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HISTORICAL MORTARS DATING FROM OSL SIGNALS OF FINE GRAIN FRACTION ENRICHED IN QUARTZ

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Abstract: In the last years the mortar dating through Optically Stimulated Luminescence (OSL) techniques has become a viable support for chronological estimations (date of construction or restoration episodes) of historical buildings. However, the dating of mortar has still open issues mainly regarding the assessment of the bleaching degree of quartz, the analysis of the OSL processes for this type of samples and the need to do appropriate tests for the most correct evaluation of the equivalent dose. This paper discusses the results obtained by OSL dating (blue diode stimulation) on the polymineral fine grain phase, enriched in quartz, extracted from lime mortar samples collected from different sites. Thermal transfer effects, through the behaviour of Equivalent Dose (ED) and recovery tests, degree and time of bleaching were studied. For each mortar sample the adjacent brick was collected; in some cases, sampling of the bricks bracketing a mortar layer was a possibility, thus obtaining a direct comparison with the standard thermoluminescence (TL) dating on the bricks.
The results obtained show, for this set of samples, the possibility of dating the mortars through the use of the fine grain fraction provided of a suitable chemical-physical preparation procedure and the verification of the bleaching conditions.

Keywords: historical mortars, luminescence dating, partial bleaching test, preheating test, age model.

1. INTRODUCTION

Luminescence measurements of mortars that were initially applied in retrospective dosimetry made use of the OSL emissions of sand-sized quartz (90-180 μm), (Bøtter-Jensen *et al.*, 2000b). These confirmed that the grains were bleached to a considerable degree during mixing. The results show the potential of using mortars for dosimetric purposes, although they cannot be taken as evidence that, in principle, the mortars are datable. A first attempt to perform optical dating on mortars was carried out by Zacharias *et al.* (2002) on 90-250 μm quartz inclusions extracted from samples taken from two Byzantine churches. The authors also performed TL dating on brick fragments associated with the mortars. The results were characterized by quite large uncertainties and the lack of material prevented from further research. Goedicke (2003), using a model-calculation on samples of known

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age, suggested the use of OSL-decay curves to evaluate the bleaching of different quartz grain size fractions during manufacture and the possibility of using the conventional SAR protocol for dating.

The difficulty to extract pure coarse grain quartz fractions of sufficient quantity from sand used as inert in mortar, has led other authors (Gueli *et al.*, 2010) to study the luminescent emission from polymineral fine grain phase "enriched in quartz" through HF etching procedures during preparation phase of the samples (Prasad, 2000; Mauz and Lang, 2004). The set of studies so far cited, have confirmed the potential of mortars for dating but have not tackled in detail other issues related to the methodology for the equivalent dose determination and, also, the related experimental steps such as preheating temperature, recovery, recuperation and partial bleaching tests, typically addressed for sediment samples.

Goedicke (2011) has discussed all the problems related to mortar dating including the most suitable methods of equivalent dose and dose rate determination, showing all the adversary properties known from sediments: for instance partial bleaching, low signal and slow decay.

In this paper, in accordance with directions of Goedicke (2011) for mortar dating, OSL measurements by SAR method on polymineral fine grain fraction enriched in quartz are made. The thermal transfer effects were studied through the behaviour of ED and recovery test values against preheating temperature (Choi *et al.*, 2003; Thomas *et al.*, 2003; Kiyak and Canel, 2006). In order to investigate the degree of bleaching, the ED frequency distributions (Olley *et al.*, 1998; Murray and Olley, 2002; Goedicke, 2003; Saini and Mujtaba, 2010) and time of bleaching in the easy-to-bleach OSL traps (Agersnap Larsen *et al.*, 2000) were studied. The OSL ages were obtained using the Central Age Model (CAM) (Galbraith *et al.*, 1999) and the results were compared with TL dating from bricks associated with the mortars.

2. EXPERIMENTAL DETAILS

The studied samples were taken from three historical buildings located in different geographical areas. The monastery of Castelletto Cervo (Vercelli, Italy) and the church of Notre Dame de la Place (Bordeaux, France) were dated within a wider project of the GdRE European Group (TCA – Terres Cuites Architecturales et nouvelles methods de datation). The Convento di San Francesco alla Collina (Paternò, Italy) was dated within a large restoration program (Gueli *et al.*, 2010). **Table 1** shows, for each structure, the presumed age, the ID and the kind of sample as well as the details of sampling points.

Artificial luminescence signals were induced by the ^{90}Sr-^{90}Y calibrated beta sources integrated in the Risø systems delivering, respectively, 6 Gy/min and 1.32 Gy/min for both mortars and bricks. ^{241}Am calibrated alpha source delivering 2.7 Gy/min was used to determine the luminescence efficiency coefficient k necessary to correct the alpha dose contribution to the annual dose (Aitken, 1985; Guibert *et al.*, 2009).

TL for bricks and OSL and IRSL (InfraRed Stimulated Luminescence) for mortars, were performed using the semi-automated Risø readers (TL-DA-10 and TL-DA-15) with EMI 9235QA photomultipliers (Bøtter-Jensen, 1997; Bøtter-Jensen *et al.*, 2000a). TL glow curves were recorded in the TL-DA-10 detection system using Corning 7-59 and Schott BG-12 optical filters. OSL and IRSL signals were obtained using TL-DA-15 reader equipped, respectively, with 41 blue LEDs (470±30 nm) and with a laser diode (830±10 nm). The stimulation units delivered ~30mWcm^{-2} for OSL and ~240mWcm^{-2} for IRSL at 90% power. Both OSL and IRSL emissions were detected using an Hoya U340 optical filter.

3. SAMPLE PREPARATION

After removal of two external millimetres, the mortar samples were pre-treated with 10% H_2O_2 for four days, to remove organic material, and with 20% hydrochloric acid for 120 min to dissolve carbonates. With the aim of finding the most effective HF treatment to isolate the fine-grained enriched in quartz fraction and eliminate feldspar component, different etching treatments were tested (Prasad, 2000; Mauz and Lang, 2004).

