



Quartz luminescence response to a mixed alpha-beta field: Investigations on Romanian loess

Constantin, Daniela; Jain, Mayank; Murray, Andrew S.; Buylaert, Jan-Pieter; Timar-Gabor, Alida

Published in:
Radiation Measurements

Link to article, DOI:
[10.1016/j.radmeas.2015.01.001](https://doi.org/10.1016/j.radmeas.2015.01.001)

Publication date:
2015

Document Version
Peer reviewed version

[Link back to DTU Orbit](#)

Citation (APA):
Constantin, D., Jain, M., Murray, A. S., Buylaert, J-P., & Timar-Gabor, A. (2015). Quartz luminescence response to a mixed alpha-beta field: Investigations on Romanian loess. *Radiation Measurements*, 81, 110–115. <https://doi.org/10.1016/j.radmeas.2015.01.001>

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Quartz luminescence response to a mixed alpha-beta field: investigations on Romanian loess

D. Constantin^{1,2,3}, M. Jain⁴, A.S. Murray⁵, J.P. Buylaert^{4,5}, A. Timar-Gabor^{1,2*}

¹ Faculty of Environmental Sciences and Engineering, Babeş-Bolyai University, Cluj-Napoca, Romania

² Interdisciplinary Research Institute on Bio-Nano-Science, Babeş-Bolyai University, Cluj-Napoca, Romania

³ Faculty of Biology and Geology, Babeş-Bolyai University, Cluj-Napoca, Romania

⁴ Center for Nuclear Technologies, Technical University of Denmark, DTU Risø campus, Roskilde, Denmark

⁵ Nordic Laboratory for Luminescence Dating, Department of Geoscience, Aarhus University, Risø campus, Roskilde, Denmark

* Corresponding author: alida.timar@ubbcluj.ro

Abstract

Previous SAR-OSL dating studies using quartz extracted from Romanian and Serbian loess samples report SAR-OSL dose-response curves on fine grained (4-11 μm) quartz that grow to much higher doses compared to those of coarse-grained (63-90, 90-125, 125-180 μm) quartz. Furthermore, quartz SAR-OSL laboratory dose response curves do not reflect the growth of the OSL signal in nature. A main difference in coarse- and fine-grained quartz dating lies in the alpha irradiation history, but the effect of mixed alpha-beta fields has so far received little attention. In the present study we investigate whether the alpha dose experienced by fine grains over geological cycles of irradiation and bleaching may have an effect on the saturation characteristics of the laboratory dose response. By applying time resolved optically stimulated luminescence we confirm that the OSL signals induced in quartz by alpha and beta radiation follow the same recombination path. We also show that a mixed alpha-beta dose response reproduces the beta dose response only up to about 800 Gy. Assuming an a-value of 0.04 we have shown that laboratory alpha and beta dose response curves overlap up to effective alpha doses of ~ 50 Gy. Based on these results, we conclude that exposure of fine grains to alpha radiation during burial and transport cycles prior to deposition, as well exposure to the mixed radiation field experienced during burial are not responsible for the age discrepancies previously reported on fine and coarse grained quartz extracted from Romanian and Serbian loess.

Keywords: alpha radiation, beta radiation, quartz, OSL

1. Introduction

Heavy charged particles such as alpha particles (helium ions) deposit a large amount of energy per unit track length when interacting with matter and cause dense ionization and, in the case of luminescence dosimeters, local charge saturation effects in the vicinity of their tracks. Because of these saturation effects, the OSL output

1 1 per unit dose from alpha particles is lower than that from high-energy electrons (beta irradiation) or photons
2 2 (gamma irradiation or X rays). This difference in OSL response is quantified by the alpha efficiency parameter
3 3 relative to beta irradiation (a-value). The lower luminescence response to alpha irradiation with respect to that
4 4 of beta and gamma radiation in quartz, both in the case of TL and OSL has been known for a long time
5 5 (**Zimmerman, 1972; Aitken and Bowman, 1975**). This effect becomes especially important in ‘fine-grain’
6 6 dating, because external alpha particles can irradiate the entire volume of the grain. Although, there has been an
7 7 increasing focus on dating fine grained quartz in the last decade (e.g. **Lowick et al. (2010); Sugisaki et al.**
8 8 **(2010); Timar et al. (2010)**) using the single aliquot regenerative dose protocol (**Murray and Wintle, 2000**),
9 9 the subject of alpha efficiency has received little attention (**Tribolo et al., 2001; Galloway, 2002; Burbidge et**
10 10 **al., 2009; Polymeris et al., 2011**). Reported estimates of alpha efficiency for fine grained quartz extracts is
11 11 known to be around 0.1 in the case of thermoluminescence (**Zimmerman (1972); Aitken (1985 appendix K)**),
12 12 while in the case of OSL reported values vary between 0.03 and 0.04 (**Rees-Jones, 1995; Mauz et al., 2006;**
13 13 **Lai et al., 2008**).

14 14 The range of alpha particles is ~20 µm in quartz (**Aitken, 1985**). Thus, they deposit energy in the entire volume
15 15 of fine (4-11 µm) grains, while only in an outer shell in coarse (63-90 µm) grains. In the latter case the alpha-
16 16 irradiated shell is etched away before measurement. The question we ask here is whether this difference in alpha
17 17 affected volume can explain the differences in the OSL behaviour of the two size fractions from the loess in the
18 18 Lower- as well as the Carpathian-Danube basin (**Timar-Gabor et al., 2011; Timar-Gabor et al., 2012;**
19 19 **Timar-Gabor and Wintle, 2013; Constantin et al., 2014a**). These differences in OSL behaviour are observed
20 20 in two ways: 1) on the one hand as observed in the Romanian loess, the laboratory dose response curves of fine
21 21 quartz (D_0 of ~175 Gy and ~1800 Gy) saturate much later than the etched coarse quartz (D_0 of ~55 Gy and ~600
22 22 Gy), (see **Timar-Gabor et al. (2012)**); this behaviour has also been observed in other sites (e.g. **Kreutzer et al.**
23 23 **(2012); Lomax et al., 2014; Timar-Gabor et al., 2014**). 2) there is a general disagreement between fine and
24 24 coarse quartz ages (**Constantin et al., 2014a,b; Timar-Gabor et al., 2011; Timar-Gabor et al., 2012**) which is
25 25 likely to be due to the difference between the naturally and laboratory generated dose response curves (**Timar-**
26 26 **Gabor et al., 2011; Timar-Gabor et al., 2012; Timar-Gabor and Wintle, 2013; Constantin et al., 2014a;**
27 27 **Constantin et al., 2014b**). The divergence observed between the natural and the laboratory dose responses is
28 28 observed for both grain fractions but is more pronounced in the case of fine grains (see **Timar-Gabor and**
29 29 **Wintle (2013), fig 6 and fig S1 a**).

30 30
31 31
32 32 Assuming that the quartz has the same origin and physical characteristics irrespective of grain size, an important
33 33 dosimetric difference between natural and laboratory irradiations lies in the dose rate and radiation field in the
34 34 sediment matrix. The dose rate is ~11 orders of magnitude lower in nature compared to that in the laboratory.

1 1 Similarly natural irradiations occur in a mixed radiation field (alpha, beta, gamma, x-rays) while laboratory
2
3 irradiations usually consist entirely of either beta particles or photons (gamma or X rays). These differences
4
5 may be reflected in calibration of luminescence response to dose and may arise from the different interaction
6
7 mechanisms of alphas and betas, and the possibility of additional defect creation due to alphas.

8
9 In the present study we compare the dose response curves and sensitivity changes measured using continuous
10
11 wave OSL (CW-OSL), under different combinations of alpha and beta irradiations designed to simulate the
12
13 effect of a natural radiation field. We use time-resolved optically stimulated luminescence (TR-OSL) as a tool
14
15 to gain insights into recombination pathways leading to the emissions generated by the two different types of
16
17 radiations. These results are reported below.

