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Autor(en): **Davey, Simon D. / Jenkyns, Hugh C.**

Objekttyp: **Article**

Zeitschrift: **Eclogae Geologicae Helvetiae**

Band (Jahr): **92 (1999)**

Heft 2

PDF erstellt am: **08.08.2024**

Persistenter Link: https://doi.org/10.5169/seals-168658

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0012-9402/99/020163-8 $1.50 + 0.20/0
Birkhäuser Verlag, Basel, 1999

Eclogae geol. Helv. 92 (1999) 163–170

Carbon-isotope stratigraphy of shallow-water limestones and implications for the timing of Late Cretaceous sea-level rise and anoxic events (Cenomanian-Turonian of the peri-Adriatic carbonate platform, Croatia)

SIMON D. DAVEY[1,2] & HUGH C. JENKYNS[1]

Keywords: Carbon-isotope stratigraphy, sea-level rises, Cenomanian-Turonian boundary, carbonate platform, Croatia

ABSTRACT

A carbon-isotope stratigraphy of shallow-water Cretaceous limestones from the Adriatic/Dinaric carbonate platform, Croatia, has enabled accurate definition of the Cenomanian-Turonian boundary by reference to well-dated ammonite-bearing sections in England and the United States. The sediments close to the boundary show pelagic influence registered by the presence of planktonic foraminifera and calcispheres, but also contain rudistid and echinodermal grains: above and below are typical peritidal carbonates. In one section the stage boundary is drawn within the deeper-water facies, in another it is tentatively placed within overlying oncoid-rich packstones and wackestones. These results spotlight the potential use of carbon isotopes as a dating and high-resolution correlative tool in shallow-water carbonate rocks, and help elucidate the timing of oceanographic events that affected the Adriatic/Dinaric carbonate platform. In particular it is suggested that the highest rate of relative, possibly eustatic sea-level rise took place during the latest Cenomanian, that this was followed by the global oceanic anoxic event during Cenomanian-Turonian boundary time, and that peak transgression or maximum flooding was achieved during the early to mid-Turonian.

RIASSUNTO

La stratigrafia isotopica ($\delta^{13}C$) effettuata su calcari cretacici appartenenti alla piattaforma carbonatica adriatico/dinarica ci ha permesso di definire con accuratezza il limite Cenomaniano-Turoniano, calibrandolo con serie coeve ad ammoniti dell' Inghilterra e degli Stati Uniti. I sedimenti in prossimità del limite mostrano un influenza pelagica, con foraminiferi planctonici e calcisfere associate a resti di rudiste ed echinodermi. Gli strati immediatamente sovrastanti e sottostanti sono costituiti invece da carbonati peritidali. In una delle sezioni, il limite si colloca all' interno della facies con influenza pelagica; in un'altra, invece, esso cade all' interno della facies micritica con oncoliti che giace al di sopra dello stesso intervallo. Questi risultati enfatizzano 1) la possibilità di usare gli isotopi del carbonio come strumento stratigrafico ad alta risoluzione nelle facies carbonatiche di bassa profondità e 2) consentono di datare eventi oceanografici che hanno impresso il loro segnale sulla piattaforma carbonatica adriatico/dinarica. In particolare, si propone che l'innalzamento eustatico del livello del mare sia coinciso con il Cenomaniano terminale, che l' evento anossico globale si sia verificato nel tardo Cenomaniano-Turoniano precoce e che la massima ingressione marina sia avvenuta nel Turoniano precoce e medio.

Introduction

Carbon-isotope stratigraphy has shown itself to be a powerful tool in the correlation of Mesozoic pelagic sediments. Its use in trans-continental correlation of Cretaceous chalk and limestone sequences, first demonstrated by Scholle & Arthur (1980), has subsequently been shown to be valid down to temporal increments of hundreds of thousands of years or less (Gale et al., 1993). The best studied interval is that which straddles the Cenomanian-Turonian boundary (93 Ma, Obradovich, 1994; Kowallis et al., 1995). This has been documented isotopically from Europe, North America, Africa and the Atlantic, Pacific and Indian Oceans (Pratt & Threlkeld, 1984; Hilbrecht & Hoefs, 1986; Schlanger et al., 1987; Jarvis et al., 1988; Kuhnt et al., 1990; Thurow et al., 1992; Curiale, 1994; Jenkyns et al., 1994, 1995; Paul et al., 1994; Lamolda et al., 1994; Accarie et al., 1996). Expanded stratigraphic sequences across this interval show a complex multi-faceted positive carbon-isotope excursion whose profile is conventionally taken to reflect global burial patterns of organic carbon. Paul et al. (1994) estimate the duration of the isotopic event as some 250,000 to 270,000 years.

