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Title: Radiocarbon dating reveals the timing of formation and development of pedogenic calcium carbonate concretions in Central Sudan during the Holocene

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Abstract: Calcitic soil horizons are common in arid and semi-arid lands and represent the result of the progressive accumulation of calcium carbonate in the soil profile over time. This process leads to the occurrence of several pedogenetic phases of calcium carbonate dissolution/precipitation. For this reason, timing the formation and development of calcic horizons through radiometric dating is not straightforward as time-averaging effects, due to the superimposition of the same process over time, occur. On this basis, this study aims to define the timing and dynamics of formation and development of pedogenic calcitic concretions from semi-arid Central Sudan, highlighting the relevance of a multi-disciplinary approach and the effectiveness of radiocarbon dating (coupled to an accurate sampling strategy) applied to pedogenic carbonates. Calcium carbonate-rich soil horizons (Bk) were sampled during the archaeological excavation of site 16-D-4 at Al Khiday (Central Sudan) and studied by optical, cathodoluminescence and scanning electron microscopy, as well as by chemical-physical and stable isotopes (C and O) analyses. Radiocarbon ages are obtained for distinctive calcitic pedofeatures and mostly refer to the Early Holocene for calcitic-cemented nodules (11.5, 9.9 and 9.6 cal. ka) and to the Middle Holocene for powdery calcitic-rich matrix (7.9, 7.7, 6.3 and 6.1 cal. ka) samples. Our data are also compared with the available information from detailed physiographic and palaeoenvironmental studies carried out in the region and from the study of the tight interaction between soil horizons and evidence of anthropic activities at Al Khiday site, as the archaeological record provides informative chronological constraints to interpret the ^{14}C ages obtained on selected calcitic pedofeatures. Pedogenic calcitic features dated to the Early Holocene can be related to short arid phases during general wetter climate conditions, whereas those dated to the Middle Holocene can be related to short humid phases. Results show that Bk horizons were characterized by alternate periods of calcite accumulation (precipitation) and dissolution, and periods of quiescence or extremely slow growth rates. Thus, the formation and development of Bk horizons has been a long-lasting process significantly influenced by climatic fluctuations. This study represents a further step in the comprehension of the development of calcium carbonate concretions

and (at a wider perspective) calcrete; the paper also contributes to the definition of a reliable method to radiometrically date the formation of calcitic pedofeatures. In a broader perspective, this work may offer a significant tool for archaeometric studies, where the interaction between secondary calcite and archaeological material is significant (e.g., radiocarbon dating of bioapatite in bones), and palaeoenvironmental studies in arid lands, where Bk horizon development is a function of past rainfall.

Padova, 07/05/2018

Dear Editor,

We accepted all the observations made by the Associate Editor and by two Reviewers to the manuscript:

GCA-D-17-00867

Title: Radiocarbon dating reveals the timing of formation and development of pedogenic calcium carbonate concretions in Central Sudan during the Holocene

Authors: Gregorio Dal Sasso, Andrea Zerboni, Lara Maritan, Ivana Angelini, Chiara Compostella, Donatella Usai, Gilberto Artioli

The useful comments and suggestions helped us to improve the quality of this manuscript; a detailed description of the changes made to the previous submission is reported in the "Response to Reviewers". I hope that it could be suitable for publication in *Geochimica et Cosmochimica Acta*.

Sincerely,

Gregorio Dal Sasso

Response to Reviewers

Comments from Frank McDermott, Associate Editor

Dear Dr Dal Sasso,

I have now received two reviews of your manuscript 'Radiocarbon dating of calcrete development records the steps of pedogenesis in arid lands'. Whilst one review is broadly positive, both reviewers have expressed fairly substantial concerns with various aspects of the current manuscript. Having read the manuscript and considered the reviews, I have come to the conclusion that substantial revisions will be required before the manuscript can be re-considered. After that, any revised version will have to be sent out for further reviews. An important point is that both reviewers are concerned that the scope of the paper is in fact much narrower than implied by the current rather 'all encompassing' title, a sentiment with which I concur.

- We modified the title

It is also clear that the current abstract is not adequate; it is far too general (and uninformative), and it does not contain any specific results (e.g. dates) that summarise the main conclusions.

- The abstract has been replaced with a more informative one.

There is also a concern that while the term 'calcrete' is useful, it may not be strictly applicable for the range of calcareous soil deposits described in the manuscript.

- Accordingly, we avoid this term, replaced by calcitic horizon. In any case, we noticed that Adamson et al. 1982 (suggested by Rev. #1) occasionally used the term 'calcrete' to define similar soil horizons found in a region of central Sudan adjoining to Al Khiday.

*Given the geochemistry-focus of *Geochimica et Cosmochimica Acta*, some basic geochemical data seem to be lacking in the current manuscript. Examples of missing information include the CaCO₃ content, pH and organic content of the soils and how these change as a function of depth in the profile.*

- As suggested by Rev. #2, we added the results of geochemical analyses on soil samples. We discuss in the revised version of the manuscript the significance of CaCO₃ content, pH, organic matter, total carbon, and total nitrogen (sections 3, 4.3, 5, new figure 3).

Linked to this is (a) the absence of a detailed field description and (b) interpretation of the soil profile, e.g. the reasons for the occurrence of two calcic horizons. Is the lower calcic horizon a buried soil for example? The question as to whether the depth of the calcic horizon is linked to annual rainfall in the region should also be addressed.

- We added field description of the sequences of horizons here considered (Tab. 1) and we proposed a genetic interpretation for the occurrence of two calcic horizons (section 4.1). Then we also try to correlate this evidence with the annual amount of rainfall, as suggested by Rev. #2.

On reading the manuscript I particularly missed detailed descriptions and preferably several photographs or photomicrographs showing the exact stratigraphic or micro-stratigraphic relationships between the exact dated material in the context of the profile. For example, the current sentence 'Therefore, three samples of cemented nodules (A3a-cn, 299 A3c-cn and A3d-cn) and three samples of calcitic-rich matrix (A3a-mt, A3c-mt and A3d300mt) from blocks A3a, A3c, and A3d, respectively, were selected and dated' does not convey a sense of the spatial relationships between the dated materials that I would have expected to see in such a manuscript. This absence of detailed documentation of the dated material (e.g. nodule size, internal structure and microfabrics, mineralogy, micro-stratigraphical context) with respect to the profile as a whole,

is frustrating for the reader, and makes the ^{14}C results difficult to evaluate. I would expect to see greatly improved documentation of the precise context of the dated materials in any revised manuscript.

- Accordingly, we improved sections 3 and 4.1 by adding more detailed information on the sampling points as well as in table 1 and in figure 3. In sections 4.4, 4.5, after showing the results of micromorphological analyses, we clarified the sample strategy for radiocarbon dating and stable isotopes analysis. Additionally, the new figure 7 was added to clarify the relationship between the analysed materials and the graphical representation of results was improved in the new figure 9.

Please consider these comments and those of both reviewers carefully. I hope that the reviews will be helpful in improving the manuscript. If you decide to revise your manuscript and re-submit to GCA, please be aware that the level of revision sought will require further additional reviews. You should also include a separate file documenting in detail how you addressed all of the points above, as well as those of the reviewers. Thank you for submitting your work to Geochimica et Cosmochimica Acta.

Comments from Reviewer #1

Review of Radiocarbon dating of calcrete development records the steps of pedogenesis in arid lands by Gregorio Dal Sasso, Andrea Zerboni, Lara Maritan, Ivana Angelini, Donatella Usai and Gilberto Artioli, submitted to Geochimica et Cosmochimica Acta.

General comments

Using radiocarbon dating to determine the ages of pedogenic calcium carbonate nodules and concretions in arid and semi-arid soils has always been something of a problem, and has been the subject of sustained debate by soil scientists, Quaternary geologists, geomorphologists and geochemists in North America, Australia, India and Africa for over fifty years. There are several reasons why this is so. The carbonate concretions often consist of concentric layers, precipitated at different times. The internal fabric of the concretion sometimes reveals one or more phases of partial carbonate dissolution and ensuing carbonate precipitation. Certain very hard calcareous nodules embedded in a softer calcareous matrix may have been transported by runoff or fluvial action from much older soils or sediments.

Background to the present study

The work described in this paper arose from sustained archaeological excavation at the site of El Khiday immediately west of the lower White Nile, which contains a sequence of pre-Mesolithic, Mesolithic, Neolithic and Meroitic human burials and associated cultural remains, for which there is an independent and credible chronology based on both AMS 14C and OSL ages. This means that the AMS ages obtained on the carbonate nodules can be compared to the ages already obtained for the prehistoric cultural phases, and use can be made of cross-cutting stratigraphic relationships (i.e. whether or not the burials and cultural remains of different ages pre-date or post-date phases of carbonate precipitation) to determine maximum and minimum ages of calcite precipitation.

Achievements

The authors of this manuscript have carried out geochemical, petrographic, micro-morphological and isotopic analyses (carbon, oxygen) from samples of soil carbonate and soil matrix in a test trench dug at archaeological site 16-D-4 near the village of El Khiday, located 3.5 km from the left bank of the lower White Nile and 22 km south of the Blue and White Nile confluence at Khartoum, Sudan. They also obtained 7 AMS 14C ages for authigenic carbonate exposed in the trench wall, and one AMS 14C age on a transported late Pleistocene carbonate nodule contained within a softer calcareous matrix.

The results showed several episodes of carbonate precipitation interspersed with phases of quiescence and phases of carbonate dissolution and re-precipitation, all of Holocene age. The oldest phase of carbonate precipitation pre-dated the Mesolithic burials recovered from this site and dating back to 8.5-8.0 ka. The pre-Mesolithic burials were coated in carbonate; the youngest burials (Meroitic) showed no sign of any secondary carbonate precipitation. The oldest 14C ages from cemented nodules in the lower of two calcareous horizons are 11.5, 9.9 and 9.6 ka, coeval with short arid intervals within a time of high White Nile flood levels and wetter regional climate. The youngest ages are 7.9, 7.7, 6.3 and 6.1 ka, coeval with two short humid phases. (For simplicity and clarity, I have rounded off the ages). The authors point out, correctly, that there have no doubt been multiple episodes of carbonate precipitation and carbonate recycling during the Quaternary in this region (and, indeed, in other arid regions of the world). They also note that in their study area at least six possible sources of carbonate need to be considered. The result of their work is a nuanced interpretation of the scope and limitations of using AMS 14C dating of pedogenic carbonate to help reconstruct past environmental changes.

Title

The focus of the present paper concerns a single carbonate unit at a single Holocene archaeological site

close to the left bank of the lower White Nile in central Sudan. The title is therefore somewhat misleading, because it makes no mention of geographical location, and purports to generalise for soil development in arid lands.

- As suggested we changed the title removing the term calcrete and adding chronological and geographical indications.

Some relevant previous work

In regard to central Sudan, Adamson, Williams and Gillespie (1982) discussed these same issues and the associated dating problems in a detailed chapter written nearly forty years ago, which the authors may wish to consider.

Adamson, D.A., Gillespie, R. and Williams, M.A.J. (1982). 'Palaeogeography of the Gezira and of the lower Blue and White Nile valleys.' In: *A Land between Two Niles: Quaternary Geology and Biology of the Central Sudan*, M.A.J. Williams & D.A. Adamson (eds.), Balkema, Rotterdam, pp. 165-219.

An additional and earlier paper, dealing with different modes of soil carbonate formation in this area, is: Williams, M.A.J. (1968). 'A dune catena on the clay plains of the west central Gezira, Republic of the Sudan'. *Journal of Soil Science*, 19(2), 367-378.

- We considered the papers suggested by Rev. #1 and we added specific references in the background section and in the discussion. Especially, we use soil carbonate ages published in Adamson et al. (1982) as comparison to ours (see new figure 8).

Specific comments

I am not convinced that the two calcareous horizons discussed in this work do indeed represent a single calcrete unit, as claimed, nor do I consider the term calcrete to be an appropriate one to use for what are carbonate nodules and a soft calcareous matrix. Perhaps the authors should reconsider their use of the term calcrete.

- As suggested here and by Rev. #2, we revised our hypothesis discussing the existence of two superimposed soil sequence, including two distinct calcitic horizons (Bk and 2Bk), as well as the use of the term calcrete.

References

The reference to Gatto and Zerboni (2015) is incomplete. Please specify the journal and editor of the Special Issue in which this paper appears, together with volume and page numbers.

- We added full reference of this paper.

Clarity, local importance, and wider international appeal

The paper is in general clearly written, logically structured, and well illustrated. The conclusions relating to phases of carbonate precipitation and non-precipitation are supported by the local site evidence presented, and are consistent with independent evidence of high and low White Nile levels and wetter or drier regional climatic phases. Other possible interpretations are weighed judiciously before being discounted.

This paper is a very useful contribution to the study of the geochemistry, mineralogy and stable isotopic composition of carbonate concretions within Holocene soils in the now arid lower White Nile valley. It will also be of interest to soil scientists working in the arid zones of the world.

Comments from Reviewer #2:

This paper presents interesting and important dataset on the characteristics and age estimation of a calcic soil in an archaeological site in Sudan. The findings of the paper stress several periods of pedogenesis, all within the Holocene. The paper is an important addition to our knowledge in this remote, relatively un-explored area of the world. However, it is not focused and suffers from several problems that should be accounted for before it can be accepted.

1. Abstract - The abstract needs to be re-written. As it is now, it is very general. Although the title of the paper deals with radiocarbon dating, the abstract does not include even a single age! (Beside "Holocene"). Also, do not use sentences that say nothing, like "radiocarbon ages are critically discussed....", instead, explicitly write the main results and their implications.

- The abstract has been replaced with a more informative one

2. Lack of laboratory analyses - When studying calcic soils, CaCO₃ content is of major importance and I am surprised it lacks from the paper. pH and organic matter are also important, see for example Zerboni et al 2011, Geomorphology. Add these important soil characteristics and discuss them.

- As suggested, we performed and discussed in the revised manuscript the results of several geochemical analyses: CaCO₃ content, pH, organic matter content, total carbon, total nitrogen. Methods and results are reported in the manuscript (sections 3, 4.3, 5), in table 1 and figure 3.

3. Lack of field description - I am sure that the soil profile was fully described but the full description is not included. As the entire paper lean on a single soil profile, a full description of the profile is a must. See for example Zerboni et al 2011, Geomorphology.

- As suggested by the reviewer, we added field description of soil horizons in table 1.

4. Discussion - the discussion is not focused and it was hard for me to follow it. Below are several important points that do not appear in the current version of the discussion:

** The authors found a B horizon (not defined as a calcic horizon) in between two Calcic horizons (Bk). This is not a usual scenario and deserves an explanation. In most cases we find a single Bk horizon that represents the common depth of the wetting front over large time periods. Usually, separated Bk horizons represent buried soils. Does the lower horizon (2Bk) represents a buried soil? If not, how the two separated horizons can be explained?*

- We improved the discussion section and we reconsidered our first interpretation by introducing the existence of a buried 2Bk horizon superimposed by a Bk horizon.

**In addition, the depth of the calcic horizon is known to be related to the average rainfall of the site, see Yaalon 1983; Retellack 2005, Geology; and many others. Thus, the depth of the calcic horizon according to the 130 mm average rainfall should be around 10-30 cm. How the findings of this study are related to this common relationship between rainfall and depth to calcic horizon? Can it be that the upper horizon (Bk) was eroded after it was formed?*

- In this case study the relationship between rainfall and depth of calcic horizon is not easily definable for two main reasons: the first is that the rainfall amount significantly changed over the large span of time here considered (and for which few data are available), the second is that erosional process occurred at the site. Therefore we chose to rely on $\delta^{18}\text{O}$ analysis to retrieve information on palaeoenvironmental conditions and suggest a model for calcium carbonate accumulation in the soil profile. We added few remarks on this issue in the discussion and conclusion sections.

* Related to the above mentioned points, the authors should discuss in much detail their findings in relationship to the general scheme calcic soil development (Machette 1985), considering the ages, stages, morphology, and climate.

- We add in many parts of the manuscript references to the papers by Machete and his ideas on the development of calcitic horizons.

Minor issues:

1. Introduction - There are almost no citations of classic calcic soil studies (Machette, Gile, etc.).

- References were added.

2. Table 1 - Add two more columns: Depth and horizon.

- Data was added

3. Figure 2 - Add the exact location of the soil pit in both figure 2a and figure 2b.

- The location of the soil profile was added to new figure 1b

4. Figure 3 - Add the depths to the schematic soil section on the left.

- Data was added

5. Figure 8 - Add another figure of the stable isotopes with depth.

- We added a figure (new figure 9) and table 2 with this information.

1 **Radiocarbon dating reveals the timing of formation and development of**
2 **pedogenic calcium carbonate concretions in Central Sudan during the Holocene**

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13

14 **Abstract**

15 Calcitic soil horizons are common in arid and semi-arid lands and represent the result of the
16 progressive accumulation of calcium carbonate in the soil profile over time. This process leads to
17 the occurrence of several pedogenetic phases of calcium carbonate dissolution/precipitation. For
18 this reason, timing the formation and development of calcic horizons through radiometric dating is
19 not straightforward as time-averaging effects, due to the superimposition of the same process over
20 time, occur. On this basis, this study aims to define the timing and dynamics of formation and
21 development of pedogenic calcitic concretions from semi-arid Central Sudan, highlighting the
22 relevance of a multi-disciplinary approach and the effectiveness of radiocarbon dating (coupled to
23 an accurate sampling strategy) applied to pedogenic carbonates. Calcium carbonate-rich soil
24 horizons (Bk) were sampled during the archaeological excavation of site 16-D-4 at Al Khiday
25 (Central Sudan) and studied by optical, cathodoluminescence and scanning electron microscopy, as
26 well as by chemical-physical and stable isotopes (C and O) analyses. Radiocarbon ages are obtained
27 for distinctive calcitic pedofeatures and mostly refer to the Early Holocene for calcitic-cemented
28 nodules (11.5, 9.9 and 9.6 cal. ka) and to the Middle Holocene for powdery calcitic-rich matrix (7.9,
29 7.7, 6.3 and 6.1 cal. ka) samples. Our data are also compared with the available information from
30 detailed physiographic and palaeoenvironmental studies carried out in the region and from the study
31 of the tight interaction between soil horizons and evidence of anthropic activities at Al Khiday site,
32 as the archaeological record provides informative chronological constrains to interpret the ¹⁴C ages
33 obtained on selected calcitic pedofeatures. Pedogenic calcitic features dated to the Early Holocene
34 can be related to short arid phases during general wetter climate conditions, whereas those dated to
35 the Middle Holocene can be related to short humid phases. Results show that Bk horizons were
36 characterized by alternate periods of calcite accumulation (precipitation) and dissolution, and
37 periods of quiescence or extremely slow growth rates. Thus, the formation and development of Bk
38 horizons has been a long-lasting process significantly influenced by climatic fluctuations. This
39 study represents a further step in the comprehension of the development of calcium carbonate

40 concretions and (at a wider perspective) calcrete; the paper also contributes to the definition of a
41 reliable method to radiometrically date the formation of calcitic pedofeatures. In a broader
42 perspective, this work may offer a significant tool for archaeometric studies, where the interaction
43 between secondary calcite and archaeological material is significant (e.g., radiocarbon dating of
44 bioapatite in bones), and palaeoenvironmental studies in arid lands, where Bk horizon development
45 is a function of past rainfall.