18

19 2. Samples and measurement facilities

20
21 Analyses have been performed on 4-11 μm and 63-90 μm quartz grains of sample CST 3 (**Timar-Gabor and**
22
23 **Wintle, 2013; Constantin et al., 2014a**). Additional measurements have been carried out on 4-11 μm quartz
24
25 from sample LCA 26 (**Constantin et al., 2014b**). Relevant information on the samples is presented in **Table**
26
27 **S1**. All investigations have been performed using large (9-mm diameter) aliquots that have been tested for
28
29 purity by OSL IR depletion ratio (**Duller, 2003; Wintle and Murray, 2006**). The natural OSL signal was
30
31 bleached by exposure to blue LEDs for 200 s at 20 °C prior to experiencing two cycles of regenerative beta
32
33 dose of 350 Gy (preheat at 220 °C for 10 s), test dose of 17 Gy (cutheat 180° C) and elevated temperature
34
35 bleach (280 °C for 40 s). In the second cycle, an IR diode exposure for 100 s at 125 °C preceded reading of the
36
37 OSL signal induced by the regenerative dose.

38 Experiments have been carried out at DTU Nutech (DTU Risø Campus, Denmark) on a standard TL/OSL-DA-
39
40 20 reader (**Thomsen et al., 2006**) using an in-built $^{90}\text{Sr}/^{90}\text{Y}$ beta source (dose rate of 0.172 Gy s⁻¹ to 4-11 μm
41
42 grains deposited on aluminium disks and 0.219 Gy s⁻¹ to 180-225 μm quartz grains mounted on stainless steel
43
44 disks) and an uncalibrated ^{241}Am source under vacuum conditions. The OSL signal induced by 5000 s of alpha
45
46 irradiation was found to be 2.22 times higher than that resulting from ~8.5 Gy of beta irradiation, giving an
47
48 effective alpha dose rate of 0.095 Gy/s. This can be converted to a tentative absolute alpha dose rate of 2.370
49
50 Gy/s assuming an *a*-value of 0.04 for fine grained quartz (**Rees-Jones, 1995**).

51 All investigations into the optical emission characteristics of fine (4-11 μm) and coarse (63-90 μm) quartz used
52
53 the SAR protocol (**Murray and Wintle, 2003**). The luminescence signals induced by alpha, beta and mixed
54
55 alpha-beta radiation were stimulated with blue LEDs in continuous-wave (CW) mode for 40 s at 125 °C and the
56
57 net CW-OSL signal was determined from the initial 0.32 s of the decay curve, less a background integrated
58
59 between 1.76 s and 2.40 s (**Ballarini et al., 2007; Cunningham and Wallinga, 2010**). For the sake of
60
61 consistency with previously reported data (**Timar-Gabor et al., 2011; Timar-Gabor et al., 2012; Timar-**

1 **Gabor and Wintle, 2013**) a preheat at 220 °C for 10 s and a cutheat to 180 °C was used, together with
2
3 stimulation at 280 °C for 40 s at the end of each SAR cycle. Sensitivity changes induced by alpha and beta
4
5 irradiation were monitored using the response to a constant 17 Gy beta test dose.

6
7 Time-resolved optically stimulated luminescence (TR-OSL) was measured on a Risø TL/OSL-DA-20 reader
8
9 equipped with an integrated pulsing option to control the stimulation LEDs and a photon timer attachment with
10
11 a detection resolution (bin-width) of 100 ps (**Lapp et al., 2009**). TR-OSL experiments have been carried out
12
13 using the same parameters of the SAR protocol as in the CW-OSL measurements. The luminescence signals
14
15 were stimulated and recorded at 125 °C for 100 s using pulsed optical stimulation (POSL). Each pulse consisted
16
17 of an *on*-time (the duration of each LED stimulation pulse) of 50 µs and an *off*-time time (the following period
18
19 when the LEDs are not illuminated) of 450 µs. The photon counter was set to count photons during both *on* and
20
21 *off*-time. The dose rates for beta irradiation on this particular reader were 0.099 Gy s⁻¹ to 4-11 µm quartz and
22
23 0.113 Gy s⁻¹ to 180-225 µm quartz.

24 25 **3. Results and discussions**

26 27 **3.1. The effect of alpha irradiation history on the beta dose-response curve**

28
29 One major difference when applying OSL dating to the fine and coarse quartz extracts is the fact that for the
30
31 coarse fraction the outer alpha irradiated rim is etched away using HF. Thus, in the case of coarse quartz the
32
33 material used for dating has not experienced alpha irradiation. Due to the different interaction mechanisms of
34
35 alpha and beta particles with matter, the multiple cycles of irradiation and light exposure in the geological past
36
37 of the fine grains and the fact that it is not proven without doubt whether alpha irradiation could cause defects
38
39 involved in luminescence production we have aimed to investigate whether the higher saturation characteristics
40
41 usually reported for fine grains compared to coarse grains (**see e.g. Kreutzer et al. (2012); Timar-Gabor et al.**
42
43 **(2012); Lomax et al. (2014)**) could be at least partially, a consequence of past alpha history of the 4-11 µm
44
45 fraction. Three 63-90 µm quartz aliquots (sample CST 3) used in this experiment were first tested for IR
46
47 depletion, followed by measurement of three consecutive SAR beta dose response curves up to 2.8 kGy to
48
49 check the reproducibility. Subsequently a total effective alpha dose of 9552 Gy (total alpha dose of ~240 kGy)
50
51 was delivered to the sample in 50 pulses of 182 Gy, each pulse being followed by a bleach (exposure to blue
52
53 LEDs for 200 s at 20 °C) in order to simulate natural irradiation and transport cycles as closely as possible.
54
55 After these alpha irradiations, the SAR beta dose response was constructed, and compared to the first three
56
57 growth curves. Finally, a fifth beta dose response was constructed to check the reproducibility. These five
58
59 laboratory dose response curves are presented in **figure 1**. There is no significant variation between the pre- and
60
61 the post-α irradiation dose response curves, and the saturation values for these curves agree with previously
62
63 reported values for coarse quartz extracted from Romanian loess (**Timar-Gabor et al., 2012**). Assuming that
64
65

1 1 OSL originates from the entire grain volume, the alpha skin contribution is expected to be less than 20% to the
2 2 total OSL (assuming a spherical grain of about 80 μm diameter). But in practice the surface plays a far greater
3 3 role than the entire volume because of self-absorption of the photons emitted from the grains' core. Based on
4 4 the results presented in **figure 1** we therefore consider it unlikely that that prior alpha exposure plays a
5 5 significant role in affecting the shape of the subsequently measured beta dose response curves (see **figure S1 a**).

6 10 7 11 8 12 **3.2. Luminescence lifetimes in quartz following alpha and beta irradiation**

9 13 **Galloway (2002)** suggested that the trapped electrons produced by alpha and beta irradiations use the same
10 14 recombination pathways upon excitation with light. We have tested this by performing a time resolved
11 15 experiment. One aliquot of CST 3 was first tested for purity using an IR depletion test. This aliquot was then
12 16 bleached with blue LEDs for 200 s at 20 $^{\circ}\text{C}$ and irradiated for 120 ks using the alpha source under vacuum
13 17 conditions; this irradiation generates a luminescence response corresponding to that produced by a beta dose of
14 18 455 Gy. Assuming an a value of 0.04, the expected total alpha dose is ~ 11.4 kGy. The measurement conditions
15 19 for the TR-OSL were chosen to be similar to those employed in CW-OSL: blue LEDs stimulation at 125 $^{\circ}\text{C}$ and
16 20 a 220 $^{\circ}\text{C}$ preheat for 10 s, beta test dose of 19 Gy, ramp heating to 180 $^{\circ}\text{C}$ (cutheat), elevated temperature OSL.
17 21 The same measurement sequence was repeated using a beta regenerative dose of 581 Gy. The TR-OSL spectra
18 22 recorded are presented in **figure 2**. The shapes of the photon-arrival time distributions are typical of quartz
19 23 (**Chithambo, 2003; Denby et al., 2006; Thomsen et al., 2008**). The 54-500 μs interval from the off time was
20 24 fitted with an exponential function plus a constant (**Figure 2 inset**). Lifetimes of 35.3 ± 0.2 μs and 35.6 ± 0.2 μs
21 25 were calculated for the alpha and beta induced OSL signals ($R^2=0.99$). These results are also typical of quartz
22 26 (**Chithambo, 2003; Ankjærgaard et al., 2010**), and compare well with the previous findings of **Galloway**
23 27 (**2002**) on the TR-OSL spectra of synthetic quartz.

