# Biomineralization Processes During the Formation of Modern Oceanic Sulfide Ore and Ore-bearing Sediments

**Irina F. Gablina, Irina G. Dobretsova and Evgenia A. Popova**

**Abstract** Sulfide ores were investigated along with ore-bearing and metalliferous sediments of the hydrothermal fields in the northern near-equatorial Mid-Atlantic Ridge (MAR) zone: Semenov (13°30–31′N), Ashadze-1 (12°58′N), Zenit-Victoria (20°08′N), and Peterburgskoe fields (19°52′N), discovered during legs 26, 32, and 33 of the R/V *Professor Logachev* FSUE PMGE. Biogenic carbonate and background sediments of this region were also examined. Lithological, biostratigraphic, and geochemical physical-chemical investigations methods were used. Mineragraphic and precision structural and chemical research of typomorphic minerals were carried out at various stages of lithogenesis. It was found out that most sulfide constructions in the Zenit-Victoria and Peterburgskoe fields, as well as the eastern field of the Semenov cluster, are located in biogenic carbonate sediments of the Holocene and Late Pleistocene ages and represent a new type of sulfide mineralization, unknown earlier in the MAR zone. This mineralization was formed by metasomatic replacement of biogenic carbonate sediments by ore minerals, simultaneously with diffuse percolating of hydrothermal solutions through the sediments.

**Keywords** Ocean · Mid-Atlantic ridge · Modern sulfide ores · Ore-bearing sediments · Biomineralization · Hydrothermal · Metasomatic

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## 1 Introduction

Massive sulfide ores from the ocean are chimneylike ore mounds (black smokers) formed under highly forced hydrothermal fluids, which are vented on the seafloor in various geological settings, mainly in spreading zones of the mid-oceanic ridges. Sulfide mineralization hosted in recent sediments has been discovered near continents and in intracontinental rifts, such as in the Red Sea.

It is suggested that most ore material is supplied into marine sediments from hydrothermal plumes, which form in the ocean above active hydrothermal vents. The effect of hydrothermal solutions on sediments while the solutions seep through them has been much less studied.

Ore-bearing sediments formed under the effect of hydrothermal solutions have been studied most completely in the Red Sea rift zone (Baturin 1971; Pushelt and Laschek 1984; Bogdanov et al. 1986; Butuzova 1998, 2003) and in the rifts adjoining the North American continental block in the west: the Guaymas Basin in the Bay of California, the Middle Valley of the Juan de Fuca Ridge, Escanaba Trough, etc. in the Pacific Ocean (Curray et al. 1982; *Gidrotermal'nye* … 1993; Goodfellow and Franklin 1982; Kurnosov et al. 1994; Zierenberg et al. 1993; Lisitsyn et al. 1989; *Geologicheskoe* … 1990; Bogdanov et al. 1989, 2006). These processes have remained poorly studied until now in mid-ocean ridges, which are distant from continents and covered only with thin sedimentary cover.

We believe that the main reason for this situation is the generally agreed opinion that the formation of ore-bearing sediments predominantly occurs by sedimentation (due to destruction of surface hydrothermal structures and precipitation of the ore substance from suspensions of hydrothermal plumes). Another cause is the unilateral approach to studying ore-bearing and metalliferous sediments: commonly, specialists (lithologists, paleontologists, geochemists) explore their own field of expertise, applying methods used in that field. These methods do not always allow them to determine the nature of mineral components. These components can be hydrothermal-sedimentary (precipitating from hydrothermal plumes or formed by destruction of hydrothermal mounds and vents), authigenic (appearing in situ as products of postsedimentation transformations, including hydrothermal metasomatic alteration), terrigenous (coming from continents), and edaphogenic (products of ocean bed destruction).

