SYMBIOSIS BETWEEN GEOCHRONOLOGISTS AND QUATERNARY GEOSCIENTISTS

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Abstract: There is an apparent conflict between the high output of absolute dates of automated devises installed in geochronological laboratories and the growing need of an individual and thorough interpretation of absolute physical dates determined for geoscientists and archaeologists. This conflict can only be solved by a symbiotic co-operation with universal geochronologists. The benefit will be an increase of the reliability and precision of the geochronological time marks, an improvement of the dating methods and an extension of the fields of application for the many absolute dating methods based on cosmogenic isotopes, radiation damage or uranium–series disequilibrium

1. SITUATION OF THE ABSOLUTE GEOCHRONOLOGY

The present situation of the absolute geochronology is determined by diverging trends. One trend is characterised by a rapid progress of analytical and measurement throughput of modern dating techniques. Examples are represented by the accelerator mass spectrometers (AMS), which deliver up to 3500 ^{14}C dates per year, automated luminescence readers, which allow up to 100 OSL/TL analyses per week, and high-capacity ICP mass spectrometers (inductively coupled plasma quadruple mass-spectrometer), which may enable high rates of thorium and uranium isotope determinations in near future. The increased output of geochronological results degrades absolutes dates to an easily accessible though costly consumption product for Quaternary geologists, geographers and archaeologists.

The other trend is reflected in the growing number of publications with, often, large series of absolute dates and frequently superficial interpretation due to an insufficient understanding of basic principles of the applied physical dating methods. This negative development of scientific quality of geochronological publications is strengthened by the usual funding policy to assess scientific efforts solely on the number of papers rather than on their quality. Papers which contain comprehensive descriptions of sampling sites, details of field work, applied physical and chemical laboratory protocols, complete lists of laboratory results used for the interpretation, details of their statistical treatment and finally of models used for the interpretation have become rare. A reduction of a number of papers in favour of their quality will enable to avoid wasting of time for writing fragmented papers and gain time to strengthen the scientific dialog.

The theoretical and analytical stuff of the absolute dating methods is already so extended that the absolute geochronology represents an own geoscientific discipline within environmental physics. It requires full scientific attention and enthusiasm of young physicists and geochemists in order to become once an expert in absolute dating. Most frequently, young geochronologists specialize on one or seldom two methods – radiocarbon, TL/OSL, ESR, or $^{230}Th/U$. In contrary, universal geochronologists are required to bridge the interdisciplinary gaps between the wide palette of methodically different geochronological methods. Only such experts are able to explain deviations of absolute dates obtained from the same sample as a result of different basic principles of the applied methods. Single dating methods may become defamed due to an insufficient knowledge of the methodical peculiarities. One example is the ^{14}C method applied to date calcrete (Eitel and Zöller, 1997). Later, complementary radiocarbon, TL and $^{230}Th/U$ demonstrated the specific potential of each of these dating methods (Geyh and Eitel, 1998). This example once more confirmed that any improvement of an absolute dating method requires parallel application of many methods for the absolute age determination. Multidisciplinary approaches are indispensable.

There is a challenge for geochronologist to become a universal expert in absolute age determination besides his own specialisation in one dating method. He must be able to improve absolute dating methods, to extend the range of application, to explain methodically deviating dates and to estimate the minimum and maximum ranges of the actual age of the sample.

Geoscientists, on the other side, nowadays cope with a wealth of complicated analytical techniques and new computer programs while their primary task of extensive and minute field work has not shrinked. Meanwhile many of them overtax their ability to handle also the multifaceted field of absolute geochronology. There is a challenge for geoscientists and archaeologists to assess their own scientific capability. They are best educated to explore suitable sites for absolute datings and to improve the selection of suitable samples rather than to overtake strange physical, mathematical or chemical jobs necessary for any reliable interpretation of raw geochronological data (Geyh and Schleicher, 1990; Wagner, 1995).

A solution of the described problems is the symbiotic co-operation between absolute geochronologists, geoscientists and archaeologists. It supports the methodical development of various dating methods, a more sophisticated selection of suitable dating material and a thorough handling of absolute dates. The following paragraphs deal with aspects related to the improvement of special dating methods.

2. SPECTRUM OF ABSOLUTE DATING METHODS

The manifold origin of differently preserved Quaternary deposits with a variable demand in age resolution to explore earth's history is the result of multiple changes between glacial and interglacial climates. Only a large spectrum of absolute dating methods can do the ensuing task (Geyh and Schleicher, 1990; Wagner, 1995). Each method has its own potential and limits, is solely applicable to specific kinds of datable material and covers a limited dating range with variable precision. Absolute dating of Holocene material, for instance, has usually to be more precise than that of the older Pleistocene material; interglacial and interstadial material is chiefly of biogenic origin which requires other dating methods than glacial material of its predominant clastic nature.

