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1 **Non-Poisson variations in photomultipliers and implications for** 2 **luminescence dating**

3
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12 **Abstract**

13 Previous studies have suggested that excess variations from single-photon counting systems
14 used in luminescence dating may result in underestimation of errors and profoundly influence
15 age models. In this study ten different photon counting systems have been investigated to
16 explore this effect with a greater number of photomultiplier types and instrumental
17 architectures. It is shown that radiation induced phosphorescence from F1 feldspar produces a
18 controllable low-level light source whose local variance approximates Poisson expectations.
19 However excess variation in dark counts was observed to varying extents from all systems.
20 The excess variance is slightly anti-correlated with the age of the system, with older devices
21 conforming more closely to Poisson behaviour. This observation does not seem to fit the
22 hypothesis that enhanced levels of helium diffused into older tubes increase non-Poisson
23 components. It was noted that a significant part of the non-Poisson behaviour was associated
24 with multi-event pulse streams within time series. Work was also undertaken to develop
25 mitigation methods for data analysis and to examine the implications for dating uncertainties
26 in a test case. A Poisson-filtering algorithm was developed to identify and remove
27 improbable multi-event streams. Application to data from signal-limited single grains of
28 sediments from a Neolithic chambered tomb in Corsica has shown that, for this case,
29 removing non-Poisson components improves the robustness of retained data, but has less

30 influence on overall dating precision or accuracy. In signal limited applications use of this
31 algorithm to remove one source of excess variation is beneficial. The algorithm and test data
32 are appended to facilitate this.

33

34 **Keywords:** Single photon counting; Poisson variations; Phosphorescence; OSL single grain
35 dating; Poisson filtering

36

37 **Highlights**

- 38 • New investigation of behaviour of 10 diverse photon counting systems
- 39 • Low level phosphorescence close to Poisson counting statistics
- 40 • Dark counts show non-Poisson counting statistics
- 41 • New Poisson filter algorithm applied to low-sensitivity single grain data
- 42 • Non-Poisson dark counts affect rejection criteria but not overall dating errors

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1. Introduction

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Luminescence methods measure the number of photons emitted from a sample under stimulation, and use single photon counting photo-multiplier tubes (PMTs). The measurement backgrounds include an intrinsic detector background from the PMT in the absence of any light sources, the dark count. Generally low dark count PMTs are selected for use in luminescence instruments. A recent study (Adamiec et al., 2012) characterised the behaviour of luminescence dating systems using the EMI QA 9235 photomultiplier under low light level conditions. Three of the four systems studied showed variations in dark count in excess of the expected Poisson distribution for non-correlated random events. It was suggested that this could result in an underestimation of measurement uncertainties and have implications for age models.

The study reported here was devised to characterise a larger number and range of PMT types and instrumental architectures to produce a significant body of new data that could help provide a broader understanding of the behaviour. The implications for the accuracy and uncertainty of luminescence measurements were also studied using a case study where low levels of luminescence signal reached critical levels, and using the system within the Scottish Universities Environmental Research Centre (SUERC) dating lab which showed the greatest level of excess dark count variation. This involved single grain Optically Stimulated Luminescence (OSL) analysis of sediments from the constructional layers associated with a Neolithic chambered tomb in Corsica. The technical part of the study included development of an algorithm to identify non-Poisson behaviour associated with multi-event bursts in luminescence measurements and remove their associated artefacts by interpolation. The use of this algorithm on a set of low intensity single grain luminescence measurements allows an

70 assessment of the impact of non-Poisson dark counts on measurement accuracy and
71 precision.
72

73 Dark counts are ‘false’ photon counts induced by various mechanisms including thermal
74 emission of electrons from the photo cathode and first dynode, cosmic ray interactions,
75 ambient radiation in the environment and potentially electrical interference. Signal counts
76 from single photo-electron emission at the photocathode coupled with electron multiplication
77 and pulse height selection form the basis of photon counting. If these are uncorrelated
78 random events they should follow a Poisson distribution described by the equation, $P(\lambda) =$
79 $e^{-\bar{n}} \frac{\bar{n}^\lambda}{\lambda!}$, where $P(\lambda)$ is the probability of observing λ events in a given interval, \bar{n} is the
80 average number of events per interval and λ is the number of events (Poisson 1837, Stigler et
81 al., 1982). A behavioural characteristic of the Poisson distribution is that the standard
82 deviation of the distribution, σ , is equal to the square root of the number of observations $\sqrt{\bar{n}}$
83 within a given time interval. For this study n is the number of photon counts per detection
84 channel, as is the case in routine luminescence measurements. Deviations from a Poisson
85 distribution would indicate correlated or anticorrelated components in the counting data. .
86

87 Recent work studying the behaviour of a small number of dating instruments (Adamiec et al.,
88 2012) under dark conditions and varying light levels has shown that under illumination the
89 observed behaviour followed Poisson statistics, however excess variance was seen in some
90 devices under dark conditions. This work introduced a parameter denoted k , which was the
91 ratio of the observed standard deviation to the expected standard deviation based on Poisson
92 statistics. Three Risø readers using the EMI QA 9235 PMT showed dark-count k values in
93 excess of unity, indicating non-Poisson behaviour, with an older Daybreak system using the
94 same PMT showing a dark-count k value near unity, once the prescaling factor was

95 considered. It is unclear from this study whether the observed non-Poisson behaviour is
96 common in different PMT types and ages, nor what causes this behaviour. Further work
97 elaborating the methods of estimating the equivalent dose and their uncertainties when non-
98 Poisson variances are present is given by Bluszcz et al., 2015.

99

100 It has been suggested that afterpulses generated by electron interactions with gases inside the
101 photomultiplier may explain the observed excess variance. Such after pulses are described in
102 detail by Morton et al., 1967, showing that in 8575 photomultipliers under low light
103 conditions approximately 5% of pulses have an associated afterpulse $\sim 0.3 \mu\text{s}$ after the main
104 pulse. This was further explored by Coates (1973) using 8850 and 8852 photomultipliers,
105 noting that the afterpulse time distribution enables ions of different masses to be separated
106 (with the PMT acting like a crude time of flight (TOF) mass spectrometer) hence allowing
107 the physical nature of afterpulses to be determined. Coates confirmed that the principle
108 features of the TOF spectrum of the afterpulses was consistent with helium in the
109 photomultipliers. Finite-difference Monte Carlo modelling of afterpulses in the 9235QA tube
110 with a partial pressure of helium matching atmosphere, by Tudyka et al 2016, produced
111 results concordant with the observations of an old 9235QA tube, where it was assumed
112 helium diffusion had had sufficient time to equilibrate with atmosphere (Adamiec et al.,
113 2012), and suggested that helium diffusion into PMTs may be a factor in the excess variance
114 observed with such systems.

