

Article

"Thermoluminescence Dating of Late Pleistocene Sediments, St. Lawrence Lowland, Québec"

Michel Lamothe et David J. Huntley

Géographie physique et Quaternaire, vol. 42, n° 1, 1988, p. 33-44.

Pour citer cet article, utiliser l'information suivante :

URI: <http://id.erudit.org/iderudit/032707ar>

DOI: 10.7202/032707ar

Note : les règles d'écriture des références bibliographiques peuvent varier selon les différents domaines du savoir.

Ce document est protégé par la loi sur le droit d'auteur. L'utilisation des services d'Érudit (y compris la reproduction) est assujettie à sa politique d'utilisation que vous pouvez consulter à l'URI <https://apropos.erudit.org/fr/usagers/politique-dutilisation/>

Érudit est un consortium interuniversitaire sans but lucratif composé de l'Université de Montréal, l'Université Laval et l'Université du Québec à Montréal. Il a pour mission la promotion et la valorisation de la recherche. Érudit offre des services d'édition numérique de documents scientifiques depuis 1998.

Pour communiquer avec les responsables d'Érudit : info@erudit.org

THERMOLUMINESCENCE DATING OF LATE PLEISTOCENE SEDIMENTS, ST. LAWRENCE LOWLAND, QUÉBEC*

Michel LAMOTHE** and David J. HUNTLEY, respectively: Terrain Sciences Division, Geological Survey of Canada, 601 Booth Street, Ottawa, Ontario K1A 0E8, and Department of Physics, Simon Fraser University, Burnaby, British Columbia V5A 1S6.

ABSTRACT This paper presents the results of a thermoluminescence (TL) dating program applied to waterlaid sediments of Late Pleistocene age, collected in the central St. Lawrence Lowland, in eastern Canada. The apparent TL ages were obtained using a partial bleach method (R-Gamma) in which the TL from light-sensitive traps is separated from the total TL. Fluvial (modern), marine (late-glacial) lacustrine (interstadial) and glaciolacustrine (stadial) sediments yielded apparent ages ranging from 4 to 135 ka, and in general, these ages agreed with available stratigraphic evidence. Whilst testing the zeroing assumption, apparent TL ages obtained from modern fluvial and late glacial sediments suggest that laboratory overbleaching or insufficient natural bleaching may result in overestimation of the apparent TL ages by 4-5 ka. All samples display anomalous fading, a problem that can be overcome by introducing a three month delay between artificial irradiation and glowing. Future research should focus on sediments for which the age as well as the sedimentology are well documented, such as the late glacial Champlain Sea sediments.

RÉSUMÉ Datation par thermoluminescence de sédiments du Pléistocène supérieur des basses terres du Saint-Laurent, Québec. Cet article présente les résultats d'un projet de datation par thermoluminescence (TL) de sédiments des basses terres du Saint-Laurent datant du Pléistocène supérieur. Les âges TL apparents ont été obtenus en utilisant une méthode de lessivage partiel (R-Gamma) par laquelle la TL émanant de pièges cristallins sensibles à la lumière a pu être séparée de la TL totale. Les sédiments, d'origines fluviales (modernes), marine (tardiglaciaires), lacustre (interstadias) et glaciolacustre (stadias) ont livré des âges entre 4 et 135 ka et, en général, les âges obtenus correspondent aux données stratigraphiques. Des expériences menées sur le phénomène de « remise à zéro » ont été menées sur des sédiments fluviaux modernes et marins tardiglaciaires. Elles démontrent que, dans les cas où le lessivage solaire a pu être insuffisant ou le lessivage simulé en laboratoire trop intense, les âges TL apparents peuvent être surestimés de 4 à 5 ka. Tous les échantillons montrent un affaiblissement anormal du signal de thermoluminescence. Ce problème peut être résolu par l'introduction d'un délai de trois mois entre l'irradiation artificielle et la mesure de l'échantillon. À l'avenir la recherche devrait se faire sur des sédiments dont la chronologie et la sédimentologie sont bien connues, tels les sédiments tardiglaciaires champlainiens.

ZUSAMMENFASSUNG Datierung mittels Thermolumineszenz von Sedimenten aus dem späten Pleistozän, Sankt-Lorenz Tiefebene, Québec. Dieser Artikel stellt die Ergebnisse eines Datierungsprojekts mittels Thermolumineszenz (TL) dar, angewandt auf angeschwemmte Sedimente aus dem späten Pleistozän, die in Ostkanada, in der zentralen Sankt-Lorenz Tiefebene gesammelt wurden. Die offensichtlichen TL-Alter wurden mittels einer partiellen Bleichmethode (R-Gamma) gewonnen, in welcher die TL der lichtempfindlichen Fallen von der totalen TL getrennt wurde. Die Sedimente von Fluß (modern), Meer (Spätglazial), See (Interstadial) und Glazio-See (Stadial) lieferten offensichtliche Alter zwischen 4 bis 135 ka, und im allgemeinen stimmten diese Alter mit den verfügbaren stratigraphischen Zeugnissen überein. Wohingegen beim Testen mittels der 'Null-Annahme' die offensichtlichen TL-Alter, die aus modernen Flußsedimenten und späten glazialen Sedimenten gewonnen wurden, vermuten lassen, daß zu intensives Bleichen im Labor oder ungenügendes Bleichen in der Natur zu einer Überschätzung der offensichtlichen TL-Alter von 4 bis 5 ka führen. Alle Proben weisen eine anormale Abschwächung des TL-Signals auf. Dies Problem kann durch die Einführung eines Aufschubs von drei Monaten zwischen der künstlichen Bestrahlung und dem Messen der Probe gelöst werden. Die künftige Forschung sollte sich auf Sedimente konzentrieren, deren Alter als auch Sedimentologie gut dokumentiert sind, wie z.B. die spätglazialen Champlain-Meer-Sedimente.

* Geological Survey of Canada Contribution 30586

** Present Address: Département des sciences de la Terre, Université du Québec à Montréal, C.P. 8888, Succ. «A», Montréal, Québec H3C 3P8

Manuscrit reçu le 30 juillet 1986; manuscrit révisé accepté le 16 septembre 1987

INTRODUCTION

For the last three decades, radiocarbon dating has provided Quaternary geologists with isotopic ages from which most of the current chronostratigraphy has evolved. Nevertheless, this dating technique is generally limited to the last 50 000 years of the Quaternary Period. Consequently, this decade has been a period of initiation of alternative techniques, such as amino-acid racemization, uranium-thorium and thermoluminescence (TL) dating, none of which has yet been proven fully reliable. In order to be able to test such techniques, a sequence of samples of similar mineralogy but of different and known ages, deposited in various sedimentary environments is needed. This type of sequence is not common. This paper presents the results of a thermoluminescence dating

program on such a sequence from the Pleistocene of the St. Lawrence Lowland, in Eastern Canada (Fig. 1). Our objectives, herein, are (1) to stress the type of problems one would encounter in dating sediments by thermoluminescence and (2) to discuss the geochronologic significance of a suite of apparent TL ages measured on sediments deposited during the last glaciation in this part of North America.