Massive sulfide deposits—ancient analogues of the contemporary oceanic massive sulfide ores—are thick stratiform bodies embedded, as a rule, in sedimentary and volcano-sedimentary rocks (Maslennikov 2006). This is similar to other type of deposits (cuprous sandstones and shales) formed under underground waters that emanated to submarine bottom sediments (Hoyningen-Huene 1963; Lur'e and Gablina 1972; Lur'e 1988; Rose 1976; White 1971). Both types are characterized by high concentrations of copper, lead, zinc, and rare elements and are the main sources for them. Enrichment in metals is explained by protracted circulation of ore-bearing fluids in sediments that serve as a barrier for metals (Gablina 1997), whereas venting of hydrothermal ore-bearing fluids in seawater results in their dispersion. Less than 5 % in the initial stage and up to 50 % in further periods are precipitated in sulfide mounds, which are formed on the seafloor near hydrothermal vents (Rona 1986; Gurvich 1998). The destruction of unstable massive sulfide ores under seawater began just after their formation (*Gidrotermal'nye*... 1993; Gablina et al. 2000).

In recent years, we and other researchers (Gablina et al. 2010, 2011, 2012; Dobretsova and Laiba 2009; Rusakov et al. 2013) obtained new data about the metasomatic effect of hydrothermal solutions on modern biogenic carbonate sediments of the Ashadze-1, Eastern (Semenov ore cluster), Zenit-Victoria, and Peterburgskoe ore fields of the Mid-Atlantic ridge (MAR), which led to the substitution of microfaunal shells of sediments by sulfides and other hydrothermal minerals, as well as to the formation of sulfide structures in them. This information is very important from both scientific and practical points of view, as the seepage of hydrotherms through the sediments might evoke precipitation of a larger part of the net load as well as the formation and preservation from destruction of a rich accumulations of sulfide ores.

## 2 Materials and Methods

Sulfide ores were investigated along with ore-bearing and metalliferous sediments of hydrothermal fields in the northern near-equatorial MAR zone: Semenov (13°30–31′N), Ashadze-1 (12°58′N), Zenit-Victoria (20°08′N), and Peterburgskoe fields (19°52′N), discovered during legs 26, 32, 33 of the R/V *Professor Logachev* FSUE PMGE, and carbonate, background sediments of this region. The investigations are based on a systematic approach to studies of the conditions of mineral and ore formation, which consists of a combination of lithological, biostratigraphic, and geochemical investigations. Mineragraphic, precision structures, and chemical research of typomorphic minerals, ore inclusive, were conducted at various stages of lithogenesis. The findings were compared with the data of direct measurements of Eh-pH parameters of ore-bearing and background sediments obtained during the leg. The main object of the research is the biogenic material of sediments, the mineral composition of which is uniform at the stage of sedimentogenesis within the ocean zone under consideration; these are calcite shells of dead plankton and benthos. Their different transformations may be associated with various conditions of diagenesis or superimposed processes.

