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Petrography and stable isotope aspects of cold-vent activity imprinted on Miocene-age "calcari a *Lucina*" from Tuscan and Romagna Apennines, Italy

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Abstract Over 20 occurrences of discontinuous limestone blocks, locally called "calcari a *Lucina*," were mapped in the Tuscan–Romagna region of the northern Italian Apennines. The limestones, consisting of a variable mixture of authigenic carbonates (calcite, dolomite, and aragonite), sulfides (primarily pyrite), and allogenic silicates, occur in association with turbidite and hemipelagite units that were deposited in foredeep basins during early to late Miocene times. The limestone blocks are interpreted to represent relicts of carbonate buildups formed around methane-rich fluid vents on the basis of their (1) striking petrographic similarities to carbonates from cold vents in the modern oceans; (2) unique chemosynthetic-like fauna, and (3) anomalously negative $\delta^{13}C$ values ($\delta^{13}C = -16‰$ to $-58‰$ PDB). The contemporaneous tectonism of the Apennine orogeny is likely to be the primary cause for the expulsion of the methane-rich fluids to the seabed in a manner analogous to the fluid-flow processes occurring at modern accretionary prisms.

Introduction

The occurrence of carbonate buildups (chemoherms, see Aharon 1994 for definition) associated with fluid and hydrocarbon venting at the sea floor have been reported over the last decade from a variety of modern settings including the tops of salt diapirs along passive continental margins in the Gulf of Mexico (Brooks et al. 1987; Roberts et al. 1990; Roberts and Aharon 1994) and in the North Sea (Hovland et al. 1987), at the base of the Florida Escarpment (Paull et al. 1984), and the accretionary prisms associated with subduction activity along active continental margins of Oregon (Ritger et al. 1987; Kulm et al. 1988) and Japan (Sibuet et al. 1988). The most common recognizable features of the vent deposits are the faunal assemblages using chemosynthesis as the principal metabolic pathway (MacDonald et al. 1989) and the authigenic carbonates and sulfides forming hard substrates that include crusts, slabs, mounds, and chimneys. More recently, fossil chemoherm occurrences that show striking resemblance to the modern ones have been reported from Phanerozoic rock formations occurring at different locations around the world (Beauchamp et al. 1989; Goedert and Squires 1990; Campbell 1992).

A modern chemoherm can be recognized on the basis of its intimate association with fluid and hydrocarbon venting on the sea floor, the presence of chemosymbiotic communities, and the anomalously light $\delta^{13}C$ compositions of its authigenic carbonate constituents (MacDonald et al. 1989; Aharon et al. 1992; Aharon 1994; Roberts and Aharon 1994). In contrast, the recognition of fossil vents in the geologic record is complicated because of the absence of physical evidence of venting and because the specific enzymes characteristic of chemosynthetic metabolism (Childress and Fisher 1992) had vanished with the decay of the soft organic tissues of the paleovent fauna. Under these circumstances there are only two reliable indicators of paleoventing. One is the anomalously negative carbon isotope compositions of the carbonates derived from oxidized hydrocarbons (Aharon et al. 1993). The other is the mineral skeletal remains of mussels and clams permitting taxonomic identification and derivation of common lineages between the fossil fauna and the living chemosymbiotic counterparts at extant vents (Taviani 1994).

Blocks of limestones locally called "calcari a *Lucina*" and scattered throughout the Tertiary sediments of the Italian Apennines have been known for many years, but their exact stratigraphic position and origin have been the subject of controversy (Ricci Lucchi and Vai 1994). These limestone blocks have recently been reinterpreted as cold-vent deposits (Clari et al. 1988; Terzi 1992; Aharon et al. 1993). The objective of this study is to report the results of

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the most recent field observations and laboratory analyses of "calcari a *Lucina*" samples, which support our interpretation of the limestone blocks as vestiges of Miocene-age submarine venting on the western Tethys sea floor.

Geologic setting

The "calcari a *Lucina*" limestones occur within Miocene-age sediments running the length of Italy from Piedmont in the north to Sicily in the south (Fig. 1). The limestone blocks, of about 2–3 m average dimensions and up to a maximum of 10 m (Fig. 2A), are characterized by their typical carbonate lithology embedded in the siliciclastic formations and by the abundance of large fossil clams (up to 20 cm long) of the genus *Lucina*. These limestones are best described as blocks because they are discontinuous and do not show clear stratigraphic relation with the surrounding rocks consisting of erosion-prone marls and sandstones.