In any case, the radioactive decay is the common chronometer for all absolute dating methods (**Fig. 1**). Chronostratigraphic dating methods e.g. based on geomagnetism or a stable isotope composition of ice or calcareous marine sediments (MIS) are indirectly related to the radioactive decay. The corresponding chronologies are calibrated with radiometric dates. Chemical dating methods using e.g. amino-acid or obsidian hydration analyses solely yield relative dates which may have, however, an important potential for stratigraphic differentiation. But they seldom yield reliable local chronologies although calibrated with absolute dates. Some of chemical dating methods, as e.g. the cation-exchange method, are questioned at all (Bierman and Gillespie, 1994).

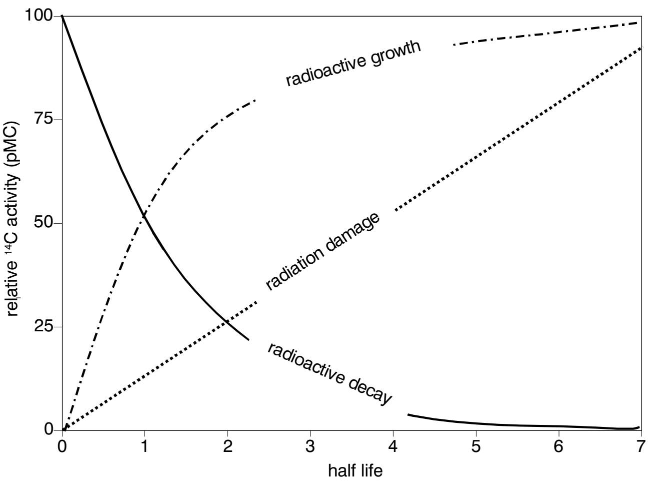

Fig. 1. *Three fundamental absolute chronometers are based on the radioactive decay (^{14}C, ^{32}Si), the radioactive growth ($^{230}Th/U$), and the constant rise of radiation damage by environmental radiation (TL/OSL, ESR).*

Many of the radiometric dating methods are well established since decades (Geyh and Schleicher, 1990; Wagner, 1995). In spite of this, the basic principles as well as the scale of applications have still a potential for an improvement, especially if geochronologists, geoscientists and archaeologists closely co-operate with each other.

Dating with cosmogenic isotopes

Beside the well-known isotope ^{14}C, the cosmogenic radioactive isotopes ^{10}Be, ^{26}Al, ^{32}Si, and ^{36}Cl cover a growing field of geochronological applications. Still restrictedly used isotopes as ^{32}Si may unpredictably gain attention when the analytical or measurement techniques are improved or new fields of applications are found (e.g. Morgenstern *et al*., 2001).

^{14}C dating

The ^{14}C method is the most widely applied and well-known absolute chronometer in Quaternary geology and archaeology. It is mainly used for dating organic matter, but also secondary carbonate is a suitable material. The corresponding samples preferentially were formed in warm and wet climatic periods.

Recent applications are dominated by the AMS technique (accelerator mass spectrometry). Numerous of the related papers deal, however, with the rediscovery of phenomena which were already described several decades ago based on conventionally determined radiometric dates. Many geoscientists apply this technique under the wrong assumption that AMS dates are superior to radiometric dates. They do not realise that both conventional and AMS techniques belong to the same dating method though they have different potential and limits. Two examples may elucidate the situation.

Olsson (1973) found that material with little carbon contains often a large proportion of fossil macrofossils resulting in apparently too old conventional ^{14}C dates. The corresponding AMS dates either are precise and reliable or completely erroneous. Hence, several AMS dates have to be determined in such cases. Another example belongs to ^{14}C dating of various fractions extracted from peat or soil. Widely differing dates are obtained (e.g. Shore *et al.*, 1995). AMS laboratories carried out similar studies without new insights in the well-known phenomenon.

There is no dispute that a large number of applications require AMS dating, especially if only small sample sizes are available. In all cases, however, where sufficient sample material can be provided, it should be seriously assessed which dating technique provides the best suiting absolute dates at minimum costs.

