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AMS RADIOCARBON DATING PROBLEMS BETWEEN 10 AND 8 KA BP IN LACUSTRINE DEPOSITS FROM LAKE GUN NUR, NORTHERN MONGOLIA

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ABSTRACT. Accelerator mass spectrometry (AMS) radiocarbon dating a continuous core from Lake Gun Nur, northern Mongolia, shows a period between 10 and 8 ka BP that could not be dated accurately. Further dating on alkali-insoluble residue and humic acid from the same samples in the Gun Nur core suggest that this AMS ^{14}C date anomaly is neither analytical nor material related. We hypothesize that the ^{14}C anomaly may be derived from increasing production rates of ^{14}C caused by diminished solar activity, a low $^{14}CO_2/^{14}CO$ ratio in the atmosphere, or an unstable ^{14}C flux in the lower atmosphere caused by changing geomagnetic field strength. Our results imply that the ^{14}C data used for ^{14}C age calibration cannot correct the age-depth regression between 8 and 10 ka BP to fit the age-depth model along with other time intervals.

INTRODUCTION

Radiocarbon dating of lacustrine sediments formed during the early Holocene is problematic. For example, ^{14}C dates in core SO89-17 from Lake Soppensee, central Switzerland, are distributed unevenly and changed dramatically between 7000 and 9000 ^{14}C yr BP, especially between 8200 and 9000 ^{14}C yr BP (Hajdas et al. 1993). In Lake Suigetsu, Japan (Kitagawa and van der Plicht 1998), the largest age reversals appeared between 1040~1354 cm in depth, corresponding to 8000~10,000 cal yr BP. A similar problem occurs in the ^{14}C dates in core IK97-11P from Lake Issyk-Kul, Kyrgyzstan, from 337–340 cm core depth (^{14}C dated at 8940 ± 65; 8670 ± 60 yr BP), and IK97-10P, depth 339–342 cm (^{14}C dated at 8310 ± 45; 8410 ± 45 yr BP), before being corrected, are unreasonable, though there is no indication of a problem from the isotope or trace element results (Ricketts et al. 2001). In the Huguang Maar Lake in southern China, leaf samples Kia8832 and Kia8833 were dated at 7670 ± 100 and 7880 +230/–220 yr ^{14}C BP, which are considered to be too young (Mingram et al. 2004). This dating problem may also be present in marine deposits (Bard et al. 1989), because the ^{14}C ages of deep-water and surface-water reservoirs, which are currently assumed to be ~400 yr in the tropics and ~1200 yr in Antarctica, might vary through time (Bard et al. 1994) and from place to place (Sikes et al. 2000). In a similar manner, we also observed the ^{14}C age uncertainty between 10 and 8 ka BP when we investigated Lake Gun Nur, northern Mongolia (50°15′N; 106°37′E), for a paleoclimate change study.

METHODS

A 748-cm-long continuous core from Lake Gun Nur in northern Mongolia was retrieved using piston-coring equipment. Two parallel cores were taken at the center of the lake at 4.6 m water depth. The cores were cut in half and sediments found to be largely undisturbed. After all segments were described carefully, they were combined into 1 complete core section based on their shared depositional characteristics, such as sedimentary structures, color, fossils, etc. Except in the upper 6 cm, the cores were sampled at 1-cm intervals for geochemical analyses and at 2-cm intervals for pollen and microfossil analyses. ^{14}C dating by accelerator mass spectrometry (AMS) yielded a high-resolution chronology. In total, 45 samples were dated by 3 AMS laboratories: the Leibniz Laboratory for Radiometric Dating and Stable Isotope Research (Kiel, Germany); Beta Analytic Inc. (Miami, USA); and the University of Arizona AMS facility (Tucson, USA).

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Ten samples were sent to Kiel in which ages for both the alkali residue (AR) and humic acid (HA) fractions were obtained. All were sampled at 1-cm intervals. X-ray and SEM analyses were conducted in the Institute of Physical Geography, Free University of Berlin, Germany.

