Chapter 2

Experimental procedures and protocols

2.1 Introduction

This chapter presents a general discussion of methods and protocols used in the thesis along with a description of instruments used. The discussion includes sample collection and preparation, measurement techniques, analysis protocols and related aspects.

2.2 Sample collection and preparation methods

After freshly exposing the section the samples were collected in specially designed cylindrical tubes (~20 cm long and 2.5 cm diameter) made of aluminum or galvanized iron (Chandel et al., 2006). Due care was taken during the collection to ensure that the sample did not get any exposure to daylight. In the laboratory, the sample tubes were opened under subdued red light conditions and the outer ~ 5 cm from both sides of the tube were used for
the dose rate estimation. Central 10 cm portion was chemically processed for luminescence analysis. The laboratory equivalent dose ($D_L$) were estimated either using fine grain fraction (poly mineral) or coarse grain fraction (quartz). The grain size was chosen considering the availability of fraction and mean grain size. A brief description of fine grain and coarse grain method is given in section 2.2.1 and section 2.2.2

### 2.2.1 Fine Grain Method

The fine grain method was developed by (Zimmerman, 1971a). In this method fine grain fraction (4−11 μm) are extracted after treating the naturally collected sample with 1N HCl and 30% $H_2O_2$ to remove carbonates and organic matter respectively. This is followed by a de−flocculation treatment in 0.01N sodium oxalate solution. The de−flocculation helps to remove the clay size fraction. Multiple cycles of these steps are needed and at each step the samples were kept in ultrasonic bath to facilitate desegregation. Finally, the fine grain fraction is extracted by Stokes’ settling method by suspending the sample in 6 cm column of alcohol for 1.5 minutes (to remove >11 μm) and for 15 minutes (to remove < 4 μm grain size). The 4−11 μm fraction is re−suspended in alcohol and equal volume of ~1 ml is pipetted onto 9.65 mm diameter aluminum discs kept in glass vials of ~1 cm diameter. These vials were then dried at temperature < 50 °C for about 16 hours. Since it is physically difficult to extract either quartz or feldspar from this grain size the luminescence signal is cumulative for polymineralic assemblages, where the signal is mostly dominated by K−feldspar on account of its significantly higher luminescence sensitivity. The use of fine grain luminescence implies the use of full age equation and the need to determine the alpha efficiency. This implies additional measurements using a vacuum alpha irradiation. Despite the tedium of additional measurements, the alpha dose dilutes the uncertainty due to environmental dose.

### 2.2.2 Coarse Grain Method

The method proposed by Ichikawa (1965) was further developed by Fleming (Fleming, 1970; Fleming, 1979). The chemical pretreatments here comprise treatment by 1N HCl and 30% $H_2O_2$ as for the fine grain method. After the removal of carbonate and organic matter the sample is dried at temperature < 50 °C and sieved to obtain the desired grain size. The quartz and feldspar separation is done either by density separation method using sodium
polytungstate \((\rho = 2.58 \text{ gm/cm}^3)\) where quartz \((\rho = 2.65 \text{ gm/cm}^3)\) sinks and feldspar \((\rho = 2.56 \text{ gm/cm}^3)\) floats; or by magnetic separation (Porat, 2006) using a Frantz Magnetic Separator (Model LB–1 of S. G. Frantz Company Inc.). The quartz fraction after separation is etched by 40% HF for 80 minutes (equivalent to removal of 15–20 μm alpha skin) followed by 12N HCl treatment for 30 minutes to dissolve the fluorides and oven dried at temperature < 50 °C for final measurement. Etched, cleaned and dried grains were deposited on stainless steel discs of diameter 9.65 mm using silicon oil (silkospray™). The advantage of using quartz is that it has no internal radioactivity and does not suffer athermal fading of its luminescence signal but the tradeoff is its lower saturation range. On the other hand the feldspar luminescence provides a possibility of both higher and lower dating range due to high sensitivity and saturation dose but has the difficulty associated with athermal fading.

