P A C T

6 1982

Revue du groupe européen d'études pour les techniques physiques, chimiques, et mathématiques appliquées à l'archéologie

Journal of the European Study Group on Physical, Chemical and Mathematical Techniques Applied to Archaeology

A SPECIALIST SEMINAR ON THERMOLUMINESCENCE DATING

Oxford, Research Laboratory for Archaeology and the History of Art, 1980

By courtesy E. Unfried and N. Vanna, Vienna, Austria.

STRASBOURG

CONSEIL DE L'EUROPE — COUNCIL OF EUROPE

Assemblée parlementaire — Parliamentary Assembly

TLS II-46: S.G.E. Bowman, R.P.W. Loosemore, G. de G. Sieveking and F. Bordes

Preliminary dates for Pech de l'Aze IV

Abstract

Preliminary thermoluminescence dates are given for burnt flint from Pech de l'Aze IV, a Middle Paleolithic site in the Dordogne. In comparison with the archaeological evidence these ages are significantly low.

1. *Introduction*

Four separate sites are recognised at Pech de l'Aze in the Dordogne (France). Pech de l'Aze I and II are separate entrances to the same cave and were occupied in the Riss and early Würm periods (Bordes, 1954, 1955) until the cave was filled to the roof with debris.

Pech de l'Aze IV (Bordes, 1975) is a destroyed rock shelter adjacent to the cave. It contains deposits of the early Würm period and appears to be the site to which settlement moved when the cave was no longer habitable.

No Upper Palaeolithic occupation occurred at Pech IV. The shelter was entirely filled with Middle Palaeolithic (Mousterian Culture) occupation deposits and sealed by the collapse of the roof. As the radiocarbon date of the boundary between the latest Mousterian cultures and the succeeding Upper Palaeolithic culture occurs at 35,000 BC (Van der Hammen *et al.*, 1967; Waterbolk, 1971) this forms a convenient upper age for the site.

In the regional climatic sequence the uppermost levels in the shelter (levels F3 and F4 are ascribed to Würm II, while earlier levels (H, I, J, X and Z) are assigned to different stages of the (Perigord) regional sequence for the Würm I period (Lavalle, 1978). The chronological relationship between this regional Würm sequence and the better-known climatic sequence in the Netherlands is the subject of dispute, but the dates put forward by Waterbolk (1977) for Würm I i.e. circa 55,000-45,000 BP may be regarded as minimal, since the beginning of this period is probably much earlier.

The ages of the Pech de l'Aze IV deposits attributed to this part of the sequence should thus not be less than 45,000 years BP, and earlier levels (X, Z) that are attributed to early stages of Würm I should be correspondingly older and separated from the latest levels (H and J). The regional sedimentological and climatic sequence thus provides a useful control for the probable age of the site in addition to the archaeology.

The age of Pech de l'Aze IV and the presence of burnt flint make it an obvious candidate for thermoluminescence (TL) dating. Preliminary results are presented for seven flints originating from six different archaeological levels.

2. *Experimental procedure*

2.1 *Sample preparation*

A minimum of 2 mm was removed from the outside of each flint using a low concentration diamond wafering blade (Buehler 11-1180 low-speed saw, Buehler Ltd, Evanston, Illinois). For three of the flints, less than 160 mg of sample remained and for one of these (Z) only 50 mg. For such small samples, preparation of a sequence of slices, as suggested by Göksu and Fremlin (1972) to avoid spurious TL, was impractical.

The samples were crushed in a steel percussion mortar prior to deposition of the 1-8 μm grains on aluminium discs (Zimmerman, 1971). To provide sufficient material to deposit, it was necessary to crush the smaller samples several times, and even so only about seven discs were obtained.

2.2 *TL measurements*

The TL equipment (Littlemore Scientific Engineering, Oxford) has been described elsewhere (Aitken and Fleming, 1972). An EMI 9635 photomultiplier was used in conjunction with Corning 7-59 and Chance-Pilkington HA3 filters. A heating rate of 10° C s^{-1} was employed.

