



| 1 | Late Neogene terrestrial climate reconstruction of the Central | | | | | | |
|--------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--|--|--|--|--|--|
| 2 | Namib Desert derived by the combination of U-Pb silcrete and | | | | | | |
| 3 | TCN exposure dating | | | | | | |
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| 16 | Keywords: Namib Desert, U-Pb dating, groundwater sil-/calcretes, cosmogenic nuclides | | | | | | |
| 17 | <u>Abstract:</u> | | | | | | |
| 18 | The chronology of the Cenozoic, so called 'Namib Group' of the Namib Desert is rather poor in | | | | | | |
| 19 | terms of direct radiometric dating. Most of the chronological information is based on the ostrich | | | | | | |
| 20 | shell biochronostratigraphy. The widespread occurrence of calcretes and silcretes in the Namib | | | | | | |
| 21 | Desert makes it possible to date important phases of landscape stability and to retrieve critical | | | | | | |
| 22 | paleoclimatic and -environmental information on desertification and its paleoclimatic variability. | | | | | | |
| 23 | The application of the U-Pb laser ablation dating technique to Plio/Pleistocene sil- and calcretes | | | | | | |
| 24 | provides critical insights into groundwater calcrete formation and climate variability in the | | | | | | |
| 25 | Central Namib. Microscale silcrete formation due to pressure solution by expanding calcrete | | | | | | |
| 26 | cementation provides the opportunity to date multiple phases (multiple generation of silcrete as | | | | | | |
| 27 | growing layers or shells) of silcrete formation and to trace their paleoclimatic and -environmental | | | | | | |
| 28 | fingerprints. Groundwater sil- and calcrete formation occurred during the Pliocene. TCN exposure | | | | | | |
| 29 | ages from flat canyon rim surfaces indicate the cessation of groundwater calcrete formation due | | | | | | |
| 30 | to incision during the Late Pliocene/Early Pleistocene and mark a large-scale landscape | | | | | | |
| 31 | rejuvenation due to climate shifts towards the Pleistocene. This study demonstrates the | | | | | | |

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application of U-Pb laser ablation to groundwater sil- and calcretes in desert environments and
 opens up the possibility of dating numerous sedimentary sequences containing sil- and calcretes
 in arid environments. In particular, the use of silcretes (as described above) reduces potential
 effects of detrital components and bulk-signal measurements by using massive calcretes. Our
 study redefines and improves the generally accepted Late Cenozoic chronostratigraphy of the
 Namib Desert (Miller, 2008).

38 **1. Introduction**

In Namibia, widespread calcretes, together with (spatially) more restricted silcretes, are among the most auspicious features of the Cenozoic surface cover along deeply incised ephemeral or fossil drainage systems in terms of their ability to record past environmental change (Miller, 2008; Candy et al., 2004; Summerfield, 1983a; Van Der Wateren and Dunai, 2001; Ward, 1987). As well as being an important component in explaining the generally low denudation rates due to their protective function (Stokes et al., 2007; Nash and Smith, 1998), these sil- and calcretes also indicate relatively long periods of landscape and climate stability during their formation.

46 In general, calcretes are thought to form under semi-arid to arid conditions, with varying 47 interpretations and associations with specific precipitation ranges (Goudie, 2020; Summerfield, 48 1983a; Alonso-Zarza, 2003). The formation of calcretes is largely dependent on the local climate 49 and the availability of calcium and carbonate ions in the system, produced by weathering and 50 leaching in the catchment. Various models have been proposed to explain the different types of 51 calcrete (pedogenic, non-pedogenic, Goudie, 2020). In this study, we will mainly focus on the 52 non-pedogenic, groundwater-related calcrete formation, based on the *per-ascensum* hypothesis 53 (Goudie, 1996; Goudie et al., 2015). One of the most prominent calcrete formations is related to 54 calcretes capping the Karpfenkliff Conglomerate of the Kuiseb Canyon in the Central Namib and 55 the Kamberg Calcrete Formation (Fig. 1, Ward, 1987). Secondary silcrete formation by pressure solution and reprecipitation was synchronous with calcrete formation in the Karpfenkliff 56 57 conglomerates. It consists of microscale silcrete with discrete multiple layers of silcrete encrusting quartz clasts within this formation. The Karpfenkliff Conglomerate overlies the 58 59 Tsondab Sandstone and was probably deposited in a proto-Kuiseb and a proto-Gaub valley (Ward 60 et al., 1983; Miller, 2008; Ward, 1987).

61 Calcretes in the central Namib are thought to be at least Early Pleistocene to Pliocene in age (Ward, 62 1987; Miller, 2008). Common dating techniques used to date calcretes are radiocarbon ¹⁴C, U/Th 63 disequilibrium, or solution U-Pb. Calcrete U-Pb laser ablation has recently been used to provide 64 critical chronological information on the age-depth relationship of the calcretized sediments from 65 the Etosha Pan (Houben et al., 2020). However, the dating of calcretes using the U-Pb system may





66 be influenced/biased by detrital components from the source area of the leached carbonates. The 67 rather slow growth rate makes it difficult to obtain individual ages from multiple generations of 68 calcrete formation when using the bulk sampling approach for solution U-Pb dating, as it is 69 affected by the 'nugget' effect (Branca et al., 2005). Although calcrete formation clearly pre-dates 70 the major canyon incision that can be dated using TCN exposure dating, calcrete formation 71 post-dates sediment deposition and is not age-equivalent to the host sediments. However, the 72 time lag between sediment deposition and calcrete formation may be negligible for any expected 73 age in the range of several millions of years. To avoid contamination by detrital components from 74 the catchment and to date multiple stages of sil-/calcrete formation, syndepositional (with 75 calcrete formation), microscale silcretes produced by the secondary effect of calcrete formation, 76 pressure solution and re-precipitation in the proximity (pressure shadow) might be a valuable 77 target.

78 Evidence for climate change and major landscape change, as well as the reliability of dating of 79 Plio/Pleistocene sediments in the Namib, is relatively poor and not well constrained, and in part 80 shows discrepancies between different dating techniques and interpretations (Miller, 2008; Van 81 Der Wateren and Dunai, 2001; Goudie and Viles, 2014). In Namibia, calcretes and silcretes 82 commonly form prominent landscape features (i.e., cliffs) along deeply incised ephemeral or fossil 83 drainage systems (Miller, 2008; Van Der Wateren and Dunai, 2001; Ward, 1987). The (relative) 84 chronology of these fossil duricrusts is the backbone of the (late) Cenozoic chronostratigraphy of 85 the Namib Desert ('Namib Group' of Miller, 2008) and past climate reconstructions (Miller, 2008, 86 and references therein). A major weakness of this chronostratigraphy is its absolute chronology: 87 essentially all early Quaternary to mid- Miocene continental deposits in the Namib Desert are 88 dated with ostrich shells (Miller, 2008) or are age-correlated with deposits dated with such shells 89 (Miller, 2008). The catch is that only the oldest shells (Aepyonithoid, Senut, 2000) are 'dated' to 90 16-20 Ma (Pickford et al., 1999), whereas the ensuing eight ostrich species are arbitrarily assigned 91 to 2 to 3 Myr long periods (Senut, 2000) without any direct age control. The ostrich shell 92 biostratigraphy provides a valuable relative chronology, but its use in its current form as an 93 absolute chronology remains unverified for the time < 16 Ma. Consequently, the generally 94 accepted Late Cenozoic chronostratigraphy of the Namib Desert (Miller, 2008) requires 95 verification.

96 The use of TCN exposure dating in the Namib Desert has grown in recent years, demonstrating 97 that this method is a reliable way to measure landscape change (Van Der Wateren and Dunai, 98 2001; Vermeesch et al., 2010; Stone, 2013; Bierman and Caffee, 2001). According to Van Der 99 Wateren and Dunai (2001), major changes in the Namib Desert, i.e. rejuvenation of the landscape 90 by intermittent fluvial phases during the predominant arid to hyperarid climate, indicate major 91 changes during the Plio-/Pleistocene. However, there are doubts about the interpretation of the





exposure ages in relation to the underlying deposited sediments (Miller, 2008). The dating of
groundwater connected sil- and calcretes beneath the surfaces sampled for TCN exposure dating
allows to verify the resulting TCN exposure ages. Furthermore, the combination of both dating
techniques can be used to build a robust chronology of landscape change during the evolution and
intensification of arid conditions in the Namib Desert.