24 41 25 42 **3.3. Mixed alpha beta dose response curves**

26 43 **3.3.1. Monitoring sensitivity changes following alpha irradiation using a beta test dose**

27 44 The SAR protocol is based on the assumption that sensitivity changes can be corrected by the response to a test
28 45 dose. While laboratory irradiations are usually carried out using only beta or gamma irradiation, the following
29 46 experiments are intended to examine the behaviour of our samples in mixed alpha/beta fields. **Biswas et al.**
30 47 (**2013**) showed that a fixed β test dose provides reliable normalization of the α and β induced OSL signal
31 48 growth in polymineral fine-grains extracted from volcanic ash samples, but **Mauz et al. (2006)** reported that for
32 49 their fine-grained quartz the β test dose did not provide a reliable correction for the sensitivity changes induced
33 50 by α dosing. Because of this inconsistency, we have tested the basic assumption of the SAR protocol (**Wintle**
34 51 **and Murray, 2006**) by examining the linearity between the regenerated (alpha dose) and the test (beta dose)

1 response. The constant alpha regeneration dose amounted to an effective dose of 100 Gy, while beta test dose
2 was set to 17 Gy.

3 **Figure 3** shows the results for one aliquot of CST 3 and one aliquot of LCA 26. It can be seen that the
4 relationship is linear ($R^2=0.98$ and 0.94 , respectively) and the fitted functions pass close to the origin, indicating
5 that a beta test dose can satisfactorily monitor the factor of ~ 2 sensitivity changes induced by alpha irradiation.

6 **3.3.2. Mixed alpha-beta dose response curves**

7 As the deviation between the laboratory and natural dose responses was reported to be more pronounced for the
8 4-11 μm quartz (see **figure S1 b**), we now investigate whether α and β laboratory irradiations designed to
9 reproduce as closely as possible the natural irradiation field produce a different growth curve compared to only
10 beta irradiations.

11 A mixed alpha-beta SAR dose response (normalized to a 17 Gy beta test dose) was build up to 800 Gy on one
12 aliquot of fine quartz from sample CST 3, which was prior checked by an IR depletion test (**Figure 4**). The
13 regenerative alpha doses represent effective alpha doses and were calculated based on the luminescence output
14 following 5 ks of alpha irradiation and 50 s of beta irradiation and assuming an a-value of 0.04. Firstly a pure
15 beta growth curve (dashed line) was constructed. For the combined $\alpha+\beta$ curve (solid line), the same
16 regenerative dose levels were used. Each dose given comprised a 12 % alpha share and an 88 % beta share. In
17 order to simulate as close as possible the natural conditions the alpha and beta doses were delivered
18 alternatively, in alpha-beta pulses of an effective magnitude equivalent to 51 Gy of beta dose: 260 s beta (45
19 Gy) and 1523 s alpha (6 Gy of effective alpha dose, ~ 150 Gy of total alpha dose). A third beta dose response
20 was constructed in order to check the reproducibility (dotted line). Finally, on the same aliquot a pure alpha
21 growth curve was constructed using the alpha doses given in the mixed dose points and a test dose of 17 Gy of
22 beta (**Figure 4 inset**).

23 The mixed alpha-beta dose response curve and the beta dose-response growth curves are well fitted with a sum
24 of two exponential functions; within the dose range investigated here they are indistinguishable (**Figure 4**). Up
25 to 90 Gy (effective alpha dose) the alpha dose response is almost linear (inset **Figure 4**). The results indicate
26 that over the investigated dose range the use of a constant a-value is justified and that the differences between
27 the natural and laboratory generated dose response curves for fine grains reported by **Timar-Gabor and Wintle**
28 (**2013**) could not be accounted by an incorrect use for the value of the alpha efficiency.

29 **3.3.3. Extending the dose range of α -generated OSL dose response curves**

30 Little work has been undertaken to determine the relative alpha to beta dose response (a-value) in quartz. Values
31 such as 0.04 **Rees-Jones (1995)** or 0.03 (**Mauz et al., 2006**) are commonly adopted in quartz OSL dating

1 1 studies. In the previous section we have shown that this is an appropriate decision in the case of our samples for
2 2 moderate doses. However, the contribution of the alpha dose to the total deposited energy in the quartz crystal
3 3 increases at higher doses, where less luminescence is created per unit beta dose. Saturation in the natural mixed
4 4 field implies a superposition of alpha and beta tracks. The interpretation of classic a -values at high dose is
5 5 complicated because the alpha and beta irradiations are performed separately. This may be relevant because
6 6 **Timar-Gabor et al. (2012)** have reported natural OSL signals from an infinitely old quartz sample well below
7 7 the saturation range of the laboratory beta dose response curve.

8 8 Pure alpha growth curves were constructed on three aliquots of fine-grained quartz (2 aliquots of CST 3 and 1
9 9 aliquot of LCA 26) normalized to 17 Gy of beta irradiation. Dose points up to 1.6 kGy of effective alpha (~ 40
10 10 kGy of total alpha) were administered. For the sake of comparison, pure beta dose-response curves were
11 11 constructed on each aliquot prior to alpha growth curve measurements, with beta dose points chosen to equal in
12 12 light levels the given alpha doses. The growth of both alpha- and beta-induced OSL signal was fitted with a sum
13 13 of two saturating exponentials. **Figure 5** shows the shapes of the alpha and beta dose-response growth curves.
14 14 The inset in **figure 5** plots the two beta dose responses as a function of the average alpha dose response (i.e. the
15 15 a -value). Note that the alpha and beta dose-response curves overlap up to ~50 Gy of beta or alpha effective
16 16 dose. This confirms that the a -value is constant over this dose-range. However, it appears that for dose > 300
17 17 Gy the interpretation of the presented alpha dose-response is difficult since at high-doses either the a -value
18 18 varies or the mechanisms responsible for luminescence production are different. Nevertheless, it is important to
19 19 note that by considering an a -value of 0.04 (as used in our previous studies) (i) the alpha dose response does not
20 20 saturate earlier than the beta dose response, (ii) for doses higher than 300 Gy the light output following alpha
21 21 irradiation is higher for alpha than for beta doses of the same magnitude. This provides further evidence that the
22 22 early saturation of the natural signals when compared to laboratory generated signals presented by **Timar-**
23 23 **Gabor and Wintle (2013)** cannot be accounted by the alpha dose received in nature by the fine grains.

24 24 **3.3.4. Can alpha irradiation overprint the effect of beta irradiation?**

25 25 Because of its high ionization power, alpha irradiation saturates any electron traps close to the alpha tracks. The
26 26 presence of saturated alpha tracks in an area receiving a beta dose may affect the luminescence efficiency of this
27 27 beta dose because in that area some traps cannot trap further charges. We test whether this model is of
28 28 significance by building a mixed alpha beta growth curve for which a constant alpha irradiation (total alpha
29 29 dose of 5000 Gy) is administered prior to beta irradiation (**Figure 6**). For comparison pure beta growth curves
30 30 have been constructed on the same aliquot before and after the mixed growth curve. **Figure 6** shows that the
31 31 pure beta dose-response curves as well as the mixed radiation dose response curves are reproducible and best
32 32 fitted with a sum of two saturating exponentials. The characteristic doses are indicated for each dataset. These

1 1 results suggest that beta and beta-equivalent alpha irradiations can largely be interchanged to produce the same
2
3 luminescence response, despite the fact that there is an order of magnitude difference in the dose rate and in the
4
5 ionization efficiency or LET (Linear Energy Transfer) between alpha and beta doses. Thus the two radiations
6
7 share the same traps, and there are no adverse, irreversible effects of saturation or defect creation during alpha
8
9 irradiation.

