Geoscience Canada

Dating Methods of Pleistocene Deposits and Their Problems: I. Thermoluminescence Dating

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Volume 5, numéro 2, juin 1978

URI: https://id.erudit.org/iderudit/geocan5_2art01

Résumé de l'article
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Major problems to be investigated concern the fact that Pleistocene sediments do not always have a well established zero point for TL, and post-depositional changes may also have a profound effect on the TL.

Quartz is one of the most suitable materials for TL dating known so far, and its use has been investigated recently at the Institute of Geology in Tallin. The best results have been obtained by using quartz grains 100-140 μm in size, after their outer layer has been removed by HF to exclude the TL caused by «-radiation.

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Dating Methods of Pleistocene Deposits and Their Problems: I. Thermoluminescence Dating

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Summary
Thermoluminescence (TL), is now widely used in archeology for the absolute dating of ancient pottery. During the last decade, particularly in the USSR, it has also been applied with mixed success to the dating of Pleistocene loess, buried soils, glacioluvial, glaciolacustrine and marine deposits, and even tills. The purpose of this paper is to stimulate investigation of the application of this method to geological problems. Absolute dating methods beyond the radiocarbon dating range are urgently needed, particularly in the Pleistocene stratigraphy of North America. Therefore, even those relative or semiabsolute dating methods which have the potential eventually to provide reliable absolute dates, have to be investigated. The TL dating method belongs to this category, with its dates ranging between 10^3 and 10^6 years B.P.

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Introduction
Shortage of either absolute or relative dates beyond the radiocarbon dating range has been hampering the development of the Pleistocene stratigraphic schemes and their correlations in the continental areas. This handicap has been felt particularly in central and eastern North America, where new stratigraphic information becomes available every year. Westgate et al. (1977) have expressed their concern in following statement in the status report on Quaternary geology - 1976:

"Geochronological control for Quaternary deposits older than 50,000 years – the limit of the 14C dating method - is very poor in Canada: the application of radiometric (e.g., K/Ar, fission track) and other dating techniques (e.g., amino-acid method), tephrochronological and paleomagnetic methods to Quaternary stratigraphic problems is still very limited. We feel this may well reflect poor communication between the Quaternary geologist, geophysicist and geochronologist."

The techniques currently used in Canada will not be discussed here; instead, we would like to draw attention to a controversial method that has not yet been applied to dating terrestrial Pleistocene deposits in North America. This is the thermoluminescence dating method, abbreviated in this article as TL.

A Brief History of TL Dating
The phenomenon of TL has been known to physicists for a long time. It is used as a tool for investigating the optical properties of defects in solids, and has been widely used to measure the radiation dose given to cancer patients. TL has been applied also in mineral exploration and rock mechanics (McDougall, 1964).

Attempts have been made to use the TL method for the dating and correlation of geological events, initially in pre-Quaternary stratigraphy, e.g., by Daniels et al. (1953) on this continent. For a list of references up to 1966 see Groger (1968) and Sheikoblyas (1973); for some more recent studies see Aitken and Aldred (1972), Aitken and Fleming (1972), Goksu et al. (1974), Hutchinson (1968), Tite (1968), Wintle (1974), Zeller (1968).

During the last two decades, several groups have studied its use in dating archeological objects, particularly pottery. An excellent account of this work can be found in the book by Aitken (1974). According to Aitken (1974), the accuracy of the TL dating of ancient pottery is presently somewhat better
than 10 per cent of the age, and it should improve to about 5 per cent. Since the autumn of 1977 a quarterly newsletter, *Ancient TL*, has been helping to exchange practical information on TL dating. It has been published by the Center for Archaeometry, Washington University, St. Louis, Mo.

The Pleistocene researchers of North America have not yet shown much interest in TL as a geological dating method, and, to our knowledge, the only recently published report is by Huntley and Johnson (1976) on TL "as a potential means of dating siliceous ocean sediments." Much more attention has been paid to its application to Quaternary chronology in eastern Europe, as shown by the references listed below (see also Karasewsky, 1974). The TL method has been widely used for dating Quaternary sediments in the USSR. The pioneer research was done particularly by V.N. Sheklovsky at the Institute of Geology of the Academy of Sciences of the Ukrainian SSR during the last 15 years. Sheklovsky and Morozov (1963, 1969) reported briefly their method and the basis for TL age determinations in loess deposits, including buried soils and some other related sediments. Descriptions of this TL dating method were given by Morozov (1968) and Sheklovsky (1971, 1973, 1974a).

