DEVELOPMENT AND APPLICATION OF STIMULATED LUMINESCENCE DATING METHODS FOR SEDIMENTS

BY

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SUMMARY

The aim of this thesis was the development of infrared stimulated luminescence (IRSL) as a dating technique, with particular application to sediments which have only experienced short sunlight exposures. The main areas studied concern methods of measuring the IRSL signal from bulk sediment, thermal stability of the IRSL signal from different feldspar minerals, dating of colluvial sediments and luminescence sensitivity changes incurred after laboratory bleaching.

The principal findings from the thermal stability studies are that potassium feldspars have a greater long term stability than plagioclase feldspars and polymineral bulk sediment. Short term thermally unstable IRSL signals were observed after laboratory irradiation and preheating procedures were developed to remove the unstable component.

Loess cores from Rocourt, Belgium, were used to investigate the IRSL signals from bulk sediments. The IRSL signals from pellets allowed an overview of the sedimentary pattern to be obtained in a relatively short time and a normalization procedure was developed using UV radiation. A portable IRSL system was designed and built.

A competition model was introduced to explain the sensitivity changes of the luminescence signals that occur after laboratory bleaching. Sensitivity changes relate to the age of sample, the extent of laboratory bleaching and the sunlight exposure prior to deposition and either an increase or decrease in sensitivity can occur. The predictions of the model are in good agreement with the results of laboratory experiments and sensitivity changes reported in the literature.

Sediments which were partially bleached at deposition have been studied. Several methods of detecting insufficient bleaching of IRSL signals were introduced. Two empirical methods for the equivalent dose determination were developed, which involve single aliquot procedures.

Using the techniques developed in this study, a dating programme was carried out to establish the geochronological record for colluvial sediment from five sections in Natal, South Africa.

STATEMENT

The work presented in this thesis has not already been accepted in substance for any degree and is not being concurrently submitted in candidature for any degree. The work is the result of my own investigations unless otherwise stated.

Sheng-Hua Li

Ann G. Wintle (supervisor)

CONTENT

SUMMARY i

STATEMENT ii

CONTENT iii

ACKNOWLEDGEMENTS ix

chapter 1: INTRODUCTION

1.1 INTRODUCTION TO LUMINESCENCE DATING 1
1.1.1. Fundamentals of luminescence dating 1
1.1.2. Thermoluminescence dating of sediments 1
1.1.3. TL residual after bleaching 2

1.2 OPTICAL DATING 3
1.2.1. Importance of optical dating 4
1.2.2. Optical dating of quartz with 514 nm Ar ion laser light 4
1.2.3. Optical dating of fine grained mixed minerals using 514 nm Ar ion laser 4
1.2.4. Optical dating of feldspars using 514 nm Ar ion laser 5
1.2.5. OSL mission spectra 6

1.3 OPTICAL DATING WITH IR 6
1.3.1. The advantages of IR(800-900 nm) stimulation 6
1.3.2. Comparison between IR and 514 nm stimulation 7

1.4 RELATIONSHIP BETWEEN TL DATING AND OPTICAL DATING 9
1.4.1. Thermally and optically stimulated luminescence 9
1.4.2. TL glow curve and optical decay curve 9

1.5 SCOPE OF THIS THESIS 10

chapter 2 EQUIPMENT AND MEASUREMENT METHODS

2.1 IR EQUIPMENT 12
2.1.1. Manual IR system mounted on Daybreak TL hot plate 12
2.1.2. Daybreak 2000 OSL reader 14
2.1.3. Risø automatic OSL/TL system 15
2.1.4. IR power at disc 16

2.2 Ar LASER OSL DATING SYSTEM 17

2.3 LIGHT SOURCES 17
2.3.1. Solar simulator 17
2.3.2. U.V. lamp 18
2.3.3. Filter and optics 18

2.4 MEASUREMENTS 19
2.4.1. Short shine 19
2.4.2. Quick heating (pulse annealing) 20
2.4.3. Single disc ED determination 21
2.4.4. Normalization 22

2.5. SAMPLE PREPARATION 23
2.5.1. Fine grain (4-11μm) 23
2.5.2. K-feldspar separation 24

2.6 EQUIPMENT RELEVANT TO DOSE RATE ESTIMATION 26
2.6.1. Thick source α counter 26
2.6.2. Thick source ß counter 26
2.6.3. K_2O measurement 26
2.6.4. Irradiation equipment 26

chapter 3 FUNDAMENTAL STUDIES ON FELDSPARS AND MIXED MINERALS

3.1 INTRODUCTION 28
3.1.1. Introduction to stability of luminescence signals 28
3.1.2. Anomalous fading 29
3.1.3. Importance of stability studies 29
3.1.4. Timescales of instability 30
3.1.5. Feldspar and mixed minerals 30
3.1.6. Summary 31

3.2 FELDSPAR CRYSTALS (MUSEUM SAMPLES) 31
3.2.1. Introduction 31
3.2.2. Feldspars and apparatus used 32
3.2.3. TL glow curves and IRSL 34
3.2.4. Short term decay and quick heating results 36
3.2.5. Thermal stability 40
3.2.6. Remarks on IRSL mechanism 42
3.2.7. U.V. recharging of feldspars 44
3.2.8. Summary 46

3.3 MIXED MINERALS (LOESS) 46
3.3.1. Introduction 46

3.3.2. Samples used and minerals contained in loess 47
3.3.3. Global view of thermal stability 48
3.3.4. Thermal stability for the Luochuan loess section 51
3.3.5. Anomalous fading of IRSL in loess 51
3.3.6. TL and IRSL 53
3.3.7. Relevance to long term dating limit 53
3.3.8. Summary 54

3.4 MIXED MINERALS (K-FELDSPAR) 55
3.4.1. Introduction 55
3.4.2. Thermal stability of different feldspar fractions 55
3.4.3. Comparison with pure minerals 56
3.4.4. Comparing K-feldspar from different regions 57
3.4.5. Implications for upper limit of dating 58
3.4.6. Anomalous fading 59
3.4.7. Summary 59

3.5 REMOVAL OF UNSTABLE SIGNALS 60
3.5.1. Introduction 60
3.5.2. Existence of thermally unstable signal 60
3.5.3. Removal of unstable signal without heating 61
3.5.4. Previous study of preheating 62
3.5.5. Justification and design of a preheat procedure 62
3.5.6. Preheating for K-feldspar samples 63
3.5.7. Preheating for loess samples 66
3.5.8. Summary 67

chapter 4: IR STIMULATED SIGNALS FROM COMPRESSED SEDIMENTS

4.1 INTRODUCTION 68
4.1.1. Initial research 68
4.1.2. Deposition of loess 68
4.1.3. IR scanning of core sediments 69

4.2 IRSL FROM COMPRESSED LOESS 70
4.2.1. Introduction 70
4.2.2. Preparation of pellets 71
4.2.3. IRSL signals from pellets 71
4.2.4. Comparison with the results of TL dating 72
4.2.5. IRSL of gamma dosed pellets 73
4.2.6. Assessment of the results and implications 74

4.3 PORTABLE IRSL SYSTEM 76
4.3.1. Introduction 76
4.3.2. Equipment design 76
4.3.3. Performance of the system 78

4.3.4. Potential and improvements 79
4.3.5. Summary 80

4.4 UV REGENERATED IRSL SIGNAL 80
4.4.1. Introduction 80
4.4.2. U.V. regenerated signal on diluted loess 81
4.4.3. Effective depth and IRSL signal 84
4.4.4. Depth of IR and IRSL penetration through a feldspar chip 85
4.4.5. Effective depth in pellet 87
4.4.6. U.V. regenerated signal and effect of natural dose 88
4.4.7. Assessments of u.v. normalization 89
4.4.8. Further information provided by the u.v. regenerated signal 90

chapter 5: LUMINESCENCE SENSITIVITY CHANGE DUE TO BLEACHING OF SEDIMENTS

5.1 INTRODUCTION 91

5.2 SENSITIVITY CHANGES AFTER DIFFERENT BLEACHING PROCEDURES 92
5.2.1. Samples used and experimental details 92
5.2.2. Sensitivity changes after bleaching 93
5.2.3. Summary 95

5.3 A COMPETITION MODEL 96
5.3.1. Introduction 96
5.3.2. Two types of trapped charges and luminescence sensitivity 96
5.3.3. Sensitivity changes 97
5.3.4. Predictions of the model 99

5.4 TESTING THE MODEL WITH EXPERIMENTS 100
5.4.1. Design of experiment 100
5.4.2. Experimental conditions 101
5.4.3. Sensitivity change results 102
5.4.4. Discussion and comparison with the model predictions 104

5.5 CONCLUSIONS AND IMPLICATIONS FOR OPTICAL DATING OF SEDIMENTS 106
5.5.1. Implication to the regeneration method 106
5.5.2. Dating with a single aliquot 107
5.5.3. Sensitivity change in sediments 108

5.6 SUMMARY 108

chapter 6: IRSL DATING OF PARTIALLY BLEACHED SEDIMENTS

6.1 INTRODUCTION 110

6.2 BLEACHING OF IRSL AND TL SIGNALS 111
6.2.1. Bleaching with sunlight 111
6.2.2. Comparison to SOL2 and IR bleaches 112
6.2.3. Colluvial and alluvial sediments 113
6.2.4. TL and IRSL bleaching 114
6.2.5. Summary 115

6.3 DETECTION OF INSUFFICIENT BLEACHING 116
6.3.1. Introduction 116
6.3.2. Shine plateau method 117
6.3.3. Natural Scatter 118
6.3.4. EDs obtained by the single aliquot method 120
6.3.5. Sensitivity change after laboratory bleaching 121
6.3.6. Comparing IRSL with TL 122
6.3.7. Assessment of different methods 122

6.4 ED ESTIMATION OF INSUFFICIENTLY BLEACHED SEDIMENTS 124
6.4.1. ED and I_n observed in aeolian and colluvial deposits 124
6.4.2. Possible connections between ED and I_n 125
6.4.3. Empirical methods to assess the true dose (EDo) 127

6.5 SUMMARY 131

chapter 7: DATING APPLICATIONS

7.1 INTRODUCTION 132
7.1.1. Importance of this study 132

7.2 GEOLOGICAL DESCRIPTION 132
7.2.1. Geological background 132
7.2.2. Grain size distribution 134
7.2.3. Sampling 134

7.3 EQUIVALENT DOSE DETERMINATION 134
7.3.1. Procedures 134
7.3.2. Results from coarse grain K-feldspar fractions 138
7.3.3. Results from fine grains fractions 141
7.3.4. Bleaching and EDs obtained by different methods 141
7.3.5. The younger samples 144

7.4 DOSE RATE ESTIMATION 145
7.4.1. Results of thick source alpha counting 145

7.4.2. Beta counting results 147
7.4.3. H_2O content 148
7.4.4. K_2O content 149
7.4.5. Cosmic rays 149
7.4.6. Internal alpha irradiation 149
7.4.7. Discussion 151

7.5 DATES AND SITE DESCRIPTIONS 152
7.5.1. Introduction 152
7.5.2. St Paul's section 152
7.5.2.1. The section 152
7.5.2.2. Luminescence dates for younger samples 154
7.5.2.3. Fine grain IRSL dates 155
7.5.2.4. Coarse grain IRSL dates 156
7.5.3. Nqutu section 156
7.5.4. Masotcheni section 157
7.5.5. Hazeldene section 157
7.5.6. Matatana section 158
7.5.7. Discussion on the age differences between fine grain and coarse grain fractions 158

7.6 CONCLUSIONS 159

chapter 8: RECOMMENDATIONS FOR FURTHER STUDIES 161

Appendix: LOESS SAMPLES FROM LUOCHUAN SECTION 163

REFERENCES 164

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chapter 1

INTRODUCTION

1.1 INTRODUCTION TO LUMINESCENCE DATING

1.1.1. Fundamentals of luminescence dating

Luminescence dating includes thermoluminescence (TL) dating and optical dating. In both methods, the detected signal is the light from released charges which recombine at luminescence centres. The charges have been previously trapped and accumulated during past geological time or archaeological time. In TL dating, luminescence is stimulated by heating. In optical dating luminescence is stimulated by light. Both luminescence dating methods have the same physical basis. The intensity of the luminescence signal is proportional to the irradiation dose received. So in luminescence dating, it is necessary to determine both the equivalent dose (ED) and the annual dose contributed by the radioactivity of the sample and its surroundings. The age of a sample can then be obtained by the equation

$$\text{AGE} = \frac{\text{equivalent dose}}{\text{annual dose}}$$

Thermoluminescence dating was initially used in determining the age of heated samples, such as pottery, because all the charges are released by the heating and the archaeological clock is reset (Aitken 1985).

1.1.2. Thermoluminescence dating of sediments

Thermoluminescence (TL) dating of sediments was an extension of its application to pottery. It was applied to sediments after the discovery that the TL signal is bleachable by sunlight (Wintle and Huntley 1979). When a sediment grain is exposed to sunlight, its TL signal is bleached and the geological clock is thus reset. Hence the age of the sediment

obtained by TL dating is the time since its last exposure to sunlight. However, a sizeable residual TL signal always remains in samples even after prolonged bleaching. A zero TL signal is never reached even by prolonged bleaching.

Three techniques which correct for the residual TL were developed (Wintle and Huntley 1980). One is the "Total bleach" method. This approach assumes that the sample was exposed to sunlight for an extended period of time before deposition. The residual TL is then determined by measuring the TL remaining after exposing an aliquot of the sample to sunlight for a period of around 60 hours. This residual signal is subtracted from the natural TL. This method works well for loess and for sand dunes (Wintle and Huntley 1982).

Another technique is the "Regeneration" method. The ED is determined by regenerating the TL following optical bleaching of the sample (Singhvi et al. 1982). This approach attempts to simulate the TL history of the sediment in the laboratory. This technique cannot be used for samples exhibiting sensitivity change after laboratory bleaching (Rendell and Townsend 1988, Berger 1988a).

The "Partial bleach" technique was also introduced (Wintle and Huntley 1980) and its exponents claim that in this technique the ED is determined by looking at the light sensitive traps. Several TL workers now advocate this technique highly (Berger 1988a), whilst others criticize it.

More recently other techniques, such as the plateau method (Mejdahl 1988a), have been successfully applied to some sediments, but have failed on others.

1.1.3. TL residual after bleaching

Although the TL signal can be bleached by sunlight, the degree of bleaching in quartz and feldspar grains varies considerably for different types of sediment; thus there is a problem

with uncertainty in the residual TL signal at the time of last exposure (Forman 1989). This residual signal cannot be precisely duplicated by any laboratory means and this results in a large error in the TL dating of young sediments. The same effect also means TL dating cannot be used for sediment which has not been exposed to sunlight for a long time (>10 hours), because the residual TL signal may be as high as 50-80% of its value before exposure, depending on the glow curve temperature. Although the partial bleach method is able to take account of this uncertainty for some samples, it could be affected badly if the degree of bleaching is mistaken.

1.2 OPTICAL DATING

Optical dating was first introduced by Huntley et al. (1985) who used both fine grain (2-8 μm) and quartz fractions. Considerable interest has led to a number of new developments in optically stimulated luminescence. Not only have a wide range of mineral samples (such as quartz, feldspars, zircon, apatite and mixed minerals) been researched, but also a wide range of stimulating light sources have been investigated, from near infrared to blue light and from mono-wavelength with lasers to broad band light sources.

Because there is not a well accepted terminology in optical dating, it is necessary to define some of terms used in this thesis to avoid confusion.

Optical dating: Optical dating is a general term used for all stimulated luminescence dating by light of any wavelength.

OSL (Optically Stimulated Luminescence): OSL is mainly used when the signal is stimulated by visible light. It is frequently used for 514 nm stimulation with an argon ion laser.

IRSL (Infrared stimulated luminescence): IRSL is the term used when infrared photons are used to stimulate the luminescence.

1.2.1. Importance of optical dating

As mentioned above, the main problem in TL dating is the uncertainty in the residual TL signal. However, this is not a problem in optical dating for most samples since the signal used for measurement can be bleached quickly to a very low level by sunlight (Godfrey-Smith et al., 1988). The bleaching rate of the IRSL signal is almost ten times quicker than that for the TL signal (Li and Aitken 1989). This should allow dating of a sediment which was not exposed to sunlight for a long time prior to or during deposition. Rapid and efficient bleaching of OSL signals was found for all wavelengths (Godfrey-Smith et al. 1988).

1.2.2. Optical dating of quartz with 514 nm Ar ion laser light

Optical dating was first applied to quartz fractions using 514 nm green light from an Ar ion laser (Huntley et al. 1985). Since then, quartz stimulation with 514 nm light has been comprehensively studied and this signal has been used to date over one hundred geological and archaeological samples (Smith et al. 1986; Godfrey-Smith et al. 1988; Rhodes 1988; Smith et al. 1990, Stokes 1992). It has been claimed that the OSL signal from quartz can be correlated with the 325°C TL peak (Smith et al. 1986). Several features relevant to dating, such as thermal stability, recuperation and charge photo-transfer, have been investigated (Aitken and Smith 1988, Rhodes 1990).

1.2.3. Optical dating of fine grained mixed minerals using 514 nm Ar ion laser.

Optical dating with a 514nm laser beam has been applied to fine grained polymineral loess samples (Li 1989; Spooner and Questiaux 1989; Questiaux 1991; Xie and Aitken 1991). It was found that the OSL signal from loess is dominated by a signal which is not stable on the time scale of more than 100 ka.

The OSL signal from a fine grain sample is derived from a mixture of many minerals, of

which quartz and feldspar are the most important. Although the OSL spectra for mixed mineral samples are made up of signals from the different minerals, it should be possible to enhance the OSL signals from a particular mineral components by using different filters. However, the EDs given by different sets of filters are within the error bars of each measurement (Spooner and Questiaux 1989, Questiaux 1991). This suggests that the OSL emissions from different minerals do not have sharp wavelength peaks. Signals from a single mineral cannot be isolated by selecting a particular filter.

Since there are few large grains in loess, medium-sized grains (40-50μm) have also been used on several occasions to obtain a bright signal (Xie and Aitken 1991, Questiaux 1991). These grains may have been less affected by weathering than the fine grains. Since the grain size is greater than the range of alpha particles in the grains, and the grains may have different internal radioactivity, the dosimetry of these sized grains may be a problem in OSL dating. However, studies on medium-sized grains indicate that there is no significant difference in the EDs determined with 4-8μm fine grains and 40-50μm medium-sized grains from the same loess samples (Questiaux 1991, Aitken and Xie 1992). This suggests that this grain size can be substituted for fine grains in dating application without re-calculation of the dosimetry.

1.2.4. Optical dating of feldspars using 514 nm Ar ion laser

The name feldspar is applied to a large family of minerals and considerable work needs to be done for each member of the family. Several aspects of various feldspars have been studied with a 514 nm laser beam (Li and Aitken 1989, Spooner 1992). In respect of the upper dating limit, the potassium (K) feldspars have shown an OSL signal which is more thermally stable than that from loess fine grains. The dose, at which the OSL reaches saturation is higher for K-feldspar than for quartz. However this effect is counteracted by the internal dose rate of K-feldspar being higher than that for quartz (Li and Aitken 1989). Even so, K-feldspars are likely to be most useful for dating old samples.

Anomalous fading of the OSL signal has also been studied. It was found that the anomalous fading of OSL from some feldspars, e.g. labradorite, follows the same pattern as its TL signal (Spooner 1992).

1.2.5. OSL emission spectra

The OSL spectra of different minerals have been studied (Godfrey-Smith et al. 1989, Huntley et al. 1989, Huntley et al. 1991). For feldspars the OSL spectra are the same as the high temperature TL spectra; whereas the OSL spectrum from quartz is different to the high temperature TL spectrum.

1.3 OPTICAL DATING WITH IR

Apart from the 514 nm laser light, stimulation with various visible wavelengths have been reported (Hütt et al. 1988; Godfrey-Smith et al. 1988; Huntley et al. 1989), but other wavelengths have not yet been studied intensively. However, for feldspars it was found that near infrared (800-900 nm) can also release charges from stable traps, giving rise to a luminescence signal (Hütt et al. 1988). Use of infrared (IR) as the stimulating wavelength has several advantages.

1.3.1. The advantages of IR (800-900nm) stimulation

First, using IR allows us to have a wider choice of detection filters. In optical dating studies, the stimulating wavelength is longer than the wavelength of the emitted luminescence. Stimulation by green light, as in the case of the 514 nm laser line, limits observation of the luminescence to the blue and near UV. Use of IR for stimulation permits observation from red to near UV, which coincides with the sensitive region of the photomultiplier tube. In practice, a single 2 mm thick Schott BG 39 filter is used to stop the IR from reaching the photomultiplier tube. Because different minerals emit at different wavelengths (Huntley et al. 1991), being able to choose different filters, which pass within the transmission range of

the Schott BG 39 filter, allows us to have a degree of selectivity regarding the emission of different minerals found in a polymineral sample.

Because the IR is well away from the wavelengths of the emitted luminescence, it is possible to separate the stimulating IR and the IRSL by a simple mirror, such as cold mirror. This has enabled the construction of a portable system, which is simple to build and may be used in the field. This will be described in detail in chapter 4.

Second, suitable IR power may be obtained from commercial light emitting diodes (LEDs); these are generally cheaper by a factor of around 1000 compared with laser light sources. Because LEDs are rather small, it is convenient to build an IR system mounted above a TL oven. Hence, TL measuring equipment with IR LEDs can be used for both TL and optical dating (Poolton and Bailiff, 1989).

Third, only certain minerals exhibit IR stimulated luminescence. Up to now, no such signal has been found for quartz, though it is an important TL mineral and produces an OSL signal with green light stimulation. This fact may be used as a convenient check for feldspar contamination in a quartz separate (Spooner and Questiaux 1989, Rhodes 1990).

1.3.2. Comparison between IR and 514 nm stimulation

Near infrared (800-900 nm) and green (514 nm) are the most commonly used wavelengths for optical dating. Some features relevant to IRSL and OSL dating have been compared for various samples (Li and Aitken 1989; Spooner 1992). Both OSL and IRSL signals can be bleached to very low levels by sunlight or using a solar simulator (Godfrey-Smith et al. 1988). This has been demonstrated by bleaching experiments for K-feldspar and fine grain loess (Li and Aitken 1989).

The spectra of the stimulated luminescence produced as the result of stimulation by either

wavelength are identical for the same feldspar samples (Huntley et al. 1991).

Li (unpublished report to RLAHA) reported that similar traps can be bleached by either wavelength. This conclusion was reached by observing the green light OSL after an infrared bleach. It appears that more than 90% of the green stimulated luminescence is removed by infrared bleaching of a fine grain loess sample from China. This suggests that a considerable number of trapped charges, if not all, could be stimulated by either wavelength. A similar response was also found for the green light OSL in K-feldspar (Li and Aitken 1989).

However, it should be borne in mind that there are several differences between the two stimulation processes. The wavelength difference means that the photon energy is different. Also, the 514 nm green light is from an argon ion laser, a source of coherent, single wavelength light; whereas, the LEDs produce photons in a wavelength band with a full width at half height of about 80nm (Spooner and Franks 1990).

As mentioned above, some minerals react differently to the two wavelengths. IR cannot be used to stimulate luminescence from quartz, even though quartz gives an OSL signal with 514 nm green light. This means that after IR dating and prolonged IR bleaching of discs carrying a polymineral sample, further measurements using 514 nm may be performed. These would give an OSL signal dominated by the quartz component. The possibility of using this multiple dating approach is dependent on the degree of feldspar OSL bleaching as a result of the IR exposure. If the OSL from IR bleachable minerals can be bleached substantially (as in the case of some feldspars), OSL measurement after prolonged IR exposure could be used to obtain another date with minerals such as quartz as suggested by Spooner et al. (1990).

The stimulation processes are different for IRSL and OSL. It has been suggested that a direct transmission is involved in the generation of the OSL signal; whereas the IR

stimulates the electrons into intermediate traps from which they are released at room temperature into the conduction band (Hütt et al. 1988, Bailiff and Poolton 1989, Duller and Wintle 1991).

Slight differences in the thermal stability were found between the OSL and IRSL signals (Li unpublished data). The IRSL shows a slightly but consistently high thermal stability than the OSL of the same feldspar.

1.4 RELATIONSHIP BETWEEN TL DATING AND OPTICAL DATING

1.4.1. Thermally and optically stimulated luminescence

Many features of TL and OSL dating are very similar. The sample preparation procedures and the dosimetry are identical in both dating techniques. Other aspects are closely related, such as part of the measurement equipment. Many techniques developed in TL dating, e.g. the methods of ED determination (additive dose and regeneration etc.) are also used in optical dating.

1.4.2. TL glow curve and optical decay curve

Although many features in TL and OSL dating are very similar, the way of recording the signal is different. In TL dating, the recorded signal is called a glow curve; it is a plot of the luminescence signal versus temperature. In optical dating, the recorded signal is a decay curve (often named a shine down curve). It is a plot of the luminescence signal versus stimulation time.

Groups of traps can be identified from the TL glow curve by their TL peak temperatures, but this type of analysis cannot be applied to the shine down curve which is made up of a mixture of signals from all the traps being stimulated. Because the different traps giving rise to the OSL cannot be distinguished from the shine down curve, information from the TL glow curve is used to obtain extra information to aid the OSL studies, e.g. to select

preheating procedures for the removal of the unstable component (see chapter 3).

Much shorter optical exposure times can also be used. In particular the short shine (see chapter 2) is a shine down curve with a very short exposure time, typically 0.1-0.5 seconds. It is often used for disc to disc normalization and for dating measurements. This short exposure results in a negligible reduction of the OSL signal and allows the disc to be re-used for further measurements. In comparison TL measurement will destroy all the information in the grains on the disc.

The ED plateau method, developed in TL dating (Mejdahl 1988a), has a different meaning when applied in OSL dating (Rhodes 1990). In TL dating, the ED plateau relates to the bleaching levels of different temperature traps and the thermal stability of the these traps. However, in optical dating the ED plateau relates to the composition of easy and hard to bleach components (traps), which may, or may not, have similar stability.

1.5 SCOPE OF THIS THESIS

This thesis may be divided into three parts, each of which is closely related to the other: **fundamental studies, IRSL stratigraphic reconnaissance and dating of sediments.** Emphasis is placed on understanding the behaviour of sedimentary grains which have received only short light exposures prior to deposition.

The fundamental studies concentrate on the stability of the IRSL signals in pure feldspar, in loess and in K-feldspar extracted from sediments. Storage procedures for the removal of thermally unstable signals are developed for these samples.

Because mineralogy and dosimetric conditions are similar in several sediment sections, the relative age can be recognized by measuring the IRSL signal from the bulk sample. The experimental conditions and procedures for developing such an approach are given. A

portable system for use in the field is described.

Particular emphasis has been placed on the study of IRSL sensitivity changes due to bleaching. A competition model has been suggested. It can explain and predict the sensitivity change for a particular sample.

Part of the thesis reports the application of IRSL dating to geological samples. The IRSL method was used to date colluvial samples from South Africa; these were expected to have received relatively short light exposures prior to and during deposition.

chapter 2
EQUIPMENT AND MEASUREMENT METHODS

In this chapter I describe the equipment and several of the measurement methods used in this study. Several pieces of IRSL equipment were developed and built in the course of the study. Ancillary equipment (e.g. light sources) is also described, including the Oxford facility for laser optical dating.

2.1 IR EQUIPMENT

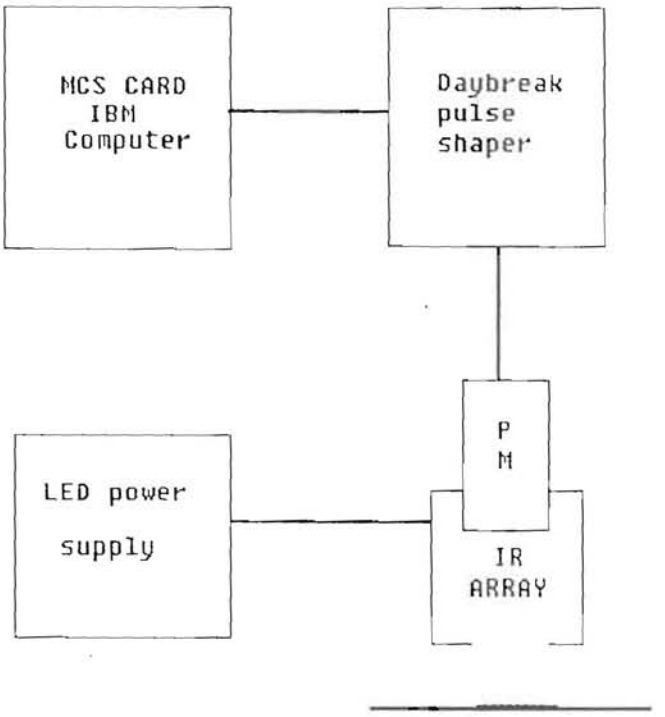
Three systems were used for IRSL measurement, namely the manual Daybreak reader, the Daybreak 2000 OSL reader and the Risø OSL/TL system.

2.1.1. Manual IRSL system mounted on Daybreak TL hot plate

A simple and easy-to-use system was built in the early stages of the study. It consisted of a manual IRSL system mounted on a Daybreak TL oven. The IR light emitting diode (LED) array was based on the design of Spooner and Franks (1990). The detection system used part of the TL set, e.g. high voltage supply, ratemeter and photomultiplier (PM) tube. A quartz light guide coated with SiO was used to enhance the signal-to-noise ratio. A 2mm thick Schott BG 39 filter was placed in front of the PM tube to prevent the stimulating IR photons reaching the photocathode. A manual shutter could be placed between the sample and the LED array. Figure 2.1.1 shows the IR LED array placed between the TL oven and the PM tube.

Instead of taking the IRSL signal to a chart recorder, the shaped pules from the EMI 9635B PM tube were fed to an EG&G Ortec ACE Multi-Channel-Scaler (MCS) card in an IBM compatible computer. This enabled data to be collected in up to 4096 channels, with channel widths from 2 microseconds to 30 minutes. Signals were then stored in the computer for

(a)



(b)

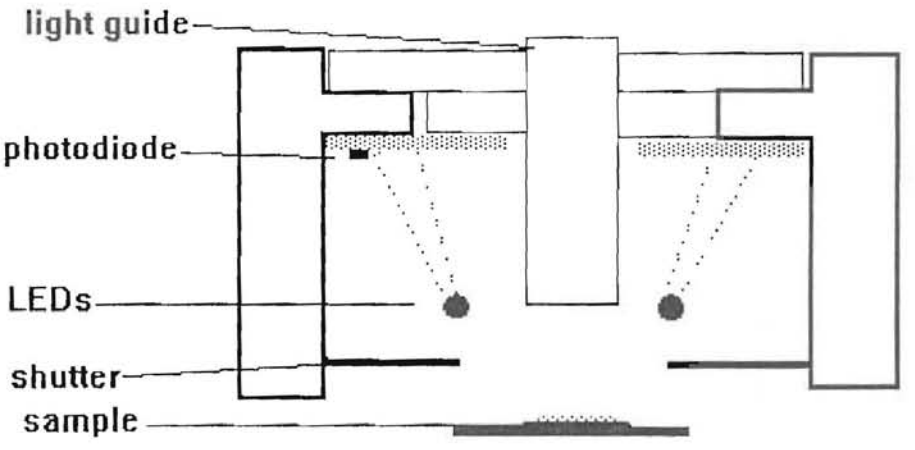


Figure 2.1.1. Schematic diagram shows the Manual Daybreak IRSL system built during this study.

further analysis. The MCS card gave considerable flexibility in data handling, a feature required for various applications in this thesis.

The LED array consisted of 12 serially connected TEMT 484 light emitting diodes (LEDs). These emit about 100 mW/Sr (100mA) at 880nm, Δ80nm, with 8° angle between half radiant intensity points, and have a temperature sensitivity of -0.53%/°C (Spooner and Franks 1990). The current through each LED is about 20mA.

Some characteristics of this system are summarized in Table 2.1.1.

Table 2.1.1. Characteristics of manual IR system

	TYPICAL SETTING	VARIABLE RANGE
power	20mW/cm^2	
LED current	20mA	0-30mA
LEDs	TEMT 484	
Photodiode	BPW 34	
Light Guide	Quartz SiO coated	
Dark noise	80 counts/s	60-120 counts/s
Length/channel	0.001s	2 μs - 30 minutes
Recording channels	200	
Trigger time	<0.001s	
LED time control	0.11 s	0.01 - 99s

The intensity of the IR illumination is monitored by a pin photo-diode (IR sensor) mounted on the LED print board which detects the reflected IR. The photo-diode type is BPW 34 which was selected for its sensitivity to the IR wavelength used. The output of the photo-diode is used to feed-back current control of the LEDs. This is a servo system that keeps the illumination constant during IR exposure of the sample. Because it monitors the reflected IR, the system is dependent on the surface conditions of the sample. This has very little effect on fine grain samples because of the uniform distribution of the sample on the discs. However, it is a factor in the study of a sediment core (see chapter 4), since the core sediment surface may not be uniform. In the later stages of this study, the system was modified to give a choice of operation, with or without feedback control.

The uniformity of the IR intensity across the sample disc (diameter 1 cm) was tested by measuring the IRSL from a disc partially covered by grains. The disc was turned successively through an angle of a few degrees and the IRSL was measured by a short shine (see below). A variation in the IRSL signal of around 20% was found when rotating the disc through successive positions. This could be for two reasons. One is the change in the feedback signal caused the change in the reflectance of the surface; this would change the current through the LEDs and change their output. The other is that the IR illumination at the disc is not uniform as a result of the differences in angle and IR intensity between the LEDs. However, these effects are not a problem if the grains uniformly cover the disc, as is the case for fine grain samples. An inconsistency in IR power may result in some scatter for coarse grain discs if the bright grains are not spread uniformly on the disc.

The illumination time of the LEDs is regulated with a pulse generated by the power supply to the LEDs. It allows a choice in IR illumination time from 0.01 second up to 99.99 seconds. The IR illumination time can also be controlled manually by either switching on and off the LEDs power or using the manual shutter.

The dark noise of the PM tube was about 80 counts/s, which varies slightly with room temperature. The background, or noise, was about 120 counts/s when the LEDs are on and a 2 mm thick BG 39 filter is in front of the PM tube.

The system has been used in a wide range of measurements. It was used for IRSL thermal stability studies, IRSL of pellet samples and IR scanning of core sediments. Fine grain (not coarse grain) discs were studied using this system.

2.1.2. Daybreak 2000 OSL reader

Towards the end of this study, a 2000 OSL reader system was bought from the Daybreak Co.. The system is similar to the manual Daybreak system (Fig 2.1.2). In particular the

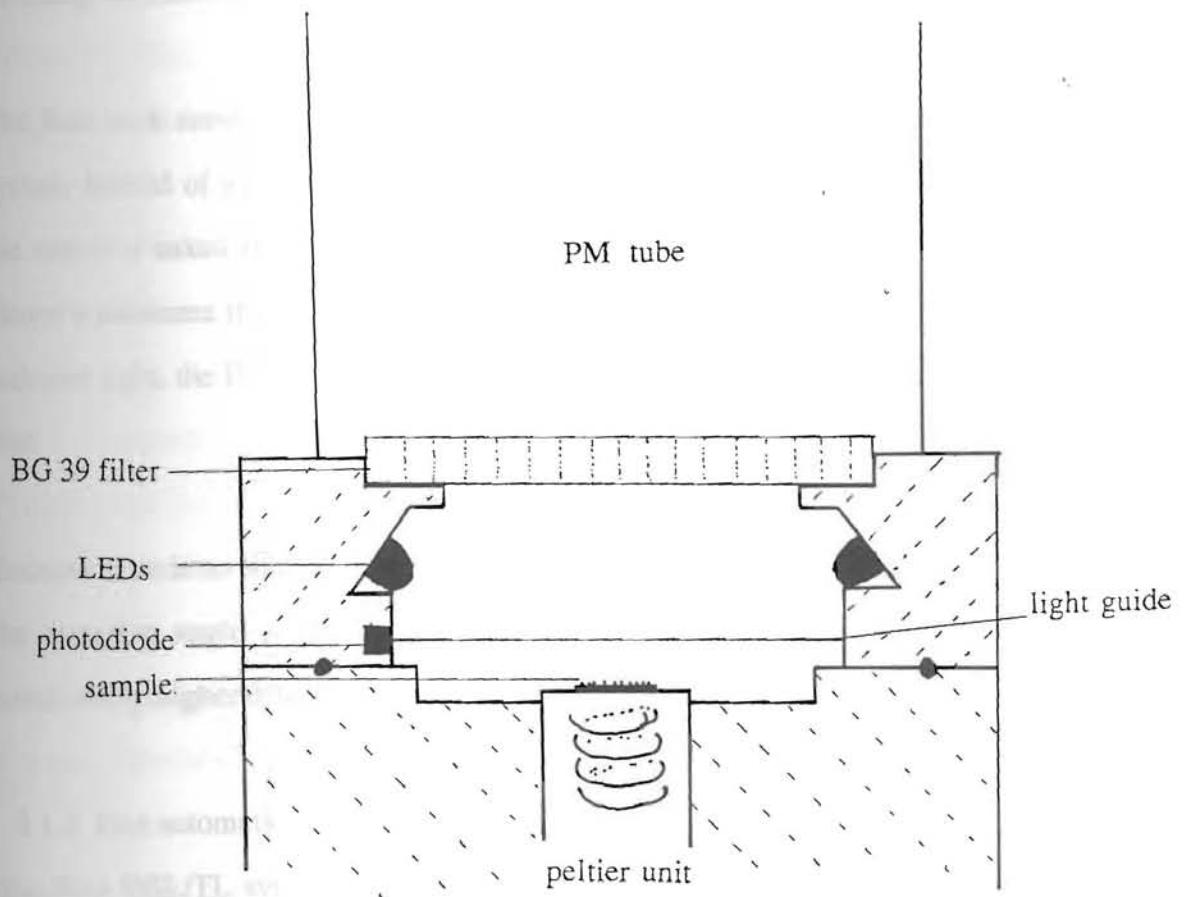


Figure 2.1.2. Schematic diagram showing the 2000 OSL Reader.

detection of the IRSL signal is identical. The temperature of the sample disc could be controlled from 5 to 50 °C by a combined peltier unit and a heater in the base plate. Because the IRSL signal changes with temperature (Duller and Wintle 1991), the system has the advantage of minimizing variation in IRSL due to temperature changes in the laboratory.

The feed-back control for the LED current is slightly different from that of the manual system. Instead of a photo-diode being placed on the printed circuit board, it is attached to the end of a naked fibre light guide which is placed between the LEDs and the sample. Hence it measures the IR flux on the sample disc. Because the IR is not controlled by the reflected light, the IR power is not affected significantly by the distribution of grains on the disc.

Because there is no shutter between the LEDs and the sample disc in the 2000 OSL system, the detection angle is larger than for the manual Daybreak system. This results in a considerably higher detection efficiency.

2.1.3. Risø automatic OSL/TL system

The Risø OSL/TL system has been described elsewhere (Bøtter-Jensen et al. 1991). The system consists of an irradiation source and an IR unit, mounted on a TL reader. This allows the system to be more flexible than the Daybreak systems. Measurements, e.g. preheating, TL and IRSL, can be performed in the system without moving the discs from the reader. The hot plate in the system also allows the disc to be held at a constant elevated temperature during IRSL measurement. This overcomes the IRSL signal variations due to temperature variations (Hütt et al. 1989, Duller and Wintle 1991).

The 32 LEDs used in the system are of the same type as those used in the manual Daybreak system. The IR power was also regulated by the IR flux detected by a photodiode. The feedback is performed by leading the IR from an additional LED to a photodiode by an IR

fibre light guide.

There is a spectrosil disc between the sample and the PM tube and this resulted in part of the IR and IRSL being reflected and absorbed. Hence the detection efficiency is not as high as for the 2000 OSL system. Because there are 32 LEDs in the system and the current is about 40 mA for each LED, the IR power is higher than the systems mentioned above.

This system was used for TL, quick heating and preheating measurements. Single disc IRSL ED determination was also performed in this system.

2.1.4. IR power at the disc

The power of the IR illumination falling on sample disc is a characteristic of the system. The power can be calibrated by direct measurement or by comparing the IRSL signal with that observed for a system of known power. Direct measurements were made using a calibrated photodiode placed at the disc position. The power of the Risø OSL/TL system was about 40mW/cm^2 and for the manual Daybreak system was of about 20 mW/cm^2.

Although direct measurement is preferable, it is not always possible to place the photodiode in the exact position of a sample disc. In this case the most suitable way, at least for calibrations for IRSL measurements, should be by comparing IRSL signals provided that there is a connection between the power and the IRSL signal.

In a sunlight bleaching study, Godfrey-Smith (1991) found that the rate of bleaching of the OSL for quartz was directly proportional to the total ambient light level. If there is a similar relationship between IRSL bleaching and IR power, then the time needed for a certain percent IRSL reduction will be inversely proportional to the power of the IR. Hence the IR power of different systems can be estimated by comparing the decay curves of identical discs. Using this method the IR power of the Risø OSL/TL system was 1.9 times that of the

manual Daybreak system. This is in line with the results of the direct measurements with the photodiode. Applying this method to the 2000 OSL reader suggested that its power was about 3 mW/cm^2.

Because measurements used for a particular ED determination are made at the same IR intensity for each group of discs, the absolute power is not important for the results.

2.2 Ar LASER OSL DATING SYSTEM

The system has been described elsewhere (Smith et al. 1986, Rhodes 1990). 514.5 nm monochromatic light from an argon ion laser (Coherent Innova 70-2), was used for stimulation. Two filters, one Corning 3-71 filter and one 514 nm interference filter were placed in the beam path to remove plasma light produced by the laser. The intensity of the beam was measured by a photodiode which monitored a portion of the beam. An electronically controlled shutter was placed in the beam before reaching the measurement chamber. The power reaching the sample disc was about 5 mW/cm^2.

Four Corning 7-59 filters were used to remove the scattered green light and one Schott BG 39 to reject the red fluorescence. Those filters were placed in front of the EMI 9635QB PM tube, which is vertically above the sample disc.

The laser system was used to study the removal of the thermally unstable signal (chapter 3).

2.3 LIGHT SOURCES

Apart from the IR diode arrays and the 514.5 nm Ar ion laser light used for stimulation, other light sources were used either for bleaching or regenerating the signals.

2.3.1. Solar simulator

Because optical dating methods determine the age of the sample since its last exposure to

sunlight, sample bleaching in the laboratory is an important procedure. Although some of the bleaching experiments were done using natural sunlight, it is not always accessible in some laboratories, e.g. Aberystwyth.

A solar simulator was one of the light sources used to emulate natural sunlight. In this study SOL2 from Dr Hönle, Martinsried, Germany was used. According to the manufacturer, this lamp has a similar spectral distribution to sunlight (Fig. 2.3.1), and produces a constant intensity up to 6.5 times that of natural sunlight.

A sample disc placed under the SOL2 will reach a 55°C equilibrium temperature in about 30 minutes.

2.3.2. U.V. lamp

In order to normalize bulk samples, e.g. pellets and cores (see chapter 4), an ultra-violet (u.v.) lamp was used as a means of regenerating the IRSL signal after previous sunlight bleaching. The u.v. lamp is a mineral light lamp of UVGL-25 from UVP INC. U.S.A.. It has two output wavelength bands peaked at 254 nm and 365 nm respectively. Only the 254nm band was used.

The distance between the pellets and the u.v. source is 10 cm, which is fixed by a sample stand for all u.v. exposures. In order to minimize the u.v. power differences, samples were placed within a relatively small area (5x5 cm) beneath the u.v. lamp. The power variation was less than 10% within the area as tested with identical feldspar sample discs.

2.3.3. Filter and optics

Because this study concentrates on the IRSL signal, it is essential to prevent the IR reaching the photocathode of the PM tube. This can be achieved by a 2mm thick Schott BG 39 filter placed in front of the PM tube. This filter passes most wavelengths in the visible range

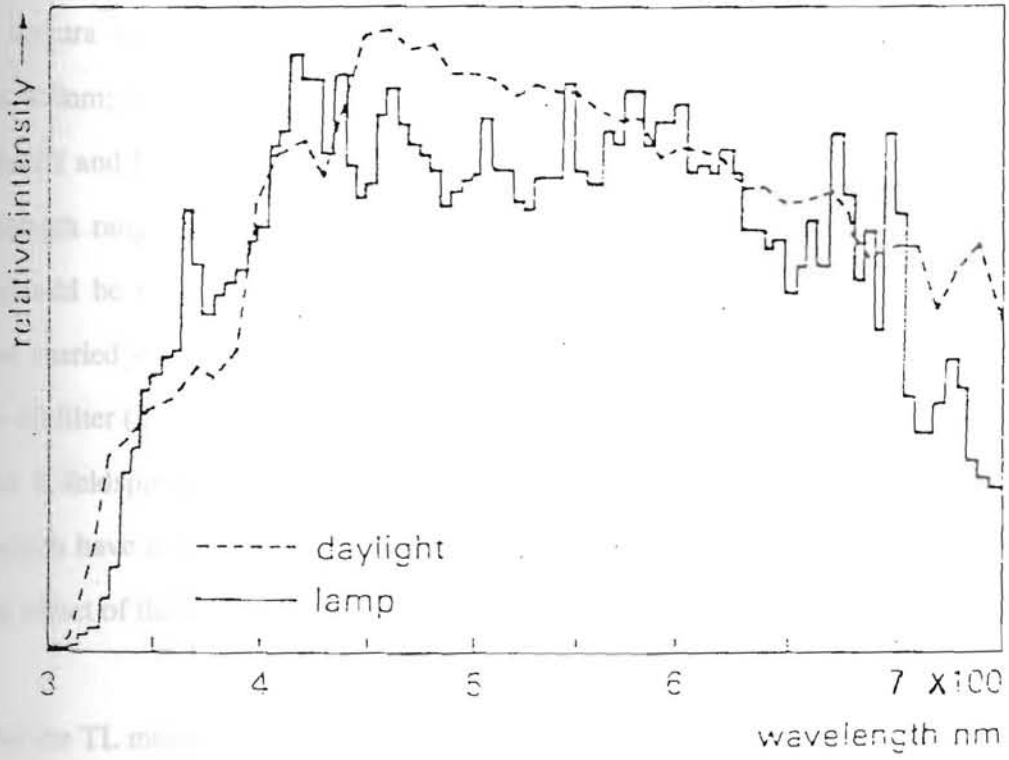


Figure 2.3.1. Spectrum of the SOL2 solar simulator (from the company brochure).

(Figure 2.3.2). Unless stated, this BG 39 filter was used for all IRSL measurements.

Emission spectra studies have indicated that K-feldspars have a major emission band peaking at 400nm; whereas Na-feldspar and plagioclase have an emission band peaking at 570nm (Bailiff and Poolton 1991, Huntley et al. 1991). The BG 39 filter transmits well in this wavelength range. Although IRSL emission spectra are not within the scope of this study, it should be noted that the same ED was given for the K-feldspar samples when dating was carried out using either the BG 39 only or the BG 39 with one 2mm thick Corning 5-58 filter (Li unpublished data). This suggests that filter selection is not crucial for a separated K-feldspar, provided that the filter passes the main emission wavelengths. For samples which have a very bright IRSL signal, a neutral density filter was used to prevent the pile-up effect of the PM tube.

For most of the TL measurements, the optical filters used were either a Corning 7-59 or a 5-60 combined with one heat-absorbing HA-3. The BG 39 filter was used on several occasions for comparison with IRSL. The combination of filters used are stated in the following chapters.

2.4 MEASUREMENTS

2.4.1. Short shine

The term "short shine" is applied to the measurement of the IRSL made with a short exposure to the stimulating light. In most cases the short shine exposure time was 0.1 second. Because the reduction of the IRSL is only a few percent in one second of exposure for the power levels in the equipment used, the reduction of the IRSL caused by this short shine is negligible. Figure 2.4.1 shows the result of 99 short shines (0.1 s) using the 2000 OSL Reader. As can be seen, the reduction of the IRSL signal is still very small (0.6%) even after 10 short shine measurements.

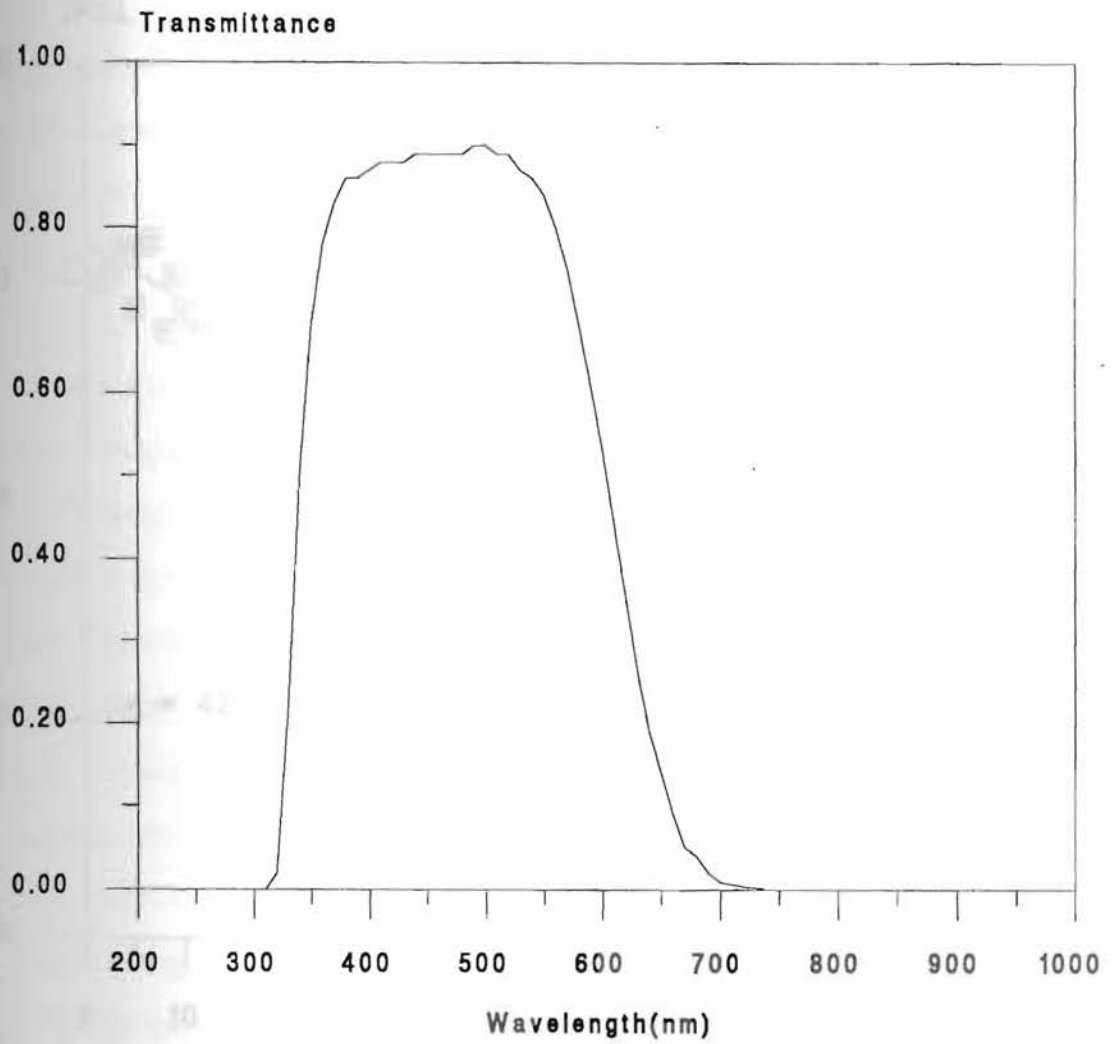


Figure 2.3.2. Spectral response of the BG39 filter (from the Schott catalogue).

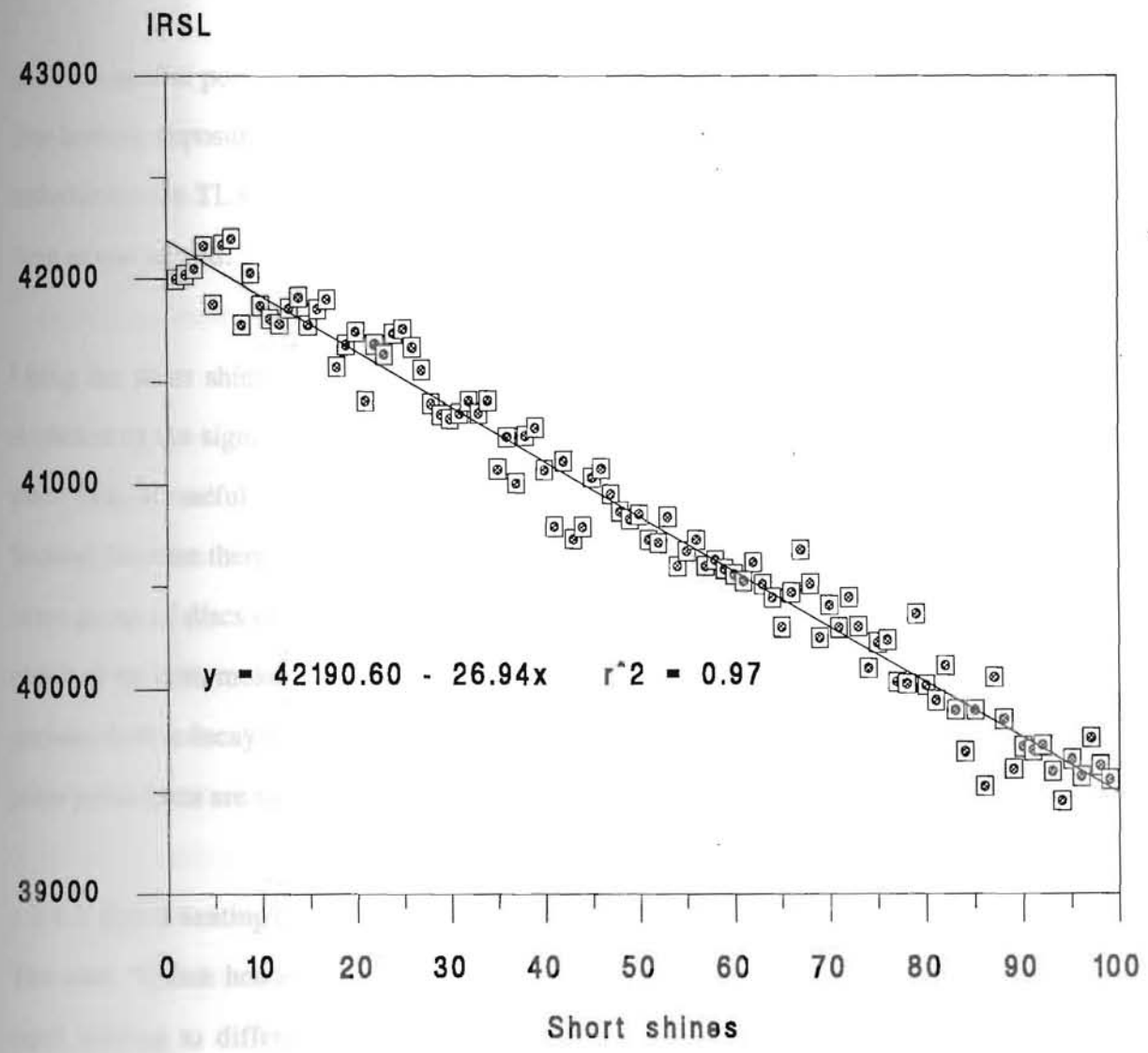


Figure 2.4.1. IRSL signal of 99 short shine (0.1 sec) measurements at room temperature. Measurements were made on the 2000 OSL Reader.

An even smaller portion of the TL is removed, even if the OSL or IRSL is reduced to a very low level by exposure to the stimulating light (Li and Aitken 1989; Duller 1992); hence, the reduction of the TL signal caused by a short shine is negligible even when the exposure is as long as one second.

Using the short shine measurement has several advantages. First, there is no significant depletion of the signal, which allows repeated measurements to be performed on the same disc. This is useful for the quick heating measurement described in the next section. Second, because there is a negligible reduction in the TL signal after each short shine, the same group of discs can be used for both IRSL (or OSL) and TL measurements. The EDs obtained by both measurements can be compared directly. On the other hand, information derived from a decay curve, e.g. a shine plateau (see chapter 6), will not be available. Short shine procedures are not suitable for young samples because detector noise is dominant.

2.4.2. Quick heating (pulse annealing)

The term "Quick heating" is applied to IRSL measurements made on a single disc after rapid heating to different temperatures. The sample is heated to a progressively higher temperature and immediately cooled to a temperature (To) for IRSL measurement. The procedure is as follows.

1. Quick heating (e.g. 5°C/s) of the sample disc to a temperature higher than To.
2. Cooling the sample disc to temperature To
3. Measuring the IRSL signal at temperature To using a short shine (0.1 s)

The temperature increase for each cycle was either in a constant step, e.g. 25°C, or randomly. The temperature (To) for IRSL measurements was 50°C. The maximum temperature reached was that which erased the IRSL totally (usually about 450°C). Similar quick heating experiments were also applied to the OSL signal from K-feldspar using the Ar ion laser system, in which the temperature for OSL measurements was 17°C.

The results of the quick heating were usually presented as of the percentage IRSL signal remaining after heating versus the previously attained temperature (Fig.2.4.2a). Occasionally the data were presented in the form of percent of IRSL reduction versus the previously attained temperature (Fig. 2.4.2b).

2.4.3. Single disc ED determination

ED determination with single aliquots has been characterized in detail elsewhere (Duller 1991). Here I briefly describe several key experimental procedures involved in the process.

1. Additive dose. The experimental procedures are: preheating the disc; measuring the IRSL signal using a short shine and adding an irradiation dose. The sequence was repeated for each of several irradiation doses which are applied in order to construct the growth curve. Because the disc is preheated in each experimental cycle, the reduction of the IRSL signal due to this preheating needs to be corrected for. This was achieved by repeating the preheating and measurement cycles for another identical disc, this time without irradiation doses. The additive dose growth curve was then constructed after the correction.

As has been mentioned by Duller (1991), this method is only suitable when the IRSL signal increases linearly with dose. This means that it is suitable for relatively young samples provided they have sufficient IRSL sensitivity.

For most samples the experimental conditions were as follows: preheating for 10 minutes at 220°C; measuring the IRSL with a short shine (0.5 s) at 50°C; added ß dose accordingly. The added ß dose is high enough that the IRSL signal created by the largest dose is more than twice the natural signal.

2. Regeneration. The regeneration method for a single disc is very similar to the regeneration methods that are used in multiple disc luminescence dating. The experimental

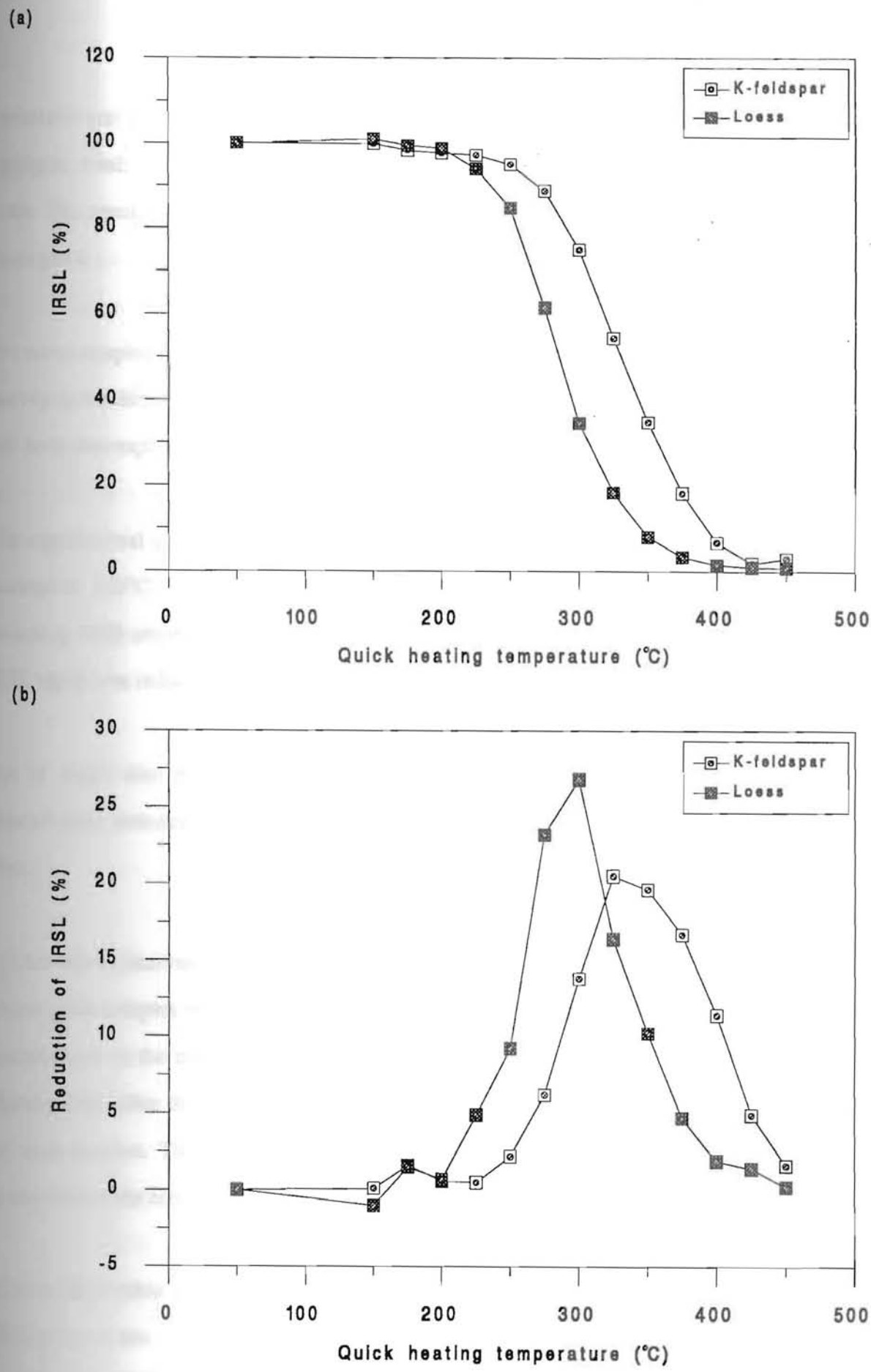


Figure 2.4.2. Quick heating curves of natural IRSL signals from loess and separated K-feldspar. (a) presented as the percent of the initial IRSL signal remaining (b) presented in form of IRSL signal reduction between each heating step. The loess sample is 88-12 from Luochuan, China. The K-feldspar is the separated fraction of sample (AF-3) from Natal, South Africa.

procedures are: preheating the disc; measuring the IRSL signal; bleaching the IRSL to a negligible level; ß dose irradiation. This experimental cycle was repeated for different ß doses. The regeneration growth curve was then constructed from the IRSL signals and the doses given.

For many samples it has been found that the sensitivity of the IRSL is changed between each cycle resulting in erroneous ED determination (Duller 1991). These sensitivity changes will be further explored in chapters 5 and 6.

The experimental conditions for most samples used in this study are: preheating for 10 minutes at 220°C; measuring the IRSL for 100 seconds with the disc held at 50°C; bleaching 1600 seconds at 50°C with the same IR power level. After this bleaching the IRSL signal was reduced to less than 1% of its initial level.

Use of single disc methods has the advantage of giving higher precision because no normalization between discs is required. The precision is better than 1% for most sample discs.

2.4.4. Normalization

Coarse grain samples were normalized by using the IRSL counts of a short shine (0.1 sec) measurement on the natural discs before any treatment. It is named 'natural normalization' (Smith 1986). After this normalization the disc-to-disc scatter was reduced to less than ±5% for most samples. The disc-to-disc scatter after normalization is smaller than for quartz grains of a similar size (Rhodes 1990, Godfrey-Smith 1991).

'Dose normalization' (Rhodes 1990) was used for the younger samples because the natural IRSL count is not high enough for the short shine measurement. In 'dose normalization', after all of the IRSL measurements had been made, the discs were bleached for 5 hours with

the SOL2, and a test ß dose of 19 Gy was added. Following a preheat for 10 minutes at 220°C, the IRSL count for 1 second IR exposure was measured and used for normalization. This method is analogous to second glow normalization for TL measurements. It was found that the disc-to-disc scatter after both 'natural' and 'dose' normalization can give information about the degree of IRSL bleaching prior to deposition. This will be discussed in chapter 6.

For fine grained (4-11μm) samples, the disc-to-disc scatter is relatively small even without any normalization. However, it was found that the disc-to-disc scatter could be as much as ±10% for some samples before normalization. Hence, most fine grain samples were 'natural normalized'. The procedure was the same as that used for coarse grain K-feldspar samples.

A normalization method with IRSL generated by u.v. exposure has been developed for sediment pellets. This will be discussed in detail in chapter 4.

2.5 SAMPLE PREPARATION

Here I briefly describe the two sample preparations routinely used in Aberystwyth laboratory.

2.5.1. Fine grain (4-11μm)

More than 2 mm was removed from the surface of the sample block using a razor blade, in order to remove grains which may have been exposed to incident light during sampling or transportation. Sample discs were prepared using the following procedures.

Extracting 4-11μm grains

1. treat with 10% HCl to remove calcium carbonate
2. wash with distilled water; put into ultrasonic bath and centrifuge (more than three times)
3. suspend in 0.005N sodium oxalate and sieve through 50μm mesh
4. settle in 20cm column of sodium oxalate for 20 minutes; pour liquid into another

cylinder; discard the coarse fraction (>11μm)

5. settle in 20cm column of sodium oxalate for 4 hours; pour off liquid; suspend solids in 20 cm sodium oxalate column for another 4 hours.

6. pour off liquid; wash with distilled water, put in ultrasonic bath and centrifuge (more than three times)

7. treat with 30% H_2O_2 (3-4 hours)

8. wash with distilled water; put in ultrasonic bath and centrifuge (more than three times)

9. dry thoroughly in 40°C oven

Making sample discs

1. weigh 0.15g of dried 4-11μm grains

2. suspend in 100ml acetone

3. ultrasonic bath

4. pipette out 1 ml into one 10mm diameter cylinder with Al disc

5. dry at room temperature (24 hours)

2.5.2. K-feldspar separation

K-feldspar fractions separated by heavy liquid were also used in the study. The separation procedure is similar to that described by Mejdahl (1985a). The 125-150μm grain size fraction was used for most of the samples from South Africa. The preparation procedures were as follows:

Grain size separation

1. treat with 10% HCl to remove calcium carbonate

2. wash with distilled water and put into ultrasonic bath (more than three times)

3. treat with 30% H_2O_2 to remove organic materials

4. wash with distilled water(more than three times)

5. wet sieving to retain grains larger than 100μm.

6. dry thoroughly

7. sieve (125-150μm was selected)

Separating light and heavy fractions

This is aimed at separating quartz and heavy minerals from feldspars. It was achieved by using sodium polytungstate solution at 2.62 s.g..

1. place subsamples of less than 5g in a 100 ml centrifuge tube
2. add about 80ml of heavy liquid
3. stir floating grains to remove trapped air, add a little more liquid
4. cover with cling film and leave for 1 hour or more
5. pour off light fraction with twisting motion
6. filter through 9 cm diameter Buchner funnel with filter paper
7. wash thoroughly with distilled water (many times)
8. dry thoroughly

repeat steps 6, 7 and 8 for the heavy fraction.

Obtaining K-feldspar

The light fraction from the above procedure was separated further by using sodium polytungstate at 2.58 s.g..

1. repeat 1-8 procedures of separating of light and heavy fractions, mentioned above. This is to separate lighter K-feldspar from plagioclase feldspar
2. etch with 10% HF for 40 minutes
3. wash with 10% HCl (more than twice) to remove fluoride precipitates
4. wash thoroughly with distilled water
5. wet sieve to remove any grains smaller than the expected grain size (100μm)
6. dry thoroughly

Making sample discs

The K-feldspar grains obtained after separation were mounted on aluminium discs using Rüsch Silkospray. After spraying with Silkospray, the disc was dipped into the grains using tweezers. Then it was tapped hard to remove the loose grains on the disc. After this treatment, the grains on the disc were found to give a single layer and were stable enough to prevent grain loss during any stage of the experiment.

2.6. EQUIPMENT RELEVANT TO DOSE RATE ESTIMATION

2.6.1. Thick source α counter

The equipment used for α counting is the Daybreak 580 α counter system. The samples were crushed in either a mortar or a ball mill. The amount of sample used is approximately 7g. Details of alpha counting have been discussed by Huntley and Wintle (1981) and Aitken (1985).

2.6.2. Thick source ß counter

The thick source ß counter was made at SURRC. Details of this equipment can be found elsewhere (Sanderson 1988). A 15 g portion of the sample used for α counting was used.

2.6.3. K_2O content measurement

The K_2O contents were measured either by AAS or ß counting. Both methods were applied to several samples as a check for consistency.

2.6.4. Irradiation equipment

Beta irradiations were made using a ^{90}Sr-^{90}Y source (27 years half life) mounted in a Daybreak automatic irradiation system. The source strength is 125 mCi and gives a dose rate of 3.81 Gy/min (March, 1992) and 3.79 Gy/min (October, 1990) for coarse grain K-feldspar and fine grain samples respectively.

Alpha irradiations were made using a 500μCi ^{244}Cm source (18 years half life) also mounted in the Daybreak automatic irradiation system. The strength of the source is 1.30/(μm^2*min) (calibrated in June 1989).

chapter 3

FUNDAMENTAL STUDIES ON FELDSPARS AND MIXED MINERALS

3.1 INTRODUCTION

3.1.1. Introduction to stability of luminescence signals

Luminescence signals result from the recombination of charge carriers, (usually thought of as electrons), at luminescence centres. The charge carriers are released from traps by the application of thermal energy. When this is applied quickly, as in the case of heating a crystal in the laboratory, the trapped charge recombines in the crystal, giving rise to thermoluminescence. At ambient temperature, e.g. in the burial environment, the trapped charge has a low, but finite probability of being released. This has been called thermal fading (McKeever 1985). The charges relating to the lower temperature TL glow peaks will escape from their traps at a faster rate than those relating to the higher temperature TL glow peaks (i.e. over 300°C). Thus in TL dating procedures the high temperature TL traps are used. The simplest way to locate a TL signal with thermal stability is to plot the ratio of the natural TL to the TL signal for a laboratory irradiation, as function of glow curve temperature. A plateau value will be attained for a temperature above which the natural signal has not undergone any decay. This is usually around 300°C.

For a single TL glow peak, it is possible to calculate its stability at the storage temperature T using measurable parameters, such as the trap depth (E) and escape frequency s. The half life time τ is given by the equation

$$\tau = s^{-1} e^{(-E/kT)}$$

where k is Boltzman's constant.

Because optically stimulated luminescence signals are measured at room temperature, there is no analogous test. The half life can only be estimated if E and s have been derived by

other measurements e.g. for identification of a related TL peak.

3.1.2. Anomalous fading

Besides thermal fading, a type of fading has been identified which does not behave according to the simple equation given above. This has been termed anomalous fading (Wintle 1973; 1974). This was first noticed in TL dating studies of feldspars from lava flows. There is substantial fading in the high temperature (>300°C) glow curve region during storage for a few hours. This fading component is related to the charge from deep traps. Anomalous fading of the TL signal has been observed for several mineral species, e.g. clay, zircon, fluorapatite and some members of feldspar family (Aitken 1985). This type of fading may be athermal (occurring at the same rate independent of storage temperature) or may show some thermal dependence, thus allowing it to be removed by storage at elevated temperatures. Models for anomalous fading have been suggested for some minerals (Templer 1986, Visocekas 1985).

3.1.3. Importance of stability studies

It is an essential requirement for luminescence dating that the signal measured is stable over the period of interest. However, this may not always be the case. For example, it has been reported that the TL signal from loess polymineral samples cannot be used for samples which are over one hundred thousand years old (Debenham 1985, Wintle 1990). It has been suggested that this is due to the lack of long term thermal stability of the signal. Similar results were also found in OSL dating of loess (Li 1989, Spooner and Questiaux 1989, Xie and Aitken 1992).

If the measured signal contains thermally unstable components, the ED, and hence the age of the sample, will be underestimated. If the signal is not stable over a certain period of time, then it will not be suitable for dating any sample older than this.

After laboratory irradiation, a considerable number of charges are trapped in shallow traps and the luminescence signal relating to these charges will decay quickly. Because of the short lifetime of this signal, it will not be found in natural samples. Removal of this unstable component is an essential step in luminescence dating methods since it is likely to contribute to or influence the signal used for ED determination. This is particularly important in optical dating, because it is not possible to determine the signal stability by looking at the signal itself.

3.1.4. Timescales of instability

Because the stability of the luminescence signal is related to the time scale concerned, it is necessary to define some terms used in this thesis.

Short term: Less than three months. If the signal is not stable in the short term, the fading can be observed in a laboratory storage experiment.

Long term: Over 100 ka. If the signal instability occurs only in the long term, signal decay will not be observed in the laboratory.

Medium term: Anything between short and long, i.e. on a time scale of months to tens of thousands of years.

3.1.5. Feldspar and mixed minerals

It is thought that the IRSL signal from a mixed mineral assemblage is dominated by the signal from the feldspar minerals (Spooner and Questiaux 1989). Feldspars are a solid-solution series in which each member has a different crystalline structure. Hence the stability of the luminescence signal should not be expected to be the same for all of them.

Coarse grain (100-300 μm) K-feldspar and fine grain (4-11 μm) mineral mixtures are two fractions widely used in luminescence dating. The coarse grained K-feldspar fraction, separated with heavy liquids, is often treated as an identifiable mineral with a characteristic signal (Mejdahl 1985a, Wintle 1992). However, after separation the sample is still a mixture

of feldspars, and could include feldspars such as sanidine, microcline and orthoclase which have different structures but similar densities (Deer et al. 1966). The fine grain (4-11 μm) (Zimmerman 1971) sample used is polymineral without any mineral separation. This fraction is often used when there is a lack of coarse grains (>100 μm), e.g. in loess and deep sea sediments, because the dosimetry is simpler.

