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# Controlling factors on clay mineral assemblages: insights from facies analysis of Pliocene to Pleistocene coastal margin deposits, Western Portugal

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## ABSTRACT

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The clay fractions in the Pliocene to Pleistocene coastal margin record are usually dominated by kaolinite and illite, with lesser amounts of vermiculite, 10-14 mixed layer clays and smectite. The high clay mineral crystallinity, the mineralogical relations to facies and depositional setting and some horizontal variations along coeval deposits suggest that clay assemblages are mainly detrital. Illite is more important in inner shelf deposits and particularly in alluvial deposits from eastern locations. The high illite content in eastern alluvial deposits is explained by the input from the neighbouring Iberian Variscan Massif that is rich in mica. The high kaolinite content in prograding sand and gravel coastal plain is partially explained by the availability of this mineral in the drainage areas. Given the arkosic nature of some of these deposits, post-depositional feldspars weathering would also contribute to an increase in kaolinite content. Vermiculite is particularly important close to the surface and to unconformities, in horizons influenced by pedogenetic processes that have more aluminous illite with relatively low crystallinity. In organic rich mud sediments low pH conditions favour post-depositional transformation of illite to vermiculite and mixed layer clays.

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**KEYWORDS** | Paleogeography. Facies. Clay minerals. Provenance.

## INTRODUCTION

The West Portuguese Pliocene to Pleistocene record constitutes a thin succession deposited in a low subsiding region. Most stratigraphic sections show several unconformities bounding strikingly different coastal and continental depositional units. Some sections include alternating facies that represent the interplay of various sedimentary environments. Furthermore, the Neogene tectonic activity and the variability in basement rock characteristics determined several morphostructural units with con-

trasting evolutions that can also influence the mineralogical data.

Original clay mineralogy depends on climate, relief, lithology of the source area, tectonic activity, among other factors (Keller, 1970; Chamley, 1989; Sáez et al., 2003). In areas where after burial clay transformation is expected to be minimal the clay mineralogy may be used as a valuable tool for untangling the environmental conditions coeval of deposition. However, care should be taken on the interpretation of clay mineralogy. One possible pro-

blem is that the clay mineral signal can lag behind the time of formation in the drainage area (Thiry, 2000). Clay mineralogy may also depend on post-depositional transformation. The extent of these transformations and involved processes depends on the facies characteristics and post-depositional evolution. It also depends on the original clay mineralogy itself, since characteristic minerals of advanced weathering stages tend to remain unaltered after exposure to different climatic conditions, whereas minerals characteristic of less aggressive climatic conditions tend to be unstable (Singer, 1980; Thiry, 2000). Even without significant burial or exposure, low pH meteoric water is capable of changing the clay mineralogy by a process of meteoric flushing (Ahlberg et al., 2003). The extent of these mineralogical changes depends on the facies (e.g. grain size and mineralogy). To discern the relative influence of the intervening factors it is essential to properly describe the facies features and understand the paleogeographic evolution.

The purpose of this paper is to interpret the variations in clay mineral assemblages on Pliocene and Pleistocene sediments from the West Portuguese Coastal Margin, with emphasis on the sediment characteristics and landscape evolution. The significance of the variations in clay mineral assemblages and their relationships to depositional systems, local morphostructural setting and depositional phases are demonstrated. The mineralogical data is integrated with sedimentological and structural information reinforcing a conceptual model for the evolution of the area.

## GEOLOGICAL FRAMEWORK

Along the studied area it is possible to recognize three morphostructural domains defined according to the observed tectonic displacements and the sedimentary cover characteristics. The Cértima Basin is a structurally subsiding basin aligned North-South near the contact with the uplifted Hercynian Massif. The Littoral Horst is a relatively uplifted unit located westward of the Cértima Basin. The late Pleistocene coastal margin is a low altitude area that extends for 90 km from Quiaios to Espinho, where late Pleistocene sediments of raised beach and fluvial terraces crop out under Holocene aeolian dunes. This unit is not studied here.

The Pliocene to Pleistocene overlies Mesozoic carbonate and siliciclastic sediments of the Lusitanian Basin and Precambrian to Paleozoic mainly metasedimentary rocks of the Iberian Variscan Massif (Figs. 1 and 2). These Plio-Pleistocene deposits on West Iberia are organized in a general prograding succession (Cunha et al., 1993; Soares, 1999; Dinis, 2004). Four informal units,

based on lithological data, can be considered here (Fig. 3). Unit 1 is characterized by a domain of inner shelf transgressive deposits. Unit 2A overlies with a regional unconformity either unit 1 or the basement. It consists mainly of fluvio-deltaic coarse sandstones. Unit 2B may overlay unit 2A or unit 1 and consists of a coarsening upward sequence that evolves from lagoon mud sediments to alluvial mudstone, sandstones and conglomerates. Unit 3 is made mainly of alluvial fan sediments that usually overlie the previous units with a regional unconformity. There is limited chronostratigraphic data for the studied deposits. The lower transgressive deposits of unit 1 are considered equivalent to the Upper Zanclean to Lower Piacenzian fossil rich beds found in southern locations of the western Portuguese coastal margin (Silva, 2001). The palynological data from a lignite bed of unit 2B points to Piacenzian age (Diniz, 1984).

There are several evidences of tectonic influence over the Pliocene-Pleistocene record. The average thickness of the sedimentary record is much greater in restricted sectors structurally bound, where it can reach 70 m. The western border of Variscan Iberian Massif was uplifted during the Neogene (Ferreira, 1991; Cabral, 1995; Soares, 1999; Dinis, 2004). One to two hundred meters offset along the contact between these two units is deduced from paleosurfaces dating and geomorphologic interpretations (Ferreira, 1991; Cabral, 1995). This offset is directly related to the accumulation of alluvial fan sediments. The complex fault displacements and tilting favoured the generation of small lakes and poorly drained basins. The neotectonic activity also affected the post-depositional evolution.

## METHODOLOGY

Samples were collected from selected sections allowing, as far as possible, an equal distribution along the study area. The selection of samples also focused on a satisfactory coverage of the different facies and stratigraphic units. Poor outcrops, where facies identification and stratigraphy are hard to establish, were rejected. Grain-size was determined by sieving in a column with 1/4 $\phi$  increment. Selected sub-samples with particle size below 2mm were analysed in a Coulter LS 320 instrument that uses laser diffraction for particle size analysis. The roundness of quartz grains was analysed in sand fractions by visual comparison.

Clay mineralogy was determined in 185 samples by X-ray diffraction (XRD). The analysis was conducted on <2 $\mu$ m fraction separated by centrifugation according to Stokes law. Oriented slides were

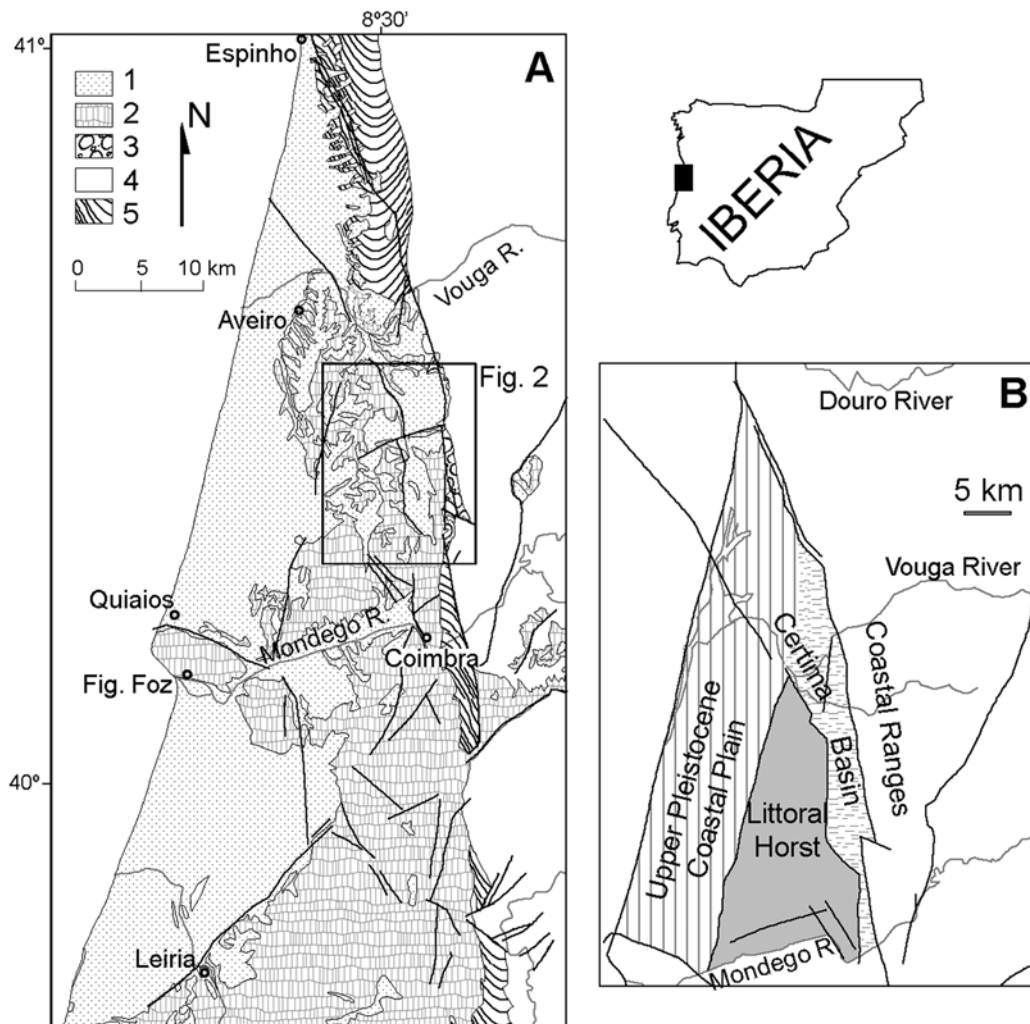


FIGURE 1 | Geological framework of the studied area (A) and morphostructural units in Central West of Portugal (B). 1: Cainozoic sediments; 2: Mesozoic sediments; 3: Intra-mountain Permian-Carboniferous sediments. 4: Central Iberian Zone; 5: Ossa-Morena Zone.

