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BARITE-IRON OXIDE-PYRITE DEPOSITS FROM APUANE ALPS (NORTHERN TUSCANY, ITALY)

(Figs. 3, Tab. 1)

Abstract: In the Apuane Alps area, where Pb—Zn±Ag, Hg, Fe—Cu and Fe—Mn mineralizations also occur, barite-Fe oxide-pyrite deposits are the only which are currently exploited. They are localized in the upper part of a low-grade metamorphic silicoclastic complex (Scisti di Fornovolasco; Middle Triassic), in particular, in association with carbonatic intercalations ("Calcare metallifero"), and in the bottom of the overlying weakly metamorphic dolostones (Grezzoni: Norian). The morphology is mostly stratiform, with minor related discordant bodies (veins or irregular masses). In some stratiform bodies, a clear zonality is recognized (from bottom upwards: pyrite + barite — barite — barite + Fe oxides). Banded textures, and mega-, meso- and microscopic evidence of tectonic deformation are commonly observed.

The main ore minerals are barite, heamatite, magnetite and pyrite, with minor galena, sphalerite, siderite and sulphosalts. Reconnaissance S-isotopic and fluid inclusion data are: δ^{34}S (sulphides) = −29.9 to +18.9 per mil; δ^{34}S (barite) = +6.5 to +30.8 per mille; homogenization temperatures of fluid inclusions in quartz and barite = 190° to 250 °C, with salinities of about 10 % wt. NaCl equivalent.

The deposits, as many others in the Apuane Alps, were previously considered to be formed from hydrothermal fluids related to hypothetical early- to synkinematic granitic intrusions during the Tertiary Apenninic orogeny. We suggest, on the contrary, that they had a Middle Triassic sedimentary-diagenetic origin, followed by tectonization, metamorphism and partial remobilization during the Apenninic orogeny.

Резюме: В районе Апуанских Альп, где встречаются и Pb-Zn±Ag, Hg, Fe-Cu и Fe-Mn оруденения, единственными месторождениями, которые в настоящее время эксплуатируются, барит-Fe окись-пиритовые месторождения. Они находятся в верхней части метаморфического силикокластического комплекса низкой степени (Scisti di Fornovolasco; средний триас), в частности, в ассоциаци с карбонатными пропластками („Calcare metallifero") и в нижней части они залегают над слабометаморфизованными доломитами (Grezzoni: норий). Морфология преимущественно стратиформная с второстепенными родственными дискордантными телами (жилы или нерегулярные массы). В некоторых стратиформных телах встречается ясная зональность (снизу вверх: пирит + барит — барит — барит + Fe окиси). Исследуются текстуры и мега-, мезо- и микроскопические доказательства тектонической деформации.

Основными рудными минералами являются барит, гематит, магнетит и пирит с второстепенным галенитом, сфалеритом, сидеритом и сульфосолями. Данные исследования S-изотопов и флюидной инклюзии следующие: δ^{34}S (сульфиды) —29,9 до +18,9 промилле; δ^{34}S (барит) = +6,5 до +30,8 промилле; температуры гомогенизации флюидных инклюзий в кварце и барите = 190° до 250 °C, с соленостью около 10 весовых процентов NaCl.

Первоначально предполагалось, что эти месторождения, как много других в Апуанских Альпах возникли из гидротермальных флюидов родственных

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гипотетическим ранним — синкинематическим гранитным интрузиям во время третичного апеннинского орогена. В отличие от предыдущего, мы предполагаем, что они среднетриасового, осадочно-диагенетического происхождения с последующей тектонизацией, метаморфизмом и частичной ремобилизацией во время апеннинского орогена.

Introduction

The Apuane Alps are a mountain group located in the north-western part of Tuscany, near the coast of the Tyrrhenian Sea (Fig. 1). The region is worldwide known for the quarries of the famous "Marmo di Carrara", but iron, manganese, lead-zinc-silver, copper, mercury, pyrite and barite deposits have also been mined (Carmignani et al., 1972).