115

116 To address the issues raised by the earlier studies, further work into non-Poisson variation for
117 a series of photon detection systems varying in age, tube type and electronic configuration,
118 used for luminescence dating and the detection of irradiated food, has been conducted. In the
119 previous work (Adamiec et al 2012) the oldest device appeared to conform more closely to

120 Poisson behaviour. The varying ages in this investigation could explore whether there is a
121 correlation with system age, in particular to explore the helium diffusion hypothesis since it is
122 expected that helium concentrations, and hence afterpulse frequency, within the tubes should
123 increase with age. To facilitate comparison with previous work, the approach of Adamiec et
124 al., 2012 to characterise non-Poisson variations using the “k” value is adopted here, although
125 it is recognised that other parameters may also be beneficial. This larger study also provides a
126 substantial data set that may be used to investigate the causes of the non-Poisson behaviour,
127 the extent to which this behaviour may influence the accuracy and precision of luminescence
128 measurements, and approaches to mitigate these effects. This has included the use of
129 phosphorescence as a low level light source, and the development of an algorithm which may
130 be used to identify non-Poisson behaviour in measurements and adjust the data to remove
131 these effects.

132

133

134 **2. Investigation**

135

136 The environmental physics group at SUERC have a large number of photon counting systems
137 that utilise different photomultipliers and architectures in different instruments. To
138 characterise the devices, measurements were conducted with luminescence and under dark
139 conditions. In this study, ten of these systems used for luminescence dating and the detection
140 of irradiated foods (Sanderson et al., 1989) have been investigated, developed between 1986
141 and 2015. These systems are:

- 142 • Two manual TL readers developed in 1986 and 1989 (Sanderson et al., 1989, Spencer
143 et al., 1994), designated SUERC TL readers TL1 and TL3. These use selected low
144 dark count photomultipliers with 2” bi-alkali photocathodes and fourteen stage linear

145 focused dynodes (type D295QA for TL Reader 1 and 9883QB for SUERC TL Reader
146 3).

147 • Two systems developed for PSL screening of food, here designated PSL 1 and PSL2.
148 PSL1 (SURRC PPSL system serial number 8) uses a 9829B PMT with a 2” bi-alkali
149 cathode, selected for low background rate, and uses the SURRC PPSL 1 board with a
150 pre-amplifier/discriminator integral ETL device (Sanderson et al., 1989). PSL2
151 (SUERC PPSL system serial number 93) has a 9814B also a 2” bi-alkali cathode,
152 again selected for low background rate, and uses the SUERC PPSL 2 control board
153 with a PIC 18 microcontroller USB2 communication to Windows (Sanderson et al.,
154 2003).

155 • Two OSL Portable readers using photo detector modules incorporating selected
156 9124B tubes with 1” photocathodes and built in HV and amplifier- discriminator
157 systems. Both use the SUERC PPSL 2 board (Sanderson & Murphy., 2010) for
158 synchronised luminescence stimulation and photon counting.

159 • Two OSL scanning imaging instruments (OSL1 and OSL PICS) built for the
160 detection of irradiated foods (Sanderson et al., 2001) and with selected 9883QB
161 (OSL1) and 9883B (OSL PICS) PMTs and EMI C604 amplifier discriminators
162 connected via ECL-TTL converters to the SURRC PPSL 1A photon counting board
163 with 24-bit 100MHz bandwidth photon counters.

164 • Two Risø readers, using the 9235QA (Risø 1) and 9235QB (Risø 3) PMTs (the
165 modern linear focussed version of the old 9635 scintillation counting venetian blind
166 dynode EMI Tube originally used in Oxford for photon counting) with proprietary
167 HV amplifier discriminator electronics with very small amplitude pulse to preserve
168 amplitude (Bøtter-Jensen et al., 2000, Bøtter-Jensen et al., 2010).

169

170 **2.1 Phosphorescence as a low level light source**

171

172 The first issue was to find a suitable random light source. Low level beta lights (such as ^{14}C)
173 may not be random due to the presence of correlated photons. The study by Adamiec et al.
174 (2012) used light emitting diodes (LEDs), however these may also be affected by non-
175 random variables related to maintaining the LED at a steady state for a prolonged period of
176 time. Other studies have used incandescent light sources with pin hole apertures to limit
177 photon emission rates into the experimental system, which may be affected by similar
178 variables as LEDs. In this work the use of phosphorescence as a low level random light
179 source was investigated. The potential advantages are that this is a light source which can be
180 readily achieved within luminescence instrumentation without incorporating additional
181 systems, and in thermoluminescence (TL) instruments control of the temperature can be used
182 to adjust the phosphorescence decay rate. In addition, the predictable decay of the light
183 source allows evaluation of PMT performance under different light conditions within a single
184 experiment. Phosphorescence was achieved by irradiating an International Atomic Energy
185 Agency (IAEA) F1 feldspar sample with a ^{90}Sr beta source, and initial investigations
186 confirmed that once irradiated and stored the phosphorescence tail could be used as a slowly
187 decaying source controllable to produce approximately 100-200 counts per second.

188

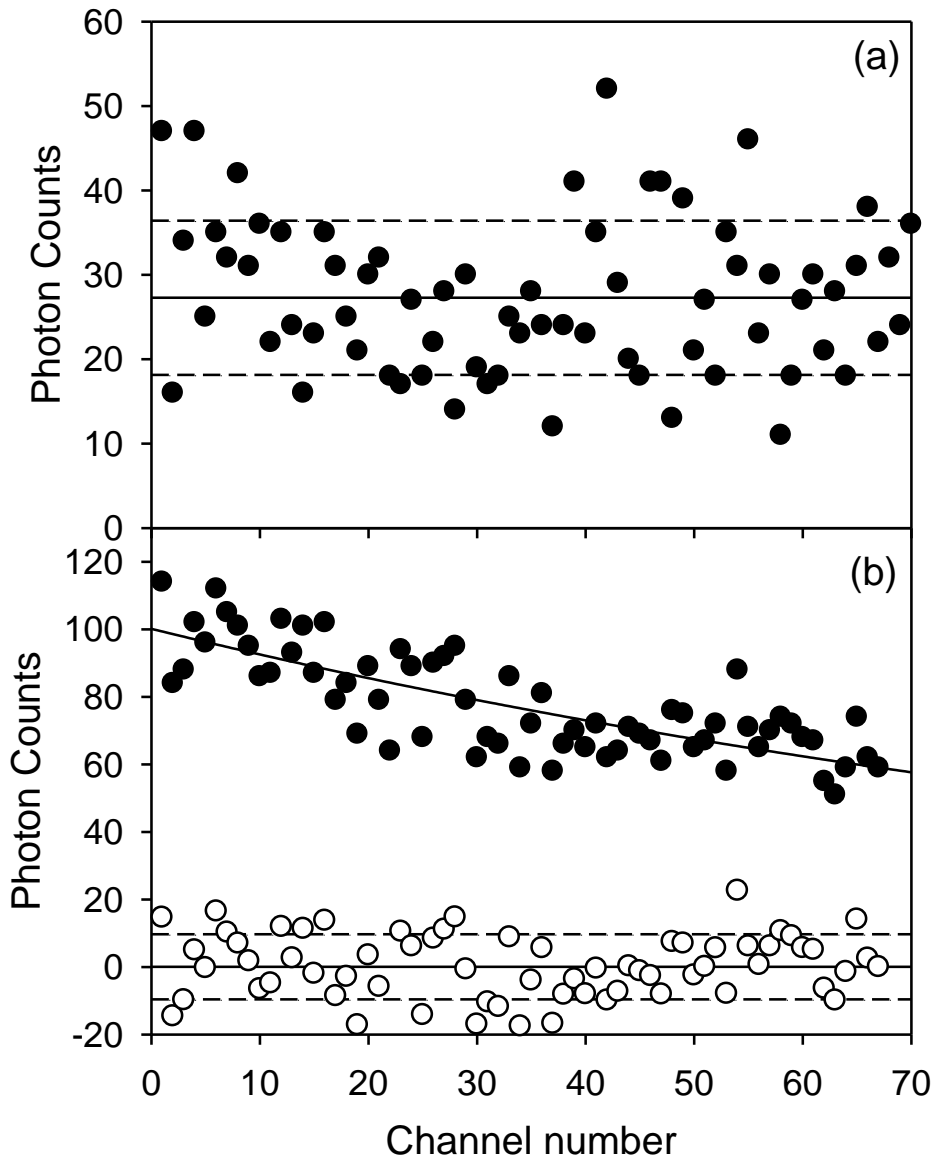
189 As a decaying source, phosphorescence results in changing light levels with varying mean
190 count rates coupled with random counting variations. However by fitting decay curves and
191 examining residuals in conjunction with local decay rates the random variation components
192 as a function of light level can be estimated. In this work phosphorescence decays were fitted
193 by single exponentials and residuals calculated by subtracting the calculated from the
194 measured counts, as illustrated in Fig.1. The standard deviation of the residuals was taken as