The limestones are particularly common in the Tuscan–Romagna region of the Apennines (Fig. 1), where they are associated with turbidite formations capped by hemipelagites and represent the infilling of Miocene foredeep basins (Ricci Lucchi and Vai 1994). The turbidite–hemipelagite paired sequences are overlain by Messinian-age evaporites (Fig. 1). The formation and demise of these elongated foredeep basins occurred during major tectonic phases of the Apennine orogeny that involved compression, basin opening at the front of the chain, infilling with turbidite deposits, and final thrusting proceeding from southwest to northeast (Boccaletti et al. 1990).

Most *Lucina*-rich limestone blocks are associated with hemipelagites and turbidites of the Marnoso-arenacea Formation of Langhian to Tortonian age and are located in close proximity to major thrusts and faults. The blocks occur at different stratigraphic levels, the highest level being only few meters below the Messinian evaporites (Fig. 1). Scattered limestone occurrences related to major fault zones have also been mapped within slump horizons of Serravallian-age (Berti et al. 1994).

Data acquisition

Over 20 occurrences of *Lucina*-rich limestones have been mapped and sampled in the Tuscan–Romagna Apennines

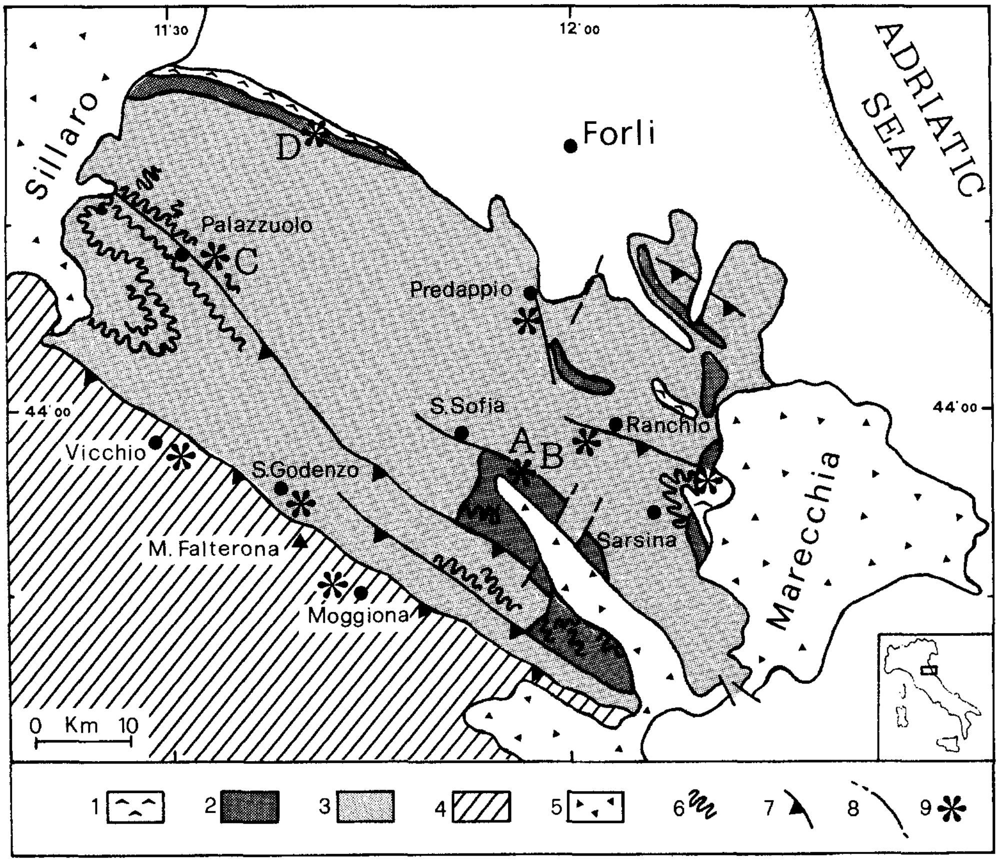

Fig. 1 Geological sketch map of the studied area in the Tuscan–Romagna Apennines of Italy (modified after Ricci Lucchi 1975) showing locations and stratigraphic positions of the "calcari a *Lucina*" limestone blocks described in this study. Legend: 1, Messinian-age evaporites; 2 and 3, Marnoso-arenacea Formation of Langhian to Tortonian age (2, hemipelagites, 3, turbidites); 4, Tuscan units; 5, allochtonous and semiallochtonous units; 6, slumping; 7, major thrusts; 8, faults; 9, occurrences of *Lucina*-rich limestones. A: Raggio; B: Case Rovereti; C: Colline; D: Rontana

