Geologica Acta, Vol.8, Nº 3, September 2010, 325-340
DOI: 10.1344/105.000001536
Available online at www.geologica-acta.com

Organic-Rich facies in the Sinemurian and Pliensbachian of the Lusitanian Basin, Portugal: Total organic carbon distribution and relation to transgressive-regressive facies cycles

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ABSTRACT

The upper Sinemurian to Pliensbachian series of the Lusitanian Basin (Portugal) correspond to marly limestone sediments rich in benthic and nektonic macrofauna. This sedimentary record includes several intervals of organic-rich facies, which are particularly well developed in the western sectors of the basin. They correspond to grey and dark marls locally showing strong lamination (black shale type) and are recognized as one of the most important potential oil source rocks. This study shows the vertical and lateral distribution of these organic-rich intervals, supported by over 550 total organic carbon (TOC) determinations. The results presented reveal two important intervals, with several black shale occurrences, in the Oxynotum(?)–Raricostatum (Polvoeira Member of Água de Madeiros Formation) and at the top of the Ibex-upper part of Margaritatus zones (top of the Vale das Fontes Formation), showing in the distal (western) sectors up to 22% and 15% TOC, respectively. TOC values decrease progressively towards the proximal sectors, the youngest organic-rich interval being the most expressive at the basin scale. This lateral TOC distribution, the facies stacking patterns and the decrease observed in benthic macrofauna confirm that these intervals are related to 2nd-order transgressive phases. 2nd-order regressive phases, developed during the uppermost Raricostatum and Spinatum zones respectively, show lower TOC values.
TOC distribution combined with other stratigraphic and sedimentological parameters enabled seven facies maps to be created for the time interval studied. At the regional scale, this study shows for the first time the good similarity between the upper Sinemurian-Pliensbachian sedimentary successions of the Lusitanian and Basque-Cantabrian basins.

KEYWORDS Lower Jurassic. Organic-rich marls. TOC. Sequence stratigraphy. Palaeogeography. Lusitanian Basin.

INTRODUCTION

In Portugal, the Lower Jurassic sedimentary record is particularly well represented in the western margin (Lusitanian Basin, hereinafter called LB) of the Variscan Iberian Massif. It corresponds to a thick carbonate succession, comprising up to 550 m of mostly marl-limestone alternations, characterizing much of the upper Sinemurian–Toarcian series of the basin (Soares et al., 1993; Duarte and Soares, 2002; Duarte et al., 2004b). These facies, comprising abundant nektonic and benthic macrofauna, are included in the Upper Triassic–Callovian 1st-order cycle (Wilson et al., 1989; Soares et al., 1993; Duarte, 1997; Azerêdo et al., 2002, 2003; Duarte et al., 2004b) and are associated with a palaeogeography controlled by an epicontinental sea, sustained by a low-gradient carbonate ramp dipping towards the northwest (Duarte, 1997, 2007; Duarte et al., 2004b). In this geological context, the upper Sinemurian–Pliensbachian interval is characterized by the occurrence of organic-rich facies regarded as a potential oil source-rock (Oliveira et al., 2006).

The first aim of this paper, through the study of several key-sections of the LB (Tomar, Rabaçal, Coimbra, Anadia, Montemor-o-Velho, Figueira da Foz, S. Pedro de Moel and Peniche; Fig. 1), involves characterizing the marly units with accompanying organic-rich levels, based on a detailed ammonite biostratigraphy and other biotic and abiotic parameters. The second objective is to quantify the total organic carbon (TOC) in these facies, showing its basin wide vertical and lateral variation. This procedure has allowed the creation of several facies maps based on organic facies and TOC distribution. Apart from constituting a key stratigraphic marker and a tool in the intrabasinal correlation of the several studied sections, TOC is also useful in terms of sequence stratigraphy interpretation of the series, particularly in the identification of 2nd-order transgressive-regressive sequences or the equivalent transgressive-regressive facies cycles (T-R facies cycles *sensu* de Graciansky et al., 1998; Jacquin and de Graciansky, 1998a, 1998b). Thus, we compared the sequential scheme already presented for the LB (e.g. Duarte et al., 2004b; Duarte, 2007) with others from neighbouring basins, emphasizing the main similarities in terms of regional transgressive and regressive events.

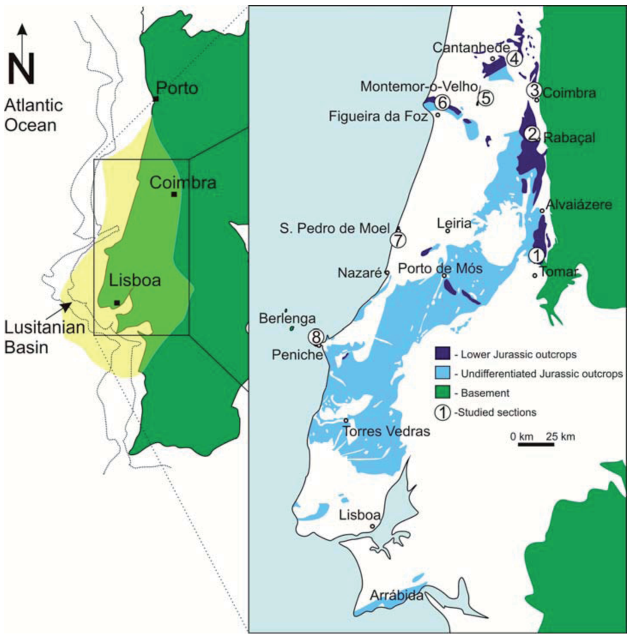

FIGURE 1 | **Geological map of Jurassic in the Lusitanian Basin. Location of the main studied Sinemurian–Pliensbachian outcrops: 1) Tomar; 2) Rabaçal; 3) Coimbra; 4) Anadia-Cantanhede; 5) Montemor-o-Velho; 6) Figueira da Foz; 7) S. Pedro de Moel; 8: Peniche.**

GEOLOGICAL SETTING AND LITHOSTRATIGRAPHY

During the Early Jurassic, the LB was characterized by carbonate sedimentation (Soares et al., 1993; Azerêdo et al., 2003; Duarte et al., 2004b). The Sinemurian-Pliensbachian series show important changes in the depositional system, from lower-upper Sinemurian peritidal facies (Coimbra Formation; Azerêdo et al., 2008; subsequently denominated as Fm.) to Pliensbachian hemipelagic deposits (including the Vale das Fontes and Lemede formations; Duarte and Soares, 2002) (Fig. 2). However, in the western sectors of the basin, such as Peniche, S. Pedro de Moel, Figueira da Foz and Montemor-o-Velho, hemipelagic deposition started earlier during the late Sinemurian (Oxynotum-Raricostatum zones; Água de Madeiros Fm.; Duarte and Soares, 2002; Duarte et al., 2004b, 2006) (Fig. 2). All these units are characterized by different marl/limestone relations, organic matter content and specific benthic/nektonic macrofauna and microfauna. As a result of the ramp morphology, controlled by regional tectonics and relative sea-level changes, the thickness of these series increases in the western sectors, which are biostratigraphically well constrained by ammonites and calcareous nannofossils (e.g. Mouterde, 1955, 1967; Antunes et al., 1981; Phelps, 1985; Dommergues, 1987; Dommergues et al., 2004; Mouterde et al., 2007; Oliveira et al., 2007).

