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Geochemistry and mineralogy of the Lower Cretaceous of the Lusitanian Basin (western Portugal): Deciphering palaeoclimates from weathering indices and integrated vegetational data

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ABSTRACT

The present study investigates the climatic influence on the geochemistry and mineralogy of the Lower Cretaceous succession of the Ericeira region (Lusitanian Basin, western Portugal). Palaeoclimatic conditions are inferred from a combination of weathering indices and independent palynological and palaeobotanical data. A composite weathering intensity index is computed from selected geochemical and mineralogical data. The concentrations of some elements (Co, Ni, Sc, Th, V, Y, Zn, Zr) indicate variable contributions of sediments from the Lusitanian Basin margin and Iberian hinterland. It is demonstrated that the weathering intensity values are largely influenced by source area geology. The increases in weathering intensity following major unconformities (Late Barremian and Late Aptian) are partially attributed to the arrival of detritus from an evolved regolith sequence in the Iberian hinterland. A combined analysis of independent proxies is found necessary to conveniently discern the roles played by distinct processes on the weathering intensity and investigate the palaeoclimatic conditions.

1. Introduction

After the discovery of rich angiosperm floras in both Lower and Upper Cretaceous strata of the Lusitanian Basin in western Iberia, this region has been considered as a key area for the study of early angiosperm radiation and diversification coupled with environmental conditions (Friis et al., 2010). One of the essential environmental variables is the climate. The isotopic composition of shells and shell fragments are among the most widely used proxies for Cretaceous climate reconstructions. However, in siliciclastic successions depleted of adequate authigenic components, such as most of the interval in the Lusitanian Basin that records the early appearance of angiosperms (Fig. 1), the application of these proxies is not possible. Hence, current understanding of the regional climatic

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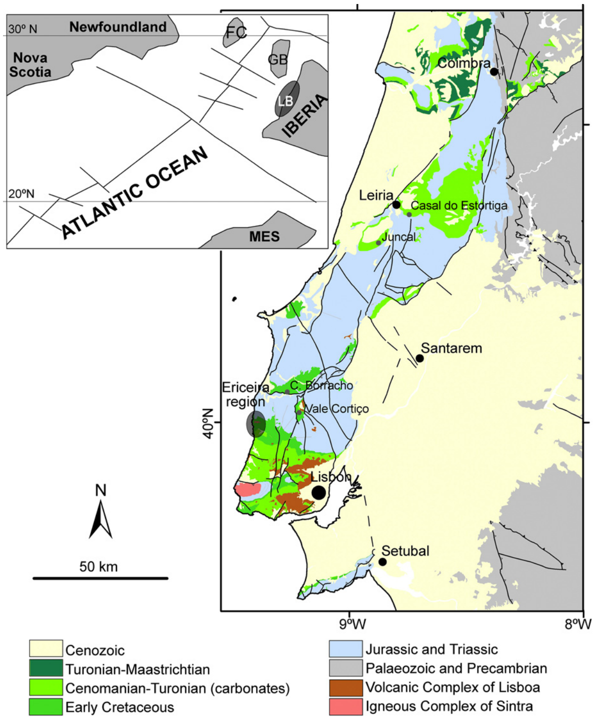

Fig. 1. Geological sketch of the onshore Lusitanian Basin (LNEG, 2010) with the location of the studied sections in the Ericeira region. Inset with location of Lusitanian Basin (LB) and reconstruction of continental areas in the Aptian (after Dercourt et al., 2000, Sahabi et al., 2004 and Sibuet et al., 2012). MES: Morocco Meseta, GB: Galicia Bank; FC: Flemish Cap.

evolution is largely based on recent palynological, palaeobotanical and mineralogical data (Heimhofer et al., 2005, 2008, 2012; Mendes et al., 2010, 2014a, 2014b).

One major difficulty concerning the interpretation of weathering intensity is the discriminations of compositional features that can be attributed to the climatic influence on newly formed sediments from those that should result from the reworking of previous cycle sedimentary units with their own weathering histories (Bauluz et al., 2000; Cox et al., 1995; Gaillardet et al., 1999). Another issue that needs to be solved is the possible occurrence of thick and evolved weathering profiles in flat areas that were relatively protected for long periods from the erosion agents, contrasting with more erosion prone geomorphic settings (Le Pera et al., 2001; Nesbitt et al., 1996) and the effect of progressive erosion of the weathering profile that may result in an inversion of the regolith sequence in the sedimentary succession (Curtis, 1990; Singer, 1980).

In this paper, we document a comprehensive analysis based on multiple geochemical and mineralogical proxies used to establish weathering intensities for the Late Hauterivian–Early Albian of the Lusitanian Basin. It is demonstrated that the abiotic compositional signal should be largely conditioned by factors other than climate conditions. Having in consideration the potential effects of recycling and shifts in provenance, regional climate interpretations are performed with the contribution of independent palynological and palaeobotanical data.

2. Geological framework

During the Early Cretaceous, the sedimentation in the Ericeira area was mixed, with siliciclastics alternating with carbonate deposits (Fig. 2). The Late Hauterivian–Albian interval comprises six formations (Rey, 1992) dated by numerous biostratigraphic indicators (echinoids, rudists, ammonites, foraminifers, algae, calpionelids) and/or by depositional sequence correlations with the southern series of the Cascais area. Various depositional environments alternated in the course of time, including: fluvial systems during the Latest Aptian–Early Albian (Rodízio Formation, exclusively siliciclastic); estuarine systems during the Late Barremian (Regatão Formation, mainly siliciclastic); tidal flats during the Late Hauterivian–Early Barremian (Ribamar Formation with mixed, siliciclastic and dolomitic deposits); inner platform environments with carbonate sedimentation, during the Early Barremian (Ribeira de Ilhas Formation) and the Early Aptian (Crismina Formation); open carbonate platform during the Middle and Late Albian (Galé Formation).

From Valanginian to Albian, the deposits are arranged in three second-order transgressive–regressive sequences (Rey et al., 2003):

- a Valanginian–Early Barremian cycle with maximum transgression during the Early Hauterivian;
- a Late Barremian–basal Late Aptian cycle, showing the maximum transgression during the Early Aptian, contemporary with the Oceanic Anoxic Event (OAE) 1;
- a Latest Aptian–Albian cycle with the transgression peak at the base of the Late Albian (Dinis et al., 2002).

The basal part of each major sequence is characterized by a strong siliciclastic influx (Regatão and Rodízio formations after intra-Barremian and Late Aptian unconformities). These second-order cycles and the associated unconformities are controlled by the geodynamic evolution of the northern Atlantic (Dinis et al., 2008). The intra-Barremian unconformity could be related with the beginning of the onset of seafloor spreading in the Iberia sector (Shillington et al., 2004; Srivastava et al., 2000), whilst the Late Aptian unconformity probably corresponds to the initiation of seafloor spreading in the Galicia sector (Schärer et al., 2000; Srivastava et al., 2000; Tucholke et al., 2007). About 15 third-order depositional sequences are recognized in the studied interval (Rey, 2006). They solely consist of transgressive and highstand systems tracts, with frequent erosional surfaces and palaeosols at the sequences boundaries. Lowstand systems tracts are missing.

3. Sampling and analytical methods

The samples were collected in the Lower Cretaceous coastal outcrops from the Ericeira region (Lusitanian Basin, western Portugal) represented in the composite log of Fig. 2. The heights within the sequence expressed in the text and figures are referred to the Jurassic–Cretaceous boundary. Mudstone beds of variable composition and thickness are found intercalated with other lithologies in both siliciclastic and mixed carbonate–siliciclastic formations.

