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Carbon isotopes as a sequence stratigraphic tool: examples from the Lower and Middle Toarcian marly limestones of Portugal

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ABSTRACT

The Lower and Middle Toarcian series of the Lusitanian Basin (Portugal) are generally characterized by hemipelagic marl-limestone alternations, included in the S. Gião Formation. The succession is informally organized into three 3rd order depositional sequences (ST1 to ST3), each bounded by regional discontinuities. These depositional sequences are characterized in terms of vertical and lateral litho- and biofacies changes. Using accurate ammonite biostratigraphic control, sequence boundaries are assigned to the lowermost *polymorphum* Zone (SBT1), *polymorphum-levisoni* Zone boundary (SBT2), intra-*bifrons* Zone (SBT3) and uppermost *bonarelli* Zone (SBT4). Maximum flooding intervals are identified in the uppermost *polymorphum* (ST1), uppermost *levisoni* (ST2) and uppermost *bifrons* (ST3) Zones. The uppermost part of *levisoni* Zone is a 2nd-order maximum transgressive phase. Carbon isotope data from bulk carbonate samples of selected sections in the Lusitanian Basin (Rabaçal, Porto de Mós, Coimbra and Figueira da Foz) reveal this analytical technique as good indicator of 3rd order sequence evolution. Positive $\delta^{13}C$ excursions correspond to transgressive (transgressive systems tracts) phases whereas negative trends/excursions are related to periods of regression (lowstand and highstand systems tracts). Furthermore, the global $\delta^{13}C$ events recognised in the Early Toarcian sections of several North-European and Tethyan basins are observed in all the studied sections of Lusitanian Basin.

Key words: Carbon isotopes, marl-limestone alternations, Portugal, stratigraphic sequences, Toarcian.

Los isótopos de carbono como herramienta de análisis de una secuencia estratigráfica: ejemplos de las calizas margosas del Toarciense Inferior y Medio de Portugal

RESUMEN

Las series del Toarciense Inferior y Medio de la Cuenca lusitánica (Portugal) se caracterizan generalmente por la existencia de alternancias de margocalizas hemipelágicas, incluidas en la Formación S. Giao. La sucesión está organizada informalmente en tres secuencias deposicionales de tercer orden (ST1 a ST3), limitadas por discontinuidades regionales. Estas secuencias deposicionales están caracterizadas por cambios verticales y laterales en las lito y biofacies. Utilizando un control bioestratigráfico preciso mediante ammonites, los límites de la secuencia son asignadas a la parte inferior de la zona polymorphum *(SBT1), el límite de la zona* polymorphum-levisoni *(SBT2), y las partes superiores de las zonas* levisoni *(ST2) y* bifrons *(ST3). La parte superior de la zona* levisoni *es una fase de máximo transgresivo de segundo orden. Los datos de los isótopos de Carbono procedentes de muestras de carbonato total de algunas secciones de la Cuenca Lusitánica (Rabaçal, Porto de Mós, Coimbra y Figueira da Foz) revelan que esta técnica analítica es un buen indicador de la evolución de la secuencia de tercer orden. Las excusiones positivas de* δ^3C *corresponden a fases transgresivas* (transgressive systems tracts), *mientras que las excursiones y tendencias negativas se relacionan con períodos de regresión* (lowstand y highstand systems tracts). *Además, los eventos globales de* δ^3C *reconocidos en las secciones del Toarciense Inferior de algunas cuencas Norte-Europeas y del Tethys, han sido observados en todas las secciones estudiadas en la Cuenca lusitánica.*

Palabras clave: alternancias de margocalizas, Isótopos del Carbono, Portugal, secuencias estratigráficas, Toarcense.

Introduction

In Portugal, Early Jurassic age sediments crop out in two different geo-structural settings, both bounded by the Iberian Variscan Massif: the Algarve Basin, in the south of Portugal and in the Lusitanian Basin, located in west-central Portugal. In the Lusitanian Basin (LB), the Toarcian is generally dominated by marl/argillaceous limestone and limestone alternations, usually characterised by a rich and diverse nektonic and benthic macrofauna (Duarte, 1997; Duarte *et al.*, 2001, 2004a).

The hemipelagic nature of the Toarcian sediments in the LB and the accurate biostratigraphic control, constitute a good context for carbon and oxygen isotope studies. This kind of pelagic sediment is suitable for using isotopes as palaeoceanographic/palaeoclimatic indicators (e.g. Schölle and Arthur, 1980; Weissert, 1989; Follmi *et al.*, 1994; Weissert *et al.*, 1998; Kump and Arthur, 1999; Jenkyns *et al.*, 2002). Carbon isotopes have proven a powerful stratigraphic tool in the study of the Toarcian of some European and Tethyan regions (Jenkyns and Clayton, 1986, 1997; Jiménez *et al.*, 1996; Röhl *et al.,* 2001; Rosales *et al.*, 2001; Jenkyns *et al.,* 2002; Schmid-Röhl *et al.,* 2002; Mattioli *et al.*, 2004, among others).

The record of stable isotopes in the Toarcian sediments of the LB also proves this parameter as a useful tool in sequence stratigraphy analysis, in particular for the definition of 3rd order sequences. Thus, the aim of this paper is to characterize the depositional systems in the whole LB during the Early and Middle Toarcian in terms of sequence stratigraphy and carbon isotope data, improving and extending some isotopic data before published in Duarte (1998).

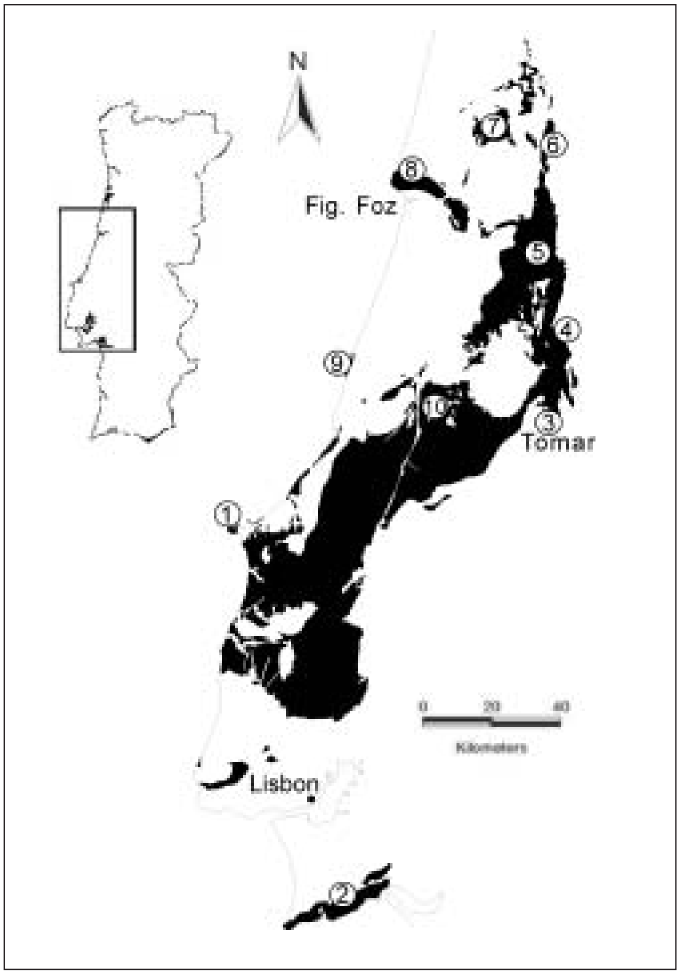

Fig. 1 - Geological map of the Jurassic in the Lusitanian Basin. Location of the main Lower Jurassic carbonate outcrops and studied sections (Rabaçal, Porto de Mós, Coimbra and Figueira da Foz): 1 – Peniche; 2 – Arrábida-Sesimbra; 3 – Tomar; 4 – Alvaiázere; 5 – Rabaçal; 6 – Coimbra; 7 – Cantanhede; 8 – Figueira da Foz; 9 – S. Pedro de Moel; 10 – Porto de Mós

Fig. 1 - Mapa Geológico del Jurásico en la cuenca de Lusitania. Situación de los principales afloramientos de carbonatos del Jurásico Inferior y de los cortes estudiados (Rabaçal, Porto de Mós, Coimbra y Figueira de For): 1.- Peniche; 2.- Arrábida-Sesimbra; 3-Tomar, 4.- Alvaiázere; 5.- Rabaçal; 6.- Coimbra; 7.- Cantanhede; 8.- Figueira de Foz; 9-S. Pedro de Moel; 10.- Porto de Mós

Geological context and lithostratigraphy

The studied successions of Toarcian age are included in the Late Triassic (Norian?)-Callovian cycle of Wilson *et al.* (1989). In this cycle, the Pliensbachian and Toarcian sediments correspond to the maximum transgressive facies interval (Soares *et al.*, 1993).

