly. Changes in the rate of exchange between the deep ocean and the atmosphere, through fluctuations in the meridional overturning circulation (i.e. the North Atlantic component of global thermohaline circulation), can strongly influence atmospheric  $^{14}\text{C}$  activity. However, little is known about the detailed spatial and temporal history of deep-ocean ventilation changes and the magnitude of resulting changes in surface-ocean and atmospheric  $^{14}\text{C}$ .

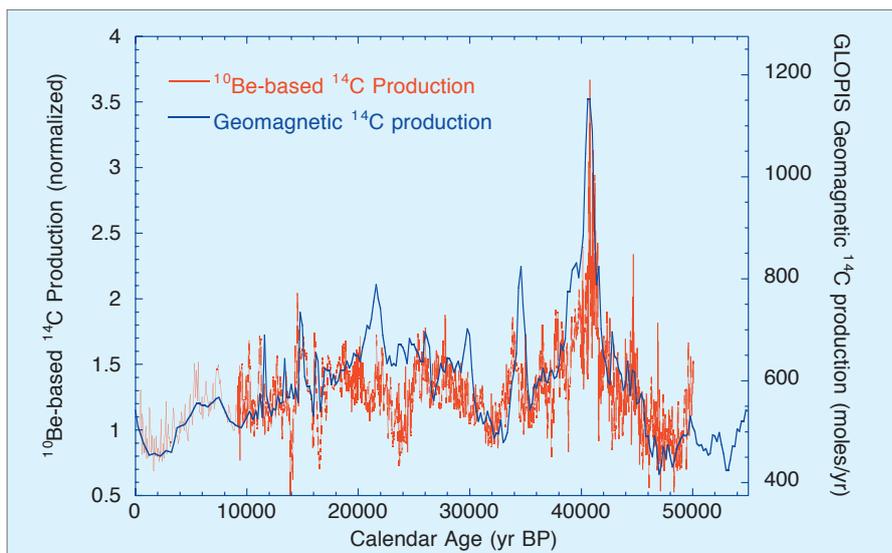


Figure 1:  $^{14}\text{C}$  production rate changes from 50 to 0 kyr reconstructed using  $^{10}\text{Be}$  flux measured in Greenland ice cores (Muscheler et al., 2005), and as a function of geomagnetic field intensity (Global Paleointensity Stack, Laj et al., 2004; Masarik and Beer, 1999). The two methods show strong agreement in temporal patterns of production change, including distinct peaks associated with the Laschamp and Mono Lake geomagnetic minima.

## Observed $\Delta^{14}\text{C}$ changes in the past

Recently, marine-based calibration data back to 50 kyr have been provided by  $^{14}\text{C}$ - and  $^{230}\text{Th}$ -dated corals with irregular sample spacing (Fairbanks et al., 2005; Cutler et al., 2004; Bard et al., 2004), and at higher resolution from marine sediments of the Cariaco Basin (Hughen et al., 2006) and Iberian Margin (Bard et al., 2004). The sediment records show distinct millennial-scale climate variability that can be reliably correlated with Dansgaard-Oeschger (D-O) events in Greenland ice cores and, more recently,  $^{230}\text{Th}$ -dated Hulu Cave speleothems. These correlations have been used to transfer the calendar chronologies onto the  $^{14}\text{C}$  series in order to provide calibration data sets. Cariaco Basin and Iberian Margin  $^{14}\text{C}$  data linked to the  $^{230}\text{Th}$  Hulu Cave chronology show excellent agreement with data from  $^{230}\text{Th}$ -dated fossil corals back to 33 kyr, and continue to agree despite increased scatter back to 50 kyr (Fig. 2).  $^{14}\text{C}$  calibration data independent from a marine-reservoir age-correction have been obtained from a  $^{230}\text{Th}$ -dated speleothem on Socotra Island in the Arabian Sea. These data show a close match to the marine sediment and coral  $^{14}\text{C}$  records between 40-50 kyr (Fig. 2). The observed convergence of data sets from dispersed archives and geographic locales will likely provide, in the near future, the basis for an extended  $^{14}\text{C}$  calibration back to 50 kyr.

