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# ESR dating evidence for early man at a Lower Palaeolithic cave-site in the Northern Caucasus as derived from terrestrial mollusc shells<sup> $\ddagger$ </sup>

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#### Abstract

Eight terrestrial shell samples from recent excavations at Treugolnaya (Triangular) Cave (Northern Caucasus) were analysed by electron spin resonance (ESR) to produce a chronology for the most ancient Acheulian-bearing layers of the cave-site. The lifetime of the 2.0012 centre used for dating is about  $3 \times 10^8$  at 5°C that allows to date the multi-level sequence of the cave-site at least in the range of the last one million years. The dating results obtained suggest that the first (from the bottom) archaeological layer, 7a, is likely to be about 583,000a old, and the next, 5b, is some 393,000 a old. These layers can be correlated with oxygen isotope stages 15 and 11, respectively. The estimates obtained imply that man presumably reached the Northern Caucasus at least as early as the beginning of stage 15, i.e. much earlier than generally recognised. The leaving of the cave by ancient man due to development of glacial environment during the subsequent stage(s) can probably be linked with the penetration of man to the southern areas of the East European Plain. © 2000 Elsevier Science Ltd. All rights reserved.

### 1. Introduction

The problem about when exactly the first inhabitants came to Eastern Europe and where they came from has not yet been solved. Probably, there were several centres of settlement which sprang up at different times. Migration of early men to southern regions of the East European Plain presumably took place from the Caucasian directions. In one's turn, the first inhabitants probably came to the Caucasus from Western Asia (Lyubin, 1984). Evidence is derived from numerous ancient dwelling places and characteristic archaeological materials found in the Transcaucasus (Lyubin, 1989). Some researchers assume the further advance of Acheulian man into the Northern Caucasus and southern regions of the East European Plain, but the Lower Palaeolithic chronology in the area has remained virtually unknown (Lyubin, 1984).

In contrast to open sites, the rock shelters could provide the basic knowledge of the ancient population because deposits in these sites are normally in primary deposition, have a more reliable stratigraphical position and store valuable lithological, palaeontological and archaeological informations. However, many archaeological sites are not datable by conventional dating methods since they are beyond the radiocarbon dating range and lack the material suitable for other dating methods.

In the present work, we report the first comprehensive palaeodosimetric study using ESR analysis on terrestrial mollusc shells collected from the oldest cultural layers of multi-level cave-site located in the Northern Caucasus. The samples come from recent excavations at Treugolnaya (Triangular) Cave which is currently regarded as the only multi-level cave-site in Russian Federation displaying finds of archaic pre-Mousterian industries (Golovanova, 1990, pers. comm.; Lyubin, 1993).

## 2. Description of the cave

Treugolnaya Cave is a karst cavity formed on the Baranach limestone plateau. The cave was discovered by L.V. Golovanova and V.B. Doronichev in 1986. The cave is situated in the basin of the Urup River at approximately 43°54′ north and 41°12′ east, at an elevation of 1510 m above sea level on the northern slope of the Greater Caucasus (Fig. 1).

Excavations revealed the presence of a long sequence of palaeontologically and archaeologically rich deposits. The deposits of cave reference section are divided into 14 lithological layers characterised by a series of units labelled (from top to down) 1–8. According to faunistic,

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Fig. 1. Location of the Treugolnaya cave-site ( $\blacktriangle$ ) and palaeogeographical scheme of Pleistocene glaciations of the Caucasus (

lithological and archaeological data, the cave deposits cover the time span from the Günz-Mindel to the Late Holocene (the Middle Ages) (Doronichev, 1991). Besides, all stratigraphic levels of the cave were formed during the Brunhes Normal Chron and, therefore, should not be older than 0.78 Ma. The lithic materials from layers 4a–b, 5a–b and 7a are classified as Early Acheulian; layers 6 and 7b are archaeologically sterile. Layer 8 is the bottom of the cave.

## 3. ESR-dating of shell fossils

Up to the present, a great number of papers have dealt with the investigations of marine malacofauna remains (see e.g. Ikeya and Ohmura, 1981; Radtke et al., 1985; Molodkov, 1988, 1989; Katzenberger et al., 1989; Skinner, 1989). But only scanty data are available on palaeodosimetric and radiospectroscopic properties of non-marine mollusc shells, and even these are mainly the results of earlier studies on fresh-water gastropods (Molodkov, 1988, 1996). The present work is one of the first efforts to use terrestrial mollusc shells as a dating material. Besides, it is the first attempt to date a Lower Palaeolithic site on the basis of ESR analysis of mollusc skeletal remains taken from Acheulian-bearing layers.

