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The Lokve Barite Deposit, Croatia: an Example of Early Diagenetic Sedimentary Ore Deposits

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Key words: Barite, Early diagenesis, Sulphur isotopes, Trace elements, Sabkha, Dinarides

Ključne riječi: barit, rana dijageneza, izotopi sumpora, elementi u tragovima, sabka, Dinaridi

Abstract

Syngenesis versus epigenesis in the Kupferschiefer genetic model is still a matter of controversy. The title of the article seems to be a paraphrase of a well-known paper by WEDEPOHL (1971), "Kupferschiefer as a prototype of syngenetic sedimentary ore deposits". It is intended, however, more as a paradigm for the genesis of the hydrogen-sulphide geochemical barrier, an important ore formation episode prior to heavy metal accumulation, which shows the utility of RENFRO's (1974) epigenetic model.

Barite mineralization in Lokve is a stratabound ore deposit conformably situated at the Permian-Triassic boundary. It bears only two ore minerals, barite and pyrite exclusively, separated into two distinct, juxtaposed horizons stretching for tens of kilometers.

The discovery of cryptalgal fabrics and other conspicuous sedimentary features in underlying siliciclastics with massive pyrite and surmounting barite-bearing dolomites supports their affiliation to a tidal flat facies and sabkha environment. Barite and pyrite accumulation were formed by an early diagenetic, bacteriogenic sulphate reduction in a peritidal muddy environment, concommitant to a widespread process of evaporative dolomitization. The early diagenetic model is supported by an analysis of the sedimentary facies, trace element geochemistry and sulfur isotope distribution along two vertical profiles across the stratabound barite and pyrite mineralization.

Sažetak

Singeneza ili epigeneza u genetskom modelu stvaranja bakarnih škriljavaca (Kupferschiefer) još je uvijek predmet znanstvenih diskusija. Iako se naslov članka čini parafrazom poznatog rada K. H. WEDEPOHL-a (1971) "Kupferschiefer as a prototype of syngenetic sedimentary ore deposits", trebalo bi ga prije shvatiti kao paradigmu o genezi sumporovodikove geokemijske barijere, važne rudotvorne epizod, koja prethodi akumulaciji teških metala, preferirajući na taj način epigenetski model RENFRO-a (1974).

Baritna mineralizacija u Lokvama je slojno rudno ležište, konkordantno smješteno na granici perma i trijasa. Mineralizacija se sastoji isključivo od dva rudna minerala, barita i pirita, a pruža se desetak kilometara u dva razdvojena horizonta. Otkriće stromatolita i različitih sedimentnih tekstura u podinskom siliciklastičnom sedimentu s masivnim piritom ili s euhedralnim piritnim zrnima, te u krovinskim baritonosnim dolomitima, govori u prilog facijesa plimskih zaravni. Baritne i piritne akumulacije su ranodijagenetska tvorevina, a njihova geneza se objašnjava bakterijskom redukcijom sulfata u blatu zone plime i osjeke, uz aktivnu evaporitnu dolomitizaciju.

1. GEOLOGY, DEPOSITIONAL ENVIRONMENT, MINERALIZATION

The Lokve barite deposits (Fig. 1) are situated along the skirts of the Upper Permian terrane, adjacent to the contact of the Lower Triassic dolomites. The largest deposits are not far from the villages of Homer and Mrzle Vodice.

The barite ore is interstratified in the Lower Triassic dolomites, always lying directly over the Permian clastics impregnated by pyrite cement. The ore bodies are equilateral layers, ranging in size from several meters to 300 m, and in thickness from 1 to 5 m. The richest ore grade yields 70 to 90% barite, but on the average it varies between 20 and 70%. Barite has been mined for decades and pyrite left behind in large masses as a gangue mineral.

Fig. 1 Location of the Lokve barite deposits.

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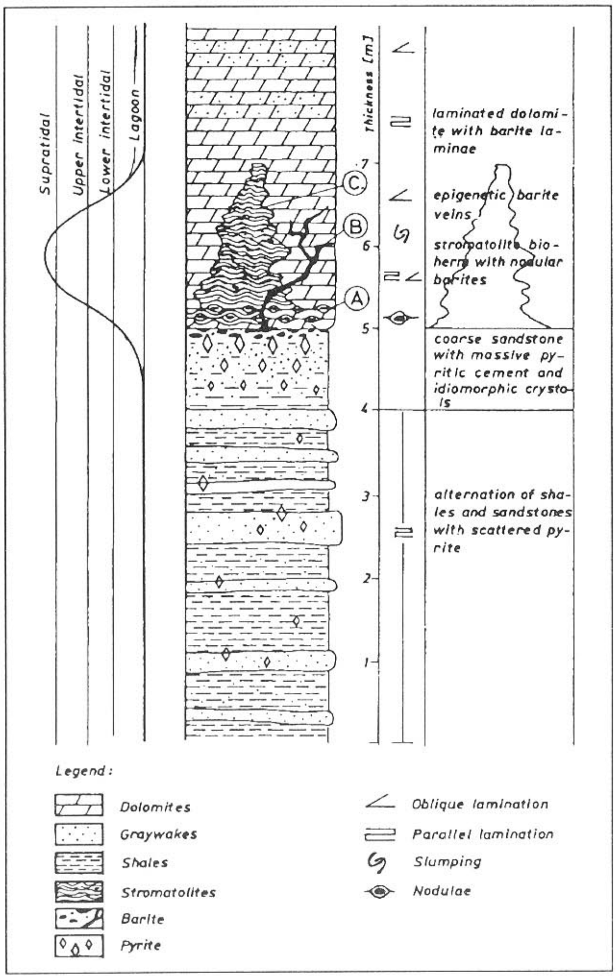


Fig. 2 Schematic stratigraphic column at the Homer locality, near the village of Lokve (after PALINKAŠ & SREMAC, 1989).

