QUATERNARY RESEARCH 34, 121–128 (1990)

Electron Spin Resonance (ESR) Dating of Hominid-Bearing Deposits in the Caverna delle Fate, Ligure, Italy

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Received January 24, 1989

Stalagmitic floors interstratified with Mousterian deposits from the Caverna delle Fate (Italy) have been dated by the electron spin resonance (ESR) method. Three samples dated fall between 60,000 and 74,000 yr and indicate the age of the Mousterian deposits. They are in good agreement with ages determined by nondestructive γ-ray spectrometry of the human remains from $^{231}Pa/^{235}U$ and $^{230}Th/^{234}U$ ratios, which are, respectively, $75{,}000^{+21{,}000}_{-14{,}000}$ and $82{,}000^{+36{,}000}_{-25{,}000}$ yr. These dates are consistent with the morphological characteristics of the bones as Neanderthal remains. © 1990 University of Washington.

INTRODUCTION

The Caverna delle Fate (Cave of the Fairies) is located 5 km northeast of Finale (Ligure, Italy) (Fig. 1). A wide opening toward the southwest reveals a large hall that extends southward, via a corridor, to a series of galleries and halls extending 150 m.

A large quantity of animal bones and lithic materials were discovered in the Mousterian layers during excavations at the end of the 19th century (Amerano, 1889). Human fossil remains were identified by G. Giacobini (Giacobini and de Lumley, 1983) during a recent revision of the bone assemblage collected by Amerano. The human remains are represented by an incomplete frontal bone of a child (Fate I), an almost complete mandible of a child (Fate II), and a fragment of an adult right half-mandible (Fate III) (Giacobini *et al.*, 1984). Examination of these remains disclosed several Neanderthal features, in keeping with the finding of a Mousterian industry in these layers. However, the remains display some modern features. At present, it is difficult to specify whether the features which are not classical Neanderthalian characteristics have evolutionary significance or are due to regional variations.

The human remains have been dated by nondestructive γ-ray spectrometry using a high-purity germanium γ-x detector (Giacobini *et al.*, 1984). Activity of radionuclides in the human bones is given in Table 1. The ages deduced from the $^{231}Pa/^{235}U$ and $^{230}Th/^{234}U$ ratios are, respectively, $75{,}000^{+21{,}000}_{-14{,}000}$ and $82{,}000^{+36{,}000}_{-25{,}000}$ yr.

Discovery of the human remains and their dating has so revived interest in this exceptional site that new excavations were carried out between 1984 and 1988 by a Franco-Italian team under the direction of G. Giacobini, G. Vicino, and H. de Lumley. Their purpose was to establish the chronostratigraphy of the deposits in relation to the human remains. This new excavation led to the discovery of further remains, including a fragment of parietal bone which can be anatomically connected with the previously discovered child's frontal bone (Fate I). Unfortunately, these new human remains were discovered in the waste of the old excavations so that their stratigraphic position has not been established. The new excavation, however, revealed that the Mousterian deposits are interstratified with several stalagmitic floors. Therefore, the radiometric dating of these floors is useful in establishing closer relationships between the human remains and the chronostratigraphy of the deposits. For this pur-

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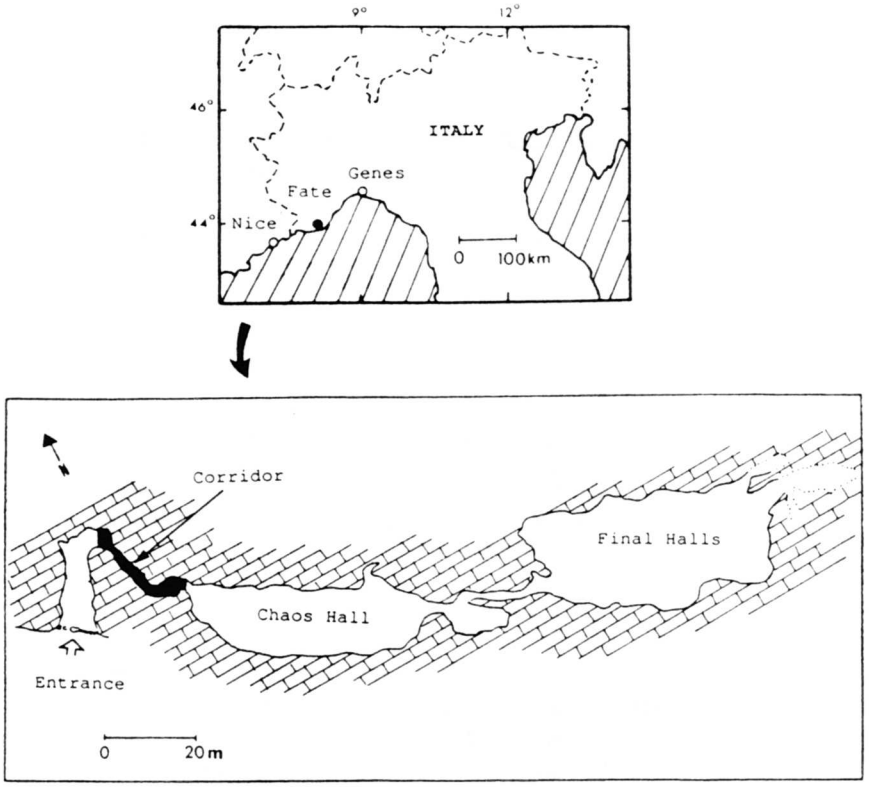


