Accepted Manuscript

Exploring IRSL$_{50}$ fading variability in bedrock feldspars and implications for OSL thermochronometry

Pierre G. Valla, Sally E. Lowick, Frédéric Herman, Jean-Daniel Champagnac, Philippe Steer, Benny Guralnik

Please cite this article as: Valla, P.G., Lowick, S.E., Herman, F., Champagnac, J.-D., Steer, P., Guralnik, B., Exploring IRSL$_{50}$ fading variability in bedrock feldspars and implications for OSL thermochronometry, *Quaternary Geochronology* (2016), doi: 10.1016/j.quageo.2016.08.004.

Exploring IRSL$_{50}$ fading variability in bedrock feldspars and implications for OSL thermochronometry

Pierre G. Valla[1,2*], Sally E. Lowick[3], Frédéric Herman[1], Jean-Daniel Champagnac[2,4], Philippe Steer[5] and Benny Guralnik[2,6]

1 Institute of Earth Surface Dynamics, University of Lausanne, 1015 Lausanne, Switzerland

2 Department of Earth Sciences, ETH Zürich, 8092 Zürich, Switzerland

3 Institute of Geological Sciences, University of Bern, 3012 Bern, Switzerland

4 Free University of Leysin, 1854 Leysin, Switzerland

5 Géosciences Rennes, CNRS UMR 6118, Université de Rennes 1, 35042 Rennes, France

6 Soil Geography and Landscape group and the Netherlands Centre for Luminescence Dating,
Wageningen University, 6708 Wageningen, The Netherlands

* Corresponding author, E-mail: pierre.valla@unil.ch

Abstract

Optically Stimulated Luminescence (OSL) is a well-established Quaternary dating method, which has recently been adapted to application in low-temperature thermochronometry. The Infra-Red Stimulated Luminescence (IRSL) of feldspar, which so far is the most promising target signal in thermochronometry, is unfortunately prone to anomalous fading. The fading of feldspar IRSL is at times not only challenging to measure, but also laborious to incorporate within luminescence growth models. Quantification of IRSL fading is therefore a crucial step in OSL thermochronometry, raising questions regarding (i) reproducibility and reliability of laboratory measurements of fading, as well as (ii) the applicability of existing fading models to quantitatively predict the level of IRSL field saturation in nature. Here we investigate the natural luminescence signal and anomalous fading of IRSL measured at 50 °C (IRSL$_{50}$) in 32 bedrock samples collected from a variety of lithologies and exhumation settings (Alaska and Norway). We report a large span of IRSL$_{50}$ fading rates between samples (g$_{2days}$ ranging from ~0.5 to ~45 %/decade), which further demonstrates (i) a good reproducibility between two common fading measurement protocols, and (ii) the ability of tunnelling models to predict the level of feldspar IRSL$_{50}$ field saturation in nature. We observe higher IRSL$_{50}$ fading in feldspar with increasing Ca content, although other factors cannot be dismissed at present. Finally, our dataset confirms that the applicability of feldspar IRSL$_{50}$ in OSL thermochronometry is limited to rapidly-exhuming settings or warm subsurface environments.

35 **Keywords:** luminescence, feldspar IRSL, anomalous fading, OSL thermochronometry

36

37 **1. Introduction**

38 1.1 OSL thermochronometry

39 Understanding the processes operating at the Earth's surface requires quantitative methods

40 to measure erosion and sedimentation rates. In that context, low-temperature

41 thermochronometric methods (see Reiners and Brandon, 2006 for a review) have been

42 increasingly used over the past decades to quantify bedrock cooling histories within the top ~2-

43 8 km of the upper crust, and subsequently to interpret them into exhumation histories (e.g.

44 Braun et al., 2006; 2012). However, linking long-term exhumation rates (10^5-10^8 year

45 timescales) to short-term erosion measurements (10^0-10^3 year timescales) remains challenging,

46 because different spatial scales and processes are involved. An ongoing debate (Herman and

47 Champagnac, 2016; Willenbring and Jerolmack, 2016), regarding the impact of Quaternary

48 climate and glacial/interglacial oscillations on global-scale erosion rates, highlights the current

49 shortage of quantitative estimates of bedrock erosion and relief evolution over Quaternary to

50 sub-Quaternary timescales (e.g. Herman et al., 2013; Champagnac et al., 2014). Recent

51 methodological developments of low-temperature thermochronometers, such as ^{4}He/^{3}He

52 (Shuster and Farley, 2005) and Optically Stimulated Luminescence (OSL) (Guralnik et al.,

53 2015a; King et al., 2016), offer an unprecedented resolution to constrain rock cooling histories

54 within the upper ~1-3 km of the Earth's crust, and thus to potentially reconstruct erosion

55 histories and/or topographic relief changes over sub-Quaternary timescales (e.g. Shuster et al.,

56 2005; 2011; Valla et al., 2011a; King et al., 2016).

57

58 OSL dating (e.g. Aitken, 1998) has recently been adapted for low-temperature

59 thermochronometry by considering the competing effects of filling and emptying of naturally-

60 occurring electron traps (linked to the OSL signal) in crystal lattices due to environmental

61 radiation and temperature, respectively (Herman et al., 2010; Li and Li, 2012; Guralnik et al.,

62 2013; King et al., 2016). While other "trapped charge dating" methods (e.g. Grün, 2001) have

63 been proposed and tested, including thermoluminescence (TL; e.g. Prokein and Wagner, 1994;

64 Tang and Li, 2015) and electron-spin resonance (ESR; Koshchug and Solovyov, 1998; Grün et

65 al., 1999), the advantage of OSL techniques in thermochronometry lies in their rapid

66 methodological development and expanding use in Quaternary science (see reviews by Wintle,

67 2008 and Rhodes, 2011). One major intrinsic limitation in trapped charge dating, common to

68 OSL, ESR and TL alike, is signal saturation (i.e. when all available electron traps become

69 filled). If the target luminescence signal for dating is close to its saturation limit, only minimum

70 ages may be provided. Consequently, when luminescence is used to decipher rock cooling

71 rates, near-saturation signals can only characterise maximum (upper-bound) rates of cooling

72 and exhumation (e.g. Guralnik et al., 2013). Fortunately, the first OSL-thermochronometry

73 applications (Herman et al., 2010; Wu et al., 2012; De Sarkar et al., 2013; Wu et al., 2015)

74 addressed settings of extremely rapid exhumation (where signal saturation should not be a

75 problem), and quantified bedrock cooling histories below ~100 °C and over sub-Quaternary

76 timescales. Despite the fact that a standard mineral/signal pair was chosen (quartz "fast OSL

77 component"; Wintle and Murray, 2006), the low sensitivity (Herman et al., 2010; Jeong and

78 Choi, 2012), anomalous characteristics (Li and Li, 2006; Preusser et al., 2009), and the

79 ubiquitous lack of the "fast component" in bedrock quartz of different petrology/mineralogy

80 (Guralnik et al., 2015b), appeared to narrow the use of bedrock quartz for OSL-

81 thermochronometry, and stimulated the continued search for alternative mineral/signal

82 combinations (Guralnik et al., 2015b).

83

84   Infra-Red Stimulated Luminescence (IRSL) from feldspar is advantageous over quartz in

85 several ways (e.g. Huntley and Lamothe, 2001), with (i) a higher luminescence sensitivity in

86 both sediment and bedrock samples (e.g. King et al., 2014), (ii) in general, a higher charge

87 storage capacity (i.e. later saturation) offering a wider dating range (e.g. Buylaert et al., 2012),

88 and (iii) a negligible signal contamination from quartz OSL, if present (e.g. Sohbati et al.,

89 2013). Recently, OSL thermochronometry using bedrock Na-feldspar was investigated in a

90 deep continental drillhole (Guralnik et al., 2015a), where IRSL signals measured at 50 °C

91 (hereafter $IRSL_{50}$) were successfully characterised in the laboratory and translated into

92 environmental palaeotemperatures in the ~40-70 °C range over a ~50 ka timescale (Guralnik et

93 al., 2015a). Moreover, the recently-introduced protocol of multiple elevated temperature post-

94 IR (MET-pIRIR) measurement of feldspar (Li and Li, 2011a,b) conceptually offers access to

95 multiple thermochronometric subsystems within a single mineral, as the different subsignals

96 display higher thermal stabilities with increasing stimulation temperatures (Li and Li, 2011b;

97 King et al., 2016).

98

99 1.2. Anomalous fading in feldspar IRSL

100   Although feldspar IRSL appears in many ways preferable to quartz OSL for

101 thermochronometry, one of the drawbacks of feldspar IRSL is its anomalous fading (e.g.

102 Spooner, 1994; Huntley and Lamothe, 2001), an athermal loss of trapped electrons via quantum

103 mechanical tunnelling to their nearest recombination centres (Visocekas, 2002; Huntley, 2006).

104 Since anomalous fading is ubiquitous in feldspar $IRSL_{50}$ (Huntley and Lamothe, 2001; Huntley

105 and Lian, 2006), recent efforts have mainly focused on extracting a more stable feldspar signal

106 using pIRIR protocols (e.g. Thomsen et al., 2008; 2011; Li and Li, 2011a; Buylaert et al.,

107 2012). Although the latter protocols exhibit a reduced, perhaps even negligible, anomalous

108 fading, it is generally at the expense of a higher thermal stability of the target electron trap (Li

109 and Li, 2011a,b; Guralnik et al., 2015c; King et al., 2016). This in turn leads to higher residual

110 doses in sediment dating (e.g. Wintle, 2008), and higher closure temperatures in

111 thermochronometry (e.g. Guralnik et al., 2015c), which may once again limit the benefit of

112 non-fading IRSL signals in low-temperature thermochronometry (e.g. Qin et al., 2015).

113 Moreover, the MET-pIRIR protocol often exhibits progressively lower IRSL sensitivity with

114 increasing stimulation temperature (Li and Li, 2011b; King et al., 2016), which can hinder

115 signal extraction for some bedrock samples with low luminescence sensitivity. Despite the

116 presence of anomalous fading in feldspar $IRSL_{50}$, and in light of recent success in avoiding it

117 via pulsed stimulation at low temperature (Tsukamoto et al., 2006; Jain et al., 2015a), $IRSL_{50}$

118 remains highly relevant for OSL thermochronometric investigations.

119

120 Over the years, several protocols have been proposed to quantify the anomalous fading of

121 feldspar over laboratory timescales (Huntley and Lamothe, 2001; Auclair et al., 2003) and to

122 correct for fading in the linear dose response range (e.g. Lamothe and Auclair, 1999; Huntley

123 and Lamothe, 2001; Lamothe et al., 2003). More recently, Huntley (2006) formulated a

124 physical description of electron tunnelling that further enabled the application of fading

125 corrections within the entire dose response range (Kars et al., 2008; Li and Li, 2008). In

126 particular, these models allow the prediction of luminescence intensities in "field saturation"

127 (Huntley and Lian, 2006), in which the natural intensity of feldspar $IRSL_{50}$ becomes insensitive

128 to time, and is governed only by the counterbalancing effects of environmental radiation and

129 anomalous fading (Kars et al., 2008).

130

131 In the present study, we investigate anomalous fading of the $IRSL_{50}$ signal in bedrock

132 feldspar, and highlight its key implications for OSL thermochronometry. As described earlier,

133 OSL thermochronometry exploits the thermal dependence of trapped electrons in minerals

134 during their exhumation towards the Earth's surface. Given zero fading, a non-saturated natural

135 $IRSL_{50}$ signal in bedrock feldspar should, in theory, reflect the sample's cooling history.

