Geologica Acta, Vol.5, Nº 1, 2007, 59-75
Available online at www.geologica-acta.com

Fifth-order cyclicity and organic matter contents relationship (Lower Eocene, Pyrenees)

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ABSTRACT

The Upper Limestone Member of the Corones Formation of the Spanish Pyrenees consists of various units (Lower and Upper Foraminifera Units, Shale Unit, Cherty-ostracode Unit, Ostracode Unit and Chara-ostracode Unit) and offers strong facies and lateral thickness (20 to 80 m) variations. Detailed facies analyses, fifth-order cycles and organic geochemical determinations in the central domain of the Corones platform carbonates (Cherty-ostracode Unit), lower Eocene in age, were carried out to establish a case of close relationship between variations in organic matter productivity and cyclicity with annual period. The Cherty-ostracode Unit displays a continuous and pervasive fifth-order cyclicity, represented by 5 cycles. Each cycle consists of a lower part (mollusc facies) and an upper part (laminated ostracode facies). The calculated fifth-order cycle period ranges from about 17,000 to 28,000 years, which falls within the Milankovitch Band. Variations in organic matter content related to these carbonate cycles have been established. The lower mollusc facies members show a low organic carbon content and Hydrogen Index (HI) below 0.6% in weight and 261, respectively. By contrast, the upper laminated ostracode facies members show high organic carbon contents (up to 2% in weight) and high HI (between 164 and 373), and are also characterized by important silicification processes (the content in chert is up to 30%). The organic geochemistry resulting from these organic rich levels reflects a contribution of algal marine input.

KEYWORDS **Cherty-ostracode facies. Organic geochemistry. Cyclicity. Corones Formation. Eocene.**

INTRODUCTION

The geometric relationships between regressive progradational deltaic systems and transgressive carbonate platforms are quite common in the geological record in different tectonosedimentary environments. In the foreland basins, siliciclastic progradations have been related to emplacement during tectonically active periods while carbonate platforms have been related to more stable tectonic periods (Puigdefàbregues et al., 1986; Tankard, 1986).

The Lower-Middle Eocene deposits in the structural Cadí Unit of the southern Pyrenean foreland basin show an alternance between regressive progradational deltaic systems and transgressive carbonate platforms. The Sagnari Formation (Fm), in part, and the Corones Fm (Upper Ilerdian - Lower Cuisian in age) constitute the best deltaic carbonate platform example in this structural Cadí Unit.

Carbonate platforms related to deltaic systems present wide facies variations throughout their geological record (Andrews and Walton, 1990), and during the Cenozoic period these platforms frequently show reefal deposits (Choi and Ginsburg, 1982; Santisteban and Taberner, 1988; López, 1991) which have been well studied. By contrast, the inner and/or restricted carbonate platform facies are often less studied. The Upper Limestone Member of the Corones Fm is a good example of a Cenozoic carbonate platform with inner and/or restricted facies.

Ostracode-rich facies generally constitute sparse thin beds associated with main varied deposits, as a result of local restricted conditions (generally brackish or hypersaline subenvironments). Thick ostracode-rich deposits are fairly uncommon in geological records. The Upper Limestone Member of the Corones Fm presents two ostracode-rich units clearly differentiated and extensively developed.

The aims of this paper are: (a) to analyse the facies, diagenesis and organic geochemistry of the Upper Limestone Member of the Corones Fm, Lower Cuisian (Lower Eocene) in age, in the Central and Eastern Pyrenees, (b) to discuss global cyclicity in the Upper Limestone Member and define fifth-order and other minor order cycles of the Cherty-ostracode Unit of this member, and (c) to determine relationships between the organic geochemical composition of the Cherty-ostracode Unit of the Upper Limestone Member and both the various studied facies and fifth-order cyclicity. This paper is also a contribution to the ongoing study of restricted carbonate platform facies.

GEOLOGICAL FRAMEWORK

Structural setting

The Central and Eastern Pyrenees are made up of two main types of thrust sheets (Muñoz, 1985; Muñoz et al., 1986; Muñoz et al., 1988; Fig. 1A). The Upper Thrust Sheets consist of a mainly Mesozoic cover emplaced during the Lower Eocene, leading to the development of the southern Pyrenean foreland basin. The Lower Thrust Sheets (Gavarnie, Les Nogueres and Cadí units) are composed of Paleozoic materials and include the Paleogene materials of the foreland basin as a result of their emplacement in piggy-back sequence. The principal outcropping structural unit of the Lower Thrust Sheets in the southeastern Pyrenees is the Cadí Unit.

The Cadí Unit, which extends over 110 km, consists of Paleozoic materials, a thin Mesozoic section, Garumnian facies and a thick Lower and Middle Eocene series (Fig. 1B). It is bounded to the north by the Serra Cavallera thrust (Muñoz, 1985) and to the south by the Vallfogona solethrust. The internal structure of the Cadí Unit is characterized by an open E-W trending syncline known as the Ripoll syncline (Muñoz et al., 1986; Vergés and Martínez, 1988).

The Albanyà fault, NNW-SSE, separates the Cadí Unit into two areas: La Garrotxa to the west and Empordà to the east. The Eocene record shows significant structural, stratigraphic and sedimentological differences between the two areas (Pujadas et al., 1989). The studied area extents along the Cadí Unit, in the Central and Eastern Pyrenees (Fig. 1).

Stratigraphic setting

The classical Lower Eocene lithostratigraphic units and/or formations in the Cadí Unit include the Cadí and Sagnari, Corones, Armàncies and La Penya, Vallfogona, and Beuda Fms. The age of these Fm ranges from Lower Ilerdian to Lower Lutetian (Fig. 2).

The Upper Cretaceous - Middle Eocene deposits (up to 3,000 m in thickness) of the Cadí Unit consist of several depositional sequences (Puigdefàbregues et al., 1986; Giménez, 1993). The siliciclastic deposits (mainly deltaic systems) record tectonically active periods, while the carbonate platforms record the more stable periods.

The Cadí Fm, defined by Mey et al. (1968), consists mainly of 300 m thick Nummulite and Alveolina limestones. It is characterized by a transgressive carbonate section which runs from peritidal restricted (ostracode-miliolid-oolite facies) to a shallow high-energy carbonate platform. It passes laterally and vertically to the Sagnari Fm (Solé Sabarís, 1958; Fig. 2), which consists mainly of marls and marly limestones with Nummulites up to 800 m thick. The upper boundary of the Sagnari Fm is transitional towards the overlying Corones Fm. Both the Cadí Fm and the Sagnari Fm are Lower-Upper Ilerdian in age (Hottinger, 1961).

The Corones Fm, defined by Solé Sabarís (1958), is composed of a sandy deltaic unit and a carbonate platform unit (Puigdefàbregues et al., 1986; Giménez, 1993). The thickness of this formation varies from 350 m in the centre of the Cadí Unit to 70 m in the east and west. The lower boundary is a sharp marine flooding surface with respect to the overlying Armàncies Fm. The age of the Corones Fm is Lower Cuisian (Giménez, 1993; Giménez-Montsant and Salas, 1997).

