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

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- 15 **Keyword:** Middle Stone Age, luminescence dating, OSL, shell midden, coastal adaptation
- 16

17 Abstract: Coastal adaptations have been considered to play an important role in the bio-cultural 18 evolution of early Homo sapiens and their dispersal out of Africa. In line with this assessment, recent 19 years have seen increasing evidence for the exploitation of seafood from the Middle Stone Age (MSA) 20 of northern and southern Africa. Yet, chronological control constitutes a key problem for better 21 understanding the evolution of these behaviours, with little well-dated evidence for their origins in the 22 late Middle Pleistocene and only few sites dated to before 100 ka. The shellfish-bearing MSA site of 23 Hoedjiespunt 1 (HDP1), located in the Western Cape in South Africa, is one of the localities tentatively 24 dated to the early Late Pleistocene. HDP1 has yielded a 1.5 m stratigraphic sequence with three phases 25 of occupation, each containing abundant lithic artefacts, shellfish, terrestrial fauna, ostrich eggshell 26 and pieces of ground ochre. Analyses of the cultural and zooarchaeological remains of HDP1 have 27 provided evidence for systematic gathering of a narrow range of marine resources coupled with stable 28 adaptations to coastal landscapes. Here we present the first complete absolute chronology for the site 29 and its entire stratigraphic sequence by applying luminescence dating (OSL) to quartz grains extracted 30 from the carbonate-rich sediments. The results show repeated human occupations during Marine 31 Isotope Stages 5e-5c (~130-100 ka) at HDP1, suggesting successful integration of coastal ecosystems 32 into the regular repertoire of behavioural adaptations by early modern humans by no later than the 33 beginning of the Late Pleistocene.

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35

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; Cunnane and Stewart, 2010; Marean, 2014; Will et al., 2019). Here we employ the broad evolutionary definition of coastal adaptations that we have proposed previously (Will et al., 2016; 2019) including various behaviors such as the regular consumption and systematic acquisition of marine food resources and the expansion of settlement systems to include coastal and near-coastal zones as occupation spots on a regular and planned basis (for a 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 al., 2007; Jerardino and Marean 2010; Ramos-Muñoz et al., 2016) and whether Homo sapiens 46 47 alone or also other species such as Neanderthals display these adaptations (Klein and Steele, 2008; Stringer et al., 2008; Cortés-Sánchez et al., 2011; Marean, 2014; Will et al., 2019; Zilhao 48 49 et al., 2020). Further research topics include the consequences of marine food consumption for human brain evolution and demography (Crawford et al., 1999; Parkington, 2001; 50 51 Broadhurst et al., 2002; Parkington, 2003, 2010; Marean, 2011; Kyriacou et al., 2014; Marean, 2016; Will et al., 2015) as well as the potential of coastal landscapes to facilitate their 52 dispersals such as for the exit of *Homo sapiens* from Africa (Stringer, 2000; Walter et al., 2000; 53 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.

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

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

88 Here is where the open-air site of Hoedjiespunt 1 (HDP1), located in the Western Cape of South Africa, comes into play. From a historical perspective, the site has already been 89 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 of the site (Berger and Parkington, 1995). These campaigns were followed in 2011 by fieldwork 94 95 of a joint team from the University of Tübingen and the University of Cape Town directed by Nicholas Conard, exclusively in the archaeological deposits. The renewed excavations 96 uncovered a short stratigraphic sequence of archaeological layers with in situ evidence for 97 MSA stone artefacts associated with shellfish and other faunal remains (Will et al., 2013, 2015; 98 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 approximately 9-6 meters higher than at present in southern Africa and globally (Miller et al., 101 102 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 (Butzer, 2004: 1777) reiterating this placement within early MIS 4. Absolute age 105 determinations based on OSL/IRSL (Optically stimulated luminescence/Infrared stimulated 106 luminescence) readings of both quartz and feldspars grains from sediments, and U-series 107 dates of ostrich eggshell from the Parkington excavations indicated a tentative age range 108 between 130 and 100 ka for one of the archaeological horizons (AH II; Woodborne, 1999, 109 2000; Parkington, 2003; pers. com. from anonymous reviewer) but this was never fully 110 confirmed and published. Based on the previous dating efforts, and in concert with 111 stratigraphical, artefactual, faunal and contextual lines of evidence from the Conard 112 excavations, the most recent assessment by Will et al. (2013; 2015) suggested an age of MIS 113 5e for much of the sequence. This would make HDP1 one of the oldest open-air site with good 114 115 evidence for coastal adaptations in sub-Saharan Africa and provide a potential age link with the isolated MIS 6 evidence from Pinnacle Point 13B. In order to test this hypothesis and to 116 base the absolute chronology of the site on secure evidence, however, modern dating by 117 118 absolute chronometric methods is required. To this end, new sediment samples were taken from the section of the Conard excavations. In this paper, we present the ages obtained with 119 120 the optically stimulated luminescence (OSL) dating method applied to quartz grains coming from nine sediment samples and discuss the further implications of these results. 121