195 the observed error of the system, at the corresponding intensity of the signal. The approach is
196 similar to that taken by Adamiec et.al. 2012, where a second order polynomial function was
197 used to de-trend light source measurements, with the variance on the residuals used as the
198 measured variance. To assess the extent to which this approximation to the statistical
199 behaviour of varying light sources can be relied on, a single phosphorescence measurement
200 (from the SUERC Portable OSL Reader) was divided into shorter time intervals, each
201 corresponding to a 1% decay in phosphorescence as determined from the fitted exponential
202 function, with the k-value calculated for each interval using the standard deviation on the
203 measured photon counts rather than the residuals. The k-values calculated for each data
204 segment are plotted in Fig 2, with the k-value calculated from the residuals for the entire
205 measurement for reference. It can be seen that the segmented values scatter around the value
206 for the entire measurement, with high values corresponding to significant excess counts at
207 approximately 100 and 450s. The mean of the k-values for all segments (1.111 ± 0.002)
208 compares favourably for the k-value from the entire measurement (1.117 ± 0.002). Rejecting
209 the segments with excess counts brings the value of k closer to unity (1.03 ± 0.001).

210

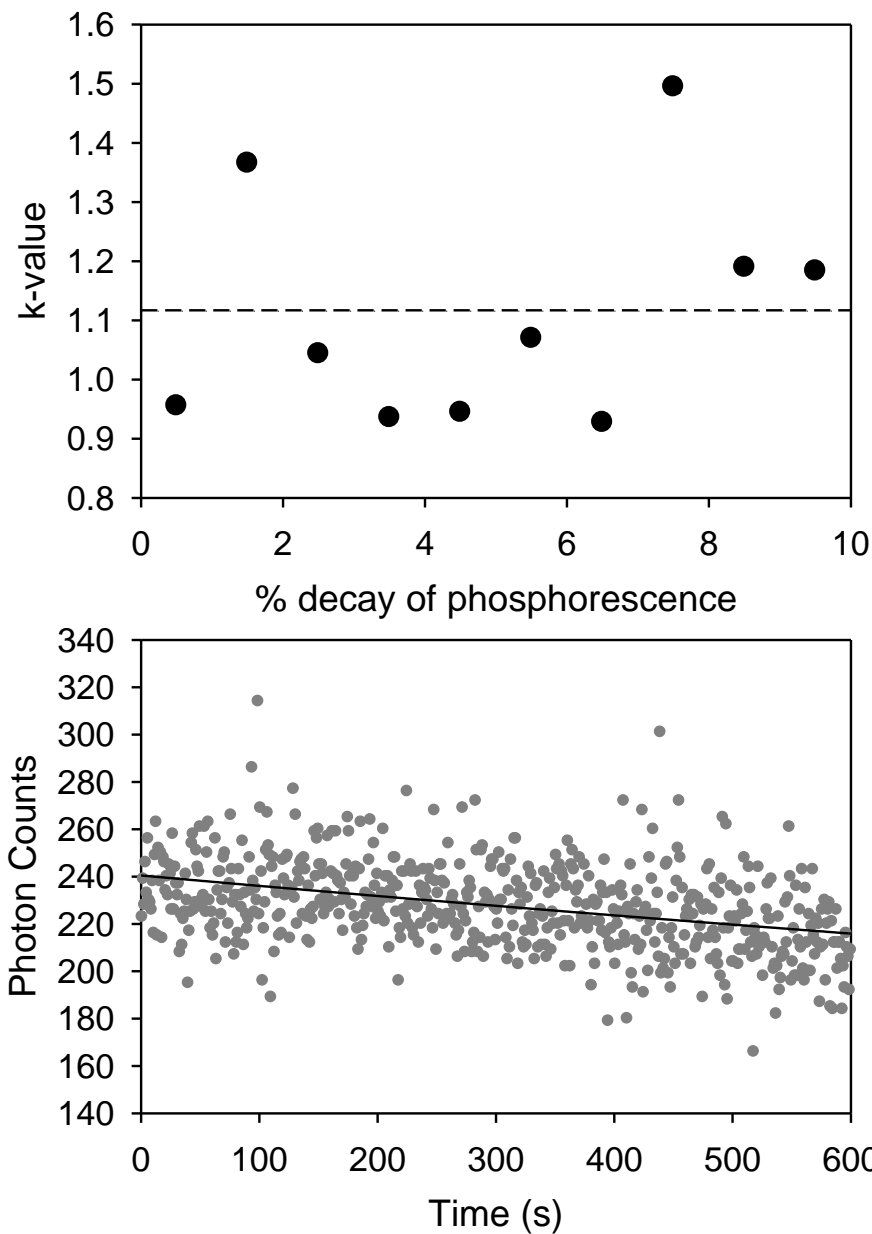
211 The data in Fig.1 has been selected as having a relatively rapid signal decay, approximately
212 25% over the measurement (from ~100 to ~75 cps), so that the curve fitting could be
213 observed. Other measurements showed much slower decays, approximately 10% over 600s.
214 Thus, it can be seen that it is possible to generate low-level phosphorescence light sources
215 which decay slowly, in order to study residual variations under controlled conditions from
216 simple and highly predictable sources of low level light.