RESULTS

Dating Results

All dates were reproduced (at 2 σ) by the 3 laboratories, except the ages between the depths of 670 and 460 cm (Figure 1a; Table 1–3). In this interval of the core, the dates differ from each other, changing the general age-depth regression trend, and forming what we informally term the Age Uncertainty Plateau, or AUP. Only 6 of 17 dates fall along the expected regression line predicted by uniform sedimentation rates (Figure 1b; Table 1–3). All dates not from the AUP interval are considered reliable because the measurements from 3 different laboratories agree well.

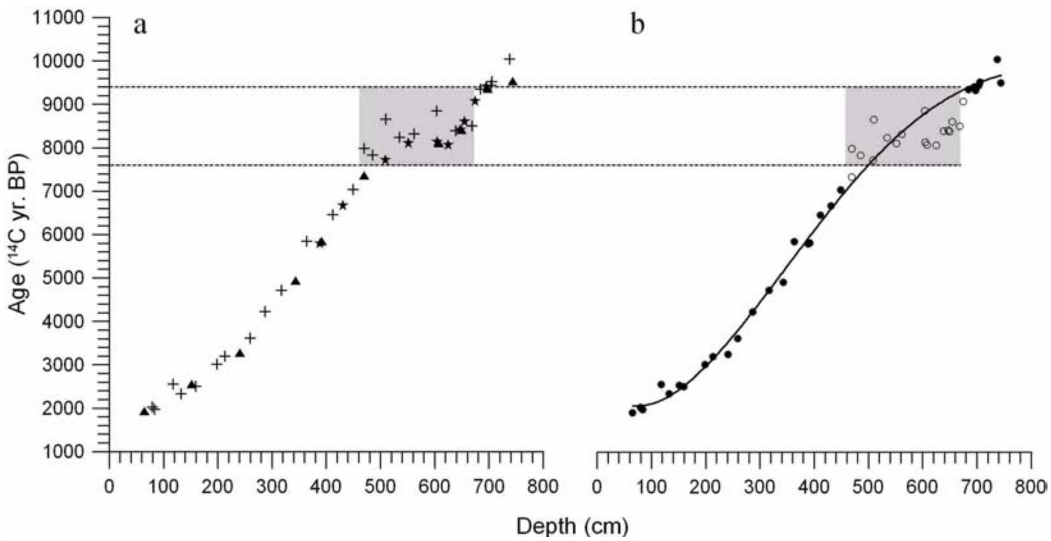

Figure 1 ^{14}C age-depth plot of the Lake Gun Nur core: a) triangles (Beta Analytic); crosses (Arizona); stars (Kiel). b) Age-depth and regression line excluding dates between 467 and 670 cm (circles). The shaded area marks the 210-cm-thick zone of anomalous ^{14}C dates.

Core Stratigraphy and Geochemistry

A detailed examination of the physical, mineralogical, and geochemical features of the Gun Nur core suggests that there is no hiatus nor unconformity, such as features from earthquakes, bioturbation, or erosion (Figure 2), especially between the depths of 460 and 670 cm (Wang et al. 2004). Because abundant benthic diatoms were observed between 450 and 720 cm (Feng et al. 2005) and carbonate-rich laminated layers were observed between 570 and 720 cm (Figure 3), which are composed of almost pure aragonite crystals, the depositional environment was deemed stable. All these observations, combined with the even, flat lake bottom and relatively uniform sedimentation rates, rule out the possibility of sudden or abnormal depositional and geochemical processes. To test the possible postdepostional translocation of humates through lake sediment, 10 paired ages of both humic acid (HA) and alkali-insoluble residue (AR) fractions of organic matter were dated. The sediments are rich in organic matter (13–21.7%), and thus are believed to overwhelm the presence of

Table 1 AMS ^{14}C dates of organic matter by Arizona AMS facility (2 σ).[a]

