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Pulsed IRSL: a stable and fast bleaching luminescence signal from feldspar for dating Quaternary sediments

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Abstract

Elevated temperature post-infrared infrared stimulated luminescence (post-IR IRSL; pIRIR) dating method using feldspar has led to a significant advancement in dating Quaternary sediments, as this signal has been shown to be stable on geological time scales. In comparison to the conventional IRSL, the pIRIR signal is nearly free from anomalous fading, but it is more difficult to reset during daylight exposure; the latter characteristic may give rise to age over-estimation in some environments because of partial bleaching. The pulsed IRSL signal has also been known to be less affected by anomalous fading, when the signal is recorded during the off-time of LED stimulation pulses. In this study, we compare the signal bleachability, thermal and athermal stability, and age estimates derived using different measurement methods, i.e., pulsed IRSL measured at 50°C (pulsed IR₅₀; subscript refers to measurement temperature), continuous wave (CW) pIRIR₂₂₅ and pIRIR₂₉₀ signals, as well as the two IR₅₀ signals measured as a part of the pIRIR protocols. This comparison is carried out for 3 samples taken from Late Quaternary fluvial sediments, which have independent age estimates from radiocarbon dating and tephrochronology. We observe that under exposure to solar simulator, the pulsed IR₅₀ signal bleaches as rapidly as the CW IR₅₀, and much more rapidly than the pIRIR signals. Furthermore, the pulsed IR₅₀ signal is also as stable as the pIRIR signals, especially when the middle part of the decay curve is used for signal intensity calculation.

1. Introduction

The post-infrared infrared stimulated luminescence (post-IR IRSL, hereafter pIRIR) dating method of feldspar, including both two-step pIRIR (Thomsen et al., 2008) and multi-elevated-temperature (MET)-pIRIR (Li and Li, 2011) have become robust methods for dating Quaternary sediments. However, the pIRIR signal appears to be more difficult to bleach than the IRSL signal measured at low temperature, typically at 50°C (IR₅₀; hereafter the subscript refers to the measurement temperature) (e.g. Buylaert et al., 2012). With increasing preheat and stimulation temperature the pIRIR signal tends to become more stable, but at the cost of poorer bleachability (Li and Li, 2011; Kars et al., 2014; Colarossi et al., 2015); thus, it appears that there is a trade-off between signal stability and its ability to reset prior to sediment burial. The post-IR IRSL signal measured at 290°C (pIRIR₂₉₀) is thought to be stable and there is no need for fading correction (e.g. Buylaert et al., 2012), whereas the pIRIR₂₂₅ signal is less stable and requires fading correction. However, pIRIR₂₂₅ signal is more bleachable than pIRIR₂₉₀, and is generally preferred for dating sediments from environments where full bleaching is less likely (e.g. Smedley et al., 2016). The pIRIR signal at a lower temperature (e.g. pIRIR₁₅₀) is generally preferred for young (e.g. Holocene) sediments (Reimann and Tsukamoto, 2012). However, there has been no consensus of how the preheat and stimulation temperatures should be selected (Zhang et al., 2015).

The pulsed IRSL signal has been also reported to be more stable than the continuous wave (CW) IRSL signal (Sanderson and Clark, 1994; Tsukamoto et al., 2006; Jain and Ankjærgaard, 2011; Jain et al., 2015), although this method has not been tested as widely as the pIRIR signal. Roskosch et al. (2015a,b) chose to use the pulsed IR₅₀ instead of the pIRIR signal for dating fluvial and glaciofluvial sediments, assuming that the slow bleachability of the pIRIR signal was not suitable for their samples. Jain et al. (2015a) showed that the residual dose from the modern sediments for the pulsed IRSL signal is much lower than that for the pIRIR signal and presumed that the pulsed IRSL signal bleaches faster than the pIRIR signal. Jain et al. (2015a) also observed that both pIRIR₂₉₀ and pulsed IRSL signals were in saturation in a sample with an age estimated 3.6 Ma. Thus, it appears that the pulsed IRSL signal has a better bleachability, but similar stability compared to the pIRIR₂₉₀ signal. In this paper, we characterise the pulsed IRSL signal in terms of bleaching, and thermal- and athermal-stability and compare these characteristics to those of the pIRIR signals (IRIR₂₂₅ and pIRIR₂₉₀) and the corresponding IRSL signals (IR₅₀) measured as part of the pIRIR protocol. We also compare the pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ ages for three fluvial sediment samples, which have independent age control.