To evaluate the archaeological dose received by each flint, artificial beta doses were given using a 40 mCi $Sr/^{90}Y$ plaque source (type SIP, Radiochemical Centre, Amersham, Bucks). The dose-rate delivered by this source was 1.26 rads s^{-1} at the sample position (15.7 mm). Six ^{241}Am alpha foil sources (Singhvi and Aitken, 1978; marketed by Littlemore Scientific Engineering, Oxford) were used for determination of a-values (Aitken and Bowman, 1975). The strengths of these sources are between 0.24 and 0.25 μm^{-2} minute^{-1}.

All the flints studied had a high temperature TL peak at about 370° C (Fig. 1). The height of this peak was used in all TL measurements. Normalisation from disc to disc was effected by a third glow monitor using the « equal pre-dose » method (as previously suggested by Wagner), which has been found to be more reliable than using the zero glow response of the 110° C peak. The « equal pre-dose » method has the advantage that, up to the fourth glow, all discs have received the same total pre-treatment and therefore can all be used to assess the supralinearity correction.

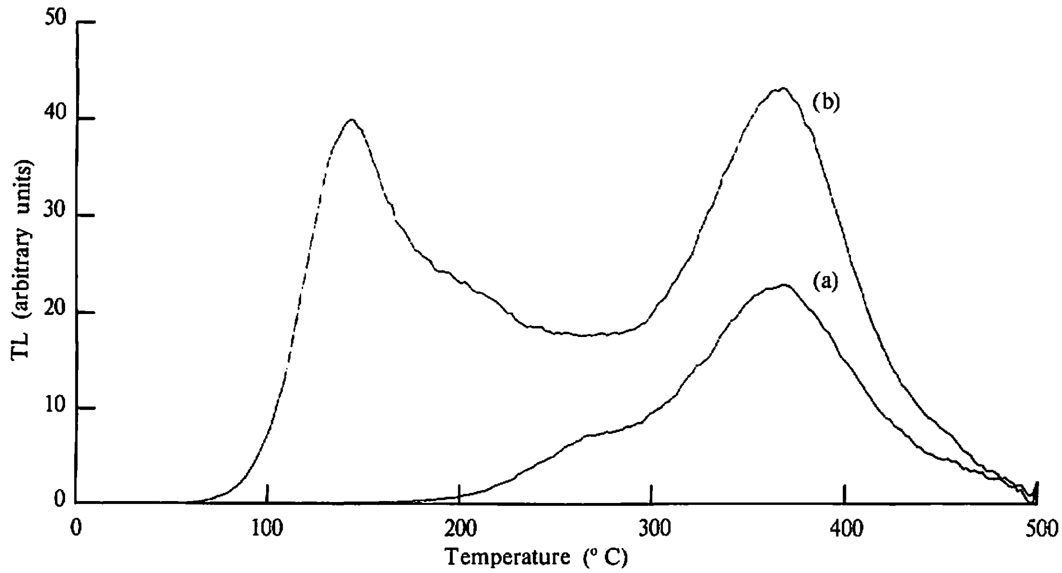

Fig. 1. Glow curves for sample J3b (ii). (a) Natural TL; (b) Natural plus beta TL (β = 4.56 krads).

Spurious TL was only observed in those samples which had been crushed a number of times before deposition. It was reduced to an acceptable level (less than 5 % of the natural TL) by evacuating the glow oven to about 0.01 torr prior to each TL measurement and each « black-body » run. During measurements nitrogen with an oxygen content of less than 1 ppm was used at a flow rate of 6 litres per minute. Relatively low levels of spurious TL for crushed flint have also been observed by Valladas (1979).

For small samples, where only a few discs were obtained, the outer 2 mm of the flint (Bowman, 1980a) were used to check the linearity of the TL growth curves at high dose prior to evaluation of the archaeological dose received by the inside of the flint (Fig. 2).

2.3 *Radioactivity analysis*

Atomic absorption was used to measure potassium content and thick sample alpha counting for the combined uranium and thorium content. The uranium : thorium ratio in flint is typically 6:1; for the purposes of this paper it has been assumed that the samples contain only uranium. For such a ratio, the error in the total dose introduced by this assumption is estimated to be about 1 %.