### 2.3 Measurement of TL and OSL: Instrument

In the present work for the measurement of luminescence, two instruments Risø TL/OSL reader (Bøtter–Jensen et al., 2000) and Daybreak 2200 Reader (Bortolot, 2000) were used. A typical TL/OSL reader is an assembly of following units

1. A detection unit (assembly of photomultiplier tube and detection filters)
2. Stimulation unit (for TL/OSL measurements)
3. Irradiation unit (for administrating laboratory dose)
4. A system controller for interface and readout system

#### 2.3.1 Detection Unit (TL OSL Reader)

The detection unit is the assembly of photomultiplier tube (PMT) and combination of filters placed in its front. The filters enable the selection of the desired band of emission and detection (normally UV 370 ± 30 nm for Quartz OSL AND TL) and 470 ± 30 nm for Feldspar (Feldspar IRSL and TL).

#### 2.3.1.1 Photomultiplier Tube (PMT)

To detect the luminescence signal from quartz/feldspar minerals, a bi–alkali type PMT (EMI 9235 QA) is used. The maximum detection efficiency of this type of PMT is ~ 380 nm
(Figure 2.1), which makes it possible to detect the entire luminescence signal from quartz or feldspar grains in Ultraviolet (UV) to Blue region. For bright samples the photon counts were kept $< 5 \times 10^5$ count/second (to avoid pileup effects) by employing either neutral density filter or by keeping low power of stimulation source. The sample to PMT cathode distance in the Risø TL/OSL luminescence reader is 55 mm, giving a detection solid angle of approximately 0.4 steradians (Bøtter–Jensen et al., 2000; Bøtter–Jensen et al., 2003a).

Figure 2.1: PMT response curve of quantum efficiency vs. wavelength. (Bøtter–Jensen et al., 2003a)

2.3.1.2 Filters

The emission of luminescence from minerals, like quartz and feldspar, ranges in the entire visible region of electromagnetic spectrum, more specifically UV to near Infrared (IR) range, after stimulation. The samples are stimulated using Light Emitting Diodes (LEDs) or lasers, in general blue (470 ± 30) nm for quartz and 890 ± 80 nm IR for feldspar. The main objectives of using a filter combination are (a) to avoid interference from stimulation source, (b) to isolate the desired emission band and (c) to reduce the blackbody radiation. To detect
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emission from quartz samples during OSL measurement a 7.5 mm thick Hoya U−340 (330 ± 35 nm) filter was used. The emission from feldspar is around 410 nm (Krbetschek et al., 1997), for which the blue emission was selected using Corning CS 7−59 (390 ± 60 nm) filter along with a combination of BG−39 (320–650 nm) filter to cut the IR. The transmission spectra of these filters is shown in Figure 2.2.

2.3.2 Stimulation Unit

A luminescence sample can be stimulated either by heat (Thermal Stimulation) or light (Optical Stimulation). The Risø TL/OSL and Daybreak 2200 Reader both have heating and optical stimulation units suitable for quartz and feldspar minerals.

2.3.2.1 Thermal Stimulation

A linear heating is obtained by placing the sample on a low−mass heater strip made of a Nickel and Kanthal (a high resistance alloy). The heater strip is shaped with a depression to provide good heat transmission to the sample and to lift sample disc securely into the measurement position. The desired temperature is obtained by feeding a controlled current through the heating element. A good control on the temperature of heater strip is obtained by mounting a Cromel−Alumel (Cr/Al) thermocouple underneath the heater strip which provides feedback to controller for controlling the temperature. The heating system is able to heat samples to 700 °C, at linear heating rates from 0.1 to 10 °C/s with step of 0.1 °C/s. The heating strip is constantly purged by nitrogen gas which not only prevents the heating system from oxidation at high temperatures but also helps in conduction of heat to the sample from the heater plate to the grains and quenches spurious non radiative luminescence. In Risø reader the precession of the temperature control is < ±4°C and the error in reproducibility of heating rate is < 1%.