3.1.6. Summary

The thermal stability of the sample is an important feature in luminescence studies. Lack of thermal stability can cause age underestimation and limits the use of luminescence dating methods for old samples. It is necessary to define what is meant by instability, which is related to the time scale and type of decay processes, e.g. thermal or anomalous fading. Removing unstable signals is an essential step in luminescence dating. Stability studies on the TL and IRSL of fine grains and K-feldspar are of particular interest because these minerals fractions are widely used in dating studies.

In this chapter, I will deal with some of these problems. This study will concentrate on the IRSL signal, but some of the features also relate to the OSL and TL signals.

3.2 FELDSPAR CRYSTALS (MUSEUM SAMPLES)

3.2.1. Introduction

Previous studies on pure feldspar minerals have led to a better understanding of different aspects of luminescence dating (Huntley *et al.* 1988, Bailiff and Poolton 1989, Huntley *et al.* 1991, Bailiff and Poolton 1991, Spooner 1992). However, the thermal stability of IRSL from feldspars has not been investigated in detail. In this section I will examine the stability for samples previously used for TL and OSL emission spectrum studies (Huntley *et al.* 1988, Huntley *et al.* 1991), allowing the results to be compared directly.

Very little has been published about the IRSL from pure feldspars, but some pertinent

studies are summarized below.

Anomalous fading: Spooner (1992) reported that short term anomalous fading was found in certain feldspars which had also shown anomalous fading of the TL signal. Anomalous fading was found to be unrelated to the composition. The decay was found to be athermal.

Emission spectrum: Investigation of IRSL in pure feldspars (Huntley et al. 1991, Bailiff and Poolton 1991), has indicated that the IRSL emission spectra of feldspars are similar to the high temperature TL spectra for the same mineral. This suggests a similar recombination mechanism is involved. K-feldspars showed a major emission peak at around 400 nm, whereas sodium feldspars had a peak at about 570nm. A weak emission peak was found below 350nm for both K-feldspars and plagioclase feldspars (Huntley et al. 1988, Huntley et al. 1991).

U.V. charge transfer: U.V. charge transfer refers to the ultraviolet light eviction of electrons from the deep traps ('donors') and results in some of them being re-trapped in shallow traps ('acceptors'). U.V. charge transfer was found for some feldspars (Bailiff and Poolton 1989; 1991). Different behaviours were observed for different feldspars.

Sensitivity: The brightest samples were found to be the high potassium and sodium feldspars (Spooner 1992), the minerals which also gave the brighter TL signals.

3.2.2 Feldspars and apparatus used

Fourteen museum samples of feldspar have been kindly given to me by Dr Godfrey-Smith, who obtained them from the National Mineral Collection of the Geological Survey of Canada and the Department of Geology, University of British Columbia. Subsamples of these feldspars have been used in the study of emission spectra of both TL and OSL (Huntley *et al.* 1988, Huntley *et al.* 1989, Huntley *et al.* 1991). The samples are listed in Table 3.2.1. These feldspars are potassic, sodic and calcitic aluminosilicates of varying proportions along the left and bottom sides of the ternary diagram (Fig. 3.2.1. Table 3.2.1). Samples Pa, K5 and K6 were used in this study but are not shown in Fig. 3.2.1. Since Pa is an albite its composition places it in the lower left corner of Fig. 3.2.1. Samples K5 and K6

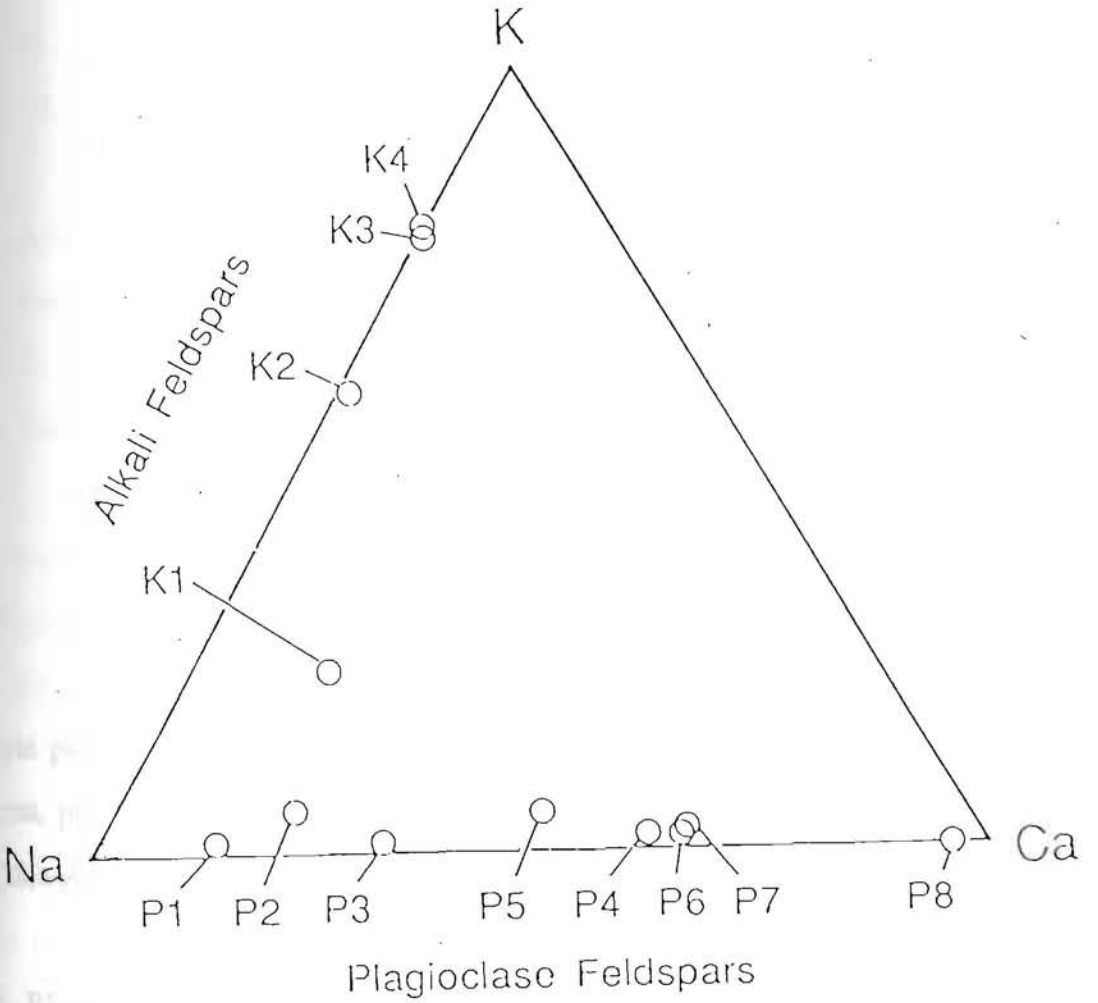


Figure 3.2.1. The feldspar ternary diagram showing the composition of fourteen feldspar minerals used in this study (from Huntley et al. 1988).

are microcline. Their composition places them in the top corner of Fig. 3.2.1.

Table 3.2.1. Pure feldspars: type and sources used in this study

LABEL	TYPE	SOURCE
K1	Anorthoclase	Norway
K2	Orthoclase	
K3	Orthoclase	Red Lodge,Montana
K4	Microcline	Colorado
K5	Microcline	
K6	Microcline	
P1	Oligoclase	Baffin Island
P2	Oligoclase	
P3	Andesine	
P4	Labradorite	Mexico
P5	Andesine/Labradorite	Nevada
P6	Labradorite	Mexico
P7	Labradorite	New York
Pa	Albite	South Dakota, USA

These sample pieces were ground in a ball mill to less than 300 μm. For luminescence measurements, prepared samples were attached to 10 mm diameter aluminium discs using silicon oil (Silkospray, Willy Rüsch AG, Germany).

Samples P4, P5, and P6 were rejected because the IRSL signal was not bright enough. For the remaining feldspars the IRSL measurements were performed in either a Risø OSL/TL system or the 2000 OSL Reader. Isothermal decay and quick heating experiments were carried out in the Riso OSL/TL system. The Daybreak system was used for the anomalous fading tests.

The TL glow curves were measured on the Riso automatic TL reader. The heating rate was 5°C/s to a maximum temperature of 450°C.

The filter used for both TL and IRSL measurements was a 2 mm thick BG 39. It covers the strong emission bands from both sodium and potassium feldspars (Fig. 2.3.2).

Samples were separated into two groups for comparison. One group is K-feldspar consisting of samples K1, K2, K3, K4, K5 and K6. The other is of plagioclase feldspars consisting of samples P1, P2, P3, P7 and Pa (Fig. 3.2.1).

3.2.3. TL glow curves and IRSL

In order to have an overview, the following experiments used one disc from each sample to record the TL, IRSL and u.v. regenerated IRSL signals. This allowed these results to be compared without normalization.

Since the samples were museum specimens with unknown irradiation and light exposure histories, all samples were heated to 450°C. The sample discs were given a 12 Gy beta dose and the TL glow curves of each sample were measured within an hour.

The shapes of the measured TL glow curves are generally different (Figs.3.2.2 and 3.2.3). No attempt has been made to normalize the TL responses of the minerals, but about the same weight is present on each disc. No relationship was found between the shape of the glow curve and the mineralogical structure or its composition with the exception that the alkali feldspar samples (K1 to K6) all showed a similar low temperature TL peak around 140-180°C. Samples P1 and P2 are both oligoclase feldspar and are close together on the ternary diagram, but their TL glow curve shapes are completely different. K-feldspar samples K5 and K6 (microcline) also had different glow curves. This suggests that neither the mineralogical structure nor composition are responsible for the shape of the TL glow curve. On the other hand, some connections between TL emission spectra and composition of K and Na elements have been reported (Huntley et al. 1988, 1991).

Following the TL measurements, the IRSL signals were then measured for 1 second in the Riso OSL/TL system after the discs were given the same beta dose (12 Gy). The

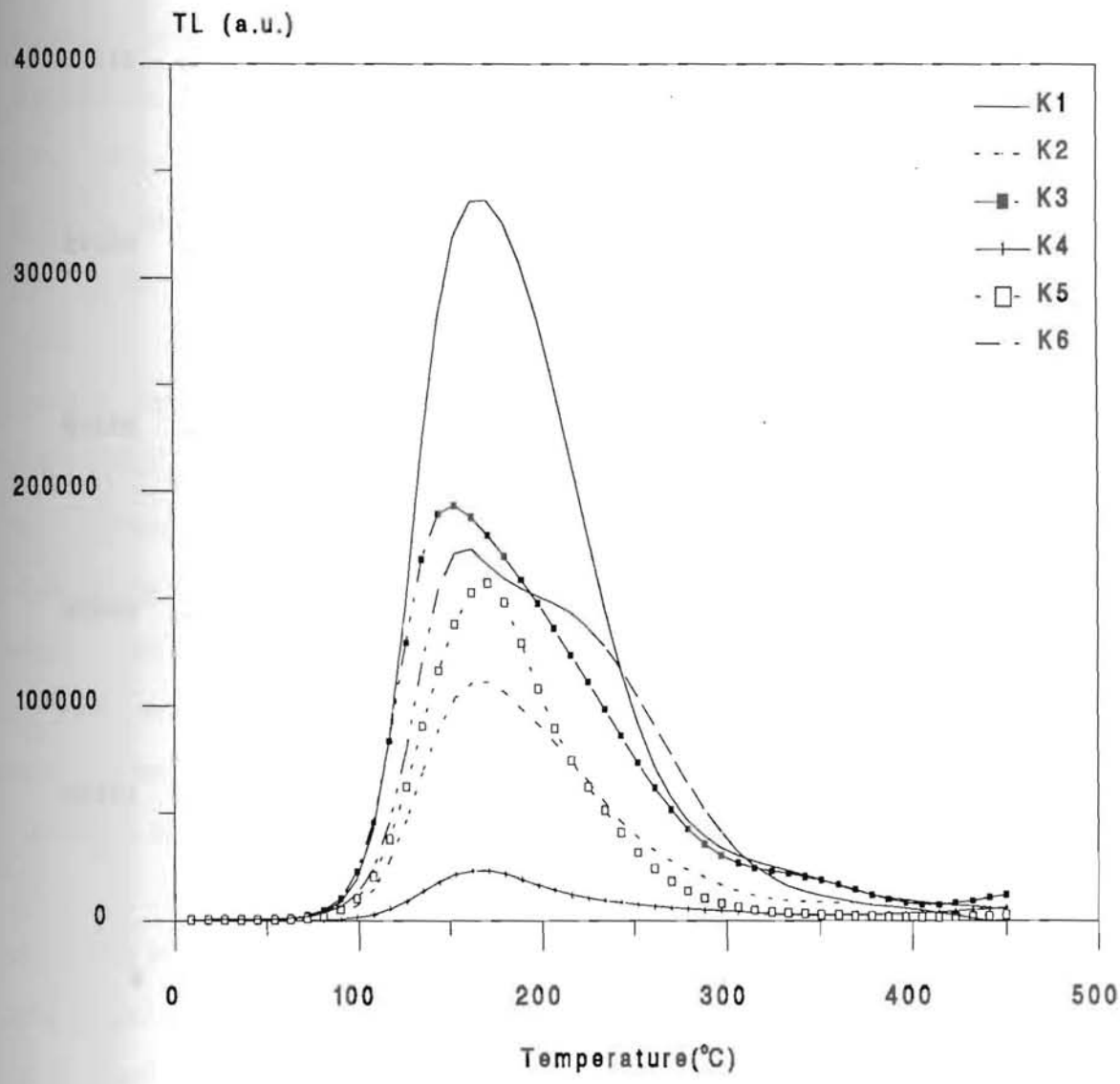


Figure 3.2.2. TL glow curves of K-feldspars. TL measurement took place with less than one hour delay after ß irradiation (12 Gy).

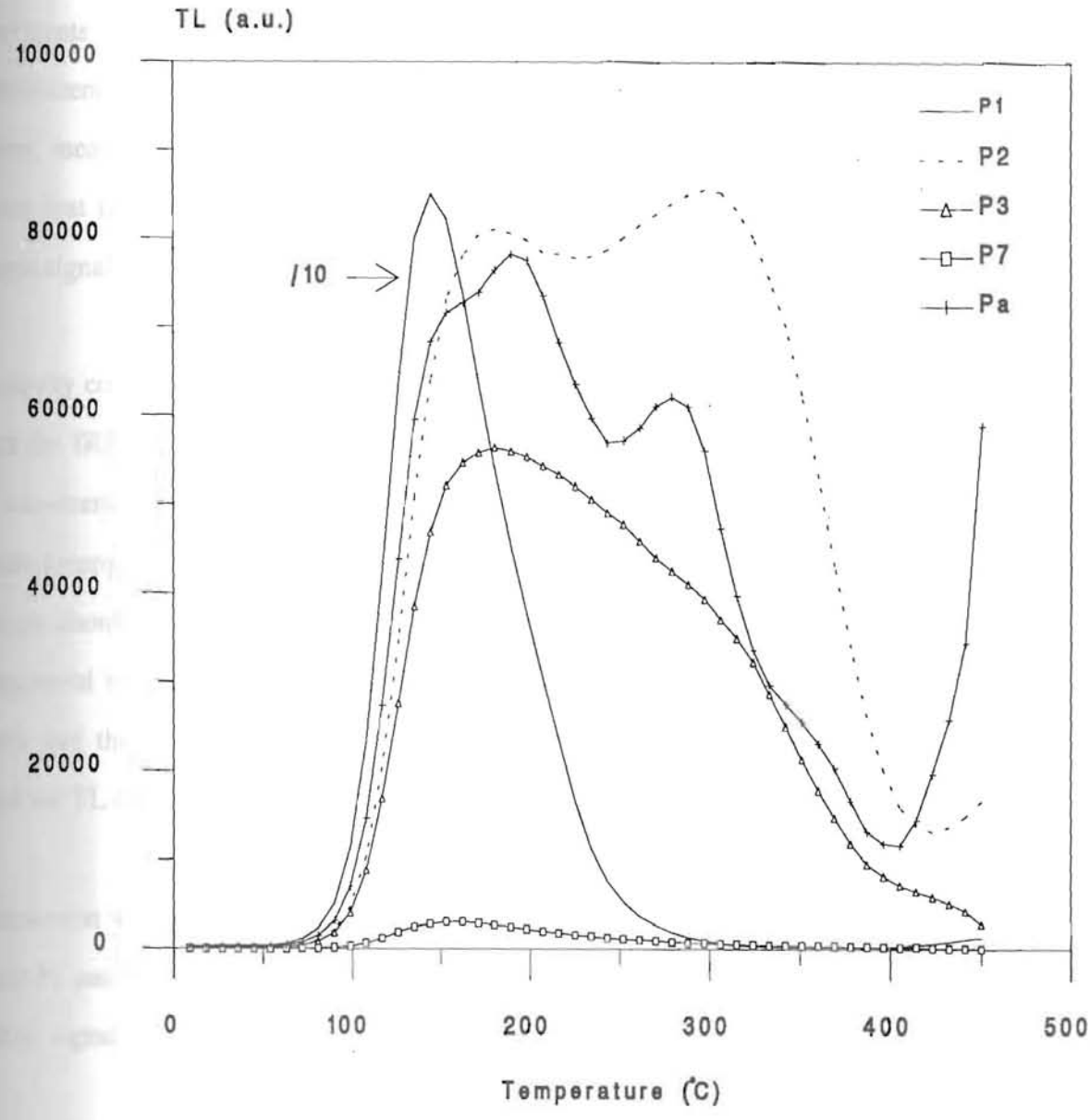


Figure 3.2.3. TL glow curves of plagioclase feldspars. TL of sample P_1 was reduced by a factor of 10.

measurements were carried out less than 1 hour after irradiation. The signal may contain phosphorescence - a decaying signal observed after the activation process is ended. However, measurements carried out for the same samples without IR stimulation have indicated that the phosphorescence has only a minor contribution (less than 10%) to the measured signal.

A sensitivity comparison of both the IRSL and TL signals was achieved by looking at the ratio of the IRSL measured with a 1 second exposure and the TL signal integrated from room temperature to 450°C (Fig. 3.2.4). There are two orders of magnitude difference in this ratio among these samples. It varied from about $2*10^{-4}$ for samples P1 and K5, to a maximum about $3*10^{-2}$ for sample P2. The samples showing a relatively high IRSL/TL ratio appeared to be those having significant TL signals at temperature above 250°C. This suggests that the IRSL signal is not produced from all of the trapped electrons which produce the TL signal.

No connection was found between the IRSL/TL ratio and the mineralogy or composition. Samples P1 and P2 had very different ratios. K5 has a sizeable TL signal but shows a very low IRSL signal.

The IRSL signal of all these samples was erased either by bleaching for 2 hour in the SOL2 solar simulator or by heating to 450°C.

After the same set of discs had been heated to 450°C, the discs were placed on a plate 20 cm below the u.v. lamp (see chapter 2) for 30 minutes. The IRSL was then measured with 1 second IR exposure on the Riso OSL/TL system.

Fig. 3.2.5. shows the ratio of u.v. regenerated IRSL and the TL signal. The differences were even larger than those observed for the IRSL created by the ß doses. The ratio varied by

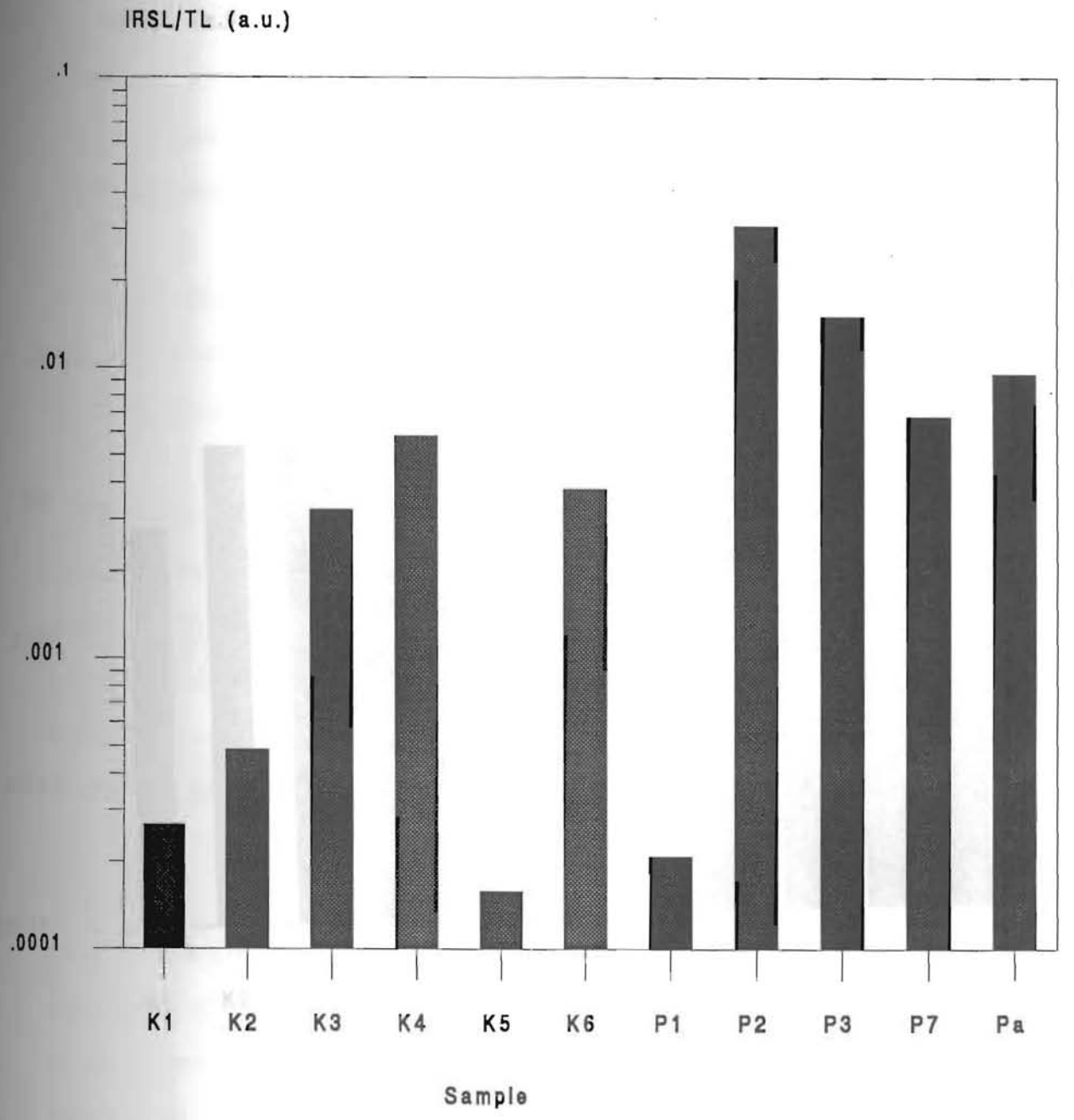


Figure 3.2.4. Ratio of beta induced IRSL from 1 second IR exposure and the TL signal integrated from 20 to 450°C for different feldspar samples.

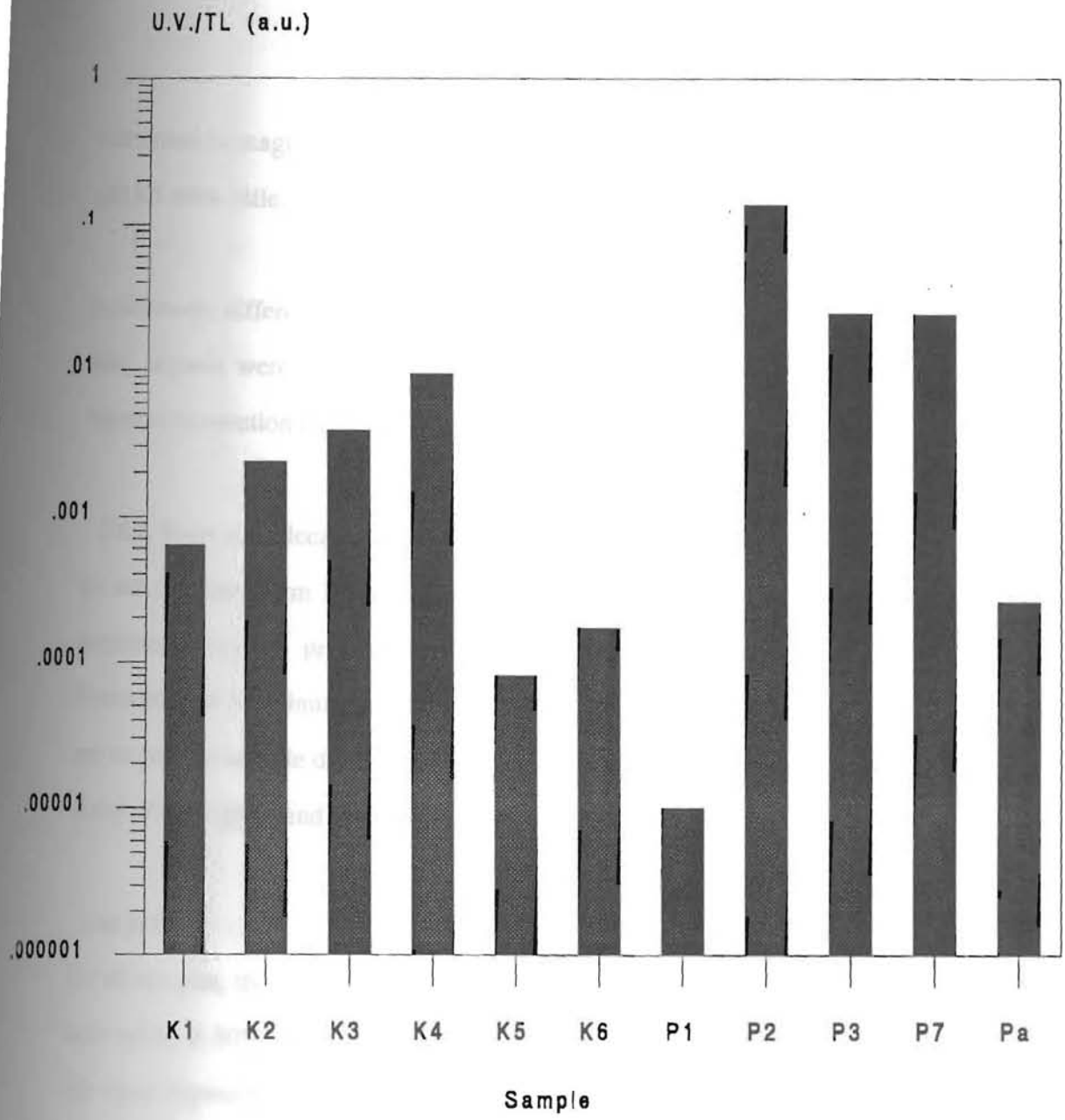


Figure 3.2.5. Ratio of u.v. (1 hour) regenerated IRSL from 1 second IR exposure and the TL signal integrated from 20 to 450°C.

four orders of magnitude from $1*10^{-5}$ for sample P1 to $1.3*10^{-1}$ for sample P2. Samples P1 and K5 show little, if any, u.v. regenerated IRSL signal.

In summary, different responses of the beta-induced TL and IRSL and the u.v. regenerated IRSL signals were found for those two groups of samples. Those responses have no apparent connection to the composition and structure of the minerals.

3.2.4. Short term decay and quick heating results

To test for short term IRSL decay at room temperature, three categories of samples were examined - (1) No preheating, (2) After preheating for 17 hours at 140°C, (3) After preheating for 10 minutes at 220°C. In addition, quick heating measurements were carried out on parallel sample discs of each category. The aim was to find a connection between the decay of the signals and the related trap depth.

1. *no preheat*

For all samples, the IRSL signal was detected using the Daybreak system. All measurements were made at 30°C with a 0.1 second IR exposure. As mentioned previously (Chapter 2), this short exposure to IR does not significantly affect the IRSL signal even after 10 such measurements. Observations of IRSL signal decay with time were made using the same group of discs with each sample represented by four aliquots.

Following two days of sunlight bleaching to remove any IRSL signal from the museum specimens, the samples were ß irradiated with between 3 and 20 Gy according to their sensitivity. The first measurements were performed less than 3 hours after irradiation. The storage time began after this measurement.

The IRSL signals from subsequent measurements were normalized to the first measurement. Fig. 3.2.6. shows the decay of the IRSL over an 84 day period. Decay of the IRSL signal

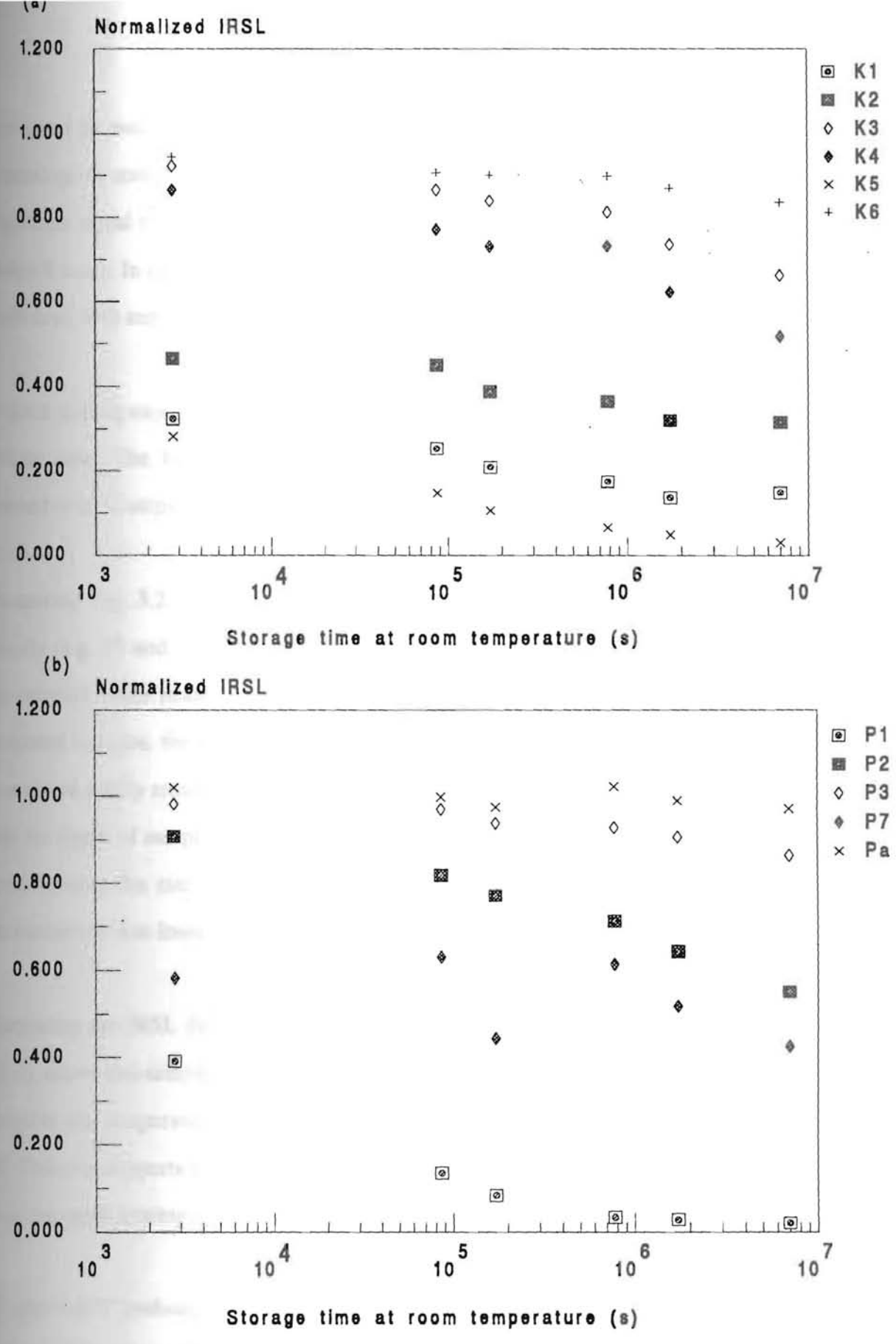


Figure 3.2.6. Decay of IRSL at room temperature. (a) K-feldspar samples (b) plagioclase feldspar samples. Samples were not preheated after irradiation. Each signal was normalized to the first measurement (see text). Each point is the mean of four aliquots.

was found for both sample groups (K-feldspar and plagioclase feldspar) regardless of their mineralogy or composition. One exception is sample Pa (albite) which does not show any significant signal reduction. On the other hand, sample P1 shows 70% fading of the signal within 8 hours. In the K-feldspar group, all samples have shown a significant loss of signal in 84 days, with sample K5 (microcline) showing the fastest decay.

A quick heating experiment, as described in Chapter 2, was conducted on a parallel set of sample discs. The heating rate was 5°C/s and a maximum temperature of 450°C was reached in 25°C steps. The results are shown in Fig. 3.2.7..

Comparing Fig. 3.2.7. and Fig. 3.2.6, it appears that the samples which decayed most quickly (e.g. P1 and K5) show a sizable IRSL signal associated with shallow traps which are emptied when heated to a relatively low temperature. For these irradiated, but un-preheated samples, the signal from sample P1 decayed quickly (Fig.3.2.6) and the signal was almost totally erased after quick heating to a temperature of only 175°C. In contrast to this, the signal of sample Pa was hardly affected until it was heated up to 275°C. A similar trend between the rate of decay and the emptying at shallow traps during quick heating measurements was found for all samples.

Comparing the IRSL decay curves (Fig. 3.2.6) and TL glow curves (Fig. 3.2.2 and Fig. 3.2.3), shows that samples exhibiting very rapid loss of IRSL signal are samples which have sensitive low temperature TL peaks. This was clearly demonstrated by samples P1, K1 and K2. This also supports the connection between fast natural fading and shallow traps deduced from the quick heating experiment.

2. After 140°C preheating

After two days bleaching with sunlight, aliquots of the 11 samples were irradiated with ß doses from 50 to 400 Gy according to the IRSL sensitivity. High doses were required for

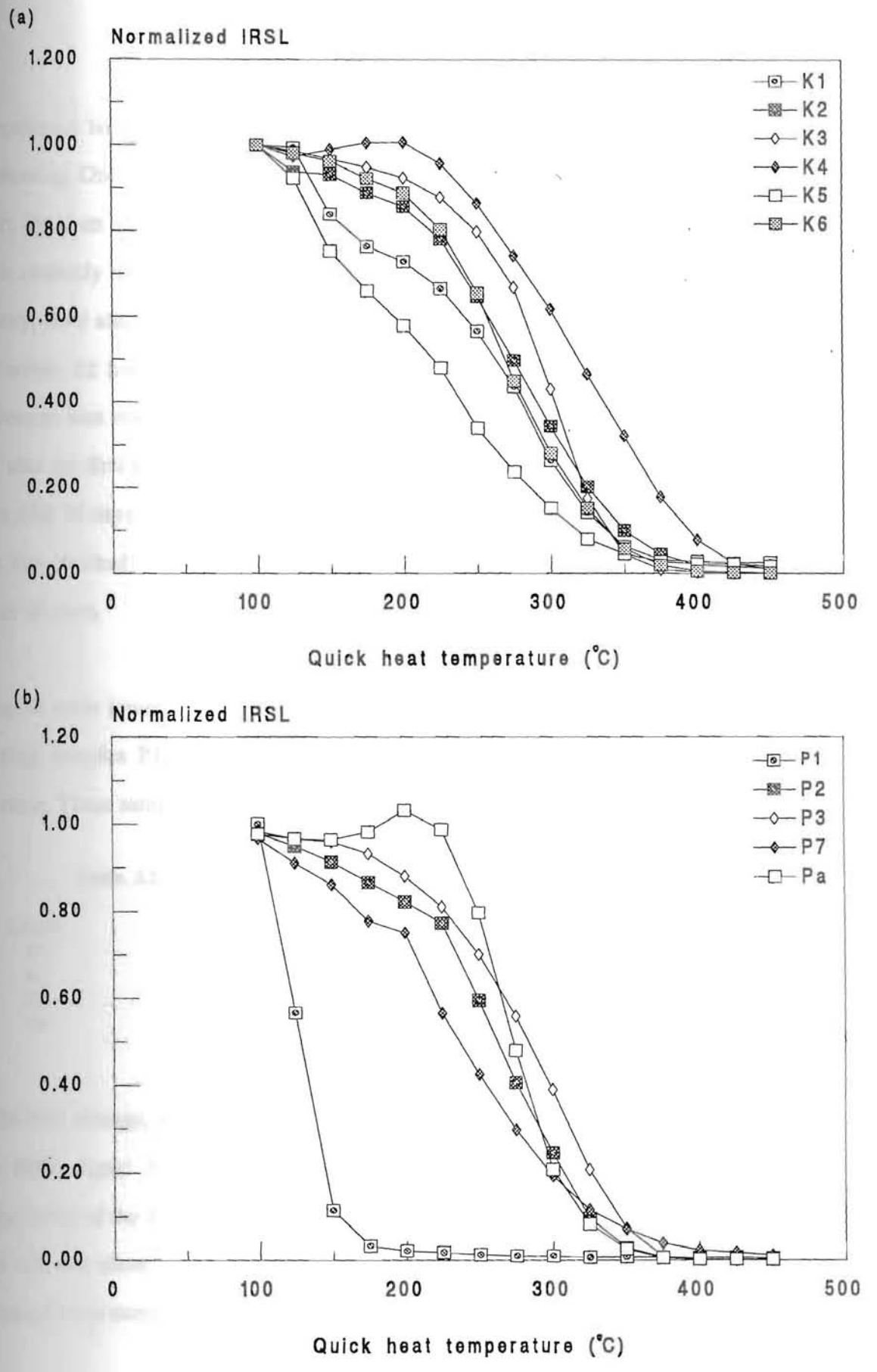


Figure 3.2.7. Quick heating of feldspar samples after ß irradiation. (a) K-feldspar samples (b) plagioclase feldspar samples. Discs were heated at 5°C/s to the temperature shown and cooled to 50°C. IRSL was measured at 50°C with a 0.1 second IR exposure.

this experiment because of the considerable reduction in IRSL intensity brought about by the preheating. One fine grained (4-11 μm) loess sample (QTL89D; see Wintle 1987) from Rocourt, Belgium was added for comparison, since preheating at 140°C for more than 16 hours is routinely used for removal of unstable components in fine grains samples in our laboratory. Four aliquots were used for each sample. Discs were preheated for 17 hours at 140°C within 12 hours of the irradiation. Shortly after preheating, the first IRSL signal measurement was made with a short IR exposure (0.1 second) at 30°C. The storage time started after the first measurement. Subsequent measurements were made on the same group of discs after 26 days of storage at room temperature (RT). The stability of the measurement system was checked by including measurements of natural IRSL from loess, both initially and after 26 days.

Because of their lower sensitivity and the removal of most of remaining signal by the preheating, samples P1, P7, K1 and K5 did not have a large enough signal for further comparison. These samples were therefore excluded from this experiment.

Table. 3.2.2. Fading (after 140 °C preheating) for 26 days storage at RT

LABEL	REMNANT IRSL (%)	LABEL	REMNANT IRSL (%)
K2	94±2	P2	87±2
K3	91±2	P3	98±0.4
K4	90±2	Pa	98±1
K6	93±2	loess	97±3

After 26 days storage, samples K2, K3, K4 and K6 in the K-feldspar group showed fading of the IRSL signal from 6 to 10 percent (Table 3.2.2). The error terms reflect the reproducibility of the 4 discs. In the plagioclase group, P2 showed a 13 percent loss, but P3 and Pa did not show any significant (i.e. >5%) reduction of signal (Table 3.2.2). The polymineral loess sample did not show significant fading of the signal.

Since the shallow traps were emptied by the preheating (Fig. 3.2.8), the loss of signal is

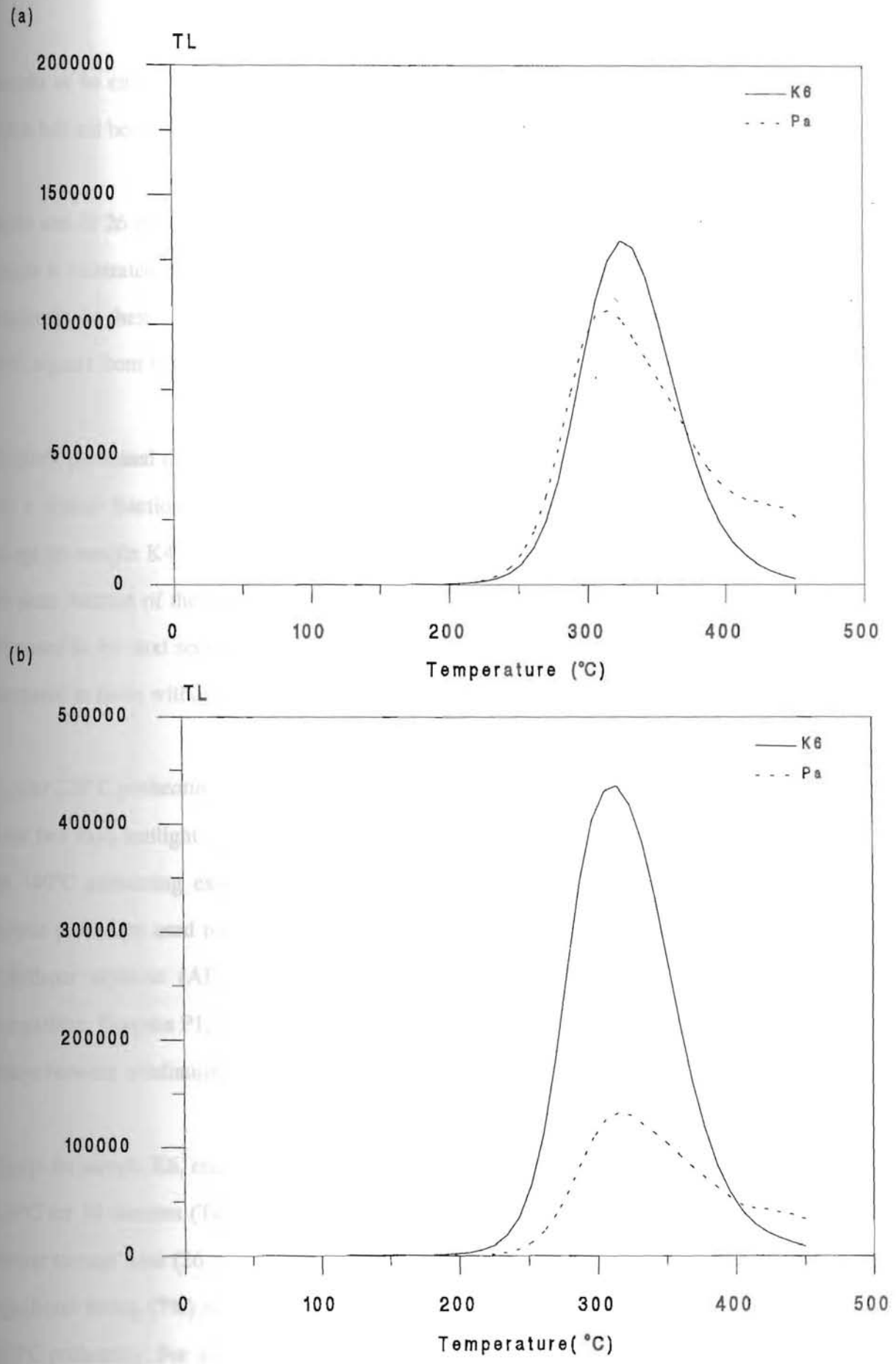


Figure 3.2.8. TL glow curves of feldspar samples after preheating. (a) after preheating at 140°C for 17 hours (b) after preheating at 220°C for 10 minutes.

thought to be caused by the anomalous fading of electrons in deep traps (Wintle, 1974) which had not been removed by the preheating.

At the end of 26 days storage, the IRSL and TL signals were measured. The ratio of both signals is illustrated in Figure 3.2.9. Except for sample K2, the ratio is of a similar order of magnitude for these samples. This suggests a relatively homogenous contribution to TL and IRSL signals from the traps.

Similarly preheated discs were used for a quick heating measurement. The results indicated that a similar fraction of the IRSL signal was removed at a similar heating temperature except for sample K4 (Fig. 3.2.10). This shows that a higher temperature is needed to erase the same fraction of the signal. This is related to thermal stability of the signal and will be discussed in the next section. The shapes of the quick heating curves are relatively uniform compared to those without preheating (Fig. 3.2.7).

3. *after 220°C preheating*

After two days sunlight bleaching, the 11 samples were given the same dose as applied in the 140°C preheating experiment. Samples were preheated at 220°C for 10 minutes, a preheat procedure used routinely in our laboratory for K-feldspar samples (Li 1991). One K-feldspar separate (AF-4) from the St Paul's section, South Africa was added for comparison. Samples P1, P7, K1 and K5 were excluded because of their lack of signal. The delays between irradiation, preheating, and the first measurement were less than 12 hours.

Except for sample K6, results from storage for 28 days at room temperature after preheating 220°C for 10 minutes (Table 3.2.3 Fig. 3.2.11 and Fig. 3.2.12) are similar to those after a similar storage time (26 days) after the 140°C preheating for 17 hours. Sample K6 shows significant fading (7%) after 140°C preheating for 17 hours but does not show fading after 220°C preheating. For all of the samples, the signal loss for discs which experienced the

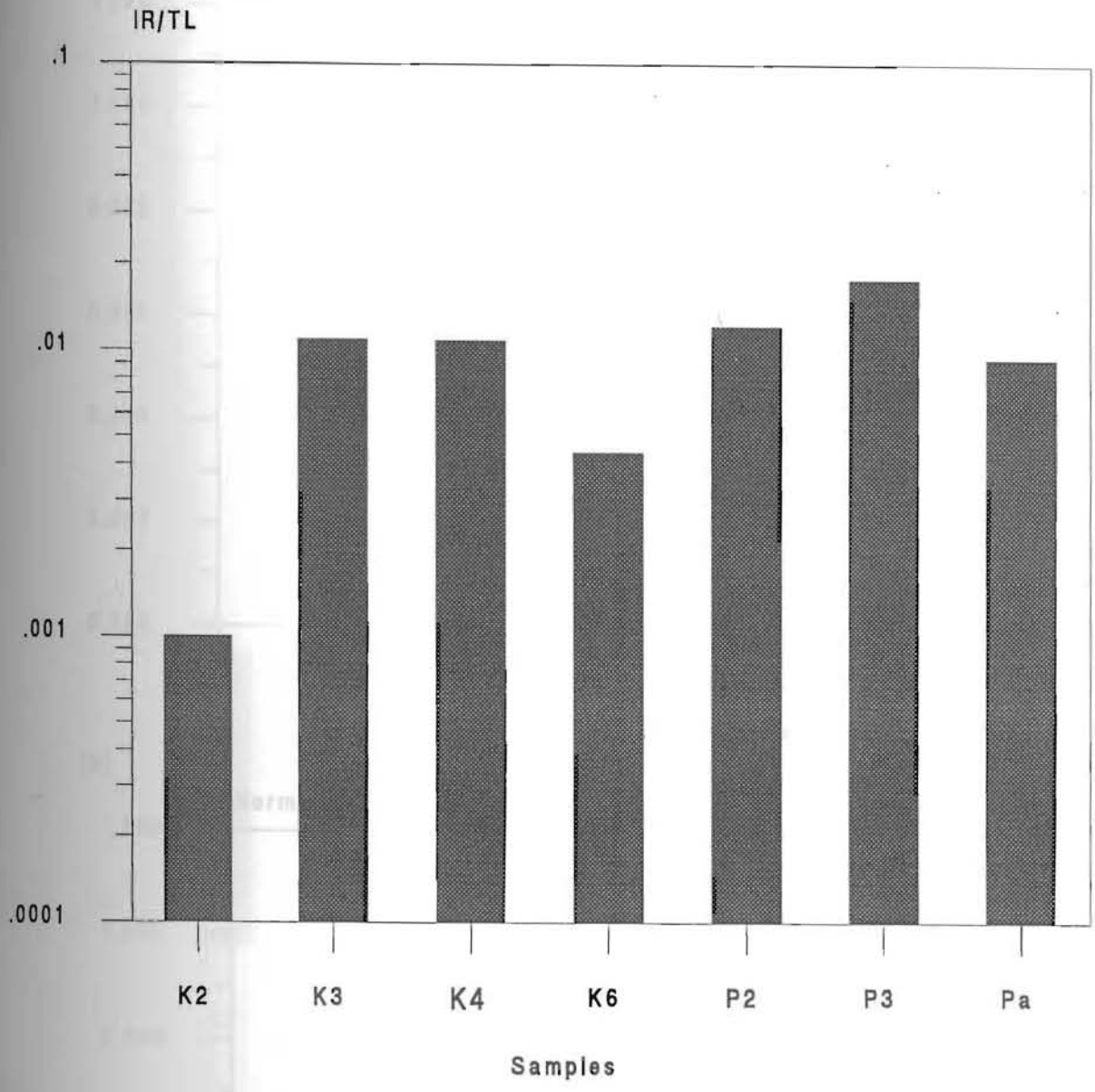


Figure 3.2.9. Ratio of IRSL from 1 second IR exposure and TL signal integrated from 20 to 450°C. Samples were preheated for 17 hours at 140°C after 12 Gy ß irradiation.

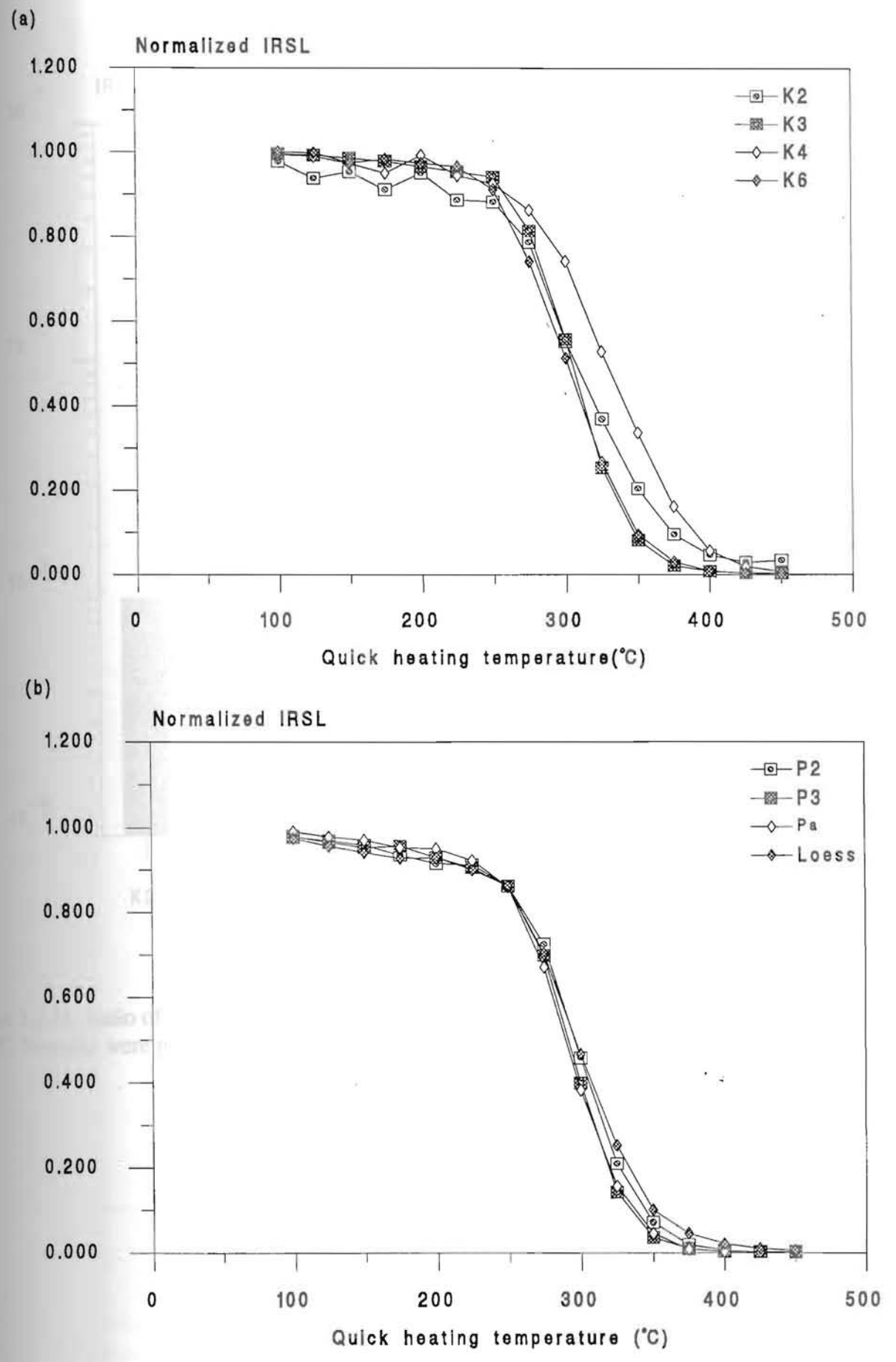


Figure 3.2.10. Quick heating of feldspar samples after 17 hours preheating at 140°C (see text). (a) K-feldspar samples (b) plagioclase feldspar samples.

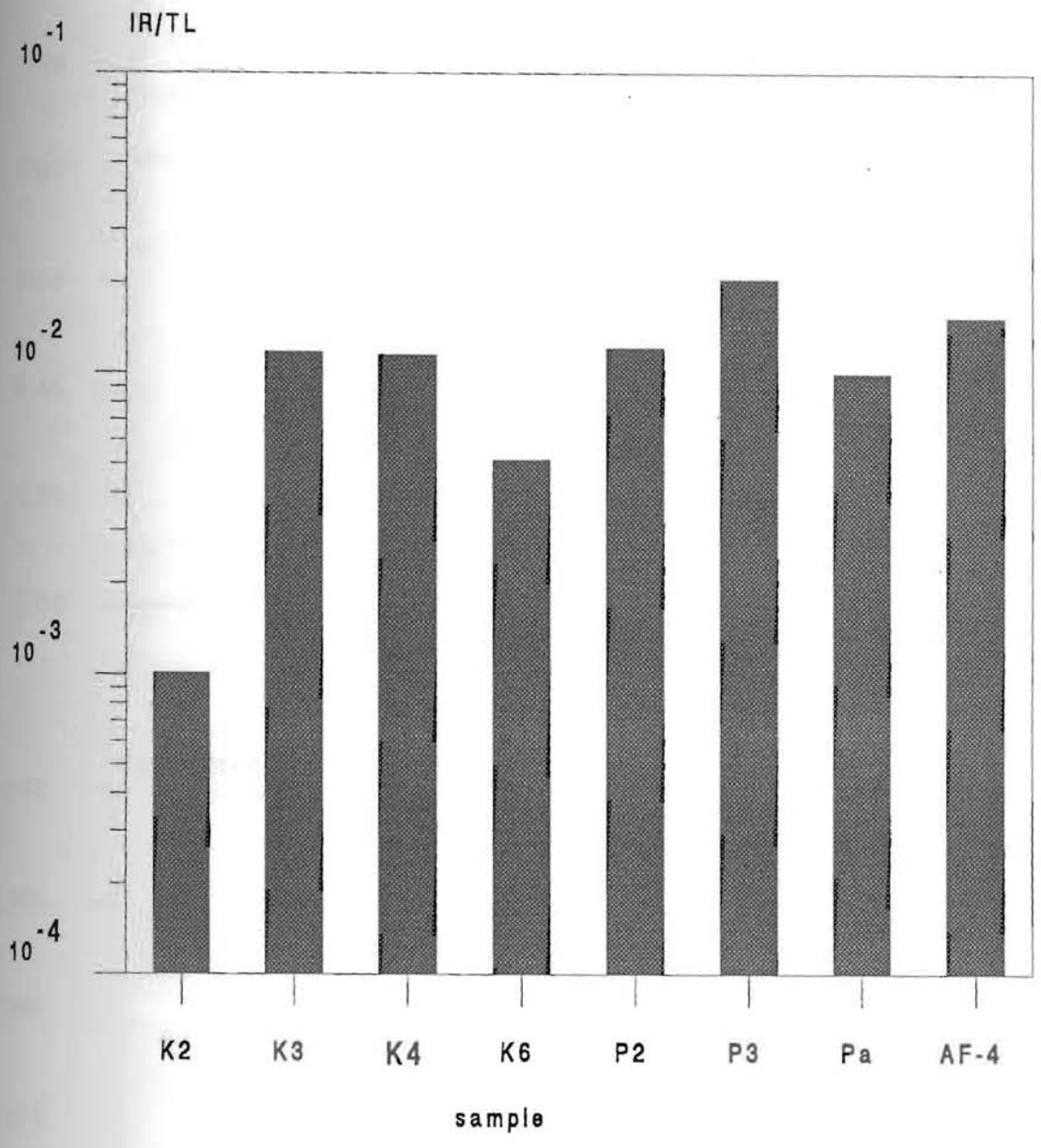
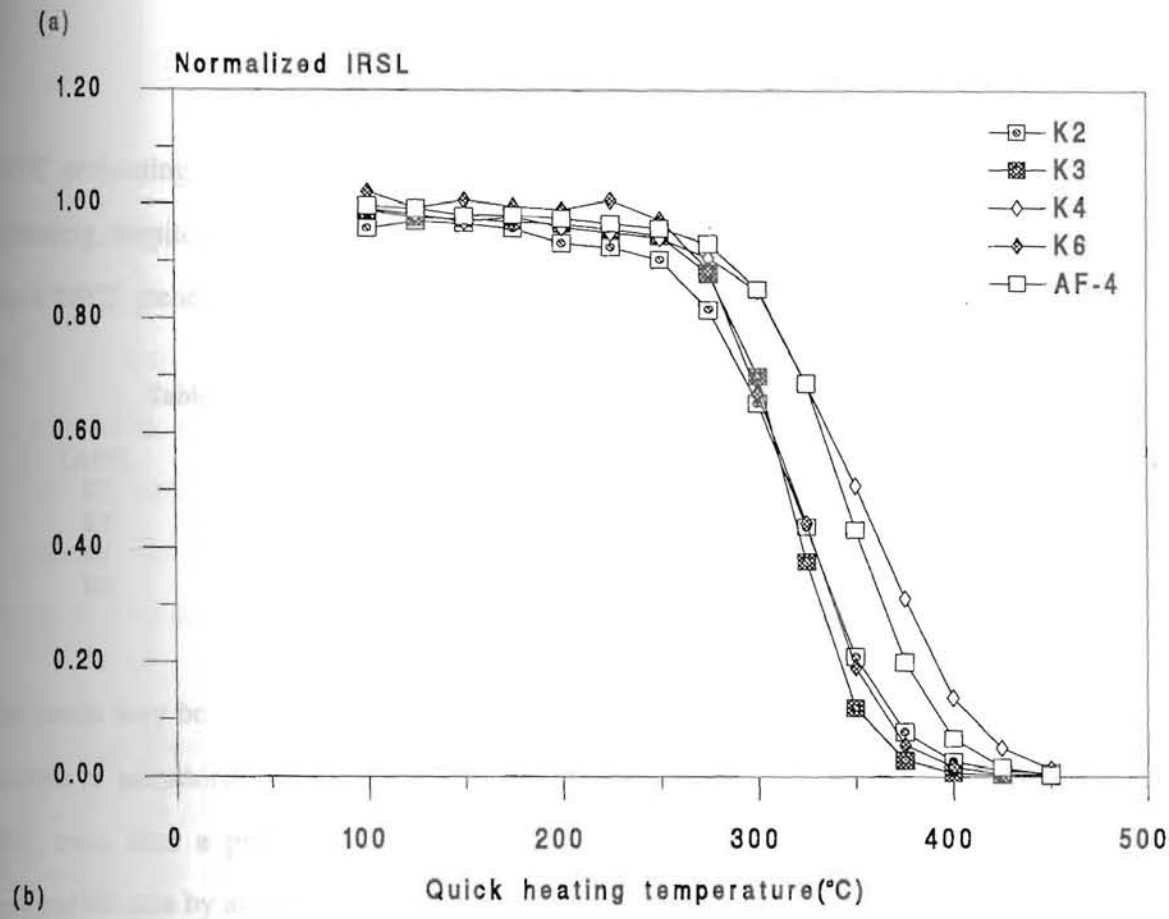


Figure 3.2.11. Ratio of IRSL from 1 second IR exposure and TL signal integrated from 20 to 450°C. Samples were preheated for 10 minutes at 220°C after 12Gy ß irradiation.



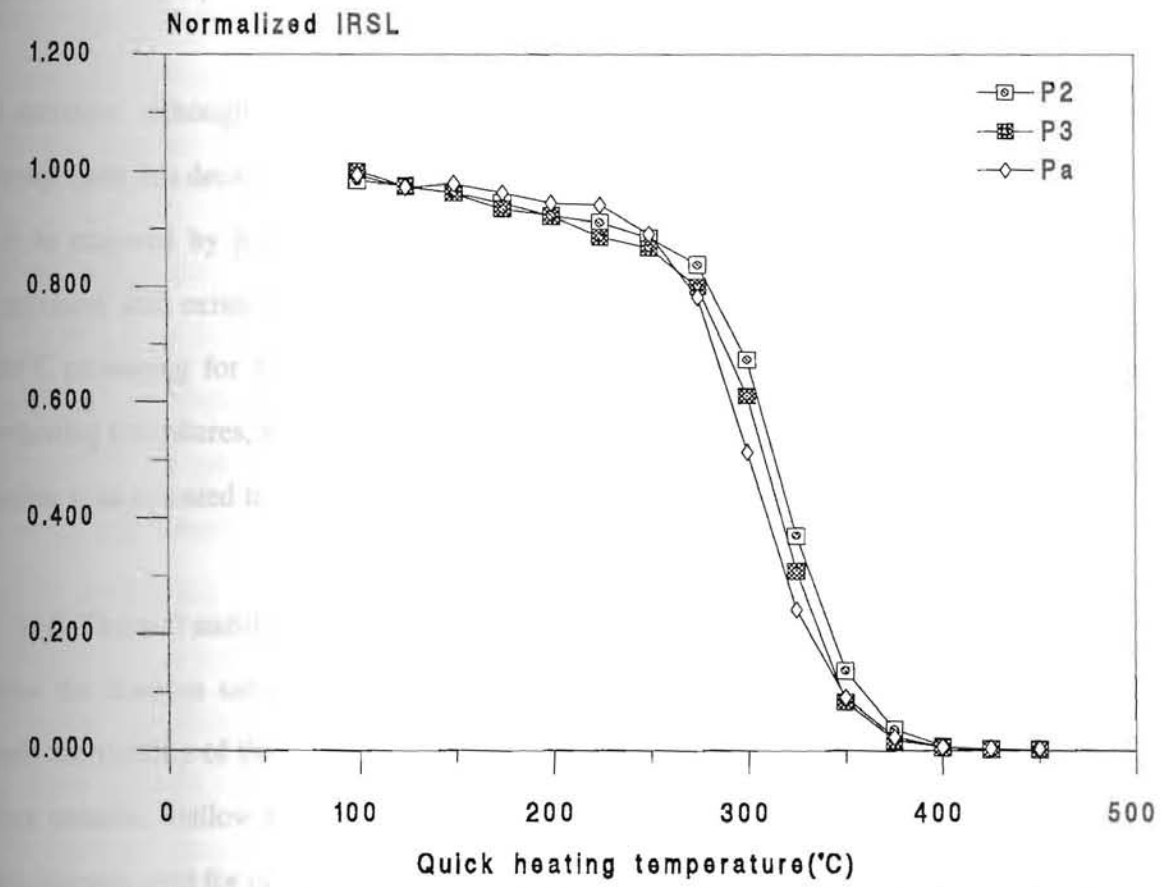


Figure 3.2.12. Quick heating results for feldspar samples after 10 minutes preheating at 220°C. (a) K-feldspars (b) plagioclase feldspars.

220°C preheating are marginally but consistently smaller than those for the 140°C preheating, despite the slightly longer (2 days) longer storage time. These results support the use of 220°C preheating for separated K-feldspar fractions (discussed further in section 3.5).

Table. 3.2.3. Fading (after 220 °C preheating) for 28 days storage at RT

LABEL	REMNANT IRSL (%)	LABEL	REMNANT IRSL (%)
K2	96±2	P2	89±1
K3	94±2	P3	100±3
K4	90±2	Pa	97±1
K6	103±3	AF-4	94±1

The results may be compared with similar decay studies carried out by Spooner (1992) on samples of labradorite and sanidine. Both showed continued loss of IRSL on storage at 10°C, even after a preheat of 1 minute at 220°C, over a 4 month period. The decay mirrored the loss by anomalous fading of high temperature TL.

In summary, although a limited number of samples were examined, some general trends emerge from this decay study. Samples in both groups show a fast fading component which can be removed by preheating samples after irradiation. However, an anomalous fading component still exists in several samples even after 140°C preheating for 17 hours, or 220°C preheating for 10 minutes. The existence of anomalous fading, after either of two preheating procedures, suggests that the anomalous fading cannot be removed by storing the sample at an elevated temperature.

3.2.5. Thermal stability

Since the museum samples had been previously exposed to sunlight, it is meaningless to study the stability of their natural luminescence signals. On the other hand, there is a signal from unstable shallow traps after irradiation which does not exist in the natural sediment any minerals used for dating. Hence experiments to study longer term thermal stability were performed on irradiated samples which had been preheated for 10 minutes at 220°C.

Thermal stability at ambient temperature may be predicted by determining the depth of the electron trap depth (Aitken 1985). Methods, such as isothermal decay, Hoogenstraaten's method and initial rise, are commonly used in TL studies (Wintle 1974, Strickertsson 1985). Since the IRSL signal is associated with many different types of traps, it is impossible to separate the signal from a particular type of trap. However, the isothermal decay method can give an idea of the relative thermal stability of the IRSL signal. In isothermal decay experiments the samples are stored at elevated temperatures.

In this experiment the isothermal decay of the IRSL signal was observed after storing sample discs at 220°C for different lengths of time using the Risø OSL/TL system. The IRSL signal was measured at 50°C for a half second IR exposure. IRSL signals were normalized to the signal after the initial 10 minute preheat at 220°C. The decay curves was plotted on a log/linear plot, which would give a straight line for a single exponential decay, and on a linear/log plot.

The isothermal decay curves are considerably different for different samples (Fig. 3.2.13). The signals from the K-feldspar group show more resistance to heating than the plagioclase group, with samples K1, K4 and K6 being significantly more stable than the others. Apart from sample K1 (an anothoclase with a relatively dim signal), K4 and K6 are microcline minerals which have a sensitive response to irradiation dose. Orthoclase sample K3 has a similar composition to K4 but shows a significantly lower thermal stability. Similar results were obtained in experiments on other microcline feldspars. Although only a few samples have been examined in this study, the higher thermal stability shown by microcline feldspars suggests that there might be a link with the mineral structure. Obviously, more work on these minerals is required to confirm this tentative conclusion.

Analogous experiments have indicated that K-feldspar fractions from sediments have a

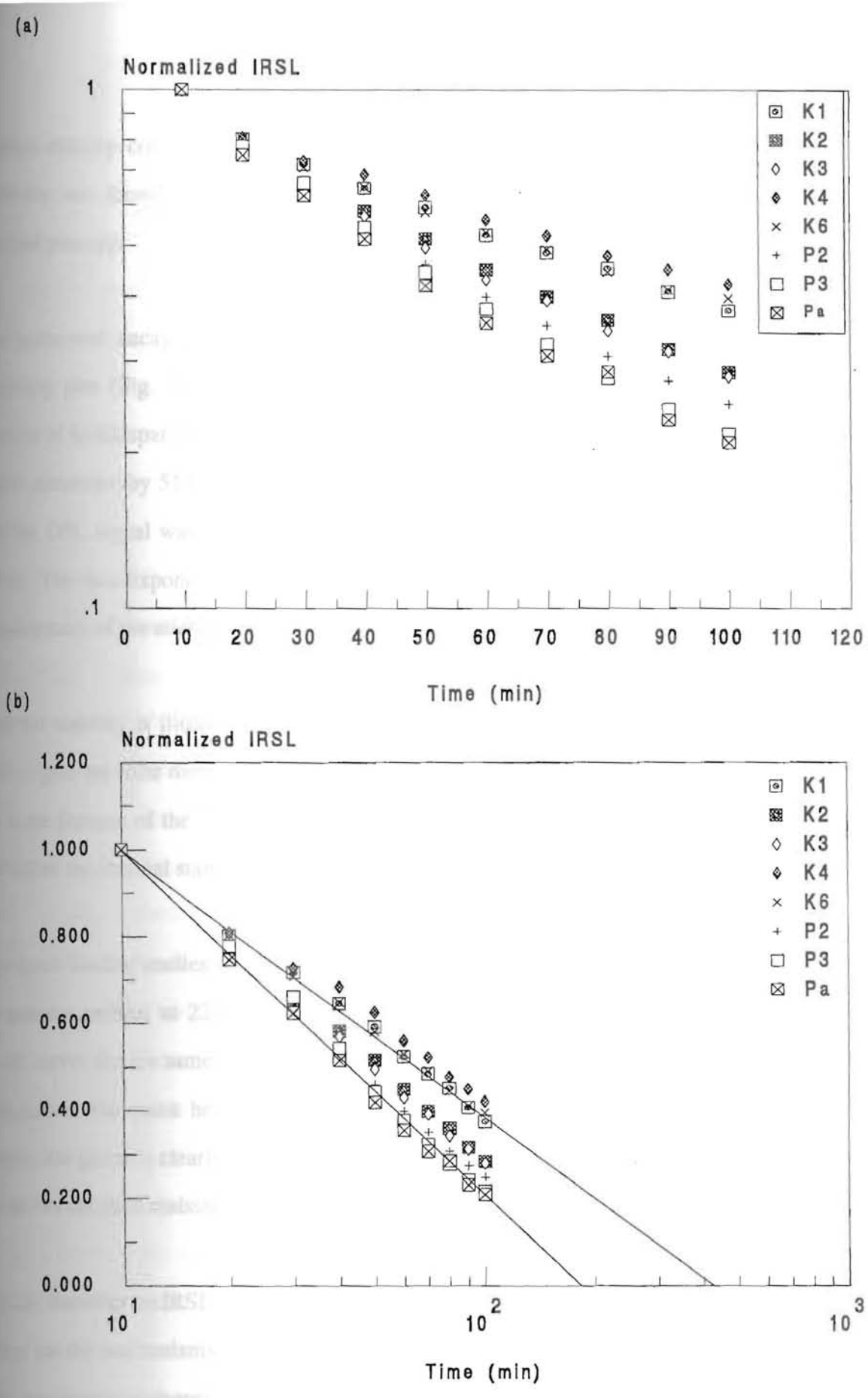


Figure 3.2.13. Isothermal decay of IRSL signal at 220°C. The signal was normalized to the measurement following the first preheat. (a) linear scale for the storage time axis and log scale for the IRSL. (b) data plotted with linear scale for IRSL but with log scale for storage time axis. (The straight lines are for guidance only).

thermal stability comparable to that of the microcline feldspars (see section 3.4). This similarity was found for K-feldspar samples from different continents regardless of the regional geology.

The isothermal decay curve is not exponential (Fig. 3.2.13a). It is almost linear on a linear/log plot (Fig. 3.2.13b). A similar form of isothermal decay has been found for samples of K-feldspar from different parts of the world. It thus differs from that for the OSL signal stimulated by 514.5 laser light from quartz (Rhodes 1990), for which it was claimed that the OSL signal was from the trap associated with the 325°C TL signal (Smith et al. 1986). The non-exponential isothermal decay of the feldspar IRSL signal led to the abandonment of the attempt to make an Arrhenius plot to determine the trap depth.

Thermal stability is illustrated in the quick heating curves. If the electrons relating to the IRSL signal are from deeper traps, a higher quick heating temperature is required to remove the same fraction of the signal. Hence, the more resistance samples show to quick heating, the higher the thermal stability of their signals.

The quick heating studies were carried out on samples which had previously experienced a 10 minutes preheat at 220°C. The results (Fig.3.2.12) are consistent with the isothermal decay curves for the same samples. The high stability shown for isothermal decay was also indicated in the quick heating curves. The difference between the K-feldspar group and plagioclase group is clearly demonstrated in the quick heating result. The microcline sample K4 shows the most resistance to quick heating.

3.2.6. Remarks on IRSL mechanism

What are the mechanisms involved in the IR stimulation processes and what is the cause of non-exponential isothermal decay of the IRSL signal? Hütt et al. (1988) suggested that the IR stimulation process involves electrons being excited to an intermediate state, thermally

moved into the conduction band and then recombining at luminescence centres. Since IRSL spectra studies (Bailiff and Poolton 1991, Huntley et al. 1991) show a similar emission spectrum for both TL and IRSL, it is likely that the electrons were excited to the conduction band and then recombined at the luminescence centres, as in TL production (Fig.3.2.14). If the electrons giving rise to the IRSL were trapped at one type of trap, as has been found for the OSL signal from quartz, the isothermal decay curve would be exponential. However, the isothermal decay curves are found to show 1/t dependence for samples of polyminerals (Li and Wintle 1992) and for the pure feldspar crystals used in this study.

In early studies of TL phenomena, it was suggested that a 1/t form of isothermal decay is possible when assuming a uniform continuous distribution of energies from zero to infinity (Randall and Wilkins 1945). More recently, another study suggested that the same 1/t decay would be given if the activation energies are represented as a uniform, continuous distribution with an energy range ΔE (Hornyak and Chen 1989). It was claimed that for activation energy distributions which are more or less bell-shaped, the 1/t dependence sets in when the ratio $kT/\Delta E$ is less than about 0.15. If this is assumed for feldspar, the minimum value for ΔE is about 0.3-0.46 eV for the temperatures from 75°C to 260°C used for isothermal (or preheating) decay experiments (Fig.3.2.14). When electrons are excited by IR from deep trapping levels to intermediate levels from which they are thermally released at room temperature into the conduction band (Hütt et al. 1988), the activation energies for this thermal release are about 0.1 to 0.2 eV (Hütt and Jaek 1989, Bailiff and Poolton 1991, Duller and Wintle 1991). This spread could be the result of a similar range of energy difference in excited electrons in the deep trapping levels. If there is a spread in the deep trapping levels, albeit somewhat greater than just indicated, this could be an explanation of the 1/t isothermal decay. In this case the thermal stability of electrons in the lower activation energy states will be different to that for the higher energy states. Another possible reason is that the IR radiation is not monochromatic but has a wavelength distribution (Δ=80 nm) which could cause a 0.07 eV energy difference.