Mineral deposits of Apuane Alps can be schematically divided into the following types:

a) iron oxide(pyrite)-quartz: Casone type and Macchione type;
b) Fe(Mn, Cr) oxides-siderite(barite)-quartz: Strettoia type;
c) barite-Fe(Mn) oxides-pyrite-quartz(siderite): Valdicastello-Fornovolasco belt type;
d) Mn carbonates and silicates (oxides)-(pyrite, haematite)-quartz: Scortico type;
e) Cu(Fe, Pb, Zn) sulphides-quartz, calcite; Vagli type and Colle Panestra type;
f) Cu(Fe, Ni, Sb, As, Bi, Pb, Zn) sulphides and sulphosalts-siderite-quartz: Frigido type;
g) Pb, Zn, Ag(Cu, Fe, Sb, As, Au) sulphides and sulphosalts-barite-fluorite, quartz, tourmaline, carbonates: Bottino type;
h) Hg(Sb, Zn, Fe) sulphides-quartz: Levigliani type and Ripa type.

It is believed that mining activity in the region started in the centuries before Christ. The maximum development was achieved in the last century and in the first half of this century; today, only the barite-iron oxide-pyrite deposits at Pollone (Valdicastello), Monte Arsiccio and Buca della Vena, in the southernmost slopes of the mountain group, are exploited (Cipriani—Tanelli, 1983).

Considering the scientific and the potential economic interest of the mineralizations of the Apuane Alps, a systematic study of their geological, mineralogical and geochemical characters has been undertaken. We report here the results of reconnaissance work specifically dealing with the barite-iron oxide-pyrite deposits.

The genesis of these deposits is quite debated. According to Bergmann (1969), they would represent Permo—Triassic exhalative deposits; a syngenetic origin was also hypothesized by Natale (1974). On the other hand, Carmignani et al. (1972; 1975; 1976) proposed an epigenetic origin during the Tertiary Alpine-Apennine orogeny, from hydrothermal fluids related to synkinematic granitic intrusions, the very existence of which, however, remains highly speculative. Recently, a Triassic (pre-Norian) origin, with subsequent metamorphism and tectonization during the Apennine orogeny, was postulated by Ciarapica et al. (1983) and Tanelli (1983).