The powder obtained was divided in different parts, the first was not treated while the others were treated with 20% HF at RT for time between 2 and 20 minutes. Sub-

Table 1. *Details for the samples studied in this work with the site of sampling, the ID number, the material and the sampling point.*

Site	ID Sample	Sample	Sampling point	
			Wall	Height* (cm)
Castelletto Cervo (Vercelli, Italy) (XI – XII centuries)	TL0_M	Mortar	Western (inside)	175
	TL0_B	Brick		
	TL5_M	Mortar	South (inside)	150
	TL5_B	Brick		
	TLOSL_B1	Brick	Bell tower, upper level (inside)	160
	TLOSL_M	Mortar		
	TLOSL_B2	Brick		
San Francesco alla Collina (Paternò, Italy) (X – XII centuries)	MPA7	Mortar	South-est (inside)	155
	BPA7	Brick		
	MPA8	Mortar	South-est (inside)	160
	BPA8	Brick		
Notre Dame de la Place (Bordeaux, France) (V – VII centuries)	NDLP_B1	Brick	South (inside)	150
	NDLP_M	Mortar		
	NDLP_B2	Brick		

**height respect to the planking level*

sequently, they were washed in 10% HCl for 25 minutes, to remove fluorides, and then the 4–11 µm fraction was selected and deposited onto 9.8 mm diameter discs.

The purity of fractions was the evaluated by the coefficient R that is the result of the ratio between postIR-OSL/T2 and OSL/T1 intensities. Where OSL/T1 represents the OSL signal normalized by T1 test dose and postIR-OSL/T2 the OSL emission obtained after IR stimulation normalized by T2 test dose.An R value close to unity indicates that the signal comes from a fraction free of feldspars (Mauz and Lang, 2004; Prasad, 2000; Shen *et al.*, 2007; Zhang and Zhou, 2007). The results obtained (**Table 2**) show, for all samples, that the most enriched quartz fraction is obtained with an HF etching at 20% for 15-20 minutes.

For brick samples a procedure to obtain polymineral fine grain phase with PH3DRA standard protocol was used (Guibert *et al.*, 2009). This procedure is divided in the different steps reported therein. The outer layers were removed by two millimetres, then the sample was crushed and sieved and the fraction below 40 µm was selected. Next, the following, etching procedures were performed: 10% HCl for 1h to remove the carbonate, 10% H_2O_2 for 48 hours to remove the organic component, 1% HF for 1 hour to remove clay and 10% HCl for 25 minutes to eliminate fluorosilicates that had possibly formed. Through a sedimentation procedure, polymineral fine grain fraction in the range of 4-11 µm was obtained and then deposited onto 9.8 mm diameter discs.

4. MORTARS DATING: OSL MEASUREMENTS

Preheating test

In dating of young samples as mortars, the OSL signal is normally measured following a relatively low (≤ 220°C) preheating treatment in order to limit the heating transfer of charge from deep energy levels to the OSL traps which influences the dating significantly. The thermal transfer can in fact cause overestimation of the equivalent dose and, therefore, an overestimation of age (Bailiff and Holland, 2000; Murray and Clemmensen, 2001; Ramzaev *et al.*, 2008; Rhodes, 2000; Wallinga *et al.*, 2001).

In this work, for five aliquots of each sample ED measurements as a function of preheating temperature (160-260°C) were made (Choi *et al.*, 2003; Thomas *et al.*, 2003; Kiyak and Canel, 2006). Each value of dose was determined using the modified SAR protocol (Murray and Wintle, 2003). The cycle of SAR protocol was repeated 7 times using increasing regeneration doses from 1.5 to 15 Gy. The integral signal of the first 2 s was used as OSL intensity, after subtraction of the background calculated from the last 3.5 s of the total stimulation time. The uncertainties were quantified on the basis of statistical counting of luminescence signals and applying error propagation.

Following the test dose, a cut heat of 160°C was applied. For minimizing the effects of charge remained in the light insensitive trap, at the end of each SAR cycle, an OSL measurement at 280°C for 40 s was carried out. On the same aliquots a recovery test in a preheating temperature range between 160°C and 240°C was done (Choi *et al.*, 2003; Kiyak and Canel, 2006). In such a test, a SAR measurement is applied to an aliquot which has been irradiated with a laboratory dose after its optical bleaching. In the absence of thermal transfer, it is expected that the ratio (R = measured / given dose) is close to unity; in the presence of thermal transfer, it takes values greater than 1. The recovery test values are considered acceptable

Table 2. *Mortar ID sample, % concentration of HF, time of etching and values of ratio (R) against time 20% HF etching determined for each sample.*

ID sample	Etching	Etching time (min)	*R*	ID sample	Etching	Etching time (min)	*R*
TL0_M	No HF	0	0.45±0.03	MPA7	No HF	0	0.48±0.03
	20% HF	2	0.56±0.03		20% HF	2	0.64±0.04
		5	0.78±0.05			5	0.75±0.05
		10	0.88±0.05			10	0.87±0.04
		15	0.99±0.03			15	1.01±0.03
		20	0.98±0.04			20	0.98±0.04
TL5_M	No HF	0	0.42±0.04	MPA8	No HF	0	0.40±0.03
	20% HF	2	0.58±0.03		20% HF	2	0.51±0.04
		5	0.72±0.04			5	0.79±0.04
		10	0.95±0.03			10	0.92±0.04
		15	1.00±0.04			15	0.99±0.03
		20	0.96±0.05			20	1.02±0.04
TLOSL_M	No HF	0	0.39±0.03	NDLP_M	No HF	0	0.56±0.03
	20% HF	2	0.50±0.03		20% HF	2	0.68±0.04
		5	0,62±0.03			5	0.87±0.03
		10	0.88±0.04			10	0.95±0.05
		15	0.98±0.03			15	1.03±0.06
		20	0.99±0.03			20	1.02±0.04

in the range between 0.90 and 1.10 (Murray and Wintle, 2003). The behaviour of ED and R (measured/given dose) values *vs.* preheating temperatures, obtained for each sample, are shown in **Fig. 1(a-c)** and **1(d-f),** respectively. The samples are grouped by site of collection.