217



219 **Fig 1.** Example selected interval spectra for the SUERC Manual TL Reader. a) dark count,
 220 showing mean and standard deviation, and b) phosphorescence, showing measured data
 221 (filled circles) and fitted exponential decay and residuals (open circles) with mean and
 222 standard deviation.

223



224 **Fig. 2.** k-values calculated for segments corresponding to 1% of the phosphorescence decay
 225 measured on OSL Portable 1, with the k-value for the entire measurement indicated by the
 226 dashed line. Larger k-values correspond to points in the measurement with higher counts
 227 (lower plot).

228

229

230 2.2 Investigations of PMT response

231

232 For this investigation multiple measurements of 600 seconds were performed under
233 phosphorescence and dark conditions for each device. Examples of low light level with the
234 fitted exponential decay and dark count spectra are shown in Fig. 1. Following the
235 characterisation process of Adamiec et al., 2012, each device was characterised a k-value
236 defined as $k = \sigma/\sqrt{\bar{N}}$, the ratio of the observed standard deviation (σ) from a series of
237 measurements and the standard deviation that is expected from Poisson statistics ($\sqrt{\bar{N}}$).
238 Adamiec et al., 2012 plot k-values with uncertainties, but the paper does not state how these
239 uncertainties were estimated. Here we estimate these uncertainties by taking the uncertainty
240 on \bar{N} to be the standard error (σ/\sqrt{n}) where n is the number of measurements in the data set,
241 and approximate the fractional uncertainty on k to half the fractional uncertainty on \bar{N} . Thus

242

$$\Delta k \approx k(0.5 (\sigma/\sqrt{n})/\bar{N})$$

243 It is noted that the uncertainties on k thus estimated are similar to those for k-values plotted
244 by Adamiec et al 2012.

245

246 For an ideal detection system and random light source Poisson statistics are expected leading
247 on average to k-values of 1. Values significantly different than one indicate non-random
248 variations, with values greater than one corresponding to excess variation..

249

250 Dark counts were measured on all devices, by running the closed systems without light
251 sources, and k-values calculated using the standard deviation and the mean counts for the
252 measurements. Then phosphorescence measurements were conducted at low levels and k-
253 values calculated using the standard deviation on the residuals and the mean counts for the
254 measurements.

255 **3. Results of PMT response investigations**

256

257 Full k parameter results under phosphorescence and dark conditions for all devices are shown
258 in Table 1 with their corresponding development ages, tube types and mean dark count rates.

259

260 Under low light conditions all devices have k values close to unity, and thus show the
261 expected Poisson behaviour. Under dark conditions, k values with the possible exception of
262 the oldest unit, are greater than unity indicating excess variance compared with Poisson
263 statistics for all photomultipliers to varying extents. These results corroborate and extend the
264 findings of Adamiec et al 2012 and indicating that similar phenomena can be observed across
265 a wider range of PMT types and architectures.

| Photomultiplier | System | PM Type | Dark count measurements | | | Phosphorescence measurements | |
|-------------------|-------------|---------|-------------------------------------|---|---------------------|-------------------------------------|------------------|
| | Development | | Number of Measurements (600s) | Mean Count Rate (cps) \pm std dev | $k_{DC} \pm \sigma$ | Number of Measurements (600s) | $k_p \pm \sigma$ |
| | Age | | | | | | |
| SUERC Manual TL 1 | 1986 | D295QA | 7 | 53 ± 10 | 1.37 ± 0.01 | 3 | 0.89 ± 0.01 |
| Reader TL 3 | 1989 | 9883QB | 5 | 28 ± 9 | 1.70 ± 0.01 | 3 | 1.00 ± 0.01 |
| PSL 1 | 1989 | 9829B | 5 | 17 ± 7 | 1.70 ± 0.01 | 1 | 1.07 ± 0.06 |
| PSL 2 | 1989 | 9814B | 5 | 25 ± 11 | 2.20 ± 0.02 | 1 | 0.97 ± 0.03 |
| OSL | 2001 | 9883QB | 6 | 27 ± 10 | 1.92 ± 0.02 | 2 | 0.92 ± 0.09 |
| OSL PICS | 2001 | 9883B | 7 | 23 ± 10 | 2.08 ± 0.01 | 2 | 0.91 ± 0.08 |
| OSL Portable 1 | 2010 | 9124B | 8 | 8 ± 8 | 2.82 ± 0.06 | 2 | 0.82 ± 0.03 |
| OSL Portable 2 | 2015 | 9124B | 8 | 10 ± 4 | 1.26 ± 0.01 | 2 | 0.89 ± 0.05 |
| Risø 1 | 1999 | 9235QA | 7 | 65 ± 18 | 2.23 ± 0.01 | 1 | 1.16 ± 0.07 |
| Risø 3 | 2008 | 9235QB | 10 | 49 ± 23 | 3.28 ± 0.03 | 1 | 1.01 ± 0.06 |

266

Table 1.

