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THE VALUE OF OSL IN DISTINGUISHING ANCIENT FROM MORE RECENT STRUCTURES IN AN ARCHAEOLOGICAL LANDSCAPE

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ABSTRACT

OSL dating of a stone built structure directly on the south of the Geometric period settlement of Zagora (Andros island, Aegean sea, Greece) was conducted to determine whether a minor access point to the site was ancient. Double single aliquot regeneration (SAR) protocol was recorded for total dose De calculation, in which samples were first stimulated with IR and the post IR blue light stimulated luminescence (BLSL) signal from quartz grains, at 220°C preheat temperature and OSL signal recorded during 40s blue light stimulation. Alpha counting, XRD, XRF were used for radioisotope content and mineralogy assessment. The construction was found to date to the 19th century CE.

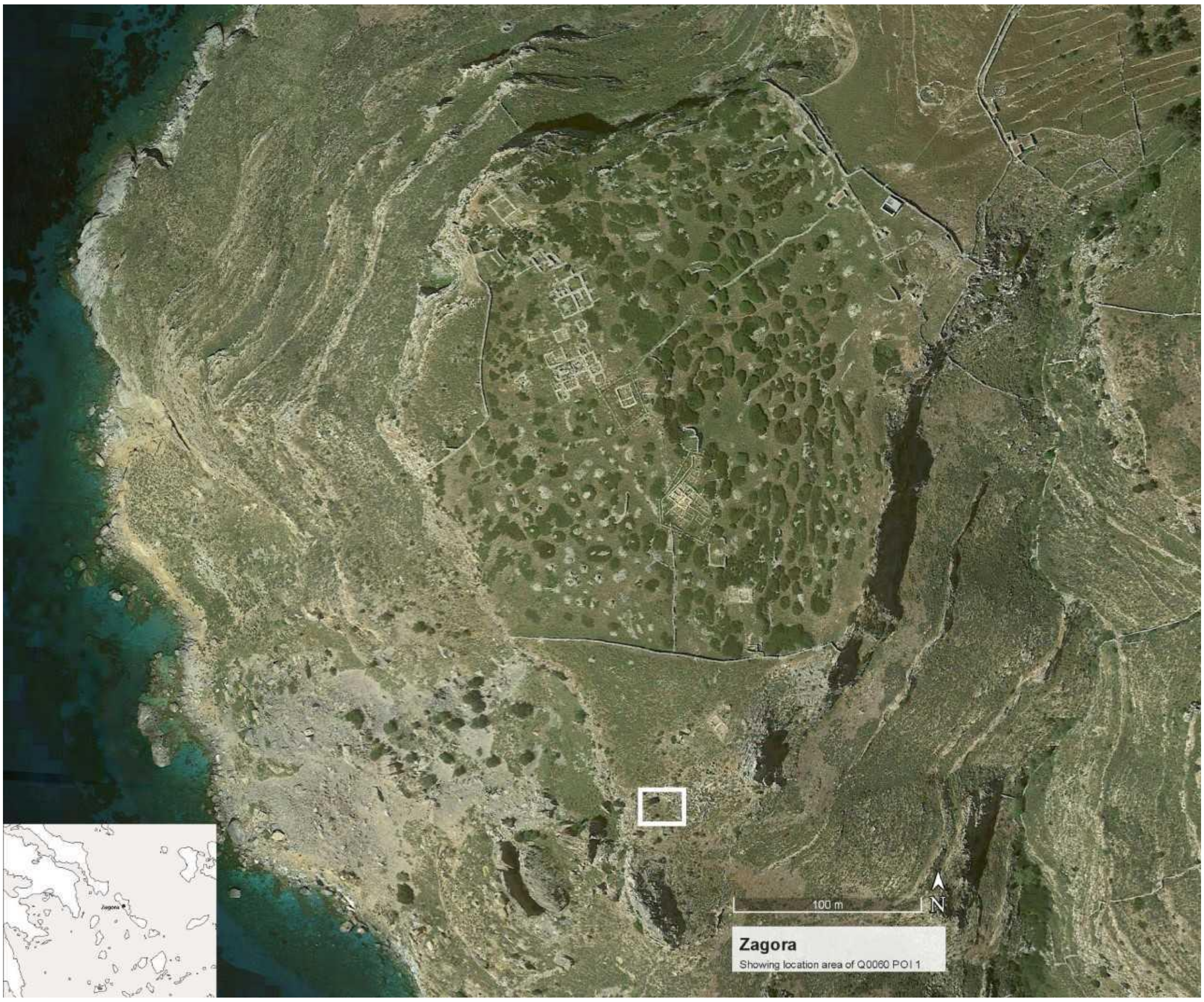
KEYWORDS: archaeology, total dose, dose rate, geometric period, OSL