obtained from sedimentation of clay suspensions on a glass slide. Relative abundance of clay minerals was estimated by empirical factors weighting the integrated peak areas of basal reflections. Accordingly, the glycolated 7 Å peak areas were multiplied by 0.5 to give kaolinite proportions, the 10 Å peak areas were multiplied by 1 to give illite proportion, the 14 Å peak areas were multiplied by 1 to give vermiculite proportions, the 15-17 Å peak areas were multiplied by 0.33 to give smectite proportions and the 12-14 Å peak areas were multiplied by 1 to give mixed layer clays (10-14 Å) proportions. Because chlorite was found just once and in fairly low proportions, its semi-quantification was not attempted. Given the uncertainties involved in quantification by XRD (Kahle et al., 2002), the results obtained by this approach are merely rough estimates of actual mineral percentages.

Crystallinity of illite was measured as the half-height width of the 10 Å peak (Kübler and Jaboyedoff, 2000). Diekmann et al. (1996) proposed four categories for illite crystallinity based on 10 Å peak width (<0.4: very well crystalline; 0.4-0.6: well crystalline; 0.6-0.8: moderately crystalline; >0.8: poorly crystalline). An approximation to illite chemistry was obtained from the ratio of intensities of 5 Å (I5) and 10 Å (I10) peaks (Esquevin, 1969; Gingele, 1996). According to Esquevin (1969), values > 0.4 correspond to Al-rich illites. The ratio decreases with Fe and Mg substituting the octahedral Al and formation of Fe, Mg rich illites. Gingele (1996) considered that an illite with I5/I10 ratio below 0.5 represents Fe and Mg rich illite, characteristic of physically eroded and unweathered materials. Ratio I5/I10 above 0.5 indicates Al-rich illites that were formed from strong hydrolysis.

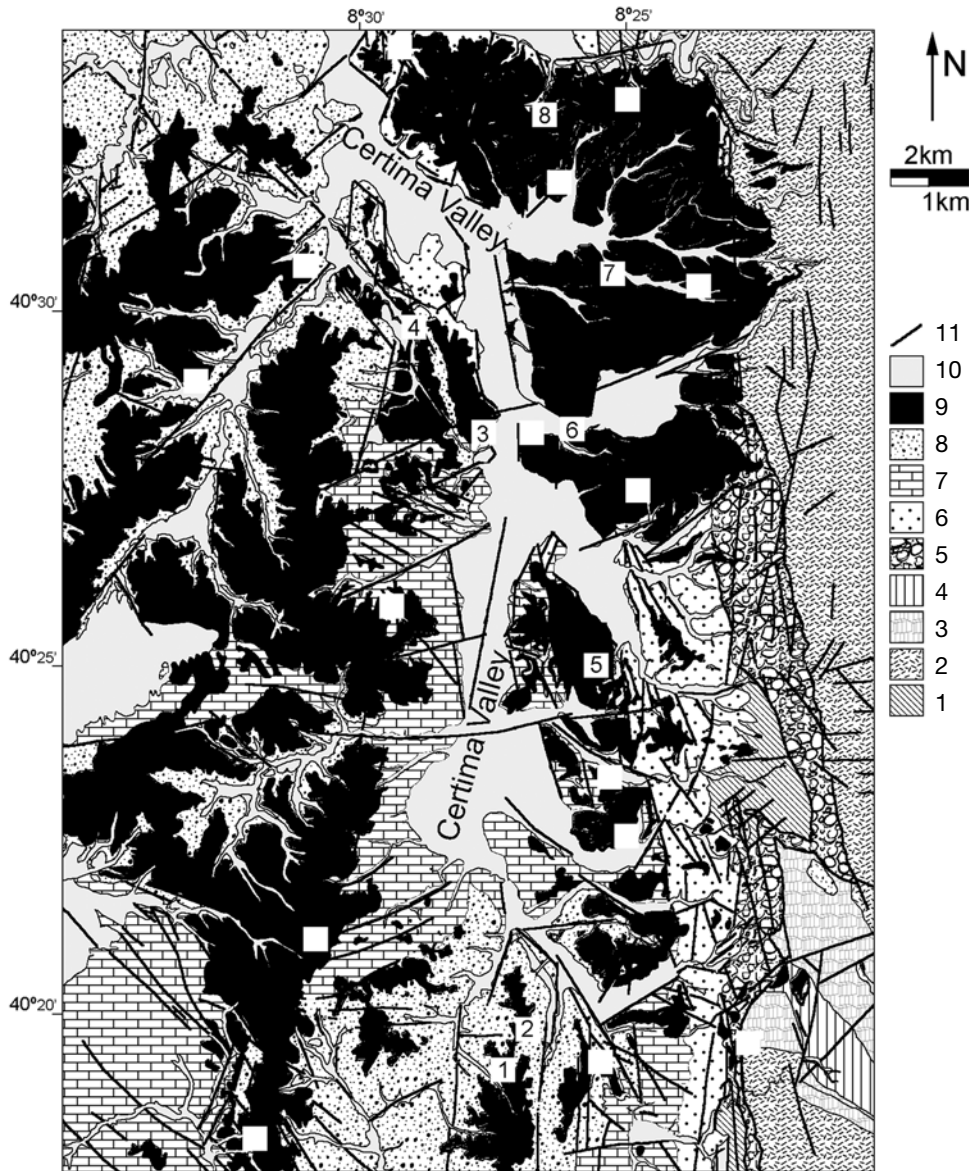


FIGURE 2 | Geological map of the studied coastal plain and surroundings (adapted from Dinis, 2004). Numbered white squares show location of sections presented in Figure 10. White squares indicate the location of other sampled sections. 1: Paleozoic and Precambrian of Ossa-Morena Zone; 2: Precambrian of Central Iberian Zone; 3: Ordovician; 4: Silurian; 5: Permian-Carboniferous; 6: Triassic; 7: Jurassic; 8: Cretaceous; 9: Studied deposits; 10: Quaternary terraces and raised beaches; 11: Fault.

**SEDIMENTARY FACIES**

**Inner shelf facies (FA)**

Facies FA (Fig. 4A) are dominated by meter scale, very well sorted sandstones with high compositional and textural maturity (Table 1). Although a structureless character is common, due to biologic obliteration of the initial structure, parallel lamination and oscillation ripples may be found. Intercalations of decimeter thick sandy-conglomerate beds with sharp erosive bases are also common. These beds consist of well rounded clasts of granule to pebble size and may be massive or show imbricated structures or cross-bedding. Thin mud part-

ings (centimeter scale) may also be intercalated in these sediments. Facies FA are barren of fossil elements. These facies are common at the base of the Neogene succession, though they can be found at higher stratigraphic levels westward.

The ubiquitous occurrence of well sorted very fine sand, the high roundness of coarse sand to gravel elements, the dominance of planar parallel lamination and the mainly tabular geometries indicate that facies FA represent inner shelf deposition. The absence of hummocky cross-bedding and the depositional sequence of conglomerates or rippled sandstones with granules followed by parallel laminated sandstones and oscillation ripple sand-

stones suggest coarse grained storm influenced deposits (Cheel and Leckie, 1992).

**Coastal subaquatic bar and dune facies (FB)**

Coarse to fine grained sands of moderate to high maturity are the most common sediments of facies FB (Table 1, Fig. 5). Intercalation of well sorted medium to fine grained sand beds and the frequency of well rounded coarse sand to centimeter size gravel dispersed in a sandy matrix or as laterally persistent thin horizons are frequent features. These sediments constitute the majority of the more westerly positioned sedimentary record. Three types of sediments can be considered. One consists of fine sands, a few decimeters to a meter in thickness, with low angle or undulated lamination, intercalated with decimeter thick beds of gravelly sands. Rhythmic interbedded onshore dipping and planar horizontal sand bodies were also found. Sometimes facies FB starts with a transgressive ravinement lag (Fig. 4B). The second type consists of coarse sand to fine gravel deposits in meter scale cross-stratified beds with unidirectional paleocurrents, usually directed to the west or northwest. The third type consists of 0.5-2 m thick beds with planar cross-stratification, that indicates bimodal or polymodal paleocurrents. These sediments may have sigmoidal foresets and millimeter to centimeter mud drapes. The second and third types usually contain outsized mud balls.

