**Mineral assemblages and low energy sedimentary processes in the FLK-Zinj, DS,
PTK and AMK complex palaeolandscape (Olduvai Gorge, Tanzania)**

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Abstract:

The mineralogical composition of the clay stratum (level 22) that contains the FLK Zinj-DS-PTK-AMK complex, the only example of pene-contemporaneous early Pleistocene sites occurring on the same palaeolandscape and simultaneously covered by the same ash fall tuff, shows significant intra-site and inter-site differences. Overall, level 22A is related to a more hydrologically closed system, with higher salinity and alkalinity than level 22B. Both levels show fresher water was entering the system from the DS area during deposition. Textural analysis of the studied samples shows great microporosity and abundant edge-to-edge and edge-to-face contacts between crystals. These results have major repercussions for the interpretation of the archaeological assemblages in the FLK Zinj-DS-PTK-AMK palaeolandscape. First, this implies that two distinct archaeological levels formed at different times with different environmental conditions in most of these localities. Second, textural analyses provide evidence that no important sedimentary compaction took place after the deposition of this layer. This affects interpretations of palaeolandscape geomorphology. Third, it shows that the sedimentary processes during all the clay sequence occurred in very low-energy conditions. This reinforces that no significant physical forces disturbed the original configurations of materials contained within these assemblages and reinforces their autochthony. Behavioural interpretations can, thus, be more solidly built upon the conception that those sites preserve most of their original *in situ* properties.

Key words: Clay minerals, zeolites, clay textures, palaeoenvironments, mixed-layer minerals

1. Introduction

The dense archaeological assemblage of anthropogenic origin discovered in 1959 at FLK-Zinj (Bed I, Olduvai Gorge) was contained in level 22, a thin clay stratum (<20 cm) underlying Tuff IC that dates back to 1.84 Ma (Hay, 1976). Since its discovery, this assemblage has been fundamental in the debates regarding early hominin subsistence and early site functionality, and it has played a central role in the hunting versus scavenging controversy in particular.

The theoretical models that emerged during the 1970s and 1980s regarding the socio-economic behaviors of early hominins were mainly built on different and diverging interpretations of this site. The home base and the food-sharing models (Isaac, 1978) were the main interpretations stemming from the side of those who advocated that early hominin foraging behavior resembled that of modern foraging groups (see also Bunn et al., 1986; Leakey, 1971). Various models were also proposed, such as the passive scavenging model (Binford, 1981; Binford et al., 1988; Blumenschine, 1986), the stone-cache model (Potts, 1984, 1988), a chimpanzee-nesting model (Sept et al., 1992), and a refuge model (Blumenschine, 1991; Blumenschine et al., 1994). All these models were representative of the paradigm of those who thought early hominin socio-economic behaviour to be more similar to that of non-human primates (see review in Domínguez-Rodrigo, 2015; Domínguez-Rodrigo et al., 2017). Domínguez-Rodrigo et al. (2007) provided a comprehensive taphonomic revision of all Bed I assemblages, which yielded solid evidence in favour of the primary access hypothesis, and established that FLK-Zinj was the only anthropogenic site of all Bed I assemblages. At the same time, their work fuelled the discussion on why hominins selected that particular spot on the

landscape for their activities. This shifted the focus of the discussion from the taphonomic dynamics of site formation to the palaeoecology of the surroundings of FLK-Zinj.

Knowing the palaeoecological context of a site is crucial in order to uncover a site's functionality: hominins were capable of carrying out certain activities at a selected location, because particular ecological conditions enabled or prompted them.

Palaeoecological reconstructions of early sites are scarce, however, and they generally describe the broad ecological characteristics of the area as open grasslands or more wooded habitats (e.g. Plummer et al., 2009; Quinn et al., 2007). In fact, the case of FLK-Zinj constitutes the only example of how interpretations of the palaeoecological conditions of a site can be linked to interpretations on its functionality (Domínguez-Rodrigo et al., 2017). While supporters of the scavenging model have situated the site on a barren lacustrine floodplain (Blumenschine and Masao, 1991) or on a peninsula that was surrounded by braided river channels (Blumenschine et al., 2012), defenders of the hunting hypothesis and of the interpretation of the site as a central or referential place have placed it on a topographically higher point in a wooded habitat, a few hundred meters away from a freshwater spring overlooking a wetland (Ashley et al., 2010; Domínguez-Rodrigo et al., 2010).