The case study below shows how a close co-operation between archaeologists and geochronologists gained new insights in the principles of even an old, well-established absolute dating method as ^{14}C. Usually it is assumed that the ^{14}C chronometer starts to run with the death of an individual. In contrary, in the case of collagen extracted from bones the ^{14}C clock of these proteins starts already slightly after the end of the puberty when the incorporation of the main proportion of proteins ceases. By this, a calibrated conventional ^{14}C age of the collagen extract of an old person is by up to 40 years apparently too large (**Fig. 2**). This finding is important for ^{14}C dating of historical events by means of bone collagen (Geyh, 2001).

An actual demand is the extension of the calibration of the ^{14}C time scale beyond 20,000 BP (e.g. Kitagawa and van der Plicht, 1998). This task can only be satisfactorily solved if geoscientists explore new geological sites with precisely and accurately datable material (as e.g. laminated lake sediments) of sufficient autochthonous, terrestrial, organic matter.

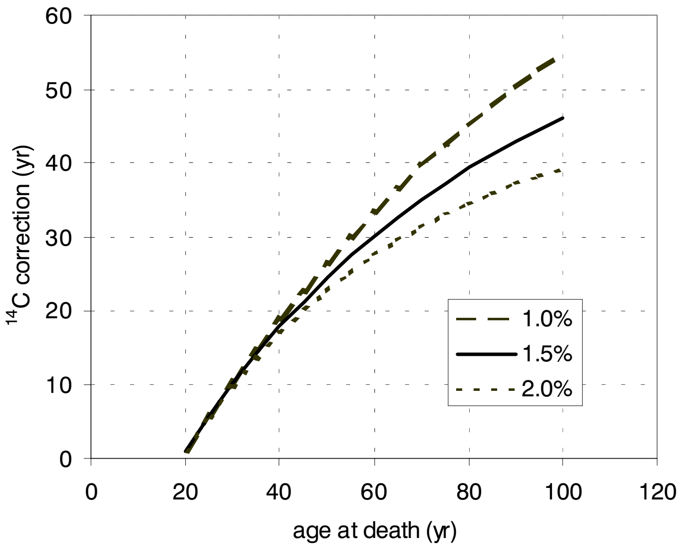

Fig. 2. The difference between the year of death and the correction term for calibrated conventional ^{14}C dates of collagen. Different carbon exchange rates in bone proteins are considered of which 1.5 %/yr is the most reasonable one (Geyh, 2001). The year of death of a 65 years old man is overestimated by 34 years.

^{32}Si and ^{210}Pb dating

^{32}Si dating has successfully been applied to date ice (Nijampurkar *et al.*, 1982) and sponge in the age range of up to 1000 years. Attempts to apply this method also to groundwater (Lal *et al.*, 1970) failed. Morgenstern *et al.* (2001) presented now new ^{32}Si dates from shallow shelf sediments from Bangladesh. They show that a natural sedimentation rate prior about 1900 AD was four times lower than that of the last 100 year derived from ^{210}Pb dates (**Fig. 3**). Large-scale deforestation in this region started in the 19th century and increased erosion and accumulation. This case study elucidates the great potential of the ^{32}Si dating method. Its dating range of up to 1000 years comprises the period of the dramatic natural climatic change from the warm Medieval period to the very cold Little Ice age and the continuos impact of industrialisation about 1850 AD ago. Based on this experience, future palaeoclimatic studies of the last millennium should include both ^{32}Si and ^{210}Pb datings. An expected success of the most recent efforts to measure ^{32}Si directly by AMS rather than indirectly via ^{32}P by liquid scintillation counting (Morgenstern *et al.*, 2001) will open a wide field of applications in paleoclimatology, geosciences, and archaeology for the ^{32}Si dating method.

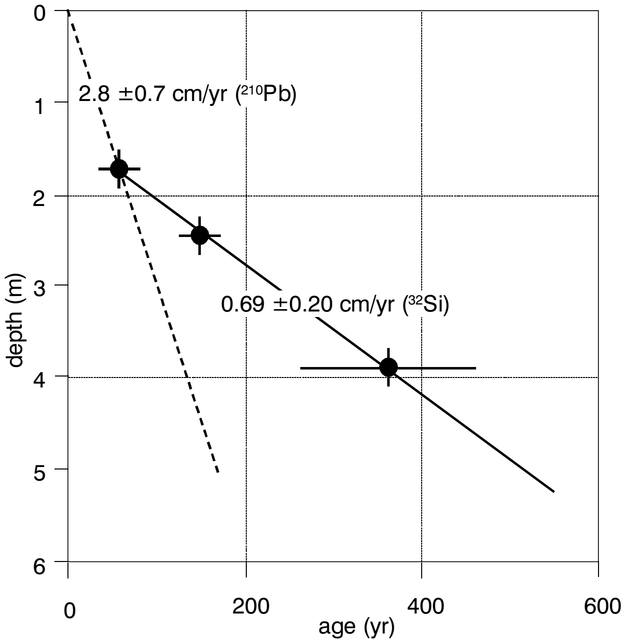

Fig. 3. ^{210}Pb and ^{32}Si dates of shallow sediments from the Shelf of Bangladesh show that the deposition rate during the last 100 yr was four times larger than that of the preceding period. Deforestation during the last two centuries intensified erosion and sedimentation (Morgenstern et al., 2001).