Throughout the basin, Toarcian hemipelagic deposits contain a rich nektonic (ammonites and belemnites) and benthic (mainly brachiopods, bivalves, crinoids and siliceous sponges) macrofauna. The integration of several stratigraphic/sedimentological analyses (e.g. lithofacies analysis, microfacies, sequential evolution and palaeontological evolution) indicates that deposition occurred on a carbonate ramp (homoclinal ramp *sensu* Read, 1982, 1985) dipping towards the northwest (Duarte 1997; Duarte *et al.,* 2001). In the LB the Toarcian is well exposed, including the two likely depth extremes of the basin: Figueira da Foz-Cantanhede at the north end and Arrábida-Sesimbra at the south end (Figure 1).

Stratigraphic Chart

Excluding the Arrábida-Sesimbra, Tomar and Peniche sectors (Figure 1 and 2), the Toarcian successions of LB are very rich in ammonites and calcareous nannofossils, allowing good biostratigraphic control of the series (Elmi *et al.*, 1989; Rocha *et al.*, 1987, 1996; Perilli and Duarte, 2006). The studied succession belongs to the S. Gião Formation (Duarte and Soares, 2002), a marl dominated unit that ranges in age from the Early Toarcian (*polymorphum* Zone) up to the Late Toarcian (base of *meneghinni*

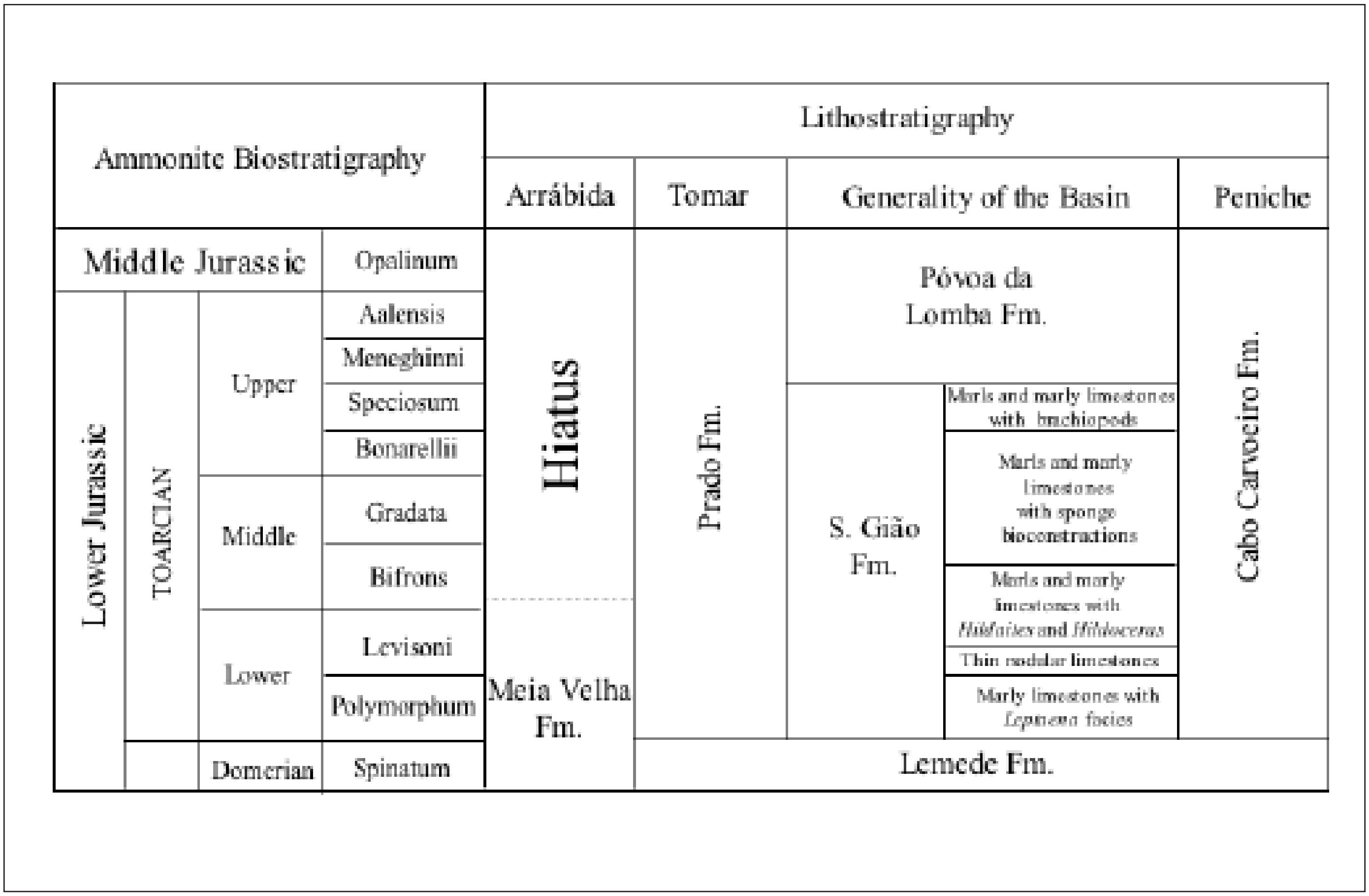

Fig. 2 - Biostratigraphic and lithostratigraphic chart of Toarcian successions in the Lusitanian Basin. Ammonite biostratigraphy from Rocha *et al.* (1996)
Fig. 2 - Columna bioestratigráfica y litoestratigráfico de las series Toarcienses de la Cuenca Lusitana. La bioestratigrafía de los amonitos según Rocha et al. *(1996)*

Zone). This unit occurs between the Lemede (Late Pliensbachian) and Póvoa da Lomba (Late Toarcian to Early Bajocian) Formations.

The S. Gião Formation can be subdivided into five informal members (Figures 2 and 3), easily recognizable across a large area of the LB (Duarte and Soares, 2002): Marly limestones with *Leptaena* facies Member (MLLF), Thin Nodular Limestones Member (TNL), Marls and marly limestones with Hildaites and Hildoceras Member (MMLHH), Marls and marly limestones with sponge bioconstructions Member (MMLSB) and Marls and marly limestones with brachiopods Member (MMLB). Generally, the thickness of each unit increases from southeast (Tomar region) towards the northwest (Figueira da Foz sector), following the direction of the dipping ramp. With the exception of Arrábida-Sesimbra sector, the ammonite records suggest that no important sedimentary gap is present in the Toarcian series of the LB.

Methods

Studied Sections

The main sections of Toarcian in the LB have been previously studied, bed-by-bed, in terms of an integrated stratigraphic analysis and sedimentary characterization (Duarte, 1997; Duarte *et al.*, 2001, 2004a). In terms of geochemical analysis four sections were selected on the basis of relative vertical continuity, accurate biostratigraphic control, and palaeogeographic context, all cropping out in northern and central part of the basin where the Toarcian is thickest and developed in hemipelagic facies (Figures 1 and 3).