2. Samples

Three fluvial sediment samples having age control were used in this study (Table 1, Fig. 2). Sample SW-II was taken from the Roman time harbour sediment in Cologne, Germany (Fig. 1), excavated by the construction works of the subway (Lauer et al., 2011a). The fluvial sediment at the former harbour of the Rhein contains broken ceramics and bricks from Roman time, and the harbour was known to have been used in the early first century (Dietmar and Trier, 2006). Three charcoal samples (RC-1, -2, and -3) which were found above and below SW-II were dated between 2010 ± 55 a BP and 2080 ± 85 a BP by radiocarbon (Lauer et al., 2011a). The calibrated radiocarbon ages are all consistent to the first century AD (Table 1). The OSL age of SW-II was calculated by various statistical methods; the mean and the 3 component minimum age model (MAM-3) were reported to be 3460 ± 350 a and 1690 ± 590 a, respectively (Lauer et al., 2011a). The latter age was recalculated with the new version of MAM-3 and yielded 1980 ± 330 a using 15% overdispersion.

Sample MHT-II was taken from an outcrop at Monheim-Hitdorf, ~20 km north of Cologne (Fig. 1), where a 6-m thick sediment is intercalated with pumices of the Laacher See Tephra. Sanidine phenocrysts of the Laacher See Tephra were dated to 12900 ± 560 a by the $^{40}\text{Ar}/^{39}\text{Ar}$ method (van den Bogaard, 1995). Hadjas et al. (1995) dated macrofossils in varved lake sediments of Holzmaar, Eifel Volcanic Field, Germany. The calibrated ^{14}C age of the Laacher See Tephra was reported to be $12,201 \pm 224$ cal. BP in the paper. However, the calibrated ages of two subsamples closest to the tephra by Intcal 13 are ranging from 12830 to 13266 cal BP and from 13071 to 13413 cal BP, in agreement with the $^{40}\text{Ar}/^{39}\text{Ar}$ age. The fluvial sediment studied by Lauer et al. (2011b) can be considered to be slightly younger than the eruption age of the Laacher See Tephra. Lauer et al. (2011b) dated three samples from the fluvial sediment sequence (MHT-I, -II, and -III) which yielded similar quartz OSL ages from 11.6 ± 0.7 to 11.7 ± 1.0 ka. MHT-I, which was taken 4 m below MHT-II, was also measured with K-feldspar using pIRIR₂₂₅ and post-IR yellow stimulated luminescence signals (Lauer et al., 2012), which are consistent with the quartz OSL age, 12.2 ± 1.2 ka and 11.5 ± 1.2 ka, respectively.

Sample LUM-3291 was collected from fluvial sediment of the Tanahara-Harataima (Th) terrace (Kaizuka and Moriyama, 1969) of Sagami River, at Muryokoji in Sagami, near Tokyo, Japan (Fig. 1). The fluvial sediment here is overlain by ~3.5 m tephric loess, which is a mixture of loess, local dust and tephra from small scale eruptions. About 40 cm from the bottom of the tephric loess, a widespread Aira Tazawa (AT) tephra from southern Kyushu has been found (Research Group of Gemorphology and Geology in Sagami, 1984). This tephra was dated to $25,120 \pm 270$ BP (from 28577 to 29831 cal BP) by radiocarbon of charcoal and humic acid samples taken close to the source area (Miyairi et al., 2004). This tephra was also found in the varved sediment in the Lake Suigetsu, and the age of AT has been estimated to $30,009 \pm 189$ a cal BP SG06₂₀₁₂ (Smith et al. 2013). The depositional

age of the fluvial sediment has been estimated to be slightly older than the age of AT tephra (Research Group of Geomorphology and Geology in Sagami-hara, 1984; Kubo, 1997).

3. Methods

Sand sized K-feldspar samples between 150 and 200 μm in diameter were prepared for all three samples. The standard preparation procedure including density separation of $d < 2.58$ was made at Leibniz Institute for Applied Geophysics for LUM-3291. Two other samples (SW-II and MHT-II) were separated using the flotation technique (Miallier et al., 1983) at University of Freiberg. All samples were measured using large aliquots containing a few hundred grains.

Gamma spectrometry was performed using 50 g of dried material for LUM-3291 for estimating the environmental dose rate. The radioactivity data for SW-II and MH-II were obtained from Lauer et al. (2011a and b). The annual dose rate for the K-feldspar samples was calculated using the natural water content, the conversion factors of Guerin et al. (2011), the internal K concentration of $12.5 \pm 0.5 \%$ (Huntley and Baril, 1997) and an a -value of 0.09 ± 0.02 (Balescu et al., 2007).