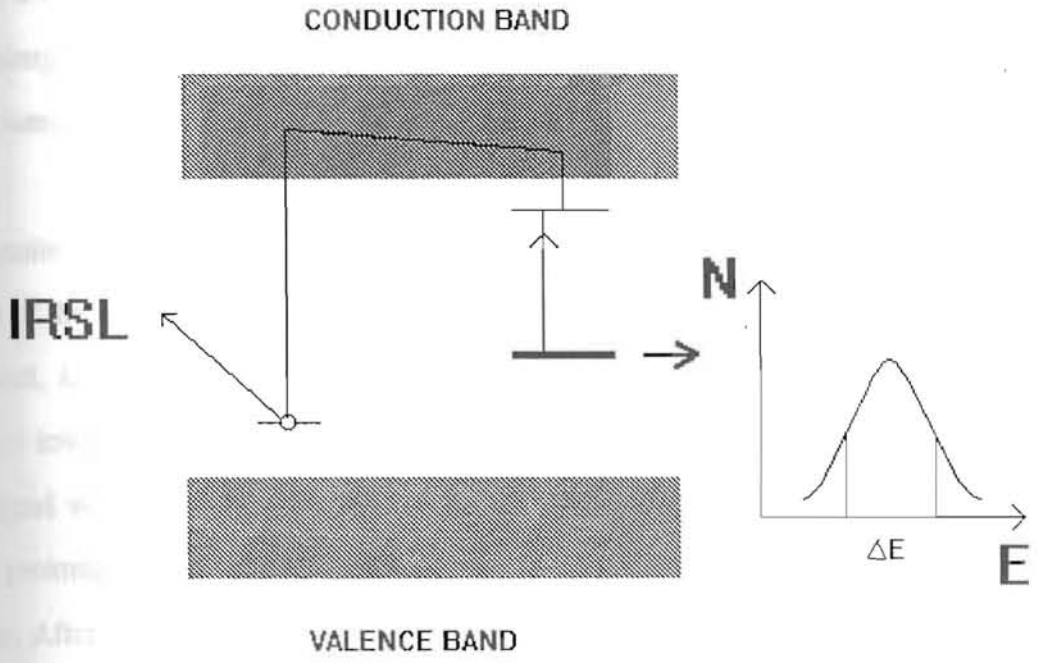


Figure 3.2.14. Diagram of IR stimulation process. The activation energy has a continuous distribution in energy range ΔE.

3.2.7. U.V. regenerated IRSL in feldspars

Since the u.v regenerated IRSL signal can be used for normalization of the signal measured in bulk samples (see section 4.4), it is of interest to study the u.v. response of individual feldspar samples.

Some results have already been mentioned (Fig.3.2.5). Further studies were carried out using all of the fourteen samples. To make sure that the natural IRSL signal of the samples was erased, sample discs were measured after prolonged sunlight exposure. These discs were then irradiated with a test ß dose (20 Gy). After an overnight delay (16 hours), the IRSL signal was measured with 20 second IR exposure on the 2000 OSL reader. After another prolonged exposure to sunlight, these discs were then exposed to u.v. radiation for one hour. After an overnight delay (16 hours), the IRSL signal regenerated by the u.v. was measured with a 20 second IR exposure. The ratio of the IRSL signals from both measurements is shown in Figure 3.2.15.

Since it was planned to use the u.v. regenerated signal for normalization, it was hoped that the u.v. regenerated signal would be proportional to the signal created by the test ß dose. However, the ratio of both measurements differs considerably among these samples (Fig.3.2.15). Samples Pa and K6 gave a very bright signal after ß irradiation but showed a relatively dim u.v. regenerated signal. Sample P2 showed the most sensitivity to u.v. irradiation whereas sample P1, P4 and P5 showed little, if no, response to the u.v.. This suggests that normalization by the u.v. regenerated signal will only be suitable for use when samples have a *similar mineralogical composition*. This will be discussed in detail in Chapter 4.

The discs used in the experiment above were exposed repeatedly to bleaching and regeneration cycles. Discs were bleached for a day by sunlight in Aberystwyth (not less than 6 hours), and then exposed to u.v. for one hour. The IRSL measurement was made in the

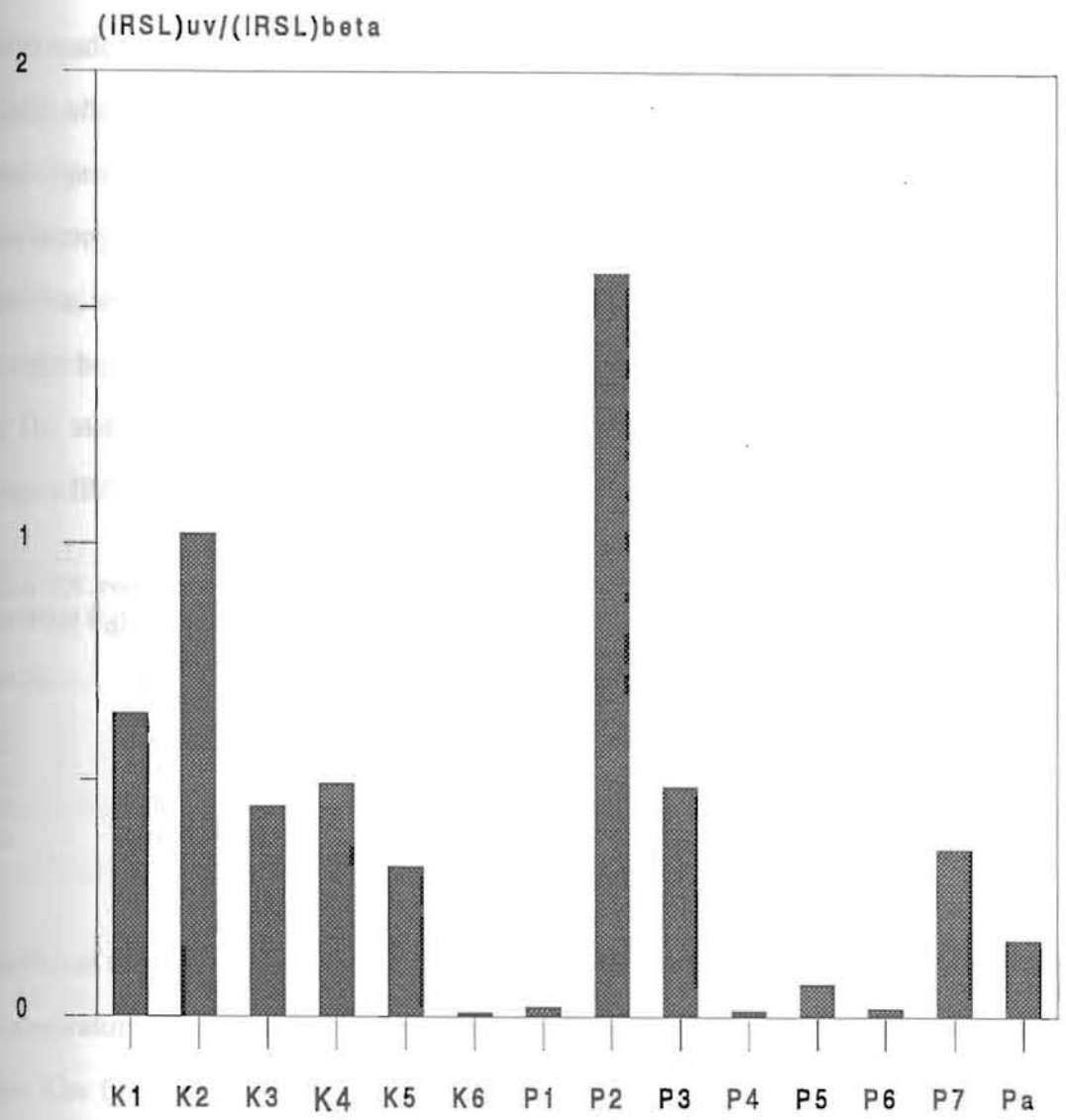


Figure 3.2.15. Ratio of u.v. (1 hour) regenerated IRSL signal and IRSL created by a ß dose (20 Gy).

OSL 2000 reader after an overnight (16 hours) delay. The u.v. regenerated signal declined consistently after each cycle for all of the feldspars. The reduction rate became less as the cycle was repeated. This signal loss was approximately 40% of the total signal, with some variation among samples. There is a constant IRSL signal (around 60% of the total) after u.v. exposure, which does not decline during the experiment. The removable fraction of the signal might be related to charge transfer, similar to that observed by Bailiff and Poolton (1991). The stable component might be the result of competition between excitation and bleaching in IRSL correlated traps (Chen et al. 1990).

Table 3.2.4. U.V. regenerated IRSL signal (c/s) measured immediately (I_0) after u.v. radiation and an overnight delay (I_d).

SAMPLES	I_0	I_d	LEFT (%)	SAMPLES	I_0	I_d	LEFT (%)
K1	1550	210	14	P1	550	30	5
K2	4055	1260	31	P2	617300	384600	62
K3	19150	7400	39	P3	42000	31170	74
K4	1170	520	44	P7	1550	260	17
K6	9500	180	2	Pa	1300	230	18

The stability of the u.v. regenerated signal was tested by observing the decay of the signal at room temperature. Subsample discs were exposed to the u.v. after prolonged sunlight exposure. The first measurement was made immediately after switching off the u.v. lamp. After an overnight (16 hours) storage in the dark at room temperature, the IRSL measurement was repeated. The signal from the measurement after an overnight delay is significantly smaller than that without a delay (Table 3.2.4). This happened to all of the samples. One possible reason is that phosphorescence makes a significant contribution to the signal immediately after u.v. irradiation. Perhaps more importantly, the u.v. regenerated signal is relatively small compared with that created with a 12 Gy ß dose, and the phosphorescence is thus comparable with the IRSL signal. The natural fading of the IRSL signal may also occur in some of these samples because some of the electrons transferred by the u.v. exposure may fill shallow traps which show rapid fading. The decay of the u.v. regenerated IRSL signal suggests that a delay between u.v. exposure and signal

measurement is necessary to minimize the phosphorescence when the signal is being used for normalization.

3.2.8. Summary

Different beta-induced TL and IRSL and u.v. regenerated IRSL responses were found for the pure feldspars used in this study.

Anomalous fading occurs for the IRSL of some K-feldspar and plagioclase feldspar minerals, and cannot be related simply to the mineralogical structure or element composition. The fading process can be accelerated by preheating, though a stable level may not be reached.

A short term unstable IRSL component related to shallow traps was found for the samples, and removal of such components is thus an essential step in IRSL dating.

It appears that alkali feldspars are more thermally stable than plagioclase feldspars. The isothermal decay curves are not simple exponentials. A continuous activation energy distribution, rather than a single trap depth, was suggested.

The u.v. regenerated IRSL was found to be unstable over 16 hours at room temperature, but signal decay was probably due to phosphorescence.

3.3 MIXED MINERALS (LOESS)

3.3.1. Introduction

Loess, an aeolian deposit, is a material which has often been used in luminescence dating studies, because the long transportation of the grains ensures that that the luminescence signal which had previously accumulated in minerals was well bleached prior to deposition.

During the past few years, many ages obtained by TL dating and optical dating methods using fine grain loess have been found to be younger than the expected ages for samples older than 100ka (Debenham 1985, Li 1989, Wintle 1990, Packman and Grün 1992). It has been claimed that the lower apparent age is due to the lack of stability of the TL and OSL signal (Debenham 1985, Li 1989, Wintle 1990, Xie and Aitken 1991). There are counter claims that the problem is caused by use of the regeneration method for samples which experience a sensitivity change after exposure to laboratory light sources (Berger 1988a, Rendell and Townsend 1988). In this section I try to ascertain whether the thermal stability differences in the IRSL signal are due to a mineral difference or local geology by looking at the IRSL signal of samples of similar age from different locations around the world. Materials from one section with different ages were also studied.

3.3.2. Samples used and minerals contained in loess

The samples used in the global loess study were from several locations around the world (Fig.3.3.1)-Lanzhou China (753i ED 170Gy); Karlich, Germany (QTL45B; ED 51Gy); Rocourt, Belgium (QTL89D; ED 55Gy); Tapiosuly, Hungary (QTL84Z; ED 83Gy) and Alaska, U.S.A. (GHC27; ED 74Gy). As obtained by TL, most of these loess samples have an age of 10 to 20 ka (Wintle 1985a, Wintle 1987, Wintle and Packman 1988, Clarke 1987), except for the sample from Lanzhou which has an age of 36ka. These young ages ensure that the natural TL and IRSL signals are not saturated and that the signal is well above the PM tube and IR background (about 100 c/s).

To examine whether the stability differs between young and old loess, samples of different ages from one section were also studied. Samples were from loess and soil units, L1, S1, L3, S3, L5, S5 and L8 in Luochuan, Shaanxi, China (see Appendix and Liu et al. 1985 for details). The estimated ages are 73, 125, 300, 300, 500, 500 and 730 ka respectively (Kukla 1987).

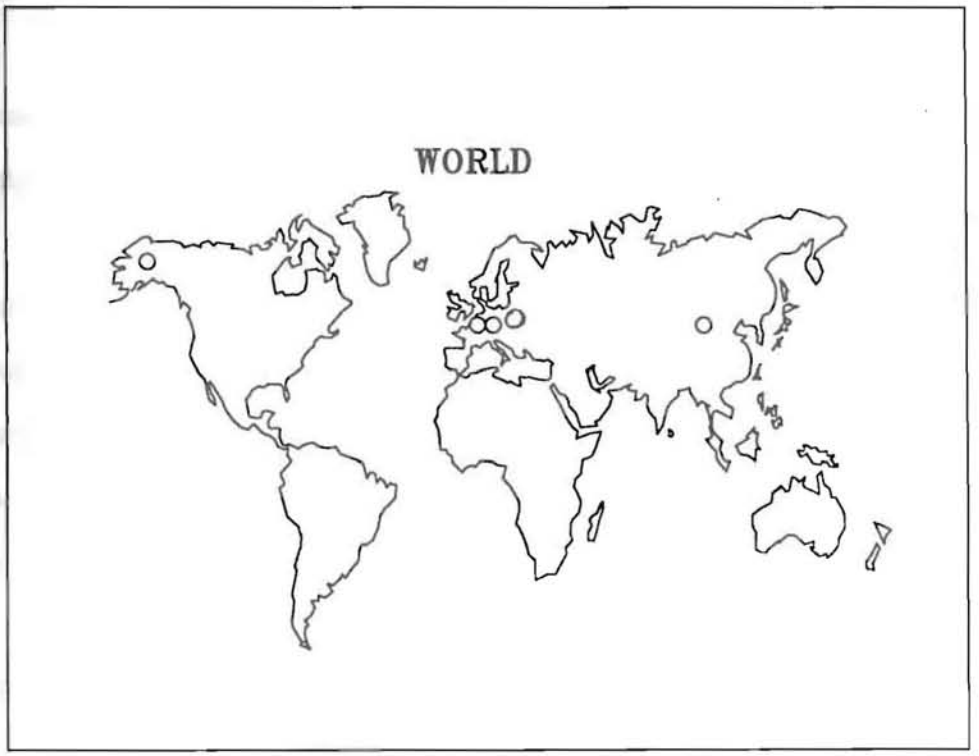


Figure 3.3.1. Sample locations 1) Lanzhou, China: 2) Karlich, Germany: 3) Rocourt, Belgium: 4) Tapiosuly, Hungary: 5) Alaska, U.S.A..

Fine grain (4-11 μm) discs were prepared by the routine method (see Chapter 2). Apart from the comparison of the IRSL from loess at Luochuan section, the IRSL signal was measured in the manual Daybreak system. Unless stated, the measurement is the signal obtained for 0.1 second IR exposure.

Loess contains different luminescent minerals, quartz, zircon and feldspar (Pye 1988), but the IRSL signal is dominated by that from feldspar minerals.

The lack of coarse grains (100-300 μm) in loess samples (Liu, et al. 1985) prevents the use of separated K-feldspar fractions, though there has been a report of TL dating of loess by using coarse grains of quartz (Lu et al. 1987). Instead, coarse grains (100-300 μm) of K-feldspar separated by heavy liquid from sand deposits in Denmark were used for comparison. The separated fraction was etched in 10% HF for 40 minutes.

3.3.3. Global view of thermal stability

The stability of the IRSL signal was evaluated by storing the samples in ovens at different temperatures for a period of time. The temperatures were chosen as 75°C, 140°C, 230°C and 260°C because these had been used for preheating either to remove an unstable TL component or for removing anomalous fading in TL signals (Wintle 1985a, Berger 1988b, Zöller and Wagner, 1989).

Three groups of discs were used for the storage experiments at 75°C and 140°C. They were natural, natural plus a ß dose, and ß dose added to natural samples following SOL2 bleaching. These groups represent those used in the dating process as natural, N+ß and regenerated signals. Only natural samples were examined after 230°C and 260°C storages. For these groups, each sample was represented by four discs. The bleaching time in the SOL2 was 5 hours. The ß dose given was 74 Gy.

For the natural samples, the discs were normalized by the natural count as measured by a 0.1 second exposure before any treatment. For the ß dosed discs used in the 75°C and 140°C storages, normalization was made using the IRSL measured immediately before being placed in the oven, and not more than 12 hours after irradiation.

1. *140°C*

140°C is the temperature routinely used for preheating of loess samples in the Aberystwyth laboratory. In this study, sample discs of the three groups were stored on the same aluminium plate in the oven from 0.5 hour up to 270 hours (more than two weeks). The isothermal decay results are given in Fig.3.3.2. For all of the natural samples the reduction of the IRSL signal is similar (Fig. 3.3.2a), except for the sample from Alaska, for which the signal is reduced more rapidly.

The natural plus ß (74Gy) and regenerated (74Gy) IRSL show a different response from the natural. This is because of an unstable signal produced by the irradiation (Fig.3.3.2b, Fig.3.3.2c).

2. *75°C*

For all samples, the natural discs gave no reduction of the IRSL signal after 7 days at 75°C, but had shown a significant loss after 30 days storage (Fig.3.3.3a).

The laboratory irradiated discs showed an even greater reduction of the signal with significant loss after only 64 hours (Fig.3.3.3b and 3.3.3c) showing the presence of an unstable component.

3. *230°C and 260°C*

The results of 230°C storage show a trend consistent with those obtained at 140°C, with a more rapid loss for the loess from Alaska (Fig.3.3.4a). In this figure the results are

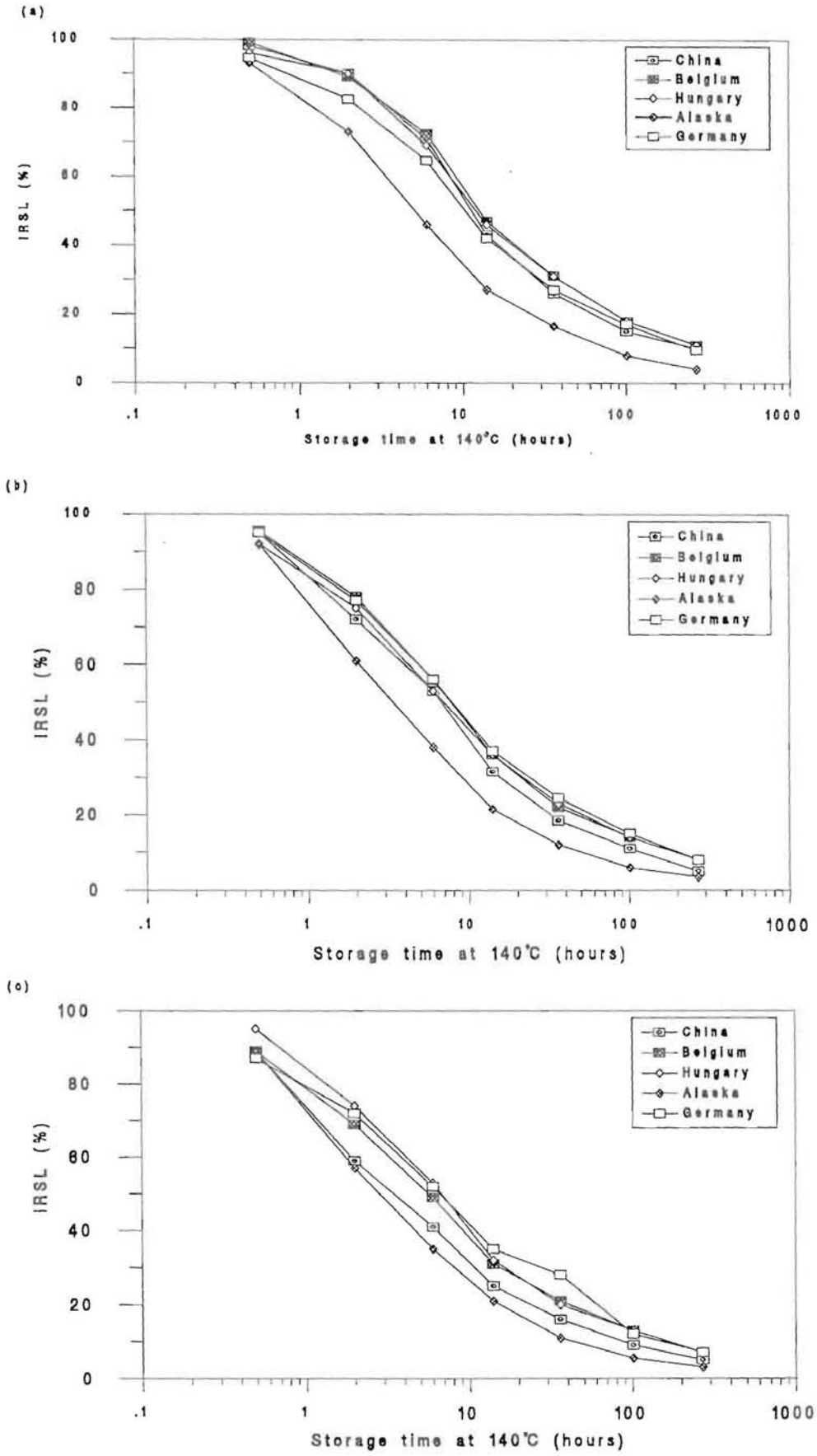


Figure 3.3.2. Results for loess samples stored at 140°C. (a) natural; (b) natural plus ß dose (74Gy); (c) ß dose (74 Gy) regenerated signal.

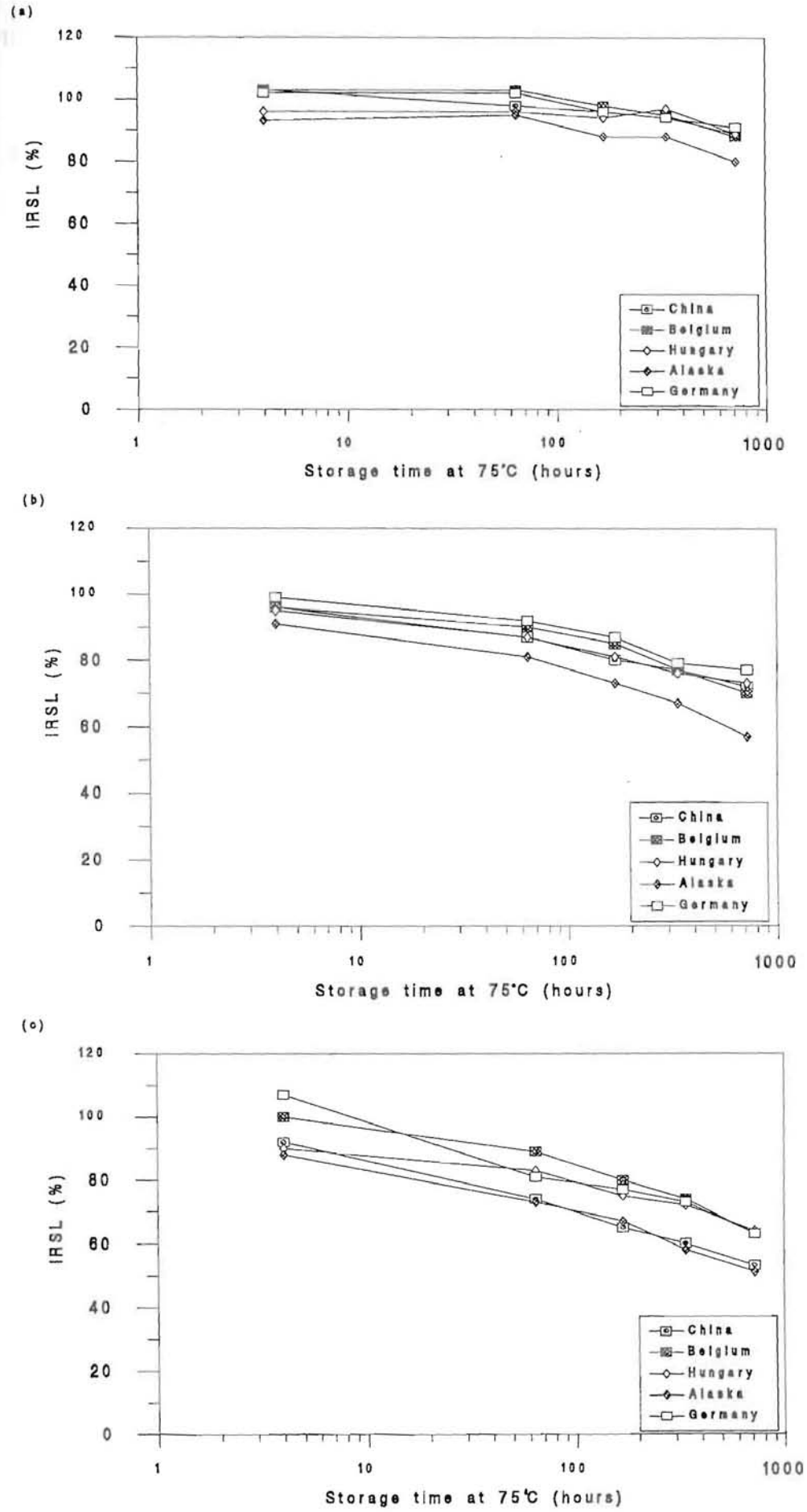


Figure 3.3.3. Results for loess samples stored at 75°C. (a) natural (b) natural plus ß dose (74 Gy) (c) ß dose (74 Gy) regenerated signal.

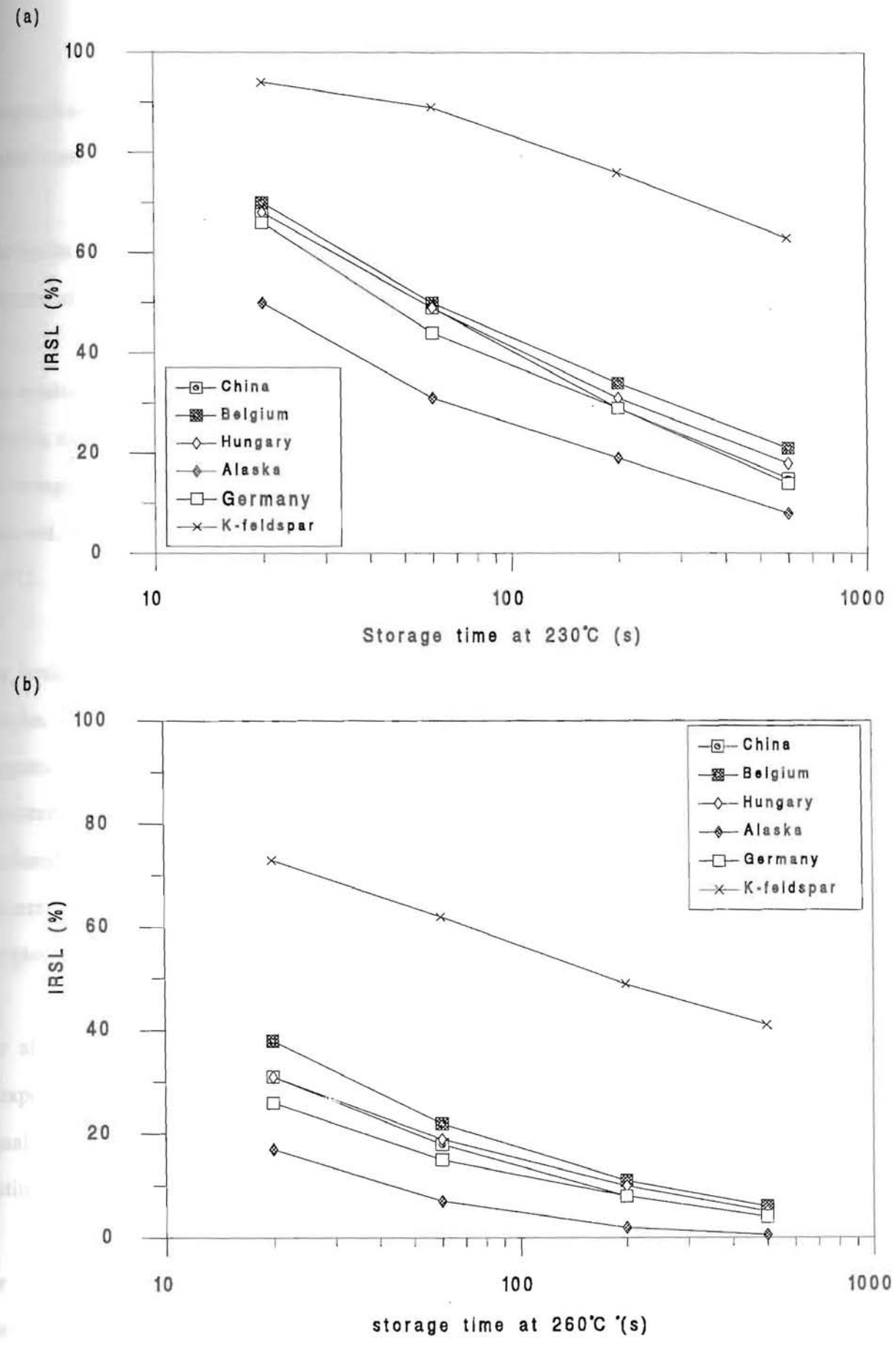


Figure 3.3.4. Loess and K-feldspar (Z29) natural discs stored at (a) 230°C and (b) 260°C.

compared with a separated K-feldspar. The latter exhibits slower thermal decay, reflecting a higher thermal stability (also see section 3.4).

The results from 260°C storage are almost identical to the results at 230°C (Fig.3.3.4b). The response of 4 out 5 of the loess samples were the same within experimental error.

The results of the quick heating experiments show a similar response for all the natural samples, except that from Alaska (Fig.3.3.5). This follows a similar trend to that found for the storage experiments. Once the sample has been heated to 350°C, no IRSL signal was observed. This suggests that the IRSL signal may be correlated with the TL traps below 350°C.

The isothermal decays at different temperatures and the quick heating curves for the loess samples are remarkably similar in spite of their different geographical locations. This suggests a similar thermal stability for these samples. The Alaskan sample shows consistently less thermal stability in the experiments on both natural and laboratory beta irradiated samples. One consideration, as far as the poorer stability of the natural IRSL is concerned, is that the ambient temperature in Alaska was relatively lower than for the other samples because of the present day existence of permafrost.

For all samples the isothermal decay curve is not a single exponential. This is not unexpected because single exponential decay was not shown by the decay of the IRSL signal of single pure feldspar (see section 3.2). Thus it is not possible to evaluate the lifetime of the IRSL signal using the isothermal decay method.

For 140°C and 75°C storages, it is shown that an unstable component exists in the laboratory irradiated samples but not in the natural sample. Because the samples are older than 10 ka, this unstable component could be explained as an IRSL signal from intermediate

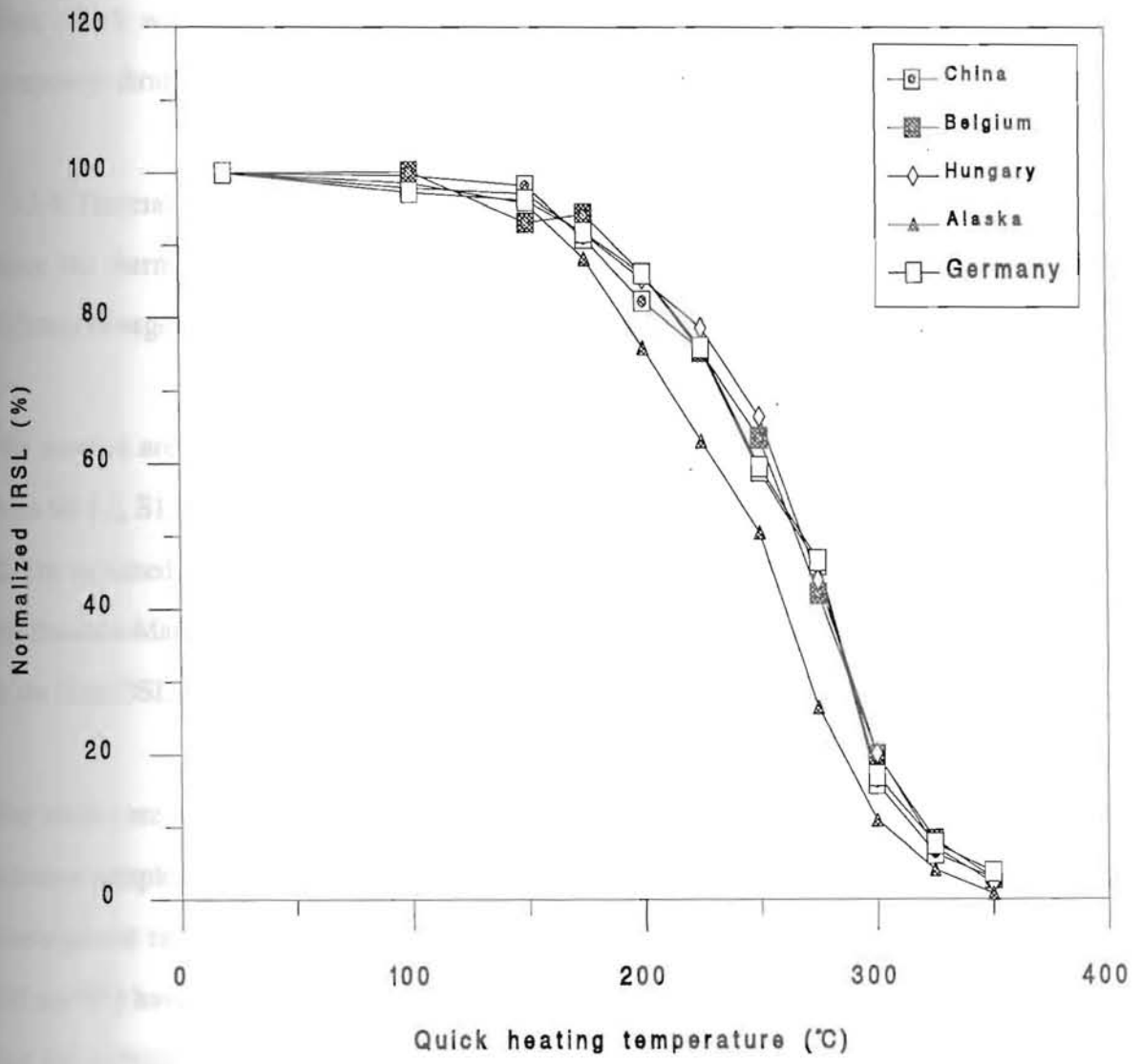


Figure 3.3.5. Quick heating of IRSL signal from natural samples.

traps, which will be emptied in a time less than 10 ka at ambient temperature. This component should be removed by preheating before ED determination.

3.3.4. Thermal stability for the Luochuan loess section

Since the thermal stability revealed in isothermal decay curves has a similar trend for different storage temperatures, only the effect of storage at 220°C was examined.

The samples are from the Potou of the Luochuan section, Shaanxi, China and were taken from the L1, S1, L3, S3, L5, S5 and L8 loess/palaeosol sequence (Liu et al. 1985, Appendix I). The expected ages are from 70 ka for youngest L1 to 730 ka for sample L8 which sits on the Brunhes-Matuyama boundary (Kukla 1987). The storage measurements were conducted in the Risø OSL-TL system.

The results are similar for all the samples (Fig. 3.3.6). There is no significant difference between samples of different age. This suggests that the stability of IRSL is an inherent characteristic rather than dominated by sample age. The signals from the palaeosol samples (S3 and S5) have a similar stability to those from the loess (L1, L3, L5 and L8), suggesting that the stability of the signal is not related to the weathering process which might have affected the mineral composition of the palaeosol samples.

3.3.5. Anomalous fading of IRSL in loess

Anomalous fading was observed experimentally by storing sample discs at room temperature after laboratory irradiation. All were normalized by the IRSL of a short shine measurement immediately before storage. The irradiation dose was 74 Gy, administered not more than 12 hours before normalization. For the five samples used in the stability study, six categories of discs were stored at room temperature (20°C±5°C). These categories represent treatment received by discs in the laboratory in routine TL and IRSL dating procedures (Table 3.3.1). They can be grouped into those with (group A), and those without

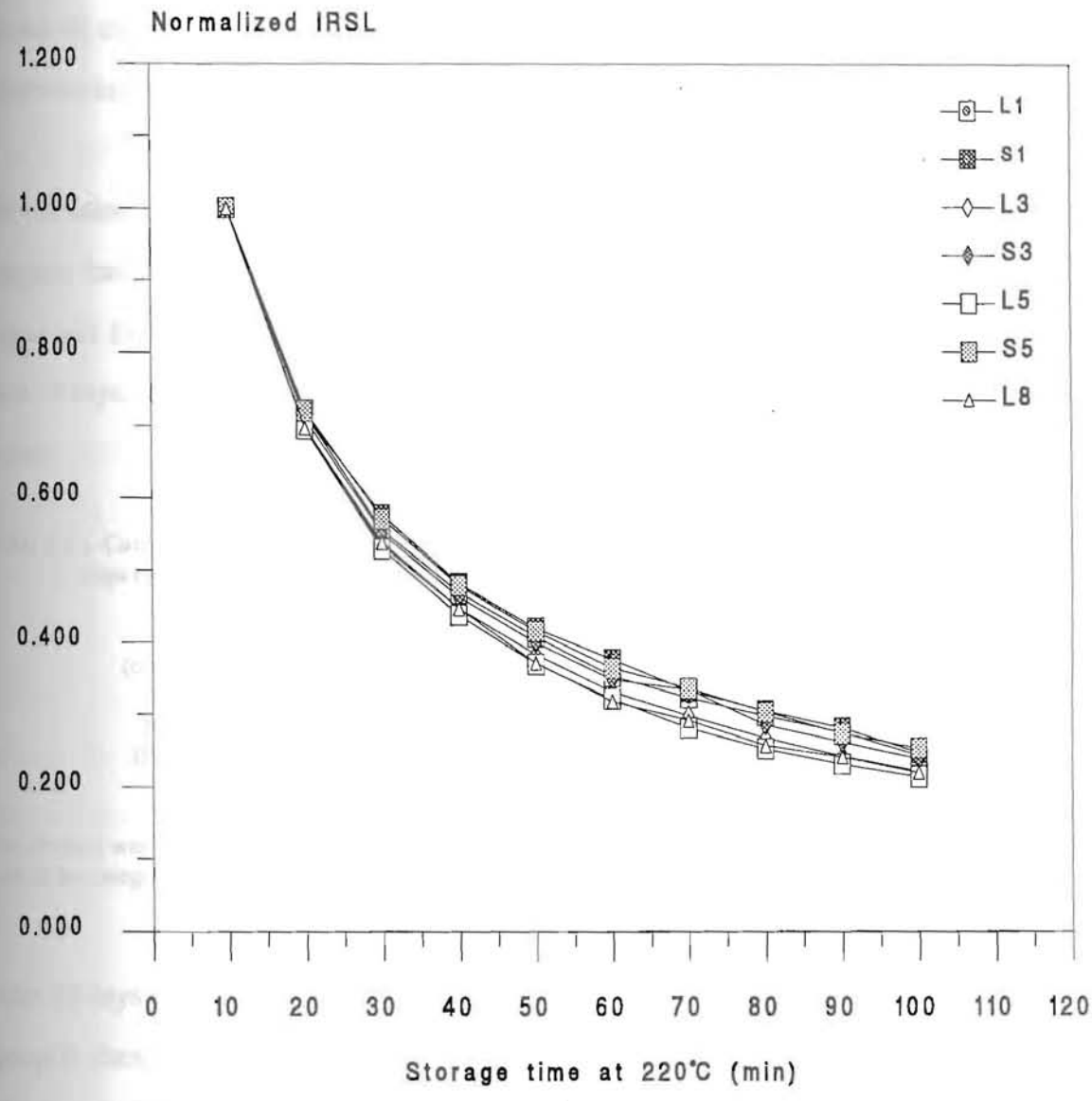


Figure 3.3.6. Effect of 220°C storage for natural loess and palaeosol samples from Luochuan section. The loess (palaeosol) samples are distinguished by L an S prefixes.

(group B) charges trapped in shallow traps produced by laboratory irradiation as achieved by preheating for 16 hours at 140°C as reflected by the TL signal below 200°C.

No reduction of the IRSL signal was found for either of the groups after 15 days. This suggests that no phenomenon equivalent to the decay of the lower temperature (<150°C) TL signal was found. It also indicates that the lifetime of the potential IRSL signal is greater than 15 days. In another words, no extremely unstable signal is observed for the loess IRSL signal.

Table 3.3.1. Categories of disc stored at room temperature with the % IRSL after storage for 50 and 620 days compared with the initial value of 100% for sample GHC27 from Alaska U.S.A.

Group A (contains unstable component)	Group B (No unstable component)
Natural + ß (91±2;80±4)	Natural (101±6; 95±5)
Bleached + ß (80±4;67±2)	Natural + preheat (108±5;104±5)
	Natural + ß + preheat (99±9;92±4)
	Bleached + ß + preheat (99±1;96±8)

note: Preheat was 140°C for 16 hours. The error limits quoted represent the standard deviation of the 4 discs in each of the categories.

After 50 days (1200 hours) at room temperature, no detectable reduction was observed for group B discs. However, both categories in group A showed a consistent loss of the IRSL signal which indicates that part of the laboratory produced IRSL signal is derived from shallow traps. This was very clearly demonstrated after 620 days storage: there is no detectable reduction for discs in group B, but significant loss of IRSL signal for discs in group A. This was found in all five samples not just GHC 27. This loss of IRSL signal in laboratory irradiated discs might relate to the unstable component observed for the 140°C and 75°C storage. In the 140°C and 75°C storage, the decay of IRSL signal for the laboratory irradiated discs was quicker than that for the natural disc.

No loss of signal in group B suggests that this short term unstable component could be removed by preheating samples and shows that preheating is an essential and important step

in dating with IR stimulation. This has also been demonstrated by comparing the ED obtained, using the additive dose method, after preheating (140°C for 16 hours) with that without preheating for loess sample QTL89D from Belgium. The ED was 84 ± 5 Gy for the former and 36± 2 Gy for the latter.

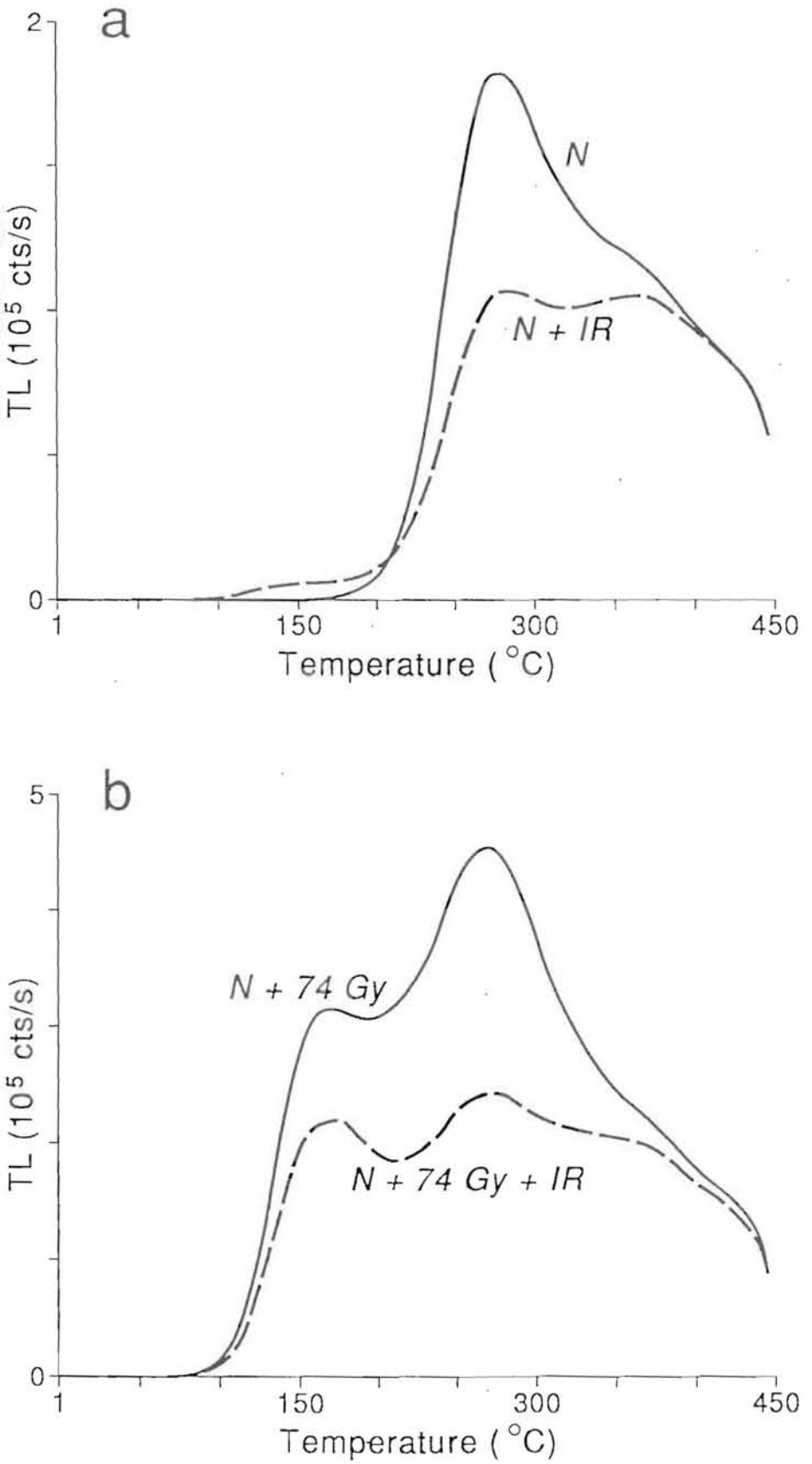
3.3.6. TL and IRSL

To study the effect of IR exposure on the TL signal, TL glow curves were obtained for samples which had been exposed to 1000 seconds of IR radiation and some which had not (Fig.3.3.7). The sample used was QTL89D from Rocourt, Belgium. The TL curves show that at least two TL peaks have been affected by the IR exposure.

As shown in the quick heating curves (Fig. 3.3.5), most of the IRSL signal is erased after heating to 325°C. This suggests that the IRSL is associated with the TL traps at temperatures lower than 325°C. The traps related to the high temperature (above 350°C) TL signal make an insignificant contribution to the IRSL signal.

3.3.7. Relevance to long term dating limit

A long term dating limitation for loess samples of about 100 ka has been widely reported in TL studies (Debenham 1985, Wintle 1990), with similar problems being encountered around the world (China, eastern Europe and north America). It has been suggested that this is due to lack of long term stability of the luminescence signals. If this is due to loss of luminescence centres, IRSL dating is likely to have the same limitation since IRSL uses the same luminescence centres as TL (as demonstrated by emission spectra studies). On the other hand, if the limitation is dominated by the fading of trapped electrons, the IRSL may be expected to have a lower thermal stability than the TL signal, since most of the IRSL is correlated to charge traps which empty below 325°C. Also, since the IRSL signal has a similar stability for samples from different parts of the world, the age limitation would not be expected to differ for different loess around the world.



3.3.7. TL glowcurves using Corning 5-60+HA3 (5°C/s) for loess from Rocourt, Belgium showing the effect of 1000 second IR exposure. (a) natural; (b) natural plus 74 Gy ß dose.

In TL dating, removal of long term unstable signal by short, strong thermal washing at high temperatures or longer preheating at lower temperatures was suggested by Zöller and Wagner (1989). This should also be suitable for the IRSL signal. However, because most of the IRSL signal is unstable in the long term, there is always a small signal left after a long preheat at a lower temperature and this may still be important compared with any long term stable signal that might be present. Unfortunately, when preheating at a higher temperature was employed, the signal left was at a comparable level with the noise level from PM tube background. Hence it is unlikely that isolation of a long term stable signal could be achieved by different preheating procedures.

3.3.8. Summary

Storage experiments at elevated temperatures showed the IRSL of all the natural loess samples to have similar thermal stabilities in spite of their different geographical locations. Similar thermal stability was found for the IRSL signal from loess of different ages from the same section.

Short term fading of the unstable signal for the laboratory irradiated samples was observed in isothermal decay experiments and for room temperature storage. Preheating is necessary to remove this component before dating measurements are made.

No anomalous fading or extremely unstable signal were found for any loess samples used in this study after a preheat of 140°C for 16 hours had been applied.

It is unlikely that further preheating could be used to isolate an IRSL signal with greater long term stability.

3.4 MIXED MINERALS (K-FELDSPAR)

3.4.1. Introduction

The K-feldspar fraction separated by heavy liquids has been used for TL dating of sediments since it has a bright TL signal which is easily bleached by sunlight exposure (Mejdahl 1985a). It has been found to have a higher saturation dose than quartz (Mejdahl 1985b). In this section, the stability of the IRSL signal from K-feldspar mineral fractions separated by heavy liquids will be discussed. Comparisons with pure minerals and loess will be made.

3.4.2. Thermal stability of different feldspar fractions

The thermal stability was examined in a similar way to that used for pure minerals and loess samples (sections 3.2 and 3.3). The sample used in this study was a coarse grain (100-200 μm) beach sand (GDNZ 17; age 6-12.5 ka) from New Zealand. Different fractions of the same grain size were inspected, namely bulk sand, etched K-feldspar, unetched K-feldspar, Na-feldspar (grains of density between 2.58-2.62 g/cm^3) and fine grain (4-11 μm) K-feldspar (obtained by grinding the separated K-feldspar). The etched K-feldspar was first separated with heavy liquids (density less than 2.58 g/cm^3) and than etched for 10 minutes with 40% HF acid. A loess (88-12, Appendix I) sample from Luochuan was used for comparison. This is represented by fine grains (4-11μm) and medium grains (50-80 μm). All the data curves are the average of four discs.

All sample discs were bleached for 5 hours with the SOL2 to erase the previous IRSL signal. After 24 hours delay, the discs were given a 100 Gy ß dose. All of the discs were preheated for 1 hour at 140°C. An IRSL signal from a short IR exposure (0.1 second) at room temperature was used for normalization. The IRSL after different storage times at 140°C was compared with this measurement (Fig. 3.4.1).

Similar to the results of 230°C and 260°C storage (mentioned in section 3.3.3), a significant

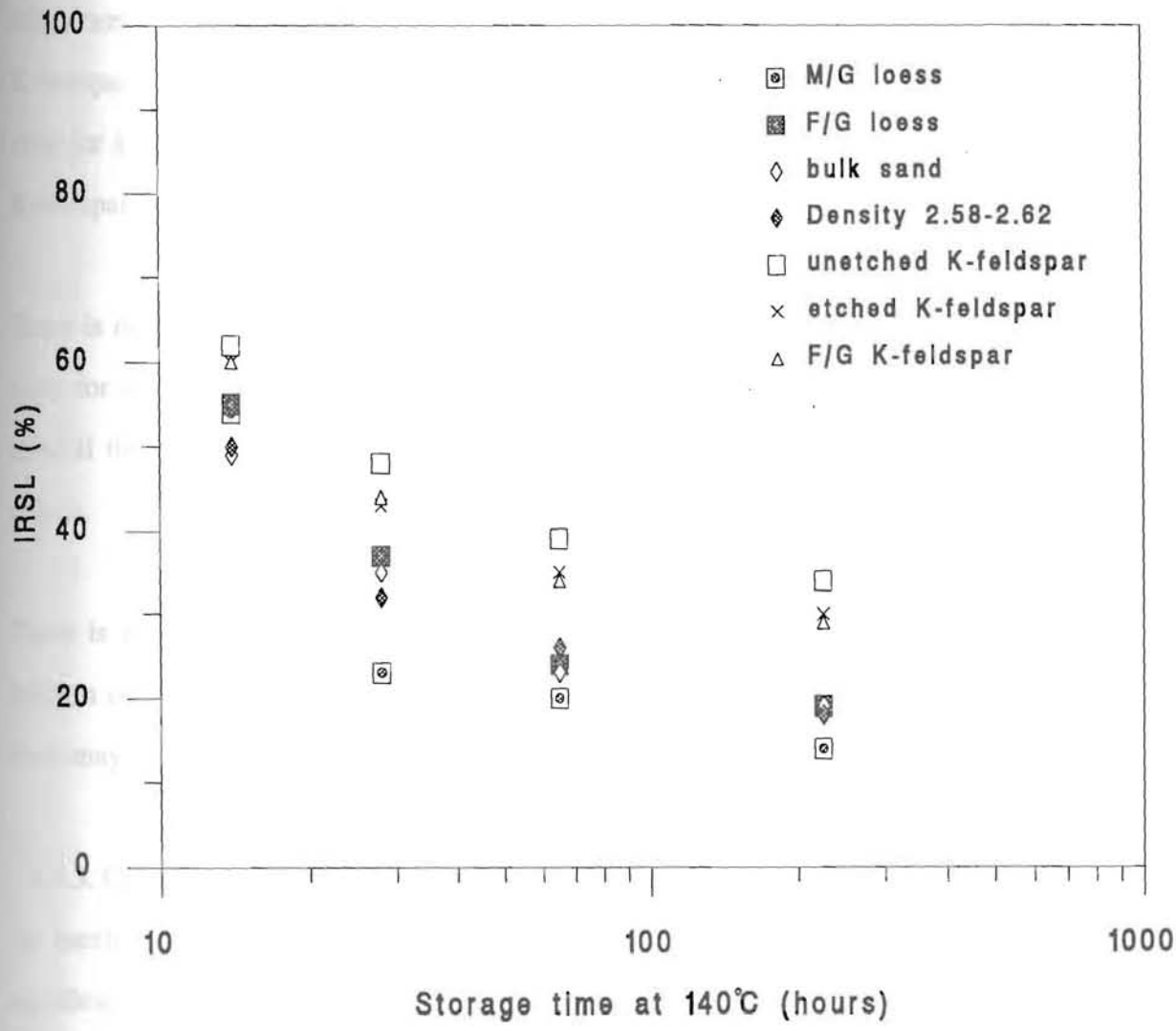


Figure 3.4.1. Comparison of the thermal stability of the IRSL signal from different mineral components of New Zealand sand and Chinese loess with different grain size fractions (as detailed in text). Samples were given five hours bleach with SOL2 solar simulator followed by a 100 Gy ß dose.

difference in the isothermal decay of the IRSL signal was found between the New Zealand K-feldspar and the loess after storing at 140°C (Fig. 3.4.1). A systematic difference was clear for all storage times from 15 to 220 hours which indicates the greater stability of the K-feldspar samples.

There is no measurable difference in the isothermal decay of the IRSL for different grain sizes for either the loess or the K-feldspar fraction (Fig.3.4.1). Hence for a particular mineral the stability of the IRSL signal is not related to the grain size or to any surface effects.

There is no difference between the unseparated sand grains for the bulk sand and the fraction consisting of Na-feldspar (density 2.58-2.62 g/cm^3). It is therefore concluded that there may be a significant contribution from Na-feldspars to the signal of the bulk sand.

3.4.3. Comparison with pure minerals

As mentioned previously (section 3.2.5), the IRSL signal from alkali (K) feldspars is significantly more stable than that from plagioclase. Comparison with pure minerals was made by looking at the isothermal decay of IRSL signal under the same storage and measurement conditions. Samples used for comparisons were K4 (microcline), Pa (albite), loess (88-12 from Luochuan) and AF-4 (K-feldspar separate from South Africa, also see chapter 7), which were mentioned in section 3.2.4. Because the stability is mirrored by the isothermal decay behaviour, there is a similar trend for different storage temperatures; hence a comparison was made only for storage at 220°C.

The IRSL of the K-feldspar from AF-4 has a comparable stability to the pure microcline feldspar (K4) and it is more stable than the plagioclase feldspar (Pa) (Fig. 3.4.2). This suggests that minerals having a more stable signal can be selected by heavy liquid separation from bulk sand grains. On the other hand, the IRSL stability of plagioclase

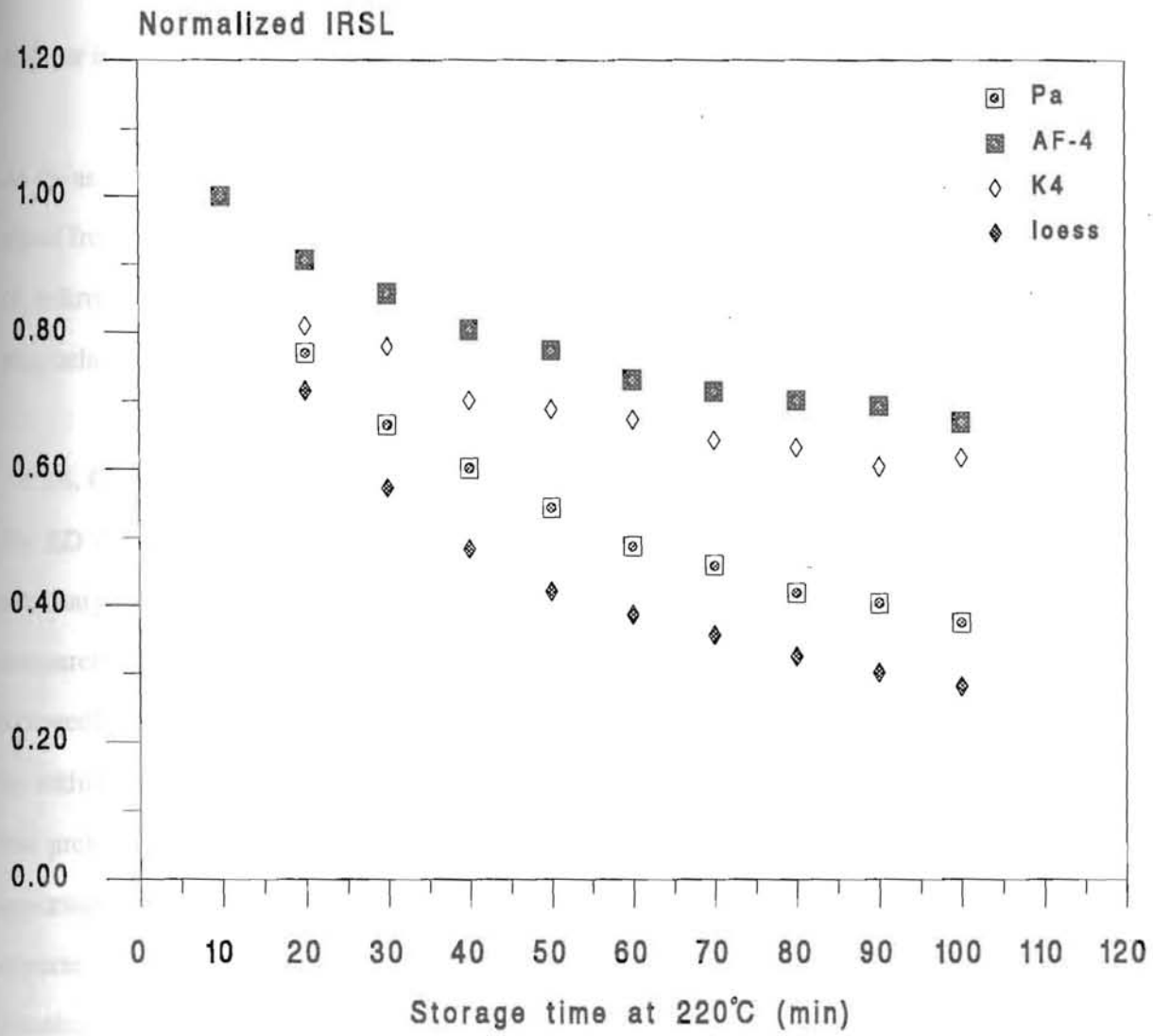


Figure 3.4.2. Comparison of the thermal stability of the IRSL signal from a K-feldspar extract and loess and the pure mineral samples (Pa and K4) used in the previous section (3.3.).

feldspar is similar to that of loess and is relatively unstable.

As far as the stability is concerned, one can conclude from these experiments that the IRSL signal from the K-feldspar separate is dominated by a signal having a stability similar to that of microcline, whereas the bulk sand and Na-feldspar separate are dominated by the plagioclase signal.

3.4.4. Comparison of K-feldspar from different regions

For ED determination by the additive dose method for single aliquots (Duller 1991), it is necessary to correct for signal loss due to preheating and short IR exposure during measurement (normally 0.5 sec). This is achieved by measuring the signal reduction repeatedly after each preheat and short IR exposure for natural sample discs which received no additional dose. This procedure is similar to that of observing the isothermal decay at that preheat temperature and hence the measurements for the preheat correction provide an opportunity to examine the thermal stability of individual samples. Under the same experimental conditions, the more stable the signal is, the higher the proportion of the remaining signal.

It appears that the stability of the IRSL signal from K-feldspar fractions is comparable for the samples investigated (Fig. 3.4.3), except for the sample from South Africa. The responses shown in figure 3.4.3. represent several samples from each region. For each region the samples have an identical thermal stability. The differences between these K-feldspar samples is smaller than the difference between K-feldspar and loess and plagioclase. Comparison of these results with those for the pure feldspars (see section 3.2.5) suggests that the sediment K-feldspar fractions are dominated by a K-feldspar with relatively high thermal stability, such as microcline. The samples from South Africa show consistently higher thermal stability than the rest of the samples and perhaps relates to the local geology.

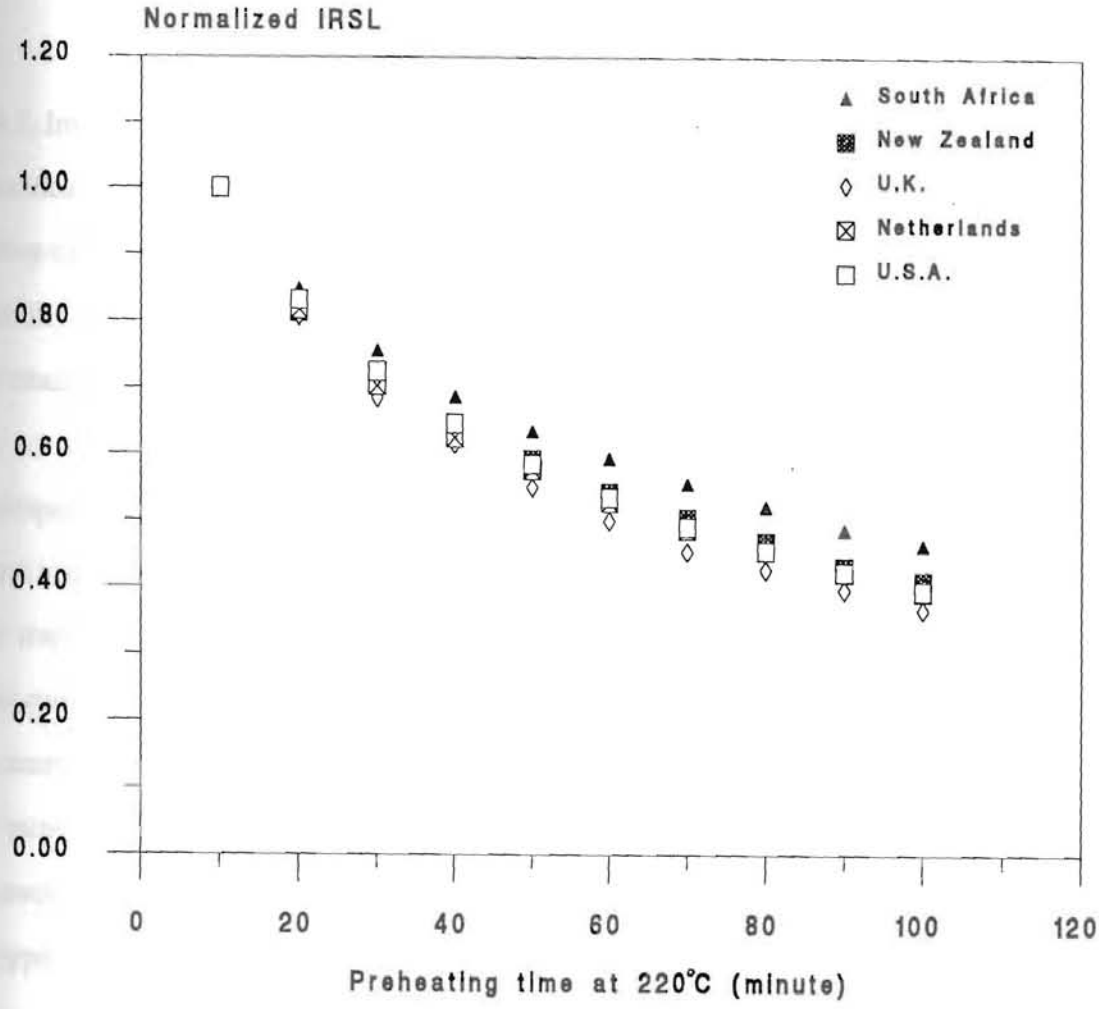


Figure 3.4.3. Comparison of the thermal stability of the IRSL signal from K-feldspar samples. Data were provided by G.A.T. Duller except for the sample from South Africa. Samples are New Zealand (D16GA, McLeavy Road, Otaki); U.S.A.(DL19, Mohave Desert); Netherlands (Lutte1, Lutterzand); U.K. (AOP, Inverness) and South Africa (AF-4, Natal).

3.4.5. Implications for the upper limit of dating

Theoretically, the upper limit of IRSL dating is dependent upon the saturation dose and the stability of the signal. The former is a characteristic of trap filling. The latter is relevant to the ability of keeping trapped charges in the traps. For a particular sample the limiting factor may relate to either feature, or both.

K-feldspar samples from sediments have complicated mineral compositions. Hence the natural signal contains contributions from different traps in different minerals. For each of these traps, the saturation level and thermal stability is likely to be different and thus the signal cannot be treated as though it was derived from a single type of trap. As far as dating is concerned, this may not be a problem for young samples because none of the traps are full and most are stable enough to avoid charges escaping during the period of interest. However, for an old sample (say 100 ka), one trap type could become full, while another trap type could have experienced some loss of charges. This results in a complex response.

Because trap filling and fading are related to the dose rate, ambient temperature and age of the sample, it is not possible to define a common upper age limit or make a general statement about the dominant behaviour for any particular sediment. For very old samples both types of behaviour could reach an equilibrium state and result in an equilibrium age.

Modelling of long term fading and saturation behaviour have been proposed and age corrections for long term fading were suggested first for the TL signal of K-feldspar (Mejdahl 1988b) and later for the OSL signal of Chinese loess samples (Xie and Aitken 1991). However, little support is given to those approaches by this study because the IRSL is from many minerals, i.e. different types of traps for both K-feldspar separates and for loess. It is unlikely that the complicated fading and saturation responses of those traps could be treated simply as that of a single type of trap.

The higher thermal stability shown by the K-feldspar fractions suggests that they might have a higher age limit than the fine grain (4-11μm) polymineral samples, since both dosimeters have a similar dose rate in most sedimentary environments.

3.4.6. Anomalous fading

Anomalous fading was found for several feldspar minerals (section 3.2.4). Thus it is possible that anomalous fading will be found in the K-feldspar fractions of sediments and hence it is worthwhile checking each sample, as suggested by Spooner (1992). If present, anomalous fading would be observed in a short time period after irradiation. From the dating point of view, measurements made a long time after irradiation would effectively minimize the observation of the fading, and reduce the error in the ED determination.

As observed for the pure minerals (section 3.2.4), preheating can accelerate the fading though it might not results in it reaching a stable level. Hence preheating might minimize the fading effect in ED determination. This behaviour is currently being studied elsewhere (Spooner, pers. comm.).

3.4.7. Summary

The IRSL signal from K-feldspar, separated from sediments by heavy liquids, showed higher thermal stability than that for fine grain loess. This is not due to a grain size or surface effect but is related to a mineralogical difference. The signal from K-feldspar separate has a similar stability to that of microcline and is higher than that of plagioclase feldspars.

Very small differences in thermal stability were found between K-feldspar samples from different regions of the world.

From the thermal stability aspect, K-feldspar may be suitable for the dating of old samples. However, there are many complex factors which influence the limit for a given sample. These include the mineral composition, dose rate differences and the interplay of trap filling and fading. No common upper age limit can be given.

3.5 REMOVAL OF UNSTABLE SIGNALS

3.5.1. Introduction

If there is a thermally unstable component in irradiated samples which is not removed by an appropriate thermal pre-treatment, the ED, and hence the age of sample, will be underestimated significantly (Li and Wintle, 1992). In this section, I will discuss the removal of thermally unstable signals. Unless otherwise stated, experiments were made by observing the OSL signal using the Ar ion laser in Oxford. Similar results were found for the IRSL signals.

3.5.2. Existence of thermally unstable signal

It is well known that there is a thermally unstable TL signal after laboratory irradiation. This is observed experimentally as the lower temperature TL peaks, which do not exist in natural samples. However, it is not so easy to observe the existence of similar unstable components in OSL and IRSL signals because the signal from traps of different stabilities are observed simultaneously. However, if thermally unstable signals exist, the following phenomena should be observed: (a) the ED obtained with these signals will be significantly lower, (b) the unstable signal may be associated with the lower TL peaks and hence OSL or IRSL signals of laboratory irradiated samples will be reduced more rapidly as the lower temperature TL peaks are removed by heat treatment (c) the low temperature TL peaks may be reduced after bleaching with the stimulating light source. These points are now examined in turn.

a. ED determination with and without a preheat

Preheating is one way to remove charges from the shallow traps. EDs were determined using identical experimental conditions apart from the preheating procedure. For the IRSL signal, the fine grain sample QTL89D gave an ED of 84±5 Gy with a preheat at 140°C for 16 hours and 36±2 Gy without. For a coarse grain (125-150μm) K-feldspar sample, STP-13, the ED was 7.1±0.1 Gy with a preheat at 220°C for 10 minutes and 2.6±0.1 Gy without.

b. Test with quick heating.

A quick heating experiment was applied to two discs, one natural and one irradiated (70Gy). The OSL was measured after the disc had been cooled to 17°C. The sample used was Z29 from Denmark (Li and Aitken 1989). Figure 3.5.1 shows the different behaviour of the two discs, which suggests that there is an OSL signal associated with the shallow traps.

c. Bleaching with stimulated light

Fig 3.3.7 shows the reduction of the lower temperature TL peak around 150°C after a long IR exposure is applied to an irradiated fine grain loess disc. A similar reduction was found after bleaching with 514.5 nm Ar laser light. This lower temperature TL signal is not stable at ambient temperature, as indicated by the fact that it was not found in the natural signal.

The observation of a fast fading signal in the museum feldspar samples supports the existence of short term unstable signals related to the shallow traps. This signal needs to be removed before ED determination.

3.5.3. Removal of unstable signal without heating

Because the unstable signal is derived from relatively shallow traps, it might be expected that the charges in these traps can be removed by using longer wavelengths, which have a lower stimulation energy. The longer wavelengths should not have enough energy to excite the charges in stable traps, which have a greater trap depth. However, because the IR is able

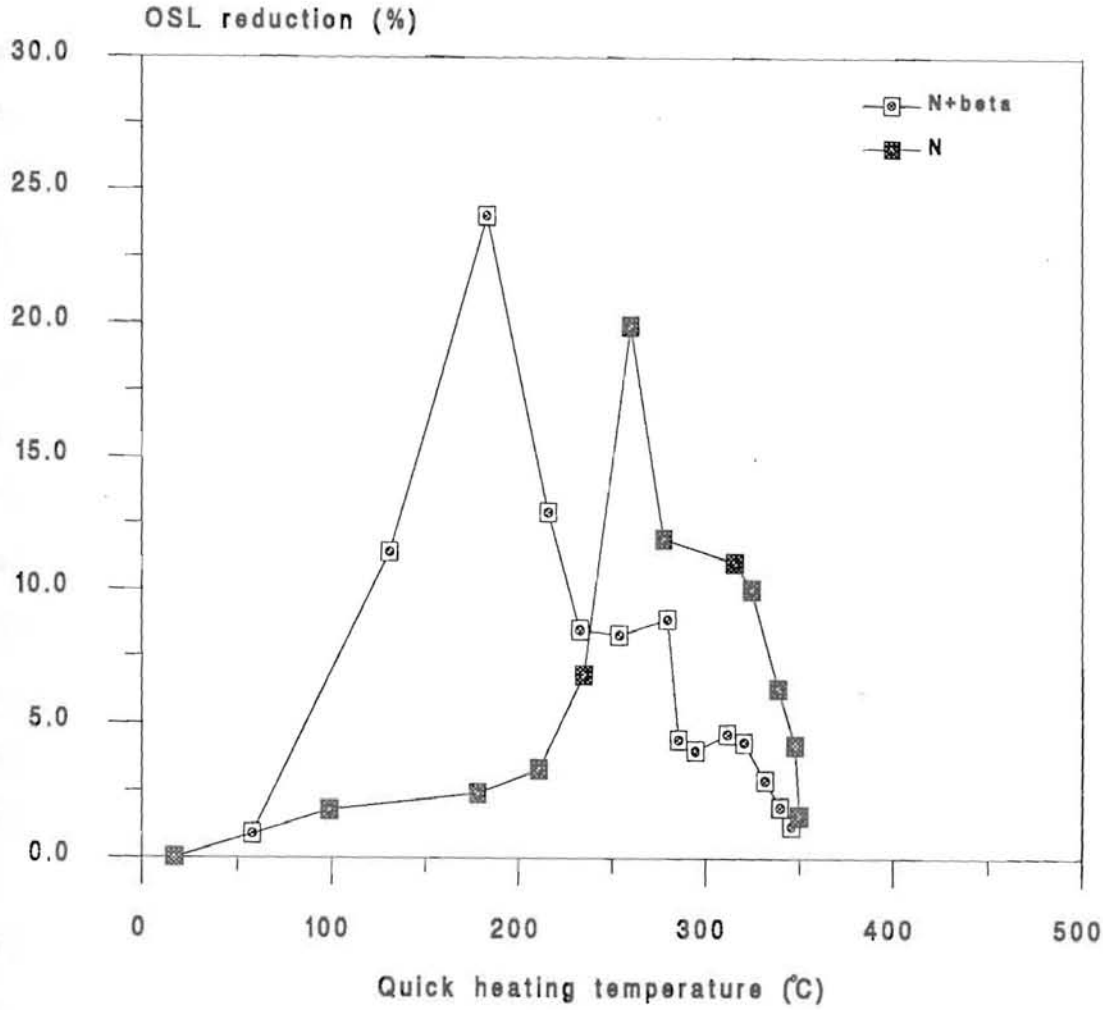


Figure 3.5.1. Quick heating of OSL signal from natural and natural plus ß dose (200 Gy) discs of sample Z29.

to stimulate charges from deep traps via intermediate traps to the conduction band (Hütt et al. 1988; Godfrey-Smith et al. 1988), thermally stable traps cannot be selected by varying the energy of the stimulating photons. Therefore it is impossible to remove the unstable signal by means of light.