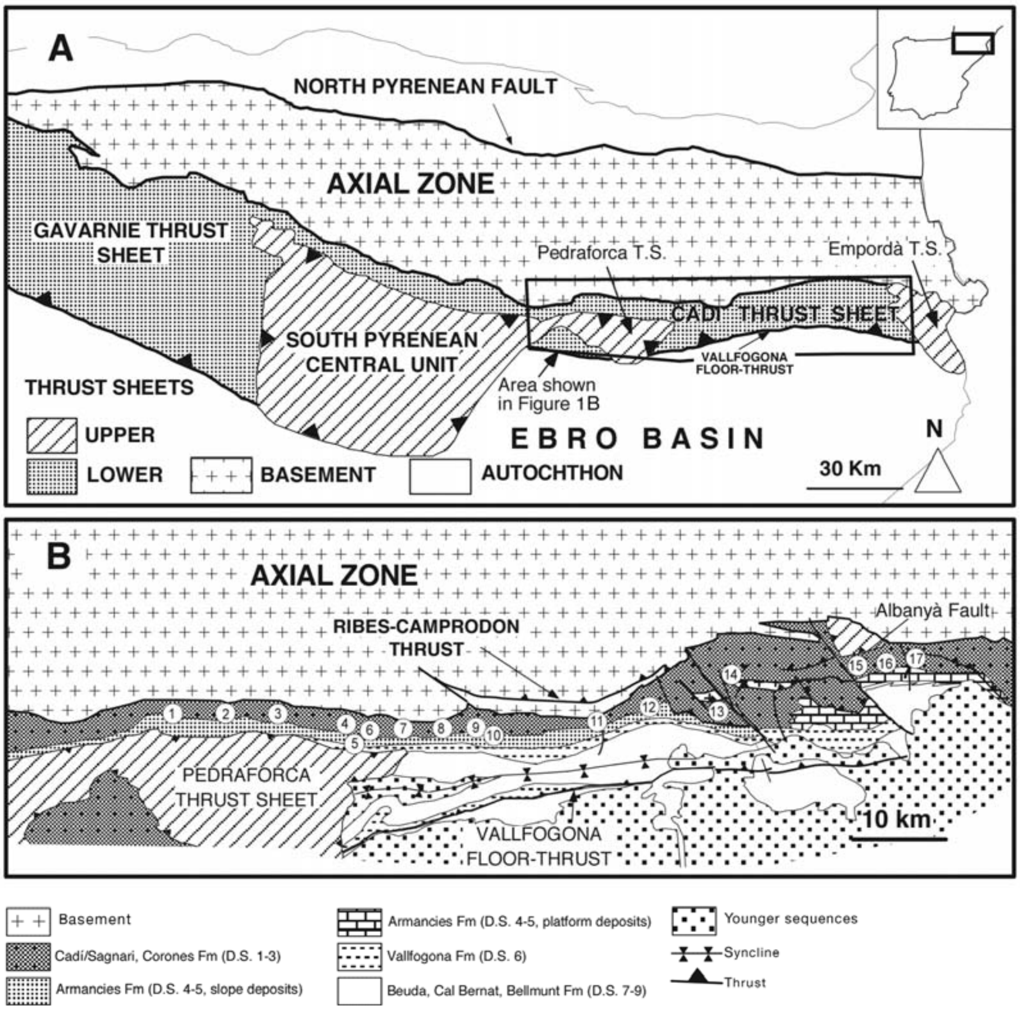


FIGURE 1 | **A) Synthethic structural map of the Eastern Pyrenees (modified from Giménez-Montsant and Salas, 1997). B) Geological map of the Cadí Unit (E Pyrenees). Stratigraphic sections represented in Figs. 2 and 3. 1: Adraen; 2: Cornellana; 3: Baridana; 4: Bagà; 5: Paller; 6: Riutort; 7: Pobla de Lillet; 8: Arija river; 9: Gombrén; 10: Ripoll; 11: Ogassa; 12: Sant Pau Seguríes; 13: Oix; 14: Lierca river; 15: Albanyà; 16: Mas; 17: Terrades. Studied sections in this paper are from Baridana (3) to Oix (13).**

The Armàncies Fm, formally defined by Gich (1969), is mainly composed of marly and carbonatic sediments, from 500 to 600 meters in thickness. This formation, which includes deposits from a middle-outer ramp, slope, and outer slope fan, as well as basin deposits, represents the flexure effect of the northern thrust sheet emplacement and the destruction of the southern carbonate platforms. The Armàncies Fm grades laterally into La Penya Fm, which was defined by Estévez (1973). It is made up of shale-sandstone and/or limestone thickening coarsening sequences up to 500 meters in thickness and is interpreted as a deltaic system followed by several carbonate platforms. The carbonate platform units are composed of Assilina-Nummulite facies and are interpreted as open marine carbonate platforms. Both Armàncies and La Penya Fms are Middle-Upper Cuisian in age (Busquets, 1981).

METHODS AND MATERIALS

Several stratigraphic sections from the Upper Limestone Mb of the Corones Fm were studied in its main outcrops (Fig. 1B), and approximately 400 samples were collected from the main stratigraphic sections (Fig. 1B; sections 3-13; that correspond to the so-called *central domain* from Fig. 3), including limestones, cherts, marls and lutites. The samples were mineralogically and petrologically studied by means of X-Ray diffraction, polished slabs and standard and dual-stained thin sections. Selected samples were examined using a cathodoluminiscence microscope (Technosyn Cold Cathodoluminiscence model 8200 MkII operating at 16-19 kV and 350 μA gun current) and a scanning electron microscope (SEM-EDS JEOL JSM-840). Selected chert-rich limestones were submerged in HF 29M for 10 min in order to evidence their internal structures.

Micropalaeontological content of the samples (mainly ostracode and foraminifera) was separated from the rock by disaggregation in water and wet-sieving through 63 μm nylon mesh. A separated fraction was dried at 40-50°C and examined in order to analyse the type/degree of transportation of particles and the classification of clastic and non-clastic content.

Fifth-order cycles from the Cherty-ostracode Unit were defined on the basis of the identified lithofacies, geochemical characteristics and associated lithologies, and the estimated interval of time for each individual cycle. Minor sixth and seventh-order cycles were also identified and discussed. Because absolute ages of both the Upper Limestone Member and the Cherty-ostracode Unit are unknown, the duration of the individual fifth-order cycles was calculated in accordance with the calculation of the accumulation rate for the whole Corones Fm, assuming that this rate remained constant during this stratigraphical interval, which, on the other hand, represents a well-constrained interval of time. The sediment accumulation rate (s.a.r.) can be calculated using the equation (1).

$$s.a.r = \frac{P}{T} \quad (1)$$

where P is the decompacted thickness in meters of a given stratigraphic interval, and T the duration of this stratigraphic interval.

Previously published sedimentation rates for the Corones Fm were also considered, presuming that this rate was constant. Decompacted thickness was estimated using decompaction curves of Sclater and Christie (1980). No partial compaction or overcompaction was assumed.

Organic geochemistry was used to determine closer relationships between the sedimentary facies, the cycles and the related environments. For this, one hundred samples from five sections were analyzed. The organic carbon contents were measured by Rock Eval Pyrolysis, following the procedure described by Espitalié et al. (1977, 1985/86). Extractable organic matter (EOM) of the samples was separated and fractionated by liquid chromatography in saturated and aromatic (SAT and ARO) hydrocarbons and polar compounds (NSO). Saturated hydrocarbons were analysed by gas chromatography.

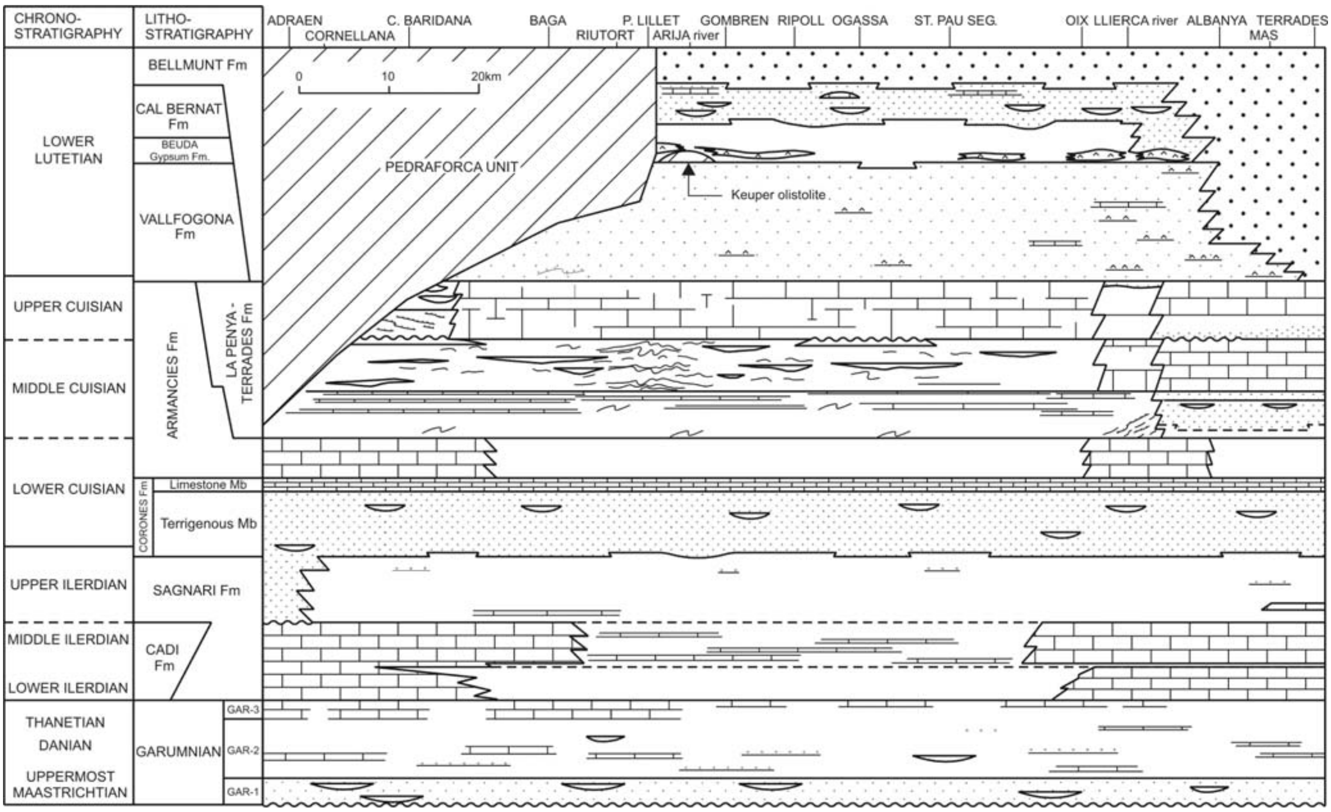


FIGURE 2 | **Lithostratigraphic and chronostratigraphic diagram of the Cadí Unit (Lower-Middle Eocene, Pyrenees; modified from Giménez, 1993 and Giménez-Montsant et al., 1999). This paper is focused in the central domain (Baridana-Oix sections).**

THE CORONES FORMATION

The Corones Fm, Lower Cuisian in age and varying in thickness from 350 m in the center of the Cadí Unit to 70 m to the east and the west, may be divided into two members (Giménez, 1993): from base to top, the Lower Terrigenous Mb and the Upper Limestone Mb (Fig. 2).

