Methane-seep brachiopod fauna within turbidites of the Sinaia Formation, Eastern Carpathian Mountains, Romania

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ABSTRACT

This study elucidates the paleoecology and paleobiogeography of the Early Cretaceous brachiopod Peregrinella known in museum collections from a few localities in Romania, supplemented with new material from a rediscovered locality first mentioned in the 1870s. Most Peregrinella fossils are enclosed in mass waste deposits, but at two sites authigenic limestones with assemblages of brachiopods preserved in life position have been recognized. Paleontological, petrographic, stable isotopic, and organic geochemical investigations of these brachiopod-bearing limestones from the Upper Sinaia Formation, Eastern Carpathian Mountains, Romania, confirm Peregrinella as having lived at methane seeps in a siliciclastic-dominated flysch basin. The seeps developed on the slope of the External Dacides Basin. The new collections of Peregrinella indicate that shells derived from contemporaneous intrabasin methane seeps and were transported downslope by turbidity currents. Previous paleoecological models that consider Peregrinella to be solely derived from transport downslope from shelf environments are questionable especially as Peregrinella has never been recovered from typical shelf faunas; in the instance documented here from the External Dacides Basin methane-seep faunas with Peregrinella are likely to be the origin of such allochthonous faunas. The Sinaia Formation was deposited in a deep-water marine basin, derived from an intracontinental rift that developed during Late Jurassic–Early Cretaceous extension. The fractured and faulted basin margin provided the backdrop for the development of the methane seepage and the associated fauna. Foraminifera from background sediments in the sequence with turbidites confirm a late Hauterivian to early Barremian age for Peregrinella within the Sinaia Formation. This is significant because it indicates that Peregrinella ranged through into the Barremian, whereas it has typically been considered to range only as high as the Hauterivian.

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1. Introduction

The Early Cretaceous brachiopod genus Peregrinella is one of the largest-sized Mesozoic brachiopods, reaching maximum dimensions of 10 cm or greater. The brachiopod has attracted much attention not only for this large size but also for its enigmatic occurrence en masse in “Peregrinella” beds (e.g., Biernat, 1957; Ager, 1965, 1968; Thieuloy, 1972; Campbell and Bottjer, 1995a; Sandy, 1995; Kiel and Peckmann, 2008) and also for its disjunct distribution and unusual paleoecology and paleobiogeography. Peregrinella is never found with other brachiopod genera and is absent from contemporaneous brachiopod shelf faunas that can typically be expected to include a few brachiopod species, including both terebratulids and rhynchonellids (Middlemiss, 1979, 1984). Peregrinella is found as monospecies or monogenic brachiopod accumulations that can be part of more diverse invertebrate assemblages that may include associated molluscs (Stanton, 1895; Biernat, 1957; Campbell et al., 1993; Kiel and Peckmann, 2008) that belong to chemosynthesis-based lineages. The disjunct distribution of Peregrinella (France, Italy, Poland, Tibet, California, Alaska, Crimea (Ukraine); e.g., Ager, 1967; Owen, 1973; pre-1995 occurrences summarized in Table 1 by Campbell and Bottjer, 1995a; Sandy and Blodgett, 1996; Posenato and Morsilli, 1999; Kiel and Peckmann, 2008), and its isolation from other brachiopods, have led to the development of intriguing paleoecological models, such as Peregrinella living restricted to rocky shorelines and subsequently washed downslope into basins—with the original rocky shoreline never preserved (Ager, 1965); however, no brachiopods are known to have had such a paleoecology (Art Boucot, personal communication, 2011). A shallowing, uplifting basin floor within the deeper water Vocontian Basin was proposed to explain the occurrence of Peregrinella in southern France (Thieuloy, 1972).

The fascination with Peregrinella has appeared warranted as its story has become even more intriguing. Peregrinella is now considered an associate of chemosynthesis-based communities (e.g.,
Campbell et al., 1993; Campbell and Bottjer, 1995a; Kiel and Peckmann, 2008). Macsotay (1980) and Lemoine et al. (1982) had first suggested hydrothermalism to explain the occurrence of Peregrinella in outcrops in the south of France. Once petrographic and isotopic work was combined with paleontological studies (for a Californian occurrence, Campbell et al., 1993, 2002), it became clear that the brachiopod was associated with hydrocarbon seeps during life. In a review of the occurrence of Peregrinella, Campbell and Bottjer (1995a) identified 17 localities globally. Of these (including new occurrences) only 2 have been confirmed as having a methane-seep origin: (1) California (Campbell et al., 1993; Campbell and Bottjer, 1995a; Birgel et al., 2006) and (2) Crimea (Kiel and Peckmann, 2008; Peckmann et al., 2009). This study marks the third confirmed association of Peregrinella with chemosynthesis-based seep environments. Presumably work will continue on trying to ascertain the paleoecology of other Peregrinella occurrences in an attempt to determine if Peregrinella is, in fact, restricted to these environments. Other representatives of the rhynchonellid superfamily Dimerelloidea, to which Peregrinella belongs, have also been shown to be associated with chemosynthesis-based environments (Sandy and Campbell, 1994; Peckmann et al., 2001, 2007, 2011; Gischler et al., 2003). Of these, Dzieduszyckia from the Devonian is known from hydrocarbon-seep carbonates but also from sedimentary strata apparently unaffected by seepage (Peckmann et al., 2007). Therefore Dzieduszyckia is considered to be a brachiopod that is more plastic in its ecological tolerances (Peckmann et al., 2007). Such an association has not been confirmed for Peregrinella, and based on current knowledge, it seems unlikely given the absence of Peregrinella from shelf brachiopod faunas.

Here we describe different modes of occurrence of Peregrinella from the Upper Sinaia Formation, Eastern Carpathian Mountains, Romania, studying the paleontological, taphonomical, sedimentological, petrographic, stable isotopic, and organic geochemical characteristics of brachiopod-bearing rocks. Our study reveals that the paleoecology of this brachiopod can only be constrained based on fossil associations preserved in authigenic rocks that formed in the brachiopods’ habitat. An attempt to reconstruct brachiopod paleoecology from accumulations of transported shells is hampered by the apparent lack of diagnostic features that help to identify the paleoenvironmental conditions of the original habitat. Criteria have yet to be identified from brachiopod shells that would allow such an assignation.