122

123 2) Site and samples

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

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

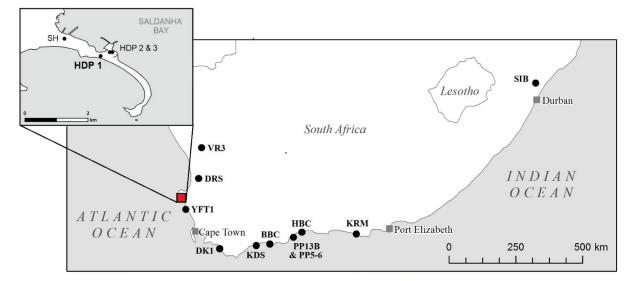




Figure 1: Location of Hoedjiespunt 1 (HDP1) in relation to other MSA sites in southern Africa (top);
picture taking during the excavation of the open-air sequence of HDP1 in 2011, showing its currentday proximity to the Atlantic Ocean (bottom). Abbreviations for archaeological sites: BBC-Blombos
Cave;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

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143 Combining both the Conard and Parkington field work, the archaeological deposits have 144 been excavated over an area of 18 m², with a volume of 1.4 m³ 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 consists of three consecutive archaeological horizons (AH I-III), each containing in situ MSA 148 lithic artefacts, ochre, shellfish, terrestrial fauna and abundant fragmented ostrich eggshells 149 (Fig. 2). The strata are in primary context, slope slightly downwards to the south and lie either 150 directly under a thick calcrete carapace or the modern surface (HUMUS). The strata, although 151 intact, have been truncated by erosion in the downslope direction toward the beach. A 152 correlation between excavated units of the Parkington and Conard excavations is possible 153 154 though not one to one (e.g., AHI consists of facies CESA, SHEM and NOSA in the old 155 excavations). Here we will use both stratigraphic designations to indicate the exact location of the dating samples (Table 1). 156

157 AH I and III are light-colored strata composed of consolidated fine sand containing many marine shells in both complete and fragmented status. Micromorphological analysis of the 158 159 deposits (Göden, 2014) show that AH I and III are composed largely (ca 85%) of rounded, sandsized, calcareous bioclasts with a more minor component of ooids, peloids and glauconite 160 grains. Sand-sized grains of quartz are also present, at around 10-15%. Coarser components 161 162 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 III (as in all other strata studied at the site) and they exhibit infillings of needle-fibre calcite 164 and well as the local formation of calcite hypocoatings. Anthropogenic components in AH I 165 and III are minor, but consist of fragments of charred organic matter, burnt and unburnt bone, 166 167 and larger shell fragments. Sandwiched between AH I and III, AH II averages 20 cm and consists of compact, dark brown clay and organic-rich sediments and yielded many, mostly broken, 168 169 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 (>710µm) sedimentary material. Find densities are highest for AH II concerning lithics and 172 173 faunal remains, yet with all AHs showing comparable densities of shellfish remains (11-13 kg/m³; Will et al., 2015: Table 3). Thin sections analysed from AH II show that this strata, like 174 175 AH I and III, consists largely of sand-sized, calcareous bioclasts. However, AH II is significantly enriched in finely comminuted organic matter, both carbonized and humified, as well as 176 177 anthropogenic components such as burnt and unburnt bone, large shell fragments, and charcoal. The groundmass appears locally phosphatized. Rootlet channels are more numerous in AH II relative to AH I or III and have more pronounced needle-fibre calcite infillings. Below the oldest archaeological horizon, AH III, lies a 2 m thick, sterile, calcareous, shelly sand (SHES) named for its abundant land snails (*Trigonephrus*). SHES rests on top of the paleontological layers of the site. Micromorphological analysis of SHES shows that it is compositionally similar to AH I and III but lacks any significant anthropogenic components.

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

192

OSL sample	elevation (mm)	unit (Parkington)	unit (Con aes)
HDP3	5 034		194
HDP2	4 763	NOSA	^{AHI} 195
HDP7	4 593		155
HDP1	4 425		196
HDP6	4 354	DAMA	AHII 197
HDP4	4 210	NOSA2	AHIII198
HDP8	4 112	NUSAZ	
HDP5	4 095	CLIFC	199 SUIS
HDP9	4 085	SHES	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.

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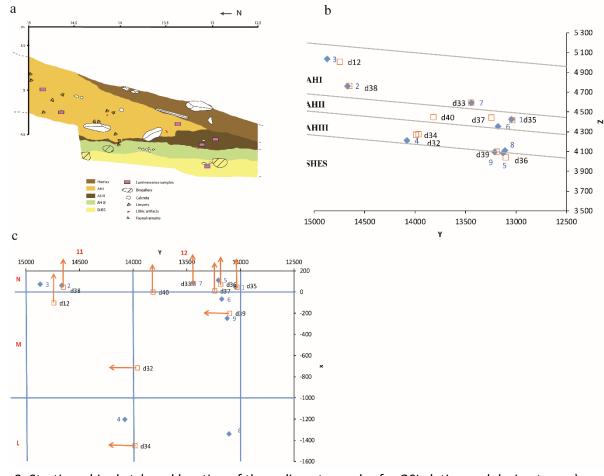


Figure 2: Stratigraphic sketch and location of the sediment samples for OSL dating and dosimeters. a) North-South profile. b) Projection on the profile of the samples (blue diamonds) and dosimeters (red squares); the approximate limits between the units are drawn (grey lines). c) Plan view, with the location of the OSL samples (blue diamonds) and dosimeters; the red squares show the edge of the dosimeter tubes that were apparent while the dosimeters were inserted into the section and were recorded with the total station; the orange arrows symbolise the tube, with the Al₂O3:C chips located at the tip.