3.5.4. Previous studies in preheating.

Preheating is often used for removal of the unstable signals in TL dating (Mejdahl and Winter-Nielsen 1982). It has also been used for the same purpose in TL dating of quartz and fine grain loess samples (Valladas and Valladas, 1982; Wintle 1985a; Zoller and Wagner 1989). In TL, the stability is indicated by the TL peak temperature. However, the signal used in optical dating comes from a range of traps and thus can contain both stable and unstable components. Hence the preheating procedure for optical dating may not be the same as that used in TL dating.

Preheating procedures have been established in optical dating of quartz (Huntley et al. 1985; Rhodes 1988). Since the OSL signal in quartz has been correlated to the 325°C TL peak (Smith et al. 1986), a preheating procedure was developed based on the TL behaviour of this peak (Rhodes, 1988). However, this procedure would not be suitable for other minerals. For K-feldspar, the OSL signal is from at least two types of trap, as demonstrated by the loss of signal from the two TL peaks (Li and Aitken 1989; Duller 1991 for IRSL).

3.5.5. Justification and design of a preheat

The aim of preheating is to remove all charges from the unstable shallow traps. To test a particular preheat procedure, the signals of natural and irradiated samples were looked at in the following ways-(a) thermal stability (b) signal response to heating (c) TL glow curve shape.

If the OSL or IRSL obeys a first order stimulation process, the number of trapped charges N

at temperature T is given by the equation

$$N=N_0 e^{(-t/\tau)} \quad \text{and} \quad \tau=S^{-1}e^{(-E/kT)}$$

where N_0 is the number of trapped charges at t=0, τ is the lifetime of the decay (s), S is a frequency constant (s^{-1}), k is Boltzmann's constant, T is absolute temperature (K) and E is the energy depth of the trap (ev).

The design of a preheat procedure involves the choice of preheating time and temperature. A very high preheating temperature allows the use of an extremely short preheating time but there is a risk that complete thermal equilibrium may not be reached and sample oxidization may occur. If the preheating temperature is very low (say 50°C), it may take months or years to remove the unstable signal, making such a procedure impossible in practice. For a particular preheating treatment to be appropriate, the thermally unstable signal should have been removed and enough stable signal must remain for measurement.

3.5.6. Preheating for K-feldspar samples

The samples used in the K-feldspar study were coarse grain (100-300μm) K-feldspar grains separated from sediments from Scandinavia. The Oxford laboratory numbers are Z10, Z29 and Z30 (Li and Aitken 1989).

In practice, two different kinds of preheating are sought. One involves a long time (a few hours) at a lower temperature, the other a short time (a few minutes) at a higher temperature. The former can be performed in an oven; the latter can be done on a TL heating plate and is useful if the same equipment is to be used for both TL and OSL measurements as the discs do not have to be moved from the hot plate between the measurements.

1. Prolonged preheating

There is a TL maximum around 160°C in the TL glow curve after laboratory irradiation (Fig. 3.5.2 curve N+200 Gy). This TL maximum is not found for the natural samples and is therefore considered to be an unstable signal. Hence 160°C was chosen as the temperature to work with.

For sample Z29, two discs, one natural and one with an additional irradiation (200 Gy) were placed in a 160 °C oven for a total period of more than 200 hours. The discs were taken out periodically for OSL measurement of 0.1 second of laser light. Fig 3.5.3. is the plot of the OSL ratio between these two discs and it shows that a plateau value was reached after only half an hour at 160°C. Use of 0.1 second exposure allows the discs to be used many times; hence no normalization is required to improve the precision. This is analogous to obtaining a plot of ED versus preheating time for a group of discs but this approach requires normalization and curve fitting which reduce the precision.

Fig 3.5.4. shows the thermal decay of the OSL that occurs during the preheating for another sample, Z10. The discs were normalized to the signal obtained for storing for 5 hours at 160°C. This shows that the thermal decay rates are the same once the two aliquots, natural and natural plus irradiation (70Gy), have been heated five hours or more, i.e. the thermally unstable signal in the dosed sample can be removed by 5 hours preheating at 160°C.

To demonstrate further the similar thermal response of the natural and irradiated samples after preheating, a quick heating experiment was performed (Fig.3.5.5.). After only half an hour at 160°C the responses were similar for samples Z10 and Z29. This implies that the OSL signal is from a similar temperature range for both discs after this preheating treatment. This was not the case for the OSL from discs which were not preheated (Fig.3.5.1).

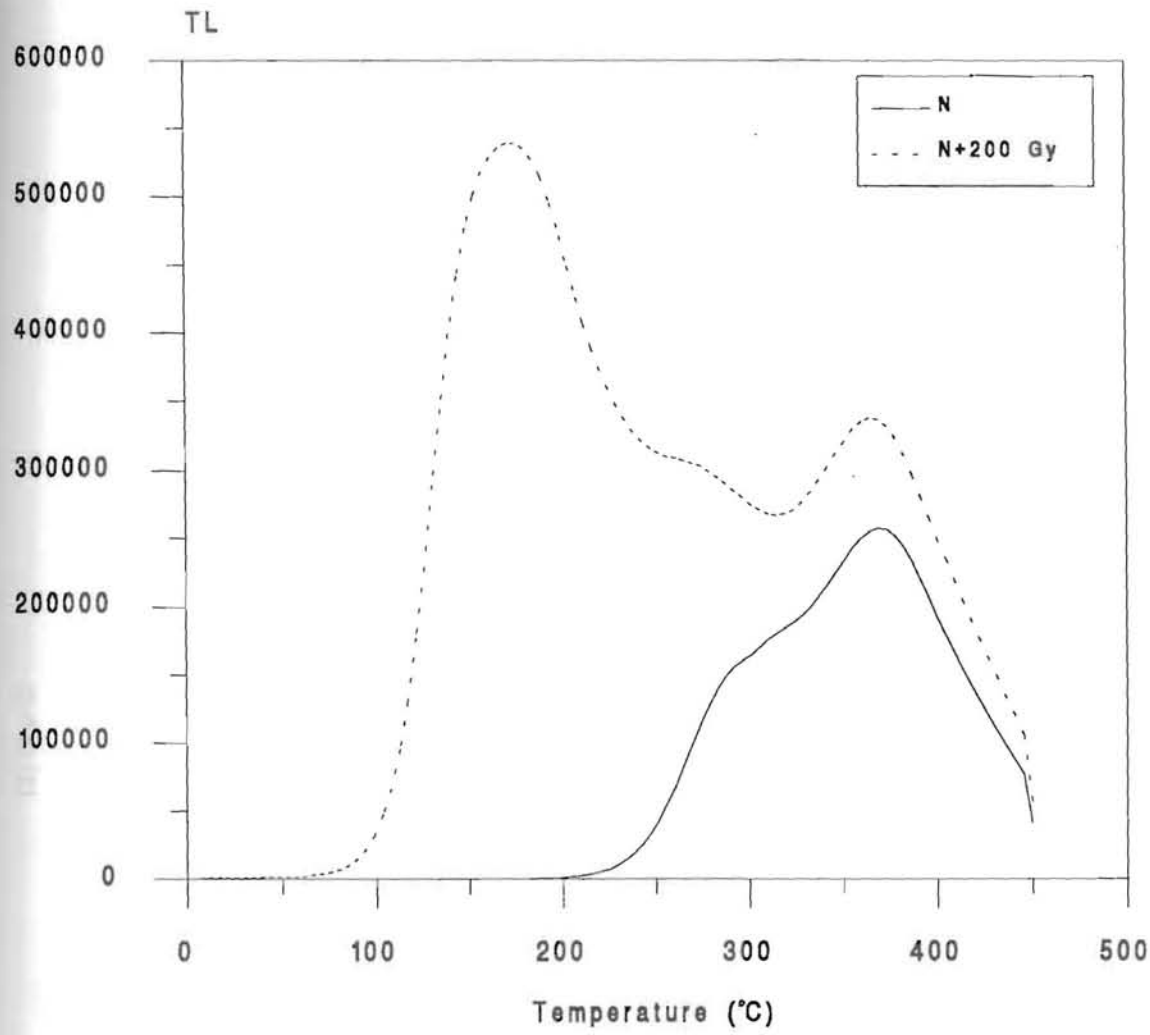


Figure 3.5.2. Typical TL glow curves of samples Z29. (a) Natural + 200Gy ß dose; (b) Natural.

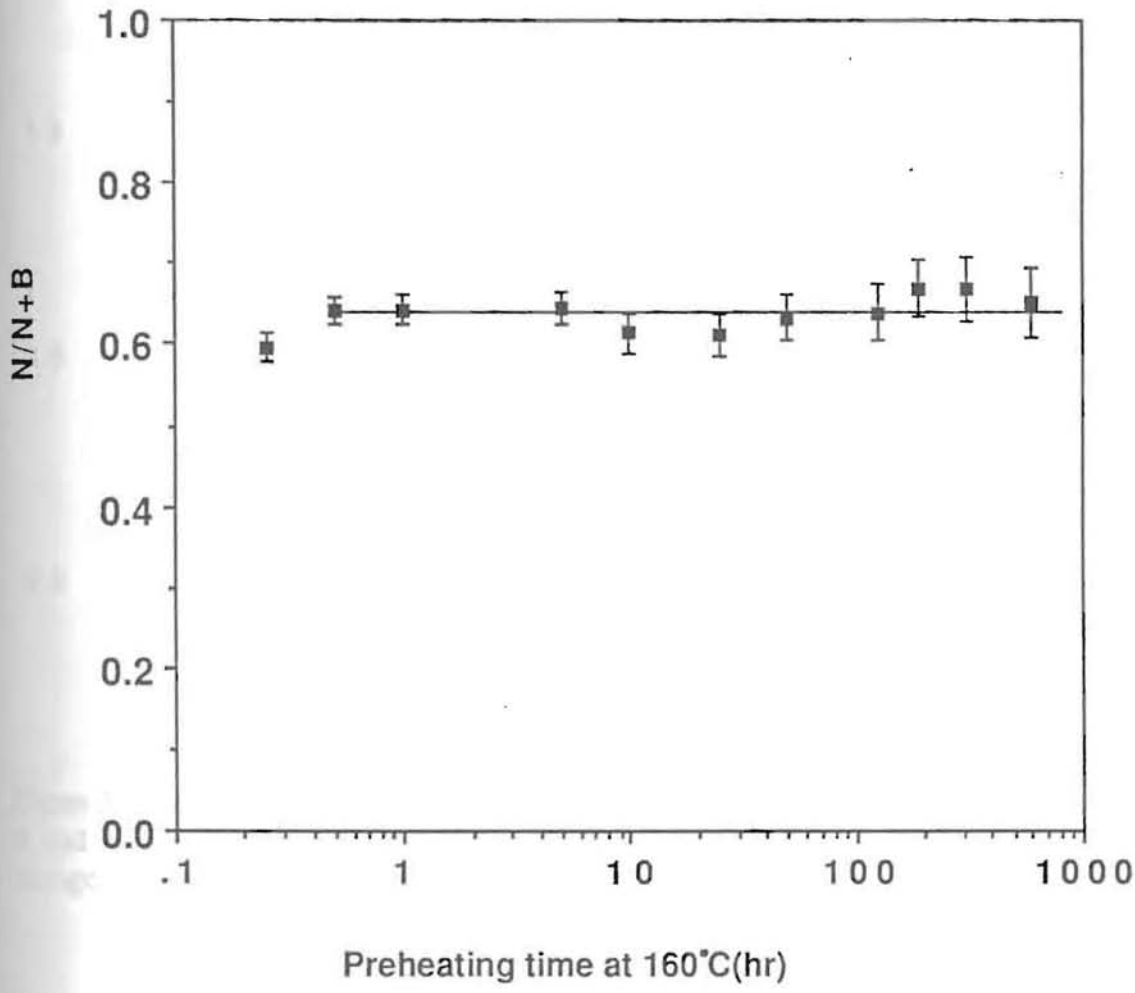


Figure 3.5.3. OSL ratio for N/N+ß versus preheating time at 160°C. The ratio is between natural and natural + ß dose (200 Gy) discs from sample Z29.

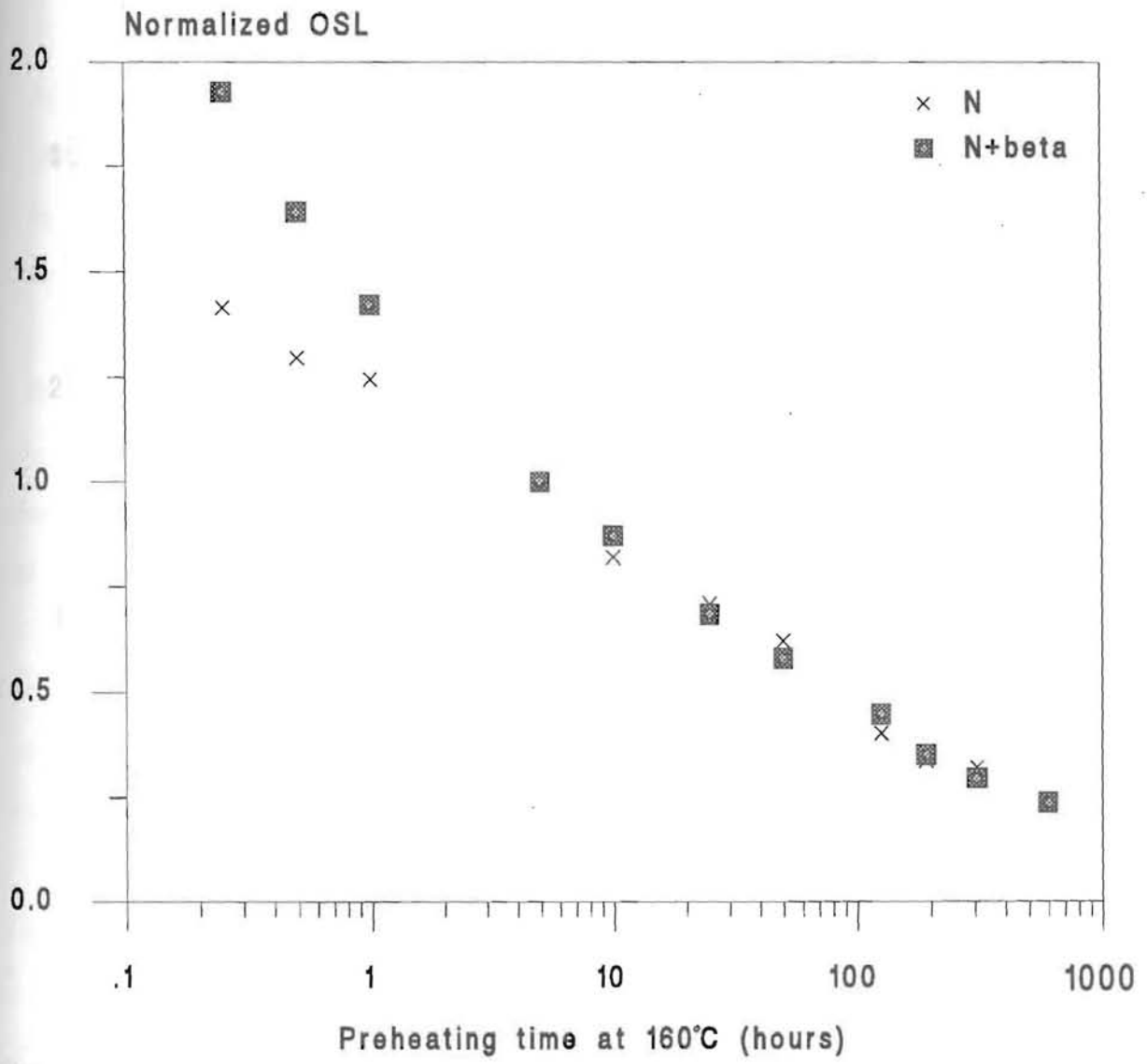


Figure 3.5.4. Isothermal decay curves for OSL signal of sample Z10 at 160°C. Samples were N and N+beta , with a ß dose 70 Gy. Normalization was for the data points for 5 hour storage.

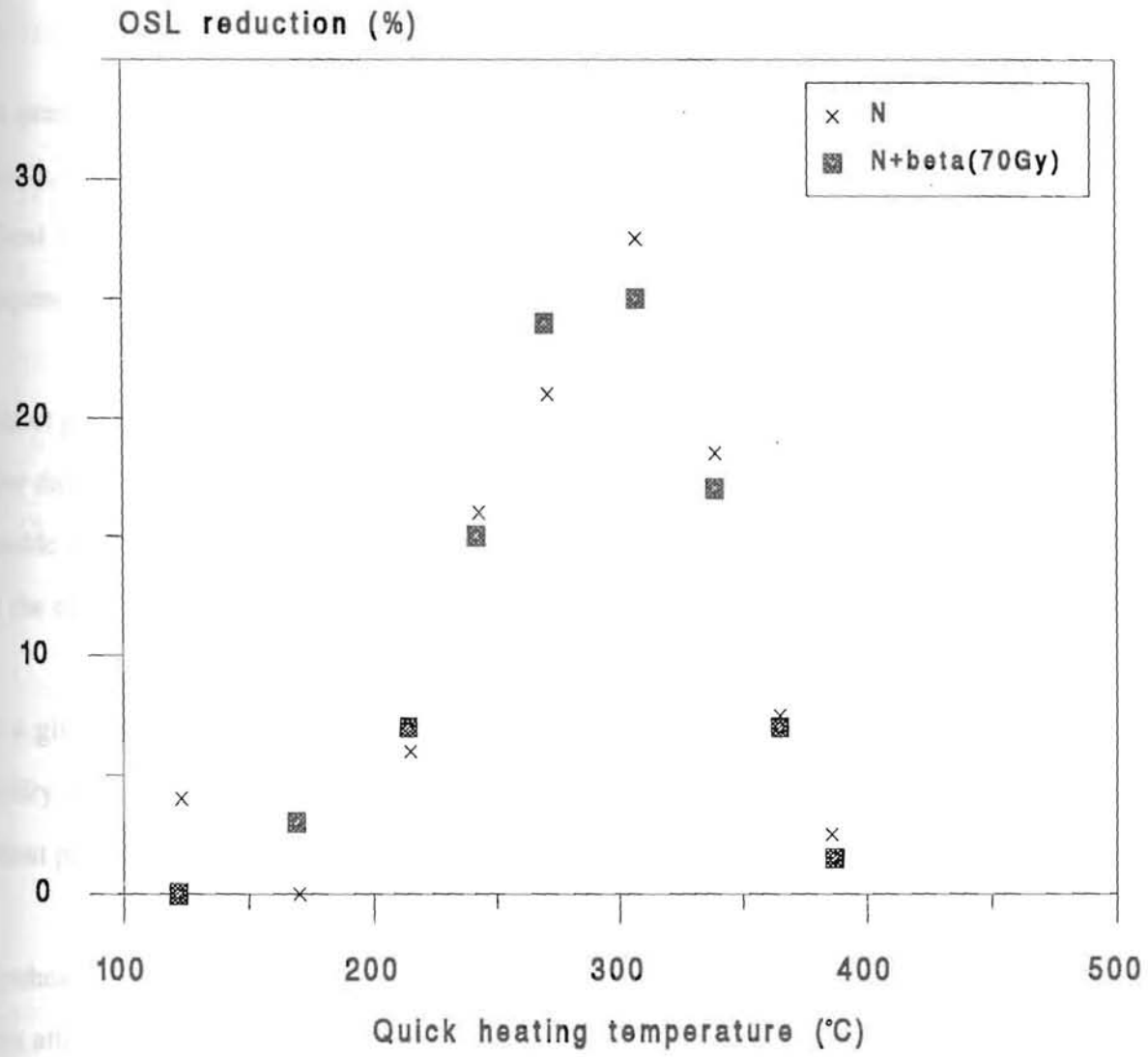


Figure 3.5.5. OSL reduction (%) by quick heating, after preheating a half hour at 160°C for natural and N+ß dosed discs from sample Z10.

The quick heating results in Fig. 3.5.5. suggest that a preheat as short as 0.5 hour at 160°C is suitable. However, the isothermal decay behaviour of the signals (Fig. 3.5.4) suggests that a preheat of longer than five hours at 160°C is recommended for removal of unstable components in K-feldspar samples.

2. Short preheating

Other dating procedures, e.g. the single disc method, require the removal of the thermally unstable signal in a short time. Ten minutes was chosen as a convenient preheating time, and the effects of different preheating temperatures were studied.

For a given sample, the higher the temperature of the TL peak , the higher is its thermal stability (Aitken 1985). To isolate the OSL signal with the highest thermal stability, a preheat procedure was developed to leave a high temperature TL peak around 360°C.

A preheating experiment was carried out on the TL heating plate before TL measurement. In an attempt to isolate a peak in the TL curve after preheating, the Width at Half Height (WHH) of the maximum TL was measured for each preheating temperature. Because the WHH is independent of the mass of grains on the disc, normalization is not necessary. Figure 3.5.6 shows the values of the full WHH (FWHH) for the natural TL and for the second glow response for the same disc sample. The left part of the WHH (LWHH) gave more consistent results. After ten minutes at 210°C, the low temperature peaks were removed, including the 280°C peak which can be seen from the natural sample (Fig.3.5.7). This experiment was repeated for all three samples. The dose used was in the range of 10 to 200 Gy and was less than the ED. Similar results were obtained.

Figure 3.5.6. indicates that a single TL component is isolated after ten minutes at temperature 210°C or higher. This was further demonstrated by a quick heating experiment

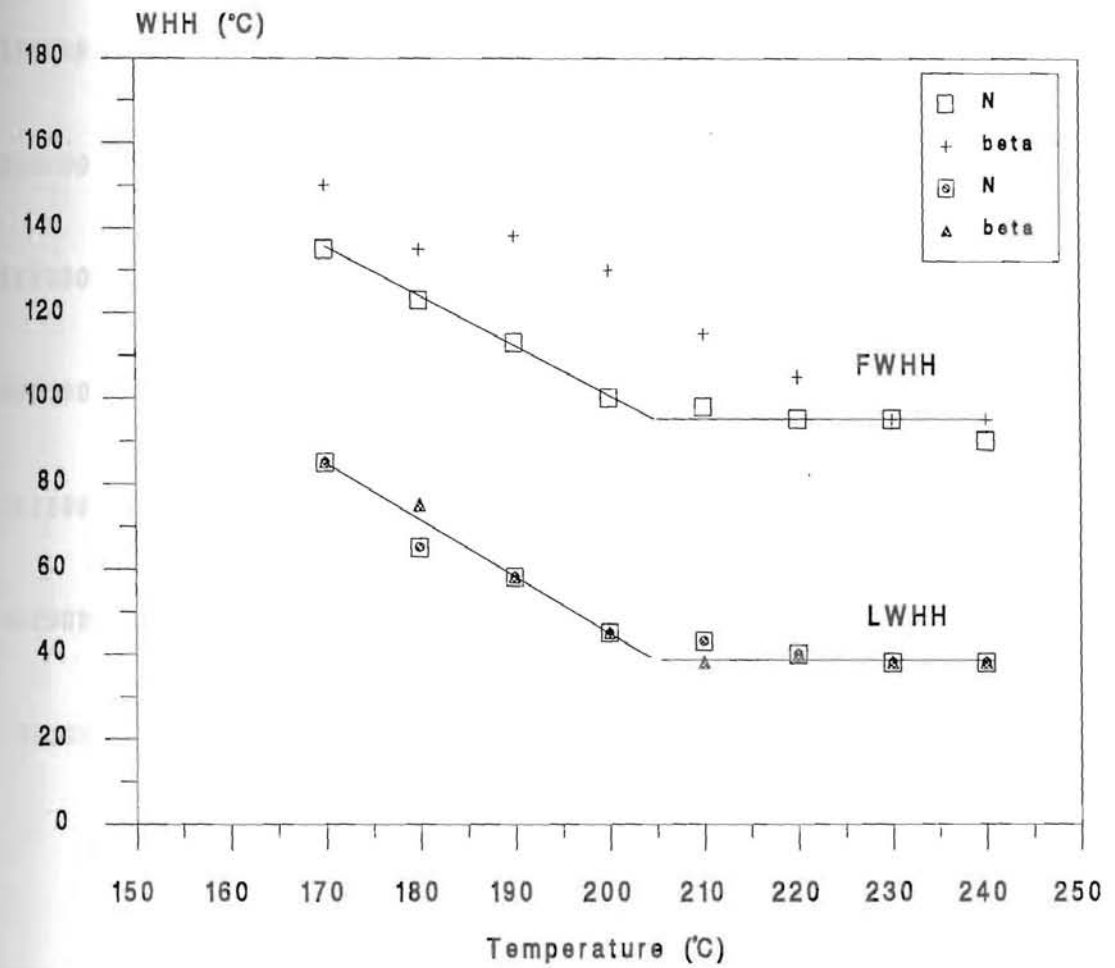


Figure 3.5.6. Plot of FWHH (see text) of TL glow curve versus preheating temperature for ten minutes. The left part of the WHH gives a better result. Results shown for Natural and second glow TL.

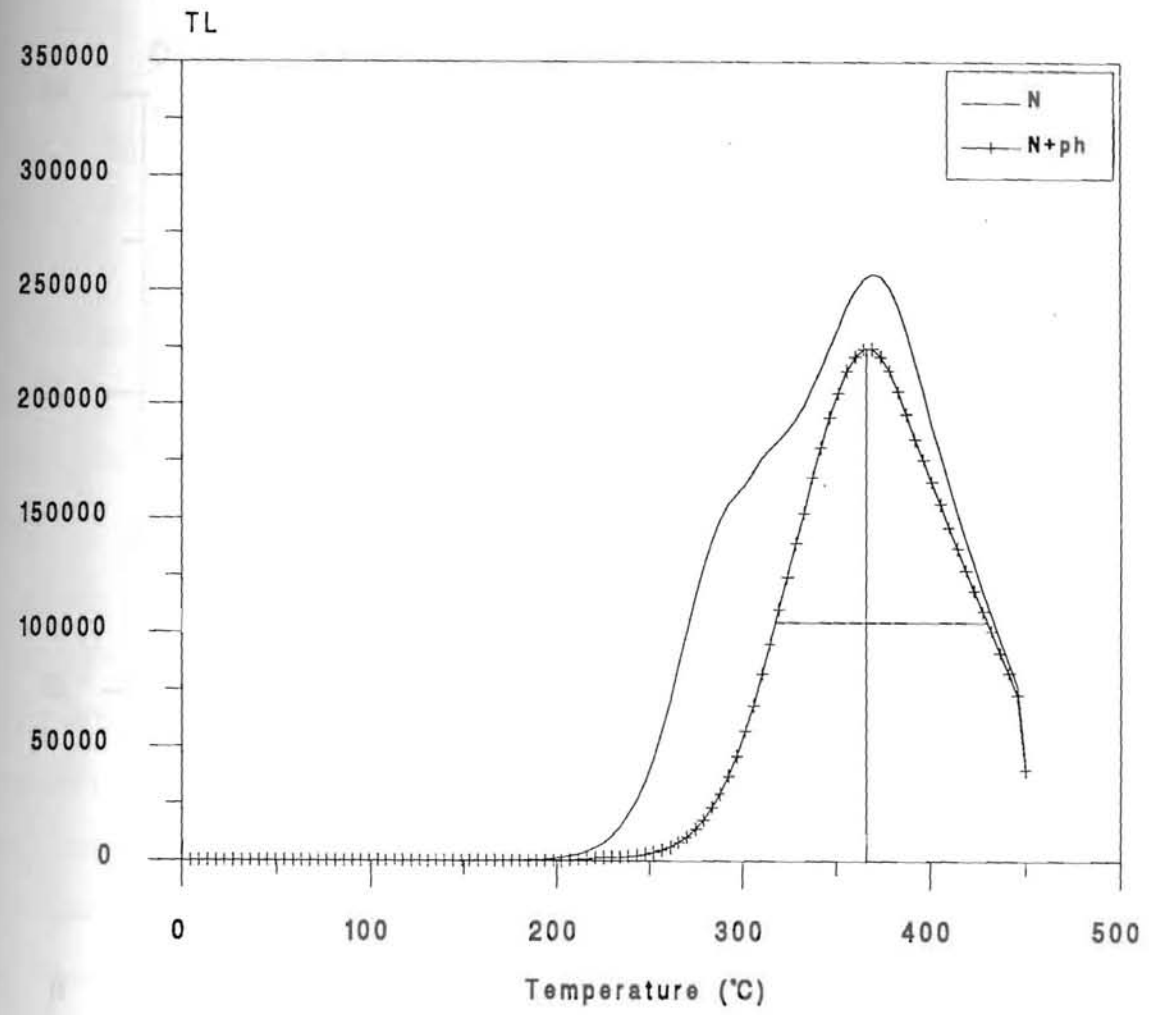


Figure 3.5.7. TL glow curves of sample Z29. The curves are natural and natural plus preheating for 10 minutes at 220°C.

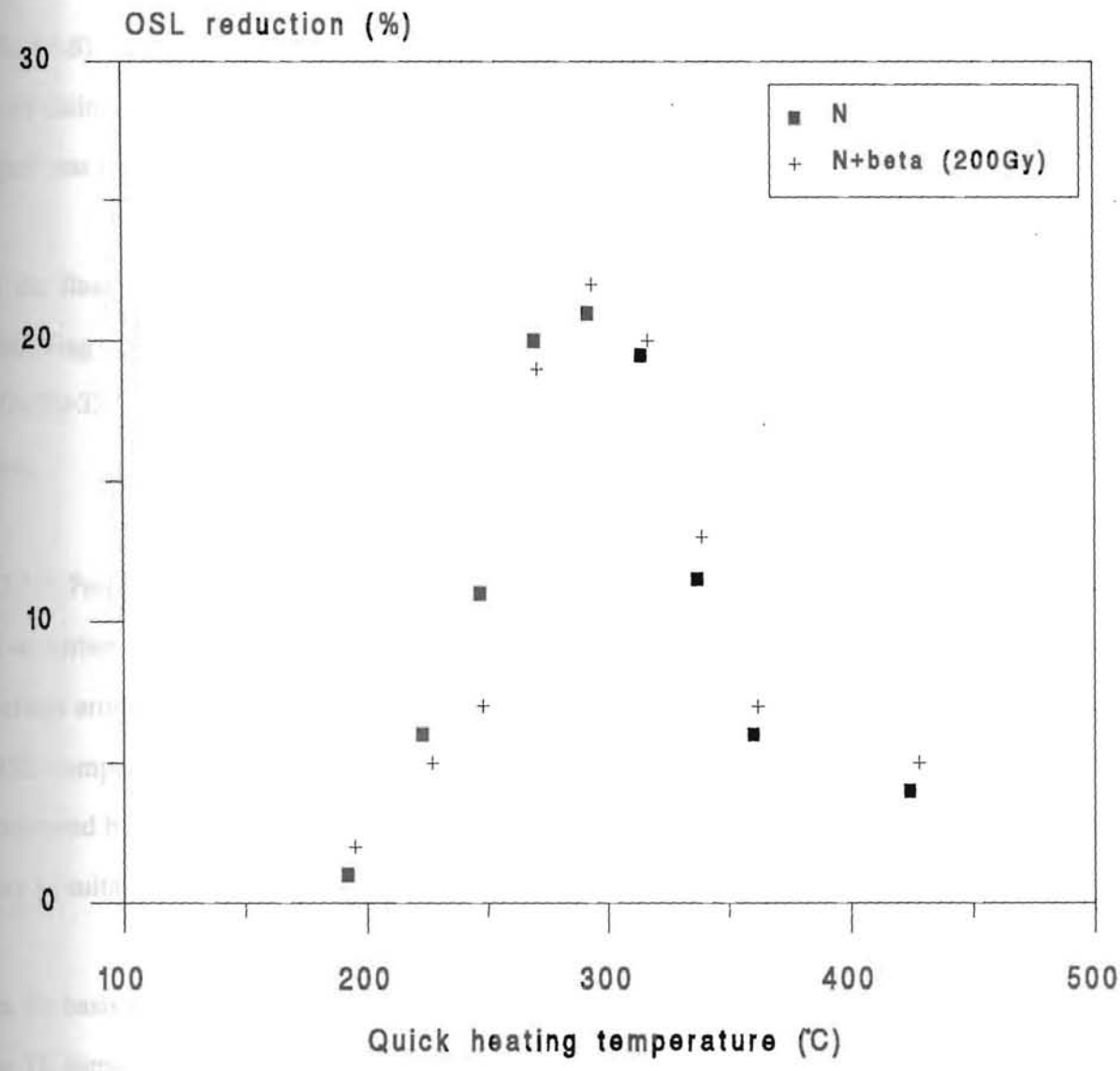


Figure 3.5.8. OSL reduction (%) by quick heating, after preheating 10 minutes at 210°C for natural and N+ß aliquots for sample Z29.

(Fig.3.5.8). which gave almost identical results for the irradiated and unirradiated samples. In all dating studies a preheating 10 minutes at 220°C was used to ensure all the unstable signal was removed.

In the final experiment, the equivalent dose was obtained for sample Z29 using both preheating procedures, 5 hours at 160°C and 10 minutes at 220°C. The values were 272+22/-32 Gy and 232+21/-17Gy respectively, and although not identical overlap at the 1σ level.

3.5.7. Preheating for loess samples

In an earlier section (3.3.3) the stability of the IRSL signal for loess samples from several sections around the world was shown to be similar. The lower thermal stability of the loess IRSL compared with that of K-feldspar suggests that the IRSL signal from loess is not dominated by the signal from K-feldspar. Hence the preheat developed for one loess section may be suitable for other loess sections, but may not be the same as that for K-feldspar.

On the basis of the results of TL dating studies on samples with independent age estimates, the TL signal from loess is unstable in the long term, but for the young loess (<50 ka), ages obtained agree with the geological evidence and independent dating results (Wintle 1990). Hence, the minimal preheating (in time or temperature) required to remove the unstable signal would diverge for samples of different age. For a young loess sample (15ka) from Rocourt, Belgium, the preheating plateau was obtained after preheating 14 hours at 140°C (Fig. 3.5.9). However, for a 70 ka old loess sample from the L1 loess in the Luochuan section, the ratio of OSL signal for natural and natural plus beta dosed discs shows an increase with the preheating time (Fig. 3.5.10), even for 160°C.

Using a similar approach to that applied to K-feldspar fractions (section 3.5.6), an attempt was made to find a suitable preheating procedure based on the thermal responses of the

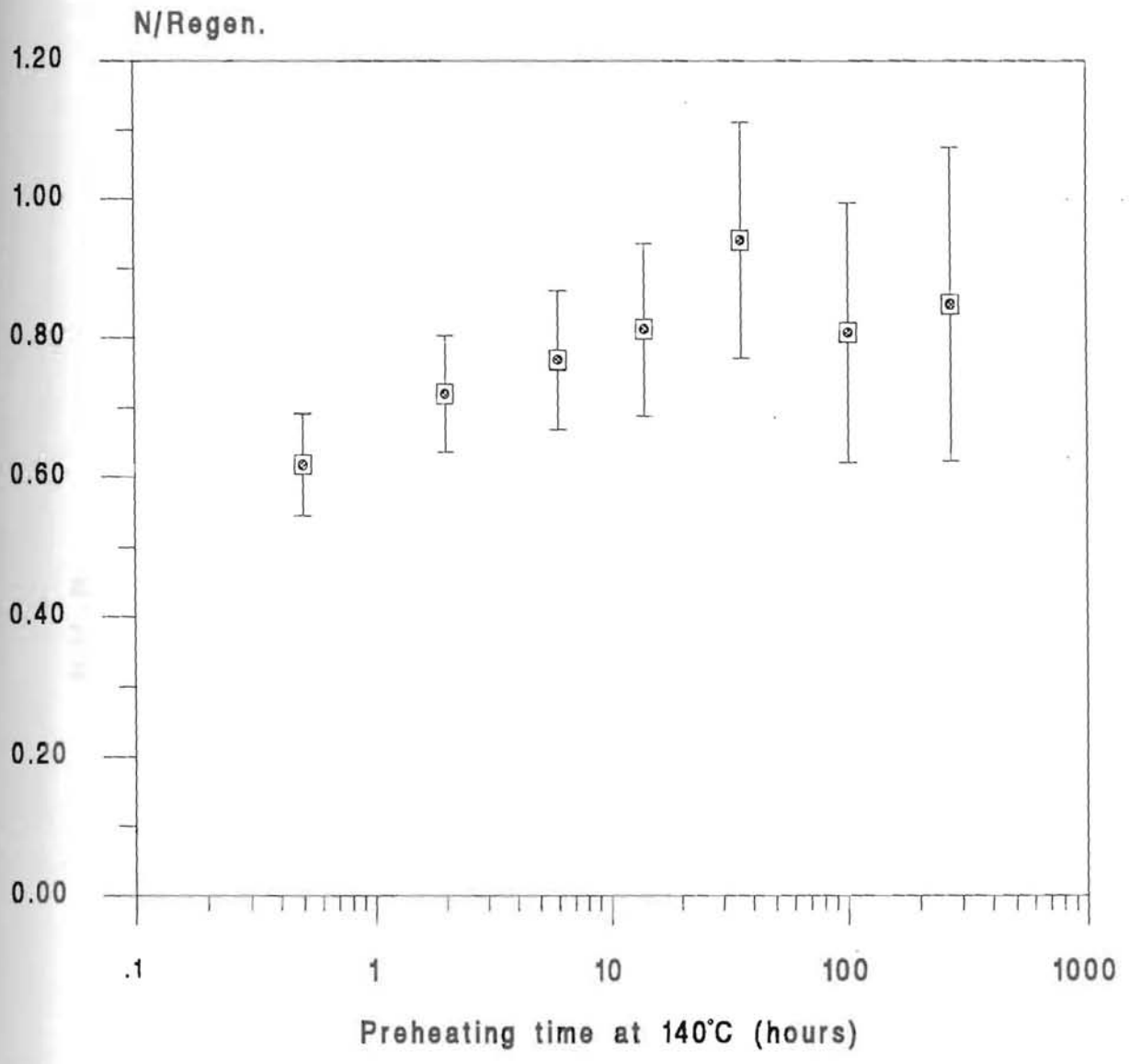


Figure 3.5.9. Preheating plateau of IRSL signals from natural and ß dosed (74 Gy) loess discs. Sample was QTL89D (ED 51 Gy) from Rocourt, Belgium. Preheating was at 140°C.

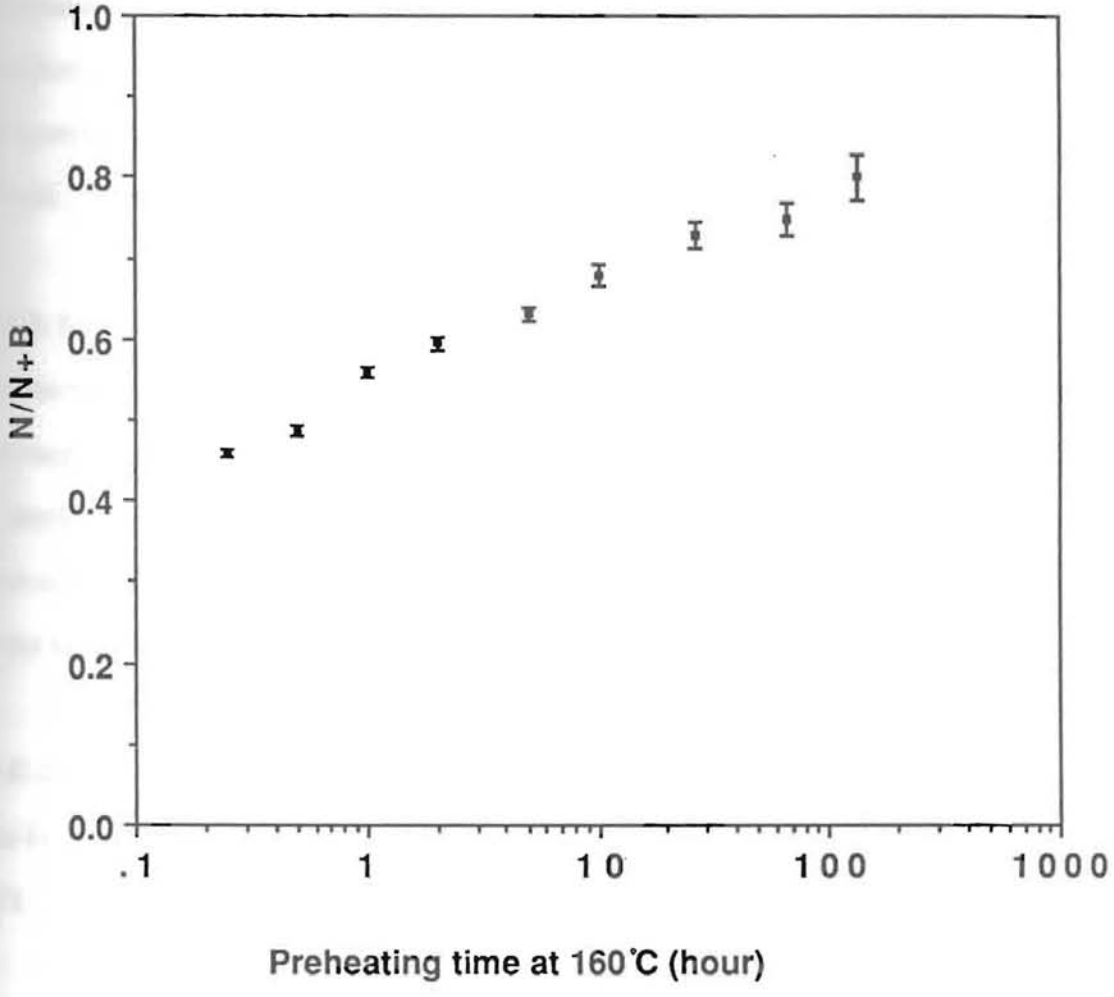


Figure 3.5.10. Preheating plateau of OSL signal for loess sample 88-12 (ED 215 Gy) from Luochuan, China. Preheating was at 160°C. The added beta dose was 200 Gy.

FWHH of TL peaks. However, the responses were complicated and no systematic behaviour was observed. No suitable preheat time was found by observing the thermal responses of the TL glow curves.

In summary: to remove the thermally unstable signal of loess, a suitable preheating procedure can be found for relatively young samples. For older samples, it is difficult to find a universal preheating procedure. More comprehensive studies are needed for old loess samples.

3.5.8. Summary

Short term thermally unstable signals were found in irradiated K-feldspar and polymineral loess samples. Removal of the thermally unstable component by preheating must therefore be employed in optical dating. A suitable preheating procedure can be derived by observation of the thermal behaviour of the signals. The preheat used may differ from one sample to another, depending on its age and mineralogy.

For K-feldspar samples, 10 minutes at 220°C, or 5 hours at 160°C, is recommended. Fine grain loess has shown a more complicated response but experiments suggest that 14 hours at 140°C may be suitable for some samples.

chapter 4

IR STIMULATED SIGNALS FROM COMPRESSED SEDIMENTS

4.1 INTRODUCTION

4.1.1. Initial research

The fact that it is possible during infrared stimulation to obtain a luminescence signal that is proportional to the sedimentary age of the mineral grains opened a new field for luminescence dating studies. Because the samples do not need to be heated, equipment different to that used for TL can be devised. The discovery that IR diodes can be used to stimulate luminescence from feldspar allows cheap and flexible IRSL systems to be built. These are capable of a wide range of uses (Godfrey-Smith et al. 1988, Spooner and Franks 1990).

During the last few years several papers have been published on dating of sediments by measuring the IRSL (Spooner et al. 1990, Aitken and Xie 1992). Spectral measurements of the IRSL indicated that it has a similar spectrum to the TL from the same feldspar (Bailiff and Poolton 1991, Huntley et al. 1991). Stability studies suggest that the IRSL signals from different loesses have a similar stability, and this is lower than that from K-feldspar (Li and Wintle 1992; chapter 3). Previous luminescence dating studies illustrated that the luminescence signal is related to several factors: these include the mineralogy of the sediments; the time elapsed since the last exposure to light; the radioactivity of the sample and its surrounding; and the extent of last exposure to sunlight.

4.1.2. Deposition of loess

During the past few years, loess has been studied because of its relevance to palaeo-climate changes (Liu et al. 1985). However, the deposition rates for a loess sequence and their variation cannot be evaluated precisely unless extensive chronological data are available.

This could be achieved by luminescence dating with intensive sampling (Li, 1986), but it is highly labour intensive and time consuming because of the long sample preparation times required.

Loess contains luminescent materials, quartz and feldspar grains, which are likely to have been exposed to sunlight prior to deposition. The mineral composition, grain size and radioactivity are relatively uniform down a section (Liu et al. 1985, Pye 1988). The similar radioactivity suggests that samples from a particular section will have a similar dose rate. The IRSL signal is thus simply related to the time since the loess was last exposed to light. and hence the signal increases with depth down the section. These characteristics make loess one of the simplest sediments with which to work.

4.1.3. IR scanning of core sediments

Clearly it would be useful if the IRSL signal could be measured rapidly along a loess core or down a section. Continuous measurement has been achieved for a glacial lacustrine sediment using an IR core scanning system built in this laboratory (Duller et al. 1992). The stimulation and detection system (manual Daybreak) was the same as that used for disc measurement (section 2.1.1). The core was moved beneath the LED array on a motor-driven core carrier.

The advantage of such a system is that it allows the IRSL signal from the core to be measured in a matter of minutes. Since the strength of the IR illumination on the core sediment is relatively low, the reduction of signal due to each measurement is negligible. Therefore repeated measurements can be made on the same material. Also, because no heating is involved, the core sediment can be used for other analyses.

It was planned to use the scanning system for loess cores from Rocourt, Belgium. Four and a half meters of loess were collected in five 1m cores with 10cm overlapping at each end of

the core. However, the fragile nature of loess prevented a smooth surface being obtained for running on the core carrier. The signal varied as the distance between the surface of the core and the LED altered because of the uneven surface. Also, the manual Daybreak IR system was designed for disc measurement, and the signal detection efficiency was not optimized. These two problems led to the abandonment of continuous measurements for these cores, but forced the development of other approaches.

In this chapter I will report another way of measuring the IRSL signal using compressed bulk sediments. A portable IRSL system will be described, which can be used for different shaped materials. A new method of normalization using the u.v. regenerated signal will be introduced.

4.2. IRSL FROM COMPRESSED LOESS

4.2.1. Introduction

The failure of the scanning system for the loess cores led to the consideration of other ways of measuring the IRSL signal along a core. Instead of continuous scanning, an approach which measures individual samples can have the same effect if sampling is sufficiently detailed. This was achieved by using compressed pellets for each sample point.

The section of the loess cores from Rocourt has been well-studied from the geological point of view (Haesaerts et al. 1981). The top 4m of the section has been TL dated using the fine grain method (Wintle 1987).

Samples, at 10 cm intervals, were taken along the cores. 4 pellets were made at each point. The results are the average of these four pellets.

The use of compressed pellets has advantages over the scanning system. First, it permits a u.v. normalization procedure to allow for possible mineral variation down the section (see

4.4). Second, it has a constant geometry to minimize the surface effect. Third, the cross section area of the pellets is the same as the discs used for conventional dating and therefore the detection efficiency is optimized.

4.2.2. Preparation of pellets

After cleaning a 2mm thick layer from the surface, loess was taken from the core and compressed using a modified car jack. This pellet-maker creates a pressure of 2 tonnes on the sample. The pellet is cylindrical, 10 mm in diameter and weighing about 0.5 g. The height of each pellet is around 0.6 cm. The pellets were sufficiently robust to be handled and the flat end surface could be cleaned with a razor blade or sandpaper.

The pellets were prepared using the natural water content, about 5%. The pellets were then dried in the dark at room temperature for more than three days in order to avoid any effects resulting from water content variation.

4.2.3. IRSL signals from pellets

The natural IRSL signal of the dried pellets was measured with 1 second IR exposure. The measurements were performed on the manual Daybreak system at room temperature. It can be seen from Fig. 4.2.1 that the natural signal increases with depth.

Because the signals can be affected by differences in feldspar concentrations between the pellets, a normalization procedure was used to minimize this effect. It involved using the signal regenerated by exposing the pellets to u.v. radiation (see 4.4) after removal of the natural IRSL. After measuring the natural signal, pellets were bleached by sunlight for 6 hours to completely erase the natural signal. This was checked by measuring the IRSL signal after the bleaching. After exposure to the u.v. lamp for 1 hour, the u.v. regenerated signal was measured at room temperature following an overnight delay (16 hours). In order to minimize any difference between experimental conditions, groups of pellets were

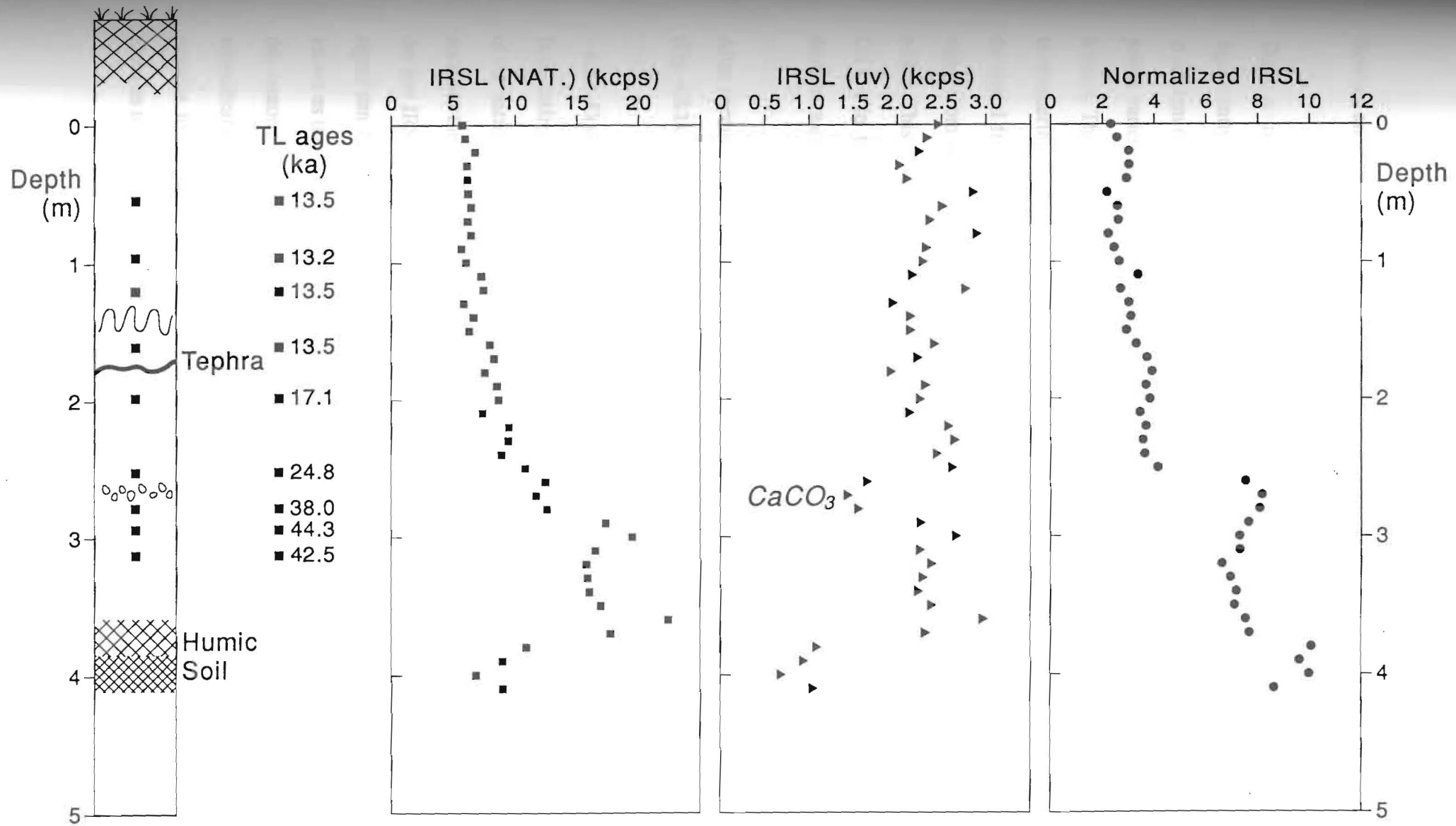


Figure 4.2.1. Schematic representation of section at Rocourt showing the tongued horizon, Eltville tephra and stoney horizon. The TL ages (errors of ±10%) are from Wintle (1987). The three logs show the natural IRSL, the response to u.v. and the normalized IRSL.

bleached and u.v. exposed in batches.

The u.v. regenerated IRSL signals were similar for most of the loess pellets, but significantly different to those from the pellets from the soil horizon and the loess layer rich in calcium carbonate (Fig. 4.2.1). It can be seen that the u.v. regenerated signal of individual pellets varies by a factor of 3, with the lowest signals observed for samples from the soil horizon. This may be caused by the feldspar concentration decreasing in the palaeosol due to weathering during the soil forming processes or increase in clay sized particles masking the signal from the feldspar grains. In the middle part of the section, the u.v. regenerated signal from three samples was also shown to be smaller than that from most of the loess pellets. This is due to the relative reduction of the feldspar concentration by the presence of $CaCO_3$ in this part of the section. The higher carbonate content of these pellets was demonstrated by a strong reaction to weak HCl.

After normalization, the IRSL signal increases with depth in steps rather than gradually (Fig. 4.2.1).

4.2.4. Comparison with the results of TL dating

In an earlier study (Wintle 1987), 9 samples were taken at 0.5m intervals down the top 4m of the section and TL dates ranging from 14 to 45 ka were obtained (Fig. 4.2.1). It can be seen that the relatively low age (13.5 ka) of the uppermost 2m of the section is reflected in the low IRSL signal (both normalized and unnormalized). A large jump in the normalized signal can be seen corresponding to the change from the loess above the fine stony layer known as the Beuningen Gravel to that below . This jump is masked in the measurements of the unnormalized signal because of the presence of calcium carbonate. Similarly, normalization also takes account of the soil horizon near the base of the section. Hence it is possible to obtain an overview of the pattern of loess deposition through time without recourse to a full dating programme.

4.2.5. IRSL of gamma dosed pellets

As observed in luminescence dating, a non-linear response of the IRSL signal with dose occurs at higher doses because of trap saturation. This results in the natural IRSL being relatively depressed compared to the equivalent dose. This response to radiation was demonstrated for pellets made from one sample taken from the uppermost part of the core.

24 pellets of the sample were given different doses of gamma radiation, measured and a growth curve for the IRSL signal with dose was constructed. Six different doses of 0, 24, 48, 96, 240 and 480 Gy were given, with four pellets at each dose. Pellets were placed in glass tubes for irradiation.

In order to remove the thermally unstable component created by the irradiation, all of the pellets were preheated for 18 hours at 140°C. The IRSL signals were measured with 1 second IR exposure, and then bleached for 6 hours by sunlight. Pellets were checked by a short shine measurement to make sure that the IRSL signal was erased by the bleaching. Then the pellets were exposed for one hour to the u.v. lamp. The u.v. regenerated signal was measured after 16 hours delay and used for normalization. The u.v. normalized IRSL signal was plotted against the added Gamma dose (Figure 4.2.2). This shows a non-linear response. The form of the plot is similar to the additive dose growth curve for TL or IRSL signals obtained for fine grain (4-11 μm) loess. Assuming zero IRSL signal for zero age samples, an ED of 45.2 Gy was obtained for the natural sample when a saturating exponential fitting was applied. The response is relatively linear up to 100Gy of added dose.

Because the loess pellets used to obtain the first IRSL column of figure 4.2.1 were measured without preheating, the normalized IRSL signals of the third column cannot be converted directly into the equivalent gamma dose by using figure 4.2.2 as a calibration curve. Therefore a correction for signal loss resulting from the preheating has to be applied. If it is

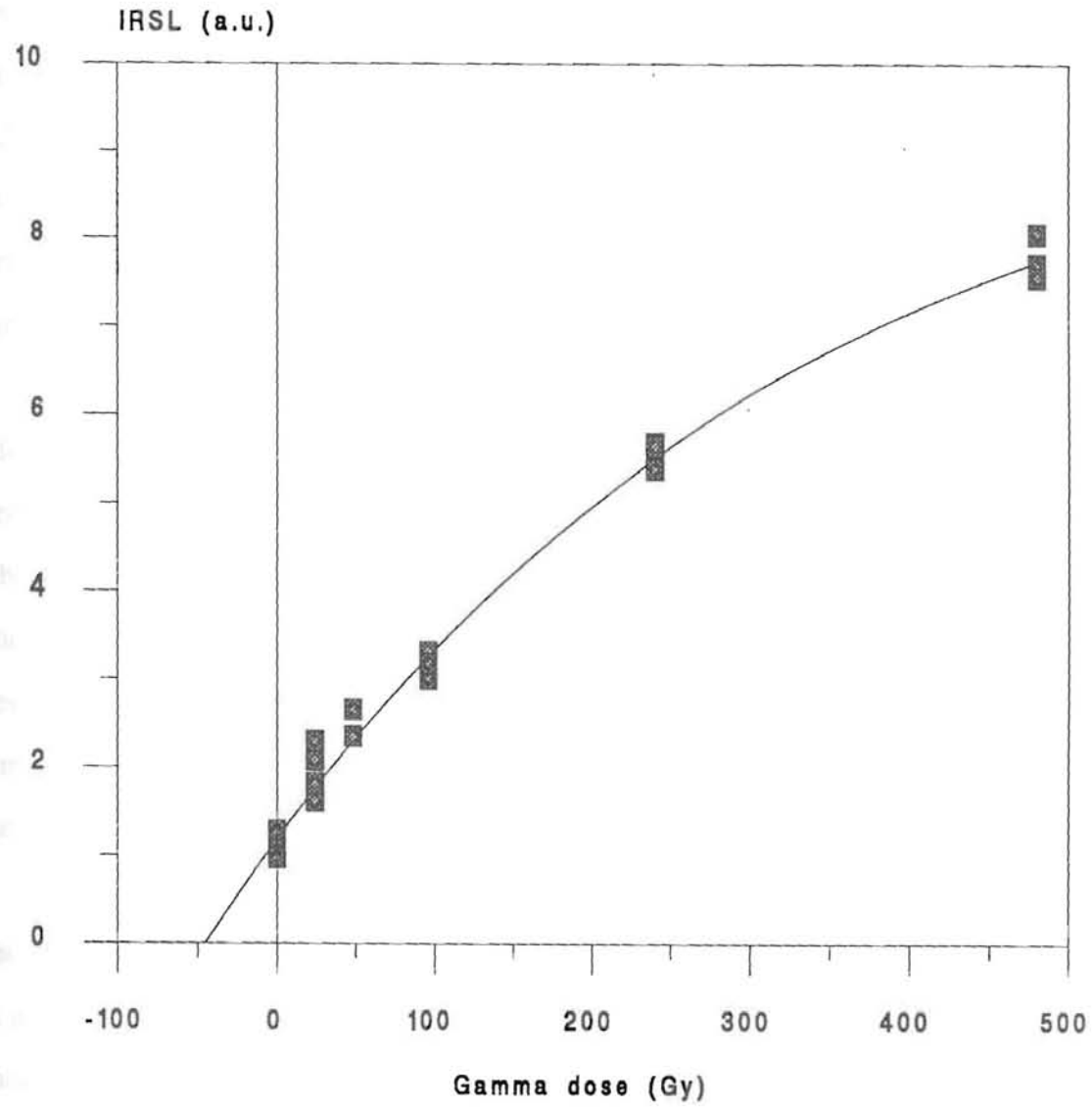


Fig. 4.2.2. IRSL from pellets of a sample as a result of γ irradiation. The sample is from the top of Rocourt loess section.

assumed that the proportion of the stable signal lost due to preheating is not dependent on the age of the sample, a normalized IRSL signal versus ED curve can be constructed (Fig.4.2.3). The correction factor used was obtained from the ratio between the normalized signal of the pellets without gamma dose, in figure 4.2.2, and the value of the natural signal measured previously for the same sample (Fig. 4.2.1). This factor was employed to for all dose points in figure 4.2.2.

If the dose response of the IRSL signal is the same down the section, estimated EDs can be obtained by comparing the u.v. normalized IRSL signal and the curve of Fig. 4.2.3. Figure 4.2.4 shows the estimated EDs versus depth. Because it is relatively young, the IRSL signal is in the almost linear region (up to 150Gy) for most samples, except for the humic soil samples at the bottom of the section. This correction is not essential for the resolution of different loess packets in this study. However it is expected that the ED correction would become more significant if older samples were studied.

Because there is a possibility of inhomogeneity in the way the gamma rays pass through the pellets in the glass tube, there might have been a different build up effect between the top and bottom of the pellets after gamma irradiation. This proved not to be the case as shown by identical results being obtained for both ends of the pellets.

4.2.6. Assessment of the results and implications

The results give an overview of the pattern of the loess deposition. The experimental procedure only takes about two weeks for a 4.5 m section. This saves much time and labour compared to a full luminescence dating programme.

A break in deposition was detected in the middle of the section. The normalized IRSL signal did not increase gradually but showed two major step changes, implying rapid and discontinuous loess deposition in this region. Some of the sedimentary features were

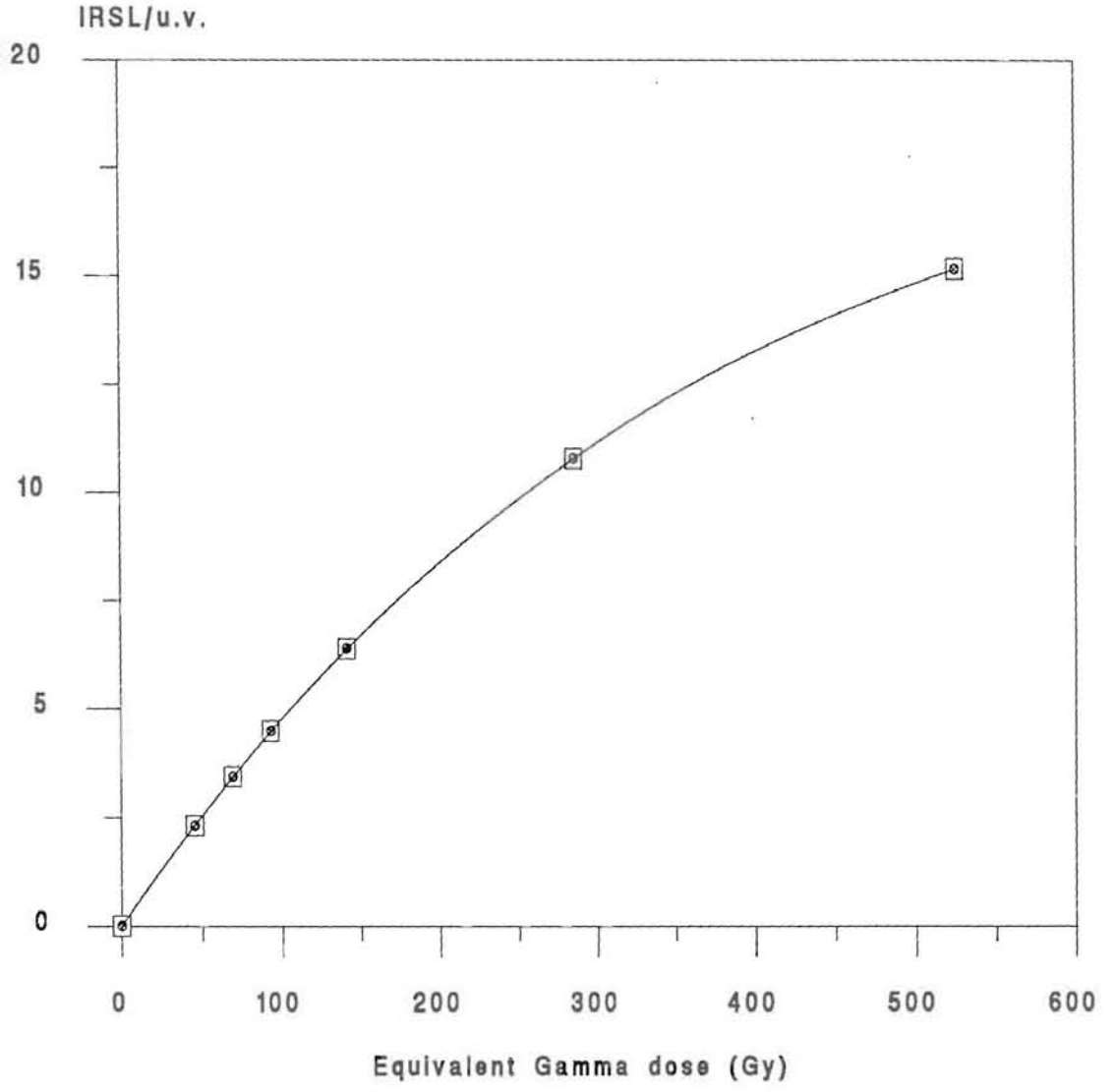


Fig. 4.2.3. IRSL responses to the γ irradiation after correction for preheating. The dose was corrected using the ED of the natural sample.

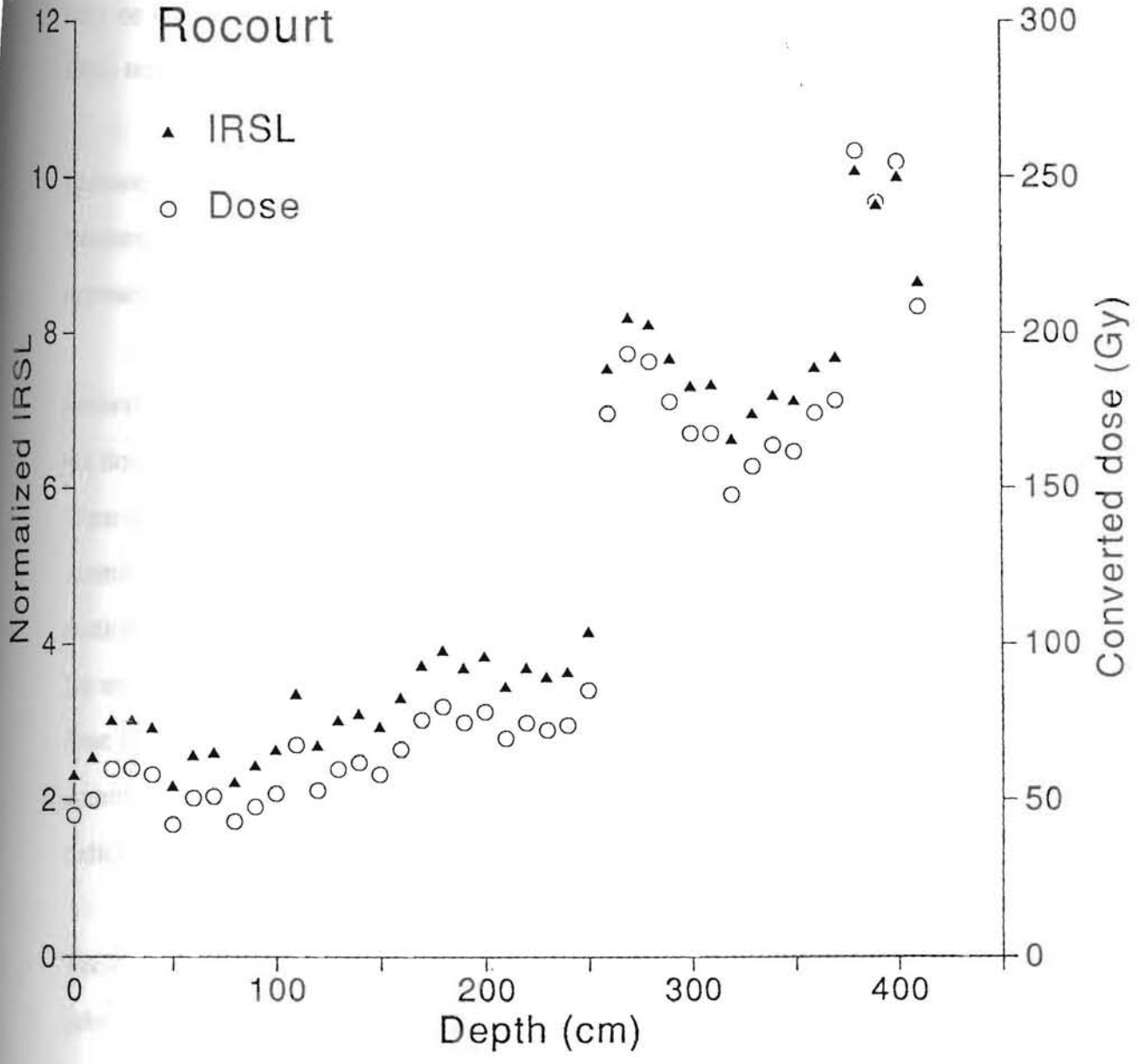


Fig. 4.2.4. (▲) Normalized IRSL as in Fig.4.2.1. (o) Values of past radiation dose obtained using the response in Fig.4.2.3.

observed in the u.v. regenerated signals; these include the humic soil horizon and the presence of $CaCO_3$ in part of the section. However, the tephra and tongued horizon were not detected in the signal. Since the tephra was in a thin layer of less than 1 cm, it could easily have been missed in sampling. Although the tongued horizon has a different structure to other loess, the response to the IRSL signal is identical. This implies that the method can only be used to detect sedimentary features which have an age difference, or a modified IRSL response.

Because the IRSL signal increases in steps, a better sampling strategy can be devised for a luminescence dating programme. Since each step corresponds to a depositional event, representative samples can be taken.

Several assumptions have to be made in the interpretation of the normalized IRSL signal. 1. the dose rate is the same for the whole section; 2. the previous signal was bleached to zero when the grains were deposited; 3. any mineral and grain size differences in pellets can be normalized by the u.v. regenerated signal. This last assumption will be discussed in the next section. Because the IRSL is very sensitive to light, it is probable that the signal is zeroed for most sediments except those deposited rapidly, such as colluvial deposits. The constant dose rate assumption may be valid only for some sections. Field measurements, e.g. with a gamma spectrometer, are needed to check this and detect the differences. Previous studies indicate that the dose rate for loess is similar for a section and over a large area (Li 1986).

Because there are so many factors involved in the generation of the signal, the accuracy of relationship between the normalized IRSL signal to the ED (or age) of the sample is not easy to evaluate. For four pellets of the same sample a reproducibility of 5% was achieved for the natural signal after u.v. normalization. This suggests that the method has a reasonable precision.

The non-linear response to dose means that the method may not be used for old samples (>100 ka). This may be analogous to the limitation found in luminescence dating (Debenham 1985, Spooner and Questiaux 1989, Wintle 1990). The method may not suitable for very young samples (<1 ka) because the natural signal is unlikely to be bright enough to measure.

Since no heating is involved, the measurements are not destructive. The pellets can be reused in other analyses or can be used to make repeat measurements because the central part of the pellets has not been exposed to light. The equipment used is simpler than that for TL measurement and this makes it possible to build a portable system (see next section) for use in the field.

4.3 PORTABLE IRSL SYSTEM

4.3.1. Introduction

Since no heating is involved, the IRSL measurement can be made without the additional equipment required for TL dating, e.g. vacuum pump, inert gas supply and hot plate. Also, because an IR LED is quite small, it is possible that a portable IRSL system can be made, which can be used for pellets, discs or sediment cores either in the field or in the laboratory. Use of a fibre light guide makes the system more flexible when used with different shaped material. In this section I will deal with a new design of IRSL equipment.

4.3.2. Equipment design

The IRSL system consists of the optical unit, the detector and the counting unit. The detector is connected to the optical unit by a fibre light guide. The counting part works in a similar way to that used in a TL reader (Bøtter - Jensen and Mejdahl 1985). The schematic diagram is shown in figure 4.3.1. Except for the detector, the units were located in a light tight box.

