

Radiocarbon dating of charcoal samples from Rakhigarhi, Haryana, India using accelerator mass spectrometer

Carbon dating is one of the most accurate methods to determine the age of any archaeological site. Here we present an analysis of seven samples from Rakhigarhi site, Haryana taken at different depths. We have used the recently acquired accelerator mass spectrometer (AMS) system at the Inter University Accelerator Centre (IUAC), New Delhi.

Rakhigarhi (29°17'19"N, 76°06'47"E) is a village in Hisar district, Haryana, India, situated in northwest about 150 km from Delhi. In 1963, archaeologists identified one of the largest Harappan sites at this place. It is situated on the bed of a now dried river¹. The site was excavated in 2015 by a group from Deccan College, Pune led by one of us (V.S.). A vertical trench was dug and various artefacts were recovered from the site. Several charcoal samples were also collected. These were analysed at IUAC, which has recently acquired a new radiocarbon dating facility. Here, we report the dates of the Rakhigarhi sites analysed at IUAC.

IUAC has recently developed a comprehensive AMS facility for ¹⁴C, ¹⁰Be and ²⁶Al radionuclide measurements. This facility is based on a 500 kV tandem ion accelerator procured from the National Electrostatic Corporation, USA. IUAC has also developed extensive facility and significant expertise in the graphitization of samples of interest for measuring them with the AMS system. The machine used for this purpose is at IUAC, but is similar in principle to the carbon dating AMS at the Institute of Physics, Bhubaneswar²⁻⁴.

For the present study, seven samples each weighing about 50 g, were provided by Deccan College which were excavated from a depth of 9.1, 15.7, 16.1, 17.3, 18.4, 19.3 and 20.6 m (Table 1). Pieces of charcoal were extracted from these samples. Typically 3 g of charcoal was taken for pre-treatment from each sample.

All the samples were first visually cleaned under stereo-zoom microscope to remove extraneous material to the extent possible. Then they were washed in deionized type-1 water (resistivity 18.2 megaohm cm). The samples were subjected to acid (HCl), base (NaOH) and acid (HCl) cycle. Then they were dipped in the solvent at 60°C for about 5 h each

and the residue was removed. In order to save the small charcoal particles, the samples were also subjected to centrifuge

(2000 rpm) for 2 min. They were then washed with the deionized water several times until the water became pH neutral,

Table 1. Details of the results obtained with charcoal samples (S1 to S8) from Rakhigarhi, Haryana

Depth (m)	No. of ¹⁴ C counts	¹⁴ C/ ¹² C × 10 ⁻¹³	Libby years (BP)	Age (years BC)		Calibrated age (BP)
				Maximum	Minimum	
9.1	16,372	5.7570	3824 ± 80	2351	2196	4273 ± 39
15.7	19,860	5.4885	4141 ± 75	2780	2628	4704 ± 38
16.1	14,787	5.7036	3900 ± 82	2481	2280	4380 ± 50
17.3	28,876	5.7017	3903 ± 224	2671	2119	4395 ± 138
18.4	11,505	5.5871	4067 ± 90	2697	2488	4592 ± 52
19.3	20,309	5.5923	4060 ± 75	2679	2480	4579 ± 50
20.6	22,870	5.5922	4060 ± 129	2762	2470	4616 ± 73

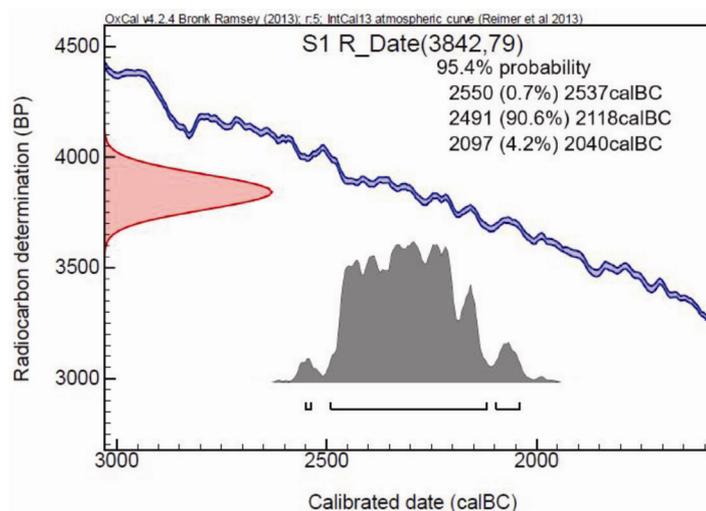


Figure 1. Correction on the date for sample S1 using OxCal 4.2.