46

47 **Keywords**

48 Holocene; Calcic concretions; North Africa; Cathodoluminescence microscopy; Radiocarbon
49 dating; Pedogenesis.

50

51 **1 Introduction**

52 Calcic and petrocalcic soil horizons, and pedogenic calcretes are the results of a near-surface
53 terrestrial accumulation of predominantly calcium carbonate, resulting from the introduction and
54 cementation of calcium carbonate into a soil profile in different forms, i.e. powdery, nodular,
55 laminar, or massive (Gile et al., 1965; Machette, 1985; Wright and Tucker, 1991; Wright, 2007;
56 Durand et al., 2010); they are present in present-day soils (WRB, 2006) as much as in paleosols
57 (Mack et al., 1993; Retallack et al., 1993; Nettleton et al., 2000). These pedogenic bodies often
58 occur in areas, where vadose and shallow phreatic groundwater is saturated with respect of calcium
59 carbonate. This general definition is applied to a large variety of calcitic horizons at different stages
60 of development, abundantly occurring worldwide, at different latitudes, within different geological
61 contexts, and under different pedoclimate settings (Gile et al., 1966; Machette, 1985; McFadden
62 and Tinsley, 1985; Alonso-Zarza and Wright, 2010). However, the most evident examples of
63 calcitic soil horizons (Bk) developed in arid and semi-arid regions (Gile et al., 1966; Adamson et
64 al., 1982; Machette, 1985). General assumption is that calcrete develops in soils, characterized by a
65 net moisture deficit, in which the circulating fluids (vadose and shallow phreatic groundwater) are
66 saturated in calcium carbonate. Calcite accumulation is promoted by several physical-chemical
67 processes as well as by biological-mediated processes (Wright et al., 1988; Verrecchia and
68 Verrecchia, 1994; Cailleau et al., 2011), which, in turn, are influenced by climatic conditions, in
69 particular rainfall amount. As a consequence, calcrete is not generally the result of a single
70 pedogenetic phase, but it corresponds to subsequent events, eventually separated by a long time
71 span, during which previously precipitated carbonates undergo a partial or complete new
72 dissolution and then re-precipitation. Calcic horizon and calcrete development, therefore, consists of
73 several, superimposed generations of micritic and/or sparitic calcium carbonate crystals. For this
74 reason, and considering also that a continuous opening of the system hampers the possibility to date
75 calcite with U/Th and ^{14}C , it is hard to define accurately the time and steps of calcrete pedogenesis.

76 Almost thirty-five years ago, Adamson et al. (1982) have carried out a huge campaign of
77 radiocarbon dating of calcitic nodules in soils from Central Sudan, highlighting the occurrence of
78 reworked carbonates and subsequent recrystallizations. For that reason and for the the large
79 occurrence of carbonate pedofeatures, soils/paleosoils of arid Sudan are an exceptional case study to
80 investigate the chronological development of carbonatic concretions. Therefore, in this research a
81 multi-analytical approach, consisting in mineralogical, micromorphological, geochemical, and
82 geochronological analyses, is applied to the study of calcitic pedogenic features from Central
83 Sudan, in order to contribute towards the understanding of dynamics and timing of their formation
84 and development. Carbonate-bearing soil horizons analysed in this study formed in a localised area
85 along the left bank of the White Nile, few km south of the Khartoum-Omdurman urban system (Fig.
86 1), where a large, multiphase archaeological site (Al-Khiday, currently under excavation) is in tight
87 relation with the carbonate concretions. In fact, part of the vestiges of the site is embedded in the
88 calcitic crust (Fig. 2a), whereas other features are clearly cut in the pre-existing carbonate-bearing
89 horizon (Fig. 2b and 2c). Detailed studies on the local physiographic and palaeoenvironmental
90 aspects (Zerboni, 2011; Williams et al., 2015), as well as on the anthropic activities, considering the
91 wide span of time covered by the archaeological evidence (almost the whole Holocene) (Usai and
92 Salvatori, 2002; Usai, 2003; Usai and Salvatori, 2005; Salvatori and Usai, 2009; Usai et al., 2010;
93 Salvatori et al., 2011; Salvatori, 2012; Dal Sasso et al., 2014a; Jakob, 2014; Salvatori et al., 2014;
94 Dal Sasso et al., 2014b; Usai et al., 2014; Dal Sasso et al., 2016; Iacumin et al., 2016; Usai et al.,
95 2017; Maritan et al., 2018), supply important information to reconstruct the timing and steps of
96 calcrete formation. In our study, results from the minero-petrographic and pedogenetic study of soil
97 samples, coupled with additional archaeological and palaeoenvironmental information, allowed a
98 more complete understanding of the processes involved in calcitic horizons and calcrete formation,
99 recycling, and development. Moreover, since these processes are strongly dependent on water
100 availability, which, in turn, depends on climatic conditions, our results are discussed taking into

101 account the regional Holocene variations in water availability, as reconstructed by several
102 palaeoenvironmental studies.

103

104 **2 Study area, palaeoenvironmental settings, and archaeological background**

105 A complex calcrete horizon has been found during the archaeological survey and excavation of
106 several archaeological sites (16-D-3, 16-D-4, 16-D-4b, 16-D-5, 16-D-6) and the geomorphological
107 survey of the region carried out since 2006 within the “El Salha Archaeological Project” (Usai and
108 Salvatori, 2002; Usai, 2003; Usai and Salvatori, 2005; Salvatori and Usai, 2009; Usai et al., 2010;
109 Salvatori et al., 2011; Salvatori, 2012). The archaeological sites are located on the western bank of
110 the White Nile, near the Al Khiday village (Central Sudan), at 3.5 km from the present-day river
111 course and about 22 Km south of its confluence with the Blue Nile (Fig. 1a). They are set on fluvial
112 sandy ridges, corresponding to the remnants of late Pleistocene longitudinal river bars, at the limit
113 of alluvial sediments deposited during the very late Quaternary flooding of the White Nile (Zerboni,
114 2011; Williams et al., 2015). These fluvial ridges and the surrounding flat areas are weakly
115 weathered by Holocene pedogenesis, despite the intense accumulation of pedogenic calcium
116 carbonate (Zerboni, 2011), thus the poorly developed local soils can be defined as aridisols
117 (Buursink, 1971).

118 The archaeological excavation and geomorphological investigation (Usai et al., 2010; Salvatori et
119 al., 2011; Zerboni, 2011) revealed a complex stratigraphy at these archaeological sites, resulting
120 from anthropic activities, sedimentary, erosional, and pedogenic processes occurring along a wide
121 span of time. The area had been inhabited several times along almost the entire Holocene. Focusing
122 on the 16-D-4 site (Fig. 1b), a Mesolithic specialised use of the area was defined by the numerous
123 pits (more than 100) that have been excavated, characterized by different types of filling material
124 and presumably with different functions (Zerboni, 2011), and radiocarbon dated to 8650-8250 cal.
125 BP (Salvatori et al., 2011). In addition, the site was used several times as a burial ground: at least
126 three different burial phases have been identified. The most ancient one, named pre-Mesolithic, is

127 characterized by individuals (at least 90) buried in a prone and elongated position. The chronology
128 of this burial phase is uncertain but constrained by the Mesolithic use of the area, as a number of
129 skeletons (16) were found to be cut by radiocarbon-dated Mesolithic pits (Salvatori et al., 2011),
130 which indicate that the pre-Mesolithic burial phase is unambiguously older than 8650 cal. BP. The
131 site was subsequently used as a cemetery during the Neolithic period (38 graves; 6500-6200 cal.
132 BP), and later on during the Meroitic period (43 graves, 2100-1800 cal. BP) (Usai et al., 2010;
133 Salvatori et al., 2011).

134 Soils in Central Sudan can be classified as Vertisols, Entisols, Aridisols, and Alfisols (Buursink,
135 1971) in the study region, the first three categories are the most represented and Aridisols are those
136 most recurrent at Al Khiday. In the whole region, pedogenic processes lead to the accumulation of
137 calcic pedofeatures, which have been described previously by several authors (Blokhuis et al., 1968;
138 Williams, 1968; Buursink, 1971; Adamson et al., 1982). Buursink (1971) in his classification of soil
139 of Central Sudan describes several petrocalcic horizons, rich in CaCO₃ nodules and/or concretions,
140 similar to the K horizons defined by Gile et al. (1966). The same pedofeatures have been reported
141 by Blokhuis et al., (1964) and Blokhuis et al. (1968), who also described the properties and
142 distribution of pedogenic carbonate in Vertisols. Soils and paleosoils with common to abundant
143 CaCO₃ concretions have been reported also by Williams (1968) along the White Nile.

144 Today, the climate of the region is arid, with mean annual temperature ranging from 22°C to 33°C
145 and average rainfall of ca. 130 mm (El-Tom, 1975; Elagib and Mansell, 2000). However, climatic
146 conditions were significantly different in the past and substantial climatic changes occurred at local
147 and regional levels (Williams and Adamson, 1980; Gasse, 2000; Nicoll, 2001; Nicoll, 2004;
148 Williams, 2009; Williams and Jacobsen, 2011; Zerboni, 2013; Gatto and Zerboni, 2015) during the
149 wide time span of the site use (from the early Holocene to the beginning of the 1st millennium AD).
150 In the early Holocene (11500-8000 cal. BP) the precipitation rate, and consequently also the
151 environmental humidity, were substantially higher, as well as the flooding level of the White Nile,
152 due to a northward expansion of the Indian monsoon domain (Gasse, 2000; Williams, 2009). The

153 formation of seasonal swamps in the surrounding area of the Al Khiday sites during the Mesolithic
154 occupation of the area is attested by geoarchaeological and geomorphological evidence (Williams
155 and Jacobsen, 2011; Zerboni, 2011). Since the middle Holocene (8000-3600 cal. BP), the
156 progressive weakening of monsoon intensity led to rainfall decrease and overall reduction of water
157 availability. In the late Holocene (since ca. 3600 cal. BP), since the Meroitic period and later on,
158 progressively drier environmental conditions occurred, up to the arid climate that now characterizes
159 central Sudan; only short-timed wetter events have been occasionally recorded (Mawson and
160 Williams, 1984). In addition to this general trend, palaeoenvironmental records identified several
161 periods of rapid climatic changes during the early Holocene wet phase, when several short arid
162 periods occurred (Williams, 2009).

163

164 **3 Materials and methods**

165 The samples considered in this research come from a test trench, exposed in the sandy deposit at Al
166 Khiday and representing the local substrate, brought to light during the excavation of a Mesolithic
167 pit (pit number 151A). The section shows a soil profile with at least 0.7 m of fine to coarse sand
168 (occasionally lenses of pebbles are present), deeply cemented by calcium carbonate. Horizons
169 description is in Tab. 1. Six sampling points, namely A1, A2, A3a, A3b, A3c, and A3d, were
170 selected at different depths from the top to the bottom of the section (see Tab. 1 and Fig. 3 for
171 details). At each sampling point, samples were collected and prepared as required for the analytical
172 techniques applied in this study. Samples were embedded under vacuum in epoxy resin (Araldite
173 2020) and prepared in petrographic thin sections (3x5 cm). Micromorphological analyses were
174 carried out by petrographic optical microscopy (OM), cathodoluminescence microscopy (CL), and
175 scanning electron microscopy (SEM).

176 OM analysis was performed with a petrographic microscope (Nikon Eclipse E660) under plane-
177 polarized (PPL) and cross-polarized light (XPL). CL analysis was performed with a petrographic
178 microscope (NIKON Labophot2-POL) equipped with a cold cathode stage (CL8200 MK3,

179 Cambridge Image Technology Ltd) and operating at 15 kV and 200 mA. SEM analysis was carried
180 out with a CamScan MX 2500 coupled to a detector for Energy dispersive X-ray spectroscopy
181 (EDS), equipped with a LaB6 cathode and operating at 20 kV and 160 nA. Thin-sections were
182 described following the terminology proposed by Bullock et al. (1985) and Stoops (2003), and the
183 interpretation mostly followed the concepts discussed in Stoops et al. (2010).

184 Samples adjoining those prepared in thin sections were selected for chemical and stable isotopes
185 analyses and radiocarbon dating. The chemical characterization of samples was performed
186 measuring CaCO₃ equivalents, pH, humified carbon content, total organic carbon, total carbon
187 content (TC) and nitrogen content (TN). Calcium carbonate equivalents were chemically performed
188 using a Dietrich–Frühling calcimeter, which measures the volume of CO₂ developed by acid
189 reacting with the bulk sample, which is proportional to the carbonate concentration. Humified
190 organic carbon was identified by means of the Walkley and Black (1934) method, using chromic
191 acid to measure the oxidizable organic carbon (titration). Total organic carbon was estimated by
192 loss on ignition - LOI - (Heiri et al., 2001); samples were air-dried (at 105°C) and organic matter
193 was oxidized at 500–550°C to carbon dioxide and ash, then the weight lost during the reaction was
194 measured by weighing the samples before and after heating. The analysis of total carbon and total
195 nitrogen were performed on dried soil samples with a Thermo Fisher Scientific Organic Elemental
196 Analyzer (OEA-Flash 2000). Samples were weighed out into tin containers to the nearest 0.001mg
197 on a Gibertini MICRO1000 electronic microbalance. Each sample was then flash-combusted at
198 1800°C in the OEA.

199 After a gentle disaggregation in an agate mortar, distinctive calcitic pedofeatures, described by thin
200 section micromorphological analyses, were identified on massive samples and accurately separated
201 by hand-picking under the stereoscopic microscope. Each pedofeature was then analysed for stable
202 carbon and oxygen isotopes and radiocarbon dated.

203 Stable isotopes analyses were carried out with a Thermo Scientific Delta V Advantage Isotope
204 Ratio Mass Spectrometer. CO₂ was developed at 70 °C by complete reaction with >99% crystalline

205 H₃PO₄ in a Gasbench II device connected to the spectrometer. Results were calibrated with two
206 internal standards (sieved Carrara marble and Millipore Suprapur carbonate), periodically calibrated
207 against the international reference carbonates NBS 19, NBS 18 and L-SVEC. A control standard
208 (sieved Monzoni marble) was also measured and reproduced with external errors of better than
209 0.1‰ (1σ) for both carbon and oxygen.

210 Radiocarbon dating by accelerated mass spectrometry (AMS-¹⁴C) was carried out at the Center for
211 Isotopic Research on the Cultural and Environmental heritage (CIRCE) laboratory (Terrasi et al.,
212 2008) of the Second University of Naples. All AMS-¹⁴C ages were calibrated according to the
213 INTCAL13 dataset (Reimer et al., 2013) using the CalPal software (Weninger and Jöris, 2008). The
214 method of multiple-group calibration was used to graphically represent and compare calibrated
215 AMS-¹⁴C ages (Weninger, 1986).

216

217 **4 Results**

218 **4.1 Field description**

219 The soil profile, here studied, corresponds to the section of the wall of the pit 151A, an
220 archaeological feature related to the Mesolithic use of the site (8650-8250 cal. BP (Salvatori et al.,
221 2011; Salvatori, 2012)), excavated on top of a fluvial sand ridge. In the area of Al Khiday,
222 pedosedimentary sequences generally correspond to poorly weathered soils interlayered to sandy to
223 silty sediments, eventually disturbed by anthropic activities (Buursink, 1971); the sequences are
224 interpreted as fluvial deposits related to an Upper Pleistocene higher level of the White Nile
225 (Zerboni, 2011). The general aspect of a typical soil profile consists of a sandy to silty top horizon,
226 enriched of calcium carbonate in form of nodules (Bk) horizon, lying on a BC horizon. An abrupt
227 limit (possibly erosional) marks the transition to the following 2Bk horizon , which consists of
228 medium to coarse sand, sometimes including lenses of coarser grains (very coarse sand to small
229 pebbles), deeply cemented by calcium carbonate; this buried petrocalcic horizon can be interpreted
230 as a former calcrete. Beneath, there is a sequence of fluvial to lacustrine fine sediments (silt and

231 sand), with few CaCO₃ nodules in the upper part (for a complete description see Williams et al.,
232 2015).

233 At the sampling point, the pedostratigraphy is very clear (Fig. 3): two distinct calcium carbonate-
234 rich horizons, separated by a sandy layer, can be observed. The uppermost Bk horizon (sampling
235 point A1) is constituted by quartz sand slightly cemented by calcium carbonate; calcite
236 concentration within the sediments is detected as coalescent irregular calcitic nodules of few
237 centimetres in diameter. A weakly cemented sandy BC horizon (sampling point A2) is interposed
238 between the first Bk horizon and a second concentration of calcium carbonate, corresponding to the
239 lower part of the section (sampling points A3a, A3b, A3c and A3d). This deepest calcitic horizon
240 (2Bk), up to 0.7 m thick, is nodular and can be interpreted as an early stage of calcrete development
241 in soil profile (Machette, 1985; Wright, 2007), resulting from the illuvial concentration of calcium
242 carbonate in a siliciclastic host material (quartz sand and silt). It is formed by cemented quartz sand
243 and carbonate concretions, where calcitic nodules can be distinguished. Abundance of carbonate
244 concretions as well as cementation increases with depth. From the pedological point of view, this
245 evidence represents a sequence of distinct calcitic horizons on distinct parent materials, probably
246 resulting from a discontinuous pedogenetic process and alternated sedimentary events.

247 The Bk horizon, as observed in the archaeological area and in naturally exposed sections is at about
248 20-50 cm depth from the current surface level. In correspondence of site 16-D-4 the close
249 relationship between calcitic pedofeatures and archaeological features of different ages can provide
250 further evidence on the timing of calcium carbonate development. Calcitic concretions partially
251 embed human bones belonging to the pre-Mesolithic burial phase, the most ancient attested at the
252 site (Fig. 2a), whereas Mesolithic pits (Fig. 2b) and Meroitic graves (Fig. 2c) are cut into it.
253 Nevertheless, pre-Mesolithic and Neolithic bones were found to be permeated by secondary calcite,
254 whereas the Meroitic bones, belonging to the most recent burial phase were not affected by
255 pedogenetic precipitation of calcite (Dal Sasso et al., 2014b; Dal Sasso et al., 2016; Dal Sasso et al.,
256 2018). In addition, the lower part of most of the Mesolithic pits is characterized by filling material

257 cemented by calcium carbonate; in particular, some pits, used as fireplaces, present a significant
258 amount of whitish and hard calcitic concretion encrusting the archaeological materials. The same
259 evidence of calcitic concretions embedding human and animal remains in archaeological contexts
260 has been described elsewhere in Sudan, as on the Singa calvaria (McDermott et al., 1996), at Abu
261 Hugar (Whiteman, 1971), and elsewhere in the Gezira (Adamson et al., 1982). The origin of this
262 calcitic cement can be associated to the processes involved in calcrete development; however, the
263 rather high level of cementation must imply a contribution to the total amount of calcite coming
264 from the recrystallization of the abundant remains of Ca-rich wood ash (Canti, 2003) originally
265 constituting the upper part of the pit infilling (Zerboni, 2011).

266

267 **4.2 Micromorphology of thin sections**

268 Thin sections from the upper part of the soil profile (A1 and A2 samples) show a groundmass with
269 abundant sand- and silt-sized quartz grains, sub-rounded to sub-angular in shape, with a bimodal
270 grain-size distribution. The authigenic calcite is heterogeneously distributed (in shape of nodules,
271 coatings, and infillings) among the sections giving to the slides a single- to double-spaced porphyric
272 c/f (coarse/fine) related distribution, locally open porphyric or gefuric. In the uppermost Bk horizon,
273 areas corresponding to typical calcitic nodules, macroscopically identified, show a micromass
274 mainly constituted by micrite and microsparite (Fig. 4a), much more abundant with respect to that
275 observed for other portions of the poorly cemented samples (Fig. 4c). Allochthonous, well-rounded
276 calcitic pedorelicts (Fig. 4b, 5c, 5d), comparable in size with sandy quartz grains, are frequent. They
277 are constituted by a micritic matrix stained by Mn and Fe oxides/hydroxides, and contain veins of
278 sparitic to micro-sparitic calcite, generally occurring within this kind of pedofeatures (Fig. 5c, 5d).