10
11

12 4. Summary and conclusion

13
14 The presence of a prior alpha dose in etched 63-90 μm quartz grains does not alter the dose-response to
15
16 laboratory beta irradiation as shown by the OSL growth curves constructed after irradiation with alpha doses of
17
18 9552 Gy. It is thus not likely that the past natural alpha dose absorbed by the 4-11 μm quartz grains is a cause
19
20 for the growth of the OSL signal to much higher doses compared to coarse quartz, as has been previously
21
22 reported for Romanian (e.g. **Timar-Gabor et al. (2012)**) and Serbian loess (**Timar-Gabor et al., 2014**). As
23
24 reported by **Galloway (2002)**, OSL signals induced by alpha irradiation and beta irradiation in fine grained
25
26 quartz have similar luminescence lifetimes; an indication that the two emissions follow the same recombination
27
28 path.

29 The differences regarding the shape and saturation characteristics of the OSL signal growth in nature and in
30
31 laboratory reported for Romanian loess (**Timar-Gabor and Wintle, 2013**) are thought not to reside in the
32
33 contribution of the alpha irradiation component in nature. We have shown that mixed alpha-beta dose response
34
35 curve simulating the nature radiation field reproduce the beta dose response curves up to 800 Gy. Using light
36
37 level matching and assuming an a-value of 0.04 we have shown that laboratory alpha and beta dose response
38
39 curves overlap up to effective alpha doses of ~50-100 Gy.

40 To conclude, exposure of quartz grains to alpha irradiation during burial and transport cycles, as well as the
41
42 mixed radiation field experienced in nature by the fine grains are not believed to be the cause for the age
43
44 discrepancies reported on fine and coarse grained quartz extracted from Romanian and Serbian loess.

45
46

47 5. Acknowledgements

48
49 A.T-G. and D.C. acknowledge the financial support from a grant of the Romanian National Authority
50
51 for Scientific Research CNCS-UEFISCDI, PN-II-RU-TE-2011-3-0062, nr. 73/05.10.2011. D.C. also
52
53 acknowledges the financial support of the project POSDRU/159/1.5/S/133391-“Doctoral and postdoctoral
54
55 excellence programs for training highly qualified human resources for research in the fields of Life Sciences,
56
57 Environment and Earth”.

58
59

60 6. References

61
62
63
64
65

- 1 1
2 2
- 2 3 Aitken, M., 1985. Thermoluminescence dating. Academic Press, London, p. 360.
- 3 4
3 5 Aitken, M.J., Bowman, S.G.E., 1975. Thermoluminescent dating: assessment of alpha particle contribution.
4 6
4 7 Archaeometry 17, 6.
- 5 8
5 9 Ankjærgaard, C., Jain, M., Thomsen, K.J., Murray, A.S., 2010. Optimising the separation of quartz and feldspar
6 10
6 11 optically stimulated luminescence using pulsed excitation. Radiation Measurements 45, 778-785.
- 7 12
7 13 Ballarini, M., Wallinga, J., Wintle, A.G., Bos, A.J.J., 2007. A modified SAR protocol for optical dating of
8 14
8 15 individual grains from young quartz samples. Radiation Measurements 42, 360-369.
- 9 16
9 17 Biswas, R.H., Williams, M.A.J., Raj, R., Juyal, N., Singhvi, A.K., 2013. Methodological studies on
10 18
10 19 luminescence dating of volcanic ashes. Quaternary Geochronology 17, 14-25.
- 11 20
11 21 Burbidge, C.I., Dias, M.I., Prudêncio, M.I., Rebêlo, L.P., Cardoso, G., Brito, P., 2009. Internal α activity:
12 22
12 23 localisation, compositional associations and effects on OSL signals in quartz approaching β saturation.
13 24
13 25 Radiation Measurements 44, 494-500.
- 14 26
14 27 Chithambo, M.L., 2003. Dependence of the thermal influence on luminescence lifetimes from quartz on the
15 28
15 29 duration of optical stimulation. Radiation Measurements 37, 167-175.
- 16 30
16 31 Constantin, D., Begy, R., Vasiliniuc, S., Panaiotu, C., Necula, C., Codrea, V., Timar-Gabor, A., 2014a. High-
17 32
17 33 resolution OSL dating of the Costinești section (Dobrogea, SE Romania) using fine and coarse quartz.
18 34
18 35 Quaternary International 334-335, 20-29.
- 19 36
19 37 Constantin, D., Cameniță, A., Panaiotu, C., Necula, C., Codrea, V., Timar-Gabor, A., 2014b. Fine and coarse-
20 38
20 39 quartz SAR-OSL dating of Last Glacial loess in Southern Romania. Quaternary International in press.
- 21 40
21 41 Cunningham, A.C., Wallinga, J., 2010. Selection of integration time intervals for quartz OSL decay curves.
22 42
22 43 Quaternary Geochronology 5, 657-666.
- 23 44
23 45 Denby, P.M., Bøtter-Jensen, L., Murray, A.S., Thomsen, K.J., Moska, P., 2006. Application of pulsed OSL to
24 46
24 47 the separation of the luminescence components from a mixed quartz/feldspar sample. Radiation
25 48
25 49 Measurements 41, 774-779.
- 26 50
26 51 Duller, G.A.T., 2003. Distinguishing quartz and feldspar in single grain luminescence measurements. Radiation
27 52
27 53 Measurements 37, 161-165.
- 28 54
28 55 Galloway, R.B., 2002. Comparison of luminescence lifetimes in quartz following alpha and beta irradiation.
29 56
29 57 Radiation Measurements 35, 591-593.
- 30 58
30 59 Kreutzer, S., Fuchs, M., Meszner, S., Faust, D., 2012. OSL chronostratigraphy of a loess-palaeosol sequence in
31 60
31 61 Saxony/Germany using quartz of different grain sizes. Quaternary Geochronology 10, 102-109.
- 32 62
32 63 Lai, Z.P., Zöller, L., Fuchs, M., Brückner, H., 2008. Alpha efficiency determination for OSL of quartz extracted
33 64
33 65 from Chinese loess. Radiation Measurements 43, 767-770.

- 1 1 Lapp, T., Jain, M., Ankjærgaard, C., Pirtzel, L., 2009. Development of pulsed stimulation and Photon Timer
2
3 attachments to the Risø TL/OSL reader. *Radiation Measurements* 44, 571-575.
4
- 3 5 Lomax, J., Fuchs, M., Preusser, F., Fiebig, M., 2014. Luminescence based loess chronostratigraphy of the
4 6
7 Upper Palaeolithic site Krems-Wachtberg, Austria. *Quaternary International* 351, 88-97.
- 5 8 Lowick, S.E., Preusser, F., Pini, R., Ravazzi, C., 2010. Underestimation of fine grain quartz OSL dating towards
9
10 the Eemian: Comparison with palynostratigraphy from Azzano Decimo, northeastern Italy. *Quaternary*
11
12 *Geochronology* 5, 583-590.
13
- 8 14 Mauz, B., Packman, S., Lang, A., 2006. The alpha effectiveness in silt-sized quartz: new data obtained by single
15
16 and multiple aliquot protocols. *Ancient TL* 24, 6.
- 10 17 Murray, A.S., Wintle, A.G., 2000. Luminescence dating of quartz using an improved single-aliquot
18
19 regenerative-dose protocol. *Radiation Measurements* 32, 57-73.
20
- 12 21 Murray, A.S., Wintle, A.G., 2003. The single aliquot regenerative dose protocol: potential for improvements in
22
23 reliability. *Radiation Measurements* 37, 377-381.
24
- 14 25 Polymeris, G.S., Afouxenidis, D., Raptis, S., Liritzis, I., Tsirliganis, N.C., Kitis, G., 2011. Relative response of
26
27 TL and component-resolved OSL to alpha and beta radiations in annealed sedimentary quartz. *Radiation*
28
29 *Measurements* 46, 1055-1064.
- 17 30 Rees-Jones, J., 1995. Optical dating of young sediments using fine-grain quartz. *Ancient TL* 13, 9-14.
31
- 18 32 Sugisaki, S., Buylaert, J.-P., Murray, A., Tsukamoto, S., Nogi, Y., Miura, H., Sakai, S., Iijima, K., Sakamoto,
33
34 T., 2010. High resolution OSL dating back to MIS 5e in the central Sea of Okhotsk. *Quaternary*
35
36 *Geochronology* 5, 293-298.
- 21 37 Thomsen, K.J., Bøtter-Jensen, L., Denby, P.M., Moska, P., Murray, A.S., 2006. Developments in luminescence
38
39 measurement techniques. *Radiation Measurements* 41, 768-773.
40
- 23 41 Thomsen, K.J., Bøtter-Jensen, L., Jain, M., Denby, P.M., Murray, A.S., 2008. Recent instrumental
42
43 developments for trapped electron dosimetry. *Radiation Measurements* 43, 414-421.
44
- 25 45 Timar-Gabor, A., Constantin, D., Markovic, S., Jain, M., 2014. Extending the area of investigation of fine
46
47 versus coarse quartz optical ages from the Lower Danube to the Carpathian Basin. *Quaternary*
48
49 *International* in press.
- 28 50 Timar-Gabor, A., Vandenberghe, D.A.G., Vasiliniuc, S., Panaitu, C.E., Panaiotu, C.G., Dimofte, D., Cosma,
51
52 C., 2011. Optical dating of Romanian loess: A comparison between silt-sized and sand-sized quartz.
53
54 *Quaternary International* 240, 62-70.
55
- 31 56 Timar-Gabor, A., Vasiliniuc, Ş., Vandenberghe, D.A.G., Cosma, C., Wintle, A.G., 2012. Investigations into the
57
58 reliability of SAR-OSL equivalent doses obtained for quartz samples displaying dose response curves
59
60 with more than one component. *Radiation Measurements* 47, 740-745.
61
62
63
64
65