The principal mineral used by him was quartz in the particle size range of 0.05 to 0.005 mm, but its extraction from loess where quartz constitutes about 85 to 90 per cent, was not described. The glow peaks were observed at 120°C. The TL dates determined by Sheklovsky (1974a, 1974b) ranged from 25,000 to 30,000 years B.P. to 923,000 ± 160,000 years B.P., but he considered it possible to extend them to 1.5 to 2 million years B.P.

Zubakov and Kotchegura (1973) are among the first users of the dates besides Sheklovsky and his co-workers. They have attempted to evaluate the TL dates, by comparing them with data from other methods. They note that about 200 Pleistocene samples have been dated by this method in the USSR up to 1972. Mainly from the Ukraine and southwestern Siberia. After a comparison of the TL age determinations with the biostratigraphic and paleomagnetic data of the sections investigated, particularly from the Pontic-Caspian region, they make the following conclusions: (a) the TL dates agree well with the C-14 dates, e.g., 36 to 45,000 years B.P. by TL, as compared with 34 to 58,000 years B.P. by radiocarbon dating; (b) the TL dates below one million years correspond closely to K/Ar dates of correlative paleomagnetic sites, with the TL dates being five to 10 per cent younger; (c) the TL dates are, however, about half as young as the K/Ar dates of the 1.5 to 2.4 million year range B.P.


Another volume edited by Zubakov (1974a) refers to the TL dating method and lists many dates in several of its chapters, mostly of them written by Zubakov, Aleshinskaya, Kochegura, Sheklovsky, and Sudakova. They do not differ much in their conclusions from those listed in the preceding paragraph. Critical remarks about some of the TL dating problems, however, appear in the chapters written by Arkhipov et al. (1974) and Veklich (1974), the latter considers the TL dates to be approximate, because of imperfections in the methodology and instrumentation.

Many Pleistocene stratigraphers, including the authors of this review, have been hesitant to refer to TL dates in their stratigraphic schemes. However, it has to be admitted that wherever several samples have been dated from individual sections or from composite geological profiles, the dates fall in the right chronological order. Further, they agree laterally, e.g., in the Likvin (Chekalin) profile (Fig. 1) of River Oka (Sudakova, 1973; Sudakova and Il'ichev, 1974), in the sections of the Yaroslavl area (Aleshinskaya et al., 1973; Sudakova and Il'ichev, 1974), and in the correlation of several sections of western Siberia and Altai mountains (Svitoch et al., 1973; Zubakov, 1974b).

Good agreement also appears in the relative vertical order of the six TL dates in the Tskur and Veseloye sections from the Azor Sea area (Zubakov et al., 1976, Fig. 3) where the dates range from 40,000 ± 5,000 to 1,200,000 years B.P. In order to test their absolute values, Zubakov et al., (1976) compared the paleomagnetic profile of the Tskur and other sections where TL age determinations and paleomagnetic investigations have been done concurrently, with the paleomagnetic scale of Cox (1969). and paleopedologic investigations in the adjoining areas. By doing this, they arrive at the conclusions that most of the TL dates of these sections are probably too young, and the previous stratigraphic interpretation in Zubakov and Kochegura (1973) may not be correct. They now prefer the following interpretation: that the paleomagnetic reversal formerly dated by TL, as about 285,000 ± 48,000 years B.P., is actually right underneath the Brunhes-Matuyama palen-
magnetic boundary of 690,000 years B.P. (Cox, 1969). The TL date of 1,200,000 years B.P. is at the boundary of reversed over normal magnetism which is probably the 2,430,000 year old Matuyama-Gauss boundary of Cox (1969).

