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Skinflint dating☆

H.P. Schwarcz*, W.J. Rink

School of Geography and Geology, McMaster University, Hamilton, Ont., Canada L8S 4M1

Abstract

When a piece of flint is broken to form an artifact, and then buried in sediment at a site, its outer surface is exposed to a higher beta-particle dose rate than is experienced by the interior of the flint. A layer about 2 mm thick on each freshly broken surface receives an excess β-dose. The difference between the internal accumulated dose and that in the skin can be used to estimate the time elapsed since formation and burial of the artifact.

1. Introduction

The principal record of human cultural evolution is preserved in the form of chipped stone artifacts. One of the great challenges in archaeology is the dating of these stone artifacts, a large proportion of which are made of flint or other siliceous fine-grained rocks. If the artifact has been heated in a fire, we are able to date it by thermoluminescence (Valladas, 1992) and possibly by ESR (Porat and Schwarcz, 1991). It would also be desirable to have a method which would allow us to date unheated chipped stone artifacts, that is, to be able to provide the date when a piece of stone was chipped to form the artifact.

A standard procedure in the preparation of burnt flints for analysis is the removal of the outer 1–2 mm of flint from the artifact, in order to remove that part of the sample which has experienced some additional α- and β-dose from the surrounding sediment. This procedure assures that only the environmental (γ- and cosmic-ray doses will contribute to the external dose experienced by the sample. Note that this outer skin of the artifact received an additional β- dose only after it was knapped (chipped) and buried in sediment at the site. Previously, the material in that skin would have been buried several millimetres inside the mass of the raw material ("blank") that was to be shaped into the artifact. The onset of accumulation of additional β-dose would typically be the same as the date of manufacture of the artifact. We can thus see that the age of manufacture of some artifacts could be determined by measuring the growth of radiation dose in the outermost 2 mm (or thinner) "skin" of flint or other raw material. We call this method "skinflint dating". The method is shown schematically in Fig. 1. Somewhat similar concepts were presented in a paper by Ikeya and Tani (1997).

The method has certain assumptions to be satisfied:

1. The radiation-induced signals in the flint artifacts are not saturated as a result of the prior geological dose history of the sample.
2. The β-dose rate from the external sediment at the site is significantly greater than arising from the flint itself.
3. The radiation-induced signal in the outer surface of the artifact was not significantly affected by the knapping process itself, or subsequent exposure to light (including the time since its recovery from the site).

Regarding the first assumption, we must consider TL and ESR signals separately. There does not appear to be any good evidence in the literature regarding the degree of saturation of TL in unburned flint. Typically, we test artifacts to determine if they have been burned by giving them a small test dose to see whether the TL signal increases. The test dose given is so small that it is difficult to see whether there is any further growth although even in this instance the analyst typically finds a small (1–5%) increase after irradiation (H. Valladas, pers. comm.). Note as well that heating flint substantially increases its TL sensitivity. Regarding ESR sensitivity, A. Skinner has noted that most unburned flint shows a substantial

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* Corresponding author.

E-mail address: schwarcz@mcmail.cis.mcmaster.ca (H.P. Schwarcz).

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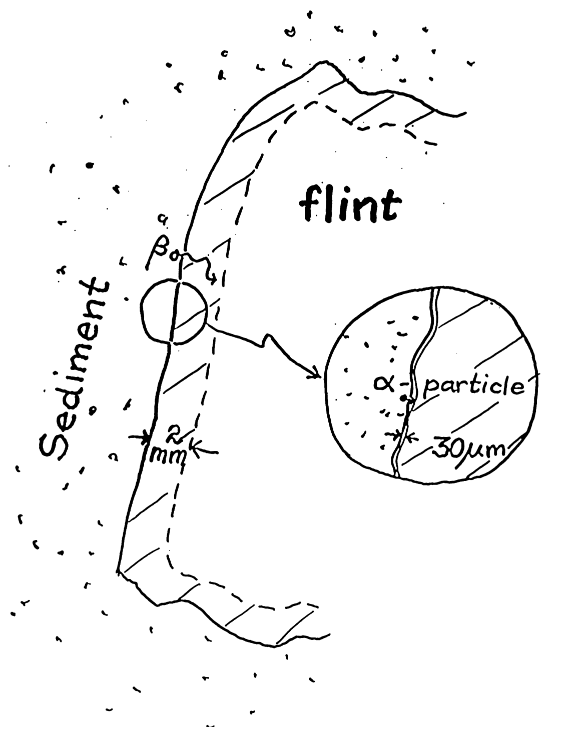