All luminescence measurements were performed with an automated Risø TL/OSL DA-20 reader, equipped with pulsed-stimulation and Photon-Timer attachments (Lapp et al., 2009). The IRSL signal is filtered through a combination of BG39 and 7-59 filters transmitting between 380 and 460 nm. Both for the time resolved (TR)-IRSL and the pulsed-IRSL measurements, the pulse width was set to 100 μs ('on'-time) and the period between two stimulation pulses was set to 400 μs ('off'-time). The off-time pulsed IRSL signal was detected after rejecting the initial 21.5 μs signal. The normal setting of the Risø pulsed OSL card only allows to reject up to 5 μs of the initial off-time signal. We added a 330 pF capacitor on top of the existing 100 pF one on the pulsing card, which made the gating time 4.3 times longer.

4. TR-IRSL signal

Fig. 3a shows a 3D plot of a TR-IRSL measurement of an aliquot of MH-II after giving a regenerative dose of 40 Gy and preheat at 250°C for 60s. The total measurement period was 500 s; thus Fig. 3b represents a sum of the individual time-resolved data corresponding to 1 million IR pulses. The TR-IRSL signal rises gradually when the stimulation light is switched on, and decays after the light is turned off. From the TR-IRSL data, two decay curves were extracted using 1) on-time (0-100 μs) data and 2) off-time data after rejecting the initial 21.5 μs . As can be seen in Fig. 3c, the signal intensity in the off-time

1 is only about ~2.5 % of that in the on-time data. In Fig. 3d, the decay curves were normalised to the
2 initial intensity. The decay rate of the off-time signal is slower than the on-time signal.

3
4 The IRSL signal from feldspar is understood to originate from decay of the excited state of the trapped
5 electrons by tunnelling or localised diffusion within the band tail states, leading to recombination with
6 the holes at the nearest-neighbouring sites (e.g. Jain and Ankjærgaard, 2011; Jain et al., 2012, 2015b).
7 As the IR stimulation continues, the distance between the trapped electrons and their nearest holes
8 should gradually increase leading to a slower decay of the trapped electron population with time; this
9 leads to a decay form following a Becquerel function (Pagonis et al., 2014; Jain et al., 2015b). Similarly,
10 since the off-time IRSL signal has been shown to arise from more distant electron-hole pairs (Jain and
11 Ankjærgaard, 2011) compared to the on-time signal, its decay should also be relatively slower; this is
12 confirmed in Fig. 3d.

13 14 5. Bleachability

15
16 Poolton et al. (2002) suggested that the bleaching rate of the IRSL signals decreases with increasing
17 stimulation temperature. This was shown experimentally by Li and Li (2011) who measured the
18 bleaching curves of the MET-pIRIR signals stimulated between 50 and 250°C. Buylaert et al. (2012)
19 presented that the pIRIR₂₉₀ signal is much harder to bleach than the IR₅₀ signal. They also showed that
20 the IR₅₀ signal with a higher preheat temperature (320°C as a part of the pIRIR₂₉₀ protocol, hereafter
21 pre-pIR₂₉₀) was more difficult to bleach than the IR₅₀ with a lower temperature preheat (250°C).
22 Colarossi et al. (2015) compared the bleachability of four IRSL signals from the pIRIR₂₂₅ and pIRIR₂₉₀
23 protocols: i.e., IR₅₀ (pre-pIR₂₂₅), IR₅₀ (pre-pIR₂₉₀), pIRIR₂₂₅ and pIRIR₂₉₀. The bleaching rates of these
24 signals varied systematically from the fastest to the slowest in the following order, IR₅₀ (pre-pIR₂₂₅)>
25 IR₅₀ (pre-pIR₂₉₀)> pIRIR₂₂₅ > pIRIR₂₉₀.

26
27 In this study, we compare the bleachability of the IR₅₀ (pre-pIR₂₂₅), IR₅₀ (pre-pIR₂₉₀), pIRIR₂₂₅, pIRIR₂₉₀
28 and pulsed IR₅₀ signals for two samples, MH-II and LUM-3291. To eliminate the possible effect of the
29 variation in natural dose across the (sub)samples, we first carried out laboratory zeroing of the signal
30 by a double exposure to IR LEDs in the Risø reader at 350°C for 200 s, followed by a regeneration dose
31 of 20 Gy; there was no detectable IRSL signal after the zeroing. Only 3 aliquots were beta irradiated at
32 a time, and they were brought immediately to a solar simulator (Hönle SOL2) for bleaching, in order to
33 minimise the delay between the irradiation and the start of the bleaching. Three aliquots per each
34 bleaching duration were bleached between 0 and 4 hours (without bleach, 3 minutes, 10 minutes, 20

minutes, 1 hour, 2 hour and 4 hour) in the SOL2, and the D_e values were measured using pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ protocols (Table 2).