1 1 Timar-Gabor, A., Wintle, A.G., 2013. On natural and laboratory generated dose response curves for quartz of
2 different grain sizes from Romanian loess. *Quaternary Geochronology* 18, 34-40.
3 3
4 Timar, A., Vandenberghe, D., Panaiotu, E.C., Panaiotu, C.G., Necula, C., Cosma, C., van den haute, P., 2010.
5 5
6 Optical dating of Romanian loess using fine-grained quartz. *Quaternary Geochronology* 5, 143-148.
7 7
8 Tribolo, C., Mercier, N., Valladas, H., 2001. Alpha sensitivity determination in quartzite using OSL single
9 9
10 aliquot procedure. *Ancient TL* 19, 4.
11 11
12 Wintle, A.G., Murray, A.S., 2006. A review of quartz optically stimulated luminescence characteristics and
13 13
14 their relevance in single-aliquot regeneration dating protocols. *Radiation Measurements* 41, 369-391.
15 15
16 Zimmerman, D.W., 1972. Relative thermoluminescence effects of alpha- and beta-radiation. *Radiation Effects*
17 17
18 14, 12.
19 19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65

FIGURE CAPTIONS

1

2

3 **Figure 1.** The effect of irradiation with a total α dose of 238.8 kGy (effective α dose of 9552 Gy assuming an a -
4 value of 0.04) prior to the construction of the β dose response curve. The dashed lines represent the β dose
5 response curves constructed on a single aliquot of 63-90 μm etched coarse quartz from sample CST 3 prior to
6 alpha irradiation. The grey solid line represents β dose response constructed on the same aliquot following α
7 irradiation. α doses were delivered in pulses of 193 Gy followed by exposure to blue LEDs for 200 s at 20 °C.
8 The dose response was best described by a sum of two exponential functions and the characteristic doses are
9 indicated for each growth curve.

10 **Figure 2.** Fine (4-11 μm) quartz TR-OSL curves following alpha (grey solid line) and beta (black solid line)
11 irradiation for sample CST 3. The TR-OSL spectra have been obtained by collecting signals during pulses (*on*-
12 time of 50 μs and *off*-time of 450 μs) that amount to a total time of 100 s. Stimulation was carried out at 125 °C.
13 The α irradiation time was 120 ks which based on alpha and beta light levels matching for this aliquot and an a
14 value of 0.04 corresponds to an effective dose of 455 Gy. The given beta dose was 581 Gy. Note that the
15 luminescence intensities are similar. Inset: close-up showing TR-OSL spectra from the 54-100 μs of the pulse.
16 The solid lines indicate the best fit of the data: a single exponential decay plus a constant and the corresponding
17 lifetimes. Please note the logarithmic scale.

18 **Figure 3.** Sensitivity changes over 10 cycles of alpha irradiation monitored using a 17 Gy β test dose on one
19 aliquot from sample CST 3 and one aliquot from sample LCA 26. In each cycle samples were exposed to a
20 constant alpha irradiation time of 26260 s, corresponding to an effective alpha dose of 100 Gy (considering an
21 a -value of 0.04). A high degree of linearity can be noticed and the functions pass through the origin (intercept
22 values are -1720 ± 1605 (CST 3) and 768 ± 2087 (LCA 26)).

23 **Figure 4.** Mixed α - β dose response curve (solid line) normalized to a 17 Gy of beta. Each dose delivered
24 combines a 12 % contribution of alpha and 88 % of β irradiation, to reproduce the natural radiation field
25 experienced by sample CST 3 (6 Gy eff α + 45 Gy β , 12 Gy eff α + 90 β , 23 Gy eff α + 179 β , 47 Gy eff α +
26 358 β , 93 Gy eff α + 717 β). In order to better reproduce natural irradiation, doses have been delivered in mixed
27 α/β pulses (6 Gy eff α + 45 Gy β). Pure beta growth curves were constructed before (dashed line) and after
28 (dotted line) the mixed dose response one. The inset presents the pure alpha growth curve build on the same
29 quartz aliquot using the alpha doses delivered in the mixed radiation experiment. Sensitivity changes were
30 monitored using a 17 Gy beta dose.

31 **Figure 5.** Comparison between beta (filled symbols) and alpha (open symbols) growth curves constructed on
32 two aliquots of CST 3 (up-triangle and down-triangle) and one aliquot of sample LCA 26 (star). The alpha dose

33 response was constructed on the same aliquot following the construction of the beta dose response. A constant
34 beta test dose of 17 Gy was used for correcting the sensitivity changes. The average beta dose response is
35 plotted against the average alpha dose response in the inset. Black solid line stands for 1:1 ratio and the dotted
36 lines bracket a 10% deviation from it. Relevant regenerative doses (beta doses/effective alpha doses) are
37 indicated by arrows.

38 **Figure 6.** Mixed $\alpha+\beta$ dose-response curve (solid line) corrected using a 17 Gy β test dose. Each mixed dose
39 point combines a fixed effective α irradiation equivalent of 200 β Gy (5000 Gy of total α) prior to a β dose. For
40 comparison, beta growth curves were constructed before (dashed line) and after (dotted line) the mixed one.
41 Recycling points are represented by stars. Saturation doses of the double exponential functions that best
42 described the data are indicated.