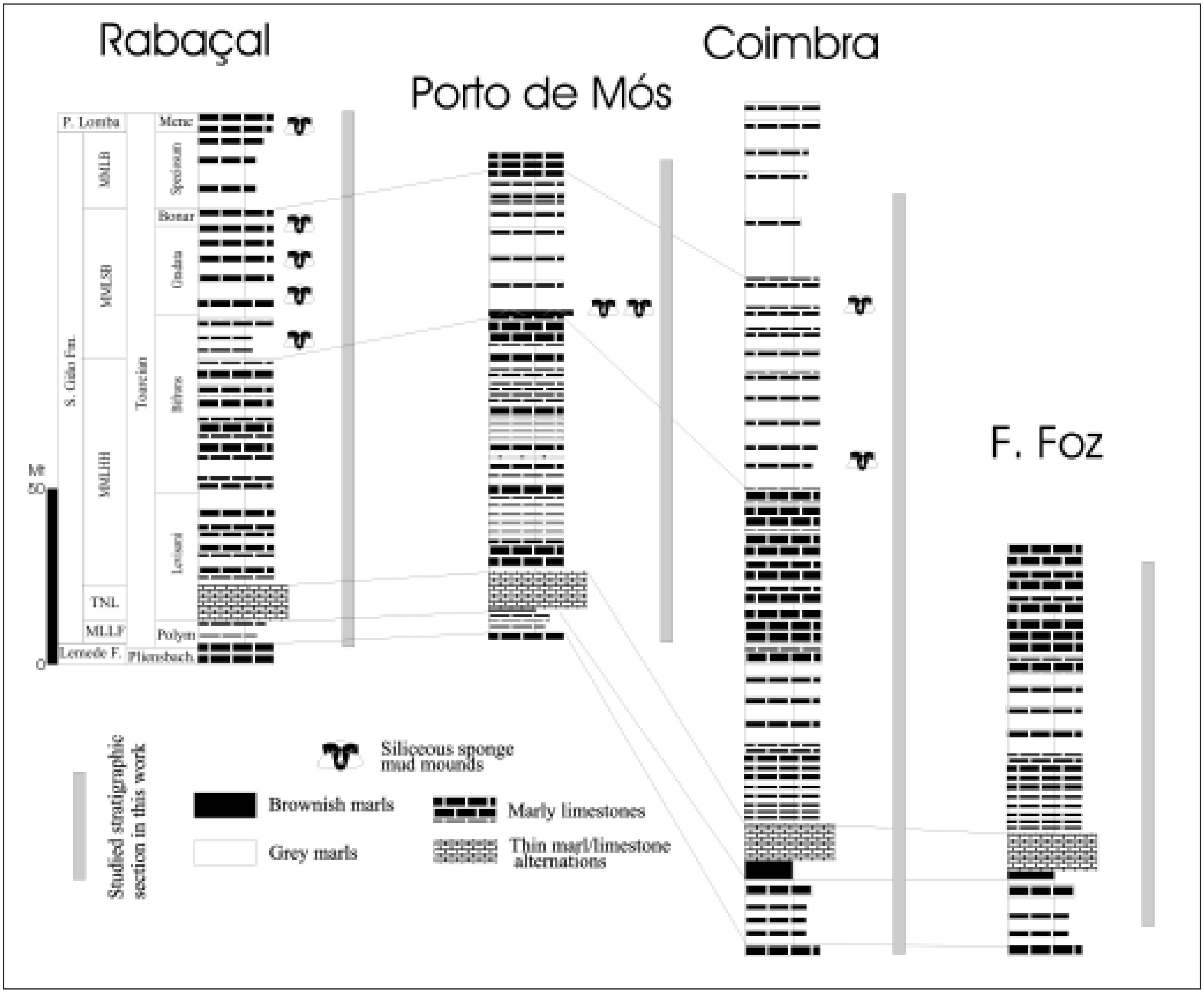

Fig. 3 – Synthetic logs, facies and stratigraphic variations of the Lower and Middle Toarcian successions (S. Gião Formation) in the four studied sectors (based on Duarte, 1997; Duarte and Soares, 2002)
Fig. 3 - Columnas sintéticas, facis y variaciones estratigráficas de las series del Toarmense inferior y medio (formación S. Ciao) en los tres sectores estudiados (basados en Duarte, 1997; Duarte y Soares, 2002)

Rabaçal Section: This constitutes the reference section for the Toarcian marly limestone facies of the LB due to its excellent exposure and relatively intensive investigation (Mouterde *et al.*, 1964/65; Duarte 1995, 1998, 2004; Duarte and Soares, 2002, and references therein). Rabaçal constitutes the type-locality of S. Gião Formation.

Porto de Mós composite-section: This is composed of the Porto de Mós I and Penas do Castelo subsections (Duarte, 1995, 1998). Only the middle part of the MMLHH (base of *bifrons* Zone; Mouterde and Ruget, 1967) is not sampled.

Coimbra composite-section: In this area, the Toarcian deposits are widespread and the succession expanded, showing well-developed hemipelagic characteristics (Duarte *et al.*, 2004a). This section is composed by the following five sub-sections: Ribeiro, Fornos, Marmeleira, Cabeço da Azevêda and S. Simão (Duarte, 1995). Only part of MMLHH/MMLSB boundary (part of *bifrons* Zone) and the middle part of MMLSB (top of *gradata* Zone) have not been studied and sampled.

Figueira da Foz composite-section: This constitutes the least studied sector of the LB, composed by

Vale das Fontes and Brenha sub-sections (Duarte, 1995). Exposure conditions restrict observations and sampling to the MLLF, TNL and middle part of MMLHH.

Carbon and Oxygen Isotopes

To investigate $\delta^{13}C$ and $\delta^{18}O$, 109 samples of bulk-rock micritic carbonate sediments from calcareous levels (with more than 60% of carbonate) belonging to the Lower and Middle Toarcian units of Coimbra and Figueira da Foz (the two most distal sectors of LB) were analysed. Although belemnites and brachiopods are particularly rich in some units of the Toarcian, they are not present in the whole succession, which prevents construction of a continuous isotopic curve based upon these particular taxa.

All rock samples were pulverized in porcelain dish to obtain particles smaller than 80 mesh. The oxygen and carbon isotopic values were obtained from individual samples using a Kiel Carbonate Device III coupled to a MAT 252 *ThermoFinnigan* mass spectrometer at Petrobras Research Center (Cenpes, Brazil). This process uses approximately 10 mg of sample, which was placed in a vacuum chamber and reacted with phosphoric acid for 6 minutes at 70°C. The water and carbon dioxide generated were captured at -170°C with liquid nitrogen. Then, this mixture was brought to -110°C when the subsequently released pure CO_2 was again collected at -170°C with liquid nitrogen. All results are reported in parts per mil (‰) notation (δ) relative to Peedee Belemnite international standard (PDB; Craig, 1957) and calibrated by routine preparation and analysis of the carbonate standard NBS-19 ($\delta^{13}C$=1.95‰, $\delta^{18}O$= -2.20‰). Day to day precision ($\pm$ 1 δ) based on NBS-19 analyses was $\pm$ 0.05 for $\delta^{13}C$ and $\pm$ 0.08 for $\delta^{18}O$.

We also include in this work other available isotope data previously published in Duarte (1998) from Rabaçal (22 samples) and Porto de Mós (24 samples) sections.

Results: Carbon and oxigen isotope stratigraphy

Carbon Isotopes

In the newly studied sections, the $\delta^{13}C$ data, presented in the Tables I and II, range between -0.87‰ and +4.02‰, results that are consistent with normal values for marine carbonate sediments. However, significant oscillations are observed between each stratigraphic unit, such as zone, member or deposi-

Sample	$\delta^{13}C$ (‰ PDB)	$\delta^{18}O$(‰ PDB)	Unit
R0	0.65	-3.48	Lemede Fm.
R1	0.49	-4.04	Lemede Fm.
R3	0.76	-3.66	Lemede Fm.
R6	0.56	-3.48	MLLF
R9	0.56	-3.43	MLLF
R11	0.98	-3.21	MLLF
R13	0.91	-2.78	MLLF
R15	1.32	-3.56	MLLF
R18	1.41	-3.63	MLLF
R25	1.10	-3.78	MLLF
R26	1.16	-3.70	MLLF
R31	1.52	-3.95	MLLF
R33	1.26	-4.43	MLLF
R41	0.94	-4.45	MLLF
R43	1.94	-3.76	MLLF
R46	1.63	-3.74	MLLF
R48	1.65	-3.83	MLLF
R50	2.15	-3.79	MLLF
R52	2.08	-3.80	MLLF
R54	2.44	-3.42	MLLF
R56	2.60	-3.79	MLLF
R59	-0.87	-4.48	TNL
R60	-0.24	-3.19	TNL
LOR2	0.06	-3.70	TNL
EP249	0.20	-2.75	TNL
VLR8	0.31	-3.68	MMLHH
VLR10	1.49	-3.61	MMLHH
VLR24	1.53	-4.06	MMLHH
EP336	1.70	-2.78	MMLHH
VLR26	2.05	-2.87	MMLHH
VLR27	2.19	-3.82	MMLHH
Malt1	2.88	-4.30	MMLHH
SA9	3.88	-3.55	MMLHH
SA15	3.93	-3.78	MMLHH
Malt2	4.02	-3.95	MMLHH
Malt3	3.87	-3.64	MMLHH
Malt4	3.88	-3.53	MMLHH
MM33	2.96	-3.65	MMLHH
MM34T	2.83	-3.68	MMLHH
MM40	2.84	-3.77	MMLHH
MM43	2.68	-3.28	MMLHH
MM44	3.02	-3.58	MMLHH
MM46A	2.68	-3.81	MMLHH
MM48A	2.79	-3.87	MMLHH
MM48B	2.93	-3.61	MMLHH
MM50	3.10	-3.41	MMLHH
MM54	2.73	-3.76	MMLHH
MM56B	2.66	-3.88	MMLHH
MM58	2.24	-4.57	MMLHH
MM64	2.66	-3.59	MMLHH
MM69	1.37	-3.46	MMLHH
MM71	2.21	-3.37	MMLHH
MM72	2.02	-3.53	MMLHH
MM77	2.38	-3.55	MMLHH
MM80	1.99	-3.72	MMLHH
MM84	1.59	-3.30	MMLHH
MM91	2.01	-3.27	MMLHH
MM95	1.79	-4.48	MMLHH
MB01	1.89	-3.00	MMLSB
MB06	1.92	-3.64	MMLSB
MB11	1.69	-2.99	MMLSB
MB13	1.55	-2.98	MMLSB
MB16	1.65	-3.60	MMLSB
MB35	1.80	-3.28	MMLSB
MB40B	1.83	-3.04	MMLSB
MB42	1.68	-3.25	MMLSB
MB44	1.43	-3.29	MMLSB
MB57	1.63	-3.49	MMLSB
MB67	1.46	-3.18	MMLSB
MB74	1.41	-3.15	MMLSB
MB76	1.84	-2.95	MMLSB
SS20	0.68	-4.99	MMLSB
SS22	1.24	-2.98	MMLSB
SS24	1.44	-3.38	MMLSB
SS26	1.11	-5.88	MMLSB
SS28	0.91	-2.25	MMLSB
SS32	0.80	-3.46	MMLSB
SS38	0.48	-4.76	MMLSB
SS45	1.19	-3.56	MMLB
SS50	1.00	-3.46	MMLB