214

215 3) Equivalent dose determination

216 <u>3.1 Preparation</u>

The samples have been sieved to 710 μ m, dried and weighted. The > 710 μ m fraction 217 and part of the $< 710 \mu m$ were saved for gamma spectrometry analyses (section 4). The rest 218 of the < 710 µm fraction was further treated in order to extract the quartz grains: wet sieving 219 at 100 and 140 µm was followed by HCl (10%), H₂O₂ (30%), heavy liquid separation 220 (heteropolytungstate of sodium at 2.72 and 2.58 g/cm³). The quartz grains were then HF (40%) 221 222 etched for 60 min followed by HCI (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,

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

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

230 <u>3.2 Measurement devices</u>

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

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

238 <u>3.3 Protocol</u>

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).

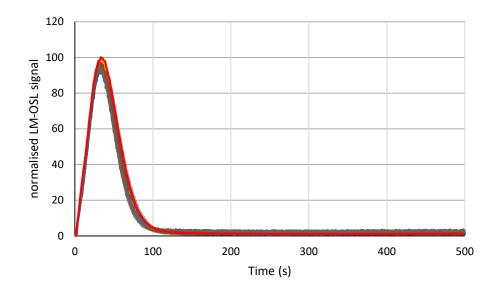


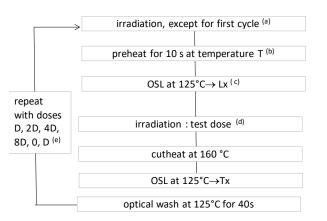
Figure 3: LM-OSL test for all the samples. One multi-grain aliquot of each sample was optically

stimulated at 125°C while increasing linearly the stimulation power from 0 to 70 mW/cm² in 500 s. The

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.

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Figure 4: SAR protocol applied for the equivalent dose determination. a) the first cycle corresponds to the measurement of the 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 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
Gy or 33 Gy; e) D was about one third of the expected De.

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A minimal requirement for SAR is that it allows to recover a known laboratory given dose. A dose recovery test (DRT) was thus performed on single grains aliquots, where the samples were first bleached for one minute in a solar simulator (Honle UVAcube 400), left in 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, was given and the protocol was applied. Analysis of single-grain data was performed with 260 Analyst v4.57 (Duller, 2015). Grains were selected based on the sensitivity of each grain (test 261 dose of the "natural" signal > 3 times background signal; relative error on "natural" test dose 262 signal < 10%), and on the recuperation signal (normalised signal for the zero Gy dose < 5% of 263 the normalised signal of the highest regeneration point). The recycling ratio was not used as 264 a rejection criteria, because one of the luminescence reader is equipped with an 265 266 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 and thus highlights the grains that have moved significantly between the two recycling cycles 268 and for which the calculation of a recycling ratio is therefore unrelevant. However, considering 269 270 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).

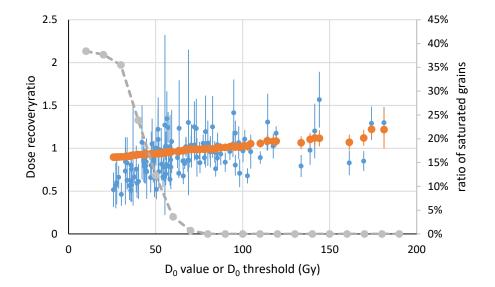
276 The dose response curve was fitted with a saturated exponential such as:

 $\frac{Lx}{Tx} = I_0 \left[1 - exp \left(-\frac{(D+b)}{D_0} \right) \right]$, where D is the regenerative dose, Lx/Tx is the normalised 277 luminescence signal and I₀, b and D₀ are fitting parameters. In addition to the grains discarded 278 because of the rejection criteria mentioned above, numerous grains (see Table 3 and 4) were 279 280 also rejected when no intercept of the normalised natural signal (Ln/Tn) onto the growth curve was possible, i.e., the natural signal is at or above the saturation limit I₀. Beside these usual 281 282 selection criteria, and following Thomsen et al., (2016), the D₀ value (saturation parameter) was also used as an additional criterion: in practice, the equivalent doses are ranked in 283 284 function of the corresponding D₀ values and the central doses (after Galbraith et al., 1999) are calculated while progressively discarding the grains with the lowest D₀ values. This leads to 285 286 increasing central dose values as a function of the D₀ value until a plateau is reached. In addition, the proportion p of grains rejected because of the saturation parameter (within the 287 population of grain that pass the first set of rejection criteria) was calculated in function of D₀. 288 289 The lowest D_0 for accepted grains was thus within the central De plateau, where less than 5%

of grains were rejected because of saturation. Figure 5 displays this analysis for sample HDP3:

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.



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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 pass the selection criteria and is not saturated. Orange dots: the dose recovery ratio calculated after the CAM in function of the minimum D_0 value considered in the distribution. Note that there are two additional points with much larger D_0 values

that have not been plotted here for the clarity of the graphic. Grey dots: percentage of saturated grains over the total number of grains passing the selection criteria, above a D_0 threshold value. The final dose recovery ratio is considered in the plateau region (here above 70 Gy), where less than 5% of the grains show saturation.

sample	tested dose (Gy)	N	n1	n2	n	D₀ (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

Table 3: Dose recovery ratios. N: number of tested grains. n_1 number of grains passing the selection criteria. n_2 : number of grains passing the selection criteria with unsaturated luminescence signals. n:

number of grains also passing the D_0 criteria and present in the final De distribution. D_0 : threshold value

304 of D₀, 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.

