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Lake Chad sedimentation and environments during the late Miocene and Pliocene: new evidence from mineralogy and chemistry of the Bol core sediments

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Abstract (164 words)

This study presents mineralogical and geochemical data from a borehole drilled near the locality of Bol (13°27'N, 14°44'E), in the eastern archipelago of the modern Lake Chad (Chad). Samples were taken from a ~200 meters long core section forming a unique sub-continuous record for Central Africa. Among these samples, 35 are dated between 6.4 and 2.3 Ma. Dominant minerals are clays (66% average) mixed with varying amounts of silt and diatomite. The clay fraction consists of Fe-beidellite (87% average), kaolinite, and traces of illite. Clay minerals originate from the erosion of the vertisols that surrounded the paleolake Chad. Sedimentological data indicate that a permanent lake (or recurrent lakes) existed from 6.7 until 2.3 Ma in the vicinity of Bol. By comparison with modern latitudinal distribution of vertisols in Africa the climate was Sudanian-like. Changes in the sedimentation rate suggest a succession of wetter and dryer periods during at least six million years in the region during the critical time period covering the Miocene-Pliocene transition.

Key words: Lake Chad, Miocene-Pliocene, Fe-beidellite, vertisol, sedimentation rate

1 Introduction

Lake Chad is a permanent and shallow freshwater body located in the Sahelian domain of Africa that fringes the southern edge of the Sahara desert (Fig.1). It is today mostly supplied from its southern watershed by the Chari-Logone rivers system. Lake Chad is a very sensitive indicator of climate and environment changes in North-Central Africa as illustrated by its dramatic recent and past shrinkage in area (Maley, 1972, 2010; Maley and Vernet, 2015). During the 1960's, Lake Chad covered 25000 km^2. It decreased less than 1500 km^2 during the 1980's (Olivry et *al*, 1996; UNEP, 2004; Don-Donné Goudoum and Lemoalle, 2014), whereas during the Holocene, 6000 years ago, it reached >350000 km^2 (Schuster et al., 2005) (Fig.1).

The Chad basin is an intracratonic sag basin, whose margins correspond to the maximum expansion of the lake during the Holocene (Schuster *et al.*, 2005; Leblanc *et al.*, 2006). The basin basement includes a suite of crystalline rocks related to the Pan-African orogeny (ca. 750-550 Ma), which is directly covered by Cretaceous sandstones (Bessoles and Trompette, 1980). On top of the Cretaceous deposits, it also includes Neogene and quaternary sediments of about 500 m thick covering an area of about 500 km in diameter (Burke et *al*., 1976).

Scarce sedimentological data coming from petroleum exploration in Niger, Central African Republic, and Chad (Genik, 1992) give a brief history of the filling of rift basins in Central

Africa, which were since completed by significant sedimentological investigations at the hominin-bearing deposits of northern Chad (Brunet et *al*., 1995, 2002, 2005). Regarding southwestern Chad, Genik reports the late Miocene in the Doba and Dosea basins as thick (200 to 800 m) non-marine sandstones, while Kusnir and Moutaye (1997) described very briefly the central basin sedimentary series as formed by Cretaceous sandstones followed by a sandy early Pliocene and a limnic argillaceous middle and late Pliocene. More recently, Swezey (2009) presented three stratigraphic sections described by Schneider and Wolf (1992), where the Pliocene sediments are briefly described as mudstones more or less diatomeous and gypsiferous. In the northern part of the basin, multiple fossiliferous sediment series were described (Schuster et *al*., 2006, 2009) and overall dated between 7.5 and 1.1 Ma using the $^{10}Be/^{9}Be$ method (Lebatard et *al*., 2010). They consist of many sequences of lacustrine (clays and diatoms), perilacustine (argillaceous sandstones with root concretions, rhizoliths and abundant termite nests) (Duringer et *al*., 2006, 2007), and aeolian deposits, suggesting successive and repeated wet and dry climatic periods during the Miocene-Pliocene. Perilacustrine sediments have yielded a quantity of fossil vertebrate remains including fishes, turtles, crocodiles, birds, and mammals among which two new species of hominins, so-called *Sahelanthropus tchadensis* and *Australopithecus barhelghazali* (Brunet et *al*., 1995, 2002, 2005) (Fig.1).

The discovery of early hominins in northern Chad has notably reinforced some people's interest in the region and its environmental past. Indeed, understanding human evolution implies to capture the environmental context of hominin occurrences in the fossil record, and to determine the factors that may have influenced their repartition and evolution through time (e.g. Levin, 2015). Many studies led in northern Chad have thus far focused on documenting the paleoenvironment at the key moments of hominin occurrences (e.g. Zazzo *et al.*, 2000; Boisserie *et al.*, 2005; Vignaud *et al.*, 2002; Jacques, 2007; Blondel *et al.*, 2010; Otero *et al.*, 2010; Pinton *et al.*, 2010; Lee-Thorp *et al.*, 2012; LeFur *et al.*, 2009, 2014) without really contextualizing their results in the broader background of paleoenvironmental and paleoclimatic changes in North-Central Africa during the Neogene. The same studies provided evidence of mosaic environments including forest patches, woodland, and grasslands in close relationship with aquatic areas at the time of *S. tchadensis* occurrence (ca 7 Ma) (Boisserie *et al.*, 2005; Vignaud *et al.*, 2002; Jacques, 2007; Blondel *et al.*, 2010; Otero *et al.*, 2010; Pinton *et al.*, 2010; LeFur *et al.*, 2009, 2014), and described the environment of *A. barhelghazali* as more open and dominated by C_4 vegetation (Zazzo *et al.*, 2000; Lee-

Thorp *et al.*, 2012). A few studies, conversely, have addressed the questions of the evolution of paleoenvironments in Chad by comparing data on a deep-time scale from the end of the late Miocene to the late Pliocene (Otero *et al.*, 2011; Novello *et al.*, 2015a, 2015b). Oxygen isotope analyses conducted on fish teeth suggest a trend toward increased regional aridity between 7 and 3 Ma in northern Chad, including a more abrupt shift at the Miocene-Pliocene transition during the time spanning across the Messinian salinity crisis in the Mediterranean region (Otero *et al.*, 2011). A multi-proxy approach was recently performed on a new sedimentary record related to a core drilled close to the locality of Bol, which is located at the limit between the northern and southern parts of the Lake Chad basin, in the northeast archipelago of the current Lake Chad (Fig.1). This sub-continuous sedimentary archive was dated between 6.3 and 2.3 Ma using the $^{10}Be/^{9}Be$ method (Novello *et al.*, 2015a, 2015b), and it therefore gives the opportunity to document the paleoenvironment and paleoclimate evolution of this part of the Lake Chad basin on a deeper time scale than the sporadic Miocene and Pliocene series of northern Chad. The diatom assemblages of this record reveal the existence (even interrupted) of lacustrine conditions at 13°N in Chad since at least 4.7±0.1 Ma, while phytoliths and pollen support the presence of grass-dominated environments in the area of Bol and probably further south in the southern part of the basin during all the Pliocene (Novello *et al.*, 2015a, 2015b). A decrease of lacustrine conditions was suggested between 3.6±0.1 Ma to 2.7±0.1 in the vicinity of Bol and deduced from the scarcity of diatom remains and the increase of marsh indicator phytoliths in the sequence (Novello *et al.*, 2015a, 2015b). This last result corroborates three sudden aridity events recorded successively in north-western Africa at about 3.5, 3.2, and 2.8 Ma (Leroy and Dupont, 1994, 1997) and suggests that dry conditions were extended all over North-West and Central Africa between ~3.6 and 2.7 Ma.

Here, we present the results obtained after a series of mineralogy analyses performed on the Bol core sequence (Moussa, 2010; Moussa *et al.*, 2013). This study is a valuable complement to the previous micro-biological data published in Novello *et al.* (2015a, 2015b), by providing new clues about the Lake Chad history, and about the paleoenvironment and paleoclimate in Central Africa from about 6 Ma. It also permits to recall the information already yielded in Novello *et al.* and to compare all the different types of data produced in order to enhance the discussions and paleoenvironmental interpretations.

2 Material

The samples studied are all cuttings, associated with a 673 m long hydrogeological core drilled in 1973 by the "Bureau de Recherches Géologiques et Minières" (BRGM, France) on the northeastern shore of Lake Chad, near the locality of Bol (Fig.1) (13°27'N, 14°44'E). The core reached the basement (Precambrian metamorphic rocks: gneiss, leptynite) at 673 m depth (Fig.2a). Cuttings and some core sections have only been preserved from between 70 and 300 m depth since the drilling. Below that, only the lithological log description of the core is still available. According to original data producing during the coring, the core can be divided into three major segments from the top to the bottom, which reflect dominant lithology (Fig.2a):

- From 0 to 71.5 m: eolian sands;
- From 71.5 to 330 m: pelites more or less mixed with lacustrine diatomites;
- From 330 to 673 m: silt/sand and clay alternation.

The–eolian sand formation is related to Kanem dunes deposited during the Last Glacial Maximum (Servant, 1983; Maley, 2010). Some pelitic samples, located between 71.5 m and 297 m, were dated using the $^{10}Be/^{9}Be$ (Fig.2, Table S1) (Novello et *al.*, 2015a, 2015b). These sediments were deposited during the late Miocene and the Pliocene. By comparison with sediments described in the northern part of the Chad basin (Vignaud et *al.*, 2002; Schuster 2002, Schuster et *al.*, 2005; Duringer et *al.*, 2006), the fine sands and clays occurring between 330 m and 580 m can be attributed to the late Miocene. The basal coarse sands may be deposited from Eocene to Miocene (Servant-Vildary, 1978; Servant 1983).