The Lower Terrigenous Mb is mainly composed of sandstones with interbedded shales, siltstones and limestones, with a thickness ranging from 30 to 290 m. It is interpreted as a progradational deltaic system in a lowstand condition (the underlying Sagnari Fm is identified as prodelta environment) and is made up of two units: the Lower Grey Terrigenous unit and the Upper Red Terrigenous unit. The Lower Grey Terrigenous unit, up to 200 m in thickness, consists of thickening coarsening-upwards sandy stackings often capped by limestones (oolitic and bioclastic wackestones and packstones). The lower boundary is transitional respect to the marls of the Sagnari Fm while the upper boundary consists of two meters of green bioturbated shales with sparse mollusc fauna. This unit is widely interpreted as a lower delta plain. The Upper Red Terrigenous unit is composed of bioturbated siltstones, massive channelized coarse sandstones and intense hydromorphic traces. Its thickness varies from 40 to 95 m and it is interpreted as a fluvial-dominated upper delta plain.

The Upper Limestone Mb is mainly composed of limestones and shales. It extends over 115 km and increases in thickness from west to east. It is a shallow carbonate platform, interpreted as a transgressive systems tract, with sharp lower and upper boundaries. From west to east, the Upper Limestone Mb presents three stratigraphic and sedimentological domains with lateral variations (Giménez-Montsant et al., 1999; Fig. 3). The westernmost domain only includes Lower and Upper Foraminifera Units, intercalated by a thin Cherty-ostracode Unit. The central domain includes four units which, base to top, are as follows: Lower Foraminifera Unit, Shale Unit, Cherty-ostracode Unit and Upper Foraminifera Unit. The easternmost domain consists of Ostracode Unit, Lower Foraminifera Unit, Shale Unit, Chara-ostracode Unit and Upper Foraminifera Unit. The thickness of the Upper Limestone Member varies considerably from the western domain (20 m) to the eastern domain (80 m). These lateral changes (facies and thickness) may be attributed to tectonic control.

THE UPPER LIMESTONE MEMBER: FACIES ANALYSIS OF THE UNITS OF THE CENTRAL DOMAIN

The Lower Foraminifera Unit, 7 to 11 m in thickness, has mainly foraminifera facies, bedded in layers from 0.5 to 3 m. This unit is often arranged in thickening coarsening shoaling upwards cycles, generally from foraminifera wackestones-packstones at the base to foraminifera grainstones at the top. The lower boundary is a sharp contact with respect to the underlying Lower Terrigenous Mb, while the upper boundary is rapid but transitional.

The Shale Unit, from 10 to 15 m thick, is the most continuous and homogeneous in the Upper Limestone Mb. It consists mainly of grey shales and siltstones, with scattered beds and common vegetal debris. The upper part often presents laminated ostracode facies levels.

The Cherty-ostracode Unit, 9 to 30 m thick, consists of well-bedded limestones, up to 0.5 m thick, with some scattered centimetric-decimetric shale intercalations. The limestones present two main facies: laminated ostracode facies and mollusc facies. These facies occur forming cycles, 0.2 to 2 m thick. Towards the east, the Cherty-

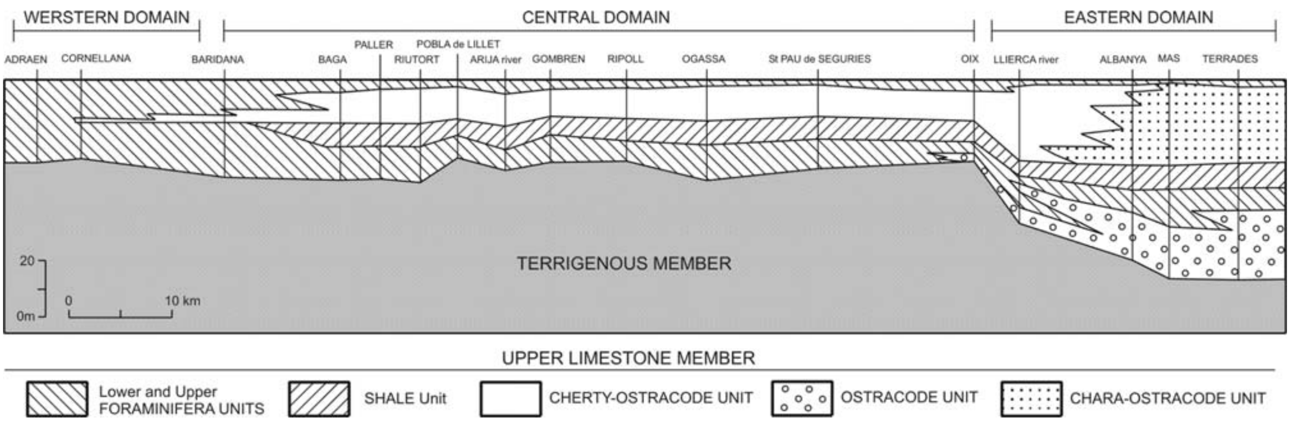


FIGURE 3 | **Stratigraphic units and sedimentological domains of the Upper Limestone Member of the Corones Fm.**

ostracode Unit grades to more lacustrine facies (Chara-ostracode Unit). Towards the westernmost zone this unit passes laterally to normal marine deposits (Upper Foraminifera Unit). The lower boundary of the Cherty-ostracode Unit is transitional with respect to the underlying Shale Unit and the upper boundary is transitional into the overlying Upper Foraminifera Unit. This transition, up to 1 m thick, consists of mixed dasyclad algae-ostracode-foraminifera packstone.

The Upper Foraminifera Unit, 2 to 4 m thick, consists principally of foraminifera facies, arranged in beds from 0.5 to 2 m in thickness. This unit does not show important lateral facies changes.

Lower and Upper Foraminifera Unit facies

The principal facies of these two units is the foraminifera facies (Fig. 4A). This facies consists of bedded grey to ochre layers from 0.3 to 4 m in thickness. Texturally this facies varies from wackestones to grainstones. The main components are miliolids (from 50% to 70%), peneroplids (specially *Spirolina*), and opertobitolids. The secondary components are aglutinated foraminifera (*Crimovulimina*), discorbids, calcareous algae (red algae as *Lithoporella*, dasyclads), bivalves, and gastropods. The echinoderms, quartz grains, briozous and ooids are the accessory components.

The Lower and Upper Foraminifera Units are generally arranged in thickening coarsening shoaling upwards cycles. The cycles present erosive bases and mollusc lags, and consist of a lower foraminifera wackestone-packstone member and upper foraminifera grainstones. Occasionally, the limestone layers are interbedded by inframetric marly levels. The foraminifera facies has been divided as follows: foraminifera wackestone to grainstone and peloidal, and bioclastic packstone to grainstone.

Cherty-ostracode Unit facies

The Cherty-ostracode Unit (Fig. 4B), 9 to 15 m thick (but up to 30 m in the Llierca river section), displays 5 well-developed cycles in all the studied sections. Each cycle is composed of two members: a lower member, which consists of mollusc facies, and an upper member, which consists of laminated ostracode facies. The top of the Cherty-ostracode Unit presents a mixed dasyclad algae-ostracode-foraminifera facies.

