

## CALIBRATION OF RADIOCARBON DATES FOR THE LATE PLEISTOCENE USING U/Th DATES ON STALAGMITES

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**ABSTRACT.** Twenty paired  $^{14}\text{C}$  and U/Th dates covering most of the past 50,000 yr have been obtained on a stalagmite from the Congo Caves in South Africa as well as some additional age-pairs on two stalagmites from Tasmania that partially fill a gap between 7 ka and 17 ka ago. After allowance is made for the initial apparent  $^{14}\text{C}$  ages, the age-pairs between 7 ka and 20 ka show satisfactory agreement with the coral data of Bard *et al.* (1990, 1993). The results for the Congo stalagmite between 25 ka and 50 ka show the  $^{14}\text{C}$  dates to be substantially younger than the U/Th dates except at 49 ka and 29 ka, where near correspondence occurs. The discrepancies may be explained by variations in  $^{14}\text{C}$  production caused by changes in the magnetic dipole field of the Earth. A tentative calibration curve for this period is offered.

### INTRODUCTION

At the International Radiocarbon Conference in Bern in 1978, it was pointed out that certain  $^{14}\text{C}$  dates for the Late Pleistocene appear to be several thousand years too young when compared with corresponding uranium series dates (Vogel 1980; Barbetti 1980). This prompted us to undertake a more detailed comparison of the two dating techniques using a slender 2.8 m stalagmite from the Congo Caves in the Cape Province of South Africa. This specimen, which stood at the far end of Cave II, was sawed into sections and removed during several visits over a period of four years. It proved to be ideal for the purpose since it had a coarse crystalline nonporous structure that ensured closed-system conditions. Its base turned out to date back nearly 50,000 yr; water was dripping onto the tip so that it was still growing at the time of removal. Preliminary results for the comparison were reported at the next Radiocarbon Conference in Seattle in 1982 (Vogel 1983).

Unfortunately there is a gap in the sequence, because the stalagmite's growth halted between *ca.* 17,000 and 7000 yr ago. This gap could partially be filled by samples of two similarly crystalline stalagmites from Lynd's Cave in Tasmania (Vogel 1987; Goede and Vogel 1991).

### METHOD

We cut samples along the growth axis of the three stalagmites and performed the  $^{14}\text{C}$  and U/Th analyses using the same pieces in each case. Carbon dioxide for measurement of the  $^{14}\text{C}$  in a  $\text{CO}_2$  proportional counter was generated by adding acid, and the remaining solution was processed for the U/Th isotope analysis, using standard  $\alpha$ -counting techniques.

### RESULTS

The results for the Congo stalagmite are given in Table 1, and those for Lynd's Cave are listed in Table 2. Two adjustments need to be made to the dates.

1. The  $^{14}\text{C}$  ages must be corrected for the apparent initial age of the calcite when it is precipitated from the dripping groundwater.

As previously shown (Vogel 1983), the first six  $^{14}\text{C}$  dates for the upper 95 cm of the Congo stalagmite show complete concordance with the U/Th dates if 1450 yr are subtracted from the initial age and the results are calibrated with the tree-ring calibration curve (e.g., Stuiver and Pearson 1993). This correction of 1450 yr is thus adapted for the older sections, too. It is, of course, possible that the apparent initial age varied with differing climatic conditions, but the shift would not have been more than a few hundred years, since this figure has been found to apply to groundwater in limestone areas worldwide. The adjustment for the Lynd's Cave stalagmites is less certain. 1500 yr are subtracted from these  $^{14}\text{C}$  dates, but an error of  $\pm 500$  should be assumed. When these results were first presented (Vogel 1987) 2160 yr was subtracted, based on a comparison with parallel electron spin resonance (ESR) dates, but subsequent tree-ring calibration data for the early Holocene (Kromer and Becker 1993) have shown this correction to be too large.

- The U/Th ages need to be corrected for any initial  $^{230}\text{Th}$  that may have been incorporated into the stalagmite during formation.

Most of the samples had very small amounts of  $^{232}\text{Th}$ . For those that did show measurable amounts of the isotope, the adjustment was  $< 230$  yr, assuming an initial  $^{230}\text{Th}/^{232}\text{Th}$  activity ratio of  $1 \pm 0.5$ . An exception was the Lynd's Cave stalagmite L1, which had a very low uranium content (ca. 0.02 ppm) and consequently required a correction of 620 yr in one case. The U/Th results from this specimen are therefore less precise than the rest.