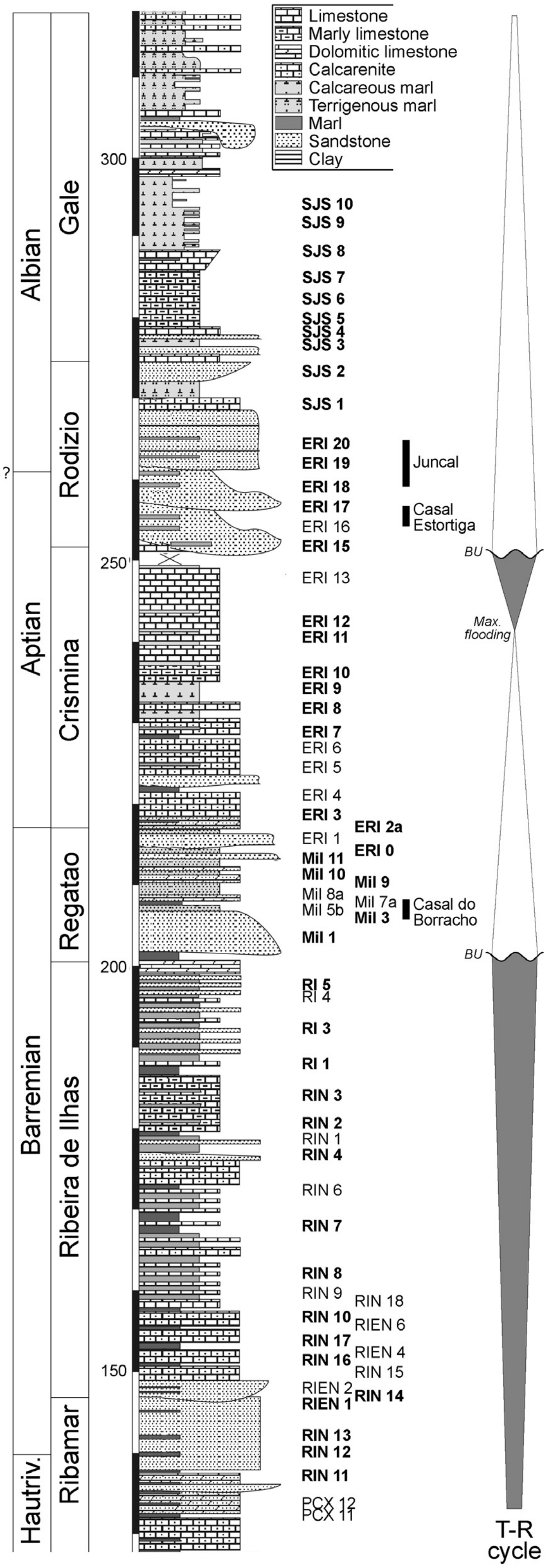

Fig. 2. Studied succession with location of the sampled beds. Bold references indicate samples with geochemical data. Black vertical bars indicate studied deposits with rich palaeobotanical assemblages. Scale heights refer to the Jurassic–Cretaceous boundary. BU: Basin-wide unconformity.

In order to minimize the effects of depositional and post-depositional processes on sediment composition, only fine-grained beds were sampled. Beds with strong cementation were omitted, and a preliminary treatment with 1 N acetic acid was applied to the samples that revealed acid effervescence.

A total of 68 sediment samples were collected from the Ericeira region and analysed for their clay mineralogy (Fig. 2). The sediments were previously washed by centrifugation whenever signs of flocculation were observed. The analyses were performed on oriented slides of the grain-size fraction < 2 μm obtained by centrifugation according to Stokes' law. Clay mineralogy was estimated by X-ray diffraction (XRD) using a Philips PW 3710 equipment, with Cu Kα radiation, belonging to the Earth Sciences Department of the University of Coimbra. Peak areas of basal reflections on glycolated diffractograms, weighted by empirical factors, were used in the approximate mineral semi-quantification (Kahle et al., 2002). Given the uncertainties in the mineralogical semi-quantification by XRD, these results should be taken as approximate estimations of actual mineral proportions (Kahle et al., 2002; Moore and Reynolds, 1997).

The chemical analyses of 48 samples were performed on the fraction finer than 63 μm. This fraction was separated by wet sieving, using deionized water, followed by centrifugation and air-drying. The proportions of major elements (oxides) were determined by X-ray fluorescence after lithium borate fusion, while trace elements were determined by ICP mass spectrometry following a lithium borate fusion and nitric acid digestion. As the analyses were performed on the fraction finer than 63 μm, ICP element underestimation due to incomplete dissolution should be limited (Walsh, 1997). Chemical results from samples with abnormally high CaO were screened out because their composition should be strongly influenced by diagenetic processes. All chemical analyses were done at the laboratories of ACME analytical.

4. Results

4.1. Clay mineralogy

The studied sediments are either dominated by kaolinite or mica-illite and may show secondary amounts of illite-smectite (I/S) mixed-layer clays (Fig. 3). The proportions of these minerals are highly variable within the six sampled formations, although, as a general rule, the abundance of kaolinite tends to be higher in coarser siliciclastic units and slightly lower in the intervened mixed siliciclastic-carbonate units. The Rodízio Formation is distinguished by its constant predominance of kaolinite while the samples from the other formations can reveal both higher abundances of kaolinite and mica-illite. The I/S mixed layers are usually most common in the middle to upper portions of the Ribeira de Ilhas and Crismina formations. The proportion of kaolinite is particularly high in the sediment beds immediately above major unconformities, namely, at the base of the Regatão and Rodízio formations. After a kaolinite maximum is recorded at the base of the Regatão and Rodízio formations, its proportion decreases upwards, defining trends that continue through the Crismina and Galé formations, respectively.

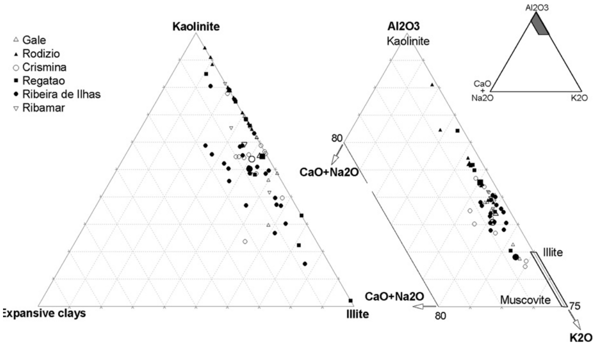

Fig. 3. Cretaceous samples plotted in ternary diagrams for clay mineralogy and A–CN–K (Nesbitt and Young, 1982). Larger symbols indicate the average composition of each formation.

4.2. Chemical composition

The concentrations of major and minor elements are presented in Supplementary file 1. Despite the variability within units, the percentages of Fe_2O_3, MgO, Na_2O and CaO tend to be higher in mud beds from mixed siliciclastic–carbonate units (Crismina, Ribeira de Ilhas and Galé formations) than in siliciclastic units (Ribamar, Regatão and Rodízio formations). TiO_2 reveals an opposite pattern, with slightly higher proportions in clay beds within the siliciclastic units. Siliciclastic units also tend to be enriched in Rare Earth Elements (REE), Y, Th, Zr and Hf, whilst mixed siliciclastic-carbonate units are usually enriched in Rb, Cs, Co, Ni, Cu and Zn (Fig. 4). The concentrations of the remaining elements are more balanced throughout the succession, although anomalously low concentrations of K_2O, Sr and Ba are observed near the base of the Rodízio and Regatão formations. The lower portions of these two formations are also particularly enriched in Y, Zr, REE, and Th and depleted in Ni, Co and Zn.