Lab nr	Depth (cm)	Depth-C (cm)	^{14}C age BP	δ^{13}C (‰)	cal ^{14}C age BP	Midpoint cal BP	Material
AA51939	79–80	79.5	2030 ± 37	–15.8	1890–2120	2005	Org.
AA51940	83–84	83.5	1975 ± 41	–17	1820–2010	1915	Org.
AA51941	117–118	117.5	2560 ± 37	–17.4	2490–2760	2625	Org.
AA51942	132–133	132.5	2338 ± 37	–20.9	2300–2500	2400	Org.
AA51943	159–160	159.5	2500 ± 140	–16.1	2300–2950	2625	Org.
AA51944	198–199	198.5	3016 ± 39	–21.6	3070–3350	3210	Org.
AA51945	212–214	213	3201 ± 38	–16.2	3340–3480	3410	Org.
AA51946	258–260	259	3617 ± 40	–19.9	3820–4000	3910	Org.
AA51947	286–288	287	4226 ± 42	–17.4	4610–4870	4740	Org.
AA51948	316–318	317	4721 ± 44	–14.8	5320–5590	5455	Org.
AA51949	363–364	363.5	5843 ± 47	–9.4	6350–6760	6555	Org.
AA51950	411–412	411.5	6458 ± 47	–15.1	7270–7440	7355	Org.
AA51951	448–450	449	7040 ± 61	–10.3	7720–7970	7845	Org.
AA51952	469–470	469.5	7991 ± 69	–8	8630–9030	8830	Org.
AA51953	485–486	485.5	7836 ± 63	–5.2	8400–9000	8700	Org.
AA51954	509–510.3	509.7	8660 ± 66	–11.5	9520–9900	9710	Org.
AA51955	534–535	534.5	8242 ± 63	–5.6	9020–9430	9225	Org.
AA51956	561–562	561.5	8324 ± 63	–5.6	9130–9490	9310	Org.
AA51957	603–604	603.5	8858 ± 83	–11	9600–10,250	9925	Org.
AA51958	638–639	638.5	8397 ± 62	–16.4	9270–9530	9400	Org.
AA51959	668–669	668.5	8500 ± 60	–15	9400–9560	9480	Org.
AA51960	684–685	684.5	9356 ± 64	–21	10,360–10,750	10,555	Org.
AA51961	703–704	703.5	9439 ± 88	–14.7	10,400–11,100	10,750	Org.
AA51962	705–706	705.5	9528 ± 58	–28.9	10,500–11,150	10,825	Org.
AA51963	737–738	737.5	10,047 ± 70	–27.6	11,200–12,100	11,650	Org.

[a]Note: Depth-C refers to the center depth of sample.

Table 2 AMS ^{14}C dates of organic matter by Beta Analytic.[a]

Lab nr	Depth (cm)	Depth-C (cm)	^{14}C age BP	δ^{13}C (‰)	cal ^{14}C age BP	Midpoint cal BP	Matter
Beta-171822	64–65	64.5	1900 ± 40	–23.1	1710–1930	1820	Org.
Beta-171823	151–152	151.5	2530 ± 40	–24.1	2470–2750	2610	Org.
Beta-171824	240–242	241	3250 ± 40	–21	3380–3570	3475	Org.
Beta-171825	342–344	343	4910 ± 40	–22.7	5590–5730	5660	Org.
Beta-171826	391–392	391.5	5820 ± 50	–20.9	6490–6750	6620	Org.
Beta-198046	469–470	469.5	7330 ± 50	–21.2	8010–8200	8105	Org.
Beta-198047	607–608	607.5	8080 ± 50	–23.5	8800–8820	8810	Org.
Beta-198048	649–650	649.5	8380 ± 50	–25.9	9500–9280	9390	Org.
Beta-198049	697–698	697.5	9330 ± 50	–24.5	10,670–10,400	10,535	Org.
Beta-171827	743–744	743.5	9500 ± 50	–26.8	10,500–11,100	10,800	Wood

[a]Note: Depth-C refers to the center depth of sample.

old residue carbon in sediment. Because organic carbon contents in the Gun Nur core are extremely high (average content >18%) and δ^{13}C values are low (< –25‰), suggesting terrestrial plants dominate the lake organic matters, the hardwater reservoir effect on our ^{14}C dating could be minimal.

Figure 2 A detailed picture of the core stratigraphy, left column: fresh cores viewed from the top (A) and from the bottom (B); right column, the core from ~4.2 m to the bottom (T-point marks the reverse point of AMS ^{14}C ages; see also Figures 3 and 4).

