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Palaeoenvironment of the Aveiro region of Portugal during the Cretaceous, based on clay mineralogy[1]

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The northern margin of the Mesozoic–Cenozoic Lusitanian (Western Portuguese) Basin is known mostly from borehole sections. Mineralogical studies of the subsurface Cretaceous are based on XRD, XRF, TEM and flame-spectroscopy of cuttings. Clay mineralogy varies stratigraphically, with kaolinite dominating in the Aptian/Albian/Cenomanian and late Turonian–early Coniacian. Illite is common in the late Cenomanian and Campanian–Maastrichtian. Smectite increases in abundance through the Late Cretaceous. Lateral variations in this division characterize palaeogeography and specific tectonic controls. The overall clay supply depended upon the varying influence of sea level, climate and palaeogeography.

KEY WORDS: Clay minerals; palaeoenvironmental reconstruction; Cretaceous; Aveiro; Portugal.

1. Introduction

The Aveiro region, located on the central–north Atlantic margin of Portugal (Figure 1), constitutes the northern section of the Western Portuguese Mesozoic–Cenozoic sedimentary basin.

During the Early Cretaceous a central graben was formed on the Portuguese Atlantic margin, flanked to the west by a horst and to the east by a system of semi-grabens being less and less developed towards the Hesperic Massif (Rey, 1982, 1984). In these grabens, carbonate deposits of platform facies accumulated, passing laterally into siliciclastic sediments of transitional and continental facies.

During the Late Cretaceous a great expansion to the NE of the basin occurred, related to thermal subsidence produced by the onset of ocean crust emplacement. Some alluvial fans developed which correspond to the base of the Cretaceous to the north of the Nazaré parallel (Pena dos Reis *et al.*, 1992). Due to both eustatic sea-level rise and a decrease in thermal subsidence, retrogradation of alluvial fans and on-lap of carbonate deposits took place. The transgression reached its maximum extent at the end of Cenomanian (Berthou, 1984).

Conversely, lowering of eustatic sea level in conjunction with tectonic uplift produced a progradation of the alluvial systems which started with micaceous clayey-sandy deposition and ended during the early Campanian with a silicification episode (Cunha, 1992; Cunha *et al.*, 1992).

During the Campanian Iberia and Africa collided, discontinuing the opening of Biscay. The nature of the sediments express intricate fluvial systems draining towards the NW, with transitional to lagoonal and marine environments (Pena dos Reis *et al.*, 1992).

Recently, the subsurface geology of the Aveiro region has been studied in

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some detail. Most of the information comes from mineralogical and sedimentological data provided by studies carried out on cutting samples from many deep boreholes drilled for water supply. Cretaceous formations are very well represented. The deposits are essentially siliciclastic. Figure 2 presents the lithostratigraphical units, with their ages according to Teixeira & Zbyszewski (1976) and Barbosa (1981). Work on sea level, palaeoclimate, tectonic and palaeogeographic interpretation of the area has been carried out previously by several authors including Teixeira & Zbyszewski (1976), Barbosa (1981), Lauverjat (1982), Berthou *et al.* (1982), Berthou (1984) and Rocha (1993).

Clay minerals and the accompanying non-clay minerals were utilized for lithostratigraphical discrimination, definition of palaeosurfaces and palaeoenvironmental reconstruction. Their nature, quantification and crystallochemical features (in particular, the crystallinity of the main clay minerals—kaolinite, illite and smectite) has led to a zonation of the Cretaceous sediments as well as to the definition of the conditions of deposition. The sedimentological and mineralogical studies were based on cuttings taken from 27 boreholes, whose locations are shown in Figure 1.