For the samples TL0_M (**Fig. 1a**) and NDLP_M (**Fig. 1c**) the ED values are independent of preheating temperature in the range of 200-240°C while at higher temperatures the ED significantly increases. This can be attributed to thermal transfer effects. Therefore the preheat temperature chosen and used for ED determination was of 220°C for 10 s.

All the other samples show ED plateau values in the range 180-220°C (**Fig. 1a** and **Fig. 1b**). Therefore the preheating temperature value used for ED determination was of 200°C for 10 s. The results of recovery test (**Fig. 1(d-f)**) confirm that the rising trend in the preheat-plateau could be the result of thermal transfer during preheating of the samples prior to OSL measurements.

ED determination and partial bleaching test

For ED determination of each sample, the modified SAR protocol (Murray and Wintle, 2003), the preheating values obtained from previous tests and a cut heat of 160°C were used. The degree of bleaching is evaluable from histograms as shown in **Fig. 2** for each measured sample (Olley *et al.*, 1998; Murray and Olley, 2002; Goedicke, 2003; Saini and Mujtaba, 2010). The individual ED values were entered into bins of 0.1 Gy and so the frequency distributions were obtained.

The frequency distributions and related Gaussian fits (with standard deviations) show a well-bleached degree for each sample. However, these diagrams do not take into account the equivalent dose uncertainties; the maximum of the histogram may therefore not correspond with the "true" natural dose, unless the errors are more or less uniform (Goedicke, 2003). So, for age calculation, we used the radial plot technique that is discussed in the next section.

A further test to evaluate the bleaching time was performed for each sample. According to the paper of Agersnap Larsen *et al.* (2000), the effect of partial bleaching of the OSL signal was studied by exposing an irradiated aliquot to light using different illumination time, prior to the OSL measurements, using the measuring sequence detailed in **Table 3**.

The test consists of a laboratory irradiation and an optical resetting event using various illumination times Δt from 0 s to 100 s. The intent of the pre-heat phase (200°C for 10 s) is to empty the shallow traps in order to reproduce as closely as possible the natural situation. A blue diode stimulation at 160°C prevents the thermal charge release from these traps during the Continuous Wave OSL (CW-OSL) readout (Agersnap Larsen *et al.*, 2000).

Fig. 3a shows, as an example for the MPA8 mortar sample, that a time of 10 s is sufficient to bleach OSL emissions. The ratio between the partially bleached and the unbleached CW-OSL curves (**Fig. 3b**) describes the bleaching effects and gives direct information of the depletion degree for each illumination time. It is therefore expected that the grains will be well bleached and negligible charge should be remaining in the easy-to-bleach OSL traps at the time of laying with a similar solar exposition time.

Radial plot analysis

To determine the well-bleached population and the best representation of the true age from this distribution, the Central Age Model (CAM) of Galbraith *et al.* (1999) was explored (**Fig. 4**). In this model, the single true *log(ED)* are not equal, but are a random sample from a normal distribution with mean *ED* and standard deviation σ. It is graphically represented through the radial plot method, where, the position on the x-axis is a measure of the precision (t/se) with which ED is known. This axis is also expressed in terms of the relative error (se/t expressed as a percentage). Radial plots were obtained using the RadialPlotter software that can be downloaded free of charge from http://pvermees.andropov.org/radialplotter (Vermeesch, 2009).

Table 4 reports, for each mortar samples, the number of aliquots, the equivalent un-weighted mean doses with standard deviations obtained by Gaussian fit and as the Central value of Radial Plots .

5. BRICK DATING: THERMOLUMINESCENCE MEASUREMENTS

The ED values for bricks were determined using TL measurements and especially the additive dose method on the polymineral fine grain phase. For each sample, 24 aliquots were prepared and grouped into six. The first group was used for measuring the natural TL signal. The remaining groups were irradiated serially and incrementally using a calibrated beta source. TL glow curves were recorded by heating the aliquots up to 500°C at a constant

Table 3. *Procedure for partial bleaching test are resumed: annealing, irradiation and preheating cycle to obtain the laboratory dose; blue diode stimulation for partial bleaching; preheating and OSL readout to obtain final curves.*

Step	Treatment		
1	TL@500°C, 100 s	*Annealing*	*Laboratory dose*
2	Irradiation β	*Irradiation*	
3	Ph@200 °C, 10 s	*Preheat*	
4	Blue-diode@160°C Δt = (0; 0.2; 1; 10; 100) s	*Bleaching*	*Partial bleaching*
5	Ph@200°C, 10 s	*Preheating*	*OSL Curve*
6	OSL@125°C, 40 s	*OSL readout*	