1. INTRODUCTION

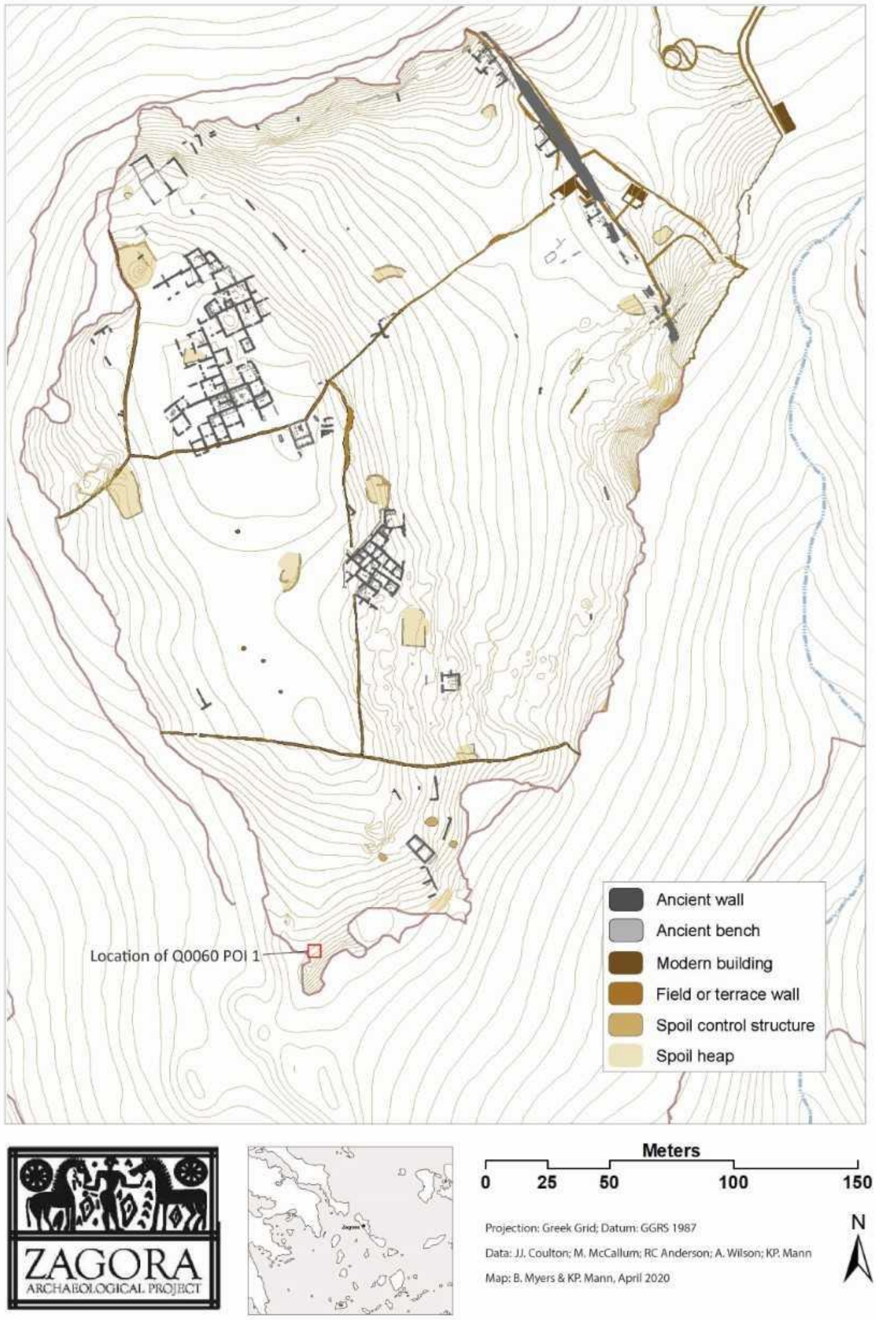
Luminescence is the emission of light from insulating solids due to the eviction of charges trapped in lattice defects. In common minerals of natural rocks (for example, quartz, feldspar, zircon, calcite) exposure to environmental radiation causes ionization and produces free charges that travel in the lattice. Some of these get trapped in lattice defects. Depending on the change in the environment of the trapped charges, their binding energy can imply a mean residence time of a second to millions of years. Given that the radiation flux from environmental radioactivity is constant, the number of charges in traps with longer mean lives, keeps increasing with time and these can be determined using measurement of their stimulated luminescence using either optical or thermal agents (Liritzis 2009a, b; Sobbati, 2013). Based on the stimulation, the emitted luminescence is called thermoluminescence or optically stimulated luminescence. The methodological references may be found elsewhere (Singhvi et al., 2011; Liritzis et al., 2013, 2020; Aitken 1998; Bateman et al., 2007) for further details on luminescence dating.

Optical stimulated luminescence (OSL) dating has developed as a reliable dating method for the late quaternary over the past three decades (Kumar et al., 2018).

In this project, OSL was used to date a stone retaining wall at Zagora, a site on the west coast of the island of Andros, Greece (http://zagoraarchaeologicalproject.org/) Beaumont et al 2012). The period of construction and occupation of the settlement and its fortification wall along its landward slope is dated by ceramic chronology to the Geometric period placed at ca 900-700 BC (Fig.1, a-b).



(1a)



(1b)

Figure 1a. Zagora Google Earth view; 1b. Zagora Site Plan with location of Survey Feature Q0060 POI 1 (database record HID 17335)

Previous research and applications to date stone constructions in an archaeological context have referred to the surface luminescence dating method, where stone surfaces exposed to sunlight set a “zero time” clock at the moment of being placed in a wall and covered by another wall element (Liritzis, 2011). The bleaching of luminescence of the quartz grains in the upper, surficial, layer of granite or limestone stones used in construction can be achieved over a period of time that ranges from a few seconds to some hours in duration, respectively. Thereafter, on construction of the wall, a stone's surface being in the dark with no exposure to sunlight begins to accumulate luminescence due to irradiation from ambient environmental radiation, arising from the radioactivity of stone itself and the surrounding rocky context (Liritzis, 2010; Liritzis et al., 2010 a,b).

The luminescence dating method, apart from providing a chronology, also serves as an authenticity test applied to constructions which are otherwise of unknown date, although suspected (or suggested) to belong to a specific period.