2. Peregrinella records in Romania

In Romania Peregrinella is known from turbidites and blocks of limestone from the Neocomian flysch deposits of the Upper Sinaia Formation, well exposed in outcrops in the southern part of the Eastern Carpathians (Fig. 1). The first mention of Peregrinella in the Eastern Carpathians was by Herbich (1878, p. 248) who recorded Rhynchonella peregrinella d’Orbigny from sandstone deposits that outcrop in the Vârghiș Valley. Subsequently Peregrinella was mentioned by Toulă (1911), Macovei (1927), Macovei and Atanasu (1933), Bâncilă (1958), Zberea (1962), Filipeşcu and Grigorescu (1966), Patrulius (1969), Păuluc (1968), Gräf (1975), Földváry (1988), Solcanu (1991, 2007), as well as Sândulescu and Dimitrescu (2004). All of these authors refer to a total of four localities: (1) Zizin Valley; (2) Vârghiș in the Sârman Valley; (3) Belin Valley (Pierșani–Baraolt area) and; (4) Cădăr-ești in the Ciughiu Valley (Fig. 1). These localities with Peregrinella are in the southern half of the Eastern Carpathians (in the Pierșani, Baraolt, Bocot, and Ciuc mountains). All previous authors mention Peregrinella from coarse sandstones and/or from isolated blocks of limestone found as loose samples in valleys where the Sinaia Formation is exposed. However, none of these works mentions the exact location of the limestones within the flysch succession. We were unsuccessful in our recent efforts to relocate these limestones. The limestone samples with Peregrinella that we studied in the present paper are samples held in the paleontological collections of the University of Bucharest (samples from Zizin, Pierșani–Baraolt area, and Cădăr-ești) and the Geological Institute of Romania (samples from Belin). During field work in 2009 and 2010 we rediscovered the turbidite outcrop with Peregrinella from Vârghiș (Herbich, 1878; Bâncilă, 1958 first mentioned the occurrence of Peregrinella from the area but not a specific outcrop). Two additional samples (one Peregrinella specimen in carbonate and one sandstone sample with external molds of Peregrinella from Cădăr-ești, Ciughiu Valley) were donated to the University of Bucharest collections in 2010 by Mihai Solcanu. We present here the first integrated study on the occurrences of Peregrinella in Romania concerning macro- and micropaleoecology, taphonomy, microfacies, sedimentology, isotopic geochemistry, and biomarkers.

3. Geological setting and stratigraphy

The stratigraphy of the Sinaia Formation in the Eastern Carpathians has been studied by numerous authors, among them, Popovici-Hateg (1898), Protesscu (1936), Oncescu (1965), and Patrulius (1969). Numerous refinements of the general geological context and stratigraphy of the Sinaia Formation have subsequently been made. However, in this paper we restrict our discussion to the authors that studied the stratigraphy of the Sinaia Formation where Peregrinella occurs. The flysch deposits of the Sinaia Formation belong to the sedimentary cover of the Ceahlău Nappe, External Dacides of the Carpathian Orogen (Fig. 1), corresponding to an external rift (extensional basin). The External Dacides consists of narrow north–south oriented nappes that developed from a Jurassic-Cretaceous paleo-rift within the European continental margin (Sândulescu, 1994). This rifting was associated with Tethyan Ocean spreading and reached its maximum extension in the Middle-Late Jurassic. In the Eastern Carpathians, the Black Flysch, Baraolt and Ceahlău nappes are floored by intraplate basalts, resulting from this extension. The basalts are overlain by flysch formations. In the Ceahlău Nappe, flysch is represented by the Sinaia Formation. In the Middle Cretaceous, compressional tectonics resulted in a thick pile of nappes, the Median Dacides (Sândulescu, 1994), which in turn were covered by obducted slabs of Tethyan oceanic crust (the Transylvanides). Both the Median Dacides and the Transylvanides were overthrust onto the Sinaia flysch of the External Dacides (Kräutner and Bindea, 2002), End-Cretaceous compression is evident in the External Dacides (Sândulescu, 1994).

The Sinaia Formation is up to 2500 m thick (Patrulius, 1969) and consists mainly of flysch represented by carbonate-rich siliciclastic and marly sediments, divided into three members based on the petrography of the units (Figs. 2, 3). The Peregrinella levels are located within the Upper Sinaia Member that is 400–500 m thick (Fig. 3) and represented by shales alternating with sandstones, marls, limestone, sandy-limestone, and silty-limestone. The succession reveals intercalations of conglomerates, breccias, and coarse gravelly sandstone toward its upper part.

Macro- and microfossils are not very common in the Sinaia Formation, therefore the stratigraphic age of the Sinaia Formation has been the subject of much debate. Patrulius (1969) made a very detailed inventory of all the paleontological arguments, concluding the range of the Sinaia Formation is Berriasian to Late Hauterivian, with the Upper Sinaia Member restricted to the Upper Hauterivian (Murgeanu et al., 1959; Patrulius, 1969). However, Avram and Matei (1964), Avram (1970), and Gräf (1975) described stratigraphically important taxa from the Upper Sinaia Member: Lamelaptychus angulosostatus (Peters), Neolissoceras gracianum d’Orbigny, Barrenites subdifflusis (d’Orbigny), Euphylliloceras tethys d’Orbigny, and Lytoceras sp. These fossils indicate a Late Hauterivian to Early Barremian age.

The section we studied with Peregrinella is located in the Sârman Valley, a tributary to the Vârghiș Valley, near the village of Vârghiș. We rediscovered the outcrop that Bâncilă (1958) briefly mentioned in his monographic work concerning the geology of the Eastern Carpathians. The flysch deposits of the Upper Sinaia Member outcrop...
on the eastern slope of the valley, 500 m upstream from the confluence with the Vârgâş Valley: N 46°08′39.5″; E 25°32′06.2″; 445 m elevation. A sedimentary succession was measured with an exposed stratigraphic thickness of about 10 m and a lateral extent of almost 8 m (Figs. 2, 4). The succession shows clear bedding (Fig. 2A–C) and is represented by successive sequences of sandstones, shales with silt intercalations and subordinate marlstones to limestones (Figs. 2C, 4). These sequences are interrupted by thick beds (20–70 cm thickness) of conglomerates and breccias. Three levels with allochthonous Peregrinella have been located within the succession (Fig. 4): the first level is at 1.14 m, on the base of a coarse- to medium-grained sandstone bed, and the overlying two levels (at 8.35 m and 9.64 m) are on the base of two beds of matrix-supported breccias. The general dip of the strata is approximately 85° East.