Table I – Isotopic data of the studied sections from Lower and Middle Toarcian in the Coimbra sector
Tabla I - Datos isotópicos de los cortes estudiados del Toarciense inferior y medio en el Sector de Coimbra

Sample	$\delta^{13}C$ (‰ PDB)	$\delta^{18}O$(‰ PDB)	Unit
VF 04	1.00	-3.94	MLLF
VF 12	1.33	-3.66	MLLF
VF 18	1.96	-3.53	MLLF
VF 23	1.94	-3.68	MLLF
VF 26	1.64	-3.71	MLLF
VF 28	2.15	-3.58	MLLF
VF 30	2.02	-3.68	MLLF
VF 31	1.94	-3.45	MLLF
VF 33	1.96	-3.70	MLLF
VF 34	1.28	-3.70	MLLF
B 6	-0.25	-3.09	TNL
B 38	0.05	-3.10	TNL
BP 68	0.12	-2.85	TNL
BP 86	-0.06	-3.18	TNL
BP 138	0.15	-2.92	TNL
BP 254	2.04	-3.57	MMLHH
BP 300	2.95	-3.22	MMLHH
BP 301	2.36	-3.16	MMLHH
BP 304	2.60	-3.47	MMLHH
BP 308	2.31	-3.52	MMLHH
BP 309	2.62	-3.80	MMLHH
BP 312	2.48	-3.54	MMLHH
BP 313	-0.36	-3.12	MMLHH
BP 314	1.29	-3.61	MMLHH
BP 315	2.42	-3.52	MMLHH
BP 316	2.09	-3.56	MMLHH
BP 318	2.46	-3.44	MMLHH
BP 321	2.30	-3.15	MMLHH
BP 322	1.05	-2.95	MMLHH

Table II – Isotopic data of the studied sections from Lower and Middle Toarcian in the Figueira da Foz sector
Tabla II - Datos isotópicos de los cortes estudiados del Toarciense inferior y medio del sector de Figueira da Foz

tional sequence, supporting the use of $\delta^{13}C$ as a stratigraphic tool in the Lower and Middle Toarcian studies of the LB.

Lowest values of $\delta^{13}C$ are observed at the base of *levisoni* Zone (TNL), where the $\delta^{13}C$ signal varies between -0.87‰ in Coimbra and -0.25‰ in Figueira da Foz (1.08‰ in Rabaçal and 0.20‰ in Porto de Mós). Highest values are observed in the middle part of *levisoni* Zone, in the base of MMLHH, showing values above 4‰, with 4.02‰ in Coimbra (4.29‰ in Porto de Mós). The highest value of 2.95‰ observed in the Figueira da Foz sector is recorded in the extremely base of *bifrons* Zone perhaps because the middle-upper part of *levisoni* Zone is not observed. Isotopic curves illustrated in Figures 4 to 7 allow the following stratigraphic observations:

MLLF, dated as *polymorphum* Zone, shows a clear positive trend in Coimbra and Figueira da Foz sectors. This trend is particularly well observed in the composite-section of Coimbra, where $\delta^{13}C$ shows an important excursion in the middle part of this member, from values below 1‰ reaching at the top 2.6‰ (1‰ to 2.15‰ in Figueira da Foz).

The greatest Toarcian changes in $\delta^{13}C$ occur between the TNL and the underlying and overlying units MLLF and MMLHH, showing that the TNL has the lowest values of the whole series. The boundary between MLLF and TNL shows an abrupt drop (from 2.6‰ to -0.87‰ in Coimbra; 2.15‰ to -0.25‰ in Figueira da Foz).

The MMLHH shows a large increase at the base (middle part of *levisoni* Zone) of this unit, followed by a continuous and gradual drop, towards the top of the unit (*bifrons* Zone), to carbon isotope values of 1.71‰ in Rabaçal, 1.79‰ in Coimbra and 1.11‰ in Porto de Mós.

Despite the differences in absolute carbon isotope values between each section observed in the MMLSB (uppermost *bifrons* Zone to *bonarellii* Zone), this interval clearly show a negative trend. The Coimbra sector shows an evolution from 1.92‰ to 0.48‰, a similar difference to the evolution occurred in Porto de Mós (2.54‰ to 1.47‰) and Rabaçal (2.52‰ to 1.7‰).

Although not analysed in detail, the MMLB shows at the base (*speciosum* Zone) a small increase in the $\delta^{13}C$, compared to the underlying unit. These values range between 1.19‰ in Coimbra, 1.77‰ in Rabaçal and 2.05‰ in Porto de Mós.

Oxygen Isotopes

Oxygen isotopic results for the Lower and Middle Toarcian of LB show a wide range of values below -2.00‰, reaching values around -5.00‰. This large range and the negative values of $\delta^{18}O$ are interpreted here to be best explained by a diagenetic overprint (early cementation or burial diagenesis), due to the known thermodependence of oxygen isotopes. For this reason, these data are not included in the aims and discussion of this work.

Sequence stratigraphy

Toarcian 2^nd order Sequence: ST

The marly limestone of the Toarcian succession in the LB comprises marl-limestone alternations, in some cases highly rhythmic and organised into hierarchical sequences. Two scales of sequences (2^{nd} to 3^{rd} order; Vail *et al.*, 1991), equivalent to stratigraphic cycle terminology of Jacquin and De Graciansky (1998a,b), have been distinguished in the Toarcian series of the LB: 2^{nd} order transgressive-regressive (T-R) facies cycles and 3^{rd} order sequence cycles. The studied series are included in the 2^{nd} order sequence ST (Duarte *et al.*, 2004a,b). This sequence embraces the whole S. Gião Fm and the lowermost part of Póvoa da

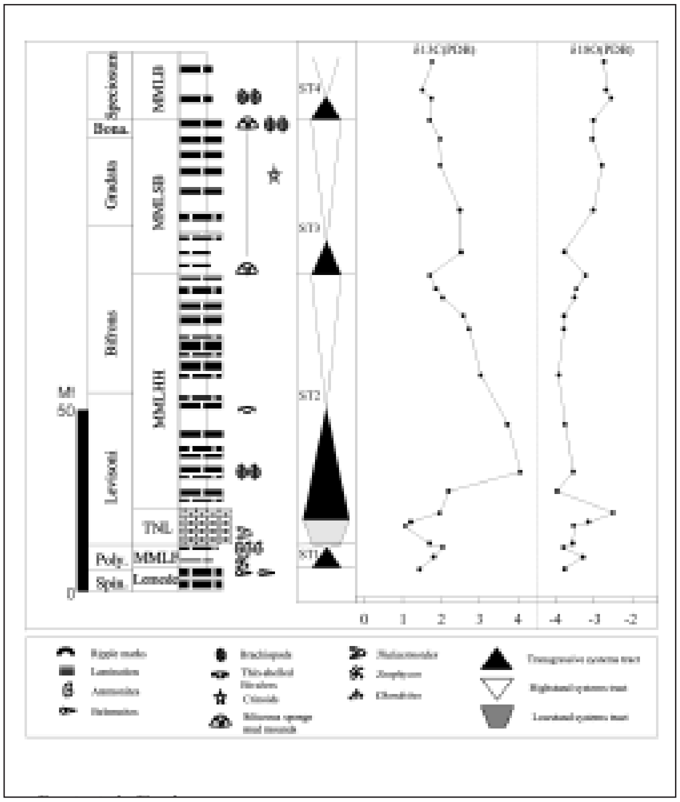

Fig. 4 - Sedimentology, sequence stratigraphy and isotope evolution of Lower to lowermost Upper Toarcian in the Rabaçal section. Isotopic data previously published in Duarte (1998). Key to the symbols used in the figure 3

