Geologie en Mijnbouw 66: 35–42 (1987)

Thermoluminescence dating of loess at Rocourt, Belgium

A.G. Wintle
The Godwin Laboratory
Sub-department of Quaternary Research, University of Cambridge, Free School Lane, Cambridge CB2 3RS, UK

Received 26 August 1986: accepted in revised form 3 February 1987

Key words: Dating, Eltviller tuff, loess, Rocourt, thermoluminescence

Abstract

Nine TL dates were obtained from a section at Rocourt. The dates were self-consistent. A date of 24.8 ± 2.1 ka for a sample from the base of the Upper Weichselian loess indicated that it was part of the massive loess sheet deposited in Europe about 25 thousand years ago. The overlying decalcified loess gave dates indicating deposition after the glacial maximum. However, the proposed chronology is in disagreement with that based on radiocarbon dates on humic material from the 'horizon à langues' at two nearby sites.

Introduction

The dating of Quaternary sediments by measuring their natural thermoluminescence signal has been reported (see Wintle & Huntley, 1982; Singhvi & Mejdahl, 1985 for reviews). Since exposure to light zeroes the earlier geological signal, wind-blown deposits such as loess have been thought to be ideal for this method, provided they are not over 100 ka. This paper reports nine TL dates from a loess section at Rocourt in Belgium. This study was primarily undertaken to obtain the TL age for an ash layer found beneath a tongued horizon at this and other sites.

Thermoluminescence dating

When exposed to ionizing radiation, such as is produced by the decay of isotopes in the uranium and thorium decay chains, TL sensitive minerals are able to record the amount of radiation to which they have been exposed. The most common TL sensitive minerals are quartz and feldspars. Free charges are produced by the interaction of the radiation with the atoms of the crystals and some of the free charges are trapped at electrically attractive defects in the crystal. If the defects are sufficiently attractive, the charges will stay in those traps for times in excess of one million years and they can be used for dating. They can be emptied out by the application of heat, as in the firing of clay to make pottery, or by exposure to sunlight during deposition of a sediment or by placing in front of a laboratory light source. When the charges are released within the crystal, a small portion of them recombine at luminescence centres within the crystal and the energy released in recombination produces photons as the centres de-excite. Photons produced in this way make up the TL signal. The signal is thus proportional to the radiation exposure experienced by the crystals since an earlier application of either thermal or optical energy 'zeroed' the previous TL signal. Sediments may go through this