4. Materials and methods

The material investigated in the present paper is represented by two categories of samples. First are limestone samples with abundant Peregrinella specimens (shells), as well as isolated specimens of Peregrinella (extracted from carbonate rocks) housed in collections of the University of Bucharest (Laboratory of Palaeontology, Bucharest: LPB) and Geological Institute of Romania, Bucharest. A total of 23 samples representing isolated brachiopod specimens and carbonate rock-samples have been studied so far. These samples are from the Sinaia Formation exposed in the Zizin Valley (LPBIII: 344, 366, 373, 374, 375; 298-2 specimens; 281-2 specimens), the Belin Valley (LPBIII: 364, 365, 367–372; IGR-P 3980: 4 samples), and the Cădăroș-Ciughiuș Valley (LPBIII 376). Second are rock samples represented by coarse gravelly sandstone and breccia with Peregrinella shell fragments (LPBIII 377–388), collected from the Vârgâş section from turbidites of the Sinaia Formation. More than 50 thin-sections and polished slabs were studied petrographically by polarized light microscopy.

The limestone beds from the Vârgâş area were analyzed by the loss on ignition method (LOI) (Dean, 1974; Heiri et al., 2001) in order to determine the amount of organic matter (LOI: 550 °C) and CO2 (LOI: 950 °C). From the amount of CO2 the content of CaCO3 was determined. The same samples were analyzed to obtain...
geochemical composition as oxides by the X-ray fluorescence method, using a HORIBA XGT 7000. The working parameters were as follows: acquisition time 100 s; XGT diameters 100 μm; X-ray tube voltage 30.00 kV; current 0.900 mA.

Six micropaleontological samples were collected from siltstone and laminated shale levels (Figs. 2B, 4). Samples were disaggregated using a sodium sulfate anhydrous solution (Na₂SO₄—“Glauber” salt), followed by several freeze/defrost cycles. Finally, samples were washed using a 63 μm sieve.

Macrofossils were prepared using an air scribe, small chisels, and ultrasonic bath system and the internal morphological characters have been observed in transverse serial sections made from one brachiopod specimen.

Powdered samples for oxygen and carbon stable isotopic analyses were taken from the surfaces of polished limestone slabs using a hand-held microdrill. Samples of individual carbonate phases were reacted with 100% orthophosphoric acid in a vacuum at 75 °C, and the evolved CO₂ gas was analyzed with a Finnigan MAT 251 mass spectrometer at the University of Bremen. The δ¹³C and δ¹⁸O values are reported relative to the Vienna PeeDee belemnite (V-PDB) standard (standard deviation < 0.03‰) and appropriate correction factors were applied.

Limestone samples from the Belin locality were too small to allow for biomarker analysis. The preparation and decalcification procedure of a limestone sample from the Zizin locality (sample weight: 330 g) was performed after a method described previously (Birgel et al., 2006). After the saponification procedure with KOH (6%) in methanol, the samples were extracted with a microwave extraction system (CEM MARS X) at 80 °C and 300 W with a dichloromethane-methanol (3:1) mixture. The total extracts were pre-cleaned by a separation into a n-hexane soluble maltene fraction and a dichloromethane soluble asphaltene fraction. Then, the maltene fraction was separated by column chromatography into four fractions of increasing polarity following the procedure described in Birgel et al. (2008a). Only the hydrocarbon fraction was found to contain compounds that can be used to describe the processes, which lead to carbonate precipitation at the ancient seep. The more polar fractions were affected by thermal maturation and biodegradation and do not contain genuine molecular fossils that reflect the sedimentary environment. The hydrocarbon fraction was measured by gas chromatography–mass spectrometry with a Thermo Electron Trace DSQ-II equipped with a 30 m Rxi-5 MS fused silica column (0.25 mm inside diameter, 0.25 μm film thickness). The carrier gas used was helium.

The temperature program was: 60 °C, 1 min isothermal; from 60 to 150 °C at 10 °C/min, from 150 to 320 °C at 4 °C/min; 22 min isothermal at 320 °C. Identification of individual compounds was based on retention times and published mass spectral data in comparison with other samples. Compound-specific carbon isotope analysis of molecular fossils was performed with a Thermo Electron Trace GC Ultra connected via a Finnigan combustion interface-II to a Finnigan MAT 252 spectrometer at the MARUM, University of Bremen. Conditions of the gas chromatograph were identical to those described above. Carbon isotopes are given as δ values in per mil relative to the Vienna PeeDee belemnite (V-PDB) standard. Each measurement was calibrated using several pulses of CO₂ with known isotopic composition at the beginning and end of the run. Instrument precision was checked with a mixture of n-alkanes (C₁₅ to C₃₅) of known isotopic composition. Analytical standard deviation was < 1‰.

Fig. 2. The outcrop of the Upper Sinaia Formation flysch deposits on the Sărmăș Valley (Vărgăș section). (A) After almost 50 yr (from Herichh, 1878; Bărcă, 1958 first mentioned the occurrence of Peregrinella in the area) the outcrop was covered by trees and a thick layer of leaves, just a few small surfaces of the beds could be observed. (B) The section after cleaning, showing clear bedding of the turbidite sequences. (C) The succession shows successive sequences of sandstones, shales with silt intercalations and subordinate marlstones to limestones.
5. Results

5.1. Peregrinella from the Sinaia Formation

The specimens from both the carbonate and siliciclastic facies can be referred to the species Peregrinella (Peregrinella) multicarinata (Lamarck, 1819) [= peregrina von Buch]. The external morphology (Fig. 5) of the Romanian specimens is very similar to the material described by Toula (1911), Biernat (1957), and Thieuloy (1972). In external morphology, the large biconvex brachiopod has a subcircular outline and rectimarginate commissure. The maximum inflation of the profile occurs at about mid-length of the shell, the hinge line is long and the umbo is strong, elevated, and incurved delimited by two distinct, sharp beak ridges. Under the umbo a flat plan area almost 2.5 mm high is well developed from early growth stages. The dorsal valve's median septum is clearly visible from the exterior. Shell ornamentation is represented by 28–34 distinct radial costae with a sharp V-shape in transverse section; the number of costae on both valves is almost the same for any one specimen. Dimensions of adult specimens range from 30 to 68 mm in length, 29.8 to 68 mm in width, and 14 to 40 mm in thickness.