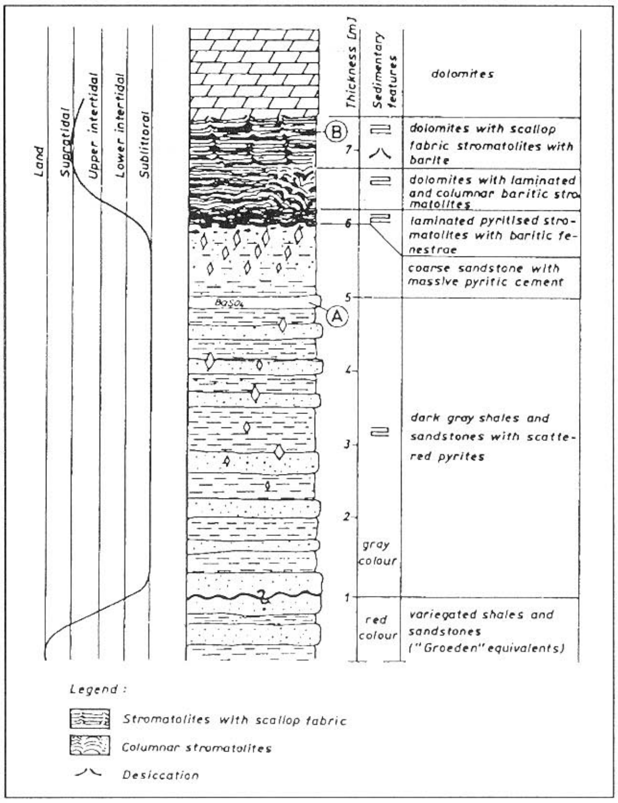


Fig. 3 Schematic stratigraphic column at the Mrzle Vodice locality (after PALINKAŠ & SREMAC, 1989).

JURKOVIĆ (1959, 1962) considered the ore deposits to be of submarine exhalative origin, while minor occurrences in the upper parts of the basal dolomite as a metasomatic product of secondary-hydrothermal remobilization. ŠUŠNJARA & ŠINKOVEC (1973) were inclined to a synsedimentary ore-forming process in shallow, partly or completely land-locked basins during regressive conditions.

Middle and Upper Permian sedimentation near Lokve village, in the Gorski Kotar area is characterized by deposition of siliciclastic material, originating from newly exposed dry land areas, uplifted during an early continental rifting phase in the Dinarides. The progressive denudation created expressive relief and sedimentary spaces were filled up by persistently maturing clastics. The final member of the Upper Permian clastic rocks is represented by green-gray and gray-violet fine-grained siltstones and pelites (BABIĆ, 1968; ŠĆAVNIČAR & ŠUŠNJARA, 1967; ŠĆAVNIČAR, 1973).

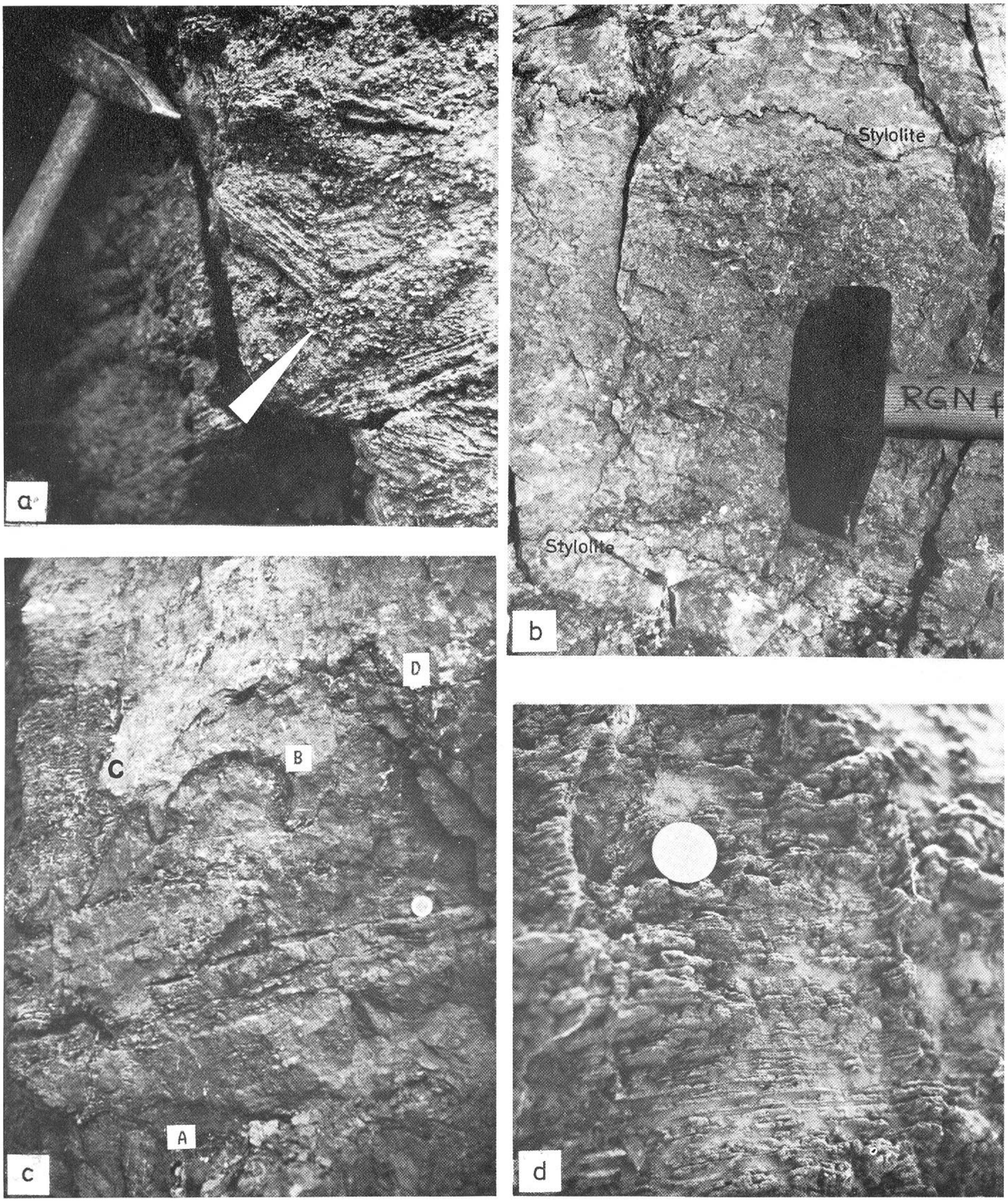
Extensive low-lying coastal areas became a site of immense evaporite deposition in lagoons and coastal and inland sabkhas. Smoothing of the relief stopped clastic deposition, which along with a concurrent change in climate from humid-arid to moderate-warm, triggered carbonate sedimentation and formation of the basal dolomites (Figs. 2, 3).