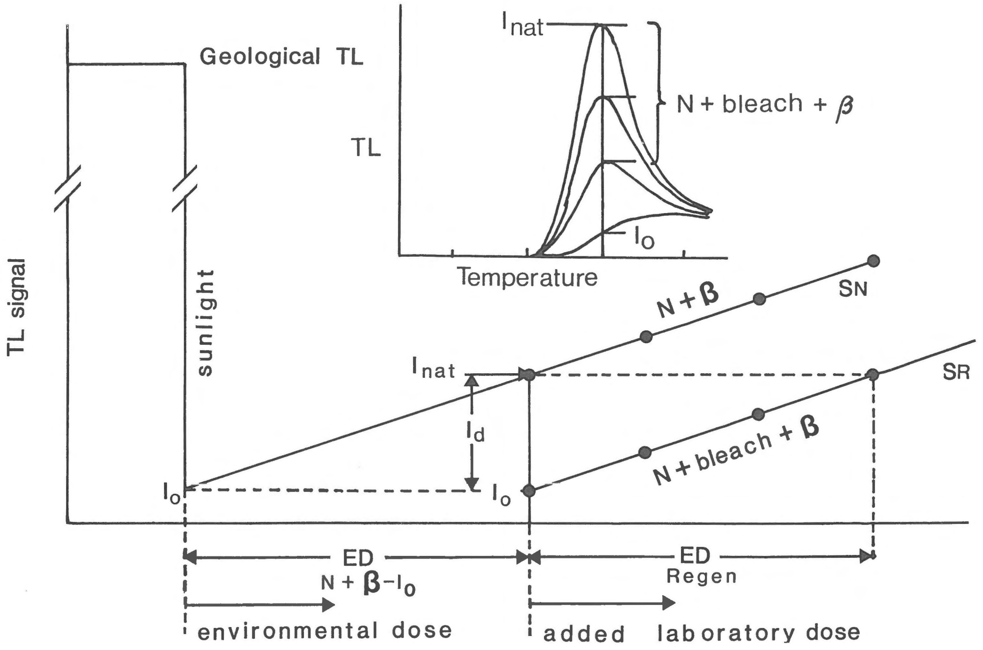

Fig. 1. The geological TL reduced to I_O by exposure to sunlight at deposition and subsequent build up to a level I_{nat} where I_d is the TL produced by radiation in the sediment. Two methods of measuring this radiation exposure, ED, are shown. The inset diagram shows typical glow curves.

cycle several times. The trapped charges may be either electrons or holes which cannot be distinguished by TL measurements, but for discussion purposes they are usually thought of as electrons.

The TL signal is observed in the laboratory with a photomultiplier tube and optical filters are chosen to select a particular part of the emission spectrum from the minerals being studied. The photomultiplier tube has a bialkali photocathode (EMI 9635) with high blue sensitivity and low sensitivity in the red-infrared so that the incandescence from the heating plate is minimized. Debenham & Walton (1983) used an image intensifier to measure the natural emission spectra of a few sediments. Based on their study a Schott UG11 optical filter was used to enhance the feldspar signal. The TL signals are observed as the samples are heated from room temperature to about 450° C at about 5° C/s in an inert atmosphere of nitrogen or argon to eliminate spurious luminescence caused by oxidation processes on the surface of the grains. The plots of TL versus temperature obtained in this way are known as glow curves. Typical glow curves for the Rocourt loess are shown in the inset diagram in Fig. 1. The glow curves have been obtained using a preheat of 16 hours at 150° C immediately before measurement (Wintle, 1985).

Measurements are made on equal aliquots of sample, the weights depending on what grain size or mineral component is being measured. For this study of loess, mixed mineral fine grains (4–11 μm) were prepared according to the procedures of Wintle & Huntley (1980).

To use the TL response for dating, the natural TL signal is compared with the TL signal produced in another aliquot by a known dose of radiation from a laboratory beta or gamma source. From this is obtained the amount of radiation equivalent to that which produced the natural TL signal, the Equivalent Dose or ED. This is put into the age equation

$$\text{Age (years)} = \frac{\text{ED (grays)}}{\text{dose rate (grays/year)}}$$

(1 gray = 1 Gy = 100 rads = 0.1 krad)

The dose rate is obtained by measurement of the radioactive content of the sediment and the methods have been discussed elsewhere (Mejdahl & Wintle, 1984; Aitken, 1985). For the experimental results presented in this paper, thick source alpha counting was used to determine the contribution from the uranium and thorium decay chains and equilibrium was assumed. No significant radon escape was observed as shown by the ratio of the sealed and unsealed alpha counts (Table 1). The ^{40}K contribution was obtained from K_2O determinations made by atomic absorption spectrometry. A cosmic dose rate of 0.14 Gy/ka was used. The alpha efficiency factor, *a*, was obtained by matching the level of alpha-induced TL to that induced by 73 Gy of beta irradiation; this factor is needed to allow for the reduced efficiency of alpha particles in producing TL.