Thus it appears that the TL dates presented or discussed by the above USSR authors are reliable for relative age determinations and for lateral correlations of lithologically similar stratigraphic units. However, their absolute values may be questioned, and the older age values, in particular, appear to be too young. The reason for this may be in the lack of rigour in the experimental techniques as pointed out recently by Hüt and Raukas (1977). In order to obtain more reliable TL dates, TL dating procedures have to be improved, by doing more research on the fundamental physics of the TL dating methods. Presently, such theoretical investigations are in progress at the Physics Department of Brookhaven National Laboratory, Upton, N.Y. (Fuller and Levy, 1977), the Department of Physics, Simon Fraser University, Burnaby, B.C., the Institute of Geology of the Academy of Sciences of the Estonian SSR in Tallinn (Hüt et al., 1977a, 1977b), at the Moscow State University (Vlassov and Kulko, 1976), and a new approach is being attempted at the All-Union Geological Scientific Research (VSEGEI) Institute in Leningrad (Pavshukov et al., 1976). The encouraging preliminary results obtained suggest that further theoretical investigations of the TL method are worth continuing.

The TL dating method, if further refined and properly applied in dating geological objects, may become a valuable tool in Quaternary geochronology. In order to create more interest in improving this dating method, brief discussions of its basic principles, dating techniques and some problems encountered will follow.

**TL, its Origin and Use for Dating**

The phenomenon of thermoluminescence has been discussed in many publications, e.g., Daniels et al. (1953); Fleming (1971), Levy (1973); Aitken (1974); Huntley and Johnson (1975); Hüt and Raukas (1977). Therefore it will be reviewed here only very briefly, with emphasis on those matters which apply to geological age determination.

If a geological sample has been irradiated, with γ-rays for example, and subsequently heated, light is emitted as a function of temperature as shown in Figure 2. This is called a "glow curve", and it is characteristic of the material heated and its radiation history; the older the sample in the TL dating range, the higher is the intensity of the glow curve (see Fig. 1 in Aitken, 1974 or Fig. 3 in Huntley and Johnston, 1976). However, the relationship of peak light intensity to age is not linear (see, for instance, Fig. 4 in Huntley and Johnston, 1976), and it depends upon various factors, many of them not yet well understood.

The radiation which causes the thermoluminescence comes from the naturally occurring radioactive isotopes (Table I). The α-radiation has a range of only 22 μm (Han and Ralph, 1971), so that it may come from the radioactive isotopes both within the sample itself, and the layer 22 μm thick surrounding the sample. The β-radiation has a range of two mm, so that it may derive, similarly, from the radio-active isotopes, in the sample, and also from the thin layer of material, up to two mm radius around the sample. The γ-radiation is very penetrating, so that most of it comes from the material surrounding the sample: 90 per cent of it from within a radius of 30 cm. A detailed discussion of the determination of sample radioactivity and dose has been given by Aitken (1974) and will not be repeated here.

Thermoluminescence may also be caused by other processes, such as pressure (piezo-TL), friction (tribo-TL), light (photo-TL) and chemical reactions (chemi-TL). For dating purposes, these other contributions are sources of error, and Aitken (1974) suggests ways to remove them.

The "TL Age" of a sample has been defined by Aitken (1974) as follows:

\[
\text{TL Age} = \frac{\text{(natural TL)}}{\text{(TL per unit dose)}} \times \text{(dose per year)}
\]

The unit of measurement of dose is the rad (Radiation Absorbed Dose) which is defined as the absorption of 100 ergs per gram.

The natural TL is measured by heating the sample and recording the glow curve. The TL per unit dose is measured by subsequently exposing the sample to a known dose from an artificial radioactive source, and then measuring the induced glow curve.

**Figure 2**

Typical TL glow curves of quartz from three sites in the USSR: (A) Karukula, Estonia, (B) Lake Tshorkan, Kerch Peninsula, (C) Pasva, Arkhangelsk District.

**Table I**

Radioactive decay schemes of U, Th, and K
There are two ways to measure the dose given per year. First, we can determine the radioactive content of the sample by chemical means, and subsequently calculate the dose from the data given in Table I. Alternatively, we can place a sensitive dosimeter at the site from where the sample was taken, and hence measure the dose directly. At the low dose rates involved, e.g., 100 millirads per year, the dosimeter has to be left in place for a year or so. It is essential that the dosimeter be placed in exactly the same location, and in the same surroundings as the sample.

