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# Luminescence dating estimates for the coastal MSA sequence of Hoedjiespunt 1 (South Africa)

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**Abstract:** Coastal adaptations have been considered to play an important role in the bio-cultural evolution of early *Homo sapiens* and their dispersal out of Africa. In line with this assessment, recent years have seen increasing evidence for the exploitation of seafood from the Middle Stone Age (MSA) of northern and southern Africa. Yet, chronological control constitutes a key problem for better understanding the evolution of these behaviours, with little well-dated evidence for their origins in the late Middle Pleistocene and only few sites dated to before 100 ka. The shellfish-bearing MSA site of Hoedjiespunt 1 (HDP1), located in the Western Cape in South Africa, is one of the localities tentatively dated to the early Late Pleistocene. HDP1 has yielded a 1.5 m stratigraphic sequence with three phases of occupation, each containing abundant lithic artefacts, shellfish, terrestrial fauna, ostrich eggshell and pieces of ground ochre. Analyses of the cultural and zooarchaeological remains of HDP1 have provided evidence for systematic gathering of a narrow range of marine resources coupled with stable adaptations to coastal landscapes. Here we present the first complete absolute chronology for the site and its entire stratigraphic sequence by applying luminescence dating (OSL) to quartz grains extracted from the carbonate-rich sediments. The results show repeated human occupations during Marine Isotope Stages 5e-5c (~130-100 ka) at HDP1, suggesting successful integration of coastal ecosystems into the regular repertoire of behavioural adaptations by early modern humans by no later than the beginning of the Late Pleistocene.

## 1) Introduction

The earliest adaptations to coastal landscapes and the exploitation of seafood resources have become important topics in the past years for understanding the bio-cultural evolution of *Homo sapiens* during the Middle/Late Pleistocene (Erlandson, 2001; Parkington, 2003;

39 Cunnane and Stewart, 2010; Marean, 2014; Will et al., 2019). Here we employ the broad  
40 evolutionary definition of coastal adaptations that we have proposed previously (Will et al.,  
41 2016; 2019) including various behaviors such as the regular consumption and systematic  
42 acquisition of marine food resources and the expansion of settlement systems to include  
43 coastal and near-coastal zones as occupation spots on a regular and planned basis (for a  
44 similar but even broader term of coastal engagement see Faulkner et al. 2021).

45 Main questions of current research concern the antiquity of these behaviors (Marean et  
46 al., 2007; Jerardino and Marean 2010; Ramos-Muñoz et al., 2016) and whether *Homo sapiens*  
47 alone or also other species such as Neanderthals display these adaptations (Klein and Steele,  
48 2008; Stringer et al., 2008; Cortés-Sánchez et al., 2011; Marean, 2014; Will et al., 2019; Zilhao  
49 et al., 2020). Further research topics include the consequences of marine food consumption  
50 for human brain evolution and demography (Crawford et al., 1999; Parkington, 2001;  
51 Broadhurst et al., 2002; Parkington, 2003, 2010; Marean, 2011; Kyriacou et al., 2014; Marean,  
52 2016; Will et al., 2015) as well as the potential of coastal landscapes to facilitate their  
53 dispersals such as for the exit of *Homo sapiens* from Africa (Stringer, 2000; Walter et al., 2000;  
54 Mellars et al., 2013; Will et al., 2019). The wide range of topics associated with coastal  
55 adaptations and the surge of studies in the past twenty years illustrate their wider relevance  
56 in understanding early human cultural and biological evolution.

57 In this contribution we are particularly interested in the first of these questions, namely  
58 the age of coastal adaptations. The oldest and longest record for the consumption of marine  
59 resources derives from coastal MSA sites in northern and southern Africa with the majority of  
60 evidence coming from South Africa. The oldest currently known occurrence comes from Benzú  
61 Cave in northern Africa (~254 ka; Ramos et al., 2008; Ramos-Muñoz et al., 2016) and Pinnacle  
62 Point Cave 13B in southern Africa at ~164 ka (Jerardino and Marean, 2010; Marean et al.,  
63 2007), the only sites in Africa with potential evidence predating the Eemian interglacial. The  
64 age of these finds remains debated and the evidence for marine resource use at both sites for  
65 the late Middle Pleistocene are scant (e.g., Will et al., 2019; Tribolo, 2020). This potential MIS  
66 8-6 evidence is followed in time by another disputed occurrence at Abdur in Eritrea dated to  
67 approximately MIS 5e (Walter et al., 2000) and more recently dated evidence at Ysterfontein  
68 1 with the oldest ages around 120-113 ka (Niespolo et al., 2021; see also Avery et al., 2008;  
69 Klein et al., 2004). Postdating the Eemian interglacial after ~110 ka, multiple sites from South  
70 Africa show ample evidence for coastal adaptations (Fig. 1.), including on the southern coast

71 Klasies River (Voigt, 1973; Thackeray, 1988), Blombos Cave (Henshilwood et al., 2001;  
72 Langejans et al., 2012), Die Kelders Cave 1 (Grine et al., 1991; Marean et al., 2000), Klipdrift  
73 Shelter (Henshilwood et al., 2014) and Herolds Bay Cave (Brink and Deacon, 1982; Inskeep,  
74 1987), and on the western coast, Ysterfontein 1 (see above), Diepkloof (Steele and Klein,  
75 2013), Varsche Rivier 003 (Steele et al., 2016), Sea Harvest (Volman, 1978) and Hoedjiespunt  
76 1 (HDP1; Volman, 1981; Parkington, 2003; Will et al., 2013). More recent research has shown  
77 that northern Africa likewise harbors evidence for coastal adaptations, mostly during MIS 5c-  
78 3 (Steele and Alvarez-Fernández, 2011; Dibble et al., 2012; Stoetzel et al., 2014; Campmas et  
79 al., 2016; Ramos-Muñoz et al., 2016). In Europe, while some authors have questioned the  
80 extent to what Neanderthals were adapted to coastlines and marine resources (Klein and  
81 Steele, 2008; Marean, 2014; but see Stringer et al., 2008; Zilhão et al., 2010; Cortés-Sánchez  
82 et al., 2011; Zilhao et al., 2020), there is now good evidence derived from ~30 sites distributed  
83 along the western Atlantic until eastern Mediterranean coasts in the timeframe of MIS 5-3  
84 (see overview in Will et al., 2019). In sum, whatever the period, there is good evidence for  
85 coastal adaptations during the Late Pleistocene in sites which escaped the high sea level of  
86 the Holocene and Eemian interglacials. Yet data concerning these behaviors remain rare and  
87 disputed before ~100 ka.

88         Here is where the open-air site of Hoedjiespunt 1 (HDP1), located in the Western Cape  
89 of South Africa, comes into play. From a historical perspective, the site has already been  
90 known since the 1970s but only from surface collections, indicating a potential association of  
91 MSA artefacts with remains of shellfish (Volman, 1978). The first excavations that revealed  
92 the importance of HDP1 took place between 1993 and 1998, conducted by John Parkington  
93 (Parkington, 2003; Parkington et al., 2004), and mostly focused on the paleontological layers  
94 of the site (Berger and Parkington, 1995). These campaigns were followed in 2011 by fieldwork  
95 of a joint team from the University of Tübingen and the University of Cape Town directed by  
96 Nicholas Conard, exclusively in the archaeological deposits. The renewed excavations  
97 uncovered a short stratigraphic sequence of archaeological layers with *in situ* evidence for  
98 MSA stone artefacts associated with shellfish and other faunal remains (Will et al., 2013, 2015;  
99 Kyriacou et al., 2015). Crucially, HDP1 lies 15-12 meters above sea level (a.s.l.) and thus has  
100 the potential to preserve sediments predating the Eemian high sea level which was  
101 approximately 9-6 meters higher than at present in southern Africa and globally (Miller et al.,  
102 2005; Hearty et al., 2007; Carr et al., 2010).

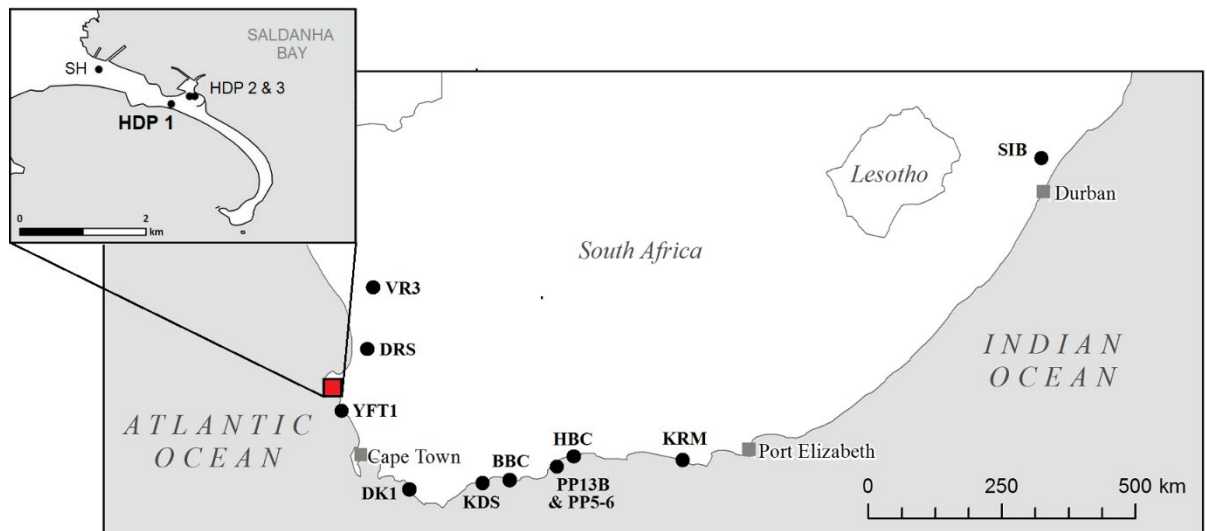
103 Absolute dating of the site has, however, proven difficult so far. Initial geological age  
104 estimates by Butzer placed the site at ~70-60 ka (in Volman, 1978) with more recent work  
105 (Butzer, 2004: 1777) reiterating this placement within early MIS 4. Absolute age  
106 determinations based on OSL/IRSL (Optically stimulated luminescence/Infrared stimulated  
107 luminescence) readings of both quartz and feldspars grains from sediments, and U-series  
108 dates of ostrich eggshell from the Parkington excavations indicated a tentative age range  
109 between 130 and 100 ka for one of the archaeological horizons (AH II; Woodborne, 1999,  
110 2000; Parkington, 2003; pers. com. from anonymous reviewer) but this was never fully  
111 confirmed and published. Based on the previous dating efforts, and in concert with  
112 stratigraphical, artefactual, faunal and contextual lines of evidence from the Conard  
113 excavations, the most recent assessment by Will *et al.* (2013; 2015) suggested an age of MIS  
114 5e for much of the sequence. This would make HDP1 one of the oldest open-air site with good  
115 evidence for coastal adaptations in sub-Saharan Africa and provide a potential age link with  
116 the isolated MIS 6 evidence from Pinnacle Point 13B. In order to test this hypothesis and to  
117 base the absolute chronology of the site on secure evidence, however, modern dating by  
118 absolute chronometric methods is required. To this end, new sediment samples were taken  
119 from the section of the Conard excavations. In this paper, we present the ages obtained with  
120 the optically stimulated luminescence (OSL) dating method applied to quartz grains coming  
121 from nine sediment samples and discuss the further implications of these results.