If a sediment contains water then a fraction of the radiation will be absorbed in the water and allowance must be made for it in the age equation. The water content, Δ, used in the dose rate calculation is defined as the (weight of water/weight of dry sediment). The effect of the water content is such that if Δ was assumed to be zero, but had in fact been 0.2 for the geological period under study, then the TL age calculated would be about 20% too low since the dose rate would have been overestimated by 20%. In this study $\Delta = 0.20 \pm 0.05$ was assumed to be typical for loess. The error of ± 0.05 allowed in this value of Δ produces a 5% error in the age. The upper limit for the water content is the saturation water content = $(1/\varrho_D - 1/S_g)$ where S_g is the specific gravity of the particles (2.65) and ϱ_D is is the dry density of the sample (Rendell, pers. comm.). For uncollapsed loess this corresponds to $\Delta = 0.40 \pm 0.05$ and if it had been applicable for the whole period since deposition, a maximum age of 20.3 ± 1.6 ka, rather than 17.1 ± 1.5 ka, would have been obtained for sample QTL89H. However, sample QTL89I immediately beneath it, would also have a correspondingly high water content and thus a TL age of 29.6 ± 2.5 ka, instead of 24.8 ± 2.1 ka.

Equivalent dose determinations

The basis of TL dating of sediments is shown in Fig. 1. The trapped charge, which would have given a large TL signal for the rock from which the sediment is derived, is reduced to a small residual value by exposure to sunlight. The TL level corresponding to this residual trapped charge is given as I_0.

Table 1. Thermoluminescence results and radioactivity data.

sample	bulk count rate ($ks^{-1} cm^{-2}$)	U (ppm)	Th (ppm)	S/U	K_2O (%)	*a*	dose rate (Gy/ka)	ED (Gy)	TL age (ka)
Niederbieber, Germany									
QTL51B	0.793 ± 0.018	3.2	11.5	1.01	1.80	0.10 ± 0.01	4.15	54.1 ± 1.2	13.0 ± 1.1
Stillfried, Austria									
QTL83C	0.865 ± 0.017	3.2	13	0.94	1.97	0.11 ± 0.01	4.66	139 ± 5	29.8 ± 2.5
Rocourt, Belgium									
QTL89B	0.716 ± 0.012	3.5	8.7	1.02	2.35	0.12 ± 0.01	4.51	60.8 ± 2.4	13.5 ± 1.1
QTL89D	0.736 ± 0.019	4.5	5.7	1.01	2.12	0.10 ± 0.01	4.18	55.3 ± 1.4	13.2 ± 1.1
QTL89F	0.750 ± 0.017	3.8	8.5	1.15	2.24	0.12 ± 0.01	4.56	61.6 ± 1.5	13.5 ± 1.1
QTL89G	0.866 ± 0.020	4.8	8.4	1.07	1.86	0.12 ± 0.01	4.72	63.5 ± 3.5	13.5 ± 1.2
QTL89H	0.815 ± 0.018	3.6	11.1	1.01	1.91	0.10 ± 0.01	4.30	73.4 ± 0.7	17.1 ± 1.4
QTL89I	0.833 ± 0.017	5.2	6.8	1.08	1.74	0.13 ± 0.01	4.65	115 ± 3	24.8 ± 2.1
QTL89J	0.747 ± 0.016	4.4	6.3	1.00	2.02	0.11 ± 0.01	4.27	162 ± 5	38.0 ± 3.2
QTL89K	0.697 ± 0.021	4.3	5.3	1.05	1.99	0.10 ± 0.01	3.95	175 ± 6	44.3 ± 3.7
QTL89L	0.699 ± 0.021	3.6	7.7	0.93	1.99	0.11 ± 0.01	4.07	173 ± 2	42.5 ± 3.5

S/U is the ratio of sealed/unsealed alpha counts

Once the sediment is covered by further deposition, the trapped charge will increase with time as the sample is exposed to ionising radiation from the radioactive decay occurring in the sediment. This causes as TL signal I_d. When the TL signal is measured in the laboratory, the total natural TL signal I_{nat} is given by $I_{nat} = I_o + I_d$ (Wintle & Huntley, 1980). A typical glow curve of the natural TL, I_{nat}, is shown in the inset in Fig. 1 along with that of a similar sample disc which has been exposed to an artificial light source for 18 hours to simulate the residual signal, I_0.

