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Carlos Arce Chamorro, Guillaume Guérin. Comparison of De values from Late Pleistocene alluvial deposits on the coast of Galicia (NW Spain) using BayLum or Analyst-based procedures. *Quaternary Geochronology*, 2024, 82, pp.101540. 10.1016/j.quageo.2024.101540 . insu-04575539

**HAL Id: insu-04575539**

**<https://insu.hal.science/insu-04575539>**

Submitted on 15 May 2024

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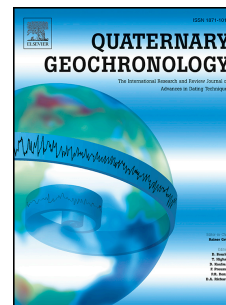


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# Journal Pre-proof

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PII: S1871-1014(24)00044-X

DOI: <https://doi.org/10.1016/j.quageo.2024.101540>

Reference: QUAGEO 101540

To appear in: *Quaternary Geochronology*

Received Date: 9 November 2023

Revised Date: 27 March 2024

Accepted Date: 7 May 2024

Please cite this article as: Chamorro, C.A., Guérin, G., Des comparisons of Late Pleistocene alluvial deposits on the Coast of Galicia (NW Spain) using BayLum or Analyst-based procedures., *Quaternary Geochronology*, <https://doi.org/10.1016/j.quageo.2024.101540>.

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1 **Des comparisons of Late Pleistocene alluvial deposits on the Coast of Galicia (NW Spain)**  
2 **using BayLum or Analyst-based procedures.**

3  
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11  
12  
13 **Abstract**

14 The coastal sedimentary record of Galicia (NW Spain) is important to study sea-level oscillations  
15 during the Late Quaternary. The alluvial deposits preserved in the Ria de Coruña and Ria de  
16 Arousa are remnants of fluvial valleys flooded by the sea during the successive Pleistocene and  
17 Holocene transgressions. A first chronological study by Optical Stimulated Luminescence (OSL)-  
18 revealed a fast-component dominated, but dim OSL signal emitted by quartz from this area.  
19 Equivalent dose distributions, as obtained with the Analyst software, yielded large overdispersion  
20 values, which were negatively correlated to the potassium concentration in the sediment, thus  
21 suggesting the influence of beta dose rate heterogeneities. Nevertheless, for most samples many  
22 aliquots were considered in saturation, leading to a palaeodose and age underestimation –  
23 regardless of the model choice for age calculation. In this paper, we present new analyses using  
24 the open-source R package BayLum, as the so-called saturated aliquots may be incorporated in  
25 age calculation. Thus, we test this model performance and more commonly used models in  
26 retrieving large doses. Then, we discuss the implications for age calculation for these continental  
27 sediments formed during the Late Pleistocene.

28  
29 **Keywords:** Coastal alluvial deposits, overdispersion, beta dose rate heterogeneities, saturated  
30 OSL signal, BayLum, Late-Pleistocene, sea-level changes, North-West Spain.

31

## 32 1. Introduction

33 The coastal alluvial deposits in NW Spain (Figs. 1; Figs. A1 and A2 in appendix) indicate the  
34 existence of long continental processes (Escuer-Sole and Vidal-Romaní, 1987), where today a  
35 marine environment is developing (see the coastal evolution map in figure 1). Therefore, they are  
36 geological evidences of the variations in sea level that occurred as a consequence of the glacial  
37 and interglacial cycles during the Late Quaternary. The first attempt to date some of these coastal  
38 alluvial deposits by optically stimulated luminescence (OSL; Murray et al., 2021) placed the  
39 formation of most of them during the Upper Pleistocene (Arce-Chamorro et al., 2022). However,  
40 the statistical treatment of data and the estimation of the palaeodose for some of the samples  
41 (MPK and ABC) using the Analyst software (Duller, 2015) could lead to an underestimation of  
42 the age by omitting much of the information due to saturation effects (see e.g., Heydari and  
43 Guérin, 2018).

44

### 45 FIGURE 1

46

47 Using multigrain quartz aliquots from these deposits, a fast-component dominated but dim OSL  
48 signal was observed (Fig.2), with values from 200 to 4,000 counts (per 0.08 seconds) at high  
49 doses (>80 Gy) and a few aliquots (<20%) whose OSL signal was more intense (up to 20,000  
50 counts/0.08s) (Arce-Chamorro, 2017). Various analyses (as interpolation, different curve fitting  
51 options such as single exponential or exponential+linear, early or late background estimation and  
52 models such as the Central Age Model (CAM), the Minimum Age Model (MAM; Galbraith et  
53 al., 1999) or the Internal External Consistency criteria (Thomsen et al., 2007)) were conducted to  
54 investigate the sensitivity of absorbed dose estimation ( $D_e$ ) to analytical choices. In all cases,  $D_e$   
55 distributions are highly scattered (individual  $D_e$ s range from 20 to >200 Gy; Fig. 2c), showing  
56 large overdispersion values (up to 63%). Part of this overdispersion may be linked with the  
57 concentration in potassium within the finest fraction (<63  $\mu\text{m}$ ) (Fig. A3), suggesting micro-beta  
58 dose-rate heterogeneities (Arce-Chamorro and Sanjurjo-Sánchez., 2020). Nevertheless, for most  
59 samples the number of aliquots in saturation is a significant issue (Fig.2b).

60

### 61 FIGURE 2

62

63 A single saturating exponential function passing through the origin ( $L/T=A[1-\exp(-D/D_0)]$ ) was  
64 used for fitting the normalized OSL signals ( $L/T$ ; natural and regenerated at increasing doses)  
65 (Fig. 2a) to describe a single trap dominated luminescence signal and signal saturation, resulting  
66 in a highly scattered Equivalent Dose ( $D_e$ ) distributions (Fig. 2b). This also results in a large  
67 proportion of aliquots in saturation, as aliquots for which the sum of  $L_N/T_N$  and its uncertainty as  
68 calculated with Analyst does not intersect the dose response curve (DRC) (Fig. 2b and A4, A5;

69 Table 1), leading to notorious difficulties for dating (Duller et al., 2012; Thomsen et al., 2016;  
70 Colarossi et al., 2020). For most of the samples, there exists a proportion of aliquots (up to 30%)  
71 for which the Analyst (Duller, 2015) does not provide a known uncertainty (in the following, we  
72 call the number of such aliquots  $N_{De/INF}$ ) (Table 1). This excludes them from calculations using  
73 frequentist statistics such as the CAM, leading to an underestimation of the burial age (Heydari  
74 and Guérin., 2018; Guérin et al., 2021; Chapot et al., 2022). The same applies to those aliquots  
75 (up to 55%) for which the Analyst provides neither equivalent dose ( $D_e$ ) nor uncertainty (we  
76 denote the number of such aliquots by  $N_{INF/INF}$ ) (Table A1; Fig.2). Alternatively, the Bayesian  
77 models of Combès et al. (2015) and Combès and Philippe (2017) implemented in the BayLum  
78 package (Philippe et al., 2019; Guérin et al., 2021) are able to use these saturated aliquots by  
79 estimating the individual  $D_e$ s and Palaeodose simultaneously from single hierarchical iteration of  
80 all  $L_N/T_N$  and  $L_X/T_X$  signal distributions of the DRCs (see Heydari and Guérin, 2018), without  
81 parameterizing individual  $D_e$  probability densities. The comparison of the results obtained with  
82 the CAM (after  $D_e$  estimation with the Analyst software) and with BayLum, according to various  
83 selections of aliquots included in the calculations ( $N_{De/SIGMA}$ , i.e., the aliquots not affected by  
84 saturation issues,  $N_{De/INF}$  and  $N_{INF/INF}$ ), as well as the estimation of a more accurate age are the  
85 main objectives of this work.