Surface-exposure dating

Surface exposure dating is based on chronometers (Lal, 1988; Dorn and Phillips, 1991) which were first proposed for ^{36}Cl in the fifties (e.g. Davis and Schaeffer, 1955). Two kinds of physical processes produce stable (^{4}He) and radioactive isotopes (^{10}Be, ^{26}Al, and ^{36}Cl): spallation and capture of thermal neutrons. The resulting spatial isotopic distributions (**Fig. 4**) as well as deviating formation rates of different isotopes within the rock in time (**Fig. 5**) allow absolute dating of up to about one million years.

The development of the AMS techniques for analyses ^{10}Be, ^{26}Al and ^{36}Cl in small samples and of mass spectrometers for ^{4}He was the prerequisite to develop this new dating method for applied geosciences. Erratics with permanently exposed surfaces in fixed positions are needed to determine the absolute ages of their exposure and to estimate erosion rates. The potential of these dating methods has been first demonstrated for objects in Antarctica. European geoscientists (e.g. Ivy-Ochs, 1996) started to apply these methods to date the exposure of extended mountainous plateaux from continental ice sheets as in Tibet which might have played a decisive role in the global climatic changes during the Quaternary.

The collection, selection and chemical handling of samples are very complicated and require considerable improvements before being standardised. Chemical treatment of the samples in the laboratory is far from that, still very tricky and requires excellent geochemical and mineralogical knowledge and skill. Efforts are necessary to assess the interrelationship of scientific experience and the reliability of the absolute dates. A close co-operation between geoscientists and geochronologists is indispensable.

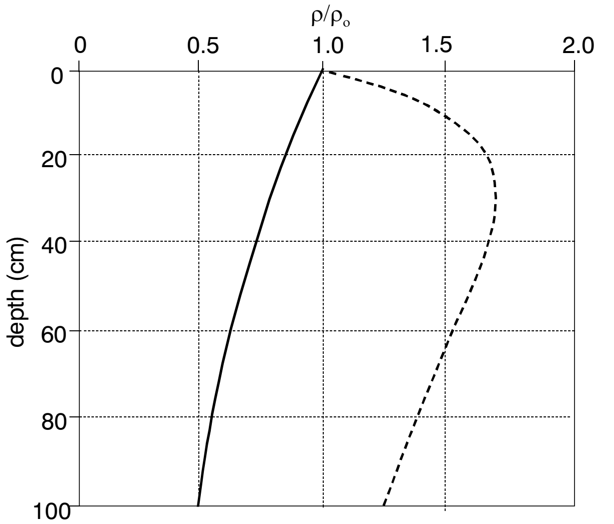

Fig. 4. *Spatial distributions of isotopes in permanently exposed rock profiles produced either by spallation or capture of thermal neutrons (dotted line).*

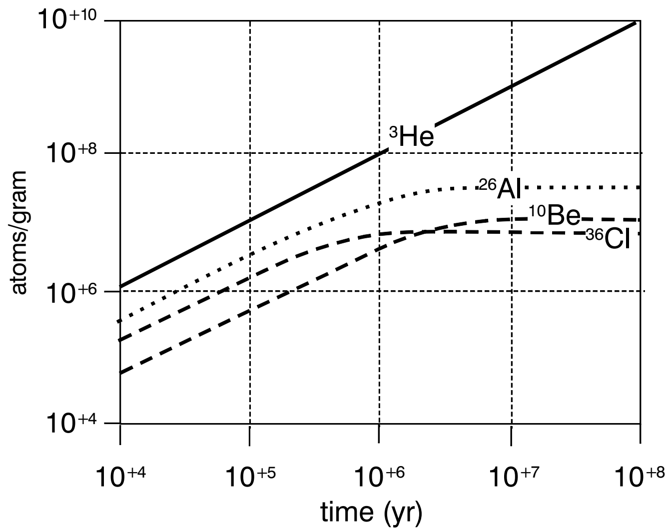

Fig. 5. *Increase of the concentration of ^3He, ^{10}Be, ^{26}Al, and ^{36}Cl in permanently exposed rocks in time.*

Other methods

There are several other absolute dating method based on cosmogenic tritium, ^{24}Na, ^{36}Cl and ^{81}Kr which are applied to date groundwater or lake water. The cosmogenic ^{41}Ca isotope is in discussion to determine the absolute age of Pleistocene bones from the ancestors of the Homo sapiens. The intrinsic question is whether Ca isotopes exchange with calcium dissolved in groundwater.