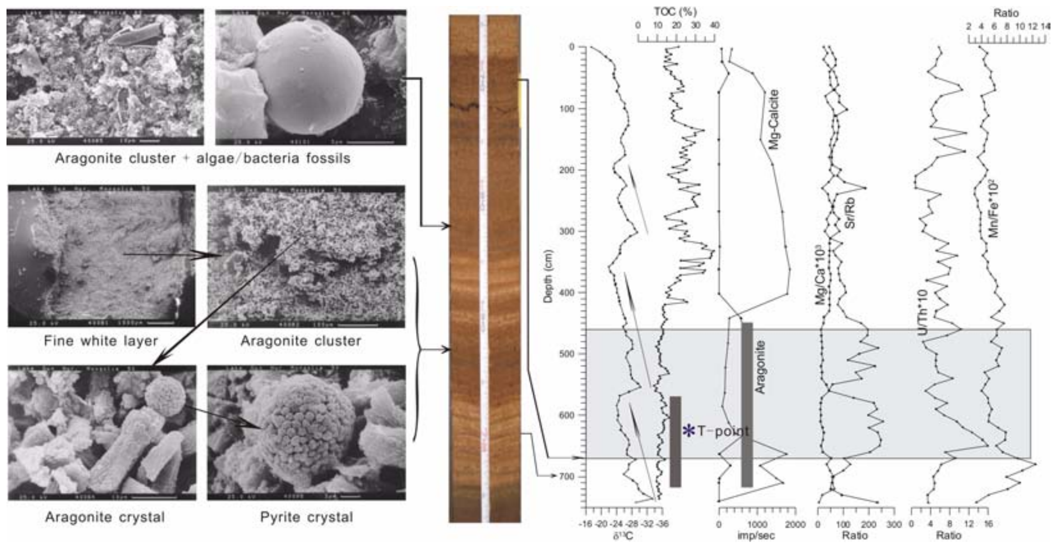

Figure 3 Left: photographs of some mineralogical and biological features of the core. Middle: well-stratified layers from the lower part of the core. Right: geochemical analyses vertical of the entire core. The shaded area denotes the 210-cm-thick portion of the core with anomalous ^{14}C dates. The longer bar denotes a zone of dominantly benthic foraminiferous layers, and the shorter vertical bar is a zone of well-stratified layers.

The dates of the humic acid (HA) fraction in all cases are older than that of the alkali-insoluble residue (AR; Table 3, Figure 4). Theoretically, the organic matter in lacustrine deposits, which are different from the soil matter, in both soluble and insoluble fractions, were formed at the same time and thus should give the same dates. Different ages given by HA and AR fractions of the same sample imply either the AR fraction was derived from the old organic matter or the HA fraction was mobile in the sediments. Therefore, the age differences between HA and AR could be used as a criterion to evaluate the reliability of AMS ^{14}C dates (Zhang et al. 2008). In the Gun Nur core, the older dates of the HA fraction indicate upward movement of the soluble organic matter, perhaps due to soft-sediment consolidation. The upward movement of HA could be absorbed by sediments, and HA could also be fused or "condensed" with the AR organic compounds through humification processes.

Table 3 AMS ^{14}C dates of humic acid (HA) and alkali residue (AR) fractions by Kiel.[a]

Lab nr	Depth (cm)	Depth-C (cm)	Age yr BP (AR)	$\delta^{13}C$ (‰)	cal age yr BP (AR)	Age yr BP (HA)	$\delta^{13}C$ (‰)	Matter
KIA25401	388–389	388.5	5795 ± 30	–17	6583	5815 ± 30/35	–17.82	Org.
KIA25402	430–431	430.5	6670 ± 30	–20.81	7533	6790 ± 45	–21.42	Org.
KIA25403	508–509	508.5	7725 ± 35	–22.82	8480	8050 ± 40	–24.01	Org.
KIA25404	551–552	551.5	8105 ± 35	–22.92	9044	8425 ± 35	–23.72	Org.
KIA23783	604–605	604.5	8135 ± 45	–23.93	9073	8340 ± 45	–24.03	Org.
KIA23782	624–625	624.5	8065 ± 40	–25.14	9033	8155 ± 40	–23.51	Org.
KIA23781	646–647	646.5	8415 ± 45	–25.32	9465	8655 ± 45	–24.59	Org.
KIA23780	654–655	654.5	8610 ± 45	–24.3	9613	8800 ± 45	–25.86	Org.
KIA23779	674–675	674.5	9075 ± 45	–26.08	10,224	9200 ± 45	–26.21	Org.
KIA23778	694–695	694.5	9409 ± 46	–21.13	10,624	9505 ± 60	–25.05	Org.