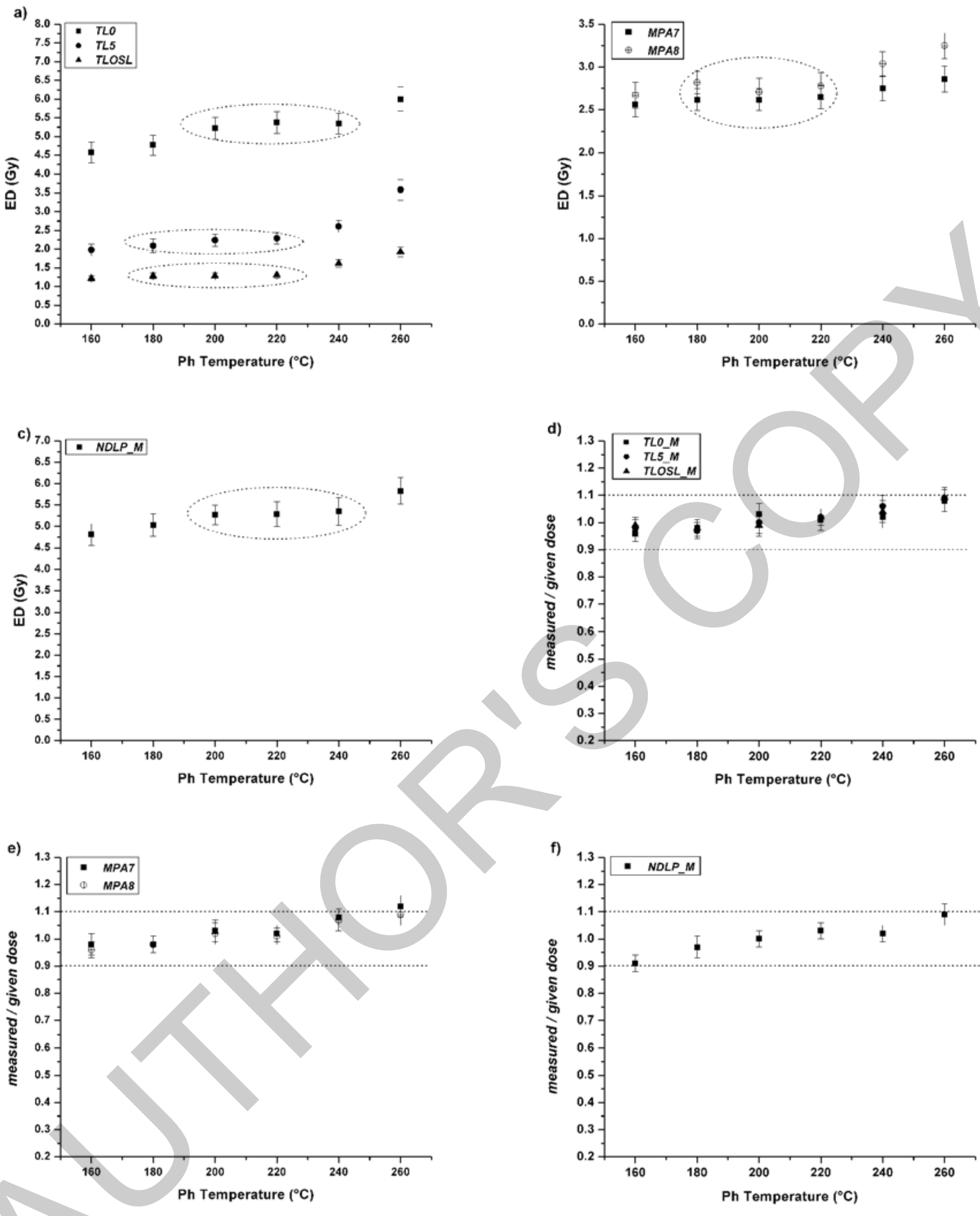

Fig. 1. *ED (a,b,c) and recovery test values (d,e,f) against preheating temperature for mortar samples.*