Relative to carbon in the 'oxidized' reservoir, organic carbon ('reduced reservoir') is enriched in the lighter isotope ^{12}C; consequently increased burial rates of organic matter produce an increase in the $^{13}C/^{12}C$ ratio in sea water which is transmitted to skeletal and inorganic carbonate (Scholle & Arthur, 1980; Berger & Vincent, 1986). Certain intervals of geological time were characterized by anomalously high burial rates of organic carbon on a global scale and these have been termed

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[1] Department of Earth Sciences, University of Oxford, Parks Road, Oxford, OX1 3PR, United Kingdom
[2] EPT-IF, Shell International Exploration and Production, PO Box 60, 2280 AB Rijswijk, Netherland

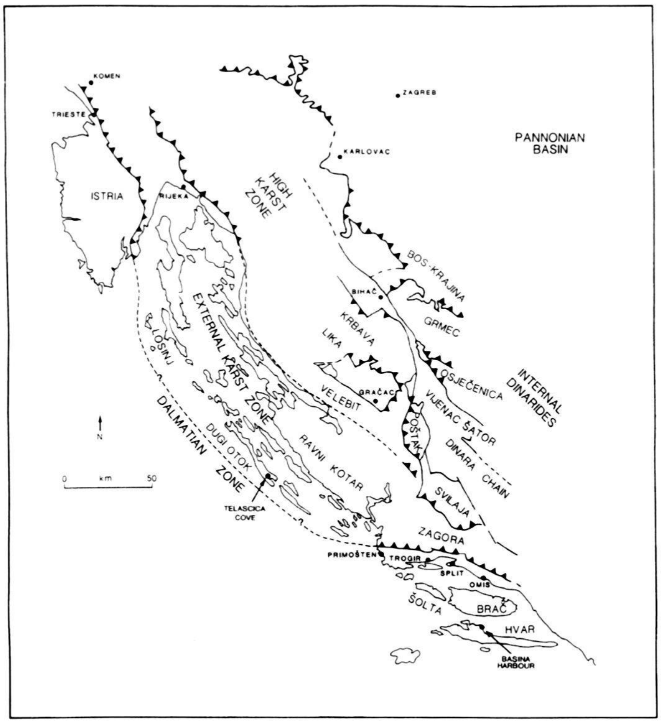

Fig. 1. Map of the Croatian region showing principal facies domains across the Adriatic-Dinaric carbonate platform during Cenomanian-Turonian boundary time, and localities mentioned in text. After Chorowicz (1977). The two localities examined isotopically are Telascica Cove on Dugi Otok and Basina Harbour on Hvar.

'oceanic anoxic events' or 'OAEs' (Schlanger & Jenkyns, 1976; Jenkyns, 1980, 1999; Arthur et al., 1990). Here we investigate the Cenomanian-Turonian isotopic excursion accompanying the Oceanic Anoxic Event as registered in the shallow-water domain of the Adriatic/Dinaric carbonate platform, and explore the stratigraphic use of isotopes in peritidal and related facies.

The Adriatic/Dinaric carbonate platform

The Adriatic/Dinaric carbonate platform, installed on the southern margin of the Tethyan Ocean, was a shallow-water system generally comparable to the Bahama Banks (Jenkyns, 1991). Subsidence on the former continental margin was balanced by prolific carbonate production, and sedimentary piles of up to 7 km were built up during the Triassic-Tertiary interval (Herak et al., 1970; D'Argenio, 1974). The facies include typical supratidal, intertidal and shallow subtidal lithologies: oolitic, oncolitic, pelletal, skeletal, stromatolitic facies and lime mudstones containing spar-filled desiccation pores. To the east, the carbonate platform is overthrust by the internal Dinaride ophiolite belt; to the west, it passes into deeper-water pelagic facies of the Adriatic region (Fig. 1).