This study focuses on the second core segment (71.5-297.2 m), which is the only sampled part of the core still available. The samples studied were taken in the core to about every 2 meters from 71.5 m to 170 m and then every 6 meters from 170 m to 297.2 m (Fig.2, Table S1). The micro-biological remains (diatoms, phytoliths, and pollen) of these samples were previously studied (Novello et *al.*, 2015a, 2015b).

3 Methods

The continuous lithology of the core (Fig.2b) was reconstructed using the core lithological log description and our direct observations of the samples. All of the 126 available samples were examined using binocular glasses and smear slides. Samples mostly consist of angular core fragments, but also in rare core sections (9 cm of diameter and 5-7 cm of height). The mineralogy of 64 samples was semi-quantified using powder X-Ray Diffraction (XRD), and then ten samples, regularly distributed along the core and previously dated (Novello et *al.* 2015a, 2015b) (except sample 01d), were studied in details by XRD and by chemical

analyses. XRD patterns were obtained with a Philips X'Pert PRO equipped with a Ni-filtered CuKα radiation generated at 40 kV and 40 mV. A <1/4 2θ anti-divergence slit and a 1/2 2θ anti-diffusion slit were used, step size is 0.017 and step time is 30 s. Scans were taken between 2.5 and 65° for randomly oriented powder. The XRD patterns were obtained from powders and oriented preparations of Na, K and, Ca-saturated samples in the air-dried state, after ethylene glycol solvation, hydrazine solvation, and heating (350ºC, 4 h). Semi-quantification of minerals amounts was obtained using the relative areas of the major peaks on powder patterns. Local chemical analyses of bulk samples were performed on core fragments (from 0.5 to 1 cm in diameter) with a JEOL JSM-5600LV scanning electron microscope (SEM) equipped with an EDX system (Bruker AXS Microanalysis). The clay fraction (<2μm) of the ten samples was analyzed for major elements and trace elements at Nancy SARM, using the ICP-MS method (Carignan et *al*. 2001).

4 Results

4.1.Lithological description

The studied samples range from claystones to siltstones. These are laminated but not varved, which gives a succession of millimetric to centimetric lamina of clays, silts/sands, and diatomites (Fig.2b). The major component is grey to light green clays. These clays occur as pelites, which are more or less mixed with silts/sands and/or diatomites. White diatomites are generally mixed with clays and/or silts/sands.

All samples consist of fine grained clayey sediments. The largest grains observed consist only in quartz grains (coarse sand) of 1 to 2 mm of diameter. Most of quartz grains, conversely, are between 200 and 500 μm and with a river transported grain morphology. Few amounts of typical aeolian quartz grains are also observed and they are always mixed with other quartz grains in some samples (2f, 8j, 25a, and 17h notably) (Table S1). Ovoid pellets of indurated mud, from 1 mm up to 1 cm, occurred in samples 21j and 35e (Table S1).

Laminated structures are observed in the samples studied all along the core. Most often millimetric lamina of clays alternate with some diatomites or silt/sand lamina. Silt/sand layers are never more than 5 mm thick. In few samples (18h, 12b, and 9e) (Table S1) few millimeters thick layers of diatomite and clays alternate.

Between 90.8 m to 92.3 m, and 100.3 to 103.7 m, all samples (5a to 5m, and 7f to 8g) (Fig.2b) include tubular holes of several centimeters long and millimetric in diameter cross the

sediments. These holes are coated by brown to red very thin deposits of amorphous iron oxy-hydroxides. These features are similar to rizoliths observed in modern soils.

In the available samples no sedimentary features like cross bedding, mud-cracks, and erosional surface are observed.

4.2. Sample mineralogy

4.2.1.Recurrent minerals

According to powder XRD data of the whole rock samples (Fig.3), the main and recurrent occurring minerals are smectite (001 reflection at 15.3 Å), kaolinite (001 reflection at 7.2 Å), illite (001 reflection at 10 Å), quartz (main reflection at 3.34 Å), K-feldspars (main reflection at 3.24 Å), and anatase (main reflection at 3.52 Å). The relative amounts of these minerals were semi-quantified (Fig.4). Clays are the most important minerals all along the core. Their relative abundance ranges from 50 to 85 %, with a mean value of 66%. Quartz is the second mineral represented, ranging from 6 and 70 % (mean value of 27%), while K-feldspars and anatase are always detected in low amounts (mean values close to 4 and 5 %, respectively). The whole mineralogy reflects the relative amounts of the silt and sand fractions (quartz, k-feldspar, and anatase) in sediments dominated by a clay fraction. XRD diagrams of ovoid pellets of indurated mud in samples 21j and 35e are similar to the ones of the surrounding pelites.

4.2.2.Sporadic minerals

Opale CT. Biogenic silica is generally made of opal-A, which has a disordered, nearly amorphous structure. It exhibits only a broad band between 19 and 25 °2θ on XRD patterns (DeMaster, 2003), which is overlapped by the dissymmetric (02-11) reflection of clay minerals. Therefore it was not possible to detect and quantify the amounts of opal-A resulting from the occurrence of diatoms and phytoliths in sediments (Novello et *al*, 2015a, 2015b). In the lower part of the core (samples 30j to 35e, from 251 m to 297 m depth), opal- CT was detected (major peak at 4.03 Å) (Fig.3). According to XRD data, the amounts of opal-CT are ranging between 20 and 40%. Opal-CT is a well-known product of the early diagenesis of opal-A (DeMaster, 2003). The biogenic silica of diatoms and phytoliths was transformed in opal-CT by diagenetic evolution of the deeper samples of the core.

Gypsum (mean peak at 7.59 Å) was detected as traces in most samples but in large quantities in samples 7f, 8a, 8e, 8o, 15h, and 21a-e (Table S1). In these samples, gypsum crystals occur as millimetric elongated laths. This crystal morphology suggests a diagenetic origin for gypsum.

Calcite and dolomite. Carbonates were rarely detected and in very low amounts (samples 1a-1d, 11d, 20b, 22n, and 30j, Table S1), except in sample 6a (10 % of calcite and 5% of dolomite). No shell fragments were observed.

Apatite. Apatite occurs as minor component in samples 5g, 6d-h, 9c, 10b, 16i, 17h, and 21j (Table S1). Apatite source is likely to be related to vertebrate fossil fragments such as fish-bones.

Jarosite (mean peak at 3.07 Å) was detected only in sample 29a (Table S1). Jarosite is a basic hydrous sulfate of iron and potassium ($KFe^{3+}_3(OH)_6(SO_4)_2$) occurring in sulfate rich environments, most often resulting from the oxidation of pyrite in acidic environments (Stoffregen et *al*., 2000). Here it is more likely that jarosite has a diagenetic origin.

4.3.Detailed studies of clay minerals

4.3.1.Relative amounts of clays

Clay minerals of samples were carefully studied using the <2 µm fraction. (Fig.5). Smectite, with a mean value of 87%, is the main clay occurring in the <2 µm fraction all along the core. The (001) peak of smectite is especially very broad in the samples having the lowest amounts of this mineral. This feature is due to a very low amount of stacked layers (about 2 layers) of smectite, which makes its quantification difficult and possibly underestimated. Kaolinite is the second clay of the <2 µm fraction, with a mean value of 12%. Illite is a minor component but it is always present in detectable amounts while quartz always occurs in very low amounts.

The clay mineralogy appears quite constant all along the core, with a light increase of smectite at the top of the core.

4.3.2.Detailed studies of ten selected samples

XRD

The (001) reflection located at 15.5 Å in air dry condition shifted to 17.8 Å after ethylene-glycol treatment, and fell down to 10.0 Å after heating at 350°C (Fig.6). This behavior is typical of a smectite without interstratification and excludes the occurrence of chlorite (Brindley and Brown, 1980). A (001) reflection at 15.5 Å in air dry condition indicates the occurrence of two water molecules in the interlayer of the smectite, and Ca as the main exchangeable cation. The (06-33) reflection of the smectite ranges from 1.50 to 1.51 Å, which is characteristic of a dioctahedral layer (Brindley and Brown, 1980). After the Hofmann-Klemen test (Brindley and Brown, 1980), the (001) reflection of the smectite is at 18 Å. This feature indicates that its layer charge originates, at least partly, from a tetrahedral charge, which is characteristic of beidellite-like smectite. After successive saturations of samples by Ca, K, and then by Ca and ethylene-glycol treatments, the (001) reflection of the smectite is at 18 Å. This behavior is typical of low to medium charge smectite layers (Brindley and Brown, 1980).

After hydrazine saturation, the (001) reflection of kaolinite partly remains at 7.14 Å while another part shifts to 10.4 Å (Fig.7). This indicates a mixture of ordered and disordered kaolinite crystals. The amount of disordered kaolinite crystals ranges from 41 to 85 % (Table 1).

No difference was found between the XRD patterns of clays in pellets and surrounding pelite for samples 21j and 35e (Table S1). Pellets are due to a reworking of pelite (rip-up clasts).

Chemical analyses

Two sets of data were collected: bulk analyses of the <2 μm fraction of samples (Table 2) and local analyses using a scanning electronic microprobe (Table 3). Both data gave consistent results. The mineralogy of the <2 μm fraction consists of smectite, kaolinite, minor illite, and quartz. The variations in SiO_2 contents are linked to the amounts of quartz and kaolinite. Because natural kaolinite crystals contain low amounts of Fe_2O_3 (Mestdagh et *al*, 1980) and illite is a minor component, the major part of Fe_2O_3 is therefore represented by smectite. Between 10 and 20% of the Al_2O_3 amounts come from kaolinite. The MgO contents are low and essentially related to smectite. As illite is a minor component of samples, a part of K_2O is associated with the smectite phase. According to these data, the smectite present in all the samples has a chemistry characterized by large amounts of Al and Fe, and by low Mg and K contents. It was however not possible to establish the structural formula of the smectite because of the complex mineralogy of the samples.