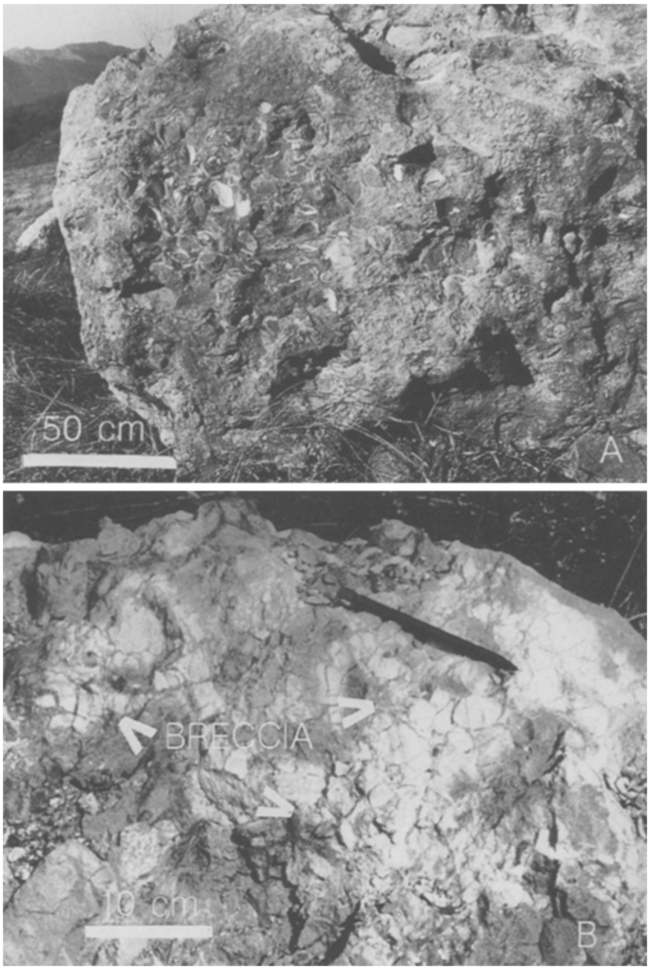

Fig. 2 Field aspects of the "calcari a *Lucina*" limestone blocks. **A** Limestone block seated in a slump belt (Colline di Palazzuolo sul Senio, Fig. 1); **B** Limestone block showing autobrecciated structures (Raggio, Fig. 1)

area (Fig. 1). Petrographic and microfacies analyses were performed on 44 thin sections. Over 100 powdered samples were analyzed by X-ray diffraction (XRD) for mineralogical compositions using a Philips APD 3520 machine and by an automated Nier-type triple-collector mass spectrometer for stable oxygen and carbon isotope compositions using standard procedures (Aharon 1988). All the XRD and stable isotope analyses were performed in the respective laboratories at Louisiana State University.

Results

Field observations

Because most of the limestone blocks show evidence of reworking, it is difficult to reconstruct with confidence the exact relation with the sediments in which they are embedded, or their original structures. Some of the structures that are still recognizable in the outcrops and may have a bearing on the origin of the limestones are described below.

Brecciated structures and rounded, often elongated, pelitic intraclasts that seem to concentrate on the external surfaces are common; the breccias are sometimes recemented by sparry calcite (Fig. 2B). Nodular (nodules 3–4 cm in diameter) and rare doughnut (7–8 cm in diameter) structures have been observed in micritic limestones (Fig. 3A). Rims and veins of acicular and botryoidal aragonite of variable thickness (Fig. 3B) are more common in the younger, Tortonian-age, occurrences than in the older ones. These structures observed in the "calcari a *Lucina*" limestones show striking similarities to the ones reported from modern chemoherms in the North Sea (Hovland et al. 1987), Oregon accretionary prism (Kulm and Suess 1990), and northern Gulf of Mexico (Roberts and Aharon 1994).