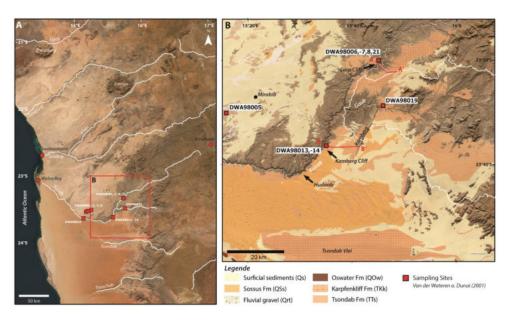
107 Here we present the new application of U-Pb laser ablation to groundwater silcretes from the 108 Namib Desert in combination with re-measured TCN exposure ages from the Karpfenkliff and 109 nearby equivalent sites. Laser ablation U-Pb dating of multiple microscale silcrete layers from the 110 Karpfenkliff Conglomerate Formation indicates groundwater cal-/silcrete formation during the 111 Pliocene. Re-measured surface clasts from Van Der Wateren and Dunai (2001) confirm and 112 substantiate the interpretation of a major landscape rejuvenation of the Central Namib during the 113 Plio-/Pleistocene transition. The combination of the two dating techniques allows a robust 114 chronological reconstruction of landscape evolution and the paleoclimate transition to increasingly arid conditions in the central Namib Desert. 115

116 **2.** Sampling Site and Samples

117 The central Namib Desert, between the Atlantic Ocean to the west and the Great Escarpment to 118 the east, is a relatively flat landscape with numerous dispersed inselbergs and locally deeply 119 incised canyons formed by ephermal rivers such as the Kuiseb or Swakop (Fig. 1). Our study 120 focuses on the Kuiseb River canyon in the central Namib. The ephermal Kuiseb River marks the 121 prominent boundary between the stone desert in the north and the Namib Sand Sea to the south. 122 The Kuiseb River receives its water from precipitation in the Great Escarpment to the east, with 123 mean annual rainfall of 200-450 mm/yr (Ward, 1987; Jacobson et al., 1995). Annual floods of the 124 Kuiseb River clean its bed of all sand transported from the Namib Sand Sea to the south. They only 125 reach the sea during exceptionally high floods (Van Der Wateren and Dunai, 2001). The Kuiseb River forms a distinctive deep and partly narrow canyon, which is up to 250 m deep and only 126 127 1000 m wide at its deepest part (Fig. 1, 2). The recent course of the Kuiseb River is southsouthwest to Hudaob, where it is thought to have been redirected north-west by the activity of the 128 129 Namib Sand Sea (Miller, 2008). Prior to this deflection, the Proto-Kuiseb River may have flowed 130 westwards, as indicated by numerous outcrops within the interdune valleys of the Namib Sand 131 Sea (Fig. 25.18 Vol. 3 in Miller, 2008; Ward, 1987; Lancaster, 1984).







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133 Fig. 1: (A) Overview map of the Central Namib Desert based on World Imagery (Earthstar 134 Geographics (TerraColor NextGen) imagery, ArcGIS Pro Version 3.1.0). Major drainage systems are 135 shown in white. Red rectangle indicates the study area. Red squares indicate sampling sites of Van 136 Der Wateren and Dunai (2001). Topographic profiles in Fig. 2 are marked as red lines. (B) Study area 137 (Earthstar Geographics (TerraColor NextGen) imagery, ArcGIS Pro Version3.1.0) including mapped 138 geology by the Geological Survey of Namibia (Geological Survey of Namibia, 2016). Relevant 139 geological formations are shown covering the Cenozoic sediment succession of the Central Namib 140 (Namib Group). Red squares indicate sampling sites from Van Der Wateren and Dunai (2001). The 141 sub-catchment of the Gaub River is shown in white.

142 Sediment Succession Kuiseb Canyon

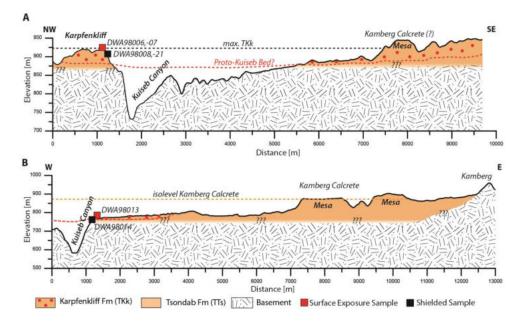
The outcrop sequence along the Kuiseb canyon in our study area comprises up to 100 m of sedimentary units (Fig. 2), consisting of basal breccias, Precambrian basement, and solidified aeolian sands assigned to the Tsondab Sandstone Formation, overlain by calcretized coarse-grained conglomerates, called the Karpfenkliff Conglomerate Formation (Ward, 1987). Well-preserved terraces, resistant to weathering due to calcretization, are exposed at the rim of the canyon (Fig. 3).

The Tsondab Sandstone Formation rests on the Namib Unconformity Surface (NUS, Ward, 1987;
Miller, 2008), and is the oldest and first terrestrial Cenozoic deposit in the Central Namib (Ward,
1987; Miller, 2008), covering large areas of the Central Namib (Fig. 1, 2). The Tsondab Sandstone
Formation consists predominantly of cemented aeolianites (Miller, 2008; Ward, 1987) and is
regarded as the precursor of the recent Namib Sand Sea (Ollier, 1977). The Tsondab Sandstone
Formation is thought to have been deposited under predominantly arid conditions (Ward, 1987),





- 155 between 20-16 Ma and 5 Ma based on the biostratigraphy of Struthious eggshells (Namoris
- 156 Oshanai, Struthio Karinagarabensis, Ward and Corbett, 1990; Pickford et al., 1995; Senut, 2000).



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158 Fig. 2: (A) Fig. 2: (A)Cross-sections of the Karpfenkliff and (B) Kamberg Cliff based on SRTM data 159 (created using ArcGIS Pro 3.1.0). Spatial information on geological units was extrapolated from 160 mapped geology (Geological Survey of Namibia, 2016). Samples were collected from the surface of 161 the Karpfenkliff (DWA980006,07) and from the subsurface at the canyon outcrop (DWA980008, -162 21). The identical sampling approach was used on the Kamberg Cliff, by sampling exposed quartz clasts (DWA98013) and shielded clasts (DWA98014). The shielded quartz clasts were used to 163 164 investigate and date secondary micro-scale silcrete layers attached to quartz clasts. Due to the 165 unknown fluvial topography of the Proto-Kuiseb, the profile is just an approximation. The occurrence 166 of the Karpfenkliff Conglomerate Formation (TKk) is used from the geological map, however, its 167 outcrop condition in profile A in the eastern sector remains speculative. The exact transition from the underlying Tsondab Sandstone (TTs) to the Karpfenkliff Conglomerate Formation is unclear and 168 169 is approximated. The elevation of the Kamberg Calcrete from its key position is marked in B and to 170 illustrate the potential discrepancy between the two formations.

171 Proto-Kuiseb Incision and aggradation of the Karpfenkliff Conglomerate Formation

The Karpfenkliff Conglomerate Formation (TKk, Ward, 1987) overlies the Tsondab Sandstone Formation and was deposited in a proto-Kuiseb and proto-Gaub River valley, a tributary of the Kuiseb River (Fig. 1, 2). Pre-depositional incision of the Kuiseb and Gaub rivers probably occurred during a wetter phase (Ward, 1987; Miller, 2008). The incision excavated a broad shallow valley and eroded the semi-consolidated Tsondab Sandstone without eroding the underlying Pre-Cambrian Damaran schists (Ward, 1987; Miller, 2008). The Karpfenkliff Conglomerate Formation consists of a medium- to fine-grained, sand-sized matrix of angular to subrounded





179 clasts (Fig. 3, Ward, 1987; Miller, 2008). Clasts are rounded to well-rounded with numerous 180 occurring percussion marks (Ward, 1987; Van Der Wateren and Dunai, 2001). The Karpfenkliff 181 Conglomerate Formation thins to the west, indicating a depositional wedge (Miller, 2008). The 182 thickest accumulations are found at the foothills of the Great Escarpment (~ 60 m, Miller, 2008), 183 decreasing to ~ 40 m (Ward, 1987) in the upper Gaub Valley, 20-30 m at the Karpfenkliff, and thinning to ~5 m at Gomkaeb (Ward, 1987; Miller, 2008). Deposition took place in a wide, shallow, 184 185 braided river system (Ward, 1987; Miller, 2008), presumably during an intermittent pluvial phase 186 despite prevailing arid conditions and synchronous with the deposition of the Tsondab aeolianites (Ward, 1987). Equivalent gravels in the Tsondab, Tsauchab and Swakop rivers are assigned to the 187 Karpfenkliff Conglomerate Formation (Miller, 2008). The conglomerate is cemented by a massive 188 189 groundwater calcrete that has caused significant volume expansion (Miller, 2008). The source 190 area of the carbonate ions is thought to be the outcropping and eroding Precambrian Nama Group 191 in the headwaters of the Kuiseb River and is therefore authogenic in origin. Calcretization caused 192 secondary precipitation of microscale silcrete by pressure solution and local re-precipitation.