Dating with uranium-series disequilibrium

Dating with the uranium-series disequilibrium methods (e.g. $^{230}Th/U$ dating) is well established (Ivanovich and Harmon, 1992). There are, however, still problems to be solved with the absolute dating of impure material as tufa, calcrete and peat. The selection of the sample fractions which are most suitable for dating is usually decisive for the reliability of the dating results (e.g. from tufa; Mallik, 2000). Other questions are not yet satisfactorily answered. The detrital correction of $^{230}Th/U$ dates (e.g. usually done by the "isochron" approach; e.g. Ku and Liang, 1984) is concerned with a complicated algorithm for the calculation of the standard deviations of "isochron" ages (Ludwig and Titterington, 1994; Geyh, 1994; 2002). Checks for open system conditions of uranium are also seldom done. Hence, absolute dating of interglacial and glacial sediments for the correlation of the terrestrial and marine chronologies remains a challenge to the $^{230}Th/U$ and OSL/TL methods.

Neglecting open system conditions of uranium and missing statistical checks of the dates may result in obtuse geochronological conclusions (Geyh, 1991). Usually the "isochron" approach is applied for $^{230}Th/U$ dating of dirty material (e.g. Ku and Liang, 1984). It is based on the assumption that only one source of allochthonous thorium is incorporated into the material during the period of its formation. Then, the specific uranium and thorium activity ratios (AR) of at least three coeval samples with differing detrital contamination yields a mixing line („isochron"). Its slope and intersection with the Y axis equal the $^{230}Th/^{234}U$ AR and the present $^{230}Th/^{232}Th$ AR which is the decisive parameter for the calculation of detritally corrected $^{230}Th/U$ dates and the detrital correction itself, respectively. It should surprise, but usually does not, that the ARs used for the $^{230}Th/^{232}Th$ – $^{234}U/^{232}Th$ plot fit the „isochron" with apparently too large correlation coefficients of >0.99. In contrary, this is generally claimed as reliable proof that the model prerequisites for $^{230}Th/U$ dating are fulfilled especially for closed system conditions of uranium. Each too perfect fit of the ARs to the mixing lines (**Fig. 6**) contradicts, however, any probabilistic error analysis and requires studies on the intrinsic phenomena (Geyh, 1994; 2002).

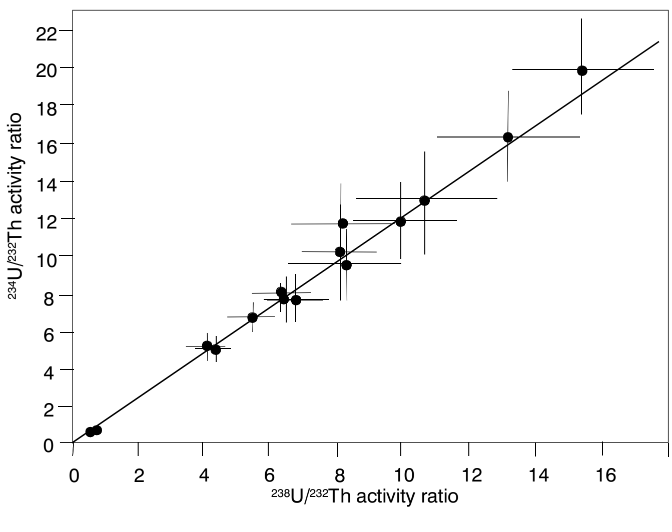

Fig. 6. *Mixing line ("isochron") with a too perfect fit of the used isotope activity ratios of uranium and thorium reflected by a correlation coefficient >0.99 (after Ivanovich and Harmon, 1992).*

Dating based on the radiation damage

Dating by radiation damage methods of eolian and fluviatile sediments preferentially deposited during glacial periods is meanwhile a routine. Automated devices allow a high throughput of TL/OSL (thermoluminescence, optically stimulated luminescence) analyses while the most time-consuming work remains the separation of pure mineral fractions, their chemical and physical treatment, the calibrated radiation and last but not least the evaluation and interpretation of raw results in order to get absolute dates. Beside many widely and continuously studied basic physical phenomena, fundamental questions are not yet satisfactorily answered. For an example the influence of palaeohydrological changes to the accuracy of the absolute dates. One of the reasons may be the limited precision of TL and OSL dates. A recently introduced technique to date single-aliquots may overcome at least some of these problems.