Fig. 4 - Sedimentología, estratigrafía secuencial y evolución isotópica desde el Toarciense superior en el corte de Rabaçal. Los datos isotópicos han sido publicados con anterioridad en Duarte (1998). La leyenda de los símbolos utilizados está en la Figura 3

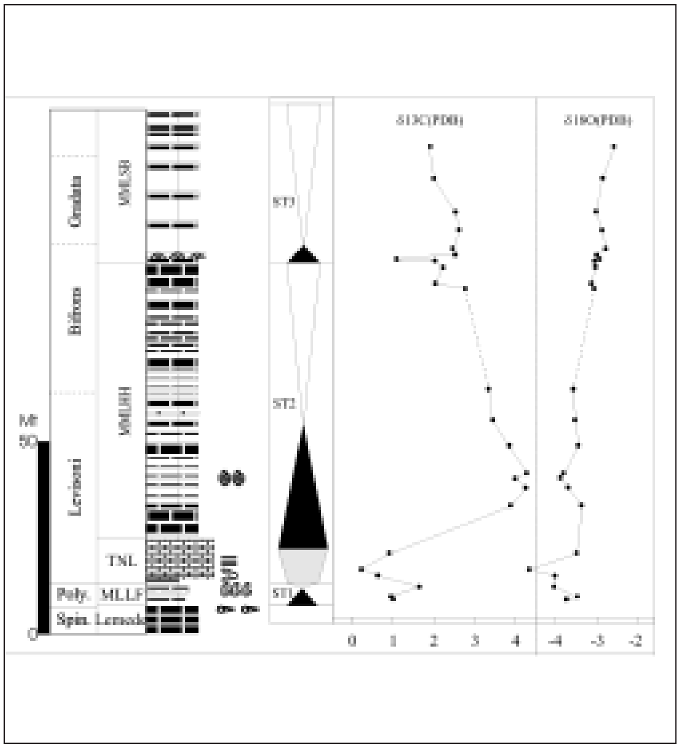

Fig. 5 - Sedimentology, sequence stratigraphy and isotope evolution of Lower to lowermost Upper Toarcian in the Porto de Mós composite-section. Isotopic data previously published in Duarte (1998). Key to the symbols used in the figures 3 and 4

Fig. 5 - Sedmentología, estratigrafía secuencial y evolución isotópica desde el Toarciense inferior a la parte basal del Toarciense superior en el corte compuesto del Porto de Mós. Los datos isotópicos han sido publicados previamente en Duarte (1998). La leyenda de los símbolos utilizados está en la Figura 3

Lomba Fm, varying between 75-80m in Tomar and around 280m thick in the Coimbra sector.

The base of ST, dated from the lowermost *polymorphum* Zone, corresponds to an abrupt flooding event, represented by marl accumulation in the whole basin. The marl dominance observed at the top of the *levisoni* Zone marks the peak transgression of the Toarcian 2nd order sequence (Duarte *et al.*, 2004a). The Middle Toarcian-Lower Aalenian succession shows a regressive trend, ending ST with an upward increase of calcareous and bioclastic content. The upper discontinuity is dated as *opalinum* Zone and shows different sedimentary records in the basin (DA1 in Duarte *et al.*, 2001).

Toarcian 3rd order Sequences

The Toarcian 2nd order sequence is subdivided into four 3rd order depositional sequences (ST1 to ST4; Figures 4 to 7), each one bounded by isochronous regional discontinuities (SBT1-SBT4; DT1-DT4 in Duarte, 1997), recognised over most of the LB (Figure 8). These sequence boundaries are dated of lowermost *polymorphum* Zone (SBT1), *polymorphum-levisoni* Zone boundary (SBT2), intra-*bifrons* Zone (SBT3) and uppermost *bonarelli* Zone (SBT4). Based on ammonite biostratigraphic data, and assuming ammonite zones of equal duration, the time span of each sequence agrees with the time range estimated by Vail *et al.* (1991) for the 3rd order depositional sequences (0.5 to 3 My).

Sequence ST1 (*polymorphum* Zone): In all the studied sections, the base of sequence ST is a clay-rich unit (MMLF), composed of decimetre- to metre-thick marl and centimetre-thick limestone alternations, very rich in benthic and nektonic macrofauna. Brachiopods (*Koninckella liasina*, *Nannirhynchia pyg-*

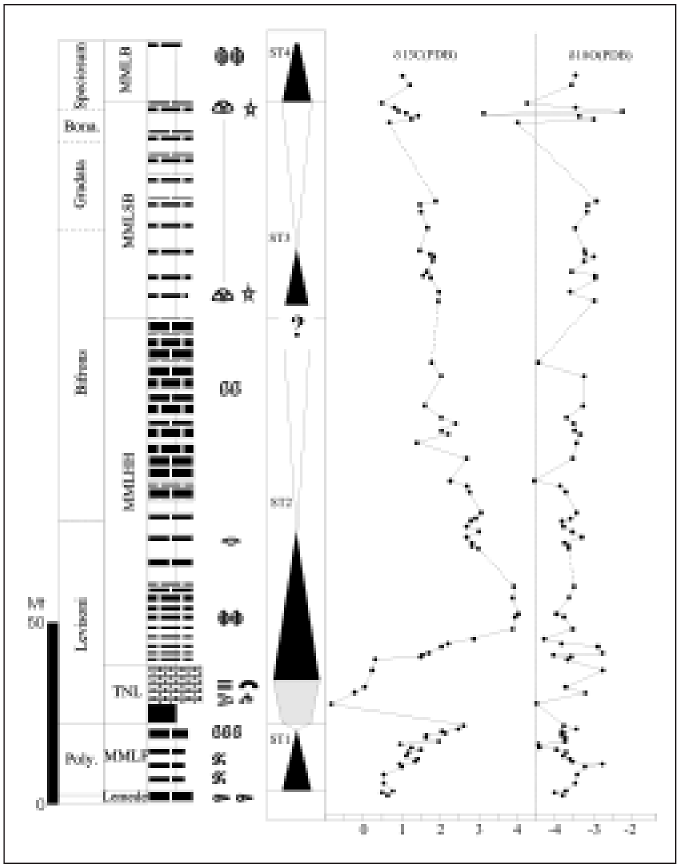

Fig. 6 - Sedimentology, sequence stratigraphy and isotope evolution of Lower to lowermost Upper Toarcian in the Coimbra composite-section. Key to the symbols used in the figures 3 and 4

Fig. 6 - Sedimentología, estratigrafía secuencial y evolución isotópica desde el Toarciense inferior a la parte basal del Toarciense superior en el corte compuesto de Coimbra. La leyenda de los símbolos utilizados están en las figura 3 y 4

moea, *Liospiriferina*), belemnites and ammonites are dominant and, in some levels, *Zoophycos* is very common, but benthic macrofauna decreases upward. This sequence increases in thickness towards north and west, reaching 20 m in Coimbra. The sedimentological features found across the *polymorphum* Zone of all the LB sectors suggest a clear deepening phase. ST1 is strongly asymmetric, and a large part of it is interpreted as a transgressive systems tract as a consequence of the increase of clay and the decrease of benthic macrofauna observed upwards. The maximum flooding surface probably corresponds to a thick limestone bed (0.50-0.80m), a condensed level of dactylioceratid ammonites (*Dactylioceras semicelatum* SIMPS), observed near the top of this sequence. This horizon is a good biologic event easily recognizable in the studied sections and in the major part of the LB.

Sequence ST2 (levisoni to bifrons Zones): One of the most marked sedimentary changes occurred in the LB during the Early and Middle Jurassic is observed between the *polymorphum* and *levisoni* Zones (Duarte 1997). This change shows different and particular sedimentological features in several points of the basin such as Peniche and Arrábida-Setúbal, related to tectonic control (Wright and Wilson 1984; Kullberg *et al*. 2001).