TABLE 1.  $^{14}\text{C}$  and U/Th data for Congo stalagmite V3. Column 2 gives the distance of the sample from the tip of the stalagmite; column 4 the apparent  $^{14}\text{C}$  age adjusted for isotope fractionation using the  $^{13}\text{C}/^{12}\text{C}$  ratio. Columns 7, 8 and 9 list the activity ratios of the U and Th isotopes and column 10 the U-series ages adjusted for initial Th.

Sample	Distance (mm)	Lab code (Pta-)	Apparent $^{14}\text{C}$ age (yr BP)	Anal. no.	U cons (ppm)	$\frac{^{234}\text{U}}{^{238}\text{U}}$	$\frac{^{230}\text{Th}}{^{232}\text{Th}}$	$\frac{^{230}\text{Th}}{^{234}\text{U}}$	U/Th age (yr BP)
V3/1a	5	2256	1530 $\pm$ 50	U-003	0.068	3.70	--	--	recent
V3/1b	14	2435	1490 $\pm$ 55	--	--	--	--	--	
V3/1c	314	2579	3270 $\pm$ 45	U-107	0.062	3.43	10	0.019	1870 $\pm$ 220
V3/1d	492	2289	4660 $\pm$ 60						
V3/2a	715	2580	5730 $\pm$ 50	U-108	0.098	3.61	28	0.044	4710 $\pm$ 350
V3/2b	887	2581	6380 $\pm$ 60	U-109	0.109	3.59	101	0.057	6240 $\pm$ 400
V3/2c	974	2436	15,400 $\pm$ 140	U-112	0.091	3.14	217	0.152	17,400 $\pm$ 800
V3/6a	989	3081	15,790 $\pm$ 50	U-263	0.109	3.15	>>	0.166	19,300 $\pm$ 800
V3/6b	1157	3087	19,180 $\pm$ 50						
V3/6c	1304	3090	21,610 $\pm$ 70	U-242	0.140	2.65	>>	0.227	27,200 $\pm$ 3000
V3/3m	1517	3084	26,710 $\pm$ 110	U-265	0.078	3.18	103	0.227	27,200 $\pm$ 1300
V3/3b	1598	3277	28,380 $\pm$ 150	U-282	0.101	3.10	256	0.264	32,100 $\pm$ 2100
V3/4a	1679	2657	30,300 $\pm$ 610	U-118	0.106	3.02	>>	0.242	29,200 $\pm$ 800
V3/4c	1759	3078	30,130 $\pm$ 180	U-264	0.083	3.22	>>	0.243	29,300 $\pm$ 3000
V3/4d	1842	3077	30,170 $\pm$ 180	U-266	0.073	2.99	153	0.282	34,400 $\pm$ 2200
V3/4b	2030	2658	30,110 $\pm$ 500	U-119	0.083	3.18	173	0.287	35,200 $\pm$ 1000
V3/5a	2101	3279	32,670 $\pm$ 330	U-269	0.077	3.03	>>	0.294	36,400 $\pm$ 1800
V3/5c	2181	3893	33,870 $\pm$ 330	U-325	0.087	3.05	155	0.305	37,700 $\pm$ 1200
V3/5b	2288	2689	35,900 $\pm$ 280	U-136	0.058	3.11	255	0.311	38,700 $\pm$ 900
V3/7a	2409	3278	36,520 $\pm$ 300	U-270	0.109	2.84	254	0.329	41,300 $\pm$ 1200
V3/7d	2491	3887	37,580 $\pm$ 490	U-324	0.117	2.85	200	0.348	44,100 $\pm$ 1400
V3/7c	2589	3884	41,330 $\pm$ 630	U-323	0.107	2.90	200	0.352	44,700 $\pm$ 1400
V3/7b	2698	3280	47,660 $\pm$ 1120	U-283	0.119	2.73	420	0.381	49,200 $\pm$ 1300

TABLE 2.  $^{14}\text{C}$  and U/Th data for the Tasmanian stalagmites. (See Table 1 for explanation.)