[a]Note: Depth-C refers to the depth of the sample center.

DISCUSSION

We attributed the anomalous ^{14}C dates between 460 and 670 cm (Figure 4a) to changes in ^{14}C production rates. Thus, we predict the same reason for similar discrepancies in other high-resolution lacustrine records discussed in the Introduction.

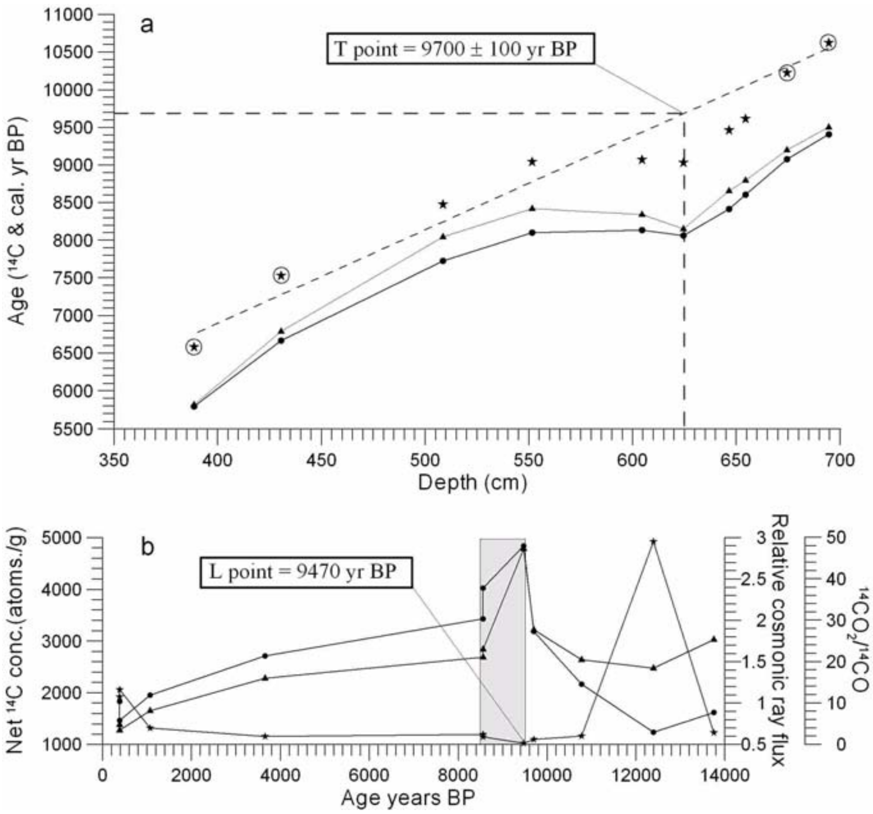

Figure 4 a) AMS ^{14}C ages between 380 and 700 cm in the core. Dotted line: age of alkali residue (AR); Triangle line: age of humic acid fraction (HA); Stars: calibrated ages given by AR fraction. The age at T-point was calculated according to the linear regression (dashed line) based on the 4 circled dates. Both ^{14}C and cal yr BP are given in the vertical axis. b) Net ^{14}C concentration (triangle line), relative cosmic-ray flux (dotted line), and $^{14}CO_2/^{14}CO$ ratio (star line) during the last 14,000 yr recorded in an ice core at Summit, Greenland. The L-point marks the lowest value of $^{14}CO_2/^{14}CO$ ratio (data after Lal et al. 2005).