In the northern sector of the basin, the lowest part of ST2 shows 3-4 m of unfossiliferous brownish clays and marls. These facies, practically absent south of a line from Coimbra to Figueira da Foz, pass upwards to a thin succession (less than 10 m thick) of centimetre-thick alternations of marlstone, brownish claystone and micritic to microsparitic limestone (TNL). The large diversity of limestone facies includes silty to fine sandy limestones, with parallel and convolute lamination, and small-scale cross stratification (climbing ripples, wave ripples and low-angle cross bedding). The occurrence of the trace fossil *Thalassinoides* is a typical feature of the base of this unit. Integrating all the information about the base of this sequence it is possible to conclude that during the time equivalent to the base of *levisoni* Zone an important sea-level fall occurred, favoured by tectonic control. The shallowing is confirmed by the lithofacies nature of TNL, interpreted by Duarte and Soares (1993) and Duarte (1997) as a consequence of a tempestitic/turbiditic event. This tectonic phase is responsible for the western input of siliciclastics in the basin, particularly observed in Peniche (Wright & Wilson 1984; Duarte 1997).

The transgressive systems tract occurs in the whole studied area, through the decrease of siliciclastics and the increase of marls. This part of ST2, coincident with MMLHH, is composed of marl/limestone decimetre- to metre-thick alternations, particularly rich in rhynchonellid and terebratulid brachiopods (e. g. *Soaresirhynchia bouchardi* and *Telothyris jauberti*). This transgressive event ends before the progressive increase of calcareous components in the series. In sectors such as Coimbra and Figueira da Foz, this unit contains horizons rich in thin-shelled bivalve (*Bositra* sp.), pelagic evidence dated as the top of *levisoni* Zone. These horizons correspond to the maximum flooding interval of ST2 and to the peak transgression of the 2nd-order sequence ST. These levels are overlain by thick aggrading/prograding package. In the whole basin, the series becomes progressively more calcareous evidencing a shallowing-upward evolution (highstand systems tract).

Sequence ST3 (bifrons Subzone – bonarelli/speciosum Zone transition): This sequence corresponds to

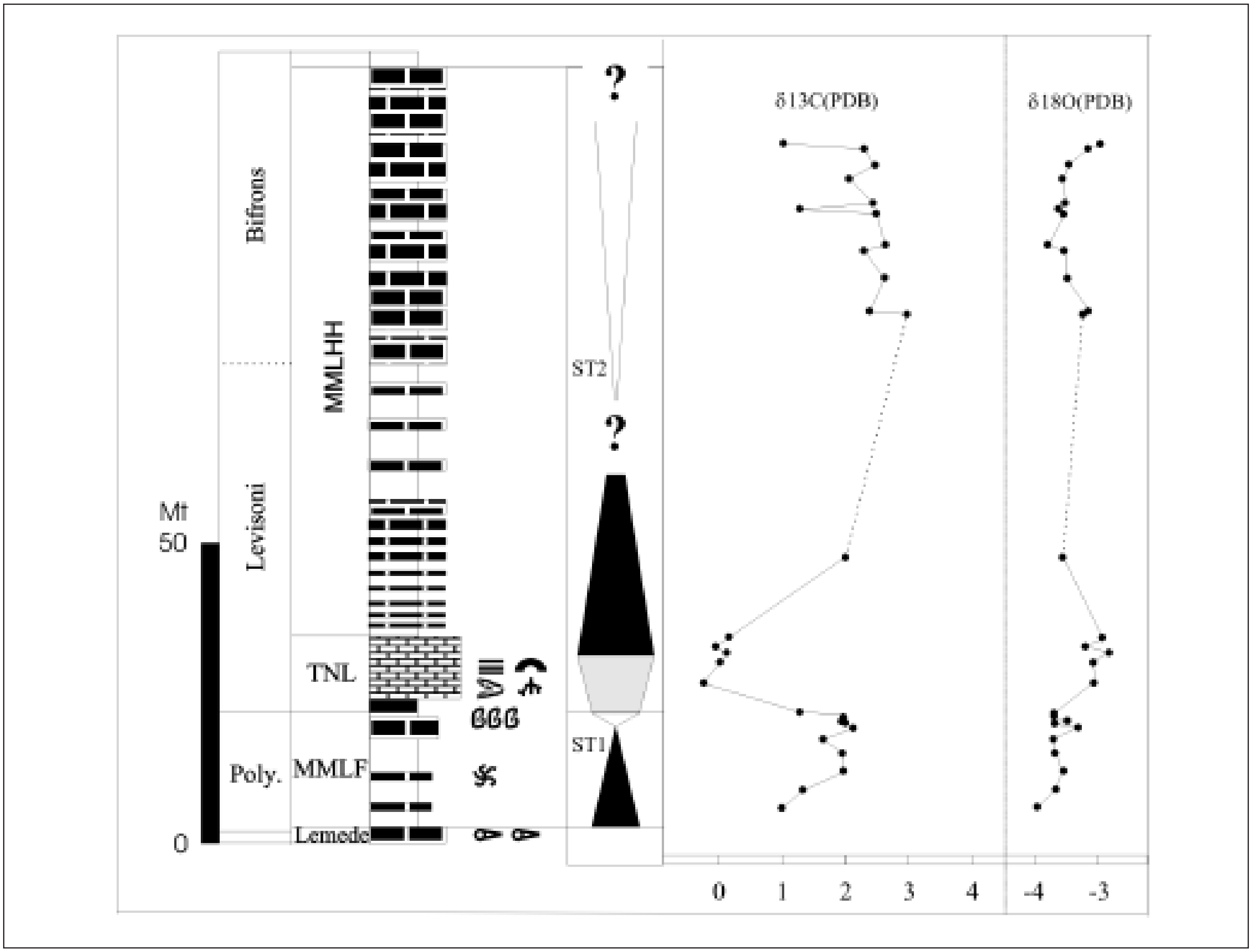

Fig. 7 - Sedimentology, sequence stratigraphy and isotope evolution of Lower to lowermost Middle Toarcian in the Figueira da Foz composite-section. Key to the symbols used in the figures 3 and 4
Fig. 7 - Sedimentología, estratigrafía secuencial y evolución isotópica desde el Toarciense inferior a la parte basal del Toarciense superior en el corte compuesto de Figueira de Foz. La leyenda de los símbolos utilizados están en las figuras 3 y 4

the MMLSB and it is characterized by marl/limestone alternations with siliceous sponge mud mounds (Duarte, 1997; Duarte *et al.*, 2001). These bioherms are observed in all sectors with exception of Tomar, Peniche and Arrábida, occurring just to the top of ST3. The strong accumulation of marls (6 to 10 m thick) recorded at the base of this sequence in northern regions is interpreted as a transgressive systems tract, resulting from an increase of accommodation space. Laterally, these marls are equivalent in Porto de Mós to a siliceous sponge biostrome condensed level that is interpreted by Duarte *et al.* (2001) as the ST3 maximum flooding interval. Overlying this surface is aggradational/progradational package belonging to the interval between *gradata* and the top of *bonarellii*/base of *speciosum* Zone. The regressive trend is well-recorded in some eastern sectors (Rabaçal-Alvaiázere) and particularly in Tomar and Peniche (Duarte, 1997). In the first case, the series becomes progressively more calcareous and bioclastic, ending the sequence with a rhynchonellid (*Pseudogibbirhynchia* sp.) bioclastic horizon. In Tomar, the bioclastic enrichment is evident in biosparites/grainstones with bivalves (*Plagiostoma* sp., *Trigonia* sp.), brachiopods (rhynchonellids and terebratulids), echinoid spines and ahermatipic corals (Figure 8). In Peniche, the marls that characterize the basin-scale uppermost *bifrons* Zone are overlain by