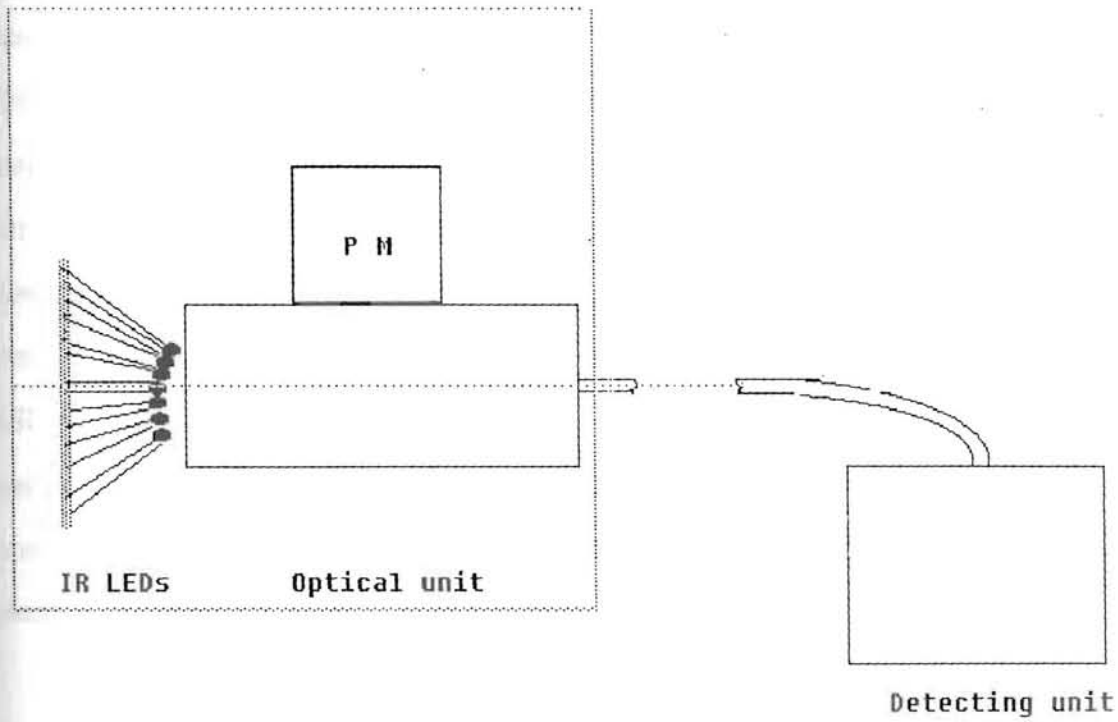


Fig. 4.3.1. Schematic diagram of the portable IR system.

1. Optical unit

This unit consists of two lenses, one concave and one focusing, a BG 39 filter and a cold mirror. Figure 4.3.2a shows the schematic diagram of the unit. When the IR from the LEDs passes through the concave lens, it becomes sub-parallel. It then passes through the cold mirror, and is focused into the fibre light guide by the focusing lens. The 45° cold mirror allows the IR to go through. Figure 4.3.2b shows the wavelength response of the cold mirror. It passes the IR and reflects light of wavelengths between 400nm to 700nm. The IRSL, which consists mainly of visible light, comes back from the sample along the fibre light guide (also see detector unit). It becomes sub-parallel after passing through the focusing lens. The cold mirror then reflects the IRSL onto the PM tube photocathode after passing through the BG39 filter.

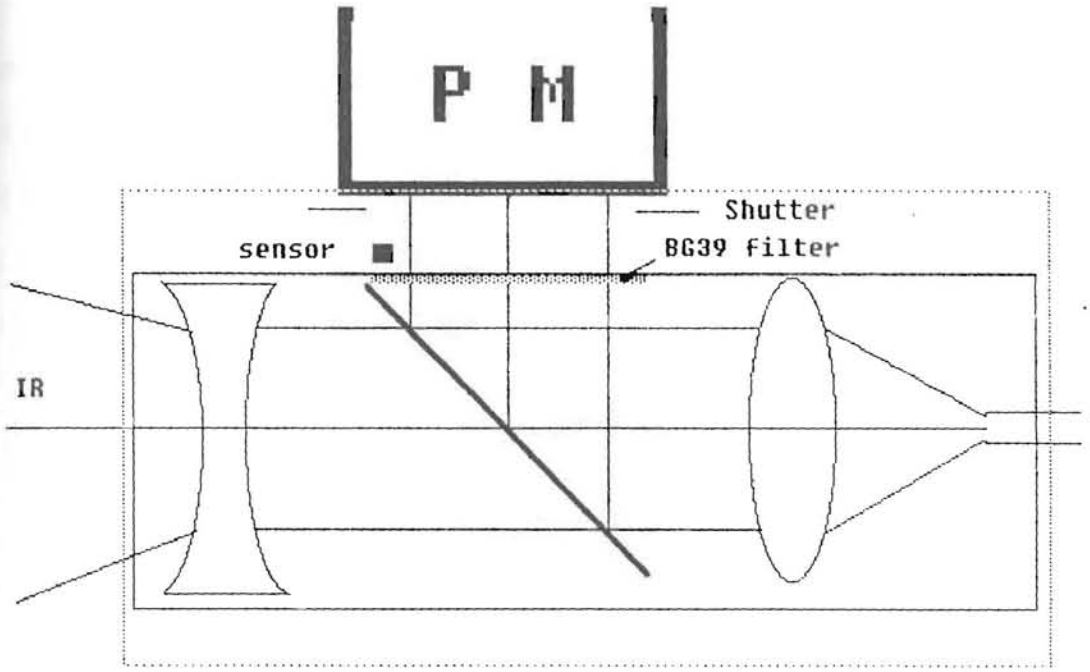
The use of the cold mirror is the heart of the design, a feature which allows the IR and the IRSL signal to share the fibre light guide.

2. Detector

The detector is on the other end of the fibre light guide (Fig 4.3.3). The IR from the fibre light guide becomes sub-parallel when it passes through the focusing lens. The IRSL from the sample is focused into the fibre light guide. Thus the same light route is used for both IR and IRSL and so minimizes the loss of IRSL by reducing the detection angle. The IRSL focused on the light guide is from all the grains exposed to the IR.

The IR power from the individual LEDs may not be exactly the same. However, since the IR is mixed up during its passage through the fibre light guide, the IR power over the sample area is relatively uniform. The sample detection area is circular and 12 mm in diameter. This is a similar size as disc samples used in the laboratory.

(a)



Optical unit

(b)

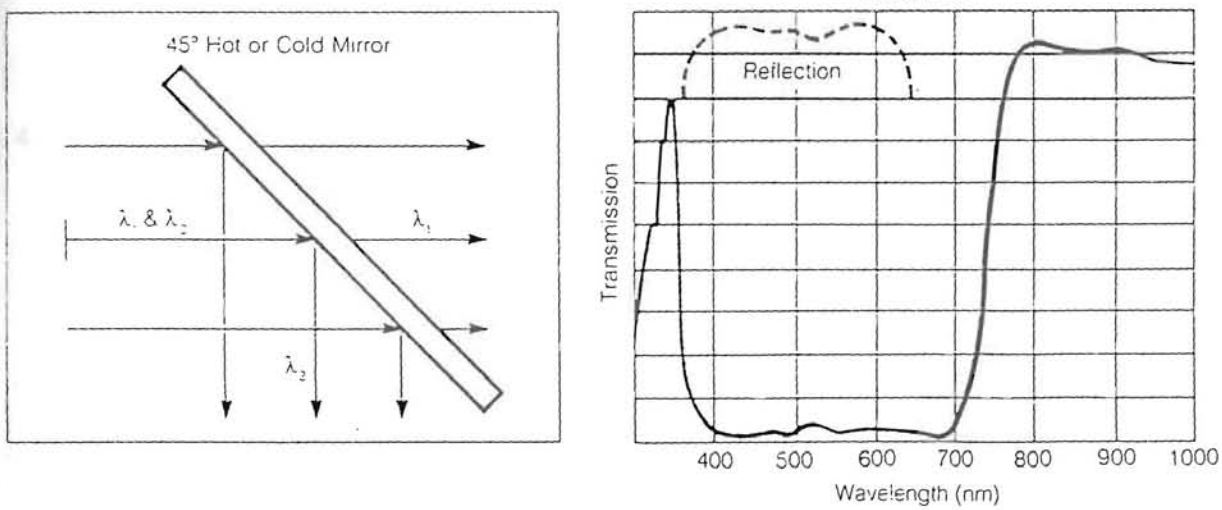


Fig. 4.3.2. (a) Schematic diagram of the optical unit (b) Characteristics of the cold mirror (from Ealing Catalogue)

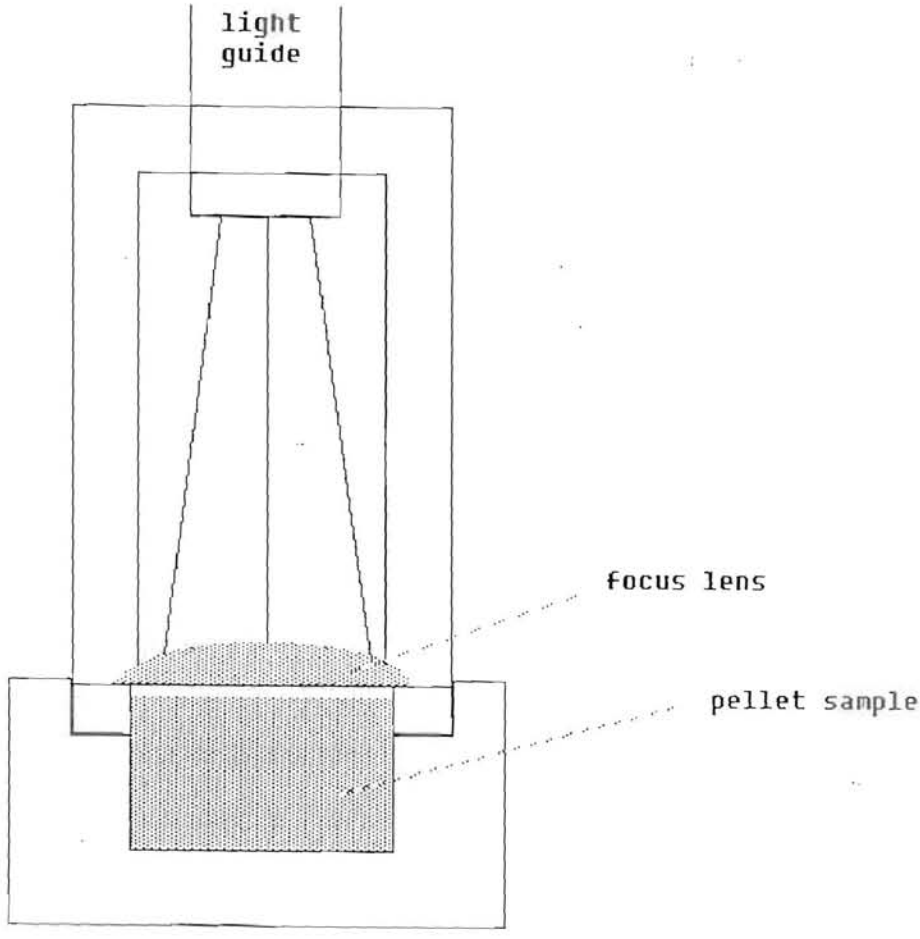


Fig. 4.3.3. Schematic diagram of the detector unit.

3. Counting unit

The PM tube used in the counting unit is similar to that used in TL and IRSL measurement. A scaler ratemeter similar to that in a portable gamma spectrometer (NE-PSR 8) is used for display. It displays the IRSL signal count for selected lengths of time; different scales can be chosen. An output interface to micro-computer was built in order to store and analyse the data.

Since the PM tube is extremely sensitive to light, it is important to avoid it being exposed accidentally to light. Several safeguards were incorporated into the design to prevent this: the use of auto-off on the high voltage supply, optical shutter in front of the PM tube and a light sensor. If the light sensor detects any count rate higher than a certain level, the shutter will be automatically closed and automatically switch off the high voltage.

Based on the above design, a portable system was built in Aberystwyth. Some of the optical components used in the system are listed in Table 4.3.1.

Table 4.3.1. Components used in the portable system

COMPONENT	TYPE	COMPONENT	TYPE
PM tube:	EMI 9924B	Fibre light guide:	27-6295
Focus lens (optical):	42-1222	Shutter:	Nikon 501 camera's shutter
Focus lens (detector)	42-1198	Optical Filter	BG39 (2mm)
Concave lens	42-0919	IR LED	TEMT 484
Cool mirror	35-6923		

note: Unless specified the number quoted is from the Ealing Electro-Optics Catalogue.

4.3.3. Performance of the system

The performance of the system was only tested for a limited number of sediment samples. Only the pellets which gave a bright signal with the manual Daybreak system were measured. For a loess sample from Dolni Vestonice, Czechoslovakia, the IRSL were 3600 c/s and 710 c/s for the manual Daybreak system and the portable system respectively. The u.v. regenerated IRSL signal was not bright enough to be detected with the portable system

because of massive signal loss through the system.

The loss of the IRSL signal was tested by monitoring the light from a C-14 light source. It was found that about 75% of the signal was lost in the fibre light guide, and about another 20% of the signal was lost in the optical unit. Although no correction has been made for the wavelength difference between the IRSL and the C-14 light source, a massive signal loss in the system was observed. It is therefore necessary to improve the signal detection.

The loss of the IR in the system has not been examined in detail but the IR arriving at the sample position was monitored using an IR laser sensor card for Newport. Higher IR power can be achieved by increasing either the number of LEDs or the current through each of them.

4.3.4. Potential and improvements

The advantage of this system is that it is portable, and measurements can be performed in the field area (e.g. in the hotel room if not actually on site). If these measurements can be compared directly to other measurements such as dose rate and magnetic susceptibility, the sedimentary pattern and environmental information can be assessed. This gives a better sampling strategy, and also provides additional sedimentary information. This system could thus become a useful tool in Quaternary studies.

The following suggestions are made to improve the system.

1. Selecting a different type of fibre light guide so that there is less loss of IR and IRSL through the light guide. The light guide used had several broken fibres and poorly polished ends when supplied from the optics company.

2. Optically optimizing the distances between the lenses and the mirror.

3. Removing the fibre light guide and the detector unit, i.e. use a more direct and more compact system. Pellet or disc measurement would still be made but it would be a less flexible system.

Although suggestions 1 and 2 would improve the detection efficiency, sediments with a very weak signal may still be unmeasurable. This is especially true for observation of the u.v. regenerated signal. For the loess samples used in section 4.2, the u.v. regenerated signal is only about 2.5 kc/s compared with 5 kc/s of natural signal for a 14 ka old sample. It is probable that other sediments, e.g. lake sediments, may have lower signal than loess because of low feldspar concentration. Suggestion 3 would be better for such sediments.

4.3.5. Summary

The portable system could be used to obtain rapid measurements in the field. The system has advantages for sampling and is flexible to use. However, the system built in Aberystwyth needs to be improved comprehensively.

4.4 UV REGENERATED IRSL SIGNAL

4.4.1. Introduction

Early on, it was found that there was a considerable difference in natural IRSL output from pellets of a similar age. It was also found that the older palaeosol sample gave a lower IRSL signal than the loess sample above it. It is therefore necessary to correct for individual pellet sensitivity.

Different normalization procedures have been introduced in luminescence dating techniques (Aitken 1985, Rhodes 1990). Except for weight and natural normalization, this was achieved by measuring the luminescence signal after α, ß or γ irradiations. For pellets a different approach must be used. Because only the very top of the pellet produces the signal, weight normalization cannot be used. Since the signal measured is not used to construct a

growth curve, natural normalization cannot be applied. Use of high energy irradiation, e.g. α, β or γ, is desirable, but it cannot be used in the field nor can it be used conveniently for pellets in the laboratory.

Exposure of feldspar grains to short wavelength radiation from the u.v. lamp (chapter 2) shows that the IRSL signal is increased for a sample previously bleached by visible wavelengths. A similar increase in IRSL was reported by Bailiff and Poolton (1989) when samples were exposed for 10 minutes to wavelengths less than 280 nm. For the short wavelength radiation from the u.v. lamp (chapter 2), the signal increased to a maximum level after 1 hour (Fig 4.4.1), and this time was chosen for subsequent experiments.

In this section, I will discuss the use of this regenerated signal for normalization of the different IRSL signals from pellets. Related factors, e.g. optical depth of IR and IRSL in the pellet, are discussed.

4.4.2. U.V. regenerated signal on diluted loess

The u.v. normalization procedure was tested by normalizing pellets which contained known proportions of IR sensitive and insensitive materials. Since no IRSL signal has been found from calcite and quartz (Spooner and Questiaux 1989) but a signal is found in most feldspars, the IRSL signal will decrease when quartz or calcite is added to a sample with a fixed concentration of feldspar. Therefore normalization using the u.v. regenerated signal can be tested using samples with known relative feldspar concentrations. Bearing in mind the research objective, a 70 ka old Chinese loess (sample 88-12) was chosen as the IRSL emitting material. Quartz or carbonate, widely found in most sediments, were added as the diluting material.

1. Diluting with carbonate material

The carbonate used was a lake sediment core from Morocco (supplied by Dr H.F. Lamb).

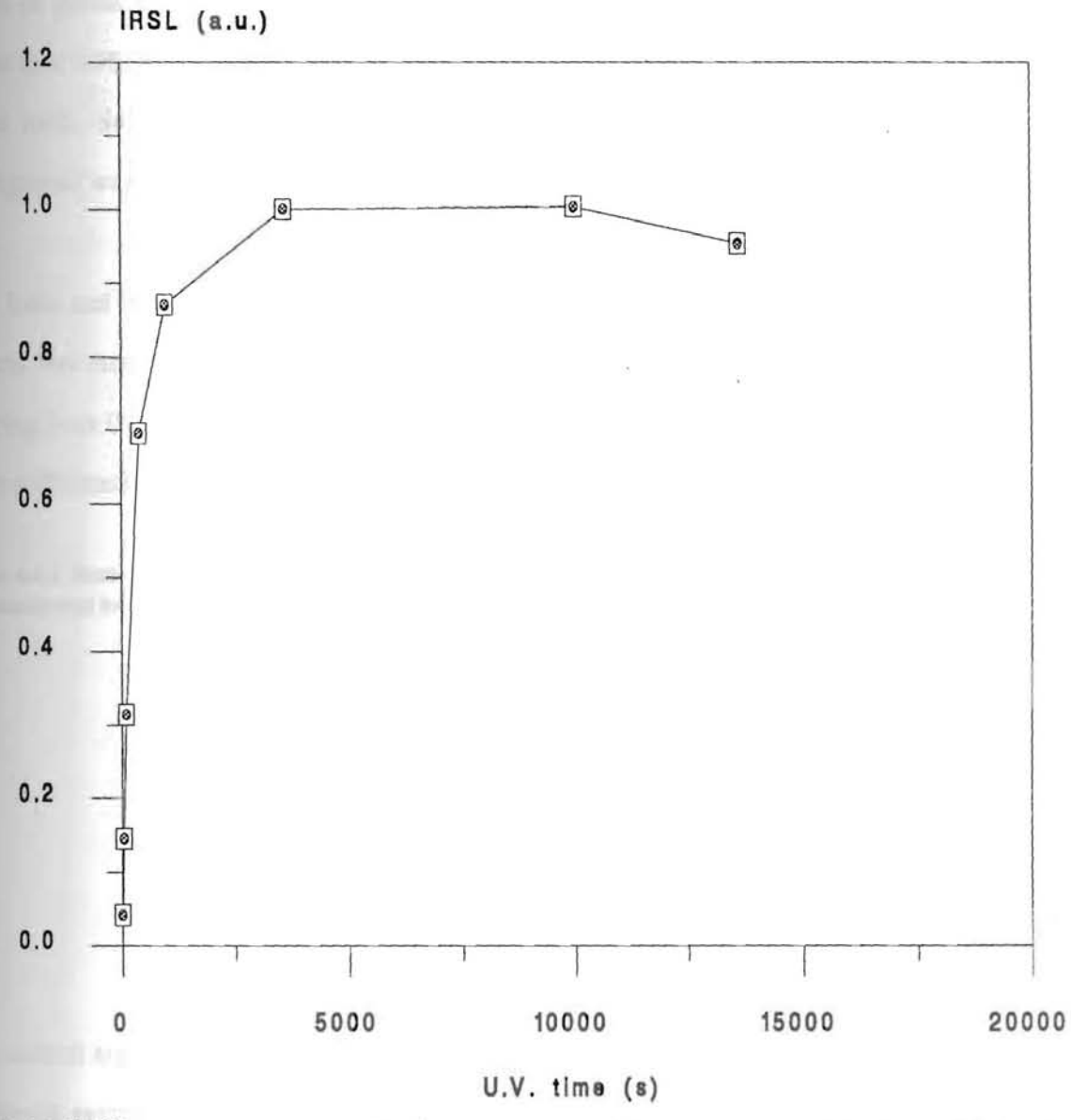


Fig. 4.4.1. IRSL response as a result of u.v. exposure. The sample used is the K-feldspar fraction separated from sediments

Most of grains were finer than silt sized. Reacting this material with HCl indicated that more than 90% of it is carbonate. The remaining part was mostly organic material such as plant roots. Subsamples were given a ß dose of about 100Gy and no IRSL above the background was detected, thus confirming that the material gives no IRSL signal.

The loess and the carbonate material were ground lightly with pestle and mortar. A set of pellets was made from well homogenized mixtures of both materials, with a loess content varying from 0 to 100% in 10% steps. The fine grain size of both materials made pellets that were sufficiently robust to be handled.

Table 4.4.1. Ratio of normalized IRSL signal relative to that from 100% loess. Pellets are mixture of carbonate and loess.

% loess	Ratio
10	1.07±0.09
20	1.13±0.18
30	1.11±0.07
40	1.06±0.02
50	1.08±0.09
60	1.09±0.03
70	0.99±0.01
80	1.01±0.06
90	1.01±0.04
100	1.00±0.08

The natural signal of these pellets was measured with 1 second IR exposure in the manual Daybreak system (Fig. 4.4.2a). The signal increases monotonically with the loess content, though not linearly. The pellets were bleached for 6 hours by sunlight and then exposed to the u.v. lamp for 1 hour. After 16 hours delay, the u.v. regenerated signal was measured under the same conditions (Fig. 4.4.2b). The u.v. regenerated signal also increased with the loess content, in a similar pattern as for the natural signal. The u.v. regenerated signal was used to correct for the response of the different loess contents. The results for duplicate pellets relative to 100% loess are given in table 4.4.1. Normalization of the natural IRSL signal was thus achieved using this approach.

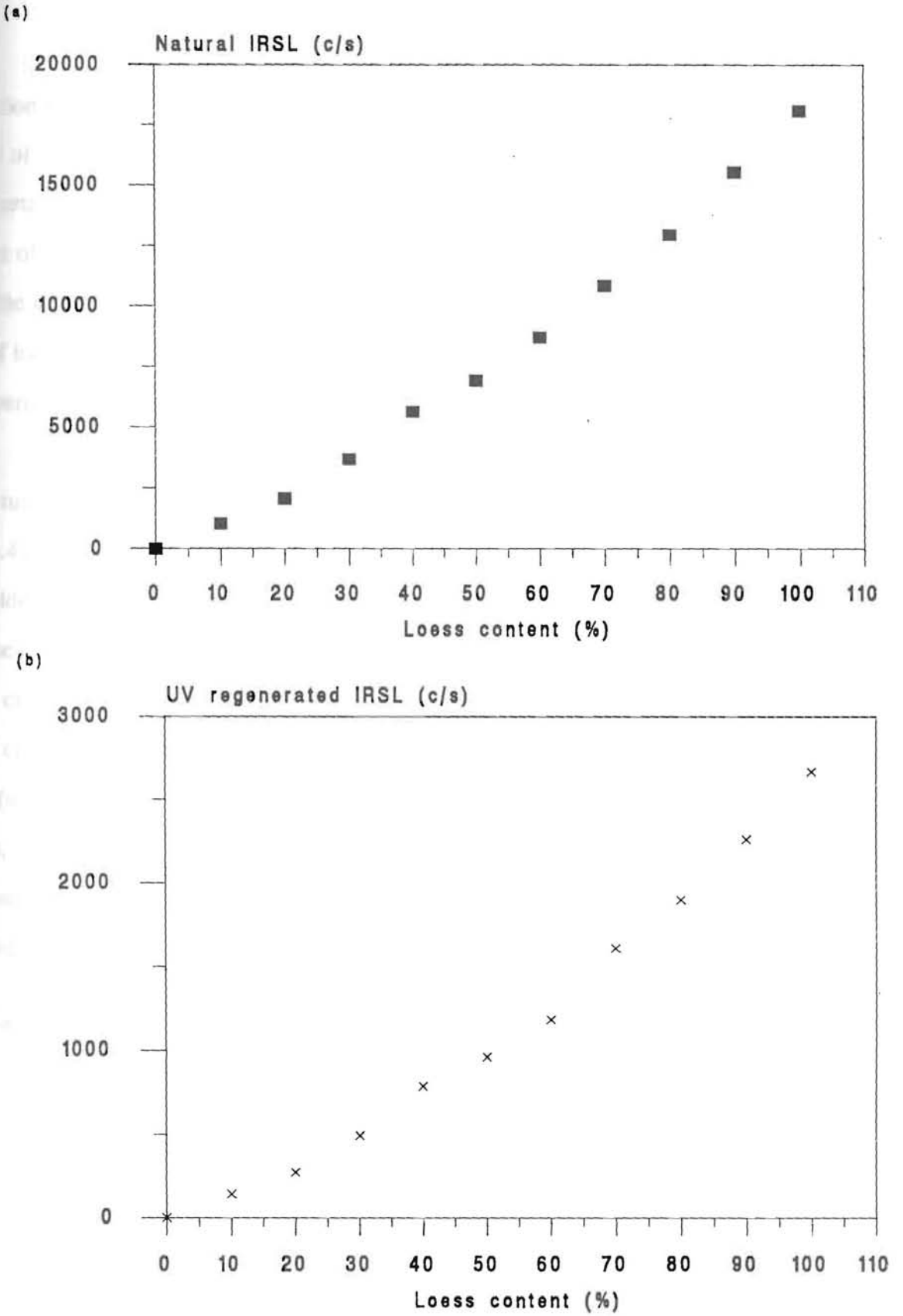


Fig. 4.4.2. IRSL signals of pellets of mixed loess and carbonate. (a) natural signal (b) u.v. regenerated signal after bleaching with visible light.

2. Dilution with quartz crystals

Instead of carbonate, crystalline quartz was added to another subsample of the same loess. The quartz was ground in a ball mill to less than 200μm. A set of pellets was made from the mixture of quartz and loess, with the loess content varying from 0 to 100% in 10% steps. Since the quartz has not been ground finer than the loess, the pellets containing 0, 10 and 20% of loess were not as robust as those mixed with carbonate and grains were lost during the experiment.

The natural and u.v. regenerated signal were measured for these pellets as described above (Fig. 4.4.3a and 4.4.3b). It shows that the signal responses are different to those for pellets with added carbonate. Both natural and u.v. regenerated signals show a rapid increase with increase of loess content; whereas the signal increased relatively slowly with loess content for the carbonate mixture (Fig. 4.4.2). For both materials the trends derived from the figures do not cross the signal axis at the origin. This is related to the effective depth of the IRSL signal (see below). Once again the natural signal was corrected by using the u.v. regenerated signals, and the results are listed in Table 4.4.2. Because of the quality of the pellets made with quartz, the normalization result is not as good as for those diluted with carbonate. Also only one pellet was used for each measurement.

Table 4.4.1. Ratio of normalized IRSL signal relative to that from 100% loess. Loess mixed with quartz.

% loess	Ratio
10	0.90
20	0.69
30	0.93
40	1.13
50	0.92
60	0.96
70	1.04
80	1.05
90	1.08
100	1.00

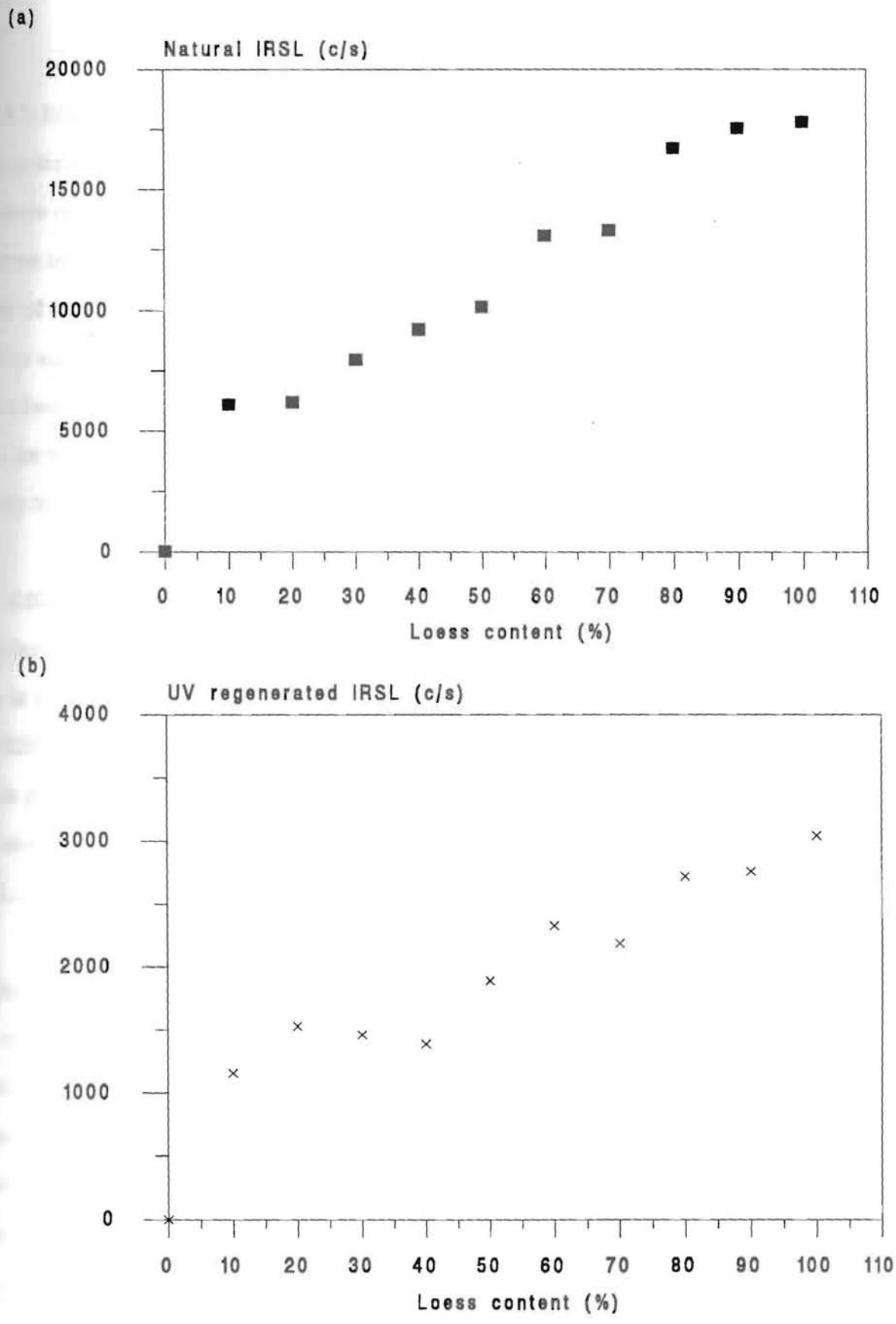


Fig. 4.4.3. IRSL signals of pellets of mixed loess and quartz crystals. (a) natural signal (b) u.v. regenerated signal after bleaching with visible light.

4.4.3. Effective depth and IRSL signal

It is generally thought that quartz has a better light transmission characteristics than most minerals over a wide range of wavelengths. Quartz can pass light in the near u.v. band whereas other minerals absorb u.v. strongly (Wintle and Duller 1991). Although the optical depth of IR (880nm) through individual minerals has not been studied specifically, optical studies suggest quartz also has high transmission at this wavelength. A study using an orthoclase feldspar chip (see below) suggests that the feldspar has a higher IR transmission than the wavelengths of the IRSL signal which, after passing through the BG39 filter, are mostly in the range of the near u.v. and visible light.

The different responses that occur with the quartz and carbonate diluting the signal can be explained by the effective depth involved in the natural and u.v. regenerated IRSL signals. This is shown schematically in figure 4.4.4. It is assumed that three types of grains affect the IRSL signal from the pellets: feldspars, carbonate and quartz. Feldspars are the minerals which give rise to the IRSL signal. The IRSL can pass through quartz but is absorbed by the carbonate. Carbonate can also be taken to represent other mineral grains which absorb the IRSL.

1. diluting with carbonate

The concentration of feldspar increases proportionately with the percentage of loess in the pellet, whereas carbonate is proportionally decreased with increased loess content. It is assumed that the IR can penetrate to a depth D (Fig.4.4.4). In the case of a high carbonate pellet (A), some of the IRSL from the grains at depth D would be absorbed by the carbonate grains before they could get through to the surface and be detected. The effective stimulation depth (d) is relatively shallow in this case, with only a few feldspar grains near the surface giving rise to the signal. In the case of high loess concentration (B), the quartz concentration also increases as loess content increases, because loess contains a high percentage of quartz (Pye 1988). More IRSL can pass through the quartz and thus get

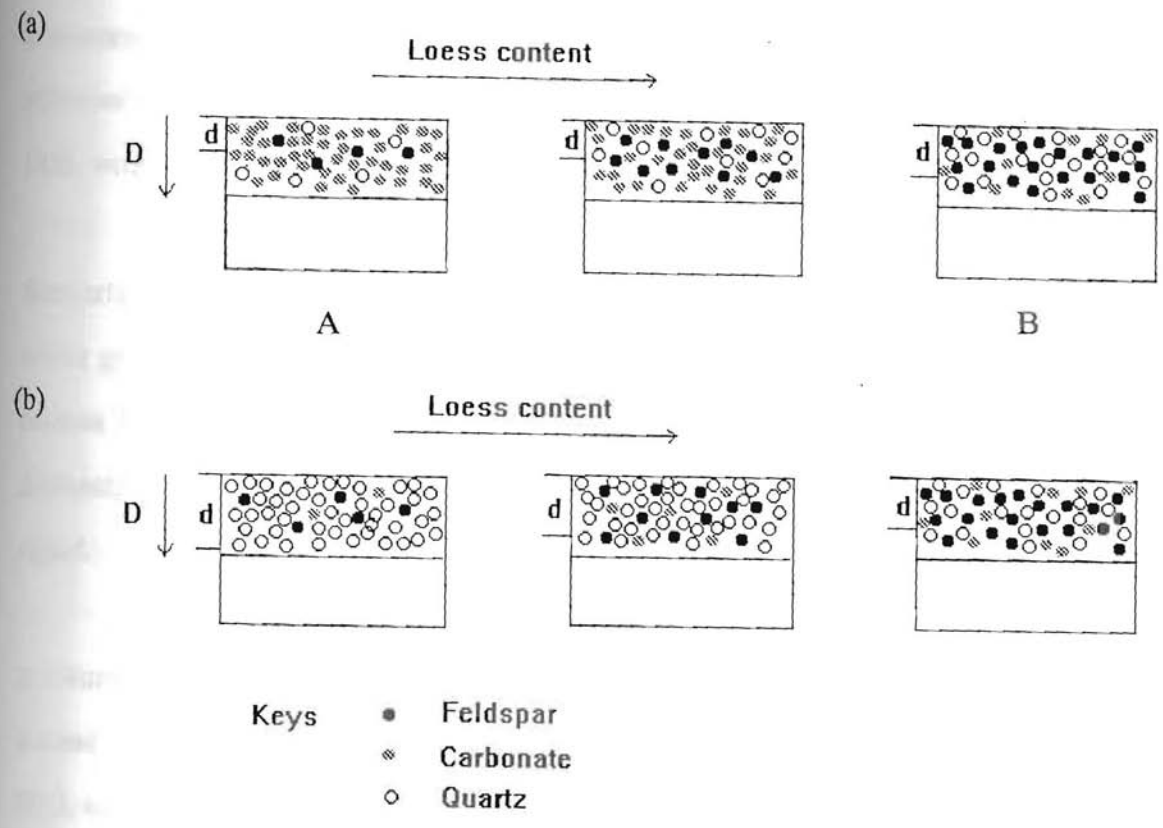


Figure 4.4.4. Schematic diagram for generation of IRSL signal from pellets. (a) loess mixed with carbonate (b) loess mixed with quartz.

D is the penetration depth of the IR.

d is the equivalent optical depth for the stimulated luminescence, and may be thought of as the effective stimulation depth.

through to the surface. The effective depth (d) is therefore greater. Thus increasing the loess concentration not only increases the IR sensitive material, feldspar, but also increases the effective depth (d) of IR stimulation. Hence a monotonic but non linear increase of the IRSL would be expected with the increase of loess content, as found in the previous section.

Similarly the u.v. regenerated IRSL signal can be affected by u.v. exposure. The depth to which grains can be reached by the u.v., may be shallower in the pellets of high carbonate content because of strong u.v. absorbtion by the carbonate. It is not clear which is the dominating factor, the absorbtion of u.v during u.v. regeneration or the absorbtion of IRSL signal.

2. Dilution with quartz

Instead of carbonate absorbing the IRSL, the higher quartz concentration allows most of IRSL to get through to the surface and results in a greater effective depth (d). Although the high quartz concentration means that there is less feldspar in the pellet, the increased sampling depth makes the IRSL decrease more slowly with the decrease in loess content. Hence the IRSL response to the loess content does not cross the origin of the axis (Fig. 4.4.3a). The u.v. regenerated IRSL signal behaves in a similar fashion to the natural IRSL signal (Fig. 4.4.3b).

4.4.4. Depth of IR and IRSL penetration through a feldspar chip

Although IR has been used as the stimulating light in optical dating for several years, the penetration depth in feldspar grains has not been studied directly. This experiment is designed to determine whether the IR can reach deeper into the crystal than the wavelengths of the stimulated luminescence.

1. sample used

The sample used in this experiment was a chip of orthoclase feldspar, approximately 700μm

thick and of an irregular but roughly circular shape. The sample was bleached in sunlight to reduce the IRSL signal to a negligible level, and it was then irradiated for an hour with a ^{244}Cm α source ($1.3/\mu m^2 min$).

2. IRSL measurements

IRSL measurements were carried out with the manual Daybreak system.

Because the alpha irradiation can only affect the chip to a depth of less than $20\mu m$, the IRSL signal would be detected if the IR could pass through the chip to reach the irradiated layer when it is placed away from the detector. The chip was placed in the disc holder (Fig 4.4.5). The IRSL was 46 kc/s when the alpha irradiated layer (a-layer) facing upwards and 22 kc/s when the chip was turned upside down. Because the measured IRSL signal involves both IR and the stimulated luminescence (IRSL) getting through the chip, it is not clear from this experiment which has the stronger absorbtion effect.

In the second experiment, the same orthoclase chip was placed between the quartz light guide and the PM tube (Fig 4.4.5). The IRSL was measured in two positions: a-layer up and a-layer down. The IRSL was 116 and 76 kc/s respectively.

From the above results, the relative absorbtion factors may be determined. It is hypothesized that the effect of absorbtion in the a-layer is negligible compared to that of the rest of the chip, and that the IRSL increases linearly with IR intensity. It is assumed that the IR intensity on the chip are ϕ_1 and ϕ_2 in the first and second experiment respectively as a result of the different positions relative to the light guide and the diodes; also, the absorbtion factors of the chip for IR and IRSL are X1 and X2. In the first experiment, when the a-layer was facing up (Fig. 4.4.5a), the IRSL can be written as $(IRSL)_a = f1 * \phi_1 = 46$, where f1 is a constant which is relevant to the efficiency of IRSL collection and IRSL sensitivity to the IR. Similarly, when the a-layer was facing away from the light guide (Fig. 4.4.5b),

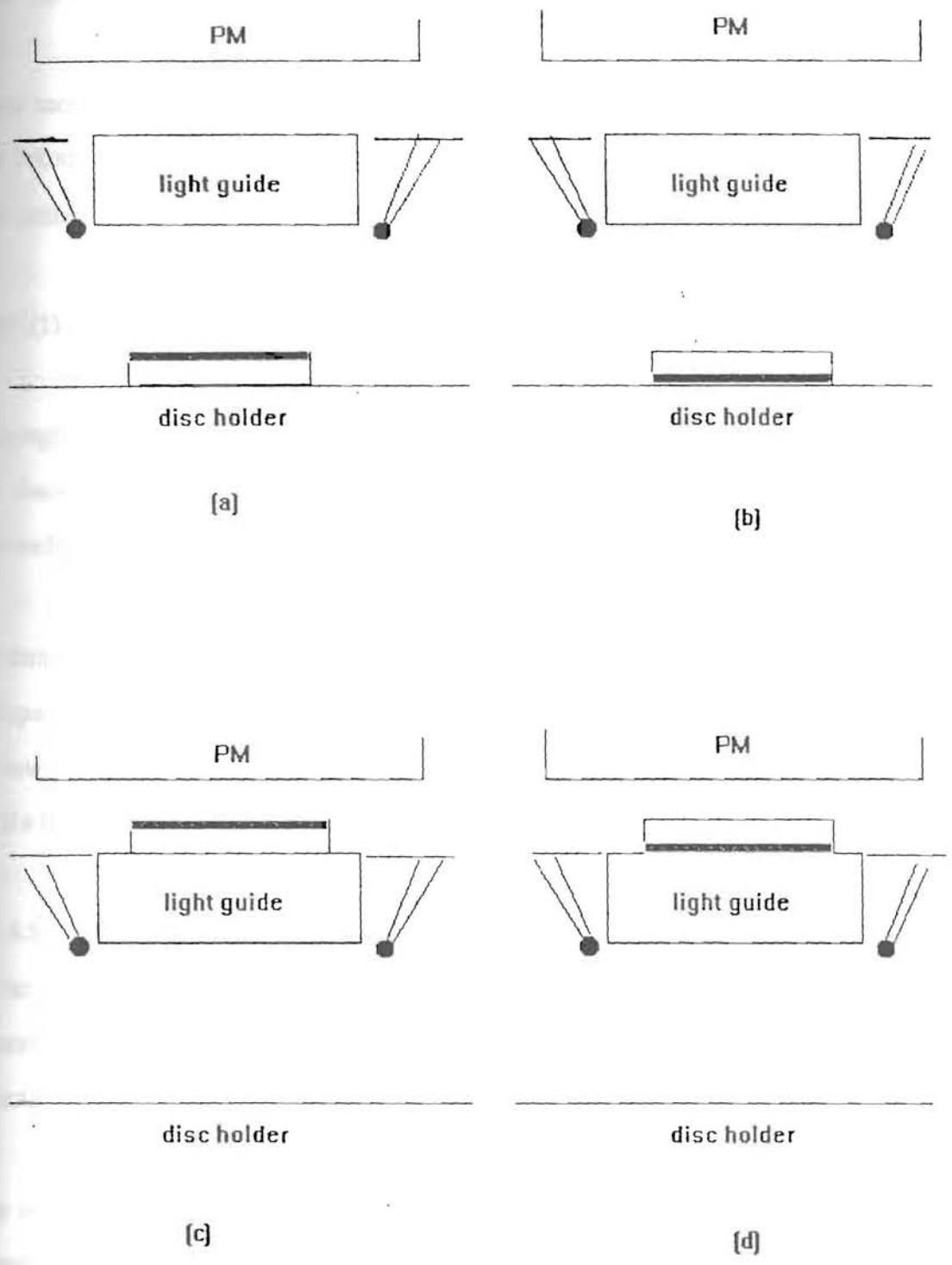


Fig. 4.4.5. Schematic diagram showing the positions of feldspar chip during measurements.

$(IRSL)_b = f1*X1*X2*\phi_1 = 22$. Hence,

$$(IRSL)_a/(IRSL)_b = 1/X1*X2 = 46/22 \qquad (1)$$

In the second experiment, when the a-layer was facing the PM tube (Fig. 4.4.5c), the IRSL has become $(IRSL)_c = f2*\phi_2*X1 = 116$. Similarly $(IRSL)_d = f2*\phi_2*X2 = 76$ when the a-layer was facing away from the PM tube (Fig. 4.4.5d). Hence,

$$(IRSL)_c/(IRSL)_d = X1/X2 = 116/76 \qquad (2)$$

From (1) and (2) we have

$$X1 = 0.85 \text{ and } X2 = 0.56$$

Although the absorbtion is likely to be more complicated than is expressed by the equations, the absorbtion factors give a relative result. They suggest that the absorbtion of IR is relatively less than the IRSL in the same feldspar chip.

A similar experiment was made for an approximately 300 μm thick piece of the same feldspar. There was no significant difference in the IRSL signal whether the a-layer was up or down. This suggests that both IRSL and IR have a negligible or similar absorbtion for such a thin chip.

4.4.5. Effective depth in pellet

In the study of loess pellets, the natural signal can be recovered after removal of the layer affected by bleaching and u.v. exposure. This enables an estimate to be made of the effective depth of light penetration through sediments.

The sample used was from the central part of the Rocourt section. Three pellets of the sample were studied. The natural signals of these pellets had been measured previously. Since the depth to be removed is small, it is not easy to measure it very precisely by means of callipers. However, since the pellets have a uniform cylindrical shape, the depth removed can be obtained from calculating the reduction in weight.

After bleaching in sunlight for 6 hours, the height and weight of the pellet were measured. The IRSL signal was close to the background level. After removing some of the grains by using sandpaper, the weight and the IRSL signal of the pellet were measured. Short shine (0.1 sec IR) was used in IRSL measurements and this was repeated several times for each pellet. Figure 4.4.6. shows the recovered IRSL versus calculated depth removed from three pellets. As can be seen, the IRSL signal can be recovered after removing the grains to a depth of approximately 0.6 mm. The removal of 2 mm from surface in sample preparation procedures therefore should be adequate.

The recovered IRSL signals are consistently (10-20%) less than the natural signal measured previously. This is explained by the bleaching of the signal on the sides of the pellets, which was not removed in the experiment. However, this does not affect the results obtained from previous measurements of loess pellets.

4.4.6. U.V. regenerated signal and effect of natural dose

From TL dating studies of the Rocourt loess section, it is known that the EDs of the samples will be different at the top and at the bottom of the section. If the charge transfer by the u.v. is from the traps which are not able to be stimulated by IR into those which are IR stimulated, and which were emptied by bleaching prior to loess deposition, the charge concentration in those traps would be different for samples from the top and the bottom of the section. If the signal is dominated by this charge transfer, a significant difference in the u.v. regenerated signal between these samples would be expected. However, there is no significant difference in the u.v. regenerated signal along the loess section (Fig. 4.4.7). This result suggests that there may be a significant contribution of u.v. excitation, which is independent of the natural dose level.

Another possibility is that the traps, from which the charges are transferred, had not been

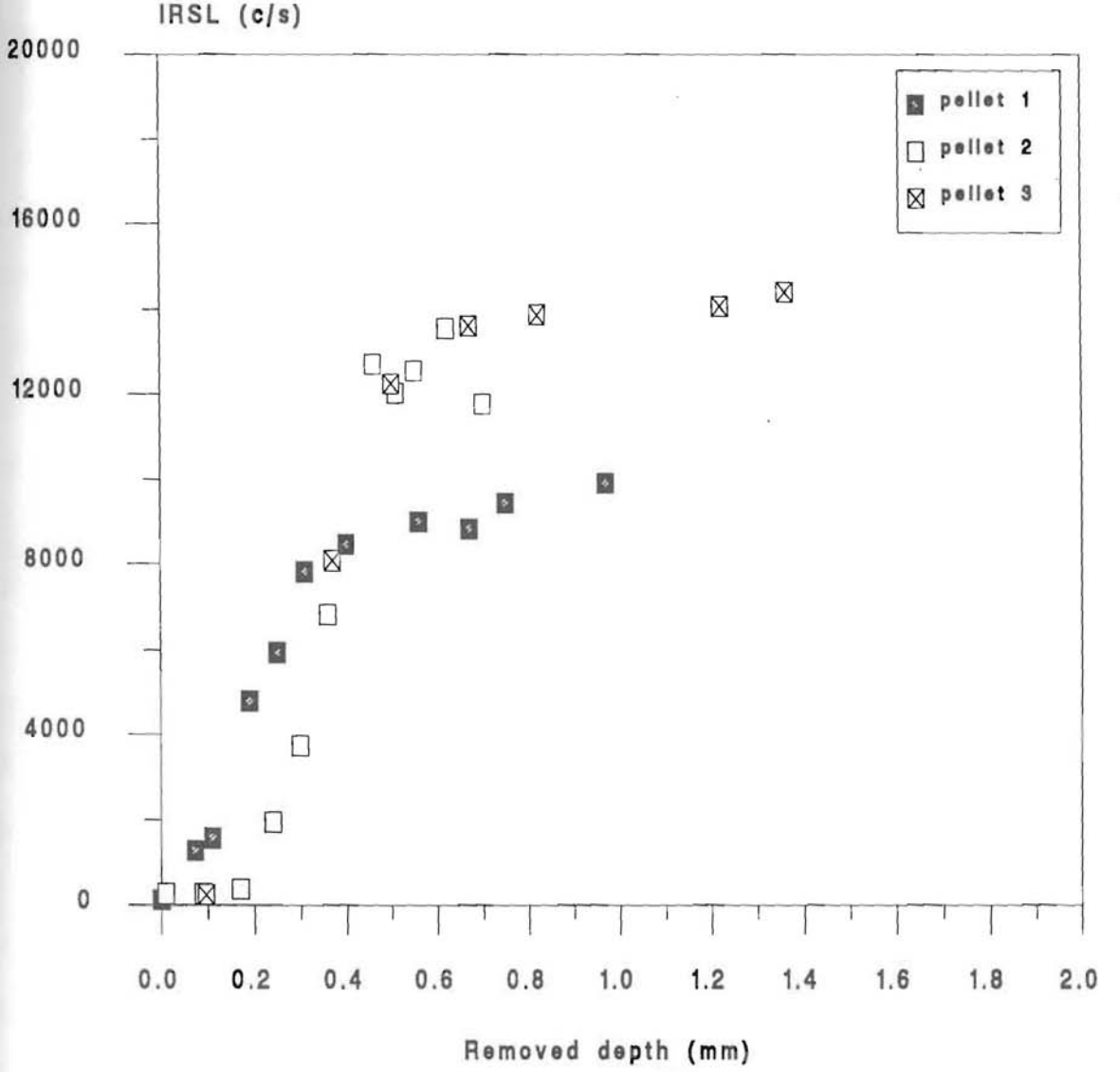


Fig. 4.4.6. Recovered IRSL signal as result of removing bleached grains.

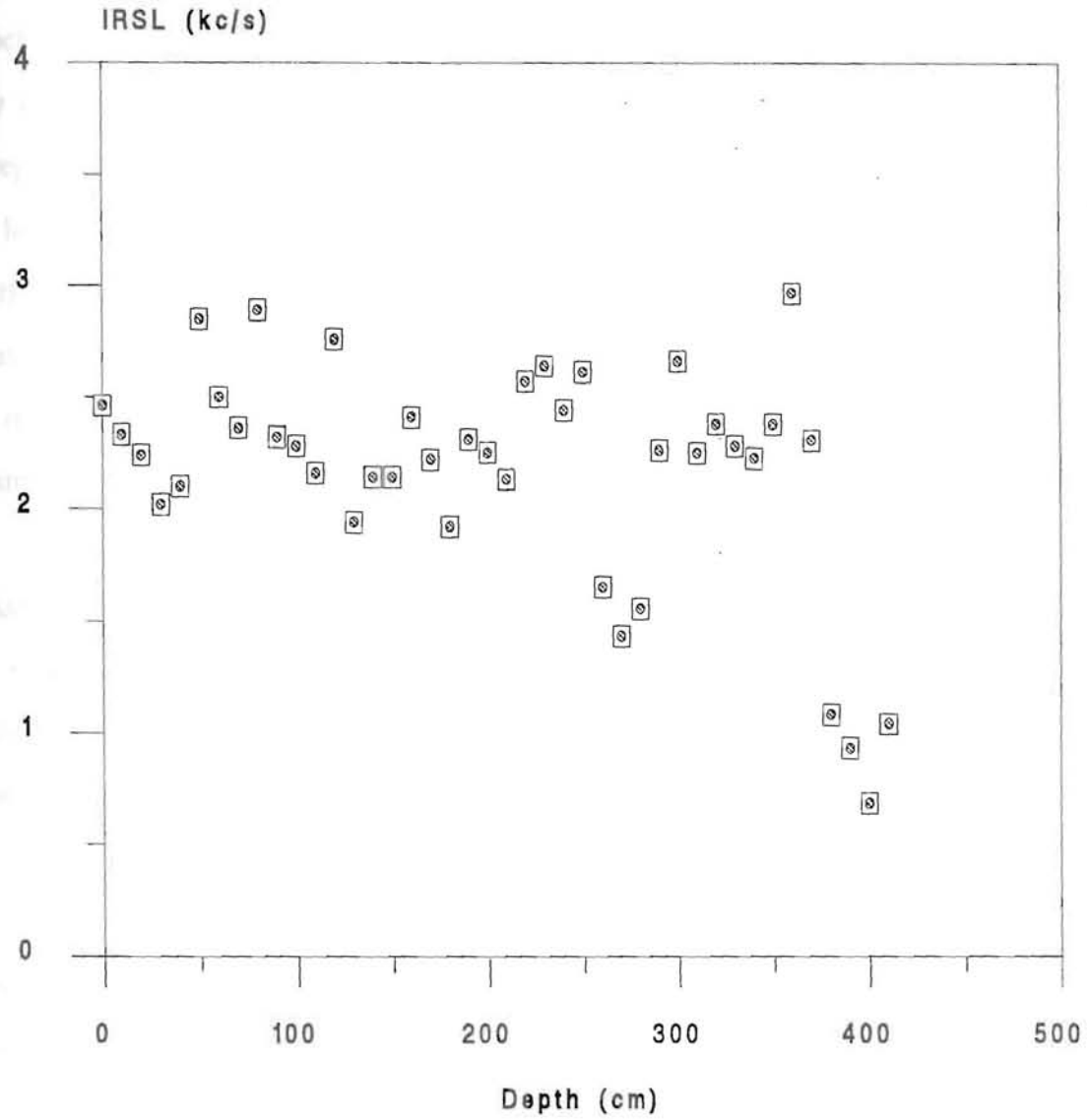


Figure 4.4.7. U.V. regenerated IRSL signal in Rocourt section.

emptied before deposition of the grains which make up of the loess. The palaeodose of these traps may be regarded as infinite and the traps considered to be saturated for all of the samples regardless of the dose since deposition. Since the wavelength of the u.v. radiation from the lamp (254 nm) is shorter than the shortest wavelength of sunlight reaching the earth's surface, these traps were not erased by the sunlight bleaching at deposition. Hence charge transfer can take place from deep traps which were not emptied before deposition. The u.v. regenerated signal probably results from a combination of u.v. excitation and charge transfer.

4.4.7. Assessment of u.v. normalization

The u.v. regenerated IRSL signal reflects the relative concentration of feldspar in the pellets. Since the same grains were sampled by the IR in the natural and u.v. regenerated IRSL signals, the absorbtion effects of the IR will not cause problems for the normalized signal.

The u.v. source can be as simple as a battery operated u.v. mineral lamp, thus allowing the experiment to be conducted near the field area.

Since only a limited number of feldspars have an u.v. regenerated IRSL signal (section 3.2.7), this normalization cannot be used if the mineral composition changes significantly down a section.

Using the manual Daybreak system, the u.v. regenerated IRSL signal for the Rocourt loess is about 200 times higher than the background level measured with a blank aluminium disc. For the lake sediment core from Morocco, the u.v. regenerated IRSL signal is comparable with the background; the signal is very low because this core has a high carbonate content. The regenerated signal may be also lower in other sediments, because of low feldspar concentration. The method is limited by the feldspar concentration.

4.4.8. Further information provided by the u.v. regenerated signal

The soil horizon shows a significantly lower (about 3 times) u.v. regenerated signal than the loess. The stronger weathering of the soil samples may cause a lower feldspar concentration and masking the IRSL by clay minerals. Therefore, the u.v. regenerated IRSL signal may provide information on weathering.

The diluting experiment showed that the higher the carbonate content is, the lower is the regenerated IRSL signal. Therefore, a relative carbonate concentration can be given by the signal. This was demonstrated in part of the Rocourt loess section, where the high $CaCO_3$ content gave a lower u.v. regenerated IRSL signal.

Because different types of feldspar have different responses to u.v. regeneration, the u.v. normalization cannot be used if the feldspar minerals composition changes through the section. The change in feldspar concentration may be the result of changes in the depositional environment or the source of the material. The change of the feldspar concentration will be reflected in the change of the u.v. regenerated IRSL, and hence the latter may provide useful information for sedimentologists.

Although limited studies were made on the u.v. regenerated signal for bulk sediment, important sedimentary information was demonstrated. It is possible that the implications and interpretation of the u.v. regenerated IRSL signal are more important than just as a normalization method.

chapter 5

LUMINESCENCE SENSITIVITY CHANGE DUE TO BLEACHING OF SEDIMENTS

5.1. INTRODUCTION

Sensitivity changes after laboratory bleaching have been reported in several luminescence dating studies. For aeolian sediments, e.g. loess and sand dune samples, the sensitivity of the luminescence signal (TL and OSL) was found to increase after laboratory bleaching (Rendell *et al.* 1983, Smith *et al.* 1990). For TL measurements this sensitivity increase was significant for old samples but negligible for young samples (Rendell and Townsend 1988, Zhou and Wintle 1989). A simple model which hypothesised that the sensitivity increase was dose-dependent was put forward by Wintle (1985), but Bowell *et al.* (1987) reported a sensitivity change which depended upon the bleaching time.

In a recent paper McKeever (1991) has attempted to model the effects of optical bleaching of quartz TL peaks. It was claimed that the bleaching process alters the distribution of trapped charge and that because several variables are involved (past irradiation, duration of bleaching and the wavelength of illumination) it is not possible to predict whether the sensitivity will increase or decrease after bleaching.

In this chapter, I will examine the implications of sensitivity changes resulting from different illumination histories. A competition model to explain and predict the sensitivity change will be introduced. The effects of the natural light exposure on subsequent ED determinations will be discussed.

5.2. SENSITIVITY CHANGES AFTER DIFFERENT BLEACHING PROCEDURES

5.2.1. Samples used and experimental details

For this study I chose to work on colluvial deposits, the grains of which were unlikely to have been exposed to light for a long time before being covered up by the arrival of further colluvium. Several reasons influenced my choice; the major ones were

a) Since sensitivity increases had already been reported for aeolian sediments, it was of interest to know whether this behaviour was universal, irrespective of the depositional environment, or related to the extent of sunlight exposure at deposition.

b) Colluvial deposits are an important category of sediment which, prior to this study, have not been considered as a material for luminescence dating applications. The samples studied in this thesis are of particular importance as they are from sections which show evidence of past climate changes. The ability to date the sections would enable a range of palaeoclimatic information (e.g. rainfall) and palaeoenvironmental information (e.g. erosion rates) to be placed in a chronological framwork.

The colluvial deposits were from Natal, South Africa (Botha *et al.* 1990). These sheet wash deposits lie on a low slope and the transport distances from source are of the order of 1 km. Four samples (AF-1, AF-2, AF-3 and AF-4) were from the St Paul's section (see chapter 7). Four groups of fine grain discs were prepared for ED determination by additive dose and regeneration methods.

Different bleaching treatments were applied to three of the groups before using the regeneration method to obtain the ED

(a) infrared bleaching for 1600 seconds (~0.5 hour) by the Riso OSL/TL system. The infrared power at the disc is about 40 mW/cm^2

(b) bleaching for 1600 seconds by the SOL2 solar simulator.

(c) bleaching for 15 hours by the SOL2.

After each bleaching treatment, the IRSL was found to have been reduced to a negligible level, close to the background level measured for a blank disc.

Discs were irradiated with the Daybreak ß irradiator. To remove the unstable component in the IRSL signal, all discs were preheated for 16 hours at 140°C (section 3.5.7). There was at least 24 hours delay after irradiation and after preheating.

The IRSL measurements were carried out using the manual Daybreak system. The IR wavelength used for stimulation was the same as that used for the IR bleaching in the Riso OSL/TL system. The filter used was a 2 mm thick Schott BG 39. An integrated photon count for 0.1 second (within a 0.12 second IR exposure) was used. This gives a negligible reduction of the TL signal. Subsequent TL measurements were carried out in the Riso reader with two optical filters, one Corning 7-59 and one HA3. The heating rate was 5°C/s up to 450°C.

Growth curves were constructed for both regeneration and additive dose methods. Because the IRSL response was not linear for the three older samples (AF-1, AF-2 and AF-3), an exponential curve fitting programme was used.

5.2.2. Sensitivity changes after bleaching

Typical IRSL growth curves obtained after the different bleaching procedures are shown in Figure 5.2.1. Each data point represents the mean of 4 measurements. The initial slope (Xo) of the growth curve represents the sensitivity of IRSL after bleaching, and was calculated with the parameters from the fitting curves.

For

$$IRSL=k-a*\exp(-b*D)$$

D is the dose. k, a and b are the fitting parameters. The initial slopes were given by

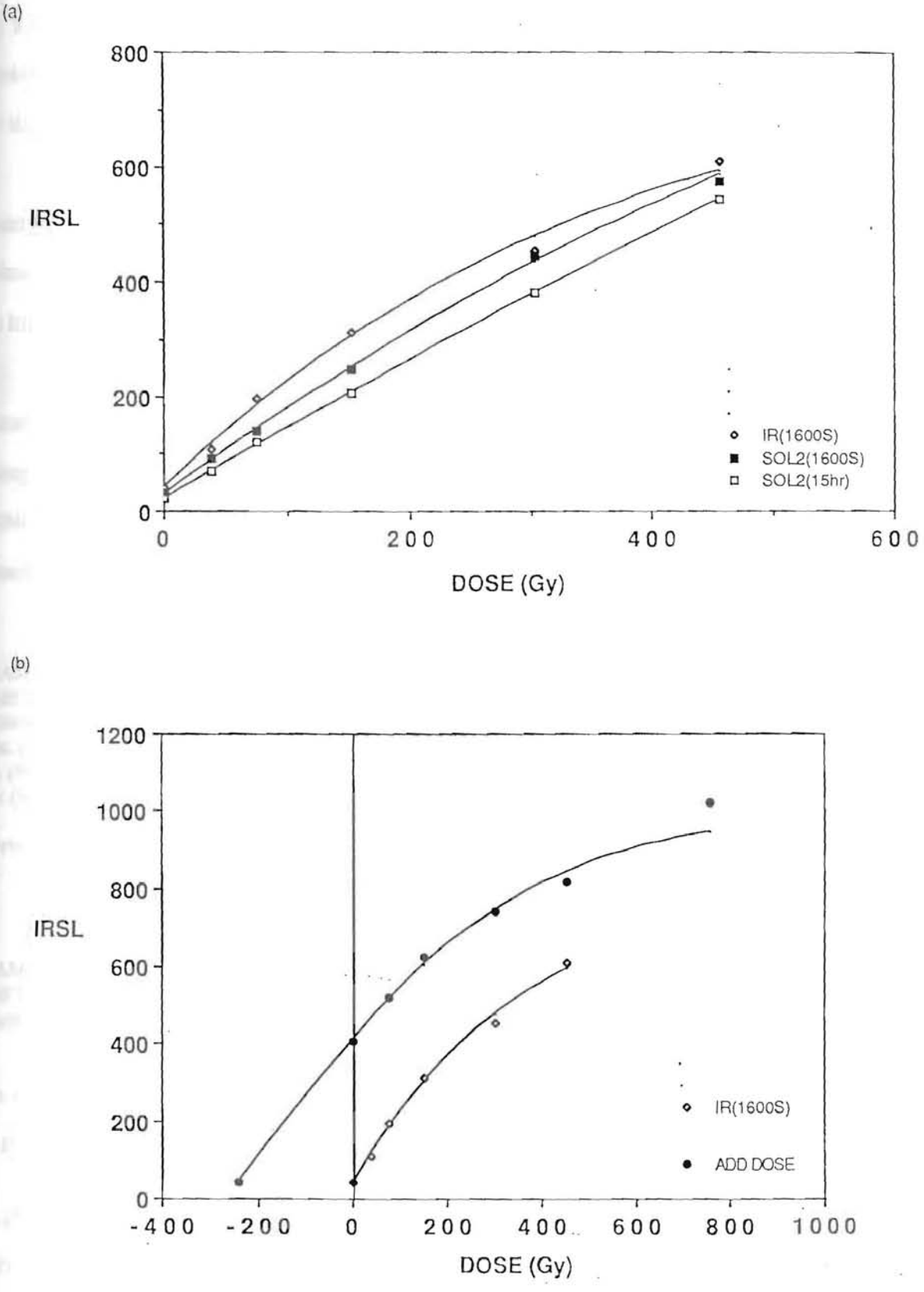


Figure 5.2.1. Growth curves for sample AF-1. a) Regeneration after exposure to IR and SOL2. b) additive dose and regeneration after IR bleaching.

$$Xo = a*b*\exp(-b*ED) \quad \text{and} \quad Xo = a*b \quad (ED=0)$$

for the additive dose method and the regeneration method respectively. The results are summarised in Figure 5.2.2a in which the sensitivity for the additive dose method is plotted against the sensitivity after bleaching for the three different treatments.

The sensitivity of the IRSL signal for the 3 older samples (AF-1, AF-2 and AF-3) decreases with bleaching time when using the SOL2 solar simulator. A short bleach (1600s) with IR gives a higher sensitivity than for either SOL2 bleach (Fig. 5.2.1a and 5.2.2a).

Significantly different EDs for the regeneration method were obtained using these different bleaching procedures (Table 5.2.1 and Fig.5.2.2b). In each case the IRSL was bleached to a negligible level before regeneration, and hence the ED differences cannot be explained by the presence of different residual levels.

Table 5.2.1a. Sensitivity of the IRSL signal

SAMPLES METHOD	AF-4	AF-3	AF-2	AF-1
Additive Dose	8.70	15.72	3.70	9.32
Regen. (IR bleach)	8.07	15.28	3.75	8.47
Regen. (SOL2 1600s)	----	12.01	2.82	6.20
Regen. (SOL2 15 hr)	6.48	10.37	2.39	4.77

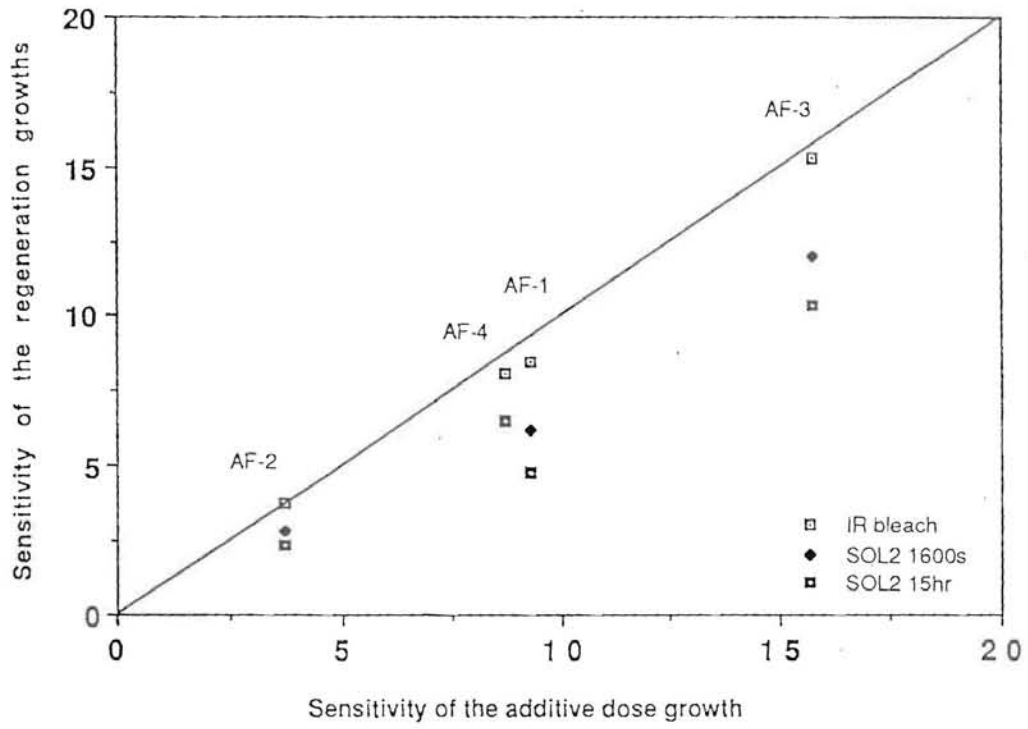
Note: Arbitrary units are used for sensitivity; the numbers are only meaningful when compared for the same sample.

Table 5.2.1b. Equivalent dose (Gy) given by IRSL and TL methods

SAMPLES METHOD		AF-4	AF-3	AF-2	AF-1
Add.Dose	IRSL ED	30±1	233±10	248±21	240±22
	TL ED	90±3	284±15	319±14	424±53
Regen. (IR bleach)	IRSL ED	23±3	212±6	243±23	238±23
Regen. (SOL2 1600s)	IRSL ED	----	245±14	297±25	277±23
	TL ED	----	218±3	291±38	295±30
Regen. (SOL2 15 hr)	IRSL ED	35±4	293±11	371±32	328±23
	TL ED	104±1	342±20	453±53	451±42

Note: TL residual after 15 hours SOL2 bleaching was used in the ED calculation of the additive dose method.

(a)



(b)

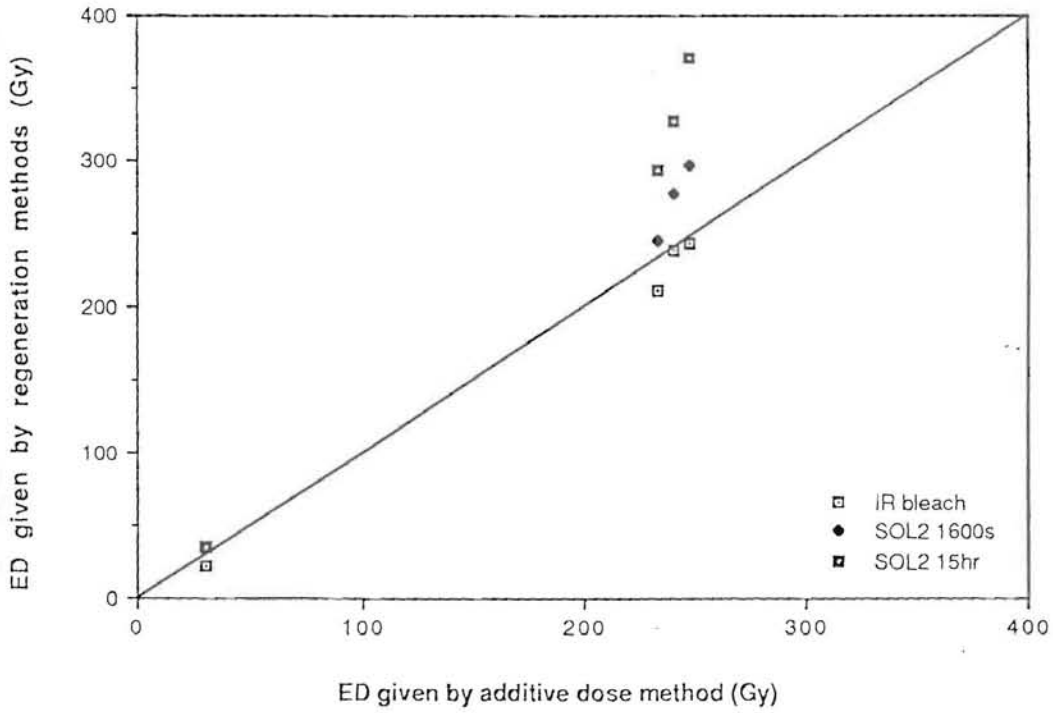


Figure 5.2.2. a) Sensitivity of initial IRSL signal measured after bleaching versus the initial sensitivity as obtained by extrapolation for the additive growth data sets. b) IRSL EDs obtained by the regeneration method using different bleachings versus that obtained by the additive dose method for 4 colluvial sediments. The line has a slope of 1.

A comparison of the regenerated IRSL ED values with those given by the additive dose method for the remaining group of discs suggests that only the ED determined after IR bleaching is similar to the additive dose ED for all four samples (Fig.5.2.2b; Table 5.2.1).

The ED values given by routine TL measurement were higher than those given by any of the IRSL measurements (Table 5.2.1b). This difference is most clearly demonstrated for the youngest sample AF-4, for which the TL ED (90±3 or 104±1 Gy) was three times larger than the IRSL ED (30±1, 23±3 or 35±4 Gy). Since the IRSL signal is more easily bleached by sunlight than the TL signal, it is suggested that this colluvial sediment was not well exposed *prior to* deposition.

Previous studies on aeolian sediments suggest that the sensitivities of TL signals (Wintle 1985; Rendell and Townsend 1988) and OSL signals (Smith *et al.* 1990) change after laboratory bleaching, with the sensitivity increase being negligible for young samples but significant for old samples (Zhou and Wintle 1989). These findings contrast with those observed in this study where the IRSL sensitivity decreased rather than increased after bleaching with the SOL2. Not only was this decrease in sensitivity found for older samples, but also for the young sample AF-4. The general difference between aeolian and colluvial sediments is the degree of sunlight exposure prior to deposition. Given the results of the above bleaching experiments, the nature of the exposure before deposition could alter the post-depositional IRSL sensitivity of the sample. The extent to which the sensitivity is affected by the predepositional bleaching can be used to give information on whether the bleaching was short or prolonged. This will discussed in chapter 6.

5.2.3. Summary

Significant differences in bleaching response were found between aeolian and colluvial deposits. For the colluvial deposits used in this study, the sensitivity of IRSL signal decreased after bleaching with SOL2 solar simulator regardless of the age of the sample.

The choice of bleaching time and wavelengths to reduce the IRSL signal to a negligible level affects the subsequent response to laboratory dose and would lead to significantly different EDs being obtained.