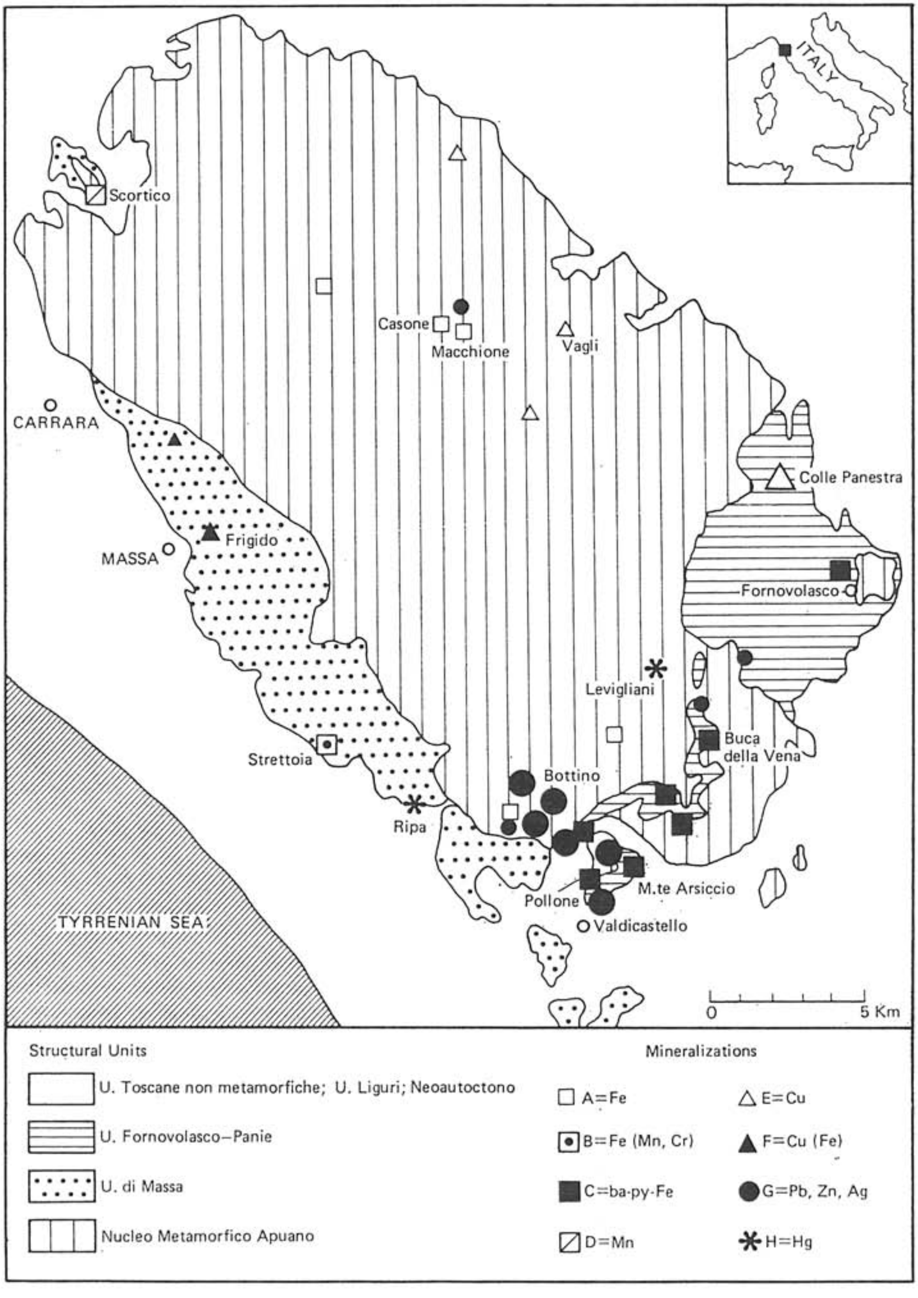

Fig. 1. Simplified structural scheme of the Apuane Alps (after Ciarapica—Passeri, 1982), with localization of the principal mineralizations.

Explanations: Larger symbols refer to deposits which are currently exploited (Ba—Fe—pyrite deposits of Buca della Vena, Monte Arsiccio and Pollone), or were exploited in the past. Smaller symbols indicate other mineral occurrences.

Geological setting of the Apuane Alps

The Apuane Alps consist of several metamorphic and non-metamorphic structural units, belonging to diverse paleogeographic domains, overthrusted during the Oligo—Miocene stages of the Alpine-Apennine orogeny (27 to 11 m. y. b. p.; Kligfield et al., 1981; Boccaletti et al., in press).

There is no unanimous consensus about the stratigraphic, paleogeographic and structural reconstruction of the region. The description given below is essentially in agreement with Ciarapica—Passeri (1982), to which the reader is referred for further details and bibliography.

The geometrically lowermost unit is a metamorphic complex known as "Nucleo metamorfico apuano", tectonically overlain by metamorphic ("Unità di Massa") and weakly metamorphic ("Unità di Fornovolasco-Panie") units, which in turn are tectonically overlain by the non-metamorphic "Unità toscane non metamorfiche" and "Unità Liguri".