193 The Karpfenkliff Conglomerate Formation is age-correlated with the occurrence of Diamantornis 194 corbetti in the Tsondab Aeolianites at Elim (Pickford and Senut, 2000; Miller, 2008), implying a 195 younger age of Diamantornis corbetti than 14-15 Ma, equivalent to the age of the Arries Drift 196 Formation (Miller, 2008). Youngest deposition age (prior to 2.81 ± 0.11 Ma) was proposed by Van 197 Der Wateren and Dunai (2001) based on ²¹Ne exposure dating of abandoned surfaces of the 198 Kuiseb River. The latter indicates the minimum depositional age for the last remnants of any 199 fluvial transport and deposition of the Karpfenkliff Conglomerate Formation. Although this age is 200 controversial according to Miller (2008, page 25-27) based on the ostrich shell biostratigraphy, it 201 clearly indicates the onset of incision by the recent Kuiseb River.

202 Calcrete within Karpfenkliff Formation and Tsondab Sandstone - Kamberg calcrete formation

203 The Kamberg Calcrete is described as a pedogenic calcrete up to 5 m in thickness (Miller, 2008; Ward, 1987; Yaalon and Ward, 1982). According to Miller (2008), it cements the upper 204 205 Karpfenkliff Conglomerate Formation in places, as well as the Tsondab Sandstone, which covers a 206 large area east of Homeb in the Kuiseb River (Miller, 2008; Ward, 1987; Yaalon and Ward, 1982). 207 The Kamberg Calcrete, as well as equivalent calcretes in the study area, represent the surface 208 predating the recent canyon incision of the Kuiseb and Gaub rivers. They are used as an important 209 stratigraphic marker horizon in the Cenozoic 'Namib Group' (Miller, 2008; Ward, 1987). Whether 210 the Kamberg Calcrete is identical to the calcrete of the Karpfenkliff can be questioned (Fig. 2). The 211 pedogenic Kamberg Calcrete may be transitional to the groundwater calcrete found at the 212 Karpfenkliff and therefore be syndepositional. If the Kamberg Calcrete at the key site at Kamberg 213 correlates with the Kamberg Cliff and the Carp Cliff at Kuiseb canyon, this would imply that it is





stratigraphically equivalent to or younger than the groundwater calcrete cementing the Karpfenkliff Conglomerate Formation (Fig. 2). A late Miocene age has been suggested for the evolution of the Kamberg Calcrete (Yaalon and Ward, 1982; Ward, 1987). The calcrete is thought to have been formed under semi-arid conditions during a relatively long period of landscape stability (Goudie et al., 2015; Ward, 1987), with seasonal precipitation of potentially 350-450 mm in the headwaters, decreasing drastically to the west (Ward, 1987).

A clear differentiation between the Kamberg Calcrete and any calcretes overlying and/or within the Karpfenkliff Conglomerate Formation is difficult. The Kamberg Calcrete is not specifically mapped in the published geological maps (Geological Survey of Namibia, 2016). For our study, we focused on near-surface clasts with silcrete at the Karpfenkliff. The clear spatial and evolutionary differentiation, as well as the connection between the two, should be the focus of future research to use their occurrence as a marker horizon in the Central Namib.

226 Kuiseb Incision – Phase of landscape rejuvenation

227 The incision of the Kuiseb River (and other adjacent rivers such as the Swakop to the north) is 228 thought to have begun at the end of the Neogene, synchronous with other major river systems in 229 South Africa (Ward, 1987; King, 1951; Partridge and Maud, 1987; Korn and Martin, 1957). The 230 recent incision was able to cut deeply into the Karpfenkliff Conglomerate Formation, the Tsondab 231 Sandstone Formation and also into the Pre-Cambrian Damaran schists (Miller, 2008; Van Der 232 Wateren and Dunai, 2001; Ward, 1987), forming a V-shaped valley and the famous Kuiseb canyon 233 (Fig. 3). The transition from the aggradation of the Karpfenkliff Conglomerate Formation and the 234 formation of calcretes, to the degradation and incision of the recent Kuiseb, Gaub and Swakop 235 rivers is thought to be related to either a tectonic- (King, 1955; Ward, 1987; Korn and Martin, 236 1957) or climatic control and forcing (Van Der Wateren and Dunai, 2001; Richards and Richards, 237 1987; Weissel and Seidl, 1998).

238 Detailed sampling sites and sampling

239 We consider that the calcrete at our sampling sites (Karpfenkliff, and Kamberg Cliff, Fig. 1, 2, 3) 240 was formed primarily by groundwater interaction, due to its direct location near the present-day 241 Kuiseb canyon. We used sampled and dated (in-situ ²¹Ne) surface quartz clasts from Van Der 242 Wateren and Dunai (2001), from abandoned exposed surfaces and shielded clasts from several 243 metres below the surface. Details of the sampling procedure and sampling sites are given in Van Der Wateren and Dunai (2001). For this study we concentrated on surface quartz clasts from the 244 245 Carp Cliff (DWA98006, -07, -08, -21) and the Kamberg Cliff (DWA98013, -14) for re-measurement 246 of cosmogenic ²¹Ne concentrations. Eight quartz clasts from the Carp Cliff with visible silcrete





- 247 cementation were prepared for U-Pb laser ablation. The following descriptions are taken from
- 248 Van Der Wateren and Dunai (2001) and partly adapted for additional samples.



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Fig. 3: Outcrop image compilation. (A) From Van Der Wateren and Dunai (2001) Kamberg Cliff, ~15 m of
Karpfenkliff Conglomerates overlying ~15 m of Tsondab Sandstones. (B) Carp Cliff (Field Campaign 2018). (C)
Close-up of the calcrete-cemented Karpfenkliff Conglomerate Formation. Rounded quartz clasts float in a
matrix-supported fabric which is cemented by calcrete. Some clasts are fractured by volume expansion of the
calcrete, resulting in pressure solution and formation of micro-scale silcrete. (D) Surface of the calcrete
cemented Karpfenkliff.

256 <u>Carp Cliff (Kuiseb highest terrace)</u>

257 DWA98006-07 (Site 6) is located on the horizontal upper surface of a mesa-shaped terrace 258 remnant 500 m west of the 200 m deep Kuiseb Canyon (Fig. 1, 2). The terrace has a surface area 259 of ~5 km² and is surrounded by steep, locally vertical and overhanging cliffs, which to the east and 260 south are nearly 50 m in height. The terrace surface consists of a desert pavement of mainly quartz pebbles overlying up to 15 cm of sandy silt. This is underlain by 10-25 m of calcretized pebble 261 and boulder conglomerates of the Karpfenkliff Conglomerate Formation (Fig. 3). Van Der Wateren 262 and Dunai (2001) collected 40 rounded (DWA98007) and (sub-)angular pebbles (DWA98006) 263 264 with diameters between 2 and 6 cm. The site is located at the top of the mesa and is almost 265 horizontal, so that post depositional transport of the sampled pebbles by the Kuiseb River or local precipitation can be excluded. 266





267 DWA98008 and DWA980021 (Site 7 and 19) are located next to small gullies running from the 268 north side of the Carp Cliff mesa. At these sites, we collected shielded samples from 5 m below the 269 terrace surface. Van Der Wateren and Dunai (2001) sampled rounded pebbles from the ceilings 270 of overhangs to ensure that the measured ²¹Ne concentrations were derived only during hillslope 271 and fluvial transport to their present site and not from subsequent exposure at the sampling site.