Reconnaissance surveys on the Zagora headland in 2012 observed a low wall constructed of field blocks that supports a terrace (Fig.1a-b). This is located to the south of the site where the slope up to the plateau on which the settlement lies is accessible. To the east, the settlement is protected by a fortification wall with a strategically-positioned gate. No such defensive fortification was required on the inaccessible north and west sides of the promontory. The retaining wall in question is set in such a way as to provide a means of access to the settlement from the gentler slopes south, invisible from those approaching from the east.

Zagora provides a rare example of an early first millennium BCE settlement fortification wall. It might make good defensive sense to have a covert rear egress from the site. Across the site as a whole the mode of construction is largely determined by the available building material–in this case schist and poor grade marble–making it impossible to date securely the construction or repairs to any wall or structure on the basis of style alone. The domestic architecture, so far excavated, is found often to have been constructed in agglutinative fashion. This allows a relative sequence of wall construction to be calculated on the basis of whether walls bond or abut; it is difficult to determine absolute dates of phasing solely on the basis of stratigraphy. The wall under consideration stands alone so that it has no relative chronology. Its construction, a blend of schist and marble rubble, is stylistically undatable. The wall might offer insight into Early Iron Age Greek defensive thinking if it could be securely dated as contemporary with the settlement's lifespan. As such, it presented an intriguing challenge.

2. SAMPLING AND METHODS

The technique developed for the dating of rock surfaces using luminescence dating was applied (Liritzis 2011; Liritzis et al., 2010a, 2013). Therefore, with care three potentially large stones at the very base of the wall were selected and sampled (Fig.2).

(A)

(B)

(C)

(D)

Figure 2. a) Location of ZAP-1 sample, b) The calibrated Geiger Counter for gamma (including cosmic) radiation, c) close up of ZAP-3 sample; the base stone sitting on the ground, and d) the sampling positions. (maximum wall height ca.1.10 m, distance between 1st and 3rd sampling 1,05 m, between 1st and 2nd is 1 m.

Sample 1 (ZAP-1) was taken centrally at the base of the wall, samples 2 and 3 (ZAP-2 and ZAP-3) were taken equidistant from sample 1, also at the base. ZAP 1 and 2 were in contact with the ground soil.

2.1 XRD mineralogy

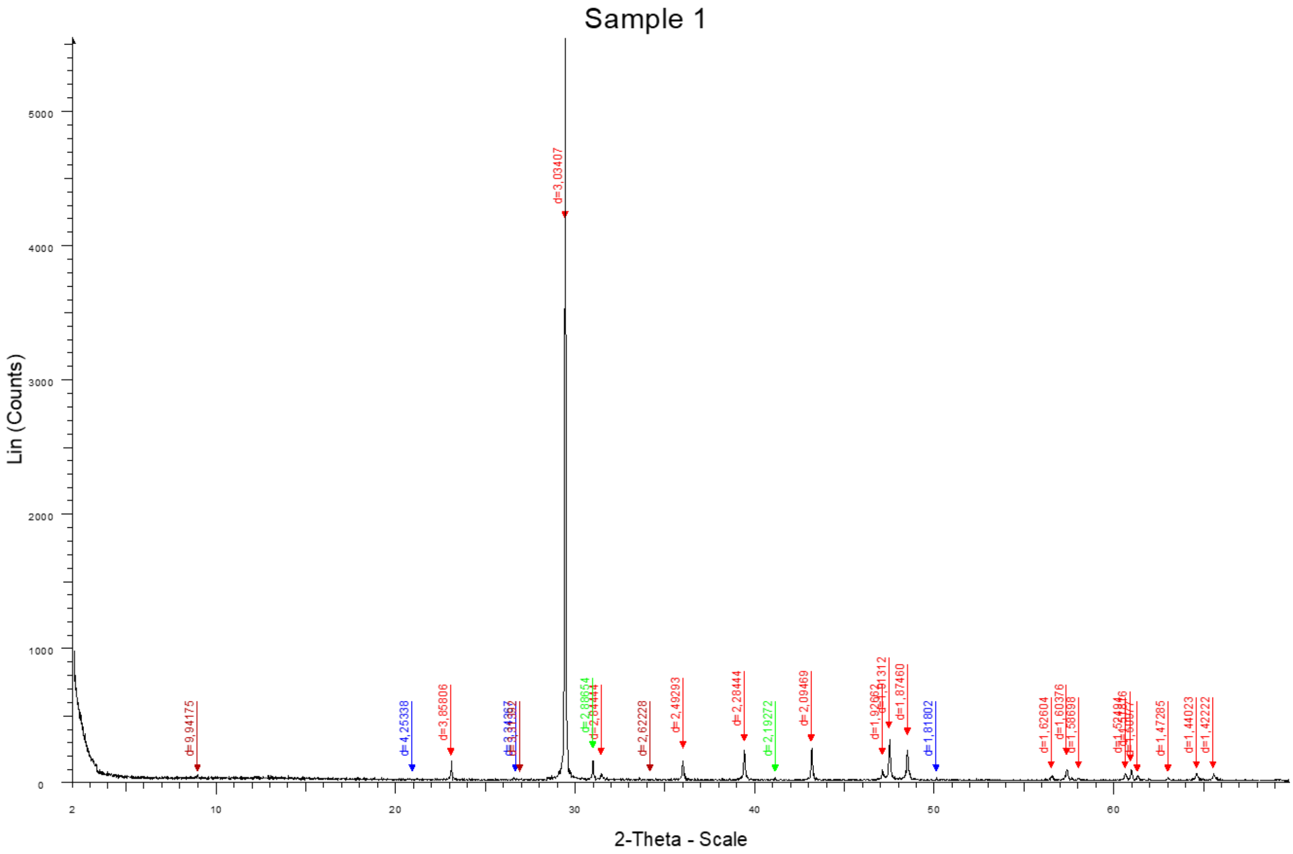
The samples were analyzed by X-Ray Diffraction technique at the Geology Department of Patras University, by Dr I. Iliopoulos). The scanning area covered the interval 2–70◦ (2θ) with a scanning angle step of 0.015◦ (2θ) and a time integration duration of 0.1 s (Iliopoulos et al., 2011).

