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Annual dose measurement for luminescence dating in Salihli, Turkey

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Abstract

Determination of the annual dose level of an area is one of the most important parameters in calculating the geological and archaeological age of the sample using luminescence techniques. Therefore, the concentrations of the natural radionuclides in soils and samples have to be determined since naturally occurring radioactivity provides a major contribution to the annual dose.

In this study, the annual dose level of Salihli, Turkey, was determined with 2 different methods: an indirect method that involves the measurement of the concentrations of the naturally occurring radionuclides and a direct method in which thermoluminescence dosimeters (TLDs) are used.

The concentrations of the natural radionuclides in soil samples were determined using HPGe γ-ray spectrometry. In the direct method, Al_2O_3:C thermoluminescence dosimeters were used. These dosimeters were chosen because of their high sensitivity and usability in dating studies. The results obtained from both methods were compared and discussed.

Key Words: Annual dose, gamma spectroscopy, TLD, luminescence dating

Introduction

Over the past decade there have been significant developments in luminescence, particularly luminescence dating. Several studies have shown that luminescence techniques applied to geological and archeological samples can be successfully used for dating (Göksu and Schwenk, 2000; Göksu et al., 2001; Hütt et al., 2001; Fattahi and Stokes, 2003; Brooke et al., 2008; Prescot and Habermehl, 2008; Veronese et al., 2008). Luminescence dating techniques make use of the mineral grains present in all types of sediments (Aitken, 1985). The luminescence is a measuring process of the light resulting from the release of electrons and holes from metastable trapping states at crystal defects. Energy originated mainly from radioactive impurities (e.g. U, Th, K) present in sediments is stored at metastable traps. When a sample is subsequently stimulated by heating or by exposure to strong light, electrons and holes are ejected and radioactively recombine with carriers of opposite charge. Measurement

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of the photons thus emitted is known as the natural luminescence, which gives a measure of the trapped charge accumulated since the last excitation in antiquity (McFee and Tite, 1998). This is called paleodose (P) or equivalent dose (ED) (Aitken, 1985; Wintle, 1997; Adamiec and Aitken, 1998).

The time elapsing since the last emptying of the traps is determined by the equation

$$\text{Age } (ka) = \text{ Paleodose } (Gy)/\text{Annual dose } (Gy.ka^{-1})$$

where annual dose is the dose sample received in a year.

The total radiation exposure received by the sample (ED) is estimated by dose measurements, while the levels of radioactivity (annual doses) in sites are determined by testing in areas near finds of datable materials. Ages are obtained as the ratio of total dose to annual dose, where the annual dose is measured and assumed to have been similar in the past.

Grain size plays an important role in the luminescence dating of sediments. Fundamentally, there are 2 methods in the luminescence dating of sediments according to their size: fine grain (0-10 μm) and coarse grain (>90 μm) (Fleming, 1970; Zimmerman, 1971). Coarse grain minerals are commonly used in luminescence dating in sediments. In this technique annual dose consists of the contribution of gamma and cosmic radiations and beta particles, but that of alpha particles is negligible because of their low penetration (Aitken, 1985, 1998; Wintle, 1997; Adamiec and Aitken, 1998).

One of the important factors in luminescence dating of sediments is the assessment of the annual radiation dose (induced by the radionuclides U, Th, and their decay series, K, and to a lesser extend by Rb and the cosmic radiation) with adequate precision and accuracy. Annual radiation dose determination techniques are classified into 2 categories, according to whether they result in indirect or a direct measurements (Aitken, 1985; Göksu et al., 1998; Göksu and Schwenk, 2000; Hossain, 2003).

Details of the annual dose procedures in dating are described in various studies (Aitken, 1985; Roberts, 1997; Aitken, 1998; Veronese, 2008). Annual radiation dose analyses in the indirect determination category include neutron activation analysis (NAA), inductively coupled plasma mass spectrometry (ICP-MS), alpha-counting, beta-counting, and gamma-spectrometry, to name a few. These techniques are termed indirect because they yield a radionuclide concentration (in mg/kg or Bq/kg) or a count-rate that subsequently needs to be evaluated in terms of energy absorbed per unit of mass and time (Gy/a). This "conversion" is accomplished by the use of tabulated factors (Aitken, 1985), which are based on measurements of the energy of emitted particles and radiations as published in nuclear data tables. The annual dose can also be directly determined (i.e. directly in terms of Gy/a), by means of highly sensitive luminescent dosimeters. The phosphor must be sufficiently sensitive to allow the measurements to be carried out on laboratory timescales. An example of a suitable dosimeter is Al_2O_3:C (Akselrod et al., 1990; Göksu et al., 1999, 2002; Kalchgruber et al., 2002; Goedicke, 2006; Kalchgruber and Wagner, 2006).