A deposition in coastal settings is proposed for facies FB on a basis of geometry (high lateral extent, scarcity of channel forms), textural features (presence of well sorted mature sand beds and roundness of sand and gravel elements) and basinward location in relation to mud dominated sediments (Dalrymple et al., 1992; Nishikawa and Ito, 2000; Anthony et al., 2002; Bhattacharya and Giosan, 2003).

**Fluvial channel facies (FC)**

Facies FC comprises submature sand and gravel deposits with frequent lenticular geometries (Table 1, Fig. 4E). The sedimentary bodies in plane view define ribbons with diverse sinuosity. Their geometry in transverse view (0.5 to 2 m thick) is characterized by a concave base surface and a planar top surface, but when several sand and conglomerate channel bodies stack over each other this geometry may be difficult to recognize. The paleocurrent data of facies FC depends on the location within the study area and stratigraphic position. Along the basin edge the gravelly channel deposits are mostly orientated east-west. The orientation of channels and cross-stratification on the western locations of the Cértima Basin tend to indicate north or northwest directed flows. High variability in paleocurrent directions is found northward (downstream) when sand dominated channel deposits are intercalated with thick mud deposits. In western locations of the Lit-

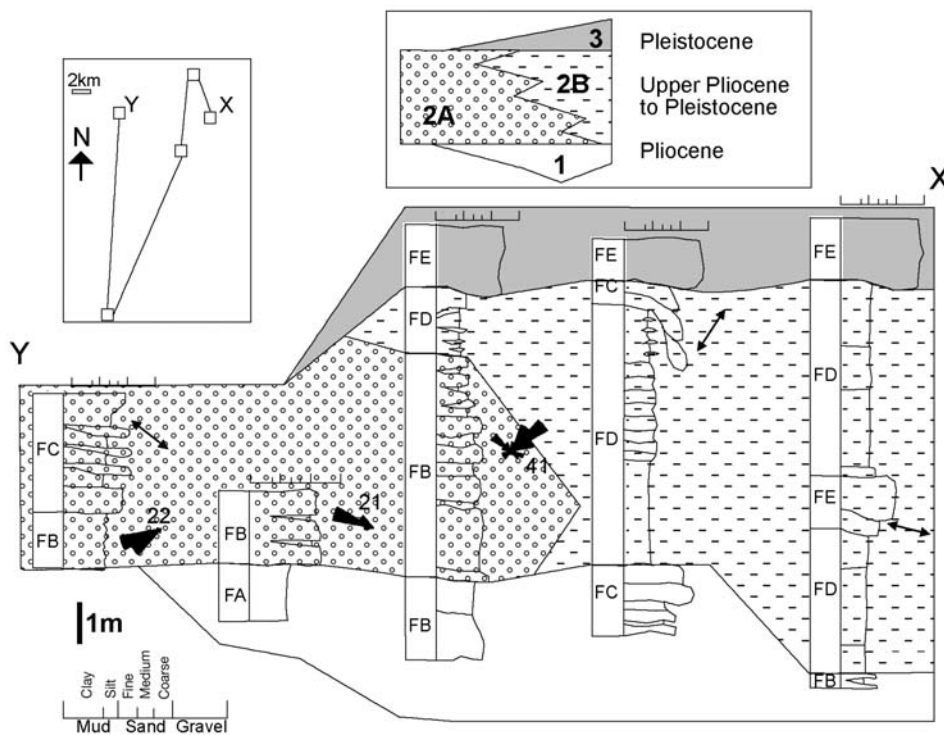


FIGURE 3 | Geometrical relations between the studied Pliocene and Pleistocene sedimentary units and facies. Number next to rose diagrams indicates the number of measurements in cross-stratified sediments. Double end arrowhead for mean channel orientation.

toral Horst, amalgamated channel bodies dominate the alluvial record.

The facies architecture seems to be primarily dependent of the accommodation conditions and regional dip. In the eastern fringe, at the contact with the uplifted Variscan Iberian Massif, the east-west channel orientation reflects a relatively high regional dip. The deflections to northward flows along the Cértima Basin were probably influenced by active tectonics. Locally the

channel bodies may be emplaced in narrow grabens. Although there are no clear evidences of lateral accretion deposits at broader subsiding locations of the Cértima Basin downstream, the variability in paleocurrent data and the thickness of intercalated mud floodplain deposits indicates relatively higher sinuosity systems in these positions. In the western locations the low variability in paleocurrents and limited proportion of fine grained deposits suggest channel amalgamation in a braided system (Friend, 1983).

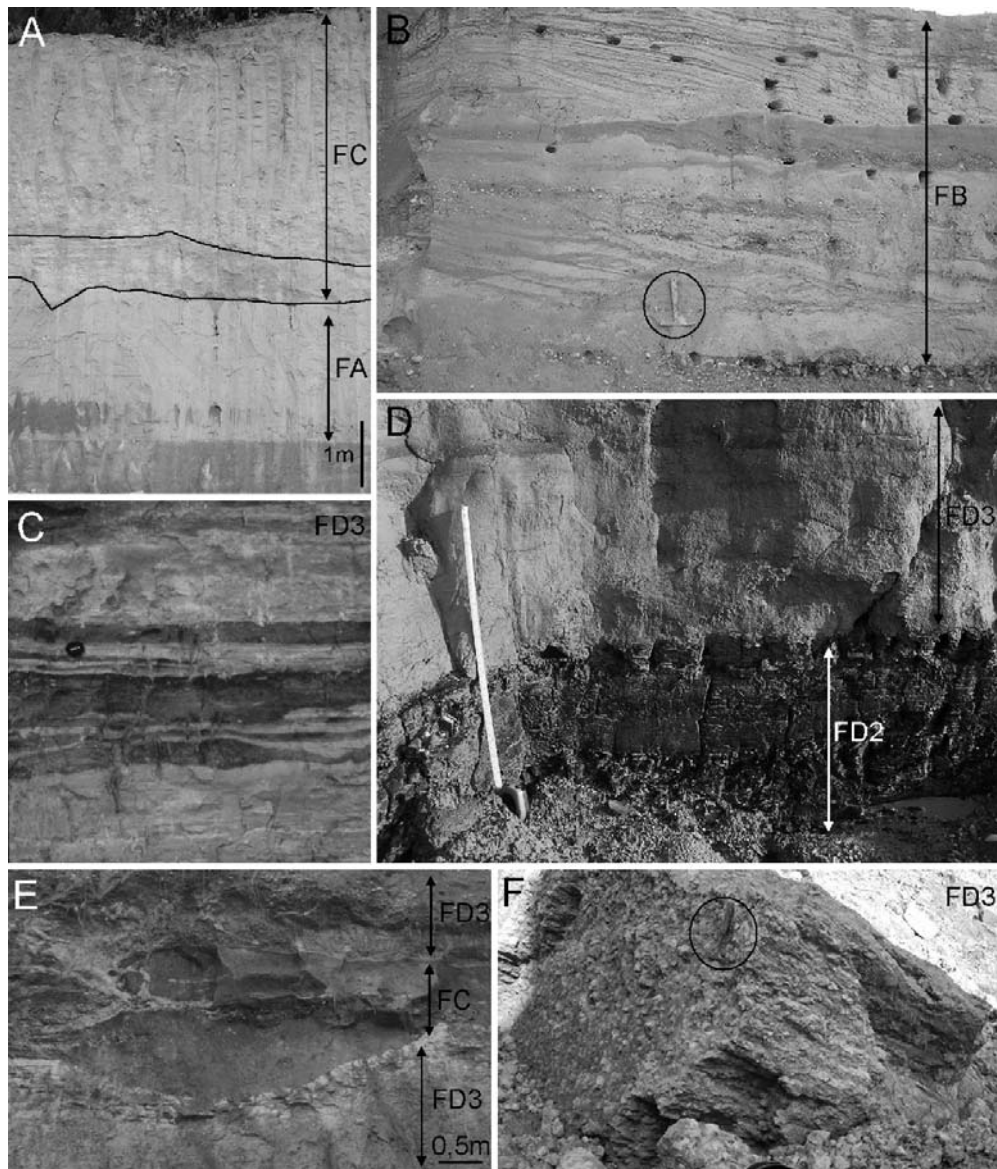


FIGURE 4 | Photographs of facies. A) Facies FA over Cretaceous friable sediments basement and followed, over unconformity, by facies FC. FA consists of intervened horizontally laminated fine-grained sandstones and thin horizons of coarse-grained sandstones or gravelly sandstones. FC is characterized by trough and planar cross-stratified sandstones and conglomerates, usually with erosive concave-up base (outlined), that define a fining-upward succession. B) Facies FB overlying a transgressive ravinement lag (hammer for scale). Here FB is characterized by low angle fine-grained sandstones interbedded with gravelly sandstones, sometimes with ripples. The paleocurrent pattern derived from these structures is bipolar. C) Laminated mud facies from floodplain deposition (FD3). D) Lignite bed between mud-rich grey sediments recording swamp deposition (FD2). (E) Channel bodies (FC) enclosed in mud sediments (FD). F) Vertical pedotubules that disrupt the depositional structure of facies FD3 (hammer for scale).

TABLE 1 | Facies characteristics.