Dolomitization, as penecontemporaneous alteration of aragonitic mud sediments by hypersaline ground water, is a preponderant process in peritidal environment. All the Lower Triassic carbonate lithotypes with stratabound barite mineralization in the Gorski Kotar adjacent to confomable contact with the Permian clastics, originated in a sedimentary environment near the mean sea level.

The basal dolomites bear typical peritidal sedimentary features like oolitization, cross-, oblique-, ribbon- and parallel-lamination, ripplemarks, intraformational collapsing, desiccation cracks, the presence of bioher-mas and biostromes, etc. Early diagenetic barite pervaded all sedimentary structures in the dolomite, making them distinctly observable due to its higher weathering

Fig. 4. **a** Early diagenetic dolomitization of lime sediments, caused by evaporative transpiration in tidal flats, induced partial stiffening of newly formed dolomite laminae, and sedimentary features were afterwards baritized. The heavy load of the overlying carbonate rocks on the soft, water-rich mud layer deformed primary ore structures and brought about intraformational collapsing and cracking of the laminae (original sediment was probably of tidal channel origin).

b Stylolites as a late diagenetic feature, passing through patches of dolomite and barite in mottled ores (upper part of the basal dolomites), represent a clear sign of the early diagenetic origin of mineralization.



c The Školski brijeg locality, different stromatolitic morphotypes (diameter of coin - 2.5 cm):

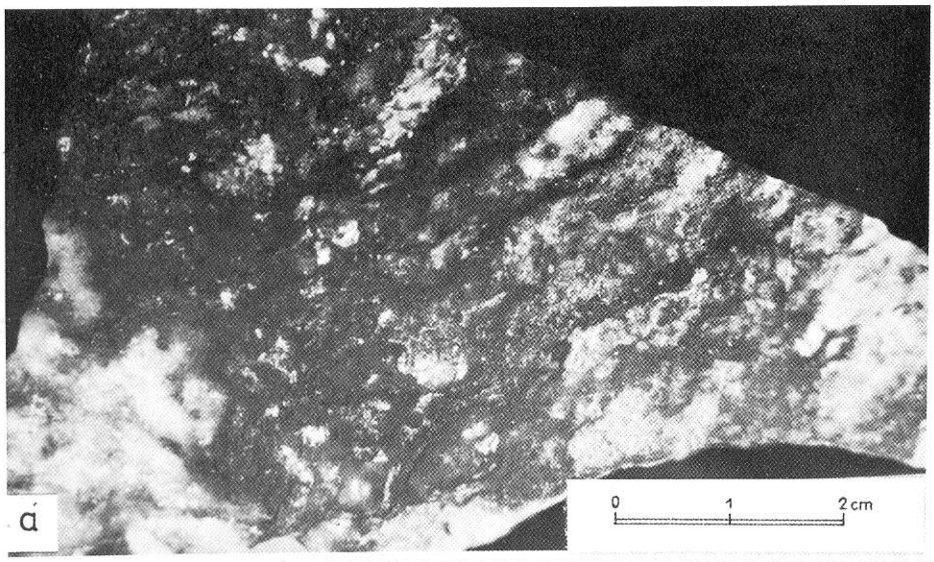
A - Low relief biostromal formation with pyrite crust.

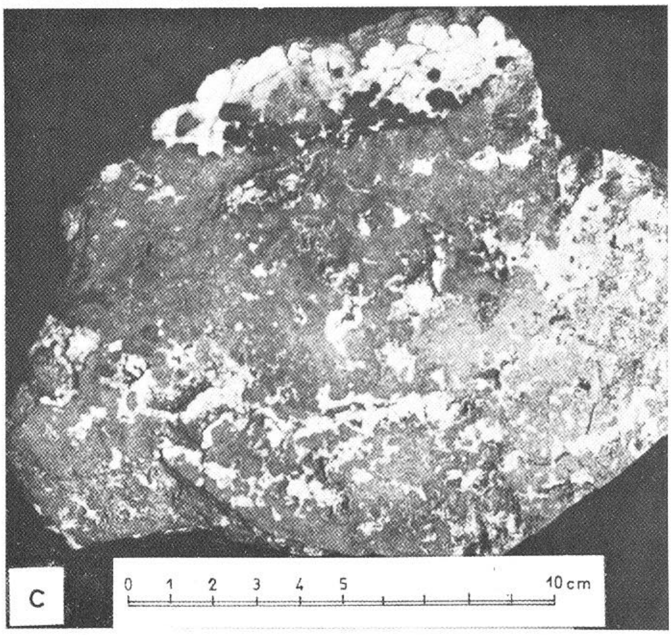
B - Discrete columnar stromatolites with prevailing conical and domal forms. The outer crust is made of approx. 5 cm thick pyrite crown at the top of the column.

C - Club-shaped columnar stromatolite with missing pyrite crest and crudely developed baritic, fenestral fabric. Internal structure minutely baritized.

D - Ridge-and-rill stromatolites formed in lowered water energy.

d The Mrzle Vodice locality, flat-laminated barite stromatolites with scallop fabric due to desiccation process in the upper intertidal, (diameter of coin 2.5 cm).