272 <u>Kamberg Cliff (Kuiseb highest terrace)</u>

273 DWA98013 and DWA980014 are from a terrace on the Karpfenkliff Conglomerate Formation 274 immediately adjacent to the nearly 250 m deep Kuiseb Canyon, 30 km downstream of Carp Cliff 275 (Fig. 1, 2, 3). The terrace surface is very similar to that of Carp Cliff, with a desert pavement of 276 pebbles and cobbles on a sandy silt overlying 25 m of calcretized conglomerates. The Karpfenkliff 277 Conglomerates rest on 30-50 m of the Tsondab Sandstone Formation, which forms the bulk of the 278 cliff adjacent to the canyon. DWA98013 sampling site is on the horizontal surface of the terrace, 279 where we sampled angular pebbles. At DWA98014, rounded pebbles (DWA98014) were sampled 280 from the ceiling of an overhang in the cliff face 6 m below.

3. Formation of Calcretes and microscale silcretes

282 In general, two types of calcretes can be differentiated, pedogenic and groundwater calcretes 283 (Alonso-Zarza and Wright, 2010), following the per descensum or the per ascensum evolutionary 284 model (Goudie, 2020). They occur preferentially in arid to semi-arid climates (Alonso-Zarza, 285 2003; Candy and Black, 2009; Goudie, 2020). Specific climatic and environmental conditions are 286 required for calcrete formation; (1) precipitation in the headwater/source area to promote 287 carbonate dissolution, (2) intermittent or seasonal precipitation downstream favour 288 groundwater systems capable of (3) causing high evaporation and evapotranspiration for 289 chemical precipitation of carbonate (Mann and Horwitz, 1979). Calcrete formation is dependent 290 on the supply of carbonate ions leached from the drainage bedrock. In this study we focus on 291 groundwater calcretes formed along the Kuiseb- and Gaub rivers. Groundwater calcretes form at 292 or above shallow groundwater tables/aquifers (Mann and Horwitz, 1979; Netterberg, 1969) and 293 do not require subaerial exposure, although shallow contacts and stable surfaces favour the 294 evolution of groundwater calcretes (Alonso-Zarza, 2003). They were originally called 'valley 295 calcretes' (Butt et al., 1977) because of their relationship with drainages. Groundwater calcretes 296 are rather restricted to local drainages, although groundwater calcretes can have lateral extents 297 of more than 100 km long and 10 km wide, depending on the drainage topography (Mann and 298 Horwitz, 1979). Groundwater calcretes do not have characteristic features compared to 299 pedogenic calcretes and are rather massive bodies (Alonso-Zarza, 2003). The permeability 300 (coarse channel sediments) of the host rock favours their formation (Alonso-Zarza and Wright, 301 2010). Calcretes have been frequently used to obtain paleo precipitation information, but the





specific ranges are still under discussion. The upper limit may be between 600 and 1000 mm/yr
(Mack and James, 1994). The lower limit may be as low as 50 mm/yr (Goudie, 1973; Retallack,
1994).

305 Silcretes can form as duricrust due to the accumulation of secondary silica within a soil or host 306 rock (Milnes and Thiry, 1992; Summerfield, 1983a). Prominent examples include silcretes from 307 Australia (Milnes et al., 1991; Taylor and Eggleton, 2017) or the Kalahari Desert (Summerfield, 308 1983b; Nash and Shaw, 1998). In this study, we focus on microscale silcretes, which are formed by pressure solution (Mcbride, 1989; Rutter, 1983; Sorby, 1863; Wilson, 2020) due to calcrete 309 310 cementation and volume expansion within the host rock or sediment, and therefore cannot be directly compared to the commonly used term 'silcrete'. Microscale silcrete formation is therefore 311 312 thought to be linked to paleo-environmental and climatic conditions favourable to calcrete formation. Calcretization involves the precipitation of CaCO₃ within the pore spaces of the host 313 314 rock or sediment, causing significant volume expansion. A secondary effect of this process is to 315 increase the differential pressure within the host rock or sediment, causing clast shattering, 316 relocation and pressure solution at intergranular contacts (Sorby, 1863; Rutter, 1983). Increased 317 stress at grain boundaries and intergranular contacts leads to dissolution, e.g., silica mobilisation. 318 Mobilized solutions migrate to regions of lower compressive stress, the 'pressure shadow', to 319 reprecipitate. Theoretically, depending on the remaining pore space, multiple pressure solution 320 and reprecipitation cycles can be archived in the host rock as multiple silcrete layers or shells 321 attached to quartz clasts.





322 **4. Methods**

323 Raman Spectroscopy

324 Raman spectra were collected with up to 1300 wavenumber (cm⁻¹), using a WITec alpha 300R 325 confocal Micro-Raman microscope, at the Goethe University Frankfurt (GUF). The objective used 326 was 50x, an excitation laser of 532 nm (using 10 mW laser power before the objective), and 327 spectra integration time of 0.2 s with 5 accumulations in total. Maps (400 x 400 μ m²) were 328 performed applying a step size of 1.3 µm with a holographic grating of 600 grooves mm⁻¹. The 329 instrument was calibrated using an Ar-Hg spectral lamp and was checked regarding its 330 performance before the measurements with respect to the 1300 cm⁻¹ line of silicon. The spectrum 331 of each sample layer was confirmed at several locations on the same layer. Raman spectra of 332 reference compounds are found in the Rruff database (https://rruff.info/).

333 Dating of Silcretes – U-Pb Laser Ablation ICP-MS

334 Quantifying the timing and duration of calcrete formation is quite difficult. Clear stratigraphic 335 relationships with the overlying and underlying sediments are not straightforward, as 336 groundwater calcrete, for example, forms within sediments deposited close to the surface. 337 Numerous studies propose only relative age controls and estimates of the formation time, such as the application of the ostrich shell biochronostratigraphy used for the Namib Group (Pickford and 338 339 Senut, 2000; Senut, 2000; Miller, 2008). Many attempts have been made to date deposits and 340 determine formation times using radiocarbon 14C (e.g. Geyh and Eitel, 1997), U/Th (Kelly et al., 2000; Candy et al., 2004; Candy and Black, 2009) or U-Pb dating (Rasbury and Cole, 2009; Houben 341 342 et al., 2020). The latter dating technique allows the investigation of much older calcretes than 343 radiocarbon ¹⁴C or U/Th.

344 Silcretes are enriched in U relative to calcretes and occur in most soils in arid and semi-arid 345 environments. Uranium decays to Pb isotopes through a chain of intermediate daughter isotopes, 346 and ages of thousands- to millions-of-years-old samples can be estimated using parent-daughter pairs ²³⁸U-²⁰⁶Pb, ²³⁵U-²⁰⁷Pb, ²³⁴U-²³⁰Th, and ²³⁸U-²³⁴U. The use of a particular isotope pair depends 347 348 on how old the sample is compared to the half-life of the selected radioactive isotope within the U 349 decay chain (Neymark, 2011; Neymark et al., 2002, 2000). Considering that the samples are 2.85 350 Ma old or older (Van Der Wateren and Dunai, 2001), the U-Pb method using the parent-daughter pairs ²³⁸U-²⁰⁶Pb and ²³⁵U-²⁰⁷Pb was chosen to date the samples in this work. 351

Many studies attempting to date massive cal-/silcretes are hampered by the dilution or averaging effect of bulk analysis and by bias from non-carbonate detrital minerals or secondary reprecipitated carbonate due to diagenesis. The "limestone dilution effect" (as a result of contamination with detrital carbonate components of the host rock, Alonso-Zarza, 2003) or the





"averaging effect" (averaging of different phases of mineral precipitation, Candy and Black, 2009;
Neymark et al., 2000) are minimised (or even avoided) by the higher spatial resolution of laser
ablation compared to bulk analysis techniques. The possible effect of detrital components (e.g.
Zircon or clay minerals) on the U-Pb analyses is also neglected, as the signals from these inclusions
can be filtered out of the time-resolved analyses.