Facies	Lithology	Geometry	Structures	Environment
FA	Very well sorted fine-grained sand and some conglomerates	Tabular, laterally persistent	Massive, horizontally laminated or ripple laminated	Inner shelf to lower shoreface
FB	Mature to sub-mature coarse sandstone. Some conglomerates and medium to fine sandstone	North-south ribbons. Westward and under thick mud sediments	Oblique cross stratification. Cases of polipolar pattern and of Gilbert type profile with Unidirectional basinward accretion	Dune and bar in coastal submarine and estuarine settings
FC	Varied sandstones and conglomerates.	Channel packages	Structureless or cross stratification. Normal grading	Fluvial channel
FD1	Plastic grey mudstone with intercalation of thin mature and immature sand beds.	Lenticular bodies. Intercalated with FB	Structureless, ripple lamination or parallel lamination	Lagoon and bay-head delta
FD2	"Dark mud with low sand content; peat beds"	1-5 meters thick lenticular bodies	Structureless	Ponds and shallow lakes
FD3	Mudstones and immature sandy mudstones	Sheets, lenses or lobes	Structureless, horizontal lamination, ripple lamination	Floodplain
FE	Immature conglomerate and sandstones	Thick sheets (1-2 m)	Structureless or horizontal lamination	Alluvial fan

### Distal alluvial, floodplain, palustrine and lagoon facies (FD)

Mud dominated sediments constitute facies FD (Fig. 5). These can be subdivided in three types (Table 1): coastal lagoon (FD1), ponds and swamps (FD2) and distal alluvial fans and floodplains (FD3). Due to the poor exposure it is not always easy to distinguish the three types of mud facies.

Facies FD1 are characterized by the presence of decimeter to meter thick beds of clayey mudstones with rare plant remains and intercalation of thin (1-30 cm) mature fine sand horizons. The mud sediments may interfinger with coastal bar deposits (FB) in the offshore direction. The geometrical relation with coastal deposits (facies FB) suggests that these sediments record lagoon and marginal lagoon deposition in a wide coastal plain (Anthony et al., 2002; Bhattacharya and Giosan, 2003).

Facies FD2 are characterized by the presence of meter thick grey or dark grey beds dominated by clay and silt grain size sediments, scarcity or absence of intercalated channel fill deposits and the presence of lignite or peat beds 2 to 3 m thick (Fig. 4D). These deposits are common in structurally subsiding sectors or upstream of uplifted blocks. The presence of peat beds, the occurrence of dark mudstones and the fine grained nature indicate that facies FD2 were deposited from suspension in poorly drained swamp or pond settings.

Facies FD3 are dominated by red, yellow or grey mudstones and immature sandy mudstones. A bimodal

grain-size distribution, with high frequency of 5-20 $\mu$ m and 100-250 $\mu$ m particles, is common (Fig. 5). The sediments can be structureless or have horizontal lamination or current ripple lamination (Fig. 4C). Heterolithic grey mud-red sand sediments with lenticular bedding may also be present. Although no well-organized paleosols were recognized, there are frequent signs of soil forming processes in facies FD3, such as sub-vertical tubular mottles that may disrupt the depositional structure (Fig. 4F).

### Proximal alluvial facies (FE)

Two types of proximal alluvial deposits, representative of two stratigraphic intervals, can be considered. The gravel size deposits of unit 3 are composed mostly of disorganized or crudely horizontally stratified pebble to cobble conglomerates. The clast composition varies significantly laterally, reflecting the variety of the source area. A wide range of clast roundness can be found in the same sample site. These facies constitute horizontally coalescent alluvial fans whose radius can reach 10 km. Sometimes unit 2B has gravel beds, with high mud content and high proportion of very angular slate and quartz clasts, which interfinger with mud dominated sediments (FD). Grain size and thickness of coarse grained bed decrease progressively downcurrent. These sediments are almost exclusively restricted to the Cértima Basin.

The immaturity, clay content, frequency of slate clasts, lobe geometries and progressive grain size decrease downcurrent indicate debris flow processes (Blair and McPherson, 1994). These processes are likely

to be more frequent in alluvial fan with drainage area basement rocks rich in clay (Blair and McPherson, 1994). The occurrence of water-lain sand and gravel lenses and the higher organization in unit 3 suggests mixed water-lain and debris flow processes. The presence of clasts with different roundness indicates reworking of previous deposits, of different characteristics, with coeval introduction of very angular, non-abraded materials from the Variscan Massif. This fact reinforces the supposition of mixed processes related to a higher variety in drainage basement rocks (Nichols and Thompson, 2005).

## PALEOGEOGRAPHIC RECONSTRUCTION

The Neogene record is organized in an overall regressive sequence. It usually starts with inner shelf deposits (FA), tends to be erosively overlain by coastal plain deposits (FB, FC and FD) and proximal alluvial facies (FE), whose thickness decreases westward. In western sections of the Littoral Horst, facies FA can be found intercalated with facies FB. In the subsiding areas east of river Cértima, above transgressive and prograding coastal sediments, the mudstone sediments (FD) usually start with lagoonal plastic mud layers that evolve upwards to floodplain and palustrine mudstones. These fine grained deposits are intercalated with alluvial sand and gravel (FC and FE), which tend to increase in thickness and grain size upwards.

Four paleogeographic phases are proposed on the basis of the stratigraphic and sedimentological data (Fig. 6). The first phase (Fig. 6A) is marked by inner shelf deposition until near the currently uplifted Variscan Massif. After this initial phase, high sediment input led to progressive seaward migration of the environments (Fig. 6B). The record is dominated by coastal mature and sub-mature sand and gravel deposits. The third phase is mani-

fested by deposition of thick alluvial and lacustrine deposits mainly sourced from the Variscan Massif and the development of alluvial systems directed northward in the Cértima Basin (Fig. 6C). These deposits are preserved just in places where subsidence was relatively pronounced. During this state the coastal deposition was displaced westward. The final phase (Fig. 6D) is marked by alluvial deposition that surpasses the more subsiding areas along the Cértima Basin edge and extends through a broader part of the coastal margin.

## CLAY MINERAL ASSEMBLAGES

Kaolinite and illite are the most abundant clay minerals (Table 2). Vermiculite, smectite and 10-14Å mixed layer clays are found widespread, usually in this rank proportion. Goethite and gibbsite are common accessory minerals. It is possible to distinguish three clay mineral assemblages based on the relative proportions of kaolinite, illite and remaining clay minerals (Fig. 7): 1) Kaolinite assemblage, where kaolinite is equal to or exceeds 90% of the clay fraction. 2) Kaolinite and illite assemblage, where kaolinite represents less than 90% and illite equals or surpasses the vermiculite, smectite and mixed clay content. 3) Mixed composition, where kaolinite represents less than 90% and vermiculite, smectite and mixed clay together exceeds illite proportions.

### Facies FA

Facies FA are usually kaolinite dominated, although a relatively high proportion of illite (can reach 60%) may occur (Fig. 8). The deficiency in other clay minerals is also characteristic of facies FA. Exceptions to this tendency occur near the topographic surface and in sediments close to regional unconformities. Moreover, higher proportions of kaolinite are observed in the vicinity of

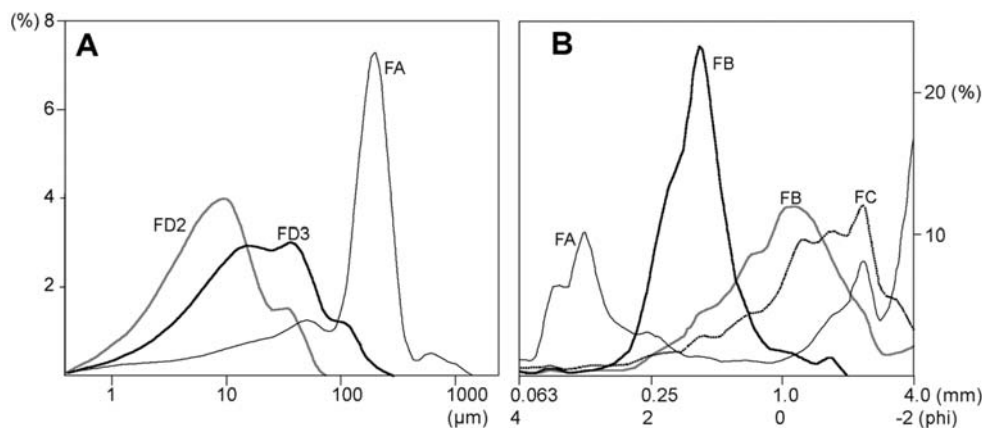


FIGURE 5 | Characteristic grain-size distributions analysed by laser diffraction (A) and sieving in 1/4φ column (B).