Moreover, Blumenschine et al. (2012) only described the clay level 22 in the surroundings of FLK-Zinj, arguing it had been eroded elsewhere throughout the gorge junction. However, since the Zinj clay stratum can be traced horizontally throughout a large portion of the gorge, a more extensive and detailed geomorphological, sedimentological, and stratigraphic reconstruction was carried out, which also yielded

the discovery of several new sites on the Zinj palaeolandscape (Uribe Larrea et al., 2014). The discovery of PTK (Philip Tobias Korongo) and AMK (Amin Mturi Korongo) in 2012, and later also of DS (David's Site) in 2014, further reinforced the claim that FLK-Zinj was located on the edge of a higher platform rather than the lower parts of the lacustrine floodplain, and that FLK-Zinj would have been used as a central place in the landscape where hominins would have transported and processed animal carcasses (Domínguez-Rodrigo et al., 2017; Uribe Larrea et al., 2014).

The clay stratum of level 22 offers an excellent opportunity to understand the site formation processes and the environmental characteristics that shaped the described assemblages, since it is a discrete layer with very good preservation of materials, and can be traced laterally over an extensive area. Despite its importance and lateral continuity, a detailed mineralogical analysis of the level has not been carried out since the initial general descriptions made by Hay (1976).

Other levels, especially in lowermost Bed II, have been mineralogically and geochemically studied (Deocampo et al., 2002; Hover and Ashley, 2003; Deocampo, 2004; Mees et al., 2005, 2007; McHenry, 2009; McHenry, 2010) using authigenic zeolite and clay mineral assemblages to infer depositional and diagenetic conditions. The co-existence of these zeolites and clay minerals present in Olduvai Gorge has been widely studied (Hay, 1963, 1964, 1970, 1973, 1976, 1980, 1986, 1996; Hay and Kyser, 2001; Hover and Ashley, 2003; McHenry, 2009; McHenry, 2010).

In addition to the study of authigenic minerals in order to determine saline and alkaline lake water conditions (Keller, 1970; Wilson, 1999; Deocampo, 2002; Deocampo, 2015;

Deocampo et al., 2017), clay minerals can provide further palaeoenvironmental and palaeoclimatic information. The use of clay mineral associations to make environmental and climatic inferences has been widely studied in areas with varied characteristics (Singer, 1980, 1984, Chamley, 1989). These studies, however, have certain limitations (Thiry, 2000), given that diagenesis and authigenesis can make palaeoclimatic inferences unfeasible. Nevertheless, several studies have proven successful in the application of clay mineral studies to palaeoclimatic analyses (Bolle and Adatte, 2001; Ehrmann et al., 2005; Carrasco et al., 2008; Do Campo et al., 2010; Arostegui et al., 2011; Foerster et al., 2018).

Recent field work at FLK Zinj, DS, PTK, and AMK has shown that what was once considered a single archaeological layer can be divided into a minimum of two (levels 22A and B) situated on the upper and lower sections of this thin clay stratum (Domínguez-Rodrigo et al., 2010; Aramendi et al., 2017).

The main objective of this paper is to exhaustively describe the mineralogy of the clay deposits of levels 22A and 22B and to infer palaeoenvironmental and depositional information from the mineral assemblages and their micro-textural evidence. This study will further improve our knowledge and understanding of the FLK-Zinj, PTK, AMK, and DS palaeolandscape complex.

2. Regional setting

Olduvai Gorge is located in northern Tanzania (Fig. 1A), on the western margin of the southern bifurcation of the eastern branch of the East African Rift (Hay, 1976). The

gorge lies northwest of the Ngorongoro Volcanic Highlands and it is cut into the Serengeti Plains (Fig. 1B). The main branch of the gorge originates in lakes Ndutu and Masek, and is joined 9 km downstream by a secondary gorge draining from the western slopes of the Lemagrut volcano (Hay, 1976; Fig. 1B, C). The highest density of archaeological and palaeontological sites, including the studied sites in this paper (Fig. 1C), can be found in the junction between the main and secondary gorges.