279 Samples from the upper units also evidenced the occasional occurrence of wood ash (calcite
280 pseudomorphs after calcium oxalate crystals (Canti, 2003)), fish bone fragments and small
281 charcoals (Fig. 4d). Anthropogenic features were observed in small localised areas of the thin
282 section (Fig. 4d), and possibly migrated from the infilling of the nearby Mesolithic pit, due to

283 intense bioturbation (Zerboni, 2011). SEM analysis on calcitic nodules from the Bk horizon
284 revealed that the microsparitic groundmass is heterogeneous in terms of compositions and
285 cementation degree: some areas are characterized by densely packed calcite crystals (Fig. 6a) with
286 respect of other areas, where a loose calcite crystals distribution is observed (Fig. 6c). In other
287 areas, calcite crystals occur within a clayey matrix (Fig. 6b). The great heterogeneity in terms of
288 calcite crystals distribution is detected also by CL, where the microsparitic micromass appears with
289 an orange luminescence, whose intensity is proportional to the density of calcite crystals (Fig. 5a).
290 Poorly cemented areas are characterized by low luminescence due to scarcity or lack of calcite in
291 the matrix (Fig 5b).

292 Thin sections from the lower 2Bk horizon (samples A3a, A3b, A3c, A3d) show areas characterized
293 by slightly cemented quartz grains, sub-rounded to sub-angular in shape, and sand- and silt-sized,
294 embedded into a micritic micromass (Fig. 4e). The distribution of mineral grains and calcite
295 generally results in a single spaced to open porphyric, occasionally gefuric, c/f related distribution.
296 Conversely, highly carbonated areas (Fig. 4f, 4g), corresponding to older calcitic nodules embedded
297 in the micritic micromass, are predominantly constituted by microsparitic calcite, and quartz sand
298 grains are extremely rare. Different textural pedofeatures were identified within calcitic nodules:
299 some portions are characterized by well-cemented calcite crystals forming hard nodules (hereafter
300 called cemented nodules) of sub-rounded to sub-angular shape, identifiable by OM (Fig. 4f, 4g), CL
301 (Fig. 5e) and SEM (Fig 6e, 6f) analyses. These features (Fig. 7) are embedded in a loose matrix
302 mainly constituted by calcite crystals similar in size, but less packed than in the cemented nodules,
303 sometimes associated to clay minerals. Towards the bottom of the 2Bk horizon, the dimension of
304 calcitic-cemented nodules decrease and their margins, in contact with the surrounding matrix, are
305 less defined (Fig. 5f). CL analysis indicates a significant, but not systematic, luminescence
306 variations of calcite crystals along the section, both dispersed and well packed. In some cases,
307 homogeneously luminescent crystals were observed (Fig. 5g), whereas in others the innermost part
308 of crystals is less luminescent with respect to their margins and inter-grain cement (Fig. 5h). OM,

309 CL, and especially SEM images show an overall increase of calcite crystal size from the upper to
310 the lower Bk horizon. Allochthonous calcitic pedorelicts, sub-rounded in shape and permeated by
311 Fe oxides/hydroxides (different from the Mn and Fe-bearing oxides/hydroxides permeating the
312 rounded grains identified in the upper unit) were observed in unit A3c and, less frequently, in unit
313 A3d. Both the two types of pedorelicts show a high concentration of oxides near their margins and a
314 thin layer of clay minerals coating the outer surface (Fig. 4h, 6d).

315

316 **4.3 Chemical and physical analyses**

317 Tab. 1 and Fig. 3 illustrate the results of chemical analyses on soil samples. Calcium carbonate
318 equivalent content and pH show similar trends, being the latter generally basic and buffered by the
319 high content of carbonates; the B horizon is the less rich in carbonate. Humified organic carbon
320 contents show trends antithetic of CaCO₃ content and the highest peak in organics (B horizon)
321 corresponds to the less basic horizon. LOI result indicates a higher content of organics in the
322 lowermost part, possibly suggesting a lower preservation of organic matter in the uppermost part of
323 the sequence, where in fact humified carbon is more abundant. TC content is mainly ruled by the
324 occurrence of calcium carbonate, whereas TN variations correspond to the variation of the humified
325 carbon, confirming that organic matter is the main source of nitrogen.

326

327 **4.4 AMS-¹⁴C dating**

328 Due to the lack of well identifiable organics in the soil matrix, AMS-¹⁴C radiocarbon dating was
329 performed on different calcium carbonate-bearing pedofeatures, identified by micro-morphological
330 analysis, and mechanically selected under the stereoscopic microscope. As for the uppermost Bk
331 horizon (A1 sampling point, Fig. 3), calcitic nodules, constituted by micritic/microsparitic
332 micromass (as shown in Fig. 4a) were identified in the massive sample as powdery aggregates, up
333 to 1 cm in size. A fraction of micritic/microsparitic micromass was selected for radiocarbon dating

334 (A1-mt samples). Allochthonous pedorelicts (A1-pedorelict) were also selected from the same
335 sampling point (A1) and dated.

336 In the lower 2Bk horizon, micromorphological analyses of calcitic nodules (up to 2 cm in size)
337 showed the occurrence of different textural pedofeatures, previously addressed as cemented nodules
338 and loose calcitic-rich matrix (as shown in Fig. 4f, 4g and 5e). In Fig. 7, OM photomicrographs and
339 the segmented false-colour image show a representative example of the relationship between these
340 pedofeatures. On the massive samples, cemented nodules were found to be much more lithified than
341 the surrounding loose calcitic-rich matrix, therefore, after a gentle disaggregation of the samples,
342 these two adjoining pedofeatures were effectively separated by hand-picking under a stereoscopic
343 microscope. This sampling strategy was then applied to massive samples collected at A3a, A3c and
344 A3d points (Fig. 3), so that for each of them cemented nodules (labelled as A3a-cn, A3c-cn and
345 A3d-cn) and calcitic-rich matrix (labelled as A3a-mt, A3c-mt and A3d-mt) samples were selected
346 for radiocarbon dating. As expected, an old radiocarbon age, 29540 ± 440 cal. years BP, was
347 obtained for the allochthonous pedorelicts (A1-pedorelict). The cemented nodules from A3a, A3c
348 and A3d sampling points show older ages (9620 ± 100 , 9900 ± 580 and 11480 ± 240 cal. years BP
349 for A3a-cn, A3c-cn and A3d-cn, respectively) with respect to the nearby loose calcitic-rich matrix
350 (6340 ± 80 , 7920 ± 100 and 7730 ± 80 cal. years BP for A3a-mt, A3c-mt and A3d-mt, respectively)
351 and from the calcitic nodules from A1 sampling point (6080 ± 240 cal. years BP) (Tab. 2, Fig. 8a,
352 9a).

353 Apart from a single radiocarbon age dating to the Upper Pleistocene (for the allochthonous
354 pedorelict), the radiocarbon dates are Holocene in age and can be tentatively clustered to the Early
355 and Middle-Holocene. Statistical treatment of dating results can better delineate the distribution of
356 radiocarbon ages on the sampled calcitic horizons. The method of multiple-group calibration
357 (Weninger, 1986; Weninger and Jöris, 2008) was used to represent radiocarbon ages, in order to
358 discern if close radiocarbon ages might refer to the same or different pedogenetic phases as well as
359 to ease the comparison with other regional radiocarbon age datasets. Our AMS- ^{14}C ages on

360 carbonates were compared (Fig. 7) with the ^{14}C ages obtained on calcium carbonate deposits in soils
361 from Central Sudan (Adamson et al., 1982), and on fossil shells (Williams, 2009; Williams et al.,
362 2015) The latter ages indicates the late Quaternary high flow levels of the White Nile, representing
363 a proxy for wetter climatic conditions. In Fig. 7 the radiometric chronology of the Holocene human
364 occupation of the Al Khiday archaeological sites (Salvatori et al., 2011) is also reported. A further,
365 indirect piece of the chronological puzzle of calcrete development at Al Khiday is represented by
366 the radiometric dating of the archaeological structures cutting the carbonatic crusts; most of them
367 belong to the Mesolithic phase and date to the 8650-8250 cal years BP.

368

369 **4.5 Stable isotopes analysis**

370 C and O stable isotope analyses were carried out on an aliquot from the same samples that were
371 radiocarbon dated; moreover, additional samples of calcitic nodules from A1 and cemented nodules
372 (cn) and calcitic-rich matrix (mt) from A3a, A3b, A3c and A3d sampling points (Fig. 3) were
373 separately selected and analysed (Tab. 2). Overall $\delta^{18}\text{O}$ ranges between -8.27‰ and -1.85‰ (V-
374 PDB) and $\delta^{13}\text{C}$ between -1.48‰ and 1.06‰ (V-PDB). $\delta^{13}\text{C}$ values show a narrow range of variation
375 and no significant differences were observed between cemented nodule and matrix samples.
376 Conversely, a wider range of variation was observed for $\delta^{18}\text{O}$, with matrix samples showing more
377 negative values with respect to cemented nodules (Tab. 2, Fig. 9b, 9c). A good consistency was
378 observed between results obtained from the same samples double-measured, in particular for those
379 from the matrix samples.

380

381 **5 Discussion**

382 The authigenic accumulation of calcium carbonate in siliciclastic host sediments, composed of
383 quartz sand lacking in calcite, indicates an external source for calcium carbonate. Conceivable
384 sources may be identified in the geological bedrock of the area, as the limited quantity of calcite,
385 occurring in the local sandstone and shale, is firstly dissolved, then mobilized and finally re-

386 precipitated thanks to circulating pore water. Further significant sources of carbonate may be
387 represented by alkaline atmospheric dust and calcium carbonate precipitated over a long period in
388 the Pleistocene lake sediments identified in the region (Williams et al., 2015). Similar sources of
389 carbonates have been claimed by Cremaschi et al. (2010) to explain the origin of spring calcareous
390 tufa in a sandstone-bearing central Saharan massif. Less important contributions are likely those
391 from the Nile overbank sediments, and from the anthropogenic calcitic ash related to the Mesolithic
392 occupation of the site, the latter being available only after the onset of pedogenetic processes related
393 to the formation of calcrete. Moreover, the dissolution and mobilization of calcium carbonate
394 accumulated in pre-existent Pleistocene Fe-enriched paleosols, which are highly cemented and
395 heavily affected by pedogenesis (Fig. 2d), may have contributed to the formation of the calcrete
396 horizon at 16D4 site. However, the origin of calcium carbonate in these paleosols is not well-
397 defined and its accumulation likely results from long-lasting processes of dissolution/reprecipitation
398 of calcite, possibly mediated by biological activity. Evidence of this type of paleosols, widely
399 outcropping in the region (Zerboni et al., 2016), can be also found few hundred meters west from
400 the 16D4 site. These Pleistocene paleosols are also the most probable source of the pedorelicts
401 identified in the calcrete section (those separated from sample A1 and dated to the 29540 ± 440 cal.
402 years BP). In fact, they probably derive from surface transportation, as suggested by their sub-
403 rounded to rounded shape and by the occurrence of clayey material coating their external surface,
404 which can be interpreted as a rolled pedofeature (Zerboni, 2011). Also Adamson et al. (1982)
405 obtained Upper Pleistocene dating on rolled carbonates, suggesting their single or multistep
406 reworking of older carbonate-impregnated clays and silts from former soils.

407 Calcitic nodules formed since the accumulation of microcrystalline calcite; the small size of crystals
408 suggests a relatively fast precipitation of calcite, associated with high rates of evapotranspiration. In
409 this case, the contribution of microbially induced calcite precipitation is assumed to be minimal, as
410 micromorphological features characteristic of biological activity (Newman et al., 1997; Richter et
411 al., 2008) are not observed. The upper Bk horizon is characterized by weak cementation and

412 nodules are formed by microcrystalline calcite precipitated in the porosity between sand- and silt-
413 sized quartz grains. The lower 2Bk horizon shows a significant increase in the accumulation of
414 calcium carbonate and the formation of larger concretions. The occurrence of very few quartz
415 grains much more dispersed in the microsparitic groundmass within nodules, with respect to those
416 observed in less cemented portions of the sample, may indicate a displacive calcite crystal growth
417 (Armenteros, 2010). Moreover, higher permeating cementation of sediments was observed in this
418 unit, thus representing subsequent development stages of the calcitic horizon. However, features
419 characteristic of successive stages of calcite precipitation/recrystallization observed in well-
420 developed calcrete profiles (Wright, 2007) are lacking in our case study, thus suggesting an
421 intermediate development stage for calcitic horizons. At Al Khiday, Bk and 2Bk horizons can be
422 described as nodular calcitic horizons as suggested by Alonso-Zarza and Wright (Alonso-Zarza and
423 Wright, 2010), or at least comparable to a Stage III of calcrete development (Machette, 1985).

424 Cathodoluminescence of carbonates, in terms of colours and intensities, is mainly attributed to the
425 occurrence of Mn^{2+} and Fe^{2+} ions in trace quantity in the calcite crystal structure, which, in turn, is
426 related to geochemical conditions during precipitation. Therefore, even if only qualitatively,
427 luminescence variations of calcite crystals observed in CL images indicate variations in the
428 chemical composition and/or redox conditions during precipitation and subsequent recrystallization
429 (Hiatt and Pufahl, 2014). These data, coupled with information on the diachronic interaction
430 between calcrete horizon and archaeological records, indicate that calcrete formation and
431 subsequent development is a long-lasting and discontinuous multi-step process, acting at least since
432 the Early Holocene. Notwithstanding the evidence of calcite accumulation in Late Pleistocene
433 fluvial sediments, the widespread occurrence of calcium carbonate features in paleosols and
434 sediments of the region, may suggest that environmental conditions suitable for calcite
435 mobilization, redistribution and precipitation in soils may have occurred many times since the
436 Pleistocene, as described also elsewhere in North Africa (Szabo et al., 1995; Brookes, 2010;
437 Zerboni et al., 2011). Yet, Adamson et al. (1982) suggest multiple events of carbonate dissolution

438 and precipitations in soil and sediments of Central Sudan since the Late Pleistocene and probably
439 earlier.

440 On these bases, timing the formation and development of carbonate concretions in the soil profile
441 by radiometric dating techniques requires an accurate sampling strategy and a careful evaluation of
442 results. Radiocarbon dating of calcrete and recrystallization-prone carbonates is not a routinely
443 performed type of analysis, even if some studies are reported in literature (Wang et al., 1996; Geyh
444 and Eitel, 1998; Deutz et al., 2002; Geyh and Thiedig, 2008; Vogel and Geyh, 2008; Achyuthan et
445 al., 2010). In fact, AMS-¹⁴C dating of calcite forming calcrete horizons might not lead to a
446 straightforward interpretation of results. When calcite precipitates, its ¹⁴C activity is in equilibrium
447 to that of circulating pore water, which, in turn, may not be in equilibrium with atmospheric CO₂
448 due to the so defined “reservoir effect” (Geyh and Eitel, 1998). This can cause an overestimation of
449 the actual age of calcium carbonate. Moreover, when dissolution and recrystallization occurs, ¹⁴C
450 activity of former calcite is then superimposed to that of the new-generated calcite, in equilibrium
451 with pore water circulating at that time (Geyh and Eitel, 1998). Notwithstanding that, Adamson et
452 al. (1982) conclude that Sudanese soil carbonates are fairly suitable materials for dating. They are
453 also aware of the ineluctability of contamination from some of the older carbonates in some cases;
454 we may confirm this and highlight also the possibility of contamination from younger calcite
455 precipitated after the climate-driven re-opening of the system.

456 Despite these difficulties, in our case study, the well-defined geomorphological,
457 palaeoenvironmental, and archaeological contexts provide valuable information to estimate the
458 consistency of radiocarbon ages on calcrete samples. Moreover, some of these issues can be
459 overcome by the very low amount (~ 10 mg) of sample required by AMS-¹⁴C dating, enabling one
460 to apply an accurate sampling strategy on small portions of the sample, thus separately analysing
461 calcite precipitated in a single pedogenetic event. In this case, the reservoir effect is assumed to be
462 minimal, since circulating pore water is more conceivably provided by the White Nile flooding and,
463 during the Early Holocene, by rainwater rather than fossil groundwater (Dee et al., 2010). In

464 addition to the superficial location of the calcitic horizons, the high values of $\delta^{13}\text{C}$, measured both
465 on cemented nodule and matrix samples, suggest the predominant contribution of atmospheric CO_2
466 as a source of CO_2 to the groundwater, whereas the contribution of plant-sourced CO_2 can be
467 considered minimal, thus suggesting a sparse vegetation cover (Cerling, 1984; Burns and Matter,
468 1995; Deutz et al., 2001) during the phases of precipitation.

469 The main issue that has to be taken into account when dealing with radiocarbon dating on
470 pedogenic calcite is the overprinting effect, which is not negligible (Deutz et al., 2002). Calcitic
471 horizons and calcretes form over time as results of subsequent dissolution and reprecipitation
472 processes (e.g., Machette, 1985), thus ^{14}C activity is time averaged and determination of the time of
473 actual onset of calcite precipitation may be hazardous. As a matter of fact, soil carbonates can be
474 reasonably considered an open system, implying environmental conditions that promote loops of
475 calcite dissolution and reprecipitation. However, when changes in environmental conditions (mostly
476 humidity) lead to the interruption of carbonate accumulation, the closed system can be
477 hypothesised. Therefore, radiocarbon ages of calcrete samples reasonably indicate the last stage of
478 calcite recrystallization; this may provide valuable information on the timing of Bk horizons
479 development. For these reasons, radiocarbon ages cannot be used according to sample stratigraphy
480 to determine a linear sequence of depositional events; this is also obvious considering the
481 complexity of this nonlinear and multistep process (Wright, 2007). However, micro-sampling
482 should minimize the artificial homogenization of different generations of calcite, and thus
483 radiocarbon ages may provide reliable information on the long soil carbonates evolution, even if
484 each sample may still be partially affected by time-averaging blurring effect.

485 The considered sequence consists of superimposed calcitic soil horizons, which distinctly developed
486 in phases of enhanced evapotranspiration; consequently, as suggested by Machete (1985), the
487 development of calcitic horizons is a clue to their dissolution-precipitation history. According to the
488 distribution of radiocarbon ages shown in Fig. 9a, the lower part of the sequence (2Bk horizons)
489 may have been affected by old pedogenetic accumulation of calcium carbonate; then, after the

490 burying of this soil and aggradation of the uppermost part of the pedosequence, a further phase of
491 calcite accumulation took place. Its occurrence is recorded by the accumulation of calcitic
492 pedofeatures in the uppermost Bk horizon and the recrystallization or freshly accumulation of
493 calcite in the 2Bk horizon. The higher concentration of calcitic pedofeatures in the 2Bk horizon can
494 be interpreted as the result of subsequent phases of precipitation and/or of a more intense initial
495 accumulation.

496 As for authigenic calcite, in order to correlate the radiocarbon ages of different calcrete
497 pedofeatures to the climatic conditions occurring in the area, radiocarbon dating results were
498 compared with radiometric ages obtained by Adamson et al. (1982), Williams (2009), and Williams
499 et al. (2015) in a cumulative graph of calibrated radiocarbon ages (Fig. 8). Williams (2015)
500 identified several Late Quaternary periods characterized by more humid climatic conditions,
501 corresponding to higher flow level of the White Nile, Blue Nile, and main Nile River. These phases
502 occurred at 14700–13100, 9700–9000, 7900–7600, ca. 6300, and 3200–2800 cal. years BP (Fig.
503 8c). On the bases of published data we may infer several phases of carbonates development at c.
504 >30000, 26000–24200, 18200–13000, and 11000–9100 cal. years BP (Adamson et al., 1982); these
505 ages were obtained on bulk carbonate samples, not separated between matrix and cemented nodules
506 (Fig. 8b).