Água de Madeiros Formation

This unit, belonging to the Oxynotum-Raricostatum zone interval, rests over the inner-shelf Coimbra Fm., which is composed of dolomitic and calcareous facies, more or less bioclastic/fossiliferous (Azerêdo et al., 2008). The Água de Madeiros Fm. has been subdivided into two members: the Polvoeira Member (Member henceforth known as Mb.) at the base and the Praia da Pedra Lisa Mb. at the top (Figs. 2, 3). These two units are particularly well exposed along the coastal sections of S. Pedro de Moel (Água de Madeiros-Polvoeira area), where their type-sections are defined (Duarte and Soares, 2002; Duarte et al., 2004b, 2006) (Fig. 4). Here, the base of Polvoeira Mb. consists of marl-limestone alternations that become progressively more argillaceous, presenting several organic-rich facies horizons. The middle-upper part of this member is a rhythmic succession with marl/limestone ratios around 1.5 to 2. This member is rich in benthic (bivalves, brachiopods) and nektonic (ammonites and belemnites) macrofauna, with a clear inverse distribution of these two groups across the unit: abundant benthic fauna at the base (*Pholadomya*, *Gryphaea, Pleuromya, Zeilleria, Cincta, Piarorhynchia*, *Spiriferina*, etc); nektonic fauna increasing in abundance towards the top. Some horizons (mainly laminated limestones) are rich in ammonites (Echioceratids and Oxynoticeratids). Limestones generally correspond to fossiliferous wackestones that are sometimes rich in ostracods, molluscs and organic matter. At S. Pedro de Moel the thickness of this member is approximately 42 m (Fig. 4), decreasing to 10 m in Peniche and Montemor-o-Velho.

The Praia da Pedra Lisa Mb. is predominantly calcareous (wackestones to grainstones). Thickness and vertical facies arrangements are very different at several points of the basin, such as Peniche (around 30 m thick) and S. Pedro de Moel (around 16 m thick). In its type-section, located in the Praia de Água de Madeiros (Fig. 4), 2km south of S. Pedro de Moel, the Praia da Pedra Lisa Mb. is included in the uppermost Raricostatum-lowermost Jamesoni zone interval (Duarte and Soares, 2002). Here, this member begins (around 9 m) by decimetre- to centimetre-thick microspar limestones (wackestones very rich in ostracods and radiolarians), with planar lamination, *Rhizocorallium* and *Thalassinoides*. Some surfaces at the top of the unit are rich in tiny ammonites (*Gemmellaroceras* sp.), but benthic macrofauna is very scarce. The upper part, approximately 7 m thick, is marked by a gradual increase in interbedded centimetre-scale grey to dark marls and a sharp thickening of the limestone beds. These limestones consist of fossiliferous micrites to biomicrites/wackestones with molluscs and ostracods. Nektonic macrofauna is common, comprising belemnites and large Apoderoceratids, such as *Apoderoceras subtriangulare* (YOUNG and BIRD) among others, and which have been attributed by Edmunds et al. (2003) to the Jamesoni Zone, Taylori Subzone.

Vale das Fontes Formation

The Vale das Fontes Fm., ranging in age from the lowermost Jamesoni to the uppermost Margaritatus zone interval, represents the return to a marly sedimentation, widespread across the whole basin. It is particularly well exposed in the western part of the basin, where it is approximately 75-90 m thick (namely in Peniche; Fig. 4) and is subdivided into three informal members: Marls and limestones with *Uptonia* and *Pentacrinus* (MLUP) Mb., Lumpy marls and limestones (LML) Mb. and Marly limestones with organic-rich facies (MLOF) Mb.

MLUP Mb. This unit is characterized by bioturbated decimetre marl/centimetre-thick marly limestone alternations, with nektonic and benthic macrofauna, comprising crinoids, brachiopods and bivalves. The microfacies consist of fossiliferous micrites to biomicrites/wackestone. Across the basin, an increase is observed in the marly character from the proximal to the distal sectors, where this unit is dated as Jamesoni Zone, involving the Taylori-Jamesoni subzone interval. This member reaches approximately 35 m in Montemor-o-Velho and 25 m in Peniche, two points of the basin where the entire member can be observed.

LML Mb. This unit is defined by the occurrence of lumpy facies (Hallam, 1971; Dromart and Elmi, 1986; Elmi et al., 1988; Fernández-López et al., 2000), interbedded in a marl-limestone succession. The lumps

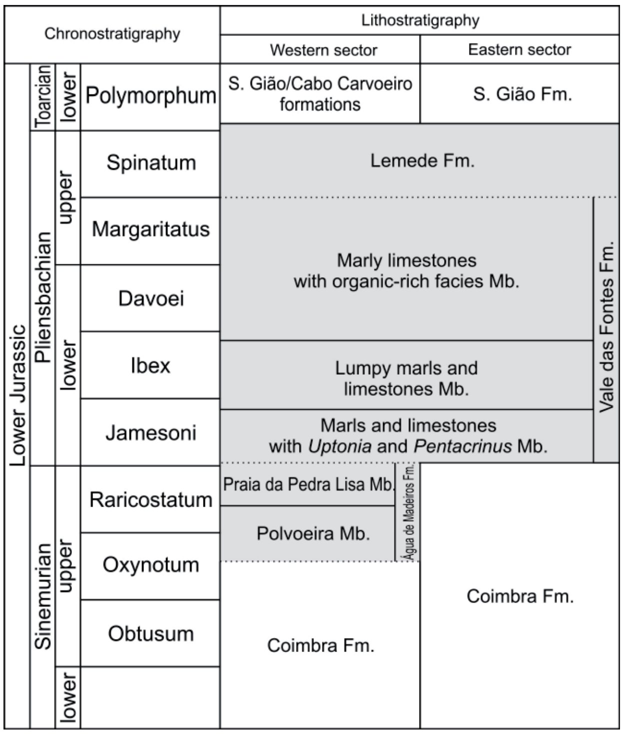

FIGURE 2 | **Lithostratigraphic chart for the Sinemurian–lowermost Toarcian series of the Lusitanian Basin (Duarte and Soares, 2002). Light grey corresponds to the studied units.**

consist of micritic grumose concretions, generally subspherical-shaped and reaching several centimetres (up to 3 cm) in size. The composition of these concretions appears to be a microbial origin, showing some cryptalgal oncolite structures (Dromart and Elmi 1986; Elmi et al. 1988). Interbedded in these facies, metric-scale grey to dark marls occur. In this member, benthic macrofauna is scarce but ammonites and belemnites are always present. This unit, with about 21 m at Peniche and 22 m at Figueira da Foz, ranges from the Jamesoni to the Luridum subzone interval.

MLOF Mb. This unit is characterized by an increase of the marly terms of the series, alternating with centimetre-thick limestone facies, locally with diverse and abundant tiny benthic macrofauna (ostreids, crinoids and brachiopods), ammonites and belemnites. In the distal regions, such as the Peniche, S. Pedro de Moel and Figueira da Foz sectors, organic-rich sediments are particularly abundant. This member comprises the Luridum Subzone (topmost of Ibex Zone) to the uppermost Margaritatus Zone interval and ranges from 10 m at Tomar to around 20 m at Rabaçal and 28/29 m at Figueira da Foz and Peniche.