136 Anomalous fading of the signal would, however, lower the measured $IRSL_{50}$ level in a given

137 bedrock sample, preventing full saturation of the signal and influencing the determination of a

138 cooling rate (if not accounted for). While the incorporation of the effects of $IRSL_{50}$ fading in

139 OSL-thermochronometric interpretation has been demonstrated as important (Guralnik et al.,

140 2015a; King et al., 2016), there is a lack of consensus at present, as to whether laboratory

141 measurements of IRSL fading are reproducible and/or reliable (see Li et al., 2014 for a review),

142 and thus whether the use of fading models (and their associated age correction schemes) is

143 appropriate. Moreover, the recent notion that all luminescence processes in feldspars are

144 intrinsically related to fading (Jain et al., 2012; 2015b) raises a renewed interest in the fading

145 model of Huntley (2006) and its later derivatives. More specifically, it raises the need for

146 validation studies, where a laboratory measurement of fading rate can be related to its natural

147 fading rate in nature. To date, only a handful of studies (Huntley and Lian, 2006; Kars et al.,

148 2008; Guralnik et al., 2015a) have quantitatively evaluated the effects of natural versus

149 laboratory fading rates. It is therefore necessary to assess (i) whether anomalous fading of the

150 feldspar $IRSL_{50}$ signal can be accurately measured, and (ii) for a given fading rate, whether the

151 field saturation of the $IRSL_{50}$ signal can be quantitatively predicted. To this aim, we extracted a

152 wide range of K- and Ca-/Na-feldspars from 32 bedrock samples in different lithologies and

153 exhumation environments, and measured their natural feldspar $IRSL_{50}$ intensities alongside the

154 anomalous fading of the laboratory-regenerated signals. A large geochemical dataset, including

155 whole-rock major and trace elements as well as selected feldspar mineralogy, enables us to

156 discuss potential causes for the wide range of $IRSL_{50}$ anomalous fading observed. Finally, we

157 use this dataset to test the theoretical ability of tunnelling models (Huntley 2006; Kars et al.,

158 2008; Guralnik et al., 2015a,c) to predict $IRSL_{50}$ field saturation levels, and to discuss the

159 implications for OSL thermochronometry.

160

161 **2. Samples and methods**

162 2.1. Sampling strategy and sample preparation

163   Bedrock samples from old crystalline or metamorphic basements have been collected in

164 different exhumation settings in Norway and Alaska to explore the applicability of feldspar

165 $IRSL_{50}$ thermochronometry over a wide range of long-term (i.e. 10^5-10^8 years) exhumation

166 rates. All sample locations and lithological descriptions are given in Table 1. Eight samples are

167 distributed along the Sognefjord and its vicinity (SOG samples; western Norway) where

168 exhumation rates are ~0.01-0.05 km Ma^{-1} (e.g. Hendriks et al., 2007; Nielsen et al., 2009). The

169 SOG samples were collected at varying elevations, from sea-level towards ~1000-m a.s.l. low-

170 relief surfaces (e.g. Steer et al., 2012). Fifteen samples have been collected in the Granite

171　Range (GRA samples; southern Alaska) where low-temperature thermochronometric data

172　indicates long-term exhumation rates of 0.1-0.5 km Ma^{-1} (e.g. Spotila et al., 2004; Berger et al.,

173　2008). In the Granite Range, our sampling strategy was to collect bedrock samples across

174　elevation profiles (Wagner and Reimer, 1972; Valla et al., 2011b). Finally, nine samples have

175　been collected in the Yakutat terrain (YAK samples; southern Alaska), all locations at sea-level

176　along the Disenchantment Bay - Russel Fjord. This area is tectonically active with reported

177　long-term exhumation rates of ~1-3 km Ma^{-1} (e.g. McAleer et al., 2009; Enkelmann et al.,

178　2015). Our sampling strategy thus covers two orders of magnitude of long-term bedrock

179　exhumation rates, from 0.01 km Ma^{-1} (SOG) up to 1-3 km Ma^{-1} (YAK). Moreover, we targeted

180　different lithologies (sandstone, gneiss and granite/diorite samples), probing a large diversity in

181　bedrock geochemistry and consequently also in feldspar mineralogy. Finally, we restricted our

182　study to cold environments (mean annual surface temperatures <10 °C, Table 1) to exclude

183　potential luminescence signal loss at surface temperature conditions in tropical to sub-tropical

184　regions (Wu et al., 2015).

185

186　　Bedrock samples were prepared under subdued orange light to ensure the extraction of

187　feldspar minerals from the interior of the sample, where the luminescence signal is unaffected

188　by surface bleaching. Initial sample dimensions were at least 10 cm (length/width/thickness) to

189　allow the removal of the light-exposed sample surface (at least 2-3 cm) using a water-cooled

190　diamond saw. The light-safe internal part was then gently hand crushed to 180-212 μm grain

191　size using a mortar and pestle to avoid mineral grinding and potential luminescence resetting

192　via triboluminescence (e.g. Bateman et al., 2012). All 180-212 μm grains were treated with

193　32% hydrochloric acid to remove potential surface carbonates and with 30% hydrogen

194　peroxide to remove any organic component. After chemical treatment, mineral fractions were

195　rinsed thoroughly with water to remove dust particles. Feldspar and quartz fractions were

196　isolated by density separation using LST Fastfloat (sodium heteropolytungstate). Densities of

197　2.58 g cm^{-3} and 2.70 g cm^{-3} were used in order to isolate potassium-rich feldspars (K-feldspars,

198　<2.58 g cm^{-3}) from quartz and other feldspars (2.58-2.70 g cm^{-3}), and from remaining heavy

199　minerals (>2.70 g cm^{-3}). For SOG samples (except SOG-22) we selected K-feldspars for

200　analysis.

201　　For GRA, YAK and SOG-22 samples, density separation at 2.58 g cm^{-3} yielded no light

202　fraction, suggesting the dominance of calcium/sodium-plagioclases (Ca-/Na-feldspars), which

203　were thus taken for further analysis. Further mineral purification was deemed unnecessary, as

204　the contribution from quartz to $IRSL_{50}$ is negligible (Sohbati et al., 2013 and references

6

205 therein). Feldspar extracts were not etched before luminescence measurements (Duller, 1992),

206 and feldspar grains were directly mounted on stainless steel discs to produce large aliquots (4 to

207 6 mm diameter, ~500-1500 grains per disc).

208

209 2.2. Luminescence equipment and protocols

210 All IRSL$_{50}$ measurements were made on automated Risø TL/OSL DA-20 readers (Institute

211 of Geological Sciences, University of Bern), each possessing a ^{90}Sr/^{90}Y beta source (~0.1-0.2

212 Gy s^{-1}) and a heater plate (20-700 °C), with systematic instrumental uncertainties of ~1.5% for

213 single-aliquot measurements (Bøtter-Jensen et al., 2010). Luminescence stimulation was

214 performed with infrared (870 ±40 nm) light-emitting diodes at 90% power (delivering ~130

215 mW cm^{-2} at the aliquot position); the emitted luminescence signal was detected through a 410-

216 nm interference and a 2-mm Schott BG-39 filters by an EMI 9235QA photomultiplier tube. All

217 luminescence protocols described below (adapted from Guralnik, 2015a) are fully detailed in

218 Table 2.

219

220 Feldspar natural IRSL$_{50}$ signals, and their subsequent laboratory dose-response curves, were

221 measured using the Single-Aliquot Regenerative-dose (SAR) protocol (Wallinga et al., 2000).

222 For each sample, at least three individual aliquots were measured using a preheat at 250 °C for

223 60 s (to empty unstable electron traps), followed by IRSL$_{50}$ for 200 s (Table 2). We used

224 laboratory beta irradiation doses from 0 up to ~2800 Gy to fully characterize the dose response

225 curve, a test dose of 44 Gy to monitor sensitivity changes throughout the protocol, and a high-

226 temperature optical wash (IRSL at 290 °C) between measurement cycles (Buylaert et al.,

227 2009). Given the recent interest in and the research into "test dose plateaus" (Yi et al., in press),

228 we believe that a test dose of 44 Gy is readily applicable to obtain equivalent doses in the ~4.4-

229 440 Gy range (cf. Yi et al., in press), and likely even beyond. While small variations in the

230 laboratory saturation level as a function of test dose have also been documented (Yi et al., in

231 press), these effects are demonstrably minor and were hence considered beyond the scope of

232 the present study.

233 All analysed aliquots fulfilled the standard performance criteria for recuperation (<5%),

234 recycling ratio (<10%) and maximum test dose error (<10%). To test the efficacy of the SAR

235 protocol parameters, a dose recovery experiment was conducted for each sample

236 (Supplementary Table S5). To this end, two to three fresh aliquots per sample were optically

237 bleached using IRSL$_{50}$ for 300 s, and were subsequently given a 200-Gy laboratory dose which

7

238   was then treated as an unknown and recovered using the same SAR protocol as the natural

239   doses.

240

241   To explore the fading variability of $IRSL_{50}$ signals in individual bedrock samples, we

242   conducted fading experiments on two sensitised aliquots from the dose response experiments.

243   We used two different approaches to measure feldspar $IRSL_{50}$ fading (Table 2), namely a SAR-

244   based long-shine method (modified from Huntley and Lamothe, 2001), and a non-SAR short-

245   shine method (modified from Auclair et al., 2003), with the objective to test the agreement

246   between the two protocols. In the long-shine protocol, after a given dose of 200 Gy, the

247   luminescence signal was measured following variable delays, each lasting between 0.3 and 48

248   hours (Table 2). In the short-shine protocol, a dose of 72 Gy was given only once, and followed

249   by a series of consecutive short-shine (0.1 s) measurements, separated from each other by

250   pauses lasting between 0.1 and 55 hours. To account for signal loss due to the short optical

251   excitation, the delayed short-shine measurements were normalized to an otherwise identical

252   series of prompt (undelayed) measurements, done once before and once after the delayed

253   experiment (Table 2).

254

255   2.3. Geochemistry and environmental dose rate determination

256   We obtained the whole-rock geochemistry (major and trace elements) of all 32 samples

257   using inductively-coupled plasma mass spectrometry (Supplementary Tables S2-S3) to

258   measure the dominant radionuclide content used for the environmental dose rate estimate (U,

259   Th and K, Table 1). For a subset of 12 samples (GRA and YAK), we used the X-ray

260   fluorescence (XRF) attachment to the Risø TL/OSL reader (Kook et al., 2012; Guralnik et al.,

261   2015a) to determine feldspar mineralogical composition (Supplementary Table S4) and infer

262   the internal potassium content (K_{int}, Tables 1 and S4). XRF data confirms that, in GRA and

263   YAK samples, the aliquot mineralogy is dominated by Ca-/Na-feldspars (with up to 50% wt.

264   quartz content), translating to an internal potassium content K_{int} between 0.2 and 4%

265   (Supplementary Table S4). Based on these measurements, we assumed for the other GRA and

266   YAK samples (incl. SOG-22) an averaged K_{int} of 1.9 ±1.4% (Table 1), in agreement with

267   previous estimates reported in the literature for Ca-/Na-feldspars (Barré and Lamothe, 2010;

268   Sohbati et al., 2013; Guralnik et al., 2015a). For SOG samples (except SOG-22), we used K_{int}

269   of 12.5 ±1% for K-feldspars following standard literature values (Huntley and Baril, 1997;

270   Barré and Lamothe, 2010).

271

272   For each sample, the whole-rock radionuclide content (U, Th and K) and feldspar K_{int} have

273   been converted into environmental dose rates using the conversion factors of Guérin et al.