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

315 <u>3.4 results</u>

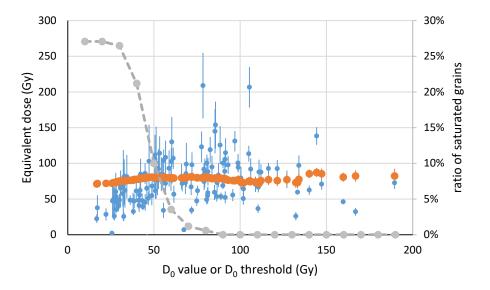
316 The equivalent doses were then measured with the same readers and protocols as for 317 the dose recovery tests. Between 2300 and 8900 grains were analysed for each sample (Table 4). Only 3 to 8% pass the first set of selection criteria and are not saturated. After the D₀ test, 318 1 to 3% of the measured grains remain in the final De distribution, down to 0.3% for HDP9. It 319 320 is worth noting that in every De 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 could be the strong wind occurring all the time at that place, which could have polluted the 325 326 samples during their extraction on the site. The D₀ threshold allows discarding most of these grains however and the grains with negative equivalent dose have been discard before the 327 328 calculations of the mean/central dose. The few remaining grains have not been discarded, because they are associated with large uncertainties and their weight in the final distribution 329 is negligible, and we wanted to avoid subjective selection. Table 4 displays the equivalent 330 331 doses, based on the CAM, together with the over-dispersion values. However, the relevance of the CAM here is disputable and further discussed in section 5. 332

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											Equivalent doses (G	y)									
Unit	sample	Ν	n1	n2	n	n	n	n	n	n	n	n	D _{0 (Gy)}	CAM		ADM		FMM (CA	M based)	logN_M	Gauss
							De	OD (%)	De	OD (%)	1	2									
	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							
NOSA/AHI	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							
DAIVIA/ AHII	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							
NUSAZ/AHIII	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							
61156	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							
SHES	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, n₁, n₂, n, D₀, 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

340 calculation with a Gaussian model



341

Figure 6: Equivalent dose (De) distribution for sample HDP3. Blue dots: De in function of D_0 value for each grain that pass the selection criteria and is not saturated. Orange dots: De calculated after the CAM in function of the minimum D_0 value considered in the distribution. Note that there are additional points with much larger D_0 values that have not been plotted here for the clarity of the graphic. Grey dots: ratio of the number of saturated grains above a D_0 threshold value, over the total number of grains passing the selection criteria. The final De is considered in the plateau region, where less than 5% of the grains show saturation.

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

353

4) Dose rate

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

4.1 cosmic and gamma dose rates

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

The gamma dose rates were calculated from in situ dosimetry: 10 tubes, each containing 365 3 Al₂O₃:C pellets, have been inserted 30 cm deep into the stratigraphic sections as close as 366 367 possible from the sediment samples (Fig. 2), and left for about one year (Bøtter-Jensen et al., 1997; Kreutzer et al., 2018). The recorded doses, corrected for the cosmic contribution, show 368 very little scatter (10% RSD on the gamma dose for the 10 tubes, 6% for the total dose over 369 the 30 chips) (Table Sup A), though they are slightly higher for the organic rich DAMA/AHII 370 layer. Each sample was attributed the gamma dose deduced from the closest dosimeter, but 371 372 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): the organic-rich unit (DAMA/AHII) displays water content around 30% (mass of water/mass of 378 379 dry sediment), while the shelly-sandy units are nearly dry (1% for SHES). In order to at least take these different power of water retention into account, no correction was applied to the 380 gamma dose rates recorded by the dosimeters (since they already take into account the 381 382 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 drastically different. The gamma doses stand between 0.30±0.04 Gy/ka and 0.41±0.04 Gy/ka. 384 They contribute for 31 to 39% of the total dose rates. 385

386 <u>4.2 beta dose rates: introduction</u>

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

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

389 equivalent to

390

- 391
- 392
- 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; δ : specific beta dose rate for either 398 U or Th or K.

 $D_{\rm s+w} = \frac{a\delta A_{\rm s}}{1 + k\left(\frac{m_{\rm w}}{m}\right)}$

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 µm fraction; see Table 5 and Table sup A), due to the presence of bioclastic sands, shell and 401 calcrete fragments of all sizes. This raises three distinct questions: 1) Are there significant 402 403 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 siliceous matrix (Nathan and Mauz, 2008; Guérin et al., 2012)? 2) Is the model of infinite matrix 405 406 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 imply for the age (actually dose) model (Mayya et al., 2006; Guérin et al., 2017)? Questions 1 408 and 2 are discussed successively in this section, question 3 will be discussed in section 5. 409

											do	se rate	s (Gy/ka	a)					
unit	sample	grain-size	water	% fine/tot	%carbonate	cos	smic	gar	nma	be	ta1	be	ta 2	t	ot1	1	tot2	me	ean
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

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Table 5: data for dose rates. Water: mass of water over mass of dry sediment. %fine/tot: mass of dry <710μm fraction over
 the total mass of dry sample. %carbonate: mass of carbonate over the mass of dry non- carbonated sediment. Cosmic dose