86

## 87 2. Material and methods.

88 Samples MPK and ABC were collected using steel cores. For OSL signal analysis from quartz  
89 grains, the raw sample from the central part of the core was dehydrated at room temperature and  
90 the 180-250  $\mu\text{m}$  grain-size fraction was separated by dry sieving (Arce-Chamorro, 2017). Quartz  
91 purification was carried out using  $\text{H}_2\text{O}_2$  and  $\text{HCl}$  (20%), as well as  $\text{HF}$ -etching (40%) during 40  
92 minutes to remove the alpha-irradiated outer layer of the grains. Ultra-dense liquid centrifugation  
93 was carried out to isolate quartz grains and silicon spray was used to prepare  $\approx 100$ -grain aliquots  
94 on steel discs ( $\varnothing=1$  mm, except for samples ABC1 and ABC4 for which 10-grain aliquots were  
95 manually prepared using a binocular and a small paintbrush). A Risø reader TL/OSL DA-15  
96 reader with a Beta  $^{90}\text{Sr}/^{90}\text{Y}$  source delivering  $0.120\pm 0.003$   $\text{Gys}^{-1}$  (calibrated with a sample given a  
97 5 Gy gamma dose using a  $^{137}\text{Cs}$  source (40 s)) was used to build the DRCs using the SAR protocol  
98 (Murray and Wintle, 2000) (Table A2), but also to test each aliquot for feldspar contamination by  
99 measuring regenerated OSL signals after an infra-red stimulation. The OSL signal was stimulated  
100 with blue LEDs at 90% of maximum power during 40 s. The fast-OSL signal was integrated from  
101 the first 0.5 s of luminescence stimulus and the background signal from the last 4 s. For this work,  
102 dose recovery (Wallinga et al., 2000) experiments have been carried out again (see Arce-  
103 Chamorro et al., 2017, for the first set of dose recovery experiments), but at given doses of 83-84  
104 Gy for MPK samples and 180 Gy for MPK8. To maximize the number of accepted aliquots and  
105 the information provided by Analyst, the acceptance criteria were a natural test dose error  $<20\%$

106 and a  $T_N$  signal  $>3\sigma$  above background, following Geach et al. (2015), Thomsen et al. (2016),  
107 Heydari and Guérin (2018), Chapot et al., (2022), etc. Frequentist statistical models such as the  
108 CAM or the Average Dose Model (ADM; Guérin et al., 2017) were implemented with the  
109 Luminescence R package (Kreutzer et al., 2017). Bayesian statistical analysis was carried out  
110 with the BayLum R Package, following the recommendations of Philippe et al. (2019) to check  
111 the validity of the estimates obtained with the Markov Chain Monte Carlo (MCMC) algorithms.  
112 For BayLum calculations (see in figure A6 the MCMC plots performed by BayLum iterations for  
113 samples ABC3 and MPK8), the number of iterations (from  $5 \times 10^3$  to  $5 \times 10^6$ ) and prior-age intervals  
114 were tested and compared using R Studio (R Core Team, 2020).

115 Dose rate estimates have been carried out throughout activity estimates of  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$   
116 decay chains, as well as  $^{40}\text{K}$ , by high resolution gamma spectrometry (HRGe) using a Camberra  
117 XTRA Ge-Intrinsic detector. Conversion factors of Guérin et al., (2011) for changing Bq/kg to  
118 Gy/ky units were used. Beta attenuation from HF etching was corrected following Brennan  
119 (2003).  $D_R$  attenuation due to water content was estimated from saturated and moisture values of  
120 the raw sample following Guérin and Mercier (2012). Auto-irradiation of quartz from alpha  
121 component was taken into account following Vandenberghe et al. (2008). Sampling depth within  
122 the sediment, altitude and latitude were considered to estimate cosmic dose rates following  
123 Prescott and Hutton (1994), as well as the density of sampled materials ( $1.8\text{-}1.9 \text{ g/cm}^3$ ).

124

### 125 **3. Results**

#### 126 **3.1 Dose rate estimates**

127 The estimated activity of the main radioisotopes is shown in table A3 for the subsequent  
128 estimation of the  $D_R$  (Gy/ka), as an indispensable parameter for the calculation of an age (section  
129 4.3) once the palaeodoses ( $D_e = \text{Gy}$ ) of each sample has been estimated ( $\text{age} = D_e / D_R$ ; Aitken, 1998).  
130 No disequilibrium was observed within the  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{228}\text{Th}$  decay chains. The water saturation  
131 percentage (W%) for MPK and ABC samples varies between 20 to 40%, as a parameter included  
132 in  $D_R$  estimates following Aitken (1998). Moisture percentage (H%) varies between 4 and 20%,  
133 thus obtaining a H/W ratio from 25 to 55% (Arce-Chamorro, 2017). This ratio is needed to  
134 estimate the water content in the sediment along burial time, since all these alluvial sediments  
135 were fully saturated when they were deposited ( $W = 100\%$ ). Subsequent incision of the fluvial  
136 network led to the abandonment of the alluvial deposits at higher altitudes, remaining perched  
137 and even giving rise to an inverted relief (Escuer-Sole and Vidal-Romaní, 1987). Thus, we  
138 assumed a sedimentary water content close to the average of H/W ratio for the analysed samples  
139 ( $W = 40 \pm 10\%$ ) (Table A3).

140

#### 141 **3.2 Dose estimates.**

##### 142 **3.2.1 Dose recovery experiments.**

143 Dose recovery experiments are meant to assess the performance of the SAR protocol and  
 144 subsequent statistical analyses in retrieving a known artificial dose administered to a population  
 145 of grains. It is generally suggested that measured to given dose ratios should lie within 10% of  
 146 unity (Wallinga et al., 2000). In our case, the dose recovery ratios have been calculated using the  
 147 CAM and BayLum (gaussian) ( $D_{e-SIGMA}$ ,  $D_{e-UNCERT.SAT}$  and  $D_{e-TOTAL}$ ) with a known dose of 84 Gy  
 148 (Table 1; Fig. 3). The choice of 84 Gy was made after observing that aliquots in saturation (i.e.,  
 149 belonging to the  $N_{De/INF}$  or  $N_{INF/INF}$  populations) begin to be observed at this level of dose when  
 150 using a single exponential fit with the Analyst software (Table 2; Fig. A7). A known given dose  
 151 of 180 Gy was also used for MPK8 sample, as  $N_{De/INF}$  or  $N_{INF/INF}$  aliquots only appear at higher  
 152 doses for this sample (see MPK8 sample in table 1).

153

154 TABLE 1

155

156 As in previous works (Duller et al., 2012; Chapot et al., 2022), a higher dose recovery inaccuracy  
 157 of CAM is observed when there is a higher proportion of saturated aliquots, in this case  $N_{De/INF}$   
 158 and  $N_{INF/INF}$  (>30%). Thus, BayLum results are strongly conditioned by the proportion of  $N_{De/INF}$   
 159 and  $N_{INF/INF}$  aliquots (Fig. 3a) providing more accurate results, like palaeodose analyses do (see  
 160 next section 3.2.2). First, by looking at Table 1 we note that the average dose recovery ratio is  
 161 equal to  $0.90 \pm 0.03$  when using the CAM,  $0.94 \pm 0.02$  when using BayLum with a Gaussian  
 162 distribution to model equivalent doses with the same number of aliquots ( $N_{De/SIGMA}$ ) and  $1.01 \pm 0.03$   
 163 with BayLum when including all aliquots and notably the so-called saturated ones. Second, to  
 164 highlight the effect of saturated grains on the dose recovery estimates (Fig. 3a), we plotted the  
 165 relative difference between the values obtained with the CAM and those calculated with BayLum  
 166 when including all aliquots, as a function of the proportion of saturated aliquots (Fig. 3b). The  
 167 clear positive trend suggests to us that indeed the CAM underestimates the given dose all the more  
 168 than the frequency of ‘saturated’ aliquots is large. Our results are similar to those of Duller et al.  
 169 (2012), Heydari and Guérin (2018) and Chapot et al. (2022), who all – among others – reported  
 170 problems of dose recovery underestimation with large doses.