## Geochemical modeling and global carbon cycle changes

The  $\Delta^{14}\text{C}$  data reveal highly elevated  $\Delta^{14}\text{C}$  values during the Glacial period. In order

to investigate the implications of the observed  $\Delta^{14}\text{C}$  record, we use a carbon cycle box model to simulate fluxes between the atmosphere, terrestrial biota plus soil/detritus, surface and deep oceans, and shallow and deep marine sediments containing organic and inorganic carbon. Reservoir inventories and rates of exchange are specified from consensus estimates for the modern (pre-industrial) carbon cycle (Hughen et al., 2004).  $^{14}\text{C}$  production rates are calculated as a function of geomagnetic field intensity (Laj et al., 2004), with a contemporary  $^{14}\text{C}$  production rate of 2.02 atom  $\text{cm}^{-2} \text{sec}^{-1}$  (Masarik and Beer; 1999). As noted previously, however, this  $^{14}\text{C}$  production rate exceeds the observed sum of  $^{14}\text{C}$  in active reservoirs. A constant scaling factor is therefore applied to the production rates in order to tune the model reservoir  $\Delta^{14}\text{C}$  values at model year 0 to observed modern values (Atmosphere 0‰, Terrestrial Biosphere -5‰, Surface Ocean -53‰, Deep Ocean -159‰). A model simulation with fixed modern exchanges ("full carbon cycle") but variable  $^{14}\text{C}$  production rate produces a temporal pattern of  $\Delta^{14}\text{C}$  change similar to paleo-observations, and matches the magnitude of  $\Delta^{14}\text{C}$  change particularly during the Holocene (Fig. 2). However, the simulation produces maximum  $\Delta^{14}\text{C}$  of only  $\sim 300$ ‰ for the interval 20-40 kyr, whereas observed  $\Delta^{14}\text{C}$  exceeds the simulated changes by as much as  $\sim 400$ ‰, most prominently around  $\sim 30$  and  $\sim 40$  kyr.

In this simple model, reducing the surface-to-deep-ocean exchange produces an additional atmospheric and surface ocean

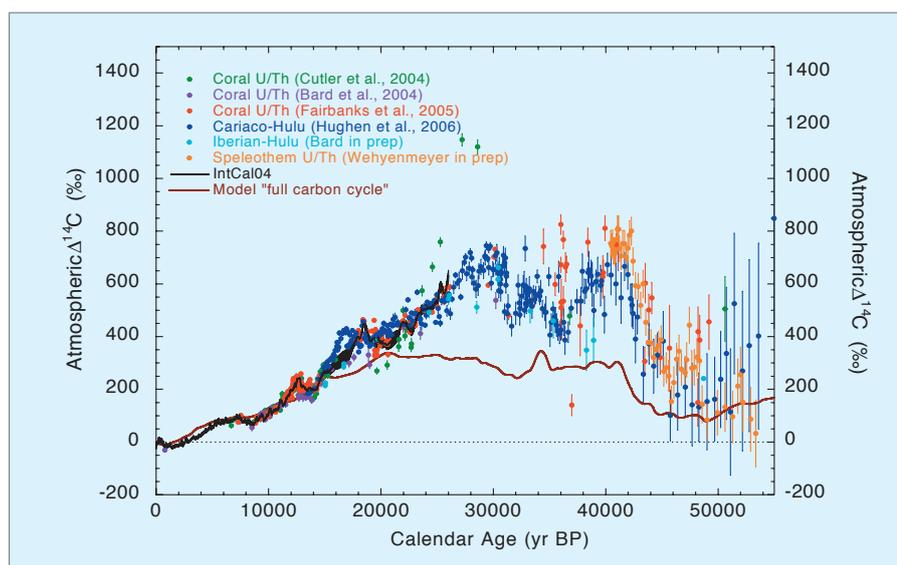


Figure 2:  $^{14}\text{C}$  activity ( $\Delta^{14}\text{C}$ ) data for the past 50,000 years. IntCal04 is shown back to its limit of 26 kyr. Marine coral and sediment  $^{14}\text{C}$  data have been corrected with a constant reservoir age, and speleothem data have been corrected with a constant dead carbon fraction. The paleo- $\Delta^{14}\text{C}$  observations are plotted compared to a carbon cycle box model simulation representing fixed preindustrial boundary conditions and changing  $^{14}\text{C}$  production ("full carbon cycle"). Error bars show 1- $\sigma$   $\Delta^{14}\text{C}$  uncertainty.

