Chapter-6

Summary and Future Outlook

6.1 Summary: Present Understanding

The present thesis investigated the amplitude and style of distribution of radiation doses in nature due to natural radioactivity and their causes. The distribution in doses occurs at microscopic level and macroscopic levels due to different ranges of ionizing radiations (α, β and γ) and heterogeneous distribution of radioactivity at a grain level. The ranges of these radiations scale in ratio 1:100:10,000 respectively, and this difference in their ranges results in distribution in doses. The present thesis tried to quantify this distribution to make the dose estimation more rigorous and explore the new possibilities, which can
help extend the dating limits and the precision of luminescence dating. In this detailed study it was seen that there are several factors responsible for the distribution in doses and their quantification provides a basis for improved age calculations, i.e.:

1. Generally, considerable distribution is found in palaeodoses obtained from well bleached ~100 μm size single grains of the quartz. During this study using Monte Carlo simulations and the mathematical model developed by Mayya et al., (2006), it was found a significant amount of distribution in the doses results from the fluctuation of K-feldspar grains around quartz grains. K-feldspar contain 40K, which is one of the major contributors to the β-dose, and the range of these particles is comparable to inter-grain distances. As different feldspar grains are located at different distance so each feldspar grain will deposit unequal doses in the quartz grains and fluctuations in configuration of feldspar grains around quartz grains result in different dose deposition. In this thesis, the nature of distribution and the minimum dose rate (δ) value was estimated as a function of concentration of K. In addition to this, estimation of dose distribution was done for different porosity and water content and it was seen that for higher porosity the dose distribution function shifts towards higher dose side. A new value of δ for different combinations was obtained that allows the estimation of age even for poorly bleached samples.

2. The distribution of doses at single grain level due to heterogeneity in β dose results in distribution in doses for single aliquots formed by several such single grains (~100). This distribution in the single aliquots doses was estimated by means of Monte Carlo simulations. The simulations allowed measurement of several useful parameters like determination of the number of the discs to be used for analysis and the resultant accuracy, maximum possible error and dispersion in doses because of β dose heterogeneity. These parameters help improve the accuracy in age determination. The maximum possible error and spread in the palaeodoses defines a limit in the precision of ages in luminescence dating, and the ratio of maximum to minimum dose value helps identify
the samples having distribution because of β dose heterogeneity from the samples having other causes of dose distribution. This then helps deciding the kind of model (FMM, MAM, CAM etc.) that could be used for age estimation. This work also suggest that mean of the dose distribution provides reliable dose estimate if β heterogeneity is the sole cause of distribution.

3. The dating range of quartz is limited by the fact that the luminescence signal of quartz saturates around ~250 Gy. This saturation of luminescence signal along with the dose rate of sediment matrix decides the limit of the dating. In this work the efforts were made to date the interior of a ~Sub-centimeter size quartz grains. The interior of such large grains provide the opportunity to date older sample hence extending the dating limits of luminescence dating. In this work, Monte Carlo simulations were used to find the beta dose profile inside the large size quartz and it was found that the contribution of beta dose from the sediment from all the sources reduces to negligible level at depths of ~2 mm. This implies that a central core inside the grains of Diameter>4 mm would always exist, that would receive dose only because of gamma radiation. For this region the dose rate is a third of the dose rate in a sediment matrix. As the dose rate in the interior is lower, the luminescence signal saturates later in time and samples 3-4 times older than normal luminescence age limit can be dated. Transparency and the bleachability experiments suggest that such large size quartz grains can be bleached by ~3 hrs of natural daylight. Thus, a carefully selected sample can be used to date older samples. A well-defined procedure is suggested for sample preparation and measurements. Experimental verification proved the proposed hypothesis and one of the sample having an age >200 ka was dated using the methodology provided.

4. A major limitation of luminescence dating has been the need for specific minerals of definite grain sizes for dating. Many times, it is difficult to separate the minerals or the proper grain sizes. This limitation can be
overcome by insitu measurements, i.e. analyzing the grain without separating them from the matrix. This can be done using surface dating technique, which involves measurement of luminescence signal corresponding to small region on the surface and analyzing that small region to estimate the dose values. However, the major issues for such studies were related to transparency of materials to be used, the dose rate estimation and measurement of spatially resolved luminescence signal. The calculations suggest that the issue of transparency can be taken care of by using the transparent material with limitation that its thickness should be smaller than the maximum β range and transparency and thickness should remain constant during the measurements. A program in Matlab enables the estimation of the natural dose rate and laboratory dose rate in terms of average dose deposited in a given thickness of sample. For estimating the natural dose rate, the radioactive concentration of the two regions should be known.

For spatially resolved luminescence measurements, an EMCCD based imaging system was designed and tested. All the units of this system (including heating and light stimulating unit, interfacing software, synchronous image acquisition, Optical arrangement and data processing software) were developed in-house. The luminescence is recorded as time series images whose intensity varies in accordance with the stimulating mode. The images acquired are processed in the programs written in Matlab to find out the Do value. These programs read the image, align the images up to single pixel accuracy and finally evaluate pixel specific or region specific dose values. The system has been tested for dose recovery test and could successfully recover the doses within ±10% of the given dose for TL of both quartz and feldspar and IRSL of feldspar. The system can now be used for routine measurement of the natural samples.