507 In this study, the oldest radiocarbon ages were obtained on cemented nodules from lower sampling
508 points A3d, A3c, and A3a (11480 ± 240 , 9900 ± 580 , and 9620 ± 100 cal. years BP, respectively),
509 corresponding to dry climatic phases (Fig. 8), according to palaeoenvironmental evidence provided
510 by Williams (2009). These older ages have a quite good correspondence (Fig. 8b) with those
511 obtained by Adamson et al. (1982). Younger radiocarbon ages, obtained from powdery calcitic-rich
512 matrix sampled at A3d and A3c points (dated to 7730 ± 80 and 7920 ± 100 cal. years BP,
513 respectively) as well as from calcitic-rich matrix sampled at A3a and from calcitic nodules sampled
514 at A1 (dated to 6340 ± 80 and 6080 ± 240 cal. years BP, respectively), show a good agreement

515 with two short periods characterized by more humid conditions, dated to 7900-7600 and about 6300
516 cal. BP (Williams, 2009; Williams et al., 2015), respectively (Fig. 8).

517 Further evidence that cemented nodules and calcitic-rich matrix possibly formed under different
518 conditions is provided by the stable isotope analysis, in particular by the significant variation of
519 $\delta^{18}\text{O}$ values observed between cemented nodules and matrix samples (Fig. 9a, 9b). Variation in
520 $\delta^{18}\text{O}$ values of calcrete samples generally reflects the variation in isotopic composition of meteoric
521 water; however, the latter may not necessarily reflect a significant variation in $\delta^{18}\text{O}$ of rainfall. In
522 fact, considering the available information on palaeoenvironmental conditions in this region,
523 previously discussed, a $\delta^{18}\text{O}$ enrichment can also be due to evaporative enrichment due to high
524 evapotranspiration rates (Cerling, 1984; Burns and Matter, 1995; Budd et al., 2002). Therefore, the
525 higher $\delta^{18}\text{O}$ values measured on cemented nodules can indicate higher evapotranspiration rates
526 related to drier environmental conditions during calcite precipitation. On the other hand, more
527 negative $\delta^{18}\text{O}$ values, as those measured on matrix samples, can reflect the $\delta^{18}\text{O}$ signature of rainfall
528 or at least a certain $\delta^{18}\text{O}$ depletion, triggered by higher rainfall intensity caused by a short-timed
529 increase of monsoon intensity (Cerling, 1984; Burns and Matter, 1995; Andrews et al., 1998).

530 The consistency of these results suggests that during the Late Pleistocene and mostly initial
531 Holocene a first onset of calcite precipitation occurred during dry climatic conditions, which is
532 compatible with the formation of a thick calcitic horizon in the lower part of the sequence. The
533 occurrence of a net moisture deficit in the vadose zone, as well as pore water saturated with respect
534 to calcium carbonate, due to decreased precipitation and the reduction of the White Nile flow,
535 possibly promoted the calcite precipitation. Subsequently, more humid climatic conditions in the
536 Early and Middle Holocene led to partial dissolution of calcite, whereas a subsequent shift towards
537 drier conditions caused the reprecipitation of calcite. Therefore, alternation of wet and dry periods
538 triggered the partial dissolution and reprecipitation of calcite, leading to a progressive calcite
539 accumulation. Cemented nodules may be interpreted as remnants of previously precipitated calcium
540 carbonate, partially dissolved during subsequent humid periods and then reprecipitated as looser

541 calcitic matrix. This hypothesis also suggests that carbonate pedogenesis occurred as a series of
542 subsequent events of calcite dissolution and precipitation, discontinuously distributed along the
543 Upper Pleistocene and Holocene, driven by local environmental conditions.
544 This interpretation is further supported by archaeological evidence. Several burial phases were
545 identified and attributed to different periods along the Holocene (Fig. 8), whereas pits, dated to the
546 Mesolithic period, were established after the pre-Mesolithic burial phase (the oldest one).
547 Mesolithic pits were cut into the deeper calcitic horizon, whereas pre-Mesolithic and Neolithic
548 human bones are permeated by secondary calcite. In the Meroitic bones, belonging to the most
549 recent burial phase (dated to 2000-1700 cal. BP; (Usai et al., 2010)), pedogenetic calcite is absent,
550 despite the fact that graves were cut within the calcrete horizon and placed deeper from the surface
551 with respect to the older ones. On the basis of these evidence, the mobilization of calcite in the last
552 2000 years, a period characterized by semi-arid to arid climatic conditions (Gasse, 2000) and lower
553 level of the White Nile (Williams, 2009), was negligible, indicating the occurrence of quiescent
554 stages during calcrete development process under arid conditions, when the amount of circulating
555 pore water is extremely limited.

556

557 **6 Conclusions**

558 In Central Sudan, the formation of calcitic soil horizons was a long-lasting process and their
559 development was significantly influenced by changes from humid to arid environmental conditions.
560 Bk horizons were characterized by alternate periods of growth due to dissolution and precipitation
561 processes, and periods of quiescence or extremely slow growth rates. These events possibly
562 occurred many times along the Quaternary, but the best-preserved evidence is the one related to the
563 Holocene. Despite the fact that radiocarbon dating of calcrete has severe limitations due to the
564 uncertain determination of the contribution of ^{14}C by different sources, accurate sampling method
565 coupled with additional information from a well-defined palaeoenvironmental context may provide
566 a reliable interpretation of the sequence of events. A higher number of carefully selected

567 radiocarbon-dated samples could supply a more statistically significant dataset for radiocarbon ages
568 as well as highlight the variability of AMS-¹⁴C determinations on this type of material. However,
569 radiocarbon ages obtained in the present work confirm the general model for accumulation,
570 formation and development of calcium carbonate in sediments/soil horizons established in previous
571 studies (Gile et al., 1966; Machette, 1985; Khormali et al., 2006; Zhou and Chafetz, 2009),
572 suggesting a progressive accumulation of calcium carbonate within sediments/soil horizons.

573 Specifically for this context, the interest in deciphering the relationship between carbonates
574 pedogenesis and archaeological records concerns the evaluation of the interaction between
575 secondary calcite and archaeological materials. In fact, the occurrence of secondary calcite in
576 archaeological materials may represent a complex issue in all those cases in which calcite may alter
577 conventional radiocarbon dates. An example can be, in the specific case, represented by the
578 radiocarbon dating of archaeological bones performed on bioapatite (the mineral fraction of bone
579 material) (Zazzo, 2014). Calcite is therefore a significant contaminant when measuring ¹⁴C activity
580 of bioapatite; despite the chemical removal of secondary calcite, the radiocarbon ages obtained (Dal
581 Sasso, 2015) are in good agreement with those obtained on pedogenic calcite, thus resulting
582 substantially younger than those hypothesized on the basis of archaeological and stratigraphic
583 evidences (Fig. 8d). Since bioapatite is not a closed system with respect to the environment
584 (Cherkinsky, 2009), carbonate exchange between calcite and bioapatite during burial may
585 significantly affect the result when dating the archaeological bones. Therefore, the study of soil
586 sediments and burial environment is fundamental in order to assess the reliability of radiocarbon
587 ages. A further example of the importance of understanding the time and steps of calcrete genesis in
588 archaeological context concerns the applicability of luminescence dating on archaeological material
589 (pottery); in fact, the dose rate of Thermoluminescence (TL) is heavily tuned by environmental
590 humidity and radiation, and the carbonate content of sediments may influence water mobilization
591 and change the natural radiation due to the increase of uranium occasionally substituting calcium in
592 calcite crystals. In TL dating, radiation is generally supposed to be constant over time, but irregular

593 development of calcrete through time may consistently vary the environmental radioactivity after
594 the burial of archaeological material (Wintle and Murray, 2006).

595 This study highlights the relevance of a multi-analytical approach to the study of carbonates
596 pedogenesis. Specifically for this case, a comprehensive study on the interaction between the
597 archaeological record and the calcrete formation and development supplies valuable information
598 both on the processes and timing of carbonates pedogenesis and on the preservation state of
599 archaeological materials.

600 Considering a wider perspective, the results of this study may represent a further step in the
601 comprehension of calcitic horizons formation and in the definition of a reliable method to date their
602 formation. This may offer a significant tool in palaeoenvironmental studies in arid lands, where
603 carbonatic concretions may represent the only archive for past climatic changes; in fact, several
604 authors had highlighted the correspondence (climofunction, *sensu* Jenny (1941) between depth of
605 Bk horizons and precipitations (e.g., Yaalon, 1983; Mack and James, 1992; Caudill et al., 1996;
606 Retallack, 2000; Retallack, 2005), being horizon's depth the results of the average amount of
607 rainfall.

608

609

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621

622 **Conflict of Interest**

623 The authors declare that they have no conflict of interest.

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626 **References**

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959

1 **Radiocarbon dating reveals the timing of formation and development of**
2 **pedogenic calcium carbonate concretions in Central Sudan during the Holocene**
3 **~~Radiocarbon dating of calcrete development records the steps of pedogenesis in~~**
4 **arid lands**

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15

16 **Abstract**

17 ~~A calcrete horizon formed in Central Sudan, sampled during the archaeological excavation of the Al~~
18 ~~Khiday 16-D-4 site, is here analysed by optical, cathodoluminescence and scanning electron~~
19 ~~microscopy. Distinct calcitic pedofeatures were identified, mechanically separated and analysed for~~
20 ~~stable carbon and oxygen isotopes and radiocarbon dated. Results are discussed taking into account~~
21 ~~all the available information from detailed physiographic and palaeoenvironmental studies carried~~
22 ~~out in this area and from the study of the tight interaction between calcrete and evidence of~~
23 ~~anthropic activities, as attested in the archaeological record. This allowed a more complete~~
24 ~~understanding of the processes involved in calcrete formation, recycling, and development in this~~
25 ~~area during almost the entire Holocene. Calcrete was characterized by alternate periods of growth~~
26 ~~and quiescence, depending on changes of environmental conditions at local and regional scale.~~
27 ~~Radiocarbon ages are critically discussed, highlighting advantages and limitations of this approach~~
28 ~~to timing calcrete development. These results show the effectiveness of a multi-analytical approach~~
29 ~~to the study of pedogenic calcrete, aiming to define the timing and dynamics of its formation and~~
30 ~~development.~~

31 ~~Considering a wider perspective, our results may represent a further step in the comprehension of~~
32 ~~calcrete formation and in the definition of a reliable method to date its formation. This may offer a~~
33 ~~significant tool for both archaeometric studies, where the interaction between secondary calcite and~~
34 ~~archaeological material may alter results and hamper their straightforward interpretation, and~~
35 ~~palaeoenvironmental studies in arid lands, where calcrete may represent the only archive for past~~
36 ~~climatic changes.~~

37 Calcitic soil horizons are common in arid and semi-arid lands and represent the result of the
38 progressive accumulation of calcium carbonate in the soil profile over time. This process leads to
39 the occurrence of several pedogenetic phases of calcium carbonate dissolution/precipitation. For
40 this reason, timing the formation and development of calcic horizons through radiometric dating is
41 not straightforward as time-averaging effects, due to the superimposition of the same process over

42 time, occur. On this basis, this study aims to define the timing and dynamics of formation and
43 development of pedogenic calcitic concretions from semi-arid Central Sudan, highlighting the
44 relevance of a multi-disciplinary approach and the effectiveness of radiocarbon dating (coupled to
45 an accurate sampling strategy) applied to pedogenic carbonates. Calcium carbonate-rich soil
46 horizons (Bk) were sampled during the archaeological excavation of site 16-D-4 at Al Khiday
47 (Central Sudan) and studied by optical, cathodoluminescence and scanning electron microscopy, as
48 well as by chemical-physical and stable isotopes (C and O) analyses. Radiocarbon ages are obtained
49 for distinctive calcitic pedofeatures and mostly refer to the Early Holocene for calcitic-cemented
50 nodules (11.5, 9.9 and 9.6 cal. ka) and to the Middle Holocene for powdery calcitic-rich matrix (7.9,
51 7.7, 6.3 and 6.1 cal. ka) samples. Our data are also compared with the available information from
52 detailed physiographic and palaeoenvironmental studies carried out in the region and from the study
53 of the tight interaction between soil horizons and evidence of anthropic activities at Al Khiday site,
54 as the archaeological record provides informative chronological constrains to interpret the ¹⁴C ages
55 obtained on selected calcitic pedofeatures. Pedogenic calcitic features dated to the Early Holocene
56 can be related to short arid phases during general wetter climate conditions, whereas those dated to
57 the Middle Holocene can be related to short humid phases. Results show that Bk horizons were
58 characterized by alternate periods of calcite accumulation (precipitation) and dissolution, and
59 periods of quiescence or extremely slow growth rates. Thus, the formation and development of Bk
60 horizons has been a long-lasting process significantly influenced by climatic fluctuations. This
61 study represents a further step in the comprehension of the development of calcium carbonate
62 concretions and (at a wider perspective) calcrete; the paper also contributes to the definition of a
63 reliable method to radiometrically date the formation of calcitic pedofeatures. In a broader
64 perspective, this work may offer a significant tool for archaeometric studies, where the interaction
65 between secondary calcite and archaeological material is significant (e.g., radiocarbon dating of
66 bioapatite in bones), and palaeoenvironmental studies in arid lands, where Bk horizon development
67 is a function of past rainfall.

68

69 **Keywords**

70 | Holocene; Calcic concretions~~Calcrete~~; North Africa; ~~Stable isotopes~~Cathodoluminescence
71 | microscopy; Radiocarbon dating; Pedogenesis.

72

73 **1 Introduction**

74 Calcic and petrocalcic soil horizons, and pPedogenic calcretes ~~refers to~~ are the results of a near-
75 surface terrestrial accumulation of predominantly calcium carbonate, resulting from the introduction
76 and cementation of calcium carbonate into a soil profile in different forms, i.e. powdery, nodular,
77 laminar, or massive (Gile et al., 1965; Machette, 1985; Wright and Tucker, 1991; Wright, 2007;
78 Durand et al., 2010); they are present in present-day soils (WRB, 2006) as much as in paleosols
79 (Mack et al., 1993; Retallack et al., 1993; Nettleton et al., 2000). These pedogenic bodies ~~is~~ often
80 occurs in areas, where vadose and shallow phreatic groundwater is saturated with respect of calcium
81 carbonate. This general definition is applied to a large variety of calcitic horizons at different stages
82 of development, abundantly occurring worldwide, at different latitudes, within different geological
83 contexts, and under different pedoclimate settings_ (Gile et al., 1966; Machette, 1985; McFadden
84 and Tinsley, 1985; Alonso-Zarza and Wright, 2010). However, the most evident examples of
85 calcitic soil horizons (Bk) developed in arid and semi-arid regions_ (Gile et al., 1966; Adamson et
86 al., 1982; Machette, 1985). General assumption is that calcrete develops in soils, characterized by a
87 net moisture deficit, in which the circulating fluids (vadose and shallow phreatic groundwater) are
88 saturated in calcium carbonate. Calcite accumulation is promoted by several physical-chemical
89 processes as well as by biological-mediated processes (Wright et al., 1988; Verrecchia and
90 Verrecchia, 1994; Cailleau et al., 2011), which, in turn, are influenced by climatic conditions, in
91 particular rainfall amount. As a consequence, calcrete is not generally the result of a single
92 pedogenetic phase, but it corresponds to subsequent events, eventually separated by a long time
93 span, during which previously precipitated carbonates undergo a partial or complete new
94 dissolution and then re-precipitation. Calcic horizon and Ccalcrete development, therefore, consists
95 of several, superimposed generations of micritic and/or sparitic calcium carbonate crystals. For this
96 reason, and considering also that a continuous opening of the system hampers the possibility to date
97 calcite with U/Th and ¹⁴C, it is hard to define accurately the time and steps of calcrete pedogenesis.

98 Almost thirty-five years ago, (Adamson et al., (1982) have carried out a huge campaign of
99 radiocarbon dating of calcitic nodules in soils from Central Sudan, highlighting the occurrence of
100 reworked carbonates and subsequent recrystallizations. For that reason and for the the large
101 occurrence of carbonate pedofeatures, soils/paleosoils of arid Sudan are an exceptional case study to
102 investigate the chronological development of carbonatic concretions. Therefore, ~~In~~-in this research,
103 a multi-analytical approach, consisting in mineralogical, micromorphological, geochemical, and
104 geochronological analyses, is applied to the study of calcitic pedogenic ~~calcrete~~-features from
105 Central Sudan, in order to contribute towards the understanding of dynamics and timing of ~~its~~-their
106 formation and development. Carbonate-bearing soil horizons~~Calcrete~~ analysed in this study formed
107 in a localised area along the left bank of the White Nile, few km south of the Khartoum-Omdurman
108 urban system (Fig. 1), where a large, multiphase archaeological site (Al-Khiday, currently under
109 excavation) is in tight relation with the ~~calcrete~~carbonate concretions. In fact, part of the vestiges of
110 the site is embedded in the calcitic crust (Fig. 1a2a), whereas other features are clearly cut in the
111 pre-existing carbonate-bearing horizon (Fig. 1b-2b and 1e2c). Detailed studies on the local
112 physiographic and palaeoenvironmental aspects (Zerboni, 2011; Williams et al., 2015), as well as
113 on the anthropic activities, considering the wide span of time covered by the archaeological
114 evidence (almost the whole Holocene) (Usai and Salvatori, 2002; Usai, 2003; Usai and Salvatori,
115 2005; Salvatori and Usai, 2009; Usai et al., 2010; Salvatori et al., 2011; Salvatori, 2012; Dal Sasso
116 et al., 2014a; Jakob, 2014; Salvatori et al., 2014; Dal Sasso et al., 2014b; Usai et al., 2014; Dal
117 Sasso et al., 2016; Iacumin et al., 2016; Usai et al., 2017; Maritan et al., 2018), supply important
118 information to reconstruct the timing and steps of calcrete formation. In our study, results from the
119 minero-petrographic and pedogenetic study of ~~calcrete~~-soil samples, coupled with additional
120 archaeological and palaeoenvironmental information, allowed a more complete understanding of
121 the processes involved in calcitic horizons and calcrete formation, recycling, and development.
122 Moreover, since these processes are strongly dependent on water availability, which, in turn,

123 depends on climatic conditions, our results are discussed taking into account the regional Holocene
124 variations in water availability, as reconstructed by several palaeoenvironmental studies.

125

126 **2 Study area, palaeoenvironmental settings, and archaeological background**

127 A complex calcrete horizon has been found during the archaeological survey and excavation of
128 several archaeological sites (16-D-3, 16-D-4, 16-D-4b, 16-D-5, 16-D-6) and the geomorphological
129 survey of the region carried out since 2006 within the “El Salha ~~archaeological~~ Archaeological
130 project” (Usai and Salvatori, 2002; Usai, 2003; Usai and Salvatori, 2005; Salvatori and Usai,
131 2009; Usai et al., 2010; Salvatori et al., 2011; Salvatori, 2012). The archaeological sites are located
132 on the western bank of the White Nile, near the Al Khiday village (Central Sudan), at 3.5 km from
133 the present-day river course and about 22 Km south of its confluence with the Blue Nile (Fig. 2a1a).
134 They are set on fluvial sandy ridges, corresponding to the remnants of late Pleistocene longitudinal
135 river bars, at the limit of alluvial sediments deposited during the very late Quaternary flooding of
136 the White Nile (Zerboni, 2011; Williams et al., 2015). These fluvial ridges and the surrounding flat
137 areas are weakly weathered by Holocene pedogenesis, despite the intense accumulation of
138 pedogenic calcium carbonate (Zerboni, 2011), thus the poorly developed local soils can be defined
139 as aridisols (Buursink, 1971).