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

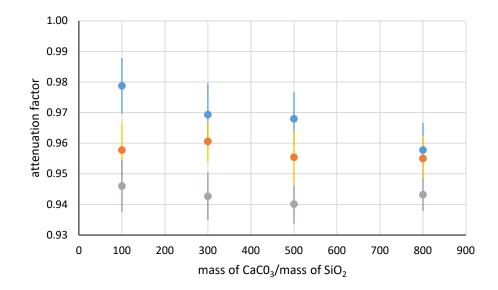
418

419 <u>4.3 determination of appropriate attenuation factors and water content correction</u> 420 <u>factors.</u>

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

The modelled matrix was composed of SiO₂ and CaCO₃, with a mass ratio (CaCO₃ / SiO₂) varying from 100% to 800%. The dose received by 120 μm quartz grains was computed in assuming that U, Th and K are uniformly distributed within the carbonate rich matrix. Note that this model differs from the one of Nathan and Mauz (2008), where the carbonate is supposed to fill pores within the sedimentary matrix and be free of any radioactive material. Moreover, these authors only considered CaCO₃ contents up to 100% in mass (mass of carbonates over mass of siliceous material).

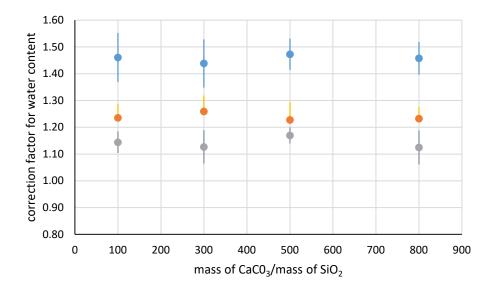
Attenuation factors *a* deduced from these calculations are shown on Figure 7. They are poorly 431 432 dependent on the proportion of carbonates. Note, however, that they are significantly different from the ones proposed by Guérin et al., (2012) for a siliceous matrix, but for the 433 potassium (0.96 in both cases): the attenuation factors are here 0.96±0.01 for U, instead of 434 0.90, and 0.94±0.01 for Th instead of 0.85. These differences are due to the differences of beta 435 absorption between the quartz grains and the matrices and to the method of calculation of 436 the attenuation factor. Guérin et al., (2012) simulated the beta self-doses of SiO₂ grains in a 437 SiO₂ matrix, and calculated the attenuation factors as the complementary of the self-dose. 438 This calculation is possible when the grain and the matrix have the same beta absorption 439 440 efficiencies. In our case the beta rays coming from the $CaCO_3$ / SiO_2 mixed matrix were simulated and the dose was directly evaluated in the SiO₂ grains. Doing this way is important 441 since the quartz self-dose is affected by the mixed matrix because the beta backscattering is 442 different than in a pure SiO₂ matrix. Similarly, the attenuation factor is affected because the 443 444 beta absorption efficiency of a CaCO₃ / SiO₂ mixed matrix is significantly different from the one 445 of a pure SiO_2 matrix.



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±0.04, 1.24±0.04 and 1.14±0.06 were applied respectively for K, U and Th beta dose rates. While they significantly differ from the 452 mean factor of 1.25 calculated by Zimmerman, (1971), but for U, we could observe that the 453 final beta dose rates calculated with both set of correcting factors do not differ by more than 454 455 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 values. As for the calculation of the gamma dose rates, the current water contents were 458 applied for the calculation of the final beta dose rates. 459

460



463 Figure 8: correction factor k for water content for beta dose from U series (orange dots), Th series (grey dots) and K (blue464 dots).

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466 <u>4.4 dealing with heterogeneities.</u>

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468 Here we deal with the fact that the presence of coarser materials with different matrix and radioactive contents than the fine (here <710µm) fraction where the quartz grains-469 470 dosimeters come from, can induce heterogeneities in the beta dose rate (e.g. Martin 2015, Martin et al., 2015b; Tribolo et al., 2017). In particular, Cunningham (2016) warned against 471 the use of the infinite matrix assumption in shell rich sediments, since the shells introduce 472 "cold spots". However we argue here that the heterogeneity of the sediment samples from 473 474 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 high resolution gamma spectrometry (Canberra, e.g. Guibert et al., 2009). Table 6 displays the 481 482 activities and radioactive contents results.