FIG. 1. Map of the Caverna delle Fate in a karst region of northwestern Italy. The dark zone represents Amerano's excavation corridor where the studied samples were extracted. This corridor is followed by a large hall, called Chaos Hall, and other complex galleries and rooms.

pose, we have dated three calcite samples by the electron spin resonance method (ESR).

SAMPLES

Amerano's excavations in the corridor (dark part) of the cave (Fig. 1) permitted the identification of archaeological layers from Mousterian to Middle Neolithic. The artifacts collected in the old layers have been classified as a typical Mousterian assemblage, rich in scrapers, with Levallois technique and of non-Levalloisian facies (de Lumley, 1969). The exact position of the human remains in the cave is not known, but we assume that they were collected in this corridor together with the other materials.

TABLE 1. ACTIVITY OF RADIONUCLIDES IN THE THREE HUMAN BONES

Nuclide	Activity (dpm/g)
^{238}U	2.70 ± 0.04
^{234}U	3.21 ± 0.60
^{230}Th	1.74 ± 0.19
^{226}Ra	1.35 ± 0.09
^{222}Rn	0.78 ± 0.01
^{210}Pb	0.78 ± 0.10
^{231}Pa	0.099 ± 0.008
^{228}Ra	0.06 ± 0.02
^{228}Th	0.06 ± 0.01

The samples used for the present study are speleothems collected from this part of the cave. They belong to two stalagmitic floors which are interstratified in the Mousterian layers (Fig. 2). Three samples (F-85-2, F-85-9, and F-85-10) were studied.

