

---

# Advantages and Limitations of Thermoluminescence Dating of Heated Flint from Paleolithic Sites

---

**Daniel Richter\***

*Department of Human Evolution, MPI for Evolutionary Anthropology, Deutscher Platz 6, 04103 Leipzig, Germany*

Thermoluminescence (TL) dating is now widely used in the age determination of Paleolithic sites. Although the basic principle of TL-dating is simple, the underlying assumptions are not trivial. One major source of error is the external dose rate, which contributes to a varying degree to the denominator of the age formula and thus has a varying influence on the dating result. The intention of this paper is to enable the user to evaluate TL age determinations of heated flint. The parameters used for age determination and some of their relationships are discussed. It is shown that the reliability of TL results of heated flint depends on the proportion of the various dose-rate parameters and that these are important for the evaluation of ages. The limitations of the method as well as the advantages are discussed. TL-dating results for two Near Eastern Paleolithic sites (Rosh Ein Mor and Jerf al-Ajla) are discussed as examples. © 2007 Wiley Periodicals, Inc.

## INTRODUCTION

The validity and power of thermoluminescence (TL) dating of burnt flint, quartz, quartzite, sandstone, chert, etc., has been shown in a variety of publications (Valladas, 1992; Valladas et al., 1991; Mercier et al., 1995a; Huxtable, 1993). The application ranges from Lower Paleolithic to Neolithic archaeological sites, with a major focus on the Middle Paleolithic, which is often beyond the range of the  $^{14}\text{C}$ -dating method. The application of TL dating on heated flint is mainly limited by the detection limits of the equipment for very young samples, the saturation of the signal measured for very old samples, and, of course, the presence of sufficiently burnt flint. It is thus, in principle, possible to date fire use over the entire human evolution. Recent reviews of luminescence dating in archaeology can be found in Roberts (1997) and Wagner (1998).

The advantages and disadvantages of TL dating of heated flint will be discussed here and special emphasis is put on potential problems and the underlying assumptions. This discussion omits the fundamental physics of the method and concentrates more on the application of this dating method.

---

\*E-mail: drichter@eva.mpg.de

## THERMOLUMINESCENCE DATING

Thermoluminescence dating of a heated flint determines the time elapsed since the last incidence of firing. In contrast to many other chronometric dating methods, it is thus possible to directly date a past human activity. Naturally occurring fires are unlikely to be responsible for heating in most Paleolithic sites (see also Alpers-Afil et al., 2007), and the penetration depth of fire in sediment is very low (Bellomo, 1994).

### Basic Principles

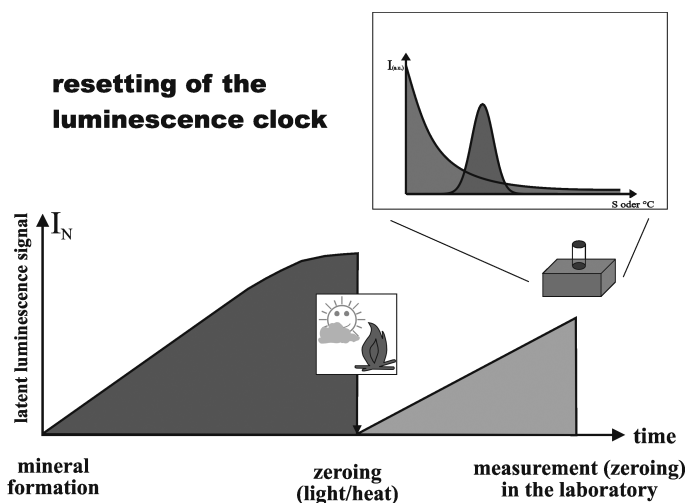
The principle of luminescence methods has been described in detail elsewhere (Aitken, 1985, 1998; Wagner, 1998; Bøtter-Jensen et al., 2003). Therefore, only a summary and a simple description of the principles are given here, with an emphasis on the issues relevant for users and for the evaluation of luminescence ages.

Luminescence dating is based on structural damage and faults to the crystal lattice of minerals by ionizing radiation. The sources for this omnipresent radiation are radioactive nuclides from the surrounding sediment and from the sample itself, as well as secondary cosmic rays. Thus a radiation dose (paleodose, or  $P$ ) accumulates in the crystal in the form of electrons in excited states, of which some are metastable and thus resident over periods of time long enough to allow a dating application. The paleodose is proportional to the dose rate ( $\dot{D}$ ), which is the ionizing radiation per time unit at the position of the sample within the sediment. This dose rate provides the clock for the dating application. Exposure to light or temperature causes the electrons to relax to a ground state, sometimes by emitting a photon, the luminescence. If the temperature is high enough (>ca. 400° C), the drainage is sufficient to relax all electrons relevant to the luminescence method used; that is, the clock is set to zero. The intensity of the luminescence signal (number of photons) increases with the total absorbed dose ( $P$ ) in a crystal and is therefore a function of exposure time to radiation.