						s (Bq/kg)				conte	nt (%)
	Sample HDP			²³⁸ U :	series			232-1		K	
		t	ор	mic		bot	tom	in in	series	К	
	3	6.0) ± 1.2	11.8	±0.4	13.5	± 2.1	7.0	± 0.2	0.24	± 0.0
	2	8.2	2 ± 1.1	9.4	± 0.3	5.9	± 2.0	5.4	± 0.2	0.18	± 0.0
	7	8.9	± 0.6	9.6	± 0.2	10.6	± 1.0	6.3	± 0.1	0.27	± 0.0
fraction	1	24.8	3 ± 1.5	15.8	± 0.4	12.7	± 2.4	15.9	± 0.3	0.62	± 0.
< 710µm	6	21.9) ± 1.2	16.3	± 0.3	12.3	± 1.8	18.6	± 0.3	0.77	± 0.
< /10µ111	4	10.4	± 1.0	13.4	± 0.3	10.3	± 1.8	6.6	± 0.2	0.26	± 0.0
	8	11.0) ± 0.7	14.0	± 0.2	9.5	± 1.2	7.0	± 0.2	0.29	± 0.0
	5	9.2	2 ± 1.0	10.4	± 0.3	8.9	± 1.8	5.3	± 0.2	0.23	± 0.0
	9	8.0) ± 0.7	9.5	± 0.2	8.4	± 1.1	4.7	± 0.1	0.23	± 0.0
	3	9.6	5 ± 1.1	11.9	± 0.3	8.9	± 1.9	5.9	± 0.2	0.17	± 0.0
	2	10.5	5 ± 1.0	9.3	± 0.3	10.2	± 2.2	4.8	± 0.3	0.12	± 0.0
	7	8.1	± 0.5	7.9	± 0.1	8.4	± 0.9	4.7	± 0.1	0.19	± 0.0
fraction	1	27.4	± 0.9	17.0	± 0.2	13.1	± 1.3	14.4	± 0.2	0.55	± 0.
> 710µm	6	21.4	± 0.7	14.5	± 0.2	10.8	± 1.1	. 14.8	± 0.2	0.62	± 0.
>710µm	4	9.7	7 ± 1.4	14.8	± 0.4	11.1	± 2.2	4.9	± 0.2	0.19	± 0.
	8	25.0) ± 1.4	25.7	± 0.4	12.9	± 2.2	6.4	± 0.2	0.39	± 0.0
	5	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
	9	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
	3	6.9	9 ± 0.9	11.8	± 0.3	12.3	± 1.6	6.7	± 0.2	0.22	± 0.
	2	9.2				-	-	-	± 0.2	0.15	± 0.
	7	8.5		8.8	± 0.1	9.5	± 0.7	5.5	± 0.1	0.23	± 0.
	1	26.7	7 ± 0.8	16.7	± 0.2	13.0	± 1.2	14.8	± 0.2	0.57	± 0.
total	6	21.6	6 ± 0.7	15.3	± 0.2	11.5	± 1.0	16.4	± 0.2	0.68	± 0.
	4	10.3	3 ± 0.9	13.6	± 0.3	10.4	± 1.6	6.4	± 0.2	0.25	± 0.
-	8	12.7	_		± 0.2				_	. 0.31	± 0.
	5	9.2	_	10.4	± 0.3	8.9	± 1.8	5.3	± 0.2	0.23	± 0.
	9	8.0) ± 0.7	9.5	± 0.2	8.4	± 1.1	. 4.7	± 0.1	. 0.23	± 0.0

⁴⁸³

485 Table 6: activities for ²³⁸U series or ²³²Th series and K contents deduced from high resolution gamma spectrometry. The 486 samples were divided into two distinct fractions: <710µm and >710µm. For the ²³⁸U series: the top activities (pre-Rn) are 487 deduced mainly from the ²³⁴Th peak, the middle activities (post Rn) are deduced from ²¹⁴Bi and ²¹⁴ Pb, the bottom activities 488 are related to ²¹⁰Pb. 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

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

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

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4.5 beta dose rates: final comments and results

Note (Table 6) that the U series show disequilibrium for most samples: for all but HDP3 505 and 7, the activity for the middle of the chain (²²⁶Ra based on ²¹⁴Bi and ²¹⁴Pb) is in excess (13 506 to 59%) compared to the bottom (based on ²¹⁰Pb). The middle of the chain is in excess as well 507 compared to the top (based on ²³⁴Th) for all samples (up to 29% for most, 97% for HDP3) but 508 509 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 processes (e.g. radon escape, leaching, etc.) could be involved. However, since U contributes 511 512 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

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

523

524 5. Ages and discussion

The equivalent dose distributions are widespread, with overdispersions (OD) values standing between 32±3 and 48±4 %, up to 64±6% and 65±10% for HDP4 and HDP9 (Table 4). While a part of this OD might be due to the low presence of modern age grains (thought this is not the case for HDP1 and 2 which have OD similar to those of the other samples), it is likely that a large part of this overdispersion can be attributed to the low K (but also U and Th) 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). These authors have shown on simplified models that, in such a case, the dose rate distribution 533 is positively skewed, and they have questioned the relevance of the central equivalent dose 534 statistical model, such as defined in the Central Age Model (Galbraith, 1999) for calculating 535 the ages (Guérin et al., 2015, 2017; Mayya et al., 2006). They have shown that the Central Age 536 Model calculates the geometric mean of the De distribution, i.e. the median of a lognormal 537 distribution, while the mean dose rate corresponds to an average (arithmetic mean), 538 539 whatever the Dr distribution. Therefore, in case the Dr and the De distributions are highly skewed, the median and average De will not match and the CAM will lead to age 540 underestimate. 541

Mayya et al., (2006) and more recently Chauhan et al., (2021) have suggested using a 542 minimum equivalent dose, based on the equivalent dose distribution, together with a 543 minimum dose rate (when the beta K contribution tends to zero; see also, David (2007) and 544 Feathers et al., (2020), for two other similar examples). However, their calculation rests on a 545 546 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, making this model difficult to apply. In addition, it would require a recalculation of the 548 549 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 of a multiple size fractions in the HDP sediments. 551

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 sigma, though the models based on the calculation of the average (ADM and BayesianGaussian) tend to give higher De than the models based on the calculation of the median (CAM
and Bayesian-logNormal median), as expected for positively skewed distributions.