361 The conventional method of calculating U-Pb isotope dates assumes that all intermediate daughter isotopes in the ²³⁸U and ²³⁵U decay chains were in secular equilibrium at the time of 362 363 formation (i.e. the radioactivity of all daughter isotopes was equal to that of the parent, Neymark, 364 2014). This is not necessarily true for calcretes and silcretes due to differences in the geochemical 365 behaviour of parent and daughter elements. The silcretes dated in this study are sufficiently old 366 (> c. 2.85 Ma) to have achieved secular equilibrium (at present), and therefore allowing all its 367 initial excess of daughter isotopes to decay, or their initial depletion to replenish (i.e. their activity ratios to be equal to 1). This does not allow any initial excess or depletion of daughter isotope to 368 be measured, making it difficult to correct for potential bias in the ²⁰⁶Pb*/²³⁸U ages. The daughter 369 370 isotopes of interest are those with longer half-lives, namely ²³⁴U and ²³⁰Th, both belonging to the 371 ²³⁸U decay chain. A deviation of 1 initial (²³⁴U/²³⁸U) activity ratio ([²³⁴U/²³⁸U]_i) unit from unity (e.g. $[^{234}U/^{238}U]_i = 0$) will cause an age difference of c. 354 ky and a deviation of 1 $[^{230}Th/^{238}U]_i$ will 372 deviate our ²⁰⁶Pb*/²³⁸U dates by c. 109 ky (considering that secular equilibrium has been reached, 373 374 calculated after Wendt and Carl, 1985).

375 Eight quartz clasts were cut in half to expose their silcrete coatings, mounted in epoxy mounts and 376 polished at the Department of Geosciences, University of Cologne (UoC). U-Pb analyses were 377 performed at the Goethe University Frankfurt (GUF) using a RESOlution 193 nm ArF excimer laser 378 (COMpex Pro 102), equipped with a two-volume ablation cell (Laurin Technic S155). The laser 379 was coupled to a ThermoScietific ElementXr sector field ICP-MS. The surfaces were cleaned with 380 8 pre-ablation laser pulses. Ablation was carried out in a He (0.3 l/min), Ar (1.01 l/min) and N 381 (0.012 l/min) atmosphere, with a high energy density (c. 5 J/cm²), a frequency of 15 Hz and round 382 50 µm diameter spots (SI2_Supporting Information).

383 Artificial silicate glasses NIST SRTM 612 and 614 were used as reference materials (RM). Plots 384 and dates are calculated using the in- house spreadsheet program (Gerdes and Zeh, 2009, 2006), 385 together with Isoplot (Ludwig, 2012). Ages are reported with and without systematic components 386 (i.e., date $\pm 2s / 2s_{sys}$). Uncertainties include internal standard errors (SE), background, counting 387 statistics, excess scatter of the primary reference material (NIST SRTM 612), and excess variance 388 (calculated from NIST SRTM 614). Systematic uncertainties also propagate systematic errors, 389 which are the long-term excess variance (1.5%, 2s), decay constant uncertainties (Horstwood et 390 al., 2016) and the uncertainty derived from the initial activity ratio. Dates are calculated as Tera-





Wasserburg lower intercepts (Tera and Wasserburg, 1972). Linear regressions are anchored to a
common-lead ²⁰⁷Pb/²⁰⁶Pb ratio of 0.837. This is the Y-intercept of sample "DWA98008-Silc4 Black
Crack", which is where this ratio is better constrained. This value is in good agreement with
modelled crustal values at the time of formation (0.836, Stacey and Kramers, 1975).

395 The samples dated are sufficiently old to have reached secular equilibrium and hence activity 396 ratios cannot be measured (assuming a closed system behaviour). Consequently, the following 397 initial activity ratios used are assumed. The silcretes dated in this study have virtually no Th (average of ~ 89 ng/g) and therefore we consider $[^{230}\text{Th}/^{238}\text{U}]_i = 0$ (initial $^{230}\text{Th}/^{238}\text{U}$ activity 398 399 ratio). Considering previous studies on calcretes and silcretes formed in semi-arid and arid environments (Oster et al., 2017; Maher et al., 2007; Neymark, 2011), the ground and surface 400 waters from which these rocks are formed often have $[^{234}U/^{238}U]_i$ values greater than 1. Therefore, 401 the data in this study are calculated with $[^{234}\text{U}/^{238}\text{U}]_i = 1.75 \pm 0.32$ (2s abs), which is a weighted 402 403 average of the [²³⁴U/²³⁸U]₁ of the above-mentioned studies. The uncertainty in this activity ratio is 404 added to the final systematic uncertainties by quadratic propagation (Scardia et al., 2019; 405 SI2_Supporting Information).

406 <u>Cosmogenic ²¹Ne Exposure Dating</u>

407 We used prepared samples from Van Der Wateren and Dunai (2001) for in situ ²¹Ne exposure 408 dating using the new noble gas mass spectrometer at the University of Cologne. The ²¹Ne analyses 409 of Van Der Wateren and Dunai (2001) were performed without an international standard (CREU). 410 For neon analysis we prepared amalgamated samples from each site containing between 35 and 40 quartz clasts (100mg/sample) using the already prepared $63-125 \,\mu\text{m}$ fraction. By also 411 analysing shielded pebbles, a pre-exposure correction (accumulated ²¹Ne concentration during 412 413 transport) can be applied to analysed surface samples (Repka et al., 1997). The presumably 414 non-atmospheric ²¹Ne concentration found in these samples can be subtracted from the concentration in their exposed counterparts. The latter also corrects for any potential nucleogenic 415 416 ²¹Ne that may be present in the samples. Samples were measured on the noble gas mass 417 spectrometer at the University of Cologne using the analytical methods outlined in Ritter et al. 418 (2021). CREU quartz standards were measured in Cologne for interlaboratory comparability and 419 quality control (Vermeesch et al., 2015). The spallogenic origin of the measured ²¹Ne excess was 420 verified using the triple isotope plot. ²¹Ne exposure ages were calculated using the 'nuclide 421 dependent scaling' after Lifton et al. (2014), calculated with "The online exposure age calculator 422 formerly known as the CRONUS-Earth online exposure age calculator." (Version 3, 423 http://hess.ess.washington.edu/math/v3/v3_age_in.html; Balco et al., 2008).





424 **5. Results**

425 Silcrete Imaging

Digital microscope images show vein-contact parallel-layering with different crystal orientation (Fig. 4A). The Raman spectra peaks at 129, 209 and 467 cm⁻¹ are indicative for quartz, which dominate the silcrete samples. Raman spectroscopy of DWA98008-Silc8 also indicate the presence of one major calcite band (dark colour in Fig. 5B, Raman spectra peaks at 156, 283, 466, 714 and 1088 cm⁻¹). The calcite crack filling might be indicative for shattering of previous silcrete and crack filling by repeated and/or ongoing calcrete formation within the Karpfenkliff Conglomerate.

433 <u>U-Pb Laser Ablation Results Silcretes</u>

Four out of the eight silcrete samples yielded meaningful dates (DWA98008 – Silc3, Silc4, Silc7, Silc8), out of which 12 dates could be calculated (SI2_Supporting Information). The dates range from 2.96 \pm 0.14 to 6.72 \pm 0.16 Ma, with maximum relative abundance peaks at around 3.4 Ma and about 5.5 Ma (see Fig. 6 and Table 1). All dates are calculated from multiple spot analyses, ranging from 9 to 28 spots per date. The majority of the analyses have U concentrations between ~30 and 70 µg/g, with an average of 42 µg/g, and very low Th concentrations, up to 200 ng/g, with an average of 89 ng/g.