OSL and TL dating

The luminescence methods – based on thermoluminescence (TL) or optically stimulated luminescence (OSL) by green (GRSL) or red light (RSL) – are suitable for reliable absolute dating of eolian but also fluviatile, glaciofluviatile and limnic sediments with ages of up to at least 100 kyr (Wintle *et al.,* 1993). Some published dates even end up with 800 kyr (**Fig. 7**; Berger *et al.*, 1992). Apart from methodical reservations (Wintle *et al.*, 1993), ages exceeding 100 kyr usually do not satisfy statistical tests.

The reliability of TL/OSL dates of young sediments with ages <100 kyr is evidenced in many geological profiles. An example of methodical intercomparison by Smith *et al*. (1997) confirms that TL/OSL dates are always larger than the corresponding conventional ^{14}C dates within the age range of 20,000 to 45,000 yr. This is methodically expected (**Fig. 8**) and it fits the ^{14}C calibration curve determined from laminated Pleistocene lake sediments (e.g. Kitagawa and van der Plicht, 1998).

It is, however, well-known that OSL/TL dates highly depend upon the analysed mineral fraction, its treatment (e.g. pre-heating), the utilised stimulation technique (additive, regeneration), the applied measurement parameters (optical filters; e.g. Preusser, 1999) and the used model for the evaluation of the optical readings. However, there is not yet physically explainable systematics which would allow to distinguish between the reliable and non-reliable dates. Prescott and Robertson (1997) state: „In the approach of luminescence dating is still a great deal of empirism, not to say necromancy". Therefore, Frechen (1994) requested to determine as many as possible dates from the same geological context and to perform stratigraphical checks of the results. Many methodical and systematic studies on the basic principles have already been done. But a lot of additional work is still necessary, requiring very close co-operation between physicists, mineralogists and geoscientists.

The possibility to check both the reliability and precision of TL/OSL dates through error propagation is seldom used. Therefore, confidence intervals are often either too large or too small compared to the actual scatter of the dates. In such cases standard deviations are without a value as they cannot be used for any quantitative evaluation of the dates. There are, however, examples where TL/OSL dates and their standard deviations are reliable (Engelmann *et al.*, 2001) though the intrinsic age information of the dates was not fully evaluated. Discarding the TL and OSL dates of the uppermost sample, the depth trend of the other dates of the underlain samples allows to calculate a reasonable sedimentation rate of 0.29 kyr/depth unit and to estimate the period of sedimentation from 46.4±2.5 to 41.7±2.9 kyr BP (**Fig. 9**). Generally, applying statistical error analysis wrong dates are easily detectable and absurd interpretations are avoidable (Geyh, 1991).

The main challenge to OSL/TL dating methods is to improve the understanding of basic principles. An objec-

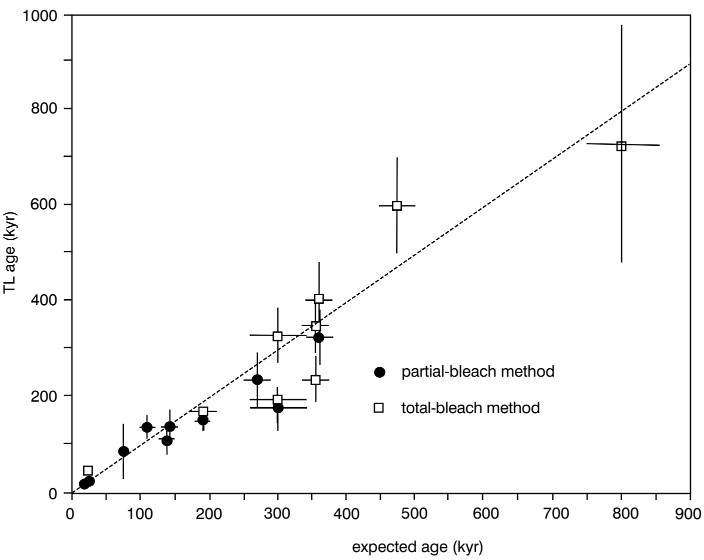

Fig. 7. *Dating of polymineral fine silt grains extracted from loess with ages of 100-800 kyr from Alaska and North Island, New Zealand (after Berger et al., 1992).*

