

*Clay minerals and geochemical evolution in
the Toarcian-lower Aalenian
of the lusitanian basin (Portugal)*

*Minerales de arcilla y evolución geoquímica
durante el Toarcense-Aalenense inferior
en la cuenca lusitaniense (Portugal)*

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ABSTRACT

The Toarcian-Lower Aalenian megasequence of the Lusitanian Basin shows quantitatively important marly-limestone sedimentation. In this type of sedimentary environment some parameters (clay minerals, Sr, $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) are very important as stratigraphic and palaeo-oceanographic markers. The analysis of these variables allowed the following conclusions to be made:

The clay mineral associations show lateral and vertical evolution in the series that show stratigraphic distribution and depositional control; the accumulation of chlorite, particularly in the sectors in the western part of the basin, indicates influx originating from the West, corroborating the palaeogeographic schemes accepted in some of the infilling phases, in accordance with the models set by Duarte (1995b).

The evolutionary spectrum of the geochemical elements shows, particularly for $\delta^{13}\text{C}$, exemplary stratigraphic distribution between lower and middle Toarcian. Its evolution along the length of the series agrees with the sequential developments of the series. The large, positive and more abrupt incursions can be seen in the transgressive depositional phases, whilst the more gradual negative tendencies correspond to regressive mechanisms.

Key words: Clay minerals, carbon and oxygen isotopes, strontium, marl/limestone alternations, Toarcian, Aalenian, Lusitanian Basin.

RESUMO

O Toarciano-Aaleniano inferior mostra na Bacia Lusitaniana uma sedimentação expressiva em sedimentos margo-calcários. Neste tipo de ambiente sedimentar, alguns parâmetros revelam-se importantes marcadores estratigráficos e paleoambientais. A análise dessas variáveis permite concluir o seguinte:

As associações dos minerais de argila apresentam uma evolução lateral e vertical na série que evidenciam uma repartição estratigráfica e um controlo de-posicional; a acumulação de chlorite, em especial nos sectores ocidentais da bacia, pressupõe um acarreio de material argiloso proveniente de Oeste, corroborando os esquemas paleogeográficos admitidos para algumas das fases de enchimento de acordo com os modelos de Duarte (1995b).

O espectro evolutivo dos elementos geoquímicos mostra, em especial para o $\delta^{13}\text{C}$, uma repartição estratigráfica exemplar ao nível do Toarciano inferior e mé-dio. A sua evolução ao longo da série é concordante com os desenvolvimentos sequenciais da série. As grandes incursões positivas, quase sempre mais bruscas, verificam-se nas fases deposicionais transgressivas enquanto que as tendê-nças negativas, mais graduais, correspondem a mecanismos regressivos.

Palavras chave: Minerais de argila, isótopos de carbono e oxigénio, estrô-ncio, alternâncias marga-calcário, Toarciano, Aaleniano, Bacia Lusitaniana.

RESUMEN

La megasecuencia correspondiente al Toarcense-Aalenense inferior en la cuenca Lusitaniense comprende una sucesión dilatada de materiales margo-sos y calcáreos. En este ambiente sedimentario, algunos parámetros (minerales de arcilla; Sr, C.¹³ y O.¹⁸) se revelan como indicadores paleoceanográficos y ambientales de gran importancia. El análisis de dichas variables ha permitido extraer las siguientes conclusiones:

Las asociaciones de minerales de arcilla muestran una evolución lateral y vertical en las series que revela una distribución estratigráfica y un control so-bre la sedimentación; la acumulación de clorita, particularmente en el sector occidental de la cuenca , indica el influjo de aportes del Oeste, lo que corrobora los esquemas paleogeográficos aceptados para algunas fases de relleno, de acuerdo con los modelos propuestos por Duarte (1995b).

El espectro evolutivo de los elementos geoquímicos muestra, especialmente en el caso del C.¹³, una distribución estratigráfica modélica entre el To-arcense inferior y medio. Su evolución a lo largo de todas las series concuerda con el desarrollo secuencial de las mismas. Las incursiones amplias positivas más abruptas, se registran durante las fases transgresivas, mientras que las tendencias negativas y más graduales se corresponden con los mecanismos o procesos regresivos.

Palabras clave: Minerales de arcilla, isótopos de Oxígeno, isótopos de Carbono, Estroncio, alternancia de margas y calizas, Toaciense, Aalenense, Cuenca Lusitanense.

INTRODUCTION

The evolutionary history of the toarcian sediments in the Lusitanian Basin is directly related to the first rifting phase of the North Atlantic (Wilson, 1988; Wilson *et al.*, 1989; Soares *et al.*, 1993a). This event includes infilling from the Middle Triassic (Rocha *et al.*, 1990) to the top of the Callovian, with sedimentary unconformities of basinal value (Soares *et al.*, 1993a,b; Soares & Duarte, 1995). In the Toarcian-Lower Aalenian there is a phase of substantial lutitic marly-limestone accumulation, with hemipelagic characteristics, despite the depositional differentiation occurring in some sectors such as Tomar and Peniche (Duarte, 1995b). These sediments show sequential organisation supported by two megasequential units (**ME1** and **ME2**, Duarte & Soares, 1993; Soares & Duarte, 1995) and four depositional sequences (mesosequences: **MST**), bounded by correlative regional unconformities (**DT**) (Fig. 1). Those units show, in differentiated contexts of a homoclinal ramp, shallowing upward evolutions; these characteristics are particularly evident in the proximal parts of the basin such as Tomar (**MSTT**) and in the southwest border at Peniche (**MSTP**) (Figs. 1 and 2).

In this sedimentary context, it is important both to analyse in detail the lateral and vertical behaviour of some chemical and mineralogical parameters and establish fine stratigraphy based on its evolutionary aspects. The type of evolution, the anomalies found and/or the correlation between some of these parameters may also contribute to the understanding of the palaeoenvironmental variables that may have controlled deposition.

STRATIGRAPHIC SECTIONS

There is quite a good understanding of the carbonate facies that characterise the Toarcian-Lower Aalenian series in all the Lusitanian Basin (Duarte, 1995b; 1997). Hemipelagic sedimentation dominates a large part of the basin, with abundant ammonoids, sometimes with very rhythmic marl/limestone alternations. The Tomar and Peniche regions contrast with this facies setting, showing associations with characteristic of a shallowing ramp with strong benthic bioclastic deposition (Duarte, 1995b, 1997) and an outer to middle submarine fan with calciturbidites (Wright & Wilson, 1984) respectively (Fig. 3).

In order to demonstrate the depositional variability in the basin during this time period, six sections were chosen (Coimbra, Figueira da Foz, Rabaçal, Porto de Mós, Tomar and Peniche), all of them referenced in the biostratigraphic scale of ELMI *et al.* (1989) (Fig. 2).