Level 22 overlies the Chapatti Tuff (CHT) with a discrete and sharp contact and it is overlain by Tuff 1C (1.84 Ma; Bed I; Hay, 1976). Level 22 consists of two clay sublevels (level 22A and level 22B), each ~10cm thick and containing separate archaeological levels. Level 22A, a dark olive grey (5Y 3/2) earthy clay, overlies level 22B, a waxier and darker (olive 5Y 4/2) clay (Hay, 1976; Uribe Larrea et al., 2014). Southern deposits of level 22 at DS and surroundings contain an intercalated, discontinuous <5cm thick silt unit between level 22A and 22B (Uribe Larrea et al., 2014).

3. Materials and Methods

Level 22 outcrops at four archaeological sites (FLK-Zinj, AMK, PTK, and DS, Fig. 1C) were freshly scraped and ~ 5-cm-thick unaltered samples were collected for each level at each of the studied sites (a total of 8 samples: DS-16-22A, PTK-16-22A, AMK-16-22A, and FLK-16-22A from level 22A and DS-16-22B, PTK-16-22B, AMK-16-22B, and FLK-16-22B from level 22B; Fig. 2).

A 10 -15 g fraction of each sample was homogenized by quartering and grinded manually with an agate mortar until it could be sieved completely through a 53µm mesh. Mineral phases were identified by bulk mineralogy powder X-ray diffractograms

(XRD) using a Philipps Analytical PW 1752 Cu K α radiation power diffractometer (graphite monochromator radiation K α 1= 1,5406 Å). Diffractograms were recorded continuously at 2 θ angles from 2° to 68° with 0.02 stepping intervals and 1 s per step.

A semiquantitative analysis was carried out according to the Chung method (1975) by using EVA software (Bruker), determining the Reference Intensity Ratios (RIR) of the existing phases, which allows the intensity calculations to be normalized on the statement that the sum of all phases in the sample is equal to 100%.

For further analysis of their clay minerals, samples were prepared for oriented aggregates XRD method (OA) by taking a further fraction of the original sample (0.1 – 0.5 g) and dispersing it within distilled water. Then, the fraction smaller than 0.5 μ m was separated and extracted according to Stokes's law and pipetted into 3 glass slides to be prepared in three different ways: 1) oriented aggregates without any further treatment (air dried, AD), 2) oriented aggregates treated with ethylene glycol for 24 h (EG) and 3) oriented aggregates heated at 550°C for 90 minutes (thermal treatment, TT).

The identification of mineral phases was carried out using a Philipps Analytical PW 1752 Cu K α radiation power diffractometer (graphite monochromator radiation K α 1= 1,5406 Å). Diffractograms were recorded continuously at 2 θ angles from 2° to 35° with 0.02 stepping intervals and 0.8 s per step.

The semiquantitative analysis was carried out by using EVA software (Bruker) according to Srödon (1984) and Moore and Reynolds (1989). This semiquantitative technique is based on the reflective power applied over the measured areas of each

mineral main reflection peak, and assuming that the sum of all clay minerals in the sample is equal to 100%.

In addition, powdered randomly oriented clay separates were prepared and studied with a $0.01^\circ 2\theta$ step size, a count time of 120 s per step size, and a count time of 3 s per step between $57^\circ 2\theta$ and $64^\circ 2\theta$ to measure 060 reflections following Moore and Reynolds (1989).

In order to enhance the mineralogical description of the samples and to confirm the occurrence of the illite/smectite mixed layers, a simultaneous thermogravimetric analyser and differential scanning calorimeter was used to perform thermal analysis in four representative samples (DS-16-22A, DS-16-22B, FLK-16-22A, and PTK-16-22A). The temperature was increased 10°C every minute from 25°C to 900°C .

For textural analyses and to compare and contrast the level 22A and 22B clays, five representative samples (DS-16-22A, DS-16-22B, PTK-16-22A, PTK-16-22B, and FLK-16-22A) were studied by means of Scanning Electron Microscopy (SEM). Small chips of each selected sample were broken from the rock blocks so as to expose fresh fracture surfaces representative of the micro-fabric and micro-texture of the rock and then gold-coated. The SEM is equipped with an energy dispersive spectrometer (EDS) system for determination of chemical composition of small particles during SEM observations.