FIGURE 1

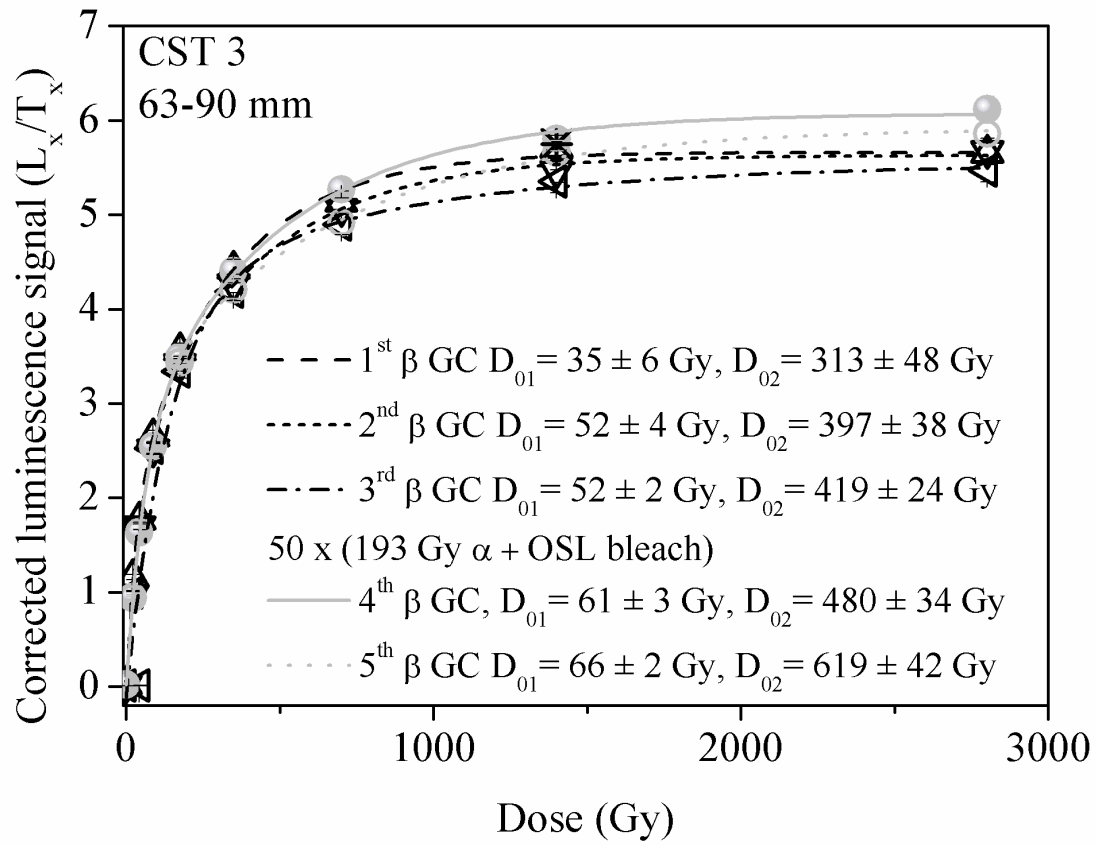


FIGURE 2

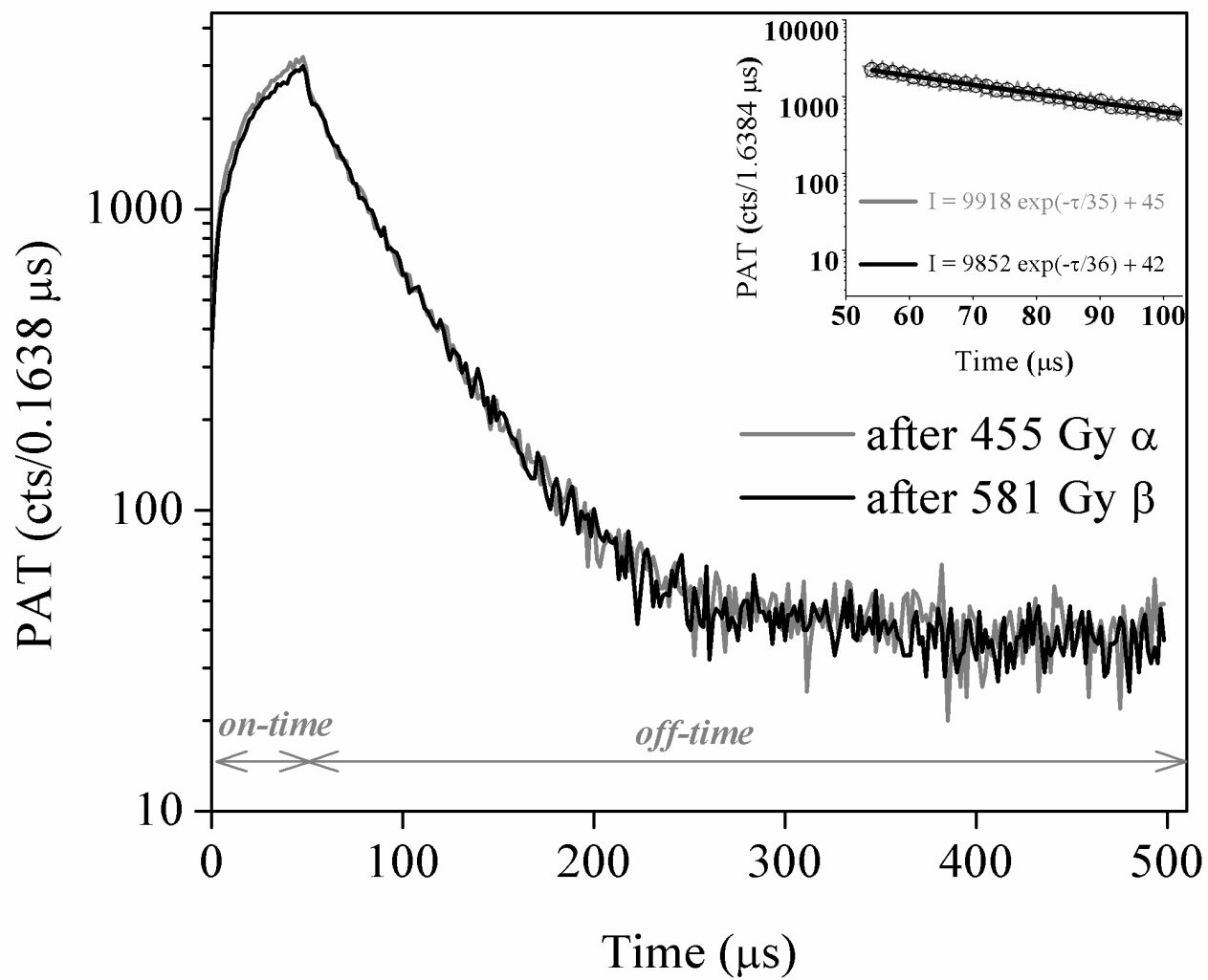


FIGURE 3

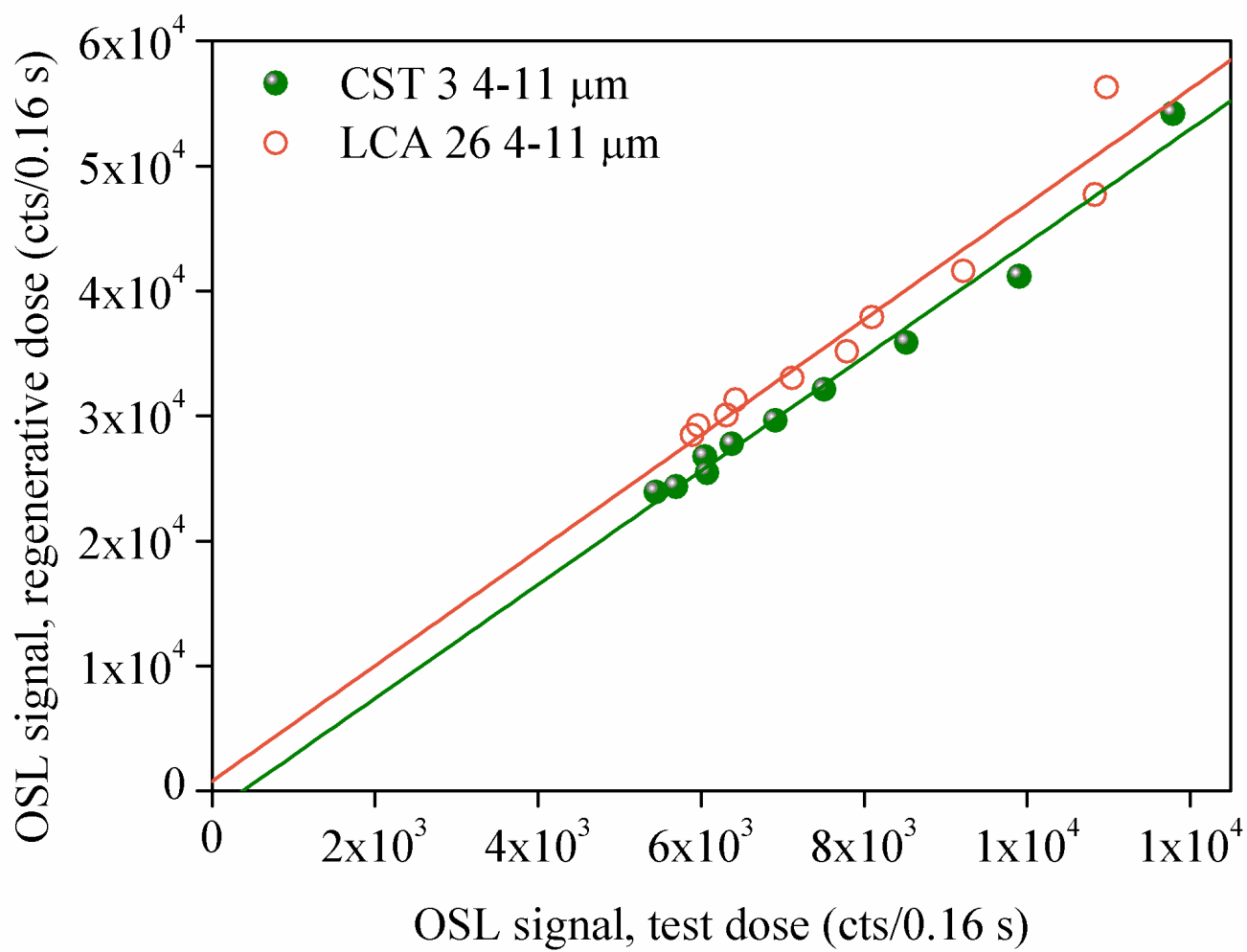