Sample	Distance (mm)	Lab code (Pta-)	Apparent $^{14}\text{C}$ age (yr BP)	Anal. no.	U cons (ppm)	$\frac{^{234}\text{U}}{^{238}\text{U}}$	$\frac{^{230}\text{Th}}{^{232}\text{Th}}$	$\frac{^{230}\text{Th}}{^{234}\text{U}}$	U/Th age (yr BP)
<i>Lynd's Cave stalagmite L2</i>									
L2/1	717	3792	10,980 $\pm$ 110	U-305	0.307	3.12	171	0.104	11,740 $\pm$ 500
L2/2	515	3791	11,800 $\pm$ 100						
L2/4	334	3707	12,770 $\pm$ 100	U-293	0.262	2.97	236	0.117	13,300 $\pm$ 360
L2/3	120	3708	13,450 $\pm$ 130	U-294	0.305	2.85	442	0.128	14,570 $\pm$ 420
<i>Lynd's Cave stalagmite L1</i>									
L1/3	1380	3762	8320 $\pm$ 90	U-301	0.019	2.32	12	0.069	7070 $\pm$ 570
L1/6	980	3731	10,970 $\pm$ 100	U-299	0.016	2.69	64	0.099	11,080 $\pm$ 800
L1/7	614	3797	12,400 $\pm$ 110						
L1/9	79	3713	14,170 $\pm$ 60	U-295	0.024	2.80	44	0.114	12,840 $\pm$ 750

## DISCUSSION

With these two adjustments to the ages, the results for the Cango stalagmite are plotted (Fig. 1.) against the distance from the tip. A few of the U/Th analyses had relatively large margins of error, but on the whole they show a nearly linear growth of the stalagmite between 44 ka and 18 ka ago, with a somewhat slower growth rate between 49 ka and 44 ka. The  $^{14}\text{C}$  ages, on the other hand, show major deviations from this linear growth curve, especially for the period beyond 30,000 yr BP, where they are consistently several thousand years younger than the uranium series dates.

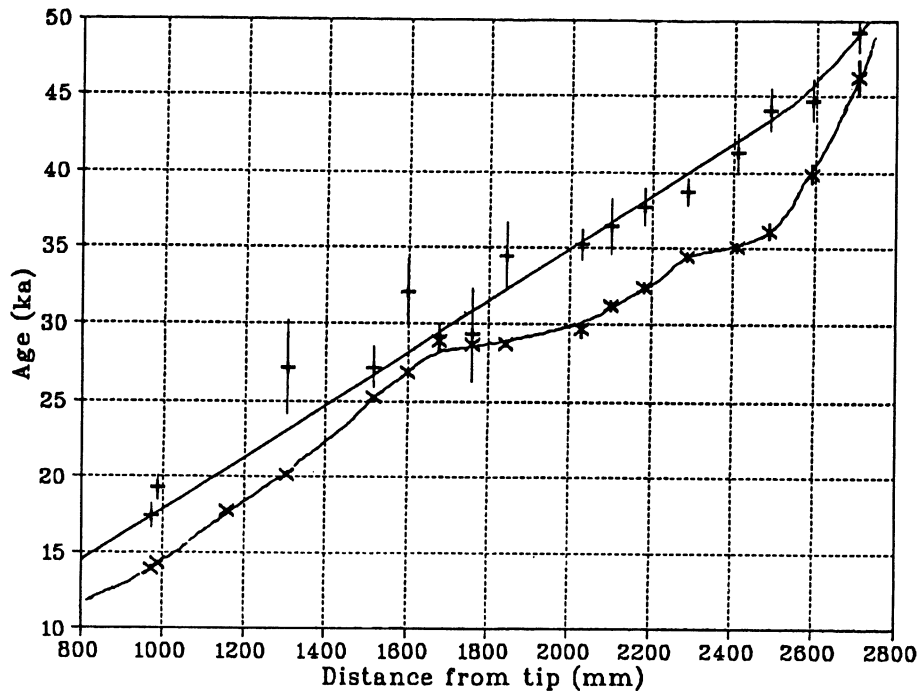


Fig. 1.  $^{14}\text{C}$  dates ( $\times$ ) adjusted for initial age of 1450 yr and parallel U-series dates (+) for the Cango stalagmite showing the large discrepancy between age-pairs between 30 ka and 45 ka

Now, it is well known that samples older than 30,000 yr are prone to produce  $^{14}\text{C}$  ages that are too young due to contamination by small amounts of more recent carbon. The fact that the  $^{14}\text{C}$  date for the bottommost sample agrees with the U/Th age to within  $1\sigma$  if the correct  $^{14}\text{C}$  half-life (5730 yr) is used ( $49,200 \pm 1300$  (U/Th) -  $47,550 \pm 1120$  ( $^{14}\text{C}$ ) =  $1650 \pm 1720$ ), indicates, however, that contamination cannot in this case explain the discrepancies between the two methods. Of special significance is near identity between the  $^{14}\text{C}$  and U/Th ages *ca.* 29,000 yr ago. This will be discussed briefly below.