The internal structures of one specimen (Fig. 6) were investigated by acetate peels of transverse serial sections (cf. Sandy, 1989). The acetate peels recorded a well-developed median septum and hinge plates (Fig. 6A), taking on the appearance of a septalium. Further toward the anterior of the specimen the hinge plates detach, but the crural bases develop from bulbous processes that develop on the lateral ventral margins of the median septum (Fig. 6D,E). The typical mergiform crura could not be fully traced as they were broken. The size and shape of the median septum, hinge plates, crural bases, and crura are all consistent with the internal structures previously recorded for Peregrinella. The external morphology is consistent with Peregrinella multicarinata as outlined above. However, it is always desirable to have internal structures available whenever possible as the phenomenon of external homeomorphy is a notorious problem when dealing with brachiopods (e.g., Rudwick, 1970; Sandy, 2001).

5.2. Taphonomic observations

The samples from the carbonate facies (Zizin Valley, Belin Valley, Perişani–Baraolt area) show dense populations with numerous tightly packed individuals of Peregrinella. By comparison with other occurrences of Peregrinella, the available blocks of limestone in this study are considered to represent fragments of more extensive brachiopod deposits (cf. Biernat, 1957; Thieuloy, 1972; Posenato and Morsilli, 1999; Kiel and Peckmann, 2008). The majority of the specimens are complete articulated shells in different growth stages. In some cases juveniles (5–20 mm width) are clustered on the lateral and anterior margins of very large specimens (40–60 mm width), with their dorsal valves oriented more or less parallel with the adult-shell substrate and the ventral valve uppermost. In one specimen more than 15 juveniles are attached around the anterior margin of a 60 mm wide adult dorsal valve (Fig. 7A,C). The pedicle foramen is quite well developed in juveniles, and from its disposition it is obvious that the juveniles were attached to adult specimens. The hard substrate necessary for the attachment of the juveniles was also provided by the shell bed generated by the mass-occurrence of these brachiopods (similar to the situation described by Kiel and Peckmann, 2008, for Peregrinella in the Crimea). Predation is evident in some specimens from the limestones, which show depression-like punctures (Fig. 8A–C) and straight, deep cuts symmetrically disposed on both valves, situated in the lateral parts of the shell, where thickness of the shell is reduced (Fig. 8D–G). The punctures are represented by four small equidistant depressions, distributed symmetrically on both valves. The fact that the shell is not fractured or broken indicates that these are not the result of crushing or compaction of brachiopods against each other. In addition, the symmetrical nature of these markings strongly suggests a biological origin. There is evidence that the depressions result from sublethal attacks; costae are deflected but continue across the zone of shell deformation. The symmetrical cuts likely reflect damage at the commissure of the living animal and subsequent mantle repair of the shell margin (Fig. 8D–G). Similar shell injuries on Peregrinella multicarinata have been reported by Biernat (1957) and Kiel and Peckmann (2008).

The Peregrinella specimens from Vârghiş section are represented by shell fragments from a few millimeters in length to complete disarticulated valves, up to 10 cm or even larger (12 cm) in length (Fig. 9), concentrated toward the base of the beds. Therefore they are allochthonous. The fragments of shells belong to different ontogenetic
stages from juveniles to adults. Although the specimens were transported and the shells broken and disarticulated, very little deformation occurred and the original shell and external characters (coarse radial costae, concentric growth lines) are very well preserved. In a few specimens the median septum is also very well preserved.

5.3. Biostratigraphy of the Peregrinella-bearing part of the Upper Sinaia Formation

The micropaleontological samples collected from the shale levels just below the Peregrinella beds yielded a foraminiferal fauna composed mainly of agglutinated species. The following taxa have been identified (Fig. 10):

- Rhabdammina sp. 1
- R. sp. 2
- Rhizammina indivisa Brady,
- Ammodiscus tenuissimus (Gümbel),
- A. siliceus (Terquem),
- Gaudryna dividens Grabert,
- G. borimensis Kovatcheva,
- Dorothia praehauteriviana Dieni and Massari,
- D. subtrochus (Bartenstein), and
- Orbitolinopsis kiliani (Prever). The large-sized Rhabdammina sp. 1 contains many pyritized radiolarian shells agglutinated to their tests. Calcareous benthic foraminifers are rare, the most common species being represented by Neotrocholina infranigrulata Noth, associated with Fischerina sp., Lenticulina nodosa (Reuss), Dentalina cf. communis d’Orbigny, Marginulinopsis schloembachi (Reuss), M. djaffaensis (Sigal), M. sp., Astacolus incurvata (Reuss), Planularia crepidularis tricarinella (Reuss), Gavelinella sp., and Fondoctularia cf. verneuiliana angustimarginata Neagu. Other microfossil remains are represented by radiolarian shells, mostly calcified or pyritized, echinoid spines, micro gastropods, and rare juvenile brachiopods.

The scarcity of microfossils is one of the main characteristics of Lower Cretaceous flysch deposits in the Carpathian area. A similar foraminiferal assemblage has been recorded in the Upper Sinaia Beds from the Carpathian Flysch (Neagu, 1972, 1975) as well as from Lower Cretaceous deposits from the Dambovicioara Basin, Romania. The micropaleontological association from Vârghiș is therefore characteristic for the Late Hauterivian–Early Barremian time interval, indicating a relatively deep water environment, but above the carbonate compensation depth.