There are several ways in which ED can be determined and two of these are also shown in Fig. 1:

1. *the total bleach method* in which a long laboratory bleach (either with a sun lamp or sunlight) is used to simulate the residual level (I_0) at deposition and additional radiation doses are given to samples containing the natural TL and then they are measured to determine the natural TL sensitivity, S_N. The ED is shown as $ED_{N+B-I0} = I_d/S_N$ where $I_d = I_{nat} - I_0$. This is obtained graphically by extrapolation of the increasing TL signal for N + B until it reaches the I_0 level; the dose intercept is then ED_{N+B-I0};
2. *the regeneration method* in which the natural TL is compared with a second growth curve regenerated for samples which have been given a long laboratory bleach to reach I_0 and then irradiated.

In the inset diagram in Fig. 1, besides the glow curves for the natural and the bleached sample discs, there are two curves obtained when different irradiations are applied to other optically bleached discs. These correspond to the two central points on the N + bleach + B curve beneath. The highest point on that curve is obtained for a signal identical to the natural TL signal and the dose given to that sample disc is thus the ED. The ED is shown as ED_{Regen} and will equal ED_{N+B-I0} provided that the regenerated TL sensitivity, S_R, equals S_N. This has been shown to be the case for samples of loess less than 20 ka (Wintle et al., 1984) and no evidence for significant sensitivity changes has been shown for samples older than this (Debenham, 1985).

Age checks in the last 30 ka

The major problem in testing the validity of the various methods of ED determination for loess from the last 150 ka has been the lack of well-dated loess deposits. It is, of course, for this very reason that TL dating is so important! Most "dated" loesses contain either weak soils or volcanic ash layers which have been correlated over large distances to sites which are fortunate enough to have one or two radiocarbon dates.

A securely dated ash is the thick Laacher See ash found in the Neuwied Basin near Koblenz; it has several radiocarbon dates about 11,000 years BP associated with it (Firbas, 1953; Bogaard & Schmincke, 1985). The eruption is thought to have occurred at the end of the Allerød) interstadial (12–11 ka). A sample of loess (QTL51B) from immediately beneath the ash was taken during excavation of the archaeological site at Niederbieber and a TL age of 13.0 ± 1.1 ka was obtained.

Loess of somewhat older age was collected from the typesite for the Stillfried 'B' soil in Austria. A sample (QTL83C) was taken from immediately beneath a thin charcoal layer for which Vogel & Zagwijn (1967) had obtained dates of 28,200 ± 290 BP (GrN-2523) and 28,340 ± 220 BP (GrN-2533). A confirmatory date of 28,900 ± 1,400 BP (GrN-11188) was recently obtained for the upper part of the Stillfried 'B' soil (Haesaerts & Otte, 1985). The TL date of 29.8 ± 2.5 ka was obtained by the regeneration method.

Age checks over 30 ka

Beyond the limit of radiocarbon dating, age estimates rely on recognition of interglacial soils developed on existing loess deposits. The assignation of these soils to a particular interglacial may be based on an established, characteristic pollen record; however, where pollen preservation is poor, a 'count down from the surface' approach may be used, with no account being taken of erosional hiatuses. At Saint Romain, France, a TL age of 115 ± 10 ka was obtained for an interglacial soil, interpreted as of Eemian age (Wintle et al., 1984).