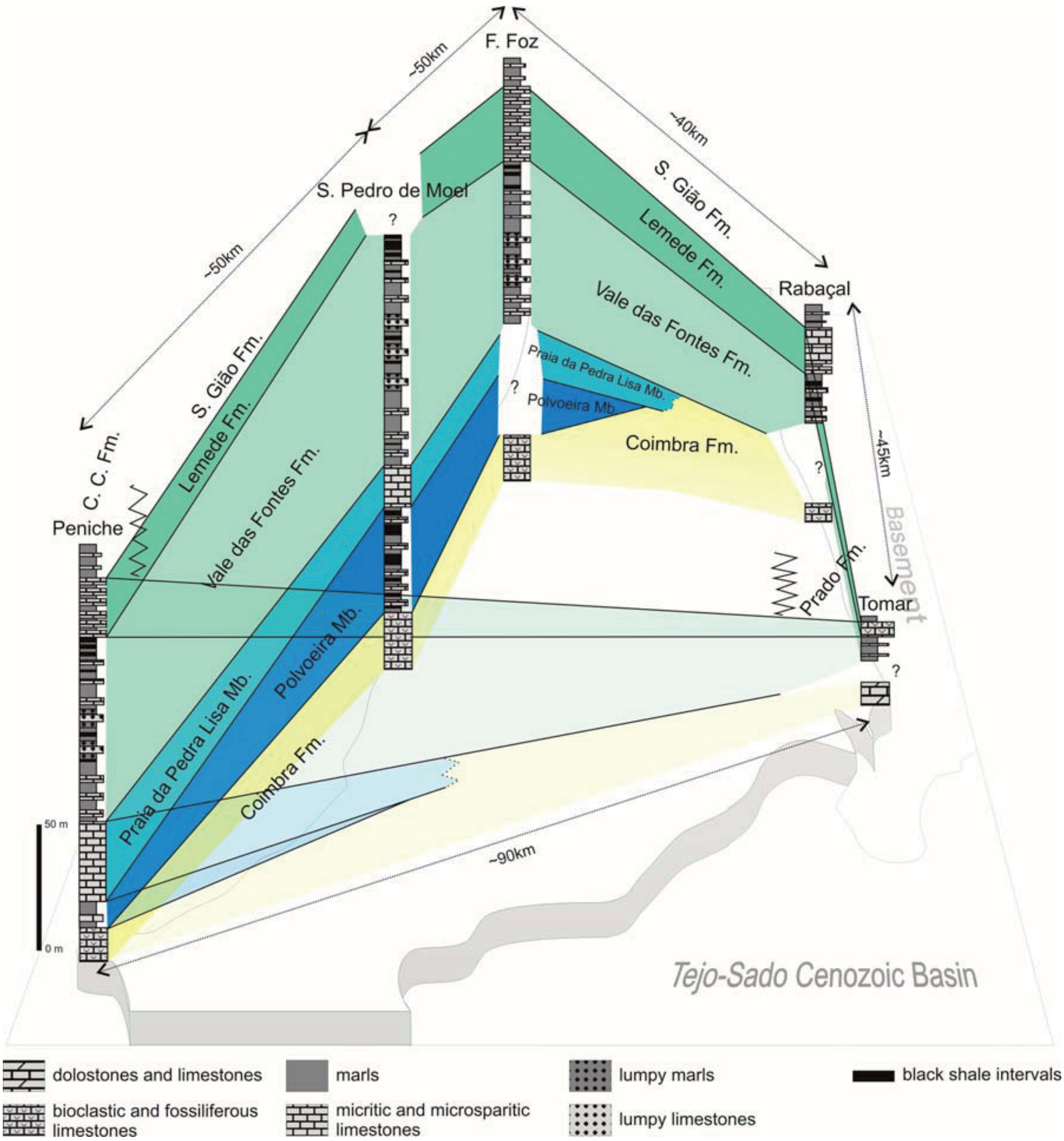

FIGURE 3 | **Lateral and vertical sedimentary variation of the uppermost Sinemurian–lowermost Toarcian marly limestone series in the Lusitanian Basin. Synthetic logs of Tomar, Rabaçal, Figueira da Foz, S. Pedro de Moel and Peniche.**

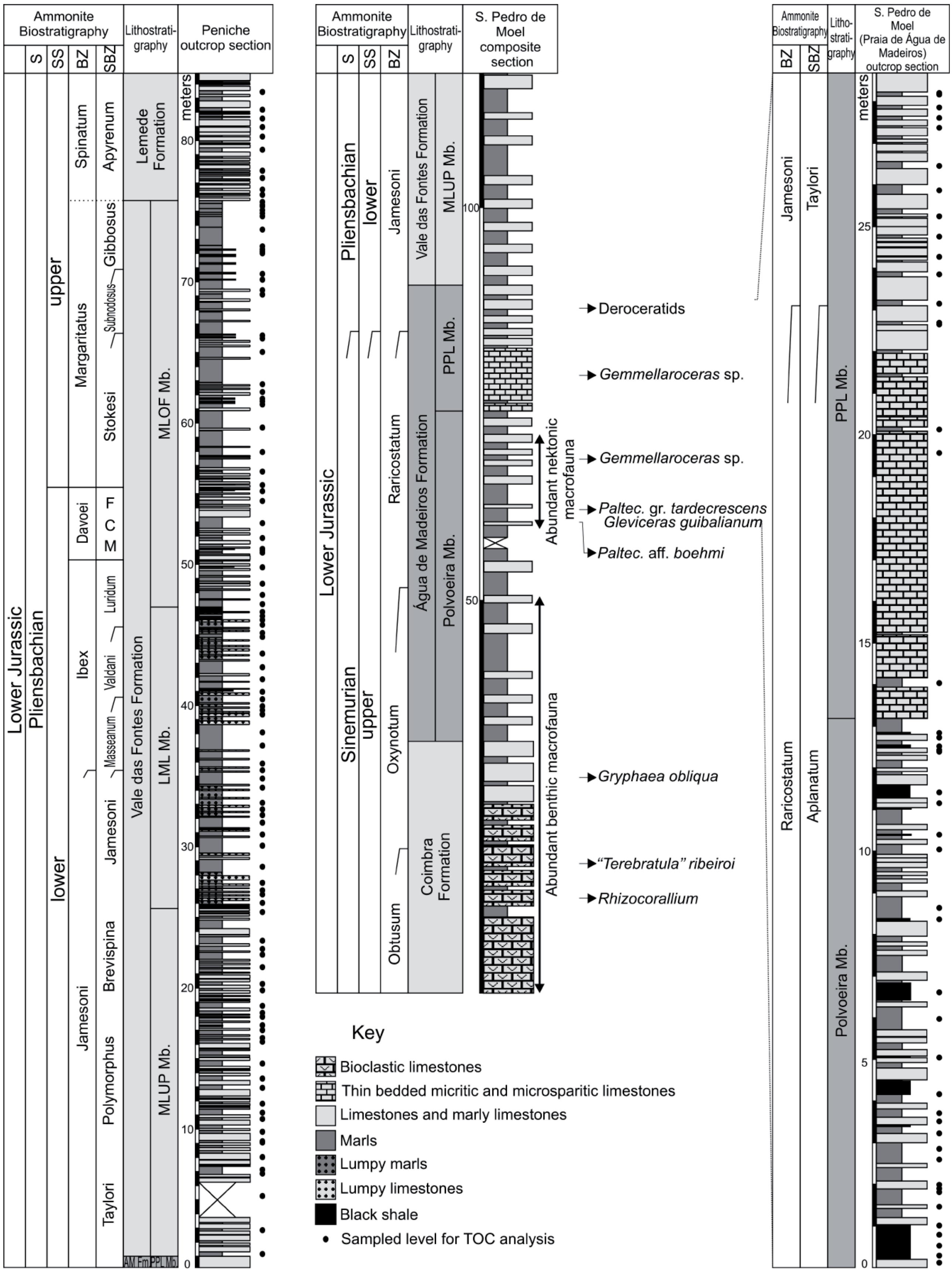

FIGURE 4 | **Type-sections of the Água de Madeiros (S. Pedro de Moel area) and Vale das Fontes (Peniche) formations in the Lusitanian Basin.**

Lemede Formation

This unit generally comprises centimetre marl/decimetre limestone bioturbated alternations, very rich in belemnites, ammonites, bivalves and brachiopods. In the southeastern part of the LB, such as Tomar, facies are much more bioclastic (packstone to grainstone) and locally dolomitic, with a diversified benthic macrofaunal component (large bivalves and brachiopods). This unit ranges in age from the Spinatum Zone to the lowermost part of Polymorphum Zone, reaching a thickness of approximately 30 m to the northwest.

STUDIED SECTIONS AND METHODS

For this contribution we have revised all the important and classic sections characterizing the upper Sinemurian and Pliensbachian hemipelagic deposits of the LB (Duarte and Soares, 2002; Duarte et al., 2004b, 2006; Oliveira et al., 2006; Silva et al., 2007). TOC content was analyzed in approximately 550 samples from eight key-sections (Fig. 1), chosen due to the occurrence of organic-rich facies, good biostratigraphic control and other stratigraphic and sedimentological (e.g. petrographic and mineralogical data) criteria. The intervals richer in organic matter were analyzed in greater detail, as in the cases of the Rabaçal (Upper Sinemurian and Davoei-Spinatum zone interval), Coimbra (Jamesoni-Margaritatus zone interval), Montemor-o-Velho (Raricostatum-Jamesoni zone interval), Figueira da Foz (Jamesoni-Margaritatus zone interval), S. Pedro de Moel (Oxynotum-Jamesoni and Davoei-Margaritatus zone intervals; Fig. 4) and Peniche (Raricostatum-Spinatum zone interval; Fig. 4) sections. Despite the good biostratigraphic data available for most of the sections (see references above), new ammonite data enabled stratigraphic boundaries to be improved, particularly in the case of the Raricostatum and Davoei-Margaritatus zone interval.