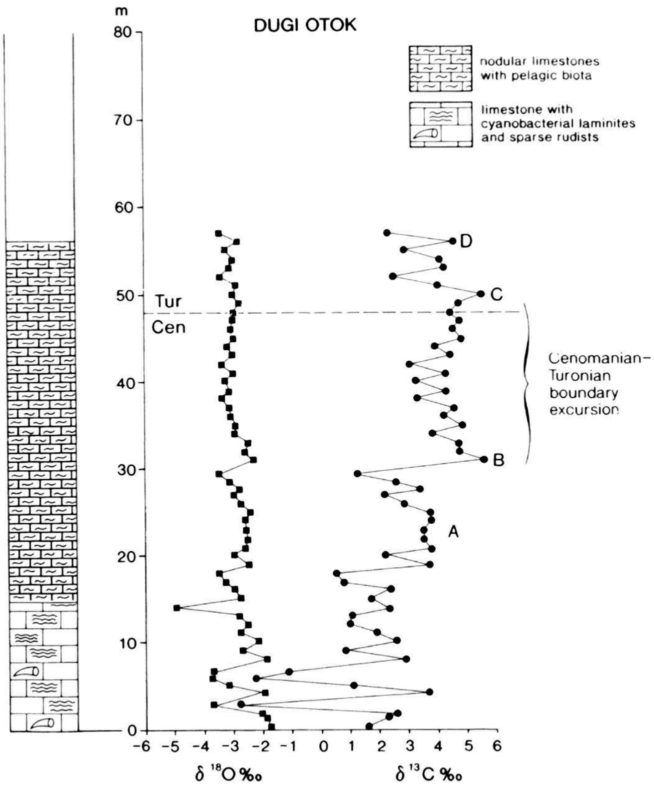

Fig. 2. Lithological section sampled at Telascica Cove, Dugi Otok, Croatia, with accompanying bulk carbon- and oxygen isotope data. Significant features of the isotope curve are labelled A, B, C and D. The basal part of the section (Milna Formation) dominantly comprises lime wackestones with cyanobacterial laminites interbedded with wackestones to mudstones with occasional rudists. Overlying these facies are the nodular limestones of the Sveti Duh Formation containing planktonic biota. Position of Cenomanian-Turonian boundary, according to carbon-isotope stratigraphy (horizontal dashed line), is also shown.

The Cenomanian-Turonian sea-level rise

The Cenomanian-Turonian has long been recognized by stratigraphers as an interval characterized by a spectacular transgression whose sedimentary record can be traced on several continents (e.g. Suess, 1888; Hancock & Kauffman, 1979; Schlanger, 1986; Haq et al., 1988; Hancock, 1989, 1993; Gale, 1996). Because of its ubiquitous influence, this transgression is commonly viewed as eustatic, with the highest stand of sea level, perhaps of the whole Phanerozoic, achieved during the early to mid-Turonian. The impact of this sea-level rise on the Adriatic/Dinaric carbonate platform, documented by Chorowicz (1977), Gušić & Jelaska (1990, 1993), and Jenkyns (1991), was manifested by an increased component of planktonic faunal elements among grains of shallow-water affinity. Locally, ammonites are recorded from the Turonian (Polšak et al., 1982; Gušić & Jelaska, 1990; Davey et al., 1992), suggestive of substantial and long-lasting deepening in some areas. The facies maps of Chorowicz indicate that this partial drowning and invasion of pelagic conditions affected only part of the platform, whereas Gušić & Jelaska (1993) have documented a more comprehensive flooding event. The strata deposited during this interval are known as the Sveti Duh Formation and, in the absence of stratigraphically diagnostic fossils, the age-assignment is conventionally taken as Cenomanian-Turonian. Above and below the Sveti Duh Formation the facies are characteristically peritidal in nature, and the overall succession illustrates the resiliance of the carbonate-platform system to changes in relative sea level (cf. Schlager, 1981).

The sections studied

Two sequences deposited on the Adriatic/Dinaric carbonate platform, dated as broadly Cenomanian-Turonian in age, have been examined for isotope stratigraphy: one on the island of Dugi Otok, the other on the island of Hvar (Fig. 1). The facies represented by the section on southeast Dugi Otok, exposed in Telascica Cove, comprise a basal 14.85 metres of lime mudstones to wackestones, locally with cyanobacterial laminites and desiccation pores, that contain benthonic foraminifera and rudistid fragments (Milna Formation, Fuček et al., 1990; Fuček et al., 1991). These strata are interpreted as having formed in a shallow lagoonal to tidal-flat environment and are characteristic of platform interiors. Overlying the Milna Formation are some 50 metres of a somewhat nodular lime mudstone (Sveti Duh Formation) with a more limited fauna dominated by calcispheres and some planktonic foraminifera, plus minor amounts of echinoid and reworked rudistid debris. Centimetre-scale brown chert nodules occur locally. This unit betrays clear pelagic influence. Capping this (but not reached in the log shown in Figure 2) is a sequence of benthic foraminiferal wackestones, rudist floatstones and strata with abundant oncoids which indicate re-establishment of shallow-water conditions (Gornji Humac Formation).