5.4. Sedimentology and petrography

5.4.1. Sedimentological characteristics of deposits with Peregrinella from Vârghiș

5.4.1.1. Depositional facies. Nine different facies have been recognized in the sequence with Peregrinella shells at the Vârghiș study site. Facies 1 (sandy gravel, massive, SyG m, Fig. 11) was identified only once in the lower part of the measured sequence. The average thickness of the unit is 50 cm. The average diameter of clasts is 2–3 cm and the largest clast is 6 cm, falling in the category of pebbles. Roundness of clasts varies from angular to sub-round. Sorting is generally poor, with the matrix being composed of sand and fine gravel. The structure is massive, having a matrix-supported character (Fig. 12A). Like the first facies, facies 2 (gravelly sandstone faintly horizontally stratified, GyS hs, Fig. 11) was only recorded once in the median part of the section. The average
thickness of the unit made up of this facies is 25 cm. The facies consists of coarse sand and fine gravel. Pebbles make up less than 20 vol.%. The largest, very angular clast is 5 cm in diameter. The long axes of the pebbles are oriented parallel to stratification (Fig. 12B). In the lower part of the unit made up by this facies, muddy clasts were found. This facies contains bioclasts of Peregrinella. The coarse facies 1 and 2 (SyG m and GyS hs) suggest deposition from hyperconcentrated density flows (sensu Mulder and Alexander, 2001), similarly to cohesionless debris flows (sensu Nemec, 1990). Facies 3 (sandy massive/normally graded, Sm / n g , Fig. 11) was recorded several times. The thickness of the respective units varies from 10 to 20 cm. The internal structure of beds made of this facies is massive or normally graded, with grain size varying from very coarse sand or even fine gravel to medium sand. Sometimes muddy clasts were encountered in the lower part of the units made of very coarse sand or even fine gravel to medium sand. Sometimes muddy clasts were encountered in the lower part of the units made of this facies. The tops of beds grade into sands with parallel lamination (Fig. 12C). This facies was deposited from concentrated density flows (sensu Mulder and Alexander, 2001), similar to high density turbidity currents (sensu Middleton and Hampton, 1973; Lowe, 1982) or sandy debris flows (sensu Shanmugam, 1996). It contains bioclasts of Peregrinella. Facies 4 (sand horizontal laminated/normally graded, S hl/ng, Fig. 11) forms many layers of an average thickness of 10–15 cm. It consists of medium-grained sand. The internal structure of beds reveals horizontal lamination and normal grading. The upper limit of units made of this facies often grades into sand with cross lamination (Fig. 12C). Facies 5 (sand convolute laminated, S cvl) was encountered only once immediately below gravelly sandstone. It consists of medium- and fine-grained sand. The unit made of this facies is 10 cm thick and reveals convolute lamination (Fig. 12B), which represents an early post-depositional deformation effect caused by overloading in a sub-consolidated stage. The primary structure from which convolute lamination resulted was parallel or cross
lamination. Facies 7 (sandy silt, horizontal laminated, SyT hl) was identified above the sand cross lamination facies (S clr). The thickness of units consisting of this facies with parallel horizontal lamination (Fig. 12C) varies from 3 to 10 cm. The facies consists of fine sand laminae in the lower part, while silt laminae dominate above. The medium to fine sandy (S hl/ng, S clr) and sandy silt facies (SyT hl) were deposited from low density turbidite currents. Facies 8 (mud, horizontal laminated, M hl) makes up much of the studied sequence. Units consisting of this facies with parallel lamination (Fig. 12C) have variable thickness, ranging from 2 to 20 cm. Finally, facies 9 (limestone and marls, massive, LM m) typically overlies facies 8. The respective units are represented by whitish beds or lenses with a thickness from 2 to 20 cm with a massive internal structure (Fig. 12D). Some of these beds and lenses reveal burrows that are filled with coarser sediment. These limestones and marls represent detrital sediments and do not resemble the limestone with *Peregrinella* accumulations. Muddy levels resulted from

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**Fig. 7.** (A)–(D) *Peregrinella multicarinata* (Lamarck) specimens from carbonates, Zizin Valley. (A)–(C) Specimen LPBIII 374, (A) ventral view, (B) dorsal view, (C) anterior view. (D) Specimen LPBIII 366 with geopetal structures inside of the shell, dorsal view. (E) LPBIII 344, Zizin Valley, limestone with *Peregrinella multicarinata* (Lamarck). (F) IGR 3908, Belin Valley, limestone with *Peregrinella multicarinata* (Lamarck).

**Fig. 8.** Predation marks on *Peregrinella multicarinata* (Lamarck). (A)–(F) Adult specimen LPBIII 376, Cădărești–Ciughiu Valley. (G) Juvenile specimen IGR 3908, Belin Valley; (A)–(C) four rectilinear punctures, symmetrically disposed on both valves; (D)–(G) specimens with straight, deep cuts symmetrically disposed on both valves.
suspension settling from the tail of low density turbidity currents, similar to the Te Bouma division, while limestones and marls (LM m) are interpreted to reflect background sedimentation, representing hemipelagites and pelagites.

5.4.1.2. Petrography of deposits from Vârghiş area. Petrographic analysis was carried out for a diagnosis of the facies to determine the possible origin of the clasts, and to make observations on the distribution of Peregrinella bioclasts (see Fig. 4 for sampling sites). The facies 6 to 8 (S cvl, SyT hl, M hl) were not analyzed in detail, because they do not contain Peregrinella. The lithoclasts of the gravelly facies 1 (SyG m) are of metamorphic and sedimentary origin. The size of the overall largest sedimentary clasts of 5–6 cm suggests an intrabasinal origin. The sandy facies 3 to 5 (S m/ng, S hl/ng, S clr) and the matrix of the facies 2 (GyS hs) facies are lithic and sublithic sandstones according to Dott (1964). The sandstones are fine to coarse-grained arenites with poor to moderate sorting. The roundness of grains is subangular-subrounded, but rounded and angular subpopulations have also been identified, suggesting different degrees of reworking and recycling. The ratio between particles and carbonate cement and matrix is 60–70% to 30–40%. The composition of the particles relative to an average of 70% is 25±2.5% polycrystalline quartz (Qp) (Fig. 13A) and subordinate monocrystalline quartz (Qm), 4±0.4% calc-sodic and potassium feldspars, and 10±1% micas, and 3±0.3% garnet (Fig. 13B). Lithoclasts (23±2.3%) are represented by sedimentary rock fragments (siltstone, shales, allochemical and orthochemical limestones of intrabasinal and extrabasinal origin; Fig. 13D), and by metamorphic rock fragments (i.e., gneiss; Fig. 13C). Bioclasts (5±0.5%) are represented by brachiopod shell fragments, bivalves, echinoid plates, and wood fragments. The largest bioclasts (up to 10 cm) with well-preserved shell structure are Peregrinella shell fragments (Fig. 13E; facies S m/ng).