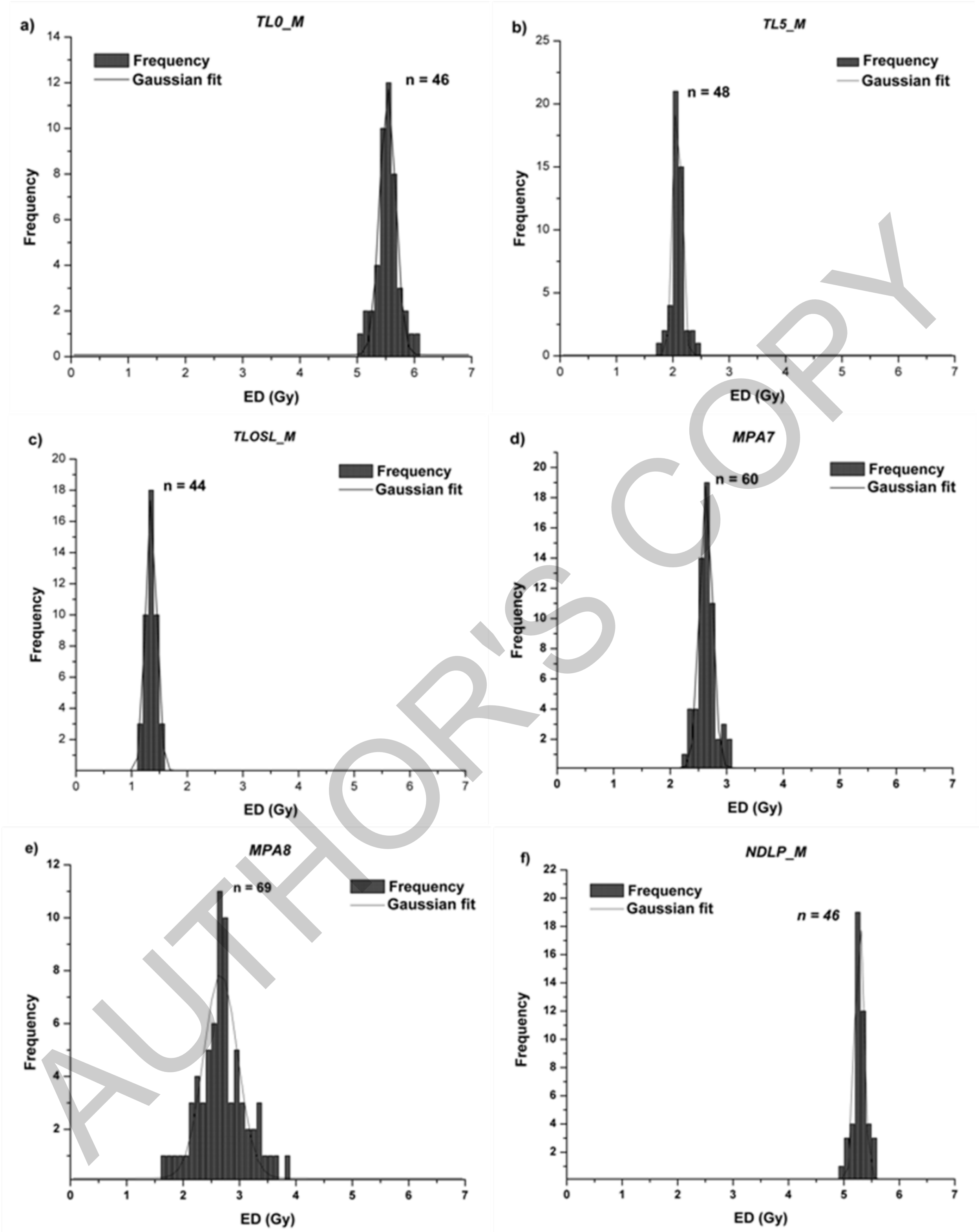

Fig. 2. *ED frequency distributions and Gaussian fit for mortar samples.*

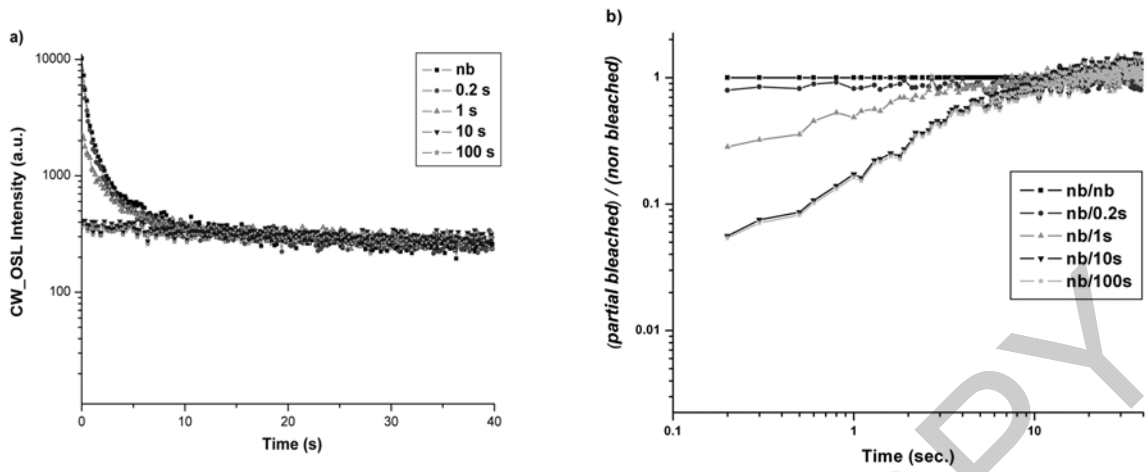

Fig. 3. *a) OSL curve for not bleached (nb) MPA8 sample and after 0.2, 1, 10 and 100 seconds of bleaching time with blue-diode@160°C; b) Ratio between the partially bleached and non-bleached (nb) CW-OSL curves.*

heating rate of 5°C/s in an ultrapure nitrogen atmosphere. The inter-aliquot intensity variations were corrected by second glow-curve normalisation. The temperature region between 300 and 350°C TL (280-390°C plateau) was used for ED calculation (Q_β). The same aliquots were then exposed to small and incrementing beta doses and a growth curve was constructed. The intercept of the curve on the dose axis is the correction value for supralinear growth (q_β) (Aitken, 1985). All samples showed a growth linear luminescent behaviour *vs.* dose. Just as an example, **Fig. 5** shows the straight growth lines of TLOSL_B2 sample from which the values of Q_β (a) and the correction q_β (b) were obtained.

For each sample measurements of natural loss of luminescence over time were performed (Aitken, 1985). The results obtained exclude the presence of anomalous fading.

6. DOSE RATE DETERMINATION

Annual dose components due to radioelements in the bricks and in the mortars samples were calculated using concentration values of U, Th, Rb and K (**Table 5**) determined by the ICP-MS technique using the conversion factors of Guérin *et al.* (2011).

The alpha contribution to the annual dose thus calculated were compared with integral thick source alpha-counting measurements (unit AL 03 system) (**Table 6**) in order to obtain indications about the radioactive equilibrium of the Uranium chain. The alpha counting in the pair coincidences counting mode can in fact be used to estimate Th contents and the alpha dose (Feathers *et al.*, 2008).

Table 4. *Number of aliquots, ED un-weighted mean by Gaussian fit, Central Value by Radial Plot and relative standard deviations obtained for mortar samples.*

Sample	Number of aliquots (*n*)	Mean ED (Gy)	SD	Central Value (Gy)	SD
TL0_M	*46*	5.53	0.20	5.53	0.06
TL5_M	*48*	2.08	0.12	2.08	0.03
TLOSL_M	*44*	1.35	0.10	1.34	0.03
MPA7	*60*	2.64	0.16	2.63	0.04
MPA8	*69*	2.69	0.43	2.66	0.10
NDLP_M	*46*	5.28	0.12	5.26	0.05