The facies sampled from the section in Basina Harbour, in the north of the island of Hvar (Fig. 3), chiefly comprise cycles of benthic foraminiferal wackestones-packstones with rudistid floatstones passing upward into cyanobacterial laminites (Milna Formation). A very thin lime mudstone unit with small amounts of pelagic microfauna occurs intercalated in this sequence between levels 6.1 m and 9.3 m in the sequence (Fig. 3). Platform-interior peritidal conditions are again indicated for most of the sequence. Above the dominantly peritidal sequence is a 10.25 m-thick unit that contains sparse pelagic microfossils but which, unlike the section on Dugi Otok, is mostly massive rather than nodular and dominated by rudistid debris in wackestone-packstone matrix. Overlying this presumed thinned equivalent of the Sveti Duh Formation is a series of wackestones and packstones with abundant oncoids (Gornji Humac Formation, Gušić & Jelaska, 1990).

In both cases, therefore, there is evidence for increased pelagic influence during Cenomanian-Turonian boundary time. However, the fundamental platform morphology in these areas was maintained and the carbonate factory was re-established after temporary deepening. A possible recent analogue for the environment on the Adriatic/Dinaric platform during the time of maximum deepening could be the deeper reaches (> 30 m) of the lagoon off Belize where pelagic biota such as coccoliths and pteropods are mixed with typical shallow-water material (Scholle & Kling, 1972; Jenkyns, 1991).

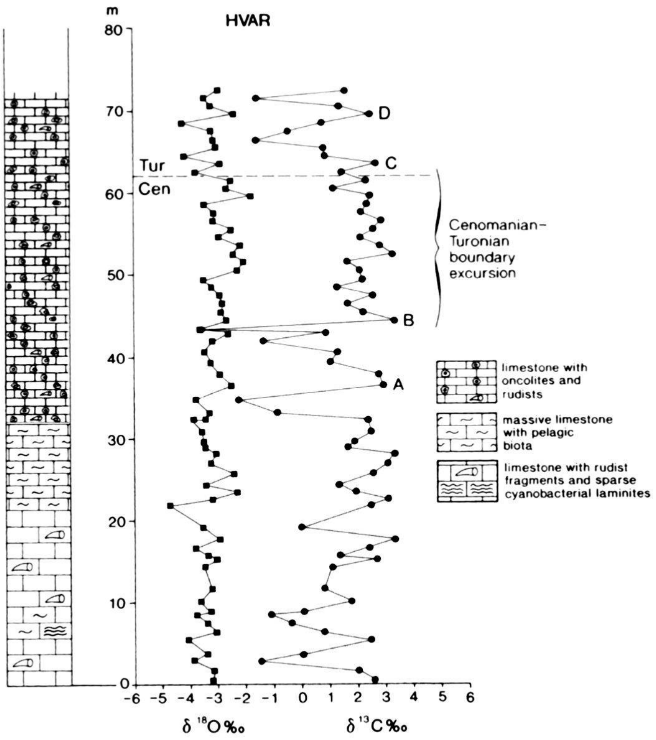

Fig. 3. Lithological section sampled at Basina Harbour, Hvar, Croatia, with accompanying bulk carbon- and oxygen-isotope data. Significant features of the isotope curve are labelled A, B, C and D as in Figure 2 (Dugi Otok) and are tentatively interpreted to represent identical positions on the isotope curve. The basal part of the section (Milna Formation) dominantly comprises rudistid lime floatstones-packstones and foraminiferal wackestones-packstones. Lime mudstones with cyanobacterial laminites are rare. Overlying these facies are the relatively massive limestones of the Sveti Duh Formation containing planktonic biota; above this unit lie the oncolitic packstones and wackestones, locally rudist-bearing, of the Gornji Humac Formation. Position of Cenomanian-Turonian boundary, according to carbon-isotope stratigraphy (horizontal dashed line), is also shown.

Isotope stratigraphy

Techniques

Samples for analyses were either obtained by careful drilling of specimens or crushing of small pieces of limestone, care being taken to avoid diagenetic vein sparite and large sketetal grains.