The environmental dose was measured using natural calcium fluoride (MBLE type super S) in nylon capsules (Aitken, 1969). Dosimeters were

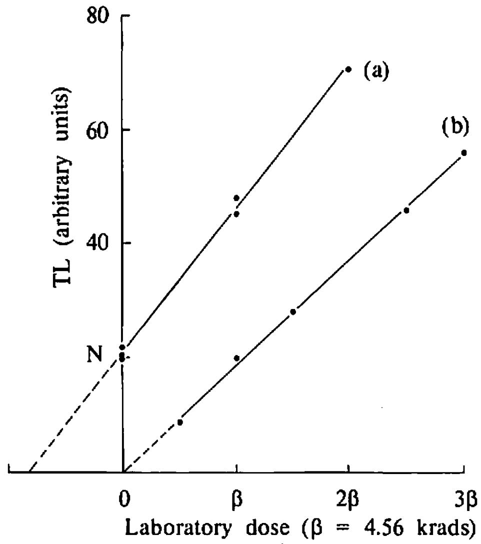

Fig. 2. Growth curves for sample J3b (ii). (a) First glow growth ; (b) Fourth glow growth.

placed in levels F and X, and at the boundary of J3a and J3b. A correction of 7.3 % has been made to allow for over-response of the phosphor (Murray *et al.*, 1979 ; Murray, 1980, personal communication).

3. *Results*

The TL and radioactivity measurements are summarised in Table 1 together with derived TL ages.

Samples from the levels assigned to Würm I all indicate ages around 43,000 years whereas the only sample from a Würm II level appears to be less than half this age.

4. *Discussion*

4.1 *Alpha count-rates*

In Table 1, the errors given on the alpha count-rates are based on counting statistics for the sample only. Most of the samples were in the form of grains (8 μm to 2 mm) together with much larger lumps and were counted in an unsealed state. Measurements of the background (typically less than 8×10^{-3} ks^{-1} cm^{-2}), however, were made with the sample holder sealed. It has been pointed out (Wintle, 1980, personal communication) that radon may give rise to higher background count rates in the unsealed state. This was confirmed when a measurement of background was found to decrease from 2.8×10^{-2} to 0.5×10^{-2} ks^{-1} cm^{-2} when the zinc sulphide screen was covered with perspex.

On the basis of the higher background (i.e. a lower true sample count

TABLE 1.

	Level	Sample	Equivalent Dose, Q. (krads)	Intercept I (krads)	a-value	Alpha count $ks^{-1}cm^{-2}$	Alpha count % error	K_2O (ppm)	Dose-rates ($rads\ a^{-1}$) Intern.	Dose-rates ($rads\ a^{-1}$) Extern.	TL Age years$\times 10^3$
Würm	F		2.05 ± 0.10	0 ± 0.10	0.051 ± 0.015	0.170	4	700	0.041	0.063	19.6 ± 1.6
Würm I	J1	ii	4.02 ± 0.28	0.48 ± 0.02	0.092 ± 0.008	0.127	4	505	0.040	0.062	44.1 ± 4.0
	J3a		4.06 ± 0.20	0.14 ± 0.01	0.085 ± 0.010	0.099	7	1020	0.034	0.062	43.7 ± 3.5
	J3b	i	4.51 ± 0.32	0 ± 0.10	0.085 ± 0.010	0.144	6	340	0.042	0.062	43.4 ± 4.3
		ii	3.81 ± 0.23	0 ± 0.10	0.104 ± 0.010	0.095	5	610	0.034	0.062	39.9 ± 3.2
	X	ii	3.25 ± 0.20	0.114 ± 0.006	0.093 ± 0.010	0.085	7	800	0.030	0.047	42.4 ± 3.4
	Z		3.97 ± 0.32	0.50 ± 0.03	0.077 ± 0.010	0.126	9	785	0.038	0.047	46.6 ± 5.0

Notes.