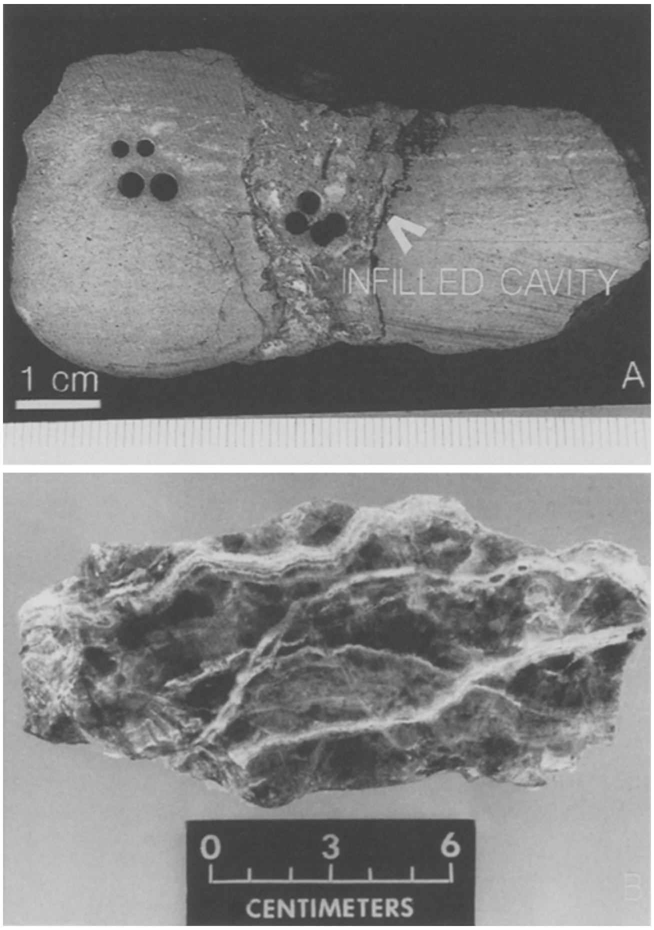

Fig. 3 Petrologic aspects of the limestone blocks. **A** Micritic limestone with central cavity infilled by coarser sediment and shell fragments (Case Rovereti, Fig. 1) resembling the doughnut-shaped structure reported from the extant carbonate buildups at Oregon vents (Kulm and Suess 1990). These structures are considered to represent fossil microconduits of methane-rich fluid venting; **B** Massive botryoidal aragonite (Rontana, Fig. 1) displaying anomalously negative $\delta^{13}C$ values (down to $-51.7‰$). These aragonites are thought to represent rapid precipitation resulting from aerobic bacterial oxidation of methane in fast flux vents (see text)

Petrography and mineralogy

The two most remarkable characteristics of the "calcari a *Lucina*" blocks are their variable lithologies and their fossil contents. The following four distinct lithofacies have been recognized on the basis of hand specimen and thin section examinations: (1) micritic limestones; (2) biomicritic marly limestones; (3) fine to very fine calcarenitic limestones, and (4) coarse calcarenitic limestones. The unifying petrologic aspect of the four lithofacies above is the pervasive micritic carbonate cement, which contributes to the hardness and the extremely low porosity of the rocks and which lends support for their generic identification as limestones. The frequently observed correspondence between the lithofacies of the limestone blocks and those of the host sediment in which they are embedded (e.g., marly limestones associated with marls and calcarenitic limestones associated with sandstones) lends credence to our contention that these blocks have undergone only minor displacement.

The marly and fine calcarenitic limestone facies are associated with abundant fossil remains consisting of thick recrystallized closed shells and molds of lucinid and vesicomyid-like clams, thin shells of modiolid-like mussels, recrystallized shells of gastropods, and unidentified vestimentiferan-like tubular fossils lacking preferential orientation. These fossil fauna show striking similarities to the chemosymbiotic communities living around vents (Taviani 1994). In contrast, micritic and coarse calcarenitic limestone facies are markedly impoverished in their fossil content. The observed relation between lithofacies and their fossil content above suggests a link between the nature of the sediment substrate, the intensity of fluid and hydrocarbon venting, and the density of vent fauna. These proposed relations deserve further exploration in future studies.