122

## 123 **2) Site and samples**

124 The open-air locality of HDP1 lies about 110 km north-northwest of Cape Town (Fig. 1;  
125 S33°01'42", E 17°57'34") and is situated on the southern coast of the Hoedjiespunt Peninsula,  
126 which separates Saldana Bay from the Atlantic Ocean. The peninsula is largely composed of  
127 fossil dune deposits and in addition to several paleontological sites, also contains other  
128 archaeological localities, such as Sea Harvest, which is located about 1 km to the west (Fig. 1).  
129 HDP1 is situated in close proximity to the modern shoreline and consists of two sites: one  
130 containing archaeological materials at 15 m a.s.l., and another that is purely paleontological  
131 at 12 m a.s.l. A ~2 m thick deposit of sterile sands separates the archaeological deposits from  
132 the paleontological layers. The paleontological strata include a Middle Pleistocene hyena den

133 rich in fauna that yielded 14 human fossils, but no archaeology or marine shells (Berger and  
134 Parkington, 1995; Churchill et al., 2000).



135

136 Figure 1: Location of Hoedjiespunt 1 (HDP1) in relation to other MSA sites in southern Africa (top);  
137 picture taking during the excavation of the open-air sequence of HDP1 in 2011, showing its current-  
138 day proximity to the Atlantic Ocean (bottom). Abbreviations for archaeological sites: BBC-Blombos  
139 Cave; DK1-Die Kelders 1; DRS-Diepkloof Rock Shelter; HBC-Herold's Bay Cave; HDP1-Hoedjiespunt 1;  
140 KDS-Klipdrift Shelter; KRM-Klasies River Mouth; PP13B/PP5-6-Pinnacle Point Cave 13B & 5-6; SH-Sea  
141 Harvest; YFT1-Ysterfontein 1; SIB-Sibhudu; VR3-Varsche Rivier 003.

142

143 Combining both the Conard and Parkington field work, the archaeological deposits have  
144 been excavated over an area of 18 m<sup>2</sup>, with a volume of 1.4 m<sup>3</sup> and exposed a maximal  
145 thickness of 1.5 m. Previous results on the stone artefacts and ochre (Parkington et al., 2004;  
146 Will et al., 2013, 2015) as well as shellfish remains (Kyriacou et al., 2015) of both the

147 Parkington and Conard campaigns have already been published. The sequence at HDP1  
148 consists of three consecutive archaeological horizons (AH I–III), each containing *in situ* MSA  
149 lithic artefacts, ochre, shellfish, terrestrial fauna and abundant fragmented ostrich eggshells  
150 (Fig. 2). The strata are in primary context, slope slightly downwards to the south and lie either  
151 directly under a thick calcrete carapace or the modern surface (HUMUS). The strata, although  
152 intact, have been truncated by erosion in the downslope direction toward the beach. A  
153 correlation between excavated units of the Parkington and Conard excavations is possible  
154 though not one to one (e.g., AH I consists of facies CESA, SHEMA and NOSA in the old  
155 excavations). Here we will use both stratigraphic designations to indicate the exact location of  
156 the dating samples (Table 1).

157 AH I and III are light-colored strata composed of consolidated fine sand containing many  
158 marine shells in both complete and fragmented status. Micromorphological analysis of the  
159 deposits (Göden, 2014) show that AH I and III are composed largely (ca 85%) of rounded, sand-  
160 sized, calcareous bioclasts with a more minor component of ooids, peloids and glauconite  
161 grains. Sand-sized grains of quartz are also present, at around 10-15%. Coarser components  
162 include shell fragments and also fragments of calcrete. The bioclastic grains exhibit micritic  
163 coatings creating a chitonic c/f-related distribution. Rootlet channels are present in AH I and  
164 III (as in all other strata studied at the site) and they exhibit infillings of needle-fibre calcite  
165 and well as the local formation of calcite hypocoatings. Anthropogenic components in AH I  
166 and III are minor, but consist of fragments of charred organic matter, burnt and unburnt bone,  
167 and larger shell fragments. Sandwiched between AH I and III, AH II averages 20 cm and consists  
168 of compact, dark brown clay and organic-rich sediments and yielded many, mostly broken,  
169 marine shells and abundant quartz artifacts. AH II is readily distinguished from the other two  
170 strata based on its darker color, due to the higher content in organic material. It is more  
171 heterogeneous than the other units, with both finer grain size and higher content in coarse  
172 (>710µm) sedimentary material. Find densities are highest for AH II concerning lithics and  
173 faunal remains, yet with all AHs showing comparable densities of shellfish remains (11-13  
174 kg/m<sup>3</sup>; Will et al., 2015: Table 3). Thin sections analysed from AH II show that this strata, like  
175 AH I and III, consists largely of sand-sized, calcareous bioclasts. However, AH II is significantly  
176 enriched in finely comminuted organic matter, both carbonized and humified, as well as  
177 anthropogenic components such as burnt and unburnt bone, large shell fragments, and

178 charcoal. The groundmass appears locally phosphatized. Rootlet channels are more  
 179 numerous in AH II relative to AH I or III and have more pronounced needle-fibre calcite  
 180 infillings. Below the oldest archaeological horizon, AH III, lies a 2 m thick, sterile, calcareous,  
 181 shelly sand (SHES) named for its abundant land snails (*Trigonephrus*). SHES rests on top of the  
 182 paleontological layers of the site. Micromorphological analysis of SHES shows that it is  
 183 compositionally similar to AH I and III but lacks any significant anthropogenic components.

184 Nine sediment samples have been taken from the available sections for OSL dating.  
 185 Samples HDP2, 3 and 7 come from the NOSA/ AH I Unit, HDP1 and 6 from the DAMA/AHII unit,  
 186 and HDP4 and 8 from the NOSAII/AHIII unit (Table 1 and Fig. 2). HDP5 and 9 were taken at the  
 187 top of the SHES unit. Within NOSA/AH1, HDP3 is located clearly above HDP2 and 7. The exact  
 188 stratigraphic relationship between HDP2 and 7, HDP1 and 6, HDP4 and 8, and HDP5 and 9 is  
 189 not clear and each couple can be considered as a replicate for its stratigraphic unit (Fig. 2). All  
 190 samples were taken by night under red/orange light, after removing the surface of the  
 191 sediment previously exposed to sunlight.

192

OSL sample	elevation (mm)	unit (Parkington)	unit (Conard)
HDP3	5 034	NOSA	194
HDP2	4 763		AH I
HDP7	4 593		195
HDP1	4 425	DAMA	196
HDP6	4 354		AH II
HDP4	4 210	NOSA2	198
HDP8	4 112		AH III
HDP5	4 095	SHES	199
HDP9	4 085		SHES
			200

Table 1: Elevation (mm above datum taken with a total station during excavations), and attribution to units (according to both Parkington's and Conard's nomenclature) of the OSL samples.

201

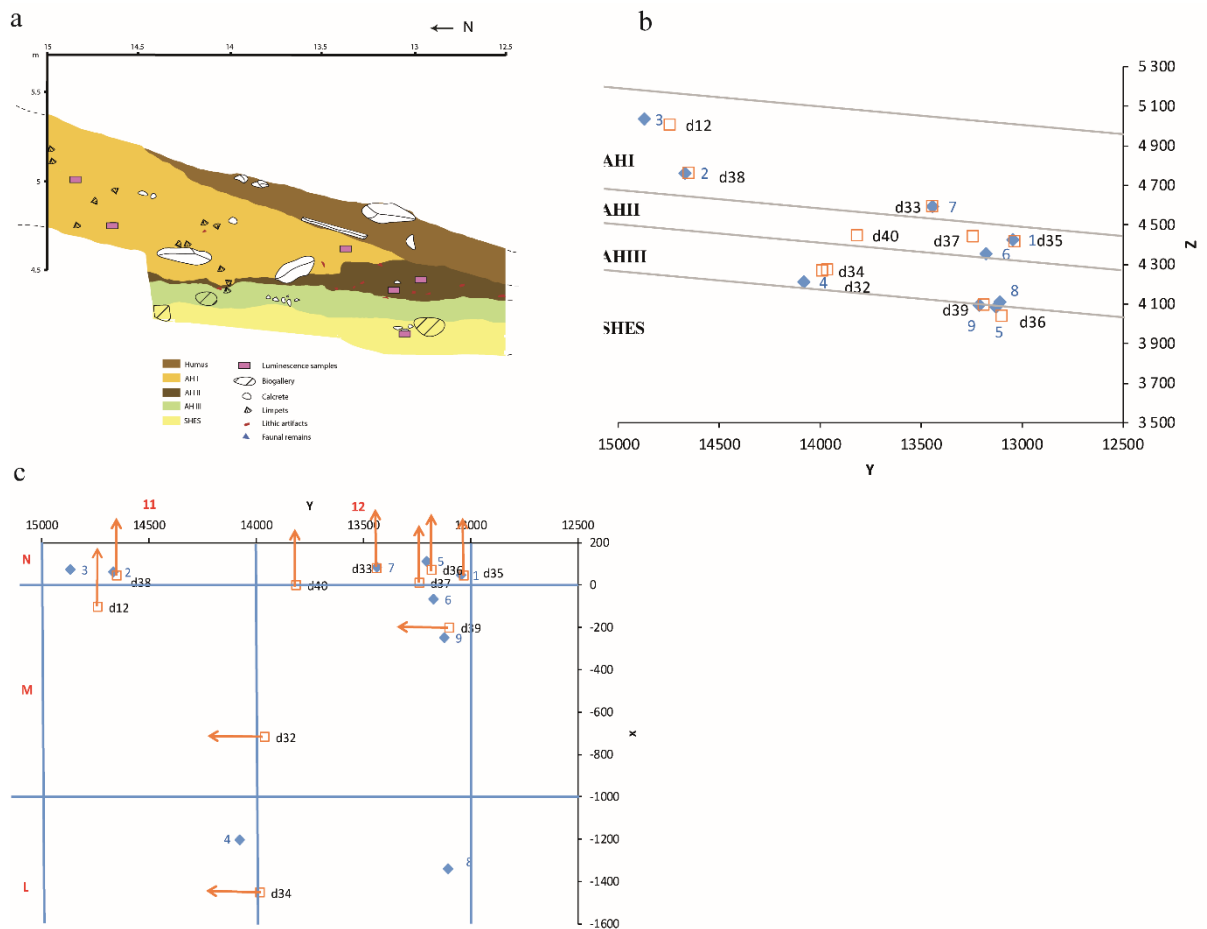
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206  
 207 *Figure 2: Stratigraphic sketch and location of the sediment samples for OSL dating and dosimeters. a)*  
 208 *North-South profile. b) Projection on the profile of the samples (blue diamonds) and dosimeters (red*  
 209 *squares); the approximate limits between the units are drawn (grey lines). c) Plan view, with the*  
 210 *location of the OSL samples (blue diamonds) and dosimeters; the red squares show the edge of the*  
 211 *dosimeter tubes that were apparent while the dosimeters were inserted into the section and were*  
 212 *recorded with the total station; the orange arrows symbolise the tube, with the Al<sub>2</sub>O<sub>3</sub>:C chips located*  
 213 *at the tip.*

214

### 215 3) Equivalent dose determination

#### 216 3.1 Preparation

217 The samples have been sieved to 710 μm, dried and weighted. The > 710 μm fraction  
 218 and part of the < 710 μm were saved for gamma spectrometry analyses (section 4). The rest  
 219 of the < 710 μm fraction was further treated in order to extract the quartz grains: wet sieving  
 220 at 100 and 140 μm was followed by HCl (10%), H<sub>2</sub>O<sub>2</sub> (30%), heavy liquid separation  
 221 (heteropolytungstate of sodium at 2.72 and 2.58 g/cm<sup>3</sup>). The quartz grains were then HF (40%)  
 222 etched for 60 min followed by HCl (10%) overnight and a final sieving at 80 μm. Note that  
 223 feldspar grains were also extracted but their study will not be presented here. The absence of  
 224 feldspar contamination in the quartz extracts was checked by applying depletion ratio tests,

225 following Duller (2003).. This was performed mainly on the dose recovery tests and once the  
 226 sample was claimed clean, this test was not repeated on the De measurements.