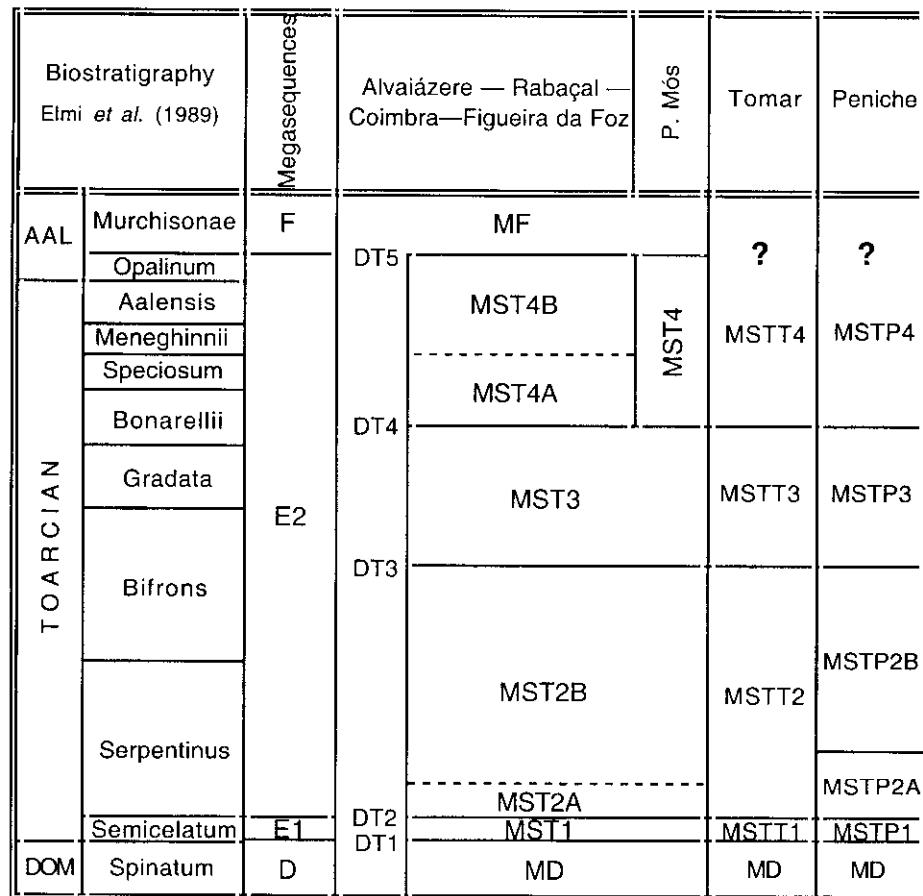
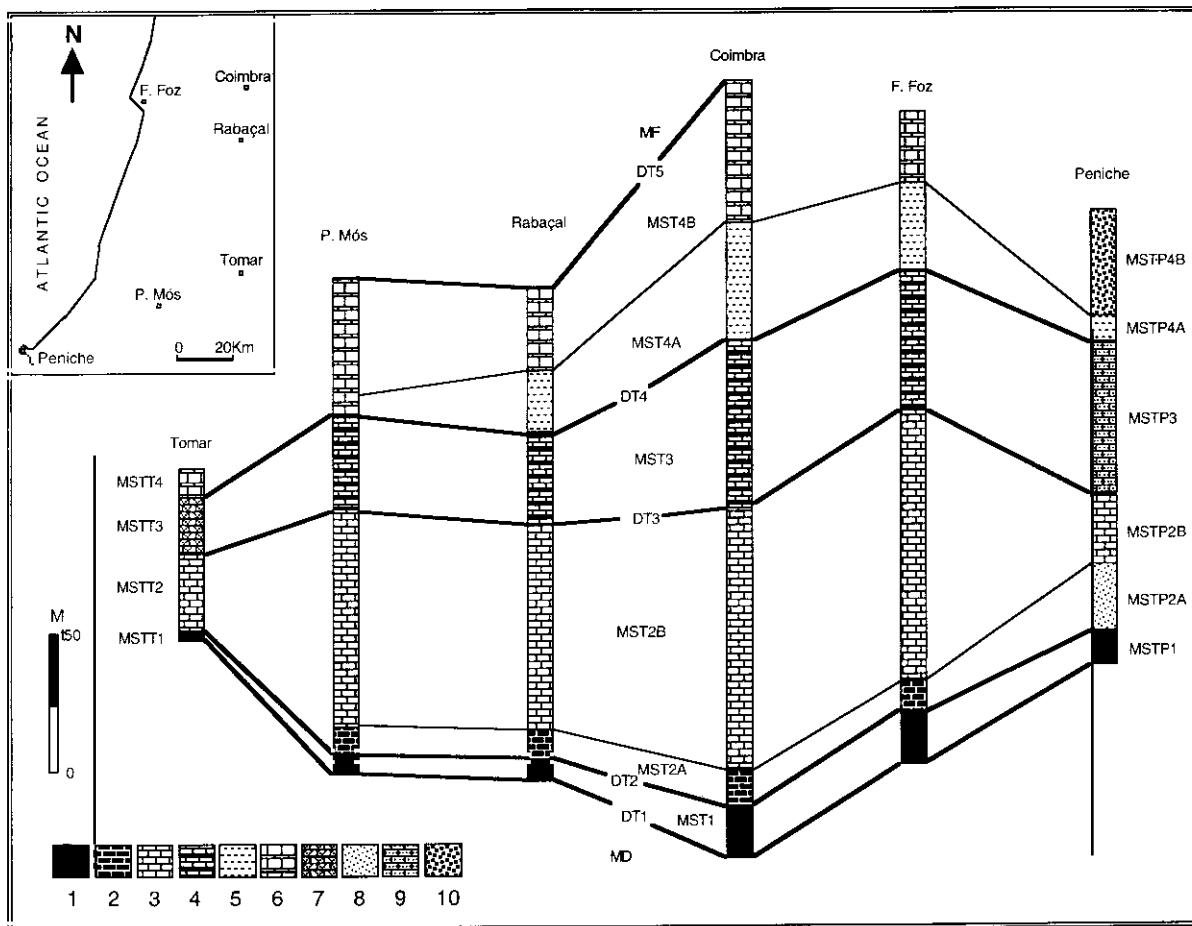


FIG. 1.—Sequential chart of the Toarcian-Lower Aalenian from the Lusitanian Basin: megasequential unities (Soares & Duarte, 1995), unconformities and depositional sequences (Duarte, 1995b).

FIG. 1.—Cuadro secuencial del Toarcense-Aalenense inferior para la Cuenca Lusitaniense: Unidades megasecuenciales, según Soares y Duarte (1995); disconformidades y secuencias deposicionales, según Duarte (1995b).

FIG. 2.—Location map of the studied sections and lateral and vertical repartitions of the depositional sequences (horizontal not to scale). 1- Marl/limestone decimetric alternations with small brachiopods; 2- Calcilutitic to calcarenitic facies alternated with marls in centimetric scale; 3- Metric to decimetric lutitic marl/limestone alternations; 4- Decimetric marl/limestone alternations with bioconstructions; 5- Marls and marly-limestones with brachiopods; 6- Marls and micritic limestones with bioconstructions; 7- Bioclastic limestones; 8- Lenticular siliciclastic facies; 9- Oolitic and peloidal facies; 10 - Oolitic and sandy limestones facies.

FIG. 2.—Mapa de situación de los perfiles estudiados y repartición horizontal y vertical de las secuencias



depositacionales (la horizontal no está a escala). 1- Alternancia decimétrica de margas y calizas con pequeños braquiópodos; 2- Facies calcilutíticas a calcareníticas alternando con margas, a escala centimétrica; 3- Alternancia métrica a decimétrica de lujitas y margocalizas; 4- Alternancia decimétrica de margas y calizas con bioconstrucciones; 5- Margas y calizas margosas con braquiópodos; 6- Margas y calizas micríticas con bioconstrucciones; 7- Calizas bioclasticas; 8- Facies siliciclasticas lenticulares; 9- Facies oolíticas y peloidales; 10- Facies de oolitos y calizas arenosas.

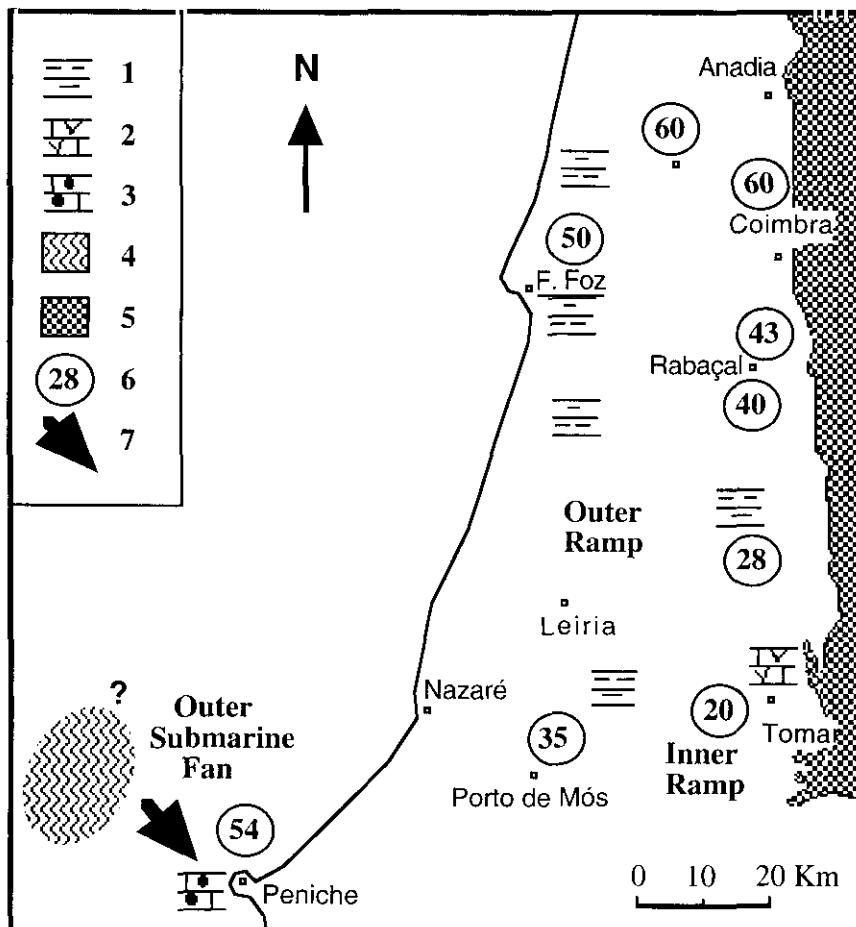


FIG. 3.—Facies and paleogeographic map during MST3 phase (Bifrons-Bonarellii zones) (Duarte, 1995b). 1 - Marly-limestone facies; 2 - Bioclastic facies; 3 - Detrital and resedimented oolitic facies; 4 - Emerged block; 5 - Hesperic Massif (submerged during MST3); 6 - Thickness; 7 - Palaeocurrents.

FIG. 3.—Facies y mapa paleogeográfico durante la fase MST3 (cronozonas Bifrons-Bonarellii; Duarte, 1995b). 1- Facies de calizas margosas; 2- Facies bioclasticas; 3- Facies oolíticas resedimentadas y detriticas; 4- Bloque emergido; 5- Macizo Hespérico (sumergido durante la fase MST3); 6- Potencia; 7- Paleocorrientes.

ANALYTICAL METHODS

The sedimentary parameters in this study include clay minerals in marly facies and some chemical components of the carbonate fraction such as strontium and the carbon and oxygen stable isotopes. The sampling took into consideration the previous characterisation of the lithofacies and their

mesosequential setting (*vide* Duarte, 1995b). Care was taken to make the process systematic, and at times even random, to best illustrate the sedimentary variability of the series (Tables 1 to 9 and Figs. 4 to 7). The stable isotopes were only analysed in the Rabaçal, Figueira da Foz and Porto de Mós sections (Figs. 8 and 9).