For HDP3, 2, 7, 1 and 6, all models point to ages covering stages 5e to 5c (Fig. 9 and Table 7). HDP2 appears to be in chrono-stratigraphic reversal. Compared to HDP7, 1 and 6 below, more grains for this sample have apparent ages over 300 ka (Fig.Sup A), despite a lower mean dose rate. Whether this is due to microdosimetric effects or to another reason is not clear for 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 measured ones. However, the OD of HDP 4 and 9 are higher and the visual inspections of the 572 573 De 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 sigma b (OD of each component) varying between 0.2 and 0.4, according to the OD observed 575 for the other samples. In all cases, the BIC and Maximum Likelihood criteria point to two 576 577 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 De close to 100 Gy. The higher components, though imprecise, are both consistent with 300 Gy. HDP4 and HDP9 are close to the stratigraphic junction between SHES and 581 NOSAII/AHIII. The De distributions suggest that there was either a natural (bioturbation) 582 583 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. However here the total dose rate of the SHES and NOSA2/AHIII samples are close enough to 587 588 allow this calculation. Consequently, the age for HDP4 would be 106±12 ka, consistent with the marine isotopic stage 5c-5d observed for the upper samples (Table 7). The age for HDP9 589 590 would be 413±64ka, rather imprecise again due to the low number of selected grains, but not 591 inconsistent with the ages obtained for HDP5 (320±28 to 346±26 ka depending on the statistic model). 592

unit	cample			·	Ages (k	a)					
unit	sample		CAM	FM	Μ	AD	M	LogN_	М	Ga	iuss
	HDP3	96	± 8			101	± 8	101	± 8	107	± 8
NOSA	HDP2	116	± 10			128	± 12	117	± 10	134	± 11
	HDP7	104	± 9			111	± 9	99	± 8	110	± 9
	HDP1	99	± 8			104	± 9	106	± 8	111	± 8
DAMA	HDP6	102	± 10			111	± 11	107	± 10	120	± 11
NOCAD	HDP4	174	± 19	106	± 12	210	± 24	178	± 18	214	± 20
NOSA2	HDP8	129	± 12	113	± 11	143	± 15	133	± 12	150	± 13
CUEC	HDP5	320	± 28			343	± 31	325	± 25	346	± 26
SHES	HDP9	232	± 36	413	± 64	283	± 46	355	± 54	387	± 53

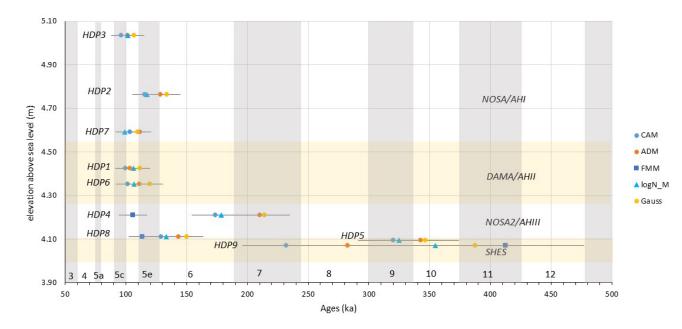
Table 7: Final ages estimates; CAM: central age model, ADM: average dose model, FMM: finite
Mixture Model, LogN_M: Bayesian calculation with a log-normal model and search of the
median, Gauss: Bayesian calculation with a Gaussian model. The shaded ages correspond to
those that might be less likely, due to possible mixing. Only one component is indicated for the
FMM, either corresponding to the main component (HDP8) or the component that seem more
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



613 614

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

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

The new absolute chronometric ages for HDP1 have further archaeological and evolutionary implications. Combining the older age estimates (Woodborne, 1999, 2000; Parkington, 2003) with our results presented here, we can now firmly place the MSA occupations of HDP1 occurring in the time frame of MIS 5e-c (ca. 130-100 ka) which suggests both a warm climate and a position of HDP1 close to the Atlantic Ocean. These results are consistent with our previous hypotheses (Will et al., 2013, 2015) based on other lines of evidence. The faunal assemblages feature frequent angulate tortoise as an indicator of warm
climates and the abundant remains of shellfish (particularly granite limpets) as well as African
penguin and Cape cormorant suggest a close proximity of the site to the sea during deposition
of the archaeological layers.

639 Regional comparisons of lithic artefacts have consistently placed HDP1 within the early part of MIS 5 (e.g., Will et al., 2013; Mackay et al., 2014; Douze et al., 2015; Schmid et al. 2016) 640 641 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 HDP1, one might speculate on the presence of similar populations or even the same group of 644 645 early modern humans exploiting the same coastal landscapes in similar ways. Finally, the geographical position of HDP1 at 15-12 meters a.s.l. means that the site would have escaped 646 647 the maximum MIS 5e sea-level high stand. The convergence of several lines of independent evidence provides a robust age assessment of HDP1 that has been lacking so far. 648

Importantly, the new ages make HDP1 one of the oldest well-dated open-air sites with 649 650 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, flexible use of lithic raw materials and the use of ground ochre (Will et al., 2013; Kyriacou et 653 al., 2016), with many similarities to the close-by site of Ysterfontein 1 with comparable 654 antiquity (Avery et al., 2008; Klein et al., 2004; Niespolo et al., 2021). The lithic assemblages 655 656 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 to the shoreline with the main aim to exploit shellfish during short-term stays, occurring in a 661 662 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 humans at HDP1 made consistent use of marine resources and coastal landscapes over many 666 thousand years between ca. 130-100 ka - with several unexcavated localities on the 667 Hoedjiespunt Peninsula occurring in similar stratigraphic positions (e.g., HDP2 and HDP3, see 668 Figure 1) – suggesting successful integration of coastal ecosystems into the regular repertoire 669 of behavioural adaptations by no later than the beginning of the Late Pleistocene. These 670 findings fit recent chronometric dating of sites (Niespolo et al., 2021) and overviews on the 671 672 increasingly important role of coastal adaptations in the bio-cultural evolution of Homo sapiens, with good and persistent evidence for these behaviours over the entire duration of 673 the Late Pleistocene MSA in both southern and northern Africa (Steele and Alvarez-Fernández, 674 2011; Campmas et al., 2016; Will et al., 2019). 675