4. Results

4.1. Mineral assemblage characterization

Samples were identified by XRD as clay mineral rich, with a significant presence of alkali feldspar, mainly anorthoclase, albite, and minor amounts of orthoclase and variable quantities of zeolites such as phillipsite, chabazite, and minor amounts of analcime (Table 1). Clinoptilolite was only found in Level 22A at FLK-Zinj. Samples also present modest amounts of pyroxenes (augite), titanite, and carbonates (calcite and dolomite-ankerite).

Fraction below 0.2 μm (clay minerals) shows an evident predominance of smectites over mica (illite) (Table 2). This predominance is more intense in level 22B than in 22A. Kaolinite is present in very low quantities ($< 3\%$). Smectite was identified with the $7.1^\circ 2\theta$ (12.4 $\text{\AA}$) peak in air dried diffractograms, expanding in ethylene glycol around $5.2^\circ 2\theta$ (16.9 $\text{\AA}$) and transformed to a mica-type pattern in the thermal treated sample. Micas (illite) appear as a wide peak around 10-11 $\text{\AA}$ in all three diffractograms (Fig. 3). Kaolinites have been described with the OA (001) reflection at 7-7.1 $\text{\AA}$, both in AD and EG patterns and disappearing after thermal treatment.

The 001 smectite d-spacings (around 17 $\text{\AA}$) and slightly irrational basal spacings suggest the presence of interstratification of illite layers between the smectitic layers. The presence of these illite-smectite mixed layers is continuous in all the studied samples, and can be easily seen in the expansion of the EG over the OA diffractograms around $8.8^\circ 2\theta$ (10 $\text{\AA}$). There are some variations in the ethylene glycol diffractograms between samples, probably because of variations in the quantity of illite present in the structure. The percentage of illite could be estimated between 10% and 50% depending on the sample, though the common values are usually around 30%-40% (Table 3;

Moore and Reynolds, 1989). Furthermore, the presence of illite-smectite mixed layers has also been confirmed by performing DTG analysis and identifying two endothermic reactions near 500° and 700° as described by Komadel and Madejová (2013).

Randomly oriented clay powder shows octahedral sheet d-spacing in 060 peak positions, and this is the common procedure to observe the octahedral sheet cation composition, allowing a further classification of the clay (Moore and Reynolds, 1989, Deocampo et al., 2009). A careful study of this region reveals large peaks around 60.90 2 θ (1.52 Å) – 61.70 2 θ (1.49 Å) for all samples, commonly associated with dioctahedral smectites (Fig. 4). These main peaks migrate from Fe dioctahedral smectites (ranging between 1.51-1.52 Å), with some Mg trioctahedral influence, in FLK towards Al dioctahedral smectites (ranging from 1.49 to 1.50 Å) in DS (values taken from Bailey, 1980; Brindley and Brown, 1980). Smectite composition has been reassured with EDS analyses (Fig. 5).

Accompanying these main peaks is another that can be observed in DS 22B at 60.70 2 θ (1.525 Å), which is related to Mg-rich (trioctahedral) species. Also in DS 22B a final peak can be detected after the main one at 61.90 2 θ (1.499 Å), which is likely mica (illite).

4.2. Micro-textural characterization

SEM-EDS observations reveal that the main clay mineral is smectite. Smectite occurs as thin particles aggregated with edge-to-face and edge-to-edge contacts between crystals. These dispositions rarely develop isolated flakes, but their aggregates form random

microfabrics with open pore structure (inter-aggregate pores, Wilson and Wilson, 2014) displaying good microporosity (Fig. 6A, D, F and 6A).

Random particle orientation shows open card-house and honeycomb type domains produced by a primary flocculated microfabric (Fig. 6A, B and 6A). Rose-shaped aggregates (Fesharaki et al., 2007; Fesharaki, 2016) of thin smectite particles can be distinguished.

In some areas, face-to-face contacts of smectite aggregates generate a better oriented structure with slit and wedge-shaped pores. These areas coincide with concentrations of the larger smectite particles (S1, Fig. 6C). In the same image, smaller smectite crystals nucleating on other larger smectite crystals with curved and warped edges (S2, Fig. 6C) can be observed alongside incipient face-to-edge and edge-to-edge junctions displaying honeycomb type microfabrics (S3, Fig. 6C). Smectite nucleation sites can often be seen as small bumps on other mineral crystal surfaces.