Table 5. *ID and type of samples, U, Th, Rb and K contents by ICP-MS technique.*

ID	Sample	U (ppm)	Th (ppm)	Rb (ppm)	K (%)
TL0_M	*Mortar*	4.40±0.21	13.30±0.85	30±1	4.08±0.01
TL0_B	*Brick*	5.30±0.25	17.00±1.09	105±5	1.50±0.01
TL5_M	*Mortar*	2.76±0.13	3.77±0.24	43±2	1.09±0.01
TL5_B	*Brick*	4.99±0.24	15.20±0.97	102±4	1.64±0.01
TLOSL_B1	*Brick*	5.26±0.25	17.00±1.09	105±5	1.50±0.01
TLOSL_M	*Mortar*	1.85±0.09	1.72±0.11	30±1	0.64±0.01
TLOSL_B2	*Brick*	5.91±0.28	17.70±1.13	102±4	1.63±0.01
MPA7	*Mortar*	3.70±0.06	1.80±0.03	34±1	0.72±0.01
BPA7	*Brick*	2.60±0.04	9.40±0.15	79±1	2.66±0.01
MPA8	*Mortar*	4.44±0.07	2.97±0.05	18±1	0.61±0.01
BPA8	*Brick*	3.30±0.05	10.80±0.17	85±1	2.93±0.01
NDLP_B1	*Brick*	2.60±0.04	10.40±0.28	130±2	1.54±0.02
NDLP_M	*Mortar*	5.70±0.09	4.80±0.13	57±1	1.47±0.01
NDLP_B2	*Brick*	2.60±0.04	9.96±0.26	129±2	1.52±0.02