140 The archaeological excavation and geomorphological investigation (Usai et al., 2010; Salvatori et
141 al., 2011; Zerboni, 2011) revealed a complex stratigraphy at these archaeological sites, resulting
142 from anthropic activities, sedimentary, erosional, and pedogenic processes occurring along a wide
143 span of time. The area had been inhabited several times along almost the entire Holocene. Focusing
144 on the 16-D-4 site (Fig. 2b1b), a Mesolithic specialised use of the area was defined by the numerous
145 pits (more than 100) that have been excavated, characterized by different types of filling material
146 and presumably with different functions (Zerboni, 2011), and radiocarbon dated to 8650-8250 cal.
147 BP (Salvatori et al., 2011). In addition, the site was used several times as a burial ground: at least
148 three different burial phases have been identified. The most ancient one, named pre-Mesolithic, is

149 characterized by individuals (at least 90) buried in a prone and elongated position. The chronology
150 of this burial phase is uncertain but constrained by the Mesolithic use of the area, as a number of
151 skeletons (16) were found to be cut by radiocarbon-dated Mesolithic pits (Salvatori et al., 2011),
152 which indicate that the pre-Mesolithic burial phase is unambiguously older than 8650 cal. BP. The
153 site was subsequently used as a cemetery during the Neolithic period (38 graves; 6500-6200 cal.
154 BP), and later on during the Meroitic period (43 graves, 2100-1800 cal. BP) (Usai et al., 2010;
155 Salvatori et al., 2011).

156 Soils in Central Sudan can be classified as Vertisols, Entisols, Aridisols, and Alfisols (Buursink,
157 1971) in the study region, the first three categories are the most represented and Aridisols are those
158 most recurrent at Al Khiday. In the whole region, pedogenic processes lead to the accumulation of
159 calic pedofeatures, which have been described previously by several authors (Blokhuis et al., 1968;
160 Williams, 1968; Buursink, 1971; Adamson et al., 1982). (Buursink, (1971) in his classification of
161 soil of Central Sudan describes several petrocalcic horizons, rich in CaCO₃ nodules and/or
162 concretions, similar to the K horizons defined by (Gile et al., (1966). The same pedofeatures have
163 been reported by (Blokhuis et al., (1964) and; Blokhuis et al.,(1968), who also described the
164 properties and distribution of pedogenic carbonate in Vertisols. Soils and paleosoils with common
165 to abundant CaCO₃ concretions have been reported also by (Williams, (1968) along the White Nile.

166 Today, the climate of the region is arid, with mean annual temperature ranging from 22°C to 33°C
167 and average rainfall of ca. 130 mm (El-Tom, 1975; Elagib and Mansell, 2000). However, climatic
168 conditions were significantly different in the past and substantial climatic changes occurred at local
169 and regional levels (Williams and Adamson, 1980; Gasse, 2000; Nicoll, 2001; Nicoll, 2004;
170 Williams, 2009; Williams and Jacobsen, 2011; Zerboni, 2013; Gatto and Zerboni, 2015) during the
171 wide time span of the site use (from the early Holocene to the beginning of the 1st millennium AD).
172 In the early Holocene (11500-8000 cal. BP) the precipitation rate, and consequently also the
173 environmental humidity, were substantially higher, as well as the flooding level of the White Nile,
174 due to a northward expansion of the Indian monsoon domain (Gasse, 2000; Williams, 2009). The

175 formation of seasonal swamps in the surrounding area of the Al Khiday sites during the Mesolithic
176 occupation of the area is attested by geoarchaeological and geomorphological evidence (Williams
177 and Jacobsen, 2011; Zerboni, 2011). Since the middle Holocene (8000-3600 cal. BP), the
178 progressive weakening of monsoon intensity led to rainfall decrease and overall reduction of water
179 availability. In the late Holocene (since ca. 3600 cal. BP), since the Meroitic period and later on,
180 progressively drier environmental conditions occurred, up to the arid climate that now characterizes
181 central Sudan; only short-timed wetter events have been occasionally recorded (Mawson and
182 Williams, 1984). In addition to this general trend, palaeoenvironmental records identified several
183 periods of rapid climatic changes during the early Holocene wet phase, when several short arid
184 periods occurred (Williams, 2009).

185

186 **3 Materials and methods**

187 The samples considered in this research come from a test trench, exposed in the sandy deposit at Al
188 Khiday and representing the local substrate, brought to light during the excavation of a Mesolithic
189 pit (pit number 151A). The section shows a soil profile with at least ~~70 cm~~0.7 m of fine to coarse
190 sand (occasionally lenses of pebbles are present), deeply cemented by calcium carbonate (~~calcrete~~).
191 Horizons description is in Tab. 1. Six oriented and undisturbed blocks were collect, sampling points,
192 namely A1, A2, A3a, A3b, A3c, and A3d, were selected at different depths from the top to the
193 bottom of the section (see Tab. 1 and Fig. 3 for details~~Fig. 3~~). At each sampling point, samples were
194 collected and prepared as required for the analytical techniques applied in this study.~~A total of 9~~
195 ~~samples (one for each block and an additional sample for A2, A3a, and A3d blocks) covering the~~
196 ~~whole section were selected.~~ Samples were embedded under vacuum in epoxy resin (Araldite 2020)
197 and prepared in petrographic thin sections (3x5 cm). Micromorphological analyses were carried out
198 ~~on thin sections~~ by petrographic optical microscopy (OM), cathodoluminescence microscopy (CL),
199 and scanning electron microscopy (SEM).

200 OM analysis was ~~carried out~~performed with a petrographic microscope (Nikon Eclipse E660) under
201 plane-polarized (PPL) and cross-polarized light (XPL). CL analysis was performed with a
202 petrographic microscope (NIKON Labophot2-POL) equipped with a cold cathode stage (CL8200
203 MK3, Cambridge Image Technology Ltd) and operating at 15 kV and 200 mA. SEM analysis was
204 ~~performed~~carried out with a CamScan MX 2500 coupled to a detector for Energy dispersive X-ray
205 spectroscopy (EDS), equipped with a LaB6 cathode and operating at 20 kV and 160 nA. Thin-
206 sections were described following the terminology proposed by Bullock et al. (1985) and Stoops
207 (2003), and the interpretation mostly followed the concepts discussed in Stoops et al. (2010).

208 Samples adjoining those prepared in thin sections were selected for chemical and stable isotopes
209 analyses and radiocarbon dating. The chemical characterization of samples was performed
210 measuring CaCO₃ equivalents, pH, humified carbon content, total organic carbon, total carbon
211 content (TC) and nitrogen content (TN). Calcium carbonate equivalents were chemically performed
212 using a Dietrich–Frühling calcimeter, which measures the volume of CO₂ developed by acid
213 reacting with the bulk sample, which is proportional to the carbonate concentration. Humified
214 organic carbon was identified by means of the Walkley and Black (1934) method, using chromic
215 acid to measure the oxidizable organic carbon (titration). Total organic carbon was estimated by
216 loss on ignition - LOI - (Heiri et al., 2001); samples were air-dried (at 105°C) and organic matter
217 was oxidized at 500–550°C to carbon dioxide and ash, then the weight lost during the reaction was
218 measured by weighing the samples before and after heating. The analysis of total carbon and total
219 nitrogen were performed on dried soil samples with a Thermo Fisher Scientific Organic Elemental
220 Analyzer (OEA-Flash 2000). Samples were weighed out into tin containers to the nearest 0.001mg
221 on a Gibertini MICRO1000 electronic microbalance. Each sample was then flash-combusted at
222 1800°C in the OEA.

223 ~~On the basis of micromorphological analysis of thin sections~~samples were gently disaggregated in
224 ~~an agate mortar; distinctive~~After a gentle disaggregation in an agate mortar, distinctive calcitic
225 pedofeatures, described by thin section micromorphological analyses, were identified on massive

226 | ~~samples along the calerete section were and~~ accurately separated by hand-picking under the
227 | stereoscopic microscope, ~~. Each pedofeature and was~~ then analysed for stable carbon and oxygen
228 | isotopes and radiocarbon dated.

229 | Stable isotopes analyses were carried out with a Thermo Scientific Delta V Advantage Isotope
230 | Ratio Mass Spectrometer. CO₂ was developed at 70 °C by complete reaction with >99% crystalline
231 | H₃PO₄ in a Gasbench II device connected to the spectrometer. Results were calibrated with two
232 | internal standards (sieved Carrara marble and Millipore Suprapur carbonate), periodically calibrated
233 | against the international reference carbonates NBS 19, NBS 18 and L-SVEC. A control standard
234 | (sieved Monzoni marble) was also measured and reproduced with external errors of better than
235 | 0.1‰ (1σ) for both carbon and oxygen.

236 | Radiocarbon dating by accelerated mass spectrometry (AMS-¹⁴C) was carried out at the Center for
237 | Isotopic Research on the Cultural and Environmental heritage (CIRCE) laboratory (Terrasi et al.,
238 | 2008) of the Second University of Naples. All AMS-¹⁴C ages were calibrated according to the
239 | INTCAL13 dataset (Reimer et al., 2013) using the CalPal software (Weninger and Jöris, 2008). The
240 | method of multiple-group calibration was used to graphically represent and compare calibrated
241 | AMS-¹⁴C ages (Weninger, 1986).

242

243 | **4 Results**

244 | **4.1 Field description**

245 | The ~~calerete soil profile~~, here studied, corresponds to the section of the wall of the pit 151A, an
246 | archaeological feature related to the Mesolithic use of the site (8650-8250 cal. BP (Salvatori et al.,
247 | 2011; Salvatori, 2012)), excavated on top of a fluvial sandy ridge. ~~Calerete horizon is established at~~
248 | ~~about 30 cm under the current surface level.~~ In the area of Al Khiday, pedosedimentary sequences
249 | generally correspond to poorly weathered soils interlayered to sandy to silty sediments, eventually
250 | disturbed by anthropic activities (Buursink, 1971); the sequences are interpreted as fluvial deposits
251 | related to an Upper Pleistocene higher level of the White Nile (Zerboni, 2011). The general aspect

252 of a typical soil profile consists of a sandy to silty top horizon, enriched of calcium carbonate in
253 form of nodules (Bk) horizon, lying on a BC horizon~~with no or very poor evidence for pedogenesis.~~
254 An abrupt limit (possibly erosional) marks the transition to the following 2Bk horizon (~~the calcrete~~),
255 which consists of medium to coarse sand, sometimes including lenses of coarser grains (very coarse
256 sand to small pebbles), deeply cemented by calcium carbonate; this buried petrocalcic horizon can
257 be interpreted as a former calcrete. Beneath, there is a sequence of fluvial to lacustrine fine
258 sediments (silt and sand), with few CaCO₃ nodules in the upper part (for a complete description see
259 Williams et al., 2015).

260 At the sampling point, the pedostratigraphy is ~~slightly more complex~~very clear (Fig. 3); ~~since~~ two
261 distinct calcium carbonate-rich calcrete horizons, separated by a sandy layer, can be observed. ~~The~~
262 ~~calcrete horizon (Bk), up to 1 m thick, is nodular and can be interpreted as an early stage of calcrete~~
263 ~~development in soil profile (Machette, 1985; Wright, 2007), resulting from the illuvial~~
264 ~~concentration of calcium carbonate in a siliciclastic host material (quartz sand and silt). Two main~~
265 ~~concentrations of calcium carbonate were identified.~~ The uppermost Bk horizon ~~part of the section,~~
266 ~~namely block~~ (sampling point A1), is constituted by quartz sand slightly cemented by calcium
267 carbonate; calcite concentration within the sediments is detected as coalescent irregular calcitic
268 nodules of few centimetres in diameter. A weakly cemented sandy ~~lens~~BC horizon; (sampling point
269 ~~corresponding to block A2~~), is interposed between the first (~~A1~~)Bk horizon and ~~the a~~ second
270 concentration of calcium carbonate, corresponding to the lower part of the section (~~namely~~
271 ~~sampling points from~~ A3a, A3b, A3c and A3d ~~blocks~~). This deepest calcitic horizon (2Bk), up to
272 0.7 m thick, is nodular and can be interpreted as an early stage of calcrete development in soil
273 profile (Machette, 1985; Wright, 2007), resulting from the illuvial concentration of calcium
274 carbonate in a siliciclastic host material (quartz sand and silt). It is formed by cemented quartz sand
275 and carbonate concretions, where calcitic nodules can be distinguished. Abundance of carbonate
276 concretions as well as cementation increases with depth. From the pedological point of view, this
277 evidence ~~can be interpreted as a single calcrete, rather than~~represents a sequence of distinct calcitic

278 horizons, ~~since calerete developed on the same distinct~~ parent materials, probably resulting from a
279 ~~long-lasting discontinuous~~ pedogenetic process and alternated sedimentary events. (Wright, 2007).
280 The ~~calerete Bk~~ horizon, as observed in the archaeological area and in naturally exposed sections is
281 at about ~~30~~20-50 cm depth from the current surface level. In correspondence of site 16-D-4 the
282 close relationship between calcitic pedofeatures~~calerete~~ and archaeological features of different
283 ages can provide further evidence on the timing of calcium carbonate calerete development. Calcitic
284 concretions ~~Calerete~~ partially embeds human bones belonging to the pre-Mesolithic burial phase,
285 the most ancient attested at the site (Fig. ~~1a~~2a), whereas Mesolithic pits (Fig. ~~1b~~2b) and Meroitic
286 graves (Fig. ~~1c~~2c) are cut into it. Nevertheless, pre-Mesolithic and Neolithic bones were found to be
287 permeated by secondary calcite, whereas the Meroitic bones, belonging to the most recent burial
288 phase were not affected by pedogenetic precipitation of calcite (Dal Sasso et al., 2014b; Dal Sasso
289 et al., 2016; Dal Sasso et al., 2018). In addition, the lower part of most of the Mesolithic pits is
290 characterized by filling material cemented by calcium carbonate; in particular, some pits, used as
291 fireplaces, present a significant amount of whitish and hard calcitic concretion encrusting the
292 archaeological materials. The same evidence of calcitic concretions embedding human and animal
293 remains in archaeological contexts has been described elsewhere in Sudan, as on the Singa calvaria
294 (McDermott et al., 1996), at Abu Hugar (Whiteman, 1971), and elsewhere in the Gezira (Adamson
295 et al., 1982). The origin of this calcitic cement can be associated to the processes involved in
296 calcrete development; however, the rather high level of cementation must imply a contribution to
297 the total amount of calcite coming from the recrystallization of the abundant remains of Ca-rich
298 wood ash (Canti, 2003) originally constituting the upper part of the pit infilling (Zerboni, 2011).

299

300 **4.2 Micromorphology of thin sections**

301 Thin sections ~~of the blocks (A1 and A2)~~ from the upper part of the ~~calerete (Bk horizon)~~soil profile
302 (A1 and A2 samples) show a groundmass with abundant sand- and silt-sized quartz grains, sub-
303 rounded to sub-angular in shape, with a bimodal grain-size distribution. The authigenic calcite is

304 heterogeneously distributed (in shape of nodules, coatings, and infillings) among the sections giving
305 to the slides a single- to double-spaced porphyric c/f (coarse/fine) related distribution, locally open
306 porphyric or gefuric. In the uppermost Bk horizon, ~~A~~ areas corresponding to typical calcitic
307 nodules, macroscopically identified, show a micromass mainly constituted by micrite and
308 microsparite (Fig. 4a), much more abundant with respect to that observed for other portions of the
309 poorly cemented samples (Fig. 4c). Allochthonous, well-rounded calcitic pedorelicts (Fig. 4b, 5c,
310 5d), comparable in size with sandy quartz grains, are frequent. They are constituted by a micritic
311 matrix stained by Mn and Fe oxides/hydroxides, and contain veins of sparitic to micro-sparitic
312 calcite, generally occurring within this kind of pedofeatures (Fig. 5c, 5d).

313 Samples from the upper units also evidenced the occasional occurrence of wood ash (calcite
314 pseudomorphs after calcium oxalate crystals (Canti, 2003)), fish bone fragments and small
315 charcoals (Fig. 4d). Anthropogenic features were observed in small localised areas of the thin
316 section (Fig. 4d), and possibly migrated from the infilling of the nearby Mesolithic pit, due to
317 intense bioturbation (Zerboni, 2011). SEM analysis on calcitic nodules from ~~these units-~~ Bk horizon
318 revealed that the microsparitic groundmass is heterogeneous in terms of compositions and
319 cementation degree: some areas are characterized by densely packed calcite crystals (Fig. 6a) with
320 respect of other areas, where a loose calcite crystals distribution is observed (Fig. 6c). In other
321 areas, calcite crystals occur within a clayey matrix (Fig. 6b). The great heterogeneity in terms of
322 calcite crystals distribution is detected also by CL, where the microsparitic micromass appears with
323 an orange luminescence, whose intensity is proportional to the density of calcite crystals (Fig. 5a).

324 Poorly cemented areas are characterized by low luminescence due to scarcity or lack of calcite in
325 the matrix (Fig 5b).

326 Thin sections from the lower 2Bk horizon (samples A3a, A3b, A3c, A3d) show areas characterized
327 by slightly cemented quartz grains, sub-rounded to sub-angular in shape, and sand- and silt-sized,
328 embedded into a micritic micromass (Fig. 4e). The distribution of mineral grains and calcite
329 generally results in a single spaced to open porphyric, occasionally gefuric, c/f related distribution.

330 Conversely, highly carbonated areas (Fig. 4f, 4g), corresponding to older calcitic nodules embedded
331 in the micritic micromass, are predominantly constituted by microsparitic calcite, and quartz sand
332 grains are extremely rare. Different textural pedofeatures were identified within calcitic nodules:
333 some portions are characterized by well-cemented calcite crystals forming hard nodules (hereafter
334 called cemented nodules) of sub-rounded to sub-angular shape, identifiable by OM (Fig. 4f, 4g), CL
335 (Fig. 5e) and SEM (Fig 6e, 6f) analyses. These features (Fig. 7) are embedded in a loose matrix
336 mainly constituted by calcite crystals similar in size, but less packed than in the cemented nodules,
337 sometimes associated to clay minerals. Towards the bottom of the ~~ealere~~2Bk horizon, the
338 dimension of ~~ealeric~~-calcitic-cemented nodules decrease and their margins, in contact with the
339 surrounding matrix, are less defined (Fig. 5f). CL analysis indicates a significant, but not
340 systematic, luminescence variations of calcite crystals along the section, both dispersed and well
341 packed. In some cases, homogeneously luminescent crystals were observed (Fig. 5g), whereas in
342 others the innermost part of crystals is less luminescent with respect to their margins and inter-grain
343 cement (Fig. 5h). OM, CL, and especially SEM images show an overall increase of calcite crystal
344 size from the upper to the lower Bk horizon. Allochthonous calcitic pedorelicts, sub-rounded in
345 shape and permeated by Fe oxides/hydroxides (different from the Mn and Fe-bearing
346 oxides/hydroxides permeating the rounded grains identified in the upper unit) were observed in unit
347 A3c and, less frequently, in unit A3d. Both the two types of pedorelicts show a high concentration
348 of oxides near their margins and a thin layer of clay minerals coating the outer surface (Fig. 4h, 6d).

349

350 4.3 Chemical and physical analyses

351 Tab. 1 and Fig. 3 illustrate the results of chemical analyses on soil samples. Calcium carbonate
352 equivalent content and pH show similar trends, being the latter generally basic and buffered by the
353 high content of carbonates; the B horizon is the less rich in carbonate. Humified organic carbon
354 contents show trends antithetic of CaCO₃ content and the highest peak in organics (B horizon)
355 corresponds to the less basic horizon. LOI result indicates a higher content of organics in the

356 lowermost part, possibly suggesting a lower preservation of organic matter in the uppermost part of
357 the sequence, where in fact humified carbon is more abundant. TC content is mainly ruled by the
358 occurrence of calcium carbonate, whereas TN variations corresponds to the variation of the
359 humified carbon, confirming that organic matter is the main source of nitrogen.