The chemistry of the <2 μm fraction in ovoid pellets (samples 21j and 35e) (Table S1) is very close to that present in the surrounding pelite (Table 2). The higher amount of SiO_2 in sample 35e° is probably due to its higher quartz composition.

SEM-EDX measurements did not allow obtaining the chemistry of isolated smectite particles. At the micron size scale, smectite, kaolinite, and quartz particles are intimately joined. Al, Fe, Mg, and K ratios measured by SEM-EDX are similar to those obtained by bulk chemical analysis (Table 3).

Rare earth elements (REE) diagrams of the <2 μm fraction (Fig.8a) are very similar between the ten samples. Similarities are also observed between all the samples for the extended diagrams of trace elements (Fig.8b).

The smectite component

Smectite is the main mineral of the clay fraction (mean value 87 %) and the main component of all the samples (main value 57%). Among the various kinds of smectite minerals existing, the smectite occurring in the Bol sediments possesses a dioctahedral feature and a tetrahedral charge, in addition to contain large amounts of iron. These are all characteristics of a Fe-beidellite (smectite) (Brindley and Brown, 1980).

5 Discussion

5.1.Source of the deposits

The recurrent occurrence of freshwater organism remains (diatoms, sponges), and notably the dominant freshwater diatom species *Aulacoseira granulata* (Novello et *al.*, 2015a, 2015b) indisputably suggests the existence of true lacustrine environments at Bol during the Miocene-Pliocene. Quartz grain morphology indicates that the major inputs of quartz to the lake originated from fluviatile transport while a few parts originated from aeolian transport only. Comparably, pollen spectra of Lake Chad also indicate significant fluviatile contributions to the lake during the Holocene (Maley, 1972, 1981) and still today (Maley, 2010).

The dominance of finely laminated clays among the sediments indicates deposition by settling. REE diagrams of the clay fraction (Fig.8a) are similar to diagrams of detrital sediments derived from post archean continental crust, with notably similar La_N/Yb_N and Eu/EU** values all along the core (McLennan, 1989). The relatively high values of La_N/Yb_N

are typical of claystones (McLennan, 1989). The similarity of REE and extended trace elements diagrams (Fig.8b) between all the samples suggests that the same basement rock was eroded and then deposited to feature in the Bol sequence. Trace element diagrams strongly suggest a detrital origin for the clay minerals and notably the Fe-beidellite. Authigenic pelloidal nontronites are not observed in the Bol sediments while they are present in modern lake Chad sediments sampled in the Chari Logone delta (Pedro et *al*., 1978). This last result coupled with the lack of cross beddings (at the scale of the core) and the absence of coarse sand deposits indicate that Bol was far from any major river delta during the total period of sediments' deposition. The changing proportions between pelite, silt-sand, and diatomite result principally from the variation in time of the rivers discharges in the Chad basin.

Today, the detrital sediments deposited in the Lake Chad basin are essentially transported by the Chari-Logone system (Fig.1) and mostly originate from the erosion of the southern part of the watershed, which is characterized by important reliefs (the Adamawa mounts, ~1900 m above the sea level) and by mean annual rainfall of 1200 mm (Olivry et al., 1996). This part of the drainage basin is currently covered by ferralitic and tropical ferruginous soils, in which kaolinite is the dominant clay (Gac 1980). The suspended clays in the Chari river, near Lake Chad, are indeed dominated by kaolinite. A smaller part of the current sediments deposited in the Chad basin also comes from the erosion of the lowlands located in the north of the Chari-Logone watershed (Gac, 1980), which are essentially covered by vertisols dominated by smectite (Fe-beidellite) clay minerals (Paquet, 1970). As a result, the current ratio kaolinite/illite/smectite in the modern Lake Chad sediments is close to 63/19/18 % (Carmouze, 1976; Carmouze et *al*., 1977; Gac, 1980; Gac et *al*, 1977). In the Bol sediments, the smectite is conversely largely dominant, with a ratio kaolinite/illite/smectite equivalent to 12/1/87 %. Paquet (1970) observed that kaolinite has a higher crystallinity in ferralitic soils of Central Africa than in vertisols in general. In the Bol sediments, badly crystallized (disordered) kaolinites occur in higher abundance than well crystallized kaolinites (Table 1). This last result suggests a partial sedimentary contribution from ferralitic soils of the southern part of the Chad watershed during the Miocene-Pliocene, while most of the contribution was originated from the erosion of vertisols of the lowlands. A higher contribution from the southern highlands than today was suggested by Novello *et al*. (2015a, 2015b) as an interpretation of the significant percentage of Afromontane pollen at 4.2 ± 0.1 Ma and the unexpected occurrence of C_3 grass indicator phytoliths in the Bol sediments. Yet, none of the

mineralogy data support a trend towards an increase of highlands contribution during the Miocene-Pliocene.

5.2.Lake morphology and dynamics

Similarities between ovoid pellets of mud and their surrounding matrix at 3.7±0.1 Ma and 6.4±0.1 Ma (samples 21j and 35e) suggest a clay rip-up clasts origin. These pellets may have been produced after the sediments eroded and deposited in the lake, probably in a context of lake regression. Hence, this would suppose rapid changes of the lake water levels at Bol during the end of the late Miocene and during the mid-Pliocene. Moreover, it is most likely that Bol was in the nearshore zone of the lake during these periods which would explain it recording such abrupt variations of the lake level. The closeness of Bol to the lake and the low lake level hypotheses during the end of the late Miocene is consistent with the phytolith assemblage observed at 5.5±0.1 Ma (sample 30j), which includes about 13% of phytoliths indicators of palms (terrestrial obligate plants), against a mere <2-5% in the other samples of the core (Novello *et al*., 2015a, 2015b). Later on between ~2.6 and 2.4 Ma, rhizoliths are observed, suggesting again that Bol was close to the lake's shore. Given the lack of true pedogenesis features in the samples, however, it is likely that these rhizoliths were associated with aquatic plants in a shallow water environment (marshy vegetation).

Hydrous sodium silicates including magadiite, kenyaite, and zeolites were described in the N'Guigmi interdunal depressions of Lake Chad (Sebag et *al*., 2001). These sodium silicates were formed from brines during the Holocene and indicate a strong evaporation rate of Lake Chad during the last 10,000 years. No such silicates were observed in the Bol samples, suggesting therefore the lack of complete, or else near complete, evaporation at Bol. Bio-silica remains (opal-A), including lacustrine indicators (diatoms and sponge spicules), were not observed in the deeper part of the core (<5.5±0.1 Ma) (Novello et *al*., 2015a, 2015b), while opal-CT was detected (this study). The siliceous remains initially made of opal-A may have been transformed into opal-CT by diagenesis, preventing their direct recognition by the diatom and phytolith specialists. From 6.4 to 2.3 Ma, therefore, water seems to have been always present at Bol even during periods of the lowest lake levels which favored marshy vegetation (Novello *et al.*, 2015a, 2015b).

The relative amounts of clays, silt/sands, and diatomites in the Bol sediments can be interpreted as indicating changes in the runoff intensity during Miocene-Pliocene. During a full year, the amount of detrital sediments transported to the current Lake Chad by the Chari-

Logone system is positively correlated with the intensity of rainfall in the drainage basin (Gac, 1980). We assumed that such a correlation had already existed during the Miocene-Pliocene and hypothesized the following scenario: during rainy periods, a severe erosion of the river watershed may have occurred, leading to the transport and then deposition of clays and silt-sands, mixed with diatomites; during periods of weak erosion of the river watershed (and then more likely dryer conditions in the drainage basin), which are also correlated with lower lake levels, diatomites may have been deposited (Lemoalle, 1978). This last hypothesis refers to the observation of significant bloomings of diatoms during periods of low Lake Chad levels and increasing aridity in the basin during the 1970's (Lemoalle, 1978).

The sedimentation rate at Bol was not strictly uniform between the upper Miocene and the lower Pleistocene. Mean sedimentation rates were calculated using the thickness of sediments between two dated samples (Fig.2b). A succession of low and high sedimentation rates occurred during the Miocene-Pliocene (Fig.9). High sedimentation rates are observed for three distinct periods: 5.6-4.7 Ma, 3.7-3.5 Ma, and 4.3-4.2 Ma (with a lower intensity). Lacustrine sediments described at the fossiliferous localities of northern Chad (Kollé, Koro Toro, and Kossom Bougoundi, 16-17°N) (Fig.1) indicate at least three transgressive events of the lake up to the northern part of the basin during the Pliocene. These events were dated to 5.4 ±0.6, 4.0 ±0.1, and 3.7 ±0.1 Ma using the $^{10}Be/^{9}Be$ method (Schuster et *al*., 2009; Lebatard et *al*., 2010). The first and last events at 5.4 ±0.6 and 3.7 ±0.1 Ma are concomitant with periods of high sedimentation rate at Bol, and would therefore be associated with an increase of humid conditions in the drainage basin during these ages. The less remarkable yet also high sedimentation rate observed between 4.3-4.2 Ma may be correlated to the transgressive event recorded at 4.0±0.1 Ma in northern Chad depending on whether or not the uncertainty on the absolute ages is taken into account. According to Schneider and Wolf's data (1992), Swezey (2009) described the lithology of three wells drilled in the Kanem region (Fig.1), which each represents a thick section >200 m of mudstones, diatomaceous, and gypsiferous mudstones. The same author attributed these wells to the Pliocene, while Schneider (1989) identified the mudstone sections as being partly late Miocene and Pliocene. It is noteworthy that a great similarity exists between the stratigraphic lithology of the Kanem wells (made of alternating mudstones and diatomitic layers with dominant mudstones) and the lithology of the Bol core. All the northern Chad deposits and cores provide evidence that Lake Chad (or a system of multiple lakes) extended as far as 16°N during the Miocene-Pliocene. Such an expansion was also observed for the Holocene, a period during which Lake Chad

reached its maximum commonly described as Megalake (Schuster *et al.*, 2005; Leblanc *et al.*, 2006; Maley, 2010). In contrast, today Lake Chad is restricted below 14°N.