Some fundamental problems confronted with annual dose measurements are the effect of seasonal changes and moisture in the sampling area, the duration of the measurements procedure, the missing of dosimeters measurements in situ, calculation of cosmic radiation dose rate, and the difficulty of low level natural radioactivity measurements for the soil samples.

In this work, 2 techniques were used; (i) indirect determination of the annual dose, namely gamma-spectrometry analysis, and (ii) direct determination using Al_2O_3:C dosimeters. Each technique has its own merits, and the possible presence of additional disturbing effects. Annual dose measurement by the direct

method takes at least 12 months but the same measurement by the indirect method takes only a couple of weeks. In direct measurement, it is taken into account of all the seasonal changes and moisture content in the sampling site. Moreover, cosmic radiation dose contribution is determined experimentally in the direct method while it is calculated theoretically in the indirect method. As the thermoluminescence dosimeters are used to determine annual dose, there is a possibility of missing of them in site and this causes a loss of money and time. However, annual dose determination using radioactivity analysis involves some difficulties resulting from measurement of low level radioactivity in soil samples. Therefore, the determination of the annual dose using several techniques is advantageous.

This region is known to be the site of widespread active continental extension and forms the eastern part of the Aegean extensional province. For example, there are many studies about the fault system in this area (Sozbilir, 2002; Bozkurt and Sozbilir 2004, 2006; Akyol et al., 2006). Therefore, determining the annual dose rate of the region is very important in dating earthquakes that previously occurred.

Materials and Methods

The sampling site in this study is located in western Turkey at 38°29′16″ N and 28°01′43″ E and 292 m a.s.l. near the Aegean Sea (Figure 1).

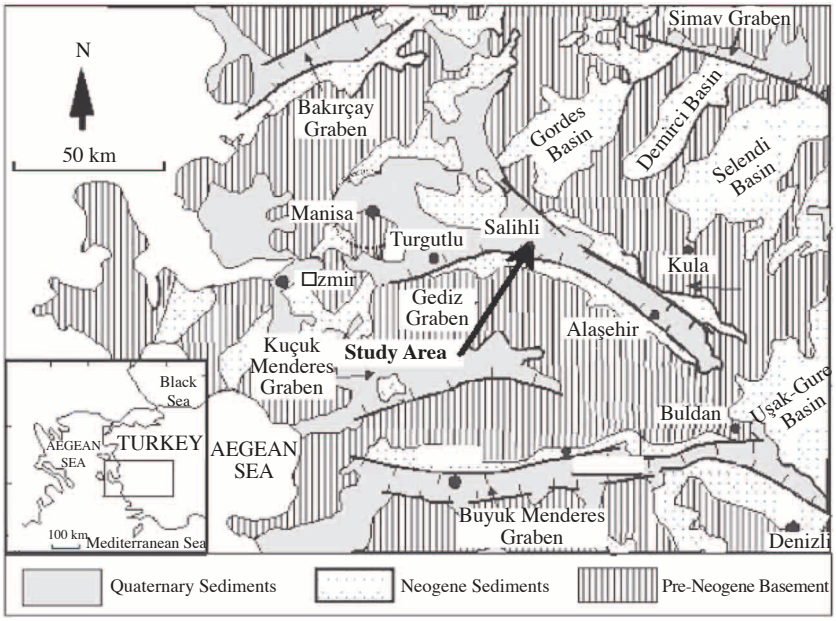

Figure 1. Regional geological map showing the major structural elements of western Turkey, specifically the Gediz and Büyük Menderes Grabens and surrounding Neogene-Quatenary basins (Sozbilir, 2002).