For the period between 10 and 20 ka we need to consider the age-pairs derived from the two Lynd's Cave stalagmites (Table 2). As previously noted (Vogel 1987), these data also show considerable discrepancies between the  $^{14}\text{C}$  ages and the U/Th ages.

More recently, a comprehensive set of age comparisons has been published using coral from Barbados (Bard *et al.* 1990, 1993), which confirm that  $^{14}\text{C}$  dates between 12 ka and 22 ka are several thousand years too young. The U/Th dates were produced by thermal ionization mass spectrometry (TIMS), which gives greater precision than the conventional  $\alpha$ -counting technique. In Figure 2 these results are presented together with our measurements in this time range; two pairs from Cango (V3) are also included to extend the age range. All except two of our data points correspond to the TIMS results to within one sigma, thus confirming that the  $^{14}\text{C}$  age discrepancy is a worldwide phenomenon and that the stalagmite data do represent the actual situation.

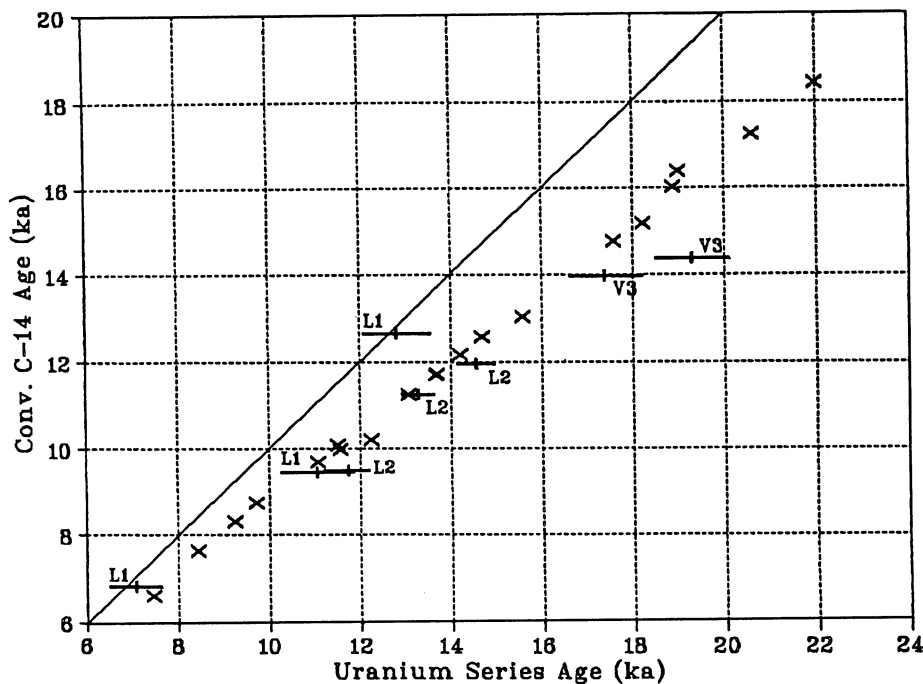


Fig. 2. Calibration curve for conventional  $^{14}\text{C}$  dates after Bard *et al.* (1993) (x) with our age-pairs for the Lynd's Cave stalagmites (L1 and L2) (Table 2) and the two Cango Cave (V3) age-pairs in this time range (Table 1)

Using the comparisons obtained from the Cango Cave stalagmite, a calibration curve for the period 25 ka to 50 ka is plotted in Figure 3. The four samples with U/Th error margins greater than  $\pm 2000$  yr (V3/6c, V3/3b, V3/4c and V3/4d) are not included in the figure because they are not very meaningful. The margins of error of this curve are still relatively large, but for the present it can be used in cases where absolute date estimates are required rather than  $^{14}\text{C}$  ages. It is evident from this figure that  $^{14}\text{C}$  dates between 35 ka and 45 ka are *ca.* 5000 yr too young. Recently, one additional age comparison has been published (Bischoff *et al.* 1994) that confirms this discrepancy.