TOC content was determined with a Leco SC-444 analyzer in the Universidade do Estado do Rio de Janeiro and Cenpes-Petrobras (Brazil), with an analytical precision of ± 0.1%. The results are presented in weight percent (wt %) and in every ten samples analyzed a duplicated analysis was performed. The equipment was calibrated daily with standards, before analyses were initiated.

ORGANIC-RICH FACIES: AGE CALIBRATION AND TOC VALUES

The organic-rich facies are quite similar in all the studied series and are characterized by a grey to dark colour, generally with a net lamination and, locally, abundant pyrite. These facies include black shales, dark-grey marls and laminated limestones. In the whole upper Sinemurian-Pliensbachian succession, the black shales are generally thin (a few cm) but in the uppermost Sinemurian (middle part of Polvoeira Mb.) of S. Pedro de Moel area they are thicker, locally reaching 50 cm. Generally, black shales are not bioturbated but, in a few organic-rich facies, the ichnogenus *Chondrites* occurs. In addition, the main inorganic components include carbonate (practically limited to calcite), clay minerals (mainly illite and 10-14 mixed layer minerals; small amounts of kaolinite, chlorite and vermiculite) and some quartz (e.g. Duarte et al., 2007b).

In terms of stratigraphic distribution, organic-rich facies occur in the latest Sinemurian to the earliest part of the late Pliensbachian, involving the Água de Madeiros and Vale das Fontes formations. Black shales occur in all members of these two formations and are recognized in all ammonite biostratigraphic zones. According to Fig. 5, which summarizes the vertical and lateral distribution of TOC in several studied sections, two intervals particularly rich in organic-rich facies and with high TOC values are observed: top of the Oxynotum-Raricostatum (Polvoeira Mb.) and top of the Ibex-Margaritatus (MLOF Mb.) zones.

Oxynotum-Raricostatum zone interval

The highest and major number of levels with high TOC values of the LB, are observed in this first stratigraphic interval, but lateral distribution of organic-rich facies in the basin is restricted to the western sectors. In the particular case of S. Pedro de Moel, where the succession is thicker, TOC values reach up to 22% in two horizons (Oxynotum(?)-base of Raricostatum Zone). Here, the TOC background values of the Polvoeira Mb. is generally above 1%, and 50% of the samples analyzed show TOC values higher than 5-6% (Fig. 5). Aside from these two peaks, several values above 10% can be found in the upper part of the Raricostatum Zone. This Zone is well constrained by an abundant ammonite record [levels with *Paltechioceras boehmi* (Hug), *Leptechioceras meigeni* (Hug), *Paltechioceras tardecrescens* (Hauer), *Gleviceras guibalianum* (D'Orbigny) and *Paltechioceras romanicum* (Uhlig)], belonging to the Raricostatum, Macdonnelli and Aplanatum subzones which, to date, have only been recognized in S. Pedro de Moel (Fig. 4). The top of the Oxynotum Zone (?), particularly rich in benthic macrofauna (bivalves and brachiopods) and very poor in ammonites, shows the first organic-rich facies, which reaches the maximum TOC value of 7.4 %.

Laterally, like the thickness of the marly facies, TOC values decrease abruptly towards the north and south of S. Pedro de Moel, reaching the maximum values of 8%

and 4% in Montemor-o-Velho and Peniche, respectively. In Montemor-o-Velho, black shales are well recognized in some levels, generally with TOC values above 2.5%. According to new ammonite data (unpublished) collected in these two sectors, the highest TOC values are located at levels that appear to be laterally correlative and dated from the Macdonnelli and Aplanatum subzones.

Top of Ibex-Margaritatus zone interval

This interval corresponds to the MLOF Mb. (Figs. 2-5), which constitutes the most organic-rich part of the Vale das Fontes Fm.. TOC contents of the studied samples in the basin show the maximum value of 15% in both the Peniche and S. Pedro de Moel sectors. Laterally, high values of this parameter are distributed widespread, reaching up to 9.8% in Figueira da Foz, 5.2% in Coimbra and 4.8% in Rabaçal. Detailed analysis of the vertical variation of TOC in distal sectors of the basin (Fig. 5) enables three intervals rich in organic facies to be defined that, in Peniche, present values of 12, 15 and 15%, respectively. These intervals are dated as Davoei (levels with *Aegoceras* and *Liparoceras*), the basal part of Margaritatus [Stokesi Subzone, levels with *Protogrammoceras* cf. *celebratum* (FUCINI)] and the upper part of the Margaritatus (between Subnodosus and Gibbosus subzones) zones, respectively. The latter interval is clearly the thickest and laterally the richest in TOC values, even in the proximal parts of the basin, such as Coimbra and Rabaçal (around 5%).

Other intervals

According to Fig. 5, TOC values are generally low in the other marly units of the basin. There are, however, some other interesting stratigraphic reference points. In the Jamesoni Zone, the TOC content is generally below 1%, but at S. Pedro de Moel can reach 5-8% in several thin black shales. In the Ibex Zone, and in spite of the low TOC background values, a thin organic-rich horizon is observed throughout the basin, showing values of 9.1% in Figueira da Foz, 7.3% in Cantanhede, 6.5% in Coimbra, and 4% in Peniche.

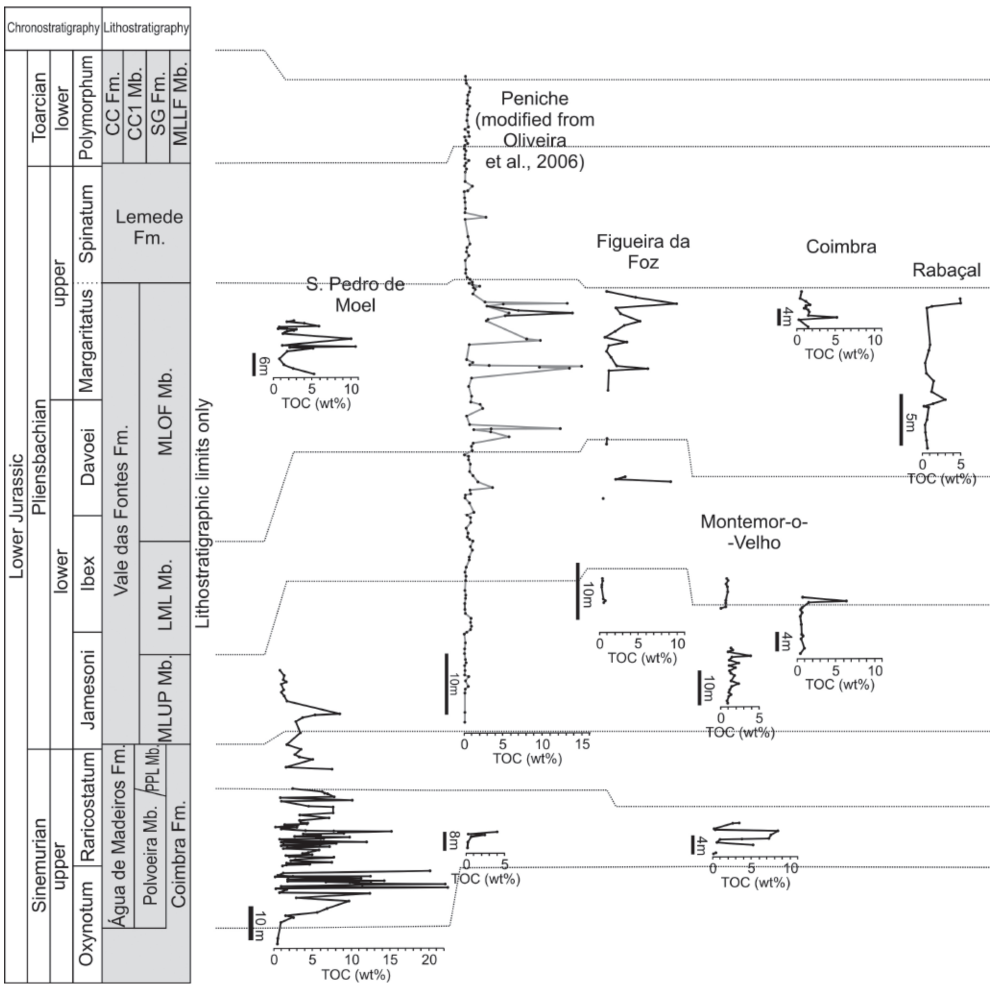

FIGURE 5 | **TOC distribution in the uppermost Sinemurian–Pliensbachian of the Lusitanian Basin (abbreviations as explained in text except for Cabo Carvoeiro Fm.: CC Fm., Cabo Carvoeiro 1 Mb.: CC1 Mb., S. Gião Fm.: SG Fm. and Marly-limestones with *Leptaena* Facies Mb.: MLLF Mb.).**