PRINCIPLES OF THERMOLUMINESCENCE DATING OF SEDIMENTS

For a review of the TL process and application, the reader is referred to Aitken (1985). This process has been used widely during the last 15 years for dating archaeological ceramics and other fired material. Since TL reflects the total

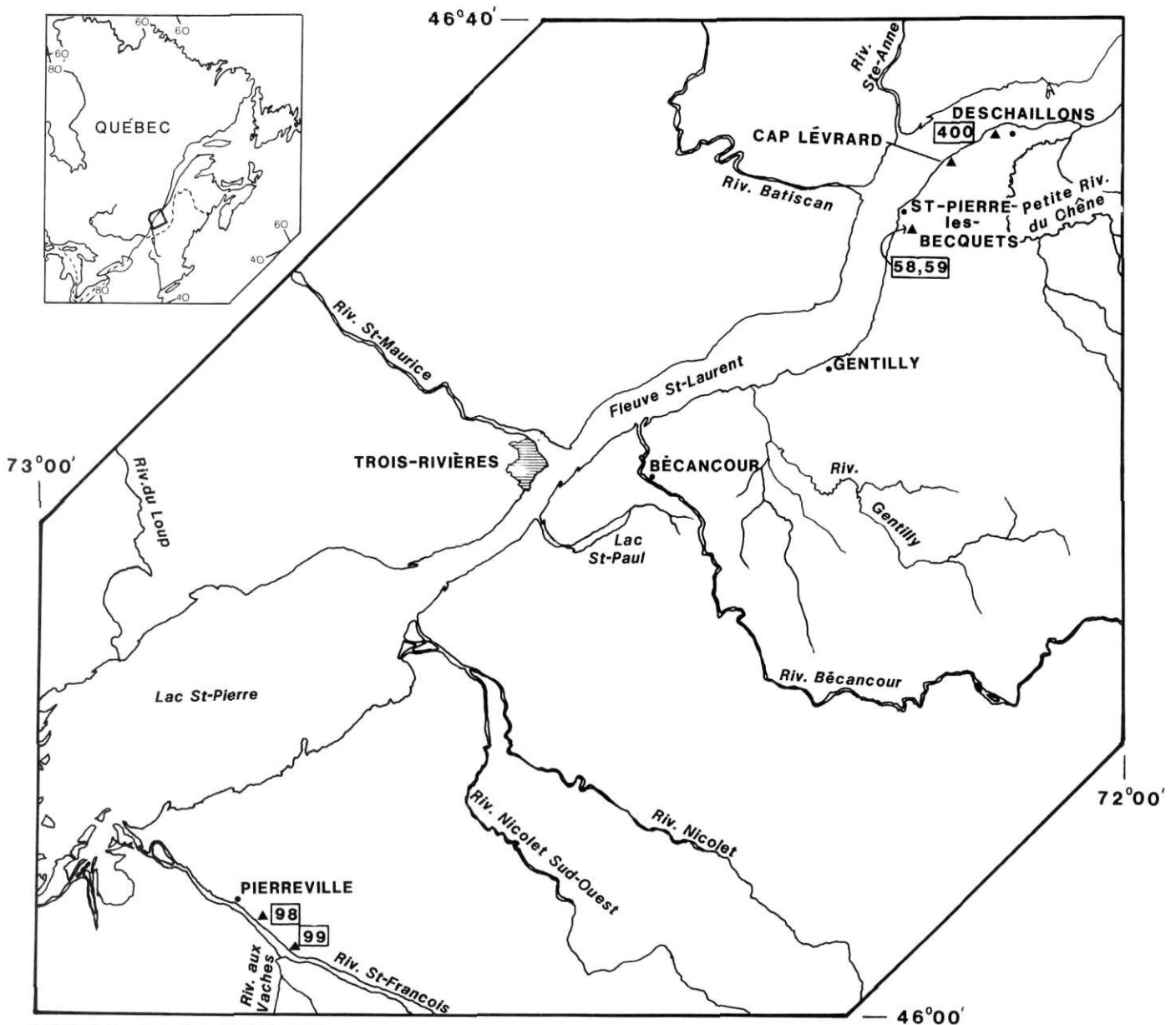


FIGURE 1. Location of study area.

Localisation de la région à l'étude

radiation dose a mineral has received since it has been heated or since its crystallization, the total TL intensity is a measure of time. Provided the radiation dose-rate is constant, an apparent age can be calculated from the following equation:

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

The equivalent dose (D_e) is defined as the artificial β or γ radiation dose that can simulate the natural TL level. The effective dose rate is defined as the rate at which energy that is manifested as TL is deposited in the mineral from α , β , γ or cosmic rays. In archaeological dating, firing in antiquity completely drains the previously acquired TL, therefore, the total D_e is used in the age equation. In contrast, for sediments, exposure to sunlight drains the previously acquired TL, but not completely, and one must determine the D_e since this event. In order to accomplish this, Wintle and Huntley (1980) suggested that traps in detrital minerals can be subdivided into two classes, light sensitive and light insensitive, and that, in order to date the last sedimentation event, one must separate the TL that is emitted by the light sensitive traps, and determine the equivalent dose for this component. It is assumed that grains were exposed to sunlight before deposition.

There are currently three different techniques in use to measure the light sensitive TL component: (1) the regeneration method, (2) the total bleach method, and (3) the partial-bleach method (R-Gamma of R-Beta). These techniques are described in several publications (e.g. Aitken, 1985). In the present study, the R-Gamma method of Wintle and Huntley (1980) has been used since this is the only one which allows for partial bleaching of the light-sensitive traps (Huntley, 1985). In this method, the reduction (R) of the TL caused by a short light exposure is measured and the equivalent dose is taken at the intersection of the unbleached TL growth curve with the optically bleached growth curve. These curves are constructed from natural and irradiated discs, some of which have been exposed to a short laboratory light exposure. In other words, the equivalent dose used in the age equation is the one that corresponds to $R = 0$.

NATURE AND STRATIGRAPHIC POSITION OF THE SELECTED SAMPLES

The Pleistocene stratigraphy of the central part of the St. Lawrence Lowland has been described by Gadd (1955, 1971). A local red till (Bécancour Till), which owes its colour to incorporation of the red Rivière Bécancour shales, is, in some sections, overlain by reddish varves (Pierreville Varves), which are overlain in turn by nonglacial sandy and peaty sediments (Saint-Pierre Sediments), glaciolacustrine sediments (Deschaillons Formation) and an upper grey till (Gentilly Till). This sequence is thought to represent one glaciation (Wisconsinan) with two major ice advances separated by a brief interstade, dated at ca. 65-75 ka BP (Gadd, 1971).

This stratigraphic concept has been refined by Lamothe (1985). For example, the stratigraphic position of the Deschaillons Formation is considered *equivocal* because its superposition over true Saint-Pierre Sediments cannot be demonstrated. However, since this stratigraphic interpretation

has not been published yet, the stratigraphic framework described above is the one used in this paper.

Most of the samples for TL dating have been collected from four critical sections that can be observed in two distinct areas (Fig. 2): the Pierreville area (sections 98 and 99) and the Saint-Pierre-les-Becquets area (sections 58 and 400). The section numbers are from Gadd (1955) except section 400 which is from Karrow (1957). Exact locations of the samples are shown on Figure 2. The samples can be classified according to their presumed age:

Modern sediments:

- CLTO: silt collected in the water of the St. Lawrence River, at Cap Lévrard, 20 cm under the surface of water.
- CCTO: gravelly sand collected on a St. Lawrence River beach, 5 cm from the surface; beach sand. The beach is located 5 km downriver from Deschaillons, at Cap Charles.

Late-glacial sediments: Those sediments were laid down in the Champlain Sea basin (9.5-12.5 ka).

- LP: clayey silt; collected 30 cm over the Gentilly Till at section 99; apparently nonfossiliferous; *Portlandia arctica* bearing silts are found on top of the same till on the other side of the Rivière Saint-François (section 109); shallow water marine clay (unit 99-G).

- MC-DES: massive pebbly silty clay collected in a St. Lawrence River section found approximately 2 km upstream from the Deschaillons brickyard; *Balanus hameri* collected in these silts yielded a ^{14}C date of $11\,130 \pm 180$ years BP (UQ-651); deepwater marine clay.

Interstadial sediments: These samples were collected in lithostratigraphic units related to the Saint-Pierre Interstadial. The age of these sediments is presumably 70 ± 10 ka (see below).

- SP: silty sand collected 30 cm below the peat, section 98; alluvial sand (unit 98-B)
- RS: clayey silt collected 30 cm above the same peat, section 98; shallow lacustrine silt; (unit 98-D).
- SS: clayey sandy silt collected in a unit correlated with RS, section 99; (unit 99-E).
- StP: sandy silt collected 30 cm above the uppermost peat layer at the Saint-Pierre type section (58); alluvial silt; (unit 58-B).