5.3.A lake older than previously thought

The $^{10}Be/^{9}Be$ dating method provided ages ranging from 6.4± 0.1 Ma for the sample at 297 m to 2.4±0.1 Ma for the sample at 90.8 m (Fig.2). Novello et *al*. (2015a, 2015b) used a model to re-estimate a few $^{10}Be/^{9}Be$ ages which were chronologically inconsistent. Sediments between 297 m and 330 m are dominated by lacustrine clay sediments which are similar to those occurring in the upper part of the core. We assume the age model used by Novello et *al*. (2015a, 2015b) to be totally relevant for estimating the age of the sediment sample located at 330 m. According to this model, the age of the 330 m-deep sample is 6.7±0.1 Ma (Fig.2). Hence a lake (or successive lakes) existed at Bol as early as 6.7±0.1Ma ago, which is about 300 ka older than the age published in Novello *et al.* (2015a, 2015b)..

5.4.Paleoenvironment and paleoclimatology

As mentioned above (see section 5.1. in the Discussion), the dominant clay mineral occurring in the Bol sediments is a smectite, and more precisely a Fe-beidellite, originated from the alteration of lowlands surrounding the lake and/or present further south in the drainage basin. This type of beidellite has been described in modern vertisols of many countries (Wilson, 2013): Turkey (Özkanand Ross 1979; Güzl and Wilson, 1981), Morocco (Badraoui and Bloom, 1990), Sudan (Wilson and Mitchell, 1979), Sardinia (Righi et *al*. 1998), and India (Math and Murphy, 1994). Paquet (1967, 1970) also described Fe-beidellite crystals with a similar chemistry in modern vertisols of Central Africa, and notably in the Chad basin. Vertisols, often named dark clay soils, are today distributed between 45°N and 45 °S of latitude (excluding the desert area) under climates displaying mean annual precipitation of 500 to 1000 mm and a dry season length of 4 to 8 months over the year (Buol et *al*., 2011). The most extensive areas of vertisols of Africa are located in lowlands of Sudan, Egypt, Ethiopia, and Chad (Buol *et al.*, 2011). In Chad, these areas are represented by the current Sudanian phytogeographical domain (White, 1983), which is dominated by mixed and open grassland vegetation. Thus, the dominance of Fe-beidellite in the clay mineral assemblages of Bol supports Sudanian-like environmental conditions in the southern part of the basin during the Miocene-Pliocene. This conclusion is consistent with phytolith and pollen remains of the Bol core (Novello et *al*., 2015a, 2015b), which indicate humid grass-dominated vegetation in the surroundings of Bol and/or in the southern part of the basin during the Pliocene. It is also

in agreement with vegetation simulations of tropical savanna in the southern Chadian basin at ~3.6-2.6 Ma (Salzmann *et al.*, 2008; Contoux *et al.*, 2013). Today, Bol area is located in a drier area, the Sahelian phytogeographical domain (White, 1983), where mean annual rainfall is <500mm/year and dry season lasts 9 to 11 months.

Sedimentation rates are not uniform between the late Miocene and the early Pleistocene. If one considers detrital sedimentation and rainfall intensity as positively correlated (see section 5.2. in the Discussion), high sedimentation rates recorded in between 5.6-4.7 Ma, 3.7-3.5 Ma, and to a lesser extent between 4.3-4.2 Ma may result from an increase of the soil erosion in the drainage basin. Intense soil erosion could be observed in a context of annual rainfall increase and/or a more pronounced monsoon period. The opposite argument would assume the low sedimentation rate observed between 3.4-2.7 Ma as a decrease of annual rainfall and/or an increase of the dry season length in the drainage basin. These hypotheses are in agreement with phytolith and diatom data, which suggest a decrease of lacustrine conditions at Bol after ~3.6 Ma and until 2.7 Ma, coupled with drier conditions in the surrounding and/or in the drainage basin (Novello et *al*., 2015a, 2015b).

Both northern Chad and Bol sedimentary deposits indicate a succession of wetter and drier conditions in Central Africa during the Miocene-Pliocene, although climatic conditions were generally more humid over the basin than today. The alternating of drier and wetter periods in the series is observed from at least ~8 Ma in the northern part of the basin (16°N), and from at least ~6 Ma in the southern part of the basin (13°N). Humid and arid phases were also recorded in central Chad during the late Pleistocene (Gasse, 2000; Maley, 2010). The long-term variability of central African climate is probably related to that recorded in West Africa during the Neogene and associated with changes in the precessional cyclicity of the Earth's orbit (deMenocal, 2004). The existence of more humid conditions over the basin during the Miocene-Pliocene would suggest a more northerly position of the ITCZ (Inter-Tropical Convergence Zone) during the summer monsoon rainfall (Contoux *et al.*, 2013; Novello et *al*., 2015a, 2015b). According to Sepulchre *et al.* (2006), the massive uplift occurring in East Africa after 8 Ma may have impacted moisture circulation between East and Central Africa. Oxygen isotope analyses performed on fish tooth remains from northern Chad (Otero *et al.*, 2011) indicate a trend towards aridification over the Chadian basin during the Pliocene, which could illustrate the eastern uplift hypothesis. The Bol data, which suggest drier conditions in the surroundings and/or in the drainage basin during the late Pliocene (~3.4-2.7 Ma) (Novello

et *al*., 2015a, 2015b; this study), may also have been indirectly affected by the eastern uplift event. Other factors such as warm temperatures, significantly elevated pCO_2 compared to present times, and/or modifications in the ocean circulation and trade wind resulting from the Northern Hemisphere Glaciation (Feakins and deMenocal, 2010; Pagani et al., 2010; Salzmann, 2011; Levin, 2015) could have played a role in the aridification of paleoenvironments in Central Africa during the Pliocene. Yet, the impact of these multiple changes is still difficult to assess and quantify at the mere scale of the Chad basin.

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Figure captions

Figure 1: Location map showing the Chad Basin, the modern Lake Chad (in blue), and the Holocene Lake Mega-Chad (broken line).

Black dot: Bol core location (13°27'N, 14°44'E). Red dots: Miocene-Pliocene fossiliferous areas of the Djurab desert: TM, Toros Menalla; KB, Kossom Bougoundi; KL, Kollé; KT, Koro Toro. Black squares: location of wells drilled in the Kanem region (from Schneider and Wolf, 1992; Swezey, 2009)

Figure 2: Bol core lithology. 2a: whole core lithology; 2b: detailed lithology for the sampled part of Bol core.
The depth (m), label, and $^{10}Be/^{9}Be$ ages (Ma) of the samples selected for micro-biological (Novello et *al*., 2015a, 2015b) and mineralogy analyses (this study) are indicated on the figure. The stratigraphic position of these samples is marked on the sequence with red stars. KB, KL, KT and TM: as in Figure 1.

Figure 3: Typical powder XRD diagram (sample 32a).
Sm: smectite; K: kaolinite; I: illite; F: K-feldpar; Q: quartz; A: anatase; Op: opale.

Figure 4: Relative amounts of the main recurrent minerals resulting from the XRD analysis. From the left to the right: A: K-feldspar; B: TiO_2; C: clays; D: quartz.

Figure 5: Relative amounts of clay minerals from XRD analysis.
Sm: smectite; K: kaolinite; I: illite.

Figure 6: Typical XRD patterns (sample 17h) of oriented aggregates (<2 μm fraction).
a: air dry; b: after ethylene glycol saturation.

Figure 7: XRD pattern of sample 01d after hydrazine saturation.

Figure 8: Chondrite normalized diagrams of the clay fraction chemistry of the Bol samples.
a: REE pattern; b: extended trace elements pattern.

Figure 9: Sedimentation rates of the Bol core.

Table captions

Table 1: Amounts of disordered kaolinite from XRD patterns of clay samples after hydrazine saturation.

Table 2: Chemistry of the clay fractions (<2 μm fraction).
°: chemistry of pellets in samples 21j and 35e.

Table 3: Local (≈ 10 νm^2) SEM-EDX chemistry of samples.

Table 4: La_N/Yb_N and Eu_N/Eu_N** values of the Bol clay samples.
Suffix "$_N$" denotes a chondrite normalized value. Eu_N/Eu_N**: $Eu_N/(Sm_N+Gd_N)/2$. °= values for pellets in samples 21j and 35e.

Supplementary material

Table S1: Location of the studied samples in the Bol core.
*: selected samples for the detailed study of clay mineralogy; **: samples dated and previously studied for micro-biological remains (Novello et *al*., 2015a, 2015b).

Table 1.

Sample	Depth (m)	Disordered Kaolinite (%)
1d	74.0	83
8n	105.0	59
13e	127.0	48
17h	147.0	45
21j	169.3	60
25a	203.0	62
27n	224.3	68
30j	251.0	50
32e	270.4	52
35e	297.2	65

Table 2.