360

361 **4.3.4 AMS-¹⁴C dating**

362 Due to the lack of well identifiable organics in the soil matrix, AMS-¹⁴C radiocarbon dating was
363 performed on different calcium carbonate-bearing pedofeatures, identified by micro-morphological
364 analysis, and mechanically selected under the ~~stereographic~~ stereoscopic microscope. As for the
365 uppermost Bk horizon (A1 sampling point, Fig. 3) units, calcitic nodules, constituted by
366 micritic/microsparitic micromass (as shown in Fig. 4a) were identified in the massive sample as
367 powdery aggregates, up to 1 cm in size. A fraction of micritic/microsparitic micromass was selected
368 for radiocarbon dating (A1-mt samples). ~~allochthonous~~ Allochthonous pedorelicts (A1-pedorelict)
369 and ~~calcitic nodules (A1-mt) from unit A1~~ were also selected from the same sampling point (A1)
370 and dated.

371 In ~~As for~~ the lower ~~unit 2~~ Bk horizon, micromorphological analyses of calcitic nodules (up to 2 cm in
372 size) showed the occurrence of different textural pedofeatures, previously addressed as cemented
373 nodules and loose calcitic-rich matrix (as shown in Fig. 4f, 4g and 5e). In Fig. 7, OM
374 photomicrographs and the segmented false-colour image show a representative example of the
375 relationship between these pedofeatures. On the massive samples, cemented nodules ~~identified in~~
376 ~~thin sections~~ were found to be much more lithified than the surrounding loose calcitic-rich matrix,
377 therefore, after a gentle disaggregation of the samples, these two adjoining pedofeatures were
378 effectively separated by hand-picking under a stereoscopic microscope. ~~and were supposed to be~~
379 less prone to the re-opening of the carbon system. This sampling strategy was then applied to
380 massive samples collected at A3a, A3c and A3d points (Fig.3), so that for each of them cemented
381 nodules (labelled as A3a-cn, A3c-cn and A3d-cn) and calcitic-rich matrix (labelled as A3a-mt, A3c-

382 ~~mt and A3d-mt) samples were selected for radiocarbon dating. Therefore, three samples of~~
383 ~~cemented nodules (A3a-cn, A3c-cn and A3d-cn) and three samples of calcitic rich matrix (A3a-mt,~~
384 ~~A3c-mt and A3d-mt) from blocks A3a, A3c, and A3d, respectively, were selected and dated.~~ As
385 expected, an old radiocarbon age, 29540 ± 440 cal. years BP, was obtained for the allochthonous
386 pedorelicts (A1-pedorelict). The cemented nodules from ~~blocks~~ A3a, A3c and A3d sampling points
387 show older ages (9620 ± 100 , 9900 ± 580 and 11480 ± 240 cal. years BP ~~for, A3a-cn, A3c-cn and~~
388 ~~A3d-cn,~~ respectively) with respect to the nearby loose calcitic-rich matrix (6340 ± 80 , 7920 ± 100
389 and 7730 ± 80 cal. years BP for A3a-mt, A3c-mt and A3d-mt, respectively) and from the calcitic
390 nodules ~~of from A1 block-sampling point~~ (6080 ± 240 cal. years BP) (Tab. ~~42~~, Fig. ~~7a8a, 9a~~).

391 Apart from a single radiocarbon age dating to the Upper Pleistocene (for the allochthonous
392 pedorelict), the radiocarbon dates are Holocene in age and can be tentatively clustered to the Early
393 and Middle-Holocene; ~~-. statistical-Statistical~~ treatment of dating results can better delineate the
394 distribution of radiocarbon ages on ~~the calcrete sampl~~sampled calcitic horizons. The method of
395 multiple-group calibration (Weninger, 1986; Weninger and Jöris, 2008) was used to represent
396 radiocarbon ages, in order to discern if close radiocarbon ages might refer to the same or different
397 pedogenetic phases as well as to ease the comparison with other regional radiocarbon age datasets.

398 ~~More in detail, Our~~ AMS- ^{14}C ages on carbonates were compared (Fig. 7) with the ^{14}C ages obtained
399 on calcium carbonate deposits in soils from Central Sudan (Adamson et al., 1982), and, ~~mostly~~
400 ~~obtained~~ on fossil shells (Williams, 2009; Williams et al., 2015); The latter ages indicating-indicates
401 the late Quaternary high flow levels of the White Nile, representing ~~(which is~~ a proxy for wetter
402 climatic conditions.), ~~and with~~ In Fig.7 the radiometric chronology of the Holocene human
403 occupation of the Al Khiday archaeological sites (Salvatori et al., 2011) is also reported (Fig. 7). A
404 further, indirect piece of the chronological puzzle of calcrete development at Al Khiday is
405 represented by the radiometric dating of the archaeological structures cutting the carbonatic crusts;
406 most of them belong to the Mesolithic phase and date to the 8650-8250 cal years BP.

407

408 | **4.4.5 Stable isotopes analysis**

409 C and O stable isotope analyses were carried out on an aliquot from the same samples that were
410 radiocarbon dated; moreover, ~~for each calcrete block along the section,~~ additional samples of
411 calcitic nodules from A1 and ~~of the~~ cemented nodules (cn) and calcitic-rich loose ~~matrix~~ (mt) from
412 A3a, A3b, A3c and A3d sampling points (Fig. 3) were separately selected and analysed (Tab. ~~42~~).
413 Overall $\delta^{18}\text{O}$ ranges between -8.27‰ and -1.85‰ (V-PDB) and $\delta^{13}\text{C}$ between -1.48‰ and 1.06‰
414 (V-PDB). $\delta^{13}\text{C}$ values show a narrow range of variation and no significant differences were
415 observed between cemented nodule and matrix samples. Conversely, a wider range of variation was
416 observed for $\delta^{18}\text{O}$, with matrix samples showing more negative values with respect to cemented
417 nodules (Tab. ~~42~~, Fig. 89b, 9c). A good consistency was observed between results obtained from
418 the same samples double-measured, in particular for those from the matrix samples.

419

420 | **5 Discussion**

421 | **5.1 Origin of calcium carbonate and calcrete formation**

422 The authigenic accumulation of calcium carbonate in siliciclastic host sediments, composed of
423 quartz sand lacking in calcite, indicates an external source for calcium carbonate. Conceivable
424 sources may be identified in the geological bedrock of the area, as the limited quantity of calcite,
425 occurring in the local sandstone and shales, is firstly dissolved, then mobilized and finally re-
426 precipitated thanks to circulating pore water. Further significant sources of carbonate may be
427 represented by alkaline atmospheric dust and calcium carbonate precipitated over a long period in
428 the Pleistocene lake sediments identified in the region (Williams et al., 2015). Similar sources of
429 carbonates have been claimed by (Cremaschi et al., (2010) to explain the origin of spring calcareous
430 tufa in a sandstone-bearing central Saharan massif. Less important contributions are likely those
431 from the Nile overbank sediments, and from the anthropogenic calcitic ash related to the Mesolithic
432 occupation of the site, the latter being available only after the onset of pedogenetic processes related
433 to the formation of calcrete. Moreover, the dissolution and mobilization of calcium carbonate

434 accumulated in pre-existent Pleistocene Fe-enriched paleosols, which are highly cemented and
435 heavily affected by pedogenesis (Fig. ~~4d~~2d), may have contributed to the formation of the calcrete
436 horizon at 16D4 site. However, the origin of calcium carbonate in these paleosols is not well-
437 defined and its accumulation likely results from long-lasting processes of dissolution/precipitation
438 of calcite, possibly mediated by biological activity. Evidence of this type of paleosols, widely
439 outcropping in the region (Zerboni et al., 2016), can be also found few hundred meters west from
440 the 16D4 site. These Pleistocene paleosols are also the most probable source of the pedorelicts
441 identified in the calcrete section (those separated from ~~block-sample~~ sample A1 and dated to the $29540 \pm$
442 440 cal. years BP). In fact, they probably derive from surface transportation, as suggested by their
443 sub-rounded to rounded shape and by the occurrence of clayey material coating their external
444 surface, which can be interpreted as a rolled pedofeature (Zerboni, 2011). Also Adamson et al.
445 (1982) obtained Upper Pleistocene dating on rolled carbonates, suggesting their single or multistep
446 reworking of older carbonate-impregnated clays and silts from former soils.
447 ~~Calcrete-Calcitic~~ nodules formed since the accumulation of microcrystalline calcite; the small size
448 of crystals suggests a relatively fast precipitation of calcite, associated with high rates of
449 evapotranspiration. In this case, the contribution of microbially induced calcite precipitation is
450 assumed to be minimal, as micromorphological features characteristic of biological activity
451 (Newman et al., 1997; Richter et al., 2008) are not observed. The upper Bk horizon is characterized
452 by weak cementation and nodules are formed by microcrystalline calcite precipitated in the porosity
453 between sand- and silt-sized quartz grains. The lower 2Bk horizon shows a significant increase in
454 the accumulation of calcium carbonate and the formation of larger ~~nodules~~concretions. The
455 occurrence of very few quartz grains much more dispersed in the microsparitic groundmass within
456 nodules, with respect to those observed in less cemented portions of the sample, may indicate a
457 displacive calcite crystal growth (Armenteros, 2010). Moreover, higher permeating cementation of
458 sediments was observed in this unit, ~~thus~~ thus ~~This may represent~~ ing a subsequent development stages
459 of ~~calcrete-the calcitic~~ horizon, ~~however~~ However, features characteristic of successive stages of

460 calcite precipitation/recrystallization observed in ~~more-well-~~developed calcrete profiles (Wright,
461 2007) are lacking in our case study, thus suggesting an intermediate development stage for ~~this~~
462 ~~ealere~~calcitic horizons. At Al Khiday, Bk and 2Bk horizons ~~It~~ can be described as ~~a-~~nodular
463 calcitic horizons as suggested by Alonso-Zarza and Wright (Alonso-Zarza and Wright, 2010), or at
464 least-and comparable to a Stage III of calcrete development (Machette, 1985).

465 Cathodoluminescence of carbonates, in terms of colours and intensities, is mainly attributed to the
466 occurrence of Mn²⁺ and Fe²⁺ ions in trace quantity in the calcite crystal structure, which, in turn, is
467 related to geochemical conditions during precipitation. Therefore, even if only qualitatively,
468 luminescence variations of calcite crystals observed in CL images indicates variations in the
469 chemical composition and/or redox conditions during precipitation and subsequent recrystallization
470 (Hiatt and Pufahl, 2014). These data, coupled with information on the diachronic interaction
471 between calcrete horizon and archaeological records, indicate that calcrete formation and
472 subsequent development is a long-lasting and discontinuous multi-step process, acting at least since
473 the Early Holocene. Notwithstanding the evidence of ~~ealere~~formationcalcite accumulation ~~o~~in
474 Late Pleistocene fluvial sediments, the widespread occurrence of calcium carbonate features in
475 paleosols and sediments of the region, may suggest that environmental conditions suitable for
476 calcite mobilization, redistribution and precipitation in soils ~~form-of-ealere~~ may have occurred
477 many times since the Pleistocene, as described also elsewhere in North Africa (Szabo et al., 1995;
478 Brookes, 2010; Zerboni et al., 2011). Yet, Adamson et al. (1982) suggest multiple events of
479 carbonate dissolution and precipitations in soil and sediments of Central Sudan since the Late
480 Pleistocene and probably earlier.

481 On these bases, timing the formation and development of carbonate concretions in the soil profile
482 by radiometric dating techniques requires an accurate sampling strategy and a careful evaluation of
483 results.

484 **5.2 Timing of ealere development**

485 Radiocarbon dating of calcrete and recrystallization-prone carbonates is not a routinely performed
486 type of analysis, even if some studies are reported in literature (Wang et al., 1996; Geyh and Eitel,
487 1998; Deutz et al., 2002; Geyh and Thiedig, 2008; Vogel and Geyh, 2008; Achyuthan et al., 2010).
488 In fact, AMS-¹⁴C dating of calcite forming calcrete horizons might not lead to a straightforward
489 interpretation of results. When calcite precipitates, its ¹⁴C activity is in equilibrium to that of
490 circulating pore water, which, in turn, may not be in equilibrium with atmospheric CO₂ due to the so
491 defined “reservoir effect” (Geyh and Eitel, 1998). This can cause an overestimation of the actual
492 age of calcium carbonate. Moreover, when dissolution and recrystallization occurs, ¹⁴C activity of
493 former calcite is then superimposed to that of the new-generated calcite, in equilibrium with pore
494 water circulating at that time (Geyh and Eitel, 1998). Notwithstanding that, Adamson et al. (1982)
495 conclude that Sudanese soil carbonates are fairly suitable materials for dating. They are also aware
496 of the ineluctability of contamination from some of the older carbonates in some cases; we may
497 confirm this and highlight also the possibility of contamination from younger calcite precipitated
498 after the climate-driven re-opening of the system.
499 Despite these difficulties, in our case study, the well-defined geomorphological,
500 palaeoenvironmental, and archaeological contexts provide valuable information to estimate the
501 consistency of radiocarbon ages on calcrete samples. Moreover, some of these issues can be
502 overcome by the very low amount (~ 10 mg) of sample required by AMS-¹⁴C dating, enabling one
503 to apply an accurate sampling strategy on small portions of the sample, thus separately analysing
504 calcite precipitated in a single pedogenetic event. In this case, the reservoir effect is assumed to be
505 minimal, since circulating pore water is more conceivably provided by the White Nile flooding and,
506 during the Early Holocene, by rainwater rather than fossil groundwater (Dee et al., 2010). In
507 addition to the superficial location of the calcrete-calcitic horizons, the high values of δ¹³C,
508 measured both on cemented nodule and matrix samples, suggest the predominant contribution of
509 atmospheric CO₂ as a source of CO₂ to the groundwater, whereas the contribution of plant-sourced

510 CO₂ can be considered minimal, thus suggesting a sparse vegetation cover (Cerling, 1984; Burns
511 and Matter, 1995; Deutz et al., 2001) during the phases of precipitation.

512 The main issue that has to be taken into account when dealing with radiocarbon dating on
513 pedogenic calcite is the overprinting effect, which is not negligible (Deutz et al., 2002). Calcitic
514 horizons and Calcretes forms over time as results of subsequent dissolution and reprecipitation
515 processes (e.g., (Machette, 1985), thus ¹⁴C activity is time averaged and determination of the time
516 of actual onset of calcite precipitation may be hazardous. As a matter of fact, soil carbonates
517 can be reasonably considered an open system, implying environmental conditions that promote
518 loops of calcite dissolution and reprecipitation. However, when changes in environmental
519 conditions (mostly humidity) lead to the interruption of carbonate accumulation
520 development, the closed system can be hypothesised. Therefore, radiocarbon ages of calcrete
521 samples reasonably indicate the last stage of calcite recrystallization; this may provide valuable
522 information on the timing of Calcrete-Bk horizons development. For these reasons, radiocarbon ages
523 cannot be used according to sample stratigraphy to determine a linear sequence of depositional
524 events leading to calcrete formation; this is also obvious considering the complexity of this
525 nonlinear and multistep process (Wright, 2007). However, micro-sampling should minimize the
526 artificial homogenization of different generations of calcite, and thus radiocarbon ages may provide
527 reliable information on the long calcrete-soil carbonates evolution, even if each sample may still be
528 partially affected by time-averaging blurring effect.

529 ~~Before discussing the significance of the most of radiocarbon ages available for Al Khiday~~
530 ~~carbonates on the basis of statistical analysis (Fig. 7), it is necessary to discuss the significance of~~
531 ~~the single result on allochthonous calcitic pedorelicts, radiocarbon dated to 29540 ± 440 cal. years~~
532 ~~BP. This sample can be interpreted as the reworked remnants of Pleistocene soil carbonates that~~
533 ~~were eroded and transported.~~

534 The considered sequence consists of superimposed calcitic soil horizons, which distinctly developed
535 in phases of enhanced evapotranspiration; consequently, as suggested by Machete (1985), the

536 development of calcitic horizons is a clue to their dissolution-precipitation history. According to the
537 distribution of radiocarbon ages shown in Fig. 9a, the lower part of the sequence (2Bk horizons)
538 may have been affected by old pedogenetic accumulation of calcium carbonate; then, after the
539 burying of this soil and aggradation of the uppermost part of the pedosequence, a further phase of
540 calcite accumulation took place. Its occurrence is recorded by the accumulation of calcitic
541 pedofeatures in the uppermost Bk horizon and the recrystallization or freshly accumulation of
542 calcite in the 2Bk horizon. The higher concentration of calcitic pedofeatures in the 2Bk horizon can
543 be interpreted as the result of subsequent phases of precipitation and/or of a more intense initial
544 accumulation.

545 As for authigenic calcite, in order to correlate the radiocarbon ages of different calcrete
546 pedofeatures to the climatic conditions occurring in the area, radiocarbon dating results were
547 compared with radiometric ages obtained by (Adamson et al., (1982), Williams, (2009), and;
548 Williams et al., (2015) in a cumulative graph of calibrated radiocarbon ages (Fig. 78). Williams
549 (2015) identified several Late Quaternary periods characterized by more humid climatic conditions,
550 corresponding to higher flow level of the White Nile, Blue Nile, and main Nile ~~river~~River. These
551 phases occurred at 14700–13100, 9700–9000, 7900–7600, ca. 6300, and 3200–2800 cal. years BP
552 (Fig. 7b8c). On the bases of published data we may infer several phases of carbonates development
553 at c. >30000, 26000–24200, 18200–13000, and 11000–9100 cal. years BP (Adamson et al., 1982);
554 these ages were obtained on bulk carbonate samples, not separated between matrix and cemented
555 nodules (Fig. 8b).

556 In this study, Tthe oldest radiocarbon ages were obtained on cemented nodules from lower ~~bloeks~~
557 sampling points A3d, A3c, and A3a (11480 ± 240 , 9900 ± 580 , and 9620 ± 100 cal. years BP,
558 respectively), corresponding to dry climatic phases (Fig. 78), according to palaeoenvironmental
559 evidence provided by Williams (2009). These older ages have a quite good correspondence (Fig.
560 8b) with those obtained by Adamson et al. (1982). Younger radiocarbon ages, obtained from
561 powdery calcitic-rich matrix ~~from sampled at~~ A3d and A3c ~~bloeks-points~~ (dated to 7730 ± 80 and

562 7920 ± 100 cal. years BP, respectively) as well as from calcitic-rich matrix ~~from-sampled at~~ A3a
563 ~~block~~ and from calcitic nodules ~~from-sampled at~~ A1 ~~block~~ (dated to 6340 ± 80 and 6080 ± 240 cal.
564 years BP, respectively), show a good agreement with two short periods characterized by more
565 humid conditions, dated to 7900-7600 and about 6300 cal. BP (Williams, 2009; Williams et al.,
566 2015), respectively (Fig. 78).