Mollusc facies

The mollusc facies, grey to pale grey in colour, is up to 0.5 m in thickness. This facies presents thin 0.1 m thick beds composed of abundant bivalves and/or gastropods, and other components such as ostracodes and sparse foraminifera (Fig. 5A). In general, this facies presents disarticulated mollusc levels which are parallel to bed stratification without erosive bases. These beds can be interpreted as being due to wave action. Locally, the mollusc facies presents erosive bases, fining-upwards sequences, presence of intraclasts, disarticulated and/or fragmented bivalve shells, preferential orientation of bivalve shells along the stratification surface, creating a coarse lamination. This graded mollusc

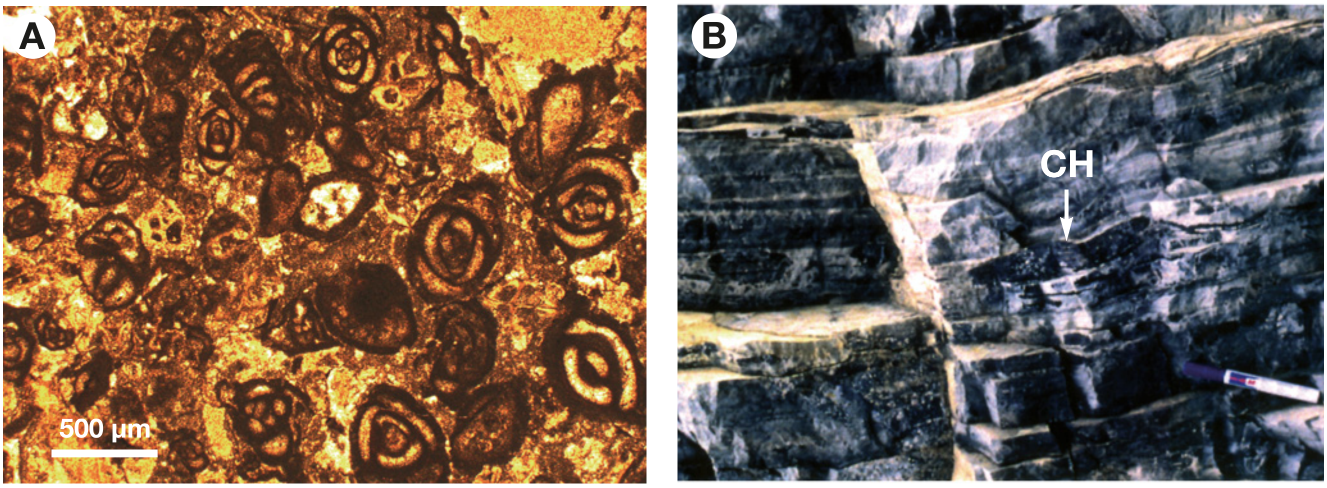


FIGURE 4 **A) Optical micrograph (plane-polarized light) of the foraminifera grainstone facies (Lower Foraminifera Unit). Sample ARC-64; Arija river section. B) Oucrop view of the Cherty-ostracode Unit (Upper Limestone Mb). CH: chert; Bagà section. The scale is given by a 14 cm long marking pen.**

facies is interpreted as an event deposit due to the influence of storms (Aigner, 1982). Mollusc facies are broadly interpreted as having been deposited in normal marine conditions.

Laminated ostracode facies

The laminated ostracode facies, generally dark grey to black in colour, is up to 1.9 m thick. This facies presents mil-

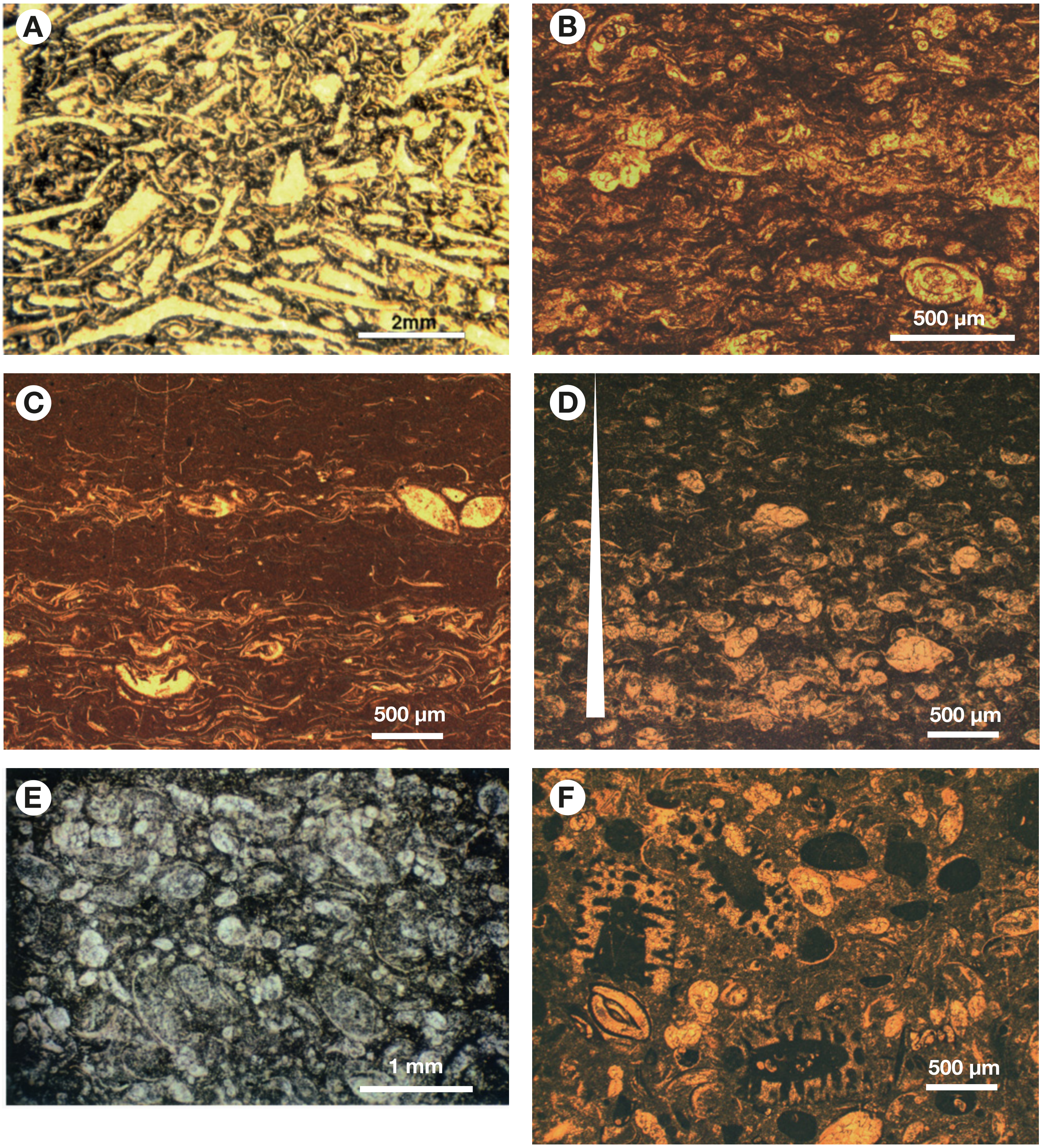


FIGURE 5 **Optical micrograph (plane-polarized light) of the facies of the Cherty-ostracode Unit; La Pobla de Lillet section. A) Mollusc facies. Sample PLC-64. B) Thin-bedded ostracode microfacies in the laminated ostracode facies. Sample PLC-70. C) Rhytmic ostracode microfacies in the laminated ostracode facies. Sample PLC-63. D) Fining-upwards sequence lamination microfacies in the laminated ostracode facies. Sample PLC-68. E) Mixed ostracode-discorbid microfacies in the laminated ostracode facies. Sample PLC-52. F) Mixed dasyclad algae-ostracode-foraminifera facies. Sample PLC-71.**

limetric and/or centimetric lamination and the laminae (lime mudstones - grainstones) are composed of monospecific accumulations of *Neocypridesis* sp. The laminated ostracode facies has been divided into four microfacies: 1) thin-bedded ostracode microfacies, 2) rhythmic ostracode microfacies, 3) fining-upwards sequence lamination microfacies, and 4) mixed ostracode-discorbid microfacies.

The thin-bedded ostracode microfacies is characterized by dense accumulations of mainly whole, and sometimes disarticulated, ostracodes (Fig. 5B). The thickness of these thin beds varies from a few millimeters to 2-5 cm. Texturally this facies is packstone-grainstone. The main component is ostracode shells (60-90%). Secondary components include peloids (locally abundant), foraminifera (common discorbid, sparse miliolid and other rare benthic foraminifera) and thin-shelled bivalves. The shells of the whole ostracodes show size gradation from 100 μm to 1 mm, the smaller sizes outnumbering the bigger ones. The carapaces are randomly distributed. This microfacies sometimes shows fining-upwards distribution (according to the number of shells) with a gradation from thin-bedded microfacies to laminated microfacies. The smaller whole ostracode shells may be interpreted as juvenile stages whereas the bigger ones may be interpreted as adult stages. The presence of whole shells and the high percentage of juvenile shells suggest sedimentation *in situ* (Reyment, 1960; Kilenyi, 1969, 1971; Oertli, 1971; Pokorny, 1978), and could also be the result of a high sedimentation rate (Oertli, 1971; Brasier, 1980). According to Wheatley (1988), high percentages of articulated juveniles indicate a high mortality rate in ontogeny. But despite the interpretation of the sedimentation *in situ*, some degree of transportation and/or reworking should not be ruled out. Kontrovitz (1975) demonstrated experimentally that the carapaces can be removed at low velocities, without disarticulation. In conclusion, the thin-bedded ostracode layers are interpreted as sedimentation *in situ* with some transport and/or reworking, according to the following characteristics: i) massive thin beds, and absence of lamination and tractive structures; ii) the ostracodes mainly present whole shells; iii) a high percentage of juvenile shells in relation to adult shells; and iv) ostracode whole shells distributed at random.