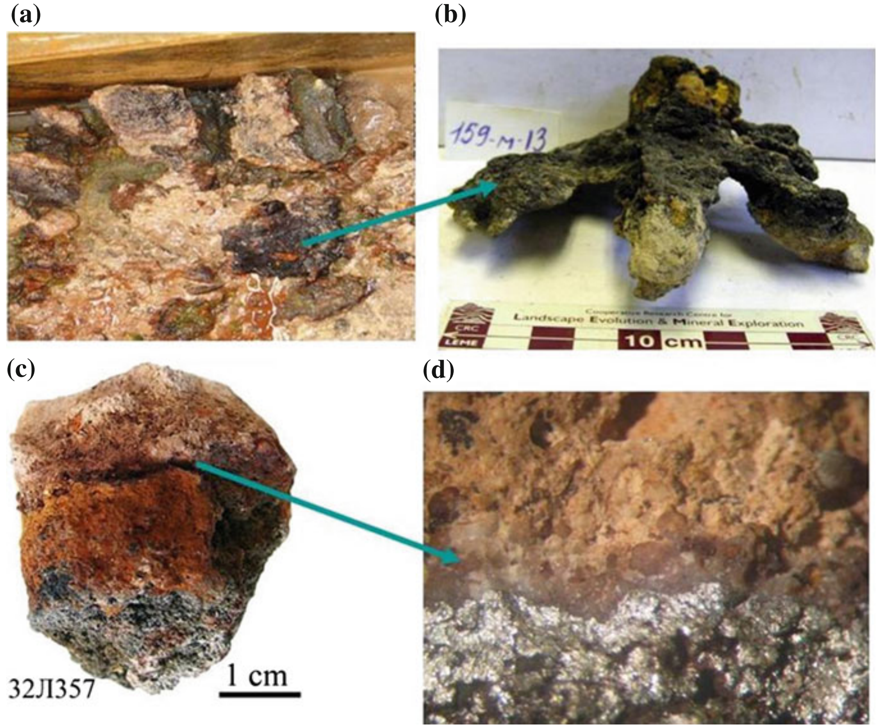


**Fig. 1** The studied area. The studied subjects are earmarked

## 3 Characterization of the Work Region

Ore fields are situated at the boards of the MAR rift valley in the area from 12° 58.4'с.ш (Ashadze-1 field) to 20°08′N (Zenit-Victoria field) at depths from ~2400 to 4200 m (Fig. 1). Ore fields are composed of basalts, gabbro, peridotites, carbonate, metalliferous, and ore-bearing sediments with inclusions of sulfide ores.

The considered ore fields occupy a specific tectonic position: they are all situated in rift valley walls, with the Peterburgskoe field being found in the second ridge of the wall. Hydrothermal fields are characterized by developing metalliferous ($Fe_{cfs}$[1] ≥ 10 %, $(Cu + Zn)_{cfs}$ < 0.25 %), ore-bearing ($Fe_{cfs}$ ≥ 10 %, $(Cu + Zn)_{cfs}$ ≥ 0.25 %), and carbonate ($Fe_{cfs}$ < 10 %, $(Cu + Zn)_{cfs}$ < 0.25 %; $CaCO_3$ ≥ 50–70 %) sediments. The thickness of carbonate sediments in this MAR sector does not exceed 1.5–3 m; ore-bearing sediments are no more than 0.5–0.6 m. Unaltered sediments are represented by calcite shells of foraminifers, coccolithes, and pteropods with admixture of clay and edaphogen material. According to the

[1]/cfs—based on carbonate-free substance.

fauna assemblage, the age of the sediments is Holocene-Late Pleistocene (0–128 ka) (Gablina et al. 2011; Rusakov et al. 2011; Shilov et al. 2012).

The Ashadze-1 hydrothermal field is represented by two ore bodies with relict and active hydrothermal constructions and ore-bearing sediments. The Semenov ore cluster contains both active constructions (North-Western hydrothermal field) and massive ores embedded in sediments (Eastern hydrothermal field). No active constructions have been revealed in the Zenit-Victoria and Peterburgskoe hydrothermal fields, whereas sulfide formations were recorded in topography in the form of small hills. The majority of the investigated sites are characterized by hydrophysical anomalies that indicate continuing hydrothermal activity. The age of sulfide ores, dated by the isotopic Th/U metod, ranges from <2–60 ka (Zenit-Victoria) to 37–124 ka (Semenov) (Shilov et al. 2012; Rusakov et al. 2013).

Most sulfide constructions in the Zenit-Victoria, Peterburgskoe, and Eastern fields of the Semenov cluster are located in sediments and represent a new type of sulfide mineralization, unknown earlier in the MAR zone. The mineralization was formed by metasomatic replacement of organic carbonate sediments by hydrothermal minerals, simultaneously with diffuse percolating of hydrothermal solutions through the sediments (Gablina et al. 2012; Rusakov et al. 2013).

## 4 Results

Sulfide ores have been most completely studied in Zenit-Victoria and Peterburgskoe hydrothermal fields. Here the ores are represented by scattered sulfide constructions composed of a porous pyrite-marcasite or sphalerite-marcasite (in Cu–Zn–Fe ores) framework. Chalcopyrite, sphalerite, copper sulfides, and, in rare cases, bornite develop in the most porous places of the framework. Opal, barite, and Fe–Si gel are most abundant among nonmetallic hydrothermal minerals.