Clay Minerals: The clay fraction of marl facies were analysed by X-ray diffraction (XRD) using a Phillips PW1840 diffractometer with CuK radiation. The method of mineralogical preparation and determination was defined in Lapa & Reis (1977). Semi-quantitative evaluations are based on the calculation of the heights and areas of the registered peaks, a method which is described in Thorez *et al.* (1970) (*in* Lapa & Reis, 1977). Vermiculite and interstratified minerals were not included in this process due to the difficulty in obtaining well defined peaks for them. The determination of the crystallinity of illite using the Kubler index (Kubler, 1964), was also carried out in the characterisation of the clay-mineral assemblages.

Strontium: The chemical analyse (about 400 samples) were carried out under the carbonate phase via atomic absorption spectrometry.

Carbon and Oxygen Isotopes: The analytical process of 70 samples consisted of the removal of carbon dioxide via the solubility of the carbonate phase using 100% phosphoric acid (McRea, 1950; Craig, 1957). The final isotopic composition was obtained with the use of a mass spectrometer, with the calculated values being in relation to PDB international standard (Craig, 1957).

CLAY MINERALS

The apparent rhythmicity demonstrated by the marl/limestone lithofacies does not agree with the associations of the clay minerals. This study of approximately four hundred samples shows that there is a clear stratigraphic division of the minerals in relation to the mesosequential units established (Figs. 1 and 2), with simple and complex associations of illite, kaolinite, smectite, chlorite, various types of mixed layers and rare vermiculite. In this context, lateral variation of the mineralogical associations can be seen in determined stratigraphic positions. This fact, although difficult to interpret due to the exclusive action of diagenesis, contributes decisively in palaeogeographic reconstructions.

The results obtained allow the discussion to be geared in two different directions: a stratigraphic (sequential) one, and a palaeogeographic one. In the latter, the variations of clay minerals (both horizontal and vertical) reveal that the basin suffered opposite detritic influences from east and west (discussion *in* Duarte, 1995b). The possible detritic origin of the minerals can serve as an important criterion in the understanding of the type of influx originating from the land borders of the basin *via* the extensive fluctuations in sea-level.

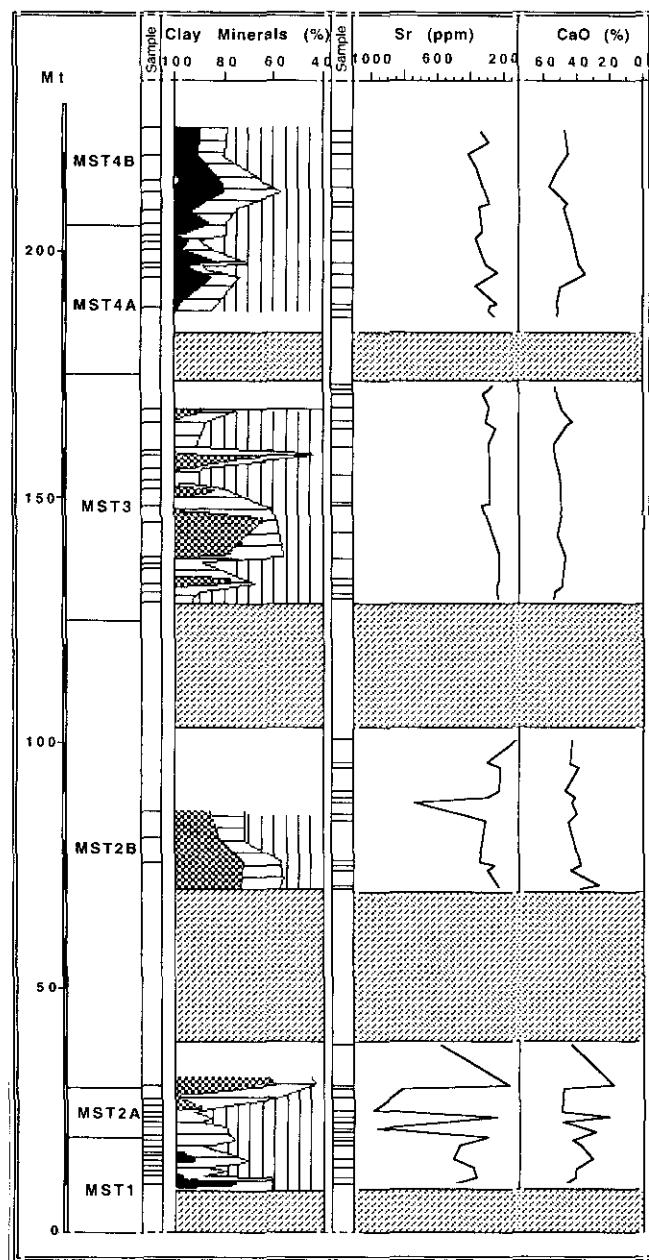


FIG. 4.—Evolution of clay minerals and geochemical data in the Figueira da Foz region (see legend in Fig. 5).

FIG. 4.—Evolución de los datos geoquímicos y de los minerales de arcilla en la región de Figueira da Foz (v. la leyenda, en la Fig. 5).

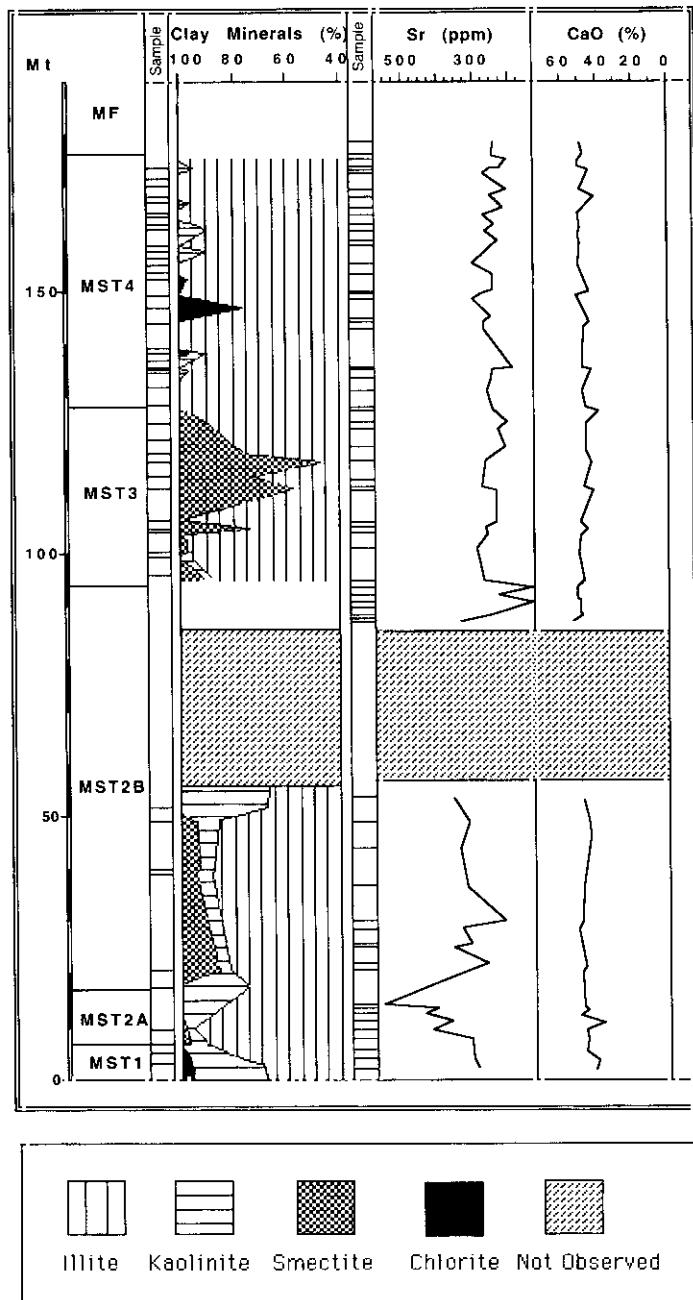


FIG. 5.—Evolution of clay minerals and geochemical data in the Porto de Mós region.

FIG. 5.—Evolución de los datos geoquímicos y de los minerales de arcilla en la región de Porto de Mós.