227 The 100-140 µm quartz grains were mounted either on multi-grains discs using silicon  
 228 oil or on single grain discs displaying 100 holes, each 150 µm diameter and 150 µm deep, so  
 229 that there was only one grain per hole.

### 230 3.2 Measurement devices

231 Measurements were performed on a Lexsyg Smart for multi-grain measurements and  
 232 on three Risoe TL/OSL DA 20 single grain devices for single grain measurements (Bøtter-  
 233 Jensen, 2000, 2003; Richter et al., 2015). Table 2 gathers the main information about the  
 234 excitation and detection systems that were then used for the Hoedjiespunt samples.

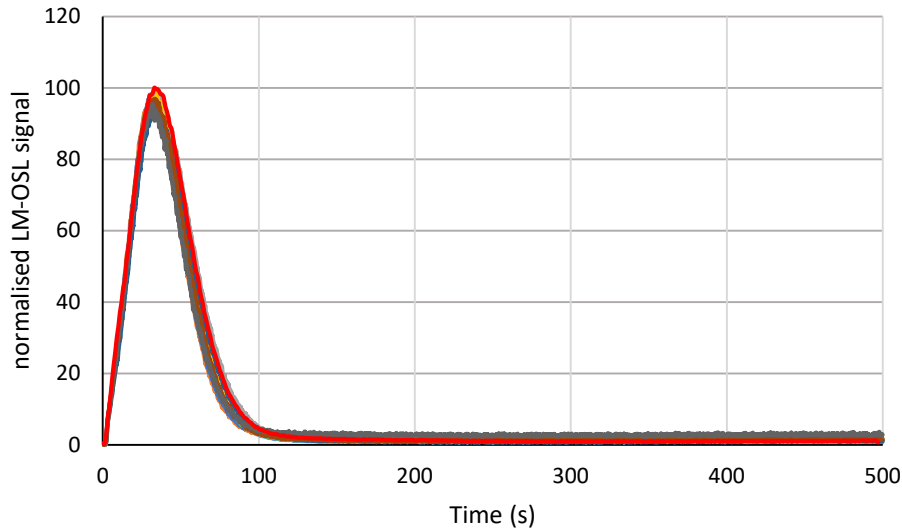
Reader	excitation	detection	source
Lexsyg Smart (multi-grain)	green LED (525 nm)	3mm Schott BG3+ 5 mm Delat BP365/50 EX - interference in front of a H7360- 02 PM tube (range 330-380 nm)	90Sr/90Y beta source, 0.19 Gy/s
Risoe TL/DA 20 (single grain)	10 mW Nd:YVO4 diode-pumped green laser (532 nm); IRSL (850nm) LEDs; blue LEDs (525 nm)	7 mmm Hoya U340 in front of Q 9235 PM tube (range: 280-380 nm)	90Sr/90Y beta source, 0.10 to 0.14 Gy/s

235 *Table 2: technical details about the stimulation, detection and irradiation systems of the Risø and Lexsyg luminescence readers*  
 236 *used in this work. The 532 nm laser stimulation was used for single grain measurements, the blue LEDs stimulation for multi-*  
 237 *grain measurements and the IRSL stimulation for testing the feldspar contamination.*

### 238 3.3 Protocol

239 The quartz grains from Hoedjiespunt are dominated by the fast component as shown by  
 240 a LM-OSL test (Fig. 3). Therefore, a Single Aliquot and Regenerative dose (SAR; Murray and  
 241 Wintle, 2000, 2003) protocol has been tested and applied (Fig. 4).

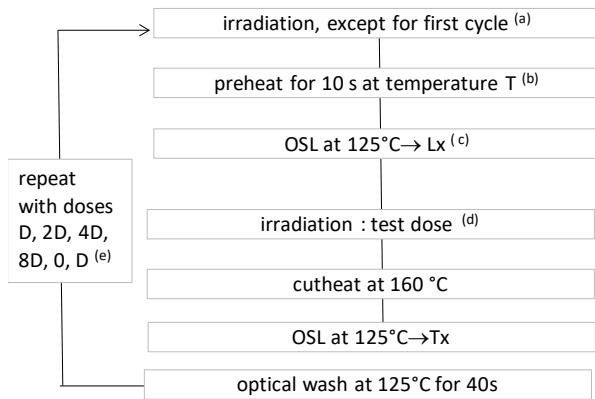
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243

244 *Figure 3: LM-OSL test for all the samples. One multi-grain aliquot of each sample was optically*  
 245 *stimulated at 125°C while increasing linearly the stimulation power from 0 to 70 mW/cm<sup>2</sup> in 500 s. The*  
 246 *red glow curve corresponds to the calibration quartz from Risø (Hansen et al., 2015), that is known to*  
 247 *be dominated by the fast component and is used as a reference.*

248



249

250 *Figure 4: SAR protocol applied for the equivalent dose determination. a) the first cycle corresponds to the measurement of the*  
 251 *natural signal; b) T was fixed at 260°C for 10 s; c) the OSL stimulation of single grains was performed for 1.6 s. Lx (and Tx) was*  
 252 *calculated from the integration of the first 0.1 s for signal and last 0.20 s for background; d) the test dose was either about 7*  
 253 *Gy or 33 Gy; e) D was about one third of the expected De.*

254

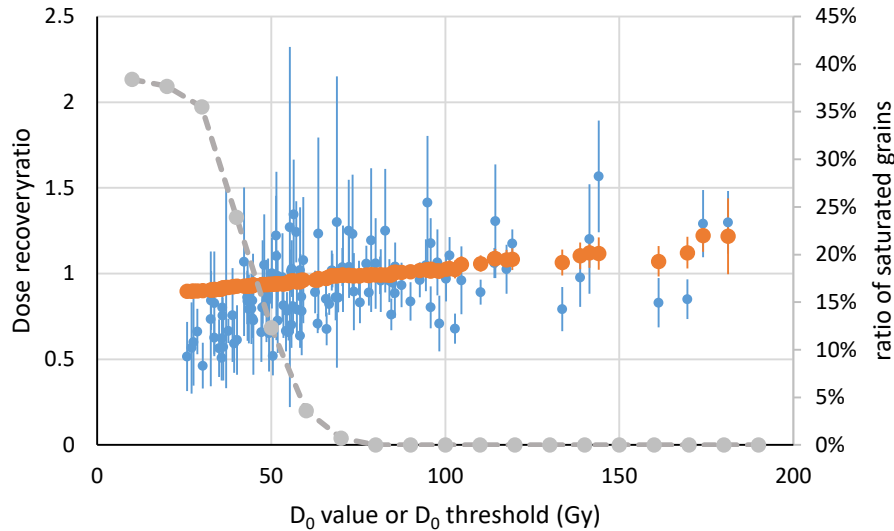
255 A minimal requirement for SAR is that it allows to recover a known laboratory given  
 256 dose. A dose recovery test (DRT) was thus performed on single grains aliquots, where the  
 257 samples were first bleached for one minute in a solar simulator (Honle UVAcube 400), left in  
 258 the dark at room temperature for at least 10000 s, and bleached again in the reader for 200 s

259 at 30°C. Then a dose, close to the equivalent dose estimated on multi-grain preliminary runs,  
260 was given and the protocol was applied. Analysis of single-grain data was performed with  
261 Analyst v4.57 (Duller, 2015). Grains were selected based on the sensitivity of each grain (test  
262 dose of the “natural” signal > 3 times background signal; relative error on “natural” test dose  
263 signal < 10%), and on the recuperation signal (normalised signal for the zero Gy dose < 5% of  
264 the normalised signal of the highest regeneration point). The recycling ratio was not used as  
265 a rejection criteria, because one of the luminescence reader is equipped with an  
266 heterogeneous radioactive source and corrections are required according to the disc position  
267 (CorrSGbin software, provided by DTU Nutech). Apparent irradiation times are re-calculated  
268 and thus highlights the grains that have moved significantly between the two recycling cycles  
269 and for which the calculation of a recycling ratio is therefore irrelevant. However, considering  
270 only the grains for which the recycling ratio could be confidently determined, one found that  
271 for these samples there is no correlation between the recycling ratios (most standing between  
272 0.75 and 1.25) and the equivalent doses or overdispersions. This observation is in agreement  
273 with Thomsen et al., (2016) and Guérin et al. (2015) who have already argued that, while  
274 reasonable, the recycling rejection criteria has no strong impact on the mean/central De  
275 calculation (at least on the samples they observed).

276 The dose response curve was fitted with a saturated exponential such as:

277 
$$\frac{Lx}{Tx} = I_0 \left[ 1 - \exp\left(-\frac{(D+b)}{D_0}\right) \right]$$
, where D is the regenerative dose, Lx/Tx is the normalised  
278 luminescence signal and I<sub>0</sub>, b and D<sub>0</sub> are fitting parameters. In addition to the grains discarded  
279 because of the rejection criteria mentioned above, numerous grains (see Table 3 and 4) were  
280 also rejected when no intercept of the normalised natural signal (Ln/Tn) onto the growth curve  
281 was possible, i.e., the natural signal is at or above the saturation limit I<sub>0</sub>. Beside these usual  
282 selection criteria, and following Thomsen et al., (2016), the D<sub>0</sub> value (saturation parameter)  
283 was also used as an additional criterion: in practice, the equivalent doses are ranked in  
284 function of the corresponding D<sub>0</sub> values and the central doses (after Galbraith et al., 1999) are  
285 calculated while progressively discarding the grains with the lowest D<sub>0</sub> values. This leads to  
286 increasing central dose values as a function of the D<sub>0</sub> value until a plateau is reached. In  
287 addition, the proportion *p* of grains rejected because of the saturation parameter (within the  
288 population of grain that pass the first set of rejection criteria) was calculated in function of D<sub>0</sub>.  
289 The lowest D<sub>0</sub> for accepted grains was thus within the central De plateau, where less than 5%

290 of grains were rejected because of saturation. Figure 5 displays this analysis for sample HDP3:  
 291 the final central equivalent dose was calculated for  $D_0 > 70$  Gy, in the plateau area, where  $p <$   
 292 5%, in order to avoid any bias due to saturation issues.



293

294 *Figure 5: Results of dose recovery tests for HDP3. Blue dots: Dose recovery ratio as a function of  $D_0$  value for each grain that*  
 295 *pass the selection criteria and is not saturated. Orange dots: the dose recovery ratio calculated after the CAM in function of*  
 296 *the minimum  $D_0$  value considered in the distribution. Note that there are two additional points with much larger  $D_0$  values*  
 297 *that have not been plotted here for the clarity of the graphic. Grey dots: percentage of saturated grains over the total number*  
 298 *of grains passing the selection criteria, above a  $D_0$  threshold value. The final dose recovery ratio is considered in the plateau*  
 299 *region (here above 70 Gy), where less than 5% of the grains show saturation.*

sample	tested dose (Gy)	N	n1	n2	n	$D_0$ (Gy)	dose recovery ratio CAM	OD (%)
HDP3	100	1000	175	122	38	80	1.00 ± 0.03	9 ± 3
HDP2	100	1000	69	50	23	80	1.04 ± 0.03	7 ± 5
HDP7	100	900	195	129	49	80	0.95 ± 0.02	8 ± 2
HDP1	114	1100	116	67	35	80	0.97 ± 0.02	5 ± 4
HDP6	159	1000	167	93	12	160	0.97 ± 0.03	2 ± 11
HDP4	143	900	58	40	16	110	1.00 ± 0.05	12 ± 5
HDP8	133	500	33	21	11	80	1.02 ± 0.05	11 ± 5
HDP5	239	1200	154	84	13	200	0.94 ± 0.05	8 ± 7
HDP9	245	1400	51	23	8	150	0.98 ± 0.11	24 ± 9

300

301 *Table 3: Dose recovery ratios. N: number of tested grains.  $n_1$  number of grains passing the selection*  
 302 *criteria.  $n_2$ : number of grains passing the selection criteria with unsaturated luminescence signals. n:*  
 303 *number of grains also passing the  $D_0$  criteria and present in the final  $De$  distribution.  $D_0$ : threshold value*  
 304 *of  $D_0$ , above which the (n) final grains are selected. CAM: the calculation of the dose recovery ratio is*  
 305 *based on the Central Age Model of Galbraith et al., (1999). OD: overdispersion.*

306

307

308 Table 3 shows the results for the dose recovery tests. The relevant “mean” dose is  
309 calculated from the Central Age Model (CAM, Galbraith et al., 1999). The ratios of obtained to  
310 expected dose are either within 5% of unity and/or consistent with unity at two sigma, and  
311 are thus claimed satisfactory. Note that the overdispersion (OD) stands between  $2\pm 11$  and  
312  $24\pm 9$  % (mean  $10\pm 6$  %). Because of the low number of accepted grains, the uncertainty  
313 associated to each OD is large, but these values are consistent with 10%-15%, which is not  
314 unusual for dose recovery tests (e.g. Thomsen et al., 2016).