Sample	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	LI	Sum
1d	47.50	20.79	7.69	0.04	1.55	1.34	0.49	1.13	0.85	0.52	17.37	99.27
8n	51.76	18.71	7.52	0.06	1.35	1.21	0.39	1.55	0.85	0.44	16.49	100.31
13e	51.53	19.76	8.21	0.06	1.07	0.79	0.35	1.34	1.00	0.14	16.37	100.62
17h	48.82	19.73	8.24	0.09	1.41	1.01	0.28	1.40	0.80	0.29	16.67	98.74
21j	58.54	17.09	8.44	0.06	0.62	0.45	0.12	0.96	0.84	0.22	12.33	99.66
21j*	52.55	16.59	8.44	0.04	0.49	1.93	0.09	0.75	0.72	0.23	16.87	98.69
25a	53.75	18.53	7.17	0.08	0.95	0.97	0.30	1.41	0.88	0.26	14.87	99.17
27n	54.99	15.55	3.72	0.04	0.48	1.96	0.10	0.69	0.57	0.07	20.61	98.76
30j	46.91	19.02	9.00	0.06	0.84	2.41	0.08	1.13	0.85	0.29	18.19	98.78
32e	53.42	17.53	7.04	0.10	1.32	1.01	0.30	1.35	0.83	0.18	15.61	98.68
35e	48.95	14.69	8.26	0.05	0.48	3.32	0.08	0.70	0.67	0.19	21.58	98.97
35e *	60.81	16.32	7.66	0.05	0.67	0.56	0.11	0.95	0.76	0.14	12.10	100.11

Table 3.

Sample	01d	08n	12L	21e	27n	30a	34e
SiO_2	60.7	59.6	57.4	63.8	63.0	55.6	59.4
TiO_2	1.0	0.9	0.7	1.0	0.7	0.8	1.2
Al_2O_3	23.5	26.0	27.4	23.7	26.2	28.0	28.0
Fe_2O_3	8.7	9.4	10.6	6.8	6.8	11.3	8.2
MnO	0.1	0.0	0.1	0.1	0.1	0.1	0.0
MgO	3.1	1.3	1.3	1.6	1.2	1.4	1.1
CaO	0.8	0.9	0.9	0.8	0.5	0.8	0.7
Na_2O	0.8	0.7	0.6	0.8	0.5	0.5	0.4
K_2O	1.3	1.0	0.9	1.4	0.9	1.5	0.9

Table 4.

Sample	La_N/Yb_N	Eu_N/Eu_N**
1d	11.6	0.74
8n	13.9	0.66
13e	20.0	0.66
17h	15.2	0.66
21j *	12.1	0.64
21j	13.7	0.66
25a	13.0	0.66
27n	15.3	0.67
30j	13.8	0.65
32e	16.3	0.59
35e	12.7	0.62
35e *	14.6	0.59

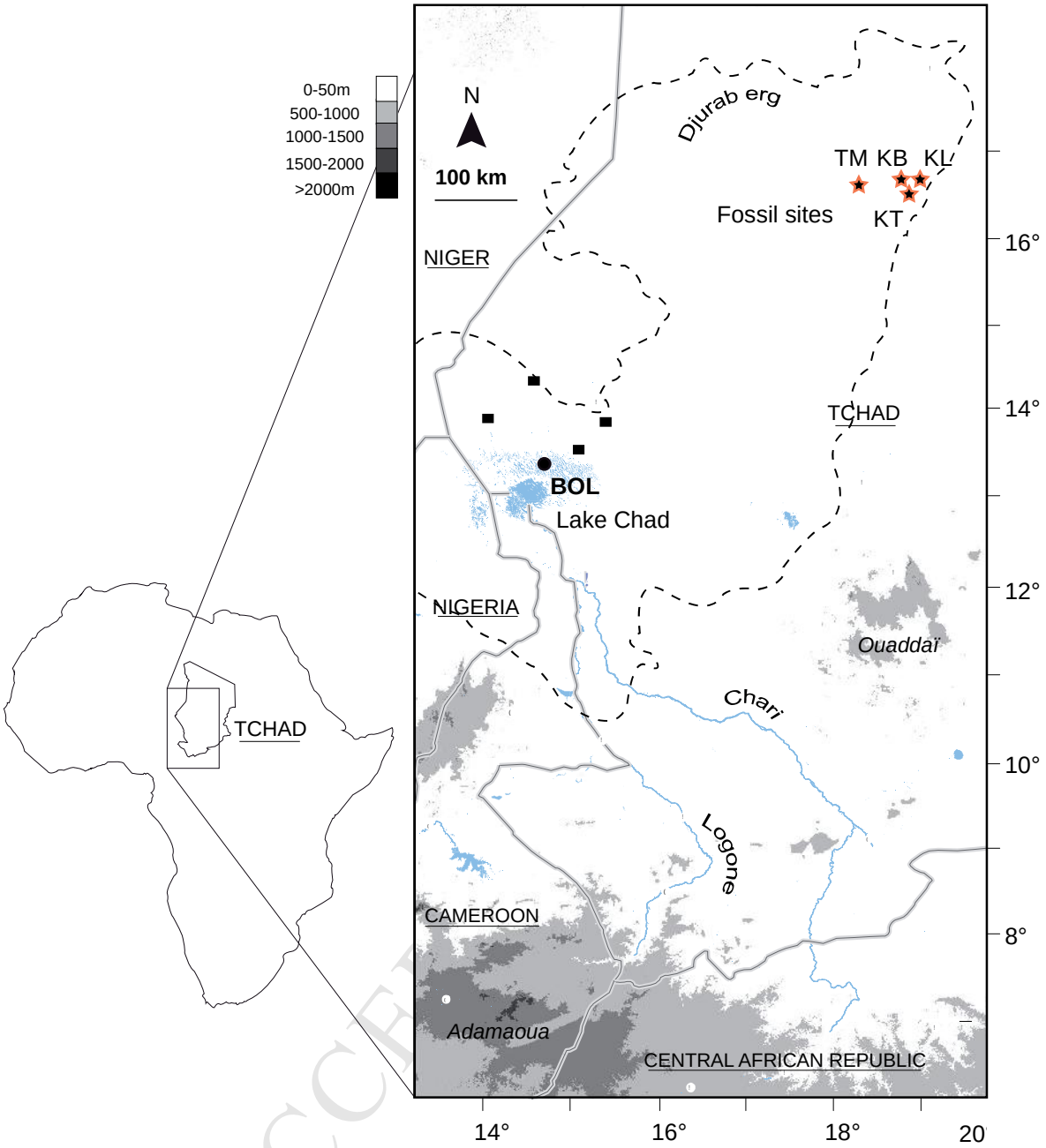

Fig. 1

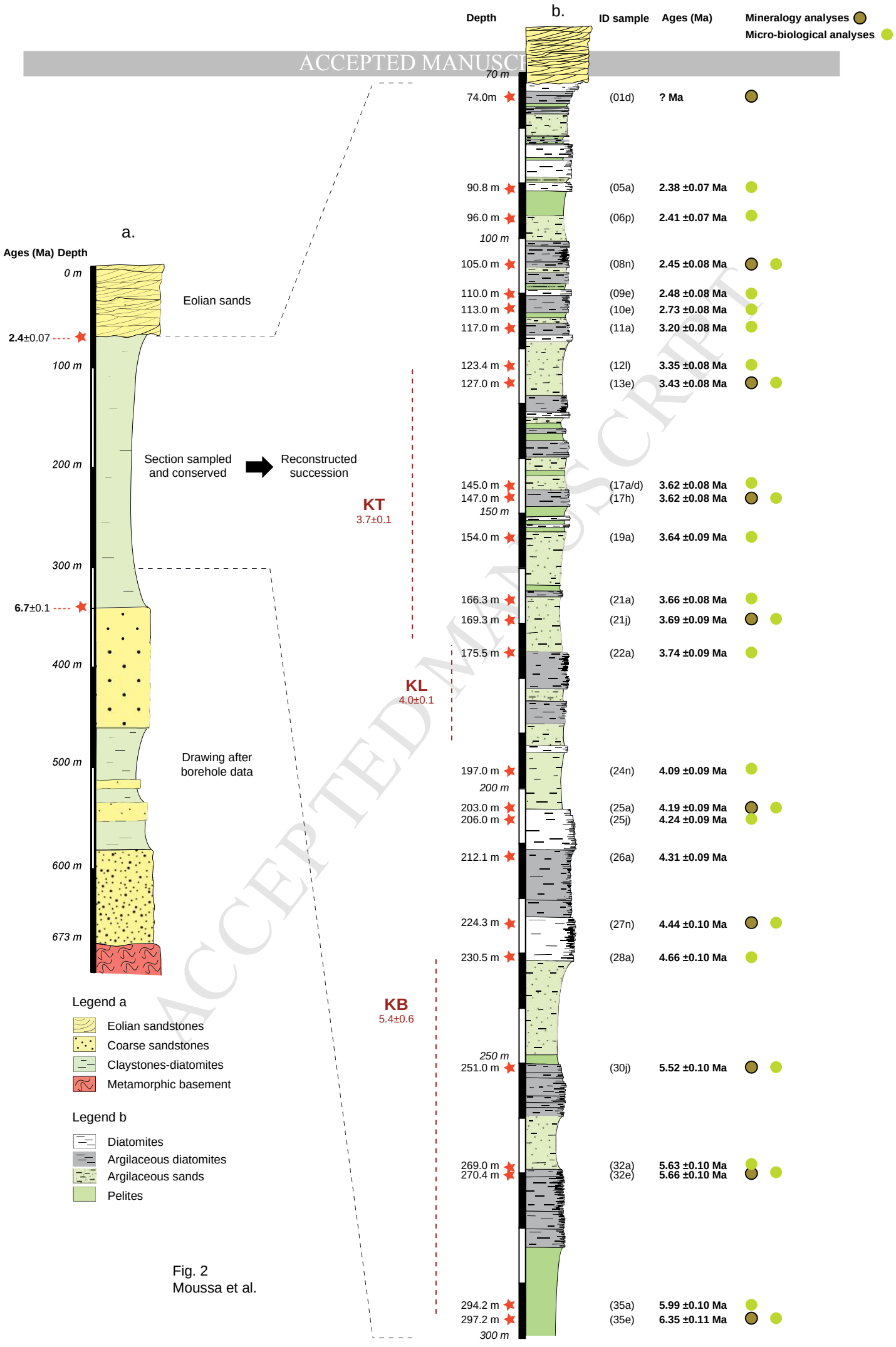

Fig. 2
Moussa et al.