This accumulation starts with the formation of the mineral. But in most geoarchaeological applications, the interest lies in the time elapsed since human activity (e.g., fire) or an event related to the human activity (e.g., sedimentation in case of Optically Stimulated Luminescence [OSL] dating). Therefore, the mineral had to be either heated or exposed to light at the time of interest in antiquity. After deposition and protection from light, a radiation dose, and thus a latent luminescence signal, accumulates again until it is measured in the laboratory (Figure 1). The age formula therefore is straight forward and simple,

$$\text{age} = \frac{\text{paleodose}}{\text{dose rate}} = \frac{P_{(\text{Gy})}}{\dot{D}_{(\text{Gy}\cdot\text{a}^{-1})}},$$

where the paleodose  $P$  is expressed in Gy and the dose rate  $\dot{D}$  in Gy per time unit (usually in ka).



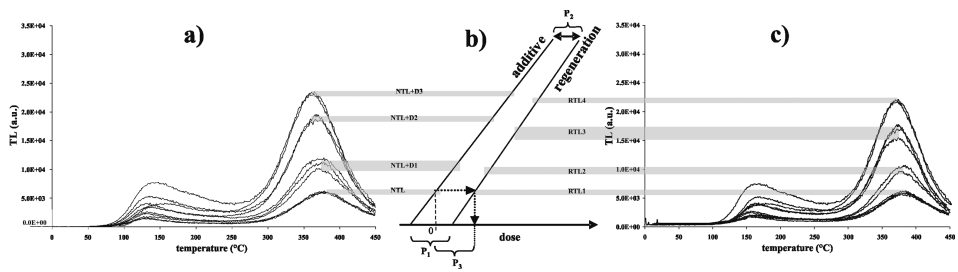
**Figure 1.** Latent luminescence signal since mineral formation until it is zeroed by exposure to heat or light in antiquity. After burial, the luminescence signal accumulates again and the sample is measured in the laboratory.

The numerator and denominator of this formula consist of several parameters, each of which will be discussed in more detail.

### The Paleo- (or Absorbed-) Dose

The absorbed dose is commonly denominated paleodose in TL dating. This paleodose is determined from the TL signal, which is measured by heating sample aliquots at a constant rate, producing the glow curves (Figure 2a and 2c). Three different types of glow curves have to be distinguished: the natural thermoluminescence (NTL) of the sample as it is (Figure 2a); the additive glow curve (NTL+ or ATL), where a radiation dose with a calibrated radioactive source is given in addition to the natural one (Figure 2a); and the regenerated signal (RTL), where the sample has been zeroed by heating and then given an artificial radiation dose (Figure 2c). The goal is to construct and reconstruct the growth of the luminescence signal with increasing radiation doses (Figure 2a–c). The resulting dose growth curves (Figure 2b) are measures of the sensitivity of a sample to ionizing radiation, consisting of several dose points, and are used to determine the paleodose.

There are essentially two ways to determine  $P$  in TL dating of heated flint (Figure 2b). Both are based on the construction of an additive (NTL and several NTL+), as well as a regeneration (several RTL) dose growth curve, where the latter is obtained on separate material which has been heated (zeroed). They differ in the way the paleodose is determined. The standard method (Aitken, 1985) performs regression analyses for both growth curves and the sum of their absolute values essentially provide,  $P$  ( $P_1$  in Figure 2b). With the normalization method (Valladas & Gillot, 1978; Valladas, 1992; Mercier, 1991), one of the two growth curves is shifted towards the other until they are matched, and the amount of the shift essentially



**Figure 2.** Glow curves and growth curves used in TL dating: (a) additive (NTL and NTL+) glow curves; (b) additive and regeneration growth curve; and (c) regeneration glow curves (RTL). In Figure 2b, the determination of the paleodose ( $P$ ) is shown, where  $P_1$  is the sum of the regression analysis of the additive and the regeneration dose curves (standard method);  $P_2$  is determined by shifting one dose curve toward the other (normalization method); and  $P_3$  is matching the NTL signal (arrows) to the regeneration dose curve (SAR method).

gives  $P$  ( $P_2$  in Figure 2b). The first method can be applied only to younger samples whose dose points are still in the quasi-linear region of the growth curve, whereas the second method is used for older samples that are at the onset of saturation of the TL signal.

A third method has been proposed recently, which employs orange-red luminescence emission, instead of the UV-blue (Richter & Krbešček, 2006; Richter & Temming, 2006). In this method a single-aliquot-regeneration (SAR) protocol with only two regeneration points (Figure 2c) is used. The NTL (Figure 2a) is simply matched to these levels in RTL luminescence in order to determine the paleodose ( $P_3$  in Figure 2b). This method is especially useful for samples too small for the other methods, as it requires little material for analysis. But the two established methods described earlier are the methods of choice, at least until this new approach has been shown to be generally applicable to a wider variety of different materials. Nevertheless, it seems to have potential and opens the possibility of dating more Paleolithic sites, previously considered not to be datable by TL on heated flint.

Common to all three approaches is the necessity to measure the effectiveness of alpha radiation, which has to be determined for each individual sample due to the high variability in sensitivity of flint samples. This is achieved by comparing the TL response of finely grained material to  $\alpha$  and  $\beta$  radiation, either by an additive procedure or by comparison in freshly zeroed material.