274   (2012), beta and gamma attenuation factors of Guérin et al. (2012) and Guérin and Mercier

275   (2012), and alpha attenuation factors of Bell (1980) and Brennan et al. (1991). For alpha

276   attenuation, an a-value of 0.15 ±0.05 was used after Balescu and Lamothe (1994). The average

277   water content in our bedrock samples was estimated at 2 ±2% (e.g. Aitken and Xie, 1990).

278   Cosmic dose rate was treated as negligible, being comparable in magnitude to uncertainty on

279   the total environmental dose rate (King et al., 2016). Based on thin-section observations, and

280   because the original grain size has been affected by rock crushing and sieving during feldspar

281   extraction, we used a 180-2500 μm grain size range for SOG and GRA samples and a 180-1000

282   μm grain size range for YAK samples.

283

284   To address the two entangled uncertainties regarding (i) the grain size of the target feldspar,

285   and (ii) the feldspar phase which is actually contributing to the $IRSL_{50}$ signal (e.g. Sohbati et

286   al., 2013), we considered several (2 or 4, Supplementary Table S1) dose rate scenarios

287   following the approach of Guralnik et al. (2015a). Specifically, for all feldspars, we calculated

288   dose rates for two end-member grain sizes (180 and 2500 μm for GRA and SOG, and 180 and

289   1000 μm for YAK), assuming a homogeneous distribution of internal potassium (K_{int}, Table 1)

290   and $IRSL_{50}$ signal contribution from the entire feldspar crystal. For Na-/Ca feldspars (GRA,

291   YAK and SOG-22), we calculated two additional scenarios, in which all the $IRSL_{50}$ signal

292   originates only from K-feldspar inclusions (1 and 100 μm, K_{int} ~12.5%) within the Na-/Ca-

293   feldspar host crystal (e.g. Sohbati et al., 2013). We then averaged these end-member scenarios

294   to obtain representative environmental dose rates, which ranged across more than an order of

295   magnitude between samples, from 0.3 to 9 Gy ka^{-1} (Table 1). This range, and the associated

296   conservative errors, reflect the variability between K-rich and K-poor feldspar phases, and the

297   averaging across the diverse dose-rate scenarios as listed above (Supplementary Table S1).

298

299   **3. Results**

300   3.1. Dose response and fading measurements

301   Dose response and fading measurements are illustrated for four representative GRA, SOG

302   and YAK samples (Figure 1), and are fully documented for all samples in the Supplementary

303   Information (Supplementary Table S6). Analysed aliquots (4- to 6-mm diameter) usually

304   yielded bright signals with typical $IRSL_{50}$ decay curves. Laboratory dose-response curves (left

305   panels in Figure 1) have been fitted using the General-Order Kinetics (GOK) growth function

9

306 (Eq. (10) in Guralnik et al., 2015c). The characteristic dose (D_0), the electron-trapping order

307 (α), as well as the equivalent dose (D_e), are reported in Tables 3 and S5. For each aliquot, we

308 also derived the natural $IRSL_{50}$ signal L_n/T_n (Figure 1) and calculated the natural trap filling

309 level $(n/N)_{nat}$ by multiplying the apparent $IRSL_{50}$ intensity relative to its laboratory maximum

310 (open stars in Figure 1) by the predicted number of occupiable traps in the laboratory (Eq. (3)

311 in Guralnik et al., 2015a). The majority of best-fit D_0 and α values are well-constrained with

312 typical relative uncertainties of <10% and <4%, respectively (Table 3). Natural trap filling

313 levels $(n/N)_{nat}$ exhibit varying uncertainties (1-70%; clearly anticorrelated with $(n/N)_{nat}$ itself),

314 and cover almost three orders of magnitude (i.e. 0.003-0.85; Table 3), thus offering a robust

315 dataset for evaluating the net effects of environmental radiation, anomalous fading and possibly

316 long-term cooling on the natural $IRSL_{50}$ intensity of bedrock feldspar.

317

318 Fading results obtained using both the long-shine (central panels in Figure 1) and short-

319 shine (right panels in Figure 1) methods are also summarised in Table 3. Two aliquots were

320 measured individually and averaged to calculate sample-specific g-values and standard

321 deviations (g_{2days} in %/decade; Huntley and Lamothe, 2001). Fitting the same datasets using

322 Eq. (5) in Kars et al. (2008), we obtained an alternative measure of fading - the nearest-

323 neighbouring hole-centre density ρ' as defined in Huntley (2006) - which we later used in the

324 kinetic model to predict the $IRSL_{50}$ field saturation levels (Guralnik et al., 2015a). Inter-aliquot

325 variability is small, confirming the good reproducibility of the fading measurement protocols,

326 with more robust estimates for the long-shine method as explained below (Table 3). Although

327 the short-shine protocol is less time-consuming (approximately half the time), the integrated

328 $IRSL_{50}$ signal at each short-shine measurement is considerably lower, and therefore associated

329 with larger uncertainties propagated into the resulting g-value (Table 3 and Figure 1). For

330 feldspars with intermediate to low luminescence sensitivity (roughly one third of our dataset),

331 the short-shine protocol yielded signals with a very low signal-to-background ratio that could

332 not be used to derive meaningful g-values (Table 3). Figure 2 shows that, on average, fading

333 rate estimates are in excellent agreement between the long- and the short-shine protocols. Thus,

334 to further reduce experimental noise, in the subsequent modelling we used averaged g-values

335 (wherever possible) from both protocols. Sample-specific $IRSL_{50}$ g_{2days} values reveal a wide

336 range from ~0.5 to ~45 %/decade (Table 3). K-feldspars from SOG samples exhibit g_{2days}

337 values between 3 and 11 %/decade, which is typical for K-feldspars from both bedrock and

338 sediment origin (Huntley and Lamothe, 2001; Huntley and Lian, 2006). Na-/Ca-feldspars show

339   a higher variability in measured g_{2days} values between samples (Figures 2 and 3), especially for

340   GRA samples (from 0.5 to >45 %/decade).

341

342   3.2. Predictions of feldspar $IRSL_{50}$ field saturation

343   To evaluate whether fading variability in our dataset is the main controlling factor for the

344   natural $IRSL_{50}$ intensities, we first examined the relationship of the laboratory-derived g_{2days}

345   values with the measured $(n/N)_{nat}$ values (Figure 3). Our results show a clear inverse correlation

346   between g_{2days} and $(n/N)_{nat}$ values, in agreement with previous observations (Huntley and Lian,

347   2006) but over a considerably wider range of fading rates. Even prior to modelling, such an

348   inverse correlation suggests that the measured $(n/N)_{nat}$ in most of our samples is mainly

349   controlled by anomalous fading. Only some YAK samples appear to deviate from the overall

350   trend, with their $(n/N)_{nat}$ values markedly below field saturation (Figure 3). However, the

351   relation in Figure 3 is only qualitative, because this empirical relationship might be influenced

352   not only by sample-specific kinetic parameters, but also by thermal loss (which cannot be

353   accounted for in this figure). In the following section, we test our current dataset against a

354   recently proposed physical model predicting feldspar $IRSL_{50}$ field saturation under anomalous

355   fading (Guralnik et al., 2015a, incorporating the tunnelling term of Huntley, 2006, and

356   extending the dose response of Kars et al., 2008 to non-first order).

357

358   Athermal IRSL signal loss in feldspars is caused by the quantum mechanical tunnelling of

359   electrons from their traps towards electron holes (Huntley and Lamothe, 2001). Here we use

360   the physical model of Huntley (2006) which is based on the assumptions that (i) the tunnelling

361   of electrons is governed by the distance to their nearest recombination centre, and (ii)

362   recombination centres within a feldspar crystal are randomly distributed with a given density.

363   This model (Huntley 2006) can be used to estimate sample-specific field saturation values (e.g.

364   Kars et al., 2008; Guralnik et al., 2015a; King et al., 2016). Following Guralnik et al. (2015a),

365   we use a Monte-Carlo approach to numerically predict, on a sample to sample basis, the $IRSL_{50}$

366   trap filling saturation values in nature $(n/N)_{ss}$ (termed "field saturation"). In the simulation

367   (1000 runs per sample), random instances of the kinetic parameters (D_0 and α, Table 3), natural

368   dose rates (Table 1), and ρ' (Table 3, averaged wherever possible between the long- and short-

369   shine experiments) are drawn from their normal distributions (as given by their best-fit values).

370   The modelling results, reported as median $(n/N)_{ss}$ and associated 68% confidence interval, are

371   given in Table 3 and visualised against measured natural trap filling $(n/N)_{nat}$ values in Figure 4.

372

373   Figure 4 highlights the quantitative agreement between predicted $(n/N)_{ss}$ and measured

374    $(n/N)_{nat}$ values, confirming that most of our samples are either in full field saturation, or hardly

375   discernible from it (<15%; Guralnik et al., 2015b). In other words, the natural feldspar $IRSL_{50}$

376   signal in the majority of our samples (n = 25) is controlled by electron tunnelling and does not

377   include a detectable thermal signature of the rock exhumation history. However, more than half

378   (66%) of YAK samples, representing the fastest-exhuming setting, exhibit $(n/N)_{nat}$ values that

379   are considerably (more than 15%) below their predicted $(n/N)_{ss}$ values, suggesting field

380   disequilibrium and thus high likelihood of a thermal signature that can be translated into a

381   cooling history. The thermal signature of two additional samples (GRA-BR and SOG-17)

382   remains in doubt, requiring further investigation that will either link them to exceptional

383   thermal histories, or dismiss them as outliers (one in each locality).

384

385   3.3. Dose recovery results

386   Calculated dose recovery ratios (Supplementary Table S5) were typically within 10% of

387   unity for all but four (GRA-13/-14/-17 and SOG-21) samples. Although imperfect, the 15-20%

388   underestimation in the dose recovery of SOG-21 is within familiar values, even when working

389   with a SOL2 solar simulator (e.g. Buylaert et al., 2012; King et al., 2016), and may relate to an

390   irreversible trapping efficiency change (Kars et al., 2014). On the other hand, it is hard to

391   blame the considerable dose recovery overestimates in GRA-13/-14/-17 (30 to 65%) on the test

392   dose/given dose ratio (here 18%), which is in the 15-80% trust zone and certainly close to the

393   recently recommended ~30% value (Yi et al., in press). To test whether the observed

394   overestimation is due to a large residual dose, we measured the latter in GRA-13 and GRA-BR

395   after an $IRSL_{50}$ bleach for 300 s. Low residuals of ~5-8 Gy correspond to less than 2% of the

396   equivalent doses, and to 2-3% of the recovered dose, and therefore cannot explain the observed

397   overestimations. To test whether the problem lies in a thermal transfer (e.g. Huntley et al.,

398   1993), we extended the dose recovery experiments further by conducting the IRSL bleach at

399   higher temperatures (100 and 200 °C). Bleaching at higher temperatures did not significantly

400   improve dose recovery in GRA-13/-14/-17, and had no effect on a well-behaving sample

401   (GRA-BR). Overall, unacceptable dose recovery affects only a minor subset (13%) of our

402   dataset and might not necessarily imply that the obtained $(n/N)_{nat}$ values are not reliable

403   (Buylaert et al., 2012). However, such dose recovery over- and underestimations do raise the

404   question of what thermal/optical bleach is most appropriate for resetting the natural $IRSL_{50}$

405   intensity in feldspars that were never previously exposed to light.