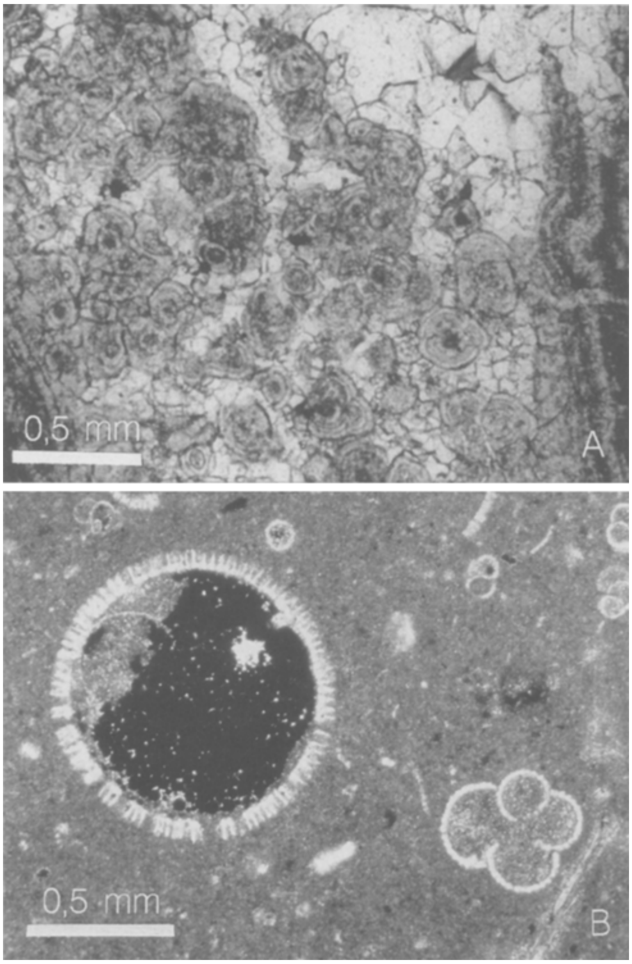

Fig. 4 Petrographic aspects of the limestone blocks (Rontana, Fig. 1). **A** Concentric texture consisting of calcite-aragonite layers resembling bacteria-mediated precipitates; **B** Authigenic framboids of pyrite infilling the test of the planktonic foraminifer *Orbulina*

Thin section and mineralogical studies by XRD indicate that both authigenic and allogenic components are present in the "calcari a *Lucina*" limestones. The authigenic components consist of carbonates (calcite, dolomite, and aragonite) and sulfides (mainly pyrite) whereas the allogenic components consist primarily of quartz and micas and a minor fraction of feldspars and sandstone clasts. The phyllosilicates often show preferred orientation in thin sections contributing to their solifluction-like appearance. Shells of planktonic foraminifera are abundant in many sections but benthic foraminifera species are rare (Aharon and Sen Gupta 1994). The authigenic carbonates forming the bulk of the micritic cement consist primarily of micro- to cryptocrystalline low-Mg calcite as well as dolomite. Sparry calcite cement is present locally in association with breccias. Radial aragonite cements and massive aragonite botryoids (Fig. 3B) are infilling veins and cavities in the limestones. Concentric features consisting of calcite and aragonite mineralogies (Fig. 4A) show striking resemblence to carbonate textures produced in the laboratory by bacteria-mediated precipitation (Buczynski and Chafetz 1991). The pyrites occur as small framboids that are either intimately associated with the micritic cement or are infilling cavities such as the inside chambers of foraminifera tests (Fig. 4B).

Stable isotopes

The oxygen and carbon isotope compositions of 116 samples pertaining to the "calcari a *Lucina*" limestones are graphed in Figs. 5 and 6, respectively. Limestones yield $\delta^{18}O$ values in the range of $-11‰$ to 5‰ relative to the PDB standard (Fig. 5). Samples that yield $\delta^{18}O$ values more negative than $-3‰$ are also the ones that have been most affected by postdepositional diagenetic alteration. Therefore the ^{18}O depletions observed for these samples can be attributed to dissolution–reprecipitation processes from continental freshwaters that were substantially depleted in the heavy oxygen isotope relative to a Miocene-age Mediterranean seawater $\delta^{18}O$ values of 0‰ to 3‰ (Aharon and Sen Gupta 1994). On the other end of the spectrum, samples heavier than 2‰ in $\delta^{18}O$ contain a substantial dolomite component (from about 15% to about 80% dolomite) coexisting with micritic calcite cement.

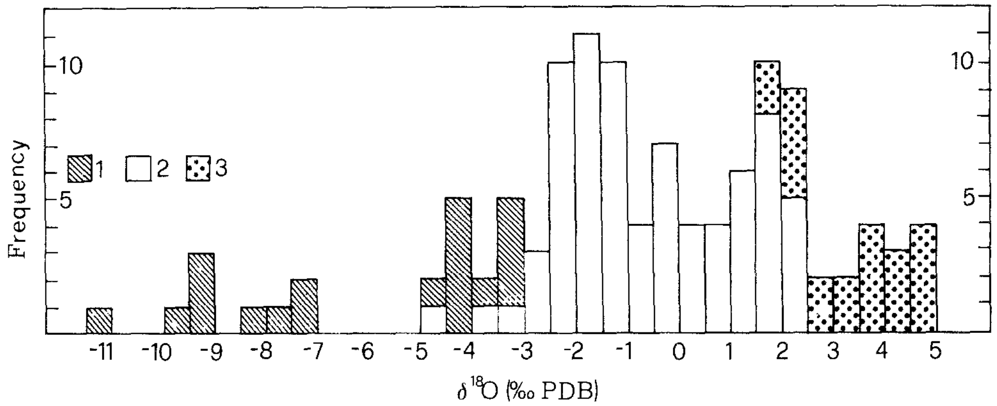

Fig. 5 Histogram showing the frequency distribution of $\delta^{18}O$ values in 116 samples analyzed in this study. Legend: 1, samples altered by meteoric diagenesis; 2, calcite samples; 3, samples containing in excess of 20% dolomite