Fig. 4a shows the bleaching curves of MH-II for the IR₅₀ (pre-pIR₂₂₅), IR₅₀ (pre-pIR₂₉₀), pIRIR₂₂₅ and pIRIR₂₉₀, and pulsed IR₅₀ plotted as remaining D_e value vs. the bleaching time. The initial signal (0-4 s) minus the background of the last 20 s of the decay curves were used to calculate the D_e values. For the pulsed IR₅₀, equivalent integration periods to the CW-IRSL data were used (i.e. 5 times CW-IRSL data). Each data point represents an arithmetic mean of 3 aliquots and 1- σ standard error. It can be clearly seen that the pulsed IR₅₀ signal bleaches as fast as the CW IR₅₀ signals (pre-pIR₂₂₅ and pre-pIR₂₉₀) and bleaches much faster than the pIRIR₂₂₅ and pIRIR₂₉₀ signals. The same tendency is observed from LUM-3291 (Fig. 4b), although the data have larger uncertainty due to the lower signal intensity for this sample.

We also examined whether there is any dependence of the D_e , measured after bleaching, on the integral section of the IRSL decay curves used for the D_e estimation. The 'remaining' D_e values for the different signal integration periods, 0-4s, 4-12s, 12-20s and 20-40s, are plotted for the sample MHT-II in Fig. 4c and d. All the signals become harder-to-bleach towards the latter part of the decay curve, however, the latter part of the IR₅₀ signal, both CW and pulsed, are still more rapidly bleachable than the initial part of the pIRIR signals.

5. Anomalous fading

Fading tests were conducted using 6 aliquots for the five signals discussed above using a generation dose of 40 Gy and a test dose of 10 Gy, and delays up to 40 hours. The g_{2days} -values of all signals for all the three samples using the initial signal (0-4 s) are shown in Fig. 5a. The pulsed IR₅₀ fading rate is slightly higher than the two pIRIR signals but lower than the CW IR₅₀ signals, when the initial signal intensity is used. It is interesting to note that the IR₅₀ (pre-pIR₂₉₀) fading rate is much lower than the IR₅₀ (pre-pIR₂₂₅) fading rate; this is possibly due to the effect of a higher preheat used in the pIR₂₉₀ protocol (Jain et al., 2015a, b). When the signal intensity from the later part of the decay curve was used (4-12s, 12-20s and 20-40s), the fading rate of two pIRIR signals and pulsed IRSL signal became indistinguishable (Fig. 5b-d). The fading rate of all signals tend to decrease towards the later stimulation time, probably due to the increasing electron-hole recombination distance with the stimulation time (e.g. Thomsen et al., 2011; Jain and Ankjærgaard, 2011).

We also explored time resolved fading in sample MH-II. A fading measurement using the TR-IRSL₅₀ signal was performed using two aliquots of MH-II. The given regeneration and test doses were the same as above (40 and 10 Gy) and repeated L/T measurements were done at 50°C for 500 s after a preheat at 250°C for 60s with delays up to 59 hours. The result is plotted against pulse arrival time in Fig. 6 for the initial part (0-4 s on time) and for the middle part (4-12 s) of the total measurement duration. The fading rate is high during the on-time, and during the off-time it does not drop immediately after the LEDs are switched off but decreases gradually; this confirms the results of Jain and Ankjærgaard (2011). Furthermore, as expected, the fading rates calculated from the middle part of the decay curve are always lower than those from the initial signal for both the on- and off-times. The g_{2days} -value becomes consistent with zero for the middle part of the decay curve (4-12s) at around ~200 μ s (~100 μ s during the off-time).

6. Thermal stability

Thermal stability measurements were conducted to compare the thermal stability of the IR₅₀ (pre-pIR₂₂₅), pIRIR₂₂₅, and pulsed IR₅₀ signals. It is already known that the pIRIR signal appears to be thermally more stable than the IR₅₀ signal (e.g. Thomsen et al., 2011; Jain and Ankjærgaard, 2011, Tsukamoto et al., 2012), even though, both these signals are argued to originate from the same trap (Jain and Ankjærgaard, 2011; Andersen et al., 2012). The interpretation of this behaviour is that the thermal stability obtained by a pulse annealing experiment is not just the measure of a trap depth, but the also the stability of an electron-hole pair as a function of the mean electron-hole separation (Jain and Ankjærgaard, 2011; Jain et al., 2015b).