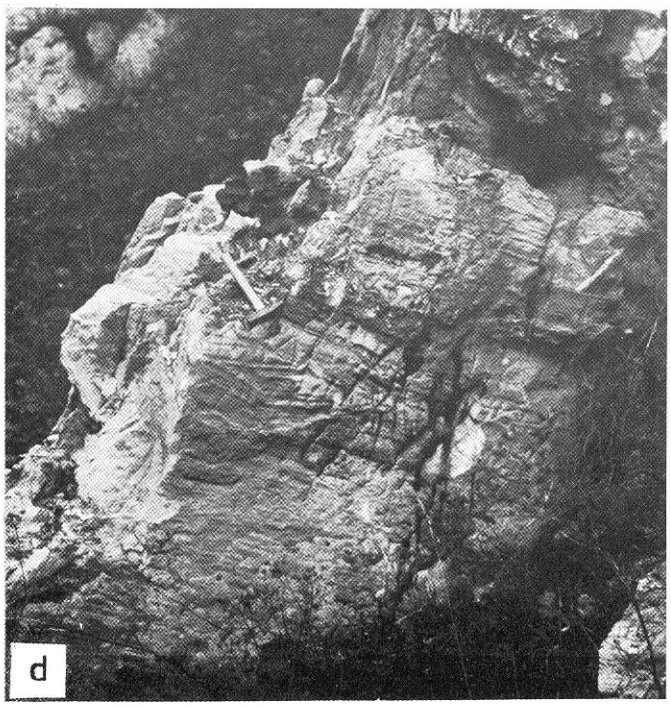


Fig. 5 **a** Microcellular and nodular stuctures (chicken-wire) typical for evaporites, in this case barite (white and pinkish masses in dolomitic laminae), growing in tidal flat sediments. Homer locality.
b Barite biostromes in the basal dolomites.
c Massive pyrite and barite ore with microcellular structure, at the clastic-dolomite contact, usually weathered into manganiferous gossan, Mrzle Vodice locality.
d High energy water sedimentary structures (cross-bedding) in the basal early diagenetic dolomites, cemented by barite, easily observable due to weathering resistance of barite in comparison to the dolomites.

resistance in comparison to the dolomite (Figs. 4a, 4c, 4d, 5b, 5d) (PALINKAŠ & ŠINKOVEC, 1986).

The schematic column at the locality Homer represents a characteristic stratigraphic sequence at Lokve village (Fig. 2). Underlying rocks are Permian clastics, subparallel alteration of shales and sandstones on a centimeter-scale. The rocks are dark, almost black, often with macroflora remnants. The coarser members are impregnated by pyritic cement which amounts to a few per cent, but increases significantly by approaching the clastics-carbonates boundary (a few tens, and at places almost one hundred per cent). In the uppermost part of the clastics, the first scattered masses of barite appear. The boundary is sharp and topped by barite-bearing dolomites.

The stromatolitic bioherms distinctly emerge as prominent masses surrounded by cross-laminated dolomites, with numerous high energy water structures (PALINKAŠ & SREMAC, 1989). At the lowermost part of the basal dolomites, a nodular, pyrite-barite ore occurs, with characteristic chicken-wire structure (Fig. 5a). Stromatolitic fabric in the dolomite is clearly demarcated by chemically resistant barite, but thin section and acetate peel examination revealed conspicous lamination with acicular barite as well. Crudely developed barite lamination and coarse fenestrae suggest an intertidal stromatolitic fabric formed from a pustular mat in the fairly agitated water of an unprotected environment.

Discrete stromatolitic morphotypes pervaded by barite have been observed at Školski brijeg (Fig. 4c). The stromatolitic series starts with low relief species having almost biostromal characteristics (Fig. 4c, point A). Coarse fenestrae, cavities, crude lamination and low relief indicate subtidal, but lower energy water. The conditions gradually changed into a more agitated water environment, causing the formation of columnar SH-stromatolites of different shapes. Conical and domal forms prevail (Fig. 4c, point B). The outer surface of the stromatolites is covered by a pyritic crust which is approximately 5 cm thick at the crest of the columns. It was a former soft, colloform mat, ideal food for desulphurizing anaerobic bacteria, digested during later anoxic conditions, likely after burial. The burial must have been fast, perhaps even torrential, since algal growth was stopped instantaneously, and organic matter was preserved from disintegration in an otherwise oxidizing water condition, (DILL et al., 1986).

On the left side of Fig. 4c, point C, one may observe a 0.5-meter-tall, clubshaped column with coarse, irregular fenestrae, formed from pustular mat. The pyrite crown is missing, since the mucilage, or organic layer, was destroyed during emergence and subaerial exposure, a common case of the disintegration process in living stromatolites in the upper intertidal regime.

The uppermost pyritic, irregular layer (Fig. 4c, point D) is a cross-section through a ridge-and-rill pseudocolumnar stromatolite shaped by wave action and tidal scouring in sublittoral, but again lower energy water.

The situation at the village of Mrzle Vodice is markedly different (Fig. 3). The Permian clastics are red with green and gray intercalations, named "Groeden equivalent". A layer of gray siltstones and sandstones, rich in pyritic cement surmounts "the red clastic series". The upper surface of the clastic is impregnated by a large quantity of pyrite, even massive at places, with delicate lamination and barite fenestrae. It was a former organically-rich, stromatolitic layer, pyritized completely during early diagenesis. It carpeted muddy shoals with stagnant, hypersaline water, as deduced by the absence of trapped detrital paritcles and grazing metazoans.

Fine, undulate lamination on a milimeter-scale at the contact of the clastics and dolomites changes to slightly coarser in the overlying dolomites, with the first sign of domal form, laterally passing into a flat laminated fabric, suggesting an increase in water energy. Stromatolitic fabrics succeeding upwards are upper intertidal to supratidal formations, and the curling of algal mats is due to desiccation processes (Fig. 4d).

Comparing the localities, there is an obvious difference in water energy. Školski brijeg might be referred to as a headland, while Homer belongs to a bight coastal type. Mrzle Vodice, with fine, laminated organic-rich stromatolites (transformed into pyrite) and no trapped particles, might have been a muddy embayment with sabkha environment characteristics (PLAYFORD & COCKBAIN, 1976).