### Heating and Bleaching

Before performing a full dating analysis, each sample must be checked to determine whether the heating in antiquity was sufficiently high for dating purposes. Samples are selected according to macroscopic signs (reddish or pink color, glossy scars or glossy [luster] surface, crazing, pot lids, or cracked faces) that indicate heating (e.g., Julig et al., 1999). The sufficiency (ca. 400°C) of the heating is checked by the heating plateau test (Aitken, 1985), where the ratio of NTL over NTL+ is required to be constant for the temperature range of the TL peak (ca. 370°C), which

indicates the zeroing of the TL signal in antiquity. Additionally, the shape and temperature of the glow peak can provide evidence of sufficiency of ancient heating (e.g., Michab et al., 1998; Richter et al., 2002).

Exposure to light can have a similar zeroing (bleaching) effect. Although there is little evidence of sensitivity to bleaching of the high-temperature TL peak used in dating heated flint (Huxtable and Aitken, 1985; Valladas, 1985a), care has to be taken with translucent samples, which might have been bleached during or after excavation. Such bleaching would give rise to severe age underestimation. It is, therefore, necessary in some cases to check whether the TL-peak used is prone to bleaching (e.g., Alpers-Afil et al., in press). It is advisable not to expose potential samples to direct sunlight, and, as a precaution, samples should be wrapped in aluminium foil soon after discovery.

### The Dose Rate ( $\dot{D}$ )

The denominator  $\dot{D}$  of the age formula consists of two independent parameters, the internal ( $\dot{D}_{\text{internal}}$ ) and the external dose rate ( $\dot{D}_{\text{external}}$ ), which will be discussed separately.

$$\text{age} = \frac{P_{(\text{Gy})}}{\dot{D}_{(\text{Gy}\cdot\text{a}^{-1})}} = \frac{P}{\dot{D}_{\text{internal}} + \dot{D}_{\text{external}}}$$

Obviously, the denominator is crucial for the accurate determination of an age. Any variability through time will produce a different age. The assumption of the constancy of the dose rate ( $\dot{D}$ ) with time can be made only within the resolution of the dating method itself. Nevertheless, it is sometimes necessary to model  $\dot{D}$ . The effect on the accuracy and precision on the dating result depends on the proportional contribution of each parameter to the total  $\dot{D}$ . These issues will be discussed below.

#### *Internal dose rate*

All rock material contains radioactive elements that give rise to an internal dose rate ( $\dot{D}_{\text{internal}}$ ). Elements of concern here are only U, Th, K, and to some extent Rb (Aitken, 1985), because other natural radioactive nuclides occur only in very small quantities or do not contribute significantly to the total absorbed dose ( $\dot{D}$ ).

$\dot{D}_{\text{internal}}$  consists of three parameters related to the  $\alpha$ -,  $\beta$ - and  $\gamma$ -radiation, where the latter is usually small in most cases.

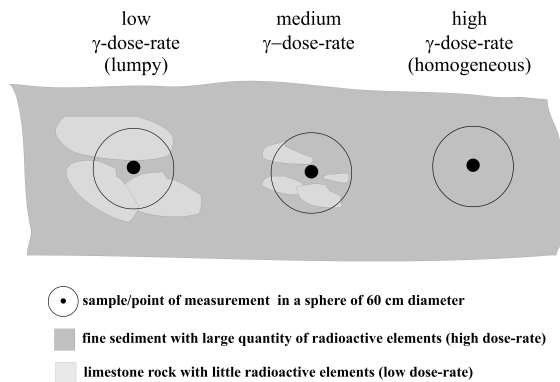
$$\text{age} = \frac{P_{(\text{Gy})}}{\dot{D}_{(\text{Gy}\cdot\text{a}^{-1})}} = \frac{P}{(\dot{D}_{\alpha} + \dot{D}_{\beta} + \dot{D}_{\gamma}) + \dot{D}_{\text{external}}}$$

Because all cortex or patinated parts are removed, the samples can be regarded as geochemical stable. The  $\dot{D}_{\text{internal}}$  thus is considered as being constant over the time span of interest in archaeological dating. To take the different TL efficiency of alpha

radiation into account, a conversion factor (expressed as a-value, b-value, or S-alpha) has to be determined for each sample as part of the internal dose rate (see above). The  $\dot{D}_{\text{internal}}$  dose rates for each sample are calculated from the element concentrations (Adamiec and Aitken, 1998), which are usually determined by neutron activation analysis (NAA) from the crushed unprepared sample. Fission track analysis has shown the presence of hot spots of radioactive elements in a few samples (Valladas, 1985b). But due to crushing, the sample is homogenized, and it is assumed that any inhomogeneity of radioactive element distribution is thus negligible (Valladas, 1985b).