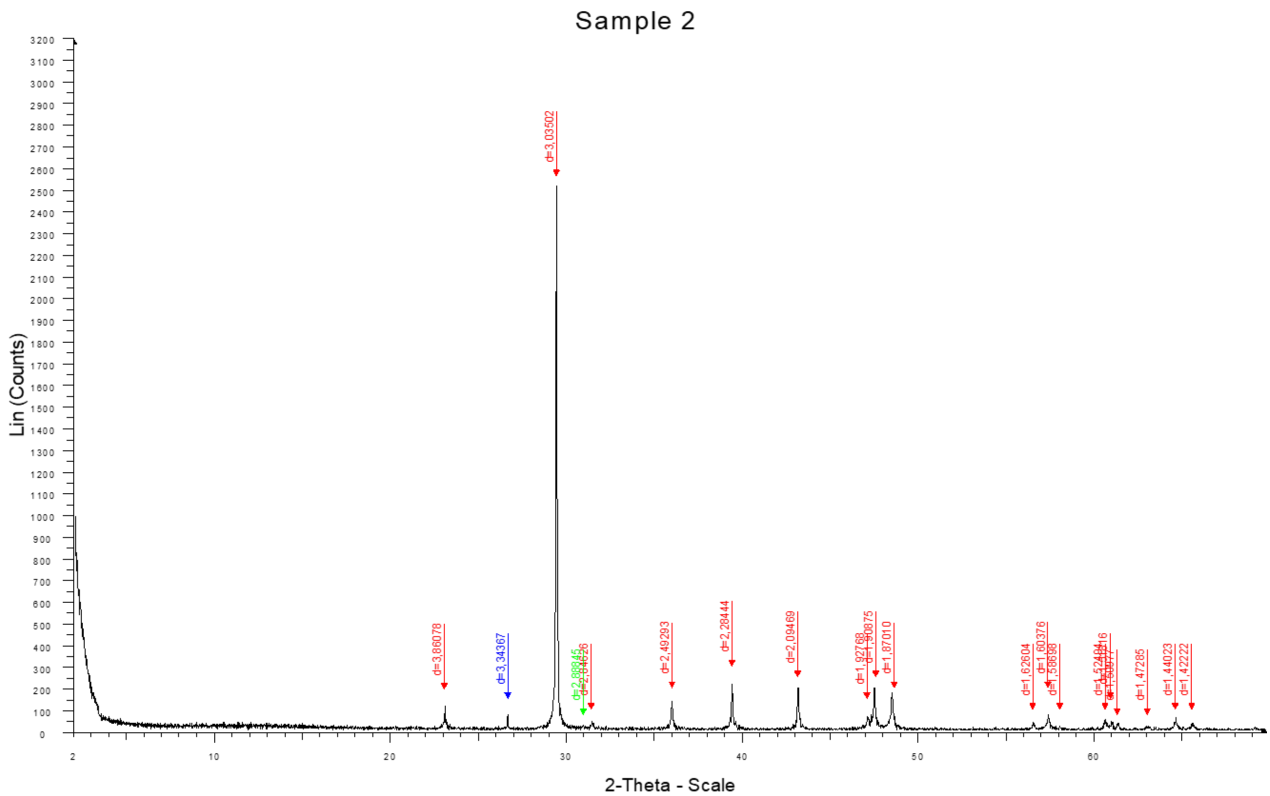
XRD results of the three stone samples were as follows in Table 1:

Table 1. XRD data for the three samples (+++ predominance, + trace, - absence)

	Calcite	Dolomite	Biotite	Quartz
ZAP1	+++	+	+	+
ZAP2	+++	+	-	+
ZAP3	+++	+	+	-

In all stones calcite is the predominate phase (limestone). Limited quantities of quartz were seen in ZAP 1 and 2 and in ZAP3, no quartz was identified (Fig.3).