The rhythmic ostracode microfacies consists of the alternation of light grainy laminae and dark muddy laminae (Fig. 5C). The laminae boundaries are clear, especially the boundary between the muddy laminae and the grainy laminae. Texturally the light grainy laminae are packstones-grainstones of many whole and disarticulated and fragmented (compaction process) ostracode shells with a thickness that varies from 1 to 4-5 mm. The dark muddy laminae consist of lime mudstones-wackestones, with sparse disarticulated ostracode shells and some peloids. The thickness of the dark laminae varies from 0.2-0.3 to 2 mm. The repetition of the laminae suggests periodic changes in the physical-chemical and biological conditions of the depositional system. Seasonal control in ostracode associations has been cited by several authors (Wheatley and Wall, 1969; Carbonel et al., 1988; Peypouquet et al., 1988; Carbonel et al., 1990; Debeney et al., 1990). The light grainy laminae (the ostracode-rich laminae) indicate a high ostracode accumulation and consequently a high rate of ostracode production. According to Carbonel et al. (1988), ostracode abundance can reflect a good food supply and an adequate oxygen supply (at least enough for the ostracodes). According to Debeney et al. (1990), the main reproduction period of the recent Cyprideis mandviensis took place when salinity was at a minimum. The dark muddy laminae (the ostracode-poor laminae) may be interpreted as sediment produced in anoxic conditions (Peypouquet et al., 1988) with a higher organic matter content. The small number of ostracodes is attributed to adverse physical-chemical and biological conditions. The period with oxygenated, low salinity waters and wet climate is interpreted to be the winter season (light grainy laminae) whereas the period with anoxic, eutrophic waters and dry climate is interpreted to be the summer season (dark muddy laminae). The monospecific accumulations can be explained by salinity fluctuations.

The fining-upwards sequence lamination microfacies is composed of fining-upwards sequence laminated stackings (Fig. 5D). Each laminae is wackestone-grainstone at the bottom (light in colour) and lime mudstone-wackestone at the top (dark in colour). Thickness varies from 2 to 8 mm. The laminae have the following characteristics: i) erosive bases (on a microscopic scale); ii) millimetric intraclasts associated with the erosive bases and derived from the underlying sediment; iii) locally ostracode shells form symmetrical ripples with a wavelength up to 3 cm and a height up to 1 cm; iv) the ostracode shells vary from whole to extremely fragmented; and v) fining-upwards sequences are defined by shell abundance. At the base there are many broken shells and at the top fewer but more disarticulated shells. The high percentage of disarticulated and even broken ostracode shells is interpreted as a transport effect. Kilenyi (1969, 1971), Oertli (1971), Peypouquet (1971) and Wheatley (1988) consider that the number of disarticulated carapaces also increased due to transportation. Kontrovitz (1975) demonstrated that the valves required stronger currents than the carapaces to be transported. The sedimentological and textural features observed in this facies suggest deposition from density currents. Each fining-upwards sequence is interpreted as a "micro-storm event". The fining-upwards sequence stackings are attributed to the repetition of the same physical processes.

The mixed ostracode-discorbid microfacies is mainly composed of ostracodes and a variable proportion of discorbids (Fig. 5E). Miliolids and other rare benthic foraminifera also occur, and molluscs are not uncommon. This microfacies is often silty, several centimetres in thickness (less than 5 cm) and presents a laminated or massive texture. The laminated texture is characterized by alternances between ostracode-rich laminae and discorbid-ostracode laminae. The occurrence of discorbids and a very small proportion of molluscs could suggest a certain opening in the restricted environmental conditions, represented by the accumulations of ostracode valves.

Mixed dascyclad algae-ostracode-foraminifera facies

This facies, composed of dasyclad algae, ostracodes, foraminifera (common miliolid, and discorbid, peneroplid) and sparse mollusc fragments (Fig. 5F), only occurs in the transition between the Cherty-ostracode Unit and the Upper Foraminifera Unit. The thickness of this facies varies from 0.5 to 1 m and it is internally arranged in decimetric beds. Bioturbation is relatively abundant in this facies, and affects the underlying Cherty-ostracode Unit up to 1 m in depth. This facies is interpreted as a practically normal marine facies, slightly restricted but in oxygenated waters.

Chert petrology

The presence of chert is an important feature of the Cherty-ostracode Unit, especially of the laminated ostracode facies (Giménez-Montsant et al., 1999; Figs. 4B and 6A). The silicified facies amount to 20 percent in the laminated ostracode facies and 2 percent in the mollusc facies. The cherty horizons, a few mm to 10 cm thick, are separated from 1 to 50 cm by carbonate host-sediment. When the Cherty-ostracode Unit passes laterally to the marine facies (western part) and to the lacustrine facies (eastern part) the abundance of chert decreases sharply.

The chert horizons present three lithofacies: nodular, layered and mottled. Nodular chert is the most common (85%). Its arrangement seems to be controlled by bedding planes, with the nodules arranged parallel to the stratification. The nodules occur in a variety of shapes from subspherical to tabular. Locally, the nodular chert grades to layered chert. The nodules are up to 10 cm high and decimetric, and even metric, in length. The layered chert (10%), which occurs in layers up to 15 cm in thickness and with an important decametric lateral extension, has sharp lower and upper boundaries and includes ghosts of original lamination, ostracodes and sponge spicules. The mottled chert (5%) presents milimetric-centimetric irregular patchy morphology.

The silicified facies are composed of four petrographic types of quartz: microquartz, length-fast (LF) chalcedony, megaquartz and microspheres. The different types of quartz occur as replacive fabric and pore-filling (cement) fabric, filling the intraparticle porosity (ostracode carapaces), sedimentary cracks, veins and fracture porosities (Giménez-Montsant et al., 1999).

The chert commonly displays dense accumulations of spherical structures, 20 to 30 μm in diameter, showing an internal core and an external cortex. Spherical structures in some nodular chert occur mainly concentrated in laminae which, outside the chert, present common calcitized and/or dissolved spheres and spicule sponges. These spherical structures are similar to the quartz microspheres described by Oehler (1975) and Meyers (1977). Neverthe-

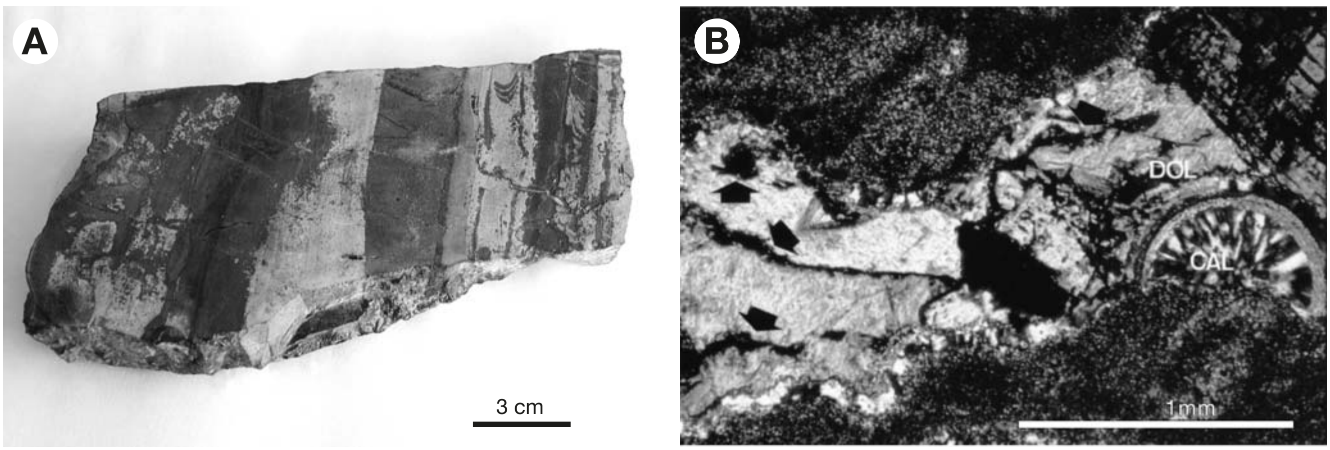


FIGURE 6 **Chert in the Cherty-ostracode Unit. A) Polished slab submerged in concentrated HF showing the alternation of rich-chert lamina (dark) and mudstone lamina (light). B) Optical micrograph (crossed-polarized light) of a chert nodule cut by late fractures, filled by LF-Chalcedony (CAL), megaquartz and saddle dolomite (DOL) with inclusions of hydrocarbons (in arrows).**

less, they also present many similarities with spherical spicules or spicular structures of certain sponges described by Cayeux (1929) (Sanfilippo, pers. comm.). The silica source of these cherts is the spicule sponge; the mobilization of the biogenic silica occurred in an early shallow diagenesis, with the consequent precipitation of chert, and continued to be precipitated into the burial diagenesis (Giménez-Montsant et al., 1999).