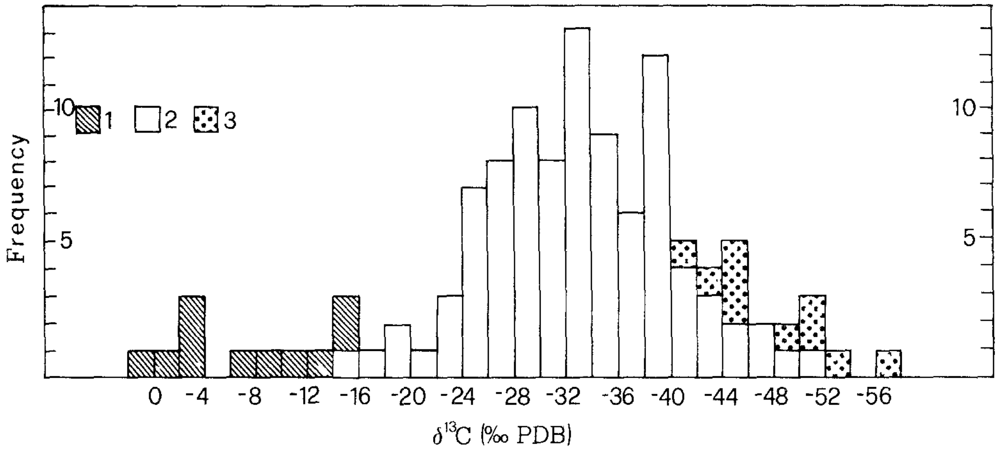

Fig. 6 Histogram showing the frequency distribution of $\delta^{13}C$ values in 116 samples analyzed in this study. Legend: 1, samples altered by meteoric diagenesis; 2, calcite and dolomite-rich samples; 3, aragonite-rich samples

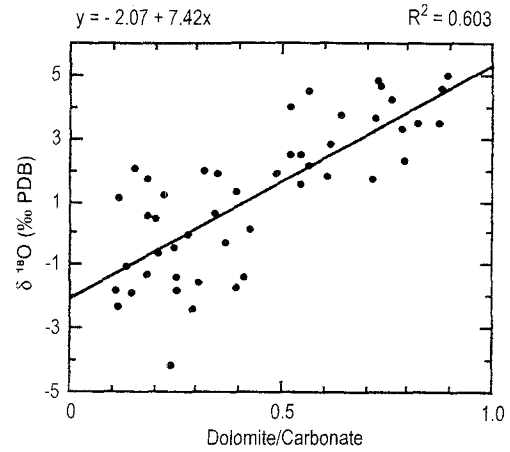

Fig. 7 Relation between the dolomite content of the samples expressed as dolomite to total carbonate ratio and the $\delta^{18}O$ values showing a statistically significant positive correlation (only samples containing over 10% dolomite were considered in this plot)

Moreover, there is a positive correlation observed between the dolomite content of the samples and their $\delta^{18}O$ values (Fig. 7). The ^{18}O enrichment of the dolomite-rich samples is consistent with the $\delta^{18}O$ fractionation of 3.4–5.6‰ between coprecipitated dolomite–calcite phases at sedimentary temperatures (Aharon 1988). Samples considered to be unaffected by diagenesis or dolomitization show a $\delta^{18}O$ range of −3‰ to 1.5‰ that are likely to represent the ambient seawater conditions during the deposition of the authigenic carbonates (Aharon and Sen Gupta 1994).

Carbon isotope compositions of the limestones range from 0‰ to −58‰ relative to the PDB standard (Fig. 6). Samples that are more positive than −15‰ show clear indications of diagenetic alteration; the remainder yield ^{13}C depletions (i.e., $\delta^{13}C$ of −16‰ to −58‰) that are anomalous relative to $\delta^{13}C$ values of normal marine carbonates (i.e., −1‰ to 3‰; Aharon et al. 1992). These anomalously negative $\delta^{13}C$ values are likely to reflect a carbon source derived from oxidation of hydrocarbons (primarily methane), which are anomalously depleted in the ^{13}C isotope relative to other carbon sources readily available in the marine environment (Aharon et al. 1992; Roberts and Aharon 1994). It is interesting to note that samples with the most negative $\delta^{13}C$ values are also the ones that have an aragonite mineralogy (Fig. 6). The distinction between the aragonite and calcite mineralogies observed in their $\delta^{13}C$ values is likely to reflect differences in their paragenesis and/or variable chemistry of the fluids involved in their deposition (see below).