Sample preparation is of prime importance; if reliable results are to be obtained. Many minerals give thermoluminescence signals, and each has different dosimetry. It is necessary therefore to isolate a particular mineral from the sample before taking the glow curve, so that the experiment is carried out on this known mineral. Thus far, the most useful candidate for this purpose seems to be quartz. Quartz usually does not contain radioactive impurities, such as U and Th, so that the radiation causing the thermoluminescence all comes from the radioactive elements in the material surrounding the quartz inclusion. However, if the quartz used contains radioactive impurities, its internal dose rate (4x + 6) also has to be considered.

The techniques of absolute dating of Pleistocene deposits and soils have been strongly influenced by the methods of the "Oxford school" used for dating ancient pottery, as their TL age determinations can be checked against known archaeological dates. They have been described, with pertinent references, by Aitken (1974), and will not be repeated here.

Problems in the TL Dating of Pleistocene Sediments

The Zero Point. Pottery, burnt stone and volcanic rock all have one thing in common: they were all heated to a high temperature at a particular moment in time, so that TL dating measures the time between this initial heating, the zero point, and the present day. The zero point for Pleistocene clastic sediments is not so well defined. If TL dating of Pleistocene materials is to be reliable, it is necessary to ensure that the zero point for the dosimeter used, e.g., quartz, coincides with the time of deposition of the sediment. It may be possible to relax this very stringent condition if the sample has been weathered at some time in its history, because sunlight will also empty the electron traps, thus providing a new, secondary, zero point, e.g., for the time of pedogenic soil formation.

Shelkopylas (1971), Aitken (1974), Sudakova and Illichiev (1974) have drawn attention to the changes in the TL produced by exposing samples to sunlight. Hutt et al. (1977a) report that the exposure of a quartz sample to diffused daylight for three days had decreased its TL by 50 per cent.

Shelkopylas (1971), Sudakova and Illichiev (1974), Goksu et al. (1974), and Huntley and Johnson (1976), suggest that increase in temperature participates in eradicating the primary TL, but no experimental data are given. The first two papers cited above suggest also, though without supporting data, that mechanical comminution of mineral grains produces a similar effect. And Morozov (1976) claims that confined pressure also reduces the TL stored by quartz, but this phenomenon has to be further investigated as pressure may cause preo-TL.

In order to investigate the effect of natural processes occurring during the sedimentation of a glacioluvial deposit, upon the pre-depositional TL of quartz, two of the authors (Hutt and Raukas, 1977) have studied quartz extracted from a sample taken from an esker near the front of a readvancing glacier on Spitsbergen. In measurements carried out in a very pure nitrogen atmosphere, no TL was observed. Those factors which have participated in the eradication of the TL need further investigation.

Post-Depositional Changes. Several changes may occur in the material to be dated. After the zero point has been attained, these have to be considered when interpreting the TL dates. Thus, for example, ground water may reduce the content of radioactive isotopes and cause seasonal fluctuations of the level of radioactivity. A further complication arises, because the decay scheme of uranium (Table I) includes the gas radon-222 with a half life of 3.84 days. Radon is chemically inert, and so it may readily diffuse away from the sample. As 98 per cent of the gamma dose rate of the U-chain comes from the members of this chain beyond radon, considerable loss of y-radiation may result. This source of error, combined with the influence of water content in soils, is discussed by Fleming (1971, p. 336), and he suggests that in situ measurements by TL phosphor capsulars be made to determine at least some of the possible loss of y-radiation.

Quartz Versus Polyminerallc Specimens as Dosimeter for Pleistocene Materials

Quartz has been investigated by archeologists, whose time scale is very short by geological standards. Detailed studies of its properties as a dosimeter for long time intervals are needed. It is also necessary to study quartz from Pleistocene deposits of different origins, because one may expect some variations, e.g., in the type and concentrations of impurities and crystal defects, which will lead to differences in the behaviour of the TL, as found by Fuller and Levy (1977) on quartz samples of different origins. Some of these variations are reported and discussed by Hutt et al. (1977a), who investigated some 20 quartz samples from Estonia; see also the differences among the glow curves of three different specimens in Figure 2. Though quartz is an obvious candidate for a dosimeter, other minerals should not be neglected. Huntley and Johnson (1976), for instance, have used silicoflagellates shells, which consist of opal.