Very intact components (mainly ostracodes) are commonly shown in the silicified facies. By contrast, the non-chertified host-sediment can present extremely flattened and compacted ostracode, with the surrounding host-sediment being deformed and adapted to the nodules. These relationships show that chertification occurred before the main phase of compaction (Namy, 1974; Geeslin and Chafetz, 1982; Eley and Jull, 1982; Maliva and Siever, 1989).

Fractures affect the cherts and the host-sediment, and are characterized by the following pore-filling sequence: isopachous LF-chalcedony first cement generation, a second generation of megaquartz cement and, finally, baroque dolomite and/or ferroan calcite cements with common inclusions of hydrocarbons (Fig. 6B); hydrocarbons are also observed filling primary (intraparticle) and secondary porosity. The presence of these carbonate cements and their relationship to hydrocarbons may be regarded as markers of a certain thermal deep burial diagenesis (Giménez, 1993).

CYCLES AND CYCLICITY

Cycles description

Third-order and fourth-order depositional cycles

The Lower Terrigenous Mb of the Corones Fm is identified as a lowstand systems tract and the Upper Limestone Mb as a transgressive systems tract. The boundary between these two members is recognised as a transgressive surface. The Upper Limestone Mb ends in a sharp surface (maximum flooding surface), and the overlying Armancies Fm deposits are interpreted as the highstand systems tract. These systems tracts constitutes a third-order depositional Sequence (Giménez-Montsant and Salas, 1997; Vergés et al., 1998).

The Upper Limestone Mb consists of two deepening upwards megacycles, up to 30 m thick, which are fourth-order cycles widely studied in the Bagà-Oix area (Giménez, 1993). The Lower Formanifera Unit belongs to the lower cycle and the upper cycle includes the Shale, Cherty-ostracode and Upper Foraminifera Units. The megacycles vary in thickness and vertical facies distribution according to the three domains of the Upper Limestone Member.

Fifth-order depositional cycles

Minor-order cycles of the Upper Limestone Mb have been studied. Because fifth-order periodicity does not

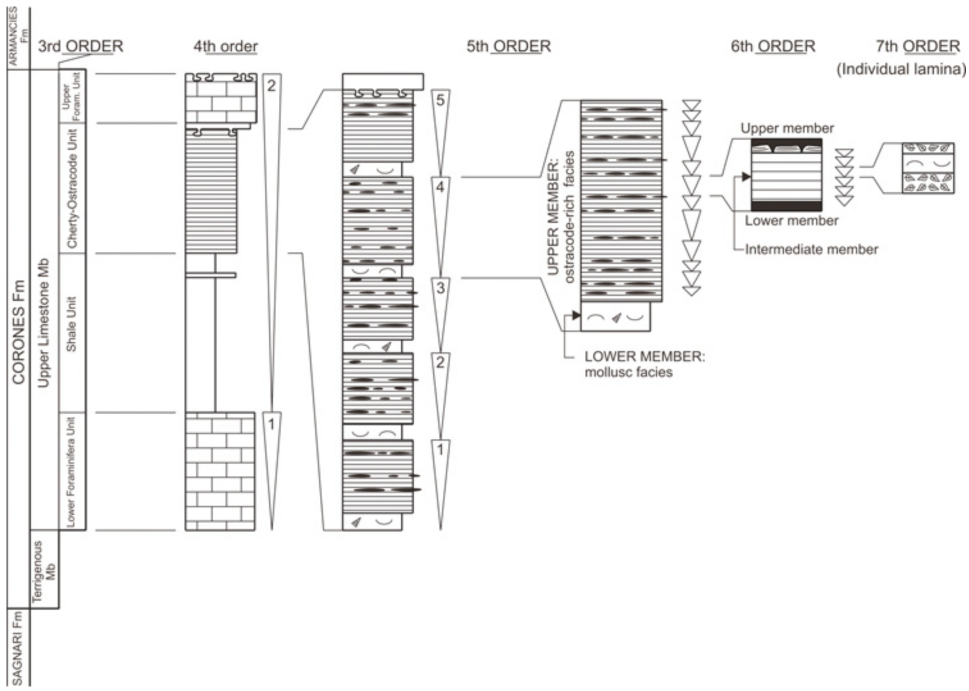


FIGURE 7 | **Cyclicity in the Cherty-ostracode Unit (Upper Limestone Mb, Corones Fm, central domain). See text and Figure 8 for scale and further explanation.**

appear as clear, cycles have been deduced from petrographic analysis (Giménez, 1993). The Cherty-ostracode Unit, nevertheless, shows an internal pervasive cyclicity arrangement, which consists of 5 cycles up to 2 m thick (Fig. 7). These cycles are very continuous in all the outcrops and extent up to 50 km. The characterization of the cycles is based on the following parameters: i) lithofacies, ii) presence or absence of chert levels, and iii) organic geochemistry analysis.

Each cycle is composed of two members: a lower member and an upper member. The lower member consists of a thin bedded mollusc facies, up to 0.5 m in thickness. Chert levels are absent or rare. The lower member facies are interpreted as shallow marine deposits (Fig. 8). The upper member, up to 1.9 m thick, is mainly composed of laminated ostracode-rich facies, with abundant chert levels. The upper member deposits are interpreted as a very shallow and restricted facies with important eutrophic conditions (Fig. 8).

The lower mollusc member is interpreted as the transgressive phase of the cycle, while the upper laminated ostracode member facies is interpreted as the regressive phase of the cycle.

Sixth-order depositional cycles

The laminated ostracode facies of each fifth-order cycle consists of several minor cycles interpreted as sixth-order cycles. Each cycle is composed of three members: lower, intermediate and upper (Fig. 7).

The lower member, which ranges from 2 cm to 10-15 cm in thickness, consists of laminated lime mudstone-wackestones with scattered sponge spicules and abundant cherts (mainly nodular and layered). The lower member facies are interpreted as normal marine deposits. The intermediate member only consists of millimetric and/or centimetric laminated ostracode facies. Texturally, this facies range from wackestone to grainstones, which are mainly composed of monospecific accumulation of *Neocypridesis* sp. The upper member is only locally present. This member shows mud-cracks, millimetric gypsum pseudomorphs, intraclasts and centimetric tepees. These structures also affect the upper part of the intermediate laminated ostracode member.

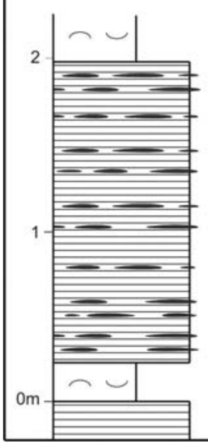
Seventh-order depositional cycles

The laminated ostracode facies of the Cherty-ostracode Unit presents several microfacies, but only the rhythmic ostracode microfacies shows a marked cyclicity, which has been interpreted as seventh-order cyclicity. The rhythmic ostracode microfacies couplet consists of two laminae: light grainy laminae and dark muddy laminae. The light grainy laminae, 1 to 5 mm thick, are packstones-grainstones of ostracode shells, whilst the muddy laminae, 0.2 to 2 mm thick, are lime mudstones-wackestones. This microfacies presents "varved" characters (Anderson, 1986; Fischer and Roberts, 1991; Ripepe et al., 1991).

Cycles quantification: Duration of the fifth-order cycles

Considering the constant accumulation rate in the Corones Fm, the absolute age of the individual cycles of the Cherty-ostracode Unit has been calculated in order to verify their fifth-order range of cyclicity:

1. Total accumulation rate. The accumulation rate has been calculated in the more stable areas of the Eocene basin in the Cadí Unit, from the Adraén to Oix sections.