441

| Sample name | Date (Ma) ₍₁₎ | 2s abs (2) | 2s sys (3) | Y-Intercept (4) | 2s (5) | Anchored (6) | MSWD (7) | n ₍₈₎ (used) | n ₍₉₎ (total) | Date (Ma) (10) | 2s abs (11) |
|-------------------|-----------------------------|------------|------------|--------------------|--------|-----------------|----------|----------------------------|-----------------------------|-------------------|-------------|
| Silc3 Rim 1 | 5.756 | 0.124 | 0.132 | 0.837 | 0.008 | TRUE | 0.45 | 9 | 9 | 5.590 | 0.120 |
| Silc3 Rim 2 | 5.246 | 0.107 | 0.115 | 0.837 | 0.008 | TRUE | 0.41 | 9 | 9 | 5.080 | 0.103 |
| Silc3 Rim 3 | 5.488 | 0.068 | 0.081 | 0.837 | 0.008 | TRUE | 1.35 | 24 | 28 | 5.322 | 0.066 |
| Silc3 Rim Outer | 3.734 | 0.064 | 0.071 | 0.838 | 0.011 | TRUE | 1.63 | 28 | 28 | 3.569 | 0.061 |
| Silc4 Black crack | 5.927 | 0.244 | 0.248 | 0.8374 | 0.0066 | FALSE | 1.50 | 21 | 25 | 5.761 | 0.237 |
| Silc4 Rim | 3.498 | 0.061 | 0.067 | 0.837 | 0.008 | TRUE | 1.24 | 16 | 17 | 3.332 | 0.058 |
| Silc4 Rim Outer | 3.456 | 0.061 | 0.067 | 0.837 | 0.008 | TRUE | 1.30 | 15 | 17 | 3.290 | 0.058 |
| Silc4 Rim Inner | 3.129 | 0.068 | 0.072 | 0.837 | 0.008 | TRUE | 1.63 | 9 | 9 | 2.964 | 0.064 |
| Silc7 | 6.881 | 0.091 | 0.106 | 0.8528 | 0.0028 | FALSE | 0.94 | 27 | 29 | 6.715 | 0.089 |
| Silc8 Rim Out 1 | 6.116 | 0.081 | 0.095 | 0.837 | 0.008 | TRUE | 0.64 | 20 | 20 | 5.950 | 0.079 |
| Silc8 Rim Out 2 | 5.131 | 0.132 | 0.139 | 0.837 | 0.014 | TRUE | 2.56 | 17 | 25 | 4.965 | 0.128 |
| Silc8 Rim Center | 3.847 | 0.131 | 0.135 | 0.837 | 0.008 | TRUE | 1.37 | 20 | 20 | 3.681 | 0.126 |

442

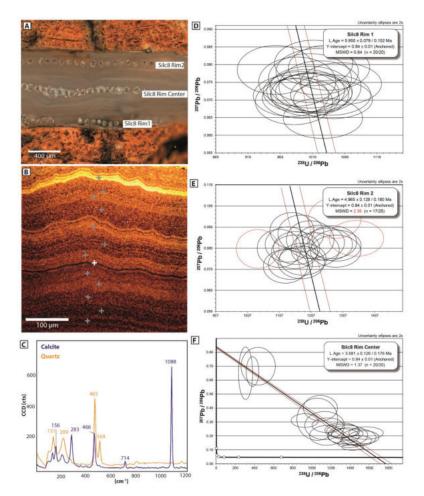
443 Table 1: 1. - Concordia curve lower intercept dates, Tera-Wasserburg diagram (Tera and 444 Wasserburg, 1972). 2. - 2s absolute uncertainties, considering within-run precision (SE of the mean 445 of the ratios), excess of scatter, background, counting statistics and excess of variance (calculated from the validating RM, SRTM NIST 614. 2.6%, 1s, on ²³⁸U/²⁰⁶Pb and 0%, 1s, on ²⁰⁷Pb/²⁰⁶Pb ratios). 446 447 3. - Previous uncertainties (2) expanded with systematic uncertainties (0.8 %, 2s, long term 448 reproducibility and decay constant uncertainties). See Horstwood et al. (2016). 4. - 207Pb/206Pb ratio 449 of the upper intercept. 5. - 2s absolute uncertainty of the upper intercept. 6. - Whether or not the 450 linear regression on the Tera-Wasserburg is anchored. 7. - Mean squared weighted deviates of the 451 regression line. 8. - Number of analyses considered. 9. - Total number of analyses. 10. - Dates 452 calculated (following Wendt and Carl, 1985) considering an initial ²³⁴U/²³⁸U activity ratio of 1.75, 453 and initial ^{230/238} activity ratios of 0. 11. - Uncertainties (2) recalculated to the new dates (10). 11. -





- 454 Uncertainties (3) recalculated to the new dates (10) and adding ²³⁴U/²³⁸U activity ratio uncertainty
- 455 (0.32, 2s abs).

456



457

Fig. 4: (A) Microscope image of sample DWA98008-Silc8 after LA-ICP-MS analysis. Laser spots are 458 459 visible, respective U-Pb Tera-Wasserburg plots are shown in (D-F). (B) Raman image of Silc8, area 460 not visible on microscope image. Grey crosses indicate measured quartz spectra. White cross marked the occurrence of Calcite, which can be traced as a black line. Raman imaging shows multiple 461 layering of silcrete filling the crack of the shattered quartz clast. Color variations are indicative of 462 differences in crystal orientations (C) Respective Raman spectra for the quartz and calcite 463 464 identification. (D-F) Tera-Wasserburg plots of Silc8 and respective U-Pb ages (red ellipses are 465 considered outliers).

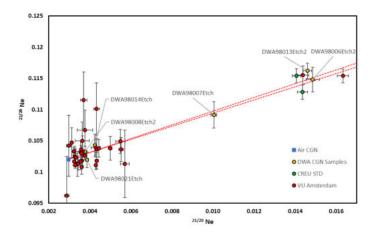
466 <u>TCN Exposure Age Results</u>

- ²¹Ne samples measured at the Noble Gas Laboratory in Cologne (Ritter et al., 2021) yield
 concentrations of 1.52 1.95 x 10⁷ atoms/gr for shielded samples (DWA98008, 014, 021) and
- 469 6.30 9.60 x 10⁷ atoms/gr for surface samples (DWA98006,007, 013). All samples are within 2





- 470 sigma of the spallation line (Fig. 5). Compared to the ²¹Ne concentrations of Van Der Wateren and 471 Dunai (2001), the etched samples measured in Cologne reveal lower concentrations of up to 13% 472 difference when comparing direct concentrations (average of five measured samples), however, 473 within $\pm 1\sigma$ it reduces to ~1.6%, agreeing within $\pm 2\sigma$ on average. We have excluded sample 474 DWA98008, as it presumably contains a high abundance of non-cosmogenic Ne, as deduced from 475 the significant concentration differences between the sample measured by Van Der Wateren and 476 Dunai (2001) and the etched counterpart measured in Cologne (SI1_Supporting Information). 477 Similar results and interpretations for sample DWA98008 were reported by Van Der Wateren and 478 Dunai (2001).
- 479 Using the mean difference of \sim 13% between VU Amsterdam and Cologne Ne concentrations
- 480 (SI1_Supporting Information), the data from Van Der Wateren and Dunai (2001) can be corrected
- for lab-specific differences. The corrected exposure ages are given in Table 2.



482

Fig. 5: Triple isotope diagram indicating single-heat-step extraction of the Cologne laboratory
(orange circles) compared to the multiple-heat-step extraction (red circles) of Van Der Wateren and
Dunai (2001). Uncertainties are 1o. The red stippled line indicates the Cologne laboratory spallation
line (Ritter et al., 2021). Green circles indicate CREU1 measured during the analysis in Cologne.

Calculated exposure ages derived from Cronus Earth (Balco et al., 2008) are summarised in 487 488 Table 2. For the Kuiseb terrace, ²¹Ne concentrations in shielded, pre-exposed samples 489 (DWA98008, DWA98021), give a mean apparent age of 0.65 ± 0.04 Ma (external uncertainty $\pm 1\sigma$). 490 The latter indicates that the non-cosmogenic component of DWA98008 has been removed by 491 etching, indicating the identical apparent exposure age as DWA98021. Correction of the ²¹Ne 492 concentration of exposed rounded pebbles (DWA98007) from the top of the Carp Cliff terrace 493 yields an exposure age of 3.2 \pm 0.2 Ma ($\pm 1\sigma$ external uncertainty), being slightly older than 494 calculated by Van Der Wateren and Dunai (2001), however, identical within the uncertainty.





495 Exposed angular clasts (DWA98006) show a younger exposure age of 2.85 ± 0.19 Ma ($\pm 1\sigma$ external 496 uncertainty). The latter is slightly older than in Van Der Wateren and Dunai (2001), which is 497 identical within their uncertainty. A similar exposure age of 2.75 ± 0.18 Ma ($\pm 1\sigma$ external 498 uncertainty) was derived from angular clasts from the Kamberg cliff (DWA98013) with. Angular 499 clasts are assumed to be derived from local sources without significant pre-exposure from long 491 transport times. Our results indicate that terrace abandonment and exposure to cosmic rays 492 started at ~2.8 Ma (Fig. 6).