171

172 FIGURE 3

173

### 174 3.2.2 Palaeodose estimates.

175 We estimated the palaeodose for each sample using different statistical models and grain  
 176 selections, to make our comparisons as thorough as possible (Fig.4). First, since the CAM and the  
 177 ADM can only work with pairs of  $D_e$  and associated uncertainty values, we are limited to  
 178  $N_{De/SIGMA}$  aliquots per sample. BayLum was ran on this population of grains, using both the  
 179 Gaussian and lognormal models for equivalent dose distribution. For the sake of comparison, we

180 used the lognormal\_M model of BayLum (Fig. 4a), i.e. the model which estimates the median of  
 181 the lognormal distribution. For purely mathematical reasons, this latter model – like the CAM –  
 182 should underestimate the average absorbed dose in the studied population of grains (Guérin et al.,  
 183 2017). In a second step of analysis, we included for each sample the number of aliquots for which  
 184 Analyst provides a  $D_e$  estimate but no bounded estimate of its uncertainty ( $N_{D_e/INF}$ ), so that we  
 185 work with a total number of aliquots  $N_{D_e-UNCERT.SAT} = N_{D_e/SIGMA} + N_{D_e/INF}$ . Finally, we also included  
 186 the aliquots for which Analyst provides no finite  $D_e$  value and thus, no uncertainty either ( $N_{TOTAL}$   
 187  $= N_{D_e/SIGMA} + N_{D_e/INF} + N_{INF/INF}$ ).

188

189 FIGURE 4

190

191 Table 2 lists all palaeodose estimates. Unsurprisingly, the CAM doses always are the lowest  
 192 values for each sample. Conversely, the largest values are systematically obtained with BayLum  
 193 when all aliquots are used (Fig.4b), including those for which the natural OSL signal (or the sum  
 194 of this signal and its uncertainty) is greater than the asymptotic limit of the DRC in Analyst. To  
 195 interpret the comparisons in more detail, one needs to disentangle two different sources of  
 196 differences, namely dose rate variability and saturation issues. It is worth mentioning that for  
 197 sample MPK8 (Table 2) a new SAR protocol was used to investigate larger regenerative doses  
 198 (120, 240, 480 and 960 Gy; Table A2), as we did not obtain convergence of the MCMC chains  
 199 using BayLum when using all aliquots ( $D_e$  TOTAL in Fig.4). (In our previous study (Arce-  
 200 Chamorro, 2017), the largest regenerative dose was 216 Gy.). For the samples MPK3 and ABC2,  
 201 whose palaeodoses are higher than the larger regenerative dose of the SAR, the results obtained  
 202 ( $D_{e-TOTAL}$  in table 2) should be interpreted with caution, although the Late Pleistocene age  
 203 calculated for these samples is consistent and reliable (see section 4.3).

204

205 TABLE 2

206

207 **4. Discussion**208 **4.1 Dose rate effects on  $D_e$ s estimates.**

209 The first question that needs to be addressed when calculating an OSL age is that of the variability  
 210 in equivalent doses measured on numerous aliquots. It is well-known that dose rates to quartz  
 211 grains are, as a general rule, variable; this variability has been observed by beta autoradiography  
 212 (e.g., Rufer and Preusser, 2009; Guérin et al., 2012b; Martin et al., 2015; Smedley et al., 2020)  
 213 and modelled (Nathan et al., 2003; Mayya et al., 2006; Cunningham et al., 2011; Guérin et al.,  
 214 2015; Jankowski and Jacobs, 2018). The question then becomes to determine which quantity  
 215 should be estimated from the population of  $D_e$  values. Again, as a general rule,  $D_e$  values may  
 216 adequately be described by a lognormal distribution, both from purely luminescence



217 measurement reasons (Galbraith and Roberts, 2012) and because radioactive hotspots such as  
 218 potassium feldspar grains are ubiquitous (Guérin et al., 2015). Both the CAM and the ADM  
 219 assume a lognormal distribution, but the CAM calculates the median while the ADM estimates  
 220 the average of this distribution – simply to match the average dose rate determined experimentally  
 221 using the infinite matrix assumption. This fundamental difference explains the discrepancy  
 222 between CAM and ADM doses, the former being 4-17% smaller than the latter (Table 2; Fig. 5a).  
 223 It also explains, at least in part, the difference between CAM doses and estimates obtained with  
 224 BayLum using a Gaussian distribution – between 5 and 30% (Table 2; Fig. 5b), with the key-  
 225 difference being the estimated parameter rather than the shape of the distribution fitted to  $D_e$   
 226 estimates.

227 The first attempts to analyze the high OD values for MPK samples (20-50%) were related with  
 228 beta dose rate variability (Arce-Chamorro et al., 2020) (Fig. A3), since partial bleaching could  
 229 not be demonstrated (Arce-Chamorro, 2017) and because intrinsic overdispersion (<5%) was  
 230 determined from dose recovery tests (Thomsen et al., 2005). In that work, it should be noted that  
 231 all  $N_{D_e/\sigma}$  aliquots interpolating close to the linear part of the DRC (i.e., below the 86.5% of the  
 232 maximum asymptote for the DRC or  $D_e > 2D_0$ ; Wintle and Murray, 2006) were excluded for  
 233 palaeodose estimates with the CAM, as individual  $D_{e,s}$  estimates from them were generally  
 234 considered as unreliable. The rejection of these aliquots led to biased  $D_{e,s}$  distributions and  
 235 underestimation of the OD from CAM. Conversely, as observed in this work, when all aliquots  
 236 with  $D_e/\sigma$  values ( $N_{D_e/\sigma}$ , following the nomenclature of this manuscript for aliquots which are not  
 237 affected by saturation) are included in the calculations, the OD from the CAM grows up from 26  
 238 to 69% (see Table 2).

239

240 FIGURE 5

241

#### 242 4.2. Saturation effects on $D_{e,s}$ estimates.

243 When using  $N_{D_e/SIGMA}$  aliquots, comparison of palaeodose estimates between CAM and BayLum  
 244 (Lognormal\_M) aliquots show similar values at low doses, as they both estimate the median of a  
 245 lognormal distribution. However, an underestimation of  $D_{e,s}$  with the CAM is observed at higher  
 246 doses (Fig. 5d), which would indicate that BayLum (Log\_M) provides better data handling in the  
 247 non-linear part of the DRC than the CAM. In addition,  $D_e$  estimates from Gaussian are higher  
 248 than Lognormal\_M procedures with BayLum (Fig. 5d) and, as commented, the differences with  
 249 CAM estimates increases with the OD (Fig. 5c). This suggests that in case of samples with widely  
 250 dispersed individual  $D_{e,s}$ , BayLum (gaussian) procedures would provide more accurate results  
 251 than the CAM (see also Heydari and Guérin, 2018).

252 For its part, when using saturated aliquots, the increase of  $D_{e-TOTAL}$  ( $N_{D_e/SIGMA} + N_{D_e/INF} + N_{INF/INF}$ )  
 253 with respect to  $D_{e-SIGMA}$  ( $N_{D_e/SIGMA}$ ) estimated by the CAM and BayLum (gaussian) is analysed as

254 a function of the number of saturated aliquots ( $N_{\text{TOTAL}} = N_{\text{De/SIGMA}} + N_{\text{De/INF}} + N_{\text{INF/INF}}$ ) included in  
 255 the calculations with the Bayesian models (Figs. 5e and 5f). For this purpose, the ratio of aliquots  
 256 was normalised as  $[(N_{\text{TOTAL}} - N_{\text{SIGMA}}) / N_{\text{TOTAL}}]$  (X axis in Figs. 5e and 5f). When this ratio tends  
 257 to zero, i.e.: when the proportion of aliquots ‘in saturation’ becomes negligible,  $D_{\text{e-SIGMA}}$  is similar  
 258 to  $D_{\text{e-TOTAL}}$ . However, when this ratio is increased,  $D_{\text{e-TOTAL}}$  is strongly increased. When this ratio  
 259 approaches unity, i.e., when most aliquots are considered saturated,  $D_{\text{e-TOTAL}}$  seems to grow  
 260 exponentially. The difference between  $D_{\text{e-TOTAL}}$  ( $N_{\text{De/SIGMA}} + N_{\text{De/INF}} + N_{\text{INF/INF}}$ ) and  $D_{\text{e-UNCERT.SAT}}$   
 261 ( $N_{\text{De/SIGMA}} + N_{\text{De/INF}}$ ) also follows this trend, showing the importance of the unknown information  
 262 (uncertainty) included within  $N_{\text{INF/INF}}$  aliquots using BayLum (inset in Fig. 5f). This observation  
 263 suggests that BayLum (gaussian) provides more accurate results when working with saturated  
 264 aliquots than any model excluding such aliquots, as is suggested in the literature (Heydari and  
 265 Guérin, 2018; Guérin et al., 2021; Chapot et al., 2022).

266

### 267 **4.3 Burial ages of MPK and ABC samples.**

268 It seems rather clear that dose estimates obtained after rejecting many aliquots due to saturation  
 269 are unreliable; only dose underestimation can result from such biased acceptance criterion (Singh  
 270 et al., 2017; Guérin et al., 2015b). Thus, in the following, we assume that BayLum dose estimates  
 271 making use of all aliquots [ $D_{\text{e-TOTAL}} = (N_{\text{De/SIGMA}} + N_{\text{De/INF}} + N_{\text{INF/INF}})$ ] are more accurate. Updated  
 272 OSL ages using these dose values are shown in table 3.

273

#### 274 **TABLE 3**

275

276 Based on these results, the studied coastal alluvial sediments in NW Spain were deposited during  
 277 different episodes throughout the Late Pleistocene (Fig. 6). The ages of the deposits  
 278 corresponding to samples MPK1, MPK5, MPK6 and ABC4 correspond to the beginning of the  
 279 regressive episode at the Upper Pleistocene (MIS5b, 5a and MIS4) (Fig. 6). BayLum (gaussian)  
 280 ages for these samples are 10 ky and 30 ky greater than those calculated in previous work (Arce-  
 281 Chamorro, 2017) based on frequentist statistics (CAM and Minimum Age Model (MAM);  
 282 Galbraith et al., 1999) (Table 3), which excluded saturated aliquots. All the rest of the samples  
 283 show much older ages (>50 ky) using BayLum. The deposit corresponding to sample MPK4  
 284 formed during the Eemian interglacial (late MIS6 and MIS5e), whereas MPK7, MPK8 and MPK2  
 285 samples are of pre-Eemian age. The deposit of the sample MPK3 was formed during MIS7, with  
 286 stratigraphic coherence between this sample located at +60 m above present sea level (apsl) and  
 287 sample MPK4 at +10 m (apsl), formed by the same secondary course of a fluvial network (see  
 288 Arce-Chamorro et al., 2023a). As for the alluvial outcrop of Catoira from which samples ABC1  
 289 of  $412 \pm 49$  ky, ABC2 of  $182 \pm 23$  ky and ABC3 of  $335 \pm 48$  ky were extracted, the presence of  
 290 different processes of aggradation and erosion in a strictly continental environment during the

291 glacioeustatic oscillations along the Late Pleistocene is observed (Arce-Chamorro and Vidal-  
292 Romaní, 2021) (Fig.A2), more specifically during MIS11-MIS10 (ABC1 and ABC3) and during  
293 MIS7-MIS6 (ABC2).

294

295 FIGURE 6

296

297 Thus, this system of alluvial deposits was not formed during the so called Villafranchian unit  
298 (within the Plio-Quaternary boundary from 3.5 to 1 My), as suggested by Nonn (1966) from his  
299 geomorphological study along the Coast of Galicia (NW Spain). Neither are they sediments  
300 whose formation age was indeterminately assigned to the Miocene (up to 5 My ago) (Arps et al.,  
301 1978), nor are they Holocene materials (Galán-Arias, 1981). From the chronology estimated by  
302 BayLum, we deduce a system of braided alluvial deposits (Escuer-Sole and Vidal-Romaní, 1987),  
303 whose sedimentation would be related to glacioeustatic levels which did not culminate during the  
304 Late Pleistocene (Fig. 8). These data provide greater reliability to the model of Escuer-Sole and  
305 Vidal-Romaní (1987), based on the geomorphological study - without the possibility of applying  
306 adequate absolute dating techniques at that time. This hypothesis, supported by the ages  
307 calculated here, helps to better understand the evolution of the Ria de Coruña which is located in  
308 the northern coast of Galicia (Fig.1), as well as the Ria de Arousa in the mid-south coast (Fig.1).  
309 And this could be most probably extrapolated to the rest of the Galician Rias, as they were fluvial  
310 valleys flooded during the transgressive episodes of the Late Quaternary (Vidal-Romaní and  
311 Grandal d'Anglade, 2018), giving rise to these characteristic wide embayments on the Northwest  
312 Coast of Spain.

313

## 314 **5. Conclusions**

315 Analyst procedures for MPK and ABC samples show widely scattered  $D_{es}$  distributions at high  
316 doses. Assuming that OD arises from dose rate variability, Bayesian procedures (or any other  
317 model estimating the average absorbed dose, such as the ADM) should provide more accurate  
318 results, as the greater the OD, the larger the Palaeodose differences between BayLum and the  
319 CAM. When working with saturated aliquots, BayLum doses calculated with 'saturated' aliquots  
320 keep increasing at much larger doses than CAM. Dose recovery experiments also suggest that  
321 BayLum provides more accurate results than CAM. Thus, the ages calculated by BayLum are  
322 older than those previously calculated by frequentist statistics (CAM or ADM). For samples  
323 MPK1, MPK5, MPK6 and ABC1, the underestimation of the CAM is between 10-30 ky. For the  
324 rest of the samples, the differences between the statistical models can reach between 50 ky and  
325 200 ky, or even more. Based on these results, they are not Late Miocene, Late Pliocene or  
326 Holocene deposits. On the contrary, they were deposited in the lowermost areas of these fluvial  
327 valleys at the end of the Middle Pleistocene and during the Upper Pleistocene.

328

329 **Acknowledgments**

330 Thanks to the Full Professor Juan Ramón Vidal Romaní for sharing his geological knowledge on  
 331 the dated alluvial sediments. Thanks to Dr. Jorge Sanjurjo Sánchez for his support with the  
 332 Luminiscence Lab of the University of Coruña. Thanks to the LED2023 Teamwork. Also thanks  
 333 to the editors of Quaternary Geochronology and to the unknown Reviewers for their suggestions.

334

335 **Funding**

336 This research has been funded and supported by Consellería de Cultura, Educación e Ordenación  
 337 Universitaria, Xunta de Galicia (programmes ED431B 2018/47 and ED431B 2021/17) through  
 338 the Grupo Interdisciplinar de Patrimonio Cultural e Xeolóxico (CULXEO).