5. The SAR protocol is widely used protocol to measure the palaeodose, De. This protocol estimates the De with continuous monitoring of the sensitivity during the measurements with the help of signal corresponding
to small test dose (TD). However, in the SAR protocol there was so far no provision of the correction for the sensitivity changes occurring during the measurement of the natural luminescence of the samples. Sensitivity monitoring experiments suggested that significant sensitivity changes occurred during the measurement of natural OSL. This sensitivity change during the measurement of natural OSL can result in a change in dose value up to 40% and the direction and amplitude of this change can be either way, i.e. it can decrease or increase. This affects $D_e$ measurements. The change is not uniform and varies from disc to disc and sample to sample. In this thesis a methodology to correct for these sensitivity changes was developed and was termed as natural sensitivity corrected SAR (NSC-SAR) protocol. This protocol includes measurement of TD TL signal before and after the natural signal measurement and the ratio of these two is used to correct for the changes in the sensitivity. The application of this protocol provides improved results, the scatter in the doses reduces significantly and the dose distribution becomes much more compact.

### 6.2 Future Outlook

Present thesis offers newer possibilities in the development of experimental techniques and for newer geological applications. Some of the new possibilities that could merit further studies are as follows,

1. The distribution in doses resulting from spatial fluctuation of the beta sources needs experimentally quantification. In the theoretical development of beta dose heterogeneity model, one of the critical assumptions was that the concentration of K should be low and that the effect get diluted for higher concentrations of K. This needs further quantification in respect of the increasing potassium and corresponding dilution of heterogeneous component.
2. It will also be interesting to combine the theory of coldspots (Jacobs et al., 2008) with the theory of K-feldspar hotspots to give a unified theory, which can accommodate all the possible dose distributions in natural environment for well-bleached sediments.

3. The dose deposited in the grains is a function of the radioactivity ranges and the size of the grains. This implies that, the rate at which the dose is deposited in the grains of variable sizes is different. Hence, for a sample of given age, the different size of the grains in the same sample will have different doses. A plot of dose as a function of dose rate (for different sizes) will give an isochron in which the slope gives the age of the sample and this method can also be tried with the added advantage that external environmental parameters such as heterogeneity of matrix and water content and their effects can be minimized.

\[ D = A \text{ge} \times \dot{D} \]  \hspace{1cm} (6.1)

4. Dating of large grains offers new prospects in dating the fluvial deposits or dating of the terraces. The river lain gravels having ~cm size quartz and which are older (~200-400ka) and undatable by normal luminescence methodology or any other technique can be dated. This will fill in much needed void in Quaternary Geology in terms of dating of the processes and climatic events in 100-1000ka range.

5. The work on large grains also offers prospects for dating lithic implements in archeology. Given that at time early man heated rock for making them amenable to mechanical working, as also that these rocks remained in day light during their use it is possible now to use a slice of these to date such implements. The exploitation of beta dose gradient additionally offers prospects of directly dating the lower Paleolithic, which at present is poorly dated in the Indian context.

6. An important element that can be developed is the non destructive dating of tools by adopting the present EMCCD system with confocal optics. This
is an uncharted area and will be worthwhile to explore this aspect and use confocal microscopic methods in luminescence dating.

7. While measuring the luminescence signal of large size quartz grains it was found that the sensitivity of the large size quartz grains was several orders of magnitude lower than the associated grains of size 90-150 µm. The sensitivity of the grains decreases as a function of increasing diameter. Such a large variation in the sensitivity with the size can be used to identify the causes of sensitivity variations in case of quartz and also use sensitivity change as a surrogate for time. As this has been a long-standing question in case of the luminescence technique.

8. Spatially resolved luminescence system developed can be further tuned in respect of its optical and technical attributes. Future improvements would include, optics for UV detection, removal of reflecting surfaces to reduce the loss of signal, and improvements of light collection using parabolic reflector around the samples and by a combination of convex and concave mirrors, which has high reflection efficiency. It can thus improve upon S/N ratio significantly.

9. Further development of the system can be done by transforming the system to a spatially resolved spectrometer, which will be capable of doing measurements in different wavelength regions. This development will make the instrument only one of its kind making such measurements.

10. The development of surface dating technique will help in many areas of geological applications. In addition, as it can date the specific regions of the surface hence it is capable of dating different geological events by dating of rock. It can be used to find terrestrial age of meteorites. It can date glacial moraines to study the dynamics of glaciers through time. It can be used to date the secondary growth of the minerals like in case of gypsum, or find out the rate of the carbonate nodule formation by dating the quartz inclusions in the carbonate nodules and hence can be used to find the rate of carbonate formation. These ideas need to be explored to identify the potential areas of refinements for routine application of surfaces.
11. The capability of the system to detect luminescence coming from different regions can be helpful for documenting the variation in the intensity of the individual grains or regions while heating or recording OSL. It will be helpful in finding the spatial distribution of the quartz and feldspars on the surfaces. Earlier such measurements were though possible but time consuming as for recording TL, each grain need to be recorded individually and then compared, similar is the case with OSL. On the other hand in present system individual measurement is not needed, and the properties of different region or grains can be studied in a single run. This aspect needs to be formally established.

12. The imaging capability of this system can be used to find out the uniformity of the phosphor development procedure, by analyzing the luminescence signal from different regions of a phosphor.