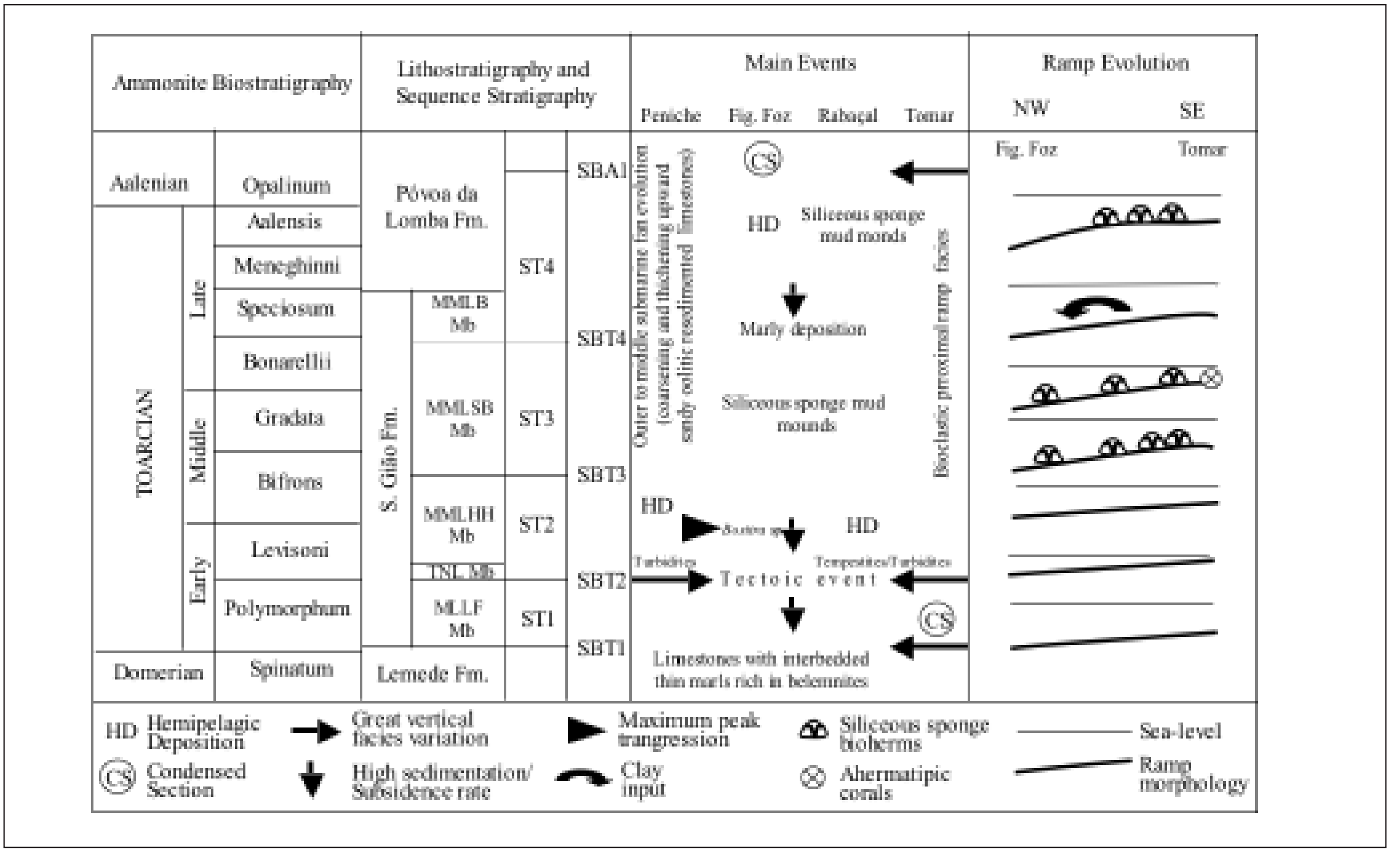

Fig. 8 - Sequence stratigraphy and sedimentary evolution of the Toarcian sediments in the Lusitanian Basin
Fig. 8 - Estratigrafía secuencial y evolución de los sedimentos del Toarciense en la Cuenca Lusitánica

an oolitic-peloidal/marl succession. This prograding trend, observed in the Peniche section, is associated with a hemipelagic to outer submarine fan evolution (Wright and Wilson 1984).

Base of Sequence ST4: This sequence includes the units MMLB (last member of S. Gião Formation) and the base of Póvoa da Lomba Formation. It belongs to the *speciosum–opalinum* Zone interval. Similarly to the preceding ST3 sequence, ST4 starts with a very strong accumulation of marls and argillaceous limestone, particularly important in the northern sectors of the basin where MMLB can reach around 40 m thick. The base of MMLB is interpreted as a transgressive systems tract, as a consequence of an increase in accommodation space. The maximum flooding interval is represented by the massive occurrence of brachiopod-rich horizons (with *Soaresirhynchia renzi* and *Nannirhynchia cotteri*), well developed in several distal parts of the basin (Almeras, 1994).

Discussion

δ^{13}C and Sequence Stratigraphy Interpretation

It is known that δ^{13}C is a useful tool in stratigraphic analysis, particularly in the case of hemipelagic marl-limestone alternation successions. This is the case of the Lower and Middle Toarcian series of the LB, which also has an accurate ammonite biostratigraphy and displays 3rd order transgressive and regressive cycles throughout. The δ^{13}C composition of bulk carbonate reflects complex relations between ocean and atmosphere (*e.g.* Weissert, 1989; Weissert *et al.*, 1998; Kump and Arthur, 1999) at time of deposition, and probably diagenetic burial reactions, but variations of this signal in the Toarcian of LB also reflect clear changes in the depositional system. In fact, 3rd order sequence boundaries are always accompanied by important, sometimes large variations, in δ^{13}C composition. These observations indicate a control-

ling mechanism of primary nature and clearly associated with transgressive or regressive phases.
Transgressive Phases

According to the interpretation above, ST1 is truncated at the top, as a consequence of an input of siliciclastics in the series, generated by tectonics (Duarte *et al.*, 2004a). This tectonic phase, dated as lowermost *levisoni* Zone, interrupts a large 3rd-order transgressive event that characterizes ST1. This transgressive trend is well correlated by the positive excursion observed in $\delta^{13}C$ at the top of this unit (top of MLLF), reaching very similar values in the distal sectors of Coimbra and Figueira da Foz (Figures 6 and 7).

Both ST2 and ST3 show a relatively short transgressive phase, more evident in the case of ST3. Considering the first sequence (ST2), the base of MMLHH (middle-upper part of *levisoni* Zone), that is interpreted as a 3rd-order deepening phase, shows a strong positive isotopic shift, reaching values of 4.29‰ in Porto de Mós and 4.02‰ in Coimbra; this excursion shows a large change of 4‰ in a time duration of only about 0.5 My.

The large increase of $\delta^{13}C$ observed in the extreme (transgressive) base of ST3 at Porto de Mós (Figure 5) is good evidence of this relation between carbon-isotope variation and sequence evolution.

Regressive Phases

In contrast to ST1, ST2 and ST3 are both characterized by strong development of regressive phases. In both cases $\delta^{13}C$ curves show a clear and significant decrease along the middle-upper part of each 3rd order sequence but with different magnitudes. Comparing the results observed in Coimbra, Rabaçal and Porto de Mós across ST2, the amplitude of decrease is very similar in the first two areas (2.23‰, 2.36‰ respectively) while in Porto de Mós it is greater than 3.16‰, reaching 1.11‰ (Figures 4 to 6). In fact, the depositional contrast between the top of ST2 and base of ST3 is better expressed in the Porto de Mós section (discussion in Duarte *et al.* 2001). The regressive trend of ST3 correlates to a slight negative trend in the $\delta^{13}C$ record: from 2.52‰ to 1.7‰ in Rabaçal, 2.54‰ to 1.47‰ in Porto de Mós and 1.89‰ to 0.48‰ in Coimbra (Figures 4 to 6). In this last sector, a negative excursion is observed at the top of ST3.

On the other hand, the lowstand deposits that characterizes the base of ST2 (TNL) are correlatable with an abrupt decrease of carbon-isotope evolution, reaching values around 0‰ (-0.87‰ in Coimbra; -0.25‰ in Figueira da Foz).

Lower Toarcian Global Events

The Early Toarcian is known as an important time of global scale events. It is argued to be an important mass extinction episode (Little and Benton, 1995; Pálfy and Smith, 2000), associated with oceanic anoxia (Jenkyns, 1988). More recently, these events have been related with the austral Karoo-Ferrar flood basalt episode (Pálfy and Smith, 2000; Jones and Jenkyns, 2001). The Lower Toarcian (around *polymorphum*/*levisoni* boundary) deposits of the LB show the most important sedimentological changes that occurred in the whole Early Jurassic (Duarte, 1997; Duarte *et al.*, 2004a), connected with a strong perturbation in the biological record (Mouterde and Ruget 1984; Gahr, 2002, 2005; Perilli and Duarte, 2006), but without the development of anoxic facies. In fact, in Portugal, the extreme base of *levisoni* Zone is marked by a strong reduction of benthic and nektonic macrofauna record. As demonstrated above, the negative and positive carbon-isotope excursions are clearly associated with 3rd order regressive and transgressive phases, respectively. However, carbon-isotope curves observed in the studied sections seem to reflect also the main global isotopic events (Figure 9).

a) The most important $\delta^{13}C$ positive excursion (magnitude up to 4‰) observed in the middle-upper part of *levisoni* Zone of LB is more or less associated with 2nd order sequence maximum flooding phase. Despite the absence of organic carbon-rich sediments in the studied sections (in Portugal, only the *levisoni* Zone of the Peniche section shows high TOC values; Duarte *et al.* 2005), this isotopic positive excursion is well observed in several European and Tethyan basins (Jiménez *et al.*, 1996; Jenkyns *et al.*, 2002). This event corresponds to a widespread deepening phase, illustrated in several eustatic curves (Hallam, 1988; 2001; Haq *et al.*, 1988; Hardenbol *et al.*, 1998), and seems to be associated to the maximum flooding interval of 2nd order transgressive-regressive facies cycles observed in some Western European basins (e.g. Paris Basin, in De Graciansky *et al.*, 1998b).