2. SULPHUR ISOTOPES, TRACE ELEMENTS, ORE FORMING MODEL

To stay within the scope of the article, our attention will concentrate on an ore forming model supported by sulphur isotope and geochemical data. In order to describe the model more illustratively, a step-wise presentation has been chosen.

The first stage responsible for barite ore formation began with clastic deposition in the Late Permian (Fig. 6a). Their dark gray or black colour, abundant plant debris and ubiquitous pyritization point out reducing diagenesis, but not necessarily an euxinic bottom water condition. On the contrary, some textural elements, grain size and flora remnants are indirect signs of shore-line proximity and high energy water. Red, green and gray clastics, underlying the few-meter-thick, pyrite-bearing reduzate horizon at Mrzle Vodice (Fig. 3) have witnessed underground oxidizing terrestrial water, or could have even been exposed subaerially as country rock during an earlier regression cycle.

An abrupt change in sedimentary style, introducing carbonate deposition as a predominant process, is **the second stage** of the model (Fig. 6b). Structural features of the basal dolomites and other lithotypes indicate a peritidal environment and represent a preferable site for oncoming baritization. At that time, the first carbonate sediments were calcareous, preferentially aragonitic in

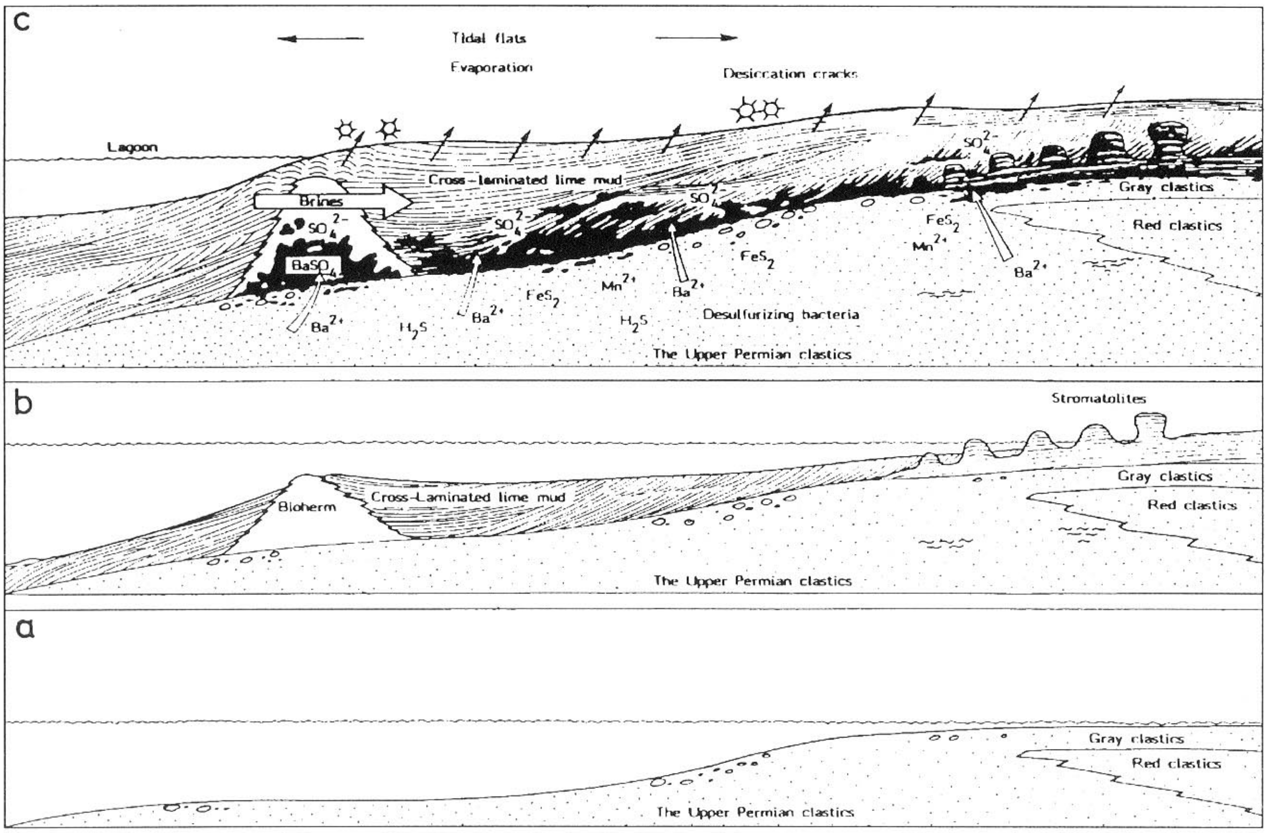


Fig. 6 Three stages of ore deposit development: **a** - deposition of Upper Permian clastics, **b** - regression and sedimentation of laminated lime mud, **c** - evaporitic dolomitisation accompanied by barite ore forming processes.

composition.

The third or "mineralization" stage is closely related to dolomitization processes (Fig. 6c). The lime sediments deposited in near-shore shallow water were soon subjected to subaerial evaporative pumping in a tidal flat environment, during a regressive phase of fluctuating sea level cycles. Early diagenetic dolomitization, as a replacing process caused by hypersaline brines with high Mg^{2+}/Ca^{2+} ratios, is followed by easily recognized concurrent manifestations such as:

- a significant increase in the porosity of dolomite in comparison to aragonite precursor, due to the remarkable difference in specific volume between aragonite and dolomite. It facilitates any subsequent infiltration process, e.g. baritization;
- early diagenetic lithification, making dolomitized layers rigid, has a characteristic geomechanical effect on underlying soft water-rich sediments. This is noticeable by frequent intraformational brecciation and colapsing structures (Fig. 4a). Subsequent upward dewatering of underlying soft deposits may have certain importance in the transport of barium to its final resting mineralization place;
- the upper surface of the tidal flat sediments, exposed to intensive drying, shows desiccation cracks and curling of the algal mat;
- the release of a high strontium concentration during dolomitization of aragonite mud may be even sufficient for the formation of celestobarite evaporitic deposits, as in the Zechstein (PUCHELT & MUELLER, 1964). Celestite may replace an older generation of evaporite minerals, or be an isomorphical substitute in barite minerals (WEST et al., 1968). Barite from the Lokve deposits is also rich in strontium. ŠIFTAR & ŠINKOVEC (1973) reported authigenic celestite as an accessory mineral in the deposits.