Comparison of the additive dose ED and EDs obtained by regeneration, suggests that IR bleaching should be used for the regeneration method.

That laboratory light exposures can affect IRSL sensitivity suggests that the extent of light exposure prior to deposition will have an effect on subsequent ED determinations.

5.3 A COMPETITION MODEL

5.3.1. Introduction

The differences in bleaching response for aeolian and colluvial samples could be due either to the differences between the predeposition bleaching experienced by the sediment grain or to difference in mineralogy. In the next section (5.4) the effect of bleaching is shown to be dominant. In this section the effects of bleaching are explored and a competition model will be introduced to explain and predict the sensitivity change after laboratory bleaching.

In order to emphasise the bleaching effects, this model will be confined to the luminescence signals which are easily bleached by sunlight. These include IRSL and OSL signals, and part of the TL signal.

5.3.2. Two types of trapped charges and luminescence sensitivity

In this section we will consider two types of trapped charges: E-type and H-type. It was hypothesized that the E-type is made up of **easy to bleach trapped charges**, i.e. those that are bleached easily by sunlight and correspond to the luminescence signal measured. This is

only part of the bleachable TL signal, I_d, as defined by Wintle and Huntley (1980). The growth curves used in optically stimulated luminescence ED determinations represent the dose responses of this type of trapped charge.

The other type, the H-type, is made up of **hard to bleach trapped charges**, i.e. those that are hard to bleach, but not non-bleached by sunlight and make up the rest of the I_d signal of Wintle and Huntley (1980). They can be bleached by prolonged sunlight exposure. The concentration of H-type charges increases with dose, but they do not contribute to the growth curves of the optically stimulated luminescence signals used for dating.

Both types of trapped charges build up with dose toward a maximum level.

In this competition model, it is hypothesised that there is competition between both types of traps during irradiation. The sensitivity of the luminescence signal (X) is related to the concentration of H-type charges (I). The higher the H-type charge concentration is, the higher is the sensitivity of the luminescence signal. Hence, the sensitivity, X, can be empirically described as

$$X = \eta * I + X_0 \qquad (1)$$

where η and X_0 are constants relating to the competition and the minimum dose response respectively.

5.3.3. Sensitivity changes

Sensitivity change was observed experimentally by comparing the additive dose growth curve with the regeneration growth curve. The sensitivity may be expressed by the initial slopes of both curves, X_A and X_R (Fig.5.3.1), and relate to the responses after deposition (A) and after laboratory bleaching(C) respectively. (B) represents the natural sample.

The sensitivity of the additive dose growth curve X_A relates to the concentration of H-type

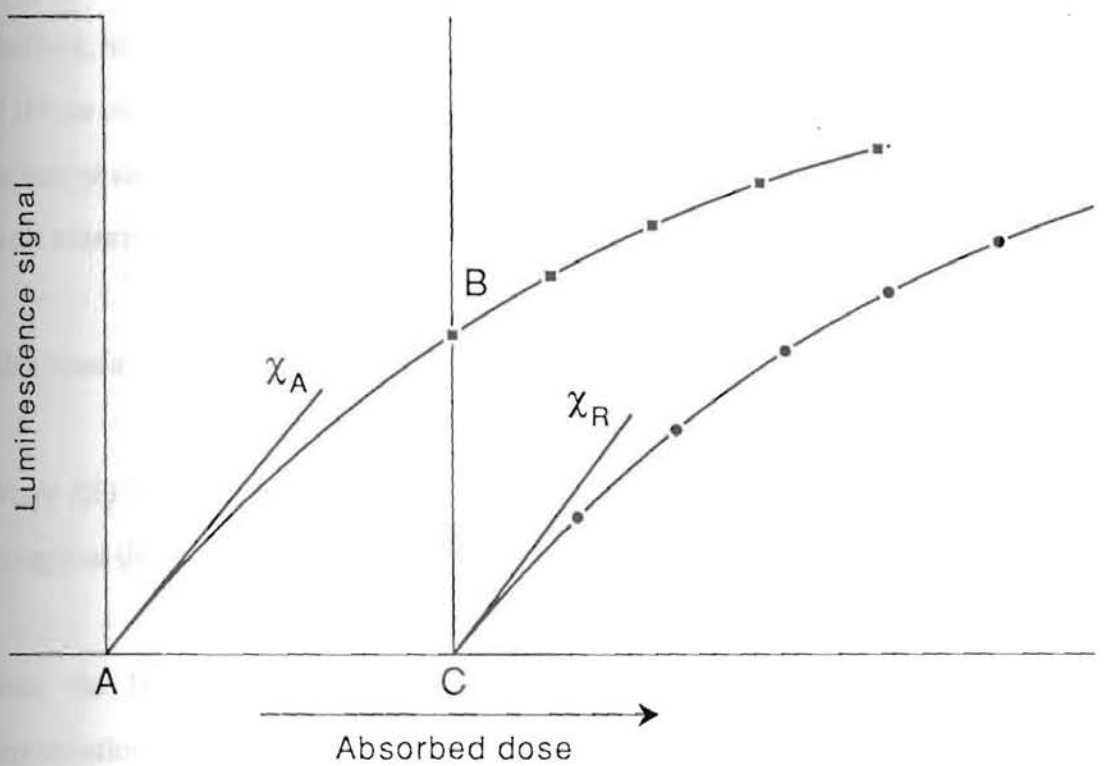


Figure 5.3.1. Schematic diagram of luminescence signal growth curves: the response to added dose (■) and response to dose after laboratory bleaching (●).

charges after the sample was last exposed to sunlight, I_a (Fig.5.3.2). Hence, equation (1) can be written as

$$X_A=\eta^*I_a + X_O \qquad (2)$$

and X_A is thus a function of the degree of sunlight exposure prior to deposition.

Similarly, the sensitivity of the regeneration growth curve X_R is related to the concentration of H-type charges after laboratory bleaching, I_c. This represents a portion of the charges in the natural sample, I_b (Fig. 5.3.2), which increased as a function of dose, F(D), and is added to the concentration I_a. Hence,

$$I_b= F(D) + I_a \qquad (3)$$

After laboratory bleaching

$$I_c = f(S)^*I_b= f(S)^*F(D) + f(S)^*I_a \qquad (4)$$

Where f(S) is a bleaching factor, which depends on the bleaching time, spectrum and the strength of the bleaching light.

Since the H-type charges can be bleached by prolonged sunlight exposure, their concentration will decrease with bleaching time. Hence, f(S) will be smaller for longer bleaching. The spectrum of light used in laboratory bleaching is likely to be either similar to sunlight, e.g. solar simulator, or a relatively narrow wavelength band. In the latter case, a particular light may or may not reduce the H-type charges when the E-type charges are removed by the light exposure. The model assumes that there is no charge migration into the H-type traps during bleaching. Therefore f(S) can be defined in the range of $1 \geq f(S) \geq 0$.

Hence, substituting (4) into (1), the sensitivity of the regeneration curve is,

$$X_R = \eta^*I_c + X_O = \eta^*f(S)^*F(D) + \eta^*f(S)^*I_a + X_O \qquad (5)$$

Comparing equations (2) and (5), the sensitivity change after laboratory bleaching is given as

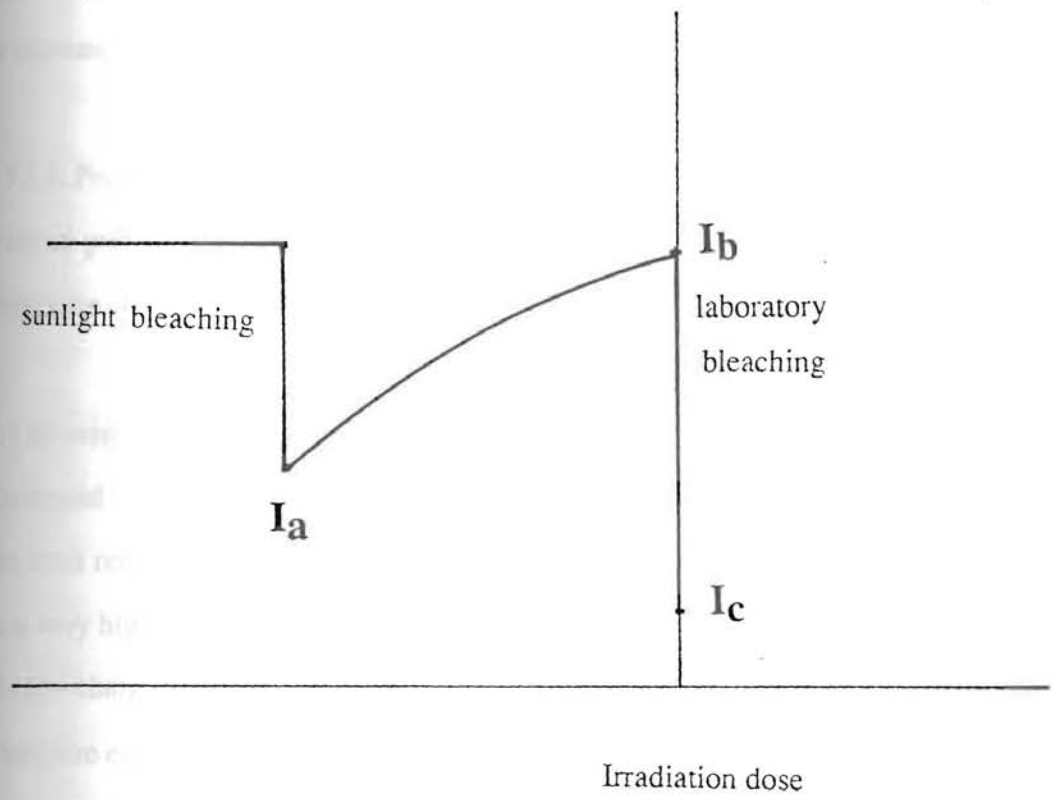


Figure. 5.3.2. Schematic diagram showing the change in concentration of H type (the hard to bleach) trapped charges as a result of natural and laboratory bleaching procedures.

$$X_R - X_A = \eta^* f(S)^* F(D) + \eta^* f(S)^* I_a - \eta^* I_a \quad (6)$$

The sensitivity change is thus related to the age of the sample F(D), laboratory bleaching f(S) and the bleaching prior to deposition as represented by I_a. The sensitivity can increase or decrease depending upon those three conditions.

5.3.4. Predictions of the model

Various predictions can be made from this model for the response of different types of sediments.

1. Colluvial sediments

Compared with aeolian sediments, H-type charges in grains from colluvial sediments have not been removed at deposition, because of the short sunlight exposure. The concentration Ia is very high, and may even be close to saturation, $I_a = I_{max}$. In all colluvial sediments the H-type charge is likely to be close to its maximum value and hence $F(D) \approx 0$, and $I_b = I_a$. Therefore equation (6) becomes

$$X_R - X_A = \eta^* I_a^* [f(S)-1]$$

Because $1 \geq f(S) \geq 0$ by definition, a sensitivity decrease will be observed after laboratory bleaching which releases H-type charges. The longer the bleaching, the greater sensitivity decrease expected.

One exception to this behaviour occurs when bleaching of the stimulated luminescence signal is carried out using the stimulating wavelength, e.g. bleaching of the IRSL by IR. The H-type traps remain full because this wavelength will remove only the E-type charges and will not affect the population of H-type charges, i.e. f(S)=1. This can be seen from the TL signal which remains after the optically stimulated luminescence is reduced to <1% by 1000 seconds of IR exposure (Li and Aitken 1989, Duller 1992). In this case the E-type traps will fill under the same competing regime as in nature and sensitivity of the signal response to dose when regenerated will be the same for any age sample.

2. Aeolian sediments

Since aeolian sediments were well exposed to sunlight at deposition, H-type traps were empty at deposition, I_a=0. Hence, equation (6) becomes

$$X_R - X_A = \eta * f(S) * F(D)$$

Since F(D) is positive $X_R - X_A \geq 0$ and hence the sensitivity will always increase after laboratory bleaching, unless a prolonged bleaching is applied or a zero age sample is studied (F(D)=0). The degree of increase is dependent upon f(S) and F(D), which relate to the degree of laboratory bleaching and the natural irradiation of the sample respectively. After a short bleach, i.e. fixed f(S), a sensitivity increase would be expected for different age samples; the older the sample, the greater the sensitivity increase expected after the same short bleach. However, for a young sample, F(D) is relatively small and may be negligible; in this case no significant sensitivity change should be observed after any laboratory bleaching. For samples of the same age, i.e. fixed F(D), the sensitivity change will decrease with increased light exposure. This model also predicts superlinear behaviour. However this is not observed and thus it is assumed that bleaching is necessary to activate the effect of H-type charges.

5.4 TESTING THE MODEL WITH EXPERIMENTS

5.4.1. Design of experiment.

Two sets of sample discs were prepared, one of which was well-exposed to light from a solar simulator (SOL2) to simulate an aeolian deposit, and the other of which was given a much shorter exposure to light source to simulate colluvium at deposition. Measurements of their IRSL and TL response after differing irradiation and bleaching treatments were then made.

All together eight sub-groups were taken from the 192 discs prepared as described in the following section. Three sub-groups (Fig.5.4.1) were exposed to the solar simulator for a relatively short time (0.5 hour) and these form the "artificial colluvial sediment". One of

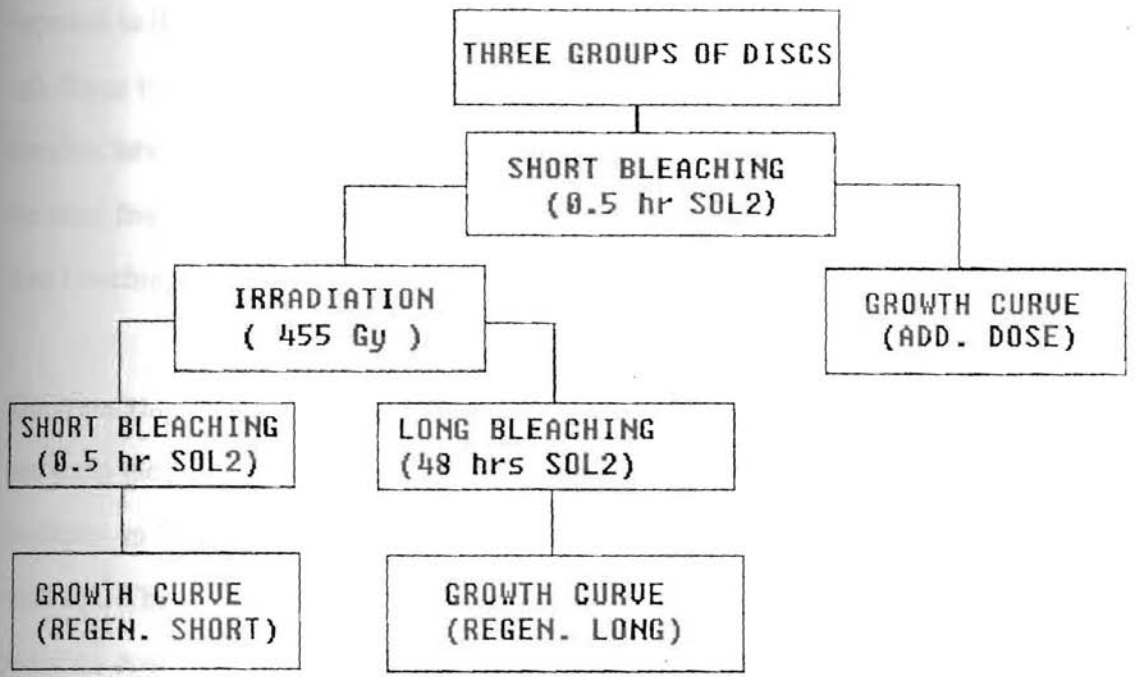


Figure 5.4.1. Experimental sequence for the "artificial colluvial sediment"

these sub-groups was given 5 additional irradiation doses to construct an "additive growth curve" and thus simulated the increase in luminescence signal with age. The other two sub-groups of "artificial colluvium" were given a single radiation dose of 455 Gy (120 minutes exposure to the beta source) and thus simulated a colluvium of a particular age (around 100 ka). These two groups were then given different light exposures (0.5 hour and 48 hours) to simulate laboratory bleaching during typical dating procedures. The discs were then given the same five irradiation doses to produce two "regeneration growth curves", after short and long bleaching respectively.

The remaining five sub-groups (Fig.5.4.2) were exposed to the solar simulator for 48 hours and form the "artificial aeolian sediment". One sub-group was given 5 additional doses to construct an "additive growth curve" and thus simulated the increase in luminescence signal with age. The remaining four sub-groups were divided into two groups which were given radiation doses of 76 Gy (20 minutes) and 455 Gy (120 minutes) and thus simulated aeolian sediment of two different ages ("young" and "old" respectively). For each of those groups, one sub-group was given a short light exposure (0.5 hour) and the other a long light exposure (48 hours) before a "regeneration growth curve" was constructed for each, as already described for the "artificial colluvial sediment".

5.4.2. Experimental conditions

The sample used in this experiment was a 700 ka old loess (88-18) from Luochuan, Shaanxi, China. The luminescence signal of this sample had reached an equilibrium level, and hence it could be treated as typical source material. Fine grains (4-11μm) discs were prepared. For all natural discs, the IRSL from a 0.1 second infrared exposure was used for normalization.

The SOL2 was used for the bleaching experiments. To ensure the bleaching was the same for all the groups, discs were placed on an aluminium plate and bleached together for each

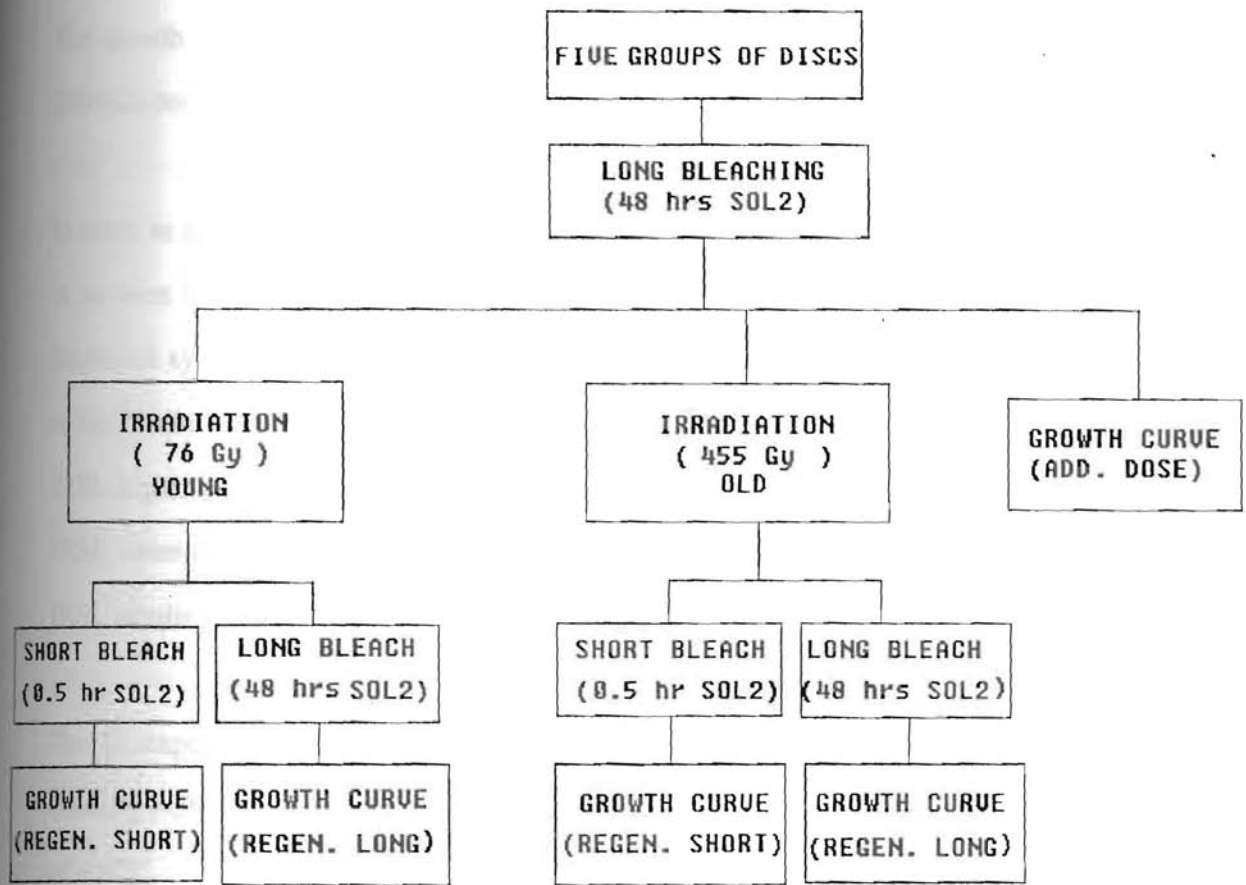


Figure. 5.4.2. Experimental sequence for the "artificial aeolian sediment"

of the long and short bleaching times.

The growth curves were constructed with the same applied ß doses, 0; 20; 40; 80; 120 and 200 minutes. The dose rate was 3.79 Gy/min.

In order to remove the thermally unstable IRSL components, discs were preheated together in an oven for 16 hours at 140°C. The IRSL measurements were performed on the manual Daybreak system. The filter used was one 2 mm thick BG39. A short infrared exposure (1 second) was used for the IRSL measurements, which resulted in a 4.5% reduction of the IRSL signal and a negligible reduction of the TL signal. Therefore, the discs used in the IRSL measurements could also be used for the TL measurements, allowing the TL and IRSL results to be compared directly.

The TL experiments were performed in the Riso TL reader. The optical filters used were a Corning 7-59 and a Chance Pilkington HA3. The heating rate was 5°C/s up to a temperature of 450°C.

For the dose range used, the IRSL and TL signals showed non-linear growth with dose and a third order polynomial fitting was used. The sensitivity of the luminescence signals after bleaching was given by the initial slope of the growth curve.

To minimize the effect due to different experimental conditions, discs were treated together at the same time for bleaching and preheating. The same irradiation doses were employed for all the growth curves. There was at least 24 hours delay between each experimental procedure.

5.4.3. Sensitivity change results

The results of the IRSL and TL measurements carried out on the same discs are summarized

in Figures. 5.4.3 and 5.4.4 and Table 5.4.1. Each data point is the mean of the measurements for four discs. In Figures. 5.4.4a,b,c. and in Table 1, the TL signal is taken as the integral of the TL signal in the region between 280-380°C. To ensure more direct comparison of the data sets, the additive dose curve has been shifted along the dose axis by the appropriate dose (20 or 120 mins). Sensitivity changes greater than 5% are taken to be statistically significant.

IRSL

1. Colluvial sediment.

Compared with the "additive dose" growth curve, the "regeneration" growth curve for the "artificial colluvial sediment" showed a significant sensitivity decrease (-27.0%) after the long bleach (Fig. 5.4.3a, Table 5.4.1), but a negligible sensitivity change (-2.1%) for the short bleach.

Table 5.4.1. Sensitivity change and equivalent doses

Groups	colluvium		aeolian (young)		aeolian (old)	
expected ED(mins)	(120)		(20)		(120)	
Bleach	Short	Long	Short	Long	Short	Long
IRSL Sensitivity change	-2.1%	-27.0%	+0.8%	+4.6%	+18.5%	-4.6%
Predicted by model	—	↓	—	—	↑	—
ED (IRSL) (mins)	119	138	16.9	19.5	115	128
ED (TL) (mins)	123	>200	15.7	23.7	103	164

2. Aeolian sediments.

"Young sample". No significant sensitivity changes were found in the young sample after

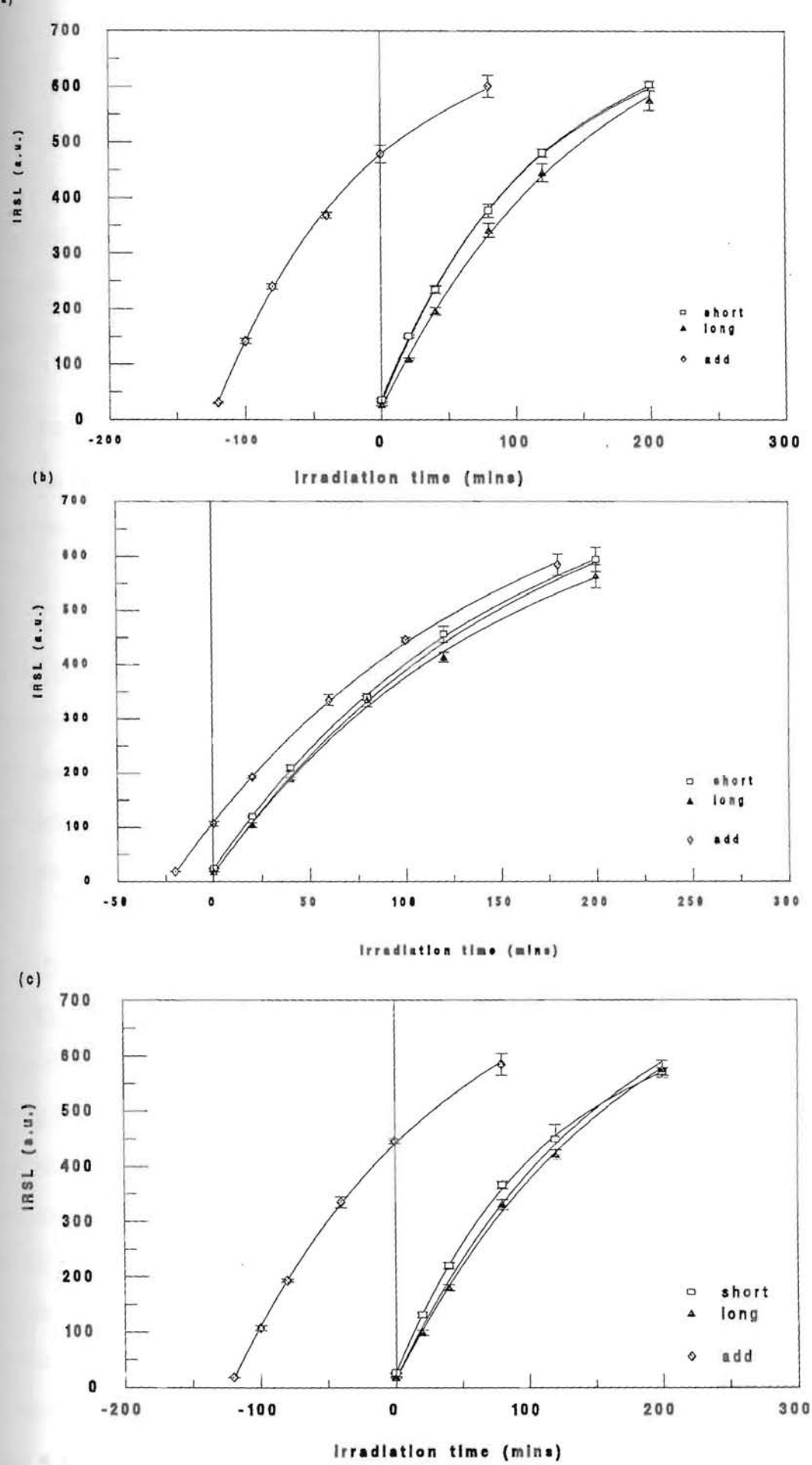


Figure 5.4.3. IRSL growth curves. (a) Colluvial sediment (b) young aeolian sediment (c) old acolian sediment. The dose rate was 3.79 Gy/min.

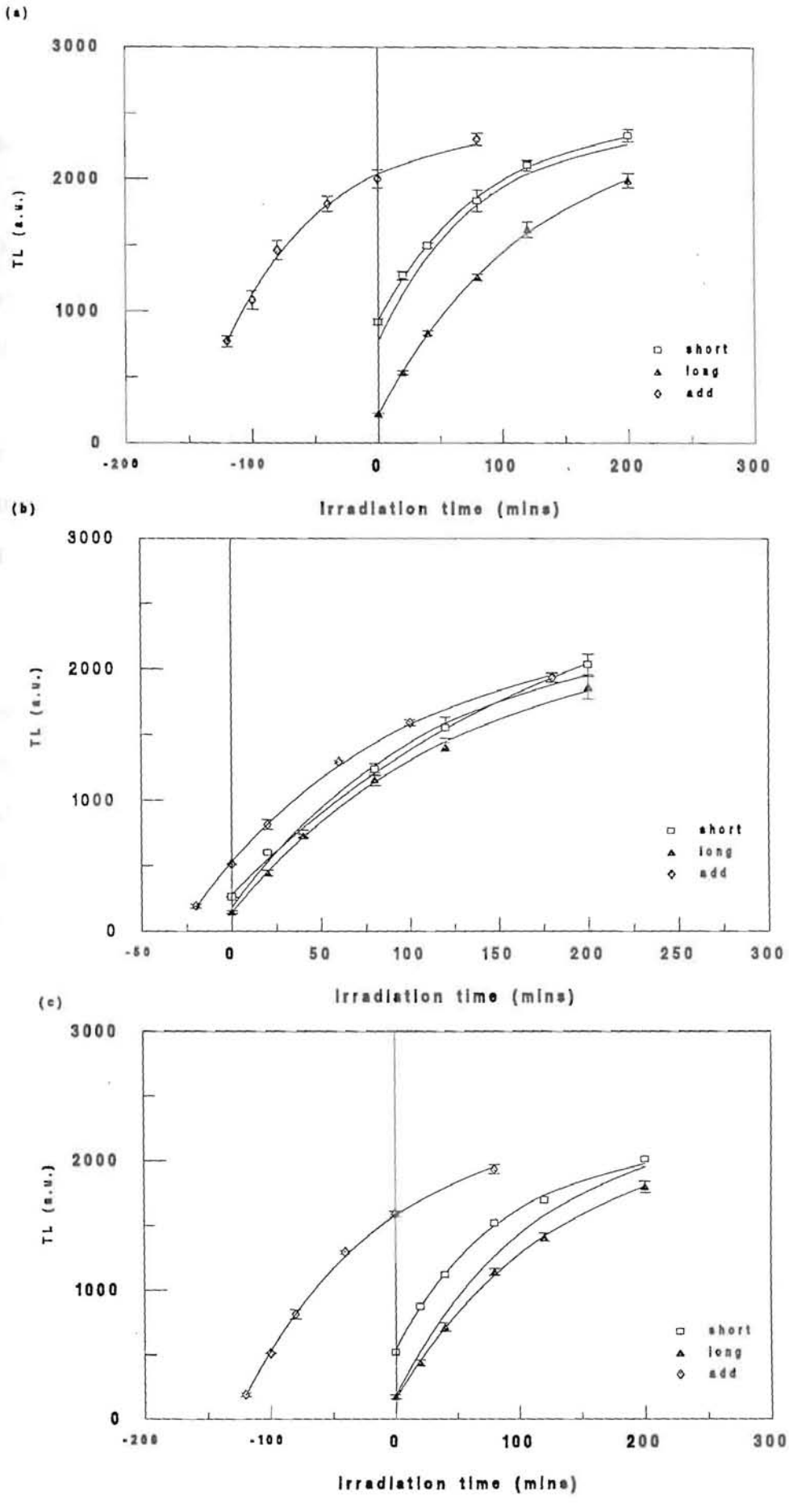


Figure. 5.4.4. TL growth curves. (a) colluvial sediment (b) young aeolian sediment (c) old aeolian sediment. Sample discs are the same as the IRSL in Fig.5.4.4. The dose rate was 3.79 Gy/min.

either the long or short laboratory bleaches (Fig. 5.4.3b, Table 5.4.1).

"Old sample". A significant sensitivity increase (+18.5%) was found for the aeolian sediments after the short laboratory bleach, but no significant change (-4.6%) after the long laboratory bleaching (Fig 5.4.3c, Table 5.4.1).

It should be noted that the sensitivity changes for the colluvial and the aeolian sediments are not due to the residual IRSL signal. Even after the short bleach, the IRSL was close to the background level, although consistently higher than the level after the long bleach.

TL

The TL results are more complicated. As expected, significantly different residual TL signals were found for the short and long bleaches (Fig. 5.4.4a,b,c). This clearly demonstrates that knowledge of the residual TL is critical when the ED is determined by the regeneration method. Because there is a finite residual signal, the sensitivity of the TL signal cannot be defined.

5.4.4. Discussion and comparison with the model predictions

The IRSL results from the experiment agree well with the model predictions (Table 5.4.1). For the "artificial colluvial sediment", the sensitivity of the IRSL signal is decreased after the long laboratory bleach, but not after the short bleach. For the "old" "artificial aeolian sediment", the sensitivity increases after a short bleach but no significant change occurs after a long bleach. The "young" "artificial aeolian sediment" did not show any significant sensitivity change. The important point is that the sensitivity change after the short bleach was substantially different for the two "artificial aeolian sediments". This implies that sensitivity changes would result in erroneous results in ED determination by the regeneration method.

We can also compare the behaviour of the "old" "artificial aeolian sediment" and the "artificial colluvial sediment", since both were given the same initial dose (120 mins). Two sensitivity changes were observed: for the "colluvial sediment", there was a sensitivity decrease after the long bleach, whereas for the "aeolian sediment" a sensitivity increase occurred after the short bleach. These different responses must be the result of the amounts of light received prior to the 120 minutes dose, i.e. equivalent to the light exposure at deposition. This suggests that not only is the sensitivity change related to the laboratory bleaching and age of the sample, but also the last exposure to light. These results for "artificial sediments" mirror sensitivity changes reported in the previous section and in the literature (Smith et al. 1990).

The TL results further confirm the importance of being able to estimate the residual signal (Aitken 1985). Unfortunately the size of the residual signals in the experiments precludes firm conclusions from being drawn concerning TL sensitivity changes particularly after the short bleach: the slope of the growth curve above the residual TL level is dependent upon the relative magnitude of the residual level, particularly for the "colluvium". The TL signal contains both bleachable and unbleachable components. The requirements of H-type charges not contributing to the measured signal by the model have not been met in the TL signal. However, comparing the shapes of the shifted "additive growth curve" and the "regeneration curve" in Fig. 5.4.4, it may be concluded that there has been a negligible change in sensitivity as a result of the 0.5 hour bleach.

In this experiment the short bleach time (0.5 hour) was used to simulate the deposition of a colluvial sediment. However, it is possible that real sediments may have been exposed to even less light, since the intensity of the SOL2 solar simulator was 6.5 times that of natural sunlight. In this case the H-type concentration may be even higher, producing an even larger sensitivity decrease after a long bleach. For IRSL dating of colluvial deposits, it would be better to bleach with IR, which would not significantly empty the H-type traps and hence

would minimize the sensitivity change. This was demonstrated experimentally for real colluvial deposits (5.2).

5.5 CONCLUSIONS AND IMPLICATIONS FOR OPTICAL DATING OF SEDIMENTS

5.5.1. Implication for the regeneration method

Either increases or decreases in sensitivity can occur after laboratory bleaching and irradiation. The sensitivity changes do not depend upon the laboratory conditions, but upon the degree of sunlight exposure of the sediment prior to deposition and the amount of radiation received by the sample since deposition. Only the latter effect has been reported for TL measurements.

Although the effect of limited sunlight exposure has long been recognized as a problem in TL dating of water lain sediments (see Berger 1988a for review), it might have been thought that, provided exposure was long enough to empty the most light sensitive (E-type) traps, OSL or IRSL techniques would yield the correct age. This seems to be unlikely if the regeneration method is applied without careful consideration of the most appropriate bleaching procedure.

Given the sensitivity changes found in these experiments and predicted by the model, the regeneration method will not be suitable for many types of sediment because of uncertainties as to the degree of bleaching of H-type charges prior to deposition. This study suggests that the regeneration method should only be applied to two types of sediments, poorly bleached colluvium in which H-type traps were close to saturation at deposition and well bleached aeolian sediments. However, without careful choice of the bleaching procedure, the results obtained with the regeneration method can only be used for comparisons.

5.5.2. Dating with single aliquot

In a recent paper, Duller (1991) introduced a method of determining the ED using a single aliquot. The method has a higher precision compared to that of the conventional method with multiple discs. For regeneration with a single aliquot, bleaching to eliminate the signal has to be carried out between each dose step. The bleaching light source can be either sunlight or the light used for optical stimulation measurements.

Figure 5.5.1 (a and b) show the type of behaviour that might be expected for a well bleached sediment when dated using the single aliquot method. If the same length of sunlight bleaching is used between each dose step, the concentration of H-type charges will relate to the previous dose and bleaching (Fig. 5.5.1a). The H-type charges will not be reduced to the same level unless using sufficiently long bleaching is used to erase the H-type charges totally. A pre-dose related sensitivity change is likely to be observed in most cases. If a sufficiently long bleach of the order of days is used, the experimental procedure will be time consuming, thus negating one of the advantages of the single aliquot method. If the bleaching light source is the same as the stimulating light, the H-type charges and thus the sensitivity will increase for most samples to an extent which is dependent on the irradiation dose in each step and the extent of light exposure. This is shown schematically in Fig.5.5.1b; although a linear response is shown, an exponential response is more likely.

For colluvial sediments, the behaviour when bleaching with the stimulating wavelength will not be expected to give a sensitivity change (Fig. 5.5.1c). The IRSL from colluvial deposits, reported in section 5.2, appeared to show no sensitivity change after IR bleaching, in accordance with Fig. 5.5.1c.. The ED obtained with the single aliquot regeneration method agreed with that obtained with the additive dose method using multiple discs. This implies that the H-type traps were full at deposition and remained full after the E-type traps were emptied with the IR bleach. The H-type charges concentration will not relate to the pre-dose

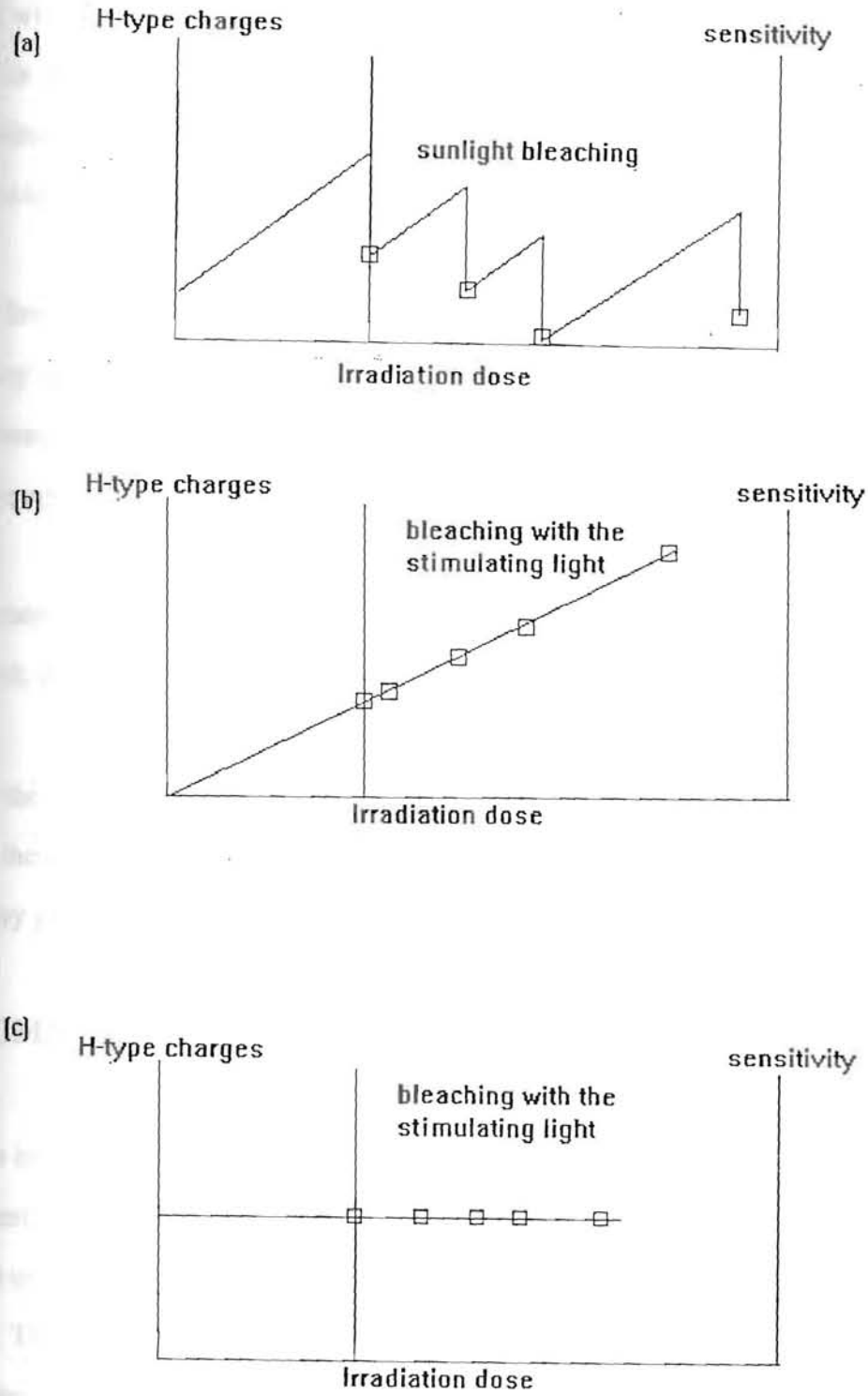


Figure. 5.5.1. Schematic diagram of H-type charge and sensitivity change involved in ED determination by the regeneration method with single aliquot. (a) bleaching with sunlight in each dose step (b) bleaching using the same wavelengths as the stimulating light (c) bleaching using the same wavelengths as the stimulating light for colluvial sediments.

and previous IR bleaching. In this case, the ED obtained by a single disc regeneration method will be correct provided that bleaching with IR is carried out between each dose step. This has been further proved by applying the method to the K-feldspar fraction and other colluvial deposits from other sections in South Africa (see chapter 7). Similar results might be obtained by using other stimulation wavelengths.

5.5.3. Sensitivity change in sediments

For many samples the degree of sunlight exposure is likely to be between those used in the experiment. Their luminescence sensitivity can therefore increase or decrease depending upon the age of the sample, laboratory bleaching and light exposure prior to deposition.

The sensitivity of a particular sample will decrease as the laboratory bleaching time is extended; this is because f(S) is smaller for longer bleaching.

Under the same laboratory bleaching conditions, the amount of sensitivity change will reflect the degree of sunlight exposure prior to deposition for sediments of the same age. This may give additional information about the degree of bleaching *prior to* deposition.

5.6 SUMMARY

Change in sensitivity of luminescence signals brought about by laboratory bleaching can be explained in terms of a competition model involving two trapped charge populations. Results of an experiment involving artificially bleached and irradiated sediment support the model. The predictions made by the model are in agreement with data reported in the literature.

Either increase or decrease in sensitivity can occur after laboratory bleaching and irradiation. The regeneration method is not suitable without careful consideration of the

most appropriate bleaching procedure.

chapter 6

IRSL DATING OF PARTIALLY BLEACHED SEDIMENTS

6.1 INTRODUCTION

In optical dating methods the stimulated luminescence signals are bleached quickly by sunlight (Huntley et al. 1985, Godfrey-Smith et al. 1988). It has therefore been suggested that these methods can be used to date materials which have not received long sunlight exposure prior to deposition. For such materials optical dating methods might be expected to have a considerable advantage over TL methods for which the residual signal must be considered.

The rapid bleaching of the OSL signal for quartz and K-feldspar has been demonstrated by Godfrey-Smith et al. (1988). Similar results for quartz were reported elsewhere (Rhodes 1990). For the OSL signal, 1% of the initial signal was reached in 10 seconds for quartz and 9 minutes for K-feldspar. For the TL signal, the same quartz needed 20 hours to reach 17% of the initial signal; the TL signal for the feldspar is more light sensitive, though much less so than for the OSL. It has been demonstrated that the bleaching of the OSL signal on an overcast day occurred 10 times more slowly than on a clear day because the light intensity on an overcast day is about 10% of that on a clear day (Godfrey-Smith 1991).

However, the residual signal for optical dating might still be a problem for the briefly bleached materials such as colluvium and alluvium (Li and Wintle 1991, Aitken and Xie 1992). Indeed such a problem was demonstrated for a fine grained colluvium sample AF-4 (also see chapter 7), for which the IRSL age derived from the ED obtained with conventional, multiple disc, additive dose method is 5.2 ka and significantly older than the ^{14}C date (1.5 ka) from an underlying soil horizon. Thus it seems that for a young, poorly bleached sample, the residual IRSL signal may still be comparable to the signal

accumulated since deposition. For such a sample the IRSL signal was not completely bleached at deposition.

Before going further, it is necessary to define what is meant by "complete bleaching". In this study the following terms are used.

prolonged bleaching: all bleachable traps are completely emptied regardless of whether they give rise to IRSL or TL signals, or H type charges (chapter 5).

short or poor bleaching: these terms are used to describe a shorter bleaching time which may not result in the IRSL signal being bleached completely.

sufficient or complete bleaching: and their converse, these refer to the IRSL signal.

old samples: samples for which the ED is greater than 100Gy.

In this chapter, I will explore the effects on the residual signal of different bleaching times and bleaching with samples having different EDs. Several methods for detection of insufficient bleaching will be reported and empirical methods of ED determination for those materials will be introduced.

6.2 BLEACHING OF IRSL AND TL SIGNALS

In order to ascertain the bleaching behaviour of the IRSL signal with natural sunlight, experiments have been carried out under different bleaching conditions. Effects of sunlight bleaching are compared with bleaching using the SOL2 solar simulator and an IR diode array, which has the same wavelength spectrum as the diodes used for stimulating the luminescence. Special attention was paid to determining the remnant signal for different initial signals.

6.2.1. Bleaching with sunlight

The sunlight bleaching experiment was carried out on a clear day in January in Aberystwyth. The sample used was the coarse grain (125-150μm) K-feldspar fraction of

sample AF-3 from South Africa. Six discs of the natural sample were used for bleaching and exposed to sunlight at the same time. The 2000 OSL Reader was used for IRSL measurements. The IRSL signal was normalized by natural normalization, i.e. using the IRSL of a short shine (0.1 second) measurement on a natural disc. The remnant IRSL signals were measured on the same group of discs with the same short shine measurement after each bleaching period. The short IR exposure allows the discs to be reused for IRSL measurements. Figure 6.2.1. shows the reduction of the IRSL with bleaching time for up to 5 hours. There is no sign of the signal having reached a stable level, though the signal was bleached to 0.2% of the initial level. The reduction rate is approximately linear in a log-log plot. This suggests an initial rapid reduction of IRSL followed by relatively slow bleaching. This is analogous to OSL bleaching behaviour reported elsewhere (Godfrey-Smith et al. 1988, Li and Aitken 1989).

Further sunlight exposure over a period of several days indicates that the IRSL signal is bleached completely after 6 days exposure. No detectable difference in IRSL signal was found for further bleaching.

6.2.2. Comparison to SOL2 and IR bleaches

Similar bleaching experiments were performed but using either the SOL2 solar simulator or the IR light. The IR bleaching and IRSL measurement were carried out at room temperature in the same system, 2000 OSL Reader. The results are compared in figure 6.2.2. The bleaching of the IRSL with the SOL2 is quicker than with sunlight. This is because the light intensity from the SOL2, as mentioned in the catalogue, is about 6.5 times higher than sunlight. If the decay rate of the IRSL is proportional to the light power, as found for the OSL signal (Godfrey-Smith 1991), the data shown in Fig.6.2.2 suggest approximately 5 times difference between SOL2 and sunlight, in agreement with the catalogue. Bleaching with IR shows the slowest reduction of the signal.

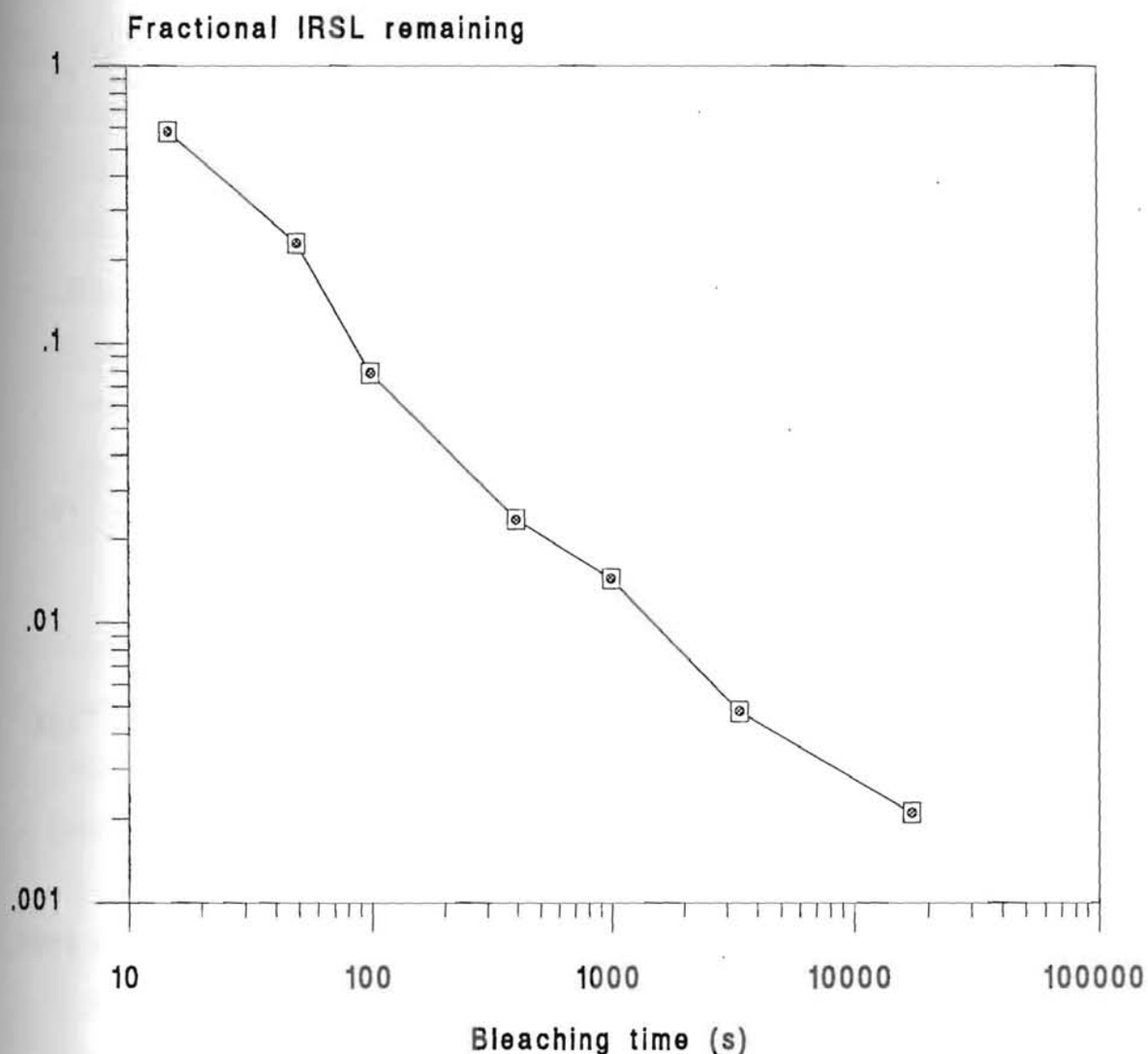


Figure 6.2.1. IRSL bleaching with sunlight on a clear day in Aberystwyth. The air temperature was 4±3°C.

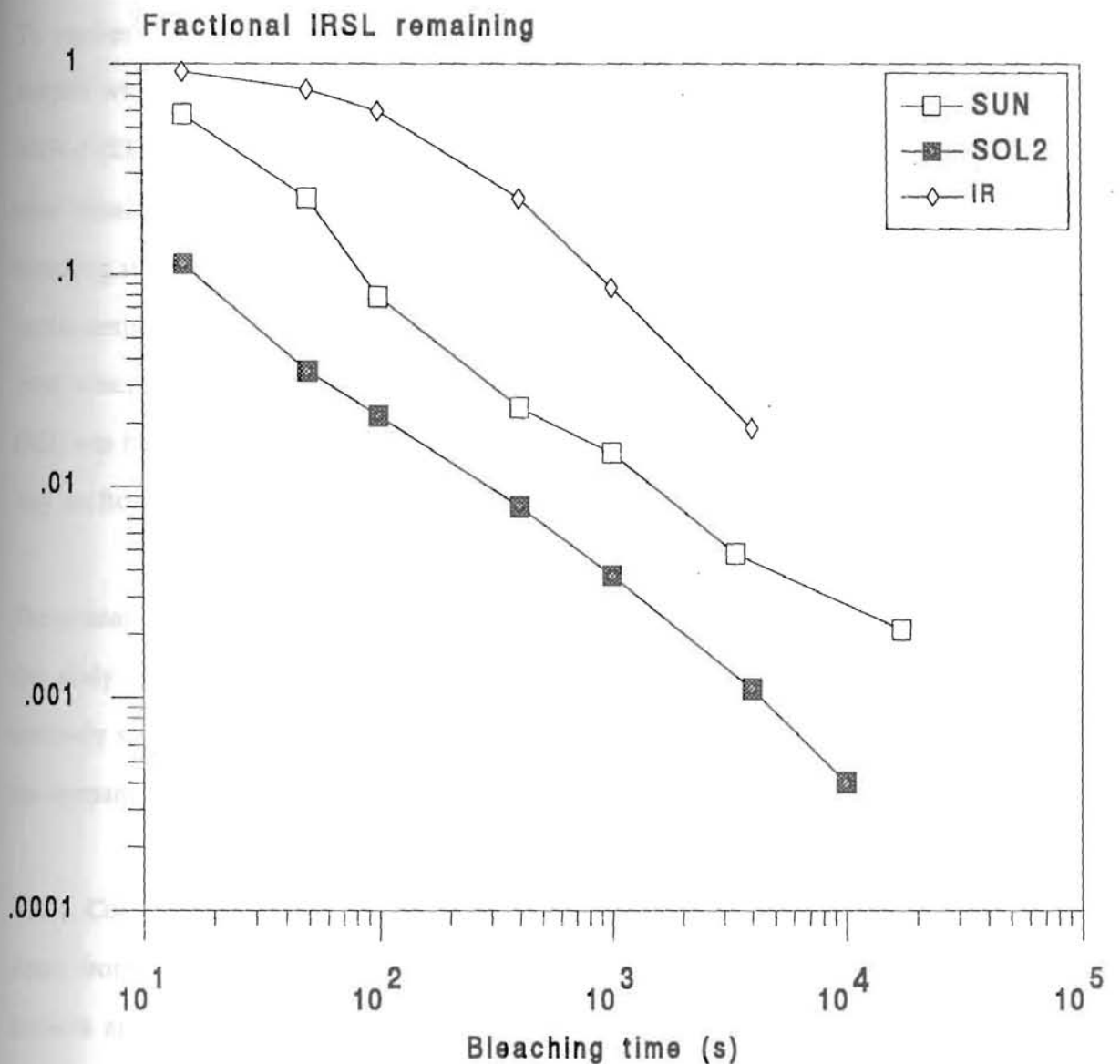


Figure 6.2.2. Comparative results of IRSL bleaching with IR, sunlight and SOL2 solar simulator.

To explore differences in signal bleaching for different initial levels, two K-feldspar samples with different EDs were used. One sample is AF-3 (ED ≈ 310Gy) and another is NEW-3 (ED ≈ 420Gy). Figure 6.2.3 shows the bleaching of both samples under the SOL2 solar simulator. Although the percentage signal reduction is almost the same for each bleaching time, the absolute level of remnant IRSL signal is significantly different. For partial sunlight bleaching, the remnant signal in a grain may thus relate to its initial IRSL level, which is related to the source of the grains and their bleaching history. Even if the IRSL was bleached to 0.1% of its initial level, there may be a sizable signal in the grains if they are from old sources, e.g. sandstone bedrock.

The remnant IRSL signal in each grain is equivalent to a dose stored in the grain, which for this study was named the 'remnant dose'. Because the dose received since deposition is relatively small for young samples, it may still be comparable to the 'remnant dose' Hence the remnant IRSL signal is particularly important for a relatively young sample.

6.2.3. Colluvial and alluvial sediments

Apart from the differences in the geological processes involved, colluvial and alluvial deposits are thought to be exposed to sunlight prior to deposition for much shorter times than aeolian deposits. This is important for luminescence signals, because the previous geological IRSL signal may not be erased completely. Even if most of the IRSL signal (99%) can be erased in a matter of minutes, the remnant signal may still be significant.

Similarly, different bleaching histories can also result in a difference in the remnant signal. It is generally thought that the grains in aeolian sediments are bleached several times during deposition and transportation. For colluvium and alluvium the bleaching cycles are fewer and thus it is necessary to check whether the sunlight exposure was sufficient to bleach the residual to a negligible level.

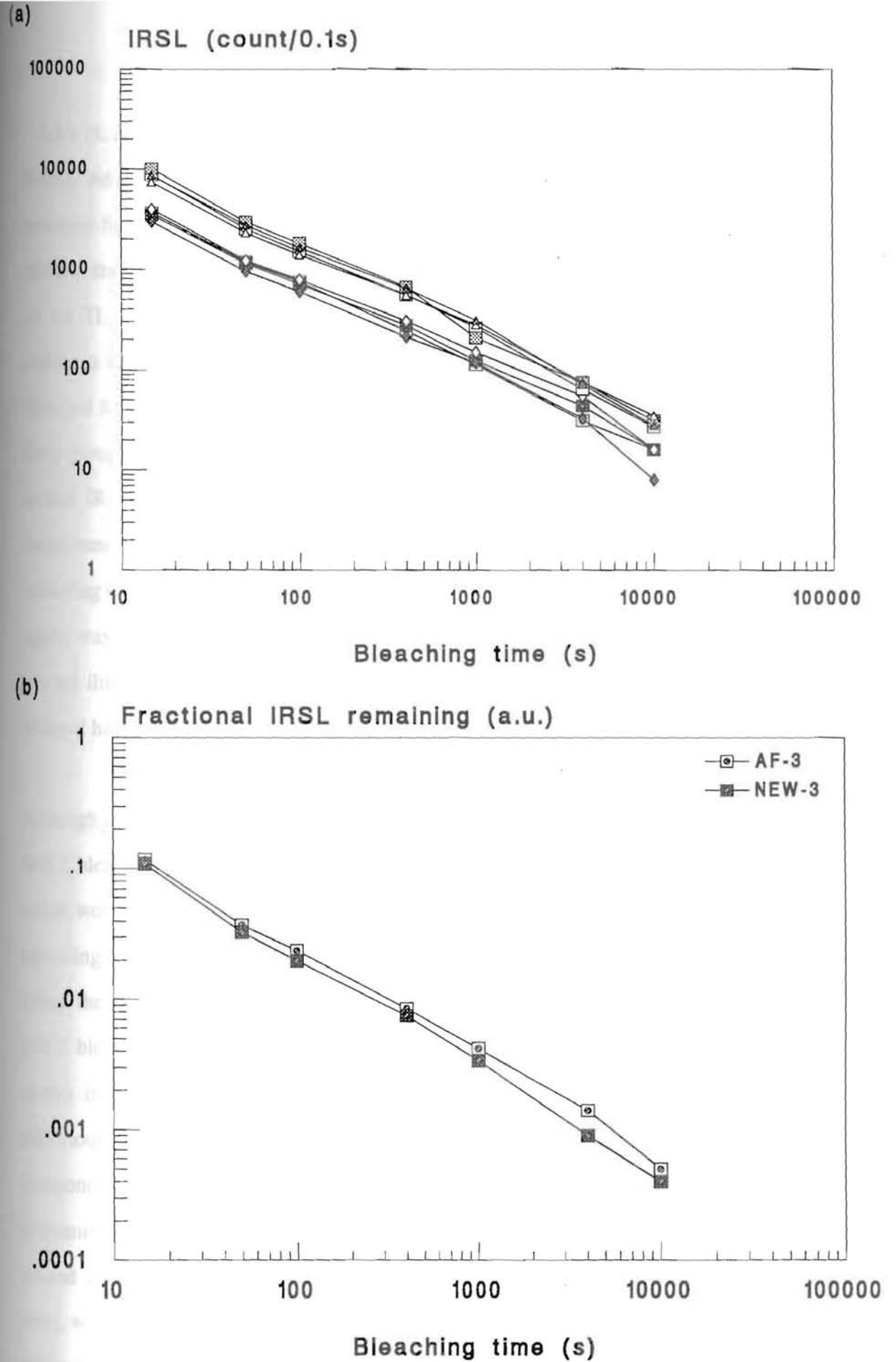


Figure 6.2.3. Bleaching of the IRSL by the SOL2 of two samples of different initial levels. (a) unnormalized discs (b) after natural normalization (each point is the average of six discs).

6.2.4 TL and IRSL bleaching

Further information can be obtained by comparing the TL and IRSL signals as a function of extended light exposure. A combined IRSL and TL bleaching study was made for the K-feldspar fraction of sample NEW-3. Because a short exposure to IR has a negligible effect on the TL signal, the same discs were used for IRSL and then TL measurements. After making a short shine (0.1s) measurement for normalization, groups of sample discs were bleached for periods of time from 3 seconds to 48 hours under the SOL2 solar simulator. Each group contains 4 discs. The IRSL was measured on the 2000 OSL system with 1 second IR exposure and subsequently the TL was measured on the Risø system. For both measurements only one (2mm thick) BG39 filter was used. Figure 6.2.4a. shows the bleaching of the TL and IRSL signal. Each data point is the average of four discs. The IRSL signal was bleached to a negligible level after 10 hours ($3.6*10^4$ s) exposure and points beyond this bleaching time are not shown in the figure. Similar results for OSL and TL of feldspar have been reported (Godfrey-Smith et al. 1988).

Although the TL signal shows a considerable residual signal even after 48 hours ($1.7*10^5$s) SOL2 bleaching, the bleaching curves give no idea about the possible difference in EDs which would be obtained using these signals. The difference between the IRSL and the TL bleaching is demonstrated more clearly if the bleachable component alone is compared. Using the data from Figure 6.2.4.a and considering the signals after 48 hours ($1.7*10^5$s) SOL2 bleaching as the unbleachable residual, the reductions of the bleachable signals are shown in figure 6.2.4b., after normalizing them to unity. The difference in bleaching behaviour can be demonstrated by calculating the difference in fractional bleachable component for the signals versus bleaching time (Fig. 6.2.4b; TL-IRSL). As can be seen, the maximum difference between IRSL and TL behaviour occurs for a bleaching time of around 25 seconds. At this bleaching time, the IRSL reached less than 10% of its initial level, whereas more than 80% of the TL signal remained. For longer bleaching times the difference between IRSL and TL rapidly becomes smaller. One important implication for

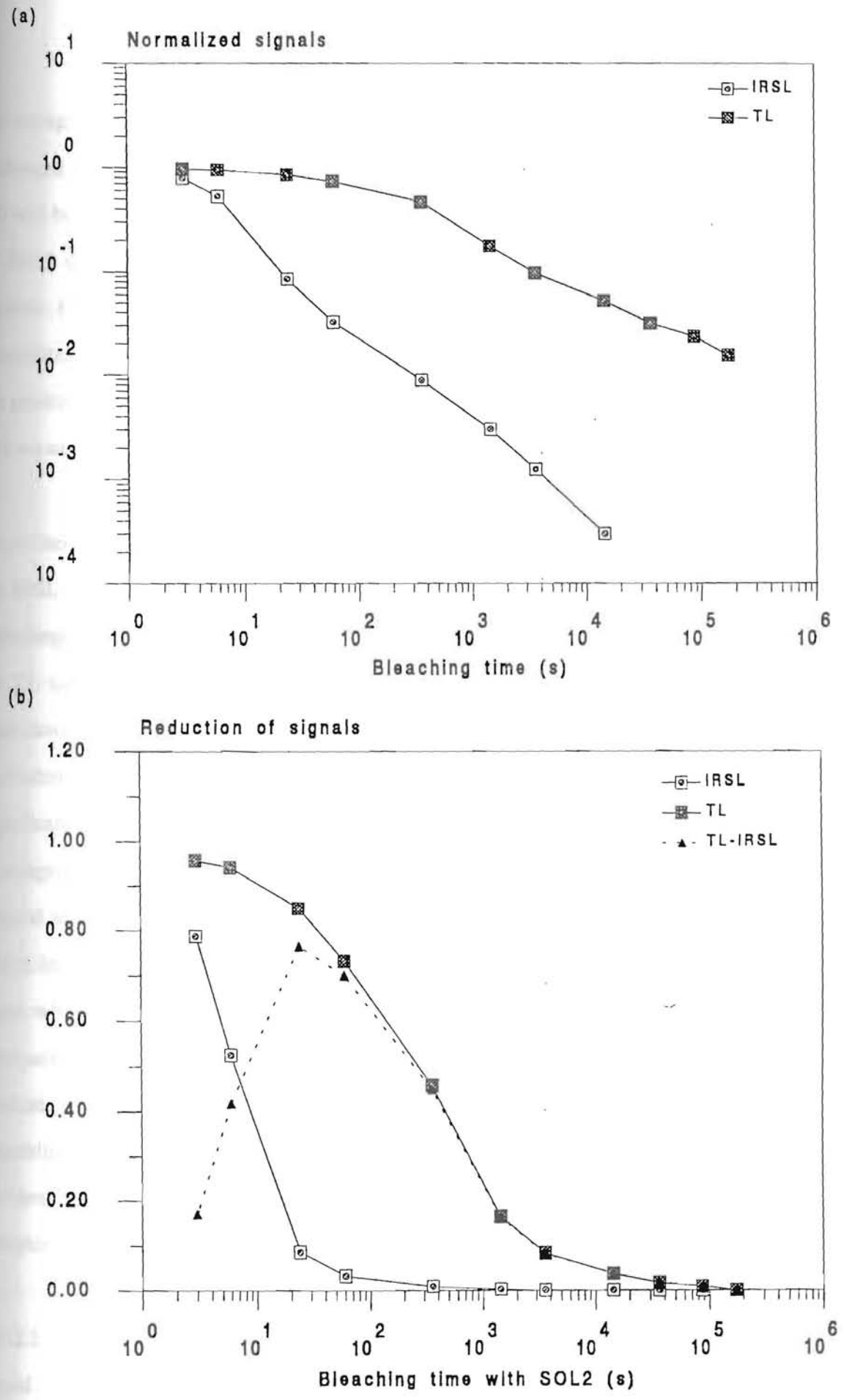


Figure 6.2.4. Bleaching of the TL and IRSL signals of K-feldspar fraction of sample NEW-3. The TL was measured after IRSL measurement with 1 second IR exposure. (a) reduction of signals (b) fractional reduction of bleachable signals and the difference (TL-IRSL) between the signals after the same bleaching. Signals after 48 hours bleaching were used as the unbleachable components and subtracted from the total signals.

the dating of sediment is that different EDs would be obtained for both signals. For a prolonged laboratory bleaching time, it would be expected that the maximum difference in ED will be observed if the predepositional sunlight bleaching was equivalent to 25 seconds of SOL2 exposure (or 150 seconds of direct sunlight). For longer sunlight bleaching, a smaller ED difference would be observed. Given the statistical error in the ED determination, normally around ±10%, the ED difference will be within the error limits if the predepositional bleaching was longer than the equivalent of 1 hour SOL2 bleaching (i.e. 6 hours of direct sunlight).

As defined and described in chapter 5, H-type charges will not start to be bleached unless the IRSL was already bleached to a negligible level. They can be bleached completely after a prolonged light exposure. Since H-type trapped charges only make up a small portion of the TL signal (i.e. that which bleaching slowly), they would be expected to be bleached even more slowly than the whole TL signal. Therefore, after an intermediate bleaching (say equivalent to 2 hours of SOL2), the ED difference between IRSL and TL would not be significant. Lack of an ED-difference might lead to the suggestion that there may have been prolonged bleaching prior to deposition. However, when applying the IRSL regeneration method after IR bleaching, a different response would be observed between those samples which have been experienced intermediate bleaching (equivalent to 2 hours of SOL2) and those which have experienced a prolonged bleaching (longer than 48 hours of SOL2). Comparing the regenerated ED after IR bleaching with that obtained for the additive dose method (without sensitivity change), similar results would be expected for the intermediate bleaching; however, for the prolonged bleaching the ED from the regeneration method after IR bleaching would be significantly smaller. These ED differences were observed in the samples used in this study and are explored in detail in chapter 7 (7.3.4).

6.2.5. Summary

Rapid bleaching of the IRSL signal was observed with sunlight exposure. Although the

signal can be bleached to be less than 1% of the initial level in a matter of tens of minutes, the remnant signal after a short bleaching could still be important for young samples.

A similar bleaching rate was found for samples with different initial signals resulting from different EDs. This implies that the remnant signal can be significantly different for grains which have received the same bleaching and will depend upon their previous geological history. For IRSL dating of young samples, it is necessary to check whether the IRSL signal was bleached completely for colluvial and fluvial sediments.

The slower bleaching of the TL, compared to the IRSL, suggests that the ED obtained by both methods will be different for samples bleached for a short time. The results of a combined TL and IRSL bleaching experiment indicates that this difference will only be observed for a limited range of bleaching time. However, the IRSL sensitivity comparison (for additive dose versus regeneration after IR bleaching) will give an indication of whether the sample has undergone prolonged bleaching prior to deposition.

6.3 DETECTION OF INSUFFICIENT BLEACHING

6.3.1. Introduction

Insufficient or uncompleted bleaching of the IRSL signal at the time of deposition will prevent correct measurement of the ED because the observed signal is a combination of the "remnant" signal and the desired "true" signal. This is particularly important for colluvial and alluvial deposits whose sediment grains may be involved in relatively few bleaching cycles prior to deposition.

From the study of sedimentary processes, it is quite unlikely that many sedimentary grains were not exposed to sunlight at least once prior to deposition. However, some deposits, e.g. aeolian deposits, were exposed for a long time, whereas others, e.g. colluvium and alluvium, were only exposed for a short time to sunlight. Because the IRSL signal can be bleached

substantially (99%) in a matter of minutes, the remnant signal after the exposure is relatively low compared to its initial signal, and is only significant for those samples which have a small signal accumulated since deposition i.e. young samples.

Two ways of detecting insufficient bleaching were suggested by Rhodes (1990) in his study of the OSL signal from quartz. These are the shine plateau and natural scatter methods, which succeeded for some samples, but failed for others.

After reviewing the shine plateau and natural scatter methods, I will introduce other methods which may be used to detect insufficient bleaching.

6.3.2. Shine plateau method

In TL dating of heated materials, an ED plateau test was introduced to confirm the full thermal resetting of the TL signal (Aitken 1985). Similarly, the concept of an ED plateau was introduced for TL dating of sediment as a way of deriving the most suitable bleaching time for obtaining the correct residual TL (Mejdahl 1988b). Analogously, a plateau in the plot of ED vs the time of the stimulated light (termed a shine plateau) would be expected to test the full zeroing of signal by light (Rhodes 1990, Aitken 1992). If the deposit was only briefly exposed to sunlight prior to deposition, the charges in the more bleachable traps would be erased and the ED obtained for electrons from these more bleachable traps will be smaller than from the traps which were not affected by the brief bleach. Therefore a poor shine plateau would be expected for poorly bleached sediments (e.g. colluvium).

The concept behind the shine plateau was first introduced for OSL dating by Huntley et al. 1985 and proposed as a method for detecting insufficient bleaching by others (Huntley et al. 1985, Rhodes 1990, Aitken 1992). Rhodes (1990) claimed that the shine plateau method could be used when the signal was bleached to be less than 11% of its initial level.

In this study a poor plateau was found for the K-feldspar fraction from a young sample (Fig. 6.3.1c). However, applying the same single disc regeneration method to fine grain polymineral discs of AF-4, the shine plateau was not flat for one disc, but was reasonably flat for another identical disc (Fig. 6.3.1a and b). This suggests that whether the IRSL signal of the sample was bleached completely cannot be demonstrated from the shine plateau alone.

As pointed out by other workers (Rhodes 1990, Aitken 1992), the plateau depends entirely on the shape of the decay curves for signals of natural and those after laboratory irradiations. The shape of each shine down curve is related to many factors, including the brightness of the disc, background level and the difference between the ED and the laboratory doses. These factors may result in the failure to detect insufficient bleaching, or give misleading information for a completely bleached sample (Aitken 1992). This method may only be used as a comparative method.

6.3.3. Natural scatter

The natural scatter method involved looking at the disc-to-disc scatter in the additive dose growth curve after different normalization procedures (Rhodes, 1990).

When colluvial sample grains were exposed to sunlight during transport along the ground surface, the amount of light to which each grain was exposed is likely to have been different. Thus for colluvial and alluvial sediments the initial "geological" dose and bleaching history of each grain is likely to be different, a fact which will also result in a different 'remnant dose' in each grain even if the depositional light exposure history is the same. It is most likely that the 'remnant dose' in each grain after a short bleach will be different. On the other hand, if all the grains are found to have the same 'remnant dose', their IRSL signal must have been zeroed at the time of deposition.