2.3.2.2 Optical Stimulation

For stimulation of quartz and feldspar samples blue and IR LEDs were used respectively. Infrared (IR) stimulation in the region 800–900 nm can stimulate luminescence from most feldspars (Hütt et al., 1988), but not from quartz at room temperature. For the stimulation of
Figure 2.2: (a) Stimulation spectra of IR− LED and the filter combinations used to detect the luminescence in blue region and (b) Stimulation spectra of Blue LED and the filter combinations used to detect the luminescence in UV region
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a single grain quartz sample, a green laser at 532 nm was used. The optical arrangement of Risø and Daybreak TL/OSL reader are as follows

**Risø TL/OSL reader:** In this system, an array of 28 blue LEDs is used. The LEDs are arranged in 4 clusters each containing seven of them. The emission wavelength of these LEDs is 470 ± 20 nm (Bøtter-Jensen et al., 2003). A long pass green filter (Schott GG–420) is incorporated in front of each blue LED cluster to minimize the amount of tail end of directly scattered blue light into the detection window (center at 330 nm). The distance between stimulation source and sample is ~20 mm. The maximum total power from 28 blue LEDs is 80 mW/cm² at the sample position (Bøtter-Jensen et al., 2003). The IR LEDs, arranged in three clusters each containing seven individual LEDs, emits at 870 ± 40 nm. The maximum power is ~145 mW/cm² at the sample position (Bøtter-Jensen et al., 2003). Basic component of OSL detection system is shown in Figure 2.3.

The luminescence from single grains is achieved by stimulating grains kept in rhodium plated aluminum disc containing 100 cylindrical holes with 300 µm diameter and 300 µm depth, arranged in the form of a 10 by 10 array with 600 µm spacing between hole centers. The individual grains are stimulated by using a 10 mW Nd:YVO₄ solid state laser beam, emitting at 532 nm focused at a spot <20 µm in diameter with maximum energy fluence rate at the sample of ~50 W/cm². The laser spot is steered by orthogonal mirrors attached to two programmed high precision motors.

**Daybreak TL/OSL reader:** The daybreak system has 20 blue LEDs, arranged in two parallel rows each containing 10 LEDs, emitting light at 470 ± 30 nm with maximum power 60 mW/cm² at sample position. For IR excitation, 10 LEDs arranged in two rows each containing 5 of them are used with the peak wavelength at 880 nm and the maximum power delivered to the sample position is 50 mW/cm² (Bortolot, 2000).

2.3.3 Irradiation Unit

2.3.3.1 Risø TL/OSL Reader

Most experiments were carried out on a Risø TL/OSL TL/DA–15 which has a mounted beta irradiator (⁹⁰Sr/⁹⁰Y beta source) on the top of the system as shown in Figure 2.3a. The distance between the radiation source and the sample is 5 mm. The calibration of the source
Figure 2.3: (a) Schematic of TL/OSL Reader, (b) detailed schematic of illumination and detection unit from (after Bøtter-Jensen et al., 2003b)
on the system using calibration quartz supplied by Risø yielded the beta source strength to be 3.29 Gy/min.

### 2.3.3.2 Daybreak TL/OSL Reader

Daybreak–2200 TL–OSL reader has the beta irradiator with source strength of 0.9 Gy/min as calibrated on March, 2012. The distance between the irradiation source and sample is 15 mm. In few cases the beta irradiations were performed using a 20 slots beta irradiator manufactured by Daybreak–Nuclear and medical systems. The calibration of Daybreak beta irradiator gave a dose rate of 0.061 Gy/sec and 0.041 Gy/sec for Quartz (90–150 μm) and fine grains (4–11 μm), respectively.

For alpha efficiency calculation a six seater alpha irradiator with $^{241}\text{Am}$ source in vacuum was used (Singhvi and Aitken, 1978). $^{241}\text{Am}$ decays with a half–life of 432.6 years emitting alpha particles of several energies ranging from 4.76 MeV to 5.54 MeV. However, most dominant decay is through 5.48 MeV (85% probability) and 5.44 MeV (13% probability) α particle (www.nndc.bnl.gov data). This isotope is commercially available in form of 1 μm thick layer backed with 200 μm silver foil and front face protected by 2 μm thick gold–palladium alloy protective covering.

### 2.4 Measurement of Natural Dose Rate

The age estimation of natural samples requires the estimation of natural dose rate that results in luminescence buildup. Environmental dose rate is the rate at which energy is deposited in the sediment from the ambient nuclear radiation flux. Major contributors to environmental dose rate are Uranium ($^{238}\text{U}$, $^{235}\text{U}$), Thorium ($^{232}\text{Th}$) and Potassium ($^{40}\text{K}$). The decay schemes for these radioactive isotopes are shown in Figure 2.4 and Figure 2.5. Given the large decay time (~Billon years) of these radionuclides the activity of these radionuclides over a Million year time scales can reasonably be assumed as constant. However in the case of samples with interaction with water there is a reasonable chance of the parent/daughter being unsupported and in such case the dose rate becomes time dependent. In this case the present activity is used to calculate the evolution of dose rate with
Figure 2.4: Decay schemes of the Uranium series, (a) Uranium–235 and (b) Uranium–238
Figure 2.5: Decay schemes of the radioactive series, (a) Thorium–232, and (b) Potassium–40 and Rubidium–87
time and this is folded in with the total dose to compute the age. The radiation from these isotopes is in the form of α, β particles and γ rays (Figure 2.6). Also a small (<2%) but finite contribution (Mejdahl, 1987) from rubidium ($^{87}$Rb) and secondary cosmic rays mesons, electrons and gamma rays also contributes to the annual dose rate. The natural dose delivered to sample arises both from the radioactivity within the individual grain and the radioactivity in the surrounding matrix. Internal dose rate is ~ few % for coarse grains (>100 µm) of feldspars (Mejdahl, 1987) but is absent in quartz grains due to its being devoid of any internal radioactivity. The concentrations of U, Th and K in the sample can be determined by many experimental techniques such as thick source alpha counters, gamma ray spectrometry using germanium detector and thallium activated Sodium Iodide (NaI) scintillation spectrometer.

The annual dose can be calculated using the conversion factors by (Adamiec and Aitken, 1998) taking into account the matrix, its grain size distribution, the distribution of radioactivity and the fraction that is being used. In present study thick source alpha counting technique was used.

In scintillation counters ionizing particles produce scintillation corresponding to each decay of parent to daughter nucleus. The scintillation are then detected and amplified by PMT and counted using electronic counting unit. The techniques for the measurement of different radioactivity concentration are briefly discussed in next section
2.4.1 Measurement of Uranium (U−238) and Thorium (Th−232) Concentration

The estimation of concentration of U and Th were done by thick source α pair counting technique (Aitken, 1985; Appendix J)

In thick source alpha counting the sample is powdered to less than 10 µm size and typically an alpha thick layer (>20 µm) of powdered sample is spread uniformly on a 42 mm diameter ZnS(Ag) scintillation screen, positioned in a sealed perspex holder. The detector gives total number of alpha counts due to both U and Th, slow pair counts for ²²⁰Rn decay series \[ ^{220}\text{Rn} \rightarrow ^{216}\text{Po} \ (T_{1/2} = 0.15 \text{ s}) \] and fast pair counts for ²³⁵U decay series \[ ^{219}\text{Rn} \rightarrow ^{215}\text{Po} \ (T_{1/2} = 0.0018 \text{ s}). \] The concentrations of U and Th are proportional to count rate and the relationship between count rate and the concentrations of U and Th for the particular geometry is discussed in Aitken, 1985. In this calculation, it is assumed that the decay series of U and Th are in equilibrium. However, disequilibrium may take place mainly due to loss of Rn \((T_{1/2} = 3.83 \text{ days})\) from U−238 decay series. Such cases can be tested using hyper pure Germanium (HPGe) detector by measuring γ rays from different members of the decay chain. The discrepancy in U concentrations, calculated from the characteristics gamma ray, emitted from different radioisotopes before and after Rn of the decay chain decides the disequilibrium phenomenon. For the present work all samples showed equilibrium.

2.4.2 Measurement of Potassium (K−40) Concentration

To estimate the concentration of Potassium in sediments γ counting is done by using NaI(Tl) as scintillator. In this the gamma ray photons emitted by sediment is compared with a known standard and concentration is found by comparing the count rate of the two.

In gamma spectrometry a 3"×3" well type thallium (Tl) activated sodium iodide crystal and lithium doped hyper pure germanium crystal were used to measure the K concentration in the sample. To measure \(^{40}\text{K}\) concentration, the Compton subtracted photo peak corresponding to 1.46 MeV gamma photon emissions were used. The background subtracted photo peak is then compared with KCl standards with known \(^{40}\text{K}\) concentration. To ensure identical geometry for sample and standard a Perspex spacer was used. To reduce the
background ~6" lead shield was used. The resolution of the NaI detector was 4 keV whereas HPGe detector had resolution of 0.25 keV.

2.4.3 Measurement of Cosmic Ray Dose Rate

Cosmic rays consist of the charged particles which get deflected by earth’s magnetic field. In earth’s equatorial region cosmic ray flux is minimum, where magnetic field lines are perpendicular to the direction of the charged particle, and in the polar region it is maximum, where magnetic field lines are nearly parallel to the direction of the charged particle. Very high energetic particle (>GeV) can enter into the earth atmosphere. Most of the cosmic rays are absorbed by the earth’s atmosphere. However the secondary particles mainly muons contribute to the natural dose rate. These particles show a dependency on altitude and geographical latitude due to earth magnetic field. The cosmic dose rate was calculated as a function of altitude and latitude using equations as suggested by Prescott and Hutton (1994).

2.4.4 Moisture content

The presence of water in the void spaces affects the dose rate significantly. The water in voids does not carry any radioactivity but attenuates the radiation dose from the radioactivity in the sediment. Therefore, a correction to the dose rate estimation is made to estimate the dose rate correctly. This is calculated considering saturation water content (W) measured in laboratory and expressed as

\[
W = \frac{\text{weight of water}}{\text{weight of dry sample}}
\]  

(2.1)

W and a factor F which is the average soil water content as a fraction of this saturation water content W is used for the dose rate estimation. It is generally considered to be (0.8 ± 0.2)W. Using these two factors and the nuclear tables the α, β and γ dose rates are corrected as suggested by Zimmerman (1971a) and the dose rates used were calculated as

\[
\dot{D}_\alpha = \frac{\dot{D}_{\alpha \text{ dry}}}{1 + 1.50 \times W \times F}
\]  

(2.2)
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\[ D_\beta = \frac{D_{\beta \, \text{dry}}}{1 + 1.25 \times W \times F} \]  
\[ D_\gamma = \frac{D_{\gamma \, \text{dry}}}{1 + 1.14 \times W_i \times F} \]

W refers to sample and \( W_i \) to soil.

2.4.5 Alpha efficiency ‘a’ value

The luminescence efficiency of alpha particles is less as compared to beta and gamma. This occurs due to local charge saturation effects and high linear energy transfer by alpha particles as compared to beta and gamma ray. The alpha efficiency factor ‘a’ can be determined by the formula suggested by Aitken and Bowman (1975)

\[ a = \frac{\beta}{13 \times s \times y} \]

Where, \( S \, (\mu m^{-2} \, \text{min}^{-1}) \), is the strength of alpha source and \( \beta \), is the beta dose (Gy) which induces the same amount of luminescence as \( y \) minutes of alpha dose. There are other formalisms for ‘a’ value estimation but overall these provide similar results (Bowman and Huntley, 1984).

2.5 Equivalent Dose (D_e)

An important component of the luminescence dating is the estimation of equivalent dose \( D_e \). The equivalent dose \( D_e \) is the amount of laboratory beta or gamma dose that produces the same amount of luminescence signal, as given by the natural sample or as received by the sample due to irradiation in the environment. Aitken (1985) summarizes several protocols devised to estimate \( D_e \). These protocols take into account the changes in sensitivity (Luminescence per unit dose per unit mass) during various readout cycles. These measurement protocol include, Multiple Aliquot Additive Dose abbreviated as MAAD, (Aitken, 1985) and Single Aliquot Additive dose abbreviated as SAAD, (Stokes et al., 2000; Wallinga et al., 2000; Zhao et al., 2003; Vandenberghe et al., 2004). In these protocols,
incremental beta doses are given in addition to natural dose to a set of identical discs and a
dose versus luminescence growth curve is constructed. The $D_e$ estimate is made by
extrapolating the growth curve on negative x axis (dose axis). Depending on the sample
requirement several other protocols like, Australian slide method (Prescott et al., 1993),
photo transferred thremoluminescence abbreviated as PTTL, (Murray, 1996), thermally
transferred OSL (TTOSL) by, Wang et al., (2007), Post IR IRSL (Buylaert et al., 2009) etc.
have been developed to make the age determination more robust. In these measurements
different normalization criterion are applied to make the luminescence signal and sample or
aliquot independent. Some of these are weight normalization, zero glow normalization,
second glow normalization and short shine normalization. Aitken (1985) provide a good over
view and assessment of the normalization methods. All these protocols applied to samples
provide a better estimation of equivalent dose. However single aliquot protocol (Murray and
Wintle, 2000) is now the most widely and routinely used protocol.