502 Table 2: TCN exposure ages.

| | | LSDn Exposure Age | |
|---------------------|----------|-------------------|---------------|
| | Age [Ma] | Int. Unc. [Ma] | Ext. Unc. [Ma |
| DWA98006Etch2 | 2.85 | 0.07 | 0.19 |
| DWA98007Etch | 3.85 | 0.07 | 0.25 |
| Corr. DWA98007Etch2 | 3.20 | 0.05 | 0.20 |
| DWA98008Etch2 | 0.66 | 0.02 | 0.05 |
| DWA98021Etch | 0.65 | 0.02 | 0.04 |
| DWA98013Etch2 | 2.75 | 0.05 | 0.18 |
| DWA98014Etch | 0.90 | 0.02 | 0.06 |
| DWA98001VU | 5.35 | 0.23 | 0.41 |
| DWA98002VU | 3.98 | 0.32 | 0.40 |
| DWA98003VU | 1.08 | 0.13 | 0.15 |
| DWA98005VU | 0.58 | 0.06 | 0.07 |
| DWA98019VU | 0.49 | 0.05 | 0.06 |
| DWA98024VU | 1.27 | 0.12 | 0.14 |

503 6. Interpretation and Discussion

504 Our U-Pb ages are stratigraphically in the correct order, with the oldest U-Pb ages at the contact 505 between quartz clast and filled rock fracture and the youngest age in the centre of the filled 506 fracture (Table 1, Fig. 4). Recurrent U-Pb ages underpin and mark the main phase of silcrete, i.e. 507 calcrete, formation. Groundwater calcrete formation, i.e., microscale silcrete formation, within the 508 sediments of the proto-Kuiseb canyon (Karpfenkliff Conglomerate) took place between the Late 509 Miocene (~7 Ma) and the Late Pliocene (~3 Ma, Fig. 6). The U-Pb silcrete ages suggest either 510 persistent or alternating periods of wetter climate for groundwater calcrete formation.

511 Based on the causal relationship between silcrete and calcrete formation, our U-Pb silcrete ages

512 indicate that environmental and climatic conditions during the Pliocene were sufficient to allow

513 for carbonate leaching, transport and calcrete formation within the coarse-grained Karpfenkliff

- 514 Conglomerate. However, whether the sampled groundwater calcrete is identical or synchronous
- 515 with the prominent Kamberg Calcrete can be questioned, but we can narrow down the timing of





major groundwater calcrete formation, previously assigned to the Late Miocene (Goudie et al.,
2015; Ward, 1987) or Plio-Pleistocene (Pickford and Senut, 2000). Calcrete formation ceased

518 during the Late Pliocene/Early Pleistocene by incision and groundwater lowering (Fig. 6).

Re-measured TCN ²¹Ne surface exposure ages from amalgamated quartz clasts agree with the 519 520 derived U-Pb silcrete chronology and are younger than the youngest U-Pb silcrete age obtained 521 (Fig. 6), i.e., in stratigraphically correct order. The surface exposure ages mark the abandonment 522 of the fluvial terraces and the onset of the Kuiseb River incision at \sim 2.8 Ma. The latter caused a 523 groundwater lowering of the water table and the cessation of calcrete formation within the 524 Karpfenkliff Conglomerate Formation. Re-measurements of the quartz clasts from the Oswater 525 terrace downstream of the Karpfenkliff and Kamberg cliff sampling sites confirm the exposure 526 ages previously obtained by Van Der Wateren and Dunai (2001). The exposure ages of the 527 Karpfenkliff and Oswater terrace constrain the period of major canyon incision to ~ 2.8 -1.3 Ma 528 (Fig. 6).

529 With the aid of absolute U-Pb silcrete and surface exposure dating, it is now possible to redefine 530 depositional ages or depositional periods for sediments in the Central Namib Desert, some of 531 which are widely used as marker horizons. Our U-Pb silcrete ages constrain the timing of sediment 532 deposition within the Kuiseb Canyon (Karpfenkliff Conglomerate Formation) to be older than 533 \sim 7 Ma, as silcrete formation within the conglomerates postdates deposition thereof (Fig. 6). The 534 incision age of the Proto-Kuiseb and the subsequent deposition by the Karpfenkliff Conglomerate 535 as proposed by Miller et al. (2021) of \sim 5 Ma, does not agree with our absolute U-Pb ages. If the relative biostratigraphic dating of Pickford and Senut (2000) is valid, the proto-Kuiseb canyon 536 was filled by the Karpfenkliff Conglomerate Formation over a time period of up to 6-7 Ma 537 538 (Diamantornis corbetti at Elim ~14 15 Ma, see Pickford and Senut, 2000). Verification and 539 absolute direct dating of the Karpfenkliff Formation is still lacking and is a target for future studies. 540 Our fluvial chronology substantially supports the chronological data obtained by Van Der Wateren 541 and Dunai (2001).

542 Pliocene Calcrete Formation – Steady State Climate

543Our U-Pb ages indicate a relatively calm or transitional phase between aggradation and backfilling544of the Proto-Kuiseb (and presumably other drainage systems such as the Swakop) and the545renewed incision by the recent Kuiseb River, throughout the Pliocene. U-Pb ages of microscale546silcrete from the same stratigraphic horizon indicate a long-term stable groundwater level, i.e. no547significant aggradation or degradation.

As the formation of groundwater calcrete is generally restricted to specific environmentalconditions, the existence and chronology of its formation in the Central Namib Desert now allows





550 us to relate these environmental conditions to specific episodes in the past and thus to obtain a 551 better and partly more quantitative paleoclimate and environmental reconstruction of the 552 Pliocene in the Central Namib Desert. However, specific ranges of precipitation are still being 553 discussed, with the upper limit between 600 and 1000 mm/yr (Mack and James, 1994) and the 554 lower limit as low as 50 mm/yr (Goudie, 1973; Retallack, 1994). Ward (1987) suggests seasonal 555 precipitation of potentially 350-450 mm in the upper reaches of the Kuiseb for calcrete formation. 556 We therefore interpret our U-Pb silcrete chronology as marking the transition of the mean annual 557 precipitation (MAP) from the upper potential limit of approximately ~600 mm/yr or the lower potential limit of 50 mm/yr in the Kuiseb catchment during the Late Miocene. Whether there was 558 559 a climate change from, or a return to, wetter conditions during the Pliocene cannot be determined from our U-Pb chronology. Calcrete formation ceased with the incision of the Kuiseb River and a 560 561 significant lowering of the groundwater table.

Marine records off Namibia (Fig. 6, ODP 1081, Hoetzel et al., 2017) suggest a shift to more arid 562 conditions over the course of the Mid to Late Miocene, controlled by a gradual increase in the 563 564 upwelling activity of the Benguela Current, initiated by a strengthening of the meridional gradient. 565 This shift is supported by pollen data (Hoetzel et al., 2015; Dupont et al., 2013), indicating the expansion of savanna grasslands (C4 expansion) in Namibia since \sim 8 Ma, with a subsequent shift 566 567 during the Pliocene to more shrubland and desert vegetation (Hoetzel et al., 2015). 568 Compound-specific hydrogen isotopes (ODP 1085, Dupont et al., 2013) indicate a change in the 569 precipitation source from the Atlantic to the Indian Ocean since ~ 8 Ma (Dupont et al., 2013). 570 Therefore, our U-Pb chronology of calcrete formation (\sim 7-3 Ma) tracks the shift to more arid 571 conditions with a corresponding reduction in the MAP to allow calcrete formation. Nevertheless, 572 this transition and aridification of the Namib was slow, and regional SST records (ODP 1082, 573 Etourneau et al., 2009; ODP 1081, Rosell-Melé et al., 2014, Fig. 6), as well as global paleoclimate 574 records (benthic δ 180, Westerhold et al., 2020, Fig. 6) indicate a relatively stable climatic period. 575 Rosell-Melé et al. (2014) proposed, based on their marine SST record off Namibia (ODP 1082), 576 that the persistently warm Pliocene, with conditions analogue to a persistent Benguela 'El Niño', 577 ended at the transition to the Pleistocene (Fig. 6).