Fig. 1. Schematic plan showing principle of skinflint dating (cartoon). The drawing shows a flint whose outer surface was trimmed by knapping to remove a layer >2 mm thick, and which was then buried for some time in sediment with a higher β-dose rate than the flint itself. The inset shows that a thin layer of α-dosed material also exists at the surface of the flint, which should be removed before analysis of the β-dosed skin.

growth in the intensity of the E′, Al and OHC signals after irradiation. Although the flint had been stored for geological time periods in the earth, apparently the ESR signals recorded in unburnt artifacts represent a steady-state (and not saturated) level of trap-filling that allows further signal growth if the sample is exposed to a higher dose rate.

We can test the second assumption by considering the average composition (U, Th, K) of flint that has been used in dating, as compared with the average composition of typical fine-grained sediments in which artifacts would be buried at sites. Based on a cursory overview of the chemical compositions of typical flints used for manufacture of artifacts, and typical fine-grained sediments found at archaeological sites, it appears that the U, Th, and K contents of sediment are all substantially higher than that of typical flint. This difference arises because of the presence in such sediments of clay minerals and Fe hydroxides typically enriched in U, Th and K. The difference is such that the full (infinite space) β-dose rate in sediment is about $5\times$ larger than that in flint. The irradiation geometry of the flint surface lowers the effect of this difference by about half but assures that in general there will be a substantial inward gradient in β-dose rate from sediment to flint. Therefore, we would expect a significantly higher accumulated dose in the skin layer of the flint than in its interior.

The third assumption is satisfied as long as we limit our measurements to ESR signals which are not significantly bleached by exposure to sunlight. The possible effect of knapping on these signals has not yet been investigated.

2. Theory of skinflint dating

We assume here that the interior of the flint is separated from the external sediment by a planar surface, but that both sediment and flint extend away from this surface, to a distance which is large with respect to β-attenuation. It is possible in principle to calculate the gradient in dose rate between the skin and interior. For example, we can assume that a layer of flint more than 1 cm thick has been removed from much of the knapped surface, exposing to the burial sediment, a surface of fresh, unaltered flint. For measurement of equivalent dose (D_E) by TL measurement, we must assume that, at the time of burial, the outer layer of the flint was not zeroed by bleaching. If the outer layer of the flint is sufficiently opaque, then below a few tens of μm depth, no significant amount of bleaching would occur. This requirement appears to be satisfied by some but not all types of flint. Use of electron spin resonance (ESR) to measure D_E eliminates this problem since ESR signals are essentially unaffected by exposure to light. Based on these assumptions we could calculate a dose gradient into the flint. In practice, however, it is easier to use the Rosy program (Brennan et al., 1999) to calculate the dose rate in the interior of the flint and in the outer 2 mm-thick layer. The difference between these dose rates we shall refer to as D_{is} . We can then measure D_E in the skin portion of the flint, and in a representative portion of the interior of the flint (outside the range of external β-particles). The difference between these two equivalent doses is called) Δ_{is}. Then the time of preparation and burial of the flint artifact is given by $t=\Delta_{is}/D_{is}$.

Because of the steep gradient in the accumulated β-dose from the outer surface, analysis of as thin a layer as possible would be advantageous. This advantage is, however, offset by two disadvantages. First, a thinner layer would contain less mass per unit area and therefore provide less material for analysis. Second, the thickness of a thinner layer would be more difficult to measure, which would lead to less accurate estimates of the dose rate in the layer.

In principle, we could determine Δ_{is} by measuring D_E in the skin and the interior using the additive dose method, and then taking the difference. In practice, however, this would require the accurate measurement of the difference between two large numbers. We can measure the difference more precisely using a variant of the "slide" method, in which we measure the offset in dose between the additive dose curves for the skin and interior. We must, of course, show that the γ- and β-sensitivities of both the regions are the same. The additive dose curves are obtained using a high dose-rate gamma source (e.g. ^{60}Co). Because the skin is close to saturation, there is some concern that the dose sensitivity will depend on γ-dose rate; this will be investigated in future tests of this method.