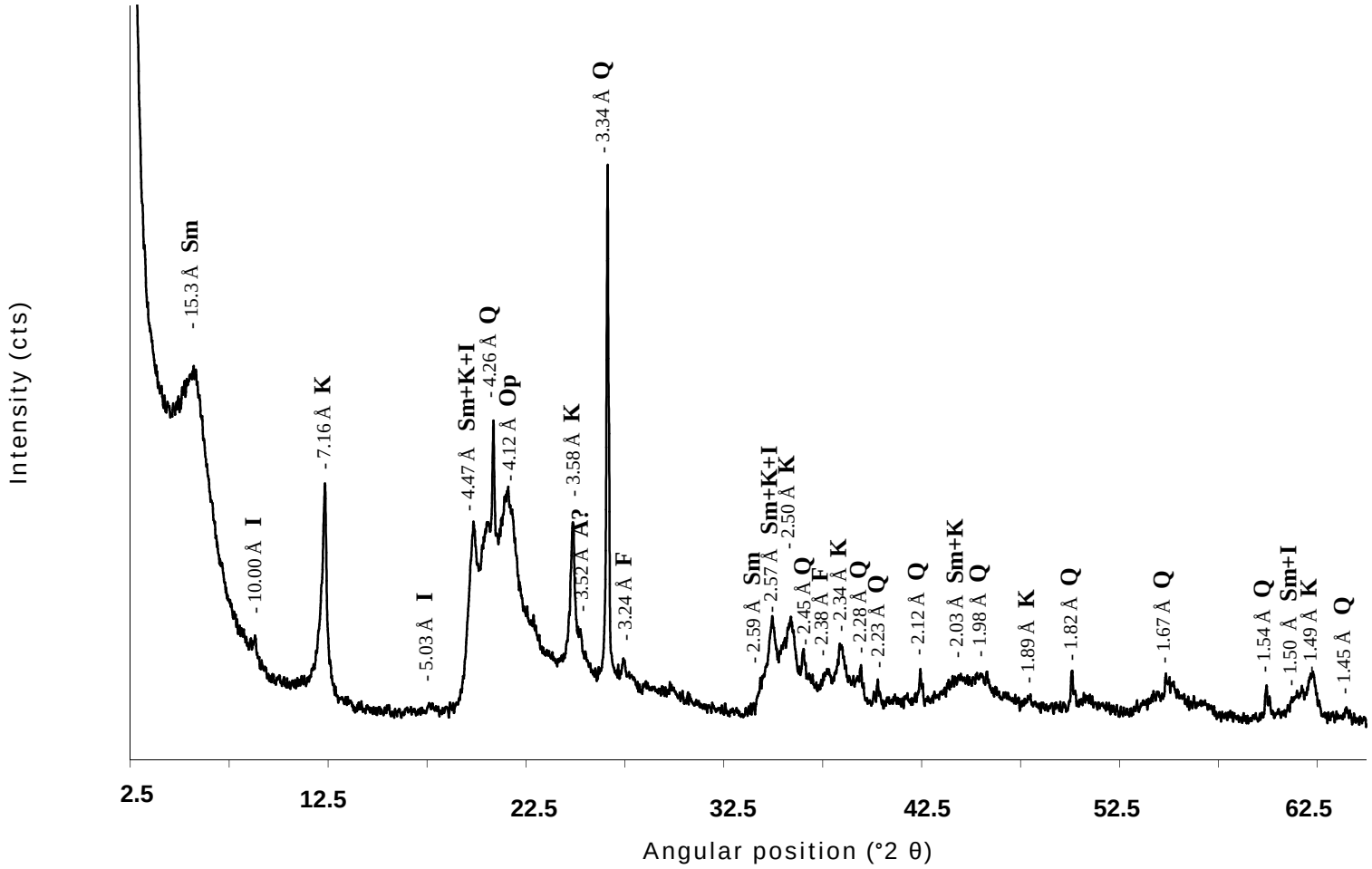

Fig.3
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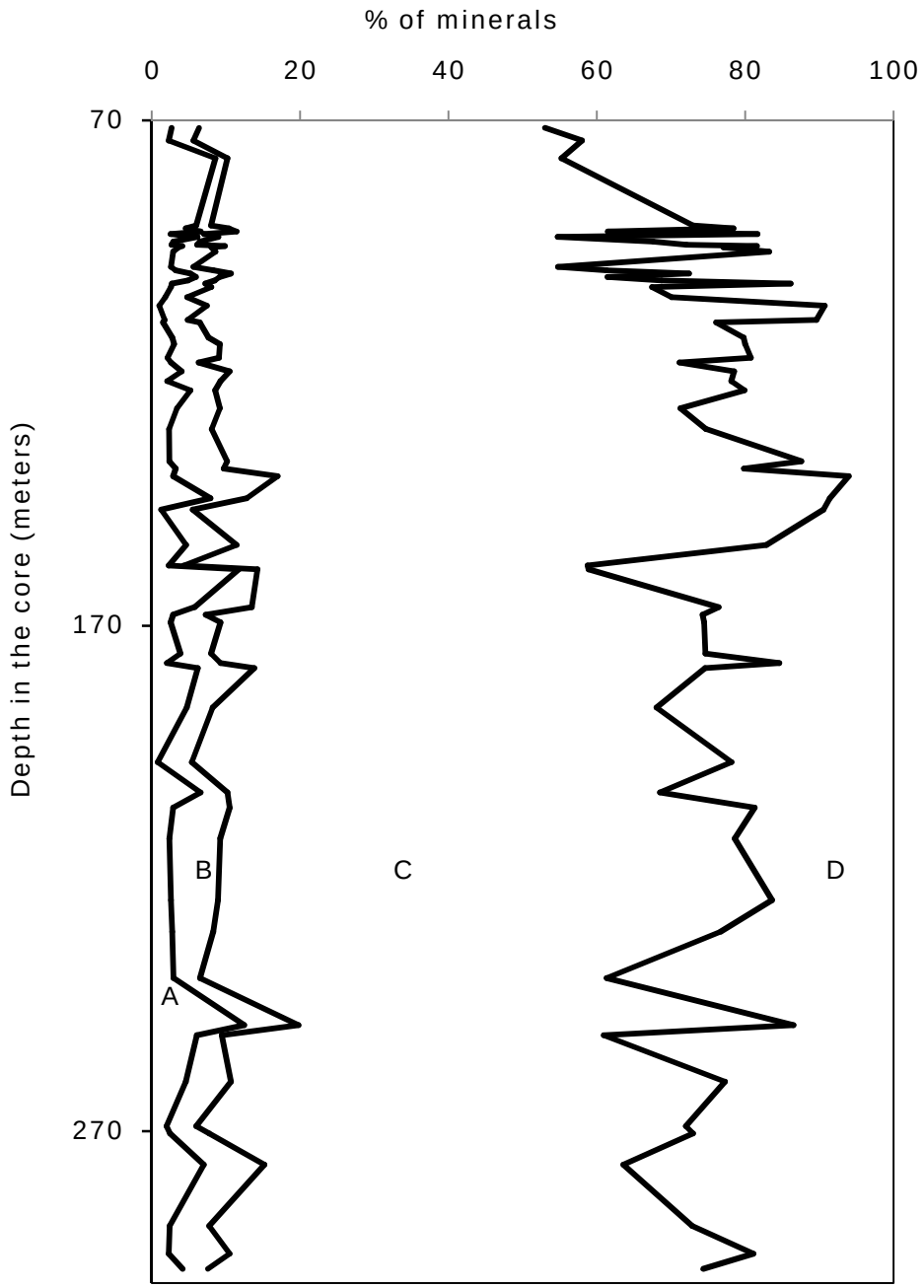