FACIES	MICROFACIES	CHERT	ORGANIC MATTER	
			Organic carbon	Hydrogen Index
LAMINATED OSTRACODE FACIES	Thin-bedded ostracode microfacies Rhythmic ostracode microfacies Fining-upwards sequence lamination microfacies Mixed ostracode-discorbid microfacies	Abundant	Up to 2%	164 - 373
MOLLUSC F.		Very scarce	Up to 0.6%	Up to 261

FIGURE 8 | **Facies and organic geochemistry of the individual fifth-order cycles of the Cherty-ostracode Unit.**

The obtained accumulation rate in the analysed stratigraphic interval is assumed to be constant. Taking into account that the thickness of the Corones Fm in the Adraén section is 200 m and in the Oix section 182 m, decompacted thicknesses have been calculated at 267 and 243 m, respectively (after the decompaction curves of Sclater and Christie, 1980). The bottom of the Corones Fm is within the Shallow Bentic Zone SBZ10 (Tosquella and Samsó, 1996; Serra-Kiel et al., 1998), thus it is not older than 52.5 my. The top of the Corones Fm occurs at 50.7 my, on the basis of the magnetostratigraphy from Vergés et al. (1998; fig. 8), making the maximum duration of this stratigraphic interval 1.8 my. The estimated accumulation rate in the Adraén section is 13.5 cm/ky and in the Oix section 14.8 cm/ky, with an average accumulation rate between the two sections at 14.2 cm/ky. In addition, Vergés et al. (1998) reported decomptacted thickness of 424 m in the Gombren section, which gives an accumulation rate of 23 cm/ky.

2. Duration of the Cherty-ostracode Unit. The duration of a given stratigraphic sub-interval, if its thickness and accumulation rate are known, may be calculated using the equation (1). The Cherty-ostracode Unit reaches its maximum thickness in the Arija section (15 m thick) and its minimum thickness in the Bagà and Oix sections (8 m thick). Calculated decompacted thicknesses range from 20 m to 12 m, respectively. Thus, the duration of the Cherty-ostracode Unit can be estimated from about 84,500 to 140,800 yr.

3. Fifth-order cycle period. The cycle period (CP) may be calculated, if the number of the cycles and the duration of a given stratigraphic sub-interval are known, using the equation (2):

$$CP = \frac{UD}{UCN} \qquad (2)$$

where the unit duration is UD and unit cycle number is UCN.

TABLE 1 | **Organic geochemical composition of Cherty-ostracode Unit facies (Organic carbon contents>0.4% in weight and/or HI>200). S1: free hydrocarbons (mgHC/g rock); S2: potential hydrocarbons (mgHC/g rock); TOC: Organic carbon content; HI: Hydrogen Index (S2/TOC); OI: Oxygen Index.**

Unit	Facies	Sample	Section	S1	S2	Tmax	TOC	HI	OI
Cherty-ostracode	Laminated ostracode	ARC114	Arija river	1.49	1.9	424	0.87	218	26
		ARC115	Arija river	0.59	6.6	441	2.03	325	12
		ARC116	Arija river	0.43	1.25	435	0.43	290	32
		ARC119	Arija river	0.27	2.92	441	1.1	265	21
		ARC123	Arija river	0.99	1.22	426	0.49	248	24
		EVALE	Bagà	0.18	2.21	445	0.68	325	23
		EVALM	Bagà	7.96	2.42	425	1.15	210	39
		EVAL21	Pobla de Lillet	0.97	0.91	432	0.47	193	87
		BCO88	Bagà	0.12	1.12	442	0.68	164	72
		PLC60	Pobla de Lillet	0.5	0.92	438	0.43	213	55
		PLC61	Pobla de Lillet	0.32	1.1	443	0.41	266	29
		PLC67	Pobla de Lillet	0.85	1.38	436	0.54	255	50
		PLC69	Pobla de Lillet	0.17	4.3	445	1.15	373	19
	Mollusc	ARC113	Arija river	0.22	0.36	438*	0.16*	225	-
		ARC118	Arija river	0.4	0.84	438*	0.35*	240	-
		ARC121	Arija river	0.43	1.12	432	0.59	189	-
		ARC122	Arija river	0.23	0.47	436*	0.18*	261	-
		EVALC	Bagà	0.15	1.00	445	0.56	179	23
		PLC52	Pobla de Lillet	0.25	0.38	436*	0.18*	211	83
		PLC55a	Pobla de Lillet	0.14	0.82	442	0.4	205	55
		PLC67	Pobla de Lillet	0.85	1.38	436	0.54	255	50
Top of Cherty-ostracode	Mixed dasyclad algae-ostracode-foraminifera	EVALR	Bagà	0.4	0.92	437	0.5	184	100
		BCO97	Bagà	0.09	1.33	442	0.72	184	56
		PLC71	Pobla de Lillet	0.51	0.9	440*	0.32*	281	25

* values below confidence level

TABLE 2 | **Saturated hydrocarbon fractions from the Cherty-ostracode Unit facies. EOM: Extractible organic matter; SAT: Saturated hydrocarbons; ARO: Aromatic hydrocarbons; NSO: Polar compounds; Pr/Ph: pristane/phytane ratio.**

Facies	Sample	Section	EOM (ppm)	SAT (%)	ARO (%)	NSO (%)	Pr/Ph	Pr/nC17	Ph/nC18
Laminated ostracode	ARC115	Arija river	2056	25.34	30.00	44.66	1.28	0.48	0.51
Laminated ostracode	ARC119	Arija river	1520	27.3	26.97	45.72	0.72	0.63	0.6
Mollusc	ARC113	Arija river	760	48.42	24.21	27.36	0.71	0.37	0.6
Mollusc	ARC122	Arija river	764	54.97	22.25	22.77	0.74	0.77	0.6
Mixed dasiclad algae-ostracode-foraminifera	EVALR	Bagà	1341	45.38	12.54	42.06	0.5	-	0.54

The Cherty-ostracode Unit consists of 5 fifth-order cycles, and consequently the cycle period offers values from 16,900 to 28,200 yr. Comparative results are obtained assuming a subsidence rate of 17 to 50 cm/ky (Giménez-Montsant and Salas, 1997; Vergés et al., 1998), where the obtained ages oscillate from 3,000 to 23,000 yr.

Even both the calculated durations in the present paper and the obtained from published data display a wide range of values, all of them fall within the range of values for the high frequency climatically-driven sea level cycles (Milankovitch Band, Fischer and Herbert, 1986; Read, 1995). An eustatic or glacio-eustatic origin for these cycles are assumed (Giménez and Calvet, 1990; Giménez, 1993). On the other hand, thickness for the individual cycles, after decompaction, has been calculated in 4 to 2.5 m/cycle, which is in agreement with the thickness of glacio-eustatic cycles (1-10 m thick; Read, 1995).

ORGANIC GEOGHEMISTRY

Studied samples belong to the following facies: 1) laminated ostracode, mollusc, and mixed dasyclad algae-ostracode-foraminifera facies of the Cherty-ostracode Unit; 2) foraminifera facies of the Lower and Upper Foraminifera Units; and 3) marly facies of the Shale Unit. The Cherty-ostracode Unit displays organic carbon values up to 2.1% in weight (Table 1), while organic carbon is nearly absent in both the Lower and Upper Foraminifera Units and the Shale Unit.

The highest organic carbon contents (up to 2% in weight) and Hydrogen Index (HI), between 164 and 373 (Table 1), correspond to the laminated ostracode facies in the Cherty-ostracode Unit, which plot within the Types II and III kerogen fields (Fig. 9). The associated chromatograms show a certain bimodal distribution of the n-alkanes with relative maximums at nC17 and nC29 (Fig. 10). The Ph/Pr ratio is the highest in the analysed facies, ranging from 0.7 to 1.3 (Table 2) but neither an even, nor odd, preference was observed. The relative maximums of nC17-nC18 alkanes reflect contribution of organic matter of algal origin (Han and Calvin, 1969; Gelpí et al., 1970; Blumer et al., 1971). These facies were deposited in relatively restricted conditions.