$\Delta^{14}\text{C}$  response of  $\sim 200\%$ , encompassing most of the Glacial age data. Reducing flux to shallow sediment reservoirs is required to match the highest observed  $\Delta^{14}\text{C}$  values. According to the model, however, the prescribed change in surface-to-deep-ocean exchange would produce a doubling of the surface-to-deep-ocean  $\Delta^{14}\text{C}$  difference. Observations do provide some evidence of decreased Glacial  $\Delta^{14}\text{C}$  in the deep western and eastern North Atlantic, as well as deep eastern equatorial and southwest Pacific (for review see Hughen et al., 2006). However, such a large change in Glacial deep ocean  $\Delta^{14}\text{C}$  has not been observed in the western equatorial Pacific (e.g. Broecker et al., 2004). It is important to note that most of the paleo-ocean  $\Delta^{14}\text{C}$  reconstructions correspond to the period around the Last

Glacial Maximum ( $\sim 21$  kyr BP), an interval when the simulated  $\Delta^{14}\text{C}$  response to production rate changes alone is close to the observations (especially if reasonable production rate uncertainties are considered). Another serious issue is that reconstructed rates of  $\Delta^{14}\text{C}$  change at the beginning of the last deglaciation,  $\sim 17$  kyr, are too large to be explained by changes in production rate alone and require a substantial dilution of  $^{14}\text{C}$  atoms in the atmosphere by a more depleted reservoir. Reconstructions of transient deglacial  $\Delta^{14}\text{C}$  changes in the intermediate depth western and deep eastern North Atlantic are consistent with a major reorganization of deep ocean circulation at that time, probably involving increased ventilation of a previously isolated deep water mass of southern or Pacific origin (e.g. Ad-

kins et al., 2002). These model simulations can place constraints on the magnitude of deep ocean  $\Delta^{14}\text{C}$  anomalies required to explain the surface marine record. In addition, the model data make quantitative predictions of the increase in surface marine reservoir age during the Glacial period. Unfortunately, however, observations of Glacial reservoir variability from low-latitude sites are rare. More sophisticated model simulations with increased spatial resolution would help identify the patterns of increase in reservoir age according to latitude and ocean basin. Regardless, it is clear that high-quality observations are needed from each of the three principal carbon reservoirs— atmosphere, surface and deep ocean—in order to constrain changes in both deep ocean  $\Delta^{14}\text{C}$  and surface marine reservoir age, and to understand the history of radiocarbon and global carbon cycle changes.

## References

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For full references please consult:  
[www.pages-igbp.org/products/newsletters/ref2006\\_3.html](http://www.pages-igbp.org/products/newsletters/ref2006_3.html)



## Assuring measurement quality: The international $^{14}\text{C}$ laboratory inter-comparison program

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### Introduction

In order to achieve reliable, precise and accurate  $^{14}\text{C}$  age measurements, laboratories routinely undertake both formal and informal quality assurance programs. Such programs may involve the repeated and routine measurement of an internal standard (such as a bulk cellulose sample), the results of which enable the laboratory to evaluate their reliability and precision. They may also routinely have access to known-age material against which to assess their accuracy. Beyond this, however, many laboratories regularly participate in in-

ter-laboratory comparisons to provide independent checks on laboratory performance.

### Reference material for $^{14}\text{C}$ calibration

High-quality  $^{14}\text{C}$  measurements also require traceability to international standards whose  $^{14}\text{C}$ -activities are known exactly by independent means, and also to reference materials whose activities are estimated and typically accompanied by associated uncertainty statements. Within the  $^{14}\text{C}$  community, there

has been an increasing realization of the need for adequate reference materials and a resultant development of both internal and external quality assurance (QA) procedures. Routinely,  $^{14}\text{C}$  laboratories make use of a number of standards and reference materials whose activities are known or are estimated from large numbers of measurements made by many laboratories (e.g. NIST OxI, OxII, IAEA C1-C8). More recent  $^{14}\text{C}$  inter-comparisons have also created a further series of natural reference materials (Scott, 2003, Scott et al, in prep).