406

407 **4. Discussion**

408 4.1. $IRSL_{50}$ and anomalous fading in bedrock feldspars

409   Our results confirm that feldspar $IRSL_{50}$ signals from bedrock extracts show consistent and

410 reproducible luminescence characteristics using the SAR protocol. This is a clear advantage

411 compared to OSL of bedrock quartz which, apart from a very few cases (e.g. Wu et al., 2015),

412 is generally deemed dim and unsuitable (Jeong and Choi, 2012; Guralnik et al., 2015b).

413 Moreover, the vast majority of our bedrock feldspars (87%, n = 28, Supplementary Table S5)

414 also fulfilled the dose recovery test, which is a fundamental acceptance criterion for

415 luminescence dating. Dose responses were successfully fitted with a General-Order Kinetics

416 model, with a median order of 2.4±0.8, in agreement with Na-feldspars from the KTB borehole

417 (Guralnik et al., 2015a), therefore capturing the non-linearity of the dose response curve in just

418 a single parameter.

419

420   Fading measurements exhibit a large variability in bedrock feldspars from ~0.5 to ~45

421 %/decade, beyond the previously published value range (Huntley and Lamothe, 2001; Huntley

422 and Lian, 2006). Moreover, we compared two established fading protocols in sediment dating

423 (Huntley and Lamothe, 2001; Auclair et al., 2003) and demonstrated that both are applicable

424 for bedrock feldspars, yielding comparable results between the two methods (Figure 2). Given

425 the considerable experimental differences between the protocols (SAR-based and non-SAR),

426 the agreement of the obtained results is striking. The dependence of the fading rate on the given

427 laboratory dose (Huntley and Lian, 2006; Li and Li, 2008; Kars and Wallinga, 2009) is not

428 evident from our dataset, although the administered laboratory doses (200 and 72 Gy for the

429 long-shine and short-shine protocols, respectively) may be too close to detect any systematic

430 deviation.

431

432   In our suite of bedrock feldspars, $(n/N)_{nat}$ values are mostly inversely correlated with

433 laboratory-measured fading rates (Figure 3), qualitatively suggesting field saturation in the

434 majority of samples. Quantitatively, we have demonstrated that field saturation can be

435 successfully predicted across almost three orders of magnitude of $IRSL_{50}$ electron trap

436 occupancy (Figure 4), using a recent extension (Guralnik et al., 2015c) of familiar electron

437 tunnelling models (Huntley, 2006; Kars et al., 2008). Despite successful prediction of field

438 saturation in both sedimentary feldspars (Kars et al., 2008; Kars and Wallinga, 2009) and

439 bedrock feldspars (Guralnik et al., 2015a; King et al., 2016), the underlying model(s) and the

440 ability to accurately measure the g-value in the laboratory remains a debateable subject (Li et

13

441   al., 2014). Here, we exploited the large variability in measured fading rates to provide a

442   quantitative validation of our predictive model over a large range of dose rates, trap

443   occupancies, and fading rates (Figure 4).

444

445   We also used the wide range of observed $IRSL_{50}$ fading rates to discuss potential

446   geochemical control, starting with the whole-rock geochemistry (Supplementary Tables S2-3)

447   as a first-order proxy. However, we found no convincing relationship between the whole-rock

448   trace element content (Supplementary Table S3) and feldspar $IRSL_{50}$ fading rate. Figure 5A

449   illustrates the lithological variability within our dataset as shown by representative major

450   oxides (SiO_2 and Fe_2O_3), suggesting that, at first order, the fading rate would increase with

451   decreasing SiO_2 content or increasing Fe_2O_3 (or CaO) content. Felsic lithologies (i.e. >63% wt.

452   SiO_2 content) all show relatively low feldspar fading rates (less than 10 %/decade) regardless

453   of the feldspar type. However, for intermediate (i.e. 52-63% wt. SiO_2 content) and mafic (i.e.

454   <52% wt. SiO_2 content) lithologies, a large spread in fading rates is observed (Figure 5A). For

455   a representative subset of GRA and YAK feldspar separates, we also performed XRF

456   mineralogical determination (Supplementary Table S4). Figure 5B shows that the feldspar

457   fading rate increases with Ca content, in agreement with previous observations (Huntley and

458   Lian, 2006; Huntley et al., 2007). In summary, neither the whole-rock geochemistry, nor the

459   trace elements, seem to be a good proxy for feldspar anomalous fading (although we did not

460   apply multivariate analysis); however, our results show that the feldspar internal chemistry (i.e.

461   Ca content, Figure 5B) may be a first-order proxy for the $IRSL_{50}$ fading rate. Further

462   investigations are required to better understand the potential causes of fading rate variability in

463   bedrock feldspars; these include both internal factors (e.g. the origin of the $IRSL_{50}$ signal in Ca-

464   /Na-feldspars; e.g. Sohbati et al., 2013) as well as external drivers, such as the metamorphic

465   grade or terrain age (Huntley and Lamothe, 2001), or weathering processes (Parish, 1992;

466   Huntley, 2011).

467

468   4.2. Implications for OSL thermochronometry

469   Our extensive bedrock dataset offers an opportunity to discuss the applicability of feldspar

470   $IRSL_{50}$ in OSL thermochronometry. First, the variability of the fading rates measured for both

471   K- and Ca-/Na-feldspars is significantly broader than those reported in previous OSL

472   thermochronometric studies (Guralnik et al., 2015a, King et al., 2016). This reinforces the

473   notion that anomalous fading in feldspar should be measured for each sample, as it can strongly

474   affect the natural $IRSL_{50}$ signal (Figure 3). Moreover, one further complexity in OSL

475   thermochronometry is that a sample's thermal history can be inverted only from the natural trap

476   filling $(n/N)_{nat}$, and not from the equivalent dose alone (D_e) as in OSL sediment dating. This

477   prevents the application of classical fading correction schemes (e.g. Huntley and Lamothe,

478   2001) and requires the quantitative prediction of the field saturation level of the luminescence

479   signal (e.g. Kars et al., 2008; Li and Li, 2008) in order to screen for a potential "thermal

480   signature" in a given sample. For most of our samples, and within a wide range of fading rates,

481   the quantitative agreement of observed field saturation levels of the $IRSL_{50}$ signal with

482   numerical results from the standard tunnelling model (Huntley, 2006), coupled with a General-

483   Order Kinetics dose response (Guralnik et al., 2015c), confirms the need to include fading

484   measurements in preliminary screening of samples before any thermochronometric applications

485   using feldspar $IRSL_{50}$. This first-step investigation of the feldspar athermal stability would

486   ultimately be combined with the experimental characterisation of each sample's thermal

487   stability (Guralnik et al., 2015a; King et al., 2016), which can vary greatly between K- and Na-

488   /Ca-feldspars (e.g. Tso et al., 1996; Li et al., 1997) and thus significantly influence

489   thermochronometric interpretation.

490

491   Our sampling strategy, covering a wide range of exhumation rates (from 0.01 to 3 km Ma^{-1}),

492   provides further information regarding the range of applicability of feldspar $IRSL_{50}$ to OSL

493   thermochronometry in diverse natural settings. All GRA and SOG samples appear in field

494   saturation (Figure 4), with just one outlier per locality (GRA-BR and SOG-17). SOG-17 was a

495   translucent bedrock sample, which may immediately be suspected of suffering optical resetting

496   (in nature and/or during sample preparation), that may have biased the observed luminescence

497   signal to below the predicted field saturation level. GRA-BR is a spatial outlier within the GRA

498   dataset, situated at low-elevation within a very wide and deeply-carved valley. Pending

499   additional experimental confirmation (beyond the scope of this study), its deviation from field

500   saturation could potentially reflect recent accelerated exhumation by efficient glacial processes.

501   Finally, more than half of the YAK samples exhibit a thermal signature in their measured

502   natural $IRSL_{50}$ signal (Table 3 and Figure 4). Samples YAK-17/-18/-50 (all with a "thermal

503   signature") are located along a deeply-carved fjord with very high long-term exhumation rates

504   (2-3 km Ma^{-1}; Enkelmann et al., 2015). Interestingly, the field-saturated YAK-09/-13/-15/-19

505   also belong to this area but are located ~20 km eastwards along another smaller fjord

506   (Enkelmann et al., 2015). These spatial differences in $IRSL_{50}$ intensities would suggest spatial

507   variations in the late-Quaternary exhumation history of this area that were not detected by

508   higher-temperature thermochronometers (McAleer et al., 2009; Enkelmann et al., 2015) but

15

509   may be evidenced in our dataset. Finally, YAK-03/-07 exhibit exceptionally low natural trap

510   filling levels (Figure 4), which may have been caused by hydrothermal reheating from crustal

511   fluid circulations in a highly-fractured area (Fairweather Fault; McAleer et al., 2009), rather

512   than by extremely rapid bedrock cooling. Further work beyond the scope of the present study

513   will focus on constraining thermal kinetic parameters for these samples to derive their thermal

514   histories.

515

516   In summary, our dataset confirms that feldspar $IRSL_{50}$ would be applicable only in very

517   rapidly-exhuming settings (>1 km Ma^{-1}; e.g. King et al., 2016) or in high-temperature

518   environments such as boreholes or tunnels (>35 °C; Guralnik et al., 2015a). Feldspar

519   anomalous fading, even if correctly accounted for, ultimately reduces the trapped charge

520   capacity in a given crystal because of athermal instability, and thus initiates a faster arrival at

521   field saturation. This might preclude the applicability of feldspar $IRSL_{50}$ in OSL

522   thermochronometry. MET-pIRIR protocols (Li and Li, 2011a,b) offer the advantage of multi-

523   thermochronometric systems in a single mineral (Qin et al., 2015), with promising outcomes in

524   providing robust constraints on bedrock cooling rates over sub-Quaternary timescales (King et

525   al., 2016). Other feldspar protocols such as thermally-transferred IRSL (Reimann et al., 2015)

526   or pulsed $IRSL_{50}$ (although the thermal stability of pulsed $IRSL_{50}$ remains to be experimentally

527   constrained; Tsukamoto et al., 2006; Roskosch et al., 2015; Jain et al., 2015b) may also be

528   interesting to explore for thermochronometric application, providing that these protocols show

529   good luminescence characteristics as well as low thermal stability for bedrock feldspar extracts.

530   Alternatively, quartz protocols based on thermally-transferred OSL (Duller and Wintle, 2012),

531   Violet Stimulated Luminescence (VSL, Ankjærgaard et al., 2013; 2015) or (red-

532   )thermoluminescence (TL or red TL, Schmidt et al., 2015; Tang and Li, 2015) could allow for

533   later saturation and thus a wider dating range in thermochronometry.

534

535   **5. Conclusions**

536   We report feldspar $IRSL_{50}$ luminescence characteristics, natural signal and anomalous

537   fading for 32 bedrock samples collected from a large variety of lithologies and exhumation

538   settings. Our results show that feldspar $IRSL_{50}$ signals from bedrock extracts are bright and

539   reproducible using established protocols in OSL dating. We measured a large variability in

540    $IRSL_{50}$ fading rates between samples. Our results show that it is essential to measure

541   anomalous fading and account for it on a sample-to-sample basis for OSL thermochronometry,

542   as it may otherwise obscure any thermochronometric information. Furthermore, we exploited

543 the wide range of observed fading rates to demonstrate the ability of electron tunnelling models

544 in quantitatively predicting the feldspar $IRSL_{50}$ level in field saturation for rather diverse

545 environmental conditions. While the potential causes for feldspar $IRSL_{50}$ fading still require

546 further investigation, our observations point towards a significant influence of Ca content in

547 feldspar on its anomalous fading rate. Finally, our results suggest that the applicability of

548 feldspar $IRSL_{50}$ in OSL thermochronometry would be limited to rapidly-exhuming settings (i.e.