Annual dose measurements were conducted with 2 different methods: direct measurement via thermoluminescence dosimeters and indirect measurement via determination of radionuclide concentrations in the immediate environment.

Direct measurements

A commercial automated TLD reader was used for luminescence measurements. In order to measure the gamma and cosmic radiation contribution to annual dose, thick Al_2O_3:C pellets 5 mm in diameter and 1 mm thick

produced by Landauer were used. The dosimeters were pre-annealed in air at 900 °C for 15 min and quenched to room temperature to remove any existing charge from traps. Since the dosimeters are sensitive to daylight they were protected from direct light during handling, irradiation, and readout. In order to assess the dose rate, thick Al_2O_3:C chips were buried at the site so that gamma and cosmic dose rate were determined. Each dosimeter was wrapped in thin lightproof aluminum foil to shield it from alpha and beta radiation and placed in a 30 cm deep hole opened in the soil within a plastic tube (Aznar et al., 2003). After keeping it for about 14 months on site, glow curves of dosimeters were measured using a TLD reader (Harshaw 3500). A heat absorbent filter (Shott KG-1) was placed beneath the photomultiplier (PM) tube in order to reduce the blackbody radiation reaching the PM tube. Thermoluminescence (TL) measurements were done by heating the sample to 350 °C at a rate of 2 °C.s^{-1} in a nitrogen atmosphere. Background measurements were made after each measurement.

In order to determine beta radiation contribution to annual dose, thin Al_2O_3:C dosimeters were used. First, sediment samples of 2 g obtained from the holes in the sampling area, opened to insert Al_2O_3:C dosimeters, were put into a plastic sample container to have a beta thick sample. The container was wrapped in a Mylar sheet. Then the samples were placed into a thick lead chamber to shield them from external gamma radiation, and left in the chamber for several weeks. Thus, the dosimeters were mainly exposed to beta radiation from the sample. To determine gamma background and subtract from the measurements, thin Al_2O_3:C dosimeters were located under the pure quartz in the chamber (Göksu et al., 1999, 2002; Kalchgruber and Wagner, 2006).

The TL signal intensities were calibrated for radiation dose using a secondary standard $^{90}Sr/^{90}Y$ source delivering an equivalent dose rate of 3.6 and 5.7 mGy s^{-1} for thick and thin Al_2O_3:C dosimeters, respectively.

Indirect measurements

One hundred grams of sediment, extracted from the hole, was used for laboratory gamma spectrometry in order to determine the gamma and beta dose rates at the location of the dosimeter. All samples were oven dried at 105 °C for 48 h, disaggregated and sieved to separate the <2 mm fraction. They were packed into a plastic holder 6 cm in diameter and 10 cm in height (100 g) for the measurement of ^{40}K, ^{232}Th, and ^{238}U activity concentration. In order to achieve secular radioactive equilibrium between radon and progeny sealed samples were stored for 40 days. The samples were analyzed for the radionuclides by direct gamma assay, using Tennelec/Nucleus HPGe (184 cc) planar type coaxial intrinsic germanium detector (FWHM: 850 eV at 122 keV, FWHM: 1.85 keV at 1.33 MeV, relative efficiency 25% and peak/compton = 57/1). The detector was situated in a chamber with 54 × 34 × 29 cm^3 dimensions and surrounded by 11 cm of lead shielding. ^{40}K, ^{238}U, and ^{232}Th activities were determined from the net area under the full-energy peak in the spectrum at 1461 keV, 1764 keV (^{214}Bi), and 2614 keV (^{208}Tl), respectively. Counting times were 30,000 s, providing a precision of ±10% at the 95% level of confidence. Minimum detectable activities based on Curie (1968) criteria for 30,000 s is determined as 27 Bq/kg for ^{40}K, 6.74 Bq/kg for ^{232}Th, and 6.69 Bq/kg for ^{238}U. U and Th standards used in the measurement were prepared in the same matrix and geometry with the samples using IAEA certificated S-13 (118 ppm) and S-16 (600 ppm) reference materials. K standard was prepared by using KCl (52.45%) (Bolca et al., 2007).

Annual dose-rates were calculated from the uranium, thorium, and potassium contents for each sample using the conversion factors given by Adamiec and Aitken (1998). A mean value of 0.2 mGy/a was taken into account for the cosmic dose-rate.

