Radiocarbon dating of charcoal samples from Rakhigarhi using AMS

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Abstract:
We have used the radiocarbon dating facility using Accelerator Mass Spectrometer (AMS) installed the Inter University Accelerator Centre, New Delhi to date the charcoal samples from different depths recovered from a Harappan site at Rakhigarhi, Haryana. We found that the samples at the depth of 9.1 meters give a (calibrated) date of 2273 ± 38 years BC while the samples from the depth of 20.6 meters yield a date of 2616 ± 73 years BC.

Introduction:
Carbon dating is one of the most accurate methods of determination of the age of any archaeological site. Here we present our analysis of seven samples from Rakhigarhi site taken at different depths. We have used the recently acquired AMS system at the Inter University Centre for Accelerator Physics, New Delhi.

Rakhigari site:
Rakhigarhi (29°17′19″N 76°06′47″E) is a village in Hisar District in the state of Haryana in India, situated in the north-west about 150 kilometers from Delhi. In 1963, archaeologists identified one of the largest Harappan sites at this place. It is situated on the dry bed of the now dried river (Amarendra Nath, 2014). The site was excavated in 2015 by a group from Deccan College led by Prof. Vasant Shinde. A vertical trench was dug into the site and various artefacts were recovered from the site. Several charcoal samples were also collected. These were analysed at the Inter University Accelerator Centre which has recently acquired a new radiocarbon dating facility. Here, we report the dates of the Rakhigarhi sites analysed at IUAC.

Details of the excavation have to be provided by Prof. Shinde.

Materials and methods:
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 graphitisation 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, Bhubneswar (Ravi Prasad, Mahapatra et al., 2004, Mahaputra et al., 1998).

For the present study, 7 samples each weighing about 50 grams, were provided by Deccan College, Pune, Maharashtra which were excavated from a depth of 9.1 meters, 15.6 meters, 16.1 meters, 17.3 meters, 18.5

1 Accepted for publication in Current Science, March 2016
meters, 19.3 meters and 20.4 meters (Table 1). Pieces of charcoal were extracted from these samples. Typically 3 grams 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. It was then washed in de-ionised type 1 water (resistivity 18.2 megaohm cm). Afterwards, samples were subjected to Acid (HCl), Base (NaOH) and Acid (HCl) cycle. The samples were dipped in the solvent at 60°C for about 5 hours each and residue was removed. In order to save the small charcoal particles, the samples were also subjected to centrifuge (2000 rpm) for 2 minutes. They were then washed with the de-ionised water several times until the water became Ph neutral before adding the next solvent and repeating the process. The carbon residues were finally washed after 2nd acid bath and then put in a controlled freeze drier at a temperature of -85°C for about 6 hours so that all the samples could be dried without combustion of carbon from the sample. About 3 milligrams of this sample was then subjected to graphitisation through a process of controlled burning at 900°C and CO₂ gas was then mixed with hydrogen and reduced to graphite in the presence of iron powder in a completely sealed environment. This graphite was then made into pallets 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 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 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 the background correction.

Data quality was monitored with secondary standard sample C7 procured from IAEA (International Atomic Energy Agency). It’s consensus pMC (percentage modern carbon) value (C7 = 49.53±0.12) was very well matching with the experimental result (C7 = 49.38±0.27). The data is presented as per Stuiver and Polach (1977). The results of the analysis are given in Table 1 and Figure 1.

Table 1: Details of the results obtained with the charcoal samples obtained from Rakhigarhi

<table>
<thead>
<tr>
<th>No</th>
<th>Depth (m)</th>
<th>No. of $^{14}$C counts</th>
<th>$^{14}$C/$^{12}$C x 10$^{13}$</th>
<th>Libby Years (BP)</th>
<th>Age Max</th>
<th>Min</th>
<th>Calibrated age (BP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>9.1</td>
<td>16372</td>
<td>5.7570</td>
<td>3824 ±80</td>
<td>2351</td>
<td>2196</td>
<td>4273 ±39</td>
</tr>
<tr>
<td>2</td>
<td>15.7</td>
<td>19860</td>
<td>5.4885</td>
<td>4141 ±75</td>
<td>2780</td>
<td>2628</td>
<td>4704 ±38</td>
</tr>
<tr>
<td>3</td>
<td>16.1</td>
<td>14787</td>
<td>5.7036</td>
<td>3900 ±82</td>
<td>2481</td>
<td>2280</td>
<td>4380 ±50</td>
</tr>
<tr>
<td>4</td>
<td>17.3</td>
<td>28876</td>
<td>5.7017</td>
<td>3903 ±224</td>
<td>2671</td>
<td>2119</td>
<td>4395 ±138</td>
</tr>
<tr>
<td>5</td>
<td>18.4</td>
<td>11505</td>
<td>5.5871</td>
<td>4067 ±90</td>
<td>2697</td>
<td>2488</td>
<td>4592 ±52</td>
</tr>
<tr>
<td>6</td>
<td>19.3</td>
<td>20309</td>
<td>5.5923</td>
<td>4060 ±75</td>
<td>2679</td>
<td>2480</td>
<td>4579 ±50</td>
</tr>
<tr>
<td>7</td>
<td>20.6</td>
<td>22870</td>
<td>5.5922</td>
<td>4060 ±129</td>
<td>2762</td>
<td>2470</td>
<td>4616 ±73</td>
</tr>
</tbody>
</table>

The data in the table consists of the sample name, the depth at which it was found, and the number of carbon atoms measured after 40 minutes of data collection. The statistical error is determined using the Poissonian statistics as square root of the total observed $^{14}$C atoms. The 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 sample were determined from the measured $^{14}$C/$^{12}$C ratio. These measures the $^{14}$C to $^{12}$C ratio are compared against a local standardised 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 dating procedure and the calibration curve. This is based on averaged worldwide data. This data in turn depends on the (galactic) cosmic ray flux that produces the $^{14}$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 programme (OxCal) takes into account the year and error.
bar from Libby dates (which come directly from the 14C/12C 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 is more or less constant around 80 years but, depending on the exact period, this translates into significantly different error.

Radiocarbon ages were then converted into calendar ages using the OxCal 4.2 (Bronk Ramsay, and Lee, 2013) online calculator developed by the University of Oxford. (https://c14.arch.ox.ac.uk/oxcal.html) As a representative case, we have reproduced the calibration curve for the sample S1 in figure 1.

The calibrated carbon age of Rakhigarhi is plotted in figure 2.

![Figure 1: Correction on the date for sample S1 using OxCal 4.2](image1)

![Figure 1: Calibrated age of Charcoal samples as function of depth from Rakhigarhi](image2)
As it can be seen from the figure that 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 the depth 15.6m and 17.3, whose value is off the line. If these sample is 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 meters had a maximum age of about 2273 ± 38 years BC while the sample extracted at 20.6 meters had a maximum age of 2762 ± 73 years BC.

Acknowledgement

The authors wish to thank Prof N.V. Chalapathi Rao for his excellent comments on the manuscript that helped us to significantly improve the manuscript.

References:


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