The carbonate background facies (LM m) contains CaCO₃ in different proportions (59.0–85.7% determined by LOI 950 °C method; and 56.8–88.6% determined by XRF method), corresponding to argillaceous limestones and subordinate marls (sensu Scolari et al., 1973) or muddy micrites (sensu Mount, 1985). Micrite underwent partial recrystallization to microsparite. Carbonates contain 1.0–2.3% organic matter (LOI at 550 °C). The bioclasts, sponge spicules, and foraminifera from these facies do not exceed 10%. Calcite veins are present. Burrows (Fig. 12D) are passively filled with coarser siliciclastic material (Fig. 13F).

5.4.2. Petrography of limestones with Peregrinella. The few available limestone blocks with Peregrinella from the Zizin and Belin localities are packed with mostly intact, articulaled brachiopod shells (Figs. 7E,F, 14A,B). Samples from the Zizin locality represent pure, authigenic limestone; whereas, samples from the Belin locality are either micritic limestone with abundant detritus or represent clasts of limestone enclosed in a detrital matrix (Fig. 14A,B). The detrital components are quartz grains, Peregrinella shell fragments, bivalve and gastropod shell debris, woody debris, and radiolarians. Articulated shells are either empty, or partially or completely filled with geopetal sediment or fibrous, banded and botryoidal cement (Figs. 7D, 14A,B).

The matrix of the Zizin limestone consists of micrite (Fig. 15A), which is either homogenous or reveals a clotted microfabric (Fig. 15...
B). Clotted micrite is particularly common in limestone from the Belin locality (Fig. 16A). The early diagenetic, fibrous, banded and botryoidal cement preferentially formed on the inside of articulated brachiopod shells (Figs. 14A, B, 15C, 16B). It appears to be much less abundant in the Zizin than in the Belin limestones, although it needs to be stressed that this comparison is based on a limited number of samples. In one of the Belin samples, banded and botryoidal cement encloses framboidal pyrite. Banded and botryoidal cement is postdated by late diagenetic equant calcite spar (Fig. 15C). Equant calcite spar fills most of the former porosity that was left after formation of the early diagenetic carbonate phases (Figs. 15A, 16A). Aggregations of fecal pellets of uncertain origin have been recognized only in the Zizin limestone (Fig. 15D).

5.5. Stable carbon and oxygen isotopes

Due to the small number of limestone samples, only a few individual carbonate phases could be sampled for stable isotope analysis. Despite this limitation, the obtained isotope patterns appear to be diagnostic for the Zizin and Belin limestones (Fig. 17). A *Peregrinella* shell from the Belin locality revealed a δ¹³C value of −0.3‰ and a δ¹⁸O value of −0.2‰. Micrite of the Zizin limestones is less ¹³C-depleted (−29.7‰ to −20.4‰; n = 4), corresponding δ¹⁸O values range from −6.0‰ to −3.0‰. Micrite of the Belin limestone is less ¹³C-depleted (−8.0‰ to −3.7‰; n = 4), but yielded similar δ¹⁸O values (−5.0‰ to −4.1‰). The Zizin limestones did not contain enough banded and botryoidal cement to allow for
5.6. Biomarkers

The hydrocarbon fraction of the studied sample from the Zizin locality is predominantly composed of n-alkanes ranging from n-C16 to n-C35. The n-alkanes do not show a preferential distribution of odd or even chains and maximize at n-C27. Overall, mid-chain n-alkanes predominate (Fig. 18). Terminal-branched alkanes with 17 to 23 carbon atoms were found in minor amounts. Apart from straight-chain and single-branched alkanes, multiple-branched alkanes (isoprenoids) are abundant. The first group of isoprenoids comprises head-to-tail linked compounds with 18 to 25 carbon atoms, peaking at 2,6,10,14-tetramethylhexacosane (phytane). Other than head-to-tail linked isoprenoids, two tail-to-tail linked isoprenoids were found: 2,6,10,15,19-pentamethylicosane (PMI), which most likely co-eluted with the head-to-tail linked 2,6,10,14,18-pentamethyllicosane peak revealed a δ13C value of −57‰, biphytane and C30-biphytane values of −42‰ and −92‰, respectively. The hopane (−60‰) and gammacerane (−90‰) are also 13C-depleted.

6. Discussion

6.1. Origin of the section at Vârghiș with Peregrinella

Many of the beds of the Vârghiș locality are similar to beds with a classic Bouma sequence (cf. Bouma, 1962), suggesting deposition from turbidity currents. In most cases, Bouma sequences are incomplete, lacking either the base, because flows contained no coarse material, or the top, because of erosion. Besides beds with incomplete Bouma sequences, beds with very different grain-size were found, represented by facies 1 and 2 (SyG m and GyS hs), suggesting hyperconcentrated density flows. Taking into account the results of micropaleontological analysis and the presence of turbidities, the section at Vârghiș is considered to represent a deep-sea environment (Fig. 19). Coarse facies probably represent channel lag deposits, while Bouma sequences suggest turbiditic lobe environments (cf. Walker, 1992). Thus, the sequence at Vârghiș represents a proximal turbiditic lobe, more accurately, the channel-lobe transition.