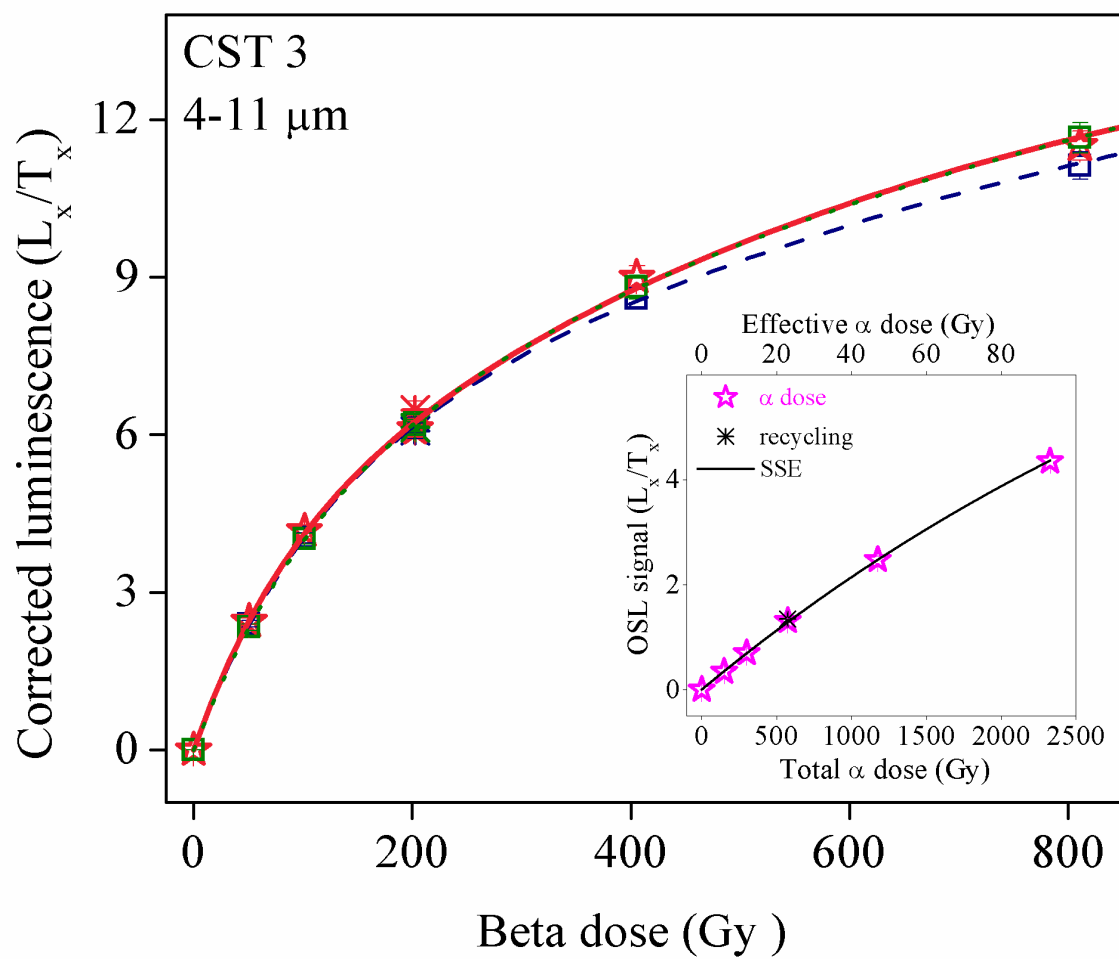


FIGURE 4



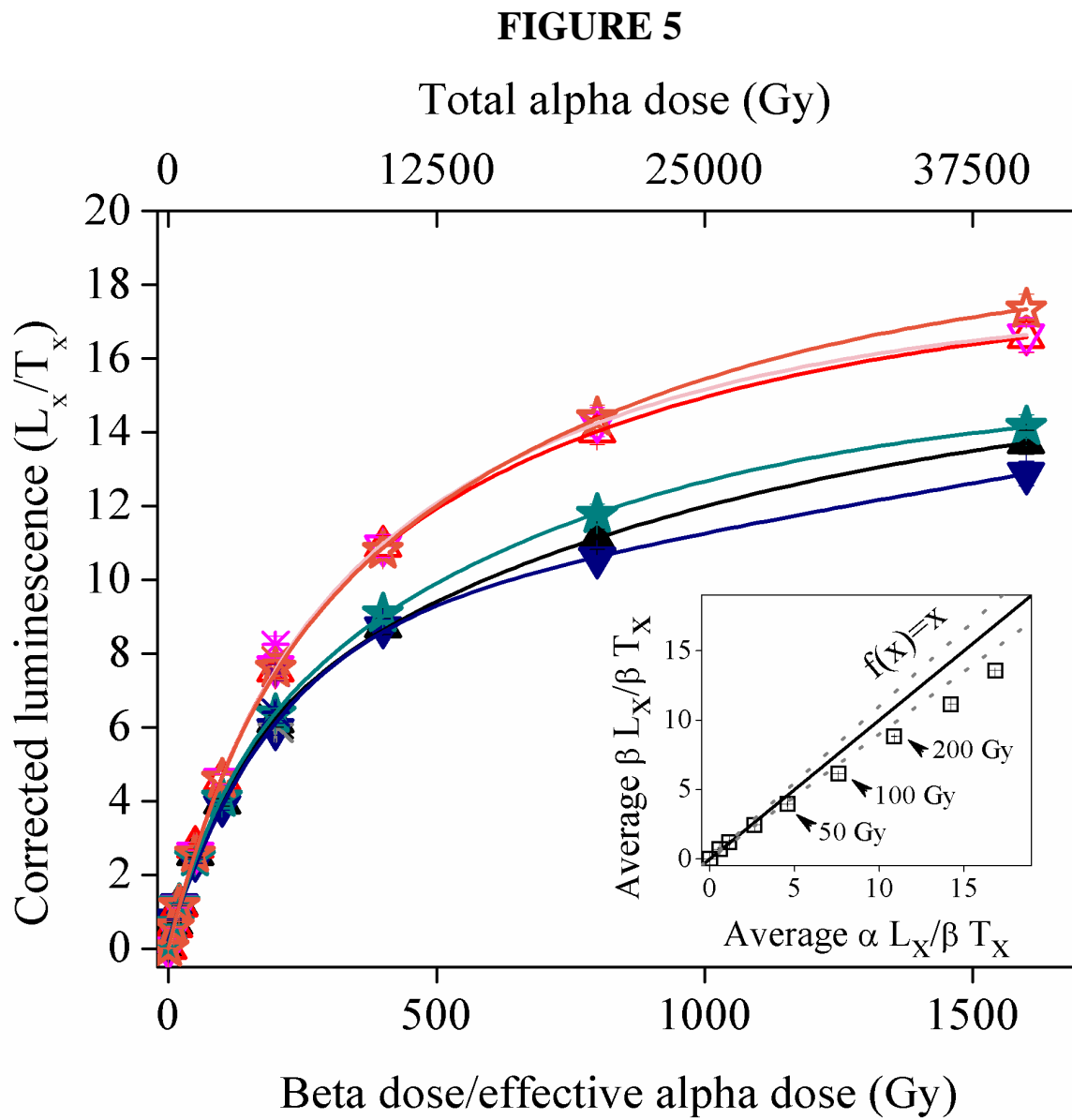
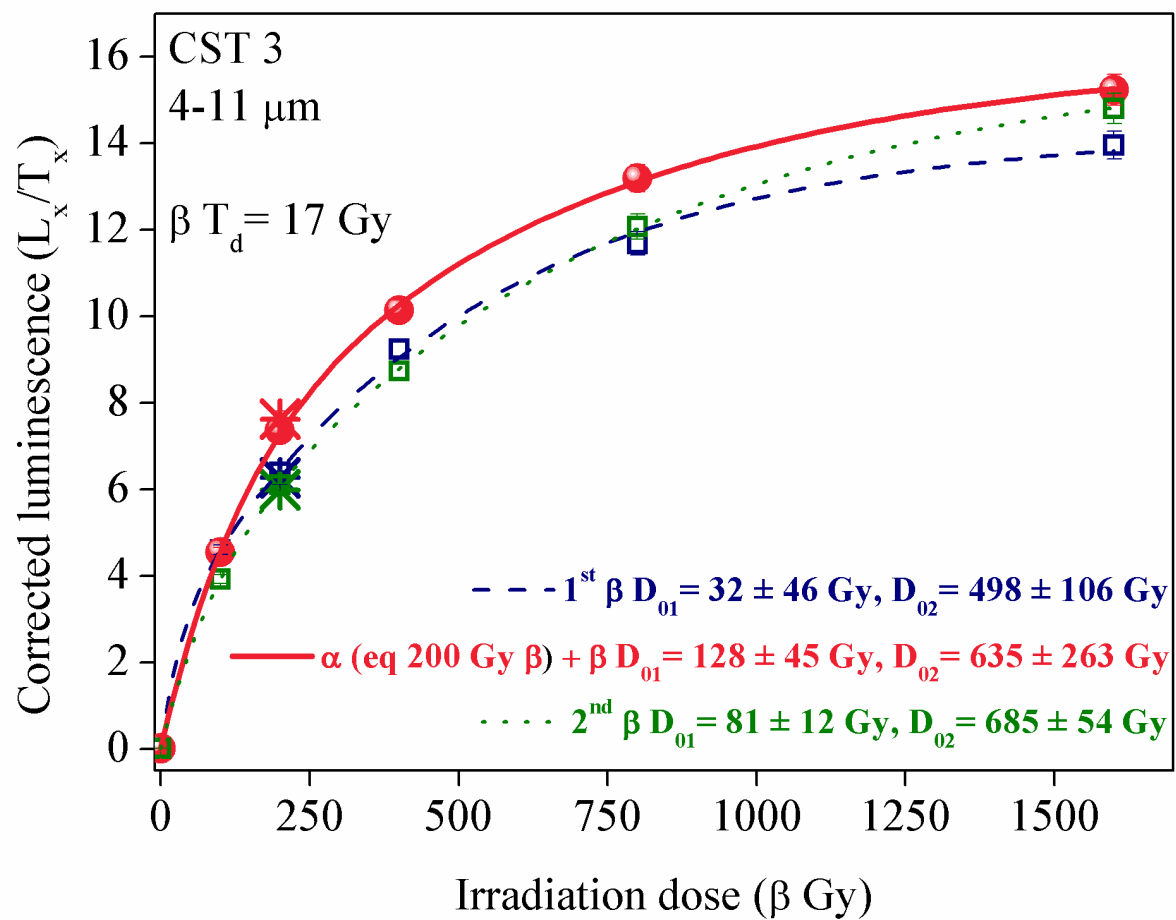