AREAL DISTRIBUTION OF THE ORGANIC RICH-FACIES

As presented above, in the LB the Sinemurian marks an important sedimentary transition, from shallow-water dolomites and limestones (Coimbra Fm.) to hemipelagic deposits. The high-resolution stratigraphy and TOC data obtained in this study allow, for the first time, the construction of detailed spatial facies maps, showing the different stages of development

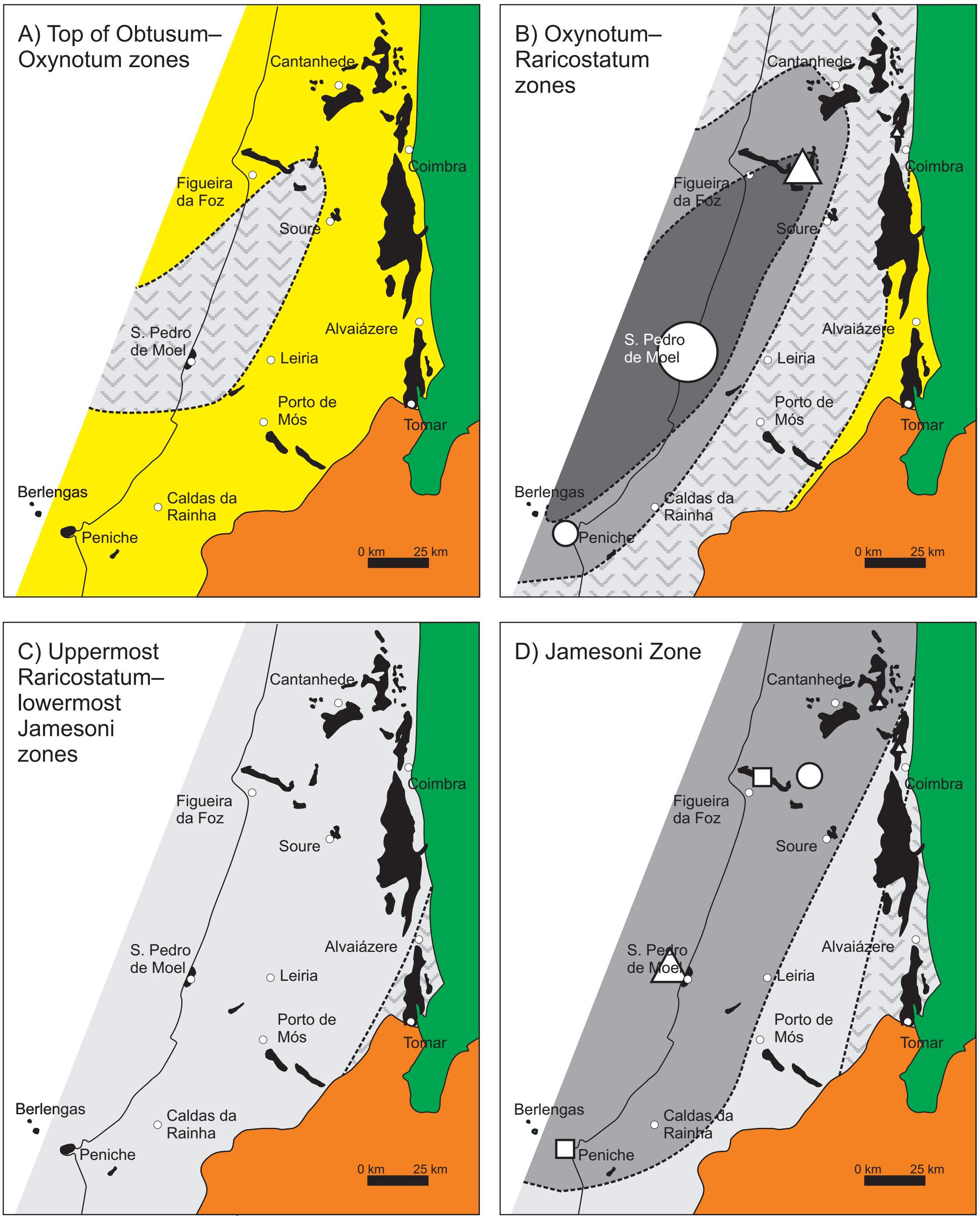

FIGURE 6 | **Facies maps and TOC distribution for the studied interval. A) top of Obtusum-Oxynotum zones (top of Coimbra Fm.); B) Oxynotum–Raricostatum zones (Polvoeira Mb.); C) uppermost Raricostatum–lowermost Jamesoni zones (Praia da Pedra Lisa Mb.); D) Jamesoni Zone (MLUP Mb.); E) Jamesoni –Ibex zones (LML Mb.); F) top of Ibex–Margaritatus zones (MLOF Mb.); G) Spinatum–lowermost Polymorphum zones (Lemede Fm.).**

of the carbonate ramp between the Late Sinemurian (Obtusum Biochron) and the top of the Pliensbachian (Spinatum Biochron). These maps correspond to the following time intervals (Figs. 6A to 6G): Top of Obtusum-Oxynotum(?) biochrons (top of Coimbra Fm.), Oxynotum(?)-Raricostatum biochrons (Polvoeira Mb.), uppermost Raricostatum-base of Jamesoni biochrons (Praia da Pedra Lisa Mb.), Jamesoni Biochron (MLUP Mb.), Jamesoni-upper Ibex biochrons (LML Mb.), uppermost Ibex-top of Margaritatus biochrons (MLOF Mb.) and Spinatum-extreme base of Polymorphum biochrons (Lemede Fm.). According to Fig. 3, the