1. Errors on Q and I are based on a least squares fit to the data.
2. A 5% error was assumed in the potassium analyses.
3. The error on the alpha count is based only on the counting statistics for the sample.
4. A 5% random error and 5% systematic error were assumed on the environmental dosimetry.
5. Overall error assessment follows Aitken (1976).
6. Conversion to dose-rate of K_2O content and alpha count-rate as in Bell (1979) and assuming no thorium content. A correction has been made for the difference in alpha particle ranges in silica and pottery (Bowman, 1980b).
7. It has been assumed that the supralinearity correction can be applied as in fine grain pottery dating.

rate) an age approximately 9% higher is obtained. For the actual counting geometry, it is likely that a background between the extremes is applicable.

It is intended that the samples are recounted as sealed powders in an attempt to avoid this problem. In addition both uranium and thorium content are to be measured by neutron activation analysis.

4.2 *Homogeneity*

Localised differences in TL sensitivity and radioactivity content have been observed in some flints (Bowman and Seeley, 1979). Such effects will not be detectable in samples prepared as fine grains. TL studies on the outer 2 mm of the Pech de l'Aze IV flints (Bowman, 1980a) however, did not indicate any gross variation of the sample characteristics. The investigation of spatial variation of TL and uranium content is nevertheless being undertaken.

4.3 *The TL Ages*

From the archaeological and other evidence outlined in the introduction the following observations can be made:

1) the TL ages for levels J, X and Z do not successively increase as expected from the stratigraphy;

2) the TL ages for these levels are lower, by a factor of between 1.5 and 2, than the generally accepted date of Würm I;

3) as expected, the TL age of level F3 is significantly lower than those of levels J, X and Z, but is roughly half the accepted age for Würm II.

4.4 *TL stability*

The effect of exposure to light has not yet been tested. However, in flint, sensitivity to light has not been observed by other workers (Wintle and Aitken, 1977). All of the samples were prepared in subdued light.

The kinetics of the high temperature TL peak of sample X(ii) have been studied (Bowman, 1980a). The values of 1.66 ± 0.03 eV for the trap depth and about $10^{13}\,s^{-1}$ for the frequency factor obtained by Hoogenstraaten's method (1958) are in reasonable agreement with values quoted in Wintle and Aitken (1977). A mean life of the order of 10^8 years, therefore, implies adequate thermal stability. This is reflected by the plateau test (Fig. 3).

Two samples [X(ii) and J3a] were tested for anomalous fading (Wintle, 1973). For each sample two discs were used. The natural TL of both was measured. One disc was then given a 9 krad beta dose and left unglowed for eight days. The TL induced by this dose was then compared with that from the second disc, given the same dose but glowed immediately. The natural TL was used to normalize the beta-induced TL. Within the experimental error of $\pm$ 3%, there was no loss of high temperature TL for either sample.

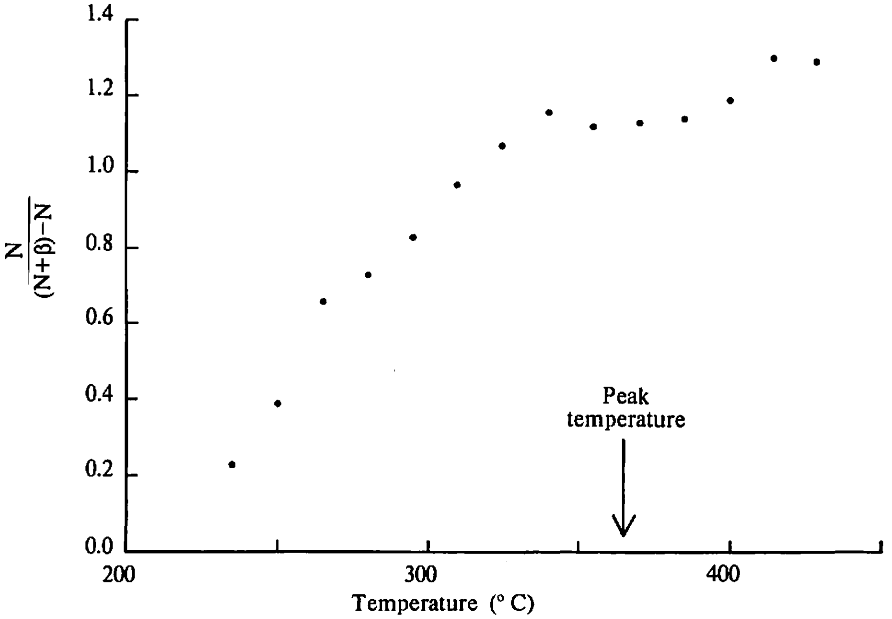

Fig. 3. Sample J3b(ii): Plateau test.