Chondrite-normalized REE data display moderate fractionation of light REE ($3.04 < La_N/Sm_N < 5.92$) and flatter, although more variable, heavy REE profiles ($0.94 < Gd_N/Yb_N < 2.68$). No clear trends within the stratigraphic succession or differences between formations were observed in light REE and heavy REE fractionations. The Eu anomaly is largely variable ($0.27 < Eu/Eu^* < 0.85$), with no obvious differences between formations (Fig. 4).

4.3. Weathering indices

During weathering, mobile elements tend to be leached, promoting the concentration of immobile elements, and liable minerals are transformed into species adapted to the exogenous condition. Several geochemical (WIP, CIA and CIX indices; Th/U, K_2O/Al_2O_3, K_2O/Th, Rb/K, Rb/Ti and Cs/Ti ratios) and clay mineralogy (kaolinite/illite ratio) proxies are used to establish the intensity of the weathering processes (Table 1). The distinct correlations between weathering indices (Table 2) reflects the different ways the compositional features change during

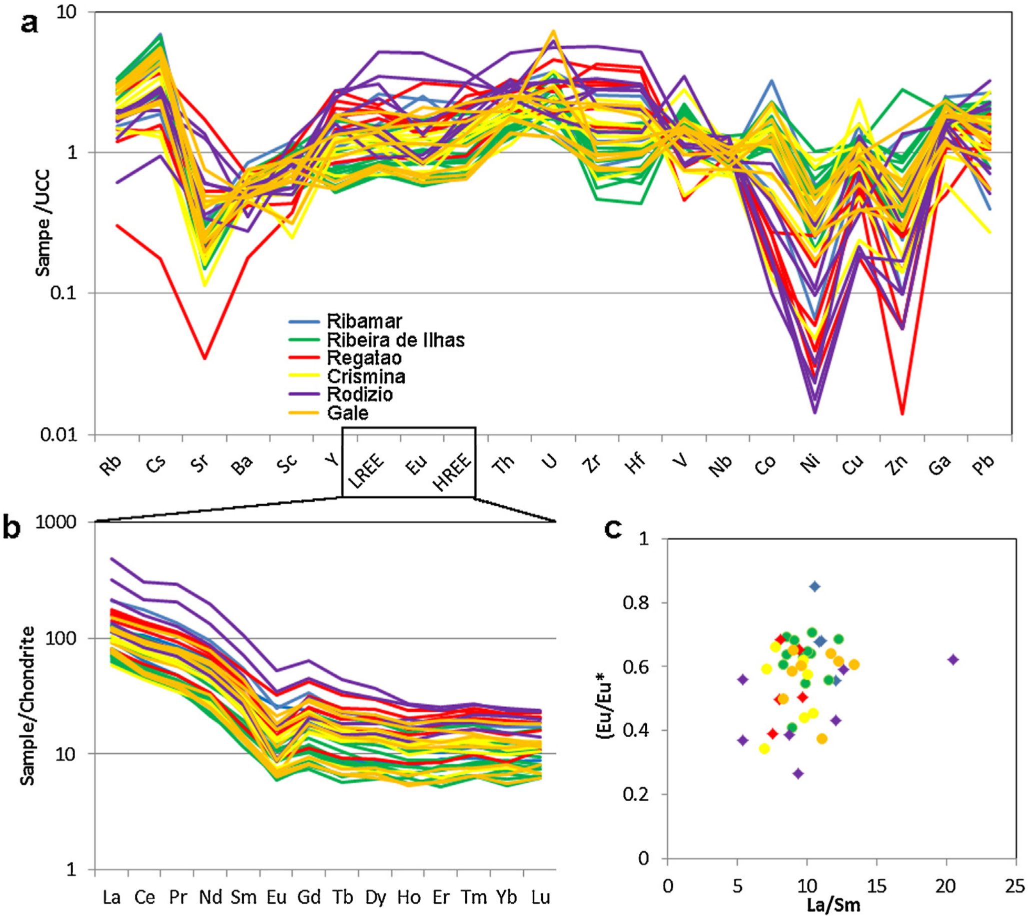

Fig. 4. Geochemical composition of the mud sediments: (a) Composition normalized to the Upper Continental Crust (UCC). Elements are arranged following the periodic table, group by group; (b) REE normalized to chondrite; (c) Light REE fractionation vs. Eu-anomaly normalized to chondrite. Symbols colours distinguish formations as in (a) and (b).

Table 1
Weathering indices considered in this study.

Weathering index	Formula	Weathering relation	Reference
ka/il	Kaolinite/Illite	Increases with weathering	Chamley, 1989; Thiry, 2000; Velde, 1995
CIA (Chemical Index of Alteration)	$Al_2O_3/(Al_2O_3 + K_2O + CaO + Na_2O) \times 100$	Increases with weathering	Nesbitt and Young, 1982
Th/U	Th/U	Increases with weathering (relation with weathering value for Th/U > 4)	McLennan et al., 1995; Gu et al., 2002
K_2O/Al_2O_3	K_2O/Al_2O_3	Decreases with weathering	Gallet et al., 1998; Deconinck et al., 2003
K_2O/Th	K_2O/Th	Decreases with weathering	Gallet et al., 1998; Deconinck et al., 2003
Rb/K	Rb/K	Increases with weathering. Not valid for low weathering	Roy et al., 2008; Buggle et al., 2011
Cs/Ti	Cs/Ti	Decreases with weathering	Yan et al., 2007
Rb/Ti	Rb/Ti	Decreases with weathering	Yan et al., 2007
WIP (Weathering Index)	$(CaO/0.7 + 2Na_2O/0.35 + 2\,K_2O/0.25 + MgO/0.9) \times 100$	Decreases with weathering. Not adequate in highly weathered sediments	Parker, 1970
CIX (modified chemical index of alteration)	$Al_2O_3/(Al_2O_3 + K_2O + Na_2O) \times 100$	Increases with weathering. Adequate for sediments with calcium carbonate	Garzanti et al., 2014

Table 2
Pearson correlation coefficients between geochemical and mineralogical weathering proxies.

	ka/il	CIA	Th/U	K_2O/Al_2O_3	K_2O/Th	Rb/K	Cs/Ti	Rb/Ti	WIP	CIX
ka/il	1.00	**0.58**	0.29	**–0.54**	**–0.52**	0.01	**–0.40**	**–0.48**	**–0.56**	**0.56**
CIA		1.00	0.15	**–0.98**	**–0.46**	0.26	–0.29	**–0.44**	**–0.59**	**0.98**
Th/U			1.00	–0.10	–0.31	0.10	–0.21	–0.26	–0.23	0.13
K_2O/Al_2O_3				1.00	**0.42**	–0.33	0.23	0.38	**0.54**	**–1.00**
K_2O/Th					1.00	0.23	**0.85**	**0.87**	**0.86**	**–0.41**
Rb/K						1.00	**0.53**	**0.46**	0.12	0.32
Cs/Ti							1.00	**0.96**	**0.79**	–0.23
Rb/ti								1.00	**0.83**	–0.38
WIP									1.00	**–0.55**
CIX										1.00

Bold values indicate correlations valid for 99% probability. The key for weathering indices symbols and formulas is given in Table 1.

weathering and suggests that other processes besides chemical alteration conditioned sediment composition. The high correlation K_2O/Al_2O_3–CIA–CIX results from the major influence of K_2O in CIA and CIX indices. Given the similarities between these parameters, they are considered redundant. Only CIX, which does not consider CaO and consequently should be less affected by diagenetic process, will be applied in the analysis of weathering. The evaluation of weathering intensities will also not take in account Cs/Ti, since this parameter strongly correlates with Rb/Ti. A selection of non-redundant weathering indices and their stratigraphic variations is represented in Fig. 5. Because only high Th/U values ($>$ 4 according to McLennan et al., 1995) are thought to be related with weathering intensity and because WIP should not be applied in highly weathered sediments (Price and Velbel, 2003; Shao et al., 2012), these indices were not considered in the subsequent integrated evaluation of weathering intensity.