The only dose rates that contribute to Δ_{is} are the β-dose rates of the flint and of the sediment immediately adjacent to the surface being dated. One can of course date more than one surface on the same artifact, but for each one a separate sample of the adjacent sediment should be analyzed. The γ-dose to the skin and to the interior of the flint are the same and cancel out of the calculation. The outer 30 μm of the skin layer also contains an excess alpha dose which can be eliminated by removing this layer before ESR or TL analysis. Alternately, if we were to use a thinner skin-layer, we could include the α-dosed portion, but this would require an accurate knowledge of the alpha efficiency of the flint for ESR dosimetry (if ESR were used for determination of D_E).

3. Preliminary data

In order to evaluate the potential utility of this method we tested whether gradients in D_E exist in the skin layers of flint artifacts. An unburned flint tool from Quneitra, Golan Heights, Israel/Syria (sample 2084,) and a heated flint artifact from Laugerie Haute, Dordogne Valley, France (2091), were analyzed by TL and ESR. Using a water-cooled diamond blade, slices were cut normal to the outer, approximately flat surface of each object. A layer 2 mm-thick layer was trimmed from each slice and lightly ground. Particles 100–150 μm were washed in HCl, dried and mounted on platinum planchettes without silicone spray. The TL signal was measured on Risø reader at $5°C\,s^{-1}$ using a Corning 7-59, HA-3 filter combination; Fig. 2 shows the intensity of the 380°C peak. ESR intensities for these samples were measured on a JEOL RE1X spectrometer at 9.44 GHz. The E′ signal ($g = 2.001$ was measured at 0.1 mW power, while the OHC ($g = 2.011$) and peroxy ($g = 2.006$) were measured at 20 mW power. Modulation amplitude was 5 gauss (Fig. 2). All equipment was in the Subdepartment for Quaternary Research, Cambridge University. The data confirm that the intensity of TL signals is higher in the outer 2 mm thick layer of flint samples from both sites, while the *burned* flint sample from Quneitra (Golan Heights, Israel) displayed a *decrease* in ESR intensity towards the skin. This is probably attributable to the incomplete zeroing of the ESR signal in this thin layer of flint during heating. (A. Skinner, 1999 pers. comm.).

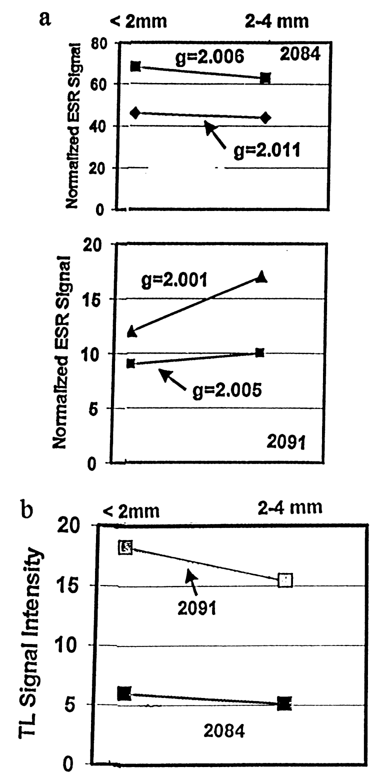

Fig. 2. TL and ESR data for flint artifacts, to demonstrate the existence of a β-dosed skin. Samples: unburned flint from Quneitra, Golan Heights, Israel/Syria (2084) and burned flint from Laugerie Haute, Dordogne Valley, France (2091): (a) ESR analyses (weight-normalized signals) and (b) TL analyses. Samples were taken from the interior of each piece (2–4 mm depth) and from the skin (< 2 mm depth), from approximately plane surfaces.

In the first study, only differences in natural intensities were recorded, and no attempt was made to determine differences in equivalent doses. In a second study, done at McMaster University, we used ESR to determine paleodose differences between skin and interior of an unburned flint artifact from layer F of the Tabun site, Israel (from the collection of the Royal Ontario Museum, Toronto). Using a water-cooled saw, slices were taken normal to a flat surface of the artifact. The outer 2 mm skin was trimmed off with a vibrating tool, and lightly ground in a steel mortar. The E′ signal of sieved fractions was analyzed on a Bruker ER100D ESR spectrometer (0.3 MW, modulation = 100 kHz, scan of 6 mT over 50 s). Between the skin layer (2 mm) and the interior, there was a gradient in intensity of about 8% for coarse flint particles (> 200 μm) while for the finer grained fraction the difference ranged between 30 and 40%. Aliquots of the > 200 μm particles powder were irradiated in the ^{60}Co gamma cell of the McMaster Nuclear Reactor, and analyzed after a delay of one week. The E′ signal which we measured may contain an unstable component (Toyoda and Schwarcz, 1997) which would produce an anomalously large apparent difference in D_E.