Reconstruction of stratigraphy of the "Nucleo metamorfico apuano" is made difficult by the presence of several heteropies, and by the scarcity of representative fossils. Schematically, the lowermost member is a Paleozoic basement, represented by phyllites, quartzites, felsic metavolcanics ("Porfiroidi") and their volcanoclastic products ("Scisti porfirici"). The basement is unconformably overlain, locally with interposition of Triassic silico-carbonatic terrains, by the dolomitic formation of "Grezzoni" (palaeontologic age: Norian and possibly Rhaetian). The sedimentary environment of the "Grezzoni" formation is interpreted as a hyperhaline carbonatic platform, passing to a shallow coastal basin. The "Grezzoni" formation is followed upward by carbonatic, argillaceous and arenaceous formations (Jurassic to Oligocene). The metamorphic grade of the "Nucleo metamorfico apuano" has been established in the greenschist facies (quartz-albite-white K-mica-chlorite assemblage), with estimated peak temperatures of 300 °—400 °C, and pressures of 3—4 kbar (Carmignani et al., 1978).

In the Apuane region, the "Unità di Massa" consists of phyllitic, quartzitic and carbonatic terrains, with minor occurrences of mafic metavolcanics. The palaeontologic age of these terrains is Middle Triassic (Anisian to Carnian), and their paleogeographic environment is described as an active tectonic trough (aborted proto-oceanic rift). The metamorphic grade of the "Unità di Massa" is slightly higher than the "Nucleo metamorfico aquano", as witnessed by the appearance of biotite.

The "Unità di Fornovolasco-Panie" starts with a phyllitic complex (sericite-quartz-chlorite-albite; lower greenschist metamorphic facies), within which beds or lenses of quartzites and of carbonatic rocks are interlayered, especially in the uppermost portion of the complex. The carbonatic beds were sometimes referred to as "Calcare metallifero", because Be-Fe-Pb-Zn-Ag mineralizations are often in association with them. The phyllites are characterized by the occurrence of an association of tourmaline and quartz, with minor pyrite and rutile, known as "tourmalinolite", which in turn may be associated to Pb-Zn-Ag and/or Ba-Fe mineralizations. The genesis of this "tourmalinolite" is quite problematic; Ciarapica—Passeri (1982) refer it to an unspecified Middle Triassic (volcanic?) event. Ciarapica — Zaninetti (1983) define the whole phyllitic complex as "Scisti di Fornovolasco", and assign it to the Ladinian—Carnian on the basis of foraminifera discovered in its upper portion. The "Scisti di Fornovolasco" are followed upward by weakly metamorphic (lower greenschist facies) dominantly carbonatic formations (Upper Triassic to Cretaceous—Eocene?). The lowermost of these formations is represented by Upper Triassic dark limestones and dolostones ("Grezzoni delle Panie").

Geology and mineralogy of the deposits

Apart from minor uneconomic Cu mineralizations in the "Unità Liguri" and Mn occurrences in the "Unità toscane non metamorfiche", all mineralizations of the Apuane Alps are associated with metamorphic terrains, mostly to formations underlying the Norian—Rhaetian "Grezzoni". The few exceptions to associated to the upper portion of the "Seisti di Fornovolasco" (especially to the latter rule are the Cu (Pb, Zn) deposits at Vagli and Colle Panestra, the Mn occurrence at Scortico, and possibly the Pb-Zn-Cu (Ag, Au) mineralization at Buca dell'Angina near Valdicastello (Fig. 2).

In particular, the barite-iron oxide-pyrite deposits are restricted to the "Unità di Fornovolasco-Panie". They present a marked stratabound character, being associated to the upper portion of the "Scisti di Fornovolasco" (especially to the carbonatic levels known as "Calcare metallifero"), sometimes extending to the contact with the overlying "Grezzoni". Notice that a strata-time--bound character links the pyrite deposits of southern Tuscany and the iron deposits of Isola d'Elba (Tanelli, 1983).

The morphology of the ore bodies is mostly stratiform or nearconformable lens-shaped, with subordinate discordant veins and masses. Texture is typically banded; massive beds and limited metasomatic-replacement textures after carbonatic rocks also occur.