Discussion

The striking similarities between "calcari a *Lucina*" limestones and modern chemoherms from extant hydrocarbon venting sites, manifested by their analogous petrographic, faunal, and stable isotope attributes presented in the preceding section, corroborates our contention that the former are relicts of Miocene-age submarine venting. Below we address questions relevant to the specific processes involved in the formation of the limestone blocks, the causes and consequences of fluid and hydrocarbon venting, and the likely relationship between the temporal distribution of the fossil chemoherms and the tectonic phases of the Apennine orogeny.

Model of deposition of the "calcari a *Lucina*" limestones

The anomalously negative $\delta^{13}C$ values of limestone samples unaffected by diagenesis (Fig. 6) and the general absence of crude oil relicts that are resistant to weathering (e.g., asphaltines) suggest that methane was the dominant hydrocarbon phase vented with the fluids to the seabed. The source of methane could have been either from a deep-sited, thermogenic origin and/or from a shallow, biogenic origin.

The carbon isotope data of the limestones does not permit us at this stage to single out with confidence the exact source of methane. This is because thermogenic and biogenic methane sources tend to overlap to a great extent in their $\delta^{13}C$ compositions, and we cannot exclude the possibility that mixing with a ^{13}C-rich carbon source derived from the dissolved inorganic carbon (DIC) in seawater had occurred prior to the deposition of the limestones (Aharon et al. 1992). We note, however, that methane dissolved in the fluids presently intercepted in the deeply buried Miocene sediments of the Po Plain in northern Italy is predominantly of biogenic origin (Mattavelli et al. 1983). It is conceivable, therefore, that the source of methane involved in the deposition of the Miocene-age "calcari a *Lucina*" limestones was also of biogenic origin and it was derived from bacterial degradation of shallow-sited organic matter trapped in the turbidites.

On the basis of petrography, structures, and carbon isotope data, we suggest that two distinct styles of venting left their mark on the limestones. The first relates to slow flux venting through the sulfate reduction zone where the fluid acquired CO_2 and H_2S derived from coupling of anaerobic bacterial methane oxidation/sulfate reduction (Table 3 in Aharon et al. 1992). The sluggish venting of the methane-derived CO_2 and sulfate-derived H_2S to the seabed would result in the enrichment of these compounds in the pore fluids and in the precipitation of micritic calcites and coexisting pyrite phases below the sediment–water interface. The massive precipitation would result in a considerable reduction of porosity of the host sediment and in the trapping of the siliciclastic and bioclastic components in the lithified limestone.

The other style relates to fast flux venting where the methane-rich fluids under pressure bypassed the sulfate reduction zone. The breakdown of methane into CO_2 may have occurred anaerobically in a bacterial–methane oxidation zone close to the sediment–water interface or in the aerobic bottom waters by bacterial consumption of dissolved oxygen (Table 3 in Aharon et al. 1992). Under these circumstances it can be predicted that carbonate deposition would occur in the absence of sulfides.

Several lines of evidence corroborate our proposed distinction between slow and fast venting imprints. First, micritic calcites are commonly associated with pyrite, suggesting that methane oxidation and sulfate reduction processes were contemporaneous. These calcites are generally only moderately depleted in ^{13}C (i.e., $\delta^{13}C$ in the range $-16‰$ to $-40‰$, Fig. 6), as would be expected if mixing of methane-derived carbon with ^{13}C-rich sources (e.g., seawater DIC) occurred during sluggish venting. Second, radial fibrous aragonite and massive aragonitic botryoids (Fig. 3B) that are infilling veins and voids (conduits?) lack sulfides and yield the most negative $\delta^{13}C$ values ($-40‰$ to $-58‰$) among the carbonate phases. We interpret the extreme ^{13}C depletions exhibited by the aragonites as a manifestation of rapid precipitation, which inhibited dilution of the methane-derived carbon with other carbon sources during fast venting. The testimony of violent gas outbursts is also imprinted in the commonly found autobrecciated structures (Fig. 2B) that are similar to the ones reported by Hovland et al. (1987) from the pockmarks of the North Sea. According to the pockmark model, the processes involved in the formation of the brecciated structures observed in the "calcari a *Lucina*" limestones were likely to be: (1) deposition of micritic carbonate crusts within the host sediment; (2) shattering of the crusts into fragments during violent gas and fluid outbursts resulting from buildup pressures, and (3) recementation of the fragmented micritic calcite crusts with fibrous aragonite. Presently most of the former aragonites are recrystallized to sparry calcites but occasional preservation of the original aragonite mineralogy suffices to confirm the veracity of the depositional model outlined above.