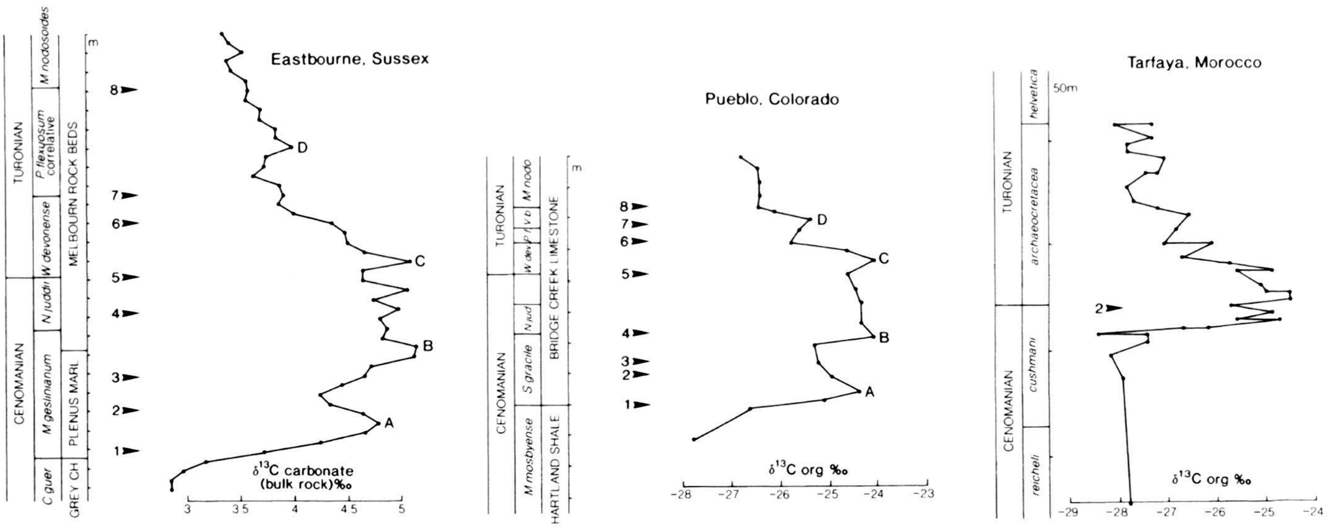

Fig. 4. Carbon-isotope stratigraphy of reference sections from three continents (Europe, North America, Africa) that span the Cenomanian-Turonian boundary. Significant features of the isotope curves are labelled A, B, C and D as in Figures 2 (Dugi Otok) and Figure 3 (Hvar). Data from Pratt & Threlkeld (1984) for organic carbon from Colorado, U.S.A. (Bridge Creek Limestone); Kuhnt et al. (1990), for organic carbon from Tarfaya, Morocco; and Gale et al. (1993) for pelagic carbonate (Chalk) from Eastbourne, Sussex, UK. These curves, derived from stratigraphically expanded sections (particularly Tarfaya; note the 50m scale increments), show the complex and distinctive shape of the positive $\delta^{13}C$ excursion which allow it to be used as a high-resolution correlative tool. The Cenomanian-Turonian boundary (datum point 5 and shaded line) is drawn at the base of the *Watinoceras devonense* Zone (Kennedy & Cobban, 1991; Bengtson, 1996). Other datum levels are: 1) appearance of ammonite *Metoicoceras geslinianum*; 2) disappearance of the planktonic foraminifera *Rotalipora cushmani*; 3) disappearance of calcareous nannofossil *Axopodorhabdus albianus*; 4) appearance of ammonite *Neocardioceras juddii*; 6) appearance of bivalve *Mytiloides* gr. *columbianus*; 7) appearance of ammonite *Fagesia catinus*; 8) appearance of ammonite *Mammites nodosoides*. Note how, in the section from Tarfaya, the Cenomanian-Turonian boundary has been placed at the extinction level of *Rotalipora cushmani* whereas a notional boundary based on macrofossils, applied using carbon-isotopic correlation, would lie higher in the section.

Powders were then cleaned using 10% H_2O_2 followed by acetone and then dried at 60°C. They were then reacted with purified orthophosphoric acid at 90°C and analysed on-line using a VG Isocarb device and Prism mass spectrometer at Oxford University. Normal corrections were applied and the results are reported, using the usual δ notation, in per mil (‰) deviation from the PDB standard. Calibration to PDB was performed via the laboratory standard calibrated against NBS19 and Cambridge Carrara marble. Reproducibility of replicate analyses of standards was generally better than 0.1 ‰ for both carbon- and oxygen-isotope ratios.