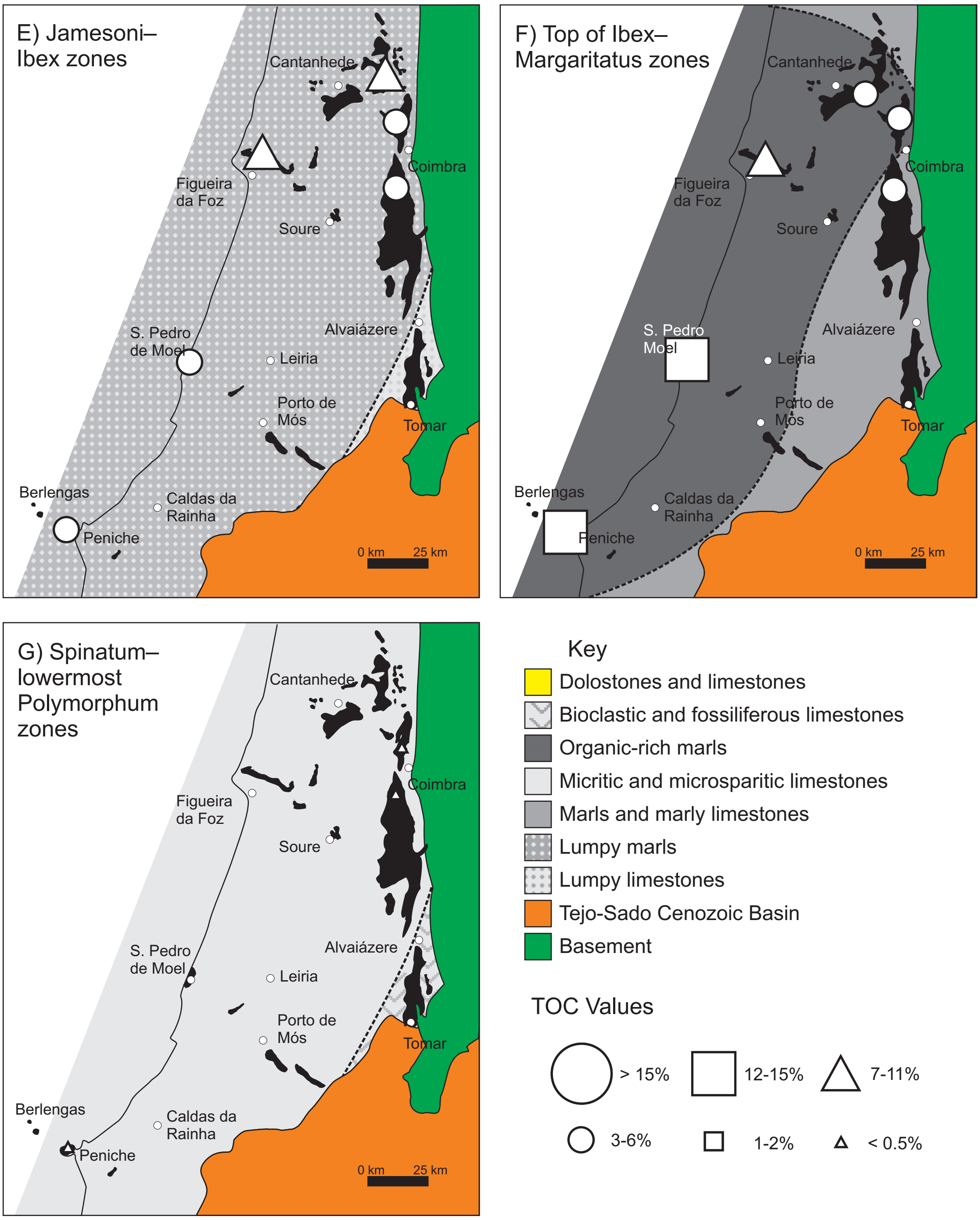

FIGURE 6 | **(See page 332).**

thickness of the different units increases from the southeastern proximal part of the basin (Tomar region) towards the northwest sector (Figueira da Foz region; distal region), following the direction of the dipping ramp. Well-developed organic-rich facies, where TOC values are always very high throughout the whole succession, are practically restricted to the western part of the basin. During the late Sinemurian, these facies were centred in the distal sectors, whereas in the Pliensbachian, black shales with higher TOC values reach several eastern sectors of the basin, such as Coimbra and Rabaçal (Figs. 6B and 6F).

TOC VARIATION RELATED TO T-R FACIES CYCLES

Apart from the importance of TOC as palaeoenvironmental and palaeogeographic markers in the studied series, this parameter is also decisive in sequence stratigraphic analysis and in the recognition of T-R facies cycles (e.g. Quesada et al., 2005). Indeed, the marly accumulation of the Polvoeira and MLOF members, the TOC-richer units of the whole Early Jurassic of the LB, are correlated with maximum flooding intervals, associated with 2nd-order transgressive phases in the Sinemurian (Sequence SS) and Pliensbachian (Sequence SP) (see Duarte et al., 2004b; Duarte, 2007). On the other hand, absence of black shales and low TOC values, observed in the base of Praia da Pedra Lisa Mb. and Lemede Fm., are both related with 2nd-order regressive phases (Fig. 7).

Sinemurian 2nd-Order T-R Facies Cycle

Despite the limited observational conditions (few and discontinuous outcrops), dolomitization, lack of biostratigraphic control and other sedimentary features, knowledge of Sinemurian carbonates in the eastern part of the basin (Coimbra Fm.; Figs. 1, 3, 7) does not allow a good sequential scheme to be constrained. However, the study of some sections located in the western part, allows interpretation of the sequential evolution (2nd-order) for the Early to Late Sinemurian. In a recent study, Azerêdo et al. (2008) demonstrated that the lower

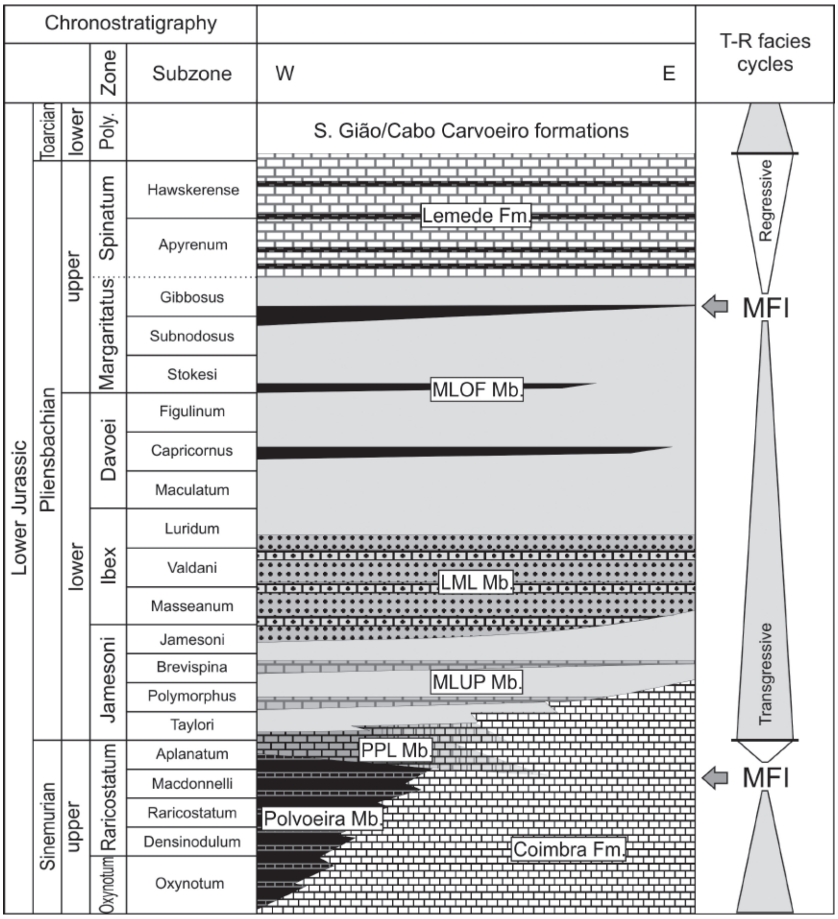

FIGURE 7 | **General facies stratigraphic chart and 2nd-order transgressive–regressive sequences for the uppermost Sinemurian–lowermost Toarcian of the Lusitanian Basin. Arrow: major black-shale intervals; MFI: maximum flooding interval (abbreviations as explained in text).**

Sinemurian-base of upper Sinemurian (Obtusum Zone) carbonate facies of the Coimbra Fm. in Praia da Concha (North of S. Pedro de Moel), correspond to the onset of a large transgressive event, with the occurrence of several microbial structures and stromatolitic mounds, which possibly marks the basal limit of the Sinemurian T-R facies cycle. The middle-top of the Oxynotum Zone (?), which marks the base of the Água de Madeiros Fm. in S. Pedro de Moel (Duarte and Soares, 2002; Duarte et al., 2004b), is represented by the first occurrence of marl/limestone alternations of the LB, very rich in benthic macrofauna (mainly brachiopods and bivalves). This lutitic sedimentation is also coupled with an increase in organic matter content and TOC values (marls generally above 1%). Despite the high TOC values (maximum of 22%) observed at some levels (Fig. 5), organic-rich facies are restricted to the S. Pedro de Moel area, which confirms a local hemipelagic deposition (Figs. 6A-B, 7). During the Raricostatum Zone (upper part of the Polvoeira Mb.), the great abundance of ammonites and calcareous nannofossils (Duarte et al., 2006), the sharp decrease in benthic macrofauna and the spreading of black shales to other parts of the basin (maximum TOC values of 15% in S. Pedro de Moel, 8.4% in Montemor-o-Velho and 4.1% in Peniche) are criteria to mark the maximum flooding interval of this T-R facies cycle. Occurrences of *Paltechioceras* gr. *tardecrescens* (HAUER) and *Gleviceras guibalianum* (D'ORBIGNY) date this event in the Aplanatum Subzone.