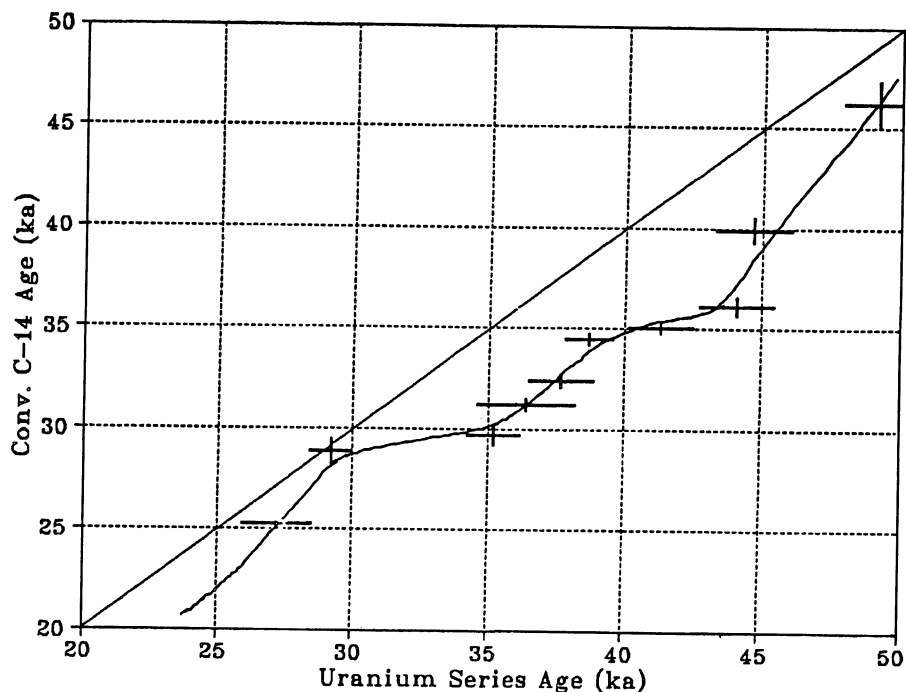


Fig. 3. Calibration curve for conventional  $^{14}\text{C}$  dates between 20 ka and 50 ka using the data for the Cango Cave stalagmite, V3 (Table 1)

The authors find that their  $^{14}\text{C}$  ages on charcoal ( $37 \pm 1$  ka) are much younger than U/Th ages on bracketing flowstone ( $42.6 \pm 1.1$  ka) in a cave near Barcelona, Spain. If the  $^{14}\text{C}$  date is calibrated, using Figure 3, it overlaps with the U/Th date.

Not shown in Figure 2 is the isolated coral sample of Bard *et al.* (1993) that gave an age-pair at variance with our results at *ca.* 30 ka (Figure 3). Their measurements show the  $^{14}\text{C}$  date to be  $4780 \pm 380$  ( $2\sigma$ ) yr younger than the U/Th date of  $30,225 \pm 160$  yr, whereas our age-pair at 29,000 yr shows almost no difference between the two techniques: at  $29,200 \pm 800$  yr (U/Th) the  $^{14}\text{C}$  age is only 350 yr younger. Other age-pairs in this time range, on the other hand, do seem to support our finding: the five comparisons listed by Barbetti (1980) between 26,000 yr and 32,000 yr individually and collectively show virtually no discrepancy between U/Th and  $^{14}\text{C}$  ages. The weighted averages for these six samples give:

$$29,690 \pm 680 \text{ yr (U/Th)} - 29,800 \pm 760 \text{ yr } (^{14}\text{C}) = -110 \pm 1020 \text{ yr} .$$

## CONCLUSION

If  $^{14}\text{C}$  dating gives the correct age at 29,000 yr, it implies that a drastic decrease in the  $^{14}\text{C}$  levels of the atmosphere occurred. This seems to be indicated by the very slow decrease in the  $^{14}\text{C}$  ages between the U/Th dates of 35,000 and 29,000 yr (Fig. 1). The effect could have been caused by a major increase in the strength of the Earth's dipole field, and there is some evidence that this did happen. Most estimates of the global dipole field intensity during the last 120,000 yr indicate considerably lower values than today, but at least one of them (Tric *et al.* 1992) shows near-normal dipole intensity between 55 and 47 ka, between 33 and 25 ka and after 12 ka, with distinctly lower values in between. The geomagnetic modulation of the  $^{14}\text{C}$  production rate could thus explain qualitatively, at least, the general trend of the data presented here.

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