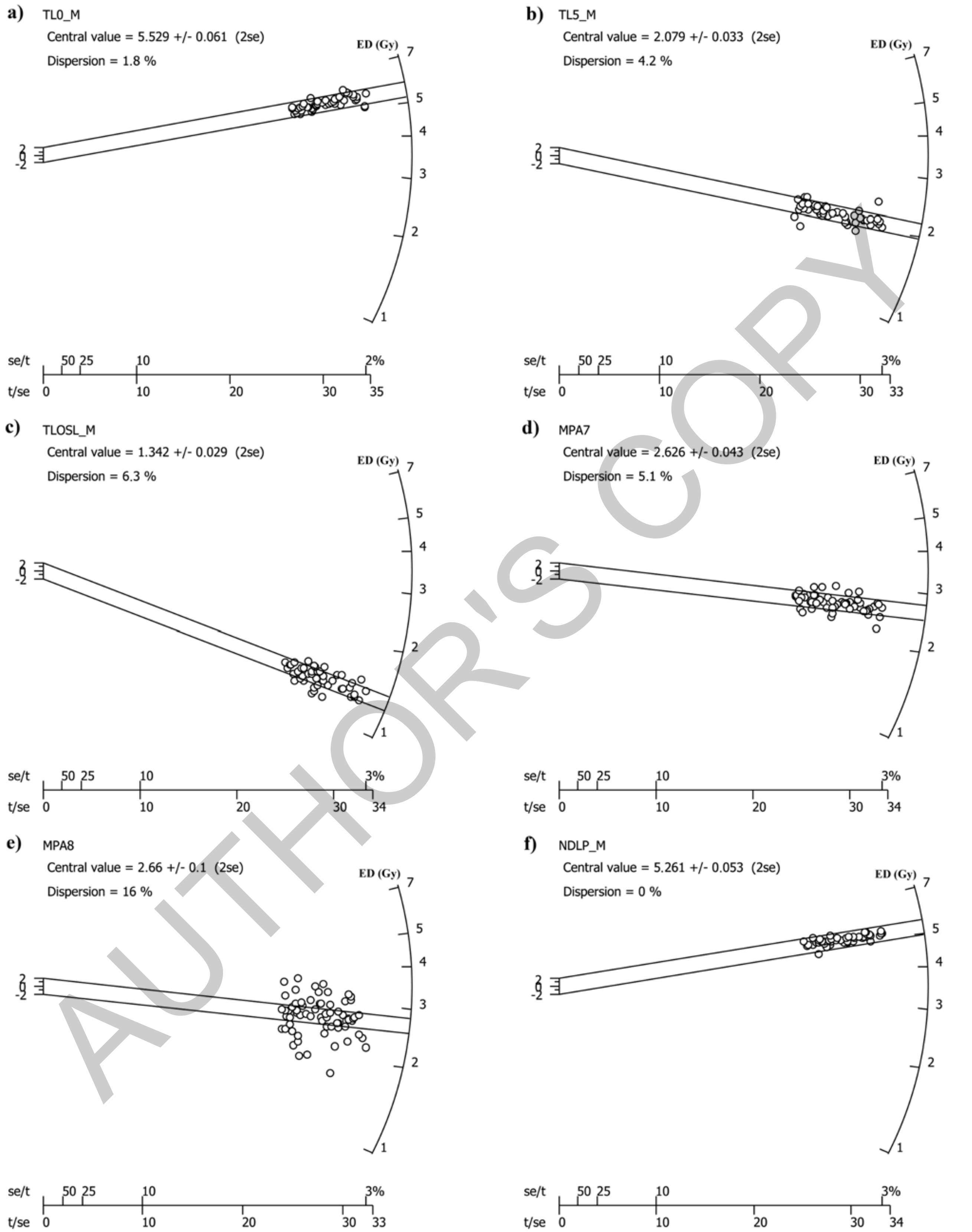

Fig. 4. *ED distribution obtained with Radial plots (CAM) for mortar samples.*

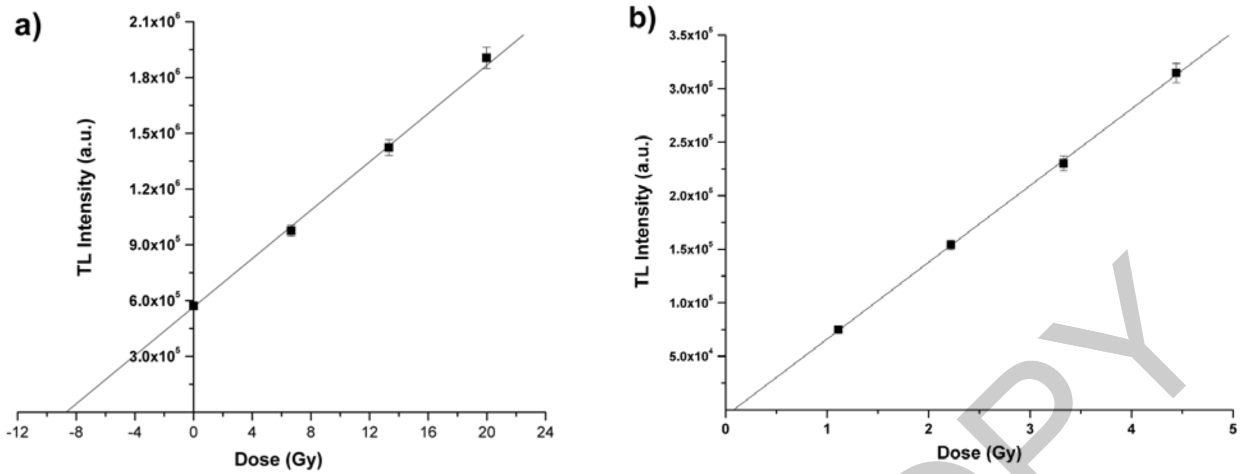

Fig. 5. *Growth curves for Q_β (a) and q_β (b) calculation for TLOSL_B2 sample.*

Table 6. *Internal Alpha dose rate: comparison between ones calculated by ICP-MS and ones by alpha counting measurements. Percentage difference between the values obtained with the two techniques (Δ%).*

ID	Sample	Alpha dose rate (mGy/a)		
		From ICPMass content	Alpha counting	Δ%
TL0_M	*Mortar*	22.11±0.86	21.58±0.91	2.40
TL0_B	*Brick*	27.35±1.07	26.86±0.17	1.79
TL5_M	*Mortar*	10.49±0.41	10.60±0.44	1.05
TL5_B	*Brick*	25.16±0.97	24.87±1.00	1.15
TLOSL_B1	*Brick*	27.24±1.06	26.83±1.09	1.51
TLOSL_M	*Mortar*	6.44±0.26	6.51±0.27	1.09
TLOSL_B2	*Brick*	29.57±1.14	29.85±1.32	0.95
MPA7	*Mortar*	11.67±0.16	11.90±0.52	1.97
BPA7	*Brick*	14.20±0.16	13.96±0.61	1.69
MPA8	*Mortar*	14.60±0.20	14.76±0.66	1.10
BPA8	*Brick*	17.19±0.19	16.95±0.69	1.40
NDLP_B1	*Brick*	14.94±0.23	14.81±0.65	0.87
NDLP_M	*Mortar*	19.47±0.27	19.02±0.79	2.31
NDLP_B2	*Brick*	14.61±0.22	14.91±0.70	2.05

Once confirmed the U chain radioactive equilibrium, the alpha dose rate values obtained from U and Th contents (ICP-MS) were used and the internal beta dose from U, Th, K and Rb was determined.

The dose contributions were corrected on the basis of the experimentally measured porosity factor (W) (**Table 7**) (Aitken, 1985) and saturation factor (F) estimated on the basis of sampling point (height, inside or outside, etc. ...) and humidity measurements of the samples to the excavation. In this particular case an F value of 0.3±0.2 for all sites was used, except for the samples collected from *Notre Dame de La Place* for which the value of 0.5±0.2 was considered.

The annual environmental dose rate was measured using TL dosimeters (GR200A) enclosed in capsules placed in situ at the sampling points (Gueli *et al.*, 2009). Around each sampling point in situ γ dose measurements (Canberra InSpector 1000 spectrometer) were made. The dose values obtained, all within a discrepancy of 3% compared to the measured value in the sampling point, show a homogeneous gamma dose distribution in the environment constituted by regular sequences of bricks and mortars. Cosmic radiation contribution is calculated according to global considerations by Prescott and Hutton (1988).

Table 7 shows the internal dose rates ($D_{\alpha int}$ and $D_{\beta int}$), for each sample, corrected for water content together with the annual environmental (gamma + cosmic) dose (D_{env}).

7. DISCUSSION

The tests made for mortars (ED and recovery test values against preheating temperature) show thermal transfer effects for preheating temperature above 220-240°C. As previously mentioned, these tests were essential to choose the true preheating temperature to avoid overestimation of the equivalent dose. Furthermore, the recovery test values close to 1, for the preheating temperature used for ED determination, validate the choice of the other experimental parameters, such as test dose and cut heat temperature.

The histograms of ED frequency distribution and the radial plot show a Gaussian behaviour of the dose values indicating that the samples are well-bleached. This is confirmed by partial bleaching test, in fact, the grains turned out well-bleached for stimulation time shorter than 10 seconds, a time comparable to sun exposure during the mixing and layer of the mortar on the structure.

For the evaluation of the annual dose was necessary to determine the alpha dose contribution starting from the contents of ICP-MS with conversion factors in dose and

Table 7. *ID and type of samples, k value, internal dose rate corrected with humidity and porosity factors (W, F) and environmental dose rates.*