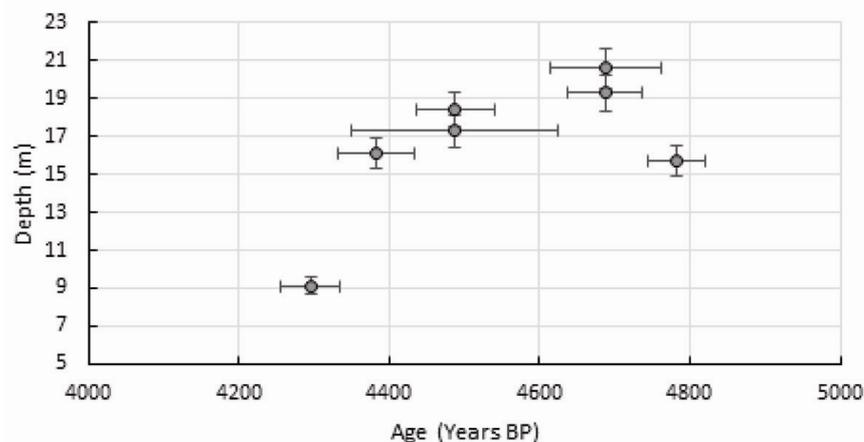


Figure 2. Calibrated age of charcoal samples as a function of depth from Rakhigarhi, Haryana.

before adding the next solvent and repeating the process. The carbon residues were finally washed after the second acid bath and then put in a controlled freeze-drier at a temperature of -85°C for about 6 h so that all the samples could be dried without combustion of carbon from them. About 3 mg of the sample was then subjected to graphitization through a process of controlled burning at 900°C , and CO_2 gas was then mixed with hydrogen and reduced to graphite in the presence of iron powder in a completely sealed environment. The graphite was then made into pellets and inserted into the ion source of the accelerator for isotopic measurements. Blank samples, i.e. dead in ^{14}C were also graphitized and measured following the same protocol as that for the charcoal samples to establish the background values.

Oxalic acid II (OX II) with known isotopic ratio for carbon, procured from the National Institute of Standards and Technology (NIST), USA, was used as primary standard. All the measured values of charcoal samples were normalized to OXII after background correction.

Data quality was monitored with secondary standard sample C7 procured from the International Atomic Energy Agency, Vienna, Austria. Its consensus pMC (percentage modern carbon) value ($\text{C7} = 49.53 \pm 0.12$) matched well with the experimental result ($\text{C7} = 49.38 \pm 0.27$). Data are presented according to Stuiver and Polach⁵. Table 1 and Figure 1 provide the results of the analysis.

Table 1 provides data regarding sample name, the depth at which it was found and the number of carbon atoms measured after 40 min of data collection. The statistical error is determined using the Poissonian statistics as square root of the total observed ^{14}C atoms. Carbon dating is done in two stages. Initially the uncalibrated dates are determined. These are then subjected to calibration data. The uncalibrated (Libby or radiocarbon age) dates of the samples are determined from the measured $^{14}\text{C}/^{12}\text{C}$ ratio. These

are compared against a local standardized source. The ratio therefore varies linearly with time, but the error is calculated with respect to the standard source and is sensitive to the local parameters. It is not purely statistical in nature.

Estimation of calibrated date is a complex function of the dating procedure and calibration curve. This is based on averaged worldwide data. These data in turn depend on the (galactic) cosmic ray flux that produces ^{14}C in the atmosphere. It is not assumed to be constant with time as it varies significantly due to changing solar conditions. Hence the calibration program (OxCal) takes into account the year and error bar from Libby dates (which come directly from the $^{14}\text{C}/^{12}\text{C}$ ratio) and a linear calibration (referred to as uncalibrated dates) and then applies a field-determined calibration curve on the same. Hence the error bars are not linearly related to period, unlike in the uncalibrated data. The error bars on the uncalibrated data are more or less constant around 80 years, but depending on the exact period, this translates into significantly different error due to varying rate of ^{14}C production as a consequence of changing interplanetary environment.

Radiocarbon ages are then converted into calendar ages using OxCal 4.2 (ref. 6) on-line calculator developed by the University of Oxford (<https://c14.arch.ox.ac.uk/oxcal.html>). As a representative case, in Figure 1 we have reproduced the calibration curve for sample S1.

Figure 2 is a plot of the calibrated carbon age of Rakhigarhi.

As can be seen from the figure, the age against the depth graph gives a strong linear fit with a least square fit of 0.4457. The error is driven primarily by two samples from a depth of 15.7 and 17.3, whose value is off the line. If these samples are assumed to have an error, the least square fit improves to 0.9775.

We conclude that the charcoal samples from Rakhigarhi extracted from a depth of 9.1 m have a maximum age of about 2273 ± 38 yrs BC, while the sample

extracted at 20.6 m has a maximum age of 2762 ± 73 yrs BC.

1. Armindra Nath, 2014; asi.nic.in/pdf_data/rakhigarhi_excavation_report_new.pdf
2. Ravi Prasad, G. V. *et al.*, *Curr. Sci.*, 2006, **90**(4), 488–490.
3. Mahapatra, D. P., Behera, A. K., Gopalan, K., Chakraborty, S., Dutta, K. and Somayajulu, B. L. K., *Nucl. Instrum. Methods in Phys. Res. B*, 2004, **223–224**, 26–30.
4. Mahapatra, D. P., Gopalan, K. and Somayajulu, B. L. K., *Curr. Sci.*, 1998, **75**(1), 18–25.
5. Stuiver, M. and Polach, H., *Radiocarbon*, 1977, **19**(3), 355–363.
6. Bronk Ramsey, C. and Lee, S., *Radiocarbon*, 2013, **55**(2–3), 720–730.

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