*External dose rate*

Sediment contains not only the flint samples, but radioactive nuclides as well. These give rise to an external dose rate in addition to the one from secondary cosmic rays ( $\dot{D}_{\text{cosmic}}$ ). Although the latter have penetration depths in rock or sediment of up to several tens of meters, the range for the ionizing radiation from the radioactive nuclides is much smaller. Whereas  $\alpha$ -particles penetrate objects only a few  $\mu\text{m}$ , the range of  $\beta$ -radiation is about 2 mm. The samples receive a  $\gamma$ -dose from the surrounding sediment of a sphere 60 cm in diameter, because the maximum range of  $\gamma$ -radiation is approximately 30 cm. This is of special concern because this dose rate has to be measured in the field, e.g., by dosimeters or a portable  $\gamma$ -spectrometer, for sediments from most Paleolithic sites. A number of measurements are required because the  $\gamma$ -dose rate depends on the composition of the 60 cm sediment sphere around the point of measurement or sample (Figure 3). This dose rate may vary to some degree within a sedimentological unit throughout a site (e.g., Brennan et al., 1997). Especially in caves in limestone regions, areas with large rocks tend to have a lower dose rate than areas where fine-grained material dominates the composition of the sediment (Figure 3). As the measurement cannot be performed at the exact location of each sample (for obvious reasons), any modeling after excavation is not only tedious but also prone to error. Nevertheless, modeling might be necessary if all sediments have been removed (e.g., Guibert et al., 1998). It is advisable to



**Figure 3.** Spatial variation of  $\gamma$ -radiation levels at sample position or point of measurement due to differing sediment composition of varying portions of rock boulders, stones, and fine sediment. This leads to high dose rate, homogeneous radiation fields or low dose rate, lumpy radiation environments.

measure the  $\gamma$ -dose rate at several locations throughout the site and use the average result as representative value, which also provides a measure of the variation within one given site due to the inhomogeneity (lumpiness) of a site. This approach is only valid for dating multiple samples, because it is assumed that the average  $\gamma$ -dose rate for the locations of the samples is similar to the average  $\gamma$ -dose rate of the measurement positions.

To eliminate or minimize the need for modeling the  $\beta$ -dose rates from the surrounding sediment ( $\dot{D}_{\beta\text{-external}}$ ), each sample is carefully stripped of its outer 2 mm surface with a water-cooled diamond saw prior to analysis. The external dose rate in TL dating of flint thus consists solely of the  $\gamma$ -dose rate ( $\dot{D}_{\gamma\text{-external}}$ ) of the sediment and the cosmic dose rate ( $\dot{D}_{\text{cosmic}}$ ).

$$\text{age} = \frac{P_{(\text{Gy})}}{\dot{D}_{(\text{Gy}\cdot\text{a}^{-1})}} = \frac{P}{\dot{D}_{\text{internal}} + (\dot{D}_{\text{cosmic}} + \dot{D}_{\gamma\text{-external}})}$$

$\dot{D}_{\text{cosmic}}$  depends on the burial depth, and because secondary cosmic radiation does not fluctuate on a significant level over the time range of approximately 500 ka (Prescott and Hutton, 1994), it can be fairly assumed to be constant. Nevertheless, care has to be taken for estimating or modeling the correct cosmic dose rate in complex geometries like caves or rockshelters in valleys, where  $\dot{D}_{\text{cosmic}}$  can become important. The  $\dot{D}_{\gamma\text{-external}}$  is a function of the mineral composition of the sediment and its moisture content. Due to the mobility of some radioactive elements, notably from the  $^{238}\text{U}$ -decay chain, the constancy of  $\dot{D}_{\gamma\text{-external}}$  can not be assumed *a priori*. High resolution  $\gamma$ -spectrometry and  $\alpha$ -spectroscopy on sediment samples can be used to determine and identify disequilibria in the  $^{238}\text{U}$ -chain and to facilitate modeling of  $\dot{D}_{\gamma\text{-external}}$ , but the detection of disequilibria may be limited and depends on the half-life of the mobile isotope (Olley et al., 1996). Furthermore, it can be reoccurring. But the absence of a measurable disequilibrium does not necessarily imply the constancy of  $\dot{D}_{\gamma\text{-external}}$  because the chain could be in equilibrium again, depending on the half-life of the isotope responsible for a temporary disequilibrium (e.g., Mercier et al., 1995b). Additionally, sediment moisture fluctuates with season and climate; thus the  $\gamma$ -dose rate varies, because water absorbs  $\gamma$ -rays. The scale of such fluctuations has to be assumed based on independent paleoclimatological data, and its uncertainty should be accounted for by an estimated error term for  $\dot{D}_{\gamma\text{-external}}$  larger than the measured uncertainty.

## DOSE RATE VARIATION AND ITS EFFECT ON AGE

Luminescence methods depend to a large degree on the geological nature of a site. Therefore, site formation processes and post-depositional disturbances should be taken into account. TL dating of heated rocks has an advantage over most other dosimetric dating methods in that at least some part of the dose rate ( $\dot{D}_{\text{internal}}$ ) can be considered to be constant over the time of interest. Its proportional contribution to the total dose rate determines the effect of any variation in the dose-rate modeling. TL dating of heated flint can be almost independent from the external dose rate and

**Table I.** Effect of the variation of the external dose rate ( $\dot{D}_{\text{external}}$ ) on extreme ratios of  $\dot{D}_{\text{external}}$  to  $\dot{D}_{\text{internal}}$  on the age (expressed as % of the true age). Doubling or cutting in half of  $\dot{D}_{\text{external}}$  has little effect when the  $\dot{D}_{\text{internal}}$  is high, but a large effect on the age when  $\dot{D}_{\text{internal}}$  is low. (Note that zero cosmic dose is assumed here for ease of comparison).