Two aliquots of each sample were irradiated with 40 Gy and preheated between 260 and 500°C (with a 20°C increment) for pIRIR₂₂₅ and pulsed IR₅₀ protocols. The result calculated from the initial signal (0-4 s) for MH-II is shown in Fig. 7a. The thermal stability of the pulsed IR₅₀ signal lies in between the IR₅₀ (pre-pIR₂₂₅) and pIRIR₂₂₅ signals, and the same was observed for the other two samples (the data are not shown). This suggests that the electron-hole separation distance for the pulsed IR₅₀ signal is greater than the CW-IR₅₀ signal but shorter than the pIRIR₂₂₅ signal. In Fig. 7b, the data are plotted with different signal integration periods, 0-4s, 4-12 s, 12-20s, and 20-40s. The latter part of the signals become more thermally stable for all signals, also reflecting the increasing distance of the electron-hole pair with the stimulation time.

7. Age comparison

Dose recovery tests were performed using all three samples and for the three protocols, pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ (Table 2). Altogether, eighteen aliquots per sample were bleached in the SOL2 for four hours. For half of the aliquots, a beta dose which is close to the estimated D_e value was given, and measured if the protocols can recover the given dose. The remaining aliquots were measured without giving dose, and the residual dose after the 4 hour bleaching was determined. The dose recovery ratio was calculated by dividing the recovered dose with the given dose, after subtracting the measured residual dose. The results are shown in Fig. 8. The dose recovery ratio of all three samples are close to unity for the pIRIR₂₂₅ and pulsed IR₅₀ signal. However, the dose recovery ratio of the pIRIR₂₉₀ signal tends to be greater than unity, while, except for LUM-3291, the dose recovery ratio of the IR₅₀ (pre-pIR₂₂₅) and IR₅₀ (pre-pIR₂₉₀) are less than unity.

Between ten and twelve aliquots for each sample were measured to calculate D_e values using pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ protocols (Table 2). Here, the IR₅₀ (pre-pIR₂₂₅ and pre-pIR₂₉₀) signals are not considered because their fading rate was large and the dose recovery ratio was not satisfactory for the two samples, SW-II and MHM-II. The mean D_e values were calculated using the initial signal (0-4 s) for the two pIRIR signals but the middle part (4-40 s) was used for the pulsed IR₅₀ signal. The fading corrections were conducted based on Huntley and Lamothe (2001). The resulting ages were then compared with independent age control. Fig. 9 shows fading uncorrected and corrected ages in comparison with the independent age control, plotted against the signal integration limit. Only uncorrected ages are shown if the fading rate is statistically consistent with zero.

SW-II was taken from the Roman time harbour sediments in Cologne, known to be in the first century AD. Both radiocarbon ages from charcoal and quartz MAM age of the sample are consistent with the historical evidence, ~2 ka, however the quartz mean OSL age of this sample is 3.46 ± 0.35 ka (Lauer et al., 2011a). Since the bleaching rate of any feldspar luminescence signal is much slower than the quartz OSL signal, we do not expect an age younger than ~3.5 ka from K-feldspar. Both fading uncorrected (3.54 ± 0.38 ka) and corrected (3.74 ± 0.41 ka) ages of pulsed IR₅₀ signal of SW-II are consistent with the quartz mean OSL age of this sample, whereas the pIRIR₂₂₅ and pIRIR₂₉₀ ages are 5.87 ± 0.37 ka (6.85 ± 0.44 ka, fading corrected) and 11.0 ± 0.7 ka (12.8 ± 0.8 ka, fading corrected), respectively (Table 3).

MHT-II was collected from the fluvial sediment intercalated with the reworked Laacher See Tephra, which was erupted at ~13 ka. Thus, the age of the sediment is expected to be slightly younger than the age of the tephra. The quartz OSL age of 11.6 ± 0.7 ka for this sample was reported by Lauer et al. (2011b). Fading uncorrected pulsed IR₅₀ age is slightly younger (9.68 ± 0.55 ka) than the quartz OSL age, due to non-negligible fading of this sample (1.6 %/decade). The fading corrected age using the

1 middle part of the pulsed IR₅₀ signal (11.1 ± 0.7 ka) is in good agreement with the quartz age. The
2 pIRIR₂₂₅ and pIRIR₂₉₀ ages again overestimated the expected age; 14.0 ± 0.7 ka (17.3 ± 1.1 ka, fading
3 corrected) for the pIRIR₂₂₅, 19.9 ± 1.1 ka (23.3 ± 1.7 ka, fading corrected) for the pIRIR₂₉₀.