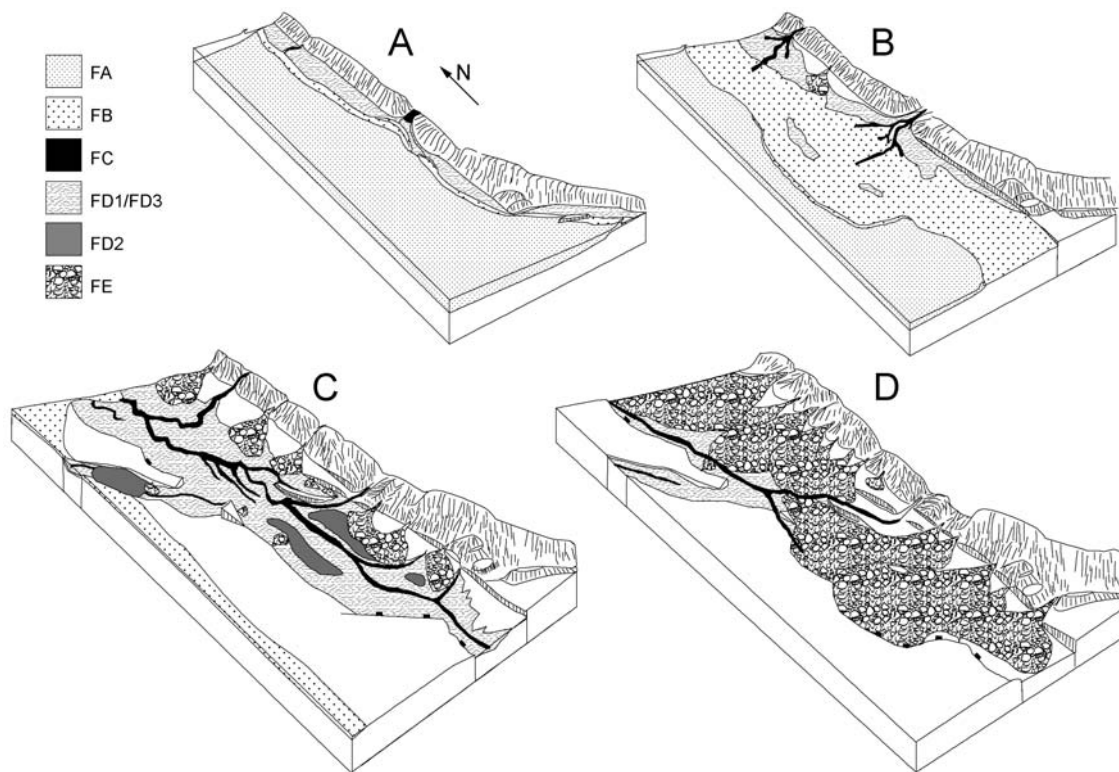


FIGURE 6 | Paleogeographic evolution of the area represented in Figure 2. See text for explanation.

unconformities when facies FA are in contact with previous kaolinite rich deposits.

Illite crystallinity in facies FA is extremely high (Fig. 9). Ninety per cent of the samples have the  $10 \text{ \AA}$  peak widths around or below 0.2. Most facies FA samples have I5/I10 ratio below 0.5 (89%), implying little chemical weathering.

### Facies FB and FC

Both facies (FB and FC) show kaolinite dominance (Fig. 8). Semiquantitative analysis indicates that kaolinite (always more than 45%) is more abundant than illite (0-44%). Significant proportions of vermiculite, smectite and mixed layer clays are frequent. Higher amounts of vermiculite and mixed layer clays were found in sedimentary levels immediately above or under unconformities. Near the topographic surface a significant enrichment in vermiculite was observed. Goethite and gibbsite are almost always present in accessory proportions.

Illite crystallinity is more variable than in the other facies (FB between 0.06 and 0.64; FC between 0.08 and 0.64). When  $10 \text{ \AA}$  peak widths are conjugated with XRD I5/I10 peak intensity ratio, two groups are well outlined for facies FB and FC (Fig. 9). Samples of wider illite peaks have higher I5/I10 relations, while samples of sharper illite peaks have lower I5/I10 relations. All sam-

ples of relatively aluminous illite and low crystallinity illite were collected in stratigraphic high horizons, close to the surface and in western flat landscape areas.

### Facies FD

Facies FD always contain kaolinite and illite, though in quite variable proportions (Fig. 8). Two groups arise from the semiquantitative analysis: one with divided domain of kaolinite and illite; the other with lower proportions of kaolinite and higher quantities of the remaining clay minerals. Kaolinite (30-77%) together with illite (23-60%) constitutes the majority of the clay fraction in

TABLE 2 | Mineralogy of clay fraction (mean values).

Facies	N	K	Il	Vr	Sm	ML	IC	KC
		%	%	%	%	%		
FA	17	68	28	4	0	0	0.11	0.23
FB	65	78	13	9	0	0	0.19	0.29
FC	16	71	22	7	0	0	0.23	0.21
FD1	12	50	37	8	1	4	0.12	0.16
FD2	8	14	78	5	3	0	0.12	0.11
FD3	49	39	50	7	2	2	0.14	0.17
FE	11	34	46	16	0	3	0.14	0.16

N: Number of samples. K: Kaolinite; Il: Illite; Vr: Vermiculite; Sm: Smectite; ML: Mixed layer clays. Values represent semi-quantitative proportion approximation based on weighted peak areas. IC: Illite crystallinity index; KC: Kaolinite crystallinity index.

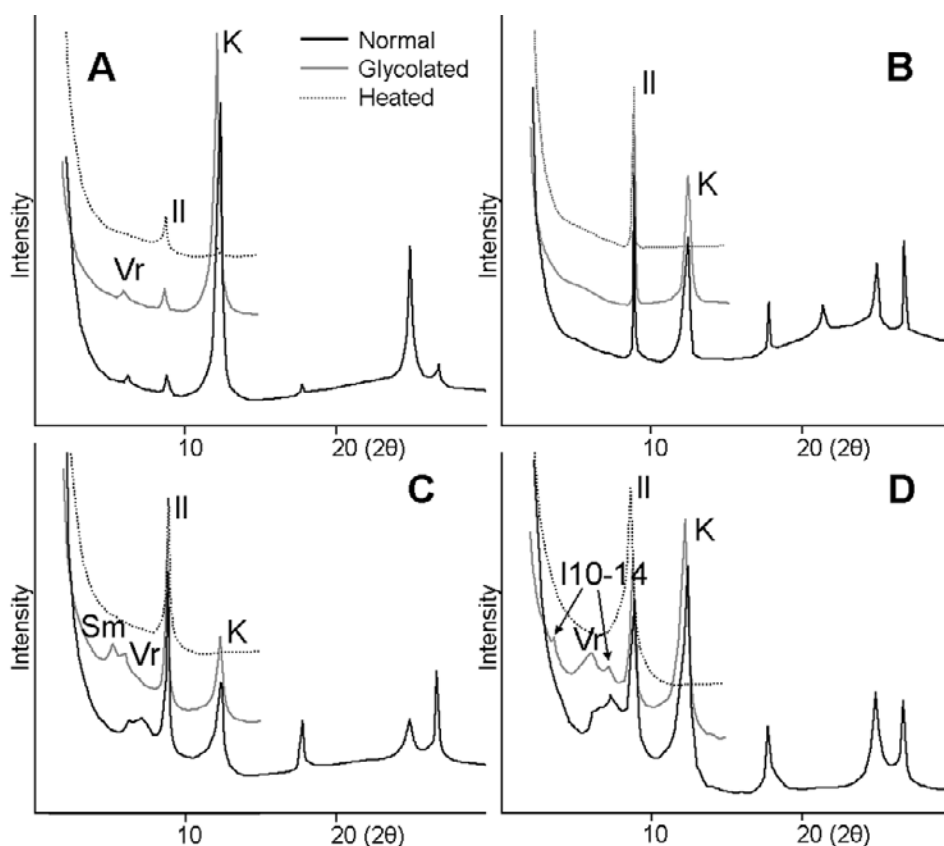


FIGURE 7 | Some typical XRD responses of kaolinite dominated assemblages (A), kaolinite-illite assemblages (B) and mixed composition (C and D). II: illite; K: kaolinite; Vr: vermiculite; Sm: Smectite; I10-14: 10-14Å mixed layer clays.

facies FD1. Sporadically, vermiculite and mixed layer clay minerals may be present in amounts comparable to illite or kaolinite. Facies FD2 always contain more illite (62-87%) than kaolinite (8-20%). Vermiculite, smectite and mixed layer clay minerals may be present in moderate proportions, although usually below kaolinite content. Facies FD3 may be kaolinite dominated (11-95%), illite dominated (4-84%) or vermiculite dominated (0-50%). An illite-smectite association was found in one sample.

The crystallinities measured in facies FD are especially high, since all but one of these facies samples have illite peak widths below  $0.3^\circ$  (Fig. 9). The exception is one sample of alluvial plain sediments that show clear pedogenic features. A relatively broad compositional range can be regarded for facies FD1 and FD3 (XRD  $5 \text{ \AA}/10 \text{ \AA}$  intensity ratio values between 0.25 and 0.75), though most samples have  $5 \text{ \AA}/10 \text{ \AA}$  ratio below 0.5 (70% of facies FD1; 80% of facies FD3). All FD2 samples have XRD  $5 \text{ \AA}/10 \text{ \AA}$  intensity ratio between 0.28 and 0.34.

### Facies FE

Clay compositions in facies FE are comparable to those of facies FD1 (Fig. 8). Kaolinite tends to be less

abundant than in other sand and gravel facies (mainly FB and FC). The more abundant mineral can be illite (9-80%), kaolinite (17-73%) or vermiculite (0-42%). Mixed layer clay minerals are secondary (0-10%) and smectite was found in trace proportions (<1%).

Illite crystallinity is very high ( $10 \text{ \AA}$  peak widths always below  $0.4^\circ$  and 90% below  $0.2^\circ$ ). The chemical composition inferred from XRD  $5 \text{ \AA}/10 \text{ \AA}$  intensity ratio is quite variable (between 0.28 and 0.85). However, a stratigraphic separation can be established, since unit 3 samples usually have higher  $5 \text{ \AA}/10 \text{ \AA}$  ratios than unit 2B.

## DISCUSSION

### Provenance

The studied sediments were deposited in a tectonically active setting and, as these conditions should favour detrital clay mineral assemblages (Chamley, 1989), the clay mineral composition should be, at least partially, explained by the source rock mineralogy. The associations controlled by sourced rock depend on the local type of basement and paleoflow pattern (Net et al., 2002). The

clay mineralogy of the Triassic to Cretaceous Lusitanian Basin rocks is dominated by kaolinite and illite, with lesser proportions of smectite and chlorite (Soares et al., 1986; Rocha, 1993). A link with the mineralogy of local basement rocks is supported by the higher kaolinite content of facies FA, when overlying Cretaceous kaolinite-rich sediments, and by the single occurrence of chlorite in sediments covering Triassic to Jurassic units that contain this mineral. The drainage basin basement rocks of long fluvial systems directed from hinterland areas comprises pelitic metamorphic rocks and granitoids. There are also some depressed areas covered by Cretaceous and Cainozoic sediments that generally have high kaolinite content at the base ("siderolithic" succession) and varied proportions of illite and smectite (Daveau et al., 1985-86; Cunha, 1992). The Variscan Iberian Massif rocks in the contact fringe with the Lusitanian Basin are mica-rich pelitic rocks. Regarding the main paleoflow, two distinct sediment paths may be deduced from the paleocurrent data, facies geometrical relationship and consequent paleoenvironmental reconstructions (Fig. 6). One is related to relatively long fluvial systems that derive sediment from hinterland areas or from the drainage basement rocks of the Lusitanian Basin. The prograding coastal plain facies FB, FC and part of FD are associated with this sediment path. It seems likely that a derivation from those kaolinite rich areas should contribute to the high kaolinite proportion in these facies. The other sediment path is related to short streams from the Variscan Massif, which is the main feeder of the endorheic basins with interbedded facies FD and FE. This sediment path should supply mostly illite. The dispersal system within inner shelf settings and the presence of facies FA both close to the Variscan Iberian Massif and basinward of a wide coastal plain should result in more complex sediment sources.

The hypothesis of significant source rock control over clay mineral assemblages is favoured by the high illite crystallinities and Fe, Mg rich chemical composition (Fig. 9). Facies FA, FD and FE have extremely high illite crystallinities. These illite characteristics in facies FD and FE from the Cértima Basin may be explained by the proximity from mica-rich source rocks. The high illite crystallinity in facies FA, despite the sample site location, indicates minimal structural deterioration during transportation to the submarine coastal setting and sedimentation.

Sea level changes may also contribute to explain some variations in clay mineralogy. In coastal sediments the increase in stream gradients during periods of low sea level may be responsible for an increase in kaolinite proportion because these streams may cause a higher stripping of the pre-existing cover (Thiry and Jacquin, 1993; Gibson et al., 2000). During phases of low sea level the erosive action in source areas is enhanced and important mobilization and re-deposition of previously formed tend to occur. Given the presence of a "siderolithic" succession covering hinterland basins, which is very rich in kaolinite, higher percentages of this mineral may be expected during low sea level deposits. Transgressive deposits may have less kaolinite because of the tendency for a decrease in stream gradients and lower sediment input. During highstand phases the prograding deposits may also have high kaolinite content, especially if sourced from hinterland areas, and the more proximal sediments tend to be enriched in kaolinite when compared to inner shelf deposits. The decrease in kaolinite basinward may also be related to selective settling, since this mineral is usually coarser than the other clay minerals and tends to settle in more proximal environments (Chamley, 1989; Šimkevičius

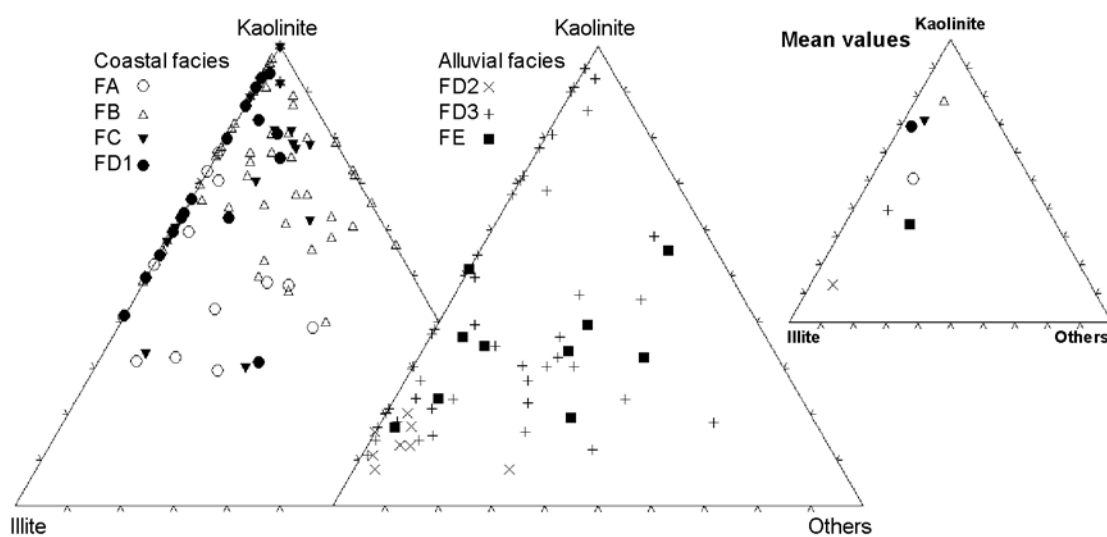


FIGURE 8 | Clay mineral composition of facies. The smaller triangle at right indicates mean values.

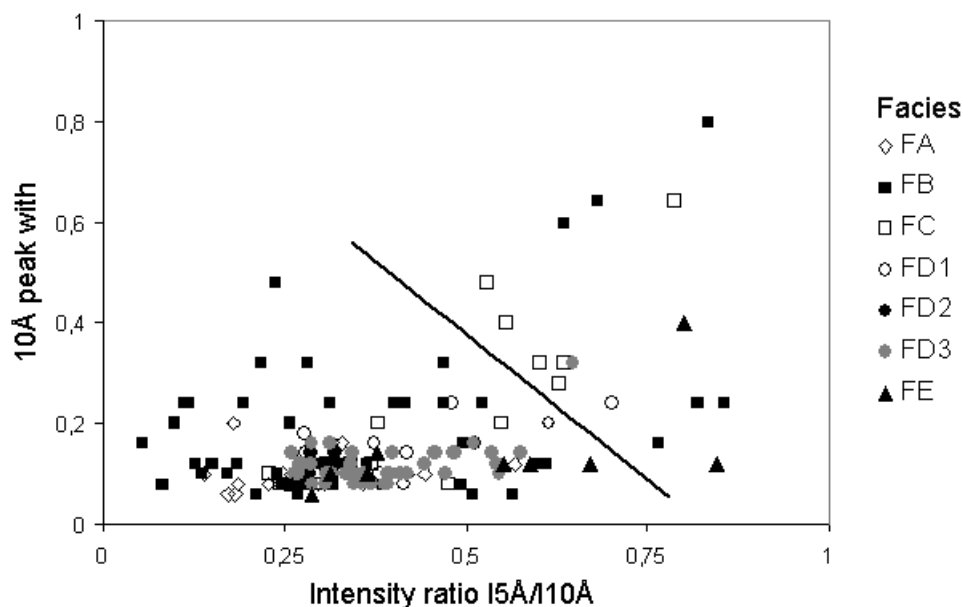


FIGURE 9 | Esquevin diagram with the relation between illite composition, inferred from intensity ratio of peaks 5 Å and 10 Å (X axis), and illite crystallinity, inferred from 10Å peak width at mean height (Y axis). Note the presence of two groups with different crystallinities and composition. Most aluminous illite (high I5/10 ratio) also has lower crystallinities and is usually found in facies FB and FC.

et al., 2003). A similar trend of illite increase seaward and kaolinite increase landward is observed in the current shelf sediment cover offshore of the study area (Oliveira et al., 2002).

## Climate

The paleofloral and paleofaunal records of Central West Iberia suggest that the climate has changed slightly during the last 3 Ma and passed through several arid and humid phases (Diniz, 1984; Pais, 1989; Antunes and Pais 1993). A global increase in temperature and humidity, recognized as “mid-Pliocene optimum”, took place around 3.0 Ma (Crowley, 1996). It was accompanied by a transgressive event that affected Europe and is recorded in several locations of the Portuguese coastal margin by Piacenzian inner shelf deposits (Antunes and Pais, 1993; Silva, 2001). Unit 1 was probably deposited during this period. After the “mid-Pliocene optimum” there was an intensification of the North hemisphere glacial-interglacial cycles (Montuire, 1999; Fauquette et al., 1999; van Dam, 2006). During the upper Pliocene and early Pleistocene the climate was affected by high frequency, 41 Ky-cycles, of changing temperature (Raymo et al., 1990; Berger et al., 1999). During the upper and middle Pleistocene the climate was influenced by longer term and amplified glacial-interglacial cycles (Raymo et al., 1990; Berger et al., 1999; Elkibbi and Rial, 2001). Unit 3 was probably deposited during a period affected by glacial-interglacial alternations of lower Pliocene or upper Pleistocene age.