Sample 1
Lin (Counts)
2-Theta - Scale


Sample 2
Lin (Counts)
2-Theta - Scale

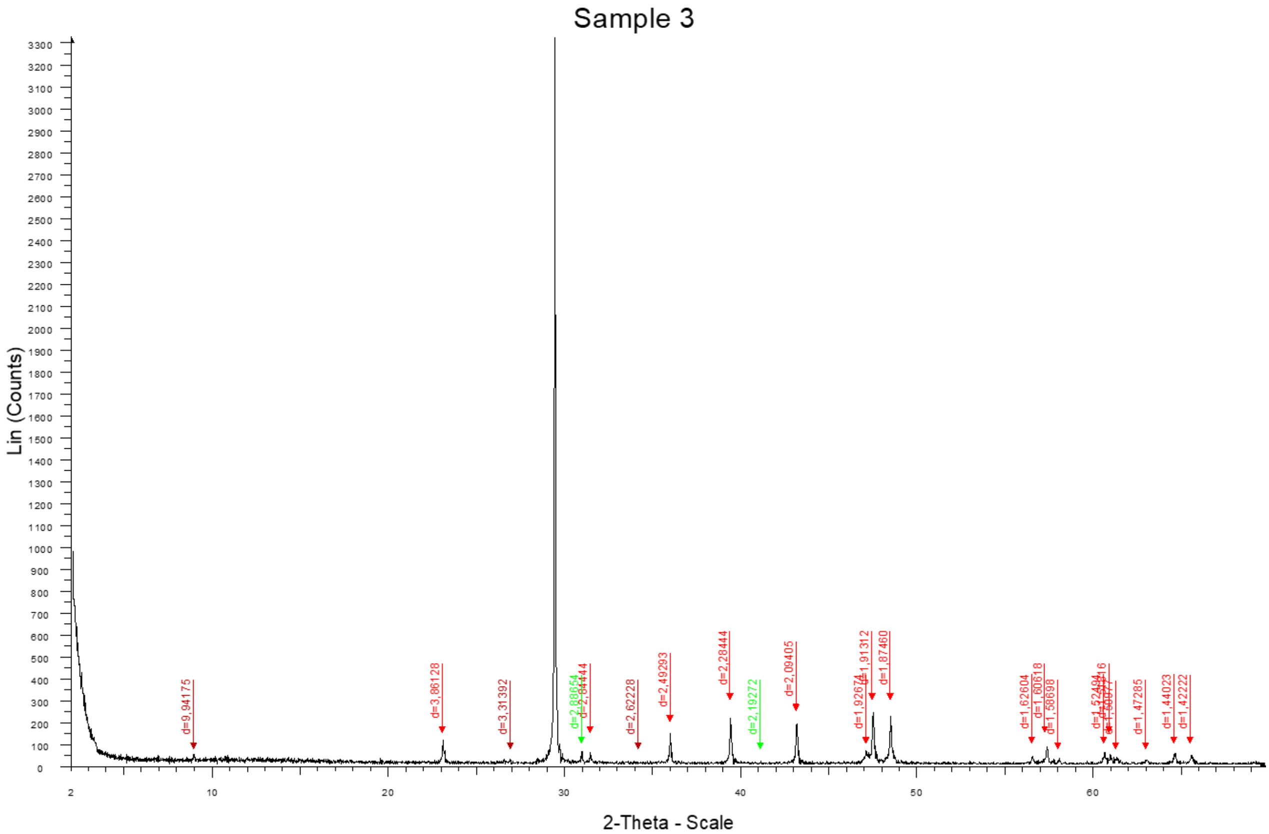


Figure 3. Typical XRD spectrum for ZAP1, 2, 3. ZAP1: with predominate of calcite (red), and traces of quartz (blue), dolomite (green), ZAP2: calcite (red), quartz (blue), dolomite (green), ZAP3: calcite (red), dolomite (green), biotite (brown).

The dolomite presence was significant in ZAP1 followed by lower amounts in ZAP3 and ZAP2. In ZAP1 and 3 small amounts of biotite were recognized too. This diffraction analysis confirmed the presence of quartz in two samples and for the third, absence of quartz did not exclude its presence in finer grain size. Thus, sample preparation for quartz extraction was applied to all 3 samples (Liritzis et al., 2010b).

2.2 Dose Rate Evaluation

The dose rate arises from the cosmic rays, the uranium, thorium, and potassium contained in of the rock and in the surrounding environment. The emitted radiation comprises of gamma rays, alpha and beta particles (Liritzis et al., 2013).

A portable Radiagem 2000 (Canberra) alpha, beta, gamma probe Geiger Counter was used, with connection of the probe directly or via cable to radiagem. The detector is pancake Geiger Muller, mica window (thickness 2mg/cm^2, metal protection grid with 75% transparency for detection area 15.5 cm^2 and lower limit of detectable gamma energy 30 KeV). Readings on counts/second were converted to mGy/yr after a successful calibration procedure based on radioactive pads and comparison with a portable calibrated NaI scintillometer (SCINTREX, model SPP-2) (Fig.2b, Fig.4).

The environmental radioactivity readings (Cg,env) were similar for the three samples as was that of stones and the soil plus cosmic radiation. Cg,env=0.52±0.06 c/s. A similar result was deduced from calculations of individual radiation components of cosmic rays, and the gamma ray dose rate of rock and ground sediment of the almost 2π geometry (see below).

The Geiger counter was calibrated on radioactive pads made at the National Center for Scientific Research "Demokritos", Athens (with Dr Y. Bassiakos). The conversion was made using the linear relationship C = 0.37*D + 0.28 (Fig.3), where C the counting rate (counts/sec) and D the dose rate in mGy/year. The errors were evaluated from: $\Delta D = [((\sigma D/\sigma C)^*\Delta C)^2]^{½}$. Background noise is 0.5c/s in an ambient of 0.1 μGy/hr.

The sediment radioactivity was measured by alpha counting pairs technique. The results were: Uranium (U) = 2.18±0.15 ppm, Thorium (Th) = 6.26±0.50 ppm, while via XRF the potassium (K) = 1%.

The radioactivity of the rock is very low due to calcitic nature (shown by XRD) (see Table 1). For the ZAP1 rock and alpha pairs technique using ZnS (Ag) the radioactive content was: U=0.34±0.03 ppm, Th=0.23±0.10 ppm, K=0.04±0.02% (Rb; negligible).