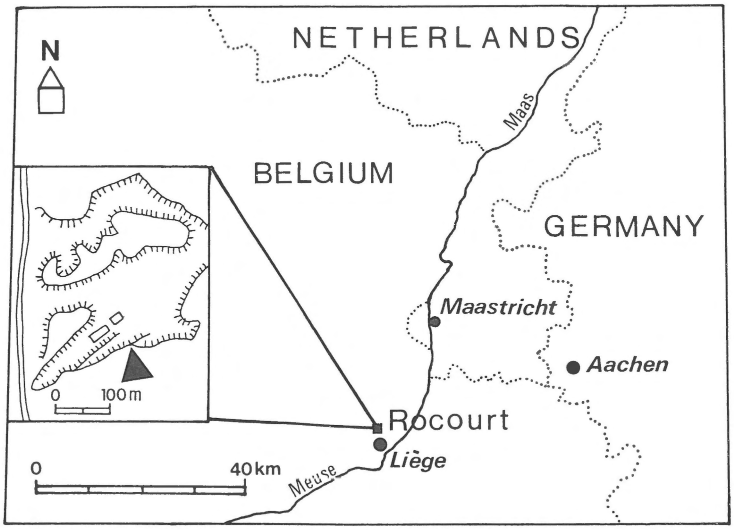

Fig. 2. Map showing location of Rocourt and plan of the sand quarry, taken from Juvigné & Semmel (1981). Sections were on the south wall of the quarry as shown.

However, TL dating of deeper horizons at this site did not yield age estimates greater than 140 ka (Wintle, 1985). Similar limitation of the fine grain TL ages obtained using the regeneration method of ED determination have been reported by Debenham (1985) for other sites in north-west Europe. Until the cause of this problem has been found and its widespread (or otherwise) nature established, TL ages over 50 ka should be regarded as minimum ages.

Rocourt

Samples for TL dating were collected through the uppermost 4.5 m of loess at the sand quarry at Rocourt (S.A. Sables et Graviers), Belgium (Figs. 2 and 3). The site was recently redescribed by Haesaerts et al. (1981). The uppermost unit is the Brabantian loess (Gullentops, 1954) in which the Holocene soil is developed. Today this is capped by humic plough soil. This loess is about 2 m thick and at its base is a darker cryoturbated layer known as the 'horizon à langues de Nagelbeek' (Haesaerts et al., 1981). This tongued horizon contains humic material which has been radiocarbon dated at two nearby sites, Lixhe, 22,190 ± 130 BP (GrN-10328) and Kesselt, 22,270 ± 380 BP (Lv-1172) (Gullentops, 1981). Beneath this is more decalcified loess overlying a carbonate rich loess. Gullentops (1954) introduced the term Hesbayen to designate the loess from the base of the tongued horizon down to the top of the humic rich horizon; the upper part of this loess has been decalcified during Holocene soil development. There is a sharp boundary between the carbonate rich and decalcified loess which can be observed by using an acid wash bottle in the field. About 50 cm below the base of the tongued horizon and within the decalcified loess at this face of the quarry is a thin, discontinuous layer of black ash which has been correlated with the Eltviller Tuff (Rohdenburg & Semmel, 1971; Juvigné & Semmel, 1981). At the base of the calcareous loess is a stoney layer which separates it from a greyish brown, non-calcareous loess. This loess contains evidence of gleying and also contains two layers where there is a concentration of heavy minerals indicating a volcanic component in the loess (Haesaerts et al., 1981). Below this is a humic horizon with radiocarbon dates of 35,900 ± 1,000 (GrN-9081) on humates, 38,550 ± 700 (GrN-9186) on the organic residue of GrN-9081 and 47,800 ± 2,100 (GrN-9080) on humates. However, this humic layer was first considered as the uppermost