Water saturation content (W) and fraction of saturation (F) were 0.18 and 0.20, respectively.

Results and Discussion

After thick Al_2O_3:C dosimeters were placed in a 30-cm-deep hole at a sampling location and kept for several months on site, thermoluminescence signals of dosimeters resulting from naturally occurring radionuclides were measured using the TLD reader. After each dosimeter was irradiated with a dose of 36 mGy by using a calibrated ^{90}Sr-β source their glow curves were recorded under the same experimental conditions.

The measurements were made using several thick Al_2O_3:C dosimeters and mean values of the intensities were recorded. Figure 2 shows an example of glow curves of one of the thick Al_2O_3:C dosimeters after being kept for 14 months inside the hole.

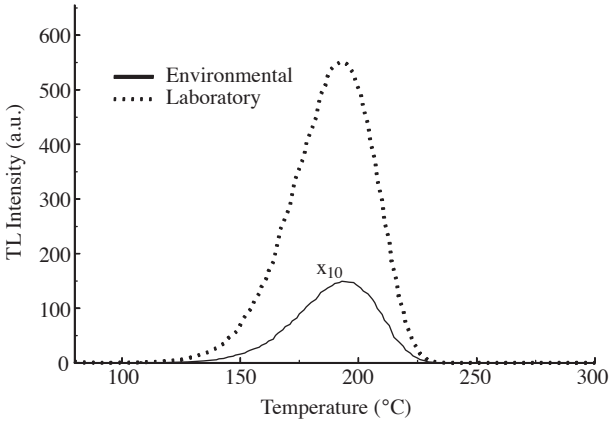

Figure 2. Glow curves of environmental radiation and laboratory irradiated thick Al_2O_3:C dosimeter.

Glow curves of environmental radiation and β irradiated dosimeters were integrated between 125 and 250 °C. Gamma (D_γ) and cosmic (D_c) radiation dose contributions were calculated using the ratio of the integrated area of glow curves of environmental radiation and laboratory irradiated dosimeters. There is no need to do moisture correction for this value since dosimeters were left in their environmental conditions. The gamma and cosmic radiation dose contribution to annual dose by the direct method was found to be 1.38 ± 0.06 mGy/a (Table).

Table. The annual dose values of Salihli region determined using the direct and indirect method.

	D_γ (mGy/a)	D_c (mGy/a)	D_β (mGy/a)	Annual Dose (mGy/a)
Direct method	1.38		2.03	3.20 ± 0.40
Indirect method	1.02	0.20	1.84	2.88 ± 0.72

The beta dose rates from the sediment were measured using the thin Al_2O_3:C dosimeters located in a thick lead shielded chamber. The background gamma dose inside the shield was determined using thin-layer dosimeters stored under pure quartz samples for the same period of time.

The glow curves of thin Al_2O_3:C dosimeters on both sediments and pure quartz were recorded (Figure 3). Then all dosimeters were calibrated using a ^{90}Sr source and the beta radiation dose (D_β) contribution to the annual dose was found to be 2.12 ± 0.26 mGy/a. Moisture correction was made for this value using water saturation content (W), fraction of saturation (F) factors, and the equations given by Aitken (1985)

and Adamiec and Aitken (1998) since the sediment samples used in this procedure were dried. The moisture corrected beta dose rate by the direct method was found to be 2.03 mGy/a (Table).

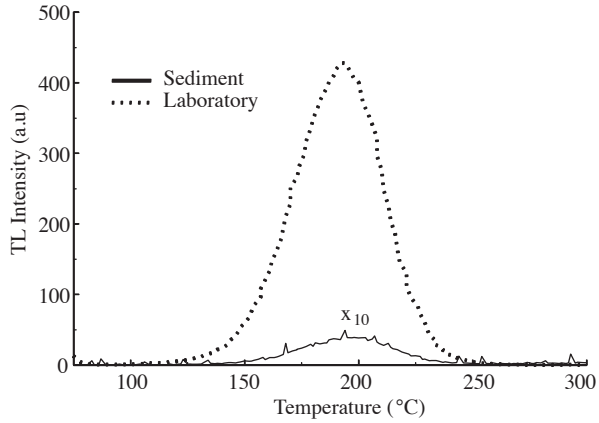

Figure 3. Glow curves of thin Al_2O_3:C dosimeter irradiated by sediment in the lead chamber and laboratory irradiated thin Al_2O_3:C dosimeter.