Mineral zoning is especially evident at Monte Arsiccio, where from the bottom contact with the "Scisti di Fornovolasco" toward the roof contact with the "Grezzoni", the following sequence is observed: banded pyrite-barite; massive barite; banded barite-iron oxides.

Mineralogy consists largely of barite, haematite, magnetite and pyrite; gangue minerals are quartz, chlorite, white K-mica, calcite, siderite, dolomite and occasionally tourmaline and garnet. Pyrite and barite may be found in the country schists, as disseminations or thin levels. Minor metallic minerals include widespread galena and sphalerite, local occurrences of arsenopyrite, pyrrhotite, stibnite, bournotite, geocronite, tetrahedrite, and a number of rare or even unique Cu-Pb-Sb-As-V minerals (Checchi — Orlandi, in press).

Microscopic mineral textures include: mutual replacement of the iron oxides; porphyroblastic structure of magnetite and pyrite; in the latter mineral, low--crystallinity relics are preserved within the porphyroblasts; triple joint texture of barite.

Isotopic and fluid inclusion studies

Previous isotopic studies on these deposits consits of two S-isotope analyses on pyrite from Monte Arsiccio reported by Carmignani et al. (1976; δ

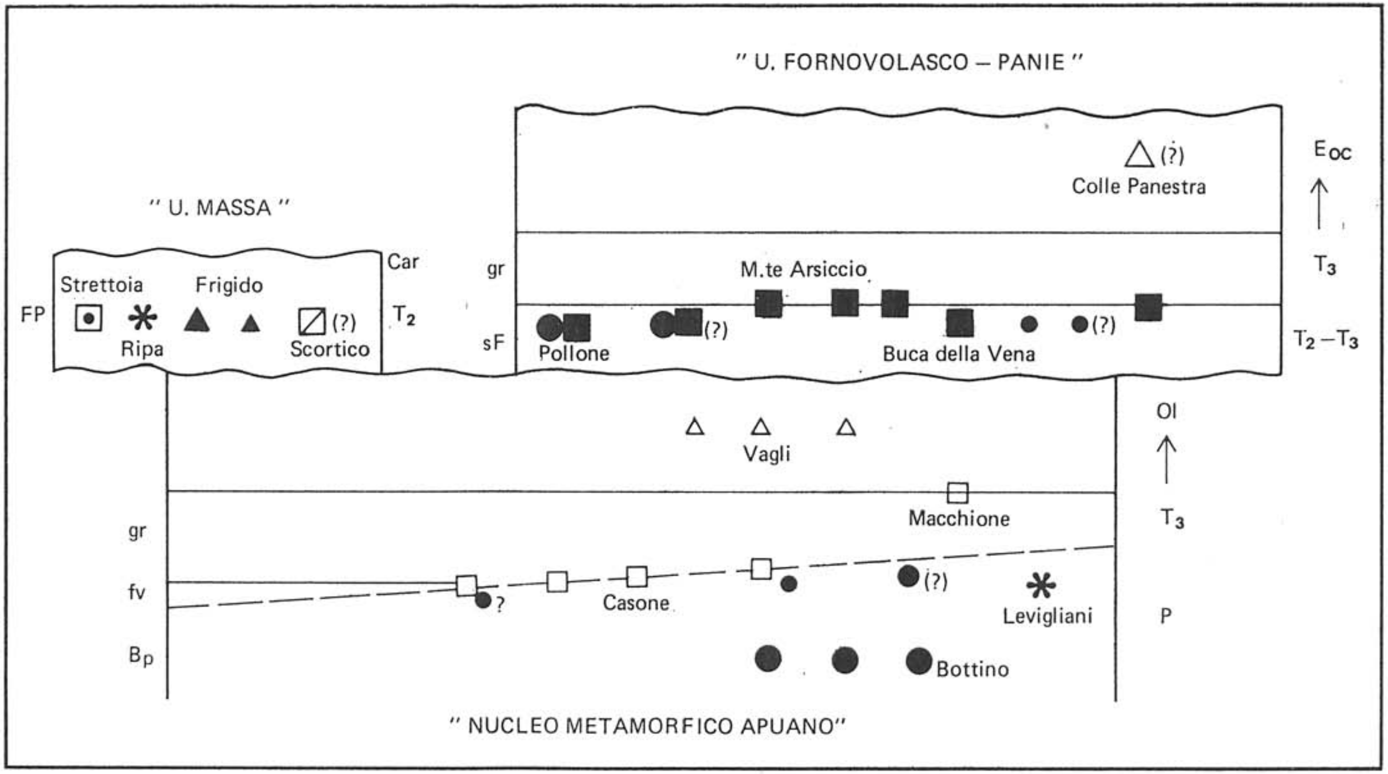

Fig. 2. Scheme of geological setting of mineralizations of the Apuane Alps.

Explanations: Symbols as in Fig. 1. Solid vawy lines = tectonic contacts; solid straight lines = conformable stratigraphic contacts; dashed straight lines = disconformities.

Bp = Paleozoic basement; fV = Formazione di Vinca; FP = Monte Folgorito and Punta Bianca groups (Middle Triassic); sF = Scisti di Fornovolasco (Ladinian–Carnian); gr = Grezzoni (Norian–Rhaetian).

P = Paleozoic; T_2 = Middle Triassic; T_3 = Upper Triassic; Car = Carnian; Eoc = Eocene; Ol = Oligocene.

$^{34}S_{CDT} = -15.6$ and 16.7 per mille), and by the Sr-isotope determinations of Barbieri et al. (1982) on barites from Pollone, Monte Arsiccio and Buca della Vena. The latter authors suggested that the source of Sr (and possibly of Ba) cannot be seawater, or marine sediments, but it is to be sought in the nearby terrigenous formations.