ESR DATING OF CALCITE SAMPLES

Method

ESR signal intensity is proportional to

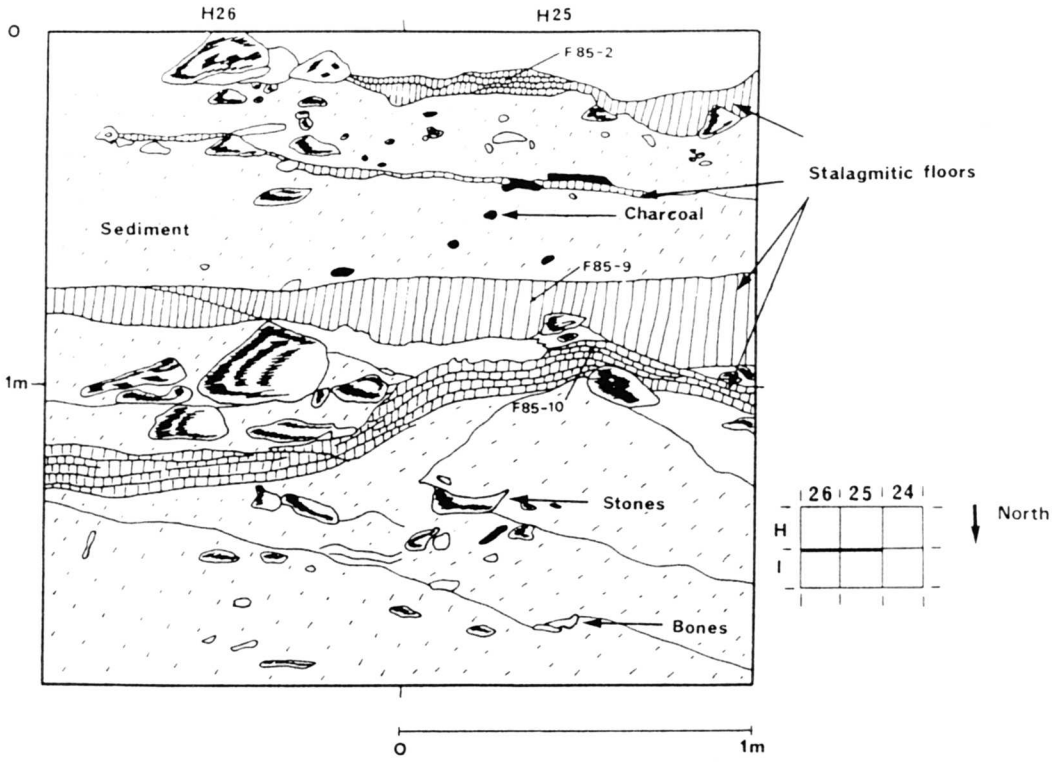


FIG. 2. Stratigraphic position of the three studied samples. This vertical section shows the Mousterian deposits interstratified with stalagmitic floors in the corridor. This section was made against the south wall of the corridor. Sediments are silty clay loam with gravels (about 15%).

the number of trapped electrons accumulated from the moment of calcite formation until the present. These trapped electrons are due to the action of the ionizing radiations (α, β, γ). Measurement of their number permits the dating. The ESR age of a sample is determined by the relation

$$ED = \int_0^t AD\, dt, \qquad (1)$$

where ED is the equivalent radiation dose received by the sample during its entire age, AD is the annual radiation dose, and t is the age. The annual dose is a sum of the doses due to different radiations,

$$AD = k \cdot D\alpha + D\beta + D\gamma + Dcosmic, \qquad (2)$$

where $D\alpha$, $D\beta$, and $D\gamma$ are the annual doses due to α-rays, β-rays, and γ-rays, respectively. Dcosmic is the annual dose due to cosmic rays, and k is the sensitivity ratio between the α-rays and the other radiations (k value).

The $D\alpha$ and $D\beta$ are due to the radioactive elements, principally uranium and its daughters, included in the calcite. Since the newly formed calcite contains only uranium without its daughters, the $D\alpha$ and $D\beta$ increase with time according to the formation of the uranium descendants ^{230}Th and ^{231}Pa and their daughters. The $D\gamma$ is essentially due to γ-rays coming from the sediment. These annual doses are determined from the uranium, thorium, and potassium contents of the calcite and the sediment (Yokoyama *et al.*, 1982).

ESR spectra of natural palaeolithic calcites usually show three radiation-induced lines termed by Yokoyama *et al.* (1983) h1