A wide variety of zeolite crystal morphologies can be seen throughout the samples. Crystals are usually short or long prisms, pseudo-prisms, subhedral trapezohedral and sometimes somewhat more tabular shapes (Fig. 6). Different stages of growth of analcime and other zeolites can be observed. For example, a spherical crystal present in Fig. 6A displays an analcime-like chemical composition. These initial morphologies are common in analcime crystals (Cheshire et al., 2013). In general, they grow on clay minerals that are forming coatings in some areas of the samples. They are also very common growing from the walls of the pores on smectites and on the original inherited phases (volcanic glass and anorthoclase). In these cases, they tend to grow in all

directions developing druses or geodes, occupying the pore space as much as possible (Fig. 6D and 7A, B).

Between the smectite particles and the rest of the minerals there are relations of growth of phases. In general, the coatings are incipient on feldspars and plagioclases, with a first layer that is usually parallel to the face of the mineral on which smectites grow and later by curving their edges a new layer is initiated in which the contacts are mainly face-to-edge or edge-to-edge (Fig. 6A, D and 7A, B). In some cases, like in the large anorthoclase crystal in Fig. 7A and B, nucleation of smectite flakes takes place with edges growing perpendicularly to crystal surfaces and along fractures (Fig. 7A, B).

The smectites crystalized prior to the zeolites and between these there is a crystallization sequence according to the different types of zeolites; usually the anhedral, spherical, or short pseudo-prismatic analcime crystals are the first to form. From these crystals, or growing on them, a new family of, in general, short, prismatic, trapezohedral, or tabular phillipsite crystals grows, being also present as long prisms, mainly filling pores and in the form of druses on previous crystals and fracture and pore walls.

SEM observations also reveal smectites and zeolite growth in the pores of the fossil remains found (Fig. 7C, D). In addition, it is observed how the original structure of the bone is coated or even replaced by these minerals, and perhaps by calcite, filling pores and primary structures of the bone (a microvertebrate, in this case).

The texture of the FLK samples is slightly different from the rest of the sites, at least in some areas of the samples, with a more granular texture formed by pseudo-cubic zeolite crystals that grow on smectites that in turn form bridges between them (Fig. 6E), although they still retain a good microporosity.

The studied sample from PTK presents a slightly more massive texture with clay coatings on original volcanic material in which there are several phases of formation of zeolites, including most prominently elongated prismatic crystals (Fig. 6F).

5. Discussion:

5.1. Palaeoenvironmental inferences

To study possible palaeoenvironmental variations spatially between the different sites and temporally between levels, the mineral assemblages and the respective percentages of each mineral are discussed.

Level 22A shows a lateral increase of albite from DS towards FLK-Zinj (Fig. 8A).

Inversely, clay minerals, augite, and anorthoclase decrease towards the northwest.

Zeolite abundance does not vary between sites. Level 22A clay mineral analysis shows slightly more smectite than mica/illite in all the studied sites. At DS, smectites have significantly higher proportions compared to other sites (Fig. 8A).

The analysis of the (060) peak reflection patterns in Level 22A samples shows similar values in the range of values indicated as dioctahedral smectites (Al-Fe-smectites), but a

continuous displacement towards more Mg-enriched positions from DS to FLK-Zinj sites can be distinguished. This peak movement pattern would be related to a northwest decrease in moisture influx and therefore different palaeohydrological conditions between the studied sites.

On the other hand, Level 22B is laterally more homogeneous, and shows only minor variations. Only clay minerals seem to be in higher proportions at FLK-Zinj and DS than in AMK and PTK (Fig. 8B). Level 22B smectite quantities are more homogeneous than in level 22A, with AMK showing the highest proportions and PTK the lowest.

As in level 22A, level 22B shows a predominance of (060) reflections in the dioctahedral smectites (Al-Fe-Smectites) range of values. Once more, a slight movement towards Mg-enriched phases can be distinguished in the same direction as in the overlying level 22A. This displacement towards an enrichment in magnesium can also suggest the same different palaeohydrological differential conditions between sites as described previously for level 22A.