Stadial sediments: The samples are from glaciolacustrine sediments overlying (VDw) and underlying (RY, VP, SPI) the Saint-Pierre Sediments. The age of the Deschaillons Formation should be close to 70 ka. The underlying stadial sediments should not be older than the Illinoian.

- VDw: clayey silt from the Deschaillons Formation; section 400; winter laminae of a sample collected in the middle part of the unit; (unit 400-B).

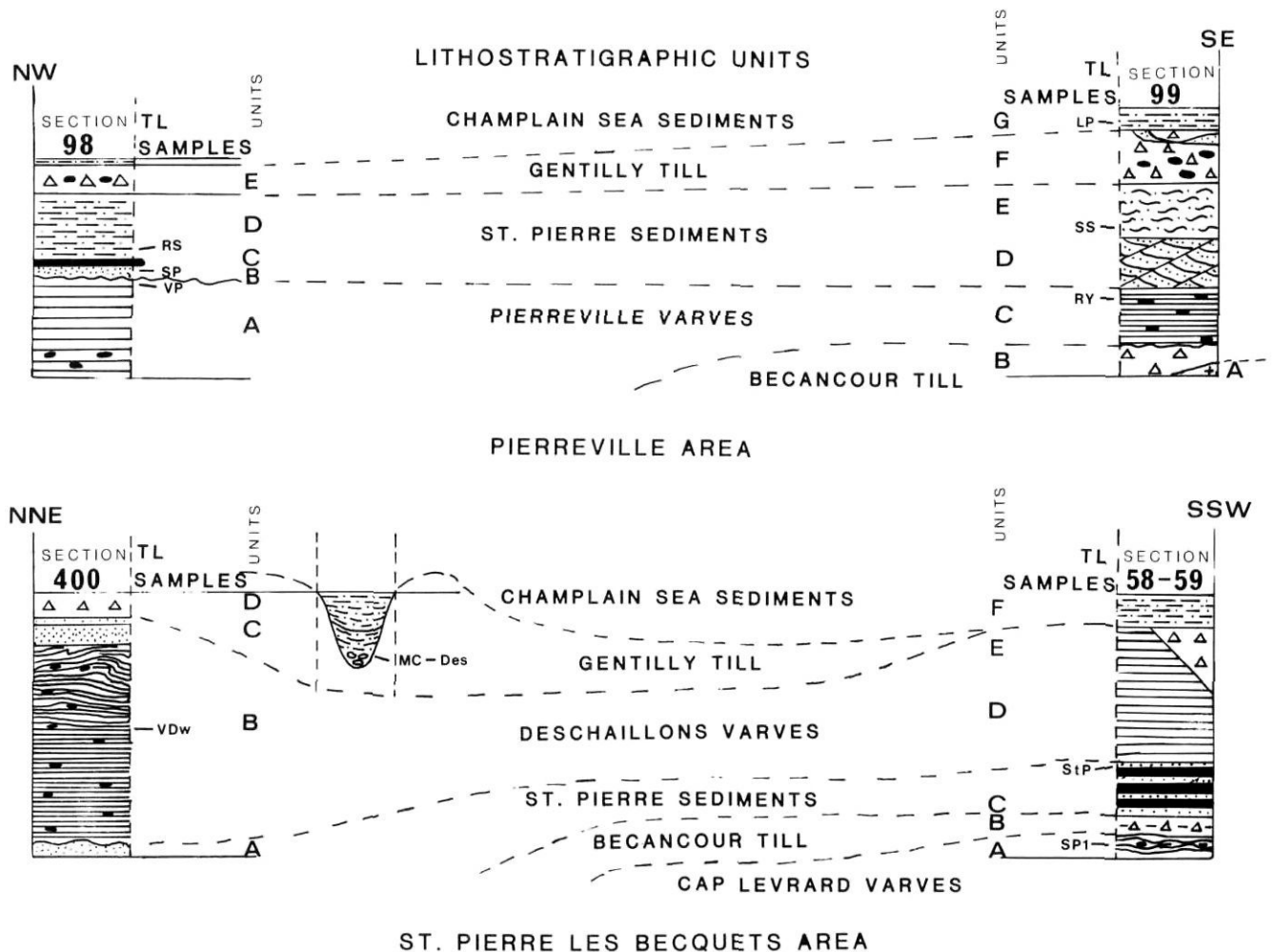


FIGURE 2. Stratigraphic position of the samples collected for TL dating. *Position stratigraphique des échantillons datés par TL.*

- RY: silty clay of 99 and 109-C. 99RY; 2 m above the brickred till, section 99 (unit 99-C). 109RY; same unit, section 109, 1 m below the overlying Saint-Pierre sand; (unit 109-C). These sediments are interpreted as "distal" glaciolacustrine clay.
- VP: clayey silt, of the Pierreville varves, section 98; 30 cm under the base of the Saint-Pierre sand; (unit 98-A).
- VPw: winter laminae of an equivalent sample. The Pierreville varves are proximal glaciolacustrine sediments.
- SPI: clayey silt collected 30 cm below the Saint-Pierre sand, section 59; glaciolacustrine sediments; (unit 59-A).

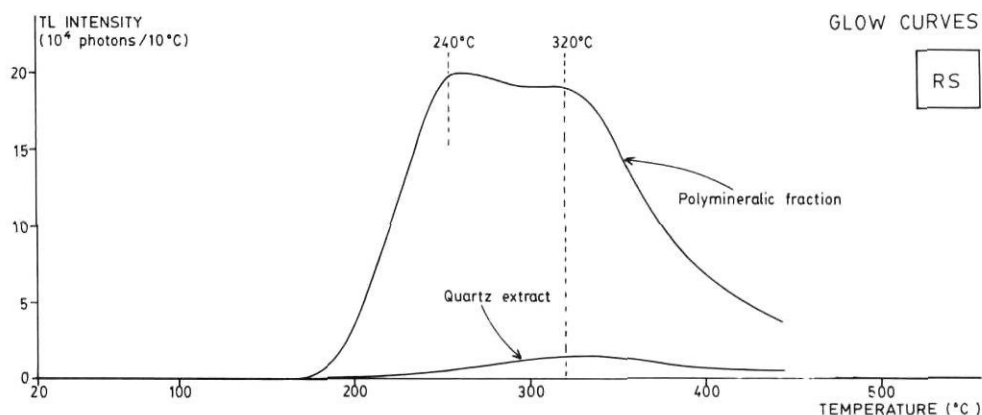
TL MEASUREMENTS: THE FINE GRAIN TECHNIQUE

The fine grain technique (Zimmerman, 1971) was used in the present test program. The TL was therefore measured on fine silt grains (4-11 μm) that were first isolated by sedimentation, soaked in dilute HCl to remove most carbonates,

and then, sedimented on 1 cm diameter aluminum disks. All TL measurements were performed at the Physics Department of Simon Fraser University. The TL apparatus used has been described by Wintle and Huntley (1980). The sample discs containing the mineral grains are heated on a Kanthal heating strip. A thermal compound (from Wakefield Engineering, Wakefield, Massachusetts, USA) was used to ensure a good thermal contact between the disc and the strip. The glow oven was evacuable and, during heating, argon flowed continuously in the oven at 1l/min at atmospheric pressure. A 5 ml beaker of P_2O_5 was placed at the bottom of the chamber to remove moisture. The heating rate was 3°C/s .

The light intensity was measured using a photomultiplier tube (EMI 9635). Two optical filters were placed between the tube and the sample, an infrared (heat absorbing) filter and a Corning 5-58 (blue transmitting) filter. Due to the large amount of light generated by the samples, neutral density filters were also added in most cases. Photon counting was employed using a multichannel scaler from which the glow curves were transferred to magnetic tape for processing.