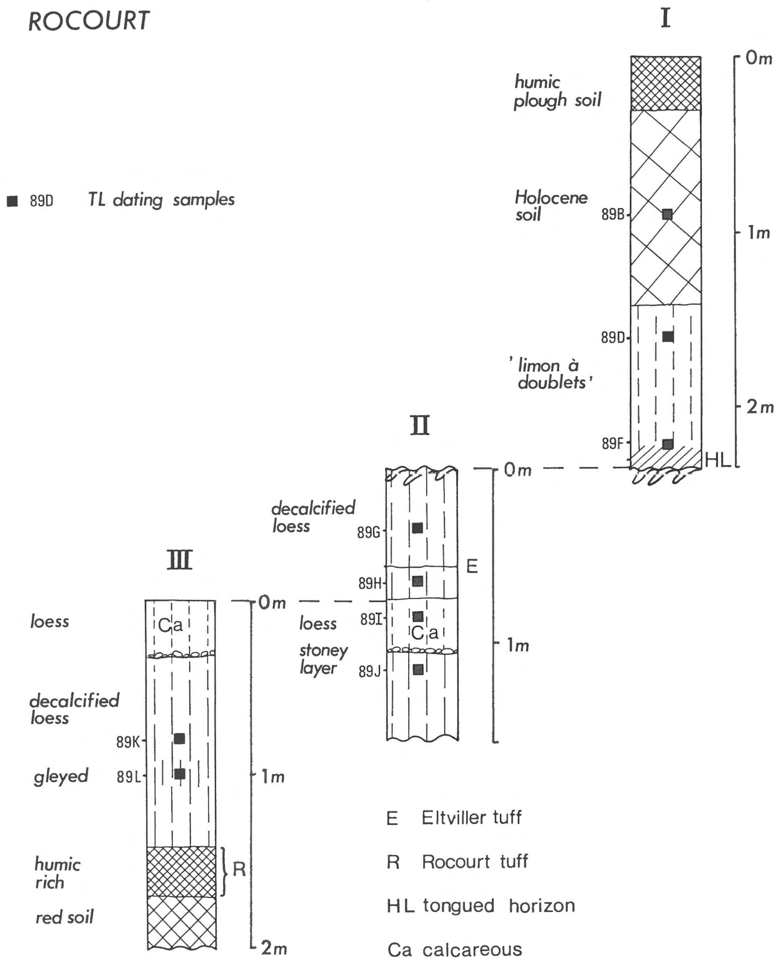

Fig. 3. Three sections at Rocourt showing samples taken for TL dating.

horizon of the Eemian soil (Sol de Rocourt; Gullentops, 1954) and has been correlated later with the Warneton Soil (Paepe & Vanhoorne, 1967), which is thought to date from the early Weichselian. Within this humic horizon there is reworked volcanic material of a widespread ash-fall called "Tuf de Rocourt" (Juvigné, 1977) and that was dated at another locality to older than 51,000 B.P. (Juvigné, 1985). Beneath this is pre-Weichselian loess in which an interglacial soil is developed. This is the lowest part of the Rocourt Soil (sensu Gullentops 1954).

TL dates have been obtained for nine samples from the uppermost 3.5 m of the section above the humic horizon (see Fig. 3). EDs were obtained using the regeneration method and are given in Table 1, along with the other data. From the TL point of view it appears that there are three periods of loess deposition after the formation of the humic-rich soil horizon. The uppermost 2.5 m of loess was deposited after the last glacial maximum and includes both the tongued horizon and the ash layer. The 30 cm of calcareous loess appears to represent a different phase of loess deposition just prior to the glacial maximum. The underlying non-calcareous loess was deposited some 13 ka earlier than this.

Radioactivity analyses – laboratory versus in situ

The dose rates were calculated on the basis of the alpha counting and atomic absorption spectrometry measurements performed in the laboratory, with a water content correction factor of △ = 0.20 ± 0.05. When the samples were collected a gamma scintillation counter was used to measure the *in situ* gamma dose rate, the NaI probe being buried 30 cm from the vertical face of the section. The counter has been calibrated using concrete blocks with known contents of uranium, thorium and potassium, and had been shown to have a negligible response to cosmic rays. The mean *in situ* water content was 0.10 ± 0.05, the samples being collected in summer from an exposed face.

In Table 2 the gamma dose rate measured by the scintillation detector can be compared with that calculated from the laboratory measurements, assuming water contents of 0.10 and 0.20 respectively. Good agreement is shown for samples QTL89G to 89K.