Shelkopylas (1971) was one of the first to emphasize the suitability of quartz as a dosimeter in the TL dating of Pleistocene deposits, but without special investigation of its dosimetric properties. He dated mainly loess, loamy soils rich in quartz and glacio-aquatic deposits (Shelkopylas, 1971, 1973, 1974a, 1974b). However, he and his school did not always separate quartz from the other minerals admixed. Errors in the technique used by the Shelkopylas school have been discussed by Hutt and Raukas (1977), and two most significant points of general interest will be mentioned here.

(1) Polyminerallc specimens should not be used for TL dating, because they are mixtures of minerals each of which has its own individual TL response; for example, the TL sensitivity of feldspar to y-radiation is about 10 times greater than that of quartz, but some varieties of feldspar lose 40 per cent of the stored TL information in three months.
(2) Shekopylas and his school are measuring the effect of the α-component of radiation only, though this radiation affects the TL storage process of quartz very little. The sensitivity of quartz to α-radiation is about 8 to 10 times less than its γ- and β-sensitivity and it plays no role in the surface activation of large particles only (about 100 μm). For these and other reasons (Hütt and Raukas, 1977), we think that some of the problems mentioned, e.g., in the evaluation of the Shekopylas’ TL dates of the Tsukur section by Zubakov et al. (1976) are at least in part due to the techniques used.

Because of the scepticism of many Pleistocene stratigraphers concerning the application of TL to the absolute dating of Pleistocene sediments, Hütt has been re-investigating the thermoluminescent and dosimetric properties of quartz extracted from sands of aquatic or glacio-aquatic origin, in view of its application as an age cell of memory of such sediments (Hütt et al., 1977a). The technique used by her (Hütt et al., 1977b) is based on the quartz inclusion method of Fleming (1970); a few grams of quartz grains of 100 to 140 μm size, with their surface removed by HF to exclude the effect of α-radiation are used for the TL measurements. The procedure is described in Hütt et al. (1977b) By using this method and continuing the investigation of the dosimetric properties of quartz, Hütt et al. (1977b) hope to be able to date Pleistocene sediments in the range of 10^3 to 10^5 years B.P. Dating of samples from several sites are in progress where the TL age determinations can be compared with those obtained by other absolute dating methods.

Conclusions

In view of the exploratory work carried out at various TL laboratories of the USSR and at Simon Fraser University in Canada during recent years, we consider that the application of the TL dating method to Pleistocene subaerial and subaquatic sediments, and paleosols has shown promising results. This is in spite of several methodological problems and uncertainties about the past history of the sediments to be dated. We are currently convinced, from some pilot studies at the Institute of Geology in Tallinn and elsewhere, that the technique of quartz inclusions is most suitable for TL age determination in geology. At the same time, it is desirable to test also other materials, for example calcite, opal, etc.

The Pleistocene sections comprise not only deposits of various genetic types, but many of them have also been affected by glio-dynamic processes, which complicate their dating. Tills are especially difficult objects for TL dating as they may form under quite different geological conditions than other sediments, and the destruction of the pregenetic TL in case of different till types requires special investigations.

In spite of various experimental problems encountered in TL dating during the past years, most dates obtained are in the right chronological order and the TL method is, at least, applicable for relative dating in the range of 10^3 to 10^5 years B.P. Under optimum conditions, some TL dates agree well with absolute dates obtained by other dating methods, while others appear to be too young.

In the next few years, it is important to date many samples of known ages but different origins, to continue investigating the theoretical background of the TL dating method and its sources of errors, and also to develop more sophisticated apparatus. This will enable us to consider more objectively the use of the TL method for dating Pleistocene deposits of various origins. However, we should not forget that further study of the mechanism of formation of Pleistocene deposits is also of great importance since without considering the peculiarities of their formation, for instance the great variety in the formation of till, the most accurate measurements of their TL will be meaningless.

Acknowledgements

We wish to acknowledge the support rendered by the Institute of Geology of the Academy of Sciences of Estonian SSR, and the national Research Council of Canada grants A-4216 to A. Dremanis, and A-4724 to P. W. Whippney.

We are grateful to D. J. Huntley, P. F. Karrow, S. C. Porter and A. G. Wintle for the critical reading of the original lengthier manuscript and their constructive comments.

The paper is a contribution to the IUGS-UNESCO International Geological Correlation Program, Project 73-1-24 “Quaternary glaciations in the northern hemisphere”.

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