The concentration of *in situ* cosmogenic ^{14}C in ice crystals at Summit in Greenland shows that on a century timescale, the cosmic-ray-induced production rate of ^{14}C was higher by about a factor of 2 during 9500–8500 yr BP (Figure 4b), attributed to the low solar activity resulting in variable modulation of terrestrial cosmic-ray flux (Lal et al. 2005). An age of 9470 yr BP for the L-point in Figure 4b is comparable to that of the T-point in Figure 4a, 9700 ± 100 yr BP. Though the data density is low, one can deduce that the high net ^{14}C concentration and relative cosmic-ray flux between 8500 and 9500 yr BP should result in a high ^{14}C value in the atmosphere over the Northern Hemisphere. However, the atmospheric ^{14}C in the ice record includes both $^{14}CO_2$ and ^{14}CO, whereas tree-ring-deduced ^{14}C is only associated with $^{14}CO_2$ because ^{14}CO cannot be taken up during plant photosynthesis. These processes can be described by the following equations:

radioactive carbon formation in the atmosphere:

$$^{14}N + n \rightarrow {}^{14}C + p \rightarrow {}^{14}CO + {}^{14}CO_2;$$

building of the carbon isotopes in plant tissues through photosynthesis:

$$6CO_2\ ({}^{14}CO_2 + {}^{13}CO_2 + {}^{12}CO_2) + 11H_2O \rightarrow {}^{(14 + 13 + 12)}C_6H_{22}O_{11} + 6O_2$$

Theoretically, the ^{14}C atom produced in the atmosphere is quickly oxidized, as the partitioning between mono- and dioxide is 9 to 1, meaning that 90% of atoms end up as ^{14}CO (Geyh and Schleicher 1990). The lifetime of carbon monoxide in the atmosphere is about 2 months, and with oxidation by a short-lived hydroxyl radical ^{14}C, eventually results in $^{14}CO_2$. Therefore, the majority of ^{14}C atoms in the atmosphere are present in the form of dioxide. Any production variations of ^{14}C would therefore be quickly reflected in atmospheric $^{14}CO_2$ concentration. This procedure, however, may be interrupted by slow atmospheric mixing and/or abrupt changes of the Earth's geomagnetic field during the period between 8 and 10 kyr BP (Figure 5).

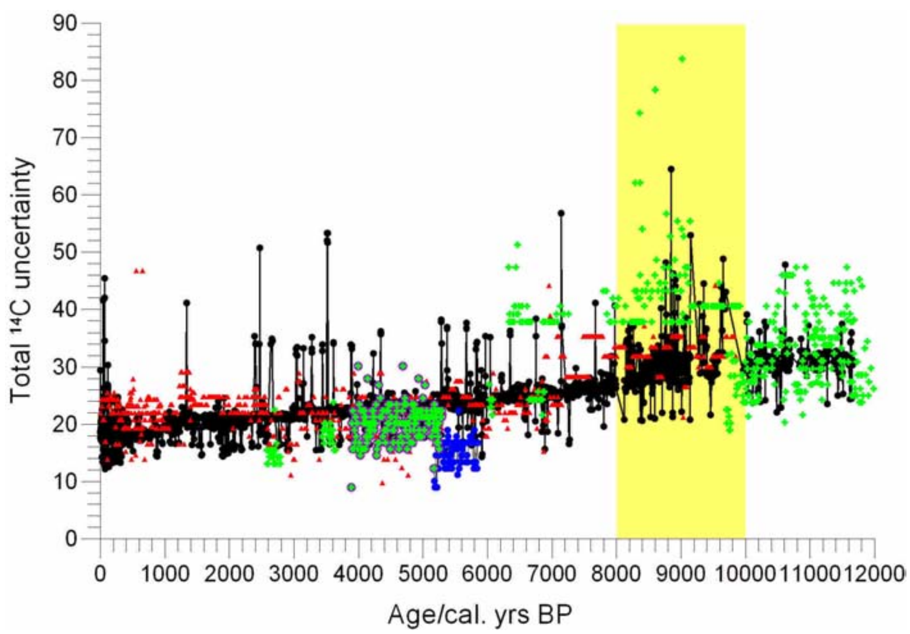

Figure 5 The total ^{14}C measurement uncertainty during last 11 kyr, in which the largest uncertainties appear between 8 and 10 kyr BP. Dark doted line: data of University of Washington (QL) tree rings; Red triangle line: data of Queen's University Belfast (UB) tree rings; Blue dotted line: data of University of Groningen (GrN) tree rings; Green crosses: data of Heidelberger Akademie der Wissenschaften (Hd) tree rings; Pink dots: data of CSIR, Pretoria (Pta) tree rings (data from Reimer et al. 2004; http://www.radiocarbon.org/IntCal04.htm). For interpretation of the references to color, the reader is referred to the online version of this article.