Investigations have shown that sulfide constructions in sediments of the Zenit-Victoria and Peterburgskoe fields have certain features that distinguish them from tubular bodies of "black smokers" described in the literature, which were generated on the surface of the ocean floor (Gablina et al. 2012). Those features include the following:

1. Unusual shape: streamlined pear- and mushroom-shaped, platy stratified (Fig. 2).
2. Multichannel inner structure characteristic of "diffusers" (Fig. 2b).
3. Hardening crusts in the roof and on the walls of structures—lithified sediments, commonly situated at channel outputs (Fig. 2a, c–f).
4. Presence of relics of sedimentary layering in platy structures (Fig. 2f).
5. Widely spread relics of microfossils replaced by ore minerals (Fig. 3).
6. Conformity of the height of structures with the thickness of enclosing sediments.

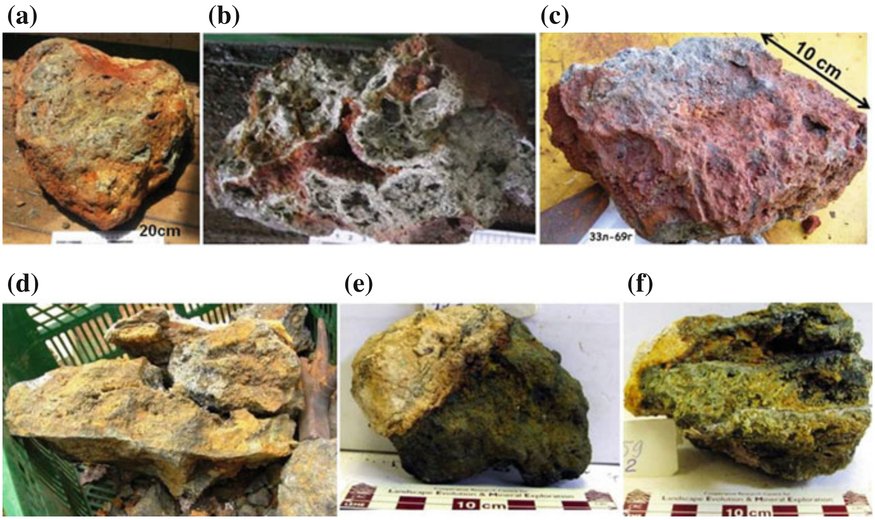


**Fig. 2** Morphology and the inner structure of sulfide edifices. Zenit-Victoria field: **a** a pear-shaped construction with a crust of lithified sediments in the roof, st. 33L70; **b** a multichannel sulfide construction ("diffuser"), st. 33L68; **c** a fragment of platy stratified edifice, st. 33L69. Peterburgskoe field, st. 33L159: **d** a large construction of stratiform type with a channel in the floor and a crust of lithified sediments in the roof; **e** a small construction with a crust of lithified sediments in the roof; **f** a fragment of sulfide construction of stratiform type with relics of the stratified texture and with a crust of lithified sediments

7. The presence of aragonite crusts with Fe and Mn hydroxides on the surface of sediments that contain sulfide formations or aragonite zones at the contact of sediments with sulfide structures (Fig. 4).

Within hydrothermal fields, metalliferous and ore-bearing sediments are spread in the form of spots, which is not consistent with the idea of a uniform precipitation of ore particles from hydrothermal plumes. They differ from background ones in their physical-chemical characteristics, by elevated content of ore components (Fe, Cu, Zn, and Mn), Si, Mg, metasomatic replacement of calcite tests by hydrothermal minerals, and development of near-ore mineral-geochemical zones.

Lateral and vertical zonation was established in the distribution of major rock- and ore-forming elements and secondary (hydrothermal) minerals in the sediments of the Ashadze-1 field. Hydrothermal minerals are represented by silicates (tremolite-actinolite, hornblende, serpentine, Fe–Mg-smectite, and, probably, polygorskite [sepiolite]), carbonates (Mg–Mn-bearing siderite), sulfides (pyrite, isocubanite), sulphates (millerite, barite, celestine), chlorides (atacamite, halite), and Fe and Mn hydroxides, which metasomatically substitute calcite in microfossil tests and form porous and crustified cement of the sediments. Pyrite, siderite, and Fe hydroxide and sulphate are most widespread newly formed minerals.