FIGURE 6



SUPPLEMENTARY MATERIAL

Quartz luminescence response to a mixed alpha-beta field: investigations on Romanian loess

D. Constantin, M. Jain, A.S. Murray, J.P. Buylaert, A. Timar-Gabor

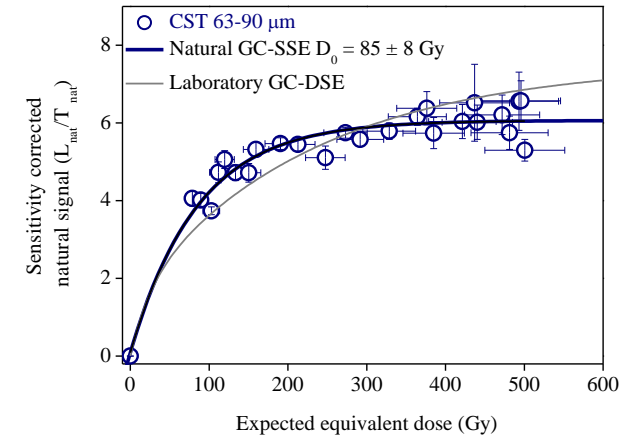
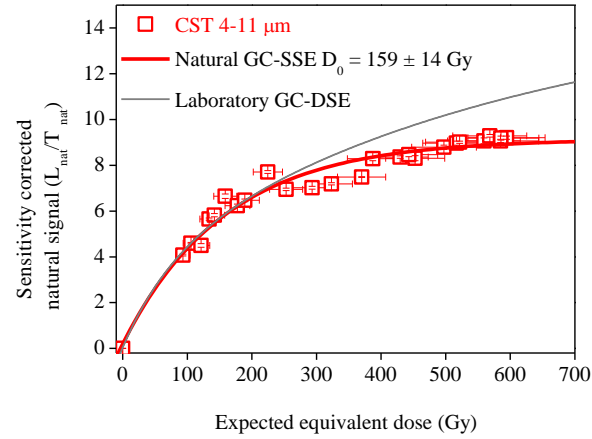
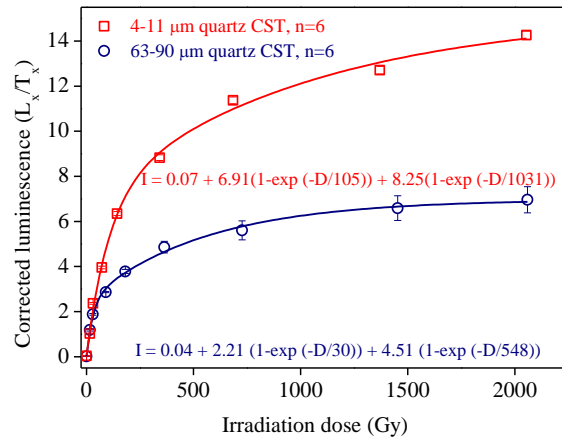
Table S1. Information relevant for OSL dating on sample CST 3 (Constantin et al., 2014). A water content of 10 ± 3 % was assumed over geological times based on the measured water content of the sample when collected.

Sample CST 3	4-11 μm	63-90 μm
Equivalent dose (Gy) SAR protocol	105 ± 3	117 ± 6
U-Ra (Bq kg^{-1})		31 ± 2
Th (Bq kg^{-1})		33 ± 1
K (Bq kg^{-1})		461 ± 7
Total annual dose rate (Gy ka^{-1})	3.09 ± 0.04	2.62 ± 0.03
Alpha contribution to the total annual dose rate (Gy ka^{-1}) corrected for water content	0.36 ± 0.07	
Age (ka)	34 ± 3	45 ± 4

SUPPLEMENTARY MATERIAL

Quartz luminescence response to a mixed alpha-beta field: investigations on Romanian loess

D. Constantin, M. Jain, A.S. Murray, J.P. Buylaert, A. Timar-Gabor



a

b

c

Figure S1. (a) Representative average sensitivity corrected SAR-OSL beta growth-curves for 4-11 μm (open squares) and 63-90 μm (open circles) quartz extracted from the Costinești loess section in Romania. The data are best fitted with a sum of two saturating exponential functions and the characteristic doses are given under each curve. Average sensitivity corrected natural signals (L_n/T_n) on all investigated aliquots of fine (b) and coarse quartz (c) plotted as function of their expected equivalent dose, derived from magnetic susceptibility ages. Data are fitted using a single saturating exponential (SSE) function. The average laboratory dose response curve fitted with a double saturating exponential (DSE) function is presented for comparison (Timar-Gabor et al., 2012; Timar-Gabor and Wintle, 2013).