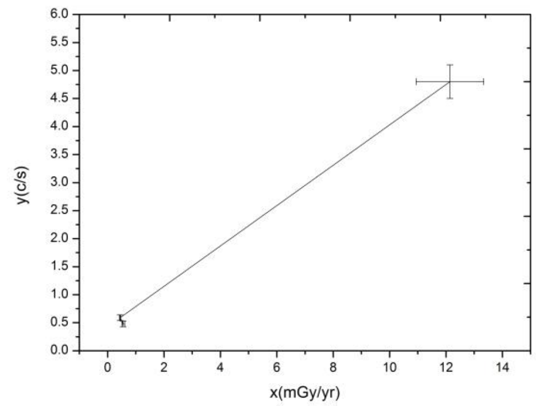


Figure 4. Linear relationship of counts versus dose rate of the radioactive pads, and floor at control room (0.48c/s).

These elements contribute 50% beta dose to the rock surface; the alphas are removed by a diluted acid wash of the surface prior to grinding and powder removal, thus only the lower than the griding stone layer provides half of alphas (powder acquisition from surface is described elsewhere, Liritzis et al., 2010, 2010a, 2010b). The dose rate for the base stone of the wall is complex; half of gamma ray comes from the ground, 1/4th from the wall itself (taken the sampling as the center of a sphere of radius ca 30 cm), the other 1/4th is air and contribution is from the ground where the dose rate at the surface is 70% of that in a depth of 30 cm of an homogeneous pad of known radioactivity, measured by a calibrated NaI (Tl) and LiF, (Liritzis and Galloway 1981). The beta dose-rate from the sediment (divided by 2) was 0.88 mGy/yr, while the contribution of alphas and betas from the rock itself was: Da=0.18 mGy/yr, Db=0.89 mGy/yr and the self-gamma-ray dose-rate from calcitic stone almost negligible.(Liritzis, 1986).

Thus, the total annual dose rate for a 20% water uptake is: $D_{annual} = d_{\gamma env}$ (+cosmic) + $da/2+db_{soil}/2+db_{rock}/2$ = 1.85 mGy/yr. Annual average water uptake from the local humidity is considered low (±20%), due to the location on a slope and winds as well as the proximity to the coast that dry the soil (Fig.1a-b).

2.3 Paleodose

The samples from the rock surfaces were taken from the outer mm of stones. The extracted quartz grains were measured at Physical Research Laboratory, Ahmedabad in India and these were processed in the laboratory under red-light conditions. The samples were initially treated with 0.01N sodium oxalate solution to de-flocculate the fine grains. The fine grain fractions (4-11 μm) were separated using density settling following Stokes' law. The separated particles were then dispensed to the aluminum discs under an alcohol medium and allowed to dry in the oven under 45°C to form as a uniform layer. Samples were then subjected to infrared (IR) stimulation to check for feldspar IR stimulated luminescence (IRSL). Generally, the signal obtained from the feldspar grains were poor but to be certain that these grains do not contribute to OSL of the quartz, a double single aliquot regeneration (D-SAR) protocol was used for paleodose measurements. In this the samples were stimulated with IR diodes at 50°C for 40 seconds and then stimulated with blue light diodes to record signal from quartz grains and 125°C for similar time interval.

Only ZAP- 1 gave a measureable signal. Twelve aliquots of sample ZAP-1 were measured using a standard single aliquot regenerative dose (SAR) protocol having 220°C preheat temperature and an OSL signal was recorded during 40s blue light stimulation with 70% power at 125°C over 250 data points (Fig. 5; Singhvi et al., 2011; Kars et al., 2014; Liritzis et al., 2013, 7, 15). The measurements were conducted using a standard Riso- TL/OSL DA-20 reader, fitted with blue diode arrays (λ = 470 nm, power = 18 mW/cm²) and calibrated $^{90}Sr/^{90}Y$ beta source which delivers a dose rate of 0.095 Gy/s was used for the measurements.. The detection optics comprised EMI 9235 QA photomultiplier and 7.5 mm, Hoya U-340 glass filters (Singhvi et al., 2011).

OSL signals were integrated over the channels 1-5 and the corresponding background signal was subtracted at the last 50 channels (Fig. 5). In general, recycling ratios were poor and aliquots with a recycling ratio up to 1.6 were accepted (Fig.6). Over half of the measured aliquots were rejected due to an exceptionally high recycling ratio (>1.5) and high recuperation. An increase in sample sensitivity was observed with higher regenerative dose points from the corresponding test dose response ratio to the test dose response of natural (Tx/Tn ratio). The De reported is the weighted mean of 5 accepted aliquots along with its Standard error (Fig. 7). Some characteristic poor blue light signal of samples ZAP-2 and 3 over time stimulation luminescence are given in Fig.8.