Sulphur isotope analyses performed during this study (Fig. 3) show a widespread of $\delta\ ^{34}S_{CDT}$ values for both sulphides (pyrite, sphalerite, galena and arsenopyrite) and barite from stratiform bodies at Pollone, Buca della Vena and Monte Arsiccio (between -29.9 and $+18.9$ per mille, and between $+6.5$ and $+30.8$ per mille, respectively). A vein mineralization at Pollone shows narrower ranges of isotopic composition ($\delta\ ^{34}S_{CDT}$ for sulphides: -3.7 to $+0.5$ per mille; for barite: $+15.8$ to $+20.8$ per mille). Barite is consistently enriched in ^{34}S with respect to coexisting pyrite. Apparent isotopic temperatures (Tab. 1) span a quite large range ($140° \pm 15°$ to $530° \pm 30°$), even for samples from the same deposit, and, therefore, they should reflect only partial equilibrium between the sulphate and the sulphide.

Reconnaissance fluid inclusion studies were carried out on barite from the vein mineralization at Pollone, and on quartz from lens-shaped pods in the country schists near the contact with the same vein.

Fluid inclusions in quartz (mostly two-phase, liquid-rich) were quite abundant. At least two generations of inclusions were recognized, the latter consisting of clearly secondary, very tiny inclusions related to healed fractures. Earlier inclusions occur as tridimensional arrays, and range from small (10—50 μm) with fairly regular elongated or hexagonal shape, to large (up to over 100 μm) with extremely irregular shape, often surrounded by aureoles of much smaller inclusions. A moderately birefringent, equant or elongate daughter mineral is sometimes present. During heating experiments, all largest inclusions were stretched or decrepitated before or just above homogenization, at temperatures around 200°—220 °C. Smaller inclusions did not undergo apparent irreversible changes, yielding reproducible homogenization temperatures between 190° and 250 °C. The daughter mineral generally dissolves slightly before final homogenization. Freezing data are quite similar for all inclusions. Salinities are around 9 % wt. NaCl equivalent, and first melting temperatures between —32° and —27 °C indicate the presence of additional components (Mg, Ca) besides Na (K)-Cl.

The largest fluid inclusions in quartz show the typical features of naturally decrepitated inclusions (Hollister et al., 1979; Pecher, 1981). They are interpreted as early metamorphic inclusions which were deformed and decrepitated during later metamorphic stages (e. g. during uplift: Hollister et al., 1979). The smaller inclusions may represent either undeformed early metamorphic inclusions, or fluids trapped in a late metamorphic stage after decrepitation of the largest inclusions. At present, we have observed no textural evidence (such as cross-cutting relationships of inclusion planes) to resolve this ambiguity.

Fluid inclusions in barite are less abundant, and mostly very small and probably secondary. A few larger (5 to 25 μm) two-phase liquid-rich inclusions occur isolated or in small groups not related to any obvious fracture plane, and may assume to be primary and therefore representative of barite-forming fluids. Microthermometric determinations on these inclusions were plaqued by

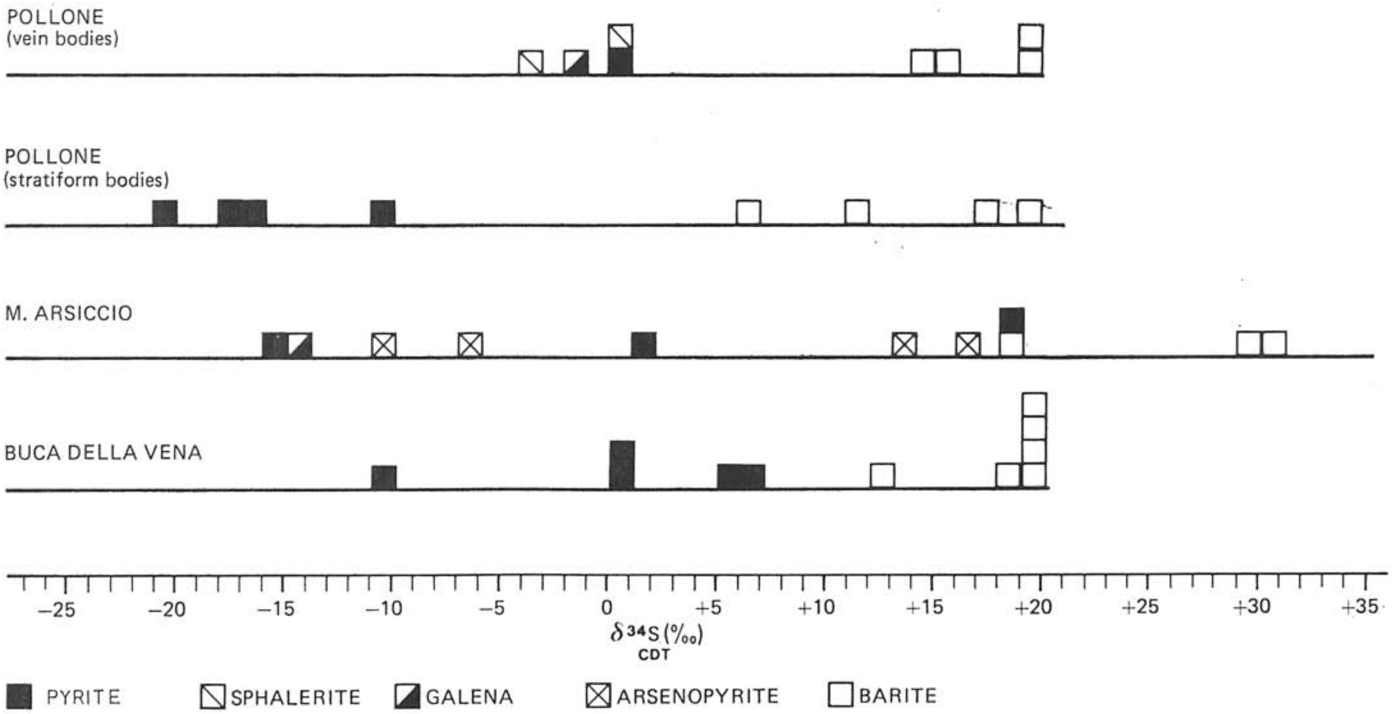

Fig. 3. Sulphur isotope analyses of barite and sulphides from the barite-iron oxide-pyrite deposits of the Apuane Alps.

