2.1 Introduction

In this chapter a detailed discussion of sample preparation methods, instruments used and methodologies to measure equivalent dose and dose rate are described. General descriptions of the protocols, normalization methods to account the sensitivity change during the luminescence measurements, analysis of the data are presented. The sample specific new or modified protocols are discussed in specific chapters.

2.2 Sample preparation

Prior to luminescence measurement of the natural samples, chemical pretreatments are required to remove inorganic and organic matters which give spurious (non radiation induced) signal during thermal and optical measurements and to isolate the desired minerals viz. quartz, feldspars or polymineralllic fine grain fraction. The sample preparation procedures for meteorites, volcanic ashes and sediments are described below.
2.2.1 Meteorite

Samples from the interior of these meteorites were gently crushed in an agate mortar and then treated with 1N HCl with frequent ultrasonic treatment, washed with distilled water and ethanol. This significantly improved the luminescence yield possibly by removing the semi-opaque coating over the luminescent minerals. Magnetic fractions were separated using a hand magnet. The coarser fractions (>200 μm) are then sieved out and typically 100-150 μm grains were spread as a monolayer on stainless steel discs using Silko-Spray silicone oil.

2.2.2 Volcanic ash

The volcanic ashes used for this thesis work were mostly clayey silt. So fine grain (4-11 μm) polyminerallic grain fraction was used, following Zimmerman (1971). The fine grain fraction was extracted after a sequential pretreatment of the bulk sample with 1N HCl and 30% H₂O₂ (to remove carbonates and organic matters respectively), followed by deflocculation in 0.01N sodium oxalate solution because the clay fractions can stick together due to static charge generated during other processes. In general multiple cycles of the above treatment were given and at each stage sample were kept for extended duration in ultrasonic bath for desegregation. This was followed by suspension of the samples in a 6 cm column of alcohol and >11 μm grain fractions were separated using gravity settling for 1.5 minutes. The desired grain fractions (4-11 μm) was then separated from the grain (<11 μm) suspension by Stokes settling for 15 minutes. The 4-11 μm size fractions was re-suspended in alcohol in such a way that the grains within ~1 ml of the solution can make just a monolayer on a circular disc of diameter ~1 cm. The equal volume (1 ml) of the solution were pipetted onto 9.65 mm diameter aluminum disc kept in a glass vials with ~1 cm diameter which were then dried at temperature <50°C. Considering the difficulty in mineral separation from silt clay dominated ash samples, the luminescence was taken to be from the entire polyminerallic assemblages, but in view of the high sensitivity, the signal from feldspar, normally dominates.
2.2.3 Sediment

The quartz mineral fraction from the sediment samples were extracted by sequentially treating the sample with 1N HCl and 30% H$_2$O$_2$ and dried. This was followed by sieving for different grain sizes range (90-150 and 150-250 µm). Quartz minerals were then separated using a Frantz Magnetic separator (Porat, 2006). The quartz fraction thus separated was etched by 40% HF (excess amount to maintain the pH during the reaction) for 80 min with constant magnetic stirring and ultrasonic to remove the outer alpha dosed skin (~20 µm), followed by a treatment with 12N HCl for 30 min to convert insoluble fluorides to soluble chlorides (Carriveau, 1977). Mejdahl (1979), suggested that due to etching, a small attenuation in external beta also occurs and proposed appropriate correction factor for various grain sizes. Typically attenuation of beta is ~5%, for a 100 µm grain size (Aitken, 1985). The contamination of BGSL signal from feldspar grains was checked with infrared stimulated luminescence (IRSL) and those indicating a contamination were re-etched using HF. The grains were spread as a monolayer on stainless steel discs using Silko-Spray silicone oil.

2.3 Instruments

For luminescence measurements the systems used were i) Risø TL/OSL reader (TL/OSL-DA-20; Bøtter-Jensen et al., 2000) and ii) Daybreak 2200 Reader (Bortolot, 2000). The basic working principles of both the machines are same. The schematic diagram of TL/OSL reader is shown in Fig. 2.1a. A typical TL/OSL reader comprises,

1. Stimulation unit: this comprises heating and optical elements and controller to stimulate the samples in a prescribed manner,
2. Detection unit: this comprises optical filters for light selection and PMT for detection.
3. Irradiation unit: this comprise radioactive source to impart dose on the sample.

2.3.1 Stimulation unit

2.3.1.1 Thermal stimulation

In the present study only a linear heating was used and this was obtained by a heating source, a low-mass heater strip made of a Nickel and Kanthal (a high resistance alloy).
Chapter 2 Experimental details and methodologies

The heater strip is shaped with a depression to provide good heat transmission to the sample and to lift sample disk securely into the measurement position. The desired temperature is obtained by resistive heating through a feedback controlled current through the heating element. A Cromel-Alumel (Cr/Al) thermocouple is spot-welded underneath the heater strip which provides feedback to control the temperature and provides the temperature information in real time. The heating system is able to heat samples to 700°C at linear heating rates from 0.1 to 10°C/s. The heating strip is constantly purged by nitrogen gas to prevent oxidation at high temperatures and to conduct heat from the heater plate to the grains. This minimized the temperature lag 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%.

2.3.1.2 Optical stimulation

For the stimulation of quartz and volcanic ash samples blue and IR LEDs were used respectively. In case of feldspar both visible light ~2.5 eV (490 nm) and infrared 1.43 eV, and 1.33 eV (866 nm, 932 nm) can stimulate the trapped electron (Hütt et al., 1988). In the first case the mechanism is connected with direct optical ionization, and in the second case with a thermo-optical stimulation. This is not the case for quartz at room temperature. Quartz cannot be stimulated by infrared light; visible (blue/green) can stimulate trapped electron of quartz. The optical arrangement of Risø and Daybreak TL/OSL reader are as follows.

Riso TL/OSL reader: In this system, an array of 28 LEDs is used. The LEDs are arranged in 4 clusters each containing seven LEDs. The emission wavelength is 470±20 nm (Bøtter-Jensen et al., 2003a). A long pass green filter (Schott GG-420) is placed in front of each blue LED cluster to reduce the tail end of directly scattered blue light into the detection window (center at 340 nm). The distance between stimulation source and sample is ~20 mm. The maximum total power at the sample position from the blue LEDs is 80 mW/cm² (Bøtter-Jensen et al., 2003a). The IR LEDs, arranged in three clusters each containing seven individual LEDs, emits at 870±40 nm. The maximum power at the sample position is ~145 mW/cm² (Bøtter-Jensen et al., 2003a). Basic component of OSL detection system is shown in Fig. 2.1b.
Figure 2.1 a) Schematic diagram of TL/OSL reader and its main components. b) Basic components of optical stimulation and detection system (redrawn after Botter-Jensen et al., 2003b).
Chapter 2 Experimental details and methodologies

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

Daybreak TL/OSL reader: For blue excitation, the system has 20 blue LEDs, arranged in two parallel rows each containing 10 LEDs, emit 470±30 nm light 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 can be delivered is 50 mW/cm² (Bortolot, 2000).

2.3.2 Detection unit

The luminescence detection unit comprises photomultiplier tube (PMT) and a filter combination in front of PMT to select desired emission band.

2.3.2.1 Filters

Luminescent minerals, like quartz and feldspar, emit photons of wavelength range in the entire visible region of electromagnetic spectrum, more specifically UV - near IR range, after irradiation and on either or both thermal/optical stimulation. Also during OSL readout the samples are stimulated by blue light for quartz and IR light for feldspars. To avoid any interference of stimulation light and to select particular emission bands, proper combination of optical glass filters are used. During OSL measurement of quartz sample a 7.5 mm thick Hoya U-340 (330±35 nm) filter was used. For TL/OSL measurement of feldspar from meteorite and volcanic ash samples both blue and red emission bands were selected. To select the blue emission from volcanic ash sample Corning Cs 7-59 (390±60 nm) filter was used. It has also emission at IR region (>680 nm). To cut the IR, BG-39 (320-650 nm) filter was used. The filter combination has emission at 395±50 nm. To detect the blue emission from meteorite sample in Daybreak 2200 reader Corning Cs 7-59 was replace by Corning Cs 5-58 (410±30) nm.
Figure 2.2 a) Transmission of all optical filters (measured using spectrophotometer). b) Combined emission in UV, blue and red emission window.
Figure 2.3 PMT response curves of a) Bialkali (EMI 9235), maximum quantum efficiency occurs at 300-450 nm, UV-blue region (redrawn after Bøtter-Jensen et al., 2003b), b) Hamamatsu (R943-02), can detect UV to IR region (200-750 nm). (redrawn after http://sales.hamamatsu.com/assets/pdf/parts_R/R943-02.pdf).
Red emission from meteorite and volcanic ash samples were restricted using interference filters combination Comar 650IU (650±20 nm) and Comar 650IW (650±37) with net transmission in the range 650±20 nm. The transmission spectra of all the filters are shown in Fig. 2.2a and combined three different emissions band, UV, blue and red are shown in Fig. 2.2b. Also for sample, like Al₂O₃:C with exceptionally high luminescence sensitivity fused silica neutral density filters were used to reduce the intensity to avoid pile up effects, both in the PM Tube and in the photon counting systems.

![Graph showing photocathode dark current density vs. cathode temperature of GaAs in red PMT, R943-02](http://www.photocool.com/tocool.htm#figure1)

**Photomultiplier Tube**

The luminescence signals were detected by two detection arrangements. For UV and blue emission detection EMI 9235 QA (bi-alkali), and for red emission Hamamatsu R943-02 PMT were used. Fig. 2.3a shows the Quantum efficiency of EMI 9235 QA PMT at different wavelength. The maximum detection efficiency at ~380 nm makes it an efficient PMT in the UV-blue region. Also the detection range (200-600 nm) permits detection of the entire luminescence signal from quartz and feldspar in the UV-near orange red signal. As the response of this PMT is poor in the red, a different red sensitive PMT Hamamatsu R943-02 PMT, which has detection range 200-800 nm (Fig. 2.3b), was used to detect red emission from the samples. It has red sensitive GaAs photocathode which is highly...
sensitive to temperature (Fig. 2.4). At room temperature (20°C) photocathode dark count is ~3000 counts/s/cm². To overcome the interference of high dark count with the signal, the PMT was cooled externally by a Peltier cooler, photocool series PMT housing (Model PC182CE). Within 2 hours the chamber temperature comes to -22°C and dark count reduced by 200-300 times compare to room temperature making it possible to make measurements.

2.3.3 Irradiation unit
For beta irradiation ⁹⁰Sr/⁹⁰Y beta source was used for both Risø and Daybreak TL/OSL reader. The source emits beta with end point energy 2.27 MeV. The distance between the irradiation source and the sample is 5 mm for Risø system and 15 mm for Daybreak system. The dose rates calibration of every system were made using calibration quartz supplied by Risø National Laboratory, Denmark. The calibrated dose rates for coarse grain (90-150 μm) quartz on stainless steel disc in Risø and Daybreak system were 0.057 Gy/s and 0.017 Gy/s respectively on June, 2009. The half life of beta particles emitted from ⁹⁰Sr/⁹⁰Y source is 28.5 yrs. So either a dose rate correction using radioactive decay equation or frequent calibration (~1 yr interval) is required. The sources were also calibrated for fine grain (4-11 μm) on aluminum disc and it was observed that the fine grain dose rate on aluminum disc is ~70% of coarse grain dose rate on stainless steel disc.

For alpha efficiency calculation a six seater alpha irradiator with ²⁴¹Am source in vacuum was used (Singhvi and Aitken, 1978).

2.4 Methodologies
For chronometric application of luminescence dosimetry, two parameters are to be estimated, equivalent dose and annual dose (dose rate), as discussed in the section 1.4.3. Procedures for estimation of these two parameters are briefly described below.

2.4.1 Measurement of Equivalent Dose
The equivalent dose (Dₑ) is the amount of laboratory beta or gamma dose which produces same luminescence signal, as produced by the natural (as received) sample (with its
signal due to irradiation in the natural radiation environment). Methods for equivalent dose estimation can be broadly classified into two categories, viz the additive and the regenerative dose method. Both the methods involve construction of luminescence signal vs. radiation dose growth curve. The two methods are described here.

2.4.1.1 Multiple aliquot additive dose (MAAD)

This method was first developed for the thermoluminescence dating of archaeological pottery (Aitken, 1985). This method uses several identical aliquots of same sample. In order to obtain appropriate paleodose (exact equivalent beta dose received by the sample), these aliquots are divided into several “identical” groups, which are given incremental beta doses ranging from zero to $\beta_1$, $\beta_2$, ..., $\beta_N$. The “fixed” natural dose is common to every disc. The luminescence yield plotted against the applied dose, provides the growth curve and this when extrapolated to zero luminescence provide the equivalent dose. The basic advantage of this method is that it does not entail any sensitivity change, a fact that makes it a robust method (Fig. 2.5a). The limitations of this method are;

1. This method assumes that all the discs are identical i.e. they have identical irradiation, thermal and optical history. Being a multi grain approach this implicitly provides an average value and factors like heterogeneous dose distribution at single grain level, different degree of bleaching (zeroing) at the time of deposition are not addressed to. The applicability of this method is better for the samples for which the geological signal was thermally zeroed, like volcanic materials, pottery etc. as compared to fluvial or aeolian sediments where optical resetting may vary at a grain level. Further to ensure the identicalness of each disc in respect of the luminescence response at disc level arising from variable sample amounts and various grain responses, various normalization techniques are used (Jain et al., 2003) and are discussed in section 2.4.1.4.

2. The extrapolation of the luminescence vs. dose growth curve, to zero luminescence value assumes that the mathematical form of the growth curve is known and that any sub- and/or supra-linear behavior can be accounted for. Recommendation on optimum measurement for minimum error in equivalent dose, was given by Felix and Singhvi, 1997, who examined this aspect in detail.
Figure 2.5 Representation of equivalent dose estimation. a) using multiple aliquot additive dose method of TL signal; extrapolation of the curve give $D_{e}$, and b) using single aliquot regeneration method of pIR-IRSL signal; interpolation of natural point provide $D_{e}$. The measurements were for TPASH sample in blue emission window.
2.4.1.2 Single aliquot regenerative dose (SAR)

Murray and Roberts (1997) and Murray and Wintle (2000) suggested the single aliquot regeneration procedure for OSL signal which presently is the most used protocol. In this method each disc (single aliquot mode) or each grain (single grain mode) provides one equivalent dose. This method provides for improved precision by combining measurements of several discs/grain as independent measurements.

In SAR method $D_e$ is estimated on a single aliquot by recording its natural luminescence and then a regeneration growth curve is made through a cycles of measurements. The intensity of natural sample is then read on the regenerated growth curve to obtain $D_e$. In this method choice of signal and pretreatment to get that signal are important. 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 $\sim$0-0.8 second of OSL signal is used for $D_e$ estimation (Murray and Wintle, 2000) and the signal in later intervals are used as background. Dose growth curves were constructed using three regeneration dose points and two points were used to estimate the recuperation effect and reliability of sensitivity corrections (recycling points) respectively. The specific details of the protocols, used for different samples are described in the corresponding section. A graphical representation of SAR protocol is shown in Fig. 2.5b.

2.4.1.3 Standard Growth Curve (SGC)

While attractive, the SAR measurement is time consuming and typically a sample analysis comprising about 40 discs would take 120 hours. The SGC-SAR approach was developed to reduce the measurement time of a sample. In the present work, this protocol was used to construct simulated growth curve in nature with fading for both meteorite and volcanic ash samples. In the normal SAR, the sensitivity corrected natural luminescence ($L_n/T_n$) is measured and it is interpolated into the dose response curve ($L_n/T_x$ vs. dose) to get the equivalent dose. Normal SAR run for each aliquot includes many cycles of irradiation, preheat and readout. Roberts and Duller (2004), suggested that instead of using individual dose response curves for the aliquots, a common dose
response curve for 5-10 aliquots can be used as standard growth curve (SGC) and then, only natural points \( (L_n/T_n) \) are needed to measure for rest of the aliquots. The assumption in this method is all the aliquots response identically with dose and for this SGC can be used if the \( R^2 \) of the fitting is greater than 0.9.

2.4.1.4 Normalization procedure

For \( D_e \) measurement using MAAD or SAR protocol, luminescence signals are not measured under identical sensitivity (luminescence/dose/mass) condition. 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 cycles 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 and widely used methods are;

**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).

**Zero glow normalization:** This method was suggested by Aitken et al., 1979 for TL MAAD application. In this method a small test dose on natural sample is given and luminescence output corresponding to 110°C TL peak which is absent in natural sample is used for the normalization of high temperature peak. A correlation between 110°C TL peak and OSL in quartz (Stokes, 1994) led to the potential of this method by Stoneham and Stokes (1991). For this thesis work this normalization technique was used for \( D_e \).
Chapter 2 Experimental details and methodologies

Estimation of meteorites using blue ITL SAR where low temperature peak was used to normalize the signal from the high temperature peak.

**Short shine normalization:** In this method a very short period (0.1 s) OSL signal is recorded before the measurement (long time, ~40 s). The short pulse depletes 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 and this aspect is not true always. This method is also used for multiple aliquot where short shine of natural sample takes 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.

**Second glow 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.

**Natural Correction Factor (NCF):** SAR protocol takes care of the sensitivity change occurs in each regeneration cycle of the measurement. But during the measurement of natural OSL, sensitivity change can happen. 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. To overcome this problem Singhvi et al. (2010), introduced a modified SAR (NCF-SAR) protocol. In this protocol the additional steps that are incorporated are, measurement of the sensitivity of 110°C peak following a test dose, both before and after the natural OSL measurement. The ratio of the two TL peak (after/before) is then multiplied with the intensity of natural OSL (\(I_n/T_n\)) and the modified sensitivity corrected natural point is then interpolated on the regeneration growth curve to retrieve the paleodose. The detail of the protocol is discussed in Singhvi et al. (2011).
2.4.1.5 **Dose Distribution**

In SAR protocol each aliquot provides a $D_e$ value and a large number of discs provide a distribution of $D_e$. The distribution of $D_e$ for a single event may be because of variation in the pre-depositional bleaching, heterogeneous distribution of $^{40}$K hotspot within the matrix (Mayya et al., 2006) and uncertainty in the measurements. For a visual appreciation of the scattering in $D_e$ values, a pictorial representation is often helpful. Several methods have been proposed so as to visualize the dose distribution. These are,

**Histogram:** Histogram is a mapping that counts number of events (frequency) fall into various disjoint equal interval of $D_e$ values, known as bin. To construct histogram selection of bin width is the most important. Normally the bin width is selected as median of the $\sigma$, where $\sigma$ is the errors of $D_e$ values (Lepper et al., 2000). The skewness of the dose distribution is used to detect poor/heterogeneously bleached sample (Murray et al., 1995; Olley et al., 1998). A histogram plot of one sample is shown in Fig. 2.6a.

**Radial plot:** Radial plot is a graphical representation of $D_e$ values, especially for comparing several estimates which have different precision. Fig. 2.6b 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,

$$\frac{\log(D_e - w')}{E'}$$  \hspace{1cm} (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 2$\sigma$ level are easily recognized, as these fall within the shaded band.

**Probability density plot:** Each $D_e$ value with an error associated with it can be represented as a Gaussian with mean is the $D_e$ value and error is the standard deviation. Similarly N number of aliquots will represent N number of gaussian. The average gaussian, simulated at different $D_e$ values, will represent as probability density plot (Fig. 2.6c).
Chapter 2 Experimental details and methodologies

Figure 2.6 Graphical representation of distribution in equivalent doses using a) Histogram, b) Radial plot and iii) probability and rank plot. These are real D_e's of TPASH sample using pIR-IRSL SAR protocols.
Chapter 2 Experimental details and methodologies

Single aliquot and single grain measurements provide a range of equivalent dose from which the true equivalent dose is extracted using a variety of statistical procedures. The dose analysis models are:

1. Common age model (Galbraith et al., 1999)
2. Central age model (Galbraith et al., 1999)
3. Minimum age model (Galbraith et al., 1999)
4. Leading edge model (Lepper and Mckeever, 2002)
5. Finite mixture model (Galbraith and Green, 1990)
6. Beta dose heterogeneity model (Mayya et al., 2006)

In the present thesis, the equivalent doses were obtained using Central age model and the reason for using this model is described in the corresponding section.