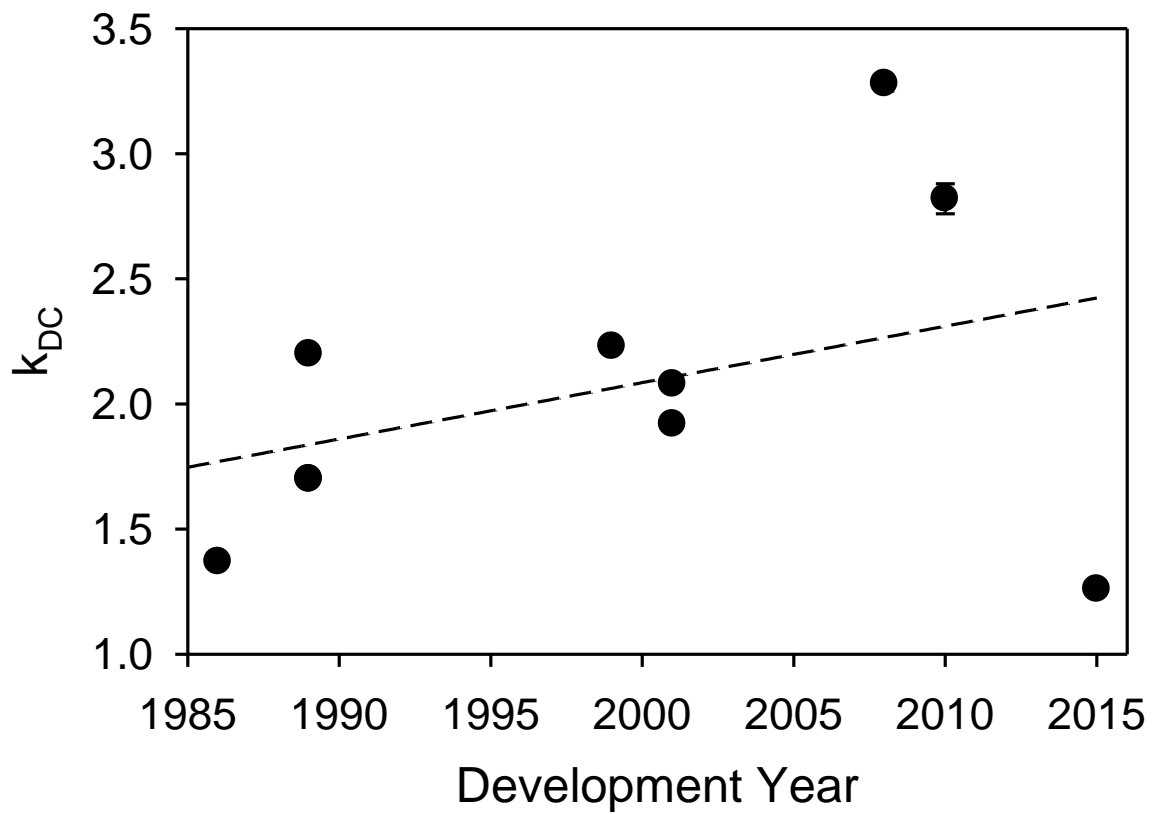
267

Summary of results for all PMT's, showing mean dark count rates, and k-values for the phosphorescence and dark count measurements.

268 Fig 3 plots the k_{DC} parameter against the year each device was produced and suggests a slight
269 correlation (younger devices showing a greater extent of non-Poisson behaviour). It has been
270 suggested that afterpulse rates associated with helium which had diffused into the PMT may
271 be responsible for the non-Poisson behaviour. In this case it might be expected that under
272 comparable diffusion rates the oldest systems should contain more helium than the younger
273 ones. Since the afterpulse intensity is a function of helium concentration then older systems
274 should be more susceptible to non-Poisson behaviour. The data do not support this
275 hypothesis. Nor is there a simple relationship between the use of quartz windows or glass
276 windows, which are expected to show different helium diffusion rates and the excess
277 variance observed. Both these observations suggest that other factors than helium
278 concentration are likely to be involved in the excess dark count variations observed.

279

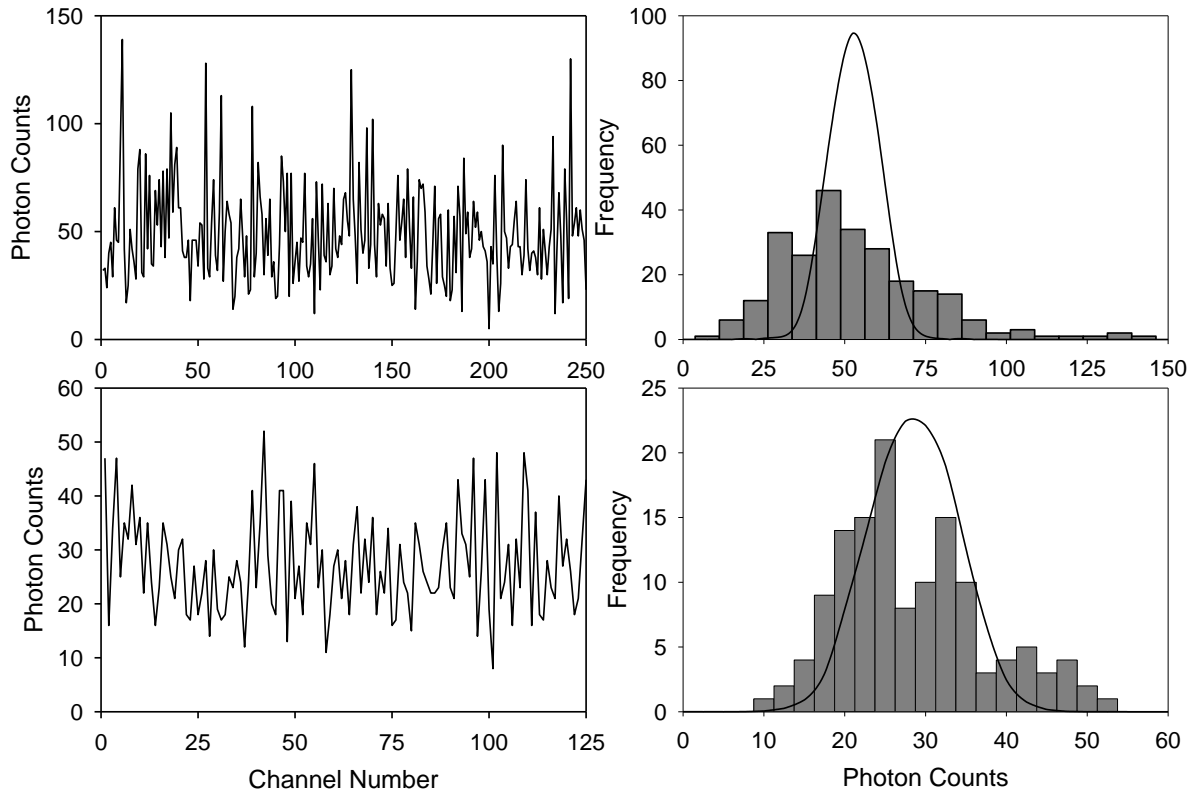
280 Fig 4 presents a comparison between spectra recorded under dark conditions for the device
281 most closely following Poisson statistics (SUERC manual TL Reader) and the least well
282 behaved device (Risø 3). The Risø 3 spectrum shows high single channel bursts, with
283 correspondingly long tail to high photon counts, above 100, in the histogram. The SUERC
284 manual TL reader has a lower dark count and does not include single channel bursts of
285 comparable amplitude. The non-Poisson component in the Risø 3 system is largely associated
286 with these single channels with photon counts significantly in excess of Poisson expectations.



287

288 **Fig 3.** Age correlation graph; readers from left to right (SUERC Manual TL Reader, TL
 289 Reader 2, PSL 1, PSL 2, OSL, OSL PICS, OSL Portable 1, OSL Portable 2, Risø 1, Risø 3)
 290 measured k_{DC} parameter plotted against the development year of the devices, with a
 291 correlation line (dashed-line plotted).

292



293

294 **Fig 4.** Comparison of the dark count spectra between the systems showing the most and least
 295 excess variance (Risø 3, top, and SUERC Manual TL Reader, bottom). Solid line is the fitted
 296 Poisson probability distribution of the counts detected.

297

298

299 **4. Poisson Smoothing**

300

301 **4.1 Development of algorithm**

302

303 Although the cause of excess variance in some systems is still unclear, it is noted that the
 304 excess variances at or close to dark count largely manifest as single channel spikes with
 305 counts significantly in excess of the expectations from Poisson statistics. This leads to the
 306 possibility of an algorithm to identify and remove these spikes, and hence reduce the excess

307 variation. A Poisson filter algorithm (illustrated in Fig. 5, with the script and test data in
308 Supplementary Information) has been written which calculates the probability of the counts
309 in a given channel falling within a Poisson distribution defined by the mean and standard
310 deviation of the spectrum. Any isolated channel counts with a Poisson probability below an
311 acceptable value (which can be input by the user) are averaged out with four neighbouring
312 channels, thus smoothing out the counts that are single channel bursts not following Poisson
313 statistics. Multiple adjacent channels below the acceptance criteria, which would include
314 signals from mineral grains, are not affected. In Figure 6 the application is implemented on
315 the Risø 3 dark count spectra, the counts that are out with Poisson criteria are removed and
316 averaged. In this case this has reduced the k_{DC} parameter for from 3.21 ± 0.05 to 2.38 ± 0.03 ,
317 thus the filter has removed some of the excess variation.