549 >1 km Ma^{-1}) or warm subsurface environments (i.e. >35 °C).

550

561

562 **References**

563 Aitken, M.J., 1985. Thermoluminescence Dating. Academic Press, London. 359 pp.

564 Aitken, M.J., Xie, J., 1990. Moisture correction for annual gamma dose. Ancient TL 8, 6-9.

565 Aitken, M.J., 1998. An Introduction to Optical Dating: The Dating of Quaternary Sediments by

566 the Use of Photon-Stimulated Luminescence. Oxford University Press. 267 pp.

567 Ankjærgaard, C., Jain, M., Wallinga, J., 2013. Towards dating quaternary sediments using the

568 quartz violet stimulated luminescence (VSL) signal. Quaternary Geochronology 18, 99-109.

569 Ankjærgaard, C., Guralnik, B., Porat, N., Heimann, A., Jain, M., Wallinga, J., 2015. Violet

570 stimulated luminescence: geo-or thermochronometer? Radiation Measurements 81, 78-84.

571 Auclair, M., Lamothe, M., Huot, S., 2003. Measurement of anomalous fading for feldspar IRSL

572 using SAR. Radiation Measurements 37, 487-492.

573 Balescu, S., Lamothe, M., 1994. Comparison of TL and IRSL age estimates of feldspar coarse

574 grains from waterlain sediments. Quaternary Science Reviews 13, 437-444.

575 Barré, M., Lamothe, M., 2010. Luminescence dating of archaeosediments: A comparison of K-

576 feldspar and plagioclase IRSL ages. Quaternary Geochronology 5, 324-328.

577 Bateman, M., Swift, D.A., Piotrowski, J.A., Sanderson, D.C.W., 2012. Investigating the effects
578   of glacial shearing of sediment on luminescence. Quaternary Geochronology 10, 230-236.

579 Bell, W.T., 1980. Alpha dose attenuation in quartz grains for thermoluminescence dating.
580   Ancient TL 12, 4-8.

581 Berger, A.L., Spotila, J.A., Chapman, J.B., Pavlis, T.L., Enkelmann, E., Ruppert, N.A.,
582   Buscher, J.T., 2008. Architecture, kinematics, and exhumation of a convergent orogenic
583   wedge: A thermochronological investigation of tectonic-climatic interactions within the
584   central St. Elias Orogen, Alaska. Earth and Planetary Science Letters 270, 13-24.

585 Bøtter-Jensen, L., Thomsen, K.J., Jain, M., 2010. Review of optically stimulated lumi-nescence
586   (OSL) instrumental developments for retrospective dosimetry. Radiation Measurements 45,
587   253-257.

588 Braun, J., van der Beek, P., Batt, G., 2006. Quantitative thermochronology: numerical methods
589   for the interpretation of thermochronological data. Cambridge University Press. 258 pp.

590 Braun, J., van der Beek, P., Valla, P.G., Robert, X., Herman, F., 2012. Quantifying rates of
591   landscape evolution and tectonic processes by thermochronology and numerical modelling
592   of crustal heat transport using PECUBE. Tectonophysics 20, 1-28.

593 Brennan, B.J., Lyons, R.G., Phillips, S.W., 1991. Attenuation of alpha particle track dose for
594   spherical grains. International Journal of Radiation Applications and Instrumentation. Part
595   D. Nuclear Tracks and Radiation Measurements 18, 249-253.

596 Buylaert, J.P., Murray, A.S., Thomsen, K.J., Jain, M., 2009. Testing the potential of an elevated
597   temperature IRSL signal from K-feldspar. Radiation Measurements 44, 560-565.

598 Buylaert, J.P., Huot, S., Murray, A.S., van den Haute, P., 2011. Infrared stimulated
599   luminescence dating of an Eemian (MIS 5e) site in Denmark using K-feldspar. Boreas 40,
600   46-56.

601 Buylaert, J.-P., Jain, M., Murray, A.S., Thomsen, K.J., Thiel, C., Sohbati, R., 2012. A robust
602   feldspar luminescence dating method for Middle and Late Pleistocene sediments. Boreas 41,
603   435-451.

604 Buylaert, J.P., Jain, M., Murray, A.S., Thomsen, K.J., Thiel, C., Sohbati, R., 2012. A robust
605   feldspar luminescence dating method for Middle and Late Pleistocene sediments. Boreas 41,
606   435-451.

607 Champagnac, J.-D., Valla, P.G., Herman, F., 2014. Late-Cenozoic relief evolution under
608   evolving climate: A review. Tectonophysics 614, 44-65.

609 De Sarkar, S., Mathew, G., Pande, K., Chauhan, N., Singhvi, A., 2013. Rapid denudation of
610   Higher Himalaya during late Pliestocence, evidence from OSL thermochronology.
611   Geochronometria 40, 304-310.

612 Duller, G.A.T., 1992. Luminescence chronology of raised marine terraces, south-west North
613   Island, New Zealand. PhD thesis, University of Wales, Aberystwyth. 236 pp.

614 Duller, G.A.T., Wintle, A.G., 2012. A review of the thermally transferred optically stimulated
615   luminescence signal from quartz for dating sediments. Quaternary Geochronology 7, 6-20.

616 Enkelmann, E., Valla, P.G., Champagnac, J.-D., 2015. Low-temperature thermochronology of
617   the Yakutat Plate corner, St. Elias Range (Alaska): bridging short-term and long-term
618   deformation. Quaternary Science Reviews 123, 23-38.

619 Grün, R., 2001. Trapped charge dating (ESR, TL, OSL). In: Brothwell, D.R., Pollard, A.M.
620   (Eds.), Handbook of Archaeological Sciences. John Wiley & Sons Inc., West Sussex,
621   England, pp. 47-62.

622 Grün, R., Tani, A., Gurbanov, A., Koshchug, D., Williams, I., Braun, J., 1999. A new method
623   for the estimation of cooling and rates using paramagnetic centers in quartz: a case study on
624   the Eldzhurtinskiy Granite, Caucasus. Journal of Geophysical Research 104, 17531-17549.

625 Guérin, G., Mercier, N., 2012. Preliminary insight into dose deposition processes in
626   sedimentary media on a grain scale : Monte Carlo modelling of the effect of water on
627   gamma dose-rates. Radiation Measurements, 47, 541-547.

628 Guérin, G., Mercier, N., Nathan, R., Adamiec, G., Lefrais, Y., 2012. On the use of the infinite
629   matrix assumption and associated concepts: a critical review. Radiation Measurements 47,
630   778-785.

631 Guralnik, B., Jain, M., Herman, F., Paris, R.B., Harrison, T.M., Murray, A.S., Valla, P.V.,
632   Rhodes, E.J., 2013. Effective closure temperature in leaky and/or saturating
633   thermochronometers. Earth and Planetary Science Letters 384, 209-218.

634 Guralnik, B., Jain, M., Herman, F., Ankjærgaard, C., Murray, A.S., Valla, P.G., Preusser, F.,
635   King, G.E., Chen, R., Lowick, S.E., Kook, M., Rhodes, E.J., 2015a. OSL-thermochronology
636   of feldspar from the KTB Borehole, Germany. Earth and Planetary Science Letters 423,
637   232-243.

638 Guralnik, B., Ankjærgaard, C., Jain, M., Murray, A.S., Müller, A., Wälle, M., Lowick, S.E.,
639   Preusser, F., Rhodes, E.J., Wu, T.-S., Mathew, G., Herman, F., 2015b. OSL-
640   thermochronometry using bedrock quartz: a note of caution. Quaternary Geochronology 25,
641   37-48.

642   Guralnik, B., Li, B., Jain, M., Chen, R., Paris, R.B., Murray, A., Li, S.H., Pagonis, V., Valla,
643   P.G., Herman, F., 2015c. Radiation-induced growth and isothermal decay of infrared-
644   stimulated luminescene from feldspar. Radiation Measurements 81, 224-231.

645   Hendriks, B., Andriessen, P., Huigen, Y., Leighton, C., Redfield, T., Murrell, G., Gallagher, K.,
646   Nielsen, S.B., 2007. A fission track data compilation for Fennoscandia. Norwegian Journal
647   of Geology 87, 143-155.

648   Herman, F., Champagnac, J.-D., 2016. Plio-Pleistocene increase in erosion rates in mountain
649   belts in response to climate change. Terra Nova 28, 2-10.

650   Herman, F., Rhodes, E.J., Braun, J., Heiniger, L., 2010. Uniform erosion rates and re-lief
651   amplitude during glacial cycles in the Southern Alps of New Zealand, as revealed from
652   OSL-thermochronology. Earth and Planetary Science Letters 297, 183-189.

653   Herman, F., Seward, D., Valla, P.G., Carter, A., Kohn, B., Willett, S.D., Ehlers, T.A., 2013.
654   Worldwide acceleration of mountain erosion under a cooling climate. Nature 504, 423-426.

655   Hijmans, R.J., Cameron, S.E., Parra, J.L., Jones, P.G., Jarvis, A., 2005. Very high resolution
656   interpolated climate surfaces for global land areas. International Journal of Climatology 25,
657   1965-1978.

658   Huntley, D.J., Hutton, J.T., Prescott, J.R., 1993. Optical dating using inclusions within quartz
659   grains. Geology 21, 1087-1090.

660   Huntley, D.J., Baril, M.R., 1997. The K content of the K-feldspars being measured in optical
661   dating or in thermoluminescence dating. Ancient TL 15, 11-13.

662   Huntley, D.J., Lamothe, M., 2001. Ubiquity of anomalous fading in K-feldspars and the
663   measurement and correction for it in optical dating. Canadian Journal of Earth Sciences 38,
664   1093-1106.

665   Huntley, D.J., 2006. An explanation of the power-law decay of luminescence. Journal of
666   Physics: Condensed Matter 18, 1359-1365.

667   Huntley, D.J., Lian, O.B., 2006. Some observations on tunnelling of trapped electrons in
668   feldspars and their implications for optical dating. Quaternary Science Reviews 25, 2503-
669   2512.

670   Huntley, D.J., Baril, M.R., Haidar, S., 2007. Tunnelling in plagioclase feldspars. Journal of
671   Physics D: Applied Physics 40, 900-906.

672   Huntley, D.J., 2011. Comment on "Isochron dating of sediments using luminescence of K-
673   feldspar grains" by B. Li et al. Journal of Geophysical Research 116, F01012.

674 Jain, M., Guralnik, B., Andersen, M.T., 2012. Stimulated luminescence emission arising from
675   localized recombination within randomly distributed defects. Journal of Physics Condensed
676   Matter 24, 385402.

677 Jain, M., Sohbati, R., Guralnik, B., Murray, A.S., Kook, M., Lapp, T., Prasad, A.K., Thomsen,
678   K.J., Buylaert, J.P, 2015a. Kinetics of infrared stimulated luminescence from feldspars.
679   Radiation Measurements 81, 242-250.

680 Jain, M., Buylaert, J.P., Thomsen, K.J., Murray, A.S., 2015b. Further investigations on 'non-
681   fading' in K-Feldspar. Quaternary International 362, 3-7.