Equivalent dose measurement methods can be broadly classified into two categories, viz
the additive dose method and the regenerative dose method, which are described here.

2.5.1 Multiple Aliquot Additive Dose Method (MAAD)

This method was first developed for the Thermoluminescence dating of archeological
pottery (Aitken, 1985). This method uses several identical aliquots of same sample. In order
to obtain appropriate $D_e$, aliquots are divided into several groups, the very first group is used
for the measurement of natural luminescence (i.e. sample as received) and other groups are
given increasing laboratory dose (e.g. $\beta_1$, $\beta_2$, $\beta_3$..... $\beta_N$) over and above the natural signal. The
luminescence signal thus obtained is plotted against the applied dose and a growth curve is
reconstructed. The equivalent dose is obtained by extrapolation of the growth–curve to zero
luminescence intensity (Figure 2.7), and the distance of this intersection point to the origin is
equal to the $D_e$. This method assumes that all aliquots have identical radiation history and
that the radiation response of the sample. This method while ensures against sensitivity
changes, requires a priori assessment of the nature of the growth curve. Further in MAAD,
growth curve is extrapolated on to the x–axis; hence the result depends quite significantly on
the choice of the mathematical function used (linear, exponential or polynomial). In addition
to this, problem arises when the extrapolation is to be made over a large dose–span, where
the growth is non-linear and/or there is a large scatter between the data points. As the construction of growth curves involves a large number of aliquots, appropriate normalization (discussed in section 2.5.2) is needed. Monte-Carlo simulations by Felix and Singhvi (1997) provide practical guidelines for construction of growth curves and extrapolations.

2.5.2 Normalization methods

For D_e measurement in laboratory using MAAD or SAR protocol, the assumption of identical environment and dose rate is not completely fulfilled. For example in MAAD protocol, the amount of sample number of bright grain in every disc may not same and different irradiations are given to different set of discs before the luminescence measurement which will cause difference in luminescence sensitivity. Similarly in SAR protocol, same disc is being repeatedly used and in each cycle irradiation (variable), preheat and luminescence measurement are done which will also cause different luminescence sensitivity at different cycle whereas all these protocol aim to measure the luminescence to construct dose response curve under identical sensitivity condition. In order to circumvent this problem, several normalization methods have been proposed (Aitken, 1985; Jain et al., 2003). The most common methods employed are;

2.5.2.1 Weight normalization

In this method aliquots are normalized by weight of sample and assume that either all grains have identical luminescence output or in every disc, number of effective bright grains is equal. Given that individual grains have variable luminescence sensitivity and that in general less than few percent of the grains provide a major fraction of luminescence of an aliquot, weight normalization often results in a scatter of ~10% or more (Aitken, 1985; Jain et al., 2003).

2.5.2.2 Zero glow normalization

This method was used initially for TL MAAD protocol (Aitken et al., 1979). A small test dose on natural sample is given and luminescence output corresponding to 110 °C TL peak which is absent in natural sample due to its short life (of about few hours) is used for the normalization of high temperature peak. A correlation between 110 °C TL peak and OSL in
quartz (Stoneham and Stokes, 1991) led to the potential of this method (Stokes, 1994). In the present work this method has been used in SAR protocol to correct for the change in the natural sensitivity during first read out of OSL (Singhvi et al., 2011) and is termed as Natural Correction Factor (NCF).