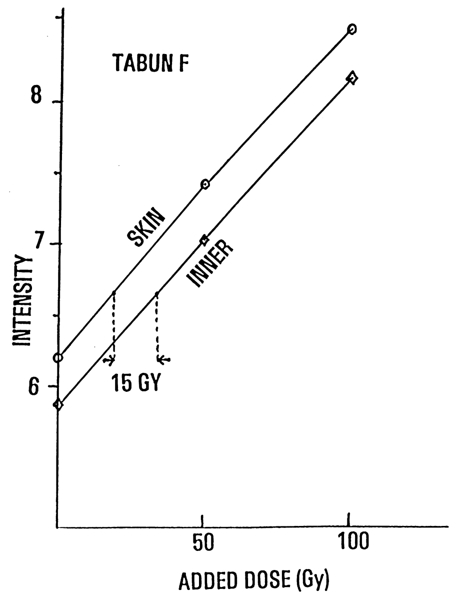

Fig. 3. Growth curves of interior and exterior parts of unburned artifact from Tabun Layer F. A 1 mm thick layer was sliced normal to the surface of the artefact, portions of the interior and of the outer 2 mm layer were removed and homogenized. Aliquots of > 200 μm particles of each of these were given additional doses of γ-rays from a ^{60}Co source. The intensities of the E′ signals were measured and weight normalized. The figure shows that the two growth curves are sensibly parallel, and offset by approximately 15 Gy, which is interpreted to be the difference in D_E between these two portions of the artefact.

Further studies of irradiated flint (J. Bartoll and H.P. Schwarcz, in progress) suggest that any unstable component, if present, makes up $< 10\%$ of the E′ signal intensity. Fig. 3, a growth curve for one of the samples, demonstrates that the first criterion listed earlier is satisfied, namely, that the intensity of the E′ signal increases with added gamma dose. We used the modified "slide" method to determine Δ_{is}, the difference in D_E between the outer 2 mm layer and a sample from the interior of the flint, by graphing growth curves for the interior and the skin samples and visually reading the distance between these lines; this gave a value of approximately 15 Gy (Fig. 3). We have not measured the dose rate in either the sediment for this site, or in the flint. However, we can use average values for the composition of sediment for the overlying layer (XIII) and for typical flint from this layer analyzed by Mercier et al. (1995) to estimate corresponding dose rates. The data are as follows (sediment, flint): U (ppm): 1.99, 0.49; Th (ppm): 4.84, 0.14; K(%): 0.27, 0.05. The typical gradient in radioactivity between sediment and flint is seen here. Using Rosy, we calculated an approximate difference in dose rates between the 2 mm-thick skin and interior, of 47 μGy/a. From this we obtained an apparent age for the skin of 320 ka. For comparison, the immediately overlying layer Ed has been dated at between 230 and 350 ka (Grün et al., 1991; Mercier et al., 1995). This degree of agreement is gratifying, considering the crudeness of the measurements.

4. Conclusions

These preliminary results and the arguments presented above suggest that it should be possible to estimate the ages of knapped flint artifacts on the basis of the excess β-dose absorbed in their skins as a result of the higher β-dose rate of sediment compared to flint. The same approach could be used, in principle, to date any stone object whose external surface was ground away during some geological process. For example, spheroidal pebbles in glacial till are assumed to have had a part of their outer surface eroded (ground) away during ice-driven transport. We could determine the time of last motion of an ice sheet by dating the skin on such an object. As noted above, analysis of thinner layers could increase the sensitivity of the method due to the steep gradient in β- and α-dose rates at the surface of the flint. This advantage is partially offset, however, by possible inaccuracies in measurement of the thickness of the layer removed, and uncertainty in alpha efficiency for ESR dating.

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