578 Plio/Pleistocene Transition

579 During the transition from the late Pliocene to the early Pleistocene, the Central Namib Desert 580 underwent large-scale landscape rejuvenation with drainage reorganisation and incision. This is 581 the same period, in which Miller et al. (2010) reconstructed the major desiccation of the Etosha 582 paleolake (Fig. 6). Based on U-Pb calcrete ages from the Kalahari basin, Houben et al. (2020) 583 proposed an intensification of arid conditions since ~3.8 Ma, older than our onset of more arid 584 conditions in the Central Namib at around 3 Ma. Higher offshore sedimentation rates off Namibia





may be associated with increased input of terrestrial material (Dupont et al., 2005) due to incision
of E-W flowing drainages into the Atlantic. The propagation of the Horingbaai fan-delta (between
Omaruru and the Ugab river) occurred approximately at the same time (2.7 -2.4 Ma) according to
Stollhofen et al. (2014), supporting the idea of large-scale landscape rejuvenation and incision of
multiple E-W flowing drainages in the Namib Desert.

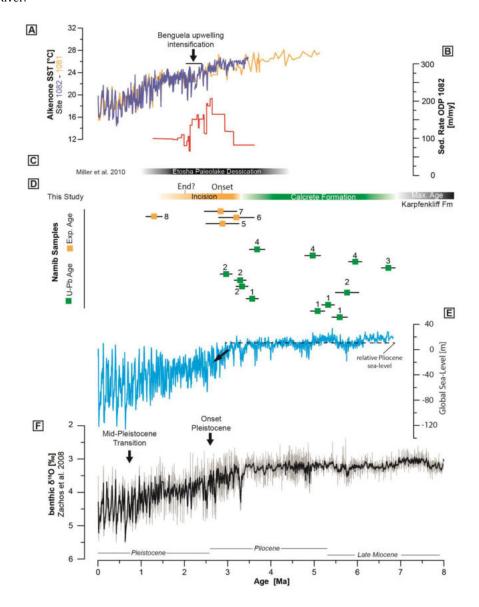
590 The underlying mechanism is still questionable, as several forcing factors could be responsible for 591 the incision of major E-W flowing river systems: climate change and variability, sea level change 592 and/or tectonic uplift. The latter was previously suggested by Ward (1987) and attributed to a 593 late Neogene epeirogenic uplift. Stollhofen et al. (2014) also suggest that uplift could be one of the 594 causes and/or at least a contributor to other factors, such as climate.

- 595 Data from the marine realm off Namibia suggest a further step towards more extreme arid 596 conditions during the Plio-/Pleistocene transition (Fig. 6). Local SST records (ODP 1082, 597 Etourneau et al., 2009; ODP 1081, Rosell-Melé et al., 2014, Fig. 6) indicate the onset of decreasing 598 SSTs at ~2.7-2.5 Ma and the significant shift towards increased upwelling activity of colder water 599 masses in the Benguela Current since ~2.2 Ma (Dupont et al., 2005; Marlow et al., 2000; Etourneau 600 et al., 2009). The significant decrease in SSTs correlates with the further intensification of 601 Northern Hemisphere glaciation since \sim 2.7 Ma (Ruggieri et al., 2009). The pollen record (ODP 602 1082) of (Dupont, 2006) shows that arid to semi-arid biomes were rather limited prior to \sim 2.7 603 Ma, and that their concentration increases with higher variability since then, reflecting the 604 intensification of arid conditions in the Central Namib Desert (Dupont, 2006).
- 605 Vegetation change may be a major cause of the exposure of landscapes to accelerated erosion. 606 Major river incision in the Central Namib Desert thus occurred during a period of climate change 607 and greater climate variability compared to the more persistently stable Pliocene (Rosell-Melé et 608 al., 2014), with the intensification of arid conditions in southern Africa, synchronous with major 609 global changes. We therefore propose that the major river incision of the Kuiseb River at the 610 Plio-/Pleistocene transition was caused by a shift to more arid conditions with decreasing 611 precipitation, resulting in reduced river discharge, river steepening and incision (e.g. Whipple and 612 Tucker, 1999; Bonnet and Crave, 2003). Catchment and river systems such as the Kuiseb (and/or 613 river systems such as the Swakop), which had reached a steady-state during the more stable 614 Pliocene, had to adapt to the new boundary conditions, which is in line with the global increase in 615 erosion rates at the Plio-/Pleistocene transition (Herman et al., 2013; Herman and Champagnac, 616 2016). A major vegetation shift towards more arid biomes and sparser vegetation cover increased 617 the susceptibility of landscapes to erosion. The global sea-level drop at the Plio-/Pleistocene 618 transition may have had an additional impact on drainage base levels.





619The incision of the recent Kuiseb River can be constrained to a period between the derived terrace620ages of ~ 2.8 and ~ 1.3 Ma (minimum age of the Oswater bedrock river terrace). The actual period621of incision may be even shorter, given the cessation of fluvial sediment deposition offshore at622 ~ 2 Ma (Dupont et al., 2005). Deposition of the Oswater Formation indicates a phase of623aggradation sometime after ~ 1.3 Ma, followed by an incision into the recent bed of the Kuiseb624River.



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Fig. 6: Compilation of paleoclimate records. (A) Alkenone SSTs from ODP 1082 (Etourneau et al., 2009) and ODP 1081 (Rosell-Melé et al., 2014). Intensification of Benguela upwelling according to





628 Etourneau et al. (2009). (B) Sedimentation rate of ODP 1082 for the Plio/Pleistocene transition 629 (Dupont, 2006). (C) Desiccation of the Etosha paleolake from Miller et al. (2010). (D) U-Pb silcrete 630 and surface exposure ages (this study). Numbers indicate identical clasts. 1 - DWA98008-Silc3, 2 -631 DWA98008-Silc4, 3 - DWA98008-Silc7, 4 - DWA98008-Silc8, 5 - DWA98013, 6 - DWA98007, 7-DWA98006, 8 - DWA98024. (E) Global Sea-Level curve from Hansen et al. (2013). Black dashed line 632 633 indicates relative mean sea-level during the Pliocene, followed by the global decrease since the Plio-634 /Pleistocene transition. (F) Global Cenozoic reference benthic foraminifer oxygen isotope dataset 635 (CENOGRID) from Westerhold et al. (2020).

636 Conclusion

637 Our study demonstrates that microscale silcrete from the Central Namib Desert can be dated using 638 U-Pb LA-ICP-MS, and that layered silcrete incrustations can be used as paleoclimate archives. 639 LA-ICP-MS U-Pb dating of silcrete has advantages over bulk carbonate analysis because it is less 640 affected by potential interferences and contamination. The combined dating approach with 641 additional ²¹Ne exposure age dating allows us to reconstruct major paleoclimate and landscape 642 changes since the Late Miocene for the Central Namib Desert. We can corroborate previously 643 obtained chronological data from Van Der Wateren and Dunai (2001) and place absolute age 644 constraints on some sediments from the Central Namib Desert, some of which are used as marker 645 horizons throughout the region. Our chronology of groundwater calcrete formation and river incision adds crucial information with absolute dates to the 'Namib Group'. Although specific 646 647 precipitation ranges for calcrete formation are still being debated, we can assign potential 648 precipitation ranges and their shifts to specific time episodes and thus provide a semi-quantitative 649 picture of the aridification of the Central Namib Desert during the Late Miocene to the 650 Plio-/Pleistocene. Our terrestrial paleoclimate record of microscale silcrete formation, i.e., 651 calcrete formation, supports the marine evidence for a persistently stable Pliocene climate in the 652 Central Namib Desert. The cessation of groundwater calcrete formation was caused by the deep incision of the Kuiseb River (presumably synchronous with other E-W flowing drainage systems 653 654 of the Central Namib Desert) at the Plio-/Pleistocene transition, which can be explained by the 655 intensification of aridity, vegetation change, and presumably global sea-level drop. Global climate change with the onset of the Pleistocene was most likely the major forcing factor for major 656 landscape rejuvenation and change in the Central Namib Desert. Precipitation decline in the 657 658 Kuiseb River catchment is identified as the tipping point for the local climate and landscape 659 response.

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667 Author Contribution:

- 668 B.R. fieldwork, sample preparation, ²¹Ne noble gas analytic, data evaluation, manuscript writing.
- 669 R.A. U-Pb dating, sample analysis thin section, Raman, data analysis, manuscript writing. A.R.
- 670 Raman, F.M.v.d.W. fieldwork, T.J.D. fieldwork, data evaluation. A.G. data analysis. All authors
- 671 reviewed the manuscript.

672 Additional Information

- 673 **Declaration of interest:** The authors declare that the research was conducted in the absence of
- any commercial or financial relationships that could be construed as a potential conflict of interest.
- 676 **Data availability statement:** All data generated or analysed during this study are included in this
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