The regressive phase of this cycle corresponds almost fully to the lower part of the Praia da Pedra Lisa Mb. However, in S. Pedro de Moel, this progradational phase appears to begin in the middle-upper part of the Polvoeira Mb. (Fig. 7), marked by a slight decrease in TOC values (Fig. 5) and an increase of limestone (Fig. 4). The above mentioned facies of the Praia da Pedra Lisa Mb. (thin bedded micritic and microsparitic limestones in Fig. 4), clearly indicate an evolution towards a shallow-marine palaeoenvironment. The last level with this kind of facies, recorded at the Água de Madeiros beach (e.g. Duarte and Soares, 2002; Duarte et al., 2006), marks the top of the Sinemurian T-R facies cycle. This limit is overlain by decimetre-thick argillaceous limestones, interbedded with centimetre-thick grey shales, and shows a gradual upward increase in the clay and macrofossil contents (base of SP in Duarte et al., 2004b). Laterally, the top of the Sinemurian T-R facies cycle is composed by bioclastic/packstones-grainstones.

Pliensbachian 2nd-order T-R facies cycle

The basal limit of the Pliensbachian T-R facies cycle, dated roughly from the Sinemurian/Pliensbachian boundary, is well observed in the S. Pedro de Moel area (Água de Madeiros beach) and at Peniche, and represents the first 2nd-order flooding event recognized at a basinal scale (Fig. 6D; see Duarte et al., 2004b; Duarte, 2007). The Pliensbachian succession shows a typical 2nd-order sequence, with dominant marly deposition at the base (lower Pliensbachian to lower upper Pliensbachian) and a calcareous dominant facies at the top (uppermost Pliensbachian to extreme base of early Toarcian).

The Pliensbachian series shows a large transgressive phase that includes marlstones, argillaceous and lumpy limestones, grey to dark marls and thin (millimetre-thick) black shales, ranging from the Jamesoni to Margaritatus zones (Figs. 6D-F). In the LML Mb., the interplay of these lithofacies (lumpy marls and limestones and grey marls) has been well observed and studied at Peniche and Figueira da Foz (Elmi et al. 1988; Fernández-López et al. 2000). According to Fernández-López et al. (2000) the series is organized into sets (4th-order sequences) of elementary sequences (5th-order sequences), each one comprising at the base a grey marly interval (sometimes with TOC above 1%) and end by an increase of calcareous and lumpy facies. Thus, TOC variation in LML Mb. seems to be coupled with this short-term cyclicity.

This 2nd-order transgressive phase is characterized by an increase in marly deposits observed in the basin (excepting Arrábida). In distal regions, facies show a marly dominance, with the development of several black-shale intervals, particularly abundant in the Davoei Zone and at the base and top of the Margaritatus Zone (Stokesi Subzone and top of Subnodosus-base of Gibbosus subzones respectively) (Figs. 5 and 7). With the exception of Arrábida and Tomar, the last black-shale interval is widespread in the basin, and corresponds to the peak transgression of the Pliensbachian T-R facies cycle (SP maximum flooding interval in Oliveira et al., 2006; Duarte, 2007)

During the Spinatum Zone (Lemede Fm.) the sedimentation returned to a calcareous regime (Figs. 6G and 7), with low TOC values and very rich in benthic macrofauna. In the southeastern sectors of the basin, this regressive phase is represented by shallow-water bioclastic/grainstone facies. The upper limit of this 2nd-order cycle, clearly observed throughout the basin (DT1 *in* Duarte, 1997; SBT1 *in* Duarte et al. 2007a), is dated from the lowermost Polymorphum Zone.

DISCUSSION: COMPARISON TO OTHER WESTERN EUROPEAN BASINS

As demonstrated above, the organic sedimentation that occurred during the late Sinemurian-Pliensbachian of the LB is associated with marine peak transgressions (mainly of 2nd-order scale), which probably caused significant oxygen oscillations in the marine bottom water. The transgressive phases that culminate during the Raricostatum and Margaritatus Biochrons appear to have favoured a restricted circulation on the sea bottom, probably as a result of water mass stratification (Hallam and Bradshaw, 1979; Wignall, 1991). Although this discussion is not the central goal of this paper, this palaeoenvironmental interpretation of oxygen-deficient conditions is particularly well supported by benthic macro- and microfauna distribution in the basin. Indeed, brachiopods (mainly terebratulids and rhynchonellids), bivalves (mainly ostreids) and gastropods, locally very abundant in several parts of the succession, are absent in the black shales and in the horizons with the highest TOC values. In addition, this relation between fauna and aerobic conditions is confirmed by three previous studies on benthic microfauna. Brunel et al. (1998) and N'zaba-Makaya et al. (2003) show for the Margaritatus Zone of the LB a clear decrease in foraminifera and ostracods from the eastern sectors towards S. Pedro de Moel. This idea tallies well with that of Duarte et al. (2006) who, in a preliminary study on the Sinemurian-Pliensbachian boundary of S. Pedro de Moel, show that ostracods are totally absent in the organic-rich facies (with TOC values above 1%) of the Raricostatum Zone. These microfossils are particularly abundant in the intercalated limestones, a fact that seems to confirm oxygen depletion on the sea floor during organic accumulation. These oxygen-deficient conditions may also be inferred by the occurrence of *Chondrites* in some organic-rich levels (see Bromley and Ekdale, 1984; Savrda et al., 1991).

In the Lower Jurassic, organic-rich marine facies are geographically widespread, associated with the Early Toarcian Oceanic Anoxic Event (T-OAE; Jenkyns, 1988; Jenkyns et al., 2002). In spite of this large organic matter sedimentation, which presents a significant record in Central and Western Europe (e.g. Jenkyns et al., 2002; among others), black shales are practically absent in the Portuguese Lower Toarcian (Duarte et al., 2004a, 2007a), probably as a consequence of the siliciclastic deposition that occurred during the base of the Levisoni Biochron (see Wright and Wilson, 1984; Duarte, 1997; Duarte et al., 2004b, 2007a; Suan et al., 2008). However, this OAE is well recognized in the LB, through the negative carbon isotopic signatures in both carbonates and wood, as confirmed by several authors (Duarte, 1998; Duarte et al., 2007a; Hesselbo et al., 2007). In this context, the organic-rich facies occurring in the late Sinemurian-Pliensbachian interval constitute the main reference of deposition and preservation of marine organic matter in the LB. These deposits correspond to an important hydrocarbon source rock (Oliveira et al., 2006).