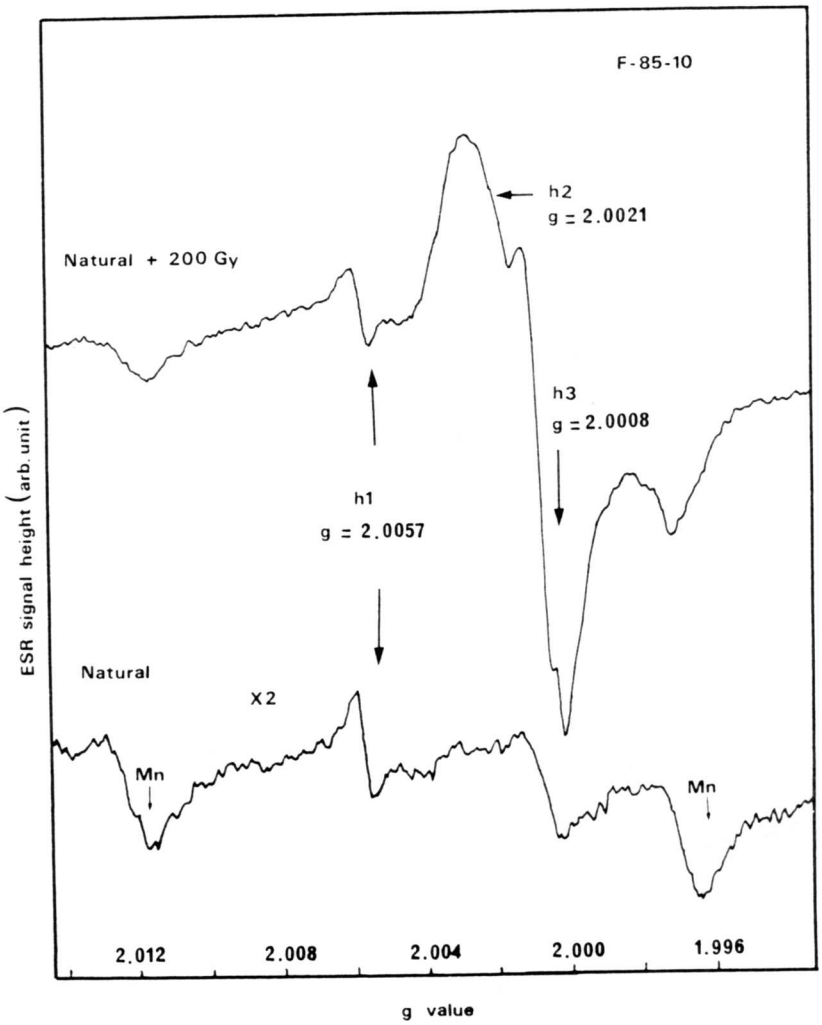


FIG. 3. ESR spectra of natural and irradiated calcite sample (F-85-10) of the Caverna delle Fate. A scanning speed of 8 min is used for these spectra.

(g = 2.0057), h2 (g = 2.0021), and h3 (g = 2.0008) (Fig. 3). Another radiation-induced line, called Broad-Line (BL) at g = 2.004, has been pointed out by Apers *et al.* (1981). These signals are due to trapped electrons and can be used for dating, except h2 which is not sufficiently stable.

Measurements were made at room temperature using a Varian E109 X-band spectrometer. The experimental conditions

TABLE 2. EXPERIMENTAL CONDITIONS FOR OBSERVATION OF CALCITE SIGNALS IN THIS STUDY

ESR signals	g value	Microwave power (mW)	Modulation (G)	Scan range (G)	Cavity temperature (°K)
h1	2.0057	5	0.5	80	293
h3	2.0008	5	0.5	80	293
BL	2.0040	0.1	5	80	293

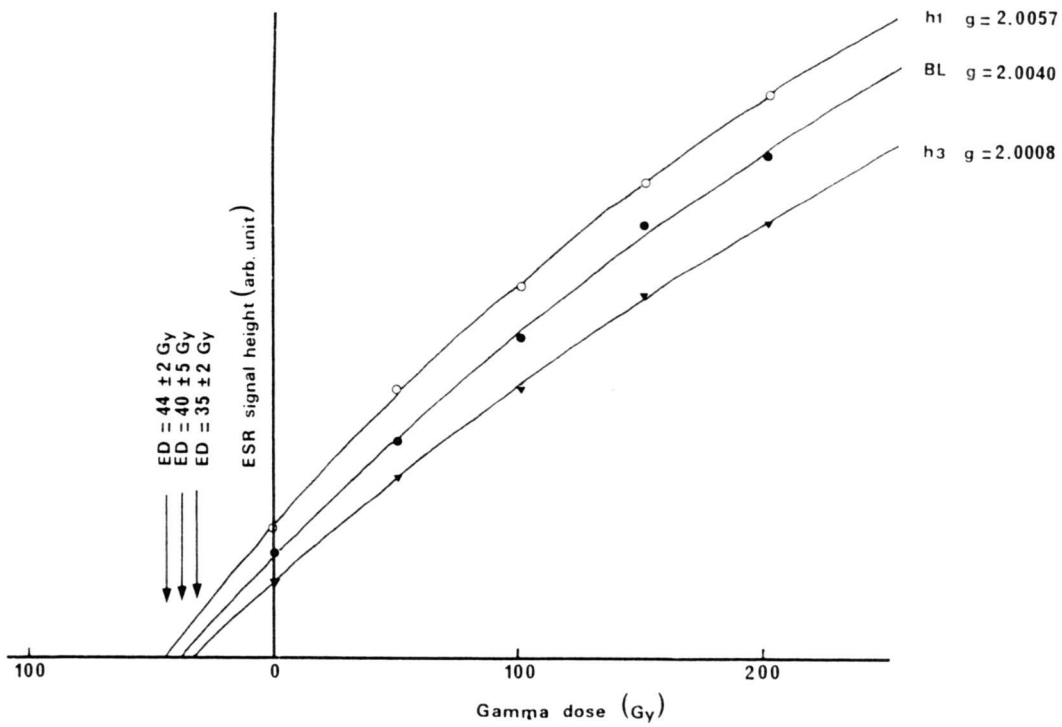


FIG. 4. Determination of the equivalent doses for the three ESR signals, h1, h3, and BL, by the additive method for F-85-10 sample.