318

319

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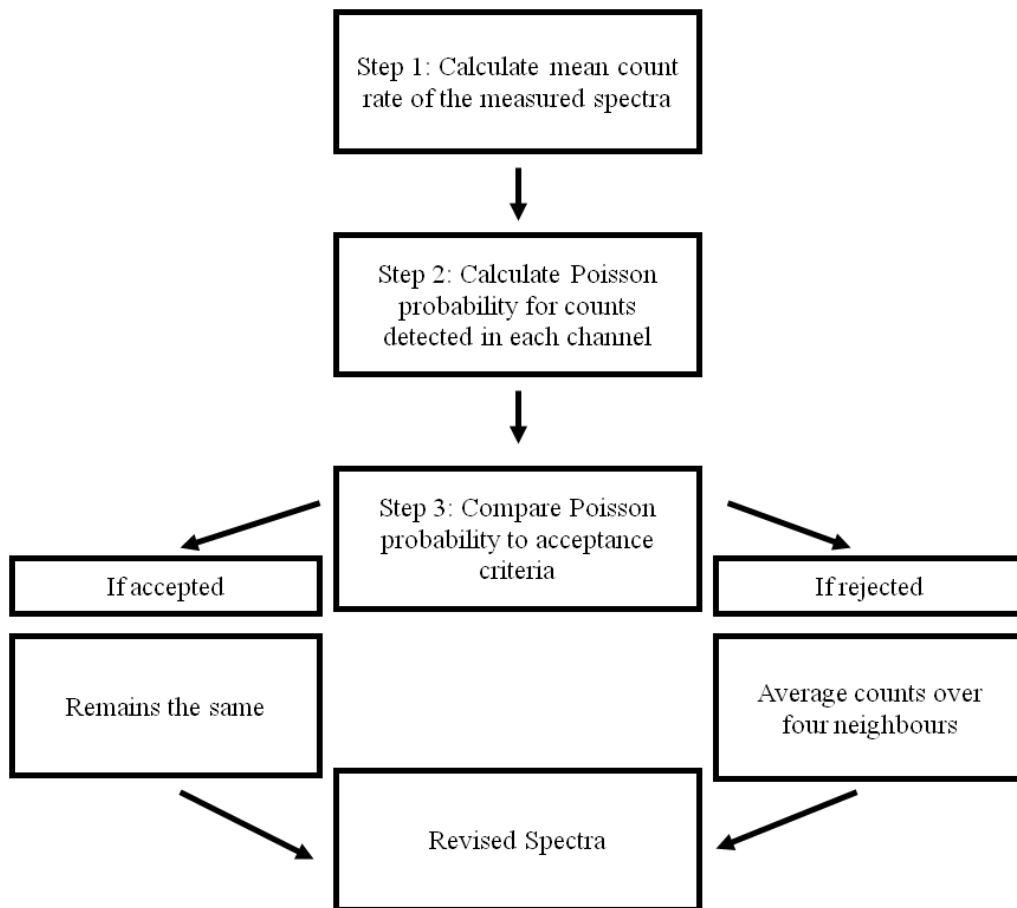
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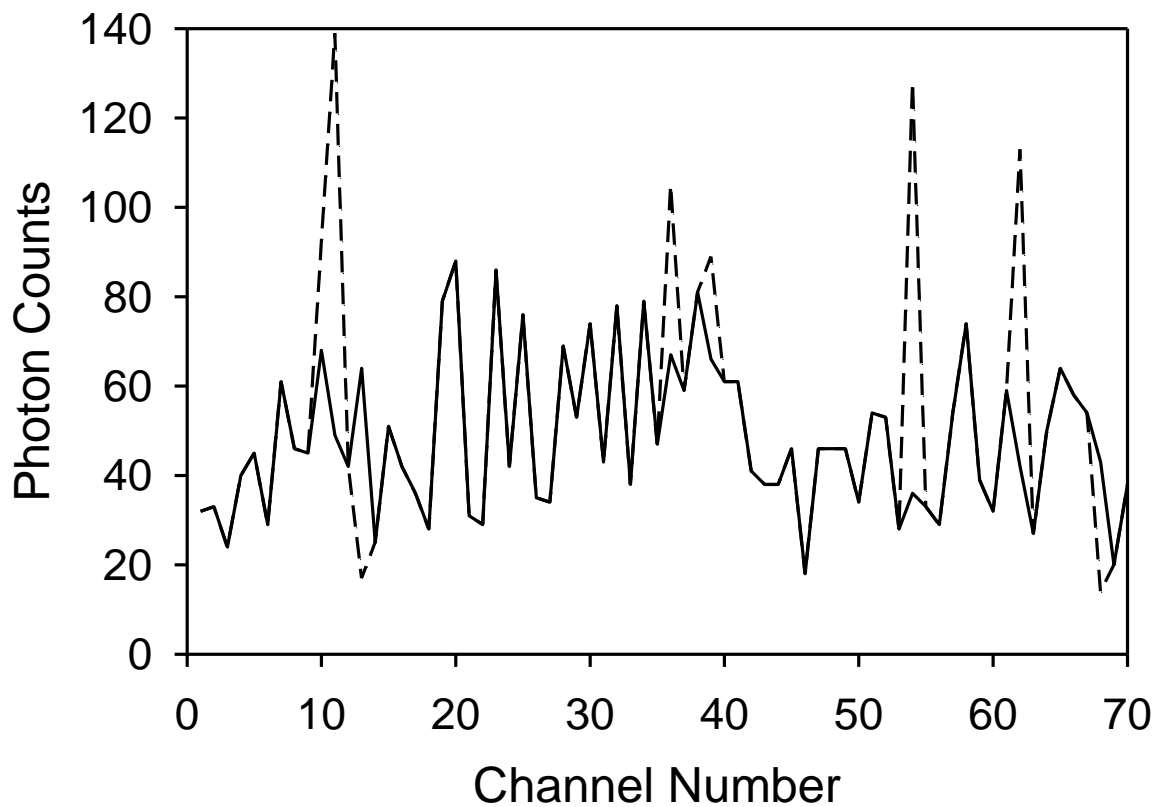
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326

327 **Fig 5.** Flow chart of the Poisson smoothing algorithm



328

329 **Fig 6.** Original (dashed) and revised (solid) spectra implementing Poisson smoothing. The
 330 large single channel noise bursts have been removed and averaged.