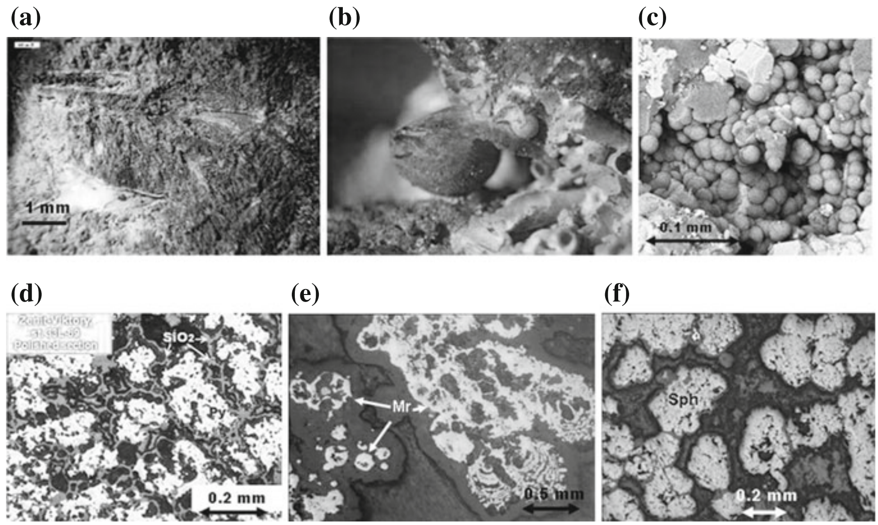


**Fig. 3** Relics of microfossils replaced by ore minerals. Microphotograph of the samples under the binocular: **a** pteropod imprints in chalcopyrite-sphalerite ores, Zenit-Victory hydrothermal field, st. 33L69; **b** opal pseudomorph after pteropod shell at the channel mouth, white below on the *right*—unaltered calcite shells, pyrite-chalcopyrite ores with Cu-sulfides, opal, Peterburgskoe field, st. 33L159. Zenit-Victory hydrothermal field, st. 33L69: **c** accumulations of opal pseudomorphs after foraminifers in the cavities of pyrite-chalcopyrite ores, electron microscopy; **d** microphotograph of a polished section of pyrite (Py) pseudomorphs after foraminiferas opal was crusted over; **e** microphotograph of a polished section of marcasite (Mr) pseudomorphs after foraminiferas in $SiO_2$ in pyrite-marcasite ores; **f** microphotograph of a polished section of sphalerite (Sph) pseudomorphs after foraminiferas in $SiO_2$ in pyrite-chalcopyrite-sphalerite ores

The following zones are identified according to the composition of the dominant secondary minerals:

(1) A sulfide zone, which coincides with ore bodies
(2) A zone of minerals with elevated magnesia content, which partly coincides with ore body 1, going beyond its contours in the west and southwest
3) A zone with developing Fe–Mn crusts along the periphery of ore bodies 1 and 2.

Vertically, sulfide paragenesis is substituted by the hydroxide-ferruginous. Farther away from the ore bodies, the processes of dissolution and replacement of carbonate shells by secondary minerals in sediments fade (Gablina et al. 2010, 2011).

The sediments of the Semenov field also exhibit metasomatic substitution of foraminiferal tests by Fe and Cu sulfides and near-ore hydrothermal alterations. Vertical zonation is also recorded in the distribution of hydrothermal minerals in host rocks. In the upward direction, from the roof of the sulfide deposit toward the surface of the ocean floor, the sulfide-barite association progressively gives