Previous works (see e.g. Molodkov, 1988, 1989, 1993) have shown, that the q = 2.0012 centres and the relevant signal (linewidth  $\sim 2.25 \times 10^{-4}$  T) are most reliable for quantitative analysis and dating of Quaternary mollusc shells including those of terrestrial origin. The signal at q = 2.0012 is usually not detected in multi-component differential spectra of the shells due to interference with the signals of other centres, including those of non-radiation origin. In the present study, quantification of the 2.0012 centre concentration was obtained from the peak-to-peak amplitude in derivative spectra by using an overmodulation (OM) detection method (Molodkov, 1988, 1993). This method has the effect of enhancing the analytical signal at g = 2.0012 and suppressing narrower interfering signals normally observed in the ESR spectra at  $g_1 = 2.0060$ ,  $g_2 = 2.0034$ ,  $g_3 = 2.0022$ ,  $g_4 = 2.0010$ 



Fig. 2. Comparison of ESR signals of terrestrial mollusc shells from Treugolnaya Cave *(Monacha caucasicala* (Lindh.)). Solid line: nature samples; dotted line: shells irradiated to dose of 5 kGy (A) or heated for 12 h at 180°C (B). The spectra were recorded with different amplification factors.

and broader signal at  $g_5 = 1.9983$ , minimising superposition effects from these lines. The method applied assures dosimetric read-out of the intensity of the true resonance lineshape (Molodkov, 1993), which is more precise compared with the overlapping peak intensities of the derivative spectra. Dosimetric reading was performed with microwave power of 2 mW and a modulation amplitude of 1 mT.

For a paramagnetic carbonate centre to be suitable for numerical dating purposes, it must have a lifetime that is sufficiently greater than the age of the sample. Kinetic studies performed on typical shell sample from the Treugolnaya Cave have shown that lifetime of the 2.0012 centres is extremely high and makes up about  $3 \times 10^8$  at  $5^{\circ}$ C which is about two orders of magnitude higher than that of marine shells (Molodkov, 1988) that make it possible to cover the whole Acheulian with such shells without making long-term fading correction (Molodkov, 1989).

Irradiation at a dose of 5 kGy and heating at  $180^{\circ}\text{C}$  for 12 h have revealed the similarity between the life times of the spectral components in the region of interest as well as the similarity of dose dependencies of the signals up to the saturation dose (Fig. 2). It secures more reliable palaeodosimetric analysis of the shell material.

The accumulated palaeodoses were determined from the enhancement of the intensity of the g = 2.0012 signal by exposure of the shell samples to increasing gamma doses. In contrast to five other lines observed in the ESR spectra of the shell substance the dose dependence of the g = 2.0012 signal is well described by exponential function in the range of the doses applied. It allows to use exponential fitting or to convert the data point into a straight line to determine the accumulated palaeodose by extrapolation of the dose dependence to zero-ESRintensity (Fig. 3).

Research into the shells from Treugolnaya Cave showed that mineralogical composition, even of the oldest shells, is represented by aragonite. It means that the original structure of the skeletal substance has preserved. This fact is of great consequence because the possible loss of accumulated age information due to inversion of initial crystalline structure is eliminated.



Fig. 3. Typical dose-response of the carbonate centre at g = 2.0012 in the shell samples from Treugolnaya Cave and evaluation of the accumulated palaeodose,  $P_s$ , by the exponential (A) and the logarithmic (B) fitting of the data points; *I* is the ESR intensity;  $I_{max}$  is the ESR intensity at saturation dose; *r* is the correlation coefficient.

#### 4. Dating results and discussion

Table 1 presents the results obtained on the samples from the cave-site. The mean ages based on dating of syngenetic samples are  $583 \pm 25$  and  $393 \pm 27$  ka for layers 7a and 5b, respectively. The ages obtained for different species show a good agreement within each layer and increase with depth.

The uranium content in the shells seems to be highly uniform ranging from 0.38 to 0.53 ppm indicating that little or no post-depositional U enrichment has occurred. The internal dose rate is much less than the external one. It allows to suggest negligible uncertainty connected with unknown uranium uptake history of shell substance.

The age of culture-bearing layer 7a has been determined on six shell samples and three samples of embedding sediments. On an average, the age is  $583 \pm 25$  ka. Finds of subfossils from the layer suggest forestation in the vicinity of the cave-site and warm climate (Baryshnikov, 1991). The numerical data obtained allow to link the first cultural layer of the cave with oxygen isotope stage 15 (Fig. 4) and to correlate it with the corresponding high-sea-level event fixed by previous ESR studies of marine deposits on Arctic islands (Molodkov et al., 1992; Bolshiyanov and Molodkov, 1999). Our dating results suggest that man made his first appearance in the Northern Caucasus somewhere in the early part of stage 15, i.e. soon after setting in the global climate amelioration and subsequent decay of the mountain ice domes in the Caucasus.