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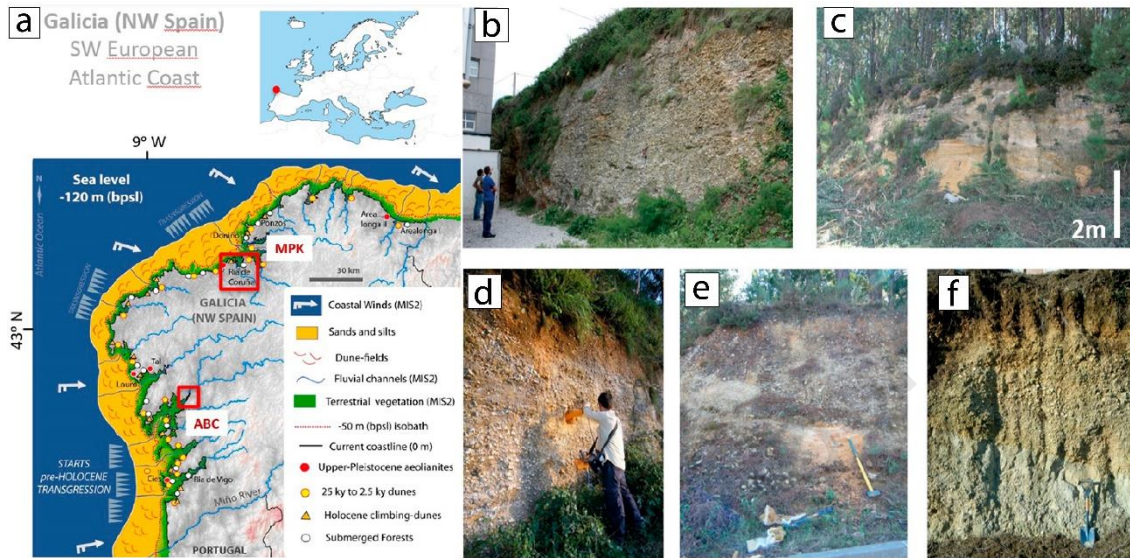
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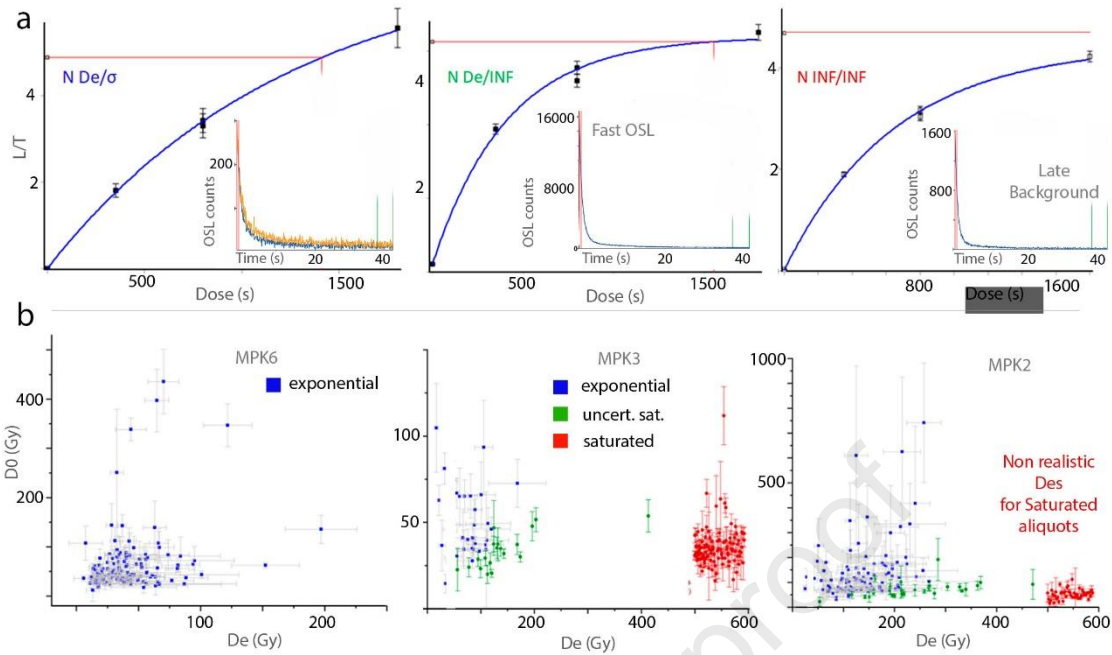


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**Figure 1.** Present day and regressive coastline (-120 m; yellow area) of Galicia (NW Spain): (a) Coastal evolution map (modified from Arce-Chamorro et al., 2023) and some of the alluvial deposits previously studied (Arce-Chamorro, 2017): (b,d) MPK2 sample ( $43^{\circ}22'21''\text{N}$ ,  $8^{\circ}20'18''\text{W}$ ); (c) ABC1,2,3 samples ( $42^{\circ}40'25''\text{N}$ ,  $8^{\circ}43'20''\text{W}$ ); (e) MPK1 sample ( $43^{\circ}16'33''\text{N}$ ,  $8^{\circ}18'46''\text{W}$ ); (f) MPK3 sample ( $43^{\circ}21'26''\text{N}$ ,  $8^{\circ}19'52''\text{W}$ ). (WGS84 coordinates).



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522 **Figure 2. (a)** Examples of dose response curves (DRCs) for MPK samples, by a single saturating exponential  
 523 fitting of the normalized OSL signals ( $L/T$ ), representing N De/ $\sigma$ , N De/INF and N INF/INF aliquots using  
 524 Analyst (Duller, 2015); Y axis: normalized OSL-signal ( $L_X/T_X$  and  $L_N/T_N$ ); X axis: given dose (seconds) at each  
 525 cycle of single aliquot regenerative dose protocol (SAR; Murray and Wintle, 2000) from  $^{90}\text{Sr}/^{90}\text{Y}$  beta source of  
 526  $0.12 \pm 0.03$  Gy of Risø TL/OSL DA-15 reader; (Inset): OSL decay curve examples with natural OSL counts (per  
 527 0.08 s) (blue line) and 3rd cycle of SAR (orange line) along luminescence stimulation time (40 s; X axis).  
 528 Integration of fast-OSL signal (red lines) and background (green lines). **(b)** Examples of characteristic dose ( $D_0$ )  
 529 for single exponential growth curves and  $D_0$ s distribution of N De/ $\sigma$  (blue squares), N De/INF (green dots)  
 530 and N INF/INF (red dots) aliquots. Non-realistic  $D_0$ s for N INF/INF aliquots.