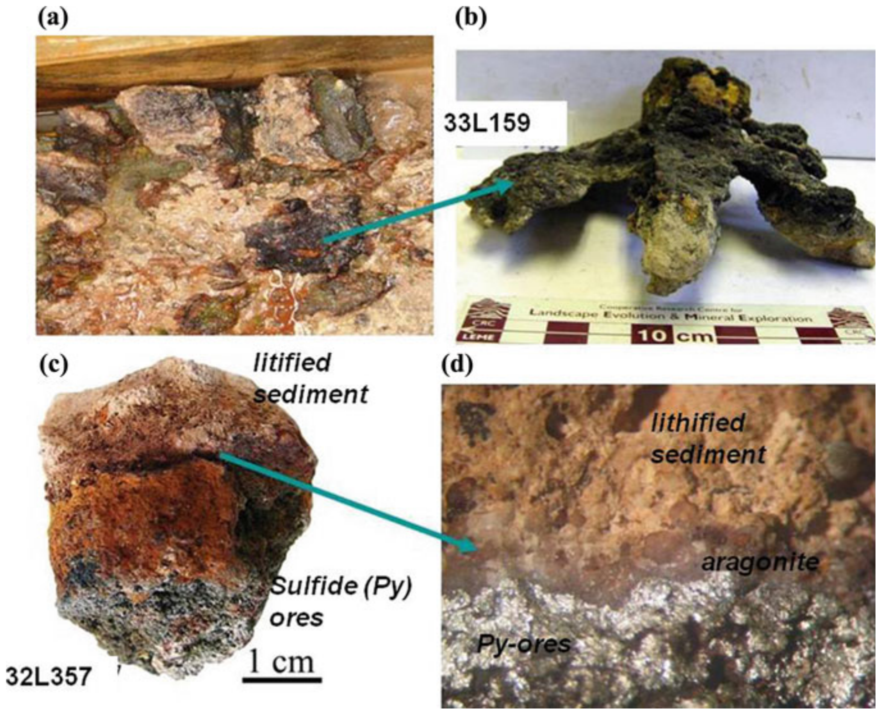


**Fig. 4** Aragonite genesis. **a**, **b** Crusts covered with a shade of Mn hydroxides on the surface of the sediment, which contains sulfide edifices (Peterburgskoe field, st. 33L159). **c**, **d** Zone at the contact of pyrite (Py) ores and lithified sediment in the roof of edifice (Semenov field, st. 32L357)

way to the hydroxide-ferruginous-sulfide-barite, barite-hydroxide-ferruginous, and atacamite-hydroxide-ferruginous associations (Rusakov et al. 2013). This zonation reflects an increase in the oxidation potential of porous solutions of sediments up the section as a result of the growing influence of seawater.

Ore-bearing sediments and host sulfide constructions of the Zenit-Victoria and Peterburgskoe fields were intensely altered, which is reflected in their lithification at the contact with ores, the development of secondary minerals in them (Fig. 5), a sharp decrease in Eh and pH values with respect to background, and an increase of porous water temperature. Simultaneously, the oxidation potential decreases (to negative values) while the acidity increases down the section, clearly indicating that acid-reducing hydrothermal fluids arrive to the sediments from below (Gablina et al. 2012).

In general, the following characteristics are typical of the studied objects:

1) Reduction in the thickness of sediments occurring above sulfide constructions or enclosing them (probably due to dissolution of primary biogenic calcite).