$\dot{D}_{\text{internal}} (\% \dot{D}_{\text{total}})$	$\dot{D}_{\text{external}} (\% \dot{D}_{\text{total}})$	$0.5 \dot{D}_{\text{external}} (\% \text{ age})$	$2 \dot{D}_{\text{external}} (\% \text{ age})$
90	10	105	91
10	90	182	53

thus from the uncertainty of fluctuations, if  $\dot{D}_{\text{internal}}$  is very high. In such cases, the fractional contribution of  $\dot{D}_{\gamma\text{-external}}$  to  $\dot{D}_{\text{total}}$  is small, and any variation of the  $\dot{D}_{\gamma\text{-external}}$  would thus be negligible.

In an extreme example such as sample JA-1 (Richter et al., 2002),  $\dot{D}_{\text{internal}}$  makes up 92% of the total dose rate. This gives great confidence in the dating result because any variation of the  $\dot{D}_{\gamma\text{-external}}$  has little effect on the resulting age. Table I gives values for changes in the resulting age (in percentage of the “true” age) for a hypothetical case where  $\dot{D}_{\gamma\text{-external}}$  is cut in half or doubled in order to model  $\dot{D}$ . For high  $\dot{D}_{\text{internal}}$  samples, there is little effect on the age, even for an extreme scenario where the external dose rate is doubled; this contrasts to the case where the dependency on the external dose rate is high (low value of  $\dot{D}_{\text{internal}}$ ), for which large scale changes result in ages different by almost a factor of two. The influence of any variation in the external  $\gamma$ -dose rate ( $\dot{D}_{\gamma\text{-external}}$ ) on the age is proportional to its fraction of the total dose rate ( $\dot{D}$ ).

The relative proportions of the three components of the dose rate ( $\dot{D}$ ) vary from site to site and from sample to sample. The precision and accuracy of thermoluminescence dating results are highly dependent on these proportions (e.g., Deckers et al., 2005).

## EXAMPLES

The influence of the parameters described above will be discussed for two Near Eastern Paleolithic sites for which the author has performed the TL analysis: Rosh Ein Mor (Israel) and Jerf al-Ajla (Syria).

### Rosh Ein Mor

The Early Levantine Mousterian site of Rosh Ein Mor is located at the edge above the Nahal Zin canyon in a shallow depression of a lower terrace. Four sedimentological units of silt and clay have been differentiated on the basis of their colors. The archaeological remains are independent of these units, as they consist of an 80 cm thick deposit made up almost entirely of flint artifacts with little fine-grained material in between. Sedimentological analysis revealed the presence of fluvial and slope wash sediments, but no correlation was found with the units. Although no vertical or horizontal sorting was observed, a distinct spatial distribution of certain artifact types over the 45 m<sup>2</sup> excavated area was found (Marks, 1977). Many artifacts below 30 cm depth

**Table II.** Percentage contribution of the external ( $\dot{D}_{\text{external}}$ ) and internal dose rate ( $\dot{D}_{\text{internal}}$ ) to the total dose ( $\dot{D}_{\text{total}}$ ) of the flint samples from Rosh Ein Mor. (Note that  $\dot{D}_{\text{external}}$  includes a small cosmic dose that could be considered as stable as well) (after Rink et al., 2003). The temperature of the natural TL peak at  $5^\circ \text{C s}^{-1}$  is given, as well as the resulting ages.

Lab. No. (FL-D15)	$\dot{D}_{\text{external}}$ (% $\dot{D}_{\text{total}}$ )	$\dot{D}_{\text{internal}}$ (% $\dot{D}_{\text{total}}$ )	Peak ( $^\circ\text{C}$ )	Age (ka)
18	30	70	360	$35.3 \pm 3.5$
32	11	89	350	$14.2 \pm 1.4$
36	49	51	370	$47.7 \pm 5.6$
38	31	69	360	$32.8 \pm 3.6$
5	21	79	360	$24.5 \pm 3.1$

showed heavy patination and edge damage. Thermoluminescence dating was performed on five samples of heated flint (Rink et al., 2003). The spread of the resulting ages is enormous, ranging from 14 to 48 ka (Table II). There is no correlation between age and vertical or horizontal distribution. However, a relationship can be observed for increasing  $\dot{D}_{\gamma\text{-external}}$ , which gives increasing age results. Sample FL-D15-32 is a clear outlier, with a high internal dose rate, which actually makes this age result more reliable than the others because of its high stable internal dose rate. Nevertheless, it obviously received a different  $\dot{D}_{\text{external}}$  than the others; thus, it has to be suspected that its heating belongs to a separate event in antiquity. As the other samples do not seem to have received similar  $\dot{D}_{\text{external}}$  it must be suspected that these samples do not belong to a common heating event either. Heating at different times would have resulted in different doses. Given the depositional environment, it is quite possible that deflation occurred, which would make several independent heating events possible. These could have been different occupations with the samples related to these events. Equally possible are later heating events, where the actual deposition of a sample is unrelated to the heating. Additionally, near-surface or exposed artifacts could have been heated more than once. This could have led to repeated full or partial zeroing, which is not distinguishable by standard TL-dating measurements.