In some sections with alluvial facies it is possible to recognize a slight increase in the illite/kaolinite relation upwards suggesting an evolution to more arid conditions (Fig. 10). However, an opposite trend also occurs, in particular when coastal plain unit 2A covers inner shelf or mud dominated deposits. The expected relative homogeneity in climatic conditions (despite the Plio-Pleistocene climatic cycles) and the clear mineralogy dependency from provenance, as alleged before, suggest that climate should not be a significant controlling agent on the variations in clay mineralogy. Actually, as coeval sediments show spatial variations of clay mineralogy (Fig. 10), other factors should be more effective in controlling the observed variability in clay assemblages.

## Post-depositional clay transformation

Although source area contribution plays the major role in clay composition some post-depositional clay formation can be expected in particular circumstances. Kaolinite formation under acidic water conditions is frequent in swamp and swamp related environments (Chamley, 1989; Sáez et al., 2003). The studied swamp sediments (facies FD2) have low kaolinite content and are rich in illite. However, these sediments usually have relatively high vermiculite and 10-14Å mixed layer clays that can be related to the deterioration of mica type minerals under low pH conditions.

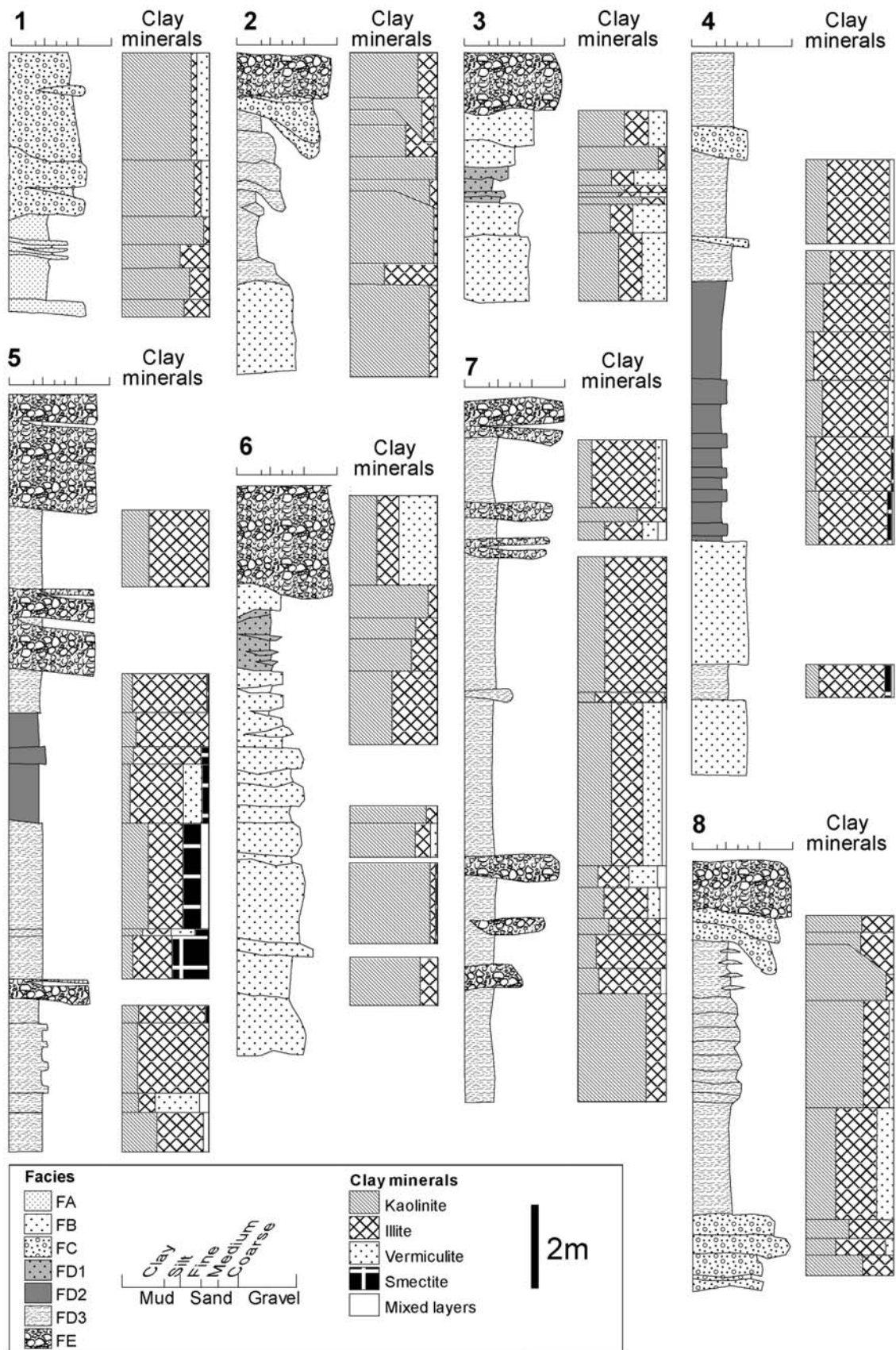


FIGURE 10 | Vertical variation of clay mineral composition in selected section. Section locations in Figure 2.

Vermiculite, smectite and 10-14Å mixed layer clays may be formed during sub-aerial exposure from the degradation of mica minerals by pedogenetic processes (Singer, 1980; Chamley, 1989; Braga et al., 2002; Pe-Piper et al., 2005). The presence of vermiculite and mixed layer clays close to unconformities and in levels with signs of pedogenesis (Fig. 10) suggest that these minerals are related to soil forming processes. The presence of vermiculite close to the land surface and the upward increase in its proportion advocates that it was also formed by current soil forming processes.

Kaolinite formation under the influence of meteoric low pH waters may occur in high porous facies FA, FB and FC. Owing to the arkosic composition of some of these sand and conglomerate sediments, the hypothesis of post-burial kaolinite formation from in situ weathering of feldspar under acidic pore water conditions must be considered. This hypothesis is supported by the presence of illite with relatively low crystallinity and high aluminous content in these sediments, suggesting chemical weathering and structural degradation of more liable minerals and in place formation of new, aluminous-rich, illites. The absence of shell fragments in coastal FA and FB deposits (Dinis, 2004), as they tend to be common in these facies and may be found in mud rich levels of coeval sediments from southern locations indirectly suggests the occurrence of low pH pore water conditions. The higher silt and clay content in facies FA when compared to present day inner shelf deposits, which may be explained by a contribution from post-depositional weathering in the former, reinforces the possibility of post depositional formation of clay minerals.

## CONCLUSIONS

The high crystallinity of clay minerals, the links between Neogene clay composition and the major sediment paths and basement rocks mineralogy indicates that the clay assemblages are primarily dependent from source sediments. Hence, climate should have a subsidiary role in the clay mineral variability. Illite content and crystallinity are especially high in intercalated mud and sand-conglomerate deposits located close to the Variscan Iberian Massif, reflecting provenance from mica-rich pelitic rocks through alluvial systems and limited mixing with other sources. The illite abundance in inner shelf deposits is also slightly greater than in coarser and more proximal coastal plain deposits that are kaolinite dominated. Post-depositional overprint on clay mineralogy is explained by three processes. Meteoric flushing in permeable sediments (e.g. sand and gravel deposits) may have led to kaolinite content increase. Weathering in paleosol horizons led to the formation of vermiculite and 10-14Å mixed layer clays. Finally, in organic-rich deposits it

occur the transformation of illite to vermiculite, smectite and/or 10-14Å mixed layer clays. The extent of mineral changes suggests relatively low weathering rates. This process was probably more active in low relief and long exposed coastal plain deposits, where the clay fraction may also contain goethite and gibbsite vestiges.

The variations in clay mineralogy are in agreement with the conceptual paleogeographic model deduced from the facies analysis. Although the clay mineralogy is chiefly dependent from provenance, it is possible to recognize overprints determined by several paleogeographic aspects like climate, tectonics, relief, hydrology and paleoflow. There are evident links between the mineral assemblages and depositional environments and the paleogeographic conditions.

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## REFERENCES

- Ahlberg, A., Olsson, I., Simkevicius, P., 2003. Triassic-Jurassic weathering and clay mineral dispersal in basement areas and sedimentary basins of southern Sweden. *Sedimentary Geology*, 161, 15-29.
- Anthony, E.J., Oyédé, L.M., Lang, J., 2002. Sedimentation in a fluviially infilling, barrier-bound estuary on a wave-dominated, microtidal coast, the Ouémé River estuary, Benin, west Africa. *Sedimentology*, 49, 1095-1112.
- Antunes, A.T., Pais, J., 1993. The Neogene of Portugal. *Ciências da Terra*, 12, 7-22.
- Berger, A., Li, X.S., Loutre, M.F., 1999. Modelling northern hemisphere ice volume over the last 3Ma. *Quaternary Science Reviews*, 18, 1-11.
- Bhattacharya, J.P., Giosan, L., 2003. Wave-influenced deltas: geomorphological implications for facies reconstruction. *Sedimentology* 50, 187-210.
- Blair, T.C., McPherson, J.G., 1994. Alluvial fans and their natural distinction from rivers based on morphology, hydrolic processes, sedimentary processes and facies assemblages. *Journal of Sedimentary Research*, A64, 450-489.
- Braga, M.A.S., Paquet, H., Begonha, A., 2002. Weathering of granites in a temperate climate (NW Portugal): granitic saprolites and arenization. *Catena*, 49, 41-56.