In indirect measurements, activity concentrations of ^{40}K, ^{238}U, and ^{232}Th radionuclides obtained by HPGe gamma spectrometer were found to be 507 ± 50, 31 ± 5, and 30 ± 5 Bq/kg, respectively. Gamma and beta radiation dose contributions to the annual dose were calculated by using these activity concentrations and components of annual dose given in the literature (Aitken 1985, 1998; Adamiec and Aitken, 1998). Moisture corrections were made for the gamma and beta radiation dose values since the samples used in activity measurements were dried. Gamma and beta radiation dose rate results for indirect measurements were 1.02 ± 0.25 and 1.84 ± 0.46, respectively (Table). The contribution of the cosmic dose to the dose rates was evaluated to be 0.20 mGy/a by assuming the elevation, latitude, and soil density (292 m, 38°, and 2.63 g cm^{-3}) of the sampling site (Prescott and Stephan, 1982; Prescott and Hutton, 1988, 1994).

Annual doses for coarse grains in the current study were calculated by the following equation and are given in the Table (Aitken 1985, 1998).

Annual Dose = $0.90D_\beta + D_\gamma + D_c$

Atlıhan and Meric (2008) studied luminescence dating of a geological sample collected from Denizli, Turkey, using the fine grain technique. They obtained alpha, beta, and gamma dose rates by determining concentrations of the major radioactive isotopes of the uranium and thorium series and of the potassium within the surrounding materials using a high-purity germanium detector. They were found an annual dose of 1.86 mGy/a in their study. This value is lower than the results of the current study, which may result from the radionuclide concentration differences between sampling areas.

In research in the Menderes Massif in Turkey, the concentrations of ^{238}U, ^{232}Th, and ^{40}K were found by using α-counting and flame photometry. Based on these values, the annual dose was found to be 0.61 mGy/a. The differences compared to the results of the present study can be explained in terms of the ^{238}U, ^{232}Th, and ^{40}K activity concentrations being much smaller, which may be due to the severe environmental conditions, such as erosive, naturally occurring chemical etching, and weathering processes (Ulusoy, 2000).

In another study conducted by Öke and Yurdatapan (2000) in Muğla, Turkey, the concentrations of uranium and thorium were determined by using the alpha counter and the potassium by atomic absorption spectrometry.

The annual dose obtained in the study was reported to be 3.93 mGy/a.

Guérin and Samper (2007) investigated the dating by thermoluminescence of quartz from 6 volcanic formations of the Saint Lucia Island (Lesser Antilles Arc). In this study, uranium, thorium, and potassium concentrations were measured by high-resolution gamma spectrometry, and cosmic ray contribution has been estimated as 0.080 mGy/a. Alpha contribution to the annual dose was ignored due to the same reason as in the current study by assuming that the HF etching had removed their outer parts. The annual dose determined in the study was between 1.63 and 2.17 mGy/a.

Conclusion

A comprehensive investigation was performed to measure the annual dose using direct and indirect measurements in the Salihli region of Turkey. In this study, a comparison of these techniques for annual dose determination was conducted for coarse quartz grains. The total annual dose obtained by direct measurement for the was 3.20 mGy/a and that by the indirect measurement was 2.88 mGy/a. Data obtained from both measurement techniques are in good agreement with each other. It was observed that the determination of annual dose by using the direct measurements takes several weeks, while the same process with direct measurements takes several months since the dosimeters had to be left at the sampling site for several months. Therefore, the indirect measurements seem more advantageous. On the other hand, in indirect measurements cosmic dose contribution can be evaluated theoretically but in direct measurements it can be determined experimentally.

One of the main contributions of this work lies in the positive new results obtained, which at least show that it is possible to use these methods to date other samples from the sites in the nearby area. Previously, no luminescence dating study had been performed in this region. Therefore, the present work was the first measurement of the annual dose. It is thought that the results obtained in this study may contribute to the promotion of the luminescence dating technique and supply valuable information for geological studies in the Salihli region of Turkey. Further research with these samples is still in progress.

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