Being close to or at the surface would also reduce the gamma dose rate significantly, which probably would not be fully compensated for by the increased cosmic dose rate ( $\dot{D}_{\text{cosmic}}$ ). The samples would have received a smaller dose from a temporarily lower  $\dot{D}_{\gamma\text{-external}}$ , but the full present-day dose rate is used for age calculation, which would lead to erroneously young ages. Furthermore, surface exposure could have led to a heating of the samples by the sun. Surface temperatures of black rocks in desert environments have been measured to reach up to  $73^\circ \text{C}$  (Warke, 2000). The resulting average temperature over the burial history thus would have been much higher than generally assumed for well-buried samples. This leads to a much reduced time of residence of the excited electrons, commonly referred to as lifetime for a TL peak of a certain temperature. Calculations based on minimum lifetimes after Aitken and Wintle (1977) show a reduction to only 120 ka of the lifetime of the  $370^\circ \text{C}$  peak at a temperature of  $70^\circ \text{C}$ . Sun bleaching of the TL signal used for dating is generally not considered to be a problem for such dark flint, but it cannot be entirely ruled out because of the high variability of the flint material in general. Signal bleaching and heating both caused by surface exposure therefore could be responsible for some

underestimation of TL ages of flint. Although the underlying data for such calculations is very thin (the values are minimum estimates except for the maximum ambient temperature), there are indications that sun exposure might be a problem at Rosh Ein Mor, because the sample with the lowest age result (FL-D15-32) has its TL peak at the lowest temperature (350°C), whereas the one for the oldest (FL-D15-32) is at the highest (370°) peak temperature of all samples.

Obviously the results do not belong to one statistical group. Due to the suspected reheating, all TL ages were discarded in the final analysis of the age of the site (Rink et al., 2003).

### Jerf Al-Ajla

A Late Mousterian Layer (C1) from Jerf al-Ajla cave was dated by means of TL on eight heated flint artifacts (Richter et al., 2002). There are typological and technological indications of the presence of Middle as well as Upper Paleolithic elements. The assemblage is comparable to layers IIBase and III2a at the open-air site of Um el Tlell (Syria) not far away. These layers at Um el Tlell have been dated to  $36.0 \pm 2.5$  ka by TL of heated flint artifacts (Boëda et al., 1996). The spread of age results (Table III) at Jerf al-Ajla is clearly smaller than the previous example. The external dose rate is small in all cases, amounting to a maximum of only 22% of the total dose rate for some of these samples (Table III). Low-level gamma spectrometry of the fine-grained component of the sediment revealed no disequilibrium in the U or Th chains (Richter et al., 2002).

The high internal dose rates ( $\dot{D}_{\text{internal}}$ ) provide great confidence in the age results because any undetected change in the gamma dose rate would lead to only a minor change in the ages. For example, a doubling of the external dose rate of sample JA-2, which gives the largest age, would result in an age of  $35.2 \pm 6.1$  ka, which is statistically indistinguishable from its original  $39.1 \pm 6.4$  ka (Table III). The ages clearly belong to the same statistical distribution, and a weighted mean can be calculated, giving an age for layer C1 of  $33.2 \pm 2.3$  ka, which is in good agreement to the results obtained for Um el Tlell.

**Table III.** Percentage contribution of the external ( $\dot{D}_{\text{external}}$ ) and internal dose rate ( $\dot{D}_{\text{internal}}$ ) to the total dose ( $\dot{D}_{\text{total}}$ ) of the flint samples from Jerf al-Ajla. (Note that  $\dot{D}_{\text{external}}$  includes a small cosmic dose that could be considered as stable as well) (after Richter et al., 2002).

Lab. No.	$\dot{D}_{\text{external}}$ (% $\dot{D}_{\text{total}}$ )	$\dot{D}_{\text{internal}}$ (% $\dot{D}_{\text{total}}$ )	Age (ka)
JA-1	8	92	$28.6 \pm 3.2$
JA-2	16	84	$39.1 \pm 6.4$
JA-3	22	77	$33.8 \pm 4.8$
JA-7	10	90	$34.3 \pm 5.0$
JA-8	18	82	$31.5 \pm 4.2$
JA-87	16	84	$32.2 \pm 3.2$
JA-324	20	80	$37.3 \pm 4.5$
JA-435	20	80	$38.1 \pm 4.1$

## DISCUSSION

All dosimetric dating methods are dependent on the sites' environment. Therefore, they are prone to error due to variation of the environment. The degree of influence of environmental parameters on the age result depends on the proportion of the varied parameter to the sum of all parameters. This has to be taken into consideration and evaluated for all results. Of all dosimetric methods, TL on heated rock material is the least sensitive, due to the stable internal dose rate in all samples. Little or no internal dose rates are present for materials used in OSL dating of sediment. In addition to the problems arising from the aforementioned variability, OSL dating suffers from potential problems in the completeness of the zeroing of the signal and the possible incorporation of sediments of different ages.