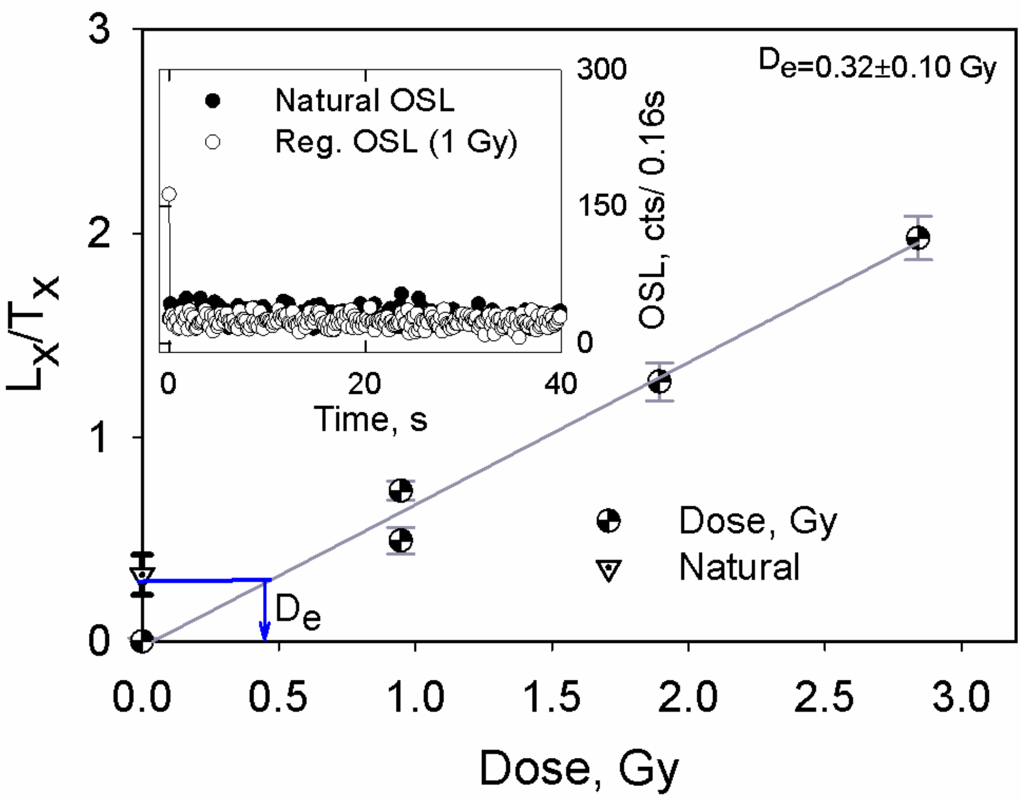


Figure 5. Diagrams showing the a) OSL growth curve of a representative sub-sample (ZAP-1), which had given a De value of 0.32±0.10 Gy. The inset figure shows the natural and regeneration dose decay curve of the same sample.

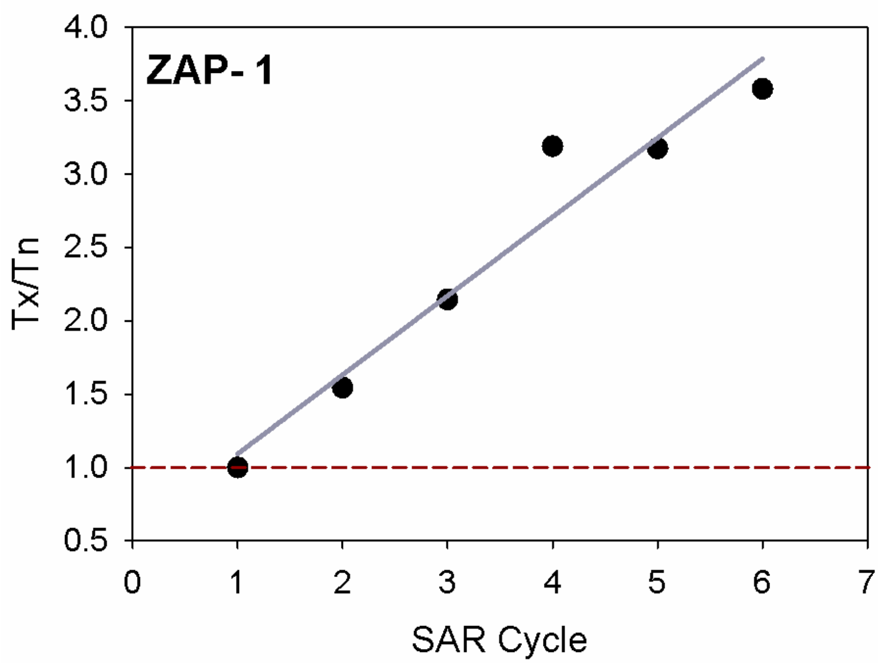


Fig.6 Recycling test for sensitivity change as a function of regenerative dose is plotted in the diagram. Test dose signal of various regenerative dose cycle at different doses over the test dose response of natural signal (Tx/Tn) are plotted in the graph. The values of test dose signal of 0.5 Gy following the regenerative doses; R1, R2, R3, R4 and R5 at 1, 2,3,0,1 Gy, respectively with the corresponding Tx/Tn values are plotted here showing increasing sensitivity of the sample with increased regenerated doses and progressive dose points.

The ZAP-2 sample with trace of quartz (like ZAP-1 sample) seems to have been exposed to light and gave a background luminescence signal. The evaluated total dose of the sample ZAP-1 was 0.33±0.16 Gy. The error is the 1σ standard deviation over the average, with individual errors per aliquot ca. ~50%.