315 3.4 results

316 The equivalent doses were then measured with the same readers and protocols as for  
317 the dose recovery tests. Between 2300 and 8900 grains were analysed for each sample (Table  
318 4). Only 3 to 8% pass the first set of selection criteria and are not saturated. After the  $D_0$  test,  
319 1 to 3% of the measured grains remain in the final  $D_e$  distribution, down to 0.3% for HDP9. It  
320 is worth noting that in every  $D_e$  distribution but for HDP1 and 2, a population of grains  
321 consistent with a modern age (dose close to 0 Gy) is present (Fig. 6). These low dose grains  
322 are not related to a low  $D_0$  value effect. While bioturbations have been observed on these  
323 sections (e. g. fine root channels, often associated with rhizolith formation) they do not come  
324 with quartz silt infilling. A more likely explanation for the presence of these 0 Gy quartz grains  
325 could be the strong wind occurring all the time at that place, which could have polluted the  
326 samples during their extraction on the site. The  $D_0$  threshold allows discarding most of these  
327 grains however and the grains with negative equivalent dose have been discard before the  
328 calculations of the mean/central dose. The few remaining grains have not been discarded,  
329 because they are associated with large uncertainties and their weight in the final distribution  
330 is negligible, and we wanted to avoid subjective selection. Table 4 displays the equivalent  
331 doses, based on the CAM, together with the over-dispersion values. However, the relevance  
332 of the CAM here is disputable and further discussed in section 5.

333

334

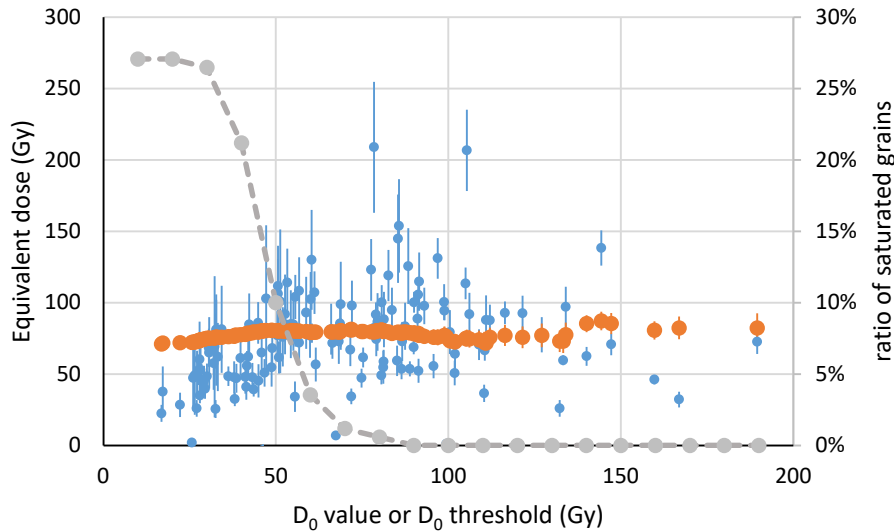
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336

Unit	sample	N	n1	n2	n	D <sub>0</sub> (Gy)	Equivalent doses (Gy)							
							CAM		ADM		FMM (CAM based)		logN_M	Gauss
							De	OD (%)	De	OD (%)	1	2		
NOSA/AHI	HDP3	2300	193	146	81	60	77.9 ± 3.5	37 ± 4	82.3 ± 3.5	33 ± 6			81.8 ± 4.4	86.2 ± 4.5
	HDP2	6700	428	288	102	80	88.9 ± 4.5	48 ± 4	98.7 ± 6.2	45 ± 5			90.0 ± 5.7	102.3 ± 6.4
	HDP7	3700	414	314	104	80	84.4 ± 3.8	41 ± 3	90.8 ± 4.1	38 ± 5			80.4 ± 4.6	89.0 ± 4.9
DAMA/AHII	HDP1	4500	377	225	93	90	116.0 ± 4.3	32 ± 3	120.7 ± 4.3	28 ± 3			123.4 ± 4.9	129.3 ± 4.7
	HDP6	7000	580	354	93	120	127.1 ± 6.4	45 ± 4	138.8 ± 7.4	42 ± 5			132.9 ± 8.3	149.1 ± 8.2
NOSA2/AHIII	HDP4	7100	629	373	63	150	150.0 ± 12.5	64 ± 6	181.8 ± 16.8	62 ± 4	91.6 ± 8.6 (54%)	271.0 ± 28.1 (46%)	153.8 ± 13.1	184.1 ± 14.6
	HDP8	7200	716	431	78	120	120.2 ± 7.0	48 ± 4	133.4 ± 9.4	46 ± 5	105.6 ± 10.3 (81%)	217.9 ± 60.0 (19%)	123.4 ± 8.3	139.3 ± 8.4
SHES	HDP5	8900	568	268	61	180	259.3 ± 14.3	40 ± 4	277.9 ± 16.5	37 ± 5			262.8 ± 14.5	279.7 ± 13.5
	HDP9	8200	785	464	24	450	181.1 ± 25.0	65 ± 10	220.8 ± 32.0	63 ± 6	104.4 ± 16.6 (51%)	322.5 ± 53.1 (49%)	275.9 ± 39.2	301.0 ± 36.2

Table 4: Equivalent dose estimates for quartz grains. N, n<sub>1</sub>, n<sub>2</sub>, n, D<sub>0</sub>, CAM, OD: see Table 3. ADM: average dose model, FMM: Finite Mixture Model, LogN\_M: Bayesian calculation with a log-normal model and search of the median, Gauss: Bayesian calculation with a Gaussian model

337  
338  
339  
340



341

342 Figure 6: Equivalent dose (De) distribution for sample HDP3. Blue dots: De in function of D<sub>0</sub> value for each grain that pass the  
343 selection criteria and is not saturated. Orange dots: De calculated after the CAM in function of the minimum D<sub>0</sub> value  
344 considered in the distribution. Note that there are additional points with much larger D<sub>0</sub> values that have not been plotted  
345 here for the clarity of the graphic. Grey dots: ratio of the number of saturated grains above a D<sub>0</sub> threshold value, over the total  
346 number of grains passing the selection criteria. The final De is considered in the plateau region, where less than 5% of the  
347 grains show saturation.

348

349 The OD for the equivalent dose measurements stand between 32±3 % and 48±4 % for  
350 all samples, but for HDP4 (64±6 %) and HDP9 (65±10 %). Visual inspection of the De  
351 distributions for these two samples suggests that they bear two distinct populations (Fig.sup  
352 A and C). This is further discussed in section 5.

353

#### 354 4) Dose rate

355 The total dose rate is the sum of the contributions from the gamma, beta and cosmic  
356 dose rates. Since the quartz grains have been HF etched, we assume that the alpha dose rate  
357 can be neglected.

358 4.1 cosmic and gamma dose rates

359 For the cosmic dose rate, we have applied the equation of Prescott and Hutton, (1994).  
360 This takes into account the burial depth of the sample, its geographical coordinates and the  
361 thickness and density of the overlaying sediment. We have assumed that the current burial  
362 depths were representative of the past mean ones. The cosmic dose rates stand between  
363  $0.18\pm 0.02$  and  $0.20\pm 0.02$  Gy/ka and represent between 16 to 25% of the total dose rates  
364 (Table 5).

365 The gamma dose rates were calculated from *in situ* dosimetry: 10 tubes, each containing  
366 3  $\text{Al}_2\text{O}_3\text{:C}$  pellets, have been inserted 30 cm deep into the stratigraphic sections as close as  
367 possible from the sediment samples (Fig. 2), and left for about one year (Bøtter-Jensen et al.,  
368 1997; Kreutzer et al., 2018). The recorded doses, corrected for the cosmic contribution, show  
369 very little scatter (10% RSD on the gamma dose for the 10 tubes, 6% for the total dose over  
370 the 30 chips) (Table Sup A), though they are slightly higher for the organic rich DAMA/AHII  
371 layer. Each sample was attributed the gamma dose deduced from the closest dosimeter, but  
372 for HDP8, for which no close dosimeter was available and thus the mean of all dosimeters was  
373 applied.

374 Variation of moisture over time induces variation of the dose rate. The mean past water  
375 content should then be taken into account. Meanwhile, it is impossible to precisely assess  
376 what it was. Moisture content was measured for the day of sampling (Table 5) and shows high  
377 variability among the samples. It seems correlated with the constituents (organic vs shelly):  
378 the organic-rich unit (DAMA/AHII) displays water content around 30% (mass of water/mass of  
379 dry sediment), while the shelly-sandy units are nearly dry (1% for SHES). In order to at least  
380 take these different power of water retention into account, no correction was applied to the  
381 gamma dose rates recorded by the dosimeters (since they already take into account the  
382 various moisture contents), but a 10% systematic error was arbitrarily added, based on the  
383 extreme values that can be calculated if the past and current water content had been  
384 drastically different. The gamma doses stand between  $0.30\pm 0.04$  Gy/ka and  $0.41\pm 0.04$  Gy/ka.  
385 They contribute for 31 to 39% of the total dose rates.

386 4.2 beta dose rates: introduction

387 The beta dose rates for each emitter (K, U, Th) can be calculated following the equation:



388 
$$D_{s+w} = \frac{D_s}{1 + k \left( \frac{m_w}{m_s} \right)}$$

389 equivalent to

390

391

392 
$$D_{s+w} = \frac{a\delta A_s}{1 + k \left( \frac{m_w}{m_s} \right)}$$

393

394 with,  $D_{s+w}$ : beta dose rate for either U or Th or K for the wet sediment;  $D_s$ : beta dose rate for either U  
 395 or Th or K for the dry sediment;  $m_s$ : mass of dry sediment;  $m_w$ : mass of water;  $A_s$ : activity of U or Th, or  
 396 K content, for the dry sediment;  $k$ : correction factor for water content for either U or Th or K;  $a$ :  
 397 attenuation factor depending on grain size for either U or Th or K;  $\delta$ : specific beta dose rate for either  
 398 U or Th or K.

399 The calculation of the beta dose rates was complicated by the high carbonate content  
 400 of the sediment (up to 800 % of mass of carbonate over non-carbonated material in the < 710  
 401  $\mu\text{m}$  fraction; see Table 5 and Table sup A), due to the presence of bioclastic sands, shell and  
 402 calcrete fragments of all sizes. This raises three distinct questions: 1) Are there significant  
 403 differences in absorption of radiative energy compare to siliceous materials, i.e. are the  
 404 attenuation factors  $a$  and corrections for water contents  $k$  different from the ones applied in  
 405 siliceous matrix (Nathan and Mauz, 2008; Guérin et al., 2012)? 2) Is the model of infinite matrix  
 406 dose rate still applicable (Cunningham, 2016)? 3) What is the impact of this carbonated matrix  
 407 on the distribution of dose rate, thus on the distribution of equivalent doses and what does it  
 408 imply for the age (actually dose) model (Mayya et al., 2006; Guérin et al., 2017)? Questions 1  
 409 and 2 are discussed successively in this section, question 3 will be discussed in section 5.