In terms of Tethyan and Boreal realms, organic-rich marine facies of the late Sinemurian-Pliensbachian time interval are very rare (Jenkyns et al., 2002). Although these facies are absent in the Iberian Range (e.g. Gómez and Goy, 2005), Betic Cordillera (e.g. Ruiz-Ortiz et al., 2004) and Algarve Basin (Azerêdo et al., 2003; Ribeiro and Terrinha, 2007), they are well developed in northern Iberia, in the Asturian and Basque-Cantabrian basins, as confirmed by several authors (e.g. Borrego et al., 1996; Herrero, 1998; Quesada et al., 1997, 2005; Perilli and Comas-Rengifo, 2002; Rosales et al. 2006; Bádenas et al., 2009). The sedimentary context of the latter occurrence is very similar to that of the LB, which constitutes an interesting palaeogeographic and palaeoceanographic feature, considering the relative proximity of these two basins. However, comparison of the studied stratigraphic parameters in each basin shows several differences between these two sectors of the Iberia Peninsula. According to Fig. 8, high-resolution stratigraphic control shows clear discrepancies in the distribution of black shales in these two basins and TOC values are notably higher in the western Iberian margin. As a consequence of this facies record, the sequence stratigraphic interpretation is also different between them (Quesada et al., 2005; Rosales et al., 2006). They also differ from the sequential schemes proposed for other Iberian basins (Aurell et al., 2003; Gómez and Goy, 2005; Quesada et al., 2005), Morocco (Souhel et al., 1998) and from the sequential (transgressive-regressive facies cycles) general schemes presented for the Boreal and Tethyan realms by Hardenbol et al. (1998) (Fig. 8). However, according to the same figure, and despite the different number and length of sequences, some interesting correspondences are evidenced. 2nd-order maximum flooding events recognized in the LB are practically synchronous in the Basque-Cantabrian Basin. The connection between western and northern Iberia is also confirmed by the ammonite palaeobiogeographic distribution. According to Dommergues and El Hariri (2002), the ammonite fauna of the latest Sinemurian and of much of the Pliensbachian of the LB shows a clear influence of NW Europe, with the occurrence of some endemic species. This boreal palaeobiological influence ended definitively in the Spinatum Zone, after which Tethyan ammonites dominated. The latter palaeobiogeographic evidence is significant, correlated with the 2nd-order regressive phase observed in the LB, and is effectively common to both Boreal and Tethyan realms (Hardenbol et al., 1998). This phase is also justified by a cooling effect (Morard et al., 2003), as documented by recent published data on oxygen isotopes from the Basque-Cantabrian and Lusitanian basins (Rosales et al., 2004, 2006; Oliveira et al., 2009) (Fig. 8). Thus, with all these stratigraphical and geochemical data, we demonstrate that, during the late Sinemurian-Pliensbachian, the LB shows more palaeoceanographic affinities with the Basque-Cantabrian basin than with any other palaeogeographic domain.

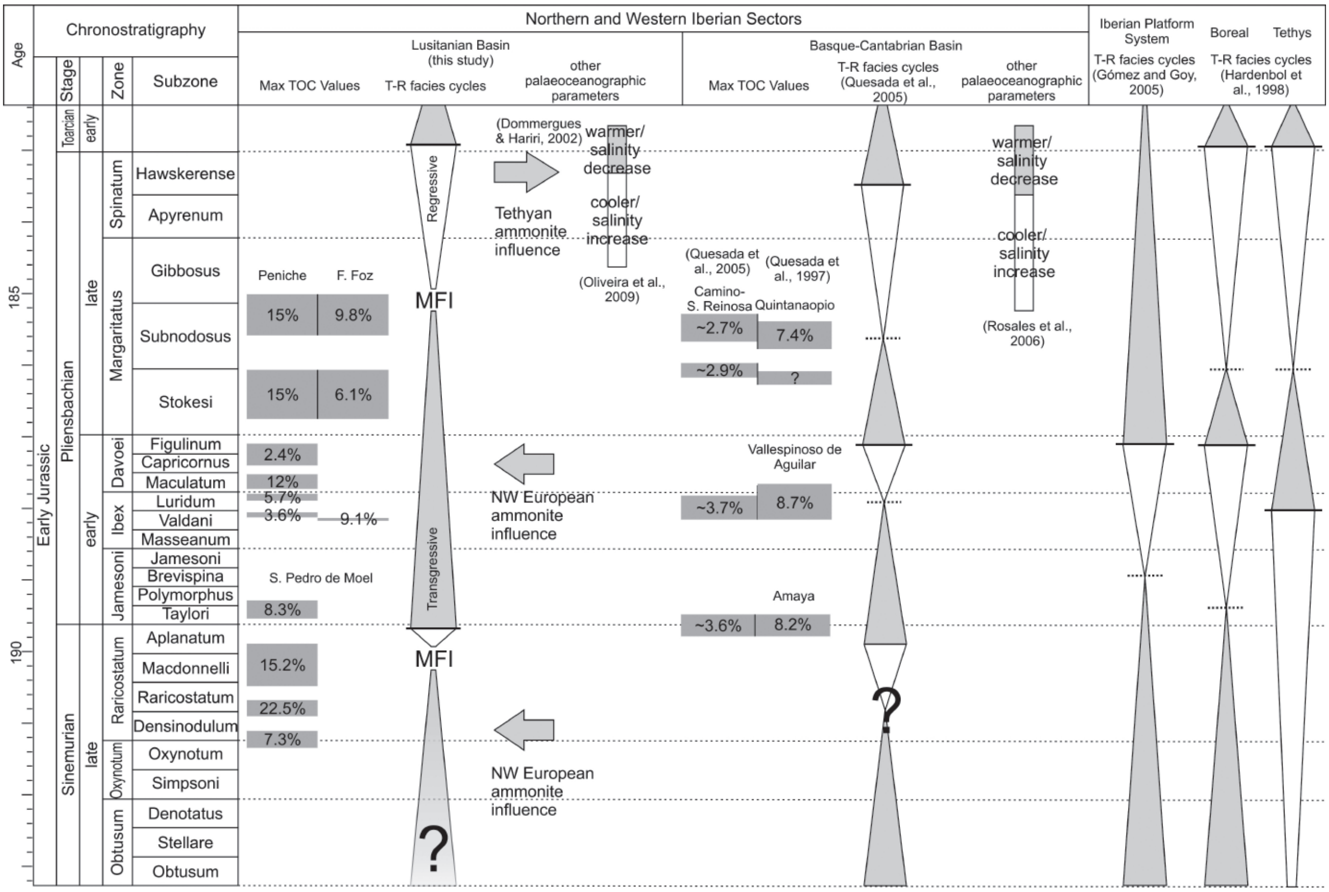

FIGURE 8 | **Correlation of black-shale (and its highest TOC values) intervals between the Lusitanian and Basque-Cantabrian basins, and comparison of the sequence stratigraphic interpretations for several domains of Iberia with the general schemes of Hardenbol et al. (1998). This chart shows the good correspondence between northern and western Iberia margins**

CONCLUSIONS

The upper Sinemurian-Pliensbachian (Água de Madeiros and Vale das Fontes formations) series of the LB is mainly composed of marly limestone sediments, deposited in a homoclinal carbonate ramp system. This sedimentary record, controlled by an accurate ammonite biostratigraphic resolution, includes organic-rich facies with several black shale levels. The stratigraphical, sedimentological and geochemical TOC study of the uppermost Sinemurian-Pliensbachian classical sections of the western Iberian margin, shows that these organic-rich facies occur practically throughout the whole succession (Oxynotum-Margaritatus zone interval), with the exception of the uppermost Pliensbachian (Spinatum Zone). The vertical and lateral distributions of TOC values show that these facies are essentially restricted to the western side of the basin and are particularly well developed in the Oxynotum-Raricostatum and the top of Ibex-Margaritatus zone intervals. Apart from the significance of these results with regard to palaeogeographical interpretation of the basin, the temporal and spatial distribution of these organic-rich facies is decisive in sequence stratigraphic interpretation of the studied series.

The observed persistence of black shales in the Oxynotum-Raricostatum and Margaritatus zones, where TOC values reach 22% and 15%, respectively, are both associated with 2nd-order transgressive phases. In addition, the density of high TOC values observed throughout the basin, associated with other sedimentological arguments, confirm the Aplanatum Subzone and the Subnodosus-Gibbosus subzone boundary as the 2nd-order maximum flooding intervals. In spite of the differences between the sequence stratigraphic schemes presented by several authors for Western Europe, these phenomena recognised in the LB are very similar to those recorded in the neighbouring Basque-Cantabrian Basin.

ACKNOWLEDGMENTS

We wish to thank René Rodrigues from the Universidade do Estado do Rio de Janeiro (Brazil) and Cenpes-Petrobras for technical support in the TOC analysis. We also thank M. Aurell, H. Jenkyns, I. Rosales and the editor L. Cabrera for constructive comments that greatly improved the manuscript. This research is a contribution to the Projects PDCTE/CTA/44907/2002 (Foundation for Science and Technology, Portuguese Government) and CGL2008-03112/BTE (Ministry of Science and Technology, Spanish Government).

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Manuscript received March 2009;
revision accepted December 2009;
published Online June 2010.