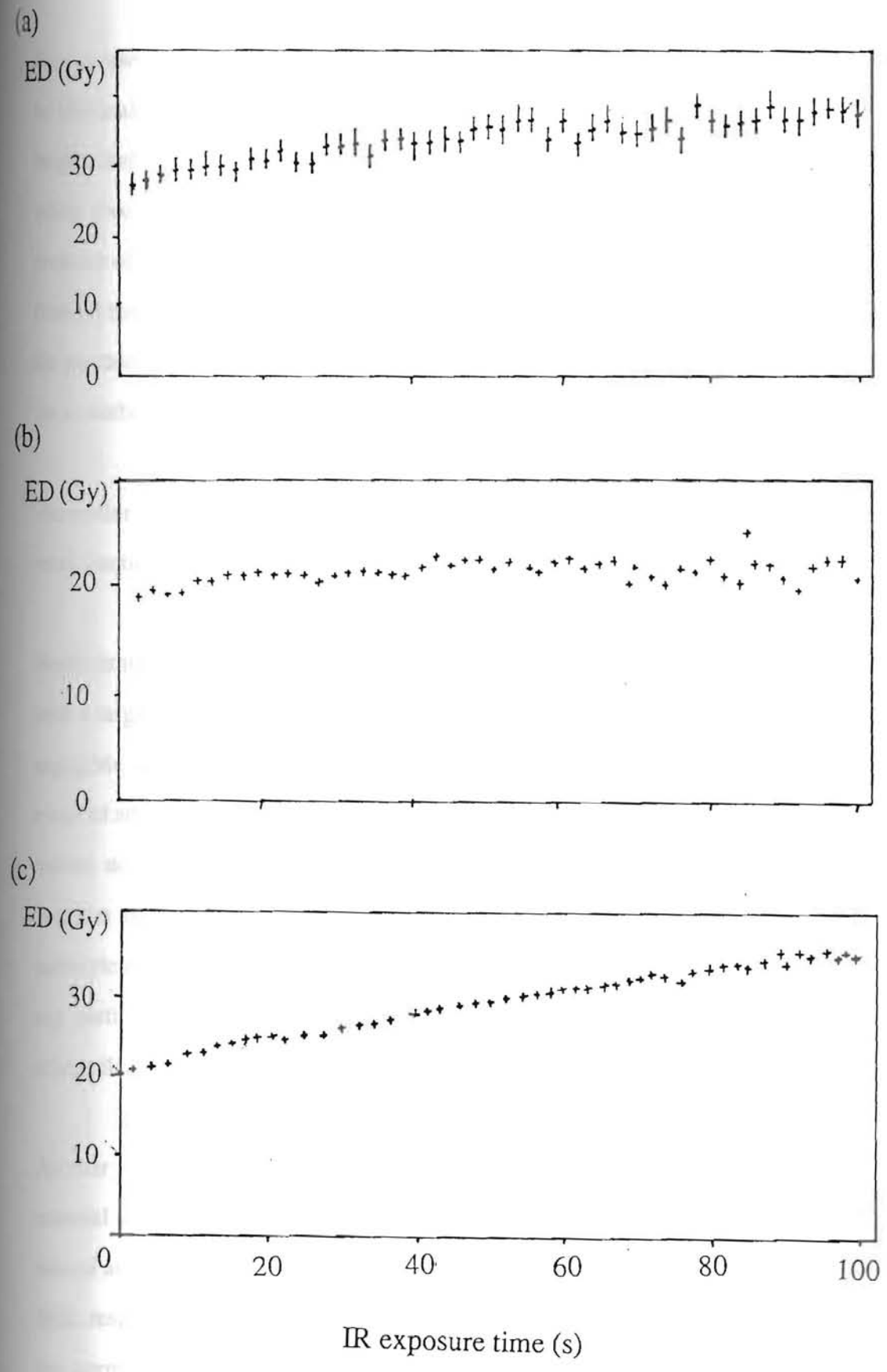


Figure 6.3.1. Shine plateau for sample AF-4. (a) fine grain disc1 (b) fine grain disc2 (c) coarse grain (125-150μm) K-feldspar disc. Single disc regeneration method was used.

For a coarse grain K-feldspar sample, the number of grains on a particular disc is unlikely to be identical. For a poorly bleached sediment there will be a few grains which have a very bright natural signal, as a result of their high 'remnant dose'. There may also be some grains which give a bright natural signal because they have an inherently higher sensitivity. The presence of the latter type of grain will not have an effect on the ED derived using the signal from all the grains on the disc. They will have little effect on disc-to-disc scatter. However, the presence of grains with a high 'remnant dose' will result in a scatter of the EDs obtained for a number of disc samples.

The scatter will become less of a problem for older samples for which the 'remnant dose' is a small fraction of the dose giving rise to the natural signal.

Normalization procedures for multiple disc methods must also be re-examined when grains with a large 'remnant dose' are present. Natural normalization procedures should result in negligible disc-to-disc variation for the natural signals. However, for discs which have received additional laboratory doses there is likely to be significant disc-to-disc scatter after natural normalization. The magnitude of the scatter will depend upon the ratio of the 'remnant dose' and the additive dose. Hence, a characteristic of a sample containing incompletely bleached grains is that for natural normalized measurements, the scatter for any particular dose point in a growth curve will increase with increasing dose. This is schematically demonstrated in Fig. 6.3.2a.

Another normalization procedure method that is applied involves the response of the material after the natural and laboratory induced signals have been erased. For IRSL, the natural and laboratory induced signals are removed by a long laboratory light exposure; the IRSL response to a test dose is then used to determine the sensitivity of the grains on disc. If this normalization procedure is applied to the mixture of sediments grains described above, the scatter given for the natural discs and for the laboratory irradiated discs will be different.

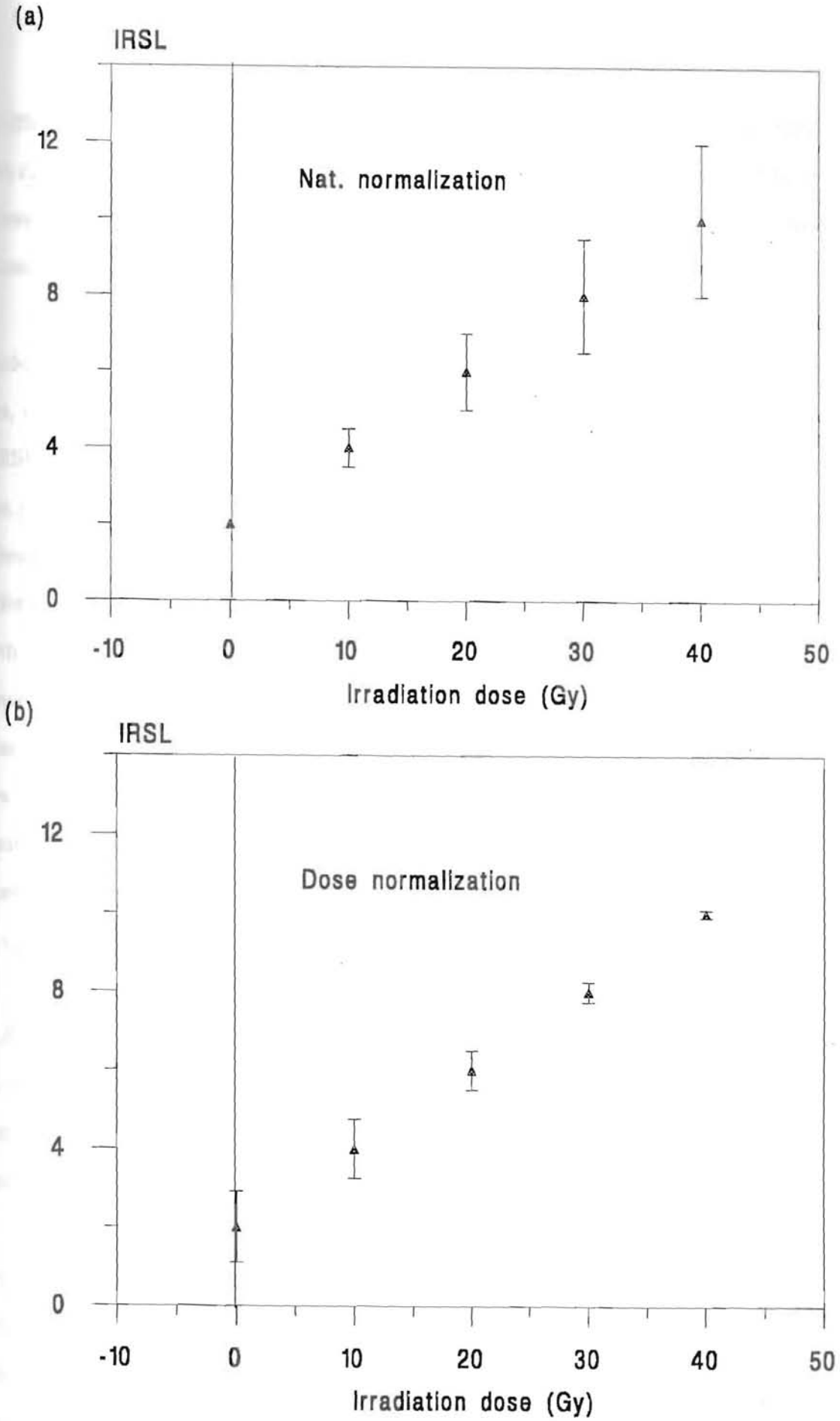


Figure 6.3.2. Schematic diagram showing the trend of disc-to-disc scatter for a insufficiently bleached sample after normalization procedures. (a) natural normalization (b) dose normalization.

The differences will not show the same pattern as when natural normalization was applied. In this case the disc-to-disc scatter will be largest for the natural dose point and will become progressively smaller for discs which have received larger laboratory doses. This is demonstrated in Fig. 6.3.2b.

Considerable disc-to-disc scatter has been reported for OSL signals from sedimentary quartz grains, even for aeolian deposits (Rhodes 1990). Very small disc-to-disc scatter was found for IRSL signals from K-feldspar for aeolian dune sands from New Zealand (Duller, pers. comm.). For the K-feldspar from the colluvial sediments in this study, significant scatter was found for the younger samples AF-4, STP-3 and HIT-2. This scatter was much larger than for the older samples. Figure 6.3.3 clearly demonstrates this effect for the additive dose growth curves of the coarse grain (125-150μm) K-feldspar fraction of sample AF-4. Figure 6.3.3a shows that the greatest scatter was obtained for the dose point with the highest laboratory dose (60 minutes i.e. 229Gy) when natural normalization was used. Figure 6.3.3b shows that use of dose nomalization (i.e. response to a test dose after a long bleach) results in least scatter for this dose point, whereas the scatter for the natural discs is larger after dose normalization. These results suggest the presence of some grains with a large 'remnant dose' on each disc.

6.3.4. EDs obtained by the single aliquot method

From the discussion of disc-to-disc scatter at the beginning of the previous section, it can be seen that the contribution of the 'remnant dose' to the measured ED can be assessed by the scatter in the ED values obtained for individual discs.

Table 6.3.1 shows the EDs obtained by the additive dose method using the single aliquot procedure (Duller 1991) for K-feldspar separates from the young colluvial samples. Similarly scattered EDs were also given by the regeneration method using IR bleaching. Since the precision of the method for a single disc is better than 1%, the measurement error

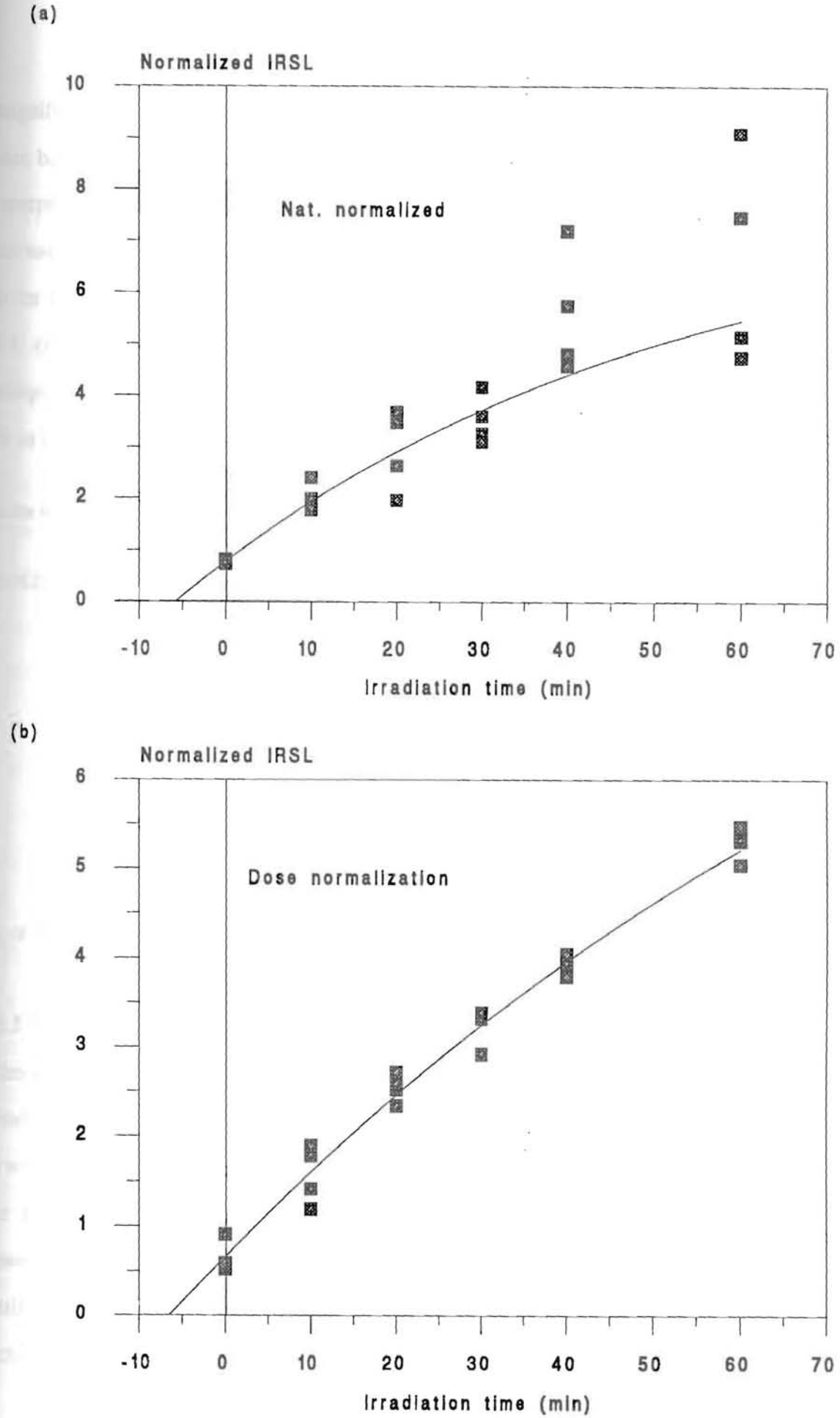


Figure 6.3.3. Disc-to-disc scatter shown for the additive growth curves of the K-feldspar fraction of sample AF-4 after normalization. (a) natural normalization (b) dose normalization (using response after a long bleach).

is insignificant when compared with the ED differences observed. The relative scatter in the ED can be seen by comparing the ED range with the mean ED of the discs. The scatter for the sample with the smallest ED, STP-13 is significantly larger than for sample HIT-2 which has a seven times larger ED. This is because the 'remnant dose' is larger compared with the post-depositional dose received by sample STP-13 (<2ka); whereas for sample HIT-2 (of early Holocene age) this portion is relatively small compared to the post-depositional dose received by the sample. (These age estimates are based on ^{14}C dates given in chapter 7).

Table 6.3.1. EDs (Gy) obtained by the additive dose method for single aliquots. The maximum and minimum values of ED are shown in bold.

SAMPLES	AF-4		STP-13		HIT-2	
	Disc	ED	Disc	ED	Disc	ED
	1	28.1	1	5.4	1	49.1
	2	33.5	2	5.6	2	42.7
	3	20.7	3	6.7	3	41.3
	4	31.6	4	8.2	4	44.9
	5	25.2	5	5.8	5	44.2
	6	33.2	6	6.2	6	**41.2**
	7	24.8	7	5.4	7	48.1
	8	21.5	8	5.1	8	43.1
	9	**15.1**	9	6.7	9	47.3
	10	28.4	10	**2.5**	10	48.0
	11	**37.0**	11	**12.5**	11	**50.3**
	12	26.6	12	9.6		
mean of ED		27.1		6.6		45.4

6.3.5 Sensitivity change after laboratory bleaching

Studies of sensitivity changes that occur after a laboratory light exposure (chapter 5) show that the changes are different for well-bleached grains and relatively poorly bleached grains. This was further illustrated in studies on the colluvial samples AF-4, AF-3, AF-2 and AF-1. Their fine grain (4-11μm) fractions showed that a sensitivity decrease occurred with increasing bleaching time (Fig. 5.2.2). It is predicted (in section 5.3 and 5.5) that the sensitivity of grains from well-bleached deposits will increase after a short laboratory bleach and that there will be no change after a prolonged bleaching; on the other hand the

sensitivity will decrease for a partially bleached deposit after a long laboratory bleach.

Because the sensitivity change is related to the concentration of H-type trapped charges (as discussed in section 5.3), sensitivity changes can only give a relative view on the bleaching experienced by the grains prior to deposition. A similar sensitivity change may be observed for several situations e.g. for a short but sufficient bleaching and also for a bleach that only partially removes the IRSL.

6.3.6. Comparing IRSL with TL

Because of the different bleaching rates for TL and IRSL signals, the EDs obtained by TL and IRSL methods will reflect the degree of bleaching prior to deposition. This is particularly clear when the same group of discs are used in both sets of measurements, a procedure which minimizes experimental differences.

Sample AF-4 has been shown to display a significant difference in EDs obtained by TL and IRSL methods; the results for fine grains (4-11μm) were discussed in chapter 5 (see Table 5.2.1). The ED given by the TL method was about three times larger than that given with IRSL. Although the alpha efficiency may be different for TL and IRSL signals (Questiaux 1992), a factor of three difference in the ED cannot be explained in this way. It is more likely that the different residual levels between TL and IRSL caused this ED difference. For well bleached aeolian samples, e.g. loess, the EDs given by TL and IRSL are very similar.

6.3.7. Assessment of different methods

The methods mentioned above (section 6.3.2 to 6.3.6) can be used to compare the behavioural differences between well- and partially-bleached deposits. Each method has particular advantages and limits in the way it can be applied to real sediments.

Out of all these methods, the most direct way of detecting insufficient bleaching is the one which involves measuring the ED for single aliquot samples. The 'remnant dose' plays a major role in the ED determination. The result can be directly linked with the extent of bleaching prior to deposition. However, for a fine grain (4-11μm) sample, the number of grains on each disc is so large that a few bright grains with a large 'remnant dose' would not add significantly to the mean ED obtained for the disc. Hence methods which look at the EDs obtained for single discs may not be suitable for looking at fine grain samples.

Similarly, the disc-to-disc scatter method also detects insufficient bleaching by involving the different 'remnant dose' for each disc. The disc-to-disc scatter would be very small for the fine grain samples. Even sample AF-4, which showed a very scattered growth curve for the coarse grain K-feldspar fraction, has little scatter in the growth curve for fine grains (4-11μm) (Fig.6.3.4).

Although the shine plateau method has no restriction related to the grain size used, it might not be a critical (or sufficient) test for dating reliability because several factors can influence the plateau (as listed in section 6.3.2).

The measured sensitivity change after the laboratory bleach can give a degree of understanding about the bleaching that occurred prior to deposition. However, it cannot indicate whether the IRSL has been bleached completely. Because the sensitivity change caused by bleaching is a relative factor and relates to the age of the samples, the degree of bleaching may not be clearly illustrated for some samples, e.g. young sediments (see chapter 5). The ED difference between TL and IRSL method is also relative, with significantly different EDs only being observed in a small range of bleaching time. The EDs can be significantly different even when the IRSL signal was not been totally bleached. The alpha efficiency between TL and IRSL can add to the uncertainty of this method. In spite of these problems, both methods (sensitivity change and ED differences) could be used for either

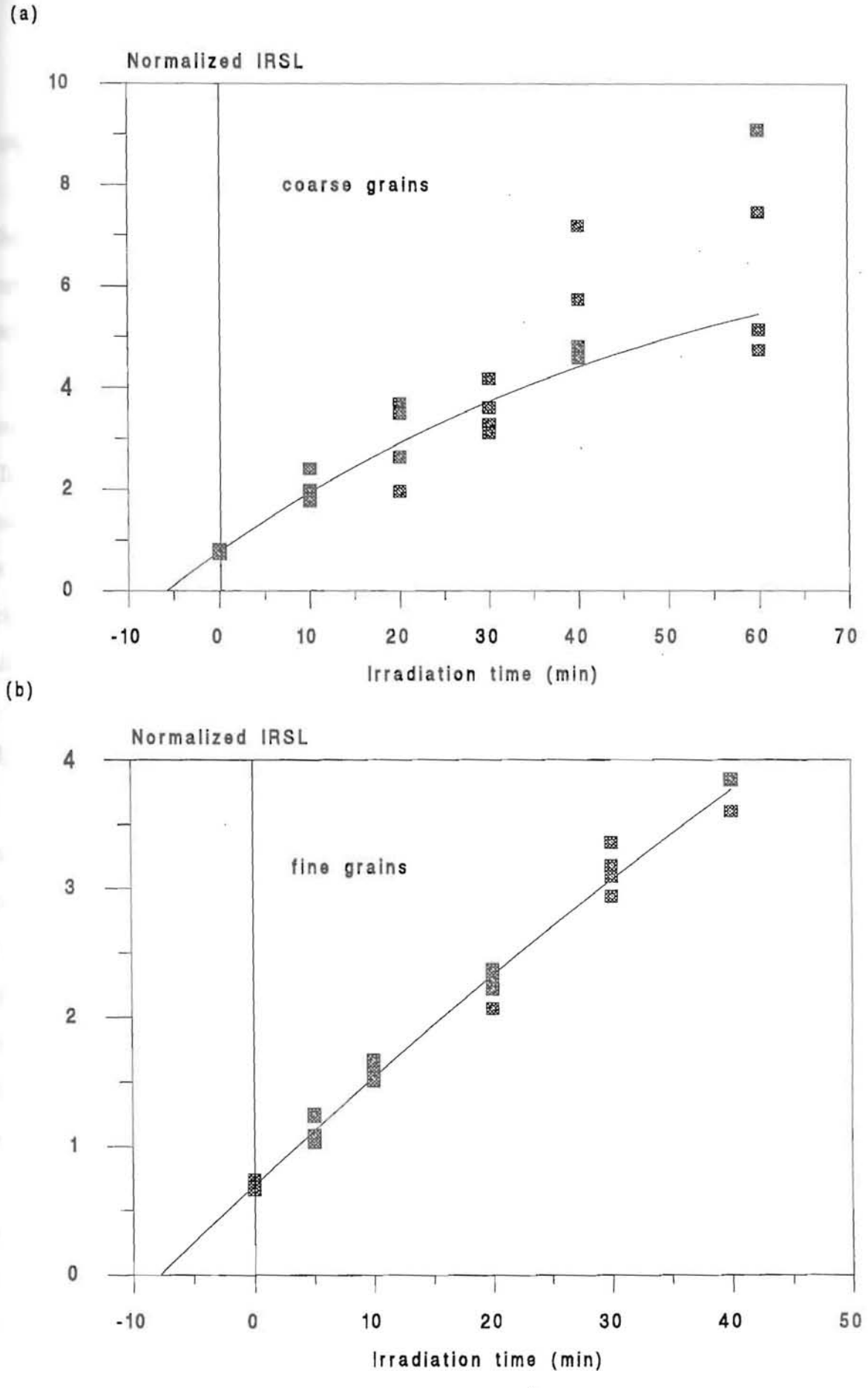


Figure 6.3.4. Comparison of disc-to-disc scatter for growth curves of sample AF-4. (a) Coarse grains (125-150μm) (b) Fine grains (4-11μm).

fine grain or coarse grain samples.

All the data used in these methods are available as part of routine laboratory dating measurements. This allows the detection of insufficient bleaching to be made without additional measurements.

Although these methods can be used to detect those sediments which have experienced insufficient bleaching, they would not detect sediments which had experienced no light exposure. If the deposit has not been exposed to sunlight, e.g. it was eroded and deposited under a thick glacier (subglacial till) or in the deep ocean (turbidites), this fact will not be detected by any one of those methods and ages older than the depositional age will be obtained.

6.4 ED ESTIMATION OF INSUFFICIENT BLEACHED SEDIMENTS

From the preceding section it appears that it is possible to identify samples which were inadequately exposed to light at deposition. For these samples the ED will be overestimated even if the IRSL signal is used for dating. This problem is particularly important for young samples for which the 'remnant dose' (as defined in section 6.2.2) is comparable to the true ED i.e. the dose received since deposition. It is therefore necessary to develop a method which enables one to obtain the best estimate of the true ED (EDo). In this section I will introduce empirical methods to evaluate EDo.

6.4.1. ED and I_n observed for aeolian and colluvial deposits

The presence of a few bright grains amongst the 5mg (or thereabouts) of K-feldspar grains on a sample disc will cause the natural IRSL intensity (I_n) to vary considerably. Disc-to-disc variation in the IRSL intensity can be brought about either by some of the grains being more sensitive to dose or by some of them containing a larger 'remnant dose' as

a result of insufficient bleaching prior to deposition. When the single disc method of ED determination is used, discs which have well-bleached grains with different sensitivities will all give the same ED. When the method is applied to poorly bleached grains, some discs will give larger EDs than others. These two types of behaviour can be demonstrated by looking at the results for an aeolian and a colluvial sample. For the well-bleached aeolian sample GDNZ32 (from New Zealand), the EDs for 18 discs were found to be very similar although there was almost a factor of 2 variation in the natural IRSL intensity, I_n (Fig. 6.4.1). When the same plot was made for 12 single disc measurements for K-feldspars of colluvial sample AF-4 (Fig. 6.4.2), the EDs varied by a factor of 2, whilst the values of I_n varied by a factor of 4.

From such plots of ED versus I_n, it is possible to determine whether the scatter in I_n values is caused by grains with different sensitivities or by the presence of grains bleached to different extents. For the aeolian sample, the ED does not increase with I_n (Fig. 6.4.1), which suggests that the difference in natural intensity is caused by sensitivity differences. On the other hand, for the colluvial sample there is a tendency for the values of ED to increase with I_n; this suggests that the scatter in I_n (and also ED) is due to differing values of 'remnant dose'. Similar behaviour patterns were found for other colluvial and aeolian samples for which the ED values were relatively small (<50Gy).

6.4.2. Possible connections between ED and I_n

In this section the links between I_n and ED (and thus the form of the plots) will be explored for grains deposited under different bleaching conditions. The information is summarized in Fig. 6.4.3 for four different scenarios and shows the single aliquot growth curves for three different discs.

For the sake of simplicity, two assumptions were made about the behaviour of the grains:- (a) the IRSL signal increase linearly with dose (b) apart from sensitivity differences the

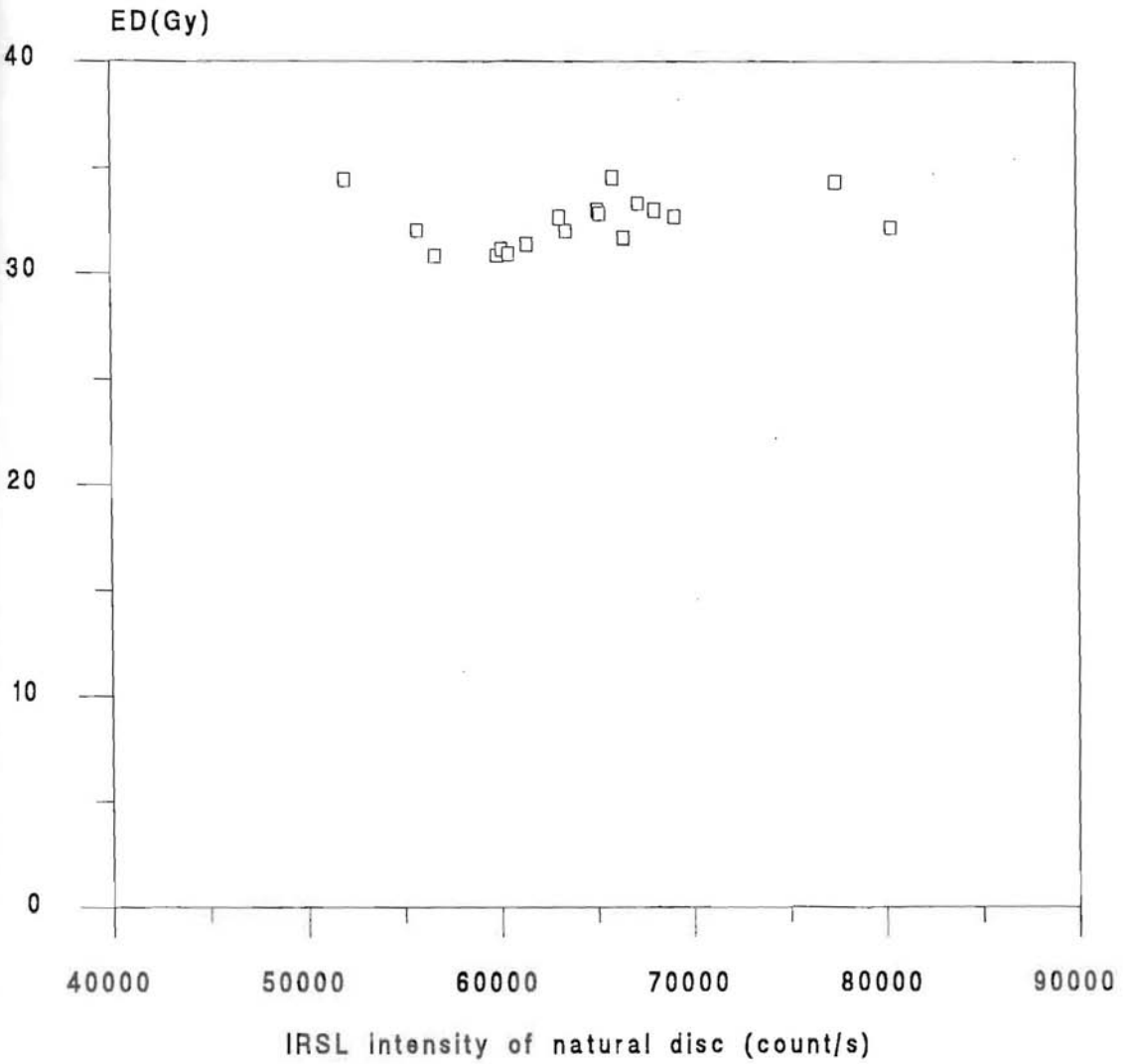


Figure 6.4.1. ED-I_n plot of aeolian sand sample GDNZ32. (Data were provided by G.A.T. Duller).

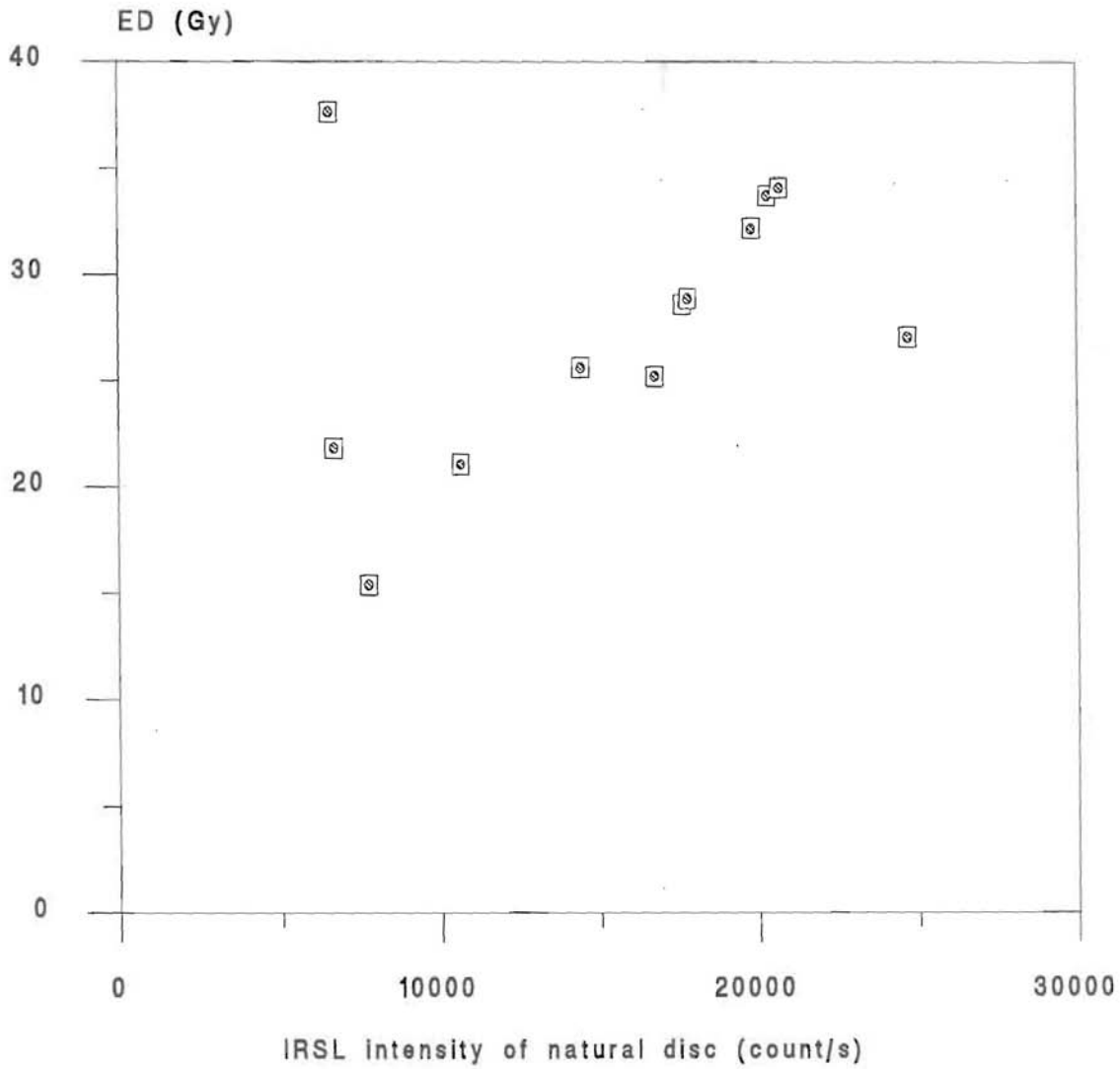


Figure 6.4.2. ED-I_n plot for a colluvial deposit sample AF-4.

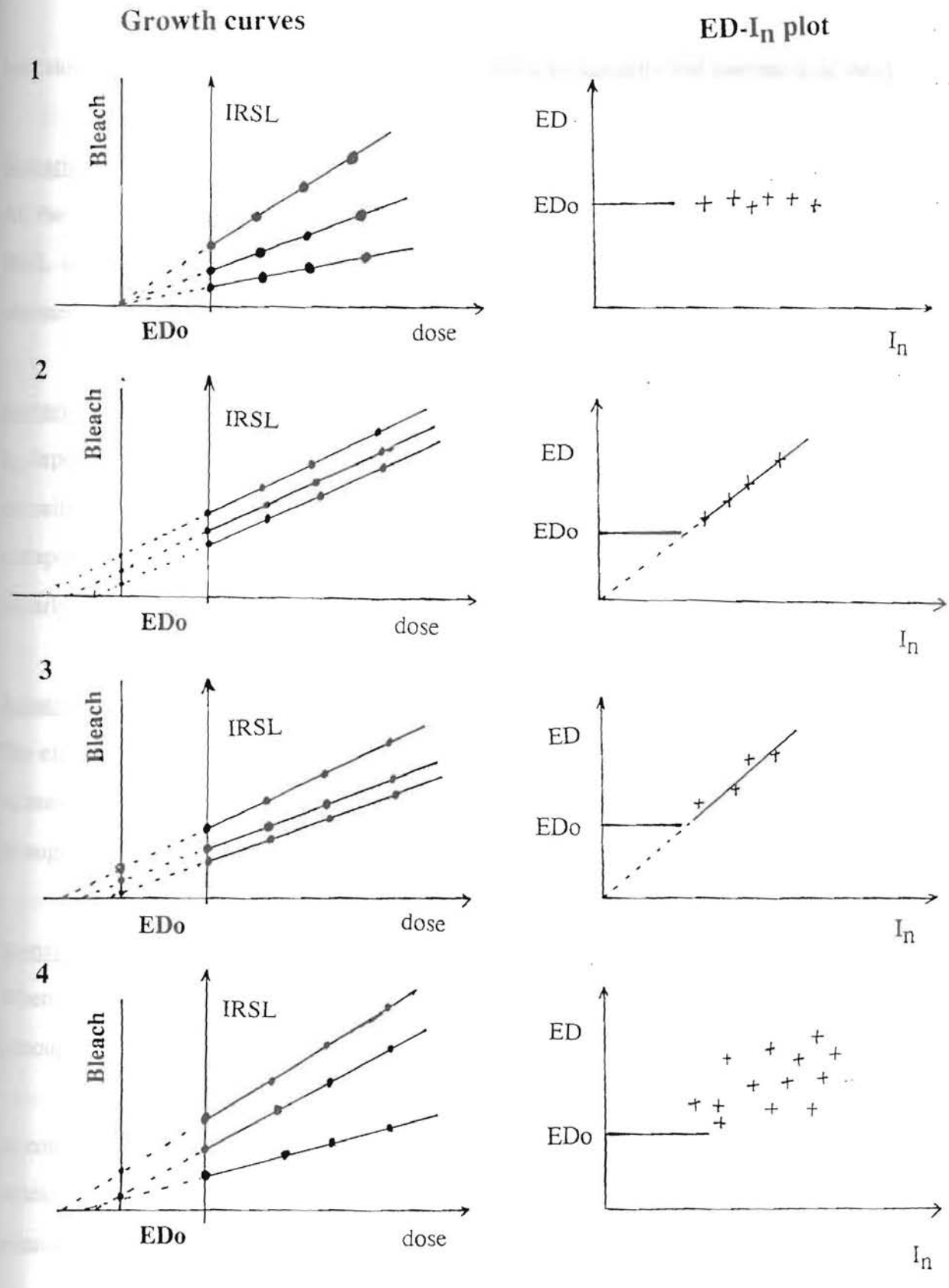


Figure 6.4.3. Possible connections between ED and I_n for different conditions of deposition.

behaviour of the grains is identical (e.g. recuperation, superlinearity and internal dose rate).

Scenario 1. Completely bleached sample with different sensitivities to dose.

All the grains were well-bleached prior to deposition. Although each disc has a different IRSL sensitivity, giving rise to different natural IRSL signals (I_n), the values of ED obtained are the same and give the true ED (EDo). This gives a flat plot of ED versus I_n.

Scenario 2. Partially bleached sample with the same sensitivity to dose.

I_n depends upon the size of the signal that would have been obtained from grains at deposition. As the grains have the same sensitivity, the growth curves are parallel and an extrapolation give values of ED larger than EDo. The plot of ED versus I_n will increase linearly and will extrapolate to the origin.

Scenario 3. Partially bleached sample with slightly different sensitivities to dose.

The effect of a finite residual signal at deposition will dominate and result in a plot with ED increasing with I_n. The sensitivity differences will cause a scatter of points about the line through the origin.

Scenario 4. Partially bleached sample with different sensitivities to dose.

When both effects play a major role, the plot of ED versus I_n will be very scattered, although a trend can still be seen, but not passing through the origin.

In conclusion, partially bleached sediments will be characterised by the trend of a linear relationship between ED and I_n. These ED-I_n plots should be born in mind when considering the best approach to determine EDo (section 6.4.3).

6.4.3. Empirical methods to assess the true dose (EDo)

1. Method 1: A value of ED from the ED-I_n plot

For a particular sample, with grains of different sensitivity which have experienced different amounts of bleaching, an approximately linear behaviour in the ED-I_n plot should be observed, provided enough discs have been measured. Such a data set is shown schematically in Fig. 6.4.4. The increase in ED with I_n is due to the presence of an increased 'remnant dose' in grains on some discs. On the other hand, for grains which were well-bleached, the ED given would be independent of the sensitivity of the grains on the disc. Thus the true dose, EDo, would be independent of the sensitivity and EDo would be given even if I_n was scarcely above the background level i.e. I_n close to zero. It thus follows that a minimum estimate for EDo could be the value of ED obtained as I_n approaches 0, i.e. the intercept on the ED axis, ED'.

As shown in Figure 6.4.2, a small number of discs did not form part of the general trend of the scatter diagram. These were considered to exhibit aberrant behaviour and a disc selection procedure was employed to make sure that the curve obtained is representative,. This was achieved by progressively rejecting discs to permit the correlation coefficient (r) to exceed a particular value. This was set at 0.8, for which the probability of being exceeded is less 0.001 when measuring 12 discs (Young 1962). The number of discs rejected should not be more than 20% of the total number discs. Figure 6.4.5a,b and c show the results for the youngest colluvial samples AF-4, STP-13 and HIT-2; rejected discs are indicated by open squares. Values of ED' were obtained from the linear fit obtained for the remaining data points; they were 10.2, 1.6 and 35.6 Gy, respectively. For older colluvial samples, e.g. HIT-1, differences in sensitivity dominate over the effect of the 'remnant dose' and an almost flat plot of ED versus I_n is obtained (Fig. 6.4.6).

Although this approach has not been justified theoretically, it is an empirical method which can give a relative result. The data are provided by single disc ED determinations used in a

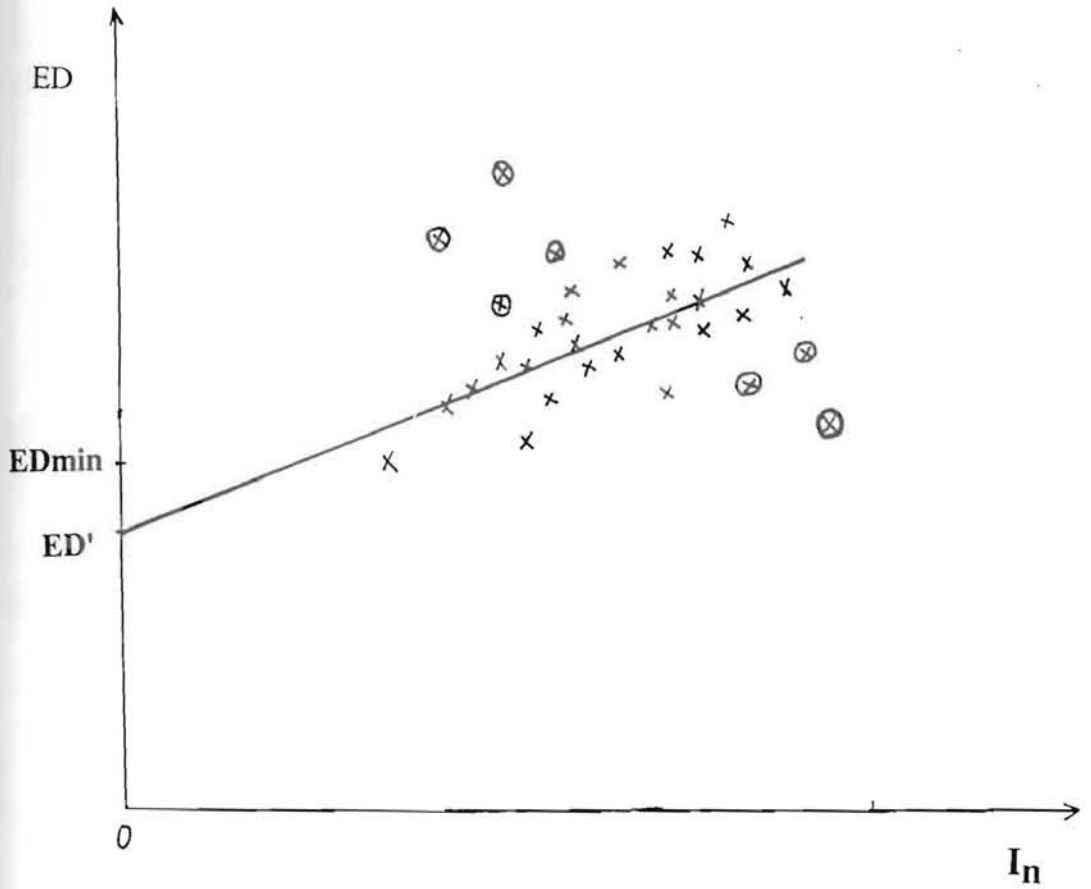


Figure 6.4.4. Schematic diagram showing the ED-I_n plot method as used for the ED determination of an insufficiently bleached sample.

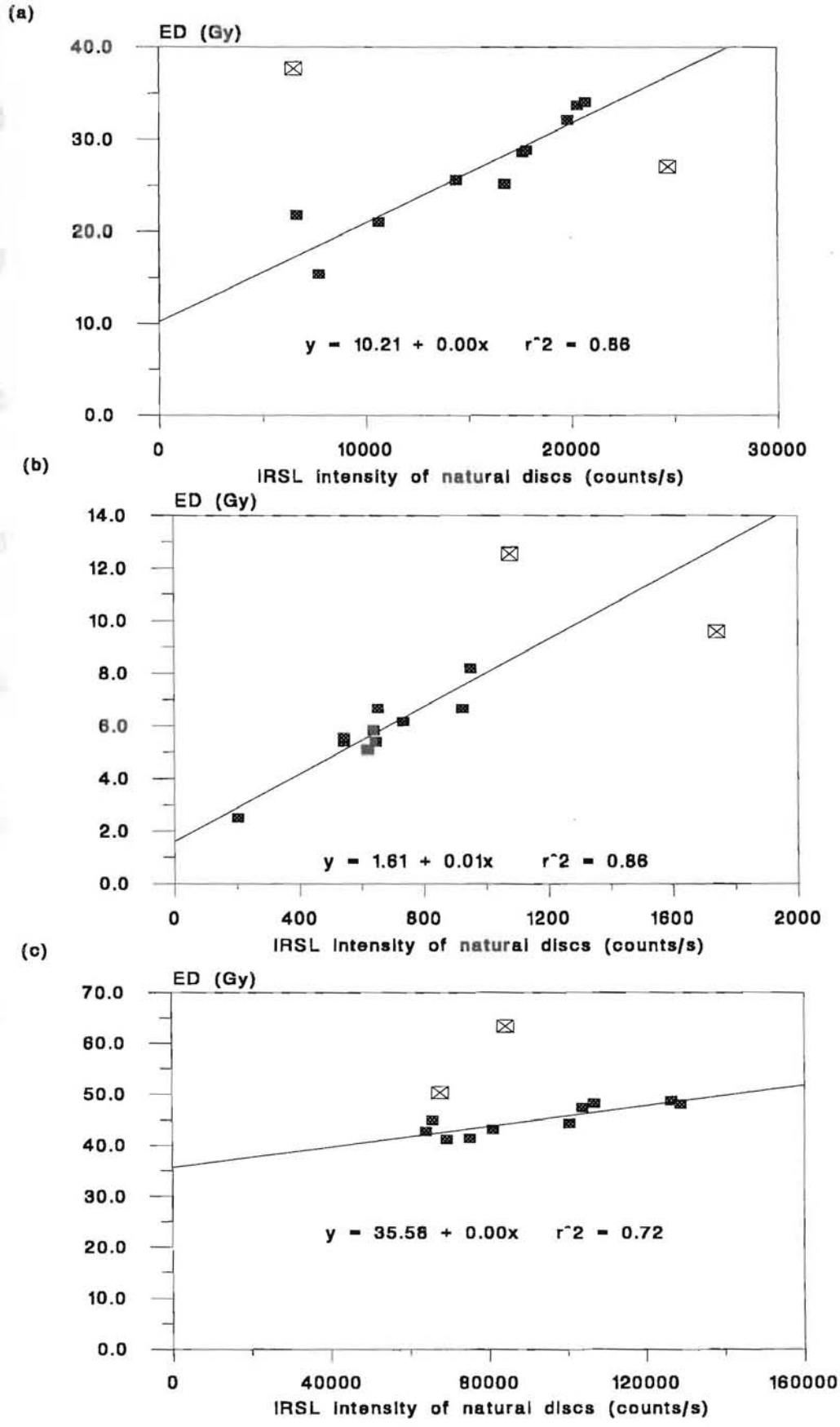


Figure 6.4.5. ED-I_n plot showing a linear relationship for partially bleached colluvial samples. (a) AF-4, (b) STP-13 and (c) HIT-2. Data points were selected to meet the criterion of a correlation coefficient greater than 0.80. (⊠) Rejected discs (■) discs used for the fitting.

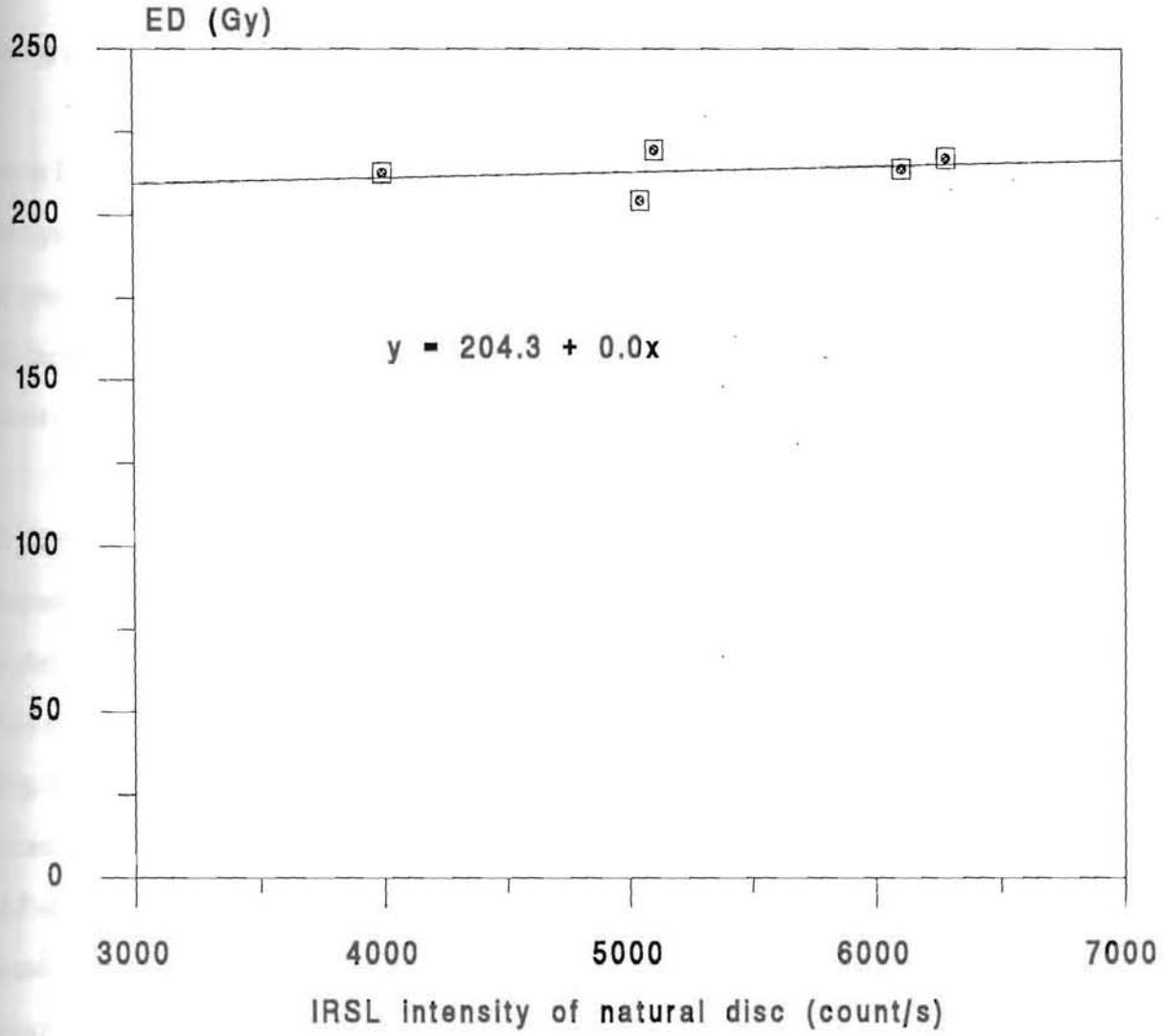


Figure 6.4.6. ED-I_n plot for an older sample, HIT-1.

dating programme and hence no additional measurements are required.

From Fig. 6.4.3 it can be seen that except for a well bleached sediment (Scenario 1), ED' is always less than EDo. In particular for scenario 2 and 3, ED' is zero. However, in the case of grains with both mixed sensitivities and mixed bleaching levels at deposition (scenario 4), the plot intercepts the ED axis above zero. Hence ED', the value for $I_n = 0$, can provide a lower limit for the ED.

2. Significance of the minimum ED

Repeated single disc ED determinations on samples of similar mass will increase the density of the data point in the ED-I_n plots but will not increase the ED or I_n limits significantly. This results in a minimum value of ED being able to be obtained from the data set, EDmin (Fig. 6.4.4). This may be best considered as a maximum estimate of the true dose (EDo) because it may still contain a component of 'remnant dose'. For the young colluvial samples (AF-4, STP-13 and HIT-2), EDmin were 15.1, 2.5 and 41.2 Gy, respectively. It should be noted that the value for STP-13 is based on a single data point which falls outside the main group, though admittedly on the same trend. A more realistic value for EDmin for STP-13 is likely to be 5Gy.

The scatter in the ED values given by the discs (in the ED-I_n plot) also provides useful information. The range (ΔED) in ED values is caused by the presence of variable amounts of 'remnant dose' from disc to disc. It can thus help to put a further limit on the evaluation of EDo.

The ratio R = ΔED/EDmin can also be used as an expression of the contribution of the 'remnant dose'. If the ED is mainly contributed to by EDo, then R would be expected to be small, whereas R would be expected to be larger if the ED has a significant contribution from the 'remnant dose'. For the three young samples, R was found to be 1.4, 4.0 and 0.2 for

samples AF-4, STP-13 and HIT-2, respectively. It should be noted that the value for STP-13 would be much lower (0.7) if the single low data point was rejected. For older colluvial samples and the young aeolian sample, R is substantially smaller. For the aeolian sample GDNZ32, R is 0.1, half the value for the colluvium of similar ED (HIT-2). For older samples, whether from aeolian or colluvial sediments, R is less than 0.2, indicating that the effect of the 'remnant dose' is negligible.

3. Method 2: Likely range of EDo

For the young colluvial samples ΔED can also be used to put further limits on the likely value of EDo. ΔED is the maximum range of the measured values of ED, as shown in Figure 6.4.7. The minimum measured value EDmin may still contain a proportion of the 'remnant dose' and thus the value EDo may be smaller than EDmin, but the difference between EDmin and EDo is likely to be less than ΔED, since EDmin is the closest value to EDo. Hence the true ED (EDo) should be in the range EDmin -ΔED to EDmin, as shown by the lower bracket in Fig. 6.4.7.

Applying this approach to samples AF-4, STP-13 and HIT-2, the possible ranges of EDo were obtained (Table 6.4.1). For samples AF-4 and STP-13 EDmin is smaller than ΔED and hence the true ED is in the range zero to 15.1 and 2.5 (5.0) Gy, respectively, indicating that both samples may be very modern. This will be discussed further in chapter 7. For sample HIT-2, EDo is in the range 32 to 41 Gy.

Table 6.4.1. The range of true ED (EDo) obtained by using EDmin and ΔED. (R is ΔED/EDmin)

SAMPLES	AF-4	STP-13	HIT-2
EDmin (Gy)	15.1	2.5 (5.0)*	41.2
ΔED (Gy)	21.9	10.2 (8.4)	7.5
R	1.4	4.0 (0.7)	0.2
possible true ED (Gy)	0-15.1	0-2.5 (5.0)	32.1-41.2

*note: for sample STP-13, the values given in brackets are those after rejecting a single data point, which falls outside the main group.

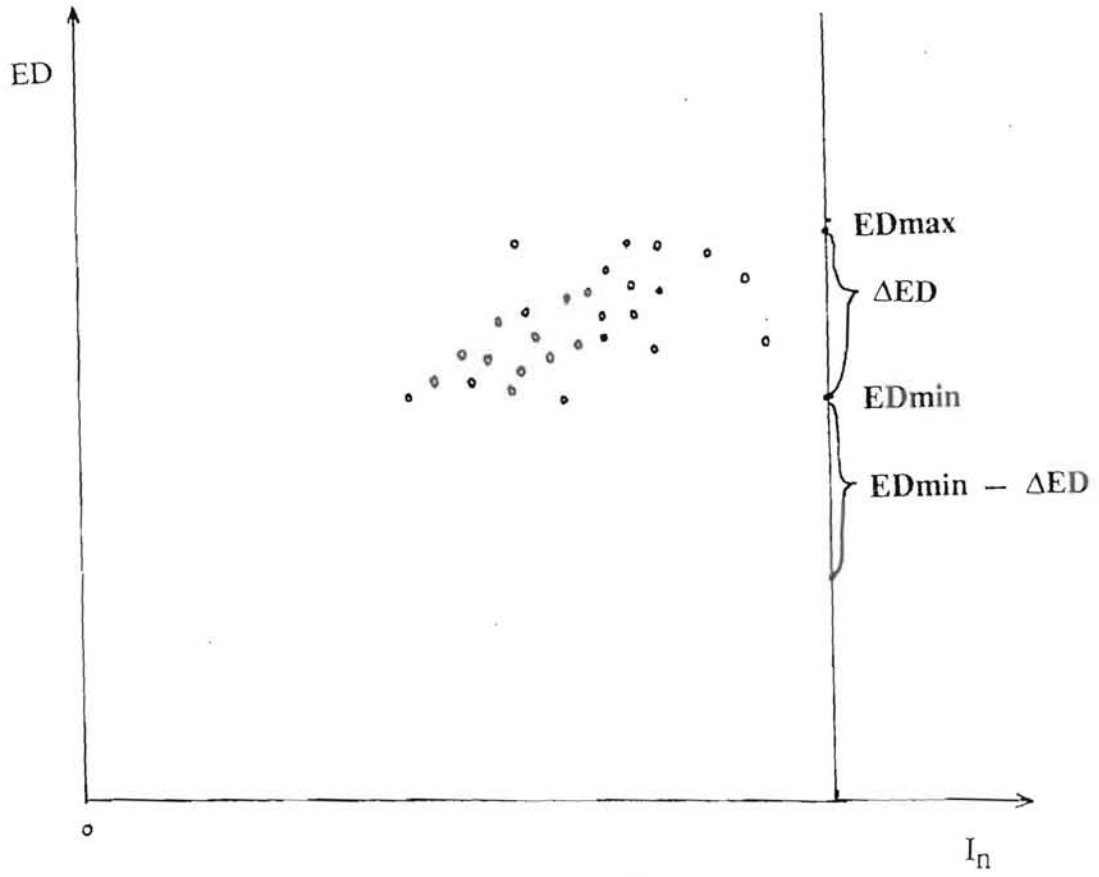


Figure 6.4.7. Schematic diagram showing method 2. The most likely value for EDo is given by the minimum ED and the range (ΔED) of the ED obtained.

The range of EDo given by this method depends on the relative strengths of EDmin and ΔED. For most very young samples, i.e. with small EDo, EDmin will be smaller than the ΔED except for completely bleached grains. Because ED must not be negative, the range of the EDo will be from zero to EDmin. Hence this method will not be suitable for very young samples of poorly bleached sediments.

4. Result of ED measurements using fewer grains

More information can be obtained by applying the single disc method of ED determination to sample discs with a smaller number of grains. It is possible that even in a colluvial sediment there are some grains which have had their IRSL signal totally erased at deposition, i.e. have no 'remnant dose'. These grains will have a greater influence on the value of ED obtained for a disc, if they are present as a higher fraction of the total number of grains. This position will be approached for a number of discs, as the number of grains per disc is reduced.

This approach was tested on the young samples AF-4, STP-13 and HIT-2. Instead of a few thousand grains being on a disc as for routine ED measurement, discs with less than approximately 500 grains were prepared. Most of the discs had only about 100 grains. Considering the limitations on this approach presented by photomultiplier tube noise, the single disc regeneration method (rather than the single disc additive dose method) was used. When matching light levels, the noise will be the same for each dose point. Figure 6.4.8. shows the EDs obtained with these discs, superimposed on the ED values already displayed in Figure 6.4.5. The smallest value of ED obtained by the discs with fewer grains and the intercept value ED' are compared in Table 6.4.2. As can be seen, the value of ED' obtained from the ED-I_n plot agrees with the smallest ED value obtained with fewer grains on the discs. These values are likely to be closest to the true ED and both doses fall in the range of dose obtained by method 2 (Table 6.4.1.).

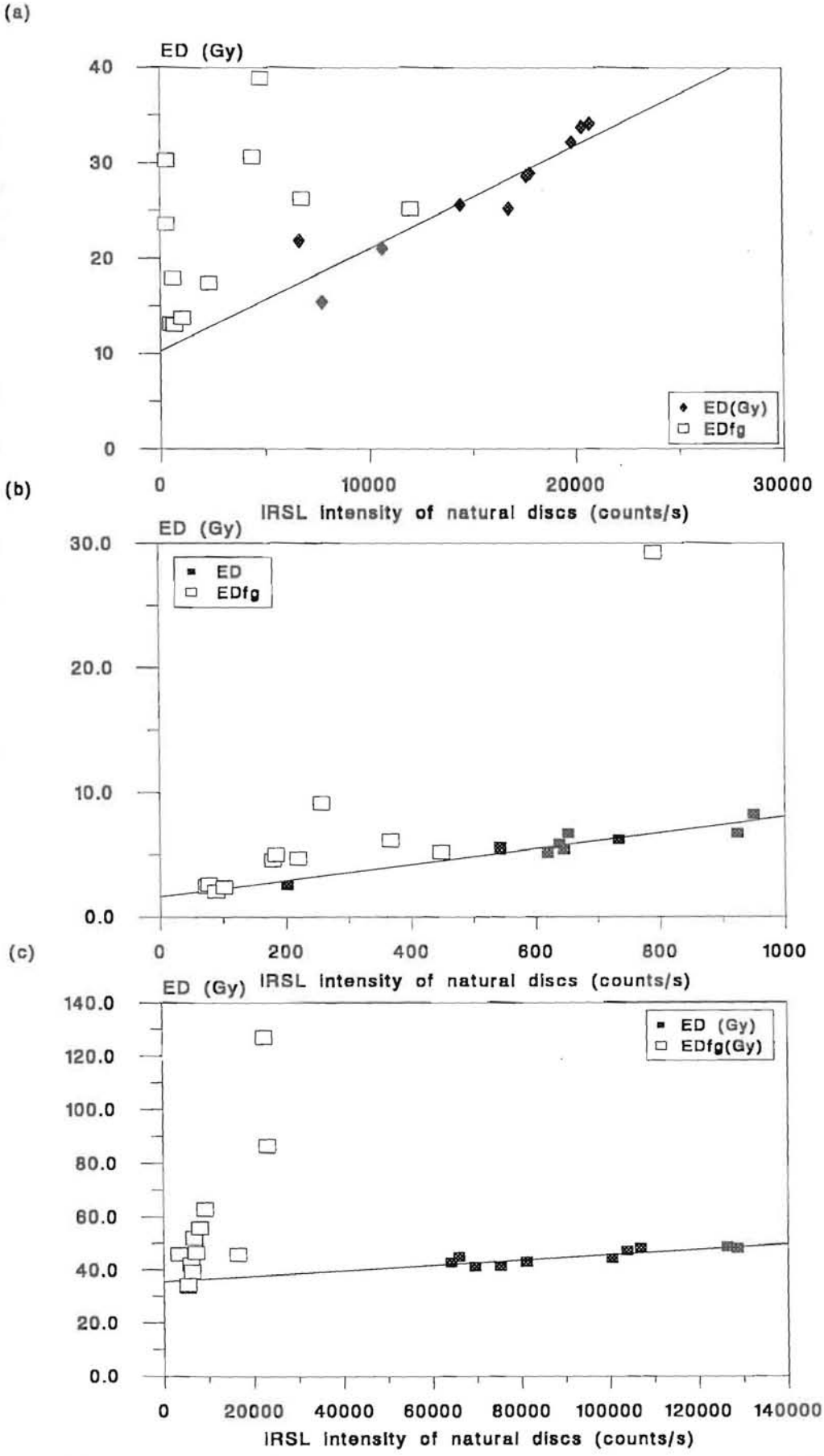


Figure 6.4.8. ED-I_n plot for discs with fewer grains compared with the values for fully covered discs. The data points for the fully covered discs were the same as in figure 6.4.5. The values of ED for the discs with fewer grains were obtained using the single disc regeneration method. The fully covered discs were measured using the additive dose single disc method.

Table 6.4.2. Comparison of ED' and the smallest ED obtained for discs with fewer grains.

SAMPLES	AF-4	STP-13	HIT-2	
ED' (Gy)	10.2	1.6	35.7	fully covered disc
smallest ED(Gy)	13.0	2.0	33.6	a few hundred grains

Apart from the low signal to noise ratio, some other effects may occur when using discs with fewer grains. The ED range for those discs will be enormous because the large 'remnant dose' in one or two grains will contribute significantly to the ED. The internal dose of the grains may be different (e.g. different K content or different U, Th content relating to the different origin of minerals when they were formed). These points will affect the ED and reduce the precision of the method.

These methods need to be applied to insufficiently bleached samples which have independent age control.

6.5 SUMMARY

Bleaching experiments indicate that the 'remnant dose' still be a problem for samples which have only been bleached for a short time prior to deposition. This effect is important for young samples but negligible for old samples.

Several methods have been introduced in order to detect insufficient bleaching and some of them can provide information on the extent of the bleaching. Others can only distinguish between samples bleached for a very long time and for a short time.

Possible values of the true ED may be evaluated from the analysis of the EDs and natural IRSL signals obtained using single disc dose determination methods.

chapter 7

DATING APPLICATIONS

7.1 INTRODUCTION

7.1.1. Importance of this study

Since the optical dating method was introduced (Huntley et al. 1985), most applications have focused on well-bleached sediments (Rhodes 1990, Godfrey-Smith 1991). However, a considerable advantage of optical dating methods is that they should be applicable to sediments which were not bleached for a long time prior to deposition. This advantage has not been demonstrated clearly in previous studies, although some interest has been expressed in dating of colluvium and alluvium (Aitken and Xie 1992). It is important to establish procedures which allow the expansion of optical dating methods to materials bleached for a short time. Such sediments exist in a wide range of geological contexts around the world.

Colluvium is defined (Watson et al. 1984) as sediment composed of poorly sorted mixtures of clay, sand and gravel sized particles which accumulates on lower hillslopes. According to Goudie and Bull (1984), colluvium is that part of the regolith that accumulates as a result of mass wasting processes on the lower parts of slopes, occurring preferentially where a topographic low was created in an earlier stage of fluvial incision.

Most samples used in this study were from the eastern part of South Africa. Few dating methods are suitable except for the ^{14}C method, which has been used to produce dates on organic buried soils which are found at some sites. The Quaternary geochronology of the region has not been established.

7.2 GEOLOGICAL DESCRIPTION

7.2.1. Geological background

Colluvial deposits mantle about 20% of the land surface of eastern southern Africa (Dardis

1990). In many parts of Natal, South Africa, hill slopes are covered with sediments which are poorly sorted. These colluvial sediments contain clay, sand and gravel sized particles which indicate that the material was transported by sheet erosion (Watson et al. 1984). The colluvial sediments in Natal occur in areas of moderate to high relief and are deposited on the foot slopes which have slopes of about 5°. The hills are of resistant dolerite, but the shallower slopes are thick sandstones, which form part of the Permian Vryheid formation. The sediments are up to 20m thick and they have been cut through by erosion gullies, known as dongas. Figure 7.2.1. shows sites where donga exposures have been described.

The sections show clear evidence of soil forming processes and the palaeosols indicate breaks in the depositional record. The sedimentary sequences are thus of importance in establishing past climatic changes (particularly with regard to rainfall) in this area of southern Africa. These records can be compared with other evidence such as calcrete formation, once a chronology has been established. These partially consolidated, bedded sediments have been formally designated the Masotcheni formation (South African Committee for Stratigraphy, 1980).

The sediments are currently being examined by G.A. Botha (Geological Survey, Pietermaritzburg), and the results are presented in his thesis (Botha, 1992 in preparation). The sediments are thought to have been deposited since the last interglacial, since Early Stone Age artifacts have been found at the base of the type site, Masotcheni, and at other sites. More recent Middle Stone Age artifacts have been found in the overlying colluvium.

The sedimentary succession and buried palaeosols have been interpreted within the framework of a cyclical model (Botha et al. 1990). Each cycle involves a period of colluvial sedimentation, followed by erosion. Studies of the sediments exposed in dongas today have led to the proposal that there have been four such cycles in the last 120ka (Botha, 1992 in preparation).

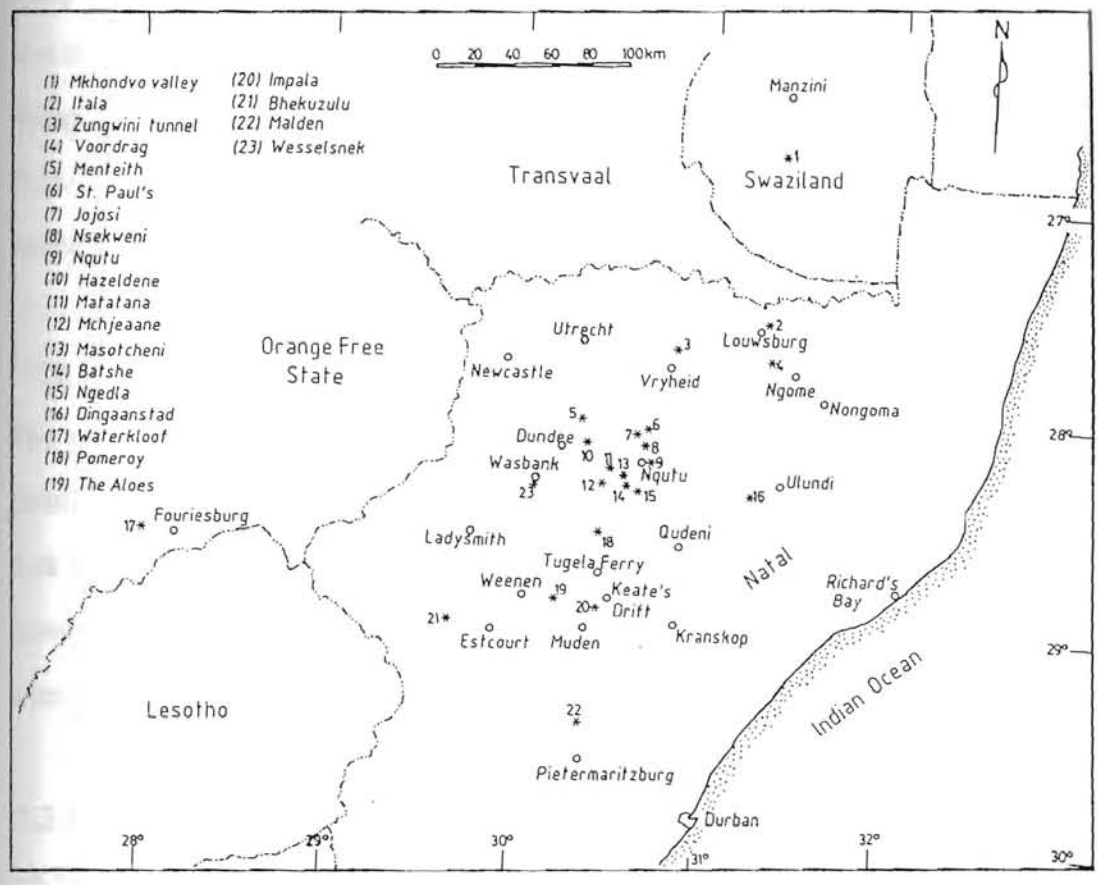


Figure 7.2.1. Site locations (from Botha 1992).

7.2.2. Grain size distribution

Samples used in this study showed a wide range in grain size from larger than 2mm stones to less than 2μm clays. The grains were continuously distributed in the range with very little change from sample to sample. For each sample there were enough grains for coarse grain (125-150μm) and fine grain (4-11μm) dating. The wide range of grain size distribution also indicates that the samples were colluvium as defined by Watson et al. (1984).

7.2.3. Sampling

The presence of clays causes the sample to be very hard and robust when dried. After cleaning at least 20cm into the section surface, large blocks (40x30x30cm) were taken to a dark room. Blocks were cut by bow-saw to remove the exposed layer in a dark room. Blocks (about 15x10x10cm) were covered with two layers of tin foil and black plastic bags. Samples were taken by Mr G.A. Botha and shipped to Aberystwyth.

7.3 EQUIVALENT DOSE DETERMINATION

7.3.1. Procedures

The samples were opened in the laboratory dark room. After removal of the sample surface for at least 2mm (in most cases only the central part of the sample block was used), K-feldspar coarse grain fractions were separated according to the procedures described in chapter 2. The fine grains (4-11μm) were separated only for four samples in the early stage of the study. These four samples are AF-1, AF-2, AF-3 and AF-4. The experimental conditions for these fine grain samples were described in chapter 5. Here I briefly describe the ED determination procedures used for the K-feldspars. Separation and disc making procedures were described in chapter 2.

Measuring the signals

For IRSL ED determination, discs of each sample were separated into several groups. These

groups include: 1. multiple disc additive dose (normally 24 discs); 2. single aliquot (about 20 discs); 3. residuals after different bleachings (about 12 discs).

1. Multiple disc additive dose

The discs were measured with a short shine (0.1 second) of IR before any treatment. This short shine IRSL signal was used for normalization. In order to construct the growth curve, sub-grouped discs, four discs in each group, were irradiated with different ß doses. The doses added to each group were exponentially increasing, e.g. 10, 20, 40, 80, 160 and 320Gy. The largest added dose was selected to be sufficient to generate an IRSL signal which is not less than twice that of the natural signal. After at least 24 hours delay, discs were preheated at 220°C for 10 minutes. The same preheating treatment was employed for the discs used for the residual signals. After more than three days (normally a week) delay, the IRSL signal was measured by a short shine (0.1 second). The filter used was a 2mm Schott BG 39. For very bright samples a neutral density filter was added.

The sample discs were checked for anomalous fading by the same short shine measurement after two or three weeks storage in the dark at room temperature. It was found that the EDs given by both measurements agreed with each other within the error of the measurement, thus indicating the lack of a short term fading component.

The TL signal of this group of discs was measured after more than a month's delay. In some cases the samples were stored in the dark for more than half of an year. The TL was read out in the Riso TL reader. The filters used for the coarse grain K-feldspar TL were a BG 39 and a neutral density filter. Since the reduction of the TL signal by the IRSL measurements is negligible, the same discs were used for both IRSL and TL measurements.