The gradual decrease of $\delta^{13}C$ up to the *bifrons* Zone is consistent with the trends observed in neighbouring basins of Iberia and Central Europe (Jenkyns and Clayton, 1986, 1997; Jenkyns *et al.*, 1991, 2002; Jiménez *et al.*, 1996; Rosales *et al.*, 2001).

b) The strong negative carbon isotopic excursion recorded at the base of *levisoni* Zone (TNL) is typical of the event that characterizes marine carbonates and organic matter dated as uppermost *tenuicostatum* (*polymorphum*) Zone and the lower part of *falciferum* (*levisoni*) Zone of some Central European basins (Jiménez *et al*., 1996; Hesselbo *et al*., 2000; Schouten *et al*., 2000; Beerling *et al*., 2002; Jenkyns *et al*., 2002; Van de Schootbrugge *et al*., 2005). Some authors such as Hesselbo *et al*. (2000) and Beerling *et al*. (2002) suggested that this rapid event was the result of large methane gas-hydrate dissociation. Others, such as Schouten *et al*. (2000) and Röhl *et al*. (2001) considered recycling and re-utilization of ^{12}C from stratified anoxic bottom waters as a possible mechanism. However, if we compare the known $\delta^{13}C$ curves in different domains of Europe we conclude that this negative excursion is not so generalised as the following positive excursion. In fact, it is not recognised in some neighbouring sectors such as the Basque-Cantabrian Basin (Rosales *et al*., 2001).

Independently of the possible global mechanisms, the lowstand deposits observed in the LB may be justified the input of ^{12}C in the oceanic carbon reservoir that could have been released by erosion and weathering of continental material. Contrary to the observation of widespread black shale deposition, the Lower Toarcian (both *polymorphum* and *levisoni* Zones) of the four LB studied sections shows low values of organic matter (TOC values below 0.5%; Duarte *et al*., 2005)

Conclusions

The Toarcian successions in the central and northern part of the LB are dominated by hemipelagic marl/limestone alternations, rich in benthic and nektonic macrofauna. The Lower and Middle Toarcian series are organised into three 3rd order depositional sequences (ST1, ST2 and ST3), bounded by four isochronous regional discontinuities, well dated by ammonites: at base of *polymorphum* Zone (SBT1), across the *polymorphum*/*levisoni* Zone boundary

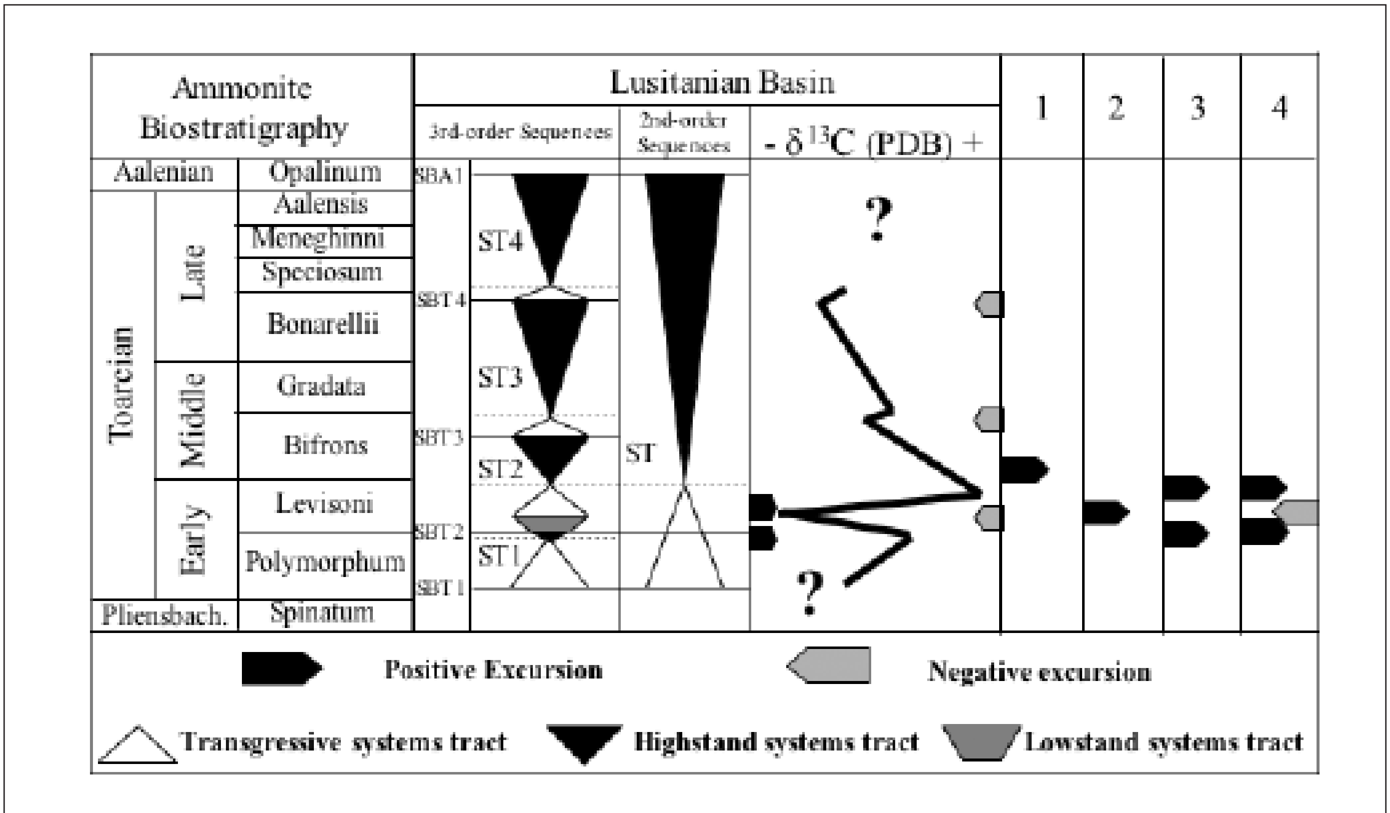

Fig. 9 – Correlation between 3rd order depositional sequences and $\delta^{13}C$ evolution in the Toarcian of Lusitanian Basin. Carbon-isotope events in carbonate of several neighbouring basins: 1 - Subbetic Cordillera (Jiménez *et al.,* 1996); 2 - Basque-Cantabrian Basin (Rosales *et al*., 2001); 3 -Yorkshire (McArthur *et al*., 2000); 4 - Mochras well in Wales (Jenkyns *et al*., 2002; Van de Schootbrudge *et al*., 2005)

Fig. 9 - Correlación entre las series deposicionales de 3er orden y la evolución del $\delta^{13}C$ en el Toarciense de la Cuenca Lusitánica. Eventos de los isótopos de carbono en los carbonatos de varias cuencas vecinas: 1.- Cordillera Subbética (Jiménez et al., *1996); 2.-Cuenca Vascocantábrica (Rosales* et al., *2001); 3.- Yokshire (McArthur* et al., *2000); 4.- Pozo Mochras, en Gates (Jenkyns* et al., *2002; Van de Schootbrudge* et al., *2005)*

(SBT2), within the *bifrons* Subzone (SBT3) and uppermost *bonarelli* Zone (SBT4), respectively. The most significant sedimentological change is observed at the ST1/ST2 boundary, where the facies of the base of *levisoni* Zone are interpreted as the result of a strong shallowing (lowstand).

Isotopic curves obtained in four reference sections of the LB, reveal that $\delta^{13}C$ is an important tool in sequence stratigraphic interpretation. The first main conclusion of the observed variations is that the link with the sequence evolution should indicate a mechanism of a primary nature. The $\delta^{13}C$ curves clearly show remarkable changes at the boundaries between the several 3rd order depositional sequences. The large positive fluctuations correspond to transgressive periods. With the exception of ST1, the uppermost parts of ST2 and ST3 show gradual negative shifts, associated with regressive phases. Two main positive $\delta^{13}C$ excursions are observed at the top of *polymorphum* Zone and in the middle-upper part of *levisoni* Zone, both peaks associated with maximum flooding intervals. Between these two peaks, the minimum value (negative excursion) is observed at the base of *levisoni* Zone, associated with shallow detrital facies deposits.

Besides, the $\delta^{13}C$ evolution observed in the Lower and Middle Toarcian of LB shows the global carbon shift recognised in other Tethyan and North-European sectors.

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