The bioclasts of *Peregrinella* were found within the channel lag facies (GyS hs) and at the base of Bouma sequences (S m/ng), interpreted as hyperconcentrated density flows and high-density turbidite currents, respectively. Large fragments of shells, represented by half or complete dorsal or ventral valves are disarticulated but otherwise very well preserved (with original shell and external and even internal characters).
If one considers the strong interlocking hinge of *Peregrinella*, it seems feasible that disarticulation of the shells without significant damage was possible during the movement of the shells within dense gravity flows. In the hyperconcentrated density flows (cohesionless debris flows; GyS hs), the dominant particle support mechanisms are matrix strength and dispersive pressure (grain-to-grain interaction). Besides these, especially in the case of high-density turbidite currents (concentrated density flows; S m/ng), fluid turbulence is involved (Middleton and Hampton, 1973; Mulder and Alexander, 2001). According to this scenario, the large *Peregrinella* shells were probably disarticulated by dispersive pressure shear stress and turbulence. Most probably, the *Peregrinella* shells were dislocated by hyperconcentrated and concentrated density flows from their original habitat situated on the slope and transported over a relatively short distance downslope.

Fig. 13. Petrography of deposits from Vârgiș. (A) Mono- and polycrystalline-quartz and metasomatic carbonate cement. (B) Lithic sandstone with heavy minerals (garnets) and metasomatic carbonate cement. (C) Metamorphic rock lithoclast, fragment of gneiss. (D) Sedimentary rock lithoclast, bioclastic limestone with intraclasts; (E) Lithic sandstone with carbonate clasts and *Peregrinella* shell bioclasts. (F) Detail of the muddy facies, showing micrite with burrows passively infilled with silts and fine arenites.

Fig. 14. (A), (B) Scanned polished slabs of carbonate rock sample (IGR 3908) with *Peregrinella multicarinata* (Lamarck), Belin Valley. Void spaces within the articulated brachiopod shells were partially cemented by a rim of acicular carbonate minerals and sparry calcite; the micritic matrix contains abundant detrital components.
6.2. Origin of the limestones with Peregrinella

The limestones from the Zizin and Belin localities reveal petrographical characteristics such as clotted micrite, fibrous, banded and botryoidal cement (Figs. 14, 15, 16), as well as framboidal pyrite that are typical of methane-seep carbonates (cf. Beauchamp and Savard, 1992; Campbell et al., 2002; Peckmann and Thiel, 2004). Such an interpretation is supported by the low $\delta^{13}$C values of early diagenetic carbonate phases and the biomarker inventory of the Zizin limestone. Whereas stable carbon isotopes of the carbonate phases can only reveal that carbonate precipitation resulted from the oxidation of methane, biomarkers document the process in which methane was oxidized. Like at other methane seeps, carbonate formation was favored by anaerobic oxidation of methane (AOM) performed by a consortium of archaea and sulfate-reducing bacteria. Molecular fossils of methanotrophic archaea are biphytanes with $\delta^{13}$C values of approximately $-90$‰ (cf. Peckmann and Thiel, 2004; Peckmann et al., 2009). Another biomarker of methanotrophic archaea, PMI, revealed a surprisingly high $\delta^{13}$C value of $-61$‰. The circumstance that PMI is much less depleted than the biphytanes is probably best explained by the co-elution of the AOM-biomarker tail-to-tail linked PMI with head-to-tail linked PMI (cf. Greenwood and Summons, 2003), and an admixture of PMI deriving from methanogenic archaea can consequently not be excluded (cf. Schouten et al., 1997). Based on its low $\delta^{13}$C value of $-86$‰, phytane apparently derived predominantly from lipids of methanotrophic archaea and not from phototrophic organisms. The relatively high content of biphytanes and the absence of crocetane may point to a low, diffusive flux of methane and the dominance of archaea of the so-called ANME-1 group (cf. Peckmann et al., 2009). Such a scenario agrees with the only moderate abundance of early diagenetic banded and botryoidal cement in the Zizin limestone compared to its greater abundance in the Belin limestone. However, based on the limited number of rock samples and only one biomarker sample, such reasoning is necessarily speculative.

Putative biomarkers for sulfate-reducing bacteria involved in AOM are terminally-branched alkanes, which have been suggested to derive from lipids synthesized by sulfate-reducing bacteria (Peckmann and Thiel, 2004; Birgel et al., 2008b). The circumstance that short-chain $n$-alkanes show the most negative $\delta^{13}$C values among $n$-alkanes agrees with a partial source of short-chain $n$-alkanes from the lipids of sulfate-reducing bacteria. However, it is difficult to
exclude that these compounds derived from oil degradation. Overall, the n-alkane pattern resembles a typical oil signature, although no evidence of accumulations of metamorphosed oil (i.e., pyrobitumen) was recognized. The lack of an odd-over-even distribution of the long-chain n-alkanes indicates high thermal maturity, which agrees with the presence of isoprenoid and terminally-branched alkane pseudohomologues that have been reported from other thermally-mature seep limestones (Birgel et al., 2006; Birgel et al., 2008b).

Based on its low $\delta^{13}C$ value of $-60‰$, the C30-hopane apparently derived from bacteria that lived at the ancient seep site. However, it is difficult to determine to which group of bacteria the source organisms belonged. The hopane could be a degradation product of C30-hopanoids, such as diplopterol, since no other hopanoids were detected. Potential source organisms are sulfate-reducing bacteria and aerobic methanotrophs. The latter source agrees with the presence of nor-lanostane, which is a biomarker of aerobic methanotrophs found in other seep deposits (Birgel and Peckmann, 2008). Gammacerane and its precursor tetrahymanol are often found at seeps (Werne et al., 2002; Birgel et al., 2006). Tetrahymanol is believed to derive from ciliates at seeps (Werne et al., 2002), which predominantly live at the oxic-anoxic interface. It is interesting that Werne et al. (2002) reported that maximum contents of diplopterol and tetrahymanol coincide in vertical profiles in sediments imprinted by seepage of methane.

The $\delta^{13}C_{\text{carbonate}}$ values as low as $-30‰$ of micrite of the Zizin limestone agree with the interpretation based on biomarkers that the carbonate resulted from the oxidation of methane. The Belin limestone $\delta^{13}C$ values of micrite as low as $-8‰$ and of banded and botryoidal cement as low as $-10‰$ are more difficult to interpret. As the sample amount was insufficient for biomarker analysis, the carbon source of this limestone cannot be determined with certainty. Because $\delta^{13}C$ values of authigenic carbonate phases forming at seeps reflect mixing of different carbon sources — mostly methane-derived carbon and marine carbonate but also carbon from a carbonate pool affected by methanogenesis — even relatively high values like those observed for the Belin limestone could still represent methane seepage (cf. Peckmann and Thiel, 2004). Alternatively, such values are typical for authigenic carbonate minerals forming at oil seeps (cf. Peckmann et al., 2007). But, with the lack of other evidence for oil seepage, like for example the presence of pyrobitumen, this possibility is difficult to evaluate. It also cannot be excluded that the relatively high values of early diagenetic phases of the Belin limestone reflect

Fig. 17. Cross plot for isotopic analyses.