Gamma irradiations at ~ 2 Gy/min were administered by an AECL Gammacell-200 Co-60 source. Alpha irradiations



GLOW CURVES FIGURE 3. Typical glow curves for the 4-11 μm fraction, sample RS (heating rate: 3°C/s).

Courbes de chauffe caractéristiques pour la fraction de 4-11 μm , échantillon RS (vitesse de chauffe: 3°C/s).

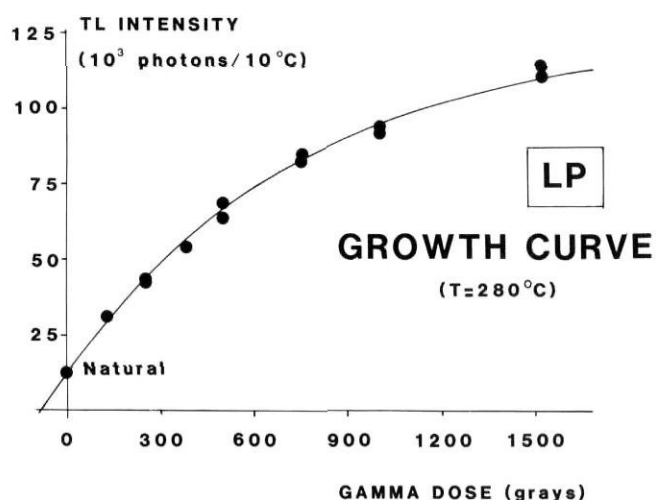


FIGURE 4. Representative growth curve (TL vs. artificial dose) for sample LP, at 280°C. The solid line is an exponential fit to the data. *Courbe de croissance (TL et dose artificielle) de l'échantillon LP, à 280°C. La ligne pleine est une courbe exponentielle.*

were administered with 12 MB_q Am-241 alpha sources for measurement of the alpha effectiveness value.

Description of the alpha-counting equipment used for the determination of U and Th contents can be found in Huntley and Wintle (1981). Potassium contents were measured by atomic absorption spectroscopy at the Département des Sciences de la Terre of the Université du Québec à Montréal.

TL CHARACTERISTICS OF THE SAMPLES

The samples are polymineralic, composed primarily of quartz and feldspars, with minor amounts of amphibole, chlorite and illite; these were identified by diffractometry. Typical glow curves are shown in Figure 3. They show two natural peaks centred at 240°C and 320°C. A hydrofluosilicic acid treatment (Berger and Huntley, 1982) was used to isolate the quartz fraction in sample RS. This fraction yields a single peak at 320-330°C, with low light intensity. Feldspars are therefore the chief source of the TL as observed elsewhere by Wintle (1982), Berger and Huntley (1982) and Berger (1984). This

has important consequences since some feldspars show anomalous fading (Wintle, 1973).

A TL growth curve generated on a young sample (LP ~10ka) shows that the growth of the TL in these sediments is best approximated by a saturating exponential (Fig. 4). The TL accumulates in traps in an almost linear fashion at low dose and approaches saturation at high dose. Apparently, traps of the polymineralic fraction of this sample were not saturated even after addition of laboratory irradiation of 1500 grays.

THE AGE EQUATION USED IN THE TL DATING PROGRAM

The apparent TL ages were calculated using the age equation shown in Table I. The nature of the upper and lower terms of this age equation and the problems encountered in the course of the dating program are discussed below.

1. The equivalent dose (D_e) and the problem of anomalous fading

The equivalent dose (D_e) is defined as the laboratory dose that yields the same TL as the natural dose. It is expressed in grays (1 gray = 1J/kg). The D_e normally increases with temperature on the glow curve (TL output vs temperature) and reaches a plateau value above 250°C. The plateau identifies the part of the glow curve which derives from electron traps for which thermal excitation of electrons has not occurred during geological time.

The equivalent dose was obtained using the R-Gamma method described earlier. Backward extrapolation of the unbleached growth curve was achieved through an exponential best-fit program (e.g. Figs. 4, 5 and 6). However, because of processing limitations, a quadratic best-fit was used for sample VP. It was found that both quadratic and exponential extrapolations gave the same result within one standard-deviation. The bleached growth curve in all samples was approximated by a linear best-fit.

All the samples measured in this test program yielded good plateaux (Fig. 7). Consequently, the condition of thermal stability of electrons in traps is met. Nevertheless, anomalous fading tests carried over 6 to 8 weeks showed a decrease of the TL signal during storage for all samples. Unfortunately,

TABLE I
Age Equation

AGE (years) = D_e (grays)

$$\begin{aligned}
 & W_{K_2O} \left[\frac{d_{\beta K}}{1 + H_{\beta}\Delta} + \frac{d_{\gamma K}}{1 + H_{\gamma}\Delta} \right] \\
 & + \lambda U \left[\frac{1.80 a}{1 + H_{\alpha}\Delta} + \frac{d_{\beta U}}{1 + H_{\beta}\Delta} + \frac{d_{\gamma U}}{1 + H_{\gamma}\Delta} \right] \\
 & + \lambda Th \left[\frac{1.74 a}{1 + H_{\alpha}\Delta} + \frac{d_{\beta Th}}{1 + H_{\beta}\Delta} + \frac{d_{\gamma Th}}{1 + H_{\gamma}\Delta} \right] \\
 & + \frac{D_c}{1 + H_{\gamma}\Delta} \quad (\text{grays-year}^{-1})
 \end{aligned}$$

- Notes
1. W_{K_2O} is the weight (%) of potassium oxide.
 2. λTh and λU are the count rates from the Th and U decay chains as measured in gross alpha counting (Huntley and Wintle, 1981) in units of $ks^{-1} cm^{-2}$.
 3. D_c is the cosmic dose-rate.
 4. a, H and Δ are described in the text; $H_{\alpha} = 1.49$; $H_{\beta} = 1.25$ and $H_{\gamma} = 1.00$ (Bowman, 1976).
 5. The values of d used are those given by Wintle and Huntley (1980).

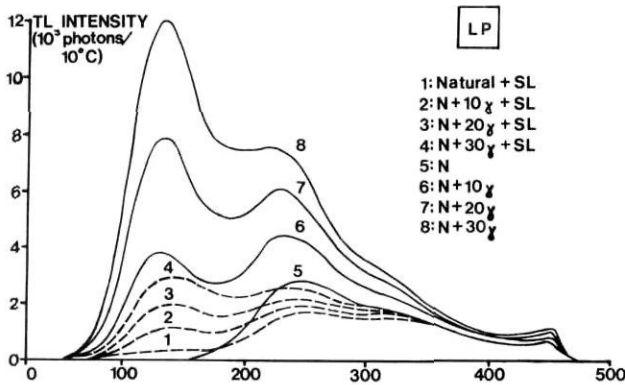


FIGURE 5. Unbleached and bleached glow curves for natural and irradiated discs of sample LP; the bleaching was 2 hours under the sunlamp (SL); λ is 2.1 Gy.

Courbe de chauffe de disques naturels ou irradiés, avant et après lessivage sous la lampe solaire (SL) pendant 2h; λ signifie 2,1 Gy.

this problem became apparent only after most of the samples had been measured, since anomalous fading tests were run after the other measurements. The D_e of these samples is therefore lower than the true equivalent dose. A delayed measurement approach adopted to solve this problem (Lamothe, 1984) demonstrated that the TL apparent ages obtained from samples measured two hours after irradiation are too young. In the tables such dates are labelled as "greater than". Apparent ages obtained after a 24 hours delay are also reported as "greater than" even if, for weakly fading TL signals, such an age may be close to the expected finite apparent TL age. Apparent TL ages obtained after a 12 weeks delay are considered as finite with one exception. In the case of VDw, fading exceeded 20 % (6 weeks/24 hrs) and, consequently, the age is reported as "greater than or equal to".