2.5.2.3 Short shine normalization

In this method a very short period (0.1 s) OSL signal is recorded before the measurement (~40 s recording of OSL). The short pulse deplete the signal by <1%. This method assumes that the luminescence sensitivity of first 0.1 s and rest of the OSL decay curve are correlated. This method is also used for multiple aliquot where short shine of natural sample take care the sample amount and the sensitivity. The applicability of this method is limited in very young or dim samples with low natural light levels.
2.5.2.4 Post normalization

This method is widely used in SAR protocol (Murray and Wintle, 2000). After each luminescence measurement a small test dose is given to the samples and then luminescence due to test dose is measured. The test dose signal at each cycle is the measure of sensitivity at each cycle. In the present work, mostly this normalization procedure was used.

2.5.3 Single Aliquot Regenerative Protocol (SAR)

The SAR protocol by Murray and Roberts (1997) and by Murray and Wintle (2000) suggested the single aliquot regeneration procedure for OSL signals. In this method each disc (single aliquot) or each grain (single grain) provides one equivalent dose. This method improves the dose precision by combining measurements of several discs or grains and provides statistical firmness.

In SAR method $D_e$ is estimated on a single aliquot by recording its natural luminescence and then a regeneration growth curve generated by giving incremental beta dose. The intensity of natural sample is then read on the regenerated growth curve to obtain $D_e$ as shown in Figure 2.8. In this method choice of signal and pretreatment to get that signal are important. At each stage a sensitivity measurement is included to ensure that any lab induced sensitivity is corrected for. For example OSL−SAR in quartz a preheat of 160−300°C is given to remove the unstable trapped electrons, which has lifetime less than or nearly same of age of the sample and first ~0−0.8 second of OSL signal is used for $D_e$ estimation (Murray and Wintle, 2000). The natural and regenerative OSL measurements are carried out at 125 °C in order to keep the 110 °C trap empty during the OSL stimulation (Murray and Wintle, 2000). Table 1 gives the details of SAR protocol, which was used. The specific details of the protocols, used for different samples will be described in the corresponding section.

2.5.4 Natural Correction Factor (NCF)

During the measurement of OSL in SAR protocol the changes in the sensitivity occurring in each regeneration cycle are taken care off by normalizing the signal with successive test dose OSL. Normalization in SAR protocol holds good if any OSL measurement and consecutive test dose OSL are measured under identical sensitivity condition or same factor of sensitivity change takes place during these two consecutive measurements for each cycle.
But natural OSL is measured in laboratory sensitivity condition whereas immediate test dose OSL is measured under laboratory condition. The conventional SAR protocol (Murray and Wintle, 2000) has in-built assumption of no change in sensitivity during natural OSL readout. However during the measurement of natural OSL (first OSL measurement), sensitivity change can happen (Singhvi et al., 2008).

To overcome this problem (Singhvi et al., 2010), introduced a modified SAR (NCF–SAR) protocol. In this protocol the additional steps are, before and after the natural OSL measurement a test dose TL up to 200°C is measured and the ratio of these two TL peak (after/before) is then multiplied with the natural point \( (L_N/T_N) \) and the modified sensitivity corrected natural point is the interpolated on the regeneration growth curve. The detail of the protocol is discussed by Singhvi et al. (2011).

Figure 2.8: Single aliquot regeneration method, sensitivity corrected luminescence is plotted against the incremental doses \( (R_1, R_2, R_3, \ldots) \). \( D_x \) is measured by interpolating the \( (L_N/T_N) \) on to the dose axis (Murray and Wintle, 2000)
### Table 2.1: Single aliquot regenerative protocol (Murray and Wintle, 2000)