567 Further evidence that cemented nodules and calcitic-rich matrix possibly formed under different
568 conditions is provided by the stable isotope analysis, in particular by the significant variation of
569 $\delta^{18}\text{O}$ values observed between cemented nodules and matrix samples (Fig. 9a, 9b). Variation in
570 $\delta^{18}\text{O}$ values of calcrete samples generally reflects the variation in isotopic composition of meteoric
571 water; however, the latter may not necessarily reflect a significant variation in $\delta^{18}\text{O}$ of rainfall. In
572 fact, considering the available information on palaeoenvironmental conditions in this region,
573 previously discussed, a $\delta^{18}\text{O}$ enrichment can also be due to evaporative enrichment due to high
574 evapotranspiration rates (Cerling, 1984; Burns and Matter, 1995; Budd et al., 2002). Therefore, the
575 higher $\delta^{18}\text{O}$ values measured on cemented nodules can indicate higher evapotranspiration rates
576 related to drier environmental conditions during calcite precipitation. On the other hand, more
577 negative $\delta^{18}\text{O}$ values, as those measured on matrix samples, can reflect the $\delta^{18}\text{O}$ signature of rainfall
578 or at least a certain $\delta^{18}\text{O}$ depletion, triggered by higher rainfall intensity caused by a short-timed
579 increase of monsoon intensity (Cerling, 1984; Burns and Matter, 1995; Andrews et al., 1998).

580 The consistency of these results suggests that during the Late Pleistocene and mostly initial
581 Holocene a first onset of calcite precipitation occurred during dry climatic conditions, which is
582 compatible with the formation of a thick calcitic horizon in the lower part of the sequenceealcrete.

583 The occurrence of a net moisture deficit in the vadose zone, as well as pore water saturated with
584 respect to calcium carbonate, due to decreased precipitation and the reduction of the White Nile
585 flow, possibly promoted the calcite precipitation. Subsequently, more humid climatic conditions in
586 the Early and Middle Holocene led to partial dissolution of calcite, whereas a subsequent shift
587 towards drier conditions caused the reprecipitation of calcite. Therefore, alternation of wet and dry

588 periods triggered the partial dissolution and reprecipitation of calcite, leading to a progressive
589 calcite accumulation. Cemented nodules may be interpreted as remnants of previously precipitated
590 calcium carbonate, partially dissolved during subsequent humid periods and then reprecipitated as
591 looser calcitic matrix. This hypothesis also suggests that ~~ealere~~calcrete-carbonate pedogenesis occurred as
592 a series of subsequent events of calcite dissolution and precipitation, discontinuously distributed
593 along the Upper Pleistocene and Holocene, driven by local environmental conditions.

594 This interpretation is further supported by ~~the~~ archaeological evidence. Several burial phases were
595 identified and attributed to different periods along the Holocene (Fig. 78), whereas pits, dated to the
596 Mesolithic period, were established after the pre-Mesolithic burial phase (the oldest one).
597 Mesolithic pits were cut into the ~~ealere~~deeper calcitic horizon, whereas pre-Mesolithic and
598 Neolithic human bones are permeated by secondary calcite. In the Meroitic bones, belonging to the
599 most recent burial phase (dated to 2000-1700 cal. BP; (Usai et al., 2010)), pedogenetic calcite is
600 absent, despite the fact that graves were cut within the calcrete horizon and placed deeper from the
601 surface with respect to the older ones. On the basis of these evidence, the mobilization of calcite in
602 the last 2000 years, a period characterized by semi-arid to arid climatic conditions (Gasse, 2000)
603 and lower level of the White Nile (Williams, 2009), was negligible, indicating the occurrence of
604 quiescent stages during calcrete development process under arid conditions, when the amount of
605 circulating pore water is extremely limited.

606

607 **6 Conclusions**

608 In Central Sudan ~~ealere~~, the formation of calcitic soil horizons was a long-lasting process and ~~its~~
609 their development ~~at the site 16D4~~ was significantly influenced by changes from humid to arid
610 environmental conditions. Bk horizons ~~Ealere~~was characterized by alternate periods of
611 growth due to dissolution and precipitation processes, and periods of quiescence or extremely slow
612 growth rates. These events possibly occurred many times along the Quaternary, but the best-
613 preserved evidence is the one related to the Holocene. Despite the fact that radiocarbon dating of

614 calcrete has severe limitations due to the uncertain determination of the contribution of ^{14}C by
615 different sources, accurate sampling method coupled with additional information from a well-
616 defined palaeoenvironmental context may provide a reliable interpretation of the sequence of
617 events. A higher number of carefully selected radiocarbon-dated samples could supply a more
618 statistically significant dataset for radiocarbon ages as well as highlight the variability of AMS- ^{14}C
619 determinations on this type of material. However, radiocarbon ages obtained in the present work
620 confirm the general model for accumulation of calcrete, formation and development of calcium
621 carbonate in sediments/soil horizons established in previous studies (Gile et al., 1966; Machette,
622 1985; Khormali et al., 2006; Zhou and Chafetz, 2009), suggesting a progressive accumulation of
623 calcium carbonate within sediments/soil horizons.

624 Specifically for this context, the interest in deciphering the relationship between calcrete-carbonates
625 pedogenesis and archaeological records concerns the evaluation of the interaction between
626 secondary calcite and archaeological materials. In fact, the occurrence of secondary calcite in
627 archaeological materials may represent a complex issue in all those cases in which calcite may alter
628 conventional radiocarbon dates. An example can be, in the specific case, represented by the
629 radiocarbon dating of archaeological bones performed on bioapatite (the mineral fraction of bone
630 material) (Zazzo, 2014). Calcite is therefore a significant contaminant when measuring ^{14}C activity
631 of bioapatite; despite the chemical removal of secondary calcite, the radiocarbon ages obtained (Dal
632 Sasso, 2015) are in good agreement with those obtained on pedogenic calcitecalcrete, thus resulting
633 substantially younger than those hypothesized on the basis of archaeological and stratigraphic
634 evidences (Fig. 7e8d). Since bioapatite is not a closed system with respect to the environment
635 (Cherkinsky, 2009), carbonate exchange between calcite and bioapatite during burial may
636 significantly affect the result when dating the archaeological bones. Therefore, the study of soil
637 sediments and burial environment is fundamental in order to assess the reliability of radiocarbon
638 ages. A further example of the importance of understanding the time and steps of calcrete genesis in
639 archaeological context concerns the applicability of luminescence dating on archaeological material

640 (pottery); in fact, the dose rate of Thermoluminescence (TL) is heavily tuned by environmental
641 humidity and radiation, and the carbonate content of sediments may influence water mobilization
642 and change the natural radiation due to the increase of uranium occasionally substituting calcium in
643 calcite crystals. In TL dating, radiation is generally supposed to be constant over time, but irregular
644 development of calcrete through time may consistently vary the environmental radioactivity after
645 the burial of archaeological material (Wintle and Murray, 2006).

646 This study highlights the relevance of a multi-analytical approach to the study of ~~calcrete~~carbonates
647 pedogenesis. Specifically for this case, a comprehensive study on the interaction between the
648 archaeological record and the calcrete formation and development supplies valuable information
649 both on the processes and timing of ~~calcrete~~carbonates pedogenesis and on the preservation state of
650 archaeological materials.

651 Considering a wider perspective, the results of this study may represent a further step in the
652 comprehension of ~~calcrete~~calcitic horizons formation and in the definition of a reliable method to
653 date ~~its~~their formation. This may offer a significant tool in palaeoenvironmental studies in arid
654 lands, where carbonatic ~~crust~~concretions may represent the only archive for past climatic changes;
655 in fact, several authors had highlighted the correspondence (climofunction, sensu (Jenny, (1941)
656 between depth of Bk horizons and precipitations (e.g., (Yaalon, 1983; Mack and James, 1992;
657 Caudill et al., 1996; Retallack, 2000; Retallack, 2005), being horizon's depth the results of the
658 average amount of rainfall.

659

660

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672

673 **Conflict of Interest**

674 The authors declare that they have no conflict of interest.

675

676

677 **References**

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950

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965 **Figure 3.** Field photograph of exposed calcrete section sampled for this study (pit 151A). ~~A model~~
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1004 **Figure 78.** Cumulative calibrated dating probability of radiocarbon ages obtained from: **a.** Calcrete
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1009 2015). More humid periods characterized by higher flow of the White Nile (~~dark grey~~light-blue
1010 bars) and the chronology established for the anthropic use of the site at Al Khiday (Salvatori et al.,
1011 2011) (~~light grey~~orange bars – Mesolithic, Neolithic and Meroitic periods) are reported.

1012 **Figure 89. a.** AMS-¹⁴C ages obtained for cemented nodules and calcitic-rich matrix at each
1013 sampling point. The sketch of the studied profile, shown in Fig. 3, is also reported; b., c. C and O
1014 Stable isotope data of for cemented nodules and and calcitic-rich matrix samples.

1015

Table 1

Soil horizon	Depth (m)	Sampling point	Sample depth (m)	Field properties	CaCO ₃ equiv. (%)	pH	Humified C (g/Kg)	LOI (%)	TC (%)	TN (%)
-	0-0.2	-	-	Disturbed layer	-	-	-	-	-	-
Bk	0.2–0.35	A1	0.27	Sandy to sandy–loamy matrix (mostly quartz grains), almost deprived of finer components; locally it showed small lenses of pebbles; common to abundant, large calcium carbonate nodules and concretions; dominant color is 2.5Y 5/4 (light olive brown), scarce mottles 10YR 6/8 (brownish yellow); scarce centimetre scale burrows, filled by brownish yellow sand are common; abrupt transition to the following horizon.	14.77	8.9	0.33	1.95	1.684	0.008
BC	0.35–0.46	A2	0.40	Sandy to sandy–loamy matrix (mostly quartz grains), almost deprived of finer components; scarce, small calcium carbonate nodules and coatings; dominant color is 10YR 6/6 (brownish yellow), scarce mottles 10YR 6/3 (pale brown); few burrows, filled by brownish yellow sand; abrupt limit to the following horizon.	3.76	8.7	1.12	1.64	0.727	0.014
2Bk	>0.46	A3a	0.51	Sandy to sandy–loamy matrix (mostly quartz grains); few, small lenses of pebbles; abundant calcium carbonate concretions; dominant color is 2.5Y 7/4 (pale yellow), scarce mottles in the uppermost part 2.5Y 5/4 (light olive brown); scarce centimetre scale burrows in the uppermost part, filled by brownish yellow sand are common; limit of the horizon not reached.	8.42	8.7	1.04	2.63	1.29	0.019
		A3b	0.61		28.58	8.9	0.35	3.81	3.499	0.015
		A3c	0.73		26.34	8.8	0.61	4.95	3.067	0.016
		A3d	0.84		37.28	8.9	0.37	5.56	4.682	0.015

Table 1. Field properties (color according to Munsell®, 1994) and results of physical and chemical analyses on collected samples.

Table 2

Soil horizon	Depth (m)	Sample name	Sample	¹⁴ C age	2σ cal. BP	δ ¹⁸ O‰ (V-PDB)	δ ¹³ C‰ (V-PDB)
Bk	0.27-	A1-p	Pedorelict	25439 ± 153	29540 ± 440		
			Pedorelict				
Bk	0.27	A1-mt	Calcitic-rich matrix	5290 ± 102	6080 ± 240	-6.25 (0.02)	0.10 (0.01)
			Calcitic-rich matrix			-8.27 (0.04)	0.23 (0.03)
2Bk	0.51	A3a-mt	Calcitic-rich matrix	5536 ± 29	6340 ± 80	-6.70 (0.04)	0.59 (0.02)
			Calcitic-rich matrix			-4.12 (0.06)	0.42 (0.05)
2Bk	0.51	A3a-cn	Cemented nodules	8663 ± 40	9620 ± 100	-4.66 (0.07)	-0.05 (0.06)
			Cemented nodules			-8.03(0.05)	0.58 (0.02)
2Bk	0.61	A3b-mt	Calcitic-rich matrix			-7.72 (0.07)	1.06 (0.05)
2Bk	0.61	A3b-cn	Cemented nodules			-4.98 (0.06)	0.07 (0.02)
2Bk	0.73	A3c-mt	Calcitic-rich matrix	7104 ± 42	7920 ± 100	-7.71 (0.04)	0.75 (0.03)
			Calcitic-rich matrix			-7.60 (0.07)	0.79 (0.04)
2Bk	0.73	A3c-cn	Cemented nodules	8823 ± 242	9900 ± 580	-5.43 (0.04)	-0.18(0.05)
			Cemented nodules			-4.15 (0.04)	0.03 (0.05)
			Cemented nodules			-2.30 (0.08)	-0.66 (0.05)
2Bk	0.84	A3d-mt	Calcitic-rich matrix	6891 ± 31	7730 ± 80	-6.26 (0.03)	0.44 (0.01)
			Calcitic-rich matrix			-6.60 (0.04)	0.21 (0.02)
2Bk	0.84	A3d-cn	Cemented nodules	10005 ± 41	11480 ± 240	-3.32 (0.04)	-1.48 (0.03)
			Cemented nodules			-1.85 (0.05)	-0.75 (0.03)

Table 2. ¹⁴C ages, calibrated ages, δ¹⁸O and δ¹³C values and their standard deviation obtained on selected calcitic pedofeatures (p: Pedorelict; mt: Calcitic-rich matrix; cn: Cemented nodules).