410

unit	sample	grain-size	water	% fine/tot	%carbonate	dose rates (Gy/ka)						
						cosmic	gamma	beta1	beta 2	tot1	tot2	mean
NOSA	HDP3	100-140	7%	69%	558%	0.20 ± 0.02	0.31 ± 0.03	0.30 ± 0.03	0.29 ± 0.03	0.82 ± 0.05	0.81 ± 0.05	0.81 ± 0.05
NOSA	HDP2	100-140	10%	70%	676%	0.19 ± 0.02	0.35 ± 0.04	0.24 ± 0.02	0.23 ± 0.02	0.78 ± 0.05	0.76 ± 0.05	0.77 ± 0.05
NOSA	HDP7	100-140	17%	50%	473%	0.20 ± 0.02	0.34 ± 0.04	0.29 ± 0.03	0.25 ± 0.02	0.83 ± 0.05	0.80 ± 0.05	0.82 ± 0.05
DAMA	HDP1	100-140	28%	25%	152%	0.19 ± 0.02	0.41 ± 0.04	0.58 ± 0.06	0.55 ± 0.05	1.18 ± 0.08	1.15 ± 0.07	1.17 ± 0.07
DAMA	HDP6	100-140	22%	42%	97%	0.19 ± 0.02	0.39 ± 0.04	0.70 ± 0.07	0.64 ± 0.09	1.29 ± 0.08	1.22 ± 0.10	1.25 ± 0.09
NOSA2	HDP4	100-140	5%	73%	409%	0.18 ± 0.02	0.33 ± 0.04	0.35 ± 0.03	0.35 ± 0.03	0.87 ± 0.05	0.86 ± 0.05	0.86 ± 0.05
NOSA2	HDP8	100-140	4%	81%	251%	0.18 ± 0.02	0.34 ± 0.05	0.39 ± 0.03	0.42 ± 0.03	0.92 ± 0.06	0.94 ± 0.06	0.93 ± 0.06
SHES	HDP5	100-140	1%	93%	742%	0.18 ± 0.02	0.31 ± 0.03	0.32 ± 0.03		0.81 ± 0.05		0.81 ± 0.05
SHES	HDP9	100-140	1%	84%	735%	0.18 ± 0.02	0.30 ± 0.04	0.30 ± 0.03		0.78 ± 0.05		0.78 ± 0.05

411 *Table 5: data for dose rates. Water: mass of water over mass of dry sediment. %fine/tot: mass of dry <710 $\mu\text{m}$  fraction over*  
 412 *the total mass of dry sample. %carbonate: mass of carbonate over the mass of dry non- carbonated sediment. Cosmic dose*

413 *rates are deduced from Prescott and Hutton (1994) equation. Gamma dose rates are deduced from field gamma spectrometry.*  
414 *Beta 1 is the beta dose rate calculated from the K, U, Th contents of the <710µm fraction, from high resolution gamma*  
415 *spectrometry, and corrected for water content. Beta 2 is the beta dose rate calculated from the K, U, Th contents of the whole*  
416 *sample, from high resolution gamma spectrometry, and corrected for water content. Tot1 and tot2 are the total dose rates*  
417 *considering beta1 and beta2 respectively. Mean is the average of tot1 and tot2.*

418

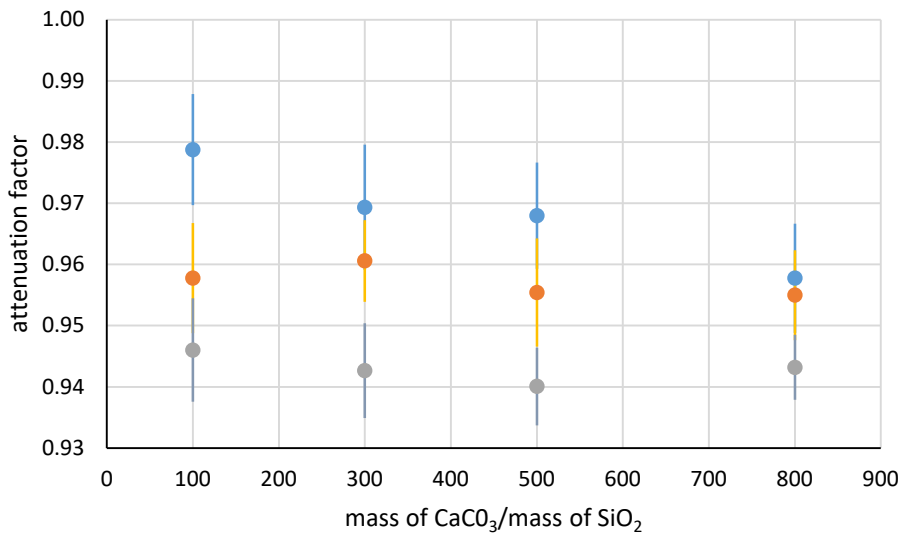
#### 419 4.3 determination of appropriate attenuation factors and water content correction 420 factors.

421 The attenuation factors  $a$  and corrections factors  $k$  in a carbonate-rich matrix were calculated  
422 with the DosiVox software (Martin, 2015, Martin et al., 2015a, Martin et al., 2015c), based on  
423 the Geant 4 toolkit version 10.01.p01 (Allison et al., 2006, 2016).

424 The modelled matrix was composed of SiO<sub>2</sub> and CaCO<sub>3</sub>, with a mass ratio (CaCO<sub>3</sub> / SiO<sub>2</sub>) varying  
425 from 100% to 800%. The dose received by 120 µm quartz grains was computed in assuming  
426 that U, Th and K are uniformly distributed within the carbonate rich matrix. Note that this  
427 model differs from the one of Nathan and Mauz (2008), where the carbonate is supposed to  
428 fill pores within the sedimentary matrix and be free of any radioactive material. Moreover,  
429 these authors only considered CaCO<sub>3</sub> contents up to 100% in mass (mass of carbonates over  
430 mass of siliceous material).

431 Attenuation factors  $a$  deduced from these calculations are shown on Figure 7. They are poorly  
432 dependent on the proportion of carbonates. Note, however, that they are significantly  
433 different from the ones proposed by Guérin et al., (2012) for a siliceous matrix, but for the  
434 potassium (0.96 in both cases): the attenuation factors are here  $0.96 \pm 0.01$  for U, instead of  
435 0.90, and  $0.94 \pm 0.01$  for Th instead of 0.85. These differences are due to the differences of beta  
436 absorption between the quartz grains and the matrices and to the method of calculation of  
437 the attenuation factor. Guérin et al., (2012) simulated the beta self-doses of SiO<sub>2</sub> grains in a  
438 SiO<sub>2</sub> matrix, and calculated the attenuation factors as the complementary of the self-dose.  
439 This calculation is possible when the grain and the matrix have the same beta absorption  
440 efficiencies. In our case the beta rays coming from the CaCO<sub>3</sub> / SiO<sub>2</sub> mixed matrix were  
441 simulated and the dose was directly evaluated in the SiO<sub>2</sub> grains. Doing this way is important  
442 since the quartz self-dose is affected by the mixed matrix because the beta backscattering is  
443 different than in a pure SiO<sub>2</sub> matrix. Similarly, the attenuation factor is affected because the  
444 beta absorption efficiency of a CaCO<sub>3</sub> / SiO<sub>2</sub> mixed matrix is significantly different from the one  
445 of a pure SiO<sub>2</sub> matrix.

446



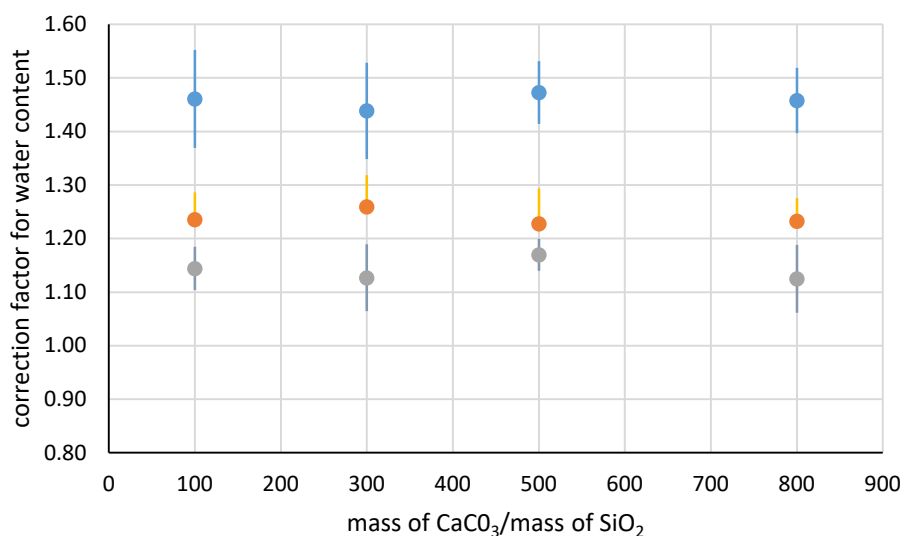
447

448 *Figure 7: attenuation factors for 120 μm grains, for beta dose from U series (orange dots), Th series (grey dots) and K (blue*  
449 *dots).*

450 Corrections factors  $k$  for water contents are shown on Fig. 8. Again, they are poorly dependent  
451 on the carbonate content. Therefore, means of  $1.46 \pm 0.04$ ,  $1.24 \pm 0.04$  and  $1.14 \pm 0.06$  were  
452 applied respectively for K, U and Th beta dose rates. While they significantly differ from the  
453 mean factor of 1.25 calculated by Zimmerman, (1971), but for U, we could observe that the  
454 final beta dose rates calculated with both set of correcting factors do not differ by more than  
455 2%. This results from mutual compensations of the difference between the two sets, and so it  
456 is likely that significant differences in the calculation of the beta dose rates will only occur in  
457 environments where the K, U, Th proportions are significantly different from the standard  
458 values. As for the calculation of the gamma dose rates, the current water contents were  
459 applied for the calculation of the final beta dose rates.

460

461



462

463 *Figure 8: correction factor k for water content for beta dose from U series (orange dots), Th series (grey dots) and K (blue*  
 464 *dots).*

465

466 4.4 dealing with heterogeneities.

467

468 Here we deal with the fact that the presence of coarser materials with different matrix  
 469 and radioactive contents than the fine (here <710µm) fraction where the quartz grains-  
 470 dosimeters come from, can induce heterogeneities in the beta dose rate (e.g. Martin 2015,  
 471 Martin et al., 2015b; Tribolo et al., 2017). In particular, Cunningham (2016) warned against  
 472 the use of the infinite matrix assumption in shell rich sediments, since the shells introduce  
 473 “cold spots”. However we argue here that the heterogeneity of the sediment samples from  
 474 HDP are not as critical as in the case described by Cunningham. All grain size fractions are rich  
 475 in carbonates (Table 5 and sup A) and the carbonate contents of the HDP samples are well  
 476 beyond the ones explored by Cunningham (<40% of total mass in his case). To sum up,  
 477 whatever the fraction, the matrix is carbonate dominated; this is different from the case  
 478 where “a few” shells are displayed in a sandy matrix. Moreover, the activities and radioactive  
 479 contents have limited variations between the <710µm and > 710µm fractions. Those have  
 480 been determined on a dried, finely crushed (i.e. homogenised) and sealed sub- samples, using  
 481 high resolution gamma spectrometry (Canberra, e.g. Guibert et al., 2009). Table 6 displays the  
 482 activities and radioactive contents results.