The age of the next culture-bearing layer, 5b, is  $393 \pm 27$  ka. According to the palaeontological data (Baryshnikov, 1991), pollen evidence and low-field magnetic susceptibility record of the cave deposits (Pospelova and Levkovskaya, 1994), the layer could be accumulated during warm interglacial optimum. These observations and dating results are in agreement with our data on the Arctic islands where transgressive deposits of Polar basin seas were dated by ESR at  $455 \pm 37$  to  $365 \pm 35$  ka (Molodkov, 1995; Bolshiyanov and Molodkov, 1999).

The archaeologically sterile layer 6 sandwiched between cultural layers 7a and 5b (Fig. 4) may be indicative of the time when man left the cave shelter. In the beginning, it might have been due to a remarkable cooling of the climate during which the mountain glaciation developed and the altitude of the snow line fell well below the cave bottom. Probably, this made man to move down to valleys and foothills (Lyubin, 1984). Subsequent climatic amelioration during the interglacial promoted the further penetration of man deep into the plain.

### 5. Conclusions

The present work is an effort to use a variety of species of terrestrial gastropods as a dating material in studying Lower Palaeolithic archaeology. The results obtained for the Treugolnaya Cave Acheulian-bearing deposits are interesting in two ways.

On the one hand, the study shows that (1) the signal at  $g = 2.0012 \pm 0.0001$  (linewidth  $\sim 2.25 \times 10^{-4}$  T) seems to be the only signal suitable for concentration analysis and ESR dating of terrestrial mollusc shells. (2) The life-time of the relevant centre is about  $3 \times 10^8$  a at 5°C. It allows to cover the whole Acheulian with such shell material even without making correction for long-term fading. (3) Life times of other radiation-induced components of the spectrum seem to have the same order of magnitude that favours the quantification of the 2.0012 centre concentration. (4) The dose–effect curve of 2.0012 signal is well described by an exponential function and reaches the level of 0.95 from saturation value,  $I_{max}$ , at about 2.5 kGy.

On the other hand, ESR results imply that ancient man came to the Northern Caucasus no later than  $583 \pm 25$  ka ago, i.e. at least near the beginning of "warm" oxygen isotope stage 15. For the second time the cave was occupied about 393,000 years ago, apparently during stage 11. Thus, the occupation of the cave during stages 15 and 11 were probably coeval with global amelioration of the climate in those periods. The absence of

Layer	Sample No.	U (ppm)	Th (ppm)	K (%)	$U_{in} \ (ppm)$	Dose rate $(\mu Gy a^{-1})$	Palaeodose (Gy)	ESR-ages (ka)
7a-1	161-062	0.91	3.55	0.61	0.51	1263	$720 \pm 48$	570 ± 54
7a-2	162-062	1.07	2.31	0.43	0.43	1017	$610 \pm 21$	$600 \pm 46$
7a-1	163-062	0.91	3.55	0.61	0.61	1221	$745 \pm 45$	$610 \pm 54$
7a-2	164-062	1.07	2.31	0.43	0.42	1027	$560 \pm 33$	$545 \pm 41$
7a-2	166-062	1.07	2.31	0.47	0.31	1000	$565 \pm 37$	$565 \pm 45$
7a-3	167-062	0.65	3.36	0.51	0.36	1080	$660 \pm 45$	$610 \pm 51$
Mean age for layer 7a: $583 \pm 25$								
5b	171-062	1.43	4.52	0.76	0.53	1668	$700 \pm 56$	$420 \pm 39$
5b	172-062	1.43	4.52	0.76	0.43	1562	$570 \pm 36$	$365 \pm 28$
Mean age for layer 5b: $393 \pm 27$								

Table 1 ESR results and radioactivity data for samples from Treugolnaya Cave<sup>a</sup>

<sup>a</sup>U, Th, K are uranium, thorium and potasium content in sediments; U<sub>in</sub> is the uranium content in shells.



Fig. 4. (A) — ESR ages of Treugolnaya Cave culture-bearing layers 7a and 5b ( $\blacktriangle$ ) and its correlation with oxygen-isotope record (Shackleton and Opdyke, 1973) and ESR ages of raised marine and lacustrine deposits ( $\textcircled{\bullet}$ ) (after Molodkov, 1985-1999); (B) — stratigraphy of Treugolnaya Cave deposits (after V. Doronichev, pers. comm.).

any sign of man's dwelling in the cave between these stages may be indicative of the setting-in of unfavourable living conditions due to a cooling of the climate in stages 14–12. This time interval can probably be linked with the penetration of ancient man into southern areas of the East European Plain.

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