331

332 **4.2. Application Case Study: Neolithic burial chamber Corsica, France**

333

334 The Poisson filter has been applied to a test set of single grain measurements. These were
 335 taken from a study of Neolithic burial chamber on Corsica (Sanderson et al., 2014, Cresswell
 336 et al., 2016). The Capu di Locu project collected 92 small samples from 10 sequences, 6
 337 associated with a menhir standing stone (Stantare) and 4 associated with a chambered tomb
 338 (Tola). Field and laboratory profiling analysis was used to target undisturbed sedimentary
 339 units with potential to date the primary construction of these Neolithic monuments. Nine
 340 samples, five from Stantare and four from Tola, were collected for OSL dating. These

341 showed low quartz OSL sensitivities of 200 - 800 integrated counts/mg/Gy for the Tola
342 samples, with higher sensitivities of 2000 - 20000 integrated counts/mg/Gy for the Stantare
343 samples. There was evidence from SAR analysis of multi-age components in the samples
344 collected from the lower fill around the Stantare menhir, and two of these samples were
345 carried forward for single grain analysis; SUTL2683 with 18 single grain disks and
346 SUTL2680L with 7 disks, in both cases using 250-500 μm quartz. A sample from the Tola
347 site (SUTL2960B), which did not show evidence for multi-age components in the SAR
348 analysis, was used as a control sample with 7 disks used for 150-250 μm and 7 disks of 250-
349 500 μm quartz grains. The single grain analyses this comprised SAR analysis of 3900 SG
350 holes each producing a series of OSL decay curves.

351

352 This case study here used measurements from the low sensitivity control sample set of seven
353 single-grain discs populated with 250-500 μm quartz grains from a thin horizon below the
354 principal slab of the Tola chambered tomb (sample SUTL2960B). Measurements were
355 conducted on a Risø DA-20 automatic reader designed for single grain luminescence dating
356 (Risø 3, which has been observed to have the largest k value in this study, Table 1). Each
357 single-grain measurement consisted of four OSL measurements; the natural and a 25 Gy
358 regenerative dose, with 5 Gy test doses. Following the 5 Gy test dose, 60% of measurements
359 produced less than 10 counts, with 37% giving 10-100 counts and only 3% giving 100-1000
360 counts. Acceptance criteria were based on the statistical significance of the net counts from
361 the regenerative dose compared with their estimated error-in signal uncertainty. Of the 700
362 measurements, examination under an optical microscope showed approximately half were
363 from unoccupied holes in the single grain discs, and 88 carried statistically significant
364 signals, when compared with a rejection threshold (expressed as number of standard
365 deviations) based on the estimated uncertainties of their net counts after late-light subtraction.

366 The estimated uncertainties in this process are based on Poisson expectations. This data set
367 was chosen because the minerals had relatively low sensitivities, and hence the signals
368 observed were small and a high proportion of observations fell below 2 sigma significance
369 levels and were rejected on the basis of Poisson criteria from the conventional analysis. It
370 was considered that a case of this type would be most sensitive to non-Poisson variation in
371 comparison with dating data sets with higher signal levels.

372

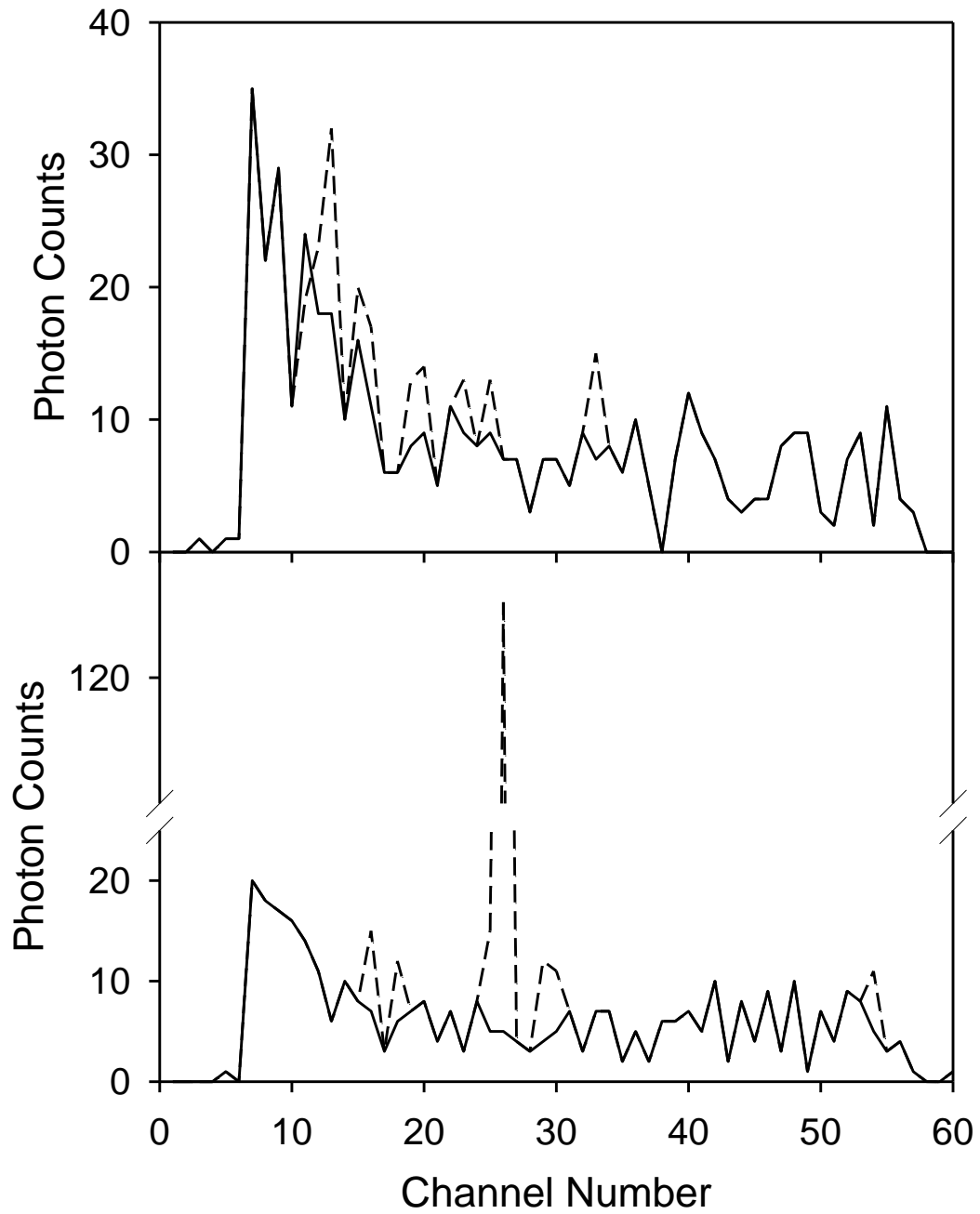
373 The analysis method integrates the counts in the rapidly decaying OSL peak in the early part
374 of each measurement, and subtracts the integrated counts in the background from the end of
375 each measurement to produce a net count. Non-Poisson artefacts in the early part of the
376 measurement would inflate the net count, and removing them would reduce the statistical
377 significance of the measurements. Conversely, they would reduce the net count if present in
378 the later part of the measurement and removing them would increase the measurement
379 significance.