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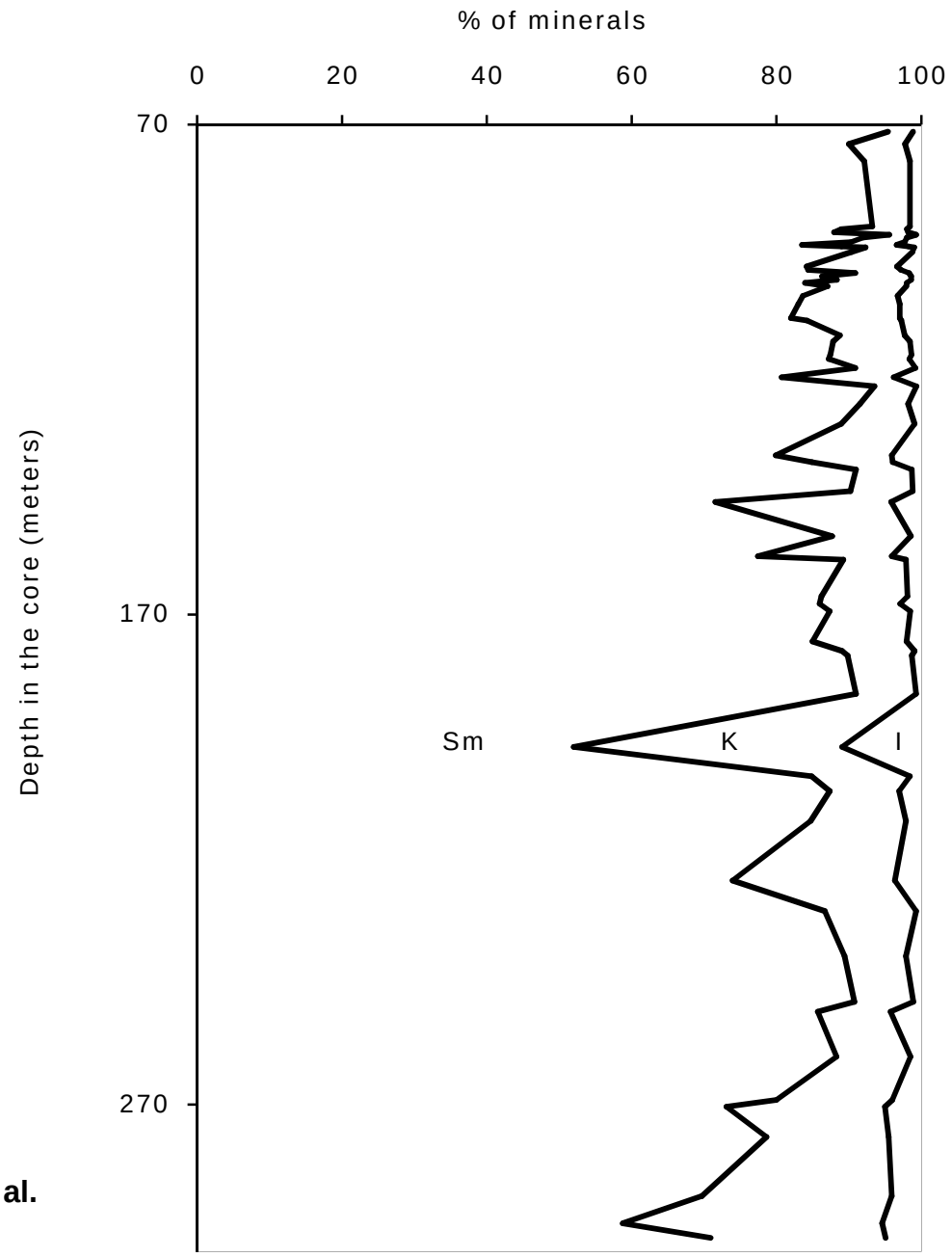

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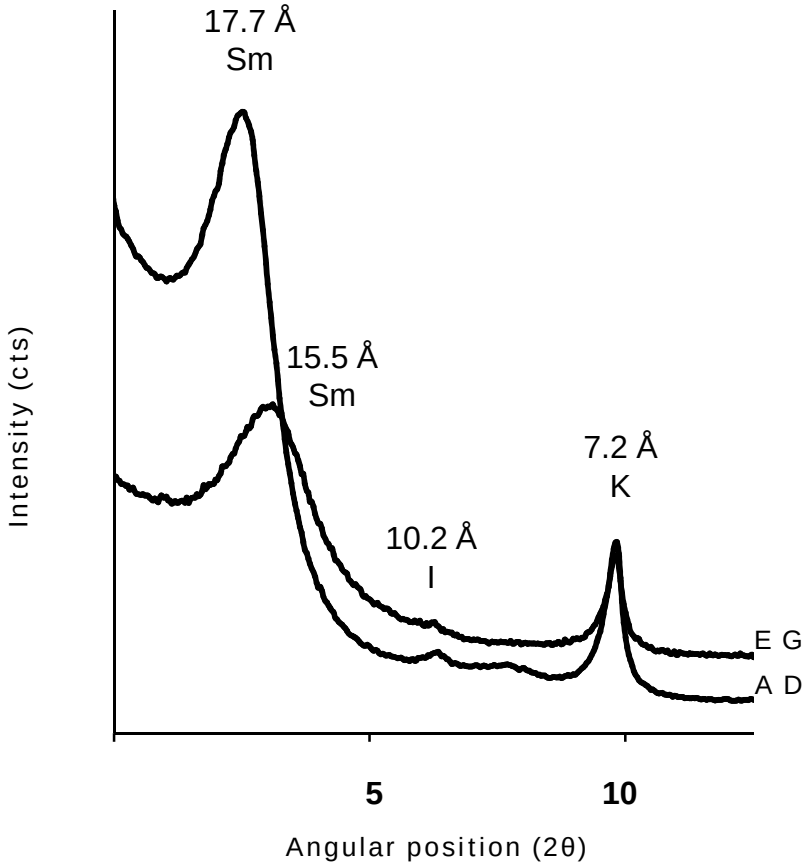

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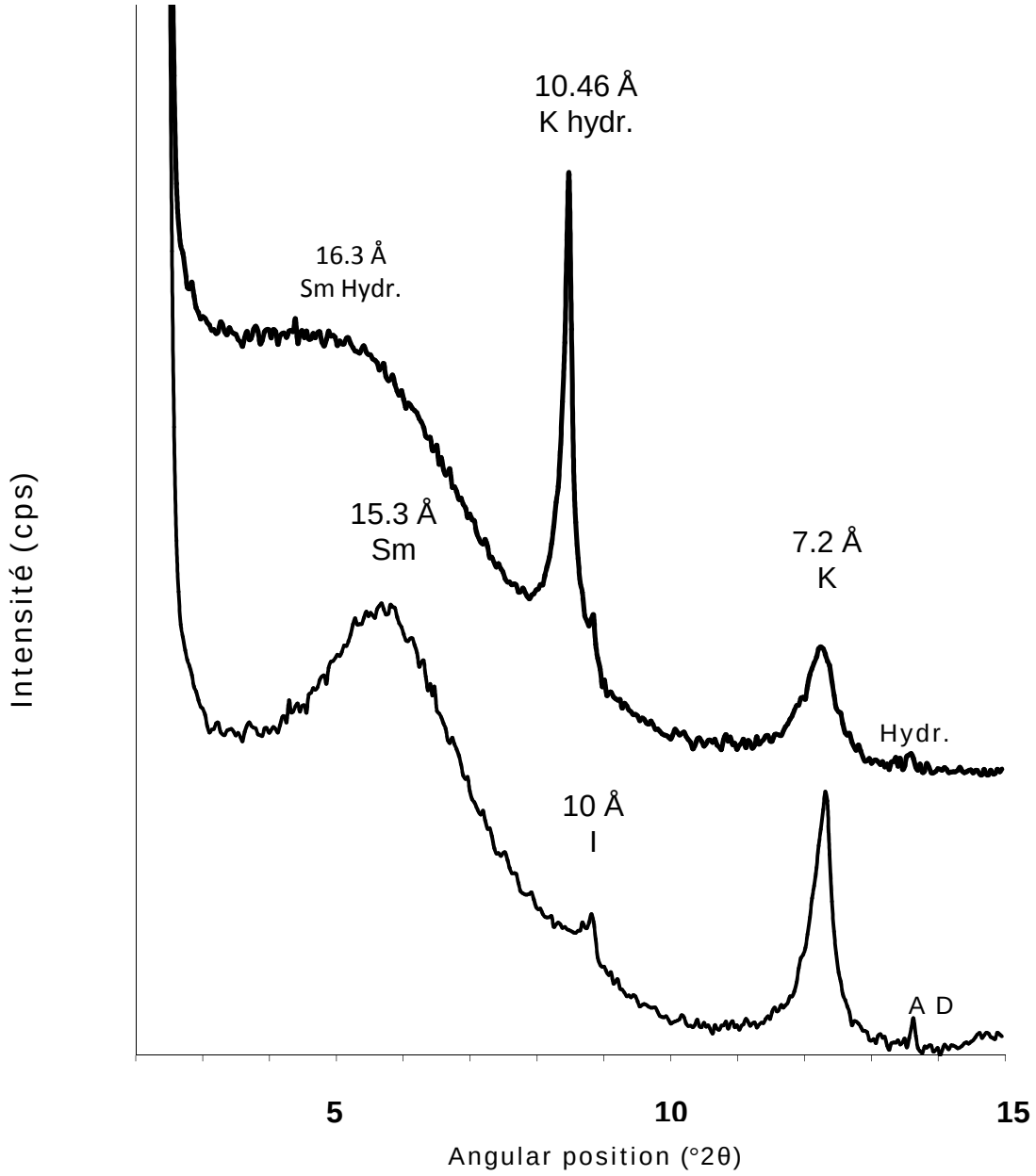

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Moussa et al.