FLK-Zinj is slightly richer in carbonates (sum of calcite and dolomite-ankerite) than the rest of the sites, followed by PTK, which is characteristic of level 22B. The proximity of a carbonate spring at FLK-NN in an equivalent stratigraphic level to level 22 (Ashley et al., 2010; Domínguez-Rodrigo et al., 2010) just a few dozen meters northward from FLK-Zinj and PTK can explain this lateral variation in carbonatic input. It is noteworthy to highlight that carbonatic enrichment could be post-sedimentary, and therefore not pene-contemporaneous with the active spring.

Considering these slight but significant mineralogical and chemical lateral variations, it can be suggested that during the deposition of levels 22A and 22B there was a differential entry of fresh water into the basin throughout the palaeolandscape. It is probable that fresher water was entering the system from the DS area during the deposition of both levels 22A and 22B.

These mineral assemblage variations throughout a palaeolake and adjoining areas due to different depositional conditions are relatively common in the geological record. For example, Turner and Fishman (1991) showed that different authigenic minerals prevail in the assemblages described toward the centre of a Jurassic palaeolake in the Colorado Plateau (USA), following the order smectite → clinoptilolite → analcime (different amounts of K-feldspars) → albite. These lateral variations were influenced by low-temperature processes taking place at surface conditions or in pores affected by highly saline and alkaline waters (Turner and Fishman, 1991).

In order to detect possible environmental changes over time, variations in the percentages of each mineral and mineral assemblages present in levels 22A and 22B have been analysed.

As a general trend in all studied sites, level 22A contains a slightly higher abundance of zeolites and augite and lower quantities of clay minerals than level 22B. Albite and anorthoclase percentages in DS and PTK levels 22A and 22B show no significant differences, whereas in AMK and FLK-Zinj, level 22A contains more albite and less anorthoclase than level 22B. Other minerals, such as orthoclase and titanite are present in the same percentages throughout levels 22A and 22B. The presence of zeolites, like

phillipsite and chabazite, point to raised alkalinity and salinity (Iijima and Utada, 2006). Their increase through level 22A is indicative of more saline and alkaline conditions.

As to the clay fraction analysis, level 22B presents more smectite and less mica (illite), whereas kaolinite, always less than 3%, remains stable throughout the studied samples. This trend is not clearly visible at DS. While mixed-layers are present in all the samples, they tend to be more illitizated in level 22A than in level 22B, from an average of 30% illitic layers in level 22B to an average of 45% illitic layers in level 22A.

Illitization of smectites changes dominant cation proportions in the octahedral sheet, which is a common process in this type of lake and lake-margin environments (Deocampo, 2015). Repeated cycles of wetting and drying can increase the potassium fixation (greater illitization) in clay mineral structures (Eberl et al., 1986, 1993; Hugget and Cuadros, 2005). This is a very plausible process in lake-margin semi-arid environments such as that of the studied sites.

In relation to (060) reflection patterns, only in DS and FLK-Zinj does there seem to be a slightly greater displacement of the peaks towards higher values of magnesium in the octahedral positions, and so more Mg-enriched minerals in level 22A than in 22B. This compositional change was confirmed with EDS analyses.

These differences in the mineral assemblages between the 22A and 22B levels suggest a more hydrologically closed system for level 22A, with higher salinity and alkalinity than level 22B. This would suggest less fresh water input pointing to a brief saline

period during one of the weaker freshening events described previously by Deocampo et al. (2017) in the palaeo-Lake Olduvai basin (1.84 Ma).

No clearly significant differences have been observed between the textural characteristics between studied sites, so the conditions in general do not appear to have been sufficiently different to create different micro-fabrics in the finest fractions. In general, open card-house and honeycomb type textures, which predominate in the studied samples, are indicative of large amounts of electrolytes in the water (Aróstegui et al., 2011). This result is in accordance with previous studies inferring high salinity and alkalinity in an alternating lake and lake-shore environment (Hay, 1976; Hay and Kyser, 2001; Deocampo et al., 2002; Hover and Ashley, 2003; Deocampo, 2015; Deocampo et al., 2017).