- Cabral, J., 1995. Neotectónica em Portugal Continental. Memórias do Instituto Geológico e Mineiro, 31, 1-265.
- Chamley, H., 1989. Clay sedimentology. Berlin, Springer Verlag, 623 pp.
- Cheel, R.J., Leckie, D.A., 1992. Coarse-grained storm beds of the upper Cretaceous Chungo Member (Wapiabi Formation), Southern Alberta, Canada. *Journal of Sedimentary Petrology*, 62, 933-945.
- Crowley, T.J., 1996. Pliocene climates; the nature of the problem. *Marine Micropaleontology*, 27, 3-12.
- Cunha, P.M., 1992. Estratigrafia e sedimentologia dos depósitos do Cretácico Superior e Terciário de Portugal Central, a Leste de Coimbra. Doctoral thesis. University of Coimbra, 262 pp.
- Cunha, P.M., Barbosa, B.P., Reis, R.P., 1993. Synthesis of the Piacenzian onshore record, between the Aveiro and Setúbal parallels (Western Portuguese margin). *Ciências da Terra*, 12, 35-43.
- Dalrymple, R.W., Zaitlin, B.A., Boyd, R., 1992. Estuarine facies models: conceptual basis and stratigraphic implications. *Journal of Sedimentary Petrology*, 62, 1130-1146.
- Daveau, S., Birot, P., Ribeiro, O., 1985-86. Les bassins de Lousã et Arganil. Recherches géomorphologiques et sédimentologiques sur le massif ancien et sa couverture a l'est de Coimbra. *Memórias do Centro Estudos Geográficos*, 8, 1-450.
- Diekmann, B., Petschick, R., Gingele, F.X., Fütterer, D.K., Abelman, A., Gersonde, R., Mackensen, A., 1996. Clay mineral fluctuations in Late Quaternary sediments of the southeastern South Atlantic: Implications for past changes of deep-water advection. In: Wefer, G., Berger, W.H., Siedler, G., Webb, D. (eds.). *The South Atlantic: Present and Past Circulation*. Heidelberg, Springer, 621-644.
- Dinis, P.A., 2004. Evolução pliocénica e quaternária do vale do Cértima. Doctoral thesis. University of Coimbra, 351 pp.
- Diniz, F., 1984. Apports de la palynologie a la connaissance du Pliocène Portugais. Rio Maior un bassin de référence pour le histoire de la flore, de la vegetation et du climat de la façade atlantique de l'Europe méridionale. Doctoral thesis. University of Montpellier, 230 pp.
- Elkibbi, M., Rial, J.A., 2001. An outsider's review of the astronomical theory of climate: is the eccentricity-driven insolation the main driver of the ice ages? *Earth-Science Reviews*, 56, 161-177.
- Esquevin, J., 1969. Influence de la composition chimique des Illites sur cristallinite. *Bulletin du Centre de Recherches de Pau*, 3, 147-153.
- Fauquette, S., Suc, J.-P., Guiot, J., Diniz, F., Feddi, N., Zheng, Z., Bessais, E., Drivaliari, A., 1999. Climate and biomes in the West Mediterranean area during the Pliocene. *Palaeogeography, Palaeoclimatology, Palaeocology*, 152, 15-36.
- Ferreira, A.B., 1991. Neotectonics in Northern Portugal. A geomorphological approach. *Zeitschrift für Geomorphologie*, v. Suppl. Bd. 82, 73-85.
- Friend, P.F. 1983. Towards the field classification of alluvial architecture or sequence. *Modern and Ancient Fluvial Systems*. In: Collinson, J.D., Lewin, J. (eds.). *Special Publication of International Association of Sedimentologists*, 6, 345-354.
- Gibson, T.G., Bybell, L.M., Mason, D.B., 2000. Stratigraphic and climatic implications of clay mineral changes around the Paleocene/Eocene boundary of the northeastern US margin. *Sedimentary Geology*, 134, 65-92.
- Gingele, F.X., 1996. Holocene climatic optimum in Southwest Africa -evidence from the marine clay mineral record. *Palaeogeography, Palaeoclimatology, Palaeocology*, 122, 77-87.
- Kahle, M., Kleber, M., Jahn, R., 2002. Review of XRD-based quantitative analyses of clay minerals in soils: the suitability of mineral intensity factors. *Geoderma*, 109, 191-205.
- Keller, W.D., 1970. Environmental aspects of clay minerals. *Journal of Sedimentary Petrology*, 40, 788-814.
- Kübler, B., Jaboyedoff, M., 2000. Illite crystallinity. *Comptes Rendus de l'Académie des Sciences*, 331, 75-89.
- Montuire, S., 1999. Mammalian faunas as indicators of environmental and climatic changes in Spain during the Pliocene-Quaternary transition. *Quaternary Research*, 52, 129-137.
- Net, L.I., Alonso, M.S., Limarino, C.O., 2002. Source rock and environmental control on clay mineral associations, Lower Section of Paganzo Group (Carboniferous), Northwest Argentina. *Sedimentary Geology*, 152, 183-199.
- Nichols, G., Thompson, B., 2005. Bedrock lithology control on contemporaneous alluvial fan facies, Oligo-Miocene, Southern Pyrenees, Spain. *Sedimentology*, 52, 571-585.
- Nishikawa, T., Ito, M., 2000. Late Pleistocene barrier-island development reconstructed from genetic classification and timing of erosional surfaces, paleo-Tokyo Bay, Japan. *Sedimentary Geology*, 137, 25-42.
- Oliveira, A., Rocha, F., Rodrigues, A., Jouanneau, J., Dias, A., Weber, O., Gomes, C., 2002. Clay minerals from the sedimentary cover from the northwest Iberian Shelf. *Progress in Oceanography*, 52, 233-247.
- Pais, J., 1989. Evolução do coberto florestal em Portugal no Neogénico e no Quaternário. *Comunicações dos Serviços Geológicos de Portugal*, 75, 67-72.
- Pe-Piper, G., Dolansky, L., Piper, D.J.W., 2005. Sedimentary environment and diagenesis of the Lower Cretaceous Chaswood Formation, southeastern Canada: The origin of kaolin-rich mudstones. *Sedimentary Geology*, 178, 75-97.
- Raymo, M.E., Ruddiman, W.F., Shackleton, N.J., Oppo, D.W., 1990. Evolution of Atlantic-Pacific  $\delta^{13}\text{C}$  gradients over the last 2.5 m.y. *Earth and Planetary Science Letters*, 97, 353-368.
- Rocha, F.J., 1993. Argilas aplicadas a estudos litostratigráficos e paleoambientais na Bacia Sedimentar de Aveiro. Doctoral thesis. University of Aveiro, 399 pp.
- Sáez, A., Inglès, M., Cabrera, L., Heras, A., 2003. Tectonic paleoenvironmental forcing of clay-mineral assemblages in nonmarine settings: the oligocene-Miocene As pontes Basin (Spain). *Sedimentary Geology*, 159, 305-324.

- Silva, C.M. 2001. Gastrópodes pliocénicos marinhos de Portugal. Sistemática, paleobiologia, paleoecologia e paleobiogeografia. Doctoral thesis. University of Lisbon, 675 pp.
- Šimkevičius, P., Ahlberg, A., Grigelis, A., 2003. Jurassic smectite and kaolinite trends of the East European Platform: implications for palaeobathymetry and palaeoclimate. *Terra Nova*, 15, 225-229.
- Singer, A., 1980. The paleoclimatic interpretation of clay minerals in soils and weathering profiles. *Earth-Science Reviews*, 15, 303-326.
- Soares, A.F., 1999. As unidades quaternárias e pliocénicas no espaço do Baixo Mondego (uma perspectiva de ordem). *Estudos do Quaternário*, 2, 7-17.
- Soares, A.F., Lapa, M.L., Marques, J.F., 1986. Contribuição para o conhecimento da litologia das unidades Mesozoóicas da bacia Lusitaniana a Norte do acidente da Nazaré (sub-zona setentrional). *Memórias e Notícias, Publicações do Museu Laboratório Mineralógico e Geológico da Universidade de Coimbra*, 102, 23-41.
- Thiry, M., 2000. Paleoclimatic interpretation of clay minerals in marine deposits: an outlook from the continental origin. *Earth-Sciences Reviews*, 49, 201-221.
- Thiry, M., Jacquin, T., 1993. Clay mineral distribution related to rift activity, sea level changes and paleoceanography in the Cretaceous of the Atlantic Ocean. *Clay Minerals*, 28, 61-84.
- van Dam, J.A., 2006. Geographic and temporal patterns in the late Neogene (12-3Ma) aridification of Europe: the use of small mammals as paleoprecipitation proxies. *Palaeogeography, Palaeoclimatology, Palaeoecology*, 238, 190-218.

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