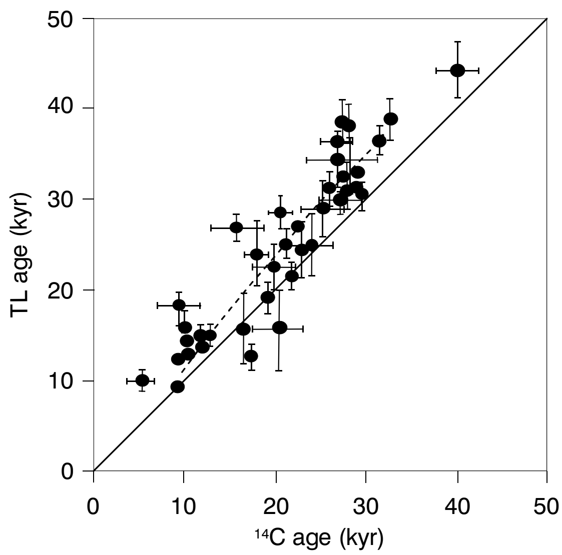

Fig. 8. *Methodical intercomparison of TL dates with conventional ^{14}C dates (Smith et al., 1997). The trend fits the preliminary ^{14}C calibration curve within the age range of 20,000 to 40,000 BP (Kitagawa and van der Plicht, 1998).*

tive judgement of the dates requires the use of different techniques to the same sample. An additional goal is the extension of the dating range hopefully to MIS 11. Combined $^{230}Th/U$, TL and OSL datings of interglacial and glacial sediments may be a suitable approach. The success of a combined application of these three dating methods is demonstrated by Geyh and Eitel (1998) who improved the understanding of the formation of calcrete and of the methodically diverging absolute dates of this material.

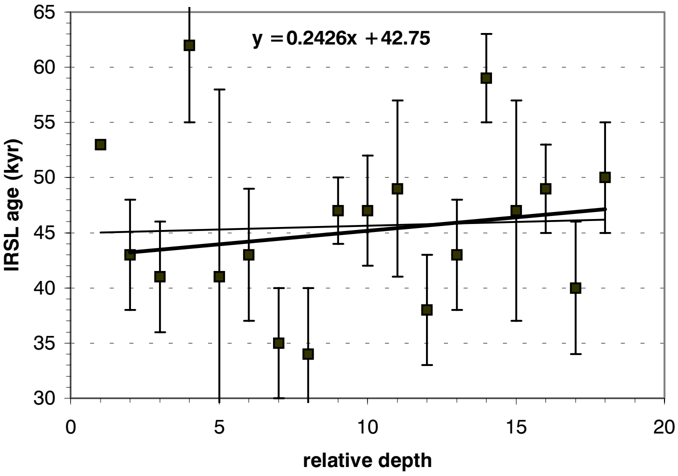

Fig. 9. *Consistent IRSL dates from a dune area in Israel (Engelmann* et al.*, 2001) after rejection of the date of the uppermost sample. The width of the probability distribution of all dates reflects their standard deviations. From the slope of the regression line follows that the sedimentation lasted from 46.4±2.5 to 41.7±2.9 kyr BP.*

ESR dating

The electron spin resonance (ESR) method is a widely used dating method especially for mollusc shells, teeth enamel, speleothems, tufa, marls, and foraminifera. The ESR geochronologists emphasise high throughput of used spectrometers, simple pre-treatment techniques of the samples, small sample size required and a width of the dating range of at least one million years (e.g. Grün, 1989; Ikeya, 1993). However, it is little convincing since widely scattering results of three international inter-laboratory checks (Hennig *et al.*, 1985; Barabas, 1993; Vanhaelewyn *et al.*, 2001) rise doubts whether ESR dating already yields absolute dates. The results highly depend on the experience and the skill of the geochronologist. It is frustrating that elaborated methodical restrictions are often ignored. Katzenberger (1989) found, for instance, that there are intrinsic problems which prevent reliable dating of mollusc shells by ESR. In spite of this, numerous ESR dates of that material have been determined and published later. It needed 1½ decade until the statistically proved reservation against such ESR and $^{230}Th/U$ dates of the same mollusc shells was accepted (**Fig. 10**; Schellmann and Radtke, 1999).

Many authors claim that most ESR dates fit the marine d^{18}O chronology (MIS). Geoscientists know that biogenic and secondary carbonates are exclusively formed in warm and humid periods. Considering this temporal restriction of their formation, ESR machines can easily, correspondingly adjusted, yield only suitable dates. The requirement of an absolute dating method of independence (Frechen, 1994) is questioned for ESR dating.