5.2. Depositional and diagenetic inferences

Smectites, the most abundant mineral found in all samples, predominantly show well defined edge-to-face and edge-to-edge contacts between crystals (Fig. 6A, B, D, and 7B). The prevalence of card-house and honeycomb type microfabrics (Fig. 6) is consistent with the high smectite content of the studied samples. Expandable layers in I/S mixed-layers range from 50 to 80% which, as indicated by Keller et al. (1986), is consistent with micro-textural features more likely resembling that of smectites than of micas (illites). However, as observed in Fig. 6C, microfabrics with face-to-face contacts between larger mixed-layer particles are observed, being more similar to illitic morphologies. This would be the case for particles more enriched in illitic layers in the

I/S mixed-layers. Such textures are indicative of flocculated clay minerals deposited in low energy sedimentary processes.

The role of water flow in level 22 at FLK-Zinj has been the subject of debate since Blumenschine et al. (2012) hypothesized a model where a great river inundated level 22, with enough energy to meddle with the original *in situ* properties of the assemblage. Later, Benito-Calvo and de la Torre (2011) and de la Torre and Benito-Calvo (2013) tested this hypothesis by using Leakey's (1971) site plans and applied GIS methods to calculate axial orientations of bones and lithic tools. They conclude the assemblages (including FLK-Zinj, one of the studied sites in this paper) had been post-depositionally disturbed with natural processes such as water disturbance. In addition, de la Torre et al. (2018) reach the same conclusion when applying grain size and shape statistical techniques. If such processes had taken place, face-to-face crystal contacts, which are more hydrodynamically stable, would be the dominant smectite texture in the studied samples. However, as stated previously, these textures are present, but in much lower proportion than edge-to-face and edge-to-edge textures. Additionally, post-depositional disturbances of clays would generate large clay aggregates, which are not present in any of the studied samples.

Smectite crystals form random microfabrics with an open inter-aggregate pore structure (Wilson and Wilson, 2014), displaying proper microporosity (Fig. 6A, D, F and 7A). This widespread microporosity throughout level 22 refutes the interpretation made by Blumenschine et al. (2012) that the clays had been significantly compacted. Inferences regarding incorrect decompactional corrections may lead to over and under-estimations of topographical features. Blumenschine et al. (2012) apply decompactional corrections

of 205% to their pre-burial landscape reconstruction, reconstructing deep and narrow fluvial channels on the FLK-Zinj palaeolandscape. However, abundant microporosity present in all samples suggests level 22 clays have not been intensively compacted, and support the palaeolandscape model proposed by Uribe Larrea et al. (2014), who inferred little to no compaction on level 22 clays.

5.3. Hydrothermalism at Olduvai Gorge

Smectite-to-illite alteration environments have been extensively described (Srodon, 2007 and references therein) and are normally associated with temperatures above 70-80°C and up to 180-190°C, where smectite percentage becomes less than 10% and cannot be measured by XRD. The most common way to reach such temperatures is through sedimentary or tectonic burial in any sedimentary basin with a common geothermal gradient, although hydrothermalism can easily reach temperatures above 70°C. Hydrothermal illitization has been widely studied (Jennings and Thompson, 1986; Eberl et al., 1987; Viczián, 1997; Ylagan et al., 2000) and have similar smectite percentage to temperature relationships than those known for burial illitization.

The Olduvai palaeo-lake basin was too shallow (100 m; Hay, 1976) to develop sedimentary burial illitization. Level 22A shows mean values of 40-50% of illitic layers in I/S mixed layers, whereas in level 22B these drop down to 20-40%. In any case, these smectite percentages relate to temperatures of 70-90° (Srodon, 2007), which could only be reached geothermally with sedimentary burial over several kilometres deep. On the other hand, hydrothermalism in this East African Rift area would not be surprising, with many hydrothermally active modern-day examples near Olduvai Gorge such as Lake

Natron, Lake Manyara, Lake Eyasi, and the Ngorongoro Crater (Ashley et al., 2004; Owen et al., 2004).

Conversely, surface illitization has been documented in extreme geochemical lake environments at pH above 9-10 (Eberl et al., 1986; Hay et al., 1991; Eberl et al., 1993; Bauer and Velde, 1999), most often producing either Fe or Al-rich mica (illite). Since the Olduvai palaeo-lake is widely known to be saline and alkaline (Hay, 1976; Hay and Kyser, 2001; Deocampo et al., 2002; Hover and Ashley, 2003; Deocampo, 2015; Deocampo et al., 2017), surface illitization due to extreme geochemical conditions cannot be ruled out. The fact that level 22A shows higher illitization and is inferred to be more saline and alkaline, suggests that water geochemistry is the main cause of such a process. Future geological and geochemical studies should be carried out to elucidate the cause of the illitization in levels 22A and 22B, whether due to hydrothermalism, extreme water alkalinity, or a combination of both.