The highest kaolinite/illite ratios are found at the top of the Ribamar Formation (c. 147 m), at the base of the Regatão Formation (c. 205 m) and in the middle position of the Rodízio Formation (c. 259 m). This ratio is highly variable in the Regatão Formation and generally moderate to low in the Ribeira de Ilhas, Crismina and Galé formations. Maximums of CIX index are detected near the bottom of the Rodízio Formation (c. 256 m). A secondary peak of CIX occurs at the base-middle part of the Regatão Formation (c. 205–210 m). The K_2O/Th and Rb/Ti ratios reveal similar oscillation patterns, characterized by high values in Ribeira de Ilhas and Galé formations, whose maximums occur in the middle–upper portions of these units, and lower values in middle part of the Regatão and Rodízio formations. Moderate values are observed in the Crismina and Ribamar formations. The Rb/K ratio displays an independent pattern of variation that seems to be independent of the sampled lithology.

A selection of non-redundant weathering parameters that should not be influenced by the carbonate content (CIX, Rb/Ti, Rb/K, K_2O/Th and kaolinite/illite) were combined to establish the weathering trends. The indices

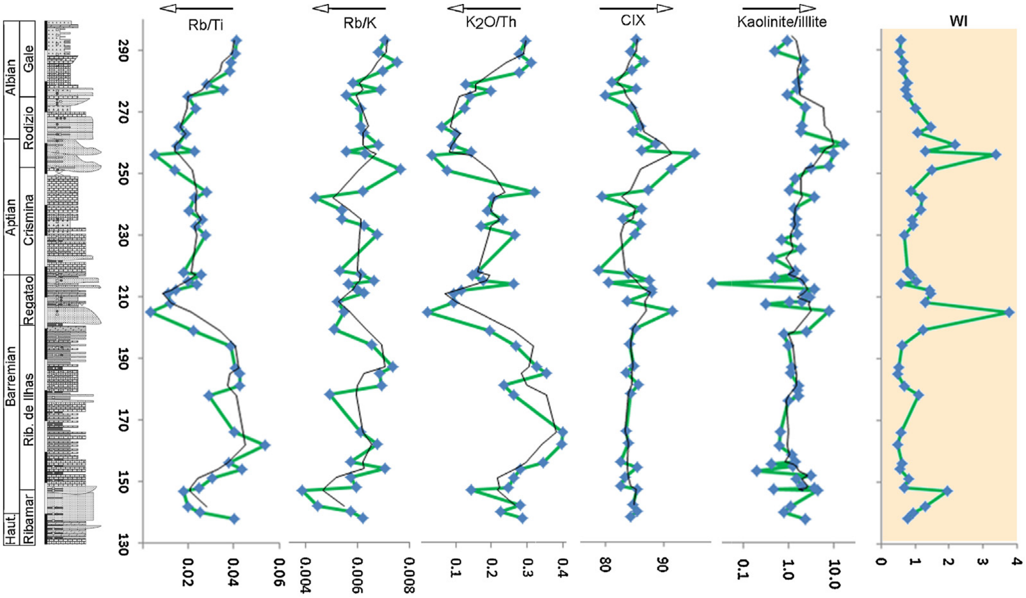

Fig. 5. Plots of the weathering intensity indicators. Arrows indicate the expected increase in weathering intensity. WI is the combined weathering index (see text for explanation). Black lines for the three and four points moving average applied to the geochemical and clay mineral data, respectively.

for which lower values reflect higher weathering intensities (Rb/Ti and K_2O/Th) were previously inverted. The weathering intensity value is then established by the summed results of the selected parameters after normalization by scaling between 0 and 1 as:

$$X_N = (X_i - X_{min})/(X_{max} - X_{min})$$

in which X_i is the value of the index at a certain position, X_{min} and X_{max} are the minimum and maximum results, respectively, for a given index and X_N is the resultant 0–1 normalized value.

The obtained pattern of variations of the weathering intensity value (Fig. 5) suggests that:

- the highest intensities occur in the lower portion of Regatão (Late Barremian) and Rodízio (Late Aptian–Early Albian) formations after abrupt rises and followed by progressive, although discontinuous, decreases;
- a secondary peak is found at the top of the Ribamar Formation (Late Hauterivian);
- wider intervals with low-moderate weathering intensity values are separated by short intervals of intense weathering;
- minor weathering intensity excursions occur within the more carbonate enriched Ribeira de Ilhas (Early Barremian) and Crismina (Early to Middle Aptian) formations.

5. Discussion

5.1. Source area geology and weathering intensity

The pattern of variation of the weathering intensity value is broadly paralleled by the transgressive–regressive cycles. The maximum flooding zones approximately correspond to the lowest weathering intensity, while the peaks tend to coincide with the beginning of transgressive cycles after major unconformities. The two basin-wide unconformities covered by this research, the intra-Barremian and Late-Aptian ones, coincide with important tectonic events associated with the Atlantic opening. Regardless of the specific genetic processes, these ruptures are coeval of major fault deformation within the Iberian sub-plate and rejuvenation of the watersheds (Dinis et al., 2008). Previous works on the Cretaceous detrital zircon age signatures point to sudden shifts in provenance with correspondence with major conformities (Dinis et al., in press). The weathering intensity values may be an inherited feature due to changes in provenance and reflecting the weathering intensity in source areas. The extensive Cainozoic cover hampers a clear knowledge of the Lusitanian Basin basement rocks in the latitude of the Ericeira region (Fig. 1). Diverse metasedimentary units (mainly metapelitic) and igneous units (including mafic lithologies) are observed in the basin edge to the north and the south of Ericeira (LNEG, 2010). Rift-related igneous rocks with tholeiitic affinity crop out in southern Portugal (Martins et al., 2008; Verati et al., 2007), but their occurrence in northern locations cannot be excluded. Inland occurs mainly Variscan and post-Variscan granitoids intruding metasedimentary units.

Immobile elements are excellent indicators of provenance. Regardless of weathering intensity, REE, Th, and Zr are abundant in granitoid-parented sediments contrasting with Co, Cr, Ni and Sc that are usually concentrated in basic rocks and their sediment products (e.g., Condie, 1993; McLennan et al., 1990, 1993). The enrichment in Y, REE, Zr and Hf coupled with a decrease in Co, Ni and Zn in the units that cover the Barremian and Aptian unconformities indicate an increased contribution of felsic sediments (Fig. 4). An oscillating contribution of mafic- and felsic-parented sediment can be evaluated with the pattern of variation of selected immobile element ratios (Fig. 6). The Barremian unconformity has correspondence with a major shift in provenance marked by an increase in the proportion of sediment associated with Variscan and post-Variscan felsic igneous rocks from the Iberia hinterland (Dinis et al., in press). It may be assumed that in Iberia inland, the regolith sequences reached more evolved stages while the catchment areas were restricted to the Iberia Atlantic margin, the increase in weathering intensity values being associated with the arrival of sediment derived from these weathering profiles. The subsequent decrease in weathering intensity is paralleled by a provenance signal marked by lower amounts of granitoid-related sediments. Sea-level rise should reduce erosion capacity and promote accommodation space for alluvial storage in lower valley realms (Holbrook et al., 2006). Consequently, an important proportion of hinterland sediment should be trapped in fluvial valleys and not reach the coastal or marine environments that developed during high-sea-level periods. In contrast, the uplifted Atlantic margin shoulders kept delivering sediment during the transgressive and highstand periods, increasing the relative proportion of mafic and probably less weathered component.