Up to now, the only convincing and most probably reliable ESR dates have been obtained from foraminifera (Sato, 1992; Mudelsee *et al.*, 1992). It is, however, not yet understood why just this material is suitable and others not.

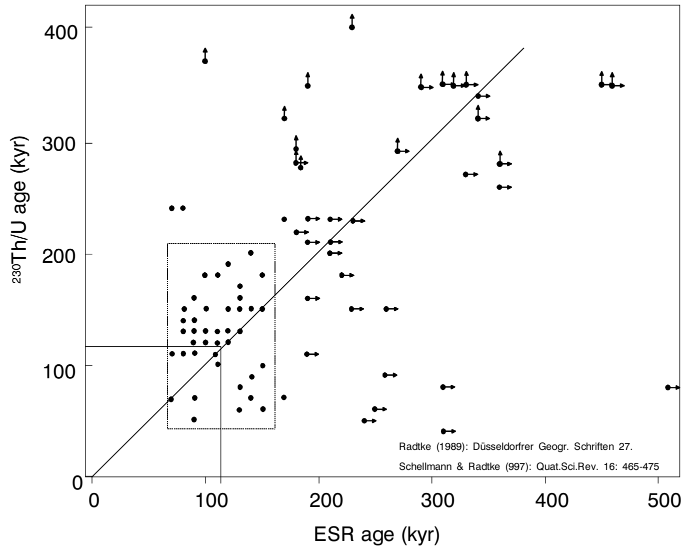

Fig. 10. *Comparison of ESR and $^{230}Th/U$ dates of mollusc shells which do not correlate with each other (Schellmann and Radtke, 1999). Katzenberger (1989) found already methodical arguments to exclude such samples for reliable dating.*

Alpha-recoil dating (ART)

The fission track-method is a well-known absolute dating method for volcanic rock minerals (Wagner, 1995). Its dating range is usually far from the Quaternary. Recently, Göken and Wagner (2000) improved the alpha-recoil dating method introduced by Huang and Walker (1967). Methodical difficulties prevented an earlier development of an applicable dating method. The breakthrough succeeded with the deciphering of the methodical principles which explain the rise of the number of visible tracks with the prolongation of the etching time. Now, dark mica and phlogopite with ages between 100 and one million years (**Fig. 11**) can be reliably dated. It is expected

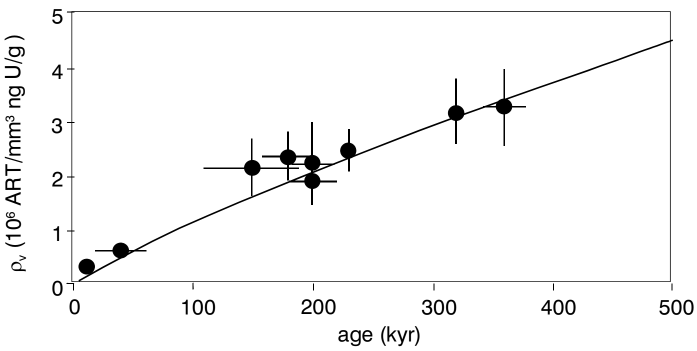

Fig. 11. *Dating of single grains of dark mica by the improved alpha-recoil method applying phase-contrast microscopy (Göken and Wagner 2001). The specific density of the alpha-recoil tracks (ART) increases with the age of the sample.*

to increase the dating accuracy from ± 20 % to ±5 % by the development of suitable uranium and thorium standards made from glass.

3. CONCLUSIONS

The absolute age determination of any material is a continuos challenge to geochronologists, geoscientists and archaeologists. The progress in the technical development of measurement devices apparently masks the slow process in solving methodical problems which is essential for any dating method. It is a grave misunderstanding of many geoscientists and archaeologists that physical dates easily allow to establish an absolute chronology often even without considering standard deviations. Dating of material with a complex genetic history and possible post-depositionary diagenetic history delivers only reliable dates if the geoscientific background is thoroughly understood and is fully taken into account in their evaluation. Only under this premise physical data may be transformable into absolute ages.

To overcome at least some of the described problems
a) Inter- and intra-laboratory comparisons should be intensified and regularly carried out for each absolute dating method within fixed periods,
b) The principles and the application of the physical dating methods should became a part of the university curriculum for environmental physics and geoscientists,
c) A thorough error analyses of all available data is an essential part of any geochronological evaluation,
d) Publications must be comprehensive, has to contain all details of analytical protocols and complete lists of the laboratory results used for the evaluation of absolute dates.

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