Two other important consequences of the Miocene-age venting are worthy of further discussion. One is the widespread distribution of dolomite in the "calcari a *Lucina*" that reaches abundance levels of up to 80% of the bulk rock. The dolomite occurs as dolomicritic cement in close association with the micritic calcite. Although we have not yet analyzed the isotopic compositions of pure dolomite, the positive correlation observed between the dolomite content and the $\delta^{18}O$ values of the carbonates (Fig. 7) suggests that dolomite was precipitated in isotopic equilibrium with the coexisting calcite. Direct precipitation of dolomite from seawater is thought to be inhibited by the presence of sulfates (Baker and Kastner 1981). In the context of the "calcari a *Lucina*" fossil vents, the prolific dolomite precipitation may have been fostered by the consumption of the sulfate during the anaerobic methane–oxidation process as also postulated for the dolomites reported from the Oregon hydrocarbon vents (Ritger et al. 1987).

Another consequence of the hydrocarbon-rich fluid venting into the benthic environment was the development of biological oases of chemosynthetic communities evidenced by the abundance of clams and mussels of taxa similar to the ones thriving around extant hydrocarbon vents (Taviani 1994). In this stage of our study we can only speculate on the chemosynthetic pathways of the fossil fauna because all the organic soft parts are lost, and the shells are recrystallized. However, the low species diversity coupled with their high density, the dominance of large individuals, and the high frequency of articulated valves offer indirect support to the chemosynthesis model.

Relation of venting with tectonic history of the Apennines

The northern Apennine orogeny was manifested during the Miocene by the migration of a thrust belt foredeep system from southwest to northeast that was accompanied by extension, block faulting, flexuring, compression, and thrusting (Ricci Lucchi and Vai 1994). These tectonic events were likely to have a profound effect on the dewatering and compaction of the turbidite and hemipelagite deposits of the foredeeps and to have caused the expulsion of hydrocarbon-rich fluids in a manner analogous to the fluid flow processes occurring at modern accretionary prisms in the Oregon and Japan subduction zones (Kulm and Suess 1990; Boulegue et al. 1986). Unlike the modern settings where the fluid conduits are clearly identified with shear planes and extension zones on top of thrusts, the paleofluid conduits associated with the deposition of the *Lucina*-rich limestones are difficult to identify with certainty because of reworking of the limestone blocks due to faulting and slumping (Berti et al. 1994).

The extensive development of the "calcari a *Lucina*" limestones during the Tortonian relative to the preceding and following Miocene stages is evidenced by the relatively high density and spatial extent of the blocks, the abundance of aragonite and dolomite phases, and the proliferation of the chemosynthetic-like fauna. If so, it may be suggested that methane-rich fluid venting was more common and more intense during the Tortonian stage than during any other Miocene time.

Conclusions

1. The "calcari a *Lucina*" limestone blocks associated with Miocene-age turbidites and hemipelagites in the Tuscan–Romagna region of the northern Italian Apennines possess petrographic, structural, and faunal characteristics that are remarkably similar to the carbonate buildups (chemoherms) recently reported from hydrocarbon-rich venting sites in the modern oceans. This conclusion is supported by the $\delta^{18}O$ and $\delta^{13}C$ compositions of the limestones, which are generally compatible with their deposition from seawater-derived fluids and with a carbon source derived from bacterial oxidation of biogenic methane dissolved in the venting fluids.

2. Systematic variations in the abundance of carbonate mineralogies accompanied by changes in stable isotope compositions, abundance shifts in sulfides, and occurrences of autobrecciated structures lead us to conclude that both slow flux venting and fast flux venting left their diagnostic marks on the limestones.

3. The two most important consequences of bacterially mediated methane oxidation and sulfate reduction on the chemistry of the vented fluids are the widespread dolomitization and the development of biological oases consisting of chemosynthetic-like communities.

4. The episodic tectonic movements that accompanied the Apennine orogeny during the Miocene is likely to be the principal cause for dewatering and compaction of the clastic sediments deposited in the foredeeps and for subsequent expulsion of methane-rich fluids to the seabed, which promoted the formation of the *Lucina*-rich limestones.

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