Figure 1

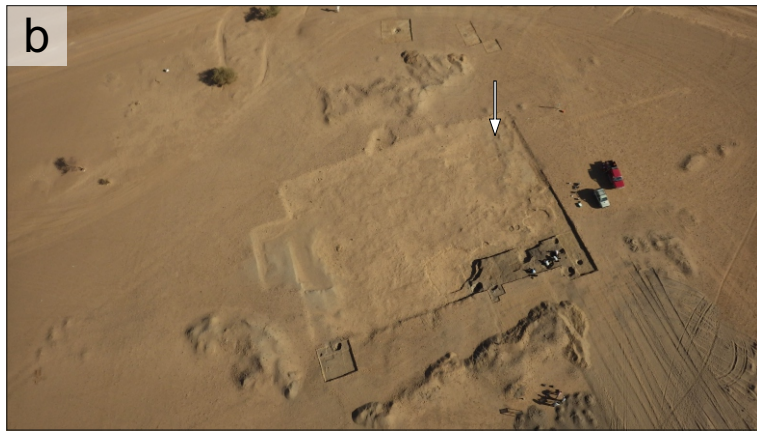


Figure 2



Figure 3

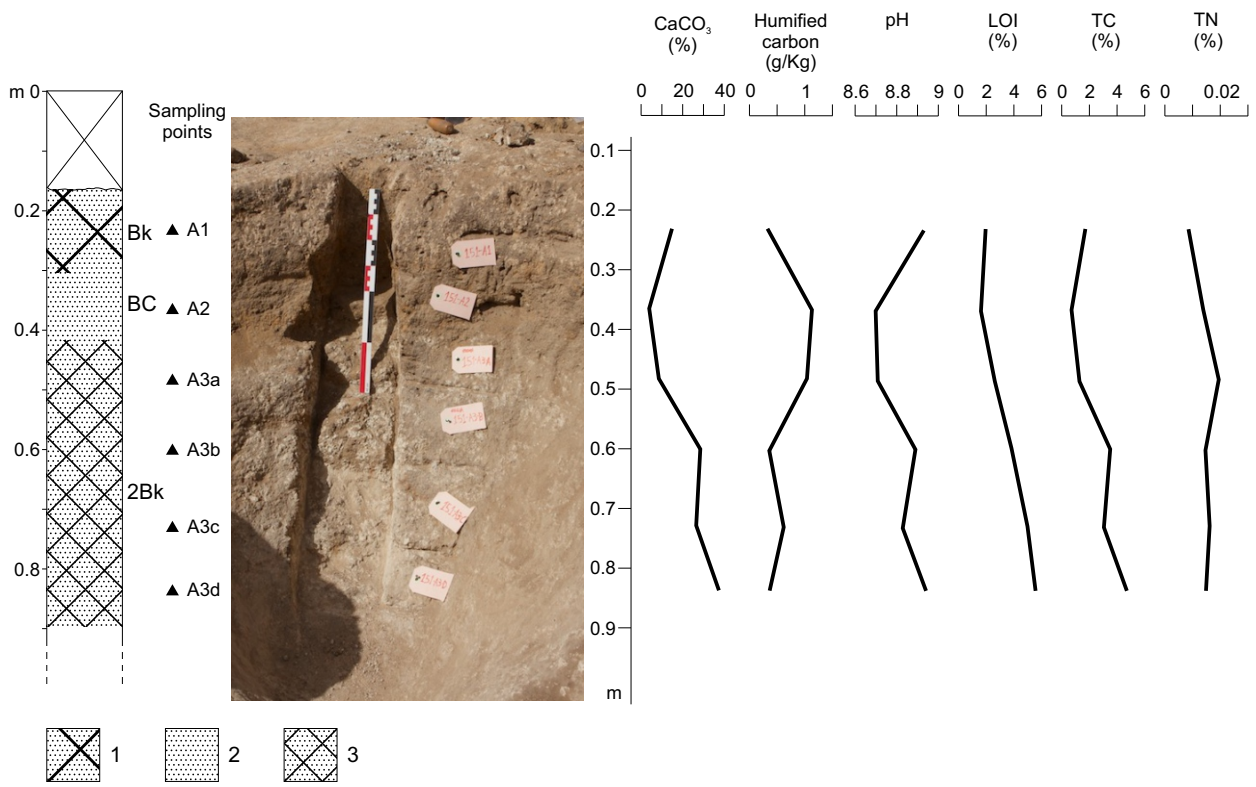


Figure 4

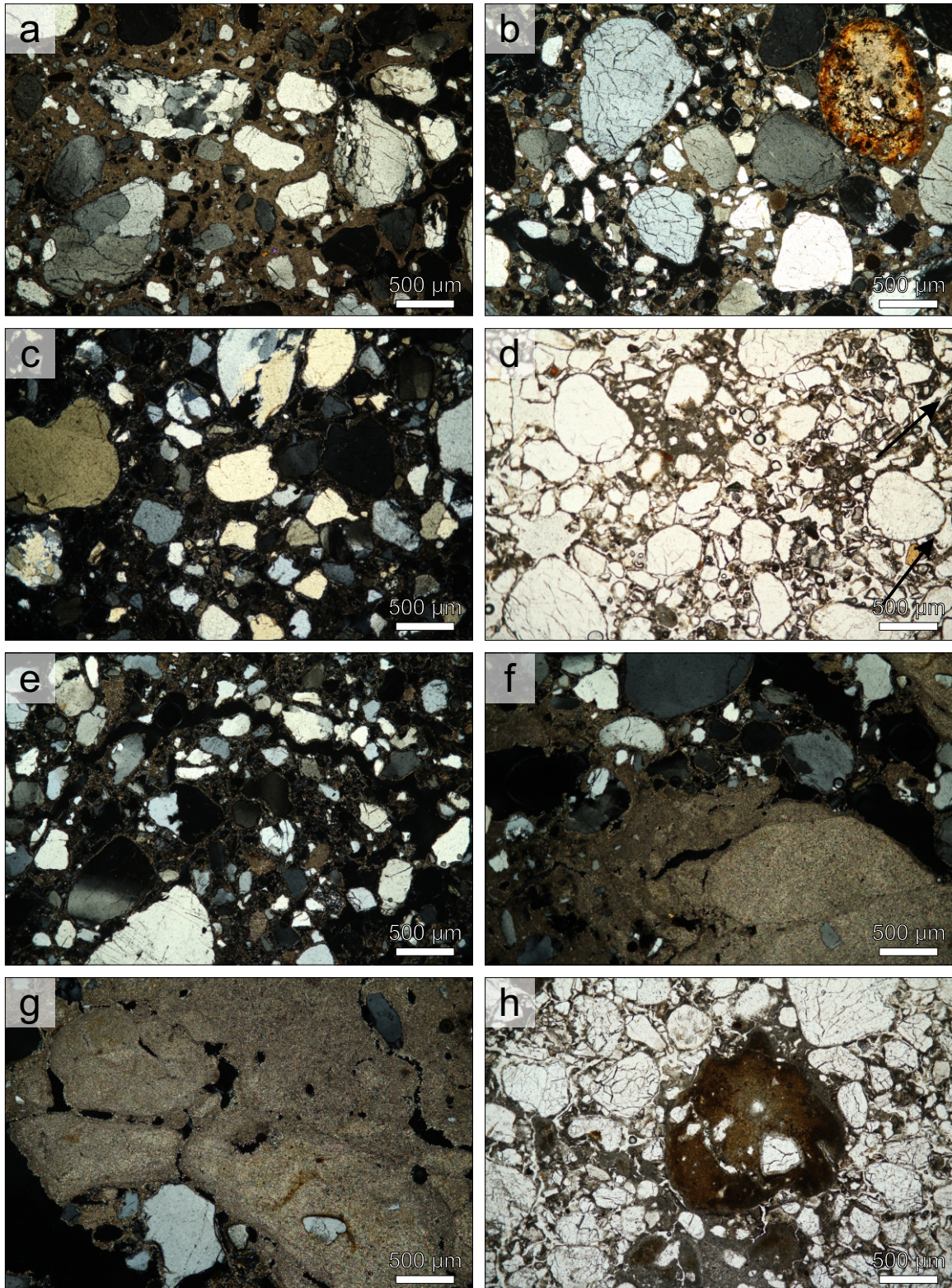


Figure 5

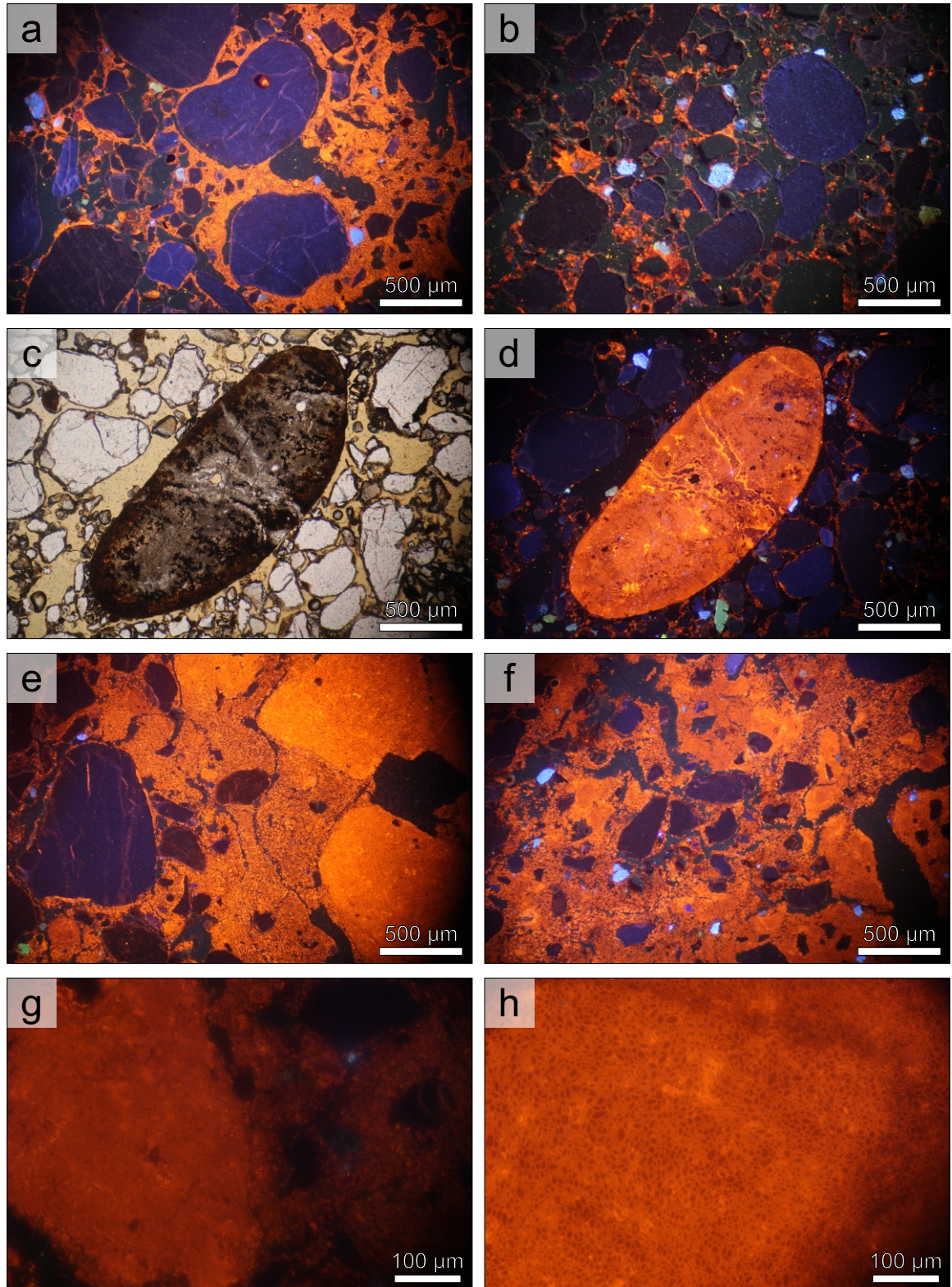


Figure 6

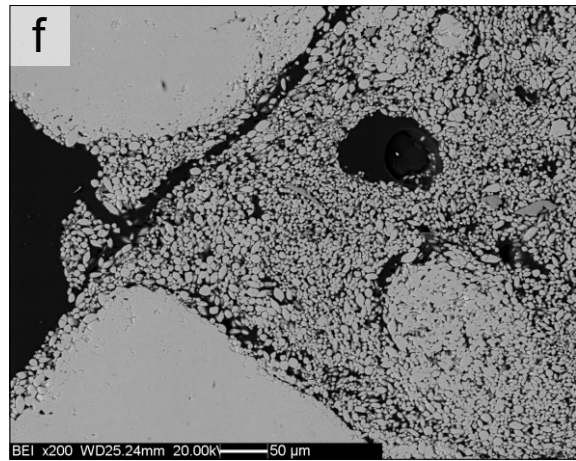
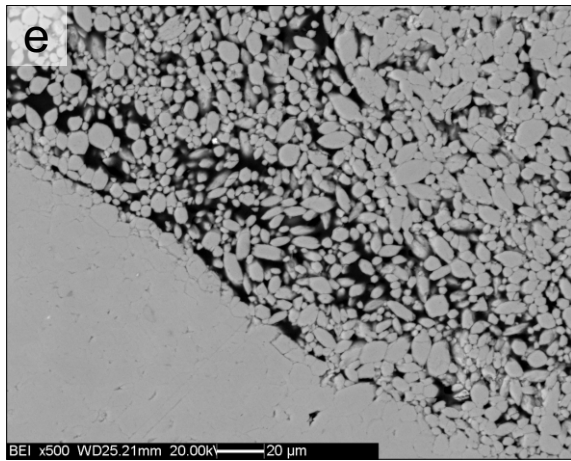
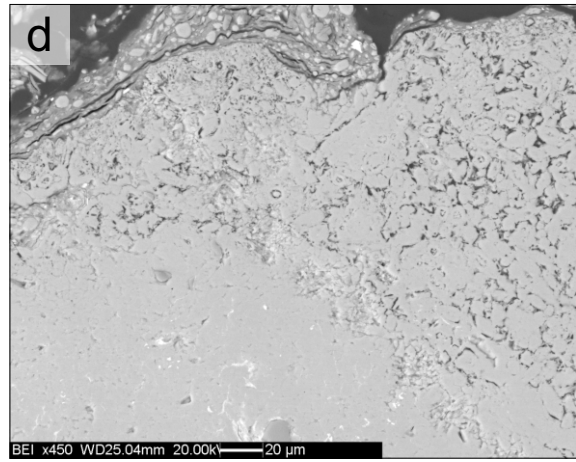
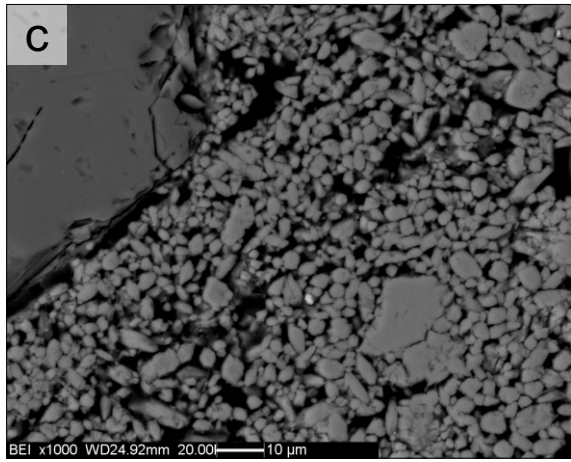
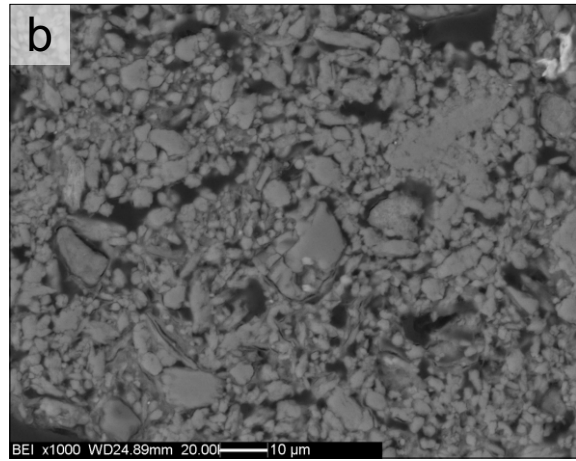
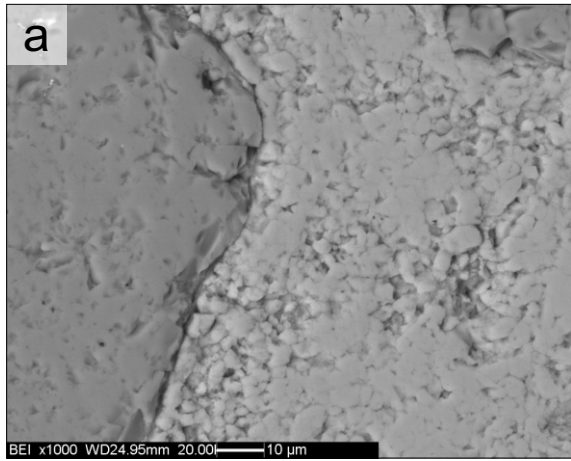
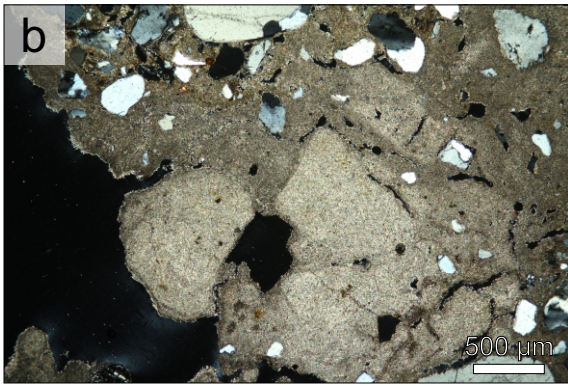
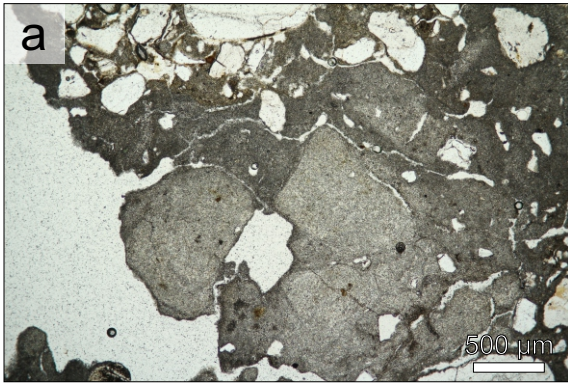


Figure 7



- | | |
|--|--|
|  Voids |  Calcitic-rich matrix |
|  Quartz |  Cemented nodules |

Figure 8

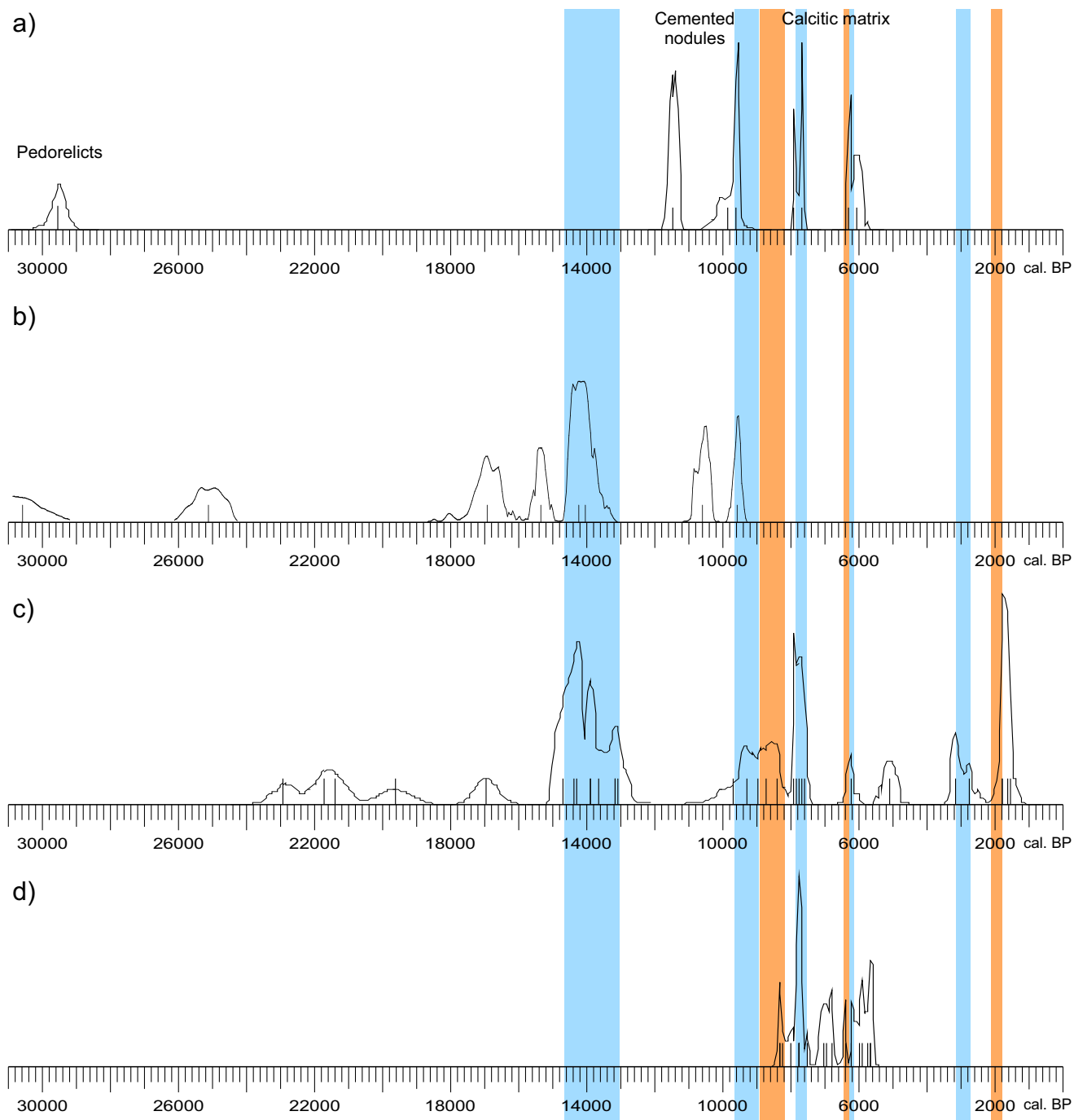
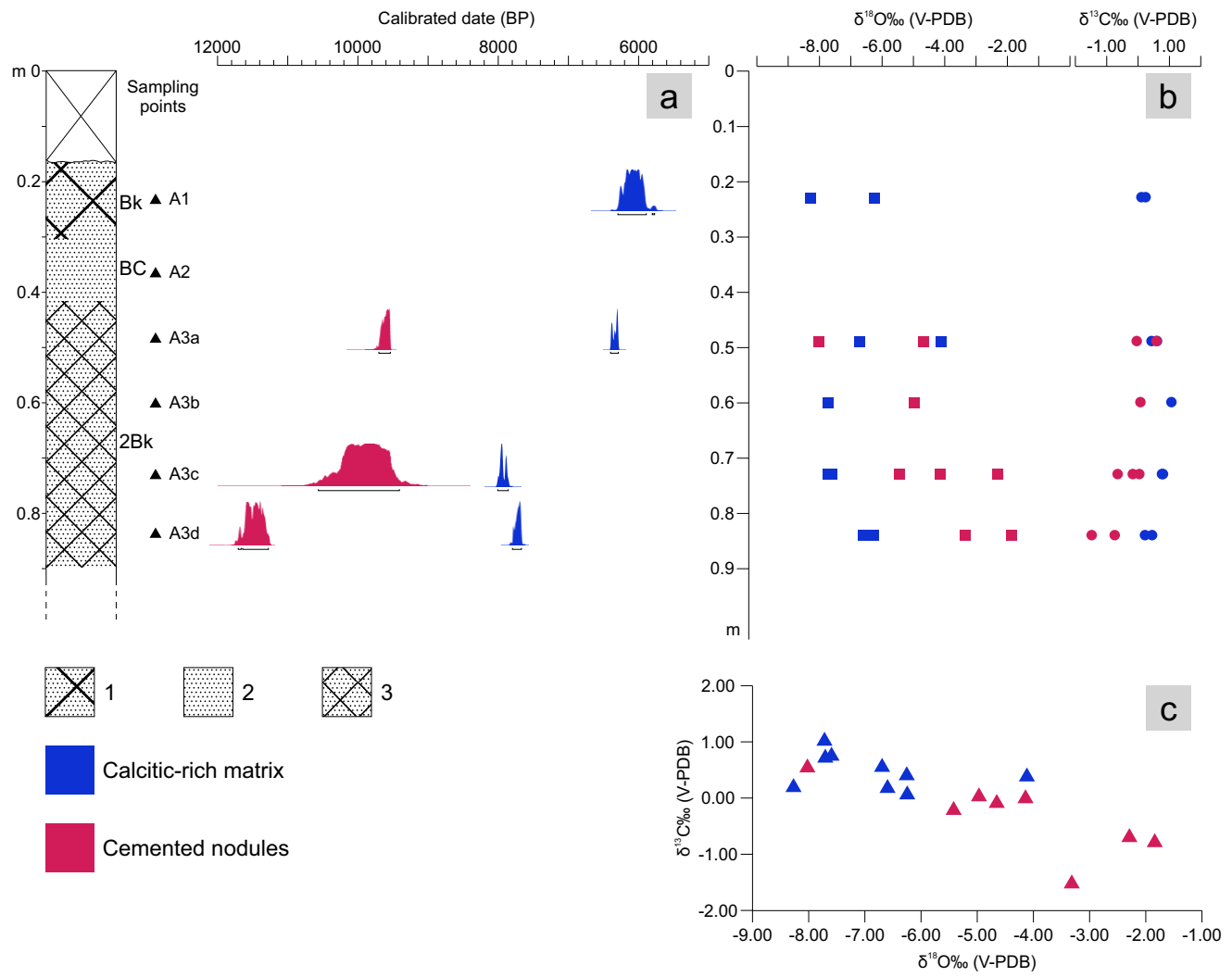


Figure 9



Interactive Map file (.kml or .kmz)

[Click here to download Interactive Map file \(.kml or .kmz\): 16D4.kmz](#)