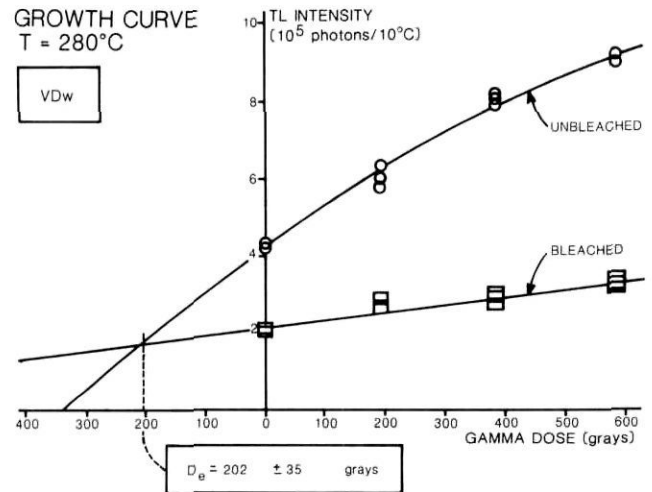


FIGURE 6. Growth curves at 280°C for sample VDw; the intersection of the unbleached and bleached curves yields the equivalent dose, D_e .

Courbes de croissance à 280°C de l'échantillon VDw; la dose équivalente D_e est déterminée par l'intersection des courbes de croissance avant et après lessivage.

2. The dose rate and the problems of water content and radon escape

The effective dose rate is the lower term in the TL age equation and it is defined as the rate at which energy is deposited in the material, with an allowance for the lower effectiveness of alpha particles for producing TL. It is expressed in $grays-year^{-1}$. In continental deposits, this energy comes from the radioactive decay of potassium 40, thorium 232, uranium 238 and 235 along with a small contribution from cosmic rays. Recent values of dose rate contributions for unit quantity of each element are given by Bell (1979) and Aitken

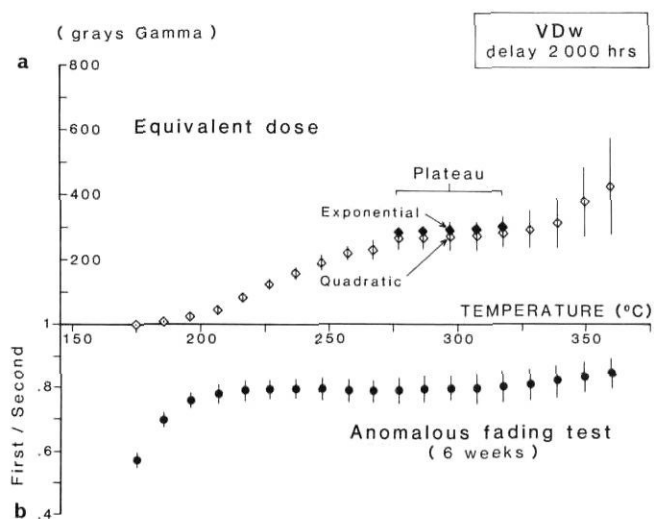


FIGURE 7. a) Equivalent dose vs. temperature for sample VDw; the equivalent doses obtained from quadratic and exponential best fits are both shown for comparison. b) Results of anomalous fading tests for this sample. Solid circles represent ratios of TL intensity (after standard artificial irradiation) left 1 day (first) after irradiation to 6 weeks (second) after irradiation.

a) Dose équivalente et température, échantillon VDw; les doses équivalentes obtenues à partir d'extrapolations exponentielle et quadratique sont comparées. b) Résultats des tests de l'affaiblissement anormal du signal TL; les cercles noirs représentent les rapports d'intensité de la TL (après irradiation artificielle standard) entre 1 jour (first) et 6 semaines (second) après irradiation.

(1985); typical effective dose rate values for sediments are in the order of a few grays per thousand years. When calculating past dose rates one must bear in mind that they may have fluctuated during geological time because of various factors, the two most critical being variations in water content of the sediment and variations in the amount of radon escape.

Water absorbs some of the radiation energy, thus decreasing the dose-rate to the sediment (Aitken, 1985). It decreases the effective dose-rate by a factor of $(1 + H \Delta)$, where (1) H is the ratio of the stopping powers of water and dry sediment for alpha and beta particles, and absorption coefficients for gamma rays and (2) $\Delta = (\text{water weight})/(\text{dry weight})$ for the sample.

When discussing dose-rate evaluations, TL workers stress the large age uncertainty that can result from an unknown past water content in the samples. For example, the apparent TL age of sample 99 RY is 86 ka, given a water content of 44 % (average value of the *in situ* and saturation water contents) as compared to 70 ka for a water content of 33 % (*in situ* value).

Estimation of the saturation water content is not straightforward in sediments because of the lack of cohesion in unconsolidated deposits. If care is not taken during laboratory saturation of the samples, the sediment may be disturbed. The saturation value is then overestimated. The following procedure was taken to minimize this effect: (1) the samples were collected with a plastic tube gently pushed into the sediment at the section site; (2) they were slowly dried in the oven (75°C during 3 to 4 days; 100°C during a few hours),



FIGURE 8. Piece of wood collected by W. W. Shilts in the fine grained, laminated lacustrine facies of Missinaibi Sediments, Hudson Bay Lowland. The present length is 41 cm; before deformation, it was 55 cm.

Morceau de bois échantillonné par W. W. Shilts dans le faciès laminé à grains fins des sédiments de Missinaibi, basses terres de la baie d'Hudson. Le morceau de bois mesure 41 cm de longueur; avant déformation, la longueur était de 55 cm.

to obtain the *in situ* value; (3) the samples were then stabilized in the tube with corks; a 5 mm hole was drilled in each cork; (4) the tube was immersed in water for 3 to 4 days; (5) the corks were removed and the outer part of the sample which could have been slightly disturbed was scraped off; (6) the saturated sample was slowly dried to obtain the saturation value. The resulting *in situ* and saturation values were then averaged for use in the age equation, since it is expected that these sediments were under ice and therefore saturated during at least half of their geological history.

Another factor may lead to an underestimation of the true water content in geological samples. The original porosity of a sediment older than the last glaciation was probably higher than the one measured after glacial loading. This reduction in sediment volume and porosity is evident on Figure 8 which shows a piece of wood reduced in length by 25 % due to glacier loading during the Wisconsinan glaciation. This wood (collected in the Hudson Bay Lowland area) was found standing vertically in the sediments (W. W. Shilts, 1986, pers. comm.). Because of all of the above, a 25 % uncertainty is attached to the water content in Table II.

Radon 222 is a gaseous product highly mobile in dry soil. Its depletion is important because 98 % of the γ contribution to the dose in the uranium decay series is produced after radon. Water enhances radon escape from minerals (e.g. Fleischer, 1983) but greatly prevents its mobility. Radon escape from a dry sample can be detected in the laboratory by comparing alpha-counts of sealed and unsealed aliquots of the same sample. In this study, radon escape was encountered in seven samples. It is tentatively assumed that this loss is counterbalanced by incoming radon from the underlying strata. Moreover, since the natural dose rate is largely dominated by potassium, radon escape values smaller than 20 % should not significantly affect the age (Aitken, 1985, p. 80).