	Sample HDP	activities (Bq/kg)								content (%)	
		<sup>238</sup> U series						<sup>232</sup> Th series		K	
		top		middle		bottom					
fraction < 710µm	3	6.0	± 1.2	11.8	± 0.4	13.5	± 2.1	7.0	± 0.2	0.24	± 0.02
	2	8.2	± 1.1	9.4	± 0.3	5.9	± 2.0	5.4	± 0.2	0.18	± 0.02
	7	8.9	± 0.6	9.6	± 0.2	10.6	± 1.0	6.3	± 0.1	0.27	± 0.01
	1	24.8	± 1.5	15.8	± 0.4	12.7	± 2.4	15.9	± 0.3	0.62	± 0.02
	6	21.9	± 1.2	16.3	± 0.3	12.3	± 1.8	18.6	± 0.3	0.77	± 0.02
	4	10.4	± 1.0	13.4	± 0.3	10.3	± 1.8	6.6	± 0.2	0.26	± 0.01
	8	11.0	± 0.7	14.0	± 0.2	9.5	± 1.2	7.0	± 0.2	0.29	± 0.01
	5	9.2	± 1.0	10.4	± 0.3	8.9	± 1.8	5.3	± 0.2	0.23	± 0.01
	9	8.0	± 0.7	9.5	± 0.2	8.4	± 1.1	4.7	± 0.1	0.23	± 0.01
fraction > 710µm	3	9.6	± 1.1	11.9	± 0.3	8.9	± 1.9	5.9	± 0.2	0.17	± 0.01
	2	10.5	± 1.0	9.3	± 0.3	10.2	± 2.2	4.8	± 0.3	0.12	± 0.02
	7	8.1	± 0.5	7.9	± 0.1	8.4	± 0.9	4.7	± 0.1	0.19	± 0.01
	1	27.4	± 0.9	17.0	± 0.2	13.1	± 1.3	14.4	± 0.2	0.55	± 0.01
	6	21.4	± 0.7	14.5	± 0.2	10.8	± 1.1	14.8	± 0.2	0.62	± 0.01
	4	9.7	± 1.4	14.8	± 0.4	11.1	± 2.2	4.9	± 0.2	0.19	± 0.02
	8	25.0	± 1.4	25.7	± 0.4	12.9	± 2.2	6.4	± 0.2	0.39	± 0.02
	5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
	9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
total	3	6.9	± 0.9	11.8	± 0.3	12.3	± 1.6	6.7	± 0.2	0.22	± 0.01
	2	9.2	± 0.8	9.3	± 0.2	7.8	± 1.5	5.2	± 0.2	0.15	± 0.01
	7	8.5	± 0.4	8.8	± 0.1	9.5	± 0.7	5.5	± 0.1	0.23	± 0.01
	1	26.7	± 0.8	16.7	± 0.2	13.0	± 1.2	14.8	± 0.2	0.57	± 0.01
	6	21.6	± 0.7	15.3	± 0.2	11.5	± 1.0	16.4	± 0.2	0.68	± 0.01
	4	10.3	± 0.9	13.6	± 0.3	10.4	± 1.6	6.4	± 0.2	0.25	± 0.01
	8	12.7	± 0.7	15.3	± 0.2	9.9	± 1.1	6.9	± 0.1	0.31	± 0.01
	5	9.2	± 1.0	10.4	± 0.3	8.9	± 1.8	5.3	± 0.2	0.23	± 0.01
	9	8.0	± 0.7	9.5	± 0.2	8.4	± 1.1	4.7	± 0.1	0.23	± 0.01

484

485 *Table 6: activities for <sup>238</sup>U series or <sup>232</sup>Th series and K contents deduced from high resolution gamma spectrometry. The*  
486 *samples were divided into two distinct fractions: <710µm and >710µm. For the <sup>238</sup>U series: the top activities (pre-Rn) are*  
487 *deduced mainly from the <sup>234</sup>Th peak, the middle activities (post Rn) are deduced from <sup>214</sup>Bi and <sup>214</sup>Pb, the bottom activities*  
488 *are related to <sup>210</sup>Pb. nd: non determined. The amount of >710µm for HDP5 and HDP9 was assumed negligible (< 16%). The*  
489 *total activities and contents are calculated from the <710µm and >710µm data, weighted by the corresponding mass ratio.*

490

491

492 As expected from the nature of each sample and in consistency with the gamma dose  
493 rate results, the activities/contents are higher (about 3 times) in the DAMA/AHII organic rich  
494 layer than in the shelly layers, for both the <710µm and >710µm fractions. The Th activities  
495 and K contents are slightly higher (9±4% to 44±24%) in the <710 µm fraction than in the  
496 >710µm fraction (but for K, HDP8), while there is no clear tendency in the U activities, but the  
497 differences remain < 25% in most cases. Consequently, there is little difference between the  
498 beta dose rate calculated from the < 710µm fraction and the beta dose rate calculated from  
499 the entire sample (<2%) (applying the conversion factors δ<sub>i</sub> from Guérin et al., 2011). We  
500 conclude that the infinite matrix assumption remains a good approximation and the mean

501 beta dose rate calculated from the fine (<710µm) fraction, and from the entire sample, has  
502 been used for the final De calculation.

503

#### 504 4.5 beta dose rates: final comments and results

505 Note (Table 6) that the U series show disequilibrium for most samples: for all but HDP3  
506 and 7, the activity for the middle of the chain ( $^{226}\text{Ra}$  based on  $^{214}\text{Bi}$  and  $^{214}\text{Pb}$ ) is in excess (13  
507 to 59%) compared to the bottom (based on  $^{210}\text{Pb}$ ). The middle of the chain is in excess as well  
508 compared to the top (based on  $^{234}\text{Th}$ ) for all samples (up to 29% for most, 97% for HDP3) but  
509 for HDP 4 and 8 from NOSA2/AHIII, where a deficit is observed (26 to 36%). The chemical origin  
510 of these disequilibria and their local evolution over time is not straightforward and different  
511 processes (e.g. radon escape, leaching, etc.) could be involved. However, since U contributes  
512 for 20 to 30% of the total dose rate, it can be estimated that in extreme cases where the beta  
513 and gamma dose rates are calculated either from the top activities only or from the middle  
514 activities only, the total dose rates would not differ by more than 5% for all samples but HDP  
515 3 (12% in this case), compared to a steady case where they are calculated with the current  
516 activities. Therefore, the current dose rates (from the current radioisotopic content for the  
517 beta dose rate and from the *in situ* dosimetry for the gamma dose rates) were applied to  
518 calculate the final age estimates.

519

520 The final beta dose rates are, contrary to the gamma dose rates, highly dispersed and stand  
521 between  $0.23\pm 0.02$  Gy/ka and  $0.42\pm 0.03$  Gy/ka for the sandy-shelly layers of AH1, AHIII and  
522 SHES, and between  $0.55\pm 0.04$  Gy/ka and  $0.70\pm 0.07$  Gy/ka for the organic rich layers of AHII.

523

#### 524 **5. Ages and discussion**

525 The equivalent dose distributions are widespread, with overdispersions (OD) values  
526 standing between  $32\pm 3$  and  $48\pm 4$  %, up to  $64\pm 6$ % and  $65\pm 10$ % for HDP4 and HDP9 (Table 4).  
527 While a part of this OD might be due to the low presence of modern age grains (though this  
528 is not the case for HDP1 and 2 which have OD similar to those of the other samples), it is likely  
529 that a large part of this overdispersion can be attributed to the low K (but also U and Th)  
530 contents observed in all the samples: emitters might be far from each other and depending

531 on the distance between the quartz grains and these emitters, the beta dose rate could be  
532 more or less affected (see Mayya et al., 2006; Cunningham et al., 2012; Guérin et al., 2015).  
533 These authors have shown on simplified models that, in such a case, the dose rate distribution  
534 is positively skewed, and they have questioned the relevance of the central equivalent dose  
535 statistical model, such as defined in the Central Age Model (Galbraith, 1999) for calculating  
536 the ages (Guérin et al., 2015, 2017; Mayya et al., 2006). They have shown that the Central Age  
537 Model calculates the geometric mean of the De distribution, i.e. the median of a lognormal  
538 distribution, while the mean dose rate corresponds to an average (arithmetic mean),  
539 whatever the Dr distribution. Therefore, in case the Dr and the De distributions are highly  
540 skewed, the median and average De will not match and the CAM will lead to age  
541 underestimate.

542 Mayya et al., (2006) and more recently Chauhan et al., (2021) have suggested using a  
543 minimum equivalent dose, based on the equivalent dose distribution, together with a  
544 minimum dose rate (when the beta K contribution tends to zero; see also, David (2007) and  
545 Feathers et al., (2020), for two other similar examples). However, their calculation rests on a  
546 simplified model of a quartz matrix sediment with K-feldspar hotspots, in which the size of the  
547 K-feldspar grains for a given K content is critical. This information is lacking in the present case,  
548 making this model difficult to apply. In addition, it would require a recalculation of the  
549 distribution of beta travel range in a carbonated matrix, since it was originally calculated for a  
550 siliceous matrix, as well as taking into account the difference in pore space due to the presence  
551 of a multiple size fractions in the HDP sediments.

552 Guérin et al. (2017) have developed another approach where the mean beta dose rate  
553 is used (contrary to a minimum beta dose for Mayya et al., 2006), but a weighted average dose  
554 is (Average Dose Model) instead of the geometric mean dose of the CAM. Heydari and Guérin  
555 (2018) have further tested Bayesian modelling (based on the Baylum R-codes; Philippe et al.,  
556 2019). They have shown that the Bayesian approach, associated with Gaussian or  
557 Lognormal\_median distributions for the prior distribution of the equivalent doses also leads  
558 to more accurate results in the case of beta dose rate heterogeneity than the CAM.

559 De calculated with the different models (CAM, ADM, FMM, Bayesian-Gaussian and  
560 Bayesian-logNormal\_median) are reported in Table 4. Fig Sup. B helps comparing single  
561 component models. For all samples but HDP9, the four De estimates are consistent within 2

562 sigma, though the models based on the calculation of the average (ADM and Bayesian-  
563 Gaussian) tend to give higher  $D_e$  than the models based on the calculation of the median (CAM  
564 and Bayesian-logNormal\_median), as expected for positively skewed distributions.

565 For HDP3, 2, 7, 1 and 6, all models point to ages covering stages 5e to 5c (Fig. 9 and Table  
566 7). HDP2 appears to be in chrono-stratigraphic reversal. Compared to HDP7, 1 and 6 below,  
567 more grains for this sample have apparent ages over 300 ka (Fig.Sup A), despite a lower mean  
568 dose rate. Whether this is due to microdosimetric effects or to another reason is not clear for  
569 now.

570 The interpretation for the four lowest samples is more complex. The results for HDP9  
571 might be biased due to the low number of selected grains despite the higher number of  
572 measured ones. However, the OD of HDP 4 and 9 are higher and the visual inspections of the  
573  $D_e$  distributions (Fig.Sup A and Sup C) suggests that they might be both bi-modal. We applied  
574 the FMM (Roberts et al., 2000), with  $k$  (number of components) varying between 2 and 4 and  
575  $\sigma_b$  (OD of each component) varying between 0.2 and 0.4, according to the OD observed  
576 for the other samples. In all cases, the BIC and Maximum Likelihood criteria point to two  
577 components, with no significant dependence with the value of  $\sigma_b$ . Results for  
578  $\sigma_b=0.4$  have been reported in Table 4. For both HDP4 and HDP9, each component  
579 represents ca. 50% of the distribution and the lower components match with the general  
580 trend of  $D_e$  close to 100 Gy. The higher components, though imprecise, are both consistent  
581 with 300 Gy. HDP4 and HDP9 are close to the stratigraphic junction between SHES and  
582 NOSAII/AHIII. The  $D_e$  distributions suggest that there was either a natural (bioturbation)  
583 mixing of the two units, or the mixing occurred during the sampling, while the lighting  
584 conditions were not optimal to distinguish the stratigraphic limit of the two phases.

585 In theory, it is not reasonable to calculate ages when such mixing occurs (e.g. Tribolo et  
586 al., 2010, Guérin et al., 2017) because the significance of the mean dose rate is questionable.  
587 However here the total dose rate of the SHES and NOSA2/AHIII samples are close enough to  
588 allow this calculation. Consequently, the age for HDP4 would be  $106\pm 12$  ka, consistent with  
589 the marine isotopic stage 5c-5d observed for the upper samples (Table 7). The age for HDP9  
590 would be  $413\pm 64$ ka, rather imprecise again due to the low number of selected grains, but not  
591 inconsistent with the ages obtained for HDP5 ( $320\pm 28$  to  $346\pm 26$  ka depending on the statistic  
592 model).



unit	sample	Ages (ka)									
		CAM		FMM		ADM		LogN_M		Gauss	
NOSA	HDP3	96	± 8			101	± 8	101	± 8	107	± 8
	HDP2	116	± 10			128	± 12	117	± 10	134	± 11
	HDP7	104	± 9			111	± 9	99	± 8	110	± 9
DAMA	HDP1	99	± 8			104	± 9	106	± 8	111	± 8
	HDP6	102	± 10			111	± 11	107	± 10	120	± 11
NOSA2	HDP4	174	± 19	106	± 12	210	± 24	178	± 18	214	± 20
	HDP8	129	± 12	113	± 11	143	± 15	133	± 12	150	± 13
SHES	HDP5	320	± 28			343	± 31	325	± 25	346	± 26
	HDP9	232	± 36	413	± 64	283	± 46	355	± 54	387	± 53

594

595 *Table 7: Final ages estimates; CAM: central age model, ADM: average dose model, FMM: finite*  
596 *Mixture Model, LogN\_M: Bayesian calculation with a log-normal model and search of the*  
597 *median, Gauss: Bayesian calculation with a Gaussian model. The shaded ages correspond to*  
598 *those that might be less likely, due to possible mixing. Only one component is indicated for the*  
599 *FMM, either corresponding to the main component (HDP8) or the component that seem more*  
600 *consistent with the chronostratigraphic order (HDP4 and 9).*

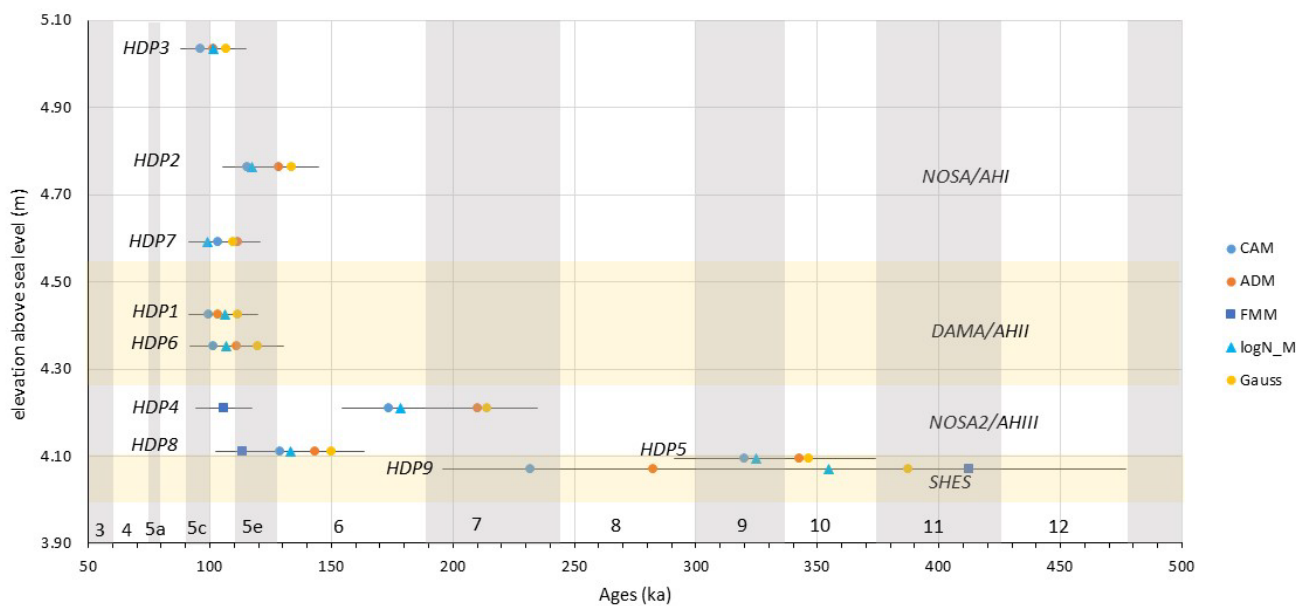
601

602 At last, hypotheses can be raised for HDP8. If a single component is considered, i.e. HDP8  
603 was well bleached and unmixed, all models point to ages consistent with the end of MIS6 /  
604 start of MIS 5e (129±12 ka to 150±13 ka). But if the same hypothesis of mixing between SHES  
605 and NOSAII/AHIII is considered, two components are suggested, the lowest one bearing 80 %  
606 of the grains and leading to an age of 113±11 ka, again consistent with the MIS 5c-5e observed  
607 for the higher samples, including HDP4.

608 A Bayesian model including the stratigraphic constraints was tentatively run, but the  
609 results (not shown) are not informative and propose an imprecise continuous increase of ages  
610 from MIS5c to MIS12.

611

612



613  
614 *Figure 9: summary of all OSL-quartz ages for different age models. The ages are plotted in function of the elevation. Note*  
615 *however that because the samples do not come from the same stratigraphic column and because of the slope of the deposit*  
616 *the stratigraphic order within a stratigraphic unit is not considered meaningful, but for HDP3 at the top. The Marine Isotopic*  
617 *Stages (MIS) are indicated at the bottom.*

618

619 In summary, the data suggest that NOSA2/AHIII to NOSA/AHI were most likely deposited  
620 during MIS5e-5c. While we could discuss at length the most relevant statistical model, with  
621 the “average” based models providing higher estimates than the “median” based models  
622 (Guérin et al., 2017, see also Chevrier et al., 2020 for another working example), the general  
623 conclusion would remain unchanged due to uncertainties related to water content or burial  
624 depth. A mixing, either natural or due to sampling, between NOSA2 and SHES is likely for our  
625 samples. SHES is much older than the upper units, likely consistent with MIS9/MIS10. Older  
626 ages might also be possible if we consider that the top of SHES remained at the surface, with  
627 possible trampling and accidental bleaching for hundreds of years before it was locked by the  
628 subsequent units.

629 The new absolute chronometric ages for HDP1 have further archaeological and  
630 evolutionary implications. Combining the older age estimates (Woodborne, 1999, 2000;  
631 Parkington, 2003) with our results presented here, we can now firmly place the MSA  
632 occupations of HDP1 occurring in the time frame of MIS 5e-c (ca. 130-100 ka) which suggests  
633 both a warm climate and a position of HDP1 close to the Atlantic Ocean. These results are  
634 consistent with our previous hypotheses (Will et al., 2013, 2015) based on other lines of

635 evidence. The faunal assemblages feature frequent angulate tortoise as an indicator of warm  
636 climates and the abundant remains of shellfish (particularly granite limpets) as well as African  
637 penguin and Cape cormorant suggest a close proximity of the site to the sea during deposition  
638 of the archaeological layers.

639 Regional comparisons of lithic artefacts have consistently placed HDP1 within the early  
640 part of MIS 5 (e.g., Will et al., 2013; Mackay et al., 2014; Douze et al., 2015; Schmid et al. 2016)  
641 and new ages for the similar stone artefact assemblages from Ysterfontein 1 at ~120-113 ka  
642 match our results well (Niespolo et al., 2021). Considering the many resemblances in  
643 behavioural traces and the close proximity in space and time between Ysterfontein 1 and  
644 HDP1, one might speculate on the presence of similar populations or even the same group of  
645 early modern humans exploiting the same coastal landscapes in similar ways. Finally, the  
646 geographical position of HDP1 at 15-12 meters a.s.l. means that the site would have escaped  
647 the maximum MIS 5e sea-level high stand. The convergence of several lines of independent  
648 evidence provides a robust age assessment of HDP1 that has been lacking so far.

649 Importantly, the new ages make HDP1 one of the oldest well-dated open-air sites with  
650 good evidence for systematic coastal adaptations. These adaptations encompassed planned  
651 and selective gathering of a narrow range of shellfish with a focus on granite limpets and black  
652 mussels, anticipated long-distance transport of fine-grained raw materials to the coasts,  
653 flexible use of lithic raw materials and the use of ground ochre (Will et al., 2013; Kyriacou et  
654 al., 2016), with many similarities to the close-by site of Ysterfontein 1 with comparable  
655 antiquity (Avery et al., 2008; Klein et al., 2004; Niespolo et al., 2021). The lithic assemblages  
656 at HDP1 are generally small and provide evidence for the import of finished silcrete tools from  
657 inland sources at least 10-30 km away. Transport of high-quality raw materials in the form of  
658 retouched pieces from inland sources, fragmented reduction chains for silcrete, low find  
659 densities and the selected harvesting of small amounts of marine food items indicate a  
660 mobility pattern at HDP1 in which well-provisioned individuals executed planned movements  
661 to the shoreline with the main aim to exploit shellfish during short-term stays, occurring in a  
662 repeated and consistent manner over a 1.5m thick stratigraphic sequence.

663

664 **Conclusion**

665 Combined with the dating results presented here, we can conclude that early modern  
666 humans at HDP1 made consistent use of marine resources and coastal landscapes over many  
667 thousand years between ca. 130-100 ka – with several unexcavated localities on the  
668 Hoedjiespunt Peninsula occurring in similar stratigraphic positions (e.g., HDP2 and HDP3, see  
669 Figure 1) – suggesting successful integration of coastal ecosystems into the regular repertoire  
670 of behavioural adaptations by no later than the beginning of the Late Pleistocene. These  
671 findings fit recent chronometric dating of sites (Niespolo et al., 2021) and overviews on the  
672 increasingly important role of coastal adaptations in the bio-cultural evolution of *Homo*  
673 *sapiens*, with good and persistent evidence for these behaviours over the entire duration of  
674 the Late Pleistocene MSA in both southern and northern Africa (Steele and Alvarez-Fernández,  
675 2011; Campmas et al., 2016; Will et al., 2019).