6. Conclusions

The mineralogical analysis of the clay unit containing the FLK Zinj-DS-PTK complex shows that the stratum contains two lithologically different subunits. This is confirmed by the presence of an intercalated silt unit in between both of them locally at DS. Therefore, this confirms ongoing work at DS and PTK showing that there are two clearly distinctive archaeological levels; something also reflected in the vertical distribution of materials in the recent excavation of FLK-Zinj (Domínguez-Rodrigo et al., 2010). The interplay between lake transgression and recession enabled, thus, a minimum of two different occupational/depositional episodes of hominin activities in

most of these sites. Both levels 22A and 22B show lateral mineralogical and geochemical variations, suggesting the presence of fresh water input from the surroundings of DS. Furthermore, mineralogical analyses hint level 22A would have deposited in more saline and alkaline conditions.

The other consequence of the mineralogical analysis of the clay that covered the FLK Zinj-DS-PTK complex is that it shows that sedimentary conditions must have occurred in very low energy conditions throughout the clay sequence of this stratum. This clearly invalidates any interpretive scenario hypothesizing about high-energy hydraulic flows disturbing the autochthonous archaeological assemblages to different degrees or rendering them lag assemblages or even more exaggerated assumptions positing the transported nature of all these sites (against Benito-Calvo and de la Torre (2011) and de la Torre and Benito-Calvo (2013)). This additionally supports taphonomic interpretations on the autochthonous nature of the sites in this complex (Domínguez-Rodrigo et al., 2012, 2014). Micro-textural analyses also suggest level 22 clays have not been intensively compacted.

An additional interesting consequence from the present analysis is the possible signal of hydrothermalism found in the mineral composition of the clays of the FLK Zinj-DS-PTK palaeolandscape. Although this could be due to extreme geochemical conditions, plausible due to the alkaline character of the lake, future analyses of biomarkers should contribute to its detection. If present, hydrothermalism would add an additional component to volcanism and highly dynamic tectonic environments for the selective criteria accounting for the evolution of hominin anatomies and behaviours.

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Table 1 – XRD results for the studied samples from levels 22A and 22B.

	Clay Minerals	Zeolites				Feldspars			Carbonates		Augite	Titanite
		Phillipsite	Chabazite	Analcime	Clinoptiolite	Albite	Anorthoclase	Orthoclase	Calcite	Dolomite-Ankerite		
DS-16-22A	33	8	7	2	0	2	31	3	0	2	9	3
DS-16-22B	40	7	5	0	0	2	36	3	1	0	5	1
PTK-16-22A	31	6	9	2	0	10	25	4	2	0	8	3
PTK-16-22B	28	9	10	2	0	1	27	4	1	5	9	4
AMK-16-22A	24	7	8	2	0	27	21	2	1	0	5	3
AMK-16-22B	33	7	8	2	0	5	33	6	1	0	2	3
FLK-16-22A	20	4	7	1	5	35	19	3	1	2	1	2
FLK-16-22B	37	8	6	2	0	2	28	3	2	6	4	2

Table 2 – Oriented aggregates semi-quantification results. PbNM= Present but not measurable.

	Smectite (%)	Mica/Illite (%)	Kaolinite (%)
DS-16-22A	86.9	13.1	0
DS-16-22B	87.3	11.9	0.8
PTK-16-22A	59.1	40.9	PbNM
PTK-16-22B	82.2	17	0.8
AMK-16-22A	48	52	PbNM
AMK-16-22B	93.7	6.3	0
FLK-16-22A	50.6	46.4	3
FLK-16-22B	87.9	11.7	0.4

Table 3 – Percentage of illitic layers in I/S mixed layers. PbNM= Present but not measurable.

	Illitic layers (%)
DS-16-22A	40-50
DS-16-22B	20-30
PTK-16-22A	40-50
PTK-16-22B	30-40
AMK-16-22A	30-40
AMK-16-22B	PbNM
FLK-16-22A	40-50
FLK-16-22B	20-30