380

381 Poisson filtering was applied to the data set of 2800 decay curves, and filtered decay curves
382 reanalysed and compared with the original unfiltered analysis. Figure 7 illustrates the natural
383 and regenerated signal for a pair of these 2800 decay curves. Single channel features in the
384 original data (dashed lines) have been removed using the filtering algorithm, and the solid
385 line shows the revised smoothed data. This shows the presence of single channel spikes
386 characteristic of non-Poisson dark count bursts which the filter has removed, leaving the
387 “corrected” OSL signal. For this particular grain, the filter has reduced the net natural signal
388 by removing a spike at channel 15, but left the net regenerated signal unchanged since the
389 spike at channel 26 does not fall within either the signal or background integrals. It can be

390 seen that the filter identifies and removes anomalous spikes, while retaining the genuine
391 signal components.

392



393

394 **Fig 7.** An example of one single grain signal and regenerative signal spectra, with original
395 signal (dashed line) and the Poisson smoothed signal (solid line).

396

397 The application of the Poisson filter to this data set is summarised in Table 2, and has
 398 removed several measurements where the apparent signal is identified as a non-Poisson
 399 artefact, reducing the number of statistically significant measurements to 56. In this instance,
 400 this has not significantly changed the age calculated for this sample, although it has brought it
 401 into closer agreement with the age from the original SAR analysis based on small aliquots.
 402 Having removed identified artefacts from the data there will be improved confidence in the
 403 equivalent dose values obtained. It is likely that other sources of over dispersion, for example
 404 micro dosimetry or partial bleaching, dominate in this instance. However, for other samples
 405 it's possible that these other sources may be less significant in which case the non-Poisson
 406 behaviour of the PMT could be more important. It is therefore recommended that a filter to
 407 reduce the effects of non-Poisson behaviour be routinely applied, especially to cases where
 408 light levels are close to detection limits.

409

410

411 **Table 2.** Tola burial chamber results, showing the single grain and SAR dates for the original
 412 analysis, and the single grain age following Poisson filtering.

| | Date | Number of accepted grains (250-500 μm) | Stored Dose Estimates (Gy) |
|----------|-------------------------|---|-------------------------------|
| Original | 2735 ± 500 BC (SG) | 88 | 25.2 ± 3.9 (SG) |
| | 2610 ± 930 BC (SAR) | | 27.0 ± 5.3 (SAR) |
| Revised | 2670 ± 900 BC | 56 | 26.0 ± 5.0 |

413

414 **5. Discussion and Conclusion**

415

416 The findings of Adamiec et al., 2012, that some photon counting systems display non-
417 Poisson dark count behaviour, have been confirmed and extended to cover a further 10
418 systems with a range in age, architecture and electronic configuration. The larger number of
419 systems studied allows a comparison between the age of the system and the extent of non-
420 Poisson variation. This shows a slight correlation, with younger devices showing larger
421 excess variation.

422

423 To assess whether the electronic architecture might be influential (in particular the pulse
424 width of the preamplifier) the PMT from our Riso3 single grain reader, which showed the
425 greatest “k” value observed here was temporarily relocated to run in the electronics of our
426 1986 TL manual reader, which had shown the smallest “k” value. The outcome of that test
427 was that the “k” value was not significantly changed by substitution to the older electronics
428 set up. This may be taken to imply that individual PMT’s have different underlying
429 behaviour.

430

431 The use of phosphorescence obtained by irradiating a feldspar sample demonstrates a simple
432 way of obtaining a controllable and predictable low level light source exhibiting random
433 variations around its value. The decay of this light source would allow evaluation of PMT
434 characteristics under different signal levels. The non-random phosphorescence decay can
435 been accounted for by fitting an appropriate exponential function, with the standard deviation
436 of the residuals, coupled with the applicable light level resulting in k value estimates. This
437 approximation to a full statistical accounting of the data has been shown to have minimal
438 effects on the k-values compared to those calculated for the measurements over shorter

439 periods where the phosphorescence decays by 1%. The use of low level light sources
440 significantly reduces the calculated k-values compared with dark counts, implying that there
441 may be different processes involved in the light detection and dark signal origins in respect of
442 non-Poisson behaviour.

443

444 In our data it was evident that significant excess variation in routine observations is
445 associated with single channel event bursts. Poisson filtering of data sets from both dark
446 counts and OSL signals, showing that the filter successfully removes the single channel
447 bursts with low random probability and reduces residual non-random components.

448 Adamiec et al., 2012 and Bluszcz et al 2015 had suggested that the excess variance in dark
449 counts results in underestimation of dating errors, and influences the outputs of some age
450 models. The implementation of the filter algorithm to a case study data set for a burial
451 chamber in Corsica, France, shows that in this case removal of excess variation via this
452 method had a limited effect on the uncertainties and calculated age of the sample. In this
453 case, and notwithstanding the low signal levels involved, where the dominant dating
454 uncertainties are derived from the variations in underlying dose distribution and
455 microdosimetry rather than the propagation of estimated measurement errors, the impact of
456 the non-Poisson component on error estimates and ages is not appreciable. Here the main
457 impact relates to definition of detection limits and rejection of insignificant data. Use of the
458 Poisson filter results in a more stringent rejection of low significance observations within the
459 dataset, and in our view produced a more robust analysis that obtained using the uncorrected
460 data set. The filtering algorithm and test data sets have been included to facilitate uptake for
461 those wishing to apply similar methods.

462

463 The origins of the non-random components in dark signals are not entirely clear at this stage.

464 Our results do not fit the hypothesis that afterpulses resulting from helium diffusion are the
465 major explanatory factor in determining the extent of non-Poisson behaviour in dark signals.
466 The data sets for the older systems, which would be expected to have acquired higher helium
467 partial pressures, show less excess variation, and as noted above there is no sign that the
468 quartz windows tubes show higher k values than the glass systems. While afterpulses
469 associated with helium have been observed in many systems, and typically account for a few
470 percent of signal events, the relationships between light and dark signals are less clear.
471 Tudyka et.al. 2016 have shown simulated trains of up to 8 helium linked afterpulses in small
472 proportions of events, but it is not clear whether event chains of 100 or more pulses could be
473 explained by such mechanisms, and if so what initiating and propagation events would be
474 implied. Dark response behaviour, and the ways in which dark signals change with time
475 following over-exposure of different tubes vary markedly from system to system. The role of
476 cosmic radiation, or sources of ionising radiation in proximity to the detectors in dating
477 systems may also warrant further attention. We therefore conclude that further research into
478 the behaviour of dark signals would be needed to clarify the origin and nature of non-Poisson
479 behaviour in these systems. Meanwhile Poisson filtering algorithms may be useful in helping
480 to deal with data sets close to detection limits.

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