A similar pattern of shift in provenance and consequent changes in weathering intensity values may be proposed for the sequence that covers the Aptian unconformity (Fig. 6). However, the progressive decrease in weathering intensity values following a maximum at the bottom of the Rodízio Formation when most immobile element ratios point to unaltered sediment provenance is attributed to changes in climatic conditions. Progressive starvation of the highly weathered upper parts of the regolith sequences may also contribute to the decrease of the weathering intensity values.

5.2. Depositional and post-depositional effects

Several authors defended that clay mineral assemblages are affected by selective settling (e.g., Chamley, 1989; Thiry and Jacquin, 1993). This process may promote kaolinite enrichment in coarser intervals. Thiry and Jacquin (1993) also showed specific clay assemblages in distinct Cretaceous depositional units associated with the Atlantic Ocean opening. Post-depositional transformation could also account for some compositional features. Kaolinite is frequently an early diagenetic mineral in sandstone deposits (Rossi et al., 2002; Thiry, 2000). Although only fine-grained beds were collected, it is possible that post-depositional transformations promoted

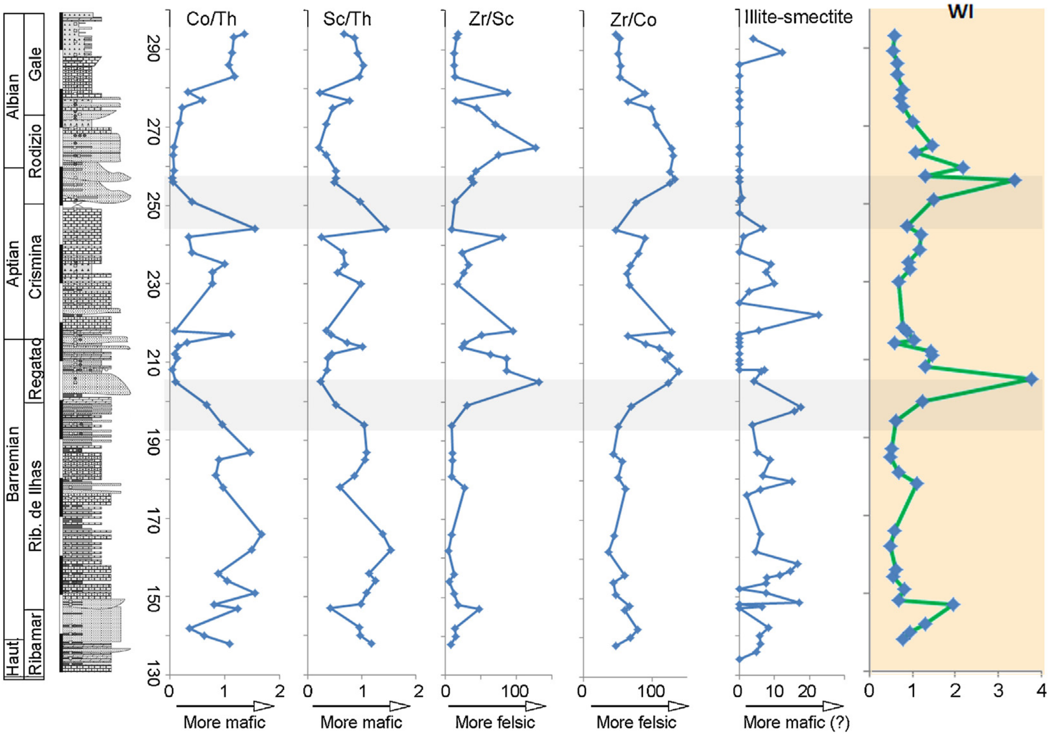

Fig. 6. Plots of the element ratios that should reflect variable contributions of felsic and mafic parented sediment and I/S mixed clays. The light shadows indicate intervals of transition to more felsic supply. Although the formation of smectite is frequently promoted by mafic bed rock (Velde, 1995), the pattern of variation of illite-smectite is distinguished from the geochemical provenance indicators, suggesting other controlling factors.

kaolinite enrichment. Hundert et al. (2006) presented a genetic model for the development of kaolinite in sediment successions below intraformational unconformities that considers water recharge where sandy sediment beds are exposed and subsequent early diagenetic kaolinite formation in these units and evolving mudstones. However, due to the lack of independent evidence of depositional and post-depositional processes, their possible effects on sediment composition will not be further discussed.

5.3. *Evolution in climate*

5.3.1. *Before the Barremian unconformity*

Having in consideration the previous remarks on the possible influence of provenance on sediment features, the compositional changes found in the succession that precedes the Barremian unconformity (Ribamar and Ribeira de Ilhas formations; Late Hauterivian to Late Barremian) are probably linked to shifts in climate. In the Vale Cortiço opencast clay pit complex (near Torres Vedras locality; Figs. 1 and 2), no angiosperms were observed in the fossil record. The spore–pollen assemblages collected from the Santa Susana Formation, which precedes the studied succession, are clearly dominated by conifers and ferns. Among conifers, *Classopollis* (Cheirolepidiaceae) is dominant and with the araucarian *Araucariacites australis* Cookson are indicative of semi-arid and arid low-lying water-margin environments under a subtropical to tropical climate (e.g., Traverse, 2007; Watson, 1988). Remains of Cheirolepidiaceae *Frenelopsis teixeirae* (Alvin & Pais) constitute the major part of the fossil flora from Vale Cortiço (Mendes et al., 2010). The Cheirolepidiaceae conifers coped well with the semiarid and even arid conditions and are normally interpreted as xeromorphic growing in near-coastal environments (Mendes et al., 2010; Vajda, 2001). These proxies point to the prevalence of a relatively warm and dry climate with a seasonally rainy period during the Early Hauterivian. The mineralogical and geochemical data suggest a subsequent period of progressive increase in humidity (Ribamar Formation) and an Early Barremian shift to drier and conditions (transition Ribamar–Ribeira de Ilhas Formation). The occasional frequency peaks of I/S mixed clays are compatible with the development of a climate with rainfall seasonality during the Early Barremian. A similar Hauterivian–Barremian pattern based on geochemical and clay mineralogical data was proposed for other places of northeastern (Ruffell et al., 2002) and northwestern (Zhang et al., 2014) Atlantic margins.