The method of Infrared Stimulated Luminescence (IRSL) dating of the K-feldspar component of sediment has a small stable internal dose rate like that in flint, but it is quite often affected by an anomalous loss of signal, which results in severe underestimation.

Electron Spin Resonance (ESR) dating suffers additionally from the uncertainties of the uptake history of U into the sample with time, whereas U-series dating of secondary carbonates require the assumption of the presence of a closed system.

The most frequently used method of radiocarbon ( $^{14}\text{C}$ ) dating provides ages for samples for which association with past human activity can be questioned quite frequently. Furthermore, the provided time scale is not linear, and the results have to be calibrated, for which an agreed method exists only up to about 20 ka. A frequent criticism of TL dating relates to the large uncertainties obtained, but the overall error of weighted average TL dating results is similar to calibrated single  $^{14}\text{C}$  data. Multiple measurements by the latter method, however, cannot be averaged after the statistical process of calibration of the individual results, thus leading to fairly large age estimate ranges for multiple dating.

## CONCLUSIONS

The accuracy of any chronometric dating of an archaeological site is dependent most of all on the relation of the sample to the archaeological event (association), but also on the depositional environment and the quality of the samples. The precision of a dating result depends on the latter two and on the method used. All dating methods require a number of assumptions to be made, and these need to be evaluated carefully for each individual site.

Thermoluminescence dating of heated flint is a useful tool to establish the time elapsed since the last heating of an object. The advantage of TL over other methods is not only the direct association of the event with past human activity on a linear time scale, but also its smaller vulnerability to unknown variation of certain parameters. Nevertheless great care has to be taken in the evaluation of TL dates, and certain standards need to be met when publishing dating results. On the one hand, these include the presentation of glow curves, heating- and  $D_E$ -plateaus, growth curve(s) (including correction for supralinearity) and the determination of the alpha sensitivity of each sample. On the other hand, equal care has to be taken in the evaluation of

parameters prone to variation with time. TL-dating results with large external dose components have to be evaluated critically, and great care has to be taken when considering the models used and errors associated with this component.  $\gamma$ - or  $\alpha$ -spectrometry should be performed on sediment samples in order to obtain at least the information on the state of the equilibrium of the U-chain for the more recent history of the external radiation field ( $\dot{D}_{\gamma\text{-external}}$ ). At many sites, however, the sum of the two constant dose rates ( $\dot{D}_{\text{internal}}$  and  $\dot{D}_{\text{cosmic}}$ ) contribute to a large extent to the total dose rate, which makes such dating results less vulnerable to variations and provide confidence in TL dating as a powerful tool in dating of Paleolithic sites.

I am grateful to all the colleagues and excavators for their contributions to this paper through discussion during my fieldwork over the recent years. I am also indebted to the two anonymous reviewers, whose comments and suggestions considerably improved the paper.

## REFERENCES

- Adamiec, G., & Aitken, M.J. (1998). Dose-rate conversion factors: Update. *Ancient TL*, 16, 37–50.
- Aitken, M.J. (1985). *Thermoluminescence dating*. London: Academic Press.
- Aitken, M.J. (1998). *An introduction to optically dating*. Oxford: Oxford University Press.
- Aitken, M.J., & Wintle, A.G. (1977). Thermoluminescence dating of calcite and burned flint: The age relation for slices. *Archaeometry*, 19, 100–105.
- Alpers-Afil, N., Richter, D., & Goren-Inbar, N. (2007). Phantom hearths and controlled use of fire at Gesher Benot Ya'Aqov, Israel. *Paleoanthropology*, 1–15.
- Bellomo, R.V. (1994). Methods of determining early hominid behavioural activities associated with the controlled use of fire at FxJj 20 Main, Koobi Fora, Kenya. *Journal of Human Evolution*, 27, 173–195.
- Boëda, E., Connan, J., Dessort, D., Muhesen, S., Mercier, N., Valladas, H., & Tisnérat, N. (1996). Bitumen as a hafting material on Middle Palaeolithic artifacts. *Nature*, 380, 336–338.
- Bøtter-Jensen, L., McKeever, S.W.S., & Wintle, A.G. (2003). *Optically stimulated luminescence dosimetry*. Amsterdam: Elsevier.
- Brennan, B.J., Schwarcz, H.P., & Rink, W.J. (1997). Simulation of the gamma radiation field in lumpy environments. *Radiation Measurements*, 27, 299–305.
- Deckers, K., Sanderson, D.C.W., & Spencer, J.Q. (2005). Thermoluminescence screening of non-diagnostic sherds from stream sediments to obtain a preliminary alluvial chronology: An example from Cyprus. *Geoarchaeology*, 20, 67–77.
- Guibert, P., Bechtel, F., Schvoerer, M., Müller, P., & Balescu, S. (1998). A new method for gamma dose-rate estimation of heterogeneous media in TL dating. *Radiation Measurements*, 29, 561–572.
- Huxtable, J. (1993). Further thermoluminescence dates for burnt flints from Maastricht-Belvédère and a finalised thermoluminescence age for the unit IV Middle Palaeolithic sites. *Mededelingen Rijks Geologische Dienst*, 47, 41–44.
- Huxtable, J., & Aitken, M.J. (1985). Thermoluminescence dating results for the Palaeolithic site Maastricht Belvédère. In T. van Kolfschoten & W. Roebroeks (Eds.), *Maastricht Belvédère: Stratigraphy, palaeoenvironment and archaeology of the Middle and Late Pleistocene deposits*. *Analecta Praehistoria Leidensia*, 18, 41–44.
- Julig, P.J., Long, D.G.F., Schroeder, B., Rink, W.J., Richter, D., & Schwarcz, H.P. (1999). Geoarchaeology and new research at Jerf al-Ajla Cave, Syria. *Geoarchaeology*, 14, 821–848.
- Marks, A. E. (1977). Introduction: a preliminary overview of Central Negev Prehistory. In A.E. Marks (Ed.), *Prehistory and Palaeoenvironments in the Central Negev Israel*, vol. II, The Avdat/Aqev area, part 2, and the Har Harif. Department of Anthropology (pp. 3–34). Dallas: Southern Methodist University Press.
- Mercier, N. (1991). Flint palaeodose determination at the onset of saturation. *Nuclear Tracks and Radiation Measurements*, 18, 77–79.