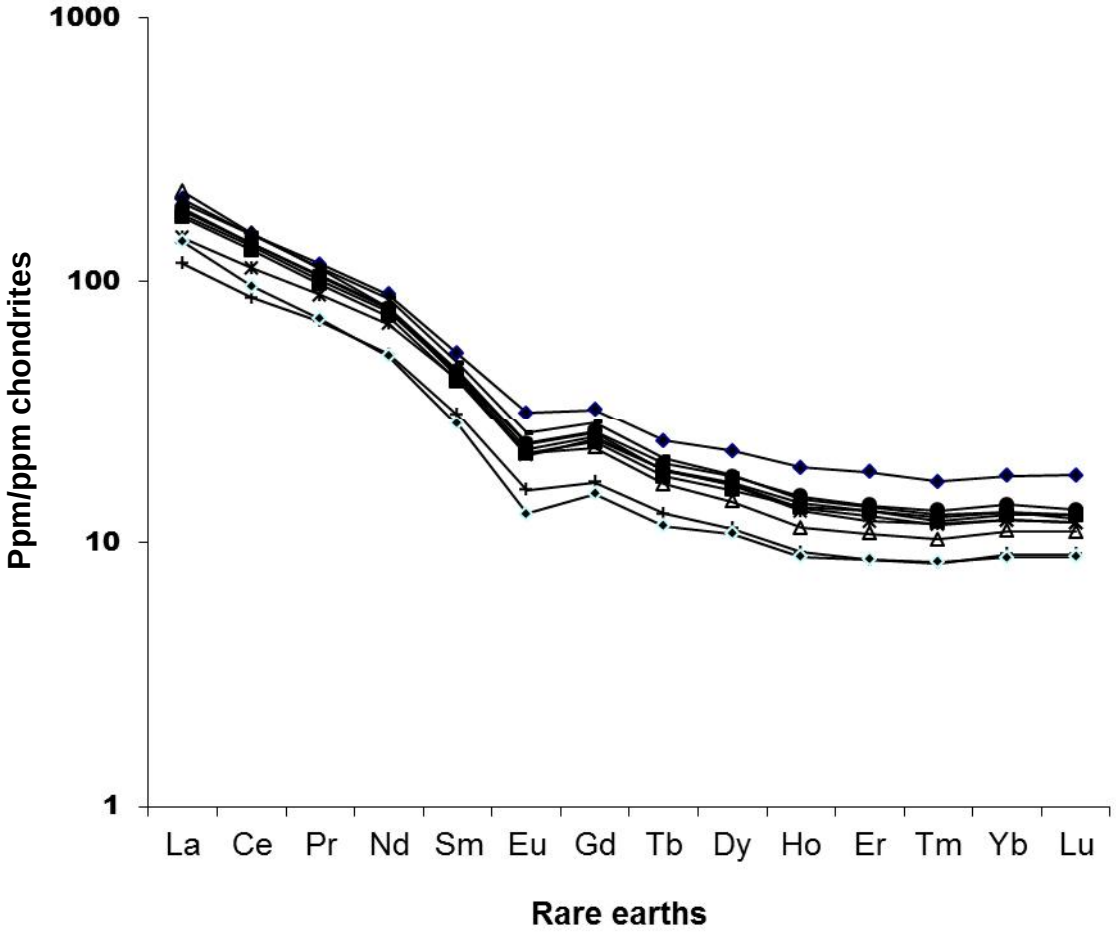


Fig.8a
Moussa et al.

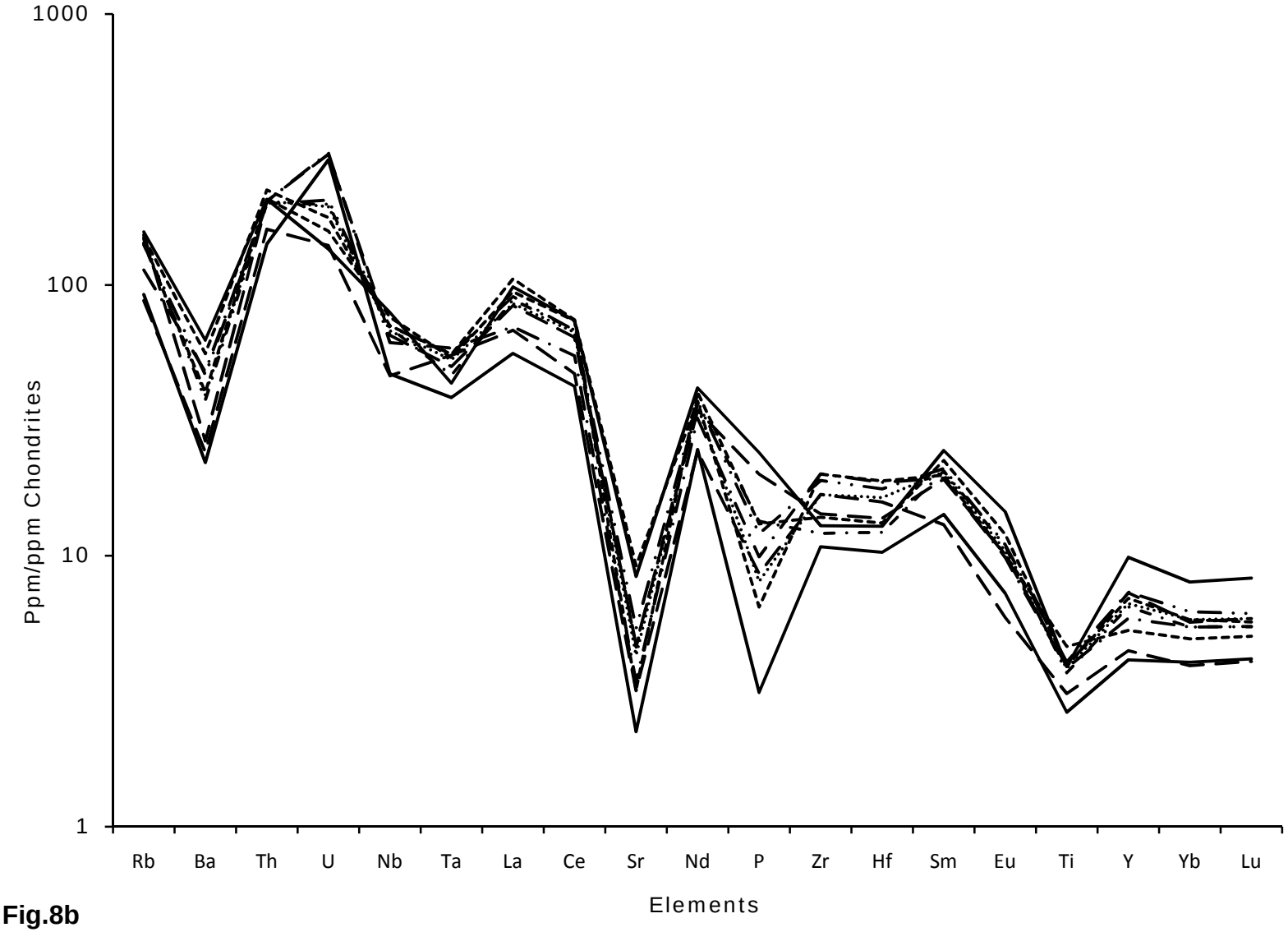
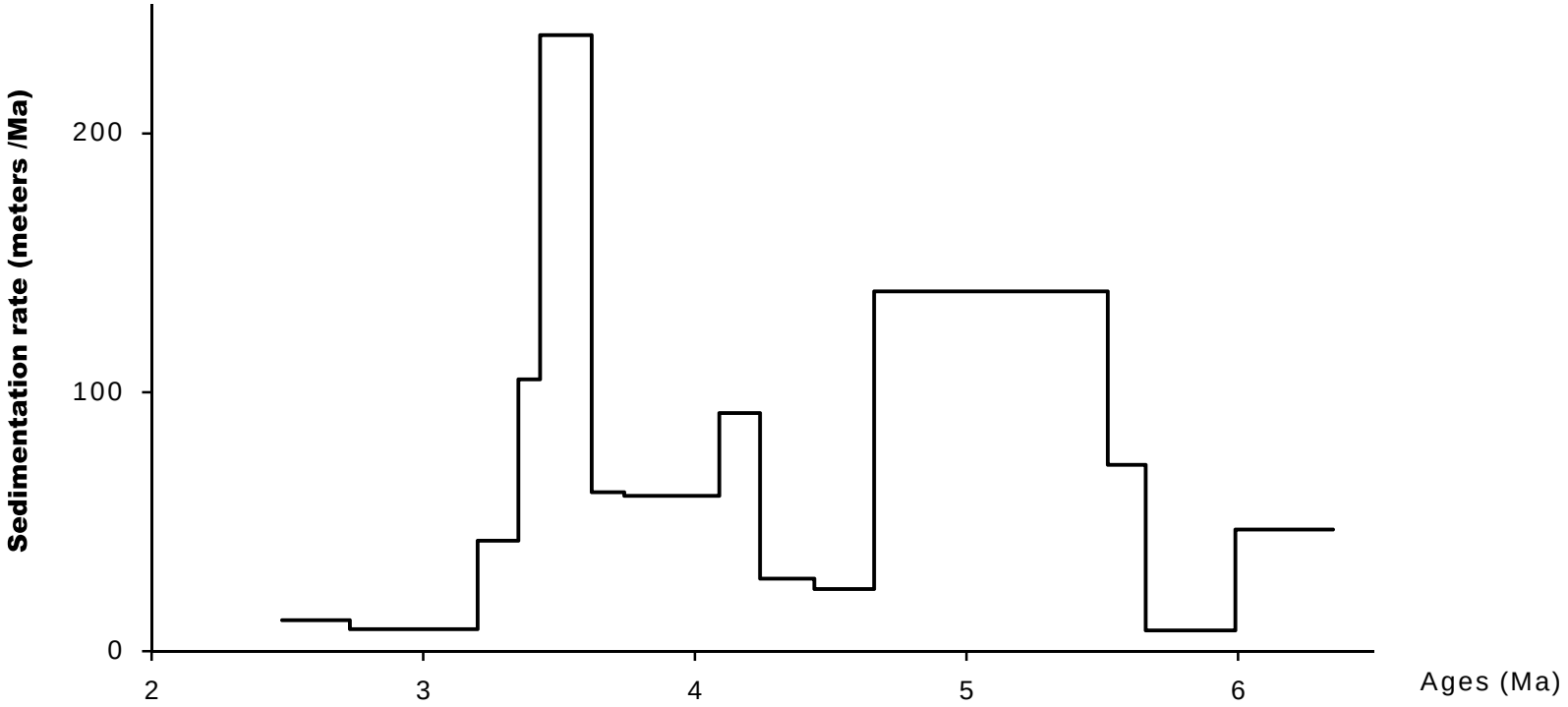


Fig.8b
Moussa et al.



Moussa et al.
Fig.9

Highlights (85 characters maximum by bullet, space included)**:**

Sediments are dominated by detrital clays, more or less mixed with silt and diatomite;

These sediments point to a permanent (recurrent) lake(s) between 6.7-2.3 Ma at Bol;

The dominant clay is a Fe-beidellite, a feature of present-day vertisols;

Vertisols near Bol suggest a Sahelo-Sudanian-like climate between 6.7-2.3 Ma;

Changes in sedimentation rate suggest an alternating of wet and dry periods.