Since a dose of alpha radiation is generally less efficient in producing TL as the same dose of gamma or beta radiation, an alpha efficiency factor, the a-value, is introduced in the age equation. It was defined by Aitken and Bowman (1975) as

$$a = X/13 S$$

TABLE II
Dose-Rates

Sample	W K ₂ O (% ± 0.05 %)	Alpha count-rate U Th ks ⁻¹ ·cm ⁻²		Water ¹ content (± 25%)	a ² value (± .01)	Rn ³ escape (%)	D _c (grays·ka ⁻¹)	Calculated dose-rate ± 10% (grays·ka ⁻¹)
LP	2.94	.437 ± .044	.178 ± .042	0.41	.143	15 ± 5	.14	3.67
MC-DES	3.86	.395 ± .040	.133 ± .037	0.30	.098	7 ± 4	.14	4.05
SP-2	2.58	.310 ± .039	.101 ± .036	0.21	.087	nd	.07	3.06
RS-2	2.91	.342 ± .060	.209 ± .057	0.25	.084	nd	.07	3.55
RS-24	3.07	.253 ± .047	.296 ± .045	0.25	.084	no	.07	3.65
SS-24	2.74	.214 ± .045	.269 ± .043	0.21	*.100	10 ± 5	.07	3.47
RS-2000	3.07	.268 ± .086	.264 ± .082	0.25	.084	no	.07	3.61
StP	2.66	.263 ± .040	.168 ± .038	0.22	*.100	22 ± 4	.07	3.17
109RY	3.98	.414 ± .057	.150 ± .053	0.44	.114	13 ± 4	.07	3.91
99RY	3.91	.241 ± .033	.277 ± .035	0.44	.114	no	.07	3.69
VP	2.70	.238 ± .052	.219 ± .049	0.31	*.100	nd	.07	3.06
VPw	2.34	.167 ± .022	.170 ± .021	0.31	*.100	nd	.07	2.53
SPI	3.44	.318 ± .086	.340 ± .082	0.34	*.100	14 ± 6	.07	4.01
VDw	3.98 (3.52) ⁴	.259 ± .048	.316 ± .046	0.33	*.100	13 ± 3	.07	4.27

Notes

1. Water content is $\Delta = (\text{water weight})/(\text{dry weight})$ in sample.
2. Data for RS-24 and RS-2000 taken from measurements on RS-2; data for 109RY taken from measurements on 99RY; *means estimated.
3. "Rn escape" is the alpha count-rate increase (in %) when the powder sample was sealed; no means less than 5%; nd means not determined.
4. Number in bracket is environmental potassium value.

where X is the number of grays of gamma radiation that produces the same TL as 1 minute of alpha irradiation from a source of strength $S \mu\text{m}^{-2}\text{min}^{-1}$.

Values were measured for samples LP, MC-DES, SP, RS and RY and a value of 0.1 was used for the remainder; these are given in Table II.

The cosmic dose rate was estimated at 0.14 grays $-\text{ka}^{-1}$ in postglacial sediments (Prescott and Stephan, 1982) and at half of this value for those sediments which have been under a thick ice-cover for part of their geological history.

The resulting calculated dose-rates are listed on Table II. They range between 3 and 4 grays $-\text{ka}^{-1}$.

APPARENT TL AGES: RESULTS AND DISCUSSION

In the course of this study, 14 samples were collected for TL dating; 19 TL dates were obtained from them. The apparent TL ages are listed in Tables III and IV. They range from 4 to 135 ka. They have been tabled according to: (1) the sampling area and (2) the unit in which they were found. Apart from the modern samples, it should be mentioned that among 19 apparent TL ages, only 5 are considered to be finite. The TL dates discussed herein carry large uncertainties which are mainly due to the estimation of the past water content.

Wintle and Huntley (1982) proposed several criteria to be met before giving a geological significance to TL dates of sediments.

(1) The first criterion is: "For each type of material and method used on it, the method should have been shown to yield zero age for recently deposited material" (*ibid.*, p. 50). The modern silt, CCTO, collected from the modern beach, gave an apparent TL age of 7 ka. The sample collected in the St. Lawrence River water (CLTO) yielded a slightly younger age of 4-5 ka. Theoretically, these samples should give a zero age. Berger *et al.* (1984) showed that the use of the R-Gamma method with a restrictive laboratory bleach could overcome this problem. However, for sample CLTO, a laboratory bleach of only 30 minutes did not reduce significantly the equivalent dose. The non-zero results may indicate that at least some of the grains were exposed to little or no sunlight in nature. In flume experiments, Gemmell (1985) demonstrated that fluvial sediments are not likely to be efficiently bleached. The possibility that some grains received no sunlight exposure has been suggested by Jungner (1983); this could happen for example if the silt grains were transported in water and sedimented with the sand grains as aggregates. In CCTO, silt aggregates can be seen under the binocular microscope. During the initial preparation of the bulk sample, prior to settling on the aluminum discs, the grain-size segregation frees these inner silts which then compose part of the measured sample. Also, the large range of light wavelengths (down to 350 nm) emitted through the optical filter that was used for simulating bleaching in the natural environment could have emptied electrons that were already trapped before deposition.

TABLE III
Apparent Thermoluminescence Ages
Pierreville

Sample (unit)	Laboratory ¹ Bleach Time (hrs)	Fading ² (% TL left)	Delay ³ (hrs)	D _e ⁴ (grays)	Dose-rate (grays-ka ⁻¹) (± 10%)	Apparent TL age (ka)
late-glacial LP (99-G)	2.0	93 ± 4 (b)	24	38.0 ± 4.0	3.67	>10.3 ± 1.7
interstadial						
SP-2 (98-B)	2.0	95 ± 3 (b)	2	105.0 ± 13.0	3.06	>34.3 ± 5.5
RS-2 (98-D)	4.0	90 ± 4 (b)	2	137.0 ± 14.0	3.55	>38.6 ± 5.4
RS-24(")	1.0	83 ± 4 (b)	24	196.0 ± 22.0	3.65	>53.7 ± 8.1
SS-24 (99-E)	1.0	84 ± 3 (b)	24	184.0 ± 20.0	3.47	>52.9 ± 7.9
RS-Q (98-D)	1.5	nd	2	217.0 ± 33.0	3.55	61.2 ± 11.0
RS-2000 (")	2.0	no (c)	2000	220.0 ± 24.0	3.61	61.1 ± 9.2
stadial						
99RY (99-C)	2.0	92 ± 4 (b)	24	267.0 ± 37.0	3.69	>72.3 ± 12.3
99RY del. (")	2.0	no (c)	2000	318.0 ± 54.0	3.69	86.3 ± 17.0
109RY (109-C)	1.0	95 ± 3 (b)	2	300.0 ± 51.0	3.91	>76.7 ± 15.0
VP (98-A)	1.0	92 ± 3 (b)	2	251.0 ± 38.0	3.06	>82.0 ± 15.0
VPw(")	4.0	no (a)	48	341.0 ± 64.0	2.53	135.0 ± 26.0

- Notes 1. All laboratory light exposures were made with a Sylvania 275W mercury "sunlamp" with a Corning 0-52 glass filter to remove wavelengths below 350 nm
2. Fading is TL left after a. 48 hrs., b. 6 weeks and c. 12 weeks of storage; no means less than 5 %; nd means not determined but no fading is expected in quartz. See Lamothe (1984) for details of measurements. Naturals were subtracted.
3. Delay is length of storage between irradiation and glowing to determine D_e.
4. D_e is weighted mean equivalent dose for the plateau over 250-350°C.

TABLE IV
Apparent Thermoluminescence Ages
St-Pierre

Sample (unit)	Laboratory ¹ Bleach Time (hrs.)	Fading ² (% TL left)	Delay ³ (hrs.)	D _e ⁴ (grays)	Dose-rate (grays-ka ⁻¹) (± 10%)	Apparent TL age (ka)
modern						
CLTO	2.0		—	18.9 ± 4.4	*4.00 ⁵	4.7 ± 0.9
	0.5		—	15.2 ± 3.4	*4.00	3.8 ± 0.8
CCTO	2.0		—	27.7 ± 10.0	*4.00	6.9 ± 2.6
late-glacial MC-Des	2.0	no (a)	48	63.5 ± 7.6	4.05	15.7 ± 3.0
interstadial StP (58-B)	2.0	no (c)	2000	87.8 ± 10.6	3.17	27.7 ± 4.3
stadial						
VDw (400-B)	4.0	79 ± 5 (b)	24	192.0 ± 24.0	4.27	>45.0 ± 8.0
VDw del. (")	2.0	10 ± 5 (c)	2000	297.0 ± 34.0	4.27	≥69.5 ± 10.5
SPI (59-A)	2.0	80 ± 2 (b)	24	240.0 ± 27.4	4.01	>59.8 ± 9.0

- Notes 1, 2, 3 and 4: see Table III. 5. * means estimated

(2) The second criterion is: "For the type of material and the method used, the method should have been shown to give correct ages for at least three suites of samples for which reliable ages have been determined independently. These should cover the time span in question" (Wintle and Huntley, 1982, p. 50).

Late glacial: LP: this sample was collected in a marine unit, suggesting a ¹⁴C age range of 9.5 to 12.5 ka. The most probable age of this unit is ~11 ka. The apparent TL age of >10.3 ± 1.7 ka is therefore geologically reasonable.

MC-DES: the apparent TL age of 15.7 ± 3.0 ka is consistent with the ¹⁴C age obtained on the *Balanus* shells embedded

in the sediment (UQ-651: $11,130 \pm 180$ years BP). The difference could be due to the uncertainties in the TL date and the deviation of ^{14}C ages from calendar ages. Also, the difference between the TL and the ^{14}C ages is similar to the apparent TL age of the modern river silt CLTO (4-5 ka), suggesting that incomplete natural bleaching may also be responsible for this apparently too large TL date.

Interstadial: The apparent TL ages obtained on the Saint-Pierre suite (SS, RS, and SP), exposed at Pierreville, have been discussed in detail elsewhere (Lamothe, 1984). They are only summarized here.

The finite TL date for the lacustrine member of the Saint-Pierre Sediments (RS-2000) is 61.1 ± 9.2 ka. This result agrees with the finite ^{14}C date measured by DeVries on pieces of wood collected from the peat layer (66.5 ± 1.6 ka; GrN-1711; Vogel and Waterbolk, 1972). However, dates obtained before 1967 may have to be disregarded since, according to Grootes (1978), contamination was present in the Groningen laboratory during these first years of radiocarbon dating. A ^{14}C date of $74\,700^{+2700}_{-2000}$ years BP (QL-198) was obtained on the same peat by Stuiver *et al.* (1978) using thermal diffusion isotopic enrichment. The TL date is believed to be coherent with this ^{14}C date since (1) ^{14}C years do not necessarily equate to calendar years because of possible fluctuations in the original ^{14}C concentration in the atmosphere; (2) the peat layer may well represent 3 to 5000 years of sedimentation (Terasmae, 1958); and (3) the large uncertainties still carried by the TL dates may themselves account for the difference.

In the Saint-Pierre area, sample StP, collected in unit 58-B, yielded a finite TL age of 27.7 ± 4.3 ka. This apparent age may be realistic even if it is much younger than the underlying peat layers. So far, only one finite ^{14}C date has been reported from these peats (67.7 ± 1.3 ka; GrN-1799; Vogel and Waterbolk, 1972). The TL sample was collected in silt overlying the uppermost peat layer. A lacuna between the peat and this silt cannot be demonstrated but may exist. A possible explanation is that the uppermost Saint-Pierre Sediments were zeroed at the end of the Saint-Pierre Interstadial, the age of which is still a matter of controversy (Lamothe *et al.*, 1983). In other words, the Saint-Pierre peat bearing sediments of the type-section were deposited 65 ka ago but the upper part of the sequence was exposed to sunlight during recycling of the sediments at the end of the interstadial. Moreover, in absence of an unusual degree of anomalous fading, it is difficult to understand how an apparent TL age would be only half the geological age for this sample.

From the above discussion, it can be concluded that the apparent TL ages reported for modern, late glacial and interstadial sediments are in relatively good agreement with the stratigraphic evidence and suggest that the optical bleaching procedures used in this program yield reasonable ages. The zero-point "error" which may be as much as 5 ka is of little significance for the older sediments.

(3) The third criterion of Wintle and Huntley (1982) is that a number of laboratory tests should be reported. Plateaux, anomalous fading, alpha effectiveness, and nonlinearity of

the TL growth curve (herein at high doses) are documented in preceding sections and need not be repeated here.

Therefore, this TL dating program partly meets the criteria listed by Wintle and Huntley (1982). Consequently, it is suggested that the apparent TL ages measured on the stadial sediments, should be geologically significant, particularly if one takes into consideration the large uncertainties attached to these numbers.

Samples 99RY, SPI and VDw yielded apparent TL ages in the order of 60 to 85 ka, that is early Wisconsinan. Since these TL ages are close to but still slightly older than the TL ages obtained on Saint-Pierre Sediments, they should be considered reliable.

Sample VP yielded two apparent TL ages, one of which is finite and in the order of 135 ka. If this result is meaningful, the underlying red till could have been deposited during the Illinoian. However, the following points need to be mentioned: (1) Only one finite apparent age is available; (2) This unit is composed of proximal rhythmites in which deposition is commonly rapid and follows grain transportation by turbidity currents. Silt-sized particles may not be efficiently bleached in this particular environment (Berger, 1985; Berger *et al.*, 1987); However, the silt laminae (VP) yielded a younger apparent age (>82 ka, *i.e.* uncorrected for anomalous fading) than the clay laminae (VPw). This age discrepancy could be explained by an overestimation of D_0 for both VPw and VP due to the short wavelengths emitted by the laboratory sunlamp combined with the fact that sample VP was not corrected for anomalous fading; (3) This could be a much older sample, or a younger unbleached one, displaying an anomalously low TL intensity, as observed by Debenham (1985) for reasons that are not yet understood. In summary, this result reflects the complexity of interpretations one can draw about an apparent TL date. This date should thus not be considered reliable.

CONCLUSION

There are a number of variables affecting the TL process itself as well as many others which are specifically related to the application of this dating technique to sediments. In their paper of 1982, Wintle and Huntley concluded that most TL dates published in the literature were unacceptable since very few methodological details were given on how the dates were obtained and which event was actually dated. They later add, "simple publication of dates alone is quite inadequate" (*ibid.*, p. 50).

This paper presents the full report of a TL dating program in which are demonstrated (1) an Early to Middle Wisconsinan age (*e.g.* 60 ka) for the Saint-Pierre Interstadial sediments, and (2) an Early Wisconsinan age (*e.g.* 60 to 85 ka) for the underlying glaciolacustrine sediments. The apparent TL ages obtained for the Pierreville Varves and the upper part of the Saint-Pierre Sediments at the type section (58) have to be confirmed.

Two critical problems encountered in this dating program must be addressed in future investigations. From the available TL literature, they seem to be of universal nature. The first

is the testing of the zeroing assumption. It is not known, at this moment, whether or not every mineral grain in each sedimentary unit has been equally exposed to sunlight. The partial bleach method using different laboratory bleaches can be used to test for this. New approaches, such as isolating different mineral fractions in sediments (Mejdahl, 1985) or "sampling" the most light sensitive traps by measuring optical luminescence (Huntley *et al.*, 1985), should also be tried.

The second major problem is the occurrence in nearly every sample of short term anomalous fading. A standard three month delay between irradiation and glowing may overcome this problem. However, for strongly fading samples such as VDw, isolation of a nonfading component (Divigalpiya, 1982; Berger, 1984) or a thermal treatment (Templer, 1985) may be needed. Such a thermal treatment has been recently used to eliminate anomalous fading in feldspar-dominated sediments (Berger, 1987). It should be mentioned that the errors produced by fading and "overbleaching" are in opposite directions and therefore at least partly cancel (Berger, 1984).

Future research should focus on a particularly well dated sequence. Modern environments in the central St. Lawrence Lowland are not likely to be suitable since they are not direct analogues to the ones that existed during most of the Quaternary. It could even be suggested that criterion 1 of Wintle and Huntley (1982) could hardly be applied in most regional work. Subtill sediments are commonly older than 25 ka. For such samples, age control is poor because the ages are close to or higher than the upper limit of the radiocarbon dating technique. Therefore, the sedimentary sequences one should sample for monitoring the TL processes are late glacial. One good example is the Champlain Sea Sediments for which the stratigraphy and sedimentology are well documented.