Long-term trends in ^{14}C concentration depend on the geomagnetic dipole moment (Stuiver et al. 1991; Stuiver and Braziunas 1993) that controls the intensity of cosmic rays reaching Earth's atmosphere and hence the ^{14}C production rate in the atmosphere. Although a geomagnetic field change does not affect the cosmic-ray flux at polar latitudes (Lal et al. 2005), the geomagnetic dipole moment changes over time (Yang et al. 2000), which modulates the cosmic-ray flux (Solanki et al. 2004). Study results show that the horizontal component of the magnetic force is greatest at low latitudes and minimal at high latitudes (Bard 1998). This leads to a weak effect on ^{14}C concentration at high latitudes and a strong one at low latitudes, which should have resulted in different impacts on the ^{14}C distribution in the atmosphere (Figure 6). If the $^{14}CO_2/^{14}CO$ ratio shift in Greenland, an indicator of cosmic-ray flux between 10 and 8 kyr BP, does relate to the AUP at middle latitudes in the Northern Hemisphere, the tree-ring-deduced ^{14}C for the ^{14}C age calibration does not appear to correct the younger ^{14}C dates of the AUP interval to fit the age-depth regression line in the record of Lake Gun Nur, northern Mongolia.

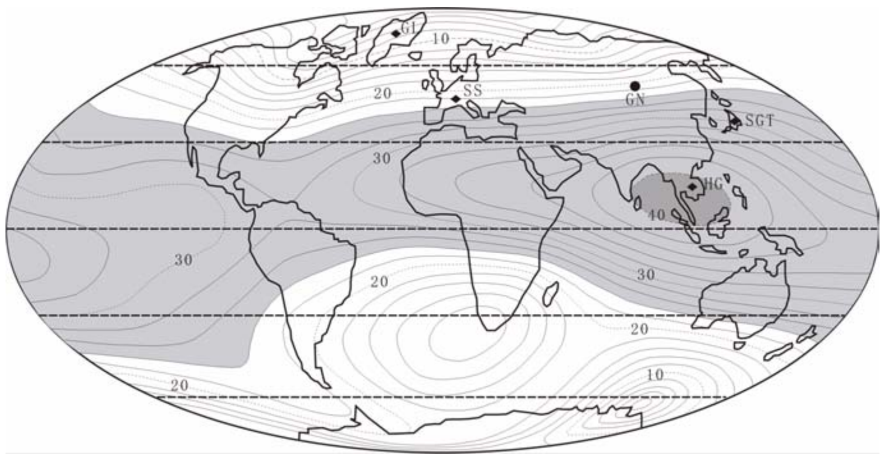

Figure 6 International geomagnetic reference field for the horizontal field component H expressed in 10^3 gamma units (modified from Bard 1998). The figure shows that the horizontal component of the magnetic force is at a maximum in low latitudes (shaded zone) and at a minimum in high latitudes. This results in a weak effect on ^{14}C in high latitudes and a strong one in the low latitudes. Several sites mentioned in the text are marked in the figure (GL: Summit in Greenland; SS: Lake Soppensee in central Switzerland; GN: Lake Gun Nur in Mongolia; SGT: Lake Suigetsu in Japan; HG: Huguang Maar Lake in south China).

Certainly, the above hypothesis is just one explanation of the dating problems we observe. More work on both geological archives in different areas and on geochemical processes in the atmosphere needs to be undertaken to test our proposal, as recent studies show that nature may behave differently than previously thought (Zhang et al. 2006a,b; Engel et al. 2009).

ACKNOWLEDGMENTS

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