Marbles with carbonatite-like geochemical signature from variegated units of the Bohemian Massif, Czech Republic, and their geological significance

Mramory geochemicky podobné karbonatitům z pestrých jednotek Českého masivu a jejich geologický význam

(4 figs, 1 table)

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Marbles with carbonatite-like geochemical signature (MCC) were recently found at several localities of ordinary calcite marbles (OMC) in the Český Krumlov (Muckov), Vratěnín (Korolupy), Olešnice (Olešnice-Lamberk) and Vranov (Vranov) Units, respectively. The rock complexes hosting the MCC are built of dominant metapelites with locally common intercalations of quartzites, calcite and dolomite marbles, minor metabasites, graphite-rich rocks and locally scapolite- and tourmaline-bearing rocks (metaevaporites?). The mineral assemblages of the OMC and MCC are very similar and include Cal+Dol+Phl, Cal+Dol+Tr, Cal+Qtz+Phl+Na,Al-enriched amphibole (tremolite, fluortremolite, edenite, pargasite, magnesiohornblend), Cal+Dol+Tr+Di. Relatively abundant diopside in the Moldanubian marbles indicates a higher grade of metamorphism relative to the Moravicum. The MCC have variable contents of Si and Al from almost pure marbles to silicate-rich rocks with up to 31.7 wt.% SiO_2 and 4.49 wt.% Al_2O_3, high SiO_2/Al_2O_3 = 21.35–6.18 is typical. The associated OMC and MCC do not exhibit any apparent differences in the major elements, disregarding slightly lower SiO_2/Al_2O_3 ratio, slightly increased Na and K contents in the MCC, nevertheless, they are enriched in Nb ≤ 393, Mo ≤ 28, REE_{total} ≤ 124, Zr ≤ 4212 and Sr ≤ 1063 (all in ppm). The carbonatite-like geochemical signature may be caused by admixture of detritus from relatively primitive volcanic rocks or participation of the evaporitic fluids mobilized from host rocks. Presence of the MCC in lithologically similar metamorphosed volcanosedimentary sequences, currently classified into distinct geological units (Moldanubicum, Moravicum), indicates that they may represent an identical unit in the Bohemian Massif.

Key words: marble, carbonatite, geochemistry, metamorphism, variegated units, Český Krumlov Unit, Vratěnín Unit, Vranov-Olešnice Units, Czech Republic

Introduction

Carbonatite-like marbles were described from the Bližná graphite mine, the Český Krumlov Unit, Variegated Unit, Moldanubicum (Šarbach et al. 1985, Drábek et al. 1986, 1999, Veselovský et al. 1987). They are strongly enriched in the elements typomorphic for carbonatites – REE's, Y, Th, Nb, Zr, Sr and Mo (e.g., Wooley – Kempe 1989, Pell 1996, Hoernle et al. 2002). Both stable (O, C) and radiogenic (Sr, Nd) isotopic signatures point to a high proportion of the material coming from a relatively primitive source. These rocks were interpreted as marbles containing large proportion of rock detritus derived from alkaline (carbonatite-like) volcanism; however, they also may represent a metamorphosed evaporite sequence (Drábek et al. 1999). The carbonate rocks subsequently underwent polyphase metamorphism in amphibolite facies.

Marbles with carbonatite-like geochemical signature (MCC) were recently found at several localities of calcite marbles in the variegated units of the Bohemian Massif: Český Krumlov, Vratěnín, Vranov and Olešnice Unit, respectively. Their mineral assemblages, chemical composition and geological setting are discussed as well as their comparison with associated ordinary marbles (OMC) and with the carbonatite-like marble from the Bližná graphite mine.

Localities of marbles with carbonatite-like geochemical signature

Localization

1) Muckov – Český Krumlov Unit (Moldanubicum)

Abandoned underground quarries are situated about 3 km NE of the graphite deposit Bližná, 600 m W of Muckov (Fig. 1). Marble layer, up to 30 m thick and 100 m long, occurs in sillimanite-biotite and amphibole-biotite gneisses.