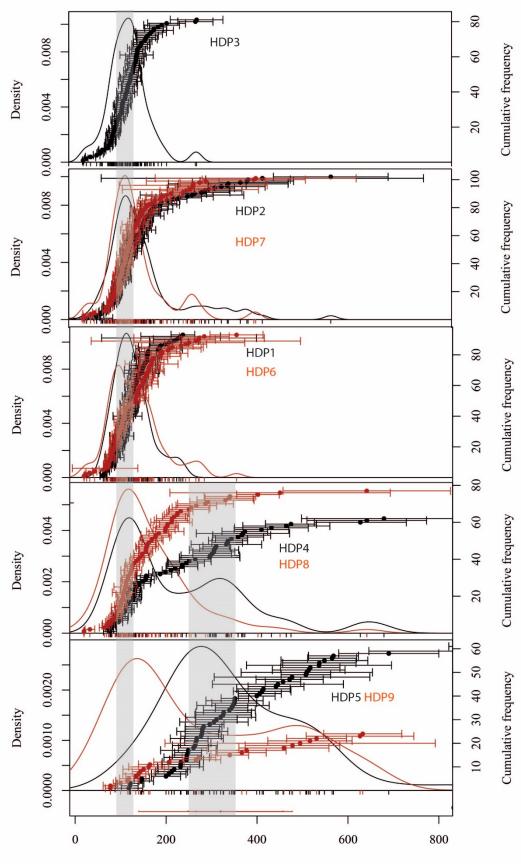
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- 677

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686

687 Supplementary material:



Age (ka)

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.

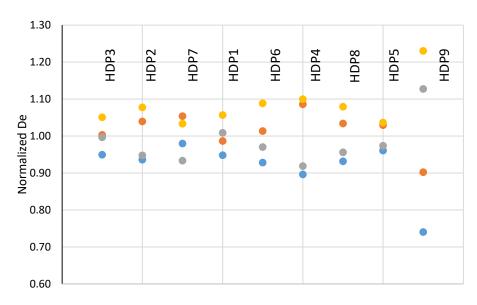


Figure Sup B : comparison of the different De estimates, normalized to the mean De for each sample. Blue dots: CAM; Grey
 dots: Bayesian-Gaussian; orange dots: ADM; yellow dots: Bayesian-Gaussian. Error bars are not plotted for sake of clarity.

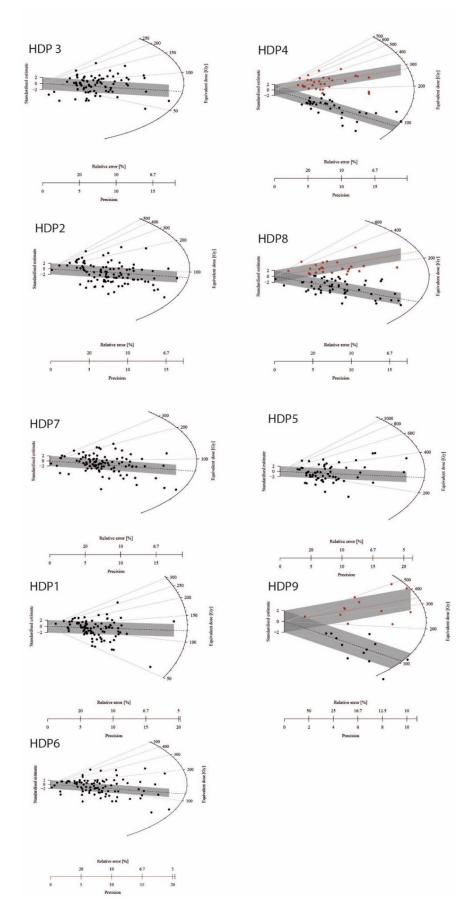




Fig Sup C: radial plots for each sample (based on the selected grains). The colored bands corresponds to either the CAM De
or to the components of the FMM.

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

703

b	mass of carbonate/mass of siliceous+carbonated material										
sample	> 10	cm	1cm -	2mm	2mm -	710 µm	<710 μm				
HDP3	95%	±22%	88%	±6%	88%	±6%	85%	1%			
HDP2	86%	±25%	90%	±7%	90%	±9%	86%	1%			
HDP7	93%	±22%	77%	±2%	90%	±5%	83%	1%			
HDP1	50%	±1%	46%	±1%	45%	±1%	60%	1%			
HDP6	60%	±1%	nd	nd	58%	±1%	49%	1%			
HDP4	92%	±12%	87%	±4%	86%	±6%	80%	1%			
HDP8	76%	±1%	89%	±4%	86%	±6%	81%	0%			
HDP5	90%	±13%	92%	±43%	88%	±31%	88%	1%			
HDP9	88%	±4%	88%	±5%	89%	±4%	88%	0%			

704

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

707

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