The source of barium should be sought in an emphatically reducing environment of an underlain organic-rich siliciclastic layer, a former stromatolitic algal mat, and clayey sediment. The thriving of desulfurizing bacteria upon burial enables a high production of dissimilatory H_2S, which lowers the Eh of connate water. Profitable H_2S production is accompanied by two effects, the formation of early diagenetic iron monosulphides and a decrease in sulphate concentration, which, along with high salinity, dissolves dispersed, tiny, authigenic barite minerals (PUCHELT, 1964; SCHERP, 1974).

The brines were being expelled from soft, sulphate depleted underlain mud by the heavy load of the rigid carbonate layer and evaporative pumping along all interstices, and solution cavities. They were expelled through porous laminated dolomites, which served as conduits and infilling space nourished by Ba-rich solutions.

Reprecipitation and encrustation of the basal dolomites by barium sulphate was **the early diagenetic process** as proved by intraformational colapsing and cracking of partly stiffened laminae (Fig. 4a). The same holds true for stylolites as a late diagenetic feature crosscutting dolomite-barite patches in mottled ores (Fig. 4b) and nodular and microcellular barite structures grown like concretions in tidal flat sediments (Figs. 5a, 5c).

Barium deposits, related to the dolomitization process have also been discovered in the English Lower Magnesian Limestones. Stratabound barite mineralization outcrops at many places from Newcastle to Nottingham along the contact of calcareous and carbonaceous mudstones and overlying Zechstein 1 carbonates, which stretch as a narrow zone for a hundred kilometers. At some sites it has a characteristic epigenetic appearance - **penetration flame structures** - illustrative and useful in supporting the proposed model as well (HARWOOD, 1981).

The last, but not the least important, contribution to the model are the geochemical data. Sulphur isotope $\delta^{34}S$ values, along two vertical profiles in the pyritiferous clastics at the Homer and Mrzle Vodice localities attest an increasing downward tendency (Figs. 7, 8; Tables 1, 2), in contrast to the Kupferschiefer profiles, which received a great deal of discussion during recent years (SWANNEY et al., 1987; SAWLOWICZ, 1989).

At the Homer locality the isotope values range between -15.47‰ and +10.44‰, and at the Mrzle Vodice locality from -15.00‰ to +20.15‰. This is conformatory evidence of a closed to semi-closed system regarding sulphate supply, already witnessed in a muddy environment (HARTMAN & NIELSEN, 1969). Nodular barite at Homer (Figs. 5a; 2, point A) has $\delta^{34}S$ values +17.06‰, stromatolitic, laminar barite +15.37‰ (Figs. 5b; 2, point C), and veinlets in biostroms +15.90‰ (Fig. 2, point B). At Mrzle Vodice, barite intercalation (Fig. 3, point A) is even at +17.40‰ and stromatolitic lamina +17.40‰ (Fig. 3, point B). All these values might be related to the Lower Triassic sea water, with a slight increase due to microbial fractionation. Using SCHWARCZ & BURNIE'S (1973) classification, and comparing the maximum values of +20.15‰ for pyrite and +18.07‰ for cogenetic barite, one may draw the conclusion that the system was open to H_2S, being permanently removed by extensive precipitation of early diagenetic pyrite.Trace element geochemistry is very similar to the Kupferschiefer in some details. The Mrzle Vodice ore bearing sequence, starting with "red beds" (Fig. 8), shows a characteristic zoning of Cu, Pb and Zn, Zn/Cu >> 1 and Co/Ni < 0.1. The maximum concentration is in massive pyrite (sample MV-13, Figs. 5c, 8, Fe_2S - 81,91%): Hg - 33.4 ppm, Zn - 320 ppm, Cu - 17 ppm, Pb - 720 ppm, Mn - 284 ppm. Manganese has the same fate as barium in a hydrogen-sulphide reducing environment. Its high mobility as Mn^{2+} gives rise to upward migration and encrustation at the clastics-dolomite contact, i.e. Eh-pH boundary (Fig. 5a).

WEDEPOHL (1980) explained the same feature as an abnormal composition of the Zechstein sea water and synsedimentary processes.

The Lokve barite mineralization, as attested by geochemical and sedimentary evidence, may be unequivocally named **"Prototype of the early diagenetic deposits"**. The major difference between the Lokve sulphur isotope profile and the typical Kupferschiefer one is the opposite tendency in $\delta^{34}S$ values (PALINKAŠ & PEZDIČ, 1989). The authors are inclined to say that the cause of this should be sought in the different hydraulic regimes during evaporative pumping due to local variations in discharge of terrestrial or lagoon water. It depends on many factors: type of hinterland, hilly or