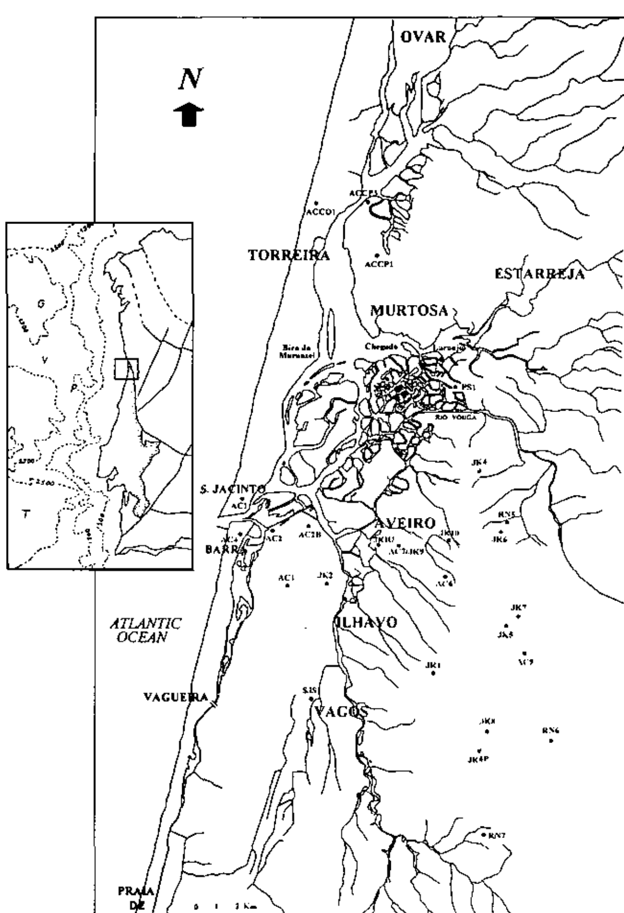

Figure 1. Map of Aveiro region showing location of studied boreholes.

2. Analytical results

Mineralogical studies were based mainly on X-ray diffraction (XRD) determinations, carried out on both the <38 μm and <2 μm fractions, using Cu Kα radiation and a Philips diffractometer (PW 1130/90 generator, PW 1050/70 goniometer, PW 1710 diffractometer control and PM 8203A register). For the semi-quantitative determination of clay minerals, criteria recommended by Schulz (1964), Thorez (1976) and Brown & Brindley (1980) were followed. Also, transmission electron microscope (TEM) studies of the <2 μm fractions were undertaken using a JEOL-JEM 100 CX II instrument along with chemical analyses of the major elements (MgO, AL_2O_3, SiO_2, CaO, TiO_2, Fe_2O_3, MnO, Na_2O and K_2O), using X-ray fluorescence (XRF) and flame-spectroscopy methods. The XRF analyses were carried out using the Cr Kα radiation and a Philips spectrometer (PW 1410/20 spectrometer, PW 1732/10 generator, PW 1390 channel control, PM 8203 register and TIAP and LiF crystals). A Corning 400 spectrophotometer was used for the flame-spectroscopy analyses. Physical and chemical properties, such as grain size distribution, colour, organic matter content, pH and flocculation-deflocculation state, were also determined.

Figure 2 shows the characterization of the Cretaceous lithostratigraphical units of the Aveiro region according to Rocha (1989, 1993) and Rocha & Gomes (1989, 1990, 1991). The units are dominated by kaolinite and illite, but the proportions of these two clay minerals vary significantly from the Early Cretaceous units to those of the Late Cretaceous.

Kaolinite is very abundant in the lower units (in particular in the 'Grés da Palhaça' and 'Grés de Oiã' Formations in which the clay fraction of some layers may contain 90% kaolinite), whereas illite is predominant in the upper unit ('Argilas de Aveiro' Formation). Late Cretaceous units (in particular the 'Grés de Verba' Formation) may contain up to 30% smectite in some layers. Irregular 10–14 interstratifications (in general, illite-vermiculite and illite-smectite) are ubiquitous but of minor importance. Chlorite is rare in all Cretaceous units. On the other hand, goethite is concentrated in some layers of the 'Grés da Palhaça' and 'Grés de Oiã' Formations.

3. Palaeoenvironmental reconstruction

In the region under study the Early Cretaceous is represented by the 'Grés da Palhaça' Formation (Aptian/Albian-early Cenomanian). This unit is very rich in kaolinite and illite. Towards the top it is characterized by a gradual increase in smectite. Goethite is concentrated at the bottom of the unit.