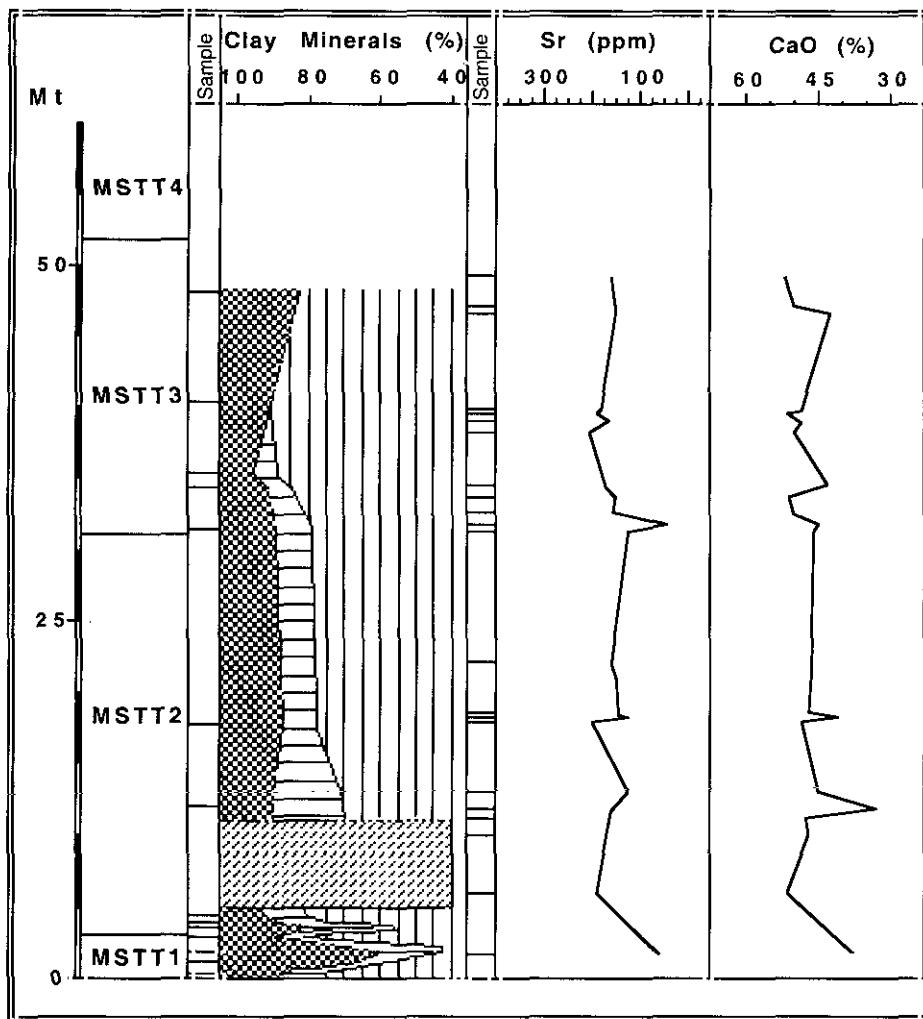


FIG. 6.—Evolution of clay minerals and geochemical data in the Tomar region (see legend in Fig. 5).
 FIG. 6.—Evolución de los datos geoquímicos y de los minerales de arcilla en la región de Tomar (v. la leyenda, en la Fig. 5).

RESULTS AND DISCUSSION

Illite: This mineral is present in all mineralogical associations. Its predominance is so evident that the lowest values, with the exception of MST2A and the base of MST2B, are always above 55%. The amounts of illite in these two exceptions, which are always associated with quite important values of smec-

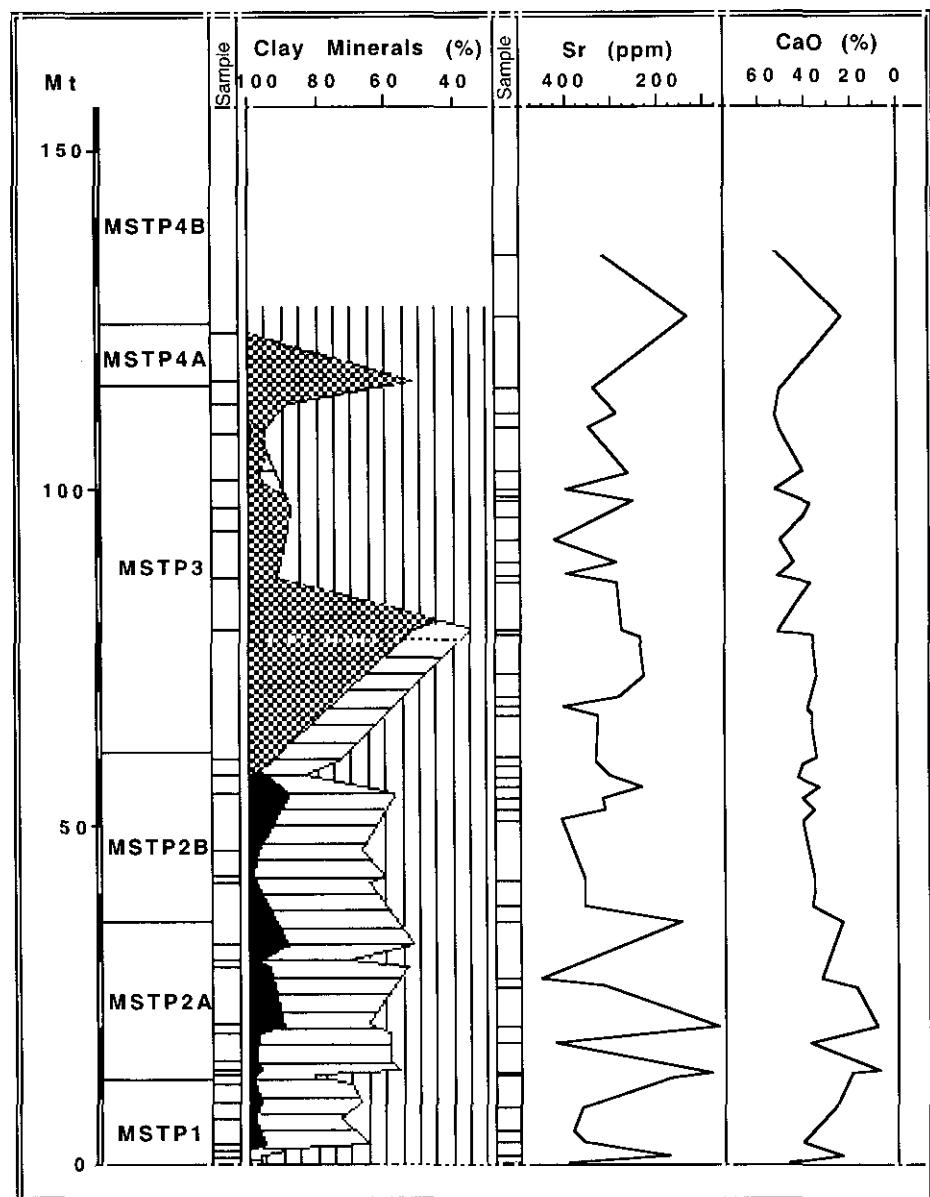


FIG. 7.—Evolution of clay minerals and geochemical data in the Peniche region (see legend in Fig. 5).
FIG. 7.—Evolución de los datos geoquímicos y de los minerales de arcilla en la región de Peniche (v. la leyenda, en la Fig. 5).

Sections	% Illite	% Kaolinite	% Smectite	% Chlorite	Sr (ppm)
Rabaçal	72-100	0-17	0	0-11	268-373
Porto Mós	70-91	6-25	0-3	0-4	290-311
Coimbra	55-86	3-19	2-38	0	406-774
F. Foz	60-94	7-22	0	0-33	330-510
Tomar	43-83	7-21	6-40	0	59
Peniche	65-97	4-30	0	0-5	358-396

TABLE 1.—Clay minerals and Strontium distribution in **MST1**, **MSTT1** and **MSTP1**.TABLA 1.—Minerales de la arcilla y distribución del Estroncio en **MST1**, **MSTT1** y **MSTP1**

Sections	% Illite	% Kaolinite	% Smectite	% Chlorite	Sr (ppm)
Rabaçal	88-97	4-11	0-3	0	181-478
Porto Mós	88-96	3-12	0	0	309-557
Coimbra	32-65	6-30	0-32	0	627-1.032
M. Velho	7-51	10-28	29-82	0	—
Cantanhede	32-65	10-21	23-48	0	—
F. Foz	60-93	7-39	0-9	0	181-972
Tomar	56-81	8-30	0-19	0	190
Peniche	51-81	19-41	0	2-12	71-461

TABLE 2.—Clay minerals and Strontium distribution in **MST2A**, base of **MSTT2** and **MSTP2A**.TABLA 2.—Minerales de la arcilla y distribución del Estroncio en **MST2A**, base de **MSTT2** y **MSTP2A**.

tite, illite-smectite mixed-layers and kaolinite, sporadically reach values of less than 10%. These low values are due to antagonistic behaviour between illite and smectite.

Its highest value is found from the top of **MST3** (generally above 75%; Tables 1 to 6) to the top of **MST4B**, becoming at times an exclusive mineral, equalling the randomly interstratified illite-smectite. This tendency seems to continue past the upper boundary for that unit (**ME2**), remaining in the sediments of the middle and upper Aalenian (**MF**).

The state of degradation of this mineral is a characteristic which remains practically constant in the whole series, showing very variable crystallinity values (between 0.5 and 1.1). However, there are some signs that indicate depositional variability. This is the case in **MSTP2A** and **MST2B** of the western regions (Figueira da Foz and S. Pedro de Moel), where the detritic contribution and the sedimentary context take on a unique appearance, and where crystallinity levels may reach values of 0.5 and 0.6, relatively higher than values obtained

Sections	% Illite	% Kaolinite	% Smectite	% Chlorite	Sr (ppm)
Alvaiázere	76-92	7-24	0-9	0-8	—
Rabaçal	86-96	4-14	0	0-7	194-339
Porto Mós	67-89	2-33	0-19	0	125-361
Coimbra	41-91	4-37	0-40	0-11	88-308
M. Velho	29-47	5-24	37-51	0	—
F. Foz	43-72	11-18	14-44	0	134-727
S. P. Moel	45-47	14-23	12-36	5-17	257-321
Tomar	9-78	7-33	0-84	0	127-197
Peniche	57-84	12-39	0-8	0-12	241-412

TABLE 3.—Clay minerals and Strontium distribution in MST2B, MSTT2 and MSTP2B.

TABLA 3.—Minerales de la arcilla y distribución del Estroncio en MST2B, MSTT2 y MSTP2B

Sections	% Illite	% Kaolinite	% Smectite	% Chlorite	Sr (ppm)
Alvaiázere	83-100	0-9	0-7	0-11	206-210
Rabaçal	71-95	5-24	0-5	0-7	209-353
Porto Mós	46-100	0-4	0-54	0	211-289
Coimbra	29-100	0-16	0-71	0	275-381
F. Foz	43-94	3-39	0-38	0-9	230-334
Tomar	79-89	0-10	4-17	0	45-205
Peniche	35-100	0-17	0-47	0-2	232-423

TABLE 4.—Clay minerals and Strontium distribution in MST3, MSTT3 and MSTP3.