For three relatively young samples, AF-4, STP-13 and HIT-2, the discs used for the IRSL multiple disc additive dose measurements were used for dose normalization, instead of the

TL measurement, in order to indicate incomplete bleaching of those samples (chapter 6). For the second glow normalization, the discs were bleached 5 hours by the SOL2 followed by a test ß dose of 19 Gy. After 24 hours delay, the discs were preheated at 220°C for 10 minutes. The IRSL signal, used for normalization, was than measured with a 1 second IR exposure. For these three samples, a new set of discs was prepared for the TL signal measurement. The procedure was similar to that of the other samples with a TL second glow normalization procedure being employed.

2. Single aliquot

The single disc methods were described in chapter 2. Natural discs were used. The regeneration method for single aliquots was employed for all of the samples, whereas the additive dose method was only applied for the relatively young samples AF-4, STP-13 and HIT-2. The experimental conditions were as follows:

Regeneration

Preheating at 220°C for 10 minutes; measuring the IRSL for 100 seconds at 50°C; bleaching with IR for 1600 seconds at 50°C; ß irradiation for an appropriate dose. This experimental cycle was repeated for each dose point. The irradiation dose was chosen randomly. The ED was obtained by matching the natural IRSL with the IRSL from the constructed regeneration growth curves.

Additive dose

Preheating at 220°C for 10 minutes; measuring the IRSL with 0.5s short shine at 50°C; ß irradiation. This cycle was repeated for different added doses.

A preheating correction file was created from measurements of identical natural discs which had not been given laboratory doses. This was used to correct the loss of IRSL caused by each preheat. The experimental conditions were the same as for the additive dose measurements but without addition of ß dose in each experimental cycle (see Duller 1991 for

details).

3. Discs for residuals

The natural discs of each samples were bleached for three different times by the SOL2 solar simulator. The bleaching times were 10 minutes, 1.5 hours and 15 hours. The effects of these bleaches on the luminescence signals reflect several possible situations in nature. 15 hours bleaching by the SOL2 is equivalent to 90 hours continuous sunlight bleaching; this can be treated as the bleaching of aeolian sediments. 10 minutes bleaching by the SOL2 is equivalent to a 1 hour sunlight bleaching, which may be similar to the bleaching experienced by some alluvial and colluvial deposits; 1.5 hours bleaching results in an intermediate effect. Four discs were used for each bleaching time. After these bleaches and a 24 hours delay, discs were preheated at 220°C for 10 minutes. The IRSL and TL signals were measured with the same conditions as for the multiple disc measurements.

ED calculation

Except for the young sample STP-13, the IRSL and TL signal showed a non-linear response to dose and the data were fitted to a saturating exponential. A computer program written by Dr R Grün for TL dating was used for the ED calculation, but the errors in the ED given by this program were unrealistic compared with those determined by a program written by Dr B.W. Smith (Smith 1983). For most samples a consistently smaller error was shown. However, the error in the ED still reflects the scatter of the data set. For ED comparisons, ED differences of greater than ±10% are taken to be statistically significant. The preheating correction for the single aliquot method was made using software written by G.A.T. Duller. As described elsewhere (Duller 1991), the precision of single aliquot methods is better than 1% for most samples.

The residual IRSL and TL signals after the different bleaches were used in the ED calculations. It was found that the residual IRSL signal after those bleaches had little effect

on the ED obtained. This is due to the rapid bleaching of the IRSL signal. However, the TL residuals were found to be significantly different. After 10 minutes SOL2 bleaching, the TL residual is about 35% of the natural signal; whereas it is about 10% and 4% after bleaching of 1.5 hour and 10 hours, respectively. Different samples showed a slightly different ratio of residual to the natural signal.

7.3.2. Results from coarse grain K-feldspar fractions

The results of the EDs obtained from the IRSL and TL signals are summarised in Table 7.3.1 and Table 7.3.2 respectively. The results for the three young samples will be discussed separately (see below).

Table 7.3.1. EDs obtained with IRSL methods with coarse grain K-feldspar fractions

Section and samples	ED (Gy) multiple disc Add. dose	Regen.	mean ED (Gy) Single aliquot Add. dose	R
St Paul's				
AF-1	256±20	259		0.135
AF-2	298±51	294		0.096
AF-3	226±14	201		0.099
AF-4	25.3±2.0	31.4	28.7	1.4
STP-1	467±24	429		
STP-9	299±44	324	278	0.10
STP-13	9.5±1.3	9.3	6.6	4.0 (0.7)
NEW-2*	519±19	383		0.095
Masotcheni				
NEW-3	460±8	423		0.015
NEW-4	202±5	193		0.024
Nqutu				
HIT-1	244±4	226	215	0.080
HIT-3*	341±13	233		0.153
HIT-4*	236±18	185		0.139
Hazeldene				
HIT-2	46.9±5.1	47.9	45.4	0.20
HIT-5*	403±7	282		0.089
Matatana				
NEW-1	434±27	364		0.062

* shows the samples for which the ED obtained by the multiple additive method disagreed with the ED obtained with the single disc regeneration method.

IRSL

The accuracy of the IRSL dating method can be assessed by comparing the EDs obtained by the different methods. Except for four samples, the EDs obtained by the multiple disc additive dose method are in agreement with those obtained by the single disc regeneration method. As discussed in chapter 5, the IRSL sensitivity will increase after IR bleaching except for poorly bleached samples. This means that the ED obtained by the single disc regeneration method can be regarded as the minimum ED. It has been reported (Li 1989) that the ED may be overestimated when using the multiple disc additive dose method, if the signal contains contributions from traps with significantly different saturation doses. Thus the agreement between EDs obtained by the multiple disc additive dose and the single disc regeneration methods suggests that the EDs obtained by the methods were accurate.

Apart from the three youngest samples, the EDs obtained are relatively large (>150 Gy), and thus the 'remnant dose' (discussed in chapter 6) is not a significant contribution to the measured ED. This is also indicated by the R value for the single disc measurements (R is the ratio between the range of the EDs given by the discs and the minimum ED found for those discs). The R values also represent the maximum ED variation caused by the 'remnant dose'. For most samples R is less than 0.1 (Table 7.3.1.) causing less than 10% variation in the value of ED. However, R is significantly larger for the young samples.

For the older samples, the ED obtained by the single disc additive dose method was only used for comparison. As mentioned in chapter 2, the ED will be underestimated if the IRSL response to the dose is non-linear.

TL

As mentioned in chapter 3, the TL glow curves are in the form of a single peak after preheating at 220°C for 10 minutes. This is regardless of the added ß dose. For analysis the

glow curves are shifted so that the peaks are aligned (around 340°C). The ED was obtained using the integrated TL signal from 275°C to 400°C. Figure 7.3.1. shows typical TL data for a K-feldspar sample.

Table 7.3.2. Comparison of EDs obtained with IRSL and TL signals for the same group of discs. The TL EDs are given for different SOL2 bleaching times.

Section and samples	IRSL ED (Gy)	TL ED (Gy)		
		15hr	1.5hr	10m
St Paul's				
AF-1#	256±20	380±15	366	295
AF-2	298±51	283±18	273	232
AF-3#	226±4	282±16	262	210
STP-9#	299±44	362±29	---	289
NEW-2	519±19	484±38	472	415
Masotcheni				
NEW-3#	460±8	575±12	551	447
NEW-4	202±5	226±12	205	153
Nqutu				
HIT-1#	244±4	290±14	---	246
HIT-3	341±13	304±11	---	197
HIT-4	236±18	251±15	240	201
Matatana				
NEW-1	434±27	456±49	434	338

shows the samples for which the TL ED (using a 15 hr bleach) is significantly larger than that obtained from the IRSL signal. The IRSL ED was for the multiple disc additive dose method in Table 7.3.1.

As expected, the TL EDs varied when different residuals were subtracted. When the residual TL after 15 hours SOL2 bleaching was used for residual subtraction, the TL ED was in agreement using the ED obtained with the IRSL signal (except for samples AF-1; AF-3; NEW-3; HIT-1 and STP-9). This is particularly interesting because the identical discs were used for both TL and IRSL measurements. For those samples having similar IRSL and TL EDs, the TL results support the results from the IRSL signal. Hence the ED value was confirmed by another method for which the luminescence signal was bleached more slowly. For those samples with different EDs, the results can be explained by the poor bleaching of the TL signal prior to deposition.

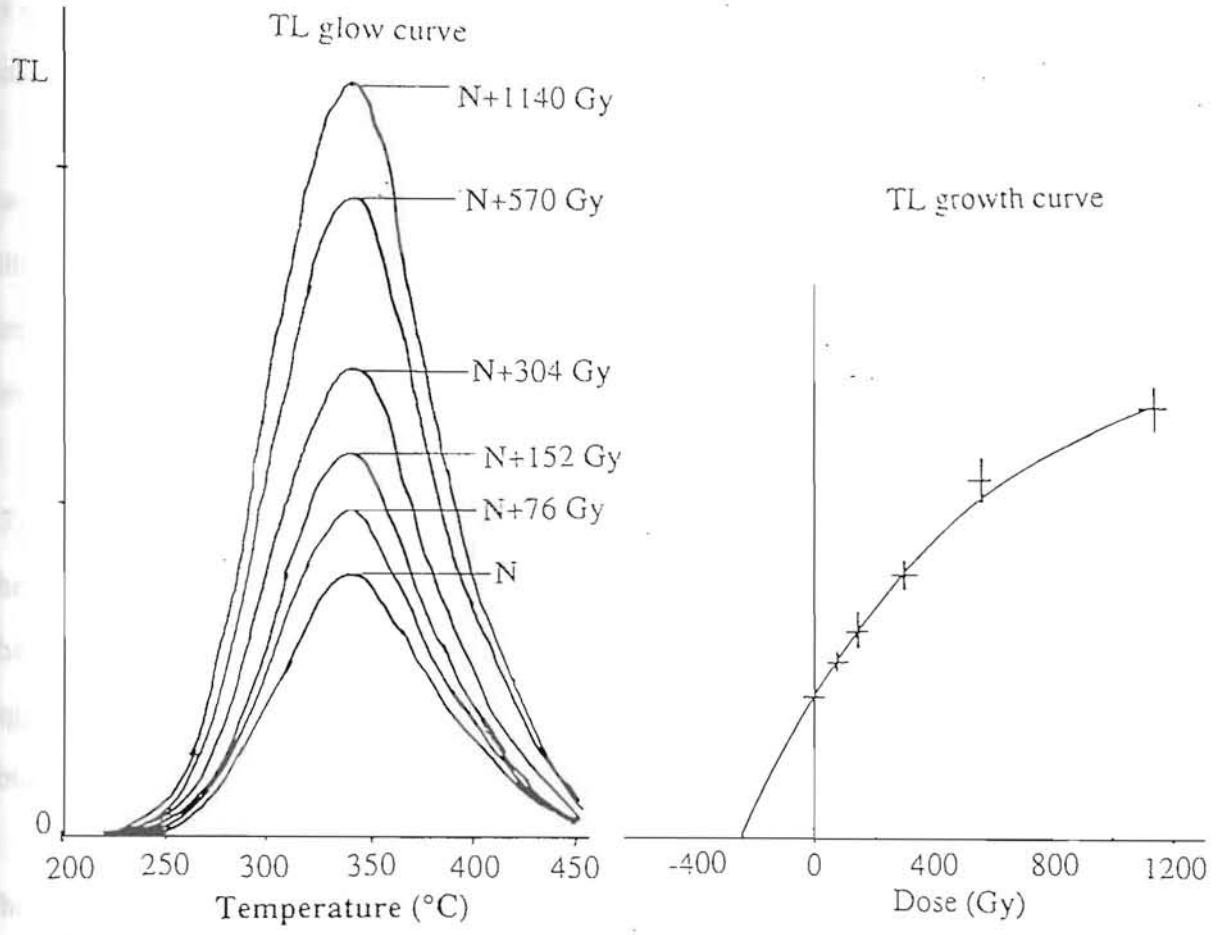


Figure 7.3.1. TL glow curves and growth curve of sample NEW-2. (a) glow curves (b) growth curve for 275-400°C . Glow curves were shifted to align the peaks at about 340°C.

For samples STP-1 and HIT-5, the TL signal was so scattered that no sensible growth curves could be constructed, though reasonable IRSL growth curves were obtained for the discs.

The values of ED obtained by the IRSL multiple disc additive dose method are in agreement with those obtained by either TL or IRSL single disc regeneration (except for sample HIT-5). Hence, the ED given by the IRSL multiple disc additive dose method, is considered as the most accurate and used in the age calculations (section 7.5).

7.3.3. Results from fine grains fractions

The fine grain (4-11μm) method was only applied to samples AF-1, AF-2, AF-3 and AF-4. The results are listed in Table 5.2.1. As with the coarse grain K-feldspar fractions, the results suggest that the ED obtained by the multiple disc additive dose method agreed with the ED obtained by the regeneration method after IR bleaching.

The TL EDs after 15 hours SOL2 bleaching were consistently larger than the IRSL EDs.

7.3.4. Bleaching and EDs obtained by different methods

In previous chapters (5 and 6), I have discussed how bleaching prior to deposition can affect the ED estimation. This can be classified in two ways; one is the sensitivity change due to the bleaching of the H-type charges, the other is the effect of different residual levels for TL and IRSL signals due to the different bleaching rates.

(1) EDs obtained by the additive dose method and single disc regeneration.

For the regeneration method using the IRSL signal, the ED will be underestimated due to the IRSL sensitivity increase after the IR bleaching, except in the case of samples which were bleached for a short period of time, and for which the H-type charges remained constant throughout the different stages of the regeneration procedure. For the additive dose method,

the sensitivity change is likely to be negligible. Hence for an old sample (say >100Gy), the ED obtained by the additive dose method will be significantly larger than the ED given by the regeneration method, if the sample was bleached for a long time prior to deposition; whereas for samples which received a short bleach prior to deposition, the EDs obtained by both methods will be in agreement.

(2) ED obtained with TL and IRSL signals

Since the bleaching rates of the TL and IRSL signals are very different, it is expected that the IRSL ED will be smaller than the TL ED for a poorly bleached sample. Hence, by subtracting the residual after the longer bleaching time (i.e. 15 hr of SOL2), the TL ED will be significantly larger than the IRSL ED. However, the bleaching experiments (see chapter 6) have suggested that this difference in ED can only be demonstrated when the bleaching is not longer than equivalent to 1 hour bleaching with the SOL2. On the other hand, the difference in the ED values is related to the true ED of the sample, because the residual TL signal is also equivalent to an effective TL dose remaining in the sample. This effective dose will be significant for younger samples.

As discussed in chapter 5, the sensitivity change is related to the H-type charges which can only be bleached by prolonged light exposure. Since the easy bleaching part of the TL signal is related to E-type charges, the H-type charges bleach more slowly than the total TL signal.

As far as luminescence dating is concerned, the bleaching condition of sediments can be divided into four categories. The bleaching conditions and subsequent effects on ED determinations are summarized in Table 7.3.3.

Table 7.3.3. Bleaching categories for sediment samples

(A) prolonged bleaching
IRSL ED_{mult} > IRSL ED_{sig} TL ED_{15h} = IRSL ED_{mult}

(B) complete bleaching of IRSL and TL signals; incomplete bleaching of H-type charges
IRSL ED_{mult} = IRSL ED_{sig} TL ED_{15h} = IRSL ED_{mult}

(C) complete bleaching of IRSL signals; incomplete bleaching TL signals and H-type charges
IRSL ED_{mult} = IRSL ED_{sig} TL ED_{15h} > IRSL ED_{mult}

(D) very short bleaching (≤30 s of SOL2); incomplete bleaching of IRSL signals, TL signals and H-type charges
All EDs are overestimated

IRSL ED_{mult} is the IRSL ED obtained with the multiple disc additive dose method.
IRSL ED_{sig} is the IRSL ED obtained with the single disc regeneration method using IR for bleaching.
TL ED_{15h} is the TL ED obtained with the multiple disc additive dose method after subtraction of the signal remaining after 15 hours SOL2 bleaching.

These effects are shown clearly for the ED results of the coarse grain K-feldspar fraction of the older samples. For samples NEW-2, HIT-3, HIT-4 and HIT-5 (Table 7.3.1.) using the IRSL signal, the EDs obtained by the multiple disc additive dose method are significantly larger than those obtained by the single disc regeneration method; whereas the rest of the samples showed agreement of the EDs. This suggests that the samples with the differing EDs might have been well bleached prior to deposition, whereas the others might only have received a short bleach. For the samples for which the IRSL EDs were already in disagreement, comparison of the ED results for the TL and IRSL signals (Table 7.3.2.) showed that agreement was obtained when the residual after 15 hours of bleaching was subtracted. This indicates complete bleaching of the TL signal, the IRSL signal and the H-type charges. Samples NEW-2, HIT-3, HIT-4 and HIT-5 thus belong to category A.

For those samples which showed agreeing IRSL EDs, the TL ED and IRSL ED were also in agreement for samples AF-2, NEW-1 and NEW-4 (Table 7.3.2). This suggests that these samples experienced light exposures which erased the TL and IRSL signals completely, but did not remove the H-type charges. These samples belong to category B.

For most of the rest of the samples (already showing agreement of the IRSL EDs) the TL ED and IRSL ED were significantly different, namely AF-1, AF-3, STP-9, NEW-3 and HIT-1 (Table 7.3.2). This also suggests poor bleaching of these samples prior to deposition but places them in category C. The TL EDs obtained using the residual after 10 minutes or 1.5 hours bleaching also gives information about the possible bleaching conditions.

Samples in category D are relatively rare. For such a sample, an unrealistically large IRSL ED (and thus age) would be obtained. Sample AF-4 may belong to this category since there are other indications of a very short light exposure experienced by this sample (see 7.3.5).

Although the use of the same group of discs for TL and IRSL measurements can minimize some differences due to different treatments on the discs, and the precision of the single aliquot method is remarkably high, the deduction of bleaching conditions from the comparison of the EDs depends strongly on the accuracy of the ED determinations. The ED obtained by the IRSL multiple disc additive dose method plays an important role in these deductions and a few cautionary comments should be made. The ED can be mis-calculated in several ways, such as by use of an inappropriate fitting program, using doses which were not high enough, high disc-to-disc scatter, inappropriate normalization procedures and anomalous fading etc.. Since these results can be obtained from routine dating measurements, the information leading to the deduction can be derived without extra experiments.

7.3.5. The younger samples

As discussed in chapter 6, for the relatively young samples AF-4, STP-13 and HIT-2, the IRSL signal was not completely bleached at deposition. Therefore, the ED would have been overestimated if conventional methods were used. For these three samples, using the minimum ED (EDmin) and the ED obtained by the ED-I_n (ED') method may be the best approaches (as discussed in chapter 6). These EDs are used in the age calculations in section 7.5. Table 7.3.4. lists the EDs obtained using IRSL and TL signals with different methods.

Table 7.3.4. Equivalent dose (Gy) obtained from IRSL and TL signals for the K-feldspar fractions of three young samples. Different sets of discs were used for the TL and IRSL measurements.

IRSL

sample	multiple disc additive dose	single disc			ED-I_n method
		Regen. (mean)	add. dose (mean)	EDmin	ED'
AF-4	25.3±1.99	31.4	28.7	15.1	10.2
STP-13	9.54±1.34	9.31	6.63	2.50	1.61
HIT-2	46.9±5.1	47.9	45.4	41.2	35.6

TL

sample	bleaching time		
	15hr	1.5hr	10m
AF-4	133±4	126	103
STP-13	16.1±0.9	12.3	9.5
HIT-2	171±12	133	57

As expected, the EDs given by the TL signal are substantially larger than those given by the IRSL. This was shown clearly for all three samples. Even using the residual signal remaining after 10 minutes of SOL2 bleaching, the ED is still overestimated. This provides further support for the suggestions of poor bleaching drawn in the previous chapter.

The TL EDs show most overestimation for sample AF-4. The TL ED after 15 hours bleaching is more than five times higher than that obtained for the IRSL signal. Because the multiple disc additive dose method was used for both signals, such a large difference in ED indicates that the grains from AF-4 were probably exposed to very little sunlight prior to deposition. It is also possible that the values of ED' and EDmin might be larger than the true ED.

7.4 DOSE RATE ESTIMATION

7.4.1. Results of thick source alpha counting

Thick source alpha counting was used for all bulk samples. This determines the radioactivity of U and Th for the materials surrounding the grains. Alpha counting measurements were made with both the sample being sealed and unsealed. Table 7.4.1. lists the results. The count

rate listed was the counting result when the sample was unsealed (open to the air).

Table 7.4.1. Thick source alpha counting results of bulk samples. The count rate was the unsealed rate. The U and Th concentrations are calculated assuming secular equilibrium.

sample	count rate (per ks*cm^2)	Th (ppm)	U (ppm)	seal/unseal	Th/U
AF-1	1.35±0.02	14.3±1.8	7.07±0.56	0.97	2.02
AF-2	1.19±0.02	11.3±1.6	6.63±0.50	1.05	1.70
AF-3	1.50±0.02	14.7±1.9	8.23±0.59	1.05	1.79
AF-4	1.00±0.02	12.7±1.6	4.61±0.49	1.20	2.75
NEW-1	0.96±0.02	11.6±1.6	4.57±0.48	1.08	2.52
NEW-2	1.43±0.02	20.9±2.1	5.78±0.63	1.00	3.61
NEW-3	0.69±0.01	6.54±0.79	3.82±0.25	0.97	1.71
NEW-4	1.08±0.02	10.6±1.8	5.95±0.57	0.97	1.78
STP-1	0.96±0.01	11.1±1.3	4.81±4.02	1.00	2.31
STP-9	1.22±0.01	13.5±1.2	6.24±0.37	0.95	2.16
STP-13	1.36±0.02	14.6±1.6	7.10±0.49	1.00	2.06
HIT-1	1.01±0.02	12.5±1.9	4.79±0.59	0.98	2.61
HIT-2	0.89±0.02	13.6±1.6	3.49±0.49	1.05	3.90
HIT-3	0.66±0.01	7.29±1.16	3.34±0.36	1.00	2.18
HIT-4	1.70±0.03	21.3±2.81	7.98±0.87	0.98	2.67
HIT-5	1.01±0.02	16.4±2.23	3.60±0.69	0.92	4.56

Although radon escape is difficult to detect without alpha or gamma spectrometry measurements, an indication of a problem may be obtained by looking at the count rate ratio between sealed and unsealed sample measurements (Aitken 1985). The results given in Table 7.4.1 indicate that radon escape is not significant, except for sample AF-4.

The alpha count rate was used for calculation of the alpha, beta and gamma dose rates assuming the usual Th/U ratio. When the count rate is converted into the effective dose rate, the dose rate distribution is related to the Th/U ratio of the sample (Aitken 1985). For the effective alpha dose rate, the upper limit error is not more than 2% for a sample whose alpha activity is calculated as being either all from thorium or all from uranium (Aitken 1985). The beta dose rate dependence on Th/U ratio is about 20%. The dependence of Th/U ratio for the gamma dose rate is about the same as in the case of the beta dose rate, but in the opposite sense. Hence, the total dose rate derived from alpha counting is largely independent of the

Th/U ratio. Nevertheless, the variation of the Th/U ratio in these samples may suggest that radioactive disequilibrium might be present in some of samples. Sample HIT-5 shows a Th/U ratio of 4.56, which is much higher than the commonly found ratio of 3.4, whereas most samples show a lower Th/U ratio. This could be studied in more detail by using a high resolution gamma spectrometer or alpha spectrometer.

The alpha counting results indicate that these sediments have a relatively high U and Th concentration. The average count rate of these samples is about 30% higher than Chinese loess, and 50% higher than for loess from Europe (Wintle 1987). This means that the dose rate will be dominated by the contribution from the sediments surrounding the grains. The cosmic ray contribution is minor.

Thick source alpha counting was also used to evaluate the alpha dose contribution of the K-feldspar coarse grains. This will be discussed in section 7.4.6.

7.4.2. Beta counting results

Further comparisons of dose rate were made using a thick source beta counting system (Sanderson 1988). The bulk samples were crushed and 15g of sample was used for each measurement. Table 7.4.2. shows the results and the comparison with those calculated from the results of thick source alpha counting and potassium content measurements.

Table 7.4.2. Beta dose rate from thick source beta counting compared with the beta dose rate calculated from thick source alpha counting and potassium measurements.

sample	ß dose rate from ß counting (Gy/ka)	ß dose rate calculated from α counting and K_2O (Gy/ka)
AF-1	2.60	2.79
AF-2	2.99	3.06
AF-3	2.52	2.79
AF-4	2.31	2.28
NEW-1	2.01	2.20
NEW-2	2.51	2.44
NEW-3	1.48	1.85
NEW-4	1.40	1.94
STP-1	2.91	2.67
STP-9	2.81	2.91
STP-13	2.41	2.95
HIT-1	1.97	2.21
HIT-2	2.29	2.43
HIT-3	1.90	1.93
HIT-4	2.86	3.19
HIT-5	2.84	2.80

For most samples the dose rate determined by the thick source beta counting agreed (within ±10% error) with that calculated from the results of alpha counting and potassium content for dry samples. These independent measurements give a cross check on the results. The result from alpha counting and K content were used for the age calculation.

7.4.3. H_2O content

The radiation dose will be attenuated by the presence of water (Aitken 1985). It is thus necessary to estimate the past water content of the samples. Because the samples were shipped to Aberystwyth, the present day water content was not able to be measured. Even if the present moisture had been measured, it might have varied considerably in the past for this type of colluvial sediment. An assumption of 10±5% of water content was made for dose rate calculation.

7.4.4. K_2O content

The potassium content of bulk samples was measured in an atomic absorption spectrometer (AAS). For the K-feldspar coarse grains fractions, a GM multicounter system was used (Bøtter-Jensen and Mejdahl 1988) and the measurements were kindly made by Dr Mejdahl and Dr McKerrell. Table 7.4.3. lists the results.

The K_2O content for the feldspar fractions is below 10% for all of these samples. This is lower than that found in samples from other parts of the world (Mejdahl 1988b), which is about 10%-14%. The reasons for this low K_2O content is not clear. However, because of the relatively high radioactive content of the bulk sample, the total dose rate has a relatively small contribution from the K_2O content of the grains, even if the luminescence signals are dominated by the grains with a high potassium content. The error caused by uncertainty in the K_2O content is less than 10% of the total dose rate.

7.4.5. Cosmic rays

Cosmic rays makes a very small percentage contribution to the total dose rate. Most samples were taken from several meters below the original land surface. 0.15 Gy/ka was used in the calculation of the dose rate according to Prescott and Hutton (1988).

7.4.6. Internal alpha irradiation

Measurements of the internal radioactivity of feldspar and quartz grains have been reported by Mejdahl (1987) who concluded that the total dose rate contribution from the Rb, U and Th content of quartz and feldspar grains is significant and must be taken into account. Although the U and Th contents in the grains makes an insiginificant contribution to the beta and gamma doses, the internal alpha dose can make an important contribution to the total dose rate.

All sixteen samples K-feldspar coarse grain fractions were crushed in a ball mill and

measured in the alpha counting system. Since the amount of K-feldspar grains was limited, smaller ZnS (Ag) screens (1.6cm diameter) were used. The screen area for counting was 2.0 cm^2. The sample weight used for each measurement was about 0.4g.

Table 7.4.3. Summary of dose rate evaluation for K-feldspar fractions.

samples	surrounding		K-feldspar grains		D_{in} (α+ß dose)*	total dose rate
	α counting (c / ks*cm^2)	K_2O (%)	α counting (c / ks*cm^2)	K_2O (%)	(Gy/ka)	(Gy/ka)
St Paul's						
AF-1	1.35±0.02	1.76	0.064	3.2	0.42	4.96±0.37
AF-2	1.19±0.02	2.52	0.100	2.7	0.65	5.34±0.37
AF-3	1.50±0.02	1.84	0.034	2.8	0.22	4.84±0.38
AF-4	1.00±0.02	1.80	0.047	2.6	0.31	4.01±0.33
STP-1	0.96±0.01	2.32	0.046	2.0	0.30	4.38±0.34
STP-9	1.22±0.01	2.2	0.116	4.0	0.76	5.31±0.35
STP-13	1.36±0.02	1.6	0.043	1.3	0.31	4.94±0.38
NEW-2	1.43±0.02	2.0	0.087	4.9	0.57	5.35±0.39
Masotcheni						
NEW-3	0.69±0.01	1.56	0.072	3.0	0.47	3.40±0.26
NEW-4	1.08±0.02	1.2	0.044	2.2	0.29	3.59±0.32
Nqutu						
HIT-1	1.01±0.02	1.68	0.128	6.4	0.84	4.62±0.34
HIT-3	0.66±0.01	1.72	0.052	7.8	0.34	3.56±0.28
HIT-4	1.70±0.03	2.16	0.036	8.0	0.23	5.78±0.46
Hazeldene						
HIT-2	0.89±0.02	2.16	0.050	5.4	0.33	4.29±0.33
HIT-5	1.01±0.02	2.64	0.229	7.1	1.50	5.97±0.35
Matatana						
NEW-1	0.96±0.01	1.76	0.051	2.9	0.33	3.93±0.32

note: (α+β dose)* is the equivalent β dose rate of internal alpha dose rate and internal β dose rate after correction for attenuation; it dose not include the internal dose rate of the potassium content.

The alpha efficiency was measured for fine grain (4-11μm) fractions of the K-feldspar grains of sample NEW-3 prepared by crushing in a ball mill. The discs were prepared as for fine grain samples. The measured alpha efficiency (k-value) was 0.15±0.02 from the comparison of the IRSL growth curves after beta and alpha irradiation (Aitken 1985). It is assumed that the alpha efficiency is the same for all samples.

Each alpha count rate was converted into effective internal alpha and beta dose rates, after invoking the alpha efficiency and beta dose attenuation factors (Mejdahl 1979), respectively. The alpha count rate and its equivalent dose rate contribution to the total dose rate are listed in Table 7.4.3. As can be seen, most of the feldspar separates have a relatively low alpha count rate compared to that of the bulk sample. In these cases, the internal alpha dose will make only a small contribution to the total dose rate. For a few samples, e.g. HIT-1, the alpha count rate was as high as 11% of the bulk sample alpha count rate. Sample HIT-5 also had a high internal alpha count rate. For those samples the internal alpha dose and the beta dose from the U and Th content are important contributions to the total dose rate.

7.4.7. Discussion

The major part of the dose rate was derived from the sediment surrounding the grains because of the higher radioactivity of the bulk samples.

Several possible sources of error should be mentioned, however:-

1. error in potassium content of the coarse grains. If the IRSL signal is dominated by that from grains with a high potassium content, the dose rate could be underestimated by 5-10%. This will not affect the results for the fine grain (4-11μm) samples.

2. external alpha dose. For the coarse grain samples, it was assumed that there was no external alpha dose contribution on the basis that the alpha irradiated layer, on the outside of the grain, was removed by the HF etching. If this was not the case, a dose rate underestimation would result.

3. water content variation. Estimating the past water content is difficult. Although a 10±5% of water content was assumed, for some samples the actual water content might have been larger than the error given, which itself results in a 5% error in the age.

7.5 DATES AND SITE DESCRIPTION

7.5.1. Introduction

In this study of colluvium from northern Natal, samples were collected from five sites - St Paul's, Nqutu, Masotcheni, Hazeldene and Matatana. They are shown in Fig. 7.5.1 (from Botha 1992) as sites a-e, respectively.The other sites listed in Fig. 7.5.1 also have radiocarbon dates on materials reported to be from the colluvial sediments of the Masotcheni formation.

The luminescence dates of the 16 samples are listed in Table 7.5.1. In this section they are presented site by site. Apart from three young samples of Holocene age (STP-13, AF-4 and HIT-2), the samples fall within the last interglacial/glacial cycle. As discussed in previous sections, the EDs were obtained by different methods for coarse grain K-feldspar separates. In addition, fine grain IRSL dates were also obtained for samples AF-1, AF-2, AF-3 and AF-4.

The dates will be discussed with reference to the available radiocarbon dates on organic matter from buried palaeosols.

7.5.2. St Paul's section

7.5.2.1. The section

The St Paul's section is adjacent to the St Paul's Mission Village, about 40 km southwest of Vryheid. The modern donga system is extensive with many side gullies joining the main donga which is about 1 km long (Wintle pers. comm.). The sections are about 25m in height and along their length show evidence of palaeosols indicating periods of much reduced sediment accumulation, and palaeodongas, indicating periods of erosion. These are indicated schematically on Figure 7.5.2. This section was the most intensely studied with 8 samples being dated by luminescence methods.

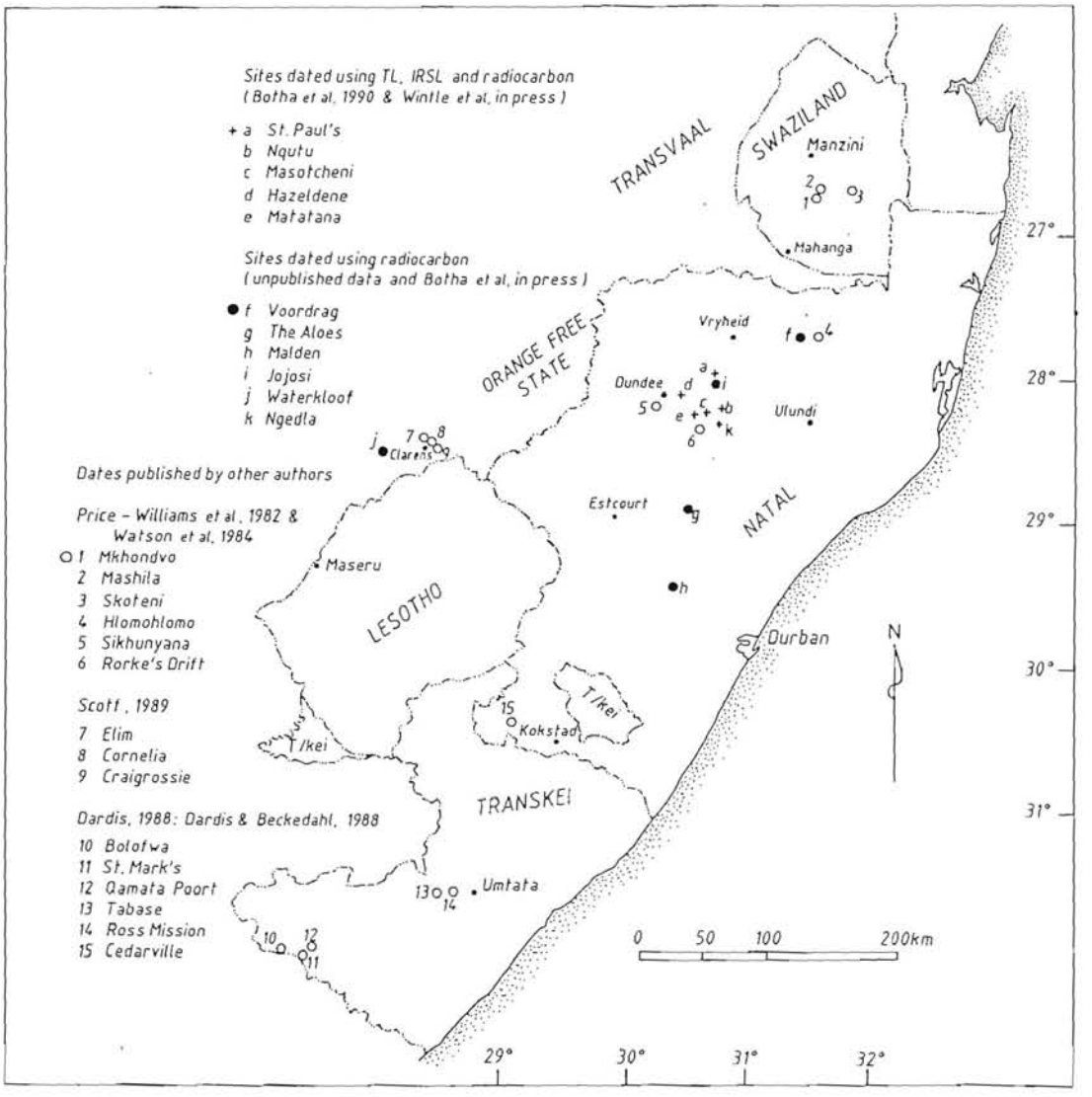


Figure 7.5.1. Geological sections (a-e) studied with luminescence dating methods (from Botha 1992)

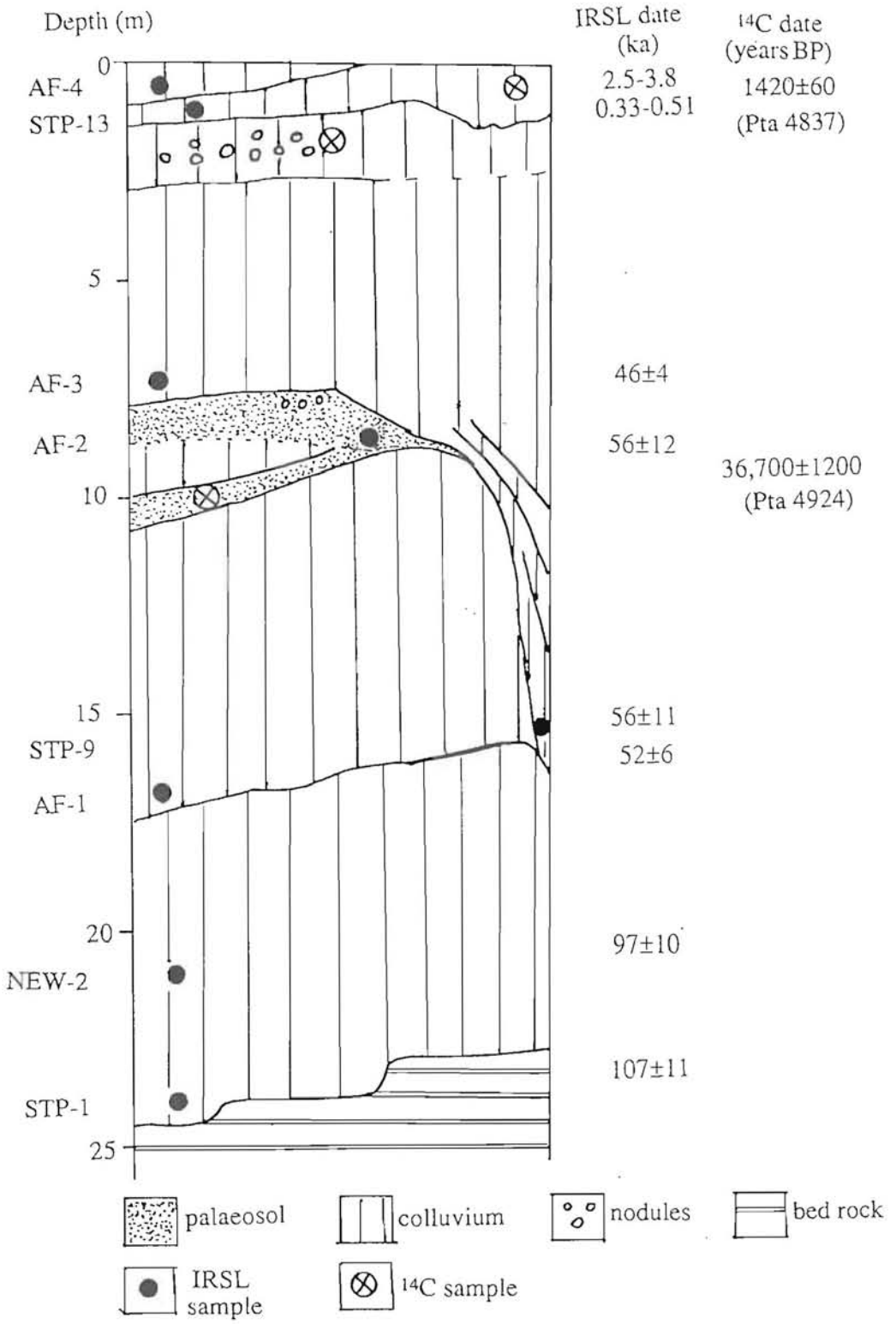


Figure 7.5.2. Schematic section showing the positions of the samples from the St Paul's sections.

Table 7.5.1. Summary of IRSL ages for the K-feldspar separates. The EDs were those obtained with the multiple disc additive dose method except for the three younger samples. The dose rate is from table 7.4.3.

Site and sample	ED (Gy)	Dose rate (Gy/ka)	Age (ka)
St Paul's			
AF-4@	10.2	4.01±0.33	2.5 (**3.77**)
STP-13@	1.61	4.94±0.38	0.33 (**0.51**)
AF-3	226±4	4.84±0.38	46±4
AF-2	298±51	5.34±0.37	56±12
AF-1	256±20	4.96±0.37	52±6
STP-9	299±44	5.31±0.35	56±11
NEW-2	519±19	5.35±0.39	97±10
STP-1	467±24	4.38±0.34	107±11
Masotcheni			
NEW-4	202±5	3.59±0.32	56±6
NEW-3	460±8	3.40±0.26	135±14
Nqutu			
HIT-4	236±18	5.78±0.46	41±5
HIT-1	244±4	4.62±0.34	53±6
HIT-3	341±13	3.56±0.28	96±10
Hazeldene			
HIT-2	35.6	4.29±0.33	10.9±1.6
HIT-5	403±7	5.97±0.35	68±7
Matatana			
NEW-1	434±27	3.93±0.32	110±13

@ For samples AF-4 and STP-13, the ages are derived from ED' and EDmin (in bold).

At the top of the section is a grey, gleyed soil which was buried by 1.5m of recent colluvium. Organic matter from this soil gave a radiocarbon age of 1420±60 years BP (Pta-4837). Sample AF-4 was taken from the overlying colluvium and STP-13 was taken from the base of the unit in which the soil is developed.

The remaining samples were taken with regard to the different units which are separated by palaeosols or erosional surfaces (Figure 7.5.2). One sample (STP-9) was accidentally collected from the fill of a palaeodonga.

7.5.2.2. Luminescence dates for younger samples

The age limits for AF-4 and STP-13 are 2.5-3.8 ka and 0.33-0.51 ka (Table 7.5.2), respectively. These results were given for the coarse grain K-feldspars using ED' and EDmin values for the single disc determinations as discussed in chapter 6. The ages derived from the EDmin and ED' may be considered as the most likely ages but should be regarded as preliminary. As discussed in section 7.3.5 and shown in Table 7.3.3, the TL results indicate that the grains from samples AF-4 were probably exposed to very little sunlight prior to deposition. This would account for their older age compared with the underlying radiocarbon dated soil and the younger age for STP-13. An IRSL age overestimate of a few thousand years will have little effect on the ages of older samples with a similar depositional history; however, the larger age overestimate obtained for the TL measurements (which assumed the grains were well bleached) would cause substantial problems even for the older samples.

Table 7.5.2. Summary of IRSL ages (ka) from different ED determinations for three young samples.

sample	multiple disc	single disc			ED-I_n method
	additive dose	Regen. (mean)	add. dose (mean)	EDmin	
AF-4	6.3±0.8	7.8	7.2	3.8	2.5
STP-13	1.9±0.3	1.9	1.34	0.51	0.33
HIT-2	10.9±1.6	11.2	11.6	9.6	8.3

The age range of 0.33-0.51 ka for STP-13 seems on the young side when compared to the radiocarbon date of 1420±60 years BP (Pta-4837) for the overlying soil. The latter date is

likely to represent the end of a soil forming process in the late Holocene, and as such, the sediment in which it is formed might be expected to be several hundred years older.

Comparison of the TL EDs for STP-13 and AF-4 suggests that the latter is not well bleached; the TL ED for AF-4 is an order of magnitude larger than that for STP-13.

If the IRSL ages were calculated using the EDs obtained with multiple disc method from Table 7.3.3, ages of 1.9±0.3 ka and 6.3±0.5 ka would be obtained for STP-13 and AF-4 respectively.

7.5.2.3. Fine grain IRSL dates

For the first four samples studied (AF-1, AF-2, AF-3 and AF-4), age determinations were also carried out on the fine grain (4-11μm) mixed mineral fraction. For the youngest sample AF-4, the multiple disc additive dose IRSL method gave an age of 5.3±0.3 ka, also demonstrating the age overestimate. The other three samples gave ages of about 36 ka (see Table 7.5.3.), in agreement with the radiocarbon date of 36,700±1200 years BP (Pta 4924). This date is probably best considered as a lower limit and for this reason, the agreement with the fine grain IRSL dates was considered fortuitous.

Table 7.5.3. Summary of dose rates and IRSL ages for fine grain (4-11μm) fractions. The EDs are given for the multiple disc additive dose method .

SAMPLE	alpha count ($c/ks*cm^2$)	K_2O (%)	Dose rate (Gy/ka)	ED (Gy)	Age (ka)
AF-4	1.00±0.02	1.80	5.61±0.15	30±1	5.3±0.3
AF-3	1.50±0.02	1.84	6.31±0.18	225±10	36±3
AF-2	1.19±0.02	2.52	6.83±0.18	248±21	36±3
AF-1	1.35±0.02	1.76	6.47±0.18	240±22	37±3
STP-1	0.96±0.01	2.32	6.06±0.15	515±21	85±7

For comparison, an IRSL ED determination was also carried out on the fine grain fraction of sample STP-1, using the multiple disc additive dose method. An age of 85±7 ka is given which is significantly younger than the IRSL age of 107±11 ka obtained for the K-feldspar

coarse grain (125-150μm) fraction (Table 7.5.1). This difference in IRSL ages may be due to an age underestimation related to the instability of the IRSL signal for fine grains (section 7.5.7).

7.5.2.4. Coarse grain IRSL dates

The ages obtained for samples AF-3, AF-2 and AF-1 using the multiple disc additive dose IRSL method are 46±4, 56±12 and 52±6 ka, respectively (Table 7.5.1). These suggest that the two upper major sedimentary units were deposited within a relatively short time and that the dark palaeosol was probably formed around 50-55 ka. This suggests that the radiocarbon age of 36,700±1200 years BP (Pta 4924) is probably a minimum estimate. Around 50-55 ka there was also erosion resulting in the formation of a donga which was rapidly refilled, as suggested by the date of 56±11 ka for STP-9. This is analogous to the situation at the top of the section where the modern donga cuts through the 1400 years old Late Holocene soil. The 10% age uncertainty precludes any further details of these events at the beginning of Oxygen Isotope Stage 3.

The dates from the lowest major colluvial unit at this site are 97±10 and 107±11 ka for NEW-2 and STP-1 respectively. These dates suggest that it was deposited after the end of Oxygen Isotope Sub-stage 5e (the last interglacial).

7.5.3. Nqutu section

Nqutu is about 30 km south southeast of the St Paul's section. The donga section has a maximum height of about 20m and extends about 160m. It shows evidence of 3 phases of colluvial deposition, past erosion and palaeosol formation. Figure 7.5.3 is a schematic section showing the three samples (HIT-4, HIT-3 and HIT-1), one from each colluvium. The oldest sediment (red), HIT-3, gives an IRSL age of 96±10 ka. HIT-1, a red sediment from an overlying colluvium filling a palaeodonga, gives an IRSL age of 53±6 ka. This colluvium overlies another palaeosol (in the adjacent donga), for which the organic matter is dated at

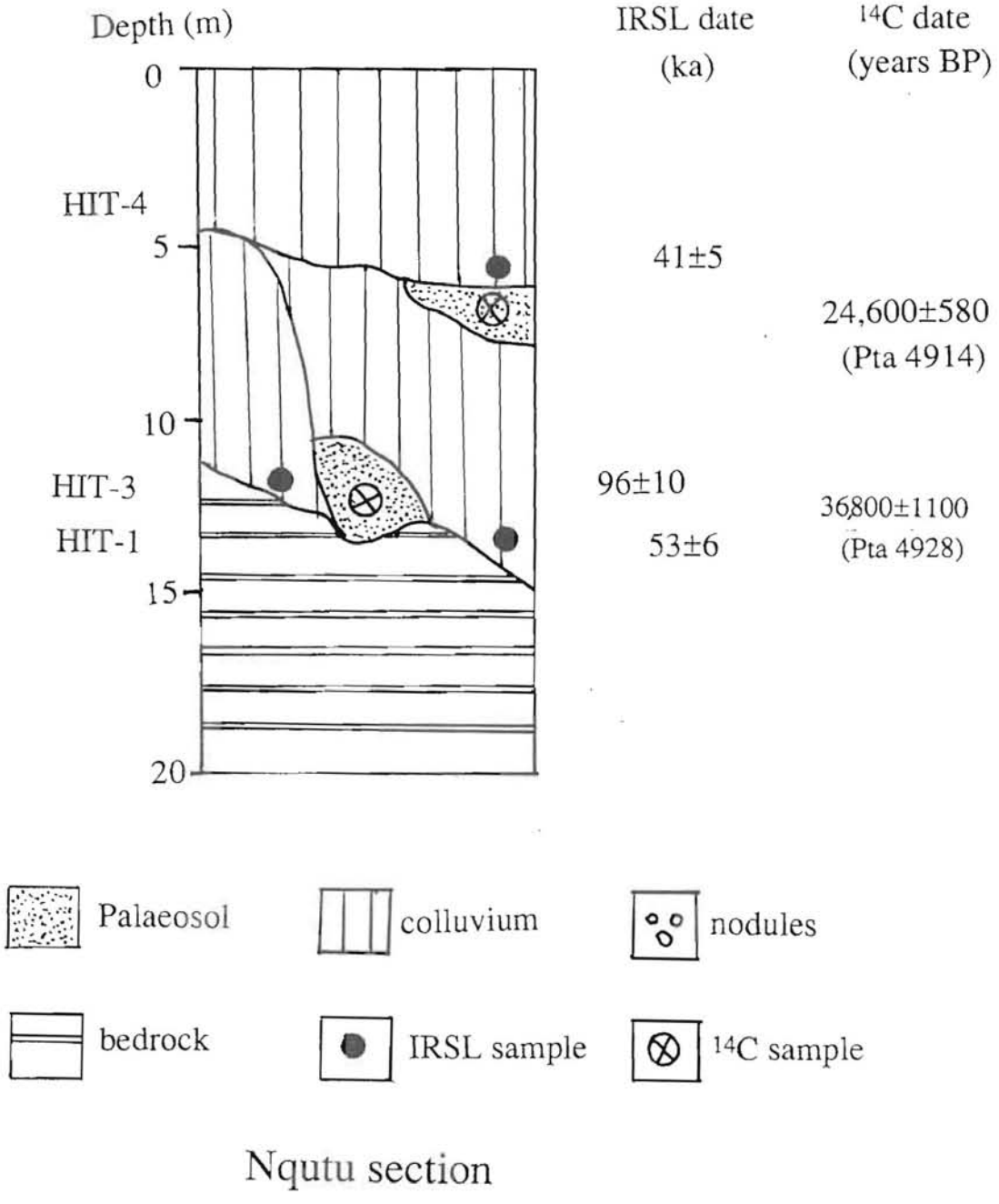


Figure 7.5.3. Schematic section showing the positions of the samples from the Nqutu section.

36,800±1100 years BP (Pta 4928). This is also likely to be a limiting radiocarbon date.

At the top of this colluvium is a dark grey palaeosol, dated at 24,600±580 (Pta 4914). The overlying sample, HIT-4, gave an IRSL age of 41±5 ka. It was taken from a light grey, leached horizon (E-horizon) overlying the palaeosol. Hence a discrepancy is observed for this section.

7.5.4. Masotcheni section

Masotcheni is about 10 km due west of Nqutu. It is the typesite for the Masotcheni formation and the section extends for 500 km as exposed in the donga. No organic-rich palaeosol is found at this site and there is no evidence of palaeodonga formation. Three colluvial units overly each other with a total maximum depth of 15 m (Fig. 7.5.4). Two samples were taken from the two lower units; NEW-3 at the base of the deepest part of the section gave an IRSL age of 135±14 ka and NEW-4 from the base of the overlying colluvium gave an IRSL age of 56±6 ka.

7.5.5. Hazeldene section

Hazeldene is about 20 km north west of Masotcheni. Three colluvia are exposed by a complex modern donga system along a 130m profile. The colluvial sediments are not so thick as at other sites, with an overall height of only 8m (Fig.7.5.5.). There is evidence of palaeodonga formation and there is a well developed dark grey palaeosol. This has a radiocarbon date of 14,070±290 years BP (Pta 4883) for soil organic matter. Two samples were collected. HIT-2 was from the yellow/ brown sediment which overlies the palaeosol and gave an IRSL age of 10.9±1.6 ka (9.6-8.3 ka for the single disc EDmin and the ED-I_n plot methods, see Table 7.5.2). This is in excellent agreement with the radiocarbon date. The second sample, HIT-5, was from a red-mottled colluvium which underlies the dark grey palaeosol. It gave an IRSL age of 68±7 ka.

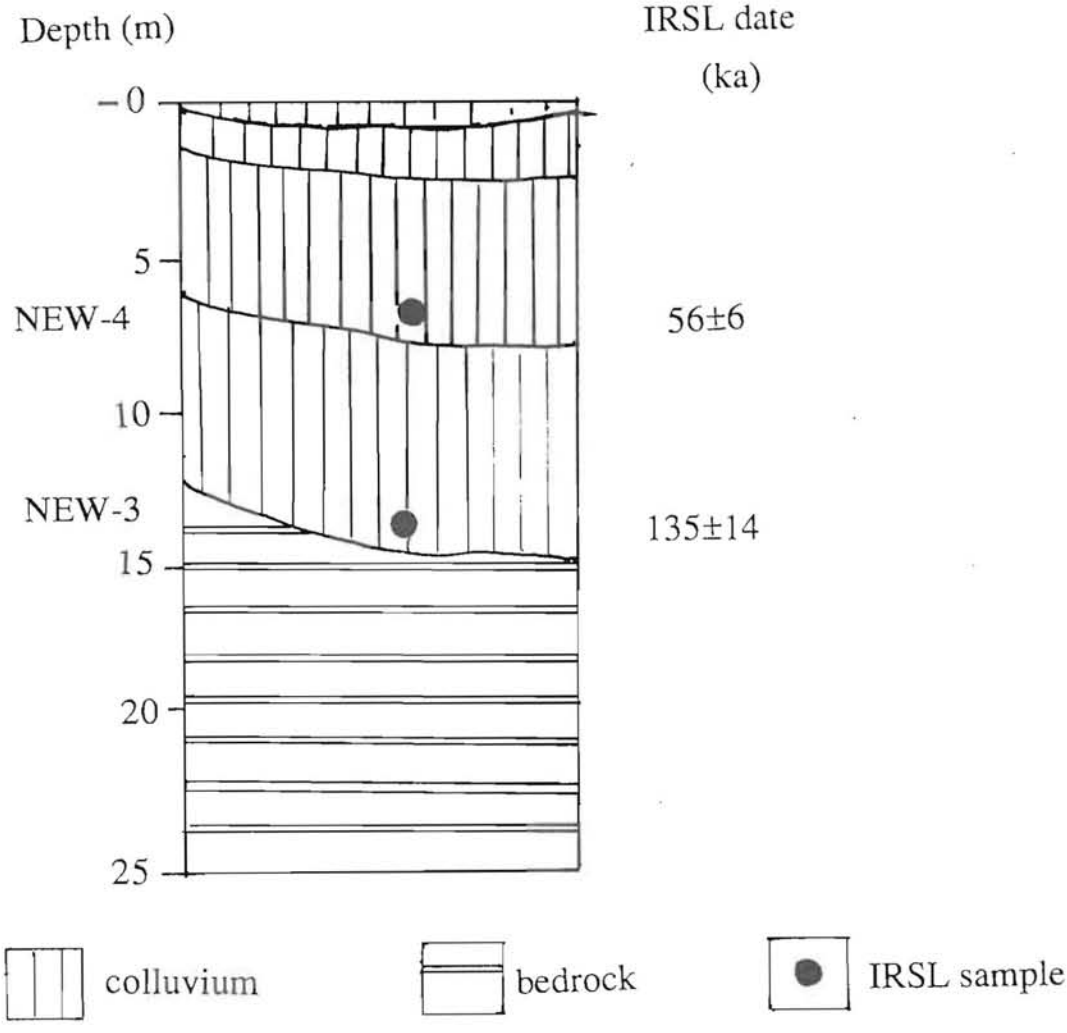


Figure 7.5.4. Schematic section showing the positions of the samples from the Masotcheni section.

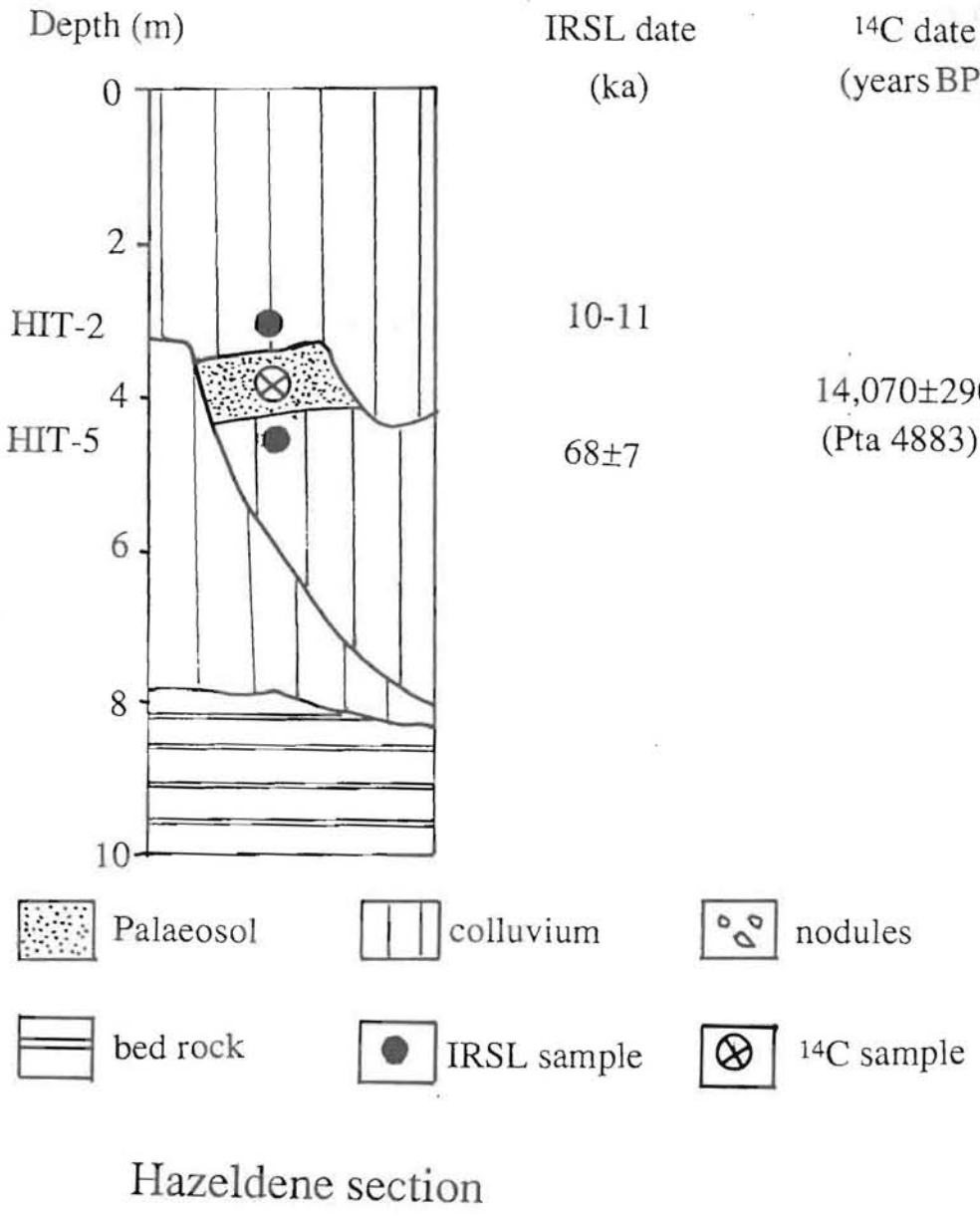


Figure 7.5.5. Schematic section showing the positions of the samples from the Hazeldene section.

For sample HIT-5, the multiple disc IRSL ED has not been confirmed by other methods as for other samples. The Th/U ratio for the bulk sample was 4.56 which is much higher than the secular ratio. Therefore, the IRSL age was considered as a preliminary result.

7.5.6. Matatana section

Only one sample was collected (NEW-1) from the base of the colluvial sequence and its IRSL age is 110±13 ka. No section diagram is given.

7.5.7. Discussion on the age differences between fine grain and coarse grain fractions

For five samples from the St Paul's section, the IRSL age determinations were carried out for both fine grain (4-11μm) and coarse grain (125-150μm) fractions. Apart from the younger sample AF-4, the samples (all older than 36 ka) showed a discrepancy in the ages obtained for those two fractions. The age differences may be explained by the different stability of the IRSL signals, although the mis-calculation of the dose rate for the coarse grain fraction might enhance the age difference (section 7.4.7).

In chapter 3, I have discussed the thermal stability for the fine grained mineral mixture from Chinese loess and the K-feldspar from New Zealand beach sand. Since these sediments were well sorted, it was not possible to obtain the two grain size fractions from the same sample for direct comparisons of the thermal stability. However, the colluvial deposites contain a wide range of grain sizes and enable some comparisons to be made.

The thermal stability of the 4-11μm mixed mineral and the 125-150μm K-feldspar fractions can be revealed by comparing their quick heating curves. Figure 7.5.6. shows the results for the natural signals of both fractions for sample AF-3 compared with the fine grains of loess (88-12) from China. As can be seen, the K-feldspar fraction shows more resistance to heat than the fine grain fraction indicating a more stable signal. Also the signals from both

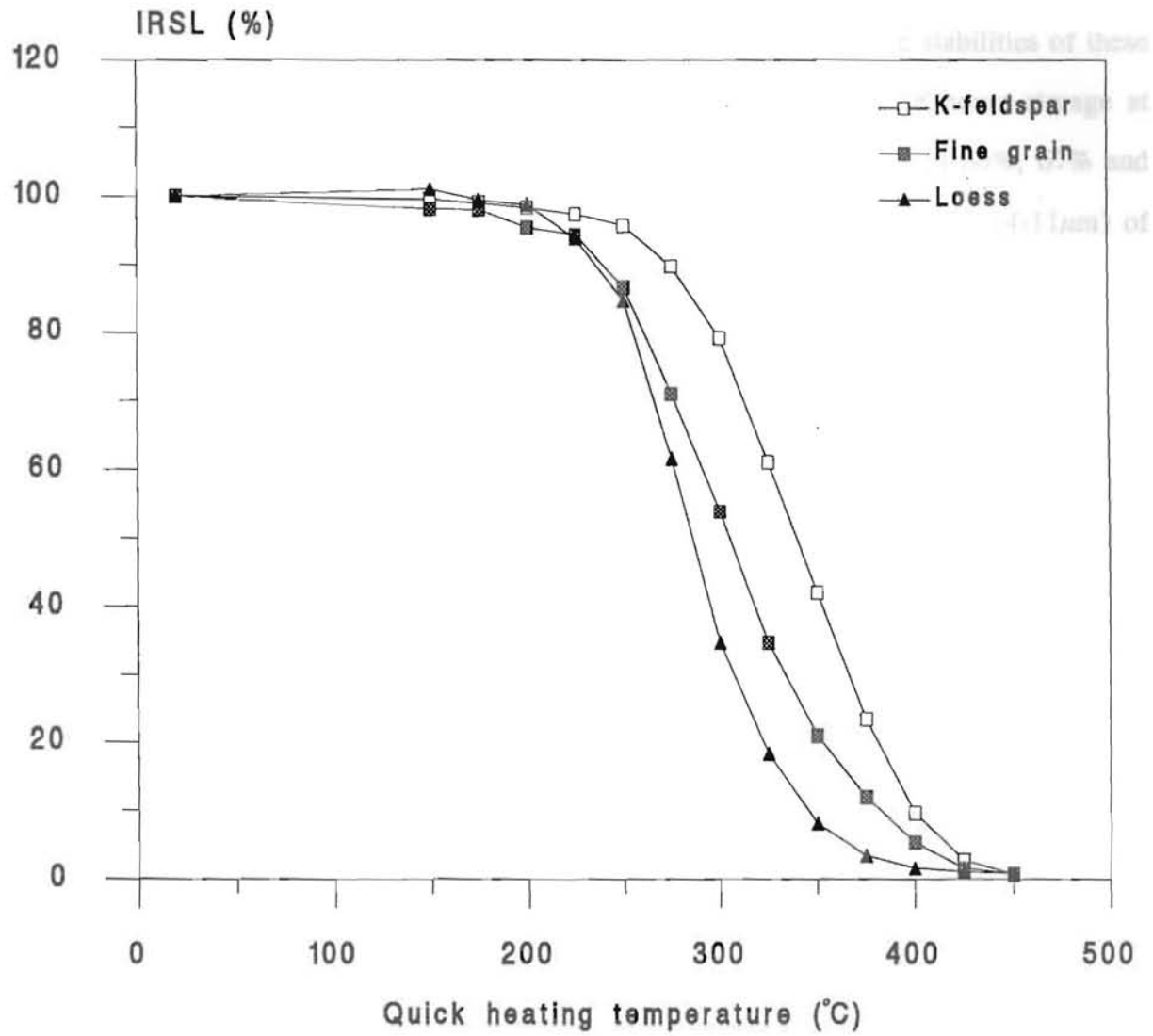


Figure 7.5.6. Quick heating curves of mineral mixtures of fine grain (4-11μm) loess (88-12), fine grain (4-11μm) mineral mixtures and coarse grain (125-150μm) K-feldspar of sample AF-3.

fractions of AF-3 are more stable than the fine grain loess. The relative stabilities of these samples were also indicated in isothermal decay experiments. After 18 hours storage at 140°C, the percentage of the remaining IRSL signals were different, with 86%, 67% and 36% for K-feldspar of AF-3, fine grains (4-11μm) of AF-3 and fine grains (4-11μm) of Chinese loess 88-12, respectively. These results support the quick heating results.

Although these thermal stability studies cannot give the lifetime of the IRSL signal for either fraction of AF-3, there is a distinct probability that the lack of stability of the IRSL signal for the fine grain fractions could have led to an underestimation of the ED (and thus the age). On the other hand, the ages for the K-feldspar fraction could have been overestimated as a result of possible underestimation of the dose rate.

7.6 CONCLUSIONS

Although a limited number of samples were examined, several general conclusions can be drawn from the IRSL ages obtained for the sixteen samples.

(a) The colluvial sediments of the Masotcheni formation were deposited during the last 120 ka, i.e. since the last interglacial, Oxygen Isotope sub-Stage 5e.

(b) The oldest colluvium was probably deposited during sub-Stage 5d.

(c) The next major period of colluvial deposition was at the beginning of Stage 3.

(d) There were several soil forming periods, the final major one being at the beginning of the Holocene, as found at Hazeldene.

(e) Further colluvial deposition took place during the Holocene.

(f) Similar ages from colluvium a few meters thick reflects the rapid deposition of colluvial sediments as a result of mass movement.

(g) IRSL ages and radiocarbon dates are in agreement in the Holocene.

(h) Radiocarbon dates over 35,000 year BP underestimate the age of soil formation in this area.

(i) The dark grey soil found at St Paul's and Nqutu is not the same as that found at Hazeldene. This was also clear from the radiocarbon dates.

chapter 8
RECOMMENDATIONS FOR FURTHER STUDIES

This study has been the first to target the dating of colluvial sediments by luminescence techniques and the first to examine approaches to allow the rapid scanning of bulk sediments. In these fields some questions were answered but others appeared which can only be answered by further studies.

I would like to suggest the following areas as being of particular interest:-

1. Sedimentary reconnaissance.

The portable system needs to be redesigned with better detection efficiency. It can then be tested on a wide range of different sediments and may be used to provide more information about the sediments, e.g. the source of the materials.

2. Luminescence sensitivity change model.

While the model has been found to be suitable for the IRSL signal from feldspars, it may also be applied to the OSL signal of quartz, which has a simpler trap distribution and about which more is known. Further studies on the model itself may act as a useful guide to the regeneration method and lead to its re-establishment. The comparison of EDs obtained by different methods has provided a way of estimating the daylight exposure prior to deposition and such studies should help to choose the most suitable bleaching time for the regeneration method.

3. Dating of incompletely bleached sediments.

The ED-I_n method proposed in this study still needs to be tested for a wider range of samples, especially those with known ages. The method, which gives the possible range of the ED (and thus an age range), may be useful for many poorly bleached samples. These

methods, with further refinement, may provide an important tool for dating samples of Holocene age (0-10 ka).

4. Studies associated with annual dose estimation.

There is some uncertainty in the dose rate determination, in particular: How does HF etching affect the feldspar grains and what effect does this have on dose rate? What is the distribution of U and Th within the grains and what effect does this have on the internal dose rate? What is the state of U series disequilibrium in colluvial deposits? Why are the K-contents of the separates with density less than 2.58 gm/cm^3 so low?

5. Dating of colluvium from South Africa.

Although most dates are in stratigraphic order and agree with the limited radiocarbon evidence, there is an unexplained discrepancy between the radiocarbon date of 24,600±580 years BP for the soil at Nqutu and the overlying IRSL age of 41±5 ka for HIT-4. Hence, more comparisons are required. Such a study is planned at Voordrag, about 50 km east of Vryheid, where there are several dark grey palaeosols with radiocarbon dates.

6. Long term stability

Further dating studies on fine grain and coarse grain fractions from the same sample would shed light on the long term stabilities of the IRSL signals from each fraction.

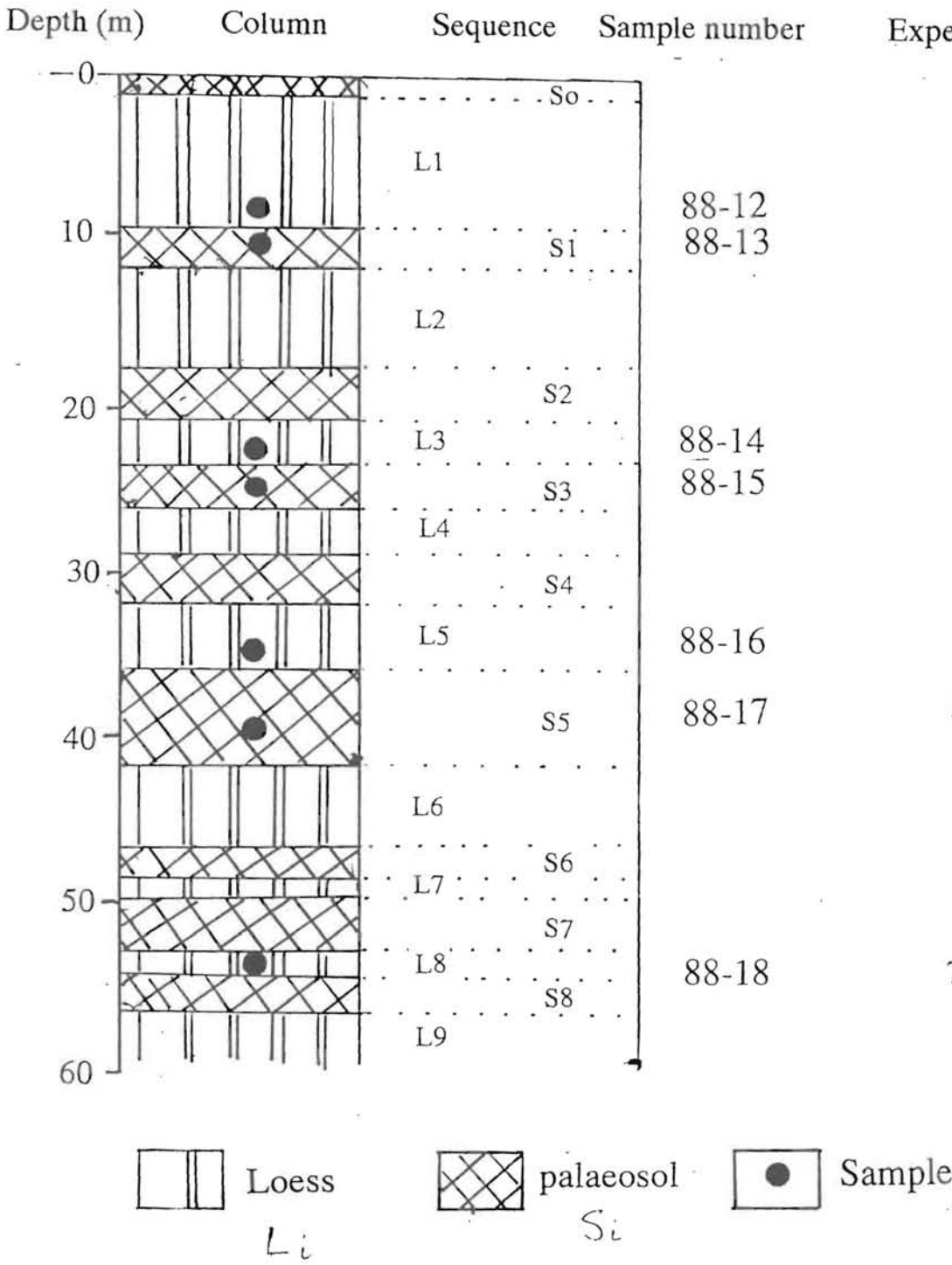
APPENDIX:
LOESS SAMPLES FROM THE LUOCHUAN SECTION

The section is located in Petou village near Luochuan, Shaanxi, China, and has been the subject of previous studies (Liu, et al. 1985, Kukla 1987). A total thickness of 135m of loess accumulated over the last 2.4 million years. The loess and palaeosol layers were numbered from the top of the section and distinguished with L and S prefixes. Further geological information can be found from elswhere (Liu et al. 1985, Kukla 1987).

The samples used in this study were taken from the upper part of the section. Samples were from the bottom of the loess layer, e.g. L1 bottom, and from the middle of the palaeosol layer. Figure A.1. shows the sampling positions and sample numbers.

The expected ages of these samples are also listed in Figure A.1. Sample 88-18 was from L8, which contains the Brunhes-Matuyama boundary according to the magnetostratigraphy of the section (Kukla 1987).

Loess and palaeosol sequence for the Luochuan section



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