4
5 LUM-3291 is a fluvial sediment consisting of Tanahara-Harataima (Th) terrace of the Sagami River. The
6 AT tephra of 30 ka (Miyairi et al., 2004; Smith et al., 2013) was found in the tephric loess sediment,
7 ~40 cm from the bottom of the tephric loess, and therefore the fluvial sediment is expected to be
8 slightly older than 30 ka. The thickness of the tephric loess above the AT tephra is 3.2 m (including
9 andosol on top) at the sampled section of LUM-3291 but typically a ~5-6 m thick tephric loess above
10 the AT tephra was observed in the Tanahara-Harataima Terrace (e.g. Kubo, 1997). Thus, a preliminary
11 loess accumulation rate of ~10-20 cm/ka can be calculated. We assume that the fluvial sediment is
12 only up to a few thousand years older than the AT. The pulsed IR₅₀ age is consistent with the expected
13 age, 33.8 ± 2.2 ka; anomalous fading was not detected from the middle part of the pulsed IR₅₀ signal
14 and, therefore, no fading correction was conducted. The pIRIR₂₂₅ and pIRIR₂₉₀ ages are much older;
15 96.2 ± 11.5 ka (101 ± 12 ka, fading corrected) for the pIRIR₂₂₅ and 107 ± 13 ka for the pIRIR₂₉₀ (no fading
16 for this signal; Table 3).

17 18 8. Discussion and conclusions

19
20 We compared the luminescence characteristics of the IR₅₀ (pre-pIR₂₂₅ and pre-pIR₂₉₀), pIRIR₂₂₅, pIRIR₂₉₀
21 and pulsed IR₅₀ signals. The bleachability of the pulsed IR₅₀ signal is comparable to the IR₅₀ (pre-pIR₂₂₅
22 and pre-pIR₂₉₀) signals, but its anomalous fading is as low as for the pIRIR₂₂₅ and pIRIR₂₉₀ signals,
23 especially, when the middle part (4-40s corresponding to the on-time) of the pulsed IR₅₀ decay curve
24 is used for signal integration. The fading correction of the pulsed IR₅₀ age was still necessary for two
25 samples, however, this could be due to the instrumental limitation of the pulsing system used in this
26 study with the maximum on-gate delay of 21.5 μ s. Non fading pulsed IR₅₀ signal may be measured, if a
27 longer on-gate delay (> 100 μ s) is used, as shown in the TR fading test (Fig. 6). The thermal stability
28 data suggest that the pulsed IR₅₀ is thermally more stable than the CW IR₅₀ but less stable than the
29 pIRIR₂₂₅. For our three dating samples, the pulsed IR₅₀ ages are all consistent with independent age
30 control, while the pIRIR₂₂₅ and pIRIR₂₉₀ ages are overestimated. The inaccuracy of the pIRIR ages likely
31 arises due to incomplete bleaching, and additionally, in case of the pIRIR₂₉₀, a poor dose recovery.

32
33 For the pIRIR signals there is a tendency that the signals become more difficult to bleach, while the
34 signal stability increases with increasing preheat and stimulation temperatures. This is presumably
35 because of a combination of thermal transfer and the fact that higher temperature pIRIR signals

originate from more distant electron-hole pairs; it may be inherently more difficult to empty traps, which have distant recombination centres, by athermal fading or by thermal/optical eviction. However, this model does not explain the unique combination of high athermal stability, but lower thermal and optical (bleaching) stability of the pulsed IR₅₀ signal compared to the pIRIR signals. Since thermal stability is both a function of thermal trap depth and electron-hole distance (Jain et al, 2015b), it is expected that higher the thermal stability, the greater the fading stability. Thus, the pulsed IR₅₀ signal should have exhibited the similar thermal stability as the pIRIR₂₂₅ signal, but in fact, the thermal stability of the pulsed IR₅₀ signal was observed to be lower than that of the pIRIR₂₂₅.