It is believed that due to similar geological context, the conclusions reached in this dating program should apply to most Quaternary sequences that have been documented in the St. Lawrence River drainage basin. This includes key sections such as the Lake Erie and the Scarborough Bluffs. The data reported by Berger (1984), for the Upper Thorncliffe and Sunnybrook sediments are indeed similar, in terms of TL characteristics, to the ones presented in this paper.

In conclusion, thermoluminescence dating is promising since it can yield preliminary absolute ages for clastic sediments older than the upper limit of the radiocarbon method. It can be performed on detrital grains that are ubiquitous and is not dependent on the presence of fossils or organic matter. However, TL ages reported herein are "apparent". They cannot be compared to calendar years at the present state of knowledge. They are "strictly analytical products" (Odin, 1982) which are believed to date a sedimentation event.

ACKNOWLEDGMENTS

This paper is excerpted from a Ph.D. thesis submitted by the senior author at the University of Western Ontario (1985). It was orally presented at the IVth International Conference on TL and ESR dating, held in Worms, Germany, in September 1984. This work has been financially supported through NSERC grants to A. Dreimanis (University of Western Ontario) and

D. J. Huntley (Simon Fraser University) and a FCAC-Équipe grant (Québec) to C. Hillaire-Marcel (Université du Québec à Montréal). This support is kindly acknowledged. G. W. Berger, W. M. R. Divigalpiya, C. Hearty, M. K. Nissen and T. A. Brown helped with the measurements. We wish to thank particularly G. W. Berger for discussing the data collected during this study. Figure 8 is a courtesy of W. W. Shiels (Geological Survey of Canada). A. Wintle and G. W. Berger critically reviewed the manuscript.

REFERENCES

- Aitken, M. J., 1985. Thermoluminescence Dating. Academic Press, London.
- Aitken, M. J. and Bowman, S. G. E., 1975. Thermoluminescent dating: assessment of alpha particle contribution. *Archaeometry*, 17: 132-138.
- Bell, W. T., 1979. Thermoluminescence dating: radiation dose-rate data. *Archaeometry*, 21: 243-245.
- Berger, G. W., 1984. Thermoluminescence dating studies of glacial silts from Ontario. *Canadian Journal of Earth Sciences*, 21: 1393-1399.
- 1985. Thermoluminescence dating applied to a thin winter varve of the late glacial South Thompson silt, south-central British Columbia. *Canadian Journal of Earth Sciences*, 22: 1736-1739.
- 1987. Thermoluminescence dating of the Pleistocene Old Crow tephra and adjacent loess, near Fairbanks, Alaska. *Canadian Journal of Earth Sciences*.
- Berger, G. W., Clague, J. J. and Huntley, D. J., 1987. Thermoluminescence dating applied to glaciolacustrine sediments from Central British Columbia. *Canadian Journal of Earth Sciences*, 24: 425-434.
- Berger, G. W. and Huntley, D. J., 1982. Thermoluminescence dating of terrigenous sediments, *PACT*, 6: 495-504.
- Berger, G. W., Huntley, D. J. and Stipp, J. J., 1984. Thermoluminescence studies on a ¹⁴C-dated marine core. *Canadian Journal of Earth Sciences*, 21: 1145-1150.
- Bowman, S. G. E., 1976. Thermoluminescent dating: The evaluation of radiation dosage. Ph.D. thesis, University of Oxford, England.
- Debenham, N. C., 1985. Use of UV Emissions in TL Dating of Sediments, *Nuclear Tracks and Radiation Measurements*, 10: 717-724.
- Divigalpiya, W. M. R., 1982. Thermoluminescence dating of sediments. M.Sc. thesis, Simon Fraser University, Burnaby, British Columbia.
- Fleischer, R. L., 1983. Theory of alpha recoil effects on radon release and isotopic disequilibrium. *Geochimica et Cosmochimica Acta*, 47: 779-784.
- Gadd, N. R., 1955. Pleistocene geology of the Bécancour map-area, Québec. Ph.D. thesis, University of Illinois, Urbana.
- 1971. Pleistocene geology of the central St Lawrence Lowland, with selected passage from an unpublished manuscript: The St Lawrence Lowland, by J. W. Goldthwaith. Geological Survey of Canada, Memoir 359.
- Gemmell, A. M. D., 1985. Zeroing of the TL signal of sediment undergoing fluvial transportation: A laboratory experiment. *Nuclear Tracks and Radiation Measurements*, 10: 695-702.

- Grootes, P. M., 1978. Carbon-14 time scale extended; comparison of chronologies. *Science*, 200: 11-15.
- Huntley, D. J., 1985. On the zeroing of the thermoluminescence of sediments. *Physics and Chemistry of Minerals*, 12: 122-127.
- Huntley, D. J., Godfrey-Smith, D. I. and Thewalt, M. L. W., 1985. Optical dating of sediments. *Nature*, 313: 105-107.
- Huntley, D. J. and Wintle, A. G., 1981. The use of alpha scintillation counting for measuring Th-230 and Pa-231 contents of ocean sediments. *Canadian Journal of Earth Sciences*, 18: 419-432.
- Jungner, H., 1983. Preliminary investigations on TL dating of geological sediments from Finland. *PACT*, 9: 565-572.
- Karrow, P. F., 1957. Pleistocene geology of the Grondines map-area, Quebec, Unpub. Ph.D. thesis, University of Illinois, Urbana.
- Lamothe, M., 1984. Apparent thermoluminescence age of St-Pierre sediments, at Pierreville, Québec, and the problem of anomalous fading. *Canadian Journal of Earth Sciences*, 21: 1406-1409.
- 1985. Lithostratigraphy and geochronology of the Quaternary deposits of the Pierreville and St-Pierre les Becquets areas, Quebec. Ph.D. thesis, University of Western Ontario, 227 p.
- Lamothe, M., Hillaire-Marcel, C. and Pagé, P., 1983. Découverte de concrétions calcaires striées dans le till de Gentilly, Basses-terres du St-Laurent, Québec. *Canadian Journal of Earth Sciences*, 20: 500-505.
- Mejdahl, V., 1985. Thermoluminescence dating of partially bleached sediments. *Nuclear Tracks and Radiation Measurements*, 10, 711-715.
- Odin, G. S., 1982. Individual radiometric studies of stratigraphically calibrated samples. *In Numerical dating in stratigraphy* (G. S. Odin, ed.) Wiley-Interscience, Chichester, 3-18.
- Prescott, J. R. and Stephan, L. G., 1982. The contribution of cosmic radiation to the environmental dose for thermoluminescent dating: Latitude, altitude and depth dependence. *PACT*, 6: 17-25.
- Stuiver, M., Heusser, C. J. and Yang, I. C., 1978. North American glacial history extended to 75,000 years ago. *Science*, 200: 16-21.
- Templer, R. H., 1985. The removal of anomalous fading in zircon. *Nuclear Tracks and Radiation Measurements*, 10: 531-537.
- Terasmae, J., 1958. Contributions to Canadian Palynology. Geological Survey of Canada, Bulletin 46, 35 p.
- Vogel, J. C. and Waterbolk, H. T. (1972): Groningen Radiocarbon Dates X. *Radiocarbon*, 14: 6-110.
- Wintle, A. G., 1973. Anomalous fading of thermoluminescence in mineral samples. *Nature*, 245: 143-144.
- 1982. Thermoluminescence properties of fine-grained minerals in loess. *Soil Science*, 134: 164-170.
- Wintle, A. G. and Huntley, D. J., 1980. Thermoluminescence dating of ocean sediments. *Canadian Journal of Earth Sciences*, 17: 348-360.
- 1982. Thermoluminescence dating of sediments. *Quaternary Science Reviews*, 1: 31-53.
- Zimmerman, D. W., 1971. Thermoluminescent dating using fine grains from pottery. *Archaeometry*, 13: 29-52.