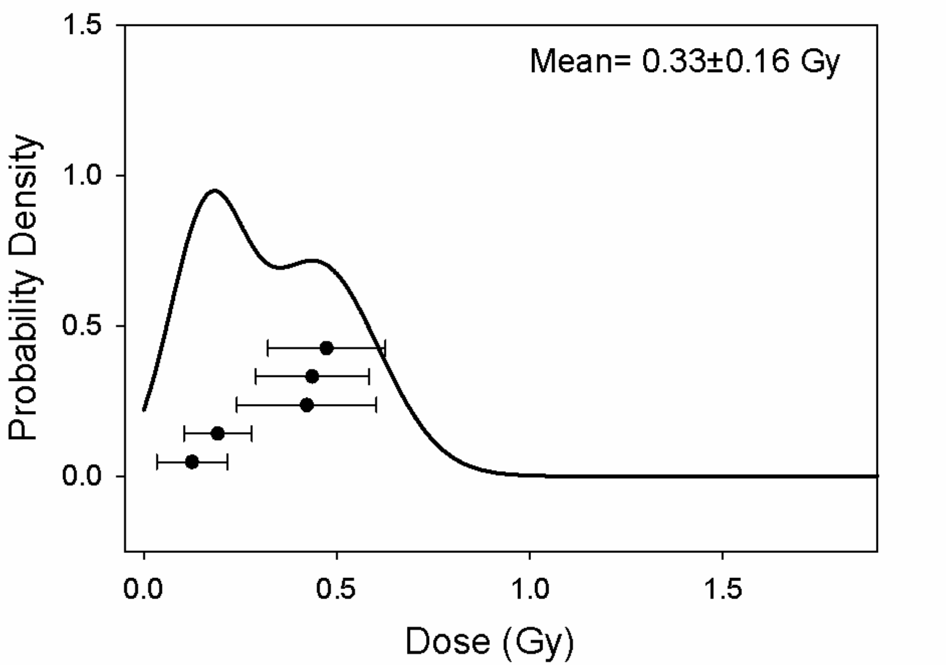


Fig.7 De distribution of sample ZAP-1 with mean value of accepted aliquots are shown. Twelve aliquots were measured for their OSL and five have passed the acceptance criteria and provided a mean equivalent dose of 0.33±0.16 Gy.

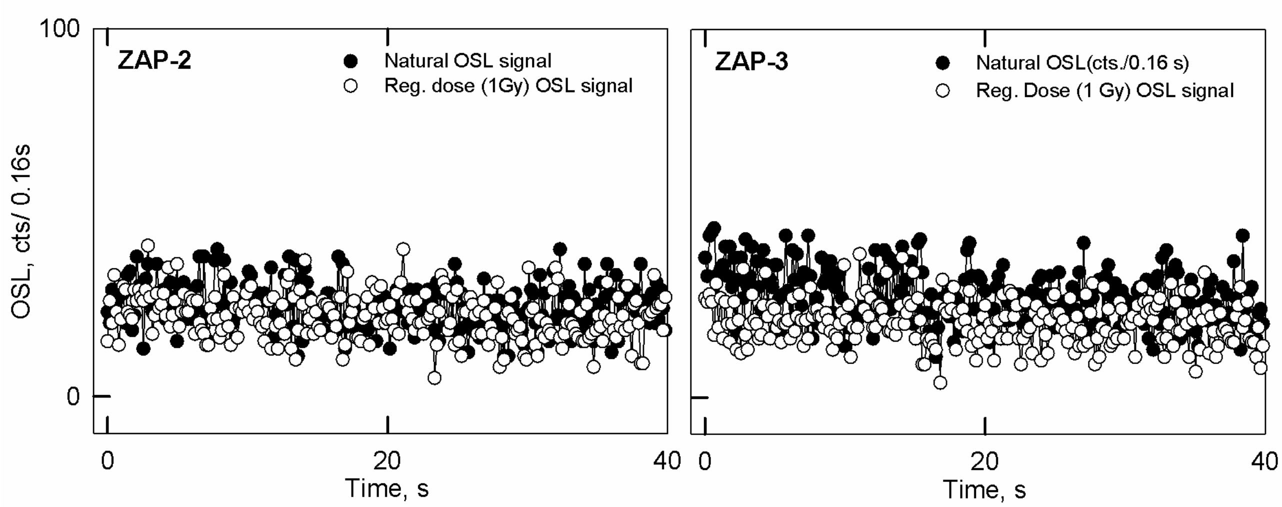


Figure 8. The OSL decay curve of samples ZAP-2 and ZAP-3 showing poor noise to signal ratio for both natural and regeneration dose. The figure also shows the insensitivity of the sample to the regeneration dose of 1 Gy (compare with the acceptable signal for ZAP-1, inset of Fig.5).

It is likely that the grains from a given rock have a high ultra fast component and low medium and slow components. With the recycling treatment of light and irradiation, the medium and slow components are sensitized and hence the over-all curve is what we see in a normal quartz. Our samples ZAP-2 and ZAP-3 have little medium and slow components which indicates that the samples were taken directly from the source rock, which had no time to be sensitized prior to incorporation in the wall. The presence of an ultrafast component in ZAP-1 suggest that the sample was from very near the source and was possibly not sensitized sufficiently. At any rate, our result is provisional as number of aliquots were small to meet the acceptance criteria and due to sensitivity issues, an often encountered problem with not heated materials. Despite this, the age would indicate a recent construction.

2.4 Age calculation

The age calculated (Total Dose / dose rate) for the sample ZAP-1 was 180 ± 15 years, which implies a recent construction made by local farmers/ shepherds during the 19th century.

3. CONCLUSION

The application of the OSL dating method as a case-study at Zagora has provided some bounds on the age of construction of the wall. It also shows that with some additional efforts, dating of built structures without associated archaeological deposits can be attempted using OSL. Sampling from the basement row of stones is essential as it is expected to represent the original construction horizon. In the present case study the recent date of the stone structure contributes to the story of modern engagement with an archaeological landscape rather than the history and development of settlement defensive systems in Geometric Greece.

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