- Mercier, N., Valladas, H., & Valladas, G. (1995a). Flint thermoluminescence dates from the CFR laboratory at Gif: Contributions to the study of the chronology of the Middle Palaeolithic. *Quaternary Science Reviews*, 14, 351–364.
- Mercier, N., Valladas, H., & Valladas, G. (1995b). Flint thermoluminescence dates from the CFR laboratory at Gif: Contributions to the study of the chronology of the Middle Palaeolithic. *Quaternary Science Reviews*, 14, 351–364.
- Michab, M., Valladas, H., Forget, L., & Mercier, N. (1998). Distinguishing burnt from partly bleached unburnt quartz pebbles of Pedra Furada, Brazil. *Ancient TL*, 16, 5–9.
- Olley, J.M., Murray, A.S., & Roberts, R.G. (1996). The effects of disequilibria in the uranium and thorium decay chain on burial dose rates in fluvial sediments. *Quaternary Science Reviews*, 15, 751–760.
- Prescott, J.R., & Hutton, J.T. (1994). Cosmic ray contributions to dose-rates for luminescence and ESR dating: large depths and long term variations. *Radiation Measurements*, 23, 497–500.
- Richter, D., Schroeder, B., Rink, W.J., Julig, P.J., Long, D.G.F., & Schwarcz, H.P. (2002). The Middle to Upper Palaeolithic transition in the Levant and new TL dates for a Late Mousterian assemblage from Jerf al-Ajla Cave near Palmyra, Syria. *Paléorient*, 27, 29–46.
- Richter, D., & Krbetschek, M. (2006). A new thermoluminescence dating method for heated flint. *Archaeometry*, 48, 695–705.
- Richter, D., & Temming, H. (2006). Dose recovery tests for heated flint. Testing equivalent dose protocols. *Radiation Measurements*, 41, 819–825.
- Rink, W.J., Richter, D., Schwarcz, H.P., Marks, A.E., Monigal, K., & Kaufman, D. (2003). Age of the Middle Palaeolithic site of Rosh Ein Mor, Central Negev, Israel: Implications for the age range of the Early Levantine Mousterian of the Levantine corridor. *Journal of Archaeological Science*, 30, 195–204.
- Roberts, R.G. (1997). Luminescence dating in archaeology: From origins to optical. *Radiation Measurements*, 27, 819–892.
- Valladas, G., & Gillot, Y. (1978). Dating of the Olby lava flow using heated quartz pebbles: Some problems. *PACT*, 2, 141–150.
- Valladas, H. (1985a). Datation par la thermoluminescence de gisements moustériens du sud de la France. Unpublished Ph.D. dissertation, University Paris VI, Paris.
- Valladas, H. (1985b). Some TL properties of burnt prehistoric flint. *Nuclear Tracks and Radiation Measurements*, 10, 785–788.
- Valladas, H. (1992). Thermoluminescence dating of flint. *Quaternary Science Review*, 11, 1–5.
- Valladas, H., Valladas, G., Bar-Yosef, O., & Vandermeersch, B. (1991). Thermoluminescence dating of Neanderthal and early humans in the Near East. *Endeavour New Series*, 15, 115–119.
- Wagner, G.A. (1998). Age determination of young rocks and artifacts. Berlin: Springer.
- Warke, P.A. (2000). Micro-environmental conditions and rock weathering in hot arid regions. *Zeitschrift für Geomorphologie* 120, 83–95.

*Received December 30, 2004*

*Accepted for publication January 16, 2006*