682 Kars, R.H., Wallinga, J., Cohen, K.M., 2008. A new approach towards anomalous fading
683   correction for feldspar IRSL dating - tests on samples in field saturation. Radiation
684   Measurements 43, 786-790.

685 Kars, R.H., Wallinga, J., 2009. IRSL dating of K-feldspars: Modelling natural dose response
686   curves to deal with anomalous fading and trap competition. Radiation Measurements 44,
687   594-599.

688 Kars, R.H., Reimann, T., Wallinga, J., 2014. Are feldspar SAR protocols appropriate for post-
689   IR IRSL dating? Quaternary Geochronology 22, 126-136.

690 King, G.E., Herman, F., Lambert, R., Valla, P.G., Guralnik, B., 2016. Multi-OSL-
691   thermochronometry of feldspar. Quaternary Geochronology 33, 76-87

692 King, G.E., Robinson, R.A.J., Finch, A.A., 2014. Towards successful OSL sampling strategies
693   in glacial environments: deciphering the residual OSL signals of quartz and K-feldspar from
694   modern glacial sediments. Quaternary Science Reviews 89, 94-107.

695 Kook, M.H., Lapp, T., Murray, A.S., Thiel, C., 2012. A Risø XRF attachment for major
696   element analysis of aliquots of quartz and feldspar separates. UK Luminescence and ESR
697   Meeting - Aberystwyth, p. 37.

698 Koshchug, D.G., Solovyov, Y.P., 1998. Accumulation of structural radiation defects in quartz
699   in cooling systems: Basis for dating. Physics and Chemistry of Minerals 25, 242-248.

700 Lamothe, M., Auclair, M., 1999. A solution to anomalous fading and age shortfalls in optical
701   dating of feldspar minerals. Earth and Planetary Science Letters 171, 319-323.

702 Lamothe, M., Auclair, M., Hamzaoui, C., Huot, S., 2003. Towards a prediction of long-term
703   anomalous fading of feldspar IRSL. Radiation Measurements 37, 493-498.

704 Li, S-H., Tso, M-Y.W., Wong, N.W.L., 1997. Parameters of OSL traps determined with
705   various linear heating rates. Radiation Measurements 27, 43-47.

706 Li, B., Li, S.-H., 2006. Comparison of De estimates using the fast component and the medium
707   component of quartz OSL. Radiation Measurements 41, 125-136.

708 Li, B., Li, S.-H., 2008. Investigations of the dose-dependent anomalous fading rate of feldspar
709   from sediments. Journal of Physics D: Applied Physics 41, 225502.

710 Li, B., Li, S.-H., 2011a. Luminescence dating of K-feldspar from sediments: a protocol without
711   anomalous fading correction. Quaternary Geochronology 6, 468-479.

712 Li, B., Li, S.-H., 2011b. Thermal stability of infrared stimulated luminescence of sedimentary
713   K-feldspar. Radiation Measurements 46, 29-36.

714 Li, B., Li, S.-H., 2012. Determining the cooling age using luminescence-thermochronology.
715   Tectonophysics 580, 242-248.

716 Li, B., Jacobs, Z., Roberts, R.G., Li, S.H., 2014. Review and assessment of the potential of
717   post-IR IRSL dating methods to circumvent the problem of anomalous fading in feldspar
718   luminescence. Geochronometria 41, 178-201.

719 McAleer, R.J., Spotila, J.A., Enkelmann, E., Berger, A.L., 2009. Exhumation along the
720   Fairweather Fault, Southeast Alaska, based on Low-Temperature thermochronometry.
721   Tectonics 28, TC1007.

722 Nielsen, S.B., Gallagher, K., Leigton, C., Balling, N., Svenningsen, L., Jacobsen, B.H.,
723   Thomsen, E., Nielsen, O.B., Heilmann-Clausen, C., Egholm, D.L., Summerfield, M.A.,
724   Clausen, O.R., Piotrowski, J.A., Thorsen, M.R., Huuse, M., Abrahamsen, N., King, C.,
725   Lykke-Andersen, H., 2009. The evolution of western Scandinavian topography: a review of
726   Neogene uplift versus the ICE (isostasy-climate-erosion) hypothesis. Journal of
727   Geodynamics 47, 72-95.

728 Parish, R., 1992. The application of sedimentological analysis and luminescence dating to
729   waterlain deposits from archaeological sites. PhD Thesis, Durham University, 263 pp.

730 Preusser, F., Chithambo, M.L., Götte, T., Martini, M., Ramseyer, K., Sendezera, E.J., Susino,
731   G.J., Wintle, A.G., 2009. Quartz as a natural luminescence dosimeter. Earth Science
732   Reviews 97, 184-214.

733 Prokein, J., Wagner, G.A., 1994. Analysis of thermoluminescent glow peaks in quartz derived
734   from the KTB-drill hole. Radiation Measurements 23, 85-94.

735 Qin, J., Chen, J., Valla, P.G., Herman, F., Li, K., 2015. Estimating rock cooling rates by using
736   multiple luminescence thermochronometers. Radiation Measurements 79, 13-18.

737 Reimann, T., Ankjærgaard, C., Wallinga, J., 2015. Testing the potential of a transferred IRSL
738   (T-IRSL) feldspar signal for luminescence dating. Radiation Measurements 81, 275-281.

739 Reiners, P.W., Brandon, M.T., 2006. Using thermochronology to understand orogenic erosion.
740   Annual Review of Earth and Planetary Sciences 34, 419-466.

741 Rhodes, E.J., 2011. Optically stimulated luminescence dating of sediments over the past
742   200,000 years. Annual Review of Earth and Planetary Sciences 39, 461-488.

743 Roskosch, J., Winsemann, J., Polom, U., Brandes, C., Tsukamoto, S., Weitkamp, A.,
744   Bartholomäus, W.A., Henningsen, D., Frechen, M., 2015. Luminescence dating of ice-
745   marginal deposits in northern Germany: evidence for repeated glaciations during the Middle
746   Pleistocene (MIS 12 to MIS 6). Boreas 44, 103-126.

747 Schmidt, C., Friedrich, J., Zöller, L., 2015. Thermochronometry using red TL of quartz? e
748   Numerical simulation and observations from in-situ drill-hole samples. Radiation
749   Measurements 81, 98-103.

750 Shuster, D.L., Farley, K.A., 2005. ^{4}He/^{3}He Thermochronometry: Theory, Practice, and
751   Potential Complications. Reviews in Mineralogy and Geochemistry 58, 181-203.

752 Shuster, D.L., Ehlers, T.A., Rusmore, M.E., Farley, K.A., 2005. Rapid glacial erosion at 1.8
753   Ma revealed by ^{4}He/^{3}He thermochronometr. Science 310, 1668-1670.

754 Shuster, D.L., Cuffey, K.M., Sanders, J.W., Balco, G., 2011. Thermochronometry Reveals
755   Headward Propagation of Erosion in an Alpine Landscape. Science 332, 84-88.

756 Sohbati, R., Murray, A., Jain, M., Thomsen, K., Hong, S.C., Yi, K., Choi, J.H., 2013. Na-rich
757   feldspar as a luminescence dosimeter in infrared stimulated luminescence (IRSL) dating.
758   Radiation Measurements 51-52, 67-82.

759 Spooner, N.A., 1994. The anomalous fading of infra-red stimulated luminescence from
760   feldspars. Radiation Measurements 23, 625-632.

761 Spotila, J.A., Buscher, J.T., Meigs, A.J., Reiners, P.W., 2004. Long-term glacial erosion of
762   active mountain belts: Example of the Chugach-St. Elias Range, Alaska. Geology 32, 501-
763   504.

764 Steer, P., Huismans, R.S., Valla, P.G., Gac, S., Herman, F., 2012. Bimodal Plio-Quaternary
765   glacial erosion of fjords and low-relief surfaces in Scandinavia. Nature Geoscience 5, 635-
766   639.

767 Tang, S.L., Li, S.-H., 2015. Low temperature thermochronology using thermoluminescence
768   signals from quartz. Radiation Measurements 81, 92-97.

769 Thomsen, K.J., Murray, A.S., Jain, M., Bøtter-Jensen, L., 2008. Laboratory fading rates of
770   various luminescence signals from feldspar-rich sediment extracts. Radiation Measurements
771   43, 1474-1486.

772 Thomsen, K., Murray, A., Jain, M., 2011. Stability of IRSL signals from sedimentary K-
773   feldspar samples. Geochronometria 38, 1-13.

774 Tso, M-Y.W., Wong, N.W.L., Li, S-H., 1996. Determination of life time of infrared stimulated
775   signals from potassium and sodium feldspars. Radiation Protection Dosimetry 66, 387-389.

776 Tsukamoto, S., Denby, P.M., Murray, A.S., Bøtter-Jensen, L., 2006. Time-resolved
777   luminescence from feldspars: New insight into fading. Radiation Measurements 41, 790-
778   795.

779 Valla, P.G., Shuster, D.L., van der Beek, P.A., 2011a. Significant increase in relief of the
780   European Alps during Mid-Pleistocene glaciations. Nature Geoscience 4, 688-692.

781 Valla, P.G., van der Beek, P.A., Braun, J., 2011b. Rethinking low-temperature
782   thermochronology data sampling strategies for quantification of denudation and relief
783   histories: a case study in the French western Alps. Earth and Planetary Science Letters 307,
784   309-322.

785 Visocekas, R., 2002. Tunneling in afterglow, its coexistence and interweaving with thermally
786   stimulated luminescence. Radiation Protection Dosimetry 100, 45-54.

787 Wagner, G.A., Reimer, G.M., 1972. Fission track tectonics: the tectonic interpretation of
788   fission track apatite ages. Earth and Planetary Science Letters 14, 263-268.

789 Wallinga, J., Murray, A.S., Wintle, A., 2000. The single-aliquot regenerative-dose (SAR)
790   protocol applied to coarse-grain feldspar. Radiation Measurements 32, 529-533.

791 Willenbring, J.K., Jerolmack, D.J., 2016. The null hypothesis: globally steady rates of erosion,
792   weathering fluxes and shelf sediment accumulation during Late Cenozoic mountain uplift
793   and glaciation. Terra Nova 28, 11-18.

794 Wintle, A.G., Murray, A.S., 2006. A review of quartz optically stimulated luminescence
795   characteristics and their relevance in single-aliquot regeneration dating protocols. Radiation
796   Measurements 41, 369-391.

797 Wintle, A.G., 2008. Luminescence dating: where it has been and where it is going. Boreas 37,
798   471-482.

799 Wu, T.S., Kunz, A., Jaiswal, M.K., Chen, Y.G., 2012. A feasibility study on the application of
800   luminescence dating for quartz from different rock types as a thermochronometer.
801   Quaternary Geochronology 10, 340-344.

802 Wu, T.-S., Jain, M., Guralnik, B., Murray, A.S., Chen, Y.-G., 2015. Luminescence
803   characteristics of quartz from Hsuchshan Range (Central Taiwan) and the implications as a
804   thermochronometer. Radiation Measurements 81, 104-109.

805 Yi, S., Buylaert, J.-P., Murray, A.S., Lu, H., Thiel, C., Zeng, L., in press. A detailed post-IR
806   IRSL study of the Niuyangzigou loess site in northeastern China. Boreas.

807

808 **Appendix A. Supplementary material**

809 Supplementary material, including details about the dose rate scenarios (Table S1), full

810 geochemical reports (Tables S2-S4), supplementary experimental results (Table S5) and all

811 sample raw data (Table S6) related to this article can be found on-line at ...