Early Cretaceous argillaceous sandstones, exposed in large areas of western Europe, are the result of intensive weathering in warm, wet conditions favourable to chemical weathering. Kaolinite is the dominant clay mineral accompanied, as a rule, by illite. This fact indicates that chemical weathering conditioning had not reached the highest possible grade. The deposits show a major continental (fluvial) facies, so-called 'Weald facies' (Millot, 1964; López-Aguayo & Martin-Vivaldi, 1973; Galán *et al.*, 1980; Weaver, 1989; Thiry & Jacquin, 1993). In Spain, Early Cretaceous kaoliniferous deposits correspond essentially (Galán *et al.*, 1980) either to 'Weald facies' (late Valanginian/Hauterivian) or to the 'Utrillas facies' (Albian). In both cases, they reflect the fluvial and lacustrine environments. In some regions, the 'Utrillas' Formation is remarkably similar in lithological and mineralogical terms to the 'Gres da Palhaça' Formation.

The high content of kaolinite in the 'Grés da Palhaça' indicates, in terms of geology and mineralogy, its similarity to the Spanish Early Cretaceous sediments of 'Weald facies' and 'Utrillas facies'. López-Aguayo & Martin-Vivaldi (1973) pointed out the great similarity of both these facies and preferred to call them 'Cretaceous siderolithic facies'. It consists of kaoliniferous sandstones (illite occurs in variable amounts together with kaolinite) which resulted from the dismantling of lateritic kaoliniferous weathering crusts. These crusts were formed during biostatic weathering stages on acidic rocks (granite and gneiss) of the Iberian Massif, under climatic conditions favourable for lateritization. The dismantling of the crusts occurred during resistatic stages and the sediments were transported and deposited in fluvial-deltaic environments under more contrasting seasonal climatic conditions (arid and rainy seasons). The deposits consist of white sandstones, and kaolinitic rich layers alternating with reddish argillaceous layers containing kaolinite + illite + iron oxides/hydroxides. Towards the top of the sedimentary sequence marly, calcareous and carbonaceous layers can occur. Wherever calcareous layers are frequent proximity to the shore is indicated (Galán, 1986).

In the Aveiro region, the 'Grés da Palhaça' Formation exhibits a continental facies of fluvial character, having been deposited in a deltaic plain, initially under very warm and wet conditions (tropical to subtropical climate) favourable for the development of ferralitic processes. Those conditions changed gradually to others characteristic of an environment with more pronounced marine influence, in a more moderate climate, but still with two contrasting seasons.

In fact, the increase in smectite to the top of the sequence indicates the gradual influence of the Cenomanian transgression (Berthou *et al.*, 1982; Weaver, 1989; Chamley, 1989). According to Berthou *et al.* (1982) and Chamley (1989), smectite reveals both climatic changes (dry periods alternating with rainy periods) and morphological changes in the source or feeding areas (lowering of relief followed by less efficient drainage).

The Cenomanian transgression had a very low amplitude in the Aveiro region. Therefore, the inundation of the sea was of short duration (Teixeira & Zbyszewski, 1976; Barbosa, 1981; Lauverjat, 1982; Rocha, 1993). The resulting sedimentary formation, the 'Formação carbonatada', is characterized by a decrease in kaolinite content and an increase in the illite and smectite contents.

Mineralogical changes correspond essentially to the influence of the transgression (Berthou *et al.*, 1982). A rise in the sea level led to an invasion of continental areas and as a result the detrital output was reduced. Soils were formed on the more level surfaces under more maritime and temperate climate. All of these conditions favoured an increase in smectite by comparison with kaolinite. In fact, in Western Europe the sediments deposited during that period are characterized by an increasing content of smectite compared to that of kaolinite (Weaver, 1989).