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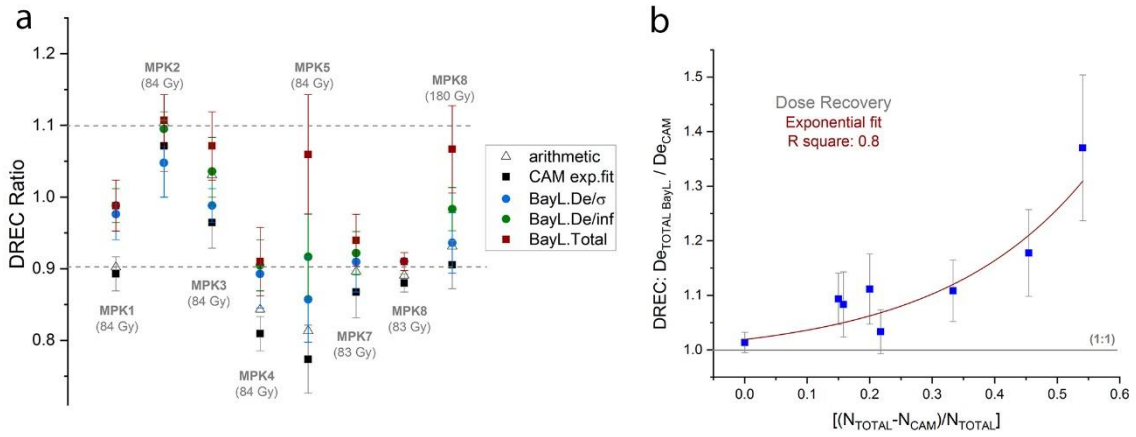
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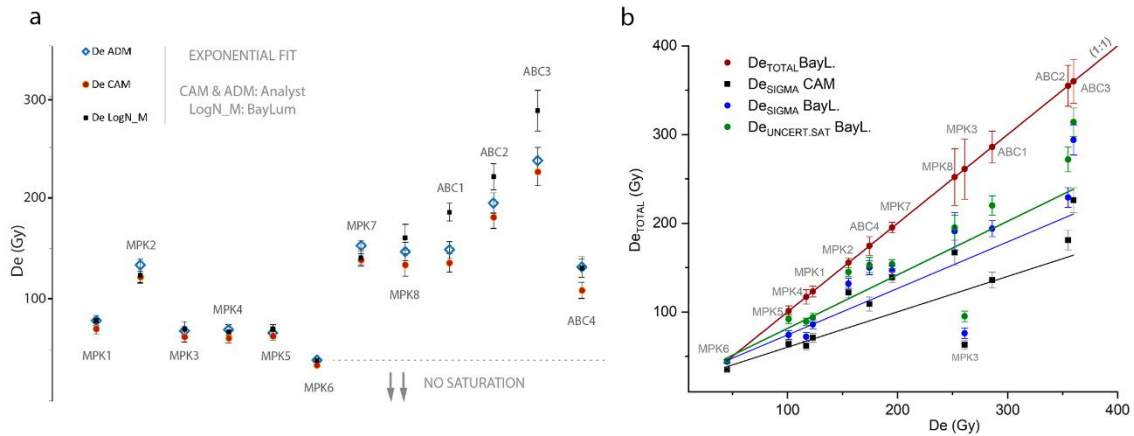
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**Figure 3.** Dose Recovery analysis using BayLum (Gaussian) and CAM. **(a)** Dose recovery ratios ( $D_{e-ESTIMATE}/D_{e-GIVEN}$ ) from CAM and BayLum (arithmetic mean; CAM using  $N_{De/SIGMA}$  aliquots; BayL- $D_{e/SIGMA}$  using  $N_{De/SIGMA}$ ; BayL- $D_{e/INF}$  using ( $N_{De/SIGMA} + N_{De/INF}$ ); BayL- $D_{e/TOTAL}$  using ( $N_{De/SIGMA} + N_{De/INF} + N_{INF/INF}$ ). Dotted grey lines show 10% criteria (0.9-1.1) for recovery test (Wallinga et al., 2000). **(b)** Differences in the  $D_{e-TOTAL}$  and  $D_{e-CAM}$  from dose recovery according to the proportion of aliquots used. Aliquots proportion:  $N_{TOTAL} = (N_{De/SIGMA} + N_{De/INF} + N_{INF/INF})$ ;  $N_{CAM} = (N_{De/SIGMA})$ .  $D_{eS}$  estimates:  $D_{e-TOTAL}$  using ( $N_{De/SIGMA} + N_{De/INF} + N_{INF/INF}$ );  $D_{e-CAM}$  using ( $N_{De/SIGMA}$ ).



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552 **Figure 4.** Palaeodose analysis for MPK and ABC samples. (a)  $D_{e,s}$  estimates from CAM, ADM and553 BayLum (lognormal\_M) using  $N_{D_{e/SIGMA}}$  aliquots. (b)  $D_{e,s}$  estimates:  $D_{e,SIGMA}$  from CAM and BayLum554 using  $N_{D_{e/SIGMA}}$  aliquots;  $D_{e,UNCERT.SAT}$  using  $N_{D_{e/SIGMA}} + N_{D_{e/INF}}$  aliquots;  $D_{e,TOTAL}$  using  $N_{D_{e/SIGMA}} + N_{D_{e/INF}}$ 555 +  $N_{INF/INF}$  aliquots); the lines (black, blue and green) show the exponential fit for each palaeodose analysis.