Table 2. Gamma dose rate measurements in Gy/ka compared with calculated values assuming two different water contents.

sample	scint	△ = 0.1	△ = 0.2
QTL89G	1.20	1.22	1.12
QTL89H	1.24	1.21	1.11
QTL89I	1.19	1.20	1.10
QTL89J	1.16	1.09	1.00
QTL89K	1.13	1.06	0.97

Carbonate analyses

All the radiocarbon dated control samples contained carbonate, as observed qualitatively in the pretreatment with dilute HCl. However, apart from sample QTL89I, negligible effervescence was observed from the samples at Rocourt. In order to quantify this, 1.7 g of samples QTL89G, H and I were subjected to carbonate analysis using Chittick Apparatus. No carbonate was found in sample QTL89G and only 1% calcite was found for QTL89H. QTL89I had 0.8% dolomite and 4.3% calcite. The presence of dolomite shows that some primary carbonate is present in this sample.

Discussion

From the TL viewpoint the ages are stratigraphically self-consistent within the error limits on individual dates and the field and laboratory dose rate measurements are in agreement. In the light of these dates an interpretation of the depositional events at Rocourt in the Upper Weichselian can be made.

The dates shown in Table 1 imply that there was more than one period of loess deposition represented by the loess between the stoney layer and the tongued horizon. The oldest loess, represented by sample QTL89I, is dated to 24.8 ± 2.1 ka which coincides with the accepted date for the beginning of Upper Weichselian loess deposition (Haesaerts, 1985). A more recent loess covered this deposit after the maximum ice advance at about 18 thousand years ago with the Eltviller tuff being incorporated in this loess around 17 thousand years ago. The tongued horizon was formed on top of this loess about 15 thousand years ago. This was followed by further loess deposition for about 2 thousand years. This is in agreement with the date for the cessation of loess deposition at Niederbieber in Germany, 13.0 ± 1.1 ka (QTL51B) given earlier in this paper and with that obtained at Saint Romain in France (Wintle et al., 1984).

This interpretation disagrees with the current geological interpretation of the Upper Pleistocene sequences in Belgium. Haesaerts (1985) considers that the main deposition of loess in Belgium took place between 20 and 25 thousand years ago and that between 15 and 20 thousand years ago a second deposition of finely stratified loess occurred. The main argument for this interpretation is based on the two radiocarbon dates on humic material from the tongued horizon at Lixhe and Kesselt which gave 22,190 ± 130 BP (GrN-10328) and 22,270 ± 380 BP (Lv-1172) respectively (Gullentops, 1981). Both these dates were obtained on 8 g of humic acids extracted from 8 kg of material taken from the humic horizons. If these are too old, for example if they contained reworked material from an older soil formation, then the chronology based on the TL dates would be viable. Besides the agreement of the two dates, the other argument for accepting them is that soil formation was followed by a cold phase with the production of ice wedges; this period is usually thought to be the time of maximum glaciation. Haesaerts also considers that the subdivision of the loess between the stoney layer and the tongued horizon to be unacceptable (personal communication). In his opinion the decalcified loess, represented by QTL89H, is of the same depositional age as the underlying carbonate-rich loess, represented by QTL89I. The clear limit of decalcification is caused by progressive downward leaching of humic acids created by post-depositional soil formation, a process which is still continuing.

In view of the possibility that TL measurements may be affected in some way by decalcification processes in such a way as to underestimate the TL age, further studies will be made of both the decalcified loess and the underlying carbonate-rich loess at other sections in the quarry at Rocourt.

Acknowledgements

TL studies in Cambridge are financed by NERC grant GR3/3174 and EEC contract nr. ST2P-0204-1-UK. The carbonate analyses were performed by Mr. H. Canfield using Chittick Apparatus at the University of Wisconsin at Madison. The author wishes to thank Dr. E. Juvigné, Professor G. Bosinski and Dr. H. Summesberger for help in collecting the samples in Belgium, Germany and Austria. I also wish to thank Dr. E. Juvigné, Dr. H. Rendell, Dr. N.J. Shackleton and Dr. J. Vandenberghe for making constructive comments on the manuscript.