In contrast, the mollusc facies in the Cherty-ostracode Unit presents low organic carbon contents (below 0.6% in weight) and HI below 261 (Table 1). The corresponding pyrolysed samples plot within the Type III kerogen field (Fig. 9), indicative of terrestrial organic matter, and at the beginning of the oil window. The associated n-alkane chromatograms show a maximum

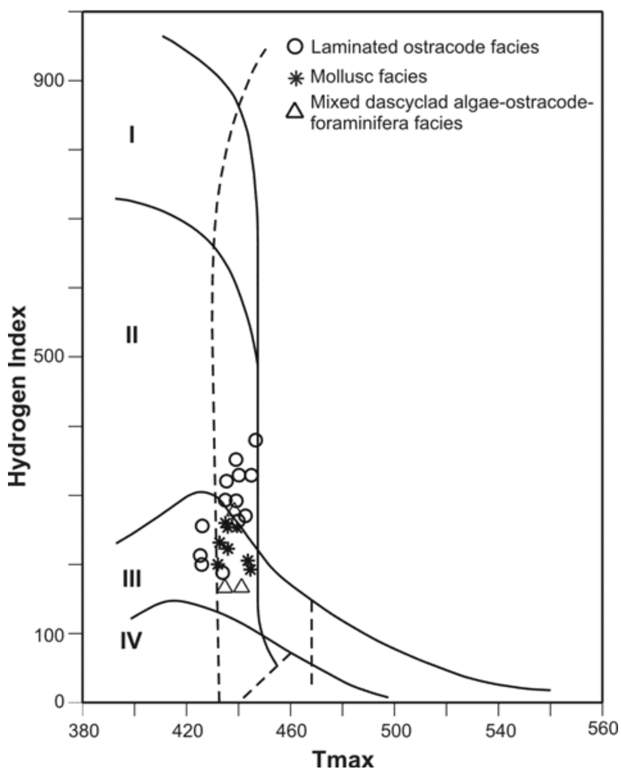


FIGURE 9 | **Rock-Eval diagrams of the Cherty-ostracode Unit facies.**

of around nC29, with no evident even/odd dominance (Fig. 10). The low concentration of the saturated hydrocarbons of medium molecular weight (nC15-nC20) could be interpreted as selective oxidation and/or biodegradation of sedimentary origin (Simoneit et al., 1984). The presence of saturated hydrocarbons of around nC29 could be explained as a selective preservation of the organic compounds from terrestrial origin, and could be considered more resistant to the primary oxidation processes. These geochemical data suggest an input of terrestrial organic matter during the mollusc facies deposition, in an oxygenated and predominantly marine environment. In these conditions, organic matter was poorly preserved.

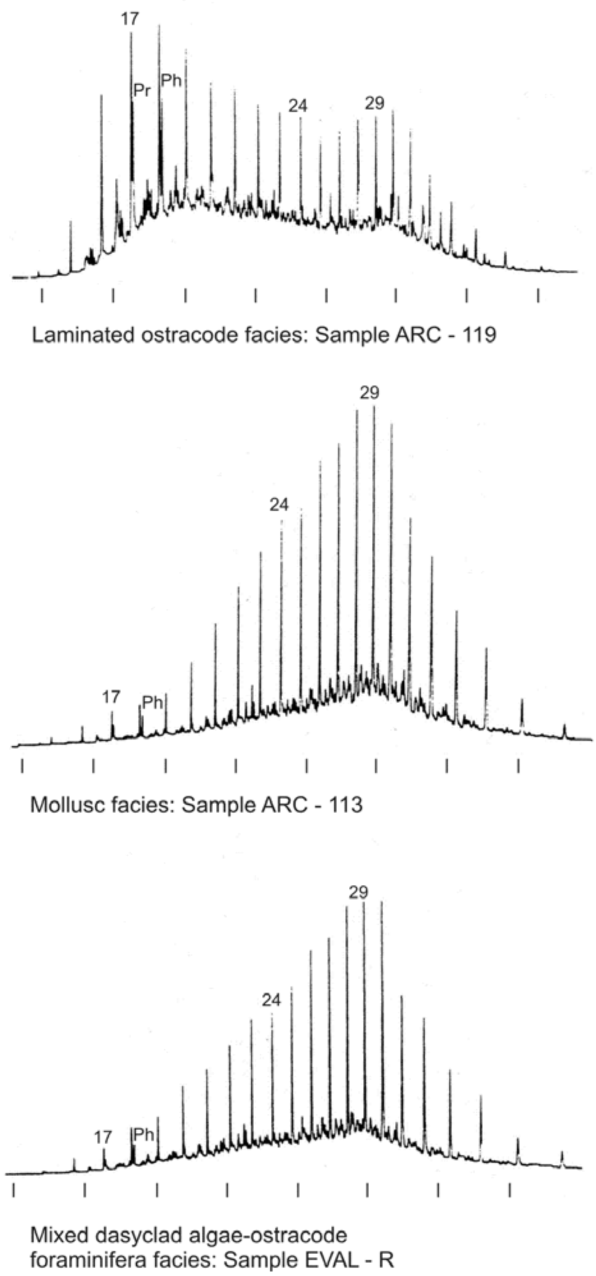


FIGURE 10 **Chromatograms of the Cherty-ostracode Unit facies with organic carbon contents >0.4% in weight.**

The higher organic carbon contents in the laminated ostracode member than in the mollusc facies implies greater productivity and/or preservation of organic matter deposited in more restricted conditions, with increasing anoxia. Moreover, the grey to dark grey colour of the ostracodes is probably due to the fixation of hydrocarbons derived from the surrounding sediments (Ainsworth et al., 1990). On the other hand, there exists a relationship between abundance of cherts and high organic matter contents in the host laminated ostracode facies, a relationship observed by both Parnell (1988), who documented the presence of cherts in organic rich levels in the Devonian Orcadian Basin of Scotland, and Gao and Land (1991).

The mixed dasyclad algae-ostracode-foraminifera facies (top of the Cherty-ostracode Unit) offers geochemical organic parameters and chromatograms very similar to those of the mollusc facies. The organic carbon contents are up to 0.72% in weight with HI up to 281 (Table 1). These results indicate a progressive opening through more marine and oxygenated waters at the top of the Cherty-ostracode Unit, which impeded the preservation of organic matter.

CONCLUSIONS

The Corones Fm is a third-order megacycle, consisting of a Lower Terrigenous Mb (interpreted as a lower system tract) and an Upper Limestone Mb (transgressive system tract). The fourth-order megacycles in the Upper Limestone Mb are composed of two deepening upwards cycles. The megacycles (third and fourth-order cycles) were due to the rhythmic variations in the rate of tectonic subsidence. Similar patterns of decametric megacycles, interpreted as having tectonic origin, composed of metric cycles interpreted as having eustatic origin have been cited by Fischer (1964) in the Upper Triassic Dachstein Fm in the Northern Calcareous Alps.

The Upper Limestone Mb of the Corones Fm, in which 6 different units are distinguished (Ostracode, Lower and Upper Foraminifera, Shale, Cherty-ostracode, Chara-ostracode Units), shows important thickness variations and lateral facies changes. Despite these two general characters, the Shale Unit preserves a constant thickness. The different behaviour of the different units of the Upper Limestone Mb can be interpreted as different rates of subsidence during the deposition of that member.

The presence of cherty horizons (nodular and layered) is a very notable feature of the Cherty-ostracode Unit, mainly represented in the central sedimentological domain of the Upper Limestone Mb, showing a close relationship with high organic matter content and high HI. The Cherty-ostracode Unit is the only unit that shows a pervasive internal cyclicity arrangement, consisting of 5 fifth-order cycles up to 2 m thick. Each single cycle is composed of a lower thin bedded mollusc facies member and by an upper laminated ostracode and chert-rich facies member. The calculated duration of the fifth-order cycle in the Cherty-ostracode Unit period ranges from about 17,000 to 28,000 years. These values are relatively similar to the processional cycle period (Berger, 1988; Fischer and Bottjer, 1991). The fifth-order cyclicity may be interpreted as eustatic or glacio-eustatic in origin, and is located in the Milankovitch Band of Fischer and Herbert (1986).

The upper laminated ostracode facies member of the fifth-order cycles consists of 5 minor cycles, corresponding to sixth-order cycles. The sixth-order cycles consist of a lower thin cherty level interpreted as the transgresssive phase and an upper laminated ostracode level interpreted as the regressive phase. The seventh-order period cycles, corresponding to the rhythmic ostracode microfacies, have been interpreted as seasonal in origin (annual period). The light grainy laminae may have been deposited during the oxygenated winter periods and the dark muddy laminae during the anoxic summer periods.

According to their organic geochemical composition, the Upper Limestone Member facies of the central domain are divided in three groups: a) the foraminifera and marly facies, with the lowest organic contents and HI, close to 0 (Lower and Upper Foraminifera, Shale Units); b) the mollusc and mixed dasyclad algae-ostracode-foraminifera facies (Cherty-ostracode Unit), with low organic carbon and HI; and c) the laminated ostracode facies (Cherty-ostracode Unit), showing higher organic carbon and HI. The lower mollusc-rich member displays low organic carbon contents and HI (below 0.6% in weight and 261, respectively), while the upper ostracode chert-rich member shows higher organic carbon contents and HI (up to 2% in weight and 373, respectively).

ACKNOWLEDGEMENTS

The authors are grateful to Dr. Ramon Salas for his comments on cyclicity and subsidence, to Dr. P. Carbonel for the ostracodes classification, Dr. Josep Serra for the foraminifera classification and paleoecology, and Dr. San Fillipo for the suggestions on sponges. The work was partially supported by the Spanish Goverment Projects CGL2005-0537, CGL2005-07445-C03.01 and CGL-2006-04860, and the "Grup Consolidat de Recerca de Geologia Sedimentària" 2001/SGR/75 (Catalan Government). We also wish to thank Dr. R. Marfil and an anonymous reviewer for their useful comments and Frances Luttikhuizen for her revision of the English version.

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Manuscript received February 2005;
revision accepted May 2006.