5.3.2. *Between the Barremian and Aptian unconformities*

The Barremian is commonly considered a dry period in western Europe (Ruffell and Batten, 1990), in general with relatively low temperatures at the Barremian–Aptian transition (Pucéat et al., 2003; Steuber et al., 2005). Although the pronounced swing in the weathering intensity value at the base of Regatão Formation (Late Barremian) is probably linked to a shift in sediment provenance, as discussed before, the deformation associated with the opening of the Atlantic Ocean may have an indirect and secondary effect on climate. As the continental break-up and seafloor spreading progressed northwards (Alves et al., 2009; Pinheiro et al., 1996), significant deformation and uplifting along the Atlantic margin shoulders is expected (Dinis et al., 2008). We assume that the Barremian unconformity marks an important moment of reorganization of the Iberian microplate, responsible for the uplifting of the basin edge that may have promoted orographic rainfall. A geomorphic context associated with the deposition of coarse-grained prograding units after the Barremian unconformity could result from this conjugation of wetter climate, due to an increase in rainfall, and steeper slopes. Late Barremian units of Casal do Borracho (coeval of Regatão Formation; Figs. 1 and 2) yield different types of angiosperm pollen (including *Mayoa portugallica* Friis, Pedersen & Crane), although subordinated in the palynoflora. Representatives of aquatic ferns were found, including *Crybelosporites pannuceus* (Brenner) Srivastava and the freshwater algae, *Ovoidites sprigii* (Cookson & Dettmann) Zippi and *Ovoidites parvus* (Cookson & Dettman) Nakoman (Mendes, work in progress). The occurrence of aquatic ferns and freshwater algae may also reflect the presence of moist environments and the influence of wet climates.

5.3.3. *After the Aptian unconformity*

Several climatic proxies suggest that the Early and Middle Aptian was a period of relatively low temperatures (McAnena et al., 2013; Pucéat et al., 2003). The temperature increased during the Late Aptian and onwards until reaching the extreme greenhouse conditions that already characterized the Upper Cenomanian (Huber et al., 2011; Pucéat et al., 2003). In several researches, it is considered that the climate of western Iberia was conditioned by the migration of the boundary between a mid-latitude warm-humid belt, to the north, and a hot-dry belt, to the south (Heimhofer et al., 2008, 2012; Mendes et al. 2014a; Trabucho Alexandre et al., 2011). These climatic belts correspond respectively to the southern Laurasian floral province with a temperate to subtropical humid vegetation and the northern Gondwana floral province with an arid to semi-arid savannah-type vegetation (Batten, 1984; Brenner, 1976; Chumakov et al., 1995; Peralta-Medina and Falcon-Lang, 2012; Philippe et al., 2004).

A study of silicified trunks ascribed to the genus *Protocupressinoxylon* Eckhold collected at the base of the Figueira da Foz Formation (coeval to Rodízio Formation) in Casal do Estortiga (Figs. 1 and 2) suggests hot summers and mild-to-warm winters (Mendes et al., 2014a). The palynological assemblage found in the upper part of the Figueira da Foz Formation at the Juncal locality (Figs. 1 and 2) is dominated by conifer pollen and fern spores. These include members of the extinct Cheirolepidiaceae represented by numerous pollen grains of *Classopollis* and conifer pollen ascribed to Araucariaceae and Pinaceae. Extinct Cheirolepidiaceae are characterized by strongly reduced leaves, thick cuticle and deeply sunken stomata that indicate water stress conditions (Watson and Alvin, 1996). Another important component in the fern assemblage is the *Deltoidospora*-type spores well known from the sporangia of the extinct dicksoniaceous fern *Onychiopsis psilotoides*. Angiosperms are represented by *Clavatipollenites*-type pollen, *Retimonocolpites* spp., *Rousea* sp., and *Senectotetradites* sp. The overall composition of this flora suggests a seasonally warm dry climate. Based on the trend for decrease in weathering intensity and the interpreted palaeovegetation from Casal do Estortiga and Juncal, it may be assumed that the region was under the influence of the mid-latitude warm-humid belt during the Late Aptian and the Earliest Albian. A subsequent transition to dryer and hotter conditions during the Albian associated with the northward migration of the tropical dry belt was proposed by Heimhofer et al. (2012).

6. Conclusions

A combined index based on independent geochemical and mineralogical data applied to the Early Cretaceous sediment succession from western Portugal suggests two major excursions in weathering intensity, both tied to recognized basin-wide unconformities: Late Barremian and Late Aptian ones. These excursions are partially attributed to changes in provenance, which are demonstrated by the evidences of variable contributions of mafic- and felsic-parented sediment. This finding illustrates how weathering intensity may be affected by source area geology and consequently the interpretation of palaeoclimates from weathering indicators is difficult to perform. We also suggest that the tectonic reorganization of the Iberian microplate associated with basin-wide unconformities may have had an indirect effect on the regional climate. Reliefs along the Atlantic margin generated during periods of tectonic activity may have promoted wetter environmental conditions due to orographic rainfall.

Palaeovegetation data strongly suggest warm and dry palaeoclimatic conditions during the Early Hauterivian. The progressive increase in the weathering intensity values during the Late Hauterivian followed by a sharp decrease during the Hauterivian–Barremian is attributed to climate changes. Wetter climates, having correspondence with the earliest well-constrained occurrences of angiosperms in the Lusitanian Basin West Iberia, occurred during the Late Barremian. The available palaeobotanical data and the progressive decrease of the weathering intensity values after the Aptian maxima point to the transition toward dryer and hotter conditions associated with the northward migration of the tropical dry belt.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.crte.2015.09.003.

References

Alves, T.M., Moita C., Cunha T., Ullnaess M., Myklebust R., Monteiro J.H., Manuppella G. 2009. Diachronous evolution of Late Jurassic-Cretaceous continental rifting in the northeast Atlantic (West Iberian margin). Tectonics. 28, TC4003. doi:10.1029/2008TC002337.

Batten, D.J., 1984. Palynology, climate and the development of Late Cretaceous floral provinces in the Northern Hemisphere: a review. In: Brenchley, P. (Ed.), Fossils and Climate. John Wiley & Sons, pp. 127–164.

Bauluz, B., Mayayo, M.J., Fernandez-Nieto, C., Lopez, J.M.G., 2000. Geochmistry of Precambrian and Paleozoic siliciclastic rocks from the Iberian Range (NE Spain): implications for source-area weathering, sorting, provenance, and tectonic setting. Chem. Geol. 168, 135–150.

Brenner, G., 1976. Middle Cretaceous floral provinces and early migration of angiosperms. In: Beck, C.B. (Ed.), Origin and Early Evolution of Angiosperms. Columbia University Press, pp. 23–44.

Buggle, B., Glaser, B., Hambach, U., Gerasimenko, N., Markovic, S., 2011. An evaluation of geochemical weathering indices in loess-paleosol studies. Quat. Internat. 240, 11–21.

Chamley, H., 1989. Clay Mineralogy. Springer, 623 p.

Chumakov, N.M., Zharkov, M.A., Herman, A.B., Doludenko, M.P., Kalandadze, N.M., Lebedev, E.L., Ponomarenko, O.A.G., Rautian, A.S., 1995. Climatic belts of the mid-Cretaceous time. Strat. Geol. Correl. 3, 241–260.

Condie, K.C., 1993. Chemical composition and evolution of the upper continental crust: contrasting results from surface samples and shales. Chem. Geol. 104, 1–37.

Cox, R., Low, D.R., Cullers, R.L., 1995. The influence of sediment recycling and basement composition on evolution of mudrock chemistry in the southwestern United States. Geochim. Cosmochim. Acta 59, 2919–2940.

Curtis, C.D., 1990. Aspects of climatic influence on the clay mineralogy and geochemistry of soils, palaeosols and clastic sedimentary rocks. J. Geol. Soc. 147, 351–357.

Deconinck, J.F., Hesselbo, S.P., Debuisser, N., Averbuch, O., Baudin, F., Bessa, J., 2003. Environmental controls on clay mineralogy of an Early Jurassic mudrock (Blue Lias Formation, southern England). Int. J. Earth Sci. 92, 255–266.