were the same for the measurement of h1 and h3 signals (Table 2). The Broad-Line signal was observed with a weaker microwave power of 0.1 mW and a wider modulation amplitude of 5 Gauss because it saturates at a low microwave power and because it is a wider signal than the others (Yokoyama *et al.*, 1985). The measurement of h1 signal was carried out after thermal pretreatment at 180°C for 16 hr (Yokoyama *et al.*, 1983).

Results

The equivalent doses (ED) of the three samples are determined by the additive method (Fig. 4). For each sample, the ED is determined for h1, h3, and BL signals. The results are in good agreement with respect to the σ errors.

Uranium content is low in these stalagmite samples. The internal annual dose is determined as a function of time because of the growth of the ^{230}Th (and ^{231}Pa) from their parents. For the age calculation, this variation has been taken into account. For the annual doses, the errors are estimated as 10%. In Table 3, the internal annual doses are given for the time of precipitation (deposition) ($t = 0$) and for the present ($t = t$).

The external doses were determined by γ-ray spectrometry of the surrounding sediments. A water content of 15% in the sediment was used for calculation of the external dose. γ-rays coming from the sediments are attenuated in the stalagmitic floors. The attenuation factor is calculated with the equation

$$A = 0.5[\exp(-m * D) + m * D * \mathrm{Ei}(-m * D)] \quad (3)$$

where A is the fraction of the γ-rays

reached at the distance D (g/cm^2) from the sediment, m is the attenuation coefficient of the γ-ray (cm^2/g), Ei is the exponential integration function, and 0.5 is the 2π geometric factor (Yokoyama *et al.*, 1987). The mean attenuation coefficient of γ-rays from the uranium series is 0.070 cm^2/g, that of the thorium series is 0.067 cm^2/g, and that of potassium 0.054 cm^2/g (Yokoyama *et al.*, 1982). We adopted a mean value of 0.065 cm^2/g. If a stalagmitic floor is sandwiched between two sediment layers, γ-rays coming from these two layers should be added.

For the three calcite samples from the Fate cave, the attenuation factors are estimated as 0.90, 0.60, and 0.50 for F-85-2, F-85-9, and F-85-10 samples, respectively. The cosmic dose is estimated as 50 mGy/yr. A k value (sensibility ratio between α-ray and β-ray) of 0.265 and a $^{234}U/^{238}U$ ratio equivalent to 1.2 ± 0.1 have been taken into account for the annual dose calculation. The resulting ages are 60,000 ± 7000, 72,000 ± 8000, and 74,000 ± 10,000 yr, respectively, for samples F-85-2, F-85-9, and F-85-10 (Table 3).

According to Bichet (1987) the sediment between the two studied stalagmitic floors corresponds to a cold period. Speleothems usually grow during humid and temperate climate conditions.

Taking into account 1 σ error, the ages suggest that the stalagmitic floor from which samples F-85-9 and F-85-10 were collected likely dates to isotope substage 5a and (or) the early part of stage 4. In consideration of the two points previously mentioned, we think that this carbonate formation most likely grew during the last part of isotope stage 5 (Fig. 5).

The speleothem that contains F-85-2 grew in stage 4 and (or) the beginning of isotope stage 3. For the same reasons, we think that this carbonate deposit probably dates to the beginning of isotope stage 3.

The ages of the human remains previously determined by nondestructive γ-ray spectrometry are compatible with the new ESR ages of the deposits. This agreement gives additional support to the assumption that the human remains were collected in Amerano's excavated corridor and probably came from deposits dating between oxygen isotopic stage 3 and substage 5a.