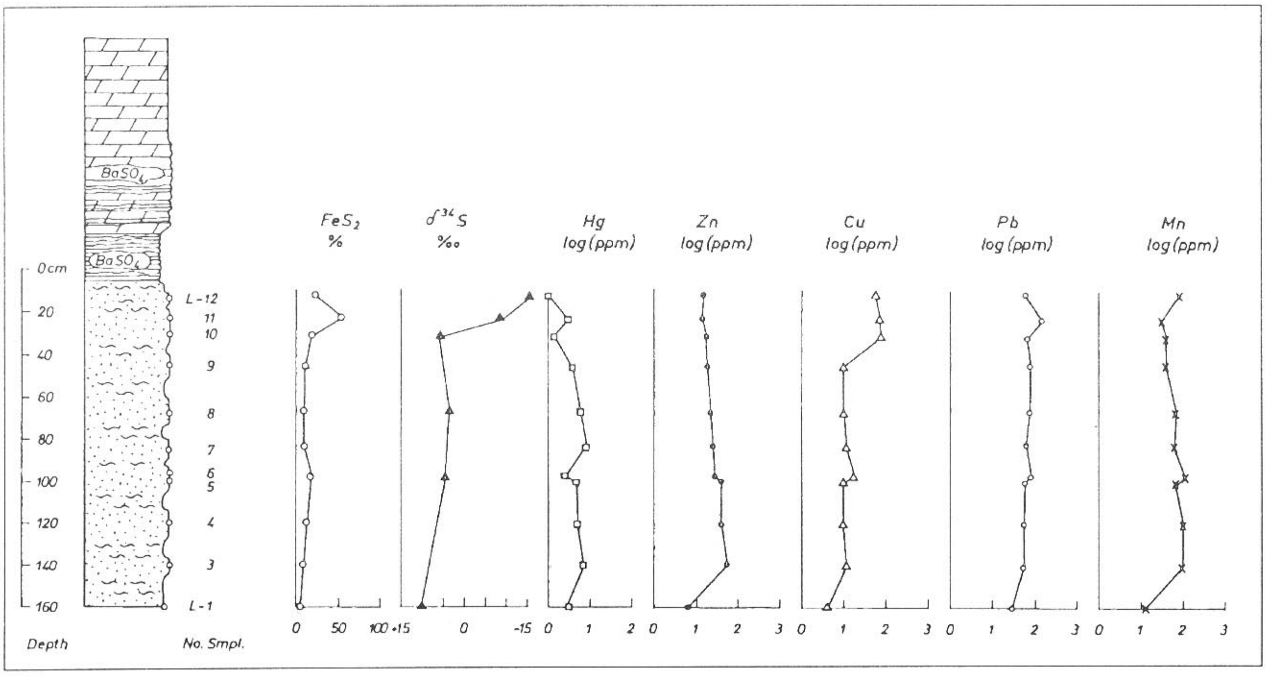


Fig. 7 Distribution of heavy metals and $\delta^{34}S$ values on the profile of the pyritiferous clastics at the Homer locality.

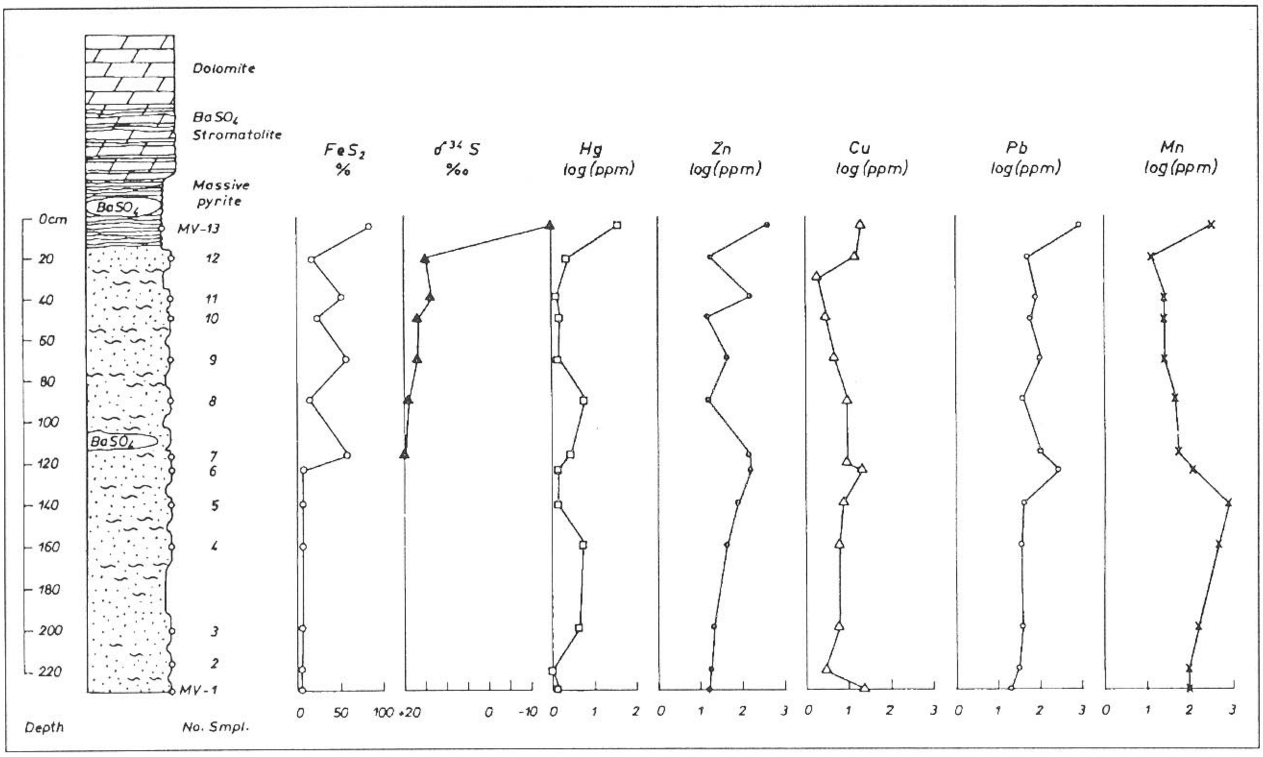


Fig. 8 Distribution of heavy metals and $\delta^{34}S$ values on the profile of the pyritiferous clastics at the Mrzle Vodice locality.

low-land, position of the Sabkha, landward or seaward, difference between high and low tide etc. Let us discuss two extreme situations:

1. Terrestrial water predominates (Fig. 9b). Water from red beds might have very light sulphate due to the oxidation of sedimentary pyrite, a good explanation for the extremly light sulphate on the famous MAROWSKY (1969) profile ($\delta^{34}S$ -22‰), and it passes through a hydrogen-sulphide generator in a stromatolitic mud layer, causing increasing tendency of $\delta^{34}S$ upwardly even in overlying dolomites, and a high concentration of base metals as in typical Kupferschiefer.