Dercourt, J., Gaetani, M., Vrielynck, B., Barrier, E., Biju-Duval, B., Brunet, M.F., Cadet, J.P., Crasquin, S., Sandulescu, M., 2000. Atlas Peri-Tethys Paleoegeographical Maps. CCGM/CGMW, Paris.

Dinis, J.L., Rey, J., Graciansky, P.C., 2002. Le Bassin lusitanien (Portugal) à l'Aptien supérieur–Albien : organisation séquentielle, proposition de corrélations, évolution. C. R. Geoscience. 334, 757–764.

Dinis, J.L., Rey, J., Cunha, P.P., Callapez, P., Reis, R.P., 2008. Stratigraphy and allogenic controls of the western Portugal Cretaceous: an updated synthesis. Cretaceous Res. 29, 772–780.

Dinis, P.A., Dinis, J.L., Tassinari, C., Carter, A., Callapez, P., Morais, M. (*in press*). Detrital zircon geochronology of the Cretaceous succession from the Iberian Atlantic Margin: palaeogeographic implications. Int. J. Earth Sc doi:10.1007/s00531-015-1221-z.

Friis, E.M., Pedersen, K.R., Crane, P.R., 2010. Cretaceous diversification of angiosperms in the western part of the Iberian Peninsula. Rev. Palaeobot. Palynol. 162, 341–361.

Gaillardet, J., Dupré, B., Allègre, C.J., 1999. Geochemistry of large river suspended sediments: Silicate chemical weathering or recycling tracer? Geochim. Cosmochim. Acta 63, 4037–4052.

Gallet, S., Jahn, B.-M., Van Vliet Lanoe, B., Dia, A., Rossello, E., 1998. Loess geochemistry and its implications for particle origin and composition of the upper continental crust. Earth Planet. Sci. Lett. 156, 157–172.

Garzanti, E., Padoan, M., Setti, M., López-Galindo, A., Villa, I.M., 2014. Provenance versus weathering control on the composition of tropical river mud (southern Africa). Chem. Geol. 366, 61–74.

Gu, X.X., Liu, J.M., Zheng, M.H., Tang, J.X., Qi, L., 2002. Provenance and tectonic setting of the Proterozoic turbidites in Hunan, South China: geochemical evidence. J. Sedim. Res. 72, 393–407.

Heimhofer, U., Adatte, T., Hochuli, P.A., Burla, S., Weissert, H., 2008. Coastal sediments from the Algarve: low-latitude climate archive for the Aptian-Albian. Int. J. Earth Sc. 97, 785–797.

Heimhofer, U., Hochuli, P.A., Burla, S., Dinis, J.M.L., Weissert, H., 2005. Timing of Early Cretaceous angiosperm diversification and possible links to major paleoenvironmental change. Geology 33, 141–144.

Heimhofer, U., Hochuli, P.A., Burla, S., Oberli, F., Adatte, T., Dinis, J.L., Weissert, H., 2012. Climate and vegetation history of western Portugal inferred from Albiannear-shore deposits (Galé Formation, Lusitanian Basin). Geol. Mag. 149, 1046–1064.

Holbrook, J., Scott, R.W., Oboh-Ikuenobe, F.E., 2006. Base-level buffers and buttresses: a model for upstream versus downstream control on fluvial geometry and architecture within sequences. J. Sedim. Res. 76, 162–174.

Huber, B.T., MacLeod, K.G., Gröcke, D.R., Kucera, M., 2011. Paleotemperature and paleosalinity inferences and chemostratigraphy across the Aptian/Albian boundary in the subtropical North Atlantic. Paleoceanography 26, 1–20.

Hundert, T., Piper, D.J.W., Pe-Piper, G., 2006. Genetic model and exploration guidelines for kaolin beneath unconformities in the Lower Cretaceous fluvial Chaswood Formation, Nova Scotia. Expl. Min. Geol. 15, 9–26.

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.

Le Pera, E., Arribas, J., Critelli, S., Tortosa, A., 2001. The effects of source rocks and chemical weathering on the petrogenesis of siliciclastic sand from the Neto River (Calabria, Italy): implications for provenance studies. Sedimentology 48, 357–378.

LNEG, 2010. Carta Geológica de Portugal 1:1,000,000. Laboratório Nacional de Energia e Geologia, Lisboa.

Martins, L.T., Madeira, J., Youbi, N., Munhá, J., Mata, J., Kerrich, R., 2008. Rift-related magmatism of the Central Atlantic magmatic province in Algarve, southern Portugal. Lithos 101, 102–124.

McAnena, A., Flögel, T., Hofmann, P., Herrle, J.O., Griesand, A., Pross, J., Talbot, H.M., Rethemeyer, J., Wallmann, K., Wagner, T., 2013. Atlantic cooling associated with a marine biotic crisis during the mid-Cretaceous period. Nat. Geosc. 6, 558–561.

McLennan, S.M., Hemming, S.R., McDaniel, D.K., Hanson, G.N., 1993. Geochemical approaches to sedimentation, provenance and tectonics. In: Johnsson, M. J., Basu, A. R. (Eds.), Processes controlling the composition of clastic sediments. Geol. Soc. Amer. 21–40.

McLennan, S.M., Hemming, S.R., Taylor, S.R., Eriksson, K.A., 1995. Early Proterozoic crustal evolution: Geochemical and Nd–Pb isotopic evidence from metasedimentary rocks, southwestern North America. Geochim. Cosmochim. Acta 59, 1153–1177.

McLennan, S.M., Taylor, S.R., Mcculloch, M.T., Maynard, J.B., 1990. Geochemical and Nd–Sr isotopic composition of deep-sea turbidites: Crustal evolution and plate tectonic associations. Geochim. Cosmochim. Acta 54, 2015–2050.

Mendes, M.M., Dinis, J., Pais, J., Friis, E.M., 2014a. Vegetational composition of the Early Cretaceous Chicalhão flora (Lusitanian Basin, western Portugal) based on palynological and mesofossil assemblages. Rev. Palaeobot. Palynol. 200, 65–81.

Mendes, M.M., Dinis, J.L., Esteban, L.G., de Palacios, P., Fernández, F.G., Pais, J., 2014b. Early Cretaceous woods of Figueira da Foz Formation in western Portugal: Palaeoenviromental, palaeoclimatic and palaeobiogeographic insights. Cretaceous Res 51, 112–120.

Mendes, M.M., Dinis, J.L., Gomez, B., Pais, J., 2010. Reassessment of the cheirolepidiaceous conifer *Frenelopsis teixeirae* Alvin et Pais from the Early Cretaceous (Hauterivian) of Portugal and palaeoenvironmental considerations. Rev. Palaeobot. Palynol. 161, 30–42.

Moore, D., Reynolds, R., 1997. X-Ray-Diffraction and the Identification and Analysis of Clay Minerals. Univ. Press, Oxford, 378 pp.

Nesbitt, H.W., Young, G.M., McLennan, S.M., Keays, R.R., 1996. Effects of chemical weathering and sorting on the petrogenesis of siliciclastic sediments, with implications for provenance studies. The J. Geol. 104, 525–542.

Nesbitt, H.W., Young, G.M., 1982. Early Proterozoic climates and plate motions inferred from major element chemistry of lutites. Nature 299, 715–717.

Parker, A., 1970. An index of weathering for silicate rocks. Geol. Mag. 107, 501–504.

Peralta-Medina, E., Falcon-Lang, H.J., 2012. Cretaceous forest composition and productivity inferred from a global fossil wood database. Geology 40 (3), 219–222.

Philippe, M., Bamford, M., McLoughlin, S., Alves, L.S.R., Falcon-Lang, H., Gnaedinger, S., Ottone, E.G., Pole, M., Rajanikanth, A., Shoemaker, R.E., Torres, T., Zamuner, A., 2004. Biogeographic analysis of Jurassic–Early Cretaceous wood assemblages from Gondwana. Rev. Palaeobot. Palynol. 129 (3), 141–173.

Pinheiro, L., Wilson, R., Reis, R.P., Whitmarsh, R., Ribeiro, A., 1996. The western Iberia margin: a geophysical and geological overview. Proc. O. D. P. Sci. Res. 149, 3–23.

Price, J.R., Velbel, M.A., 2003. Chemical weathering indices applied to weathering profiles developed on heterogeneous felsic metamorphic parent rocks. Chem. Geol. 202, 397–416.

Pucéat, E., Lécuyer, C., Sheppard, S.M.F., Dromart, G., Reboulet, S., Grandjean, P., 2003. Thermal evolution of Cretaceous Tethyan marine waters inferred from oxygen isotope composition of fish tooth enamels. Paleoceanogrphy 18, 1–12.

Rey, J., 1992. Les unités lithostratigraphiques du Crétacé inférieur de la région de Lisbonne. Com. Serv. Geol. Portugal 78, 103–124.

Rey, J., 2006. Stratigraphie séquentielle et séquences de dépôt dans le Crétacé inférieur du Bassin lusitanien. Ciênc. Terra, v. esp. 6, 1–120.

Rey, J., Graciansky, P.C., Jacquin, T., 2003. Les Sequences de dépôt dans le Crétacé inférieur du Bassin Lusitanien. Com. Inst. Geol. Min. 90, 15–42.

Rossi, C., Kälin, O., Arribas, J., Tortosa, A., 2002. Diagenesis, provenance, and reservoir quality of Triassic TAGI sandstones from Ourhoud field, Berkine (Ghadames) Basin, Algeria. Mar. Petrol. Geol. 19, 117–142.

Roy, P.D., Caballero, M., Lozano, R., Smykatz-Kloss, W., 2008. Geochemistry of late Quaternary sediments from Tecocomulco lake, central Mexico: Implication to chemical weathering and provenance. Chem. Erde Geochem. 68, 383–393.

Ruffell, A.H., Batten, D.J., 1990. The Barremian–Aptian arid phase in western Europe. Palaeogeogr. Palaeoclimatol. Palaeoecol. 80, 197–212.

Ruffell, A., McKinley, J.M., Worden, R.H., 2002. Comparison of clay mineral stratigraphy to other proxy palaeoclimate indicators in the Mesozoic of NW Europe. Phil. Trans. Royal Soc. Lond. Ser. A 360, 675–693.

Sahabi, M., Aslanian, D., Olivet, J.-L., 2004. Un nouveau point de départ pour l'histoire de l'Atlantique central. C. R. Geoscience. 336, 1041–1052.

Schärer, U., Girardeau, J., Cornen, G., Boillot, G., 2000. 138-121 Ma asthenospheric magmatism prior to continental break-up in the North Atlantic and geodynamic implications. Earth Planet. Sci. Lett. 81, 555–572.

Shao, J., Yang, Y., Li, C., 2012. Chemical indices (CIA and WIP) as proxies for integrated chemical weathering in China: Inferences from analysis of fluvial sediments. Sedim. Geol. 265–266 [110–120].

Shillington, D.J., Holbrook, W.S., Tucholke, B.E., Hopper, J.R., Louden, K.E., Larsen, H.C., Van Avendonk, H.J.A., Deemer, S., Hall, J., 2004. Data report. Marine geophysical data on the Newfoundland nonvolcanic rifted margin around SCREEECH transect 2. In: Tucholke, B.E., Sibuet, J.C., Klaus, A. (Eds.). Proc. ODP, Init. Rep. 210, 1–36.

Sibuet, J.-C., Rouzo, S., Srivastava, S., 2012. Plate tectonic reconstructions and paleogeographic maps of the central and North Atlantic oceans. R. Canad. Sc. Terre 49, 1395–1415.

Singer, A., 1980. The paleoclimatic interpretation of clay minerals in soils and weathering profiles. Earth Sci. Rev. 15, 303–326.

Srivastava, S.P., Sibuet, J.-C., Cande, S., Roest, W.R., Reid, I.D., 2000. Magnetic evidence for slow seafloor spreading during the formation of the Newfoundland and Iberian margins. Earth Planet. Sci. Lett. 182, 61–76.

Steuber, T., Rauch, M., Masse, J.-P., Graaf, J., Malko, C.M., 2005. Low-latitude seasonality of Cretaceous temperatures in warm and cold episodes. Nature 437, 1341–1344.

Thiry, M., 2000. Palaeoclimatic interpretation of clay minerals in marine deposits: an outlook from the continental origin. Earth Sci. Rev. 49, 201–221.

Thiry, M., Jacquin, T., 1993. Clay minerals distribution related to rift activity, sea level changes and paleoceanography in the Cretaceous of the Atlantic Ocean. Clay Minerals 28, 61–84.

Trabucho Alexandre, J., Van Gilst, R.I., Rodríguez-López, J.P., Boer, P.L., 2011. The sedimentary expression of oceanic anoxic event 1b in the North Atlantic. Sedimentology 58, 1217–1246.

Traverse, A., 2007. Paleopalynology. Topics in Geobiology, 28. Springer.

Tucholke, B.E., Sawyer, D.S., Sibuet, J.-C., 2007. Breakup of the Newfoundland-Iberia rift. In: Karner, G.D., Manatschal, G., Pinheiro, L.M. (Eds.), Imaging, mapping and modelling continental lithosphere extension and breakup. Geol. Soc. Sp. Publ. 282, 9–46.

Vajda, V., 2001. Aalenian to Cenomanian palynofloras of SW Scania. Sweden Acta Paleontologica Polonica 46, 403–426.

Velde, B., 1995. Origin and mineralogy of clays: clays and the environment. Springer-Verlag, Berlin.

Verati, C., Rapaille, C., Fé raud, G., Marzoli, A., Bertrand, H., Youbi, H., 2007. ^{40}Ar/^{39}Ar ages and duration of the Central Atlantic Magmatic Province volcanism in Morocco and Portugal and its relation to the Triassic–Jurassic boundary. Palaeogeogr. Palaeoclimatol. Palaeoecol. 244, 308–325.

Walsh, J.N., 1997. Inductively coupled plasma-atomic emission spectrometry (ICP-AES). In: Gill, R. (Ed.), Modern analytical geochemistry. Addison Wesley Longman, pp. 41–66.

Watson, J., Alvin, K., 1996. An English Wealden floral list, with comments on possible environmental indicators. Cretaceous Res. 17, 5–26.

Watson, J., 1988. The Cheirolepidiaceae. In: Beck, C.B. (Ed.), Origin and Evolution of Gymnosperms. Columbia Univ. Press, pp. 382–447.

Yan, Y., Xia, B., Lin, G., Cui, X., Hu, X., Yan, P., Zhang, F., 2007. Geochemistry of the sedimentary rocks from the Nanxiong Basin, South China and implications for provenance, paleoenvironment and paleoclimate at the K/T boundary. Sedim. Geol. 197, 127–140.

Zhang, Y., Pe-Piper, G., PiPer, D.J.W., 2014. Sediment geochemistry as a provenance indicator: Unravelling the cryptic signatures of polycyclic sources, climate change, tectonism and volcanism. Sedimentology 61, 383–410.