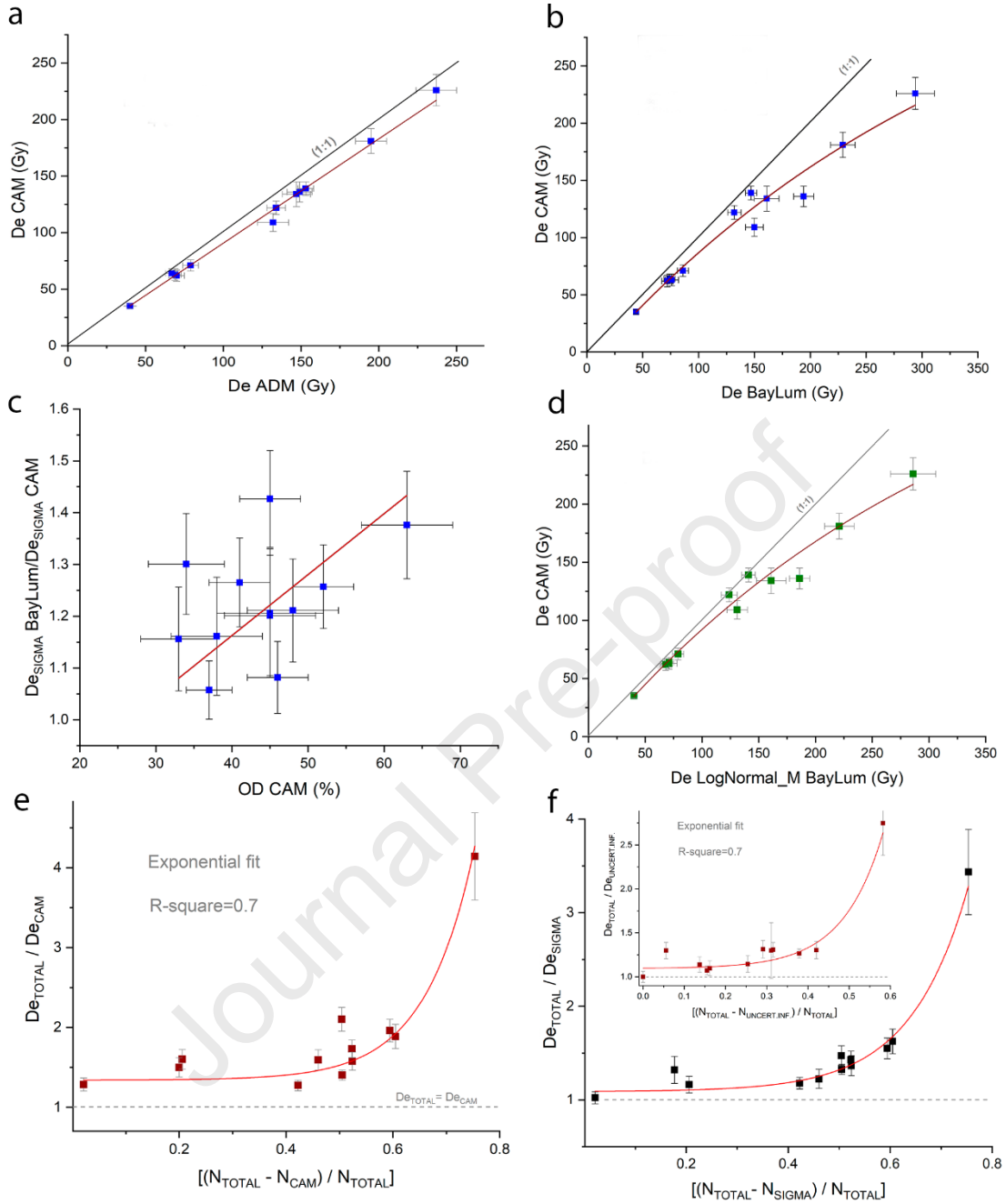
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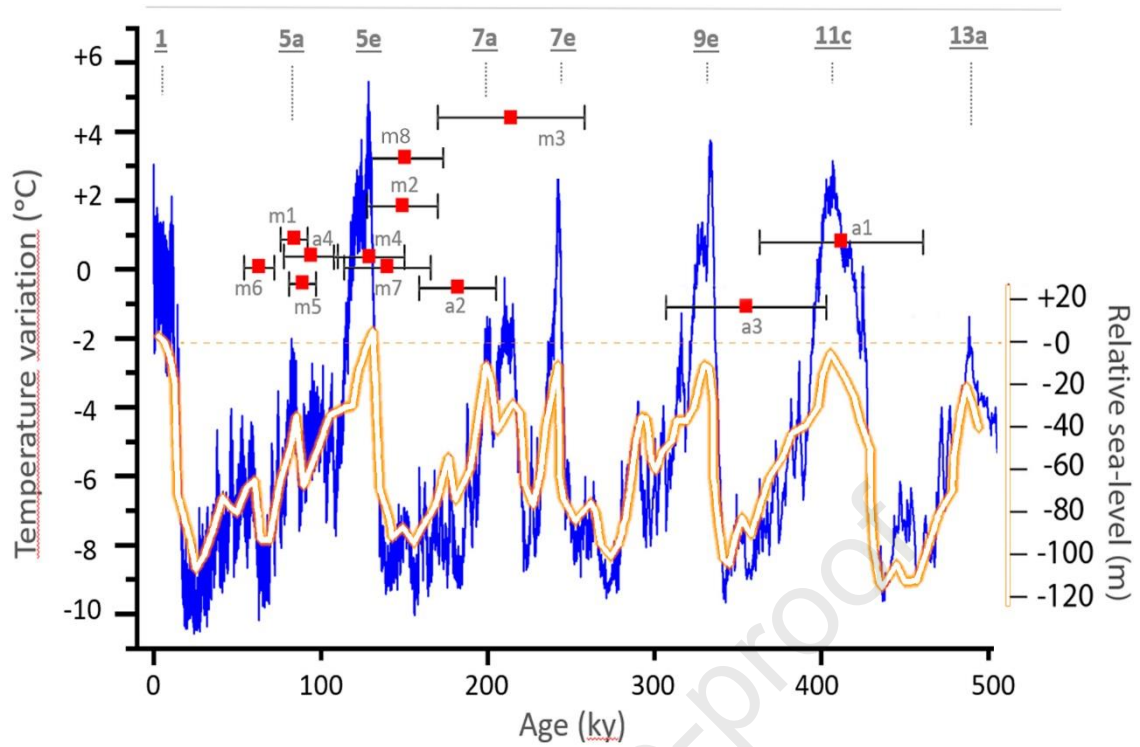
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**Figure 5.** Palaeodose comparison. **(a,b,c,d)**:  $D_{eS}$  estimates of MPK and ABC samples from CAM, ADM and BayLum using  $N_{D_{e\Sigma}}$  aliquots from Analyst. **(a)** CAM vs ADM. **(b)**: CAM vs BayLum gaussian. **(c)**  $D_{eS}$  differences from  $D_{e\text{BayLum-gaussian}}/D_{e\text{CAM}}$  vs OD-CAM (%). **(d)**:  $D_e$  CAM vs BayLum Lognormal\_M. **(e,f)** Differences in the  $D_{eS}$  estimates according to the proportion of aliquots used for each case. Aliquots proportion:  $N_{\text{TOTAL}} = (N_{D_{e\Sigma}} + N_{D_{e\text{INF}}} + N_{\text{INF/INF}})$ ;  $N_{\text{CAM}}$  and  $N_{\Sigma} = (N_{D_{e\Sigma}})$ ;  $N_{\text{UNCERT.INF}} = (N_{D_{e\Sigma}} + N_{D_{e\text{INF}}})$ . **(e)**:  $D_{e\text{TOTAL}}$  using  $(N_{D_{e\Sigma}} + N_{D_{e\text{INF}}} + N_{\text{INF/INF}})$ ;  $D_e$  CAM using  $(N_{D_{e\Sigma}})$ . **(f)**:  $D_{e\Sigma}$  using  $(N_{D_{e\Sigma}})$  with BayLum (gauss.). (Inset in f):  $D_{e\text{UNCERT.SAT.}}$  using  $(N_{D_{e\text{INF}}} + N_{\text{INF/INF}})$  with BayLum (gauss.).



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**Figure 6.** MPK and ABC ages with BayLum using all aliquots ( $D_{e-TOTAL} = N_{De/SIGMA} + N_{De/INF} + N_{INF/INF}$ ) and temperature anomaly (blue line) from Deuterium Record of Antarctica-Dome C ice-core (Jouzel et al., 2007). Orange line: Relative sea-level (Rohling et al., 2014). Marine Isotopic Stages (MIS) (Railsback et al., 2015).

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**Table 1.** Measured and accepted aliquots (Meas. aliq.; accept.) for the dose recovery analysis for MPK samples, showing the dose recovery ratios from CAM and BayLum (Gaussian) using  $N_{De/SIGMA}$  and using saturated aliquots (all aliquots= $N_{De/SIGMA} + N_{De/INF} + N_{INF/INF}$ ).

Sample	Meas. Aliq.	Accept. $N_{De/SIGMA}$	Accept. $N_{De/INF}$	Accept. $N_{INF/INF}$	CAM ( $N_{De/\sigma}$ )	OD CAM	BayLum ( $N_{De/\sigma}$ )	BayLum (ALL aliquots)
MPK1 (84 Gy)	40	34 (85%)	5 (13%)	1 (3%)	$0.89 \pm 0.02$	$10 \pm 3$	$0.98 \pm 0.04$	$0.99 \pm 0.04$
MPK2 (84 Gy)	75	54 (72%)	9 (12%)	6 (8%)	$1.07 \pm 0.04$	$16 \pm 6$	$1.05 \pm 0.05$	$1.11 \pm 0.04$
MPK3 (84 Gy)	48	32 (67%)	14 (29%)	2 (4%)	$0.96 \pm 0.04$	$3 \pm 1$	$0.99 \pm 0.02$	$1.07 \pm 0.05$
MPK4 (84 Gy)	30	22 (73%)	5 (17%)	3 (10%)	$0.81 \pm 0.02$	$2 \pm 1$	$0.89 \pm 0.05$	$0.91 \pm 0.05$
MPK5 (84 Gy)	58	19 (33%)	13 (22%)	10 (17%)	$0.77 \pm 0.05$	$15 \pm 7$	$0.86 \pm 0.06$	$1.06 \pm 0.08$
MPK7 (83 Gy)	35	29 (83%)	2 (6%)	4 (11%)	$0.87 \pm 0.04$	$18 \pm 4$	$0.91 \pm 0.04$	$0.94 \pm 0.04$
MPK8 (83 Gy)	38	38 (100%)	0	0	$0.88 \pm 0.01$	$4 \pm 2$	$0.91 \pm 0.01$	$0.91 \pm 0.01$
MPK8 (180 Gy)	44	24 (55%)	7 (16%)	13 (30%)	$0.91 \pm 0.03$	$4 \pm 1$	$0.94 \pm 0.05$	$1.07 \pm 0.06$
Average Dose-recovery ratio					$0.90 \pm 0.03$		$0.94 \pm 0.02$	$1.01 \pm 0.03$