The section on Dugi Otok

In this section, from base level to about 15 m, both carbon- and oxygen-isotope values fluctuate greatly, and a high degree of positive correlation is notable (Fig. 2). This is suggestive of a diagenetic overprint, possibly under the influence of meteoric waters (cf. Marshall, 1992). Data from below 14.85 m in the section derive from peritidal carbonates whereas data from above this level are derived from more pelagic lime mudstones of the Sveti Duh Formation. The abrupt shift in stable-isotope signature thus correlates with the facies change, and the remarkly uniform nature of the $\delta^{18}O$ profile in these higher levels of the section is notable.

The carbon-isotope data reveal a detailed positive $\delta^{13}C$ spike, with a stratigraphically lower interval (A) of relatively elevated values, generally greater than 3.5‰, followed by a high of nearly 5.6‰ at a sharply defined peak (B). This peak (B) is placed at the 30 m level in the section, some 15m above the first development of facies with pelagic affinity. Values then remain high, on a plateau with values generally greater than 4.0‰, for 19 m, before again peaking at over 5.5‰ (C). A final subsidiary peak of over 4.0‰ (D) is recorded before the end of the section. The remarkably consistent $\delta^{18}O$ values in the Sveti Duh Formation and their similarity to those of mid-Cretaceous pelagic marine calcites from many localities worldwide (Scholle & Arthur, 1980; Hudson & Anderson 1989; Jenkyns et al., 1994) suggests that both carbon- and oxygen-isotope ratios in this unit have been little modified.

The section on Hvar

In this section (Fig. 3), the variability in oxygen-isotope values is higher than in the section from Dugi Otok, although a relatively stable trend is still present. Once again, the absolute values of $\delta^{18}O$ are almost all within the typical band of Cretaceous marine calcites. Compared with the section on Dugi Otok, the $\delta^{13}C$ values, although manifesting little correlation with $\delta^{18}O$, display neither the same well-defined trend nor the same high absolute values. Nevertheless, the carbon-isotope ratios show a broad trend of increase and decrease, with several small peaks whose values are greater than 3‰. Abrupt changes in $\delta^{13}C$ values to negative figures may signify local diagenetic overprints.

The poorer definition of the curve in this section could relate to the greater proportion of large carbonate skeleta, such as echinoderms and red algae, exhibiting non-equilibrium isotopic fractionation (Keith & Weber, 1965; Weber & Raup, 1966; Weber, 1968; Anderson & Arthur, 1983). Diagenetic alteration of originally aragonitic material may also have been significant. Alternatively, the water-masses on the carbonate platform at this time may have had only limited connection with the open sea and become relatively depleted in ^{13}C because of local oxidation of organic matter. Such effects have been observed on modern carbonate platforms such as Florida and the Bahamas (Patterson & Walter, 1994). The Lower Cretaceous peritidal platform carbonates from the Gavrovo Platform, western Greece, analysed isotopically by Grötsch et al. (1998), follow global trends but are also negatively offset by about 1‰ relative to values of coeval pelagic limestones.

Although the Hvar section contains the same general stratigraphy as the section on Dugi Otok, the location of the major broad peak with high $\delta^{13}C$ values (greater than 3‰) is less obvious. Certainly it is not unambiguously displayed within the Sveti Duh Formation, where most $\delta^{13}C$ values are less than 3.0‰. Most probably the broad peak lies in the interval between 43m and 65m metres within the oncolitic facies that overlie the more pelagic interval (Fig. 3). In this part of the section there is a distinct relative maximum with values of 3.0‰ (A), in advance of a distinct peak at 3.4‰ (B), marking the beginning of a broad plateau of relatively high $\delta^{13}C$ values extending over 20 metres of section. This plateau contains some minor peaks of just under and just over 3.0‰, before values drop to background levels. Two small peaks with $\delta^{13}C$ values just under 3.0‰ (C, D) are seen before the end of the section. This pattern, albeit with lower absolute values, is very similar to that seen in the section at on Dugi Otok (A,B, C and D are assumed to be correlative) but, by contrast, the carbon-isotope excursion occurs in the shallow-water oncolitic facies above the Sveti Duh Formation. Remarkably, perhaps, the thickness of strata between points 'B' and 'C' on the sections from Dugi Otok and Hvar is almost identical.