Long-term fading checks are now being performed; the first three of these are described in Bowman (1980a). In summary, after a delay of approximately three years, a significantly low TL evaluation of two known gamma doses was obtained. However, it has not as yet been determined whether the discrepancy is due to the period of storage, crushing of the samples after dosing or some other cause, since the first two possibilities seem unlikely (Bowman, 1980a).

Summary

On the basis of the archaeological evidence the preliminary TL ages for Pech de l'Aze IV are too low to be acceptable. The discrepancy does not seem to be attributable to lack of thermal stability or to short-term anomalous fading.

Acknowledgements

We wish to thank Dr M.S. Tite for helpful discussions throughout this work; M.-A. Seeley for measuring the environmental doses and M. Cowell for the atomic absorption measurements.

References

Aitken, M.J., 1969, in *Archaeometry*, 11, p. 109.

Aitken, M.J., 1976, in *Archaeometry*, 18, p. 23.

AITKEN, M.J. and FLEMING, S.J., 1972, in *Topics in Radiation Dosimetry*, suppl. 1 (ed. F.H. Attix), Academic Press, p. 1.

AITKEN, M.J. and BOWMAN, S.G.E., 1975, in *Archaeometry*, 17, p. 132.

BELL, W.T., 1979, in *Archaeometry*, 21, p. 243.

BORDES, F., 1954, in *L'Anthropologie*, 58, p. 401. – 1955, in *L'Anthropologie*, 59, p. 1.

BORDES, F., 1975, in *Bulletin de la Société préhistorique française*, 72, p. 293.

BOWMAN, S.G.E., 1980a, *TL studies on burnt flint*, this volume.

BOWMAN, S.G.E., 1980b, *Alpha particle ranges in pottery*, this volume.

BOWMAN, S.G.E. and SEELEY, M.-A., 1979, in *PACT*, 2, p. 151.

GÖKSU, H.Y. and FREMLIN, J.H., 1972, in *Archaeometry*, 14, p. 127.

HOOGENSTRAATEN, W., 1958, in *Philips Res. Rep.*, 13, p. 515.

LAVALLE, H., 1978, *Stratigraphie et Chronologie du Remplissage des Grottes du Périgord*, in *Études quaternaires*, 3, Marseille.

MURRAY, A.S., BOWMAN, S.G.E. and AITKEN, M.J., 1979, in *PACT*, 2, p. 84.

SINGHVI, A.K. and AITKEN, M.J., 1978, in *Ancient TL*, n° 3, p. 2.

VALLADAS, H., 1979, in *PACT*, 2, p. 180.

VAN DER HAMMEN, T., MAARLEVELD, G.C., VOGEL, J.C. and ZAGWIJN, W.H., 1967, *Geologie en Mijnbouw*, 46 (3).

WATERBOLK, H.T., 1971, in *The Origin of Homo Sapiens*, (ed. F. Bordes), UNESCO, Paris.

WINTLE, A.G., 1973, in *Nature*, 245, p. 143.

WINTLE, A.G. and AITKEN, M.J., 1977, in *Archaeometry*, 19, p. 111.

ZIMMERMAN, D.W., 1971, in *Archaeometry*, 13, p. 29.

S.G.E. BOWMAN, R.P.W. LOOSEMORE, G. de G. SIEVEKING* and F. BORDES**
Research Laboratory, The British Museum, London WC1B 3DG, U.K.

* Department of Prehistoric and Romano-British Antiquities, The British Museum.
** Institut Quaterniaire, Bordeaux University 1, France.