a number of technical problems, such as poor optics and stretching during heating runs. An approximate estimate of the fluid characteristics indicates moderate salinities (around 10 % wt. NaCl equivalent), presence of components (Mg, Ca) other than Na (K) Cl, and homogenization temperatures in the order of 200°—250 °C.

Table 1

S-isotope fractionation and apparent isotopic temperatures for barite-pyrite pairs from barite-iron oxide-pyrite deposits of the Apuane Alps

Deposit	$\Delta^{34}S_{ba-py}$	Temperature* (°C)
Pollone	16.5	393±15
Buca della Vena	25.7	213±15
" " "	19.3	319±15
" " "	19.1	324±15
Monte Arsiccio	33.5	140±15
" "	28.9	178±15
" "	11.8	530±30

* calculated according to Ohmoto-Rye (1979)

Metallogenic discussion

Several lines of evidence lead us to suggest a pre-metamorphic, sedimentary-diagenetic origin, or at least pre-concentration, of the barite-iron oxide-pyrite ores of the Apuane Alps:

— they reveal a pronounced stratabound character, which is especially remarkbale considering the complicated tectonic structure of the region;

— the morphology of the ore bodies is quite often stratiform and conformable; the limited discordant-replacement bodies may be related to late-metamorphic hydrothermal remobilization;

— mega-, meso- and microscopic textures suggest that the ore bodies were affected by the regional Apenninic metamorphism (faulting and folding of the ore bodies; porphyroblastic growth of magnetite and pyrite, and growth structures in the latter mineral; triple joint annealing texture of barite).

In agreement with the paleogeographic reconstruction of Ciarapica—Passeri (1982), the environment of primary ore deposition may be considered as a coastal lagoonal domain (Ladinian—Carnian) transitional from the dominantly terrigenous sedimentation of the "Scisti di Fornovolasco" to the hyperhaline-evaporitic platform of the "Grezzoni". Such an environment is potentially apt to formation of sedimentary-diagenetic barite concentrations for reaction between sulphate of ultimate marine origin and waters carrying Ba^{2+} released from continental weathering (see e.g. Fuchs, 1980).

The source of Fe may have been continental as well, considering the occurrence of Fe concentrations in the "Nucleo metamorfico apuano". Precipitation of pyrite probably resulted by bacterial reduction of sulphate.

The above picture is in agreement both with Sr-isotopic data, which suggest a non-marine origin of Ba, and with S-isotope determinations. The large spread of S-isotopic compositions of both pyrite and barite, and the enrichment in ^{34}S of the latter mineral with respect to Upper Triassic marine sulphate ($\delta\ ^{34}S =$ about $+ 16$ per mille), are typical of isotopic effects of bacterial sulphate reduction in restricted basins. The few less positive $\delta\ ^{34}S$ values for barite may be explained with contributions of sulphate from partial oxidation of pyrite. On the other hand, the occurrence in the region of Middle Triassic volcanism suggests that possible contributions of sulphur (and other elements) from volcanic sources cannot be ruled out.

The present setting of the deposits was eventually determined during the following geological history, and especially by the Apenninic metamorphism (deformation, recrystallization and late-metamorphic remobilization). In particular, the more restricted range of S-isotopic compositions of both sulphides and barite in the vein mineralization at Pollone may arise from isotopic homogenization during remobilization.

The above suggested metallogenic picture requires further confirmation and elucidation of several aspects; with this aim, specific detailed studies on individual deposits are currently under way.

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