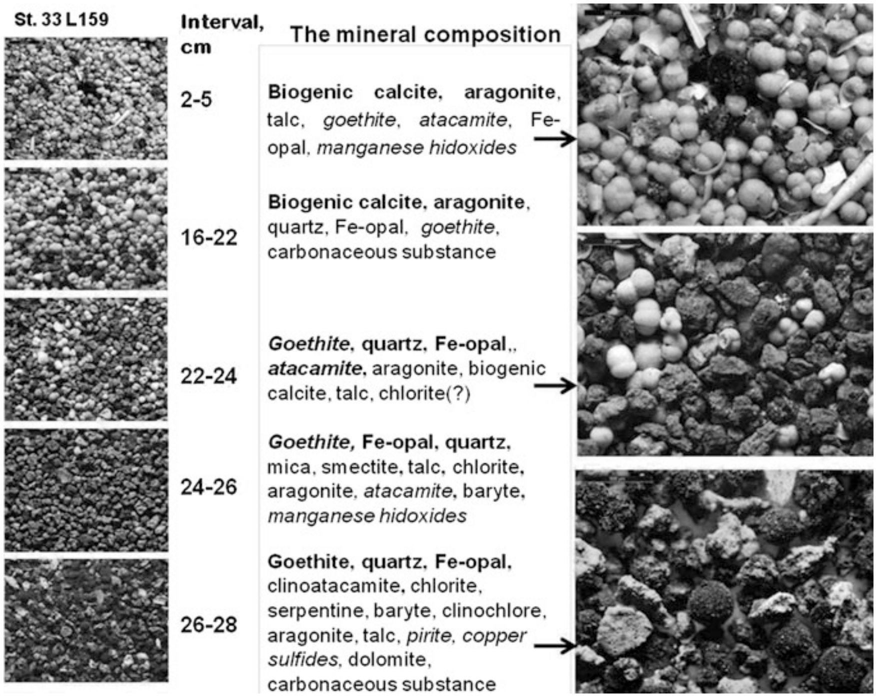


**Fig. 5** Vertical mineral zonation of biogenic carbonate sediments in the zone of near-ore alterations in the Peterburgskoe hydrothermal field (core st. 33L159). A fraction contains 0.25–0.5 mm of sediments. The mineral composition is based on the data of mineragraphic, electron microscopic, electron microprobe, and X-ray investigations. *Left* Microphotograph of the samples under the binocular. *Right* Their enlarged fragments. Predominant minerals are given in *bold type*, with mineral indicators of the physical-chemical environment shown in *italics*

2) Development of pseudomorphs of ore and other hydrothermal minerals after microfauna tests in ore-bearing sediments (Gablina et al. 2011).
3) Vertical and lateral mineral-geochemical zonation in ore-bearing sediments around ore bodies; the sedimentary section, from bottom to top, reflects an increase in the oxidation potential and pH up the sedimentary column and at the flanks of the ore fields (Fig. 5).
4) A sharp difference of ore-bearing sediments from background sediments revealed by the physical and chemical characteristics of their interstitial waters; Eh and pH of ore-bearing sediments are close to the physical-chemical parameters of hydrothermal solutions (Gablina et al. 2012).

## 5 Conclusions

The described shapes and the inner structure of sulfide constructions (“diffusers”) suggest that their formation occurred under incomplete permeability of the environment, which hampered the spread of fluids (i.e., the diffusion mechanism of flow promotion). In sulfide ores, the presence of relics of the layered sedimentary structure and pseudomorphs of ore minerals after microfauna tests suggest the metasomatic origin of ore constructions. The biogenic calcite that composed the sediments dissolved during the test and was replaced by hydrothermal minerals. The ore-forming process can be associated with the influence on sediments of acid metalliferous low pressure fluids of diffusion type, which arrive from below through permeable zones in basalts. Mineralization occurred on the geochemical barrier in the zone of interaction among metalliferous fluids, seawater saturating the sediments, and biogenic sedimentary calcite that is unstable in the acidic environment. Carbonate bottom mud served both as a geochemical barrier (with the presence of reactive carbonate material) and a physical barrier, making rapid migration of ore-bearing solutions difficult and encouraging precipitation and accumulation of useful components. At the initial stages or at low pressure of fluids (e.g., in the “exocontact” of the already formed constructions), metasomatic replacement of isolated tests leads to the formation of ore-bearing and/or metalliferous sediments. The prolonged influence of hydrotherms brings about the complete replacement of sedimentary biogenic material and the formation of sulfide constructions in its place. The recycling of dissolved biogenic calcite occurs on the surface of sediments (the sea floor) in aragonite crusts, enriched in Fe and Mn hydroxides, or at the contact of sediments with sulfide structures (Fig. 4). The presence of hardening crusts of sediments in the roof and on the walls of the sulfide constructions indicates the increased temperature of leaking fluids. The occurrence of isocubanite among the newly formed hydrothermal minerals indicates a high temperature (>260 °C) of hydrothermal solution supplied to sediments (Vaughan and Craig 1978).

Our conclusions concerning superimposed metallization on Holocene-Late Pleistocene (0–128 ka) bottom sediments do not contradict the data on the absolute age of sulfide ores (from less than 2 to ~124 ka)[2] and the availability of recent hydrophysical anomalies above the Zenit-Victoria ore field, which indicate ongoing hydrothermal activity there (Shilov et al. 2012).

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[2]The dating in the Petersburgskoe ore field of 176.2 ± 59 ka (Shilov et al. 2012) needs to be clarified.

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