TABLA 4.—Minerales de la arcilla y distribución del Estroncio en MST3, MSTT3 y MSTP3

ned in the other sectors. This evidence seems to be linked to detritic influx originating from the west border of the basin rather than a possible diagenetic overprint, hypothetically responsible for the increase in the illite crystallinity. Therefore, this mineral was probably inherited from the surrounding continental landmasses. Its almost exclusive occurrence from the top of MST3 (top of Gradata zone) is certainly related to the regressive development of the sedimentation, related to lowstand of sea-level, as is shown in DUARTE (1995b) (Fig. 10).

Kaolinite: With the exception of the presence of mixed-layers, whose identification and evaluation is always difficult, kaolinite is the second most represented type of clay mineral in the series. Parallel to the expansive minerals,

<i>Sections</i>	% Illite	% Kaolinite	% Smectite	% Chlorite	Sr (ppm)
Alvaiázere	88-100	0-12	0	0	—
Rabaçal	93-100	0-5	0	0-2	226-373
Coimbra	100	0	0	0	195-324
Cantanhede	83-100	0-13	0-15	0-8	290-292
F. Foz	69-91	4-25	0	0-18	244-370
Peniche	53-100	0	0-47	0	133-322

TABLE 5.—Clay minerals and Strontium distribution in **MST4A** and **MSTP4A**.
TABLA 5.—Minerales de la arcilla y distribución del Estroncio en **MST4A** y **MSTP4A**.

<i>Sections</i>	% Illite	% Kaolinite	% Smectite	% Chlorite	Sr (ppm)
Alvaiázere	95-100	0	0-5	0	—
Rabaçal	92-100	0-6	0	0	207-385
Porto Mós	77-100	0-10	0-8	0-23	189-298
Coimbra	94-100	0	0	0-6	248-343
Cantanhede	85-100	0-13	0-7	0	309-324
F. Foz	57-90	6-25	0	4-19	288-413

TABLE 6.—Clay minerals and Strontium distribution in **MST4B**.
TABLA 6.—Minerales de la arcilla y distribución del Estroncio en **MST4B**.

its highest representation is in **MST2A** (maximum of 39% in Figueira da Foz; Table 2, Fig. 4), in **MSTP2A** (19 to 41% with an average of 33.4%; Table 2, Fig. 7) and in the bases of **MST2B** (maximum of 37% in Coimbra; Table 3) and **MSTP2B** (maximum of 40%, with an average of 28%; Table 3). It becomes rarer from **MST3** onwards, and even disappears in the regions of Coimbra and Porto de Mós (Tables 4 to 6; Fig. 5). In **MST4A**, it is practically absent, except for the regions of Cantanhede and Figueira da Foz, occasionally present with values under 12%. In **MST4B**, this tendency continues, being intensified in the differentiated sector in the northwest part of the basin. In Tomar and Peniche (Figs. 6 and 7), this mineral disappears earlier than in the other regions (top of **MSTT3** and base of **MSTP3**). This could be an indication of an ever increasing continental landmass influence on deposition (marine environment becoming progressively shallower).

The abundance of kaolinite in the jurassic sediments is, according to Chamley (1989), an indicator of landmasses with a hot climate and constant humidity. The absence of this mineral, or its reduction, seen from the Upper

Toarcian upwards can be justified by a lowering of sea level and the development of more arid conditions. Its enrichment in the distal sector of the basin (Figueira da Foz), throughout the units **MST4A** and **MST4B**, may be the result of two mechanisms: diagenetic alteration of the other clay minerals present (illite and chlorite) or the detritic influx from the west, as is the case with chlorite.

If one can assume a strong relationship with the climate, then the presence/absence of kaolinite may be associated more with the degree of drainage on the continental areas than the transgressive or regressive character of the sedimentation. It may even be possible that a period of more arid conditions may be responsible for the practical disappearance of kaolinite in the marine environment.

Smectite: Smectite and randomly illite-smectite mixed layers show an apparently more homogeneous stratigraphic distribution, with these last explained in some cases, as the result of diagenesis (degradation).

With high values in **MSTT1** (and in **MST1** in Coimbra; Table 1), **MST2A** (23 to 48% in Cantanhede; 28 to 82% in Montemor; Table 2), base of **MST2B** (37 to 51% in Montemor; Table 3), base of **MST3** (< 71% in Coimbra, < 54% in Porto de Mós; Table 4, Fig. 5) and in **MSTP3** (Fig. 7), smectite shows quite open basal reflections, denoting low crystallinity values. The horizontal distribution of this mineral is quite variable, with extremely objective zonation in **MST2A** and the base of **MST2B** (Serpentinus zone) in the region from Montemor-o-Velho to Cantanhede and in **MST3** (Bifrons-Gradata zones) in the regions of Coimbra and Porto de Mós.

There has been a lot of discussion about the origin of these minerals. The idea defended by Chamley *et al.* (1992), that clay mineralogy based on tectonic instability, seems to coincide particularly well with the occurrence of this mineral in **MST2A**. This unit, characterised by deposition in shallow carbonate ramp with increased energy conditions (Duarte & Soares, 1993), is symptomatic of the intensity of mechanical erosion, which according to Chamley *et al.* (1992), may have contributed to the dismantling of smectite resulting from pedological activity at the surface. The privileged distribution of this mineral in **MST2A** in Montemor region, or rather along the Arunca-Montemor alignment, may be directly linked to the meridional fracturation shown in Soares *et al.* (1993a).

With the exception of **MST2A**, its highest level of occurrence appears in transgressive (flooding) conditions demonstrated in the bases of the mesosequences and, in particular in the cases of **MST2B**, **MSTP3** and **MST3**; the latter, in sectors as far away, in palaeogeographical terms, as Coimbra and Porto de Mós (Fig. 3), where smectite is exclusively associated with illite. In fact, it seems that its highest values have nothing to do with any type of diagenetic evolution between the base and top of the series, in spite of the fact that locally, diffractograms show a very degraded association of smectite and illite.

Owing to good stratigraphic distribution of this mineral, it is possible that the role of diagenesis is well expressed in mineralogical associations with ran-

domly illite-smectite mixed layers. These clay minerals can be the result of the intermediate transformation of smectite in illite (is strong the antagonism between these two minerals).

Chlorite: This mineral tends to occur completely randomly in the whole series, with values rarely reaching 10%. There are exceptions in the sections situated in the most western parts of the basin, such as **MST1** in Figueira da Foz (< 33%; Table 1, Fig. 4), **MST2B** in S. Pedro de Moel (5 to 17%; Table 3), **MSTP2A** (< 12%; Table 2) and **MSTP2B** (< 12%; Table 3) in the region of Peniche (Fig. 7). The content of this mineral in the clay assemblages from the top of **MST3** are practically constant in Figueira da Foz (<18% in **MST4A**; Table 5; < 19% in **MST4B**; Table 6). Its absence in determined stratigraphic compartments of the series such as **MST2A** (Table 2), base of **MST3** and the whole Tomar section (Fig. 6) is also important.

Not being a good stratigraphic marker, the occurrence of chlorite seems to be closely related to the geographic location of the sections in the basin, therefore being a good palaeogeographic indicator. Its frequent association with silty micaceous minerals, present in the marls of the many units in the Figueira da Foz to Peniche coast, allows us to consider a detritic origin from the West border of the basin. Assuming this, chlorite appears as a mineral directly inherited from the igneous and metamorphic rocks of the Berlenga horst, a block which was probably emerged during the Upper Toarcian (Wright & Wilson, 1984; Duarte, 1995b). Its abrupt reduction to the east of Figueira da Foz, particularly in the case of **MST4B**, may be in agreement with Zimmermann (1982) who states that this mineral has a terrigenous origin with deposition after a less important transport phase.

Vermiculite: In this study, vermiculite appears casually in mineralogical associations, and it is not possible to relate it to any type of stratigraphic position and/or geographic location. Considering that its presence is associated with sections along the length of low sloping vertices, it is possible that it is related to more recent superficial pedological alterations. This idea is corroborated by Chamley *et al.* (1992, p. 116), that explains the vermiculitic state as a result of smectite alteration due to Mg²⁺ fixation.

STRONTIUM DISTRIBUTION

RESULTS AND DISCUSSION

The values of Sr, when analysed vertically (Figs. 4 to 7), do not show any kind of tendency, but demonstrate stratigraphic zonation practically concordant with the mesosequential units (Tables 1 to 6). This fact alone apparently demonstrates a weak alteration in diagenetic processes. The variability of this element in the toarcian lithofacies allows us to draw the following conclusions:

— The main anomalies, with values of over 1000 ppm, some with more than 4% of the total rock, can be seen in particular contexts, being associated with celestite nodules (top of **MST2B**, in Rabaçal), encrinitic facies (**MST3**, in Coimbra, Rabaçal, Alvaiázere and Porto de Mós) and lateral facies of *mud-mounds* (**MST4B**, in Rabaçal).

— Isolating the lithological contexts mentioned above, the highest values of Sr in the carbonate phase can be observed in **MST2A** (Coimbra-Figueira da Foz region; Table 2). The high values observed in limestone lithology, on average higher than 600 ppm, stand out in comparison to the other values observed in the units above and below it. It is also possible to establish chemical zonation in the whole Toarcian series of Tomar section (with values systematically below 200 ppm; Tables 1 to 4; Fig. 6). **MST2B** seems to show negative evolution, a tendency which is particularly evident in Rabaçal and Porto de Mós (Fig. 5).

Values of this element may be conditioned by numerous factors including diagenetic ones. According to Renard (1984), the evolution of this element in pelagic environments seems to be, among other reasons, associated with transgression-regression cycles. However, the analysis of its behaviour in its association with the variation of sea-level in the Portuguese series needs to be carried out with some prudence. The apparent antagonism in the values of strontium, observed in the microsparites of the units **MST2A** (on average 600 ppm) and **MST2B** (120 to 250 ppm), discard the possibility of diagenetic effects and justify a primary origin. However, the well-fundamented theory put forward by Renard (1984, p. 357) that the synthesised carbonates during the transgressive phases enriched in strontium relative to those produced in regressive phases cannot be applied to the toarcian series of the Lusitanian basin. In fact, considering the particular type of sedimentation in **MST2A** (*vide* Duarte, 1995b), where benthic macrofauna is absent and the level of $\delta^{13}\text{C}$ is very low (as will be demonstrated further on), it is possible that this element may have a continental landmass origin, linked to evaporites in the drainage area. This link with the salinity of the water may justify the strong increase in benthic fauna in the base of **MST2B** (*vide* Duarte, 1995b) where the values are particularly low (on average 200-250 ppm). The same relationship between benthic fauna and Sr is visible for example, in **MSTT2** where not only is there a big percentage of organisms, but also low values of that element under 200 ppm) are evident (Fig. 6).

STABLE ISOTOPES

The stable isotope studies in the carbonate sediments have been widely used in the stratigraphic definition of the old sedimentary record and as important indicators in palaeoclimatic reconstructions. It is known that their variations, particularly in pelagic environments, witness the main chemical changes oc-

<i>Samples</i>		$\delta^{13}\text{C}$	$\delta^{18}\text{O}$
251 Z6	MD	1,43	-3,79
251 Z12	MST1	1,84	-3,28
251 Z13		2,07	-3,82
251 Z15a		1,73	-3,58
251 Z15d	MST2A	1,08	-3,52
251 FC15		1,20	-3,14
251 Z15g		1,97	-2,77
251 Z19		2,20	-3,92
251 Z22		4,07	-3,80
251 Z32		3,72	-3,74
251 Z40		3,07	-3,89
251 Z44	MST2B	2,73	-3,80
251 Z46		2,59	-3,79
251 Z48		2,09	-3,52
251 Z49		1,87	-3,45
251 Z50s		1,71	-3,19
251 Z51		2,52	-3,80
251 Z54s		2,49	-3,03
251 Z58	MST3	1,99	-2,82
251 Z59b		1,98	-3,07
251 Z64		1,70	-3,03
251 Z65		1,77	-2,57
251 Z66a		1,55	-2,68
251 Z66d	MST4A	1,79	-2,48
251 Z68c		1,92	-2,72
251 Z69b		2,19	-2,08
251 Z70s2-1		1,89	-2,59
251 Z70s2-2		1,89	-4,70
251 Z75C	MST4B	1,95	-2,51
251 Z90		1,87	-2,30
251 Z94	MF	2,08	-2,66
251 Z105		2,22	-2,65

TABLE 7.—Isotopic compositions of the samples studied from Rabaçal section.

TABLA 7.—Composiciones isotópicas de las muestras estudiadas de la columna de Rabaçal.

<i>Samples</i>		$\delta^{13}C$	$\delta^{18}O$
308 PM3	MST1	1,66	-4,05
308 PM7		0,66	-4,03
308 PM13	MST2A	0,20	-4,68
308 PM19		0,89	-3,46
308 PM31		3,87	-3,37
308 PM62		4,27	-3,68
308 PM72	MST2B	4,29	-3,83
308 PM81		3,83	-3,42
308 PM113		3,33	-3,58
308 PM160		2,79	-3,08
308 PM199		2,54	-3,01
308 PC202		2,46	-2,79
308 PC206		2,61	-2,86
308 PC218	MST3	2,55	-3,02
308 PC243		2,05	-2,84
308 PC265		1,92	-2,61
308 PC288		2,05	-2,35
308 PM509		1,48	-1,95
308 PC304		1,91	-2,71
308 PM544	MST4	1,93	-2,29
308 PM592b		2,07	-2,78
308 PM642		1,75	-2,40
308 PM894		1,68	-2,32
308 PM906	MF	2,18	-1,81

TABLE 8.—Isotopic compositions of the samples studied from the P. Mós section.
TABLA 8.—Composiciones isotópicas de las muestras estudiadas en la columna de P.Mós.

curring in oceans (Scholle & Arthur, 1980; Renard & Letolle, 1983; Renard, 1984, 1986).

When selecting sections for isotopic analysis, various factors were taken into account including, the hemipelagic character of the sediments (marl/limestone), the existence of a fine stratigraphy framework supported by ammonite scales; their geographic position in the basin, continuity of observation and weak affection to tectonic processes. Therefore, the Tomar and Peniche sections were excluded due to the fact that they show very particular sedi-

<i>Samples</i>		$\delta^{13}\text{C}$	$\delta^{18}\text{O}$
239 VF4	MST1	1,44	-3,16
239 VF30		2,34	-3,29
239 B68	MST2A	0,47	-2,54
239 B138		0,64	-2,47
239 B254	MST2B	2,37	-3,23
239 B309		3,00	-3,33
239 B314		1,68	-3,09
239 B400	MST3	2,21	-3,39
239 B439		1,80	-2,83
239 B455		1,92	-2,94
239 B489		1,76	-3,08
239 B820	MST4A	1,58	-2,29
239 B838		1,77	-2,49
239 B882	MST4B	2,06	-2,36

TABLE 9.—Isotopic compositions of the samples studied from the F. Foz section.

TABLA 9.—Composiciones isotópicas de las muestras estudiadas en la columna de F. Foz.

mentary conditions (Duarte, 1995b, 1997). Isotopic data are presented in Tables 7 to 9, their stratigraphic and vertical distributions being shown in Fig. 8 and Fig. 9.

RESULTS

Carbon Isotopes: In the sections analysed, the values of $\delta^{13}\text{C}$ vary between +0.2‰ and +4.27‰, with significant oscillations between some of the meisosequential units. The isotopic variation curve of carbon ($\delta^{13}\text{C}$) reinforces the tendencies illustrated in Duarte (1995a). As is demonstrated in Fig. 8, the large fluctuations occur mainly between the base of the Lower Toarcian and the Gradata zone (**MST1**, **MST2A**, **MST2B** and **MST3**) thus indicating its importance as a stratigraphic marker. In **MST4** (Upper Toarcian-Lower Aalenian), the isotopic values seem more homogeneous, still showing a tendency to gradual reduction.

Detailed analysis shows that the fluctuations in $\delta^{13}\text{C}$ are tendentially negative along the length of the mesosequences with nearly perfect gradual evolution schemes in **MST2B** and **MST3**. The more abrupt alterations, which always correspond to positive oscillations, were registered in the **MD/MST1**, **MST2A/MST2B** boundaries, and between **MST2B** and **MST3** (particularly in the Rabaçal section) (Fig. 8). Due to criteria of a sedimentological and palaeoecological nature, the base of these units (**MST1**, **MST2B** and **MST3**) is interpreted as being the result of a deepening phase in the sedimentation, or rather, an increase in paleobathymetry. In this context, this geochemical sign seems to clearly illustrate the changes in sea level. $\delta^{13}\text{C}$ supplies the perfect explanation for the macroscopic contrast between the **MST2A** and **MST2B** units. The abrupt increase clearly shows the change in sedimentary style operating between the two sub-units, with the deposition of marl/limestone decimetric to metric alternations, rich in benthic marine fossils in the base of **MST2B**. The extent of this tendency, shown in Fig. 8, is clearly demonstrated by the positive variation of 3% —between +0.89% (PM19) and +3.87% (PM31)— and can be seen in the Porto de Mós section (Table 8). In the others positions of the basin, although the variations are not as marked, they also show values which are statistically important —from +0.64% (B138) to +2.37% (B254) in Figueira da Foz (Table 9) and from +1.97% (Z15g) to +4.07% (Z22) in Rabaçal (Table 7).

In contrast, the boundary between **MST1** and **MST2A** shows the only abrupt reduction in the evolution of $\delta^{13}\text{C}$ in the following: from +2.07‰ (Z13) to +1.08‰ (Z15d) in Rabaçal (Table 7), from +1.66‰ (PM3) to +0.2‰ (PM13) in Porto de Mós (Table 8) and from +2.34‰ (VF30) to +0.47‰ (B68) in Figueira da Foz (Table 9). The nature of the facies and their sequential arrangement illustrate a correlative reduction of the water column in **MST2A** (Duarte, 1995b, 1997).

In the upper Toarcian, the evolution tends to become a little more complex. In any case, $\delta^{13}\text{C}$ tends to diminish along the length of **MST4** in all sections, reaching values lower than +2‰: +1.87‰ (Z90 in Rabaçal), +1.68‰ (PM894 in Porto de Mós) and +1.77‰ (B838 in Figueira da Foz) at the top of the mesosequence. If the boundary between **MST4A** and **MST4B** is not marked, in terms of significant oscillation of the carbon isotope, this is not the case between **MST4B** and the base of **MF**. An isotopic increase was observed in this megasequential boundary (**ME2/MF**), which contradicts the previous negative evolution. In Rabaçal and Porto de Mós (Tables 7 and 8), where the modification of the facies and their sequential arrangement is more noticeable, the values increase to +2.22% (Z105) and +2.18% (PM906) respectively, levels of the base of **MF**.

Oxygen Isotopes: The range in variation of the $\delta^{18}\text{O}$ varies in the three sections, from between -4.70‰ and -1.81‰ (Tables 7 to 9). The results obtained are not as clear cut as those for carbon in terms of sedimentary evolution (Fig. 9). The clear tendency for a gradual increase along the length of

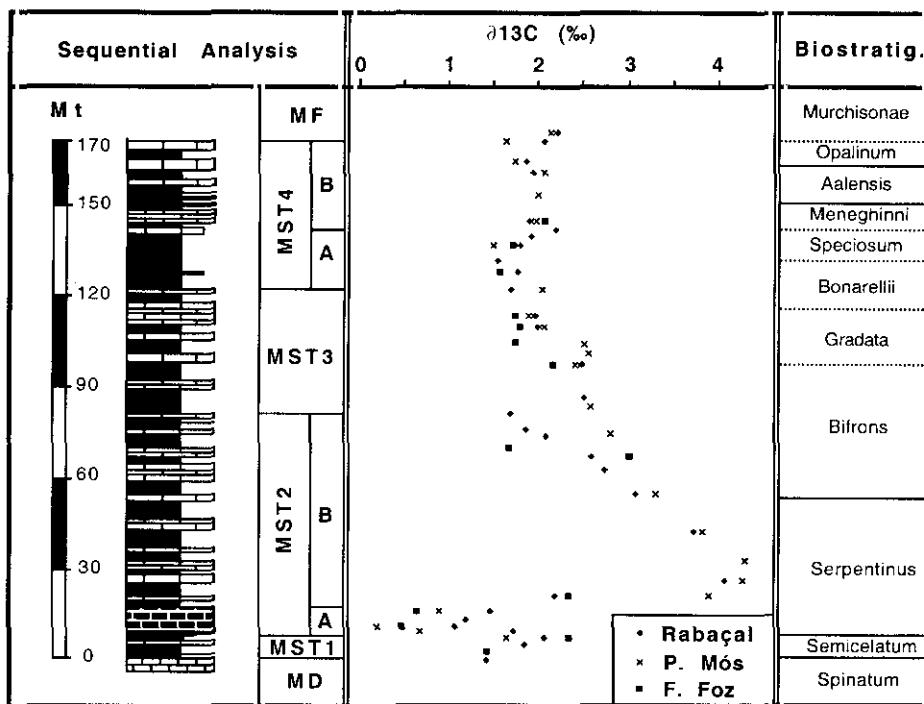


FIG. 8.— $\delta^{13}\text{C}$ distribution in the Toarcian-lower Aalenian from the Lusitanian Basin (synthetic section of Rabaçal).

FIG. 8.—Distribución del C.¹³ durante el Toarcense-Aalenense inferior en la Cuenca Lusitaniense (sección sintética de Rabaçal).

each section (values becoming less negative), is not a good indicator of an exclusively primary genetic explanation. Taking into account the thermodependence of oxygen (McCrea, 1950; Epstein *et al.*, 1953), these results, despite precautions taken not to include recrystallised limestone lithofacies at the sampling phase, do not seem to show anything more than a diagenetic effect. The best example of this can be seen in the almost uniform evolutions between **MST2A** and the other units above and below it (**MST1** and **MST2B**), contrary to the abrupt variations observed in $\delta^{13}\text{C}$. The lithification of the limestone facies may be closely linked to the negativity of the $\delta^{18}\text{O}$ values (-4.68‰ in Porto de Mós). Even so, the isotopic variability indicates a series of small oscillations, particularly in the mesosequential boundaries, that can be explained by primary mechanisms. In some cases, the stratigraphic comparison between the tendencies observed in each section can actually show evolutionary antagonisms. One example is the boundary from **MST2B** to **MST3**, where there is an increase from -3.01% (PC199) to -2.79% (PC202)

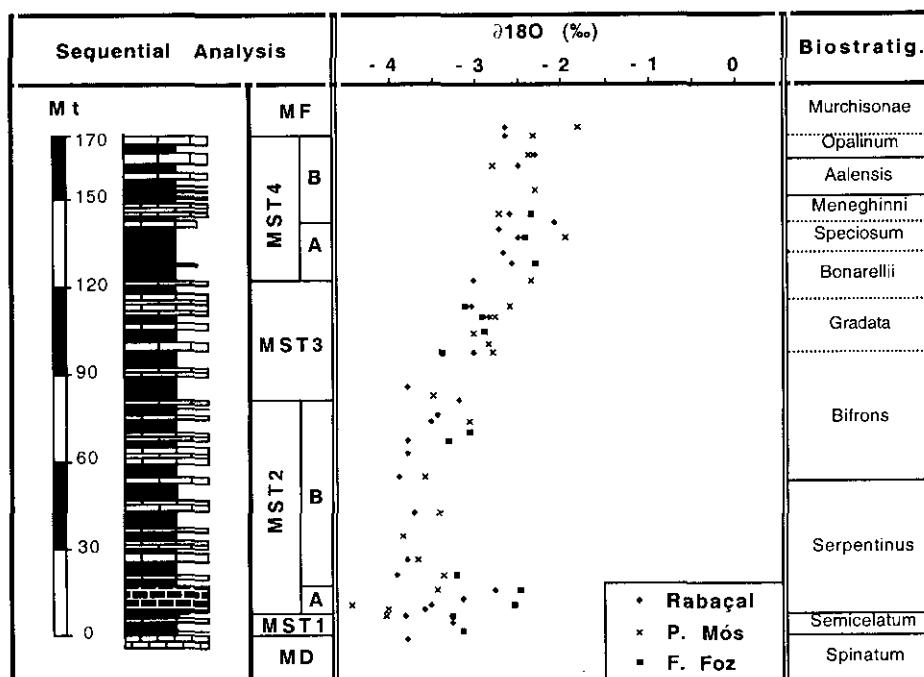


FIG. 9.— $\delta^{18}\text{O}$ distribution in the Toarcian-lower Aalenian from the Lusitanian Basin (synthetic section of Rabaçal).

FIG. 9.—Distribución del O. ¹⁸ durante el Toarcense-Aalenense inferior en la Cuenca Lusitaniense (sección sintética de Rabaçal).

in Porto de Mós (Table 8), and a reduction from -3.19% (Z50s) to -3.80% (Z51) in Rabaçal (Table 7, Fig. 8). Despite this, and taking into consideration only the Rabaçal section, which is objectively the most complete, the evolution of the isotopic data seems to closely agree, although inversely, with the $\delta^{13}\text{C}$ tendency: a gradual increase towards the top of the mesosequences with reduced variations between the boundaries.

DISCUSSION

The Origin of the Variations in $\delta^{13}\text{C}$: In sedimentary geology and particularly in the carbonate marine environment, $\delta^{13}\text{C}$ is one of the most frequently used palaeo-oceanographic and stratigraphic tools (Letolle & Renard, 1980; Cavelier *et al.*, 1981; Renard, 1984, 1986; Shackleton & Hall, 1984; Weissert, 1989; Emmanuel & Renard, 1993). We know about its relationship with primary productivity at the surface, with the oxidation of organic matter in

deeper waters (Kroopnick, 1985) and also its importance in the explanation of physiographic variations of carbonate platforms (Follmi *et al.*, 1994). Furthermore, the idea that $\delta^{13}\text{C}$ may be a direct indicator of palaeodepth in a pelagic environment (Renard & Letolle, 1983), and therefore able to modify the eustatic movement cycles, is an important reference in this study.

The association between $\delta^{13}\text{C}$ evolution in the toarcian marly-limestone of the Lusitanian Basin and the changes in sea-level is justified by the meaning of the sedimentary facies and their own vertical organisation. The variation curves for the three sections clearly show concordant tendencies for the various mesosequences. The large abrupt positive fluctuations tend to occur in the base of the **MST1**, **MST2B**, **MST3** and **MF** sequences, interpreted as flooding periods.

The variations of $\delta^{13}\text{C}$ are normally interpreted as being a consequence of two factors: the burial of organic carbon and/or through the productivity variations at the surface of the water. The link between the observed variations and the sequential analysis previously established, should indicate a mechanism of a primary nature. The possibility of carbon transfer, fixed to the organic matter of marine sediments, to oceanic waters, as a consequence of its degradation and oxidation (Weissert, 1989) does not seem to agree with the low levels of organic matter present in the sediments (data from Baudin, 1989) and does not justify the sometimes very high values of $\delta^{13}\text{C}$. Furthermore, the productivity factor does not seem to be sufficient to justify the large negative incursions, as the abrupt enrichment in ^{12}C cannot be due to a simple break in productivity.

One of the most simple possibilities, considering the link between the large isotopic variations and the sequential boundaries, is the influence of continental landmass on deposition, considerably richer in ^{12}C than marine water. We prove this idea with **MST2A**. Indicating sedimentation in a shallow ramp environment, with strong landmass influences, is in this sequence where the lowest values of $\delta^{13}\text{C}$ are observed (close to 0‰). With the exception of this unit, we can observe in each mesosequential unit, a gradual evolution of the values, with a tendency for ^{12}C enrichment (and subsequent reduction of $\delta^{13}\text{C}$). The regressive nature of each mesosequence indicates a larger continental landmass influence in the deposition (Duarte, 1995b). On the other hand, the positive anomalies observed in the base of **MST1**, **MST2B**, **MST3** and **MF**, resulting from faster transgressive effects, seem to imply the opposite.

Therefore, the results confirm the idea put forward by Letolle & Renard (1980), that the behaviour of $\delta^{13}\text{C}$ in pelagic carbonates is an indicator of palaeodepth: positive oscillations occur during transgressive events; the opposite occurring during regressive phases.

It is also possible that the differentiation of the $\delta^{13}\text{C}$ amplitudes observed between the three sections may be explained by local variations in the physiography of the sea bottom. In Porto de Mós it is visible via the facies (lithologi-

cal and biological ones), a sedimentological change which is more evident between the mesosequences than in the other locations (Rabaçal and Figueira da Foz). The definition of a unique palaeogeographic position in this region (DUARTE, 1995b), indicating a larger topographic platform gradient, can also be explained by the larger vertical variability of $\delta^{13}\text{C}$.

Diagenetic Influence: The idea that $\delta^{13}\text{C}$ is not very distorted by diagenetic processes (burial depth and recrystallisation), its thermodependence being low (Keith & Weber, 1964; Renard, 1984) is clearly confirmed in the Rabaçal section. Analyses carried out in a same level (Table 7) of this section, in the carbonate sediment of the dominating lithofacies (Z70S2-1) and the recrystallised sediments of a lateral side of a mud-mound (Z70S2-2), gave exactly the same value: +1.89%. This does not occur with $\delta^{18}\text{O}$ where there is a reduction in the recrystallised facies (from -2.59‰ to -4.70‰). The value of this sample corresponds to the lowest value obtained in the series, which shows the effects of thermic readjustments.

The Results in the European and Tethyan Realm: There have been numerous isotopic studies carried out on oxygen and carbon related to the toarcian series. The anoxic series of the lower Toarcian of Central and Northern Europe (Kauffman, 1978; Jenkyns, 1985, 1988) have sparked an increased interest in this field, particularly in the determination of $\delta^{13}\text{C}$, both in organic and carbonate composites. Analysing the information available in a carbonate phase (Jenkyns & Clayton, 1986; Baudin, 1989; Jenkyns *et al.*, 1991; Jiménez *et al.*, 1996), and removing the series localised in those particular depositional contexts (for example the Paris Basin, *in* Baudin, 1989), it is possible to show that the highest values of $\delta^{13}\text{C}$ can be found in the Falciferum zone (generally above +3‰), with a tendency to gradually reduce up to the Bifrons zone (about +2‰). This type of evolution can be seen in all the Lusitanian basin sections, where the largest positive incursions (over +4% in Rabaçal and Porto de Mós) are centered in the Serpentinus Zone (= Falciferum), in the alternating marl/limestone facies of the base of **MST2B** (Fig. 8). This increase may even reach +3%, after showing values very close to 0% in the base of the Serpentinus Zone, in a sedimentary context of a shallow ramp (**MST2A**). In the rest of the column, from the Bifrons Zone onwards, the trend is that of a gradual reduction of these values, a type of evolution that is also observed in the whole Tethyan Realm (Jenkyns & Clayton, 1986). Taking into consideration the key position of the Lusitanian Basin in palaeogeographical and biogeographical terms, the $\delta^{13}\text{C}$ results confirm an apparently global effect, at least on the tethyan scale and for the Toarcian Stage.

CONCLUSIONS

The type of analysis outlined in this study shows the importance of some sedimentary parameters as stratigraphic markers in the toarcian carbonates of

the Lusitanian Basin. The chart shown in Fig. 10 summarises the principal evolutionary tendencies of those variables, constituting strong arguments in the palaeoenvironmental discussion of the diverse sedimentary infilling phases (discussion in Duarte, 1995b).

1) Clay minerals indicate primary differentiations in deposition, both in vertical and lateral terms. It is possible to conclude the following:

- illite is practically exclusive in the sets from **MST3** onwards; its presence is ubiquitous in all mineralogical associations;
- kaolinite is practically constant in the base, disappearing throughout the whole basin at the top of the series (**MST4**);
- important contributions of smectite in **MST2A** and the base of **MST2B**, that highlight its importance as a stratigraphic marker (base of the Serpentinus zone);
- facies with chlorite in the mineralogical associations of **MST2B** and from **MST4A** onwards in the western sectors (F. Foz and S. Pedro de Muel); there is also an appreciable value of chlorite in the lower and middle Toarcian of Peniche (**MSTP1**, **MSTP2A** and **MSTP2B**). This evidence supports the idea that the origin of this mineral is from hercynian blocks which were probably emerged, situated in the western border of the basin.

2) The distribution of strontium shows good sequential control, and consequently, good stratigraphic control, which is interesting but difficult to interpret. High values observed in **MST2A** may indicate conditions of higher salinity levels. This may be associated with the proximity of a slightly evaporitic margin, as the sedimentary context and the association of the facies of that unit demonstrate a very shallow carbonate ramp (Duarte, 1995b).

3) The palaeobathymetric evolution of the series, judged from the study of these parameters and from sequential evolution, seems to identify itself best with the behaviour of $\delta^{13}\text{C}$. This element shows a close correlation with the relative sea-level changes: its highest increases, which are almost always abrupt, accompany the flooding phases of sedimentation; negative tendencies developed along the length of the mesosequences, indicate typically regressive evolution.

The stratigraphic distribution of $\delta^{13}\text{C}$ in the Toarcian-lower Aalenian series of the Lusitanian Basin is similar to that observed in other tethyan basins.

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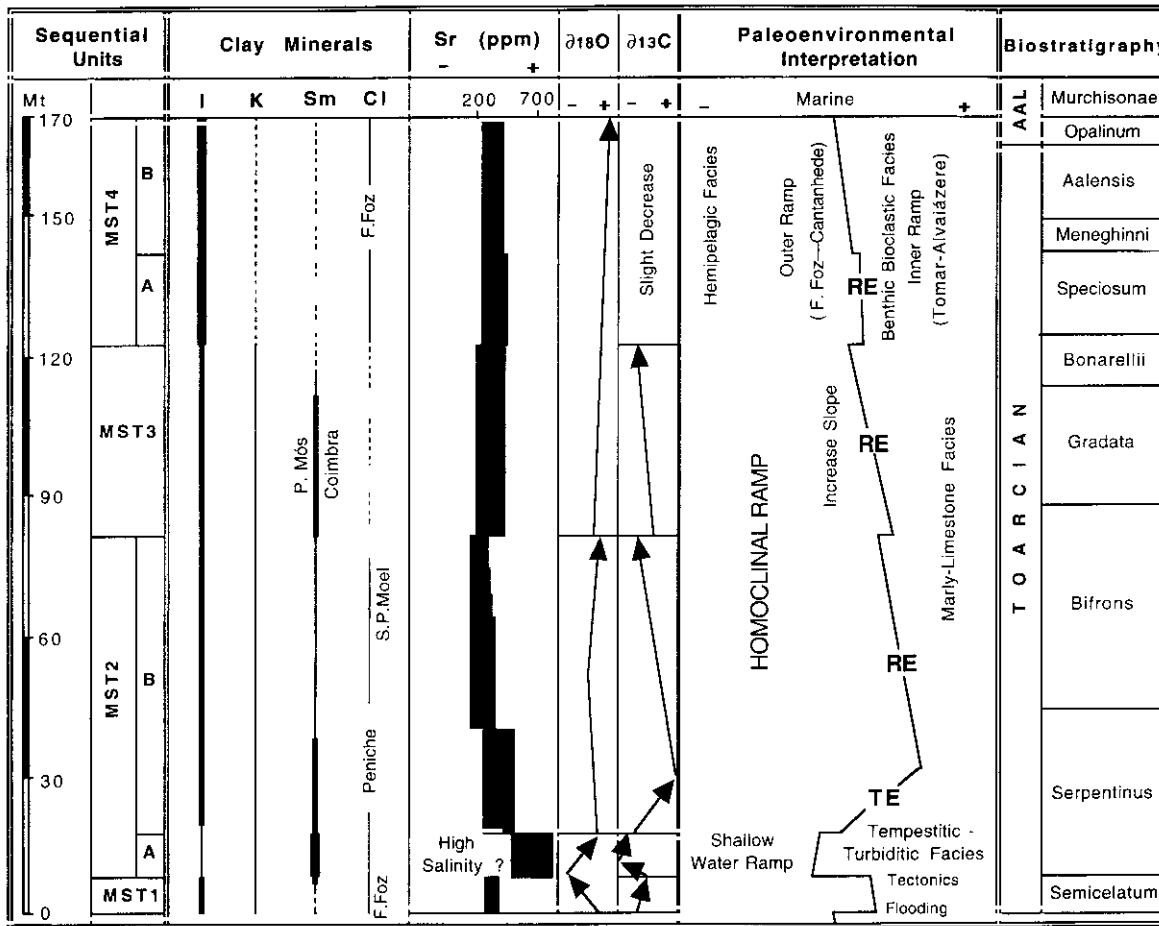


Fig. 10- Sedimentary evolution in the Toarcian-lower Aalenian series of the Lusitanian Basin. Clay minerals: I - Illite, K - Kaolinite, Sm - Smectite, Cl - Chlorite. RE - Regressive evolution ; TE - Transgressive evolution.

Fig. 10- Evolución sedimentaria de la sucesión del Toarciano-Aaleniano inferior en la Cuenca Lusitana. Minerales de arcilla: I: Illita, K: Caolinita, Sm: Esmectita, Cl: Clorita, RE: Evolución regresiva, TE: Evolución transgresiva.

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