ID	Sample	k	W	$D_{\alpha int\_corrected}$ (mGy/a)	$D_{\beta int\_corrected}$ (mGy/a)	D_{env} (mGy/a)	Dose rate (mGy/a)
TL0_M	*Mortar*	0.06±0.01	0.20±0.01	20.34±3.09	3.99±0.48	1.67±0.05	6.96±0.56
TL0_B	*Brick*	0.23±0.01	0.12±0.01	25.92±1.59	2.37±0.10		10.01±0.46
TL5_M	*Mortar*	0.07±0.01	0.30±0.01	9.26±1.46	1.12±0.14	1.73±0.05	3.45±0.20
TL5_B	*Brick*	0.13±0.01	0.13±0.01	23.76±1.46	2.38±0.10		7.13±0.32
TLOSL_B1	*Brick*	0.23±0.02	0.12±0.01	25.82±1.59	2.37±0.10	1.47±0.05	9.83±0.60
TLOSL_M	*Mortar*	0.06±0.01	0.21±0.01	5.92±0.90	0.78±0.09		2.67±0.13
TLOSL_B2	*Brick*	0.19±0.01	0.13±0.01	27.96±1.72	2.57±0.11		9.38±0.49
MPA7	*Mortar*	0.07±0.01	0.19±0.01	10.92±1.58	1.11±0.13	0.68±0.02	2.58±0.21
BPA7	*Brick*	0.10±0.01	0.15±0.01	13.06±0.66	2.61±0.11		4.59±0.18
MPA8	*Mortar*	0.06±0.01	0.12±0.01	13.93±1.98	1.17±0.14	0.70±0.02	2.64±0.18
BPA8	*Brick*	0.09±0.01	0.11±0.01	16.35±0.79	3.02±0.12		5.19±0.22
NDLP_B1	*Brick*	0.30±0.01	0.26±0.01	12.47±1.14	1.67±0.12	0.91±0.04	6.32±0.39
NDLP_M	*Mortar*	0.05±0.01	0.07±0.01	18.54±4.39	2.07±0.40		3.91±0.50
NDLP_B2	*Brick*	0.17±0.01	0.12±0.01	13.40±1.12	1.78±0.12		4.97±0.27

alpha counting measurements. The comparison between the results obtained, all within a discrepancy of ~2%, confirms the absence of problems related to radioactive disequilibrium. With regard to the environmental contribution to the annual dose, mappings with in situ γ counter show a uniform degree of dose certainly due to the regular brick-mortar construction.

The dose values thus obtained, for mortar and bricks, were corrected with the factors related to the porosity (W) and the saturation (F). The porosity was experimentally obtained whereas the saturation was estimated. This last evaluation, in agreement with the other groups involved in the dating project considers both the geographical position of the structures and the height of sampling points.

Table 8 shows, for each sample, the technique and the method used for ED determination, the number of aliquots and the ages obtained.

The ages obtained for each mortar sample are in good agreement with the associated brick and with the chronological results obtained for the studied structures, except in the case of the set of samples TLOSL# taken from the monastery of *Castelletto Cervo*. In this case, the mortar results to be younger than bricks (coeval between their), and this suggests that it has been inserted in the structure during a restoration phase. This is confirmed both by the elemental composition of bricks similar to that of the other coeval bricks and by the different composition of the mortar samples than the other taken in the structure (TL0_M

Table 8. *Details and age results for each sample.*

Site	ID	Sample	Technique	Method	n	*Age 2se (a)
Castelletto Cervo (Vercelli, Italy)	TL0_M	Mortar	OSL	SAR	46	822±55
	TL0_B	Brick	TL	Added dose	48	794±65
	TL5_M	Mortar	OSL	SAR	48	602±36
	TL5_B	Brick	TL	Added dose	48	650±40
	TLOSL_B1	Brick	TL	Added dose	48	885±68
	TLOSL_M	Mortar	OSL	SAR	44	502±26
	TLOSL_B2	Brick	TL	Added dose	48	931±65
San Francesco alla Collina (Paternò, Italy)	MPA7	Mortar	OSL	SAR	60	982±63
	BPA7	Brick	TL	Added dose	48	1018±83
	MPA8	Mortar	OSL	SAR	69	1008±79
	BPA8	Brick	TL	Added dose	48	961±54
Notre Dame de La Place (Bordeaux, France)	NDLP_B1	Brick	TL	Added dose	48	1365±116
	NDLP_M	Mortar	OSL	SAR	46	1346±172
	NDLP_B2	Brick	TL	Added dose	48	1348±91

**Age for mortars obtained applying Central Age Model (CAM)* and TL5_M) and coeval to the bricks. Considering the sampling points, in fact, from historical sources (Destefanis, 2009) it is known that the bell tower is similar to the same phase of the first architectural plant, whose chronology is anchored to the end of the 1th century, or, at the latest, by the first decades of the 12th century.

The data obtained for the two bricks, even if in good agreement with the historical reference data, are certainly affected by the presence of the mortar used for the restoration. In these cases the presence of new elements introduces a further error source in determining the age due to environmental dose rate changes that receives the analyzed sample. In the particular case, most likely this effect is minimized by thin thickness of mortar and from a homogeneous gamma dose distribution in the environment.

8. CONCLUSIONS

The results obtained confirm the possibility of dating the lime mortar layer from historical buildings, using the OSL emission of its fine-grained fraction enriched in quartz. The cross-dating based on bricks and associated mortar samples age, if related to other data, is furthermore a way to solve chronological issues regarding historical buildings and represents a valuable improvement with respect to using the only thermoluminescence dating on bricks.

From the methodological point of view, the possibility to use polymineral fine grain phases enriched in quartz from mortars, trough etching procedure during the preparation phase must be coupled with a careful examination of several tests (e.g. preheating and recovery tests, radioactive equilibrium). It is in fact necessary to study the effects of thermal transfer that, if not considered, may lead to an overestimation of the equivalent dose. This is achieved thanks to the luminescent linear behaviour *vs.* dose of samples that allows an accurate analysis of the phenomenon. Additionally, the luminescence measurements performed on fine grain fraction, partially solve the problem of the low intensity of the luminescent signals, arising from single grains, that makes difficult the analysis.

Thus, the condition of well-bleached population identified by ED frequency Gaussian distributions and therefore the appropriate Age Model used (CAM), allowed us to obtain OSL ages for mortars, validated also from good agreement with those of associated bricks obtained through routine thermoluminescence measurements.

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