<table>
<thead>
<tr>
<th>Step</th>
<th>Treatment</th>
<th>Observation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Preheat (160−300 °C) / 10 (s)</td>
<td>L&lt;sub&gt;N&lt;/sub&gt;</td>
</tr>
<tr>
<td>2</td>
<td>OSL (Natural)</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Test dose</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Cut heat (160 °C) / 10 (s)</td>
<td>T&lt;sub&gt;N&lt;/sub&gt;</td>
</tr>
<tr>
<td>5</td>
<td>Test dose OSL</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Illumination (240−280 °C) / 100 (s)</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Regeneration dose (R&lt;sub&gt;1&lt;/sub&gt;)</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>Preheat (160−300 °C) / 10 (s)</td>
<td>L&lt;sub&gt;1&lt;/sub&gt;</td>
</tr>
<tr>
<td>9</td>
<td>OSL (R&lt;sub&gt;1&lt;/sub&gt;)</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>Test dose</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>Cut heat (160 °C) / 10 (s)</td>
<td>T&lt;sub&gt;1&lt;/sub&gt;</td>
</tr>
<tr>
<td>12</td>
<td>Test dose OSL</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>Illumination (240−280 °C) / 100 (s)</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>Go to step 7 and repeat it for R&lt;sub&gt;2&lt;/sub&gt;, R&lt;sub&gt;3&lt;/sub&gt;…</td>
<td></td>
</tr>
</tbody>
</table>

![Diagram](image)

Figure 2.9: NCF correction factor model after (Singhvi et al., 2011)
Figure 2.10: (a) Histogram for the sample JR−4 (Jira village), (b) Radial plot of JR−4. Shaded region is the band of $2\sigma$ on the y−axis and (c) Probability density plot for the same sample
2.6 The nature of Distribution in $D_e$ within a sample

When the SAR protocol is repeated for number of aliquots for single event, each aliquot provide a $D_e$ value which is slightly different from other, resulting into the distribution of $D_e$. There could be several causes for this distribution of $D_e$ for a single event e.g. partial resetting of the luminescence signal, heterogeneous distribution $^{40}K$ hotspot within the matrix (Mayya et al., 2006) or because of experimental variation. To have the knowledge of degree of scattering in $D_e$ values, a pictorial representation is necessary. Several methods have been proposed in order to visualize the distribution, as follows.

2.6.1 Radial plot

A radial plot is a graphical representation of $D_e$ values (Galbraith, 1988; Galbraith, 1990), especially for comparing several estimates which have different precision.

Figure 2.10b shows the radial plot of the same sample. The X−axis represents the precision, expressed in relative error (%). The Y−axis is the standardized estimates of $D_e$, which is,

$$\log\left(\frac{D_e - w'}{w'}\right)$$

(2.1)

where, $w'$ is weighted average of all the aliquots, and $E'$ is the standard error of log$D_e$. Equivalent doses are statistically consistent at the 2σ level are easily recognized, as these fall within the shaded band.

2.6.2 Probability Density Plots

In this a Gaussian can be simulated for Each $D_e$ value with an error associated with it as standard deviation. For N number of $D_e$ will N Gaussian are simulated. The average Gaussian is constructed and represented as probability density plot (Figure 2.10c). Recent work by Chauhan and Singhvi (2011) suggested that the dose visualization methods such as radial plot and histograms may not be sufficient to draw inferences on the proper causes for the distribution in the paleodose and few other checks might be helpful. They suggested using the ratio (R) of maximum to minimum paleodose to account for the beta heterogeneity due to radioactive potassium in the sediment matrix as an additional proxy. A ratio $R \geq 10$ would
then imply sources of paleodose distribution (such as poor bleaching) other than the K heterogeneity, and a ratio less than 10 would imply well bleaching of the sample and mean $D_e$ would give the correct age of the event. The minimum number of aliquots required for the estimation of $D_e$ will also depend on the amount of radioactive potassium in the sediment matrix. As suggested for $K = 3\%$, minimum 24 aliquots would be needed to have a dose within ± 5% range of the actual value.

2.7 Estimation of errors in TL/OSL measurement

The errors in a luminescence age can be divided into two groups as in other standard cases, a) random errors and b) systematic errors. Random errors are those that comprise error in estimation of $D_e$, error in $a$–value, error in spectrometric measurement, in the computation of the annual dose and arise due to statistical fluctuation in photon counting, scintillation counting etc. Systematic errors are those that comprise error in the calibration of the alpha and beta source, error in calibration of alpha and gamma counters, parameters used in converting the U and Th concentration to dose rate, error in water content etc. The error estimation in the present case was based on the calculation proposed by Aitken (1985) and Murray and Olley (2002). The error quoted in TL/OSL ages are normally at 1 sigma level.