676

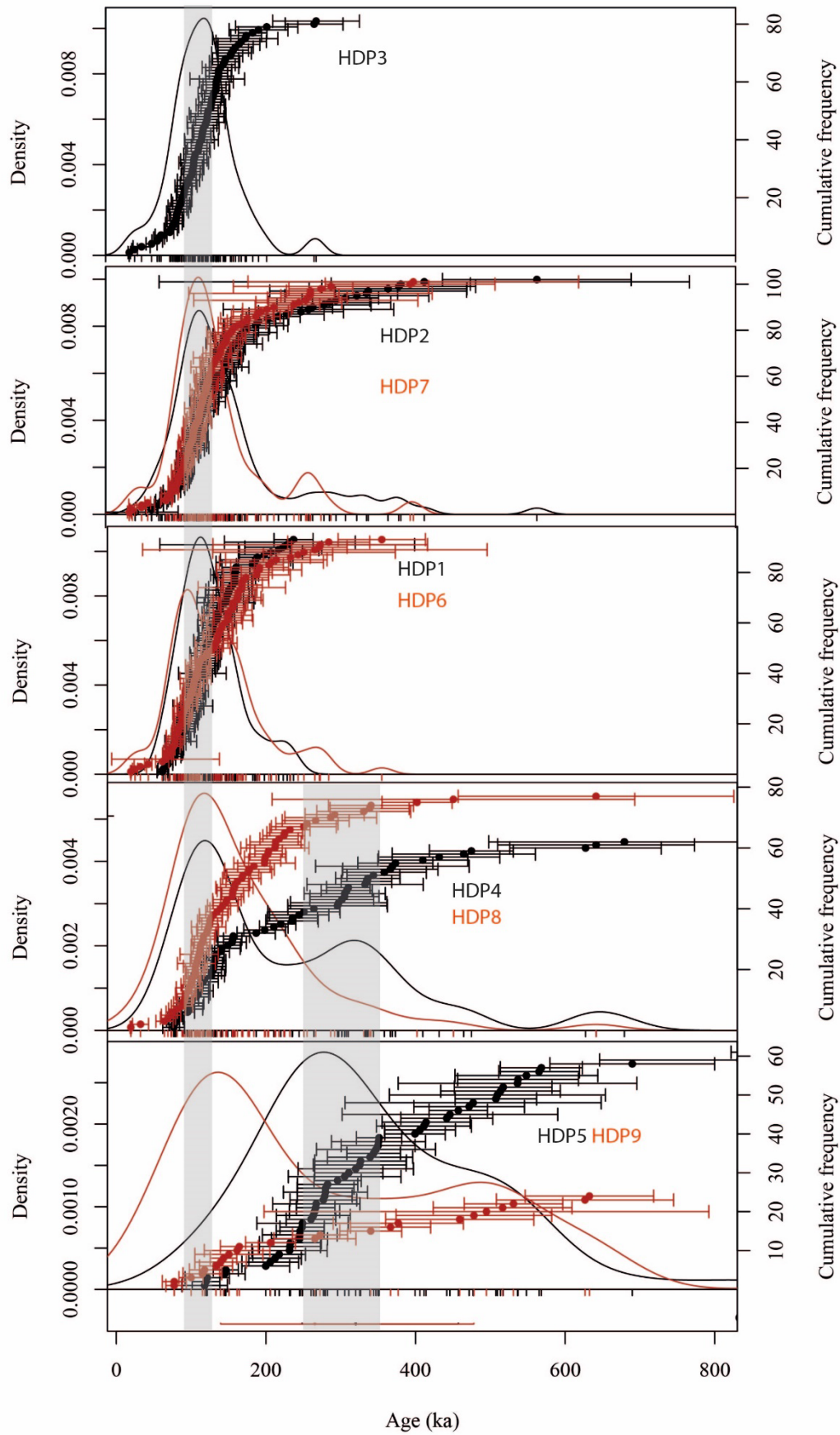
677

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685 to highlight his important and inspiring contributions on coastal archaeology in South Africa.

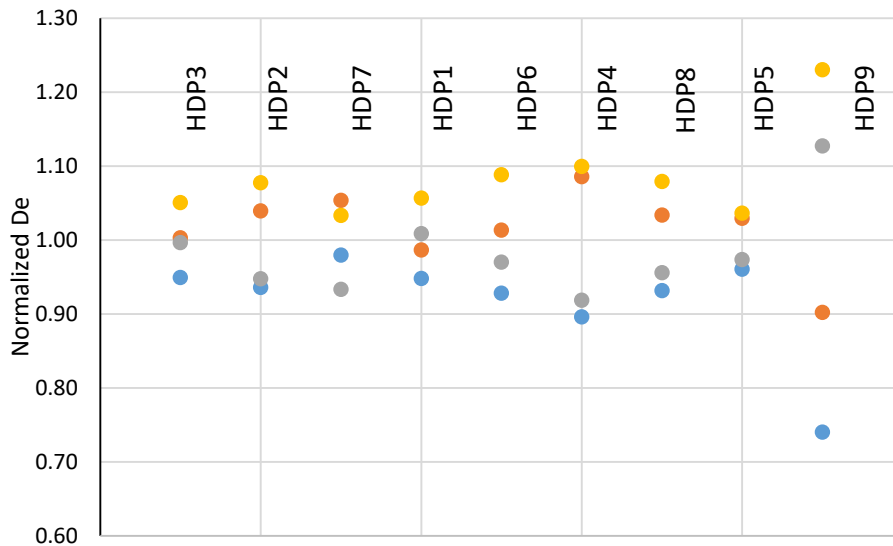
686

687 **Supplementary material:**



689 *Figure Sup A (previous page): Ages for individual grains for all samples and Kernel density plots. Note that all the peaks do*  
 690 *not necessarily have a meaning. The plots are in the stratigraphic order. The grey bars highlight the periods around 110 and*  
 691 *300 ka, corresponding to the two main peaks.*

692



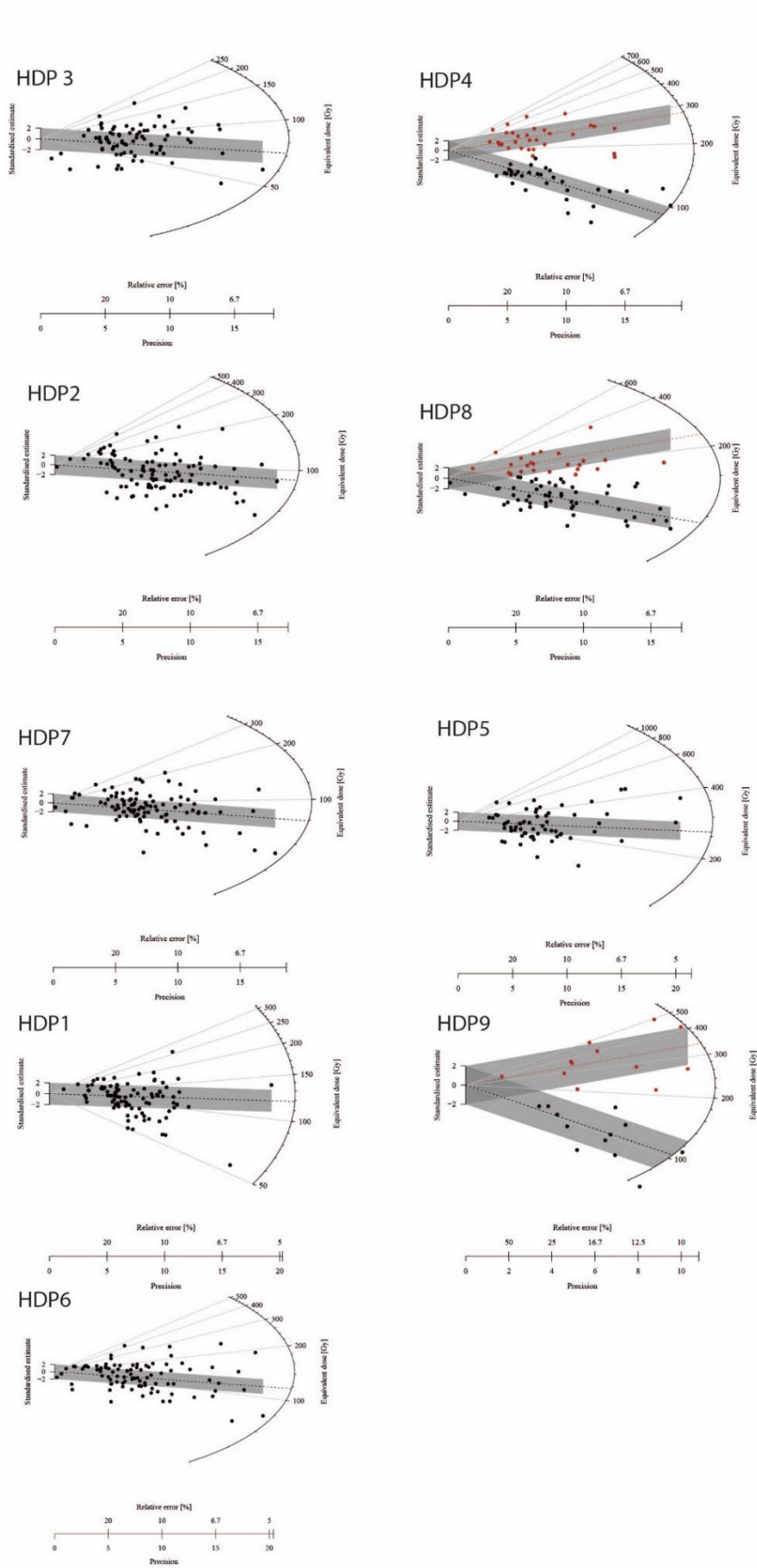
693

694 *Figure Sup B : comparison of the different De estimates, normalized to the mean De for each sample. Blue dots: CAM; Grey*  
 695 *dots: Bayesian-Gaussian; orange dots: ADM; yellow dots: Bayesian-Gaussian. Error bars are not plotted for sake of clarity.*

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701

*Fig Sup C: radial plots for each sample (based on the selected grains). The colored bands corresponds to either the CAM De or to the components of the FMM.*

702

a	mass of carbonate/mass of siliceous material							
sample	> 1cm		1cm - 2mm		2mm - 710 $\mu$ m		<710 $\mu$ m	
HDP3	1950%	$\pm$ 456%	748%	$\pm$ 51%	755%	$\pm$ 54%	558%	$\pm$ 8%
HDP2	620%	$\pm$ 178%	889%	$\pm$ 66%	914%	$\pm$ 93%	612%	$\pm$ 9%
HDP7	1367%	$\pm$ 321%	340%	$\pm$ 7%	863%	$\pm$ 45%	473%	$\pm$ 6%
HDP1	101%	$\pm$ 3%	86%	$\pm$ 2%	82%	$\pm$ 2%	152%	$\pm$ 3%
HDP6	150%	$\pm$ 3%	nd	nd	139%	$\pm$ 3%	97%	$\pm$ 1%
HDP4	1209%	$\pm$ 155%	668%	$\pm$ 34%	619%	$\pm$ 42%	409%	$\pm$ 4%
HDP8	308%	$\pm$ 5%	825%	$\pm$ 37%	640%	$\pm$ 46%	421%	$\pm$ 1%
HDP5	860%	$\pm$ 122%	1100%	$\pm$ 518%	750%	$\pm$ 267%	742%	$\pm$ 7%
HDP9	742%	$\pm$ 34%	767%	$\pm$ 40%	771%	$\pm$ 35%	735%	$\pm$ 3%

703

b	mass of carbonate/mass of siliceous+carbonated material							
sample	> 1cm		1cm - 2mm		2mm - 710 $\mu$ m		<710 $\mu$ m	
HDP3	95%	$\pm$ 22%	88%	$\pm$ 6%	88%	$\pm$ 6%	85%	1%
HDP2	86%	$\pm$ 25%	90%	$\pm$ 7%	90%	$\pm$ 9%	86%	1%
HDP7	93%	$\pm$ 22%	77%	$\pm$ 2%	90%	$\pm$ 5%	83%	1%
HDP1	50%	$\pm$ 1%	46%	$\pm$ 1%	45%	$\pm$ 1%	60%	1%
HDP6	60%	$\pm$ 1%	nd	nd	58%	$\pm$ 1%	49%	1%
HDP4	92%	$\pm$ 12%	87%	$\pm$ 4%	86%	$\pm$ 6%	80%	1%
HDP8	76%	$\pm$ 1%	89%	$\pm$ 4%	86%	$\pm$ 6%	81%	0%
HDP5	90%	$\pm$ 13%	92%	$\pm$ 43%	88%	$\pm$ 31%	88%	1%
HDP9	88%	$\pm$ 4%	88%	$\pm$ 5%	89%	$\pm$ 4%	88%	0%

704

705 *Table Sup A : carbonate contents for the different fractions a) expressed as mass of carbonate over mass of non-carbonated*  
 706 *material b) expressed as mass of carbonate over total mass of sediment.*

707

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