2. Lagoon or sea water predominates (Fig. 9a). Water passes through the porous upper layer of carbonates, carrying the sulphate of the contemporaneous sea or lagoon water over underlying reduzate mud, representing a semi-closed or closed system regarding sulphate supply, and an open system for hydrogen-sulphide, removed by precipitation of early diagenetic iron monosulphides and causing a characteristic increase of $\delta^{34}S$ downwardly, with low base metal content as in the Lokve barite deposits.

3. CONCLUSION

In order to summarize the presented ideas on the sedimentary and ore forming model, a short review of the important events in successive order is given. Deposition of the Upper Permian clastics. Regression

Sample	Depth m	FeS_2 %	$\delta^{34}S$ ‰	Hg ppm	Zn ppm	Cu ppm	Pb ppm	Mn ppm
MV-13	0.05	81.91	-15.00	33.4	320	17	720	284
MV-12	0.20	15.65	+15.56	2.4	18	14	46	11
MV-11	0.40	48.92	+14.08	1.2	46	2	72	23
MV-10	0.50	20.59	+16.86	1.5	15	3	52	23
MV-9	0.70	55.43	+17.23	0.7	40	5	83	23
MV-8	0.90	12.72	+19.34	5.3	16	9	37	38
MV-7	1.20	65.88	+20.15	2.6	141	9	82	49
MV-6	1.25	5.71		0.7	150	20	259	107
MV-5	1.40	1.63		1.5	97	8	40	792
MV-4	1.60	2.23		5.4	41	6	37	445
MV-3	2.00	1.87		4.4	20	6	34	131
MV-2	2.15	1.44		1.0	18	3	29	89

Table 1 Heavy metals and $\delta^{34}S$ values in the footwall pyrite-bearing clastics at the Mrzle Vodice locality.

Sample	Depth m	FeS_2 %	$\delta^{34}S$ ‰	Hg ppm	Zn ppm	Cu ppm	Pb ppm	Mn ppm
L-12	0.00	23.26	-15.47	1.0	15	55	58	79
L-11	0.25	52.60	-9.05	2.9	14	68	125	30
L-10	0.30	32.33	+6.38	0.7	19	72	63	38
L-9	0.40	10.81		3.4	17	10	72	38
L-8	0.70	8.47	+3.21	5.6	22	9	66	60
L-7	0.90	9.18		8.0	25	11	60	53
L-6	0.95	15.99	+4.52	2.7	29	16	77	114
L-5	1.00	8.81		4.9	38	10	59	75
L-4	1.20	10.69		5.1	40	10	55	93
L-3	1.40	8.80		6.6	60	11	55	89
L-1	1.60	9.96	+10.44	3.7	6	4	31	11

Table 2 Heavy metals and $\delta^{34}S$ values in the footwall pyrite-bearing clastics at the Homer locality.

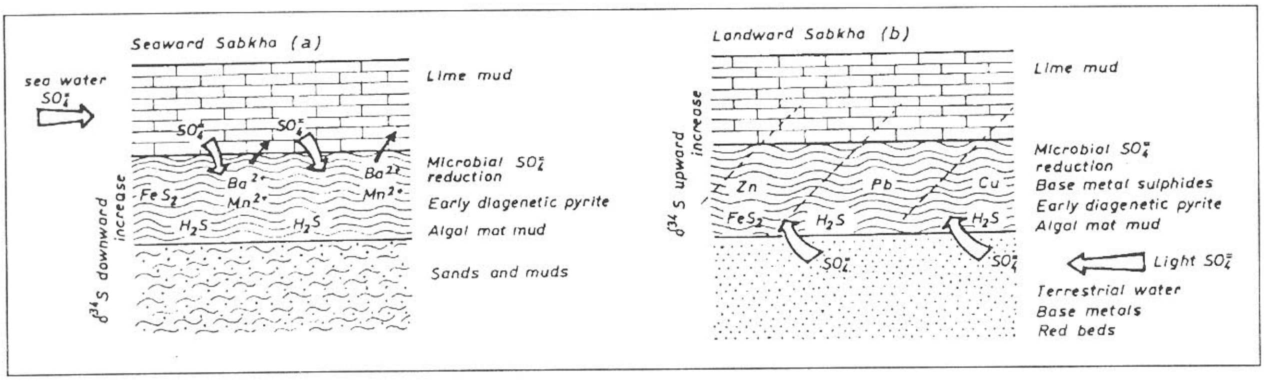


Fig. 9 Two hypothetical models for the explanation of sulphur isotope distribution and metal accumulation in Kupferschiefer-type deposits.

and carbonate deposition. Lowering of the mean sea level by evaporative drawdown and exposure of lime mud to intensive evaporation. Dolomitization and early diagenetic lithification. Loading of a rigid dolomitized horizon over a soft, organic-rich, reduzate, water-logged, clayey sediment, where reduction of sulphate, precipitation of iron monosulphides and release of barium take place. Expulsion and diffusion of Ba-rich, sulphate deficient brines into overlying porous dolomites, soaked with landward moving, hypersaline lagoonal brines, carrying sulphate, caused by overburden pressure and evaporative pumping. Barite precipitation in porous dolomite, possible replacement of former evaporites, and concretionary growth.

Stylolites as a late diagenetic feature, crossing through dolomite-barite patches in mottled ores, nodular and microcellular barite and pyrite structures and distribution of $\delta^{34}S$ values points out early diagenetic origin. Barium is remobilized in underlain, reduzate sediments by bacterial activity, lateral secretion mechanism and then reprecipitated in the dolomites again. The clear stratabound relationship of the mineralization with the sharp "clastic-dolomite" boundary, as well as its wide lateral extention, supports diagenetic origin. Dolomite-barite laminates with high energy water characteristics dismiss the possibility of direct barite precipitation from the sea water and preservation of the alternating laminae in intact position, i.e. syngenetic model.

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