Comparison with Cenomanian-Turonian boundary sections at Eastbourne, England, Pueblo, Colorado, U.S.A. and Tarfaya, Morocco

Carbon-isotope profiles from Eastbourne, Pueblo and Tarfaya (Kuhnt et al., 1990; Gale et al., 1993) are presented in Figure 4. In the case of the first two sections, the stratigraphy is well constrained by the presence of ammonites, bivalves, planktonic foraminifers and nannofossils; the latter is zoned by microfossils. The Cenomanian-Turonian boundary, drawn at the base of the *Watinoceras devonense* ammonite Zone (Kennedy & Cobban, 1991; Bengtson, 1996), can be fixed in the Tarfaya section by comparing the curves. The numbers refer to datum levels defined by the appearance or disappearance of particular biotic elements. Note how these datum levels possess a constant relationship with the peaks of the carbon-isotope curve, suggesting effective synchroneity of both chemostratigraphic and biostratigraphic divisions.

These isotopic reference curves enable us to place a stage boundary, defined on macrofossils, remarkably exactly in the section from Dugi Otok and more tentatively in the section from Hvar (Figs. 2, 3). In the section from Dugi Otok the thickness of section from the base of the Sveti Duh Formation to the Cenomanian-Turonian boundary is some 33 metres; on Hvar it is 40 metres. Isotopic correlation between the Croatian, English and American sections is illustrated in Figure 5.

Implications

These results show clearly that accurate dating and correlation of facies containing few stratigraphically diagnostic fossils is possible where the carbon-isotope reference curve is possessed of considerable structure. Isotopic work on Cretaceous platform carbonates from Oman revealed that gross trends can be used to correlate from well to well (Wagner, 1990), and more detailed correlation has now been achieved in this and adjacent areas (Vahrenkamp, 1996; Grötsch et al., 1998). Refining of Upper Cretaceous stage boundaries, using the carbon-isotope curve, has also been possible for platform carbonates exposed in the Iberian Chain, Spain (Valladares et al., 1996). Resolution to the zonal level has been achieved on shallow-water Lower Cretaceous peritidal carbonates from the mid-Pacific Mountains (Resolution Guyot, Jenkyns, 1995) and from the Southern Limestone Apennines of Italy (Ferreri et al., 1997).

The fixing of the Cenomanian-Turonian contact in the section exposed on Hvar shows that an organic-rich laminated fish-bearing facies (Hemleben & Freels, 1977), which crops out stratigraphically below the section illustrated in Figure 3, is actually late Cenomanian in age. This organic-rich level was related to the Oceanic Anoxic Event at the stage boundary by Jenkyns (1991): this event is believed to register the time of maximum global burial rate of organic carbon (Arthur et al., 1987; Jenkyns, 1999). However, the $\delta^{13}C$ profile from Hvar, defining the stage boundary, indicates clearly that deposition of the organic-rich fish beds, as suggested by Gušić & Jelaska (1993), predated the Oceanic Anoxic Event. Isotopic profiles from other localities in Europe (e.g. England, Italy) show that this burial rate increased throughout the mid to late Cenomanian before peaking at the stage boundary (Arthur et al., 1987; Jenkyns et al., 1994). The organic-rich laminated fish-bearing facies of the Adriatic/Dinaric carbonate platform can still be