While further investigations are required, our data together with the data of Jain et al. (2015a), suggest that the pIRIR signals may be mobilising 'isolated' electrons (infinite tunnelling lifetime), which become available by diffusion and redistribution only at high temperature IR stimulations. Such isolated electrons will be thermally/athermally stable and difficult to bleach, and, thus, add a stable yet unbleachable component to the pIRIR signal. The pulsed IR₅₀ on the other hand may sample a small component of a relatively stable electron population, which is just far enough from nearest holes to give an athermal stability on the geological time scales. Such electrons are likely to be less thermally stable and relatively easier to bleach compared to the 'isolated' electron population. Alternatively, there could possibly be a distribution in the trap depth of the IR trap. In this scenario, there will be a finite population of deep traps that will require a strong thermal assistance to be emptied and will be more difficult-to-bleach because of their higher optical depth; such traps may contaminate the pIRIR signals, but not the pulsed IR₅₀ signal since they will not be available at 50 °C measurement. A distribution in the trap depths is conceivable given the commonly observed disorder in feldspar crystals, however, this model further requires that there is no dependence of fading on the trap depth. Although, we incline towards the model of 'isolated' electrons, further research is required to fully understand the differences in the physical mechanisms behind pulsed IR₅₀ and pIRIR signals.

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Figure captions

Fig. 1: Map showing the locations of sampling sites at (a) Cologne and Monheim, Germany and (b) Sagamihara, Japan. Geomorphological maps were modified from Lauer et al. (2011b) and from Kubo (1997). Insets show the location of the detailed map.

Fig. 2: Graphic sedimentary logs of sampling sites at (a) Cologne, (b) Monheim and (c) Sagamihara.

Fig. 3: (a) 3D time resolved (TR) IRSL curve of MHT-II measured at 50°C for 500 s with 100 μ s on and 400 μ s off, after irradiating 40 Gy and preheating at 250°C for 60s. (b) photon arrival time curve from the same dataset, (c) IRSL decay curve for on-time and off-time from the TR measurement showing in (a). (d) Normalised on- and off-time decay curves. Note that the X-axis of the decay curve was adjusted to the on-time (500 s pulsed IR₅₀ measurement = 100 s on-time).

Fig. 4: Bleaching curves of IR₅₀ (pre-pIR₂₂₅), IR₅₀ (pre-pIR₂₉₀), pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ signals for (a) MHT-II and (b) LUM-3291 for the initial signal (0-4s). The data for MHT-II were also plotted for the different signal integration limits, 0-4s, 4-12s, 12-20s, and 20-40s for (c) IR₅₀ (pre-pIR₂₂₅) and pIRIR₂₂₅ and for (d) pulsed IR₅₀. Note that the integration period for the pulsed IR₅₀ signal is showed on its on-time.

Fig. 5: (a) Anomalous fading of IR₅₀ (pre-pIR₂₂₅), IR₅₀ (pre-pIR₂₉₀), pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ signals for all samples using the initial (0-4s) signal. The g-values of these signals calculated from the different signal integration limits (0-4s, 4-12s, 12-20s and 20-40s) are plotted for (b) SW-II, (c) MHT-II and (d) LUM-3291.

Fig. 6: Fading rate (g_{2days} -value) measured by TR-IRSL for MHT-II. Data are plotted with two different integration limits, 0-4s and 4-12s of the decay curve.

Fig. 7: Thermal stability of the IR₅₀ (pre-pIR₂₂₅), pIRIR₂₂₅ and pulsed IR₅₀ signals for (a) 0-4s signal integration. (b) The same dataset as (a) for four different signal integration limits.

Fig. 8: (a) Dose recovery ratio and (b) measured residual dose after 4 h bleaching with a solar simulator for IR₅₀ (pre-pIR₂₂₅), IR₅₀ (pre-pIR₂₉₀), pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ signals.

- 1 Fig. 9: Comparison of the pIRIR₂₂₅, pIRIR₂₉₀, pulsed IR₅₀ ages and with independent age control. The
- 2 luminescence ages were plotted against the mean quartz ages for SW-II and MHT-II and against the
- 3 tephra (AT) age for LUM-3291.

Table 1: Age control information for the studied samples.