812

813

814 **Figure captions**

815 **Figure 1.** $IRSL_{50}$ dose-response curves (left panels) and fading experiments (central and right

816 panels) for representative samples GRA-08 (A-B-C), GRA-12 (D-E-F), SOG-06 (G-H-I) and

817 YAK-09 (J-K-L). See Tables 2 and 3 for experimental details and results, respectively. The

818 dose-response curves (left panels) have been obtained following the single aliquot

819 regenerative-dose (SAR) protocol for 3 individual aliquots. The natural luminescence signal

820 (white stars) and each regenerative dose (black circles) are normalised to a given test dose, and

821 used to build the dose-response curve (fitted by the GOK approach, black line and grey

822 envelope; α: electron-trapping order, D_0: characteristic dose) and to calculate the sample

823 natural luminescence level (L_n/T_n) and natural trap filling $(n/N)_{nat}$ (Guralnik et al., 2015a).

824 Fading experiments, following the long-shine (central panels) and short-shine (right panels)

825 protocols, were used to derive the sample fading rate (sample-specific g_{2days} are given by the

826 slope of the black lines normalized to 2 days; Huntley and Lamothe, 2001).

827

828 **Figure 2.** Comparison of fading rate measurements (g_{2days}) using the short-shine and long-shine

829 methods (Table 3). The symbols represent individual GRA (black circles), YAK (grey squares)

830 and SOG (open triangles) samples. The dashed line defines the 1:1 relationship. The inset

831 shows a zoom-in to the low-fading rate region (dashed box in main panel).

832

833 **Figure 3.** Natural trap filling $(n/N)_{nat}$ and g_{2days} values (long-shine method, Table 3) for GRA

834 (black circles), YAK (grey squares) and SOG (open triangles) samples. Samples in field

835 saturation define a well-characterised inverse relationship between laboratory-measured fading

836 rates (g_{2days}) and the natural trap filling $(n/N)_{nat}$ values over a wide range of fading rates (from

837 ~0.5 to ~45 %/decade).

838

839 **Figure 4.** Predicted field saturation $(n/N)_{ss}$ vs. measured natural trap filling $(n/N)_{nat}$ for GRA

840 (circles), YAK (squares) and SOG (triangles) samples (A: lineal plot; B: logarithmic plot).

841 Symbols are coloured according to the individual sample g_{2days} values (see legend). Most of

842 analysed samples plot along the 1:1 relationship (dashed line), validating the approach of
843 Guralnik et al. (2015a) to numerically predict sample field saturation (after Huntley, 2006 and
844 Kars et al., 2008). Some samples have $(n/N)_{nat}$ below $0.85(n/N)_{ss}$ (dotted line), bearing potential
845 thermochronometric information. See text for details and discussion.

846

847 **Figure 5.** Feldspar geochemistry and laboratory-measured fading rate. (A) Whole-rock
848 representative oxides showing the lithological variability between analysed samples
849 (Supplementary Table S2). (B) Feldspar mineralogical compositions for a subset of samples
850 (Ab: albite; An: anorthite; Or: orthoclase; Supplementary Table S4). GRA: circles; YAK:
851 squares; SOG: triangles. Symbols are coloured according to the respective sample's g_{2days} value
852 (see legend). See text for discussion.

Table 1. Overview of sample locations, lithology and climate, alongside measured radionuclides and calculated natural dose rate.

Sample	Latitude/ Longitude (°N)/(°W)	Elevation (m)	Lithology	Mean Annual Temperature[1] (°C)	Whole-rock geochemistry[2]			Feldspar minimum K_{int}[3] (%)	Natural dose rate D_{rate}[4] (Gy ka^{-1})
					U (ppm)	Th (ppm)	K (wt. %)		
Granite Range (southern Alaska)									
GRA-03	60.853/ 141.451	1419	Diorite	-5.8	0.4	1.1	0.79	0.7 [a]	1.12 (±0.24)
GRA-04	60.832/141.467	1761	Diorite	-4.5	0.2	0.6	0.82	1.7 [a]	1.22 (±0.23)
GRA-05	60.835/141.469	1995	Diorite	-6.3	0.4	1.1	0.86	1.4 [a]	1.31 (±0.14)
GRA-06	60.841/141.501	2388	Diorite	-6.8	0.3	0.5	0.76	1.0 [a]	1.06 (±0.14)
GRA-08	60.905/141.970	2188	Diorite	-7.3	0.3	0.8	0.97	1.4 [a]	1.34 (±0.13)
GRA-09	60.901/141.954	1691	Diorite	-5.7	0.1	0.1	0.50	0.8 [a]	0.70 (±0.15)
GRA-10	60.900/141.934	1412	Diorite	-4.2	0.3	1.1	2.02	3.9 [a]	2.72 (±0.53)
GRA-11	60.908/141.919	970	Diorite	-3	0.6	1.1	0.81	1.4 [a]	1.33 (±0.16)
GRA-12	60.991/142.763	1554	Schist	-2.5	0.3	0.5	1.03	1.9 [b]	1.46 (±0.22)
GRA-13	61.003/142.741	1253	Granitoïde	-1.5	1.2	9.4	0.72	1.9 [b]	2.37 (±0.61)
GRA-14	60.991/142.782	1979	Granitoïde	-4.2	0.9	1.3	1.09	1.9 [b]	1.79 (±0.20)
GRA-17	60.943/142.475	1818	Granitoïde	-6.1	0.3	0.5	0.61	1.9 [b]	1.11 (±0.35)
GRA-18	60.957/142.378	1395	Schist	-4.1	0.1	0.3	0.61	1.9 [b]	1.02 (±0.39)
GRA-19	61.008/142.362	720	Granitoïde	-1.4	1.6	3.6	0.73	1.9 [b]	1.95 (±0.39)
GRA-BR	61.109/142.420	392	Monzogranite	-0.2	1.6	3.9	2.17	4.2 [a]	3.62 (±0.49)
Sognefjord (western Norway)									
SOG-02	61.096/-5.681	33	Granitic gneiss	7.4	1.8	10.4	4.18	12.5 [c]	8.45 (±3.44)
SOG-06	61.155/-6.461	883	Granitic gneiss	2.4	2.2	8.3	5.13	12.5 [c]	9.01 (±3.01)
SOG-10	61.146/-6.542	47	Granitic gneiss	6.5	1.1	10.3	4.76	12.5 [c]	8.68 (±3.24)
SOG-17	61.233/-7.593	961	Granodiorite	1.1	0.2	0.2	4.13	12.5 [c]	7.51 (±3.94)
SOG-21	61.239/-7.664	10	Anorthosite	3.5	0.7	10.5	5.15	12.5 [c]	8.87 (±3.10)
SOG-22	61.263/-7.867	1274	Gabbro	-0.9	0.1	0.8	0.52	1.9 [b]	1.00 (±0.40)
SOG-25	61.272/-7.765	48	Anorthosite	4.8	0.6	5.5	5.30	12.5 [c]	8.65 (±3.19)
SOG-38	61.151/-6.728	21	Granitic gneiss	7.0	0.4	3.7	5.13	12.5 [c]	8.40 (±3.34)

Yakutat terrane (southern Alaska)									
YAK-03	59.813/138.912	5	Diorite	0.3	0.1	0.3	0.18	0.2 [a]	0.34 (±0.17)
YAK-07	59.843/139.162	2	Diorite	1.6	0.1	0.1	0.04	1.9 [b]	0.33 (±0.22)
YAK-09	59.825/138.986	0	Diorite	2.3	2.5	4.4	1.72	2.2 [a]	3.24 (±0.70)
YAK-13	59.647/139.296	2	Sandstone	3.5	1.7	6.3	1.37	1.9 [b]	2.80 (±0.63)
YAK-15	59.851/139.313	1	Sandstone	3.0	2	4.8	1.53	1.9 [b]	2.90 (±0.63)
YAK-17	59.974/139.450	2	Sandstone	2.2	1.4	3.9	0.62	1.9 [b]	1.74 (±0.39)
YAK-18	59.967/139.531	1	Sandstone	2.9	1.2	3.5	1.87	1.9 [b]	2.80 (±0.44)
YAK-19	59.597/139.342	2	Sandstone	3.2	1.6	3.2	1.89	4.2 [a]	3.12 (±0.37)
YAK-50	59.946/139.620	0	Sandstone	2.6	1.2	3.5	1.11	1.9 [b]	2.08 (±0.37)

Notes. [1] Derived from Worldclim database (Hijmans et al., 2005). [2] Measured on a Perkin Elmer Sciex ELAN 6100/9000 ICP-MS, with an estimated analytical precision of 2.3 (U), 3.4% (Th) and 1.2% (K). See Supplementary Tables S2-S3 for all elements and further details. [3] Measured or estimated as follows: [a] measured using the Risø XRF attachment (Kook et al., 2012); [b] assumed value for Na/Ca-feldspars (average of the Risø XRF data above) [c] assumed value for K-feldspars (Huntley and Baril, 1997; Barré and Lamothe, 2010) [4] For calculation of feldspar dose rates, we used conversion factors of Guérin et al. (2012), water content of 2(±2)%, and negligible cosmic radiation. The reported dose rates cover two (or four) end-member scenarios after Guralnik et al. 2015a (see Table S1). For K-feldspars, two end-member grain sizes were considered (180 and 1000 or 2500 μm); for Na/Ca-feldspars, two additional scenarios, in which the entire luminescence signal originates from K-feldspar inclusions (1 or 100 μm) were further included (Table S1).

Table 2. Laboratory protocols for luminescence measurements of feldspar IRSL$_{50}$.

Dose response SAR protocol[a]	Anomalous fading	
	SAR protocol, (Long-shine method[b])	Short-shine method[c]
	1. IRSL at 290 ℃ (200 s)	1. IRSL at 290 ℃ (200 s)
1. Give dose D_x	2. Give dose D_{fading}	2. Give dose D_{fading}
2. Preheat (250 ℃ for 60 s)	3. Preheat (250 ℃ for 60 s)	3. Preheat (250 ℃ for 60 s)
	4. Hold at room T ℃ for t_x s	4. Hold at room T ℃ for t_x s
3. IRSL at 50 ℃ (200 s) $\rightarrow L_x$	5. IRSL at 50 ℃ (200 s) $\rightarrow L_x$	5. IRSL at 50 ℃ (0.1 s) $\rightarrow L_x$
		6. Return to step 4
		7. IRSL at 290 ℃ (200 s)
4. Give test dose D_{test}	6. Give test dose D_{test}	8. Give test dose D_{test}
5. Preheat (250 ℃ for 60 s)	7. Preheat (250 ℃ for 60 s)	9. Preheat (250 ℃ for 60 s)
6. IRSL at 50 ℃ (200 s) $\rightarrow T_x$	8. IRSL at 50 ℃ (200 s) $\rightarrow T_x$	10. IRSL at 50 ℃ (0.1 s) $\rightarrow T_x$
7. IRSL at 290 ℃ (200 s)		
8. Return to step 1	9. Return to step 1	11. Return to step 10

Protocols adapted from Guralnik et al. (2015a). [a] The irradiation doses of the SAR dose response protocol (Wallinga et al., 2000) were D_x = [0, 22, 44, 87, 175, 349, 698, 1396, 2792, 0, 87] Gy, and the test dose was D_{test} = 44 Gy. The first and the penultimate zero-doses (no irradiation) extract the natural dose and signal recuperation, respectively. [b] The irradiation and test doses of the long-shine fading experiment (Huntley and Lamothe, 2001) were D_{fading} = 240 Gy and D_{test} = 44 Gy, respectively, and the holding times were $t_x \approx$ [1200, 1200, 1200, 136660, 1200, 7970, 1200, 143450, 1200, 1200 1200] s. [c] The short-shine fading experiment (Auclair et al., 2003) consisted of a single irradiation dose D_{fading} = 72 Gy which was given, followed by delay times of $t_x \approx 10^{2.34+0.37x}$ s, where x = [1, 2, ..., 8]. The test dose was D_{test} = 72 Gy. L_x and T_x in both SAR protocols (left and central column) were derived by integrating the first 15 s of the stimulation curve (signal) after subtraction of the last 100 s (background). L_x and T_x of the short-shine experiment (right column) were derived by integrating the entire stimulation period of 0.1 s, after subtraction of a 20 ms background (immediately following the IRSL stimulation). *Note:* All preheats were done with a linear heating rate of 5 ℃ s^{-1}.