Fig. 18. Gas chromatogram (total ion current) of the hydrocarbon fraction of the Zizin valley Peregrinella limestone. Compound-specific $\delta^{13}C$ values are indicated in parentheses. Circles: n-alkanes; white triangles: head-to-tail linked isoprenoids; black triangles: tail-to-tail linked isoprenoids; gray triangles: biphytanes; black quadrangles: iso-alkanes; white quadrangles: anteiso-alkanes; Pr: pristane; Ph: phytane; PMI: 2,6,10,15,19-pentamethylicosane; Sq: squalane; istd: internal standard; bp: biphytane.
oxidation of more pristine organic matter, but given the high amount of organic matter required and the similarity of the Belin limestone with the Zizin limestone and other seep carbonates containing *Peregrinella* (Campbell and Bottjer, 1995a; Kiel and Peckmann, 2008), this scenario seems unlikely. Consequently, carbonate microfabrics as well as isotope patterns of authigenic carbonate phases and molecular fossils agree with the formation of the studied limestones at methane seeps. It cannot be verified if the seepage fluids contained crude oil in addition to methane, but the paleoenvironment of the Romanian *Peregrinella* can be unequivocally identified as one influenced by seepage.

6.3. Scenario of *Peregrinella* occurrences in the Sinaia Formation of the Eastern Carpathian Mountains

*Peregrinella* has a broad distribution, with records in Europe (Poland, France, Italy, Crimea of Ukraine, and Romania), Asia (Tibet), and North America (California and Alaska). When considering the distribution of *Peregrinella multicarinata*, in addition to the Romanian material discussed herein, the species is also recorded from France (e.g., Thieuloy, 1972), Poland (within olistolith blocks, Biernat, 1957), and Crimea (Smirnova, 1972; Kiel and Peckmann, 2008). This would indicate that there may have been paleobiogeographic and tectonic connections between these localities in the Early Cretaceous. In terms of relating the Carpathian occurrences of *Peregrinella* from the Ceahlău Nappe to others from Europe, there were presumably marine connections with the Ceahlău Nappe developing from part of the rifting interior of the European continental margin. In the Ceahlău Nappe, the deepest part of the External Dacides Basin containing flysch is preserved (the Sinaia Formation). The shallowest parts and upper slopes of the basin were eroded or are covered to the west by other, more internal geotectonic units (Sândulescu, 1984). This points to the possibility that it may not be possible to find methane-seep limestones with *Peregrinella* in place in the External Dacides Basin.

The dimerelloid brachiopod *Peregrinella* has been considered to represent part of a seep-restricted brachiopod genus and superfamily (Campbell and Bottjer, 1995a,b; Gischler et al., 2003; Kiel and Peckmann, 2008; Sandy, 2010), and an obligate (endemic) taxon of Early Cretaceous deep-sea communities (Kiel, 2010). The Romanian *Peregrinella* studied herein are considered to have lived at methane seeps (as demonstrated for Zizin and Belin localities) on the slopes of the Upper Sinaia flysch basin. Three of the four known localities with *Peregrinella* (Zizin, Belin, and Cadărești, Fig. 1) of a likely methane-seep origin are within a 100 km radius of each other but it has not been confirmed that they represent in situ seep deposits. The fourth occurrence of *Peregrinella*, from Vârghiș, is in a proximal turbidite fan, near to the base of the basin slope. Vârghiș is in a somewhat geographically intermediate position between the other 3 localities (Fig. 1). The methane-seep limestones are probably parts of larger carbonate deposits that developed due to methane seepage on the basin slope. Hardgrounds developed at these methane seeps, and thriving populations of *Peregrinella* attached to and colonized these surfaces (cf. Campbell and Bottjer, 1995a; Sandy, 1995; Kiel and Peckmann, 2008), flourishing with a plentiful local food source (Campbell, 2006).

The fact that the original locations of the methane-derived carbonates with *Peregrinella* have not been found in the field during this study could be explained by the rather small dimensions of such carbonate bodies (not more than tens of meters across by analogy with similar occurrences, e.g., Sandy, 2010), within the voluminous siliclastic sequences of the Upper Sinaia Formation, like proverbial “needles in a haystack” (cf. Campbell, 2006). Zberea (1962, p. 284) mentioned that he collected samples with *Peregrinella* from the Belin Valley from “small limestone blocks packed full with brachiopod shells, which outcrop within the breccias from the Upper Sinaia Formation”. It seems likely that such small limestone blocks have been transported downslope in sediment gravity flows (cf. Campbell and Bottjer, 1995a for limestones rafted to the basin floor at Wilbur Springs, California), but to clarify this, future detailed research in the Belin Valley is necessary. Alternatively, as discussed above, the original seep deposits may have been destroyed by erosion or tectonics. This interpretation is based on field observations at the localities from where these limestone blocks have been reported, as the Sinaia

![Fig. 19. Sedimentary succession of the Upper Sinaia Formation in the Vârghiş area, Sârman Valley; Scale: 1:50. Facies associations; incomplete Bouma turbiditic sequences intercalated with channels and limestones in a deep-sea environment.](image-url)
limestone blocks could have been transported down the basin slope from higher up the basin slope. Even if this were the case, the limestones are considered intrabasinal and not extrabasinal.

(6) This study establishes that Peregrinella from the Sinaia Formation lived in a chemosynthesis-based environment. This is an association that has also been observed for Peregrinella in California (Campbell et al., 1993, 2002) and the Crimea (Kiel and Peckmann, 2008). Whether the genus is wholly restricted to chemosynthesis-based environments (all methane seeps to date) remains to be elucidated, but seems probable as the genus has not yet been recorded from shelf faunas.

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