References

Aitken, M.J. 1985. Thermoluminescence Dating – Academic Press, London; 359 pp

Bogaard, P. & H-U. Schmincke. 1985. Laacher See Tephra: A widespread isochronous late Quaternary tephra layer in central and northern Europe – Geol. Soc. Am. Bull. 96: 1554–1571

Debenham, N.C. 1985. Use of U.V. emissions in TL dating of sediments – Nuclear Tracks 10: 717–724

Debenham, N.C. & A.J. Walton. 1983. TL properties of some wind-blown sediments – PACT 9: 531–538

Firbas, F. 1953. Das absolute Alter der jungsten vulkanischen Eruptionen im Bereich des Laacher Sees – Naturwissenschaften 40: 54–55

Gullentops, F. 1954. Contribution à la chronologie du Pléistocène et des formes du relief en Belgique – Mém. Inst. Géol. Univ. Louvain XVIII: 125–252

Gullentops, F. 1981. About the climate of the last glaciation in NW Europe – Symposium on Quaternary climatic variations in a Milankovitch Perspective, U.C. Louvain

Haesaerts, P. 1985. Les loess du Pléistocène supérieur en Belgique; comparaisons avec les séquences d'Europe centrale – Bull. Assoc. Fr. Étude Quat., 2–3: 105–115

Haesaerts, P., E. Juvigné, O. Kuyl, H. Mucher & W. Roebroeks. 1981. Compte rendu de l'excursion du 13 Juin 1981, en Hesbaye et au Limbourg Néerlandais, consacrée à la chronostratigraphie des loess du Pléistocène supérieur – Ann. Soc. Géol. Belg. 104: 223–240

Haesaerts, P. & M. Otte. 1985. Nouvelle recherches au gisement de Willendorf (Autriche) Coll. Intern. Le Cadre chronologique du Paléolithique supérieur ancien – Leon, 1983, Santillana del Mar (in press)

Juvigné, E. 1977. Zone de dispersion et âge des poussières volcaniques du tuf de Rocourt – Ann. Soc. Géol. Belg. 100: 13–22

Juvigné, E. 1985. Données nouvelles sur l'âge de la capture de la Wouche à Bévercé – Bull. Soc. Geog. Liège. 21: 3–11

Juvigné, E. & A. Semmel. 1981. Un tuf volcanique semblable à l'Eltviller Tuff dans les loess de Hesbaye (Belgique) et du Limbourg néerlandais – Eiszeitalter Gegenw. 31: 83–90

Mejdahl, V. & A.G. Wintle. 1984. Thermoluminescence applied to age determination in archaeology and geology. In: Y.S. Horowitz (ed) Thermoluminescence and thermoluminescent dosimetry, vol. III: 133–190

Paepe, R. & R. Vanhoorne. 1967. The stratigraphy and paleobotany of the late Pleistocene in Belgium – Mem. Expl. Cartes Géol. Min. Belg. 8: pp 96

Rohdenburg, H. & A. Semmel. 1971. Bemerkungen zur Stratigraphie des Wurm-Losses im westlichen Mitteleuropa – Notizbl. L.-Amt Bodenforsch. 99: 246–252

Singhvi, A.K. & V. Mejdahl. 1985. Thermoluminescence dating of sediments – Nuclear Tracks 10: 137–161

Vogel, J.C. & W.H. Zagwijn. 1967. Groningen radiocarbon dates VI – Radiocarbon 9: 63–106

Wintle, A.G. 1985. Stability of TL signal in fine grains from loess – Nuclear Tracks 10: 725–730

Wintle, A.G. & D.J. Huntley. 1980. Thermoluminescence dating of ocean sediments – Can. J. Earth Sci. 17: 348–360

Wintle, A.G. & D.J. Huntley. 1982. Thermoluminescence dating of sediments – Quat. Sci. Rev. 1: 31–53

Wintle, A.G., N.J. Shackleton & J.P. Lautridou. 1984. Thermoluminescence dating of periods of loess deposition and soil formation in Normandy – Nature 310: 491–493