Site	Sample code	Description of age control	Age control		Reference	
			Radiocarbon			Other
			¹⁴ C BP	Calibrated (cal BP)*		
Cologne subway	SW-II	quartz OSL of the same sample			3460±350 (mean), 1980±330 (MAM3)	Lauer et al. (2011a)
	SW-I	quartz OSL, 5 m west of SW-II			2220±170 (mean post-IR OSL)	Lauer et al. (2011a)
	RC-1	¹⁴ C of charcoal, above SW-II	2010±55	1832-1842 (1%), 1864-2119 (99 %)		Lauer et al. (2011a)
	RC-2	¹⁴ C of charcoal, below SW-II	2080±85	1876-2211 (88%), 2221-2308 (12%)		Lauer et al. (2011a)
	RC-3	¹⁴ C of charcoal, below SW-II	2025±60	1865-2145 (100%)		Lauer et al. (2011a)
Monheim	MHT-II	quartz OSL of the same sample			11.6 ± 0.7 ka	Lauer et al. (2011b)
	MHT-I	quartz OSL, 4 m below			11.7 ± 1.0 ka	Lauer et al. (2011b)
	MHT-III	quartz OSL, 1.5 m above			11.6 ± 1.3 ka	Lauer et al. (2011b)
	HYM-13a	¹⁴ C of macrofossil from Holzmaar at the position of Laacher See tephra	11210±95	12830-13266 (100%)		Hadjas et al. (1995)
	HYM-13b	¹⁴ C of macrofossil from Holzmaar at the position of Laacher See tephra	11380±95	13071-13413 (100%)		Hadjas et al. (1995)
		Ar/Ar of sanidine from Laacher See Tephra			12.90 ± 0.56 ka	van den Vogaard (1995)
Sagamihara		¹⁴ C of charcoal and humic acid for Aira Tanzawa (AT) tephra, 40 cm above	25,120 ± 270 BP	28577 - 29831 (100%)		Miyairi et al. (2004)
		Varve chronology combined with macrofossil ¹⁴ C in Lake Suigetsu, 15 cm above		30,009±189		Smith et al. (2013)

* All radiocarbon ages are recalibrated using Intcal 13.

Table 2: pIRIR₂₂₅, pIRIR₂₉₀ and pulsed IR₅₀ protocols, used in this study.

a pIRIR ₂₂₅			b pIRIR ₂₉₀			c pulsed IR ₅₀		
Step	Treatment	Observed	Step	Treatment	Observed	Step	Treatment	Observed
1	Dose		1	Dose		1	Dose	
2	Preheat 250°C 60s		2	Preheat 320°C 60s		2	Preheat 250°C 60s	
3	IRSL 50°C 100s	Lx	3	IRSL 50°C 100s		3	Pulsed IRSL 50°C for 500s*	Lx
4	pIRIR 225°C 200s	Lx	4	pIRIR 290°C 200s	Lx	4	Test Dose	
5	Test Dose		5	Test Dose		5	Preheat 250°C 60s	
6	Preheat 250°C 60s		6	Preheat 320°C 60s		6	Pulsed IRSL 50°C for 500s*	Tx
7	IRSL 50°C 100s	Tx	7	IRSL 50°C 100s		7	Back to 1	
8	pIRIR 225°C 200s	Tx	8	pIRIR 290°C 200s	Tx			
9	Back to 1			Back to 1				

* 100 µs on and 400 µs off (total on time: 100s)

Table 3: Dose rate, D_e values, fading rate and age of the samples.

Sample	Dose rate (Gy/ka)	Signal	D_e (Gy)	g-value (%/decade)	Fading uncorrected age (ka)	Fading corrected age (ka)
SW-II	2.81 $\pm$ 0.13	pIRIR ₂₂₅	16.5 $\pm$ 0.7	2.1 $\pm$ 0.3	5.9 $\pm$ 0.4	6.9 $\pm$ 0.4
		pIRIR ₂₉₀	30.8 $\pm$ 1.1	1.8 $\pm$ 0.1	11.0 $\pm$ 0.7	12.8 $\pm$ 0.8
		pulsed IR ₅₀	9.9 $\pm$ 1.0	0.6 $\pm$ 0.3	3.5 $\pm$ 0.4	3.7 $\pm$ 0.4
MHT-II	2.88 $\pm$ 0.13	pIRIR ₂₂₅	40.3 $\pm$ 1.4	2.2 $\pm$ 0.3	14.0 $\pm$ 0.8	17.3 $\pm$ 1.1
		pIRIR ₂₉₀	57.3 $\pm$ 1.4	1.7 $\pm$ 0.5	19.9 $\pm$ 1.1	23.3 $\pm$ 1.7
		pulsed IR ₅₀	27.9 $\pm$ 0.9	1.6 $\pm$ 0.3	9.7 $\pm$ 0.6	11.1 $\pm$ 0.7
LUM-3291	1.52 $\pm$ 0.12	pIRIR ₂₂₅	147 $\pm$ 13	0.5 $\pm$ 0.3	96.2 $\pm$ 11.5	101 $\pm$ 12
		pIRIR ₂₉₀	164 $\pm$ 14	0.05 $\pm$ 0.5	107 $\pm$ 13	n.a.
		pulsed IR ₅₀	51.5 $\pm$ 2.4	-0.8 $\pm$ 0.5	33.8 $\pm$ 2.2	n.a.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 4 (continued)

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9