ACKNOWLEDGMENTS

We thank H. de Lumley and G. Giacobini for samples and discussion. We are grateful to C. Chachaty of

TABLE 3. EQUIVALENT DOSES, ANNUAL DOSES, AND AGES OF THE FATE SAMPLES

Sample	U (ppm)	Equivalent dose (Gy)	Annual dose (mGy/yr)				Age (10^3 yr)	
			Internal		External			
			$t = 0$	$t = t$	Gamma	Cosmic		
F-85-2	0.180	h3 57 ± 5 h1 51 ± 1 BL 53 ± 4	47	123	744	50	64 ± 9[a] 58 ± 6 60 ± 8	60 ± 7[b]
F-85-9	0.166	h3 44 ± 1 h1 50 ± 4 BL 46 ± 2	43	122	496	50	69 ± 8 77 ± 10 72 ± 8	72 ± 8
F-85-10	0.129	h3 35 ± 2 h1 44 ± 2 BL 40 ± 3	34	97	414	50	66 ± 8 82 ± 9 75 ± 9	74 ± 10

[a] The errors are quadratical addition of the equivalent dose errors and the annual dose errors. For the equivalent dose errors, a minimum of 5% is used if the observed errors are smaller than this value.

[b] The average ages are calculated from the weighted average of equivalent doses of the three signals h1, h3, and BL.

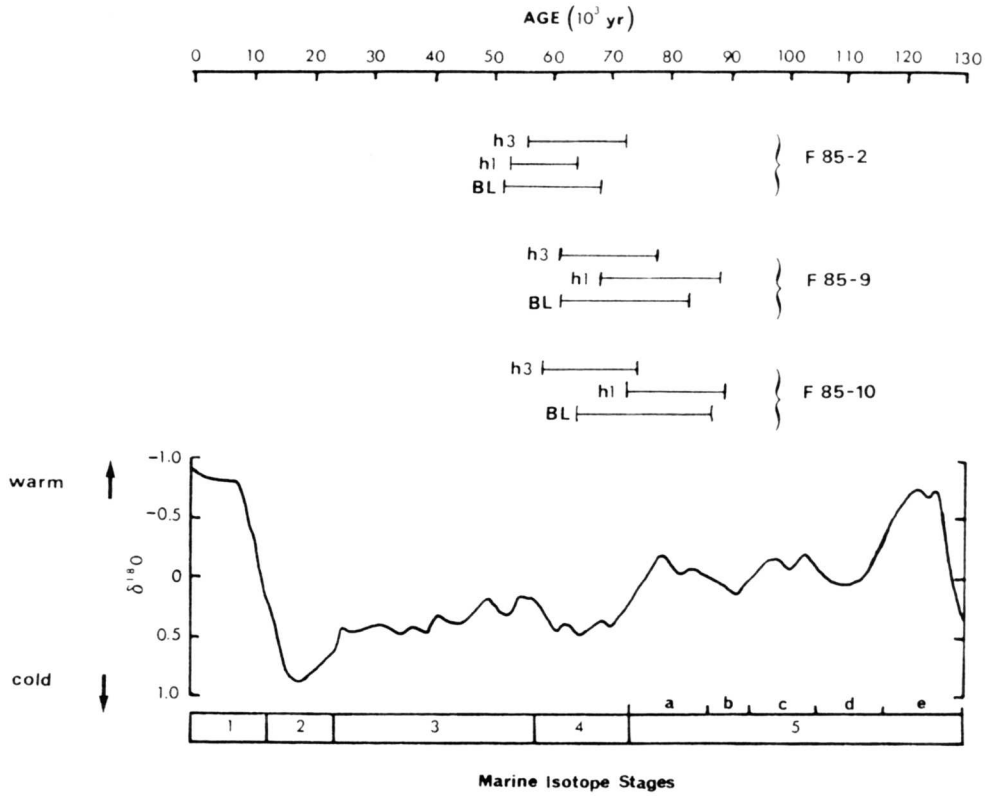


FIG. 5. The time span of growth of the studied stalagmitic floors of the Caverna delle Fate with respect to the variation of $\delta^{18}O$ of themarine isotope stages (Martinson *et al.*, 1987). For each sample, three induced signals (h1, h3, and BL) have been used to determine the age.

the Département de Physico-chimie du Centre Nucléaire de Saclay and to B. Morin of the Département de Spectrochimie de Paris VI for the use of ESR spectrometer. We thank J. J. Bahain of the Centre des Faibles radioactivités de Gif sur Yvette for his help in the experiment.

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