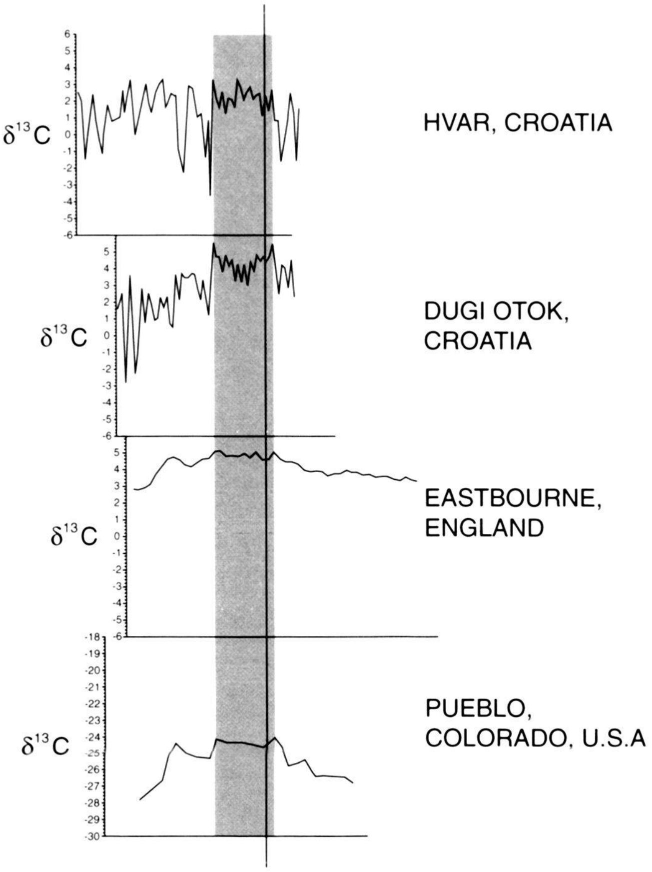

Fig. 5. Correlation of carbon-isotope profiles from Croatian limestones and the Chalk of Eastbourne, England (all $\delta^{13}C_{carb}$) and organic matter from the Bridge Creek Limestone ($\delta^{13}C_{org}$) exposed near Pueblo, Colorado, U.S.A. All sections are plotted on the same vertical scale. Horizontal scale, representing stratigraphic thickness or time, is variable. The vertical shaded band defines the zone of relatively high $\delta^{13}C$ values, with similar overall structure in terms of peaks, troughs and plateaux, characteristic of the Cenomanian-Turonian boundary. The vertical line defines the position of the Cenomanian-Turonian boundary, as fixed by international agreement, at Pueblo (Bengtson, 1996).

considered part of the global pattern of carbon-rich facies that is peculiarly characteristic of this latter part of Cretaceous time. Indeed some of these facies, in the Trieste-Komen region (Fig. 1), may be of Cenomanian-Turonian-boundary or younger age (Jurkovšek et al., 1996).

A further implication of the isotope stratigraphy is that the age and duration of the more open-marine interlude represented by the Sveti Duh Formation varies from place to place on the Adriatic/Dinaric carbonate platform. On Hvar it is latest Cenomanian; on Dugi Otok it spans the Cenomanian-Turonian boundary. If it is assumed that the deepening reflects a eustatic signal, then the most rapid rate of sea-level rise most likely took place in the early to mid point of the pelagic interlude, assuming a curvilinear trend of eustatic change (Posamentier et al., 1988). This implies that major deepening and the most rapid increase in water depth took place during the late Cenomanian, either before or in earliest *M. geslinianum*-Zone time by reference to Figures 2, 3 and 4, when carbonate production was temporarily outpaced. Thereafter, where local recovery of the platform took place (i.e. on Hvar), water depths decreased during Cenomanian-Turonian boundary time. Assuming constant subsidence rates, decrease in water depth could have been caused by a fall in the rate of rise in eustatic sea level or an increase in the rate of carbonate deposition or, more probably, a combination of the two phenomena. This simple interpretation needs to be tested against the stratigraphic record of coeval Cretaceous facies from non-tectonically active areas which were sensitive to eustatic sea-level change (c.f. Hancock, 1993; Uličný et al., 1993, 1997; Hilbrecht et al., 1996; Mitchell et al., 1996; Voigt and Hilbrecht, 1997).

If the evidence from Croatia is of regional significance, then the order of two major Late Cretaceous palaeoceanographic events was as follows: 1) maximum rate of rise of relative sea level in the late Cenomanian, prior to or in earliest *M. geslinianum*-Zone time, followed by; 2) the global oceanic anoxic event, characterized by the diagnostic positive $\delta^{13}C$ excursion (Cenomanian-Turonian boundary time), as the rate of rise declined. Peak transgression or maximum flooding was achieved during the early to mid-Turonian, as suggested by regional stratigraphic data from northern Europe (Hancock, 1993; Gale, 1996) and many other cratonic areas (Haq et al., 1988). By this time, however, many of the drowned areas of the Adriatic/Dinaric carbonate platform had built back close to sea level (Gušić & Jelaska, 1993).

Acknowledgements

Davey acknowledges a NERC/BP CASE studentship held at Cardiff and Oxford Universities. Jenkyns acknowledges support from British Petroleum for studies on European Mesozoic stratigraphy. The geochemical analyses were undertaken in the Oxford Isotope Laboratory. Julie Cartlidge is thanked for assistance. Massimo Sarti checked and corrected the Italian abstract.

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Manuscript received June 8, 1997
Revision accepted June 25, 1999