During the Turonian the sea regressed and there is evidence of a new phase of detrital deposition in the littoral areas. Indeed, during the late Cenomanian the reactivation of late-Hercynian faults produced a tilt towards the north of the northern sector of the Lusitanian Basin and uplift of the region south of the Nazaré fault. These events produced orientation changes in the drainage systems

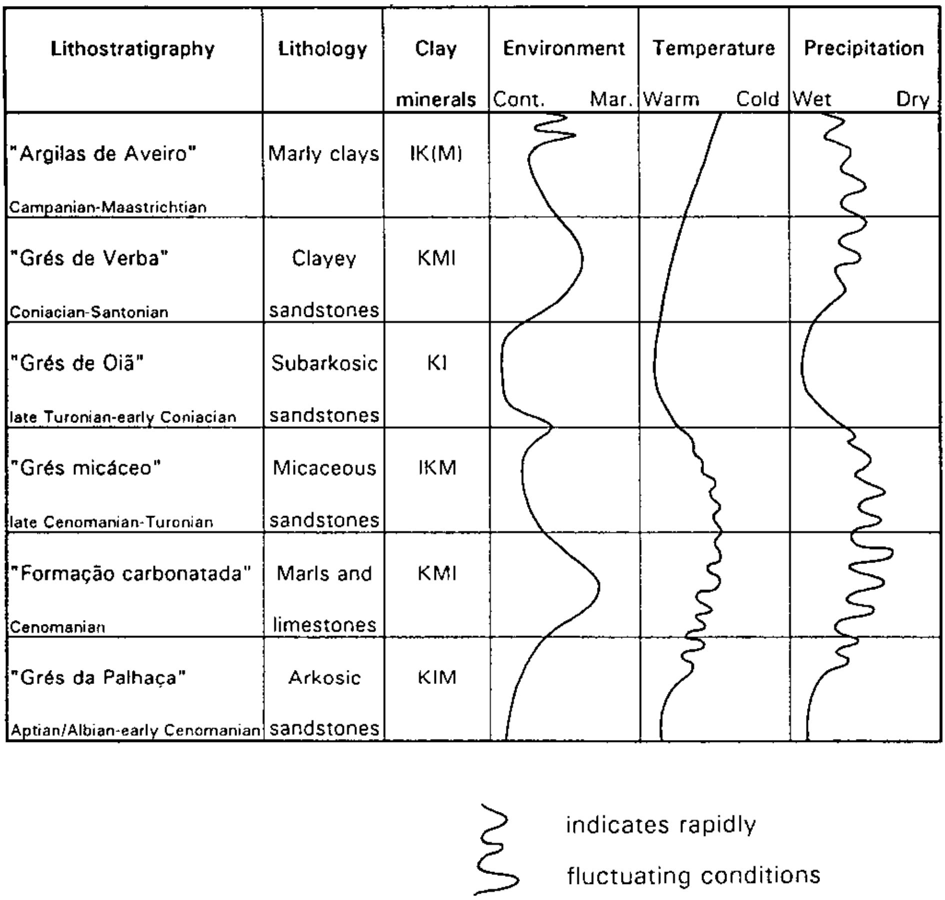

Figure 2. Palaeoenvironmental evolution of the Aveiro region during Cretaceous times.

which flowed towards the NW, as reflected in the transport of detrital materials (Berthou *et al.*, 1982). The prevailing clay mineral association found in the sediments is illite + kaolinite + smectite, and is considered, according to Ortega Huertas *et al.* (1991), to indicate climatic oscillations in the source-areas.

The deposition of the 'Grés micáceo' (late Cenomanian–Turonian) may have taken place in three geographically different palaeoenvironments: 1, a N/NE fluvio-littoral environment: kaolinite is the dominating clay mineral of a kaolinite + illite + Na-smectite assemblage which is typical of a continental influence; 2, a central-W marginal/littoral environment, characterized by an association of illite + kaolinite, which includes an increasing content of smectite towards the W; and 3, a SE environment of lagoonal-lacustrine character, typified by an association of kaolinite + smectite. Climatic conditions during the deposition of the 'Grés micáceo' were similar to those reported for the 'Formação carbonatada'.