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590 **Table 2.** Palaeodose estimates ( $D_e$  values in Gy) for MPK and ABC samples using the central dose model (CAM; Galbraith et al., 1999), the average dose model (ADM; Guérin et al.,  
591 2017) and BayLum (Guérin et al., 2021) from accepted aliquots (N) for which single exponential fit of Analyst (Duller, 2015) provides (or not)  $D_e$  and uncertainty ( $N_{De/SIGMA}$ ,  $N_{De/INF}$  and  
592  $N_{INF/INF}$ ). (g.): Gaussian distribution; (Log.M): Lognormal\_M distribution; Underest.: palaeodose underestimation; OD: overdispersion percentage from CAM. (\*) New SAR at higher  
593 doses (960 Gy) for MPK8, as no convergence of the MCMC chains using BayLum using saturated aliquots ( $N_{De/SIGMA}$ ,  $N_{De/INF}$  and  $N_{INF/INF}$ ) from previous measurements (Arce-Chamorro,  
594 2017).

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Sample	De SIGMA ( $N_{De/SIGMA}$ )					De UNCERT.SAT ( $N_{De/SIGMA} + N_{De/INF}$ )					De TOTAL ( $N_{De/SIGMA} + N_{De/INF} + N_{INF/INF}$ )				
	N	De CAM	OD CAM (%)	De BayLum (g.)	Underest. CAM vs BayL.(g.)	De ADM	Underest. CAM vs ADM	De BayLum Log_M	Underest. CAM vs BayL(log.M)	N	BayLum (g.)	Underest. BayL.(g.) ( $N_{De/SIGMA}$ )	N	BayLum (g.)	Underest. BayL. (g.) ( $N_{De/INF}$ )
	MPK1	62/141	71±5	32±6	86±5	17%	79±5	10%	79±5	10%	89/141	94±5	9%	130/141	123±6
MPK2	108/202	122±6	46±4	132±6	7%	134±6	9%	124±7	2%	158/202	145±5	9%	187/202	155±5	7%
MPK3	36/168	63±5	45±7	76±6	17%	69±4	9%	71±7	11%	61/168	95±6	20%	146/168	261±34	64%
MPK4	34/91	62±5	38±6	72±5	14%	70±5	11%	68±5	9%	61/91	89±4	19%	86/91	117±8	24%
MPK5	50/125	64±4	33±5	74±5	15%	67±4	4%	71±4	10%	88/125	92±5	20%	105/125	101±6	9%
MPK6	139/188	35±2	52±4	44±2	20%	40±2	13%	40±2	13%	142/188	45±2	2%	142/188	45±2	0%
MPK7	102/219	139±6	37±3	147±5	5%	153±5	9%	141±6	1%	128/219	154±5	5%	206/219	195±6	21%
MPK8	35/195	134±11	45±6	161±11	17%	147±9	9%	161±13	17%	85/195	217±10	26%	192/195	no data	--
MPK8*	24/32	167±14	35±7	191±21	12%	172±16	3%	186±22	10%	29/32	195±14	2%	30/32	252±32	24%
ABC1	59/133	136±9	45±4	194±8	30%	149±8	9%	186±9	27%	82/133	220±11	12%	119/133	286±18	32%
ABC2	56/168	181±11	41±4	229±11	21%	195±10	7%	218±12	17%	80/168	272±14	16%	138/168	355±23	35%
ABC3	34/83	226±14	34±5	294±17	23%	237±13	5%	286±20	21%	47/83	314±16	6%	63/83	360±25	18%
ABC4	81/112	109±8	63±6	150±8	17%	132±10	17%	130±8	16%	88/112	153±9	2%	102/112	175±11	14%

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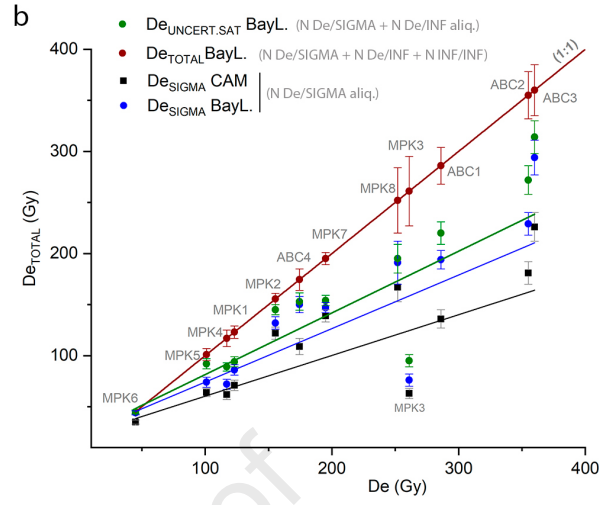
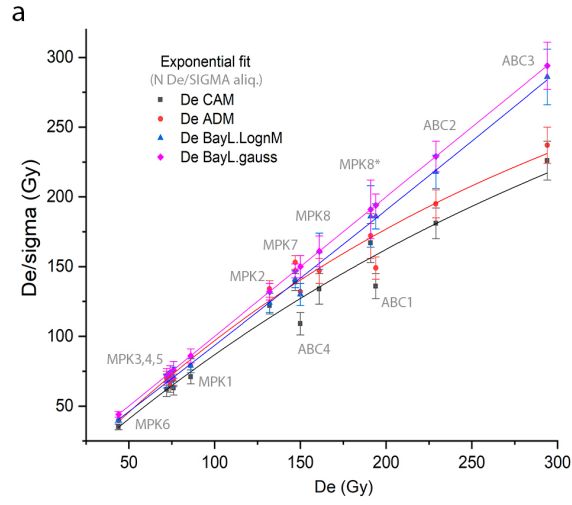
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**Table 3.** Ages for MPK and ABC samples, as function of the saturated aliquots included in bayesian calculations: De-SIGMA using ( $N_{De/SIGMA}$ ) aliquots, De-UNCERT.SAT using ( $N_{De/SIGMA} + N_{De/INF}$ ) aliquots and De-TOTAL using ( $N_{De/SIGMA} + N_{De/INF} + N_{INF/INF}$ ) aliquots. Ages using CAM with ( $N_{De/SIGMA}$ ) aliquots from previous work (Arce-Chamorro, 2017), except MPK6 and ABC3 samples for which minimum age model (MAM; Galbraith et al., 1999) were used.

Sample	DR-Total (Gy/ky)	Age CAM/MAM* De-SIGMA (ky)	Age BayL (g.) De-SIGMA (ky)	Age BayL (g.) De-UNCERT.SAT (ky)	Age BayL (g.) De-TOTAL (ky)
MPK1	1.5±0.06	65±13	58±11	64±6	84±8
MPK2	1.01±0.02	59±8	131±19	140±20	149±21
MPK3	1.21±0.04	95±19	64±17	78±14	214±44
MPK4	0.9±0.02	105±23	78±12	96±15	129±21
MPK5	1.15±0.01	77±11	63±5	80±7	89±8
MPK6	0.68±0.01	46±12	63±1	63±9	63±9
MPK7	1.37±0.07	99±22	107±22	109±21	140±26
MPK8	1.64±0.02	102±19	117±18	123±19	157±23
ABC1	0.69±0.01	180±46	243±32	272±37	412±49
ABC2	1.94±0.04	71±13	102±12	118±14	182±23
ABC3	1.07±0.02	72±20	227±32	277±39	335±48
ABC4	1.81±0.08	62±24	81±12	81±13	94±16

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**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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