Table 3. Best-fit parameters and model results for feldspar $IRSL_{50}$.

Sample	D_0 (Gy)	Electron-trapping order (α)	Long-shine g_{2days} (%/decade)	Long-shine $\log_{10}\rho'$	Short-shine g_{2days} (%/decade)	Short-shine $\log_{10}\rho'$	Observed $(n/N)_{nat}$	Predicted $(n/N)_{ss}$
			Fitted kinetic parameters				**Interpretation**	
GRA-03	1208 (±59)	4	41.3 (±1.2)	-4.69 (±0.01)	47.7 (±4.5)	-4.68 (±0.03)	0.003 (±0.003)	0.002 (±0.0002)
GRA-04	1208 (±75)	4	28.5 (±0.7)	-4.81 (±0.01)	Low signal-to-noise ratio		0.007 (±0.005)	0.007 (±0.001)
GRA-05	550 (±71)	2.56 (±0.82)	28.0 (±1.0)	-4.82 (±0.01)	23.5 (±3.1)	-4.89 (±0.04)	0.017 (±0.003)	0.017 (±0.004)
GRA-06	612 (±29)	1.64 (±0.26)	36.2 (±1.4)	-4.74 (±0.01)	23.3 (±3.2)	-4.89 (±0.05)	0.008 (±0.002)	0.012 (±0.002)
GRA-08	599 (±31)	1.60 (±0.28)	33.8 (±0.8)	-4.75 (±0.01)	29.9 (±6.3)	-4.82 (±0.07)	0.015 (±0.003)	0.009 (+0.002/-0.003)
GRA-09	718 (±115)	2.11 (±0.86)	23.6 (±0.6)	-4.88 (±0.01)	22.2 (±3.2)	-4.92 (±0.05)	0.016 (±0.007)	0.022 (±0.006)
GRA-10	519 (±36)	1.67 (±0.38)	12.9 (±0.3)	-5.10 (±0.01)	14.3 (±1.7)	-5.07 (±0.04)	0.120 (±0.027)	0.098 (+0.016/-0.012)
GRA-11	606 (±73)	4	11.1 (±0.6)	-5.16 (±0.02)	Low signal-to-noise ratio		0.074 (±0.015)	0.113 (+0.011/-0.012)
GRA-12	545 (±10)	2.92 (±0.12)	13.9 (±0.3)	-5.08 (±0.01)	14.2 (±1.3)	-5.07 (±0.03)	0.092 (±0.005)	0.074 (+0.009/-0.007)
GRA-13	132 (±3)	1.62 (±0.08)	0.5 (±0.3)	-6.50 (±0.26)	Low signal-to-noise ratio		0.845 (±0.024)	0.903 (+0.054/-0.023)
GRA-14	243 (±4)	1.76 (±0.07)	3.4 (±0.3)	-5.65 (±0.04)	3.7 (±1.1)	-5.60 (±0.13)	0.589 (±0.017)	0.512 (+0.078/-0.061)
GRA-17	171 (±2)	3.02 (±0.11)	3.0 (±0.6)	-5.70 (±0.09)	Low signal-to-noise ratio		0.432 (±0.015)	0.567 (+0.041/-0.093)
GRA-18	245 (±3)	1.99 (±0.07)	7.5 (±0.4)	-5.32 (±0.02)	Low signal-to-noise ratio		0.276 (±0.013)	0.260 (+0.013/-0.020)
GRA-19	875 (±36)	4	9.1 (±2.0)	-5.24 (±0.09)	Low signal-to-noise ratio		0.097 (±0.014)	0.153 (+0.069/-0.037)
GRA-BR	433 (±11)	4	5.0 (±0.5)	-5.49 (±0.04)	Low signal-to-noise ratio		0.234 (±0.036)	0.376 (+0.028/-0.037)
SOG-02	688 (±16)	1.57 (±0.13)	8.6 (±0.2)	-5.26 (±0.01)	Low signal-to-noise ratio		0.172 (±0.010)	0.229 (+0.015/-0.017)
SOG-06	613 (±8)	1.41 (±0.07)	4.3 (±0.1)	-5.54 (±0.01)	5.1 (±1.0)	-5.47 (±0.08)	0.409 (±0.044)	0.444 (+0.09/-0.019)
SOG-10	588 (±10)	1.75 (±0.09)	4.6 (±0.1)	-5.52 (±0.01)	4.6 (±0.7)	-5.52 (±0.06)	0.403 (±0.050)	0.444 (+0.010/-0.013)
SOG-17	384 (±6)	1.87 (±0.09)	3.7 (±0.1)	-5.61 (±0.01)	5.2 (±0.3)	-5.47 (±0.02)	0.294 (±0.019)	0.448 (+0.006/-0.005)
SOG-21	717 (±10)	2.21 (±0.08)	8.5 (±0.1)	-5.27 (±0.01)	11.1 (±0.5)	-5.16 (±0.02)	0.164 (±0.009)	0.180 (+0.011/-0.010)
SOG-22	236 (±5)	1.30 (±0.06)	4.0 (±0.3)	-5.57 (±0.03)	3.2 (±0.4)	-5.67 (±0.05)	0.519 (±0.022)	0.480 (+0.049/-0.023)
SOG-25	567 (±12)	2.11 (±0.13)	5.2 (±1.3)	-5.47 (±0.10)	4.8 (±2.1)	-5.50 (±0.18)	0.359 (±0.016)	0.408 (+0.057/-0.108)
SOG-38	552 (±9)	2.19 (±0.11)	3.9 (±1.1)	-5.58 (±0.12)	4.5 (±0.5)	-5.52 (±0.04)	0.439 (±0.020)	0.446 (+0.049/-0.027)

YAK-03	349 (±15)	1.72 (±0.23)	12.6 (±1.6)	-5.12 (±0.05)	Low signal-to-noise ratio		0.034 (±0.008)	0.094 (+0.039/-0.020)
YAK-07	276 (±6)	1.51 (±0.08)	4.1 (±0.4)	-5.56 (±0.04)	4.1 (±2.3)	-5.56 (±0.24)	0.011 (±0.008)	0.443 (+0.112/-0.132)
YAK-09	194 (±3)	1.70 (±0.06)	3.3 (±0.3)	-5.65 (±0.04)	3.1 (±0.3)	-5.67 (±0.05)	0.549 (±0.018)	0.577 (+0.022/-0.039)
YAK-13	431 (±13)	2.44 (±0.21)	5.5 (±0.4)	-5.44 (±0.03)	7.4 (±1.7)	-5.32 (±0.09)	0.250 (±0.023)	0.289 (+0.046/-0.025)
YAK-15	271 (±11)	2.32 (±0.29)	4.1 (±0.5)	-5.56 (±0.05)	Low signal-to-noise ratio		0.427 (±0.071)	0.447 (+0.045/-0.024)
YAK-17	443 (±22)	4	6.8 (±0.5)	-5.36 (±0.03)	Low signal-to-noise ratio		0.125 (±0.036)	0.254 (+0.017/-0.020)
YAK-18	405 (±7)	2.24 (±0.11)	8.1 (±0.4)	-5.29 (±0.02)	5.5 (±1.0)	-5.44 (±0.08)	0.204 (±0.007)	0.282 (+0.024/-0.020)
YAK-19	905 (±13)	4	7.2 (±0.6)	-5.33 (±0.03)	Low signal-to-noise ratio		0.121 (±0.014)	0.233 (+0.019/-0.022)
YAK-50	390 (±7)	2.36 (±0.12)	4.1 (±0.3)	-5.57 (±0.03)	3.3 (±1.1)	-5.66 (±0.15)	0.124 (±0.005)	0.476 (+0.090/-0.038)

Notes. Dose response curves (with recycling ratio within 10% of unity) have been fitted using the General-Order Kinetics GOK function (Guralnik et al., 2015a,c), to obtain the characteristic dose D_0 and the kinetic order α (note: a limiting value of $\alpha = 4$ was adopted for a few datasets which did not exhibit sufficient flattening off of the luminescence signal at high doses). Fading decay curves were fitted (a) following Huntley and Lamothe (2001) to obtain the g-value (normalized to 2 days); (b) following Huntley (2006) to obtain the density of recombination centres ρ' (reported as $\log_{10}\rho'$), assuming an escape frequency factor of 3×10^{15} s^{-1} (Huntley, 2006). Results of the short-shine method were accepted only when there existed two replicates with acceptable signal to noise ratio (luminescence signal ten times higher than background). Natural trap filling (n/N_{nat}; reported as mean and std. dev.) and predicted field saturation (n/N_{ss}; reported as median and 68% conf. int.) were derived following Guralnik et al. (2015a), the latter using a Monte-Carlo approach (1000 iterations, each with randomized input of the kinetic parameters and the dose rate; note that wherever possible, an average ρ' between the long- and short-shine experiments was used). Laboratory dose rates vary between 0.099 and 0.207 Gy s^{-1} for the different instruments used.

GRA-08
$\alpha = 1.60 \pm 0.28$
$D_0 = 599 \pm 31$ Gy
$L_n/T_n = 0.07 \pm 0.01$
$(n/N)_{nat} = 0.02 \pm 0.003$

(A)

$g_{2days} = 33.8 \pm 0.8$ %/decade (B)

$g_{2days} = 29.9 \pm 6.3$ %/decade (C)

GRA-12
$\alpha = 2.92 \pm 0.12$
$D_0 = 545 \pm 10$ Gy
$L_n/T_n = 0.21 \pm 0.001$
$(n/N)_{nat} = 0.09 \pm 0.01$

(D)

$g_{2days} = 13.9 \pm 0.3$ %/decade (E)

$g_{2days} = 14.2 \pm 1.3$ %/decade (F)

SOG-06
$\alpha = 1.41 \pm 0.07$
$D_0 = 603 \pm 8$ Gy
$L_n/T_n = 0.55 \pm 0.02$
$(n/N)_{nat} = 0.41 \pm 0.04$

(G)

$g_{2days} = 4.3 \pm 0.1$ %/decade (H)

$g_{2days} = 5.1 \pm 1.0$ %/decade (I)

YAK-09
$\alpha = 1.70 \pm 0.06$
$D_0 = 194 \pm 3$ Gy
$L_n/T_n = 0.67 \pm 0.01$
$(n/N)_{nat} = 0.55 \pm 0.02$

(J)

$g_{2days} = 3.3 \pm 0.3$ %/decade (K)

$g_{2days} = 3.1 \pm 0.3$ %/decade (L)