The 'Grés de Oiã' unit (late Turonian–early Coniacian), with kaolinite and goethite, represents a continental facies of fluvial character. The clay mineral assemblage is very similar to that observed in the 'Grés da Palhaça'. However, it is more kaolinite-rich. This fact indicates the development of intensive chemical weathering under wet tropical conditions, favourable for highly developed hydrolysis. Sediment deposition occurred in continental environments (Weaver, 1989). Therefore, we can say that the 'Grés de Oiã' Formation was deposited under a very warm and wet subtropical climate which favoured the development of ferralitic alteration. Both at the base and at the top of the unit significant amounts of Na-smectite and calcite are present.

A renewed transgression of the sea took place during the Coniacian (Teixeira & Zbyszewski, 1976; Barbosa, 1981; Lauverjat, 1982; Rocha, 1993), being responsible for the deposition of the 'Grés de Verba' unit in four distinctive geographic palaeoenvironments: 1, a northern area, where kaolinite is the dominant clay mineral of the association kaolinite + illite + smectite: such a clay assemblage characterizes a continental, fluvio-littoral, environment; 2, a central-E area under greater marine influence, of marginal/littoral character, where the sediments are smectite rich; 3, a central-W area, where the deposits have a more distal character, being marine facies and illite-rich; and 4, a meridional area, where sediments with lagoonal character contain the clay mineral assemblage kaolinite + Na-smectite + illite associated with gypsum, anhydrite and pyrite. There is a significant smectite increase in the clay mineral suite. This implies prevailing arid periods alternating with rainy periods and a climate characterized by high temperatures favourable for highly developed hydrolysis.

Coastal regions having poor drainage associated with transgressive conditions produce smectitic soils (Chamley, 1989). A gradual illite increase towards the west reflects the transition to more distal facies (Ortega Huertas *et al.*, 1991).

A regression caused the deposition of the 'Argilas de Aveiro' Formation (Campanian–Maastrichtian). This unit is composed of marly-sandy clays and resulted from intensive and rapid weathering and erosion that affected rejuvenated continental areas in a temperate climate. Sediments were deposited in a flat region with channels of fresh to brackish waters, characterized by low hydrodynamics. The prevailing climate in the depositional areas would have been of subtropical type. Also, a barrier island-tidal flat system would have been developed in the region. For such a depositional environment, three domains have been established: 1, a central-E, supra-tidal area with anoxic episodes, characterized by clayey-sandy sediments containing the association illite + kaolinite, with pyrite in some layers; 2, a central-W area of lagoonal character with marly illite-rich sediments deposited on a tidal flat; and 3, a meridional margin represented by clayey smectite-rich sediments.

The 'Argilas de Aveiro' Formation is characterized by a high illite content showing, as a rule, highly degraded structure and irregular interstratifications. This reflects the intensive and rapid alteration of the rocks of the source-areas, showing a relief rejuvenated because of either sea-level changes or structural reorganization of continental margins (Berthou *et al.*, 1982; Chamley, 1989) under climatic conditions less warm and wet by comparison with those that prevailed during preceding periods. However, such conditions, despite being cooler and drier, did not entirely restrain the hydrolysis responsible for both the formation of kaolinite and the degradation of illite structure. Also, the smectite, which is frequently part of the clay mineral assemblage, may indicate contrasting climatic conditions, as already discussed.

Smectite dominates in the meridional sector of the Aveiro region, associated with kaolinite and illite. In this sector, where illite and smectite show a comparatively better crystallinity, the clay mineral assemblage is significantly associated with zeolites, opal C/CT as well as gypsum and anhydrite. This mineralogy points to diagenetic activity in a more basic and magnesian confined (probably lagoonal) environment with some marine influence (Nemecz, 1981).

4. Conclusions

The clay mineral assemblages present in the six Cretaceous lithostratigraphical units of the Aveiro region have provided very useful information for palaeoenvironmental reconstruction. Figure 2 presents a synthesis of the proposed conditions that prevailed. It is consistent with the interpretation of the general evolution of the basin.

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