2.4.2 Measurement of dose rate

Apart from equivalent dose measurement, dose rate is another important parameter for the estimation of the age of a sample. In case of meteorites, cosmic rays are the major source of irradiation (details in Chapter 4). For terrestrial sample i.e. volcanic ash and sediment samples, the ambient radioactivity is the source of irradiation. The dose rate is the rate at which energy is deposited in the sediment due to the radiation field arising from the decay of its natural radioactivity viz., Uranium (\(^{238}\text{U},^{235}\text{U}\)), Thorium (\(^{232}\text{Th}\)) and Potassium (\(^{40}\text{K}\)). The radiation field arising from these isotopes consists of \(\alpha\) particles, \(\beta\) particles and \(\gamma\) rays. There is also small but finite contribution from rubidium (\(^{87}\text{Rb}\)) and secondary cosmic rays (mesons, electrons and gamma rays). As the radioisotopes have long half life (~billion yrs) it ensures that over a million year time scale, the dose rate can be assumed to be constant. However this constancy can be disturbed if a chemical fractionation occurs such that parent in the decay chain is deposited without its daughter or vice versa. In such cases the dose rate becomes time dependent, changing with the decay series moving towards an equilibrium situation. The natural dose delivered to sediment (sample) arises from radioactivity within the individual grain and from the surrounding region. Internal dose rate is significant for coarse grains (>100 \(\mu\)m). In general the internal radioactivity for quartz grains is insignificant and can be neglected.
(Murray and Roberts, 1997). The internal potassium content in the K-feldspars can be 14% and its contribution to the total dose rate can be up to a few percent (Mejdahl, 1987). The concentrations of U, Th and K in the sample can be determined by many experimental techniques such as thick source ZnS (Ag) alpha counting technique, high resolution gamma ray spectrometry using germanium detector or a low resolution spectrometry using thallium activated sodium iodide (NaI(Tl)) gamma ray spectrometers, beta counter spectrometers etc. And the annual dose is then calculated using the conversion factors by Adamiec and Aitken (1998). The techniques for the measurement of different radioactivity concentration used in the present thesis are briefly discussed below.

2.4.2.1 Thick source \( \alpha \) counting

In this technique the concentration of U and Th are estimated on the basis of \( \alpha \) counting. In thick source alpha counting the sample is powdered to less than 10 \( \mu \)m size and typically a thick layer (>20 \( \mu \)m) 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 \(^{232}\text{Th}\) decay series \(^{220}\text{Rn} \rightarrow ^{216}\text{Po}\) (\( T_{1/2} = 0.15 \) s) and fast pair counts for \(^{235}\text{U}\) decay series \(^{219}\text{Rn} \rightarrow ^{215}\text{Po}\) (\( T_{1/2} = 0.0018 \) 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 (\( T_{1/2} = 3.83 \) days) from \(^{238}\text{Uranium}\) decay series. Such cases can be tested using hyper pure Germanium (HPGe) detector by measuring \( \gamma \) rays from different members to compute the activity of each of the member of the decay chain. The difference in U concentrations, calculated from the characteristics gamma ray, emits from different radioisotopes before and after Rn of the decay chain informs the extent of the disequilibrium. For the present work all samples showed equilibrium.
2.4.2.2 Scintillation Gamma Spectrometry

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 ⁴₀K 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 an AR grade KCl standard with known stoichiometric 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.

2.4.2.3 Cosmic ray contribution

Cosmic rays comprise charged particles and hence get deflected by earth’s magnetic field. So in the equatorial region, where magnetic field lines are perpendicular to the direction of the charged particle, cosmic ray flux is minimum 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 and most of the cosmic rays absorbed by the earth’s atmosphere. However the secondary particles mainly muons contribute to the dose rate. These particles show a dependency on altitude and geographical latitude due to earth magnetic field. Prescott and Hutton (1994), have provided a simple relationship to compute the cosmic dose rate at any location by using its altitude, latitude and longitude.