2) Korolupy – Vratěnín Unit (Moldanubicum)

Abandoned quarry is located about 0.5 km W of the village. Marble body, up to 30 m thick and 70 m long, is enclosed in muscovite-biotite gneiss with intercalations of quartzite and amphibolite.

3) Olešnice-Lamberk – Olešnice Unit (Moravicum)

Several abandoned quarries occur about 2 km NW of the town. Marble layers, up to 20 m thick and about 100 m long, are hosted in fine-grained biotite gneiss.

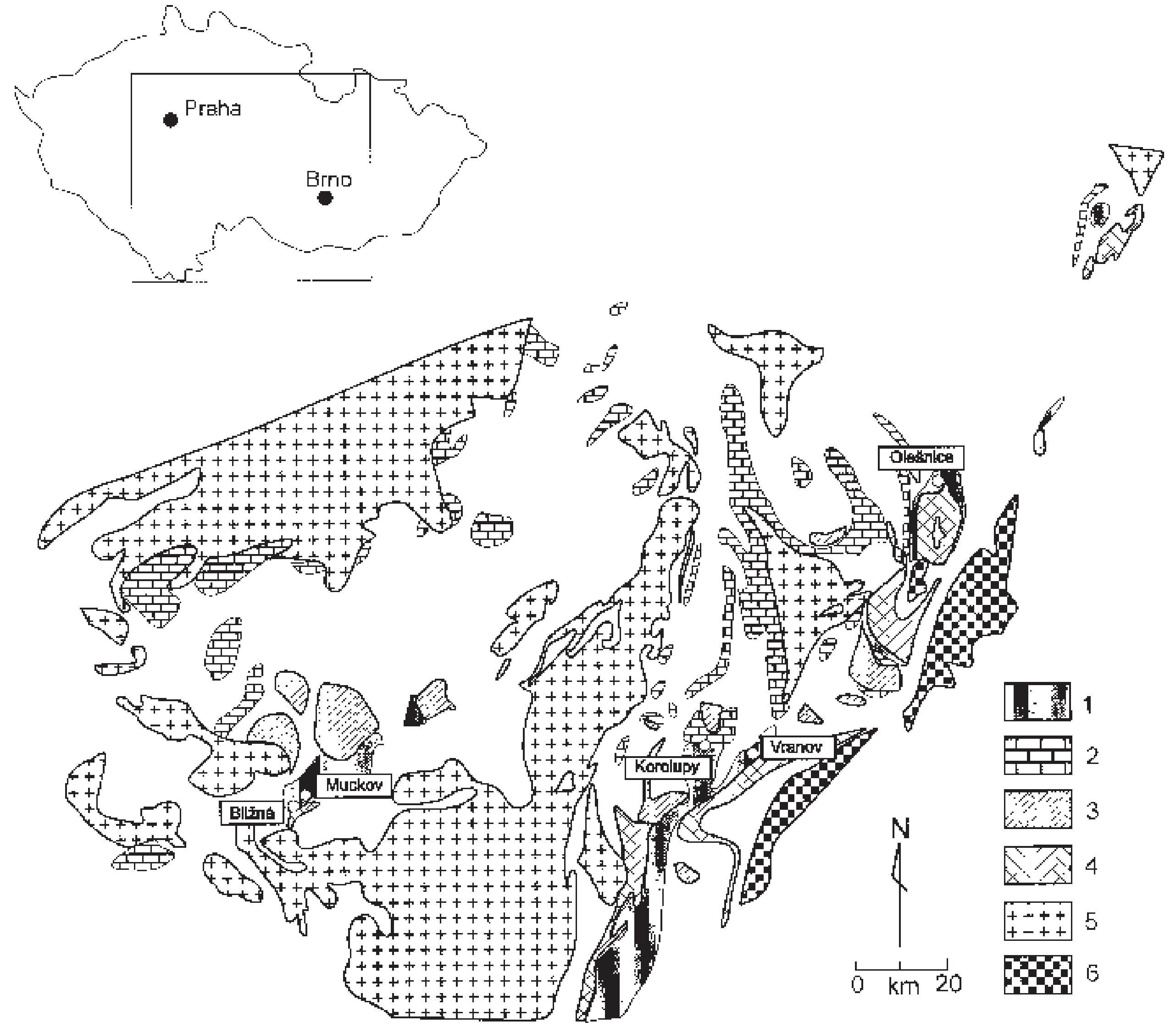

Fig. 1 Regional setting of the studied localities. 1 – tremolite marbles (including carbonatite-like marbles), 2 – marble-rich variegated units, 3 – granulites, 4 – Proterozoic orthogneisses, 5 – Variscan granitoids, 6 – Cadomian granitoids.

4) Vranov – Vranov Unit (Moravicum)

Abandoned quarries are located in the Junák valley, 1.5 km WNW of the town. Marble layer, up to 10 m thick and 200 m long, is enclosed in muscovite-biotite gneiss and in mica schist with intercalations of quartzite and amphibolite.

Geological setting

The rock complexes hosting the OMC with MCC are built of dominant metapelites (mica schists, muscovite-biotite and biotite gneisses) with locally common intercalations of quartzites, calcite and dolomite marbles, minor metabasites and graphite-rich rocks, typically closely associated with marbles (Zoubek 1946, Dudek 1960, Kadounová 1987, Kříbek 1988). Intercalations of scapolite-bearing gneiss and albite-dolomite rocks in marble-bearing rock sequences are typical in the Olešnice Unit (Sekanina 1965).

The MCC occur as rather small, isolated layers perhaps several cm to dm thick, in large marble (OMC) bodies. These bodies typically contain layers of tremolite marbles with large gray amphibole porphyroblasts. They represent a specific lithotype occurring exclusively in the variegated crystalline complexes located along the eastern margin of the Bohemian Massif such as Vratěnín, Olešnice, Vranov and Velké Vrbno Units, respectively; however, they also occur in the Český Krumlov Unit (Houzar et al. 2000).

Petrography

Český Krumlov and Vratěnín Unit (Moldanubicum)

The OMC from the Moldanubicum are generally grayish, medium- to coarse-grained and commonly silicate-poor, they only locally contain high amounts of silicates. In the locality Muckov, the simple mineral assemblage Cal+Dol+Phl ± accessory rutile (abbreviations after Kretz 1983) is typical in silicate-poor marbles, the assemblage Cal+Qtz+Phl+Na,Al-enriched amphibole (edenitic amphibole or rare fluortremolite to Na,Al-enriched fluortremolite) in silicate-rich marbles, respectively. Amphibole porphyroblasts are overgrown by diopside (Fig. 2). The MCC are white, medium-grained, calcite-rich with variable proportions of silicates (dominantly Phl). The typical mineral assemblages in Korolupy include Cal+Dol+Tr+Phl+Ttn; Na,Al-enriched amphibole (edenite) and diopside represent later assemblage, scapolite was found at contact with metabasites. The associated MCC are white to grayish, coarse-grained, calcite-rich and silicate-poor (Phl > Amp).

Olešnice and Vranov Unit (Moravicum)

The OMC from Olešnice-Lamberk is white to gray, medium- to coarse-grained, with variable amount of silicates. The mineral assemblages include Cal+Qtz+Tr+Ttn+Dol and later Na,Al-enriched amphibole (pargasitic hornblende, Fig. 3), locally accessory graphite, pyrite, plagioclase (An_{9-12}), scapolite (Me_{58}), apatite and dravite are present. The associated MCC are white to gray, medium- to coarse-grained, calcite-rich with minor Phl and Tr. The OMC from Vranov are gray, medium-grained with vari-

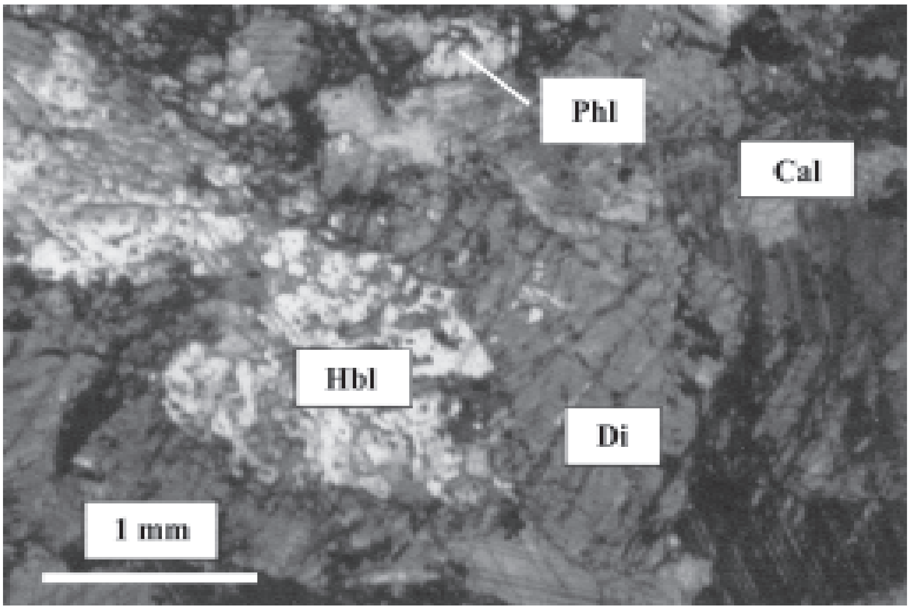

Fig. 2 Edenitic amphibole overgrown by diopside grains – Muckov, Český Krumlov Unit, Moldanubicum.

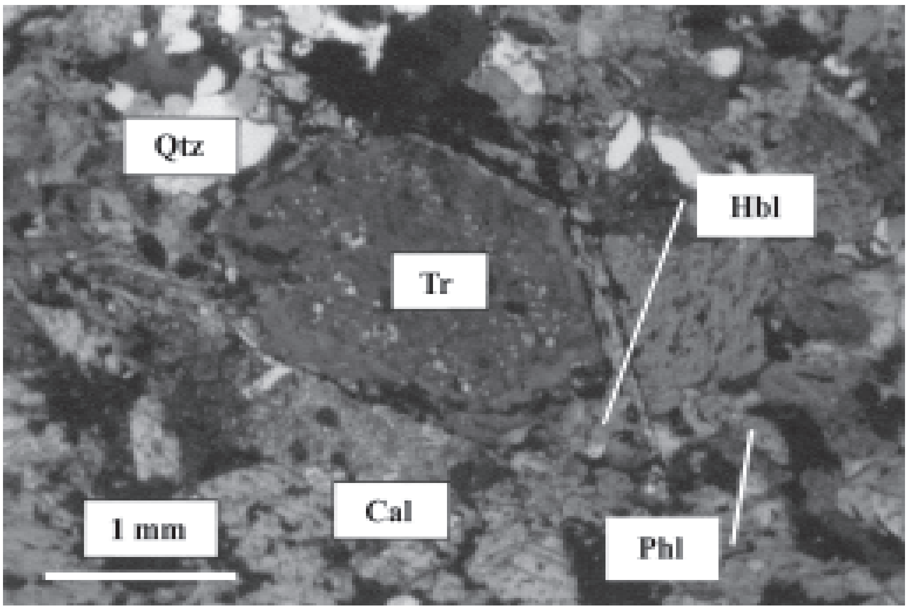

Fig. 3 Tremolite overgrown by pargasitic amphibole – Olešnice, Olešnice Unit, Moravicum.

able proportions of silicates. The mineral assemblages include Cal+Dol+Tr+Phl and Cal+Tr+Di+Phl. The associated MCC are gray, medium-grained, and calcite-rich with abundant Phl and Tr.

Generally, the MCC are highly variable in texture and appearance (medium- to coarse-grained, gray to white) and mineral assemblages (silicate-poor to phlogopite- and/or tremolite-rich). However, they do not exhibit any apparent differences from the host OMC occurring in the same bodies except lower abundance of graphite, more abundant Na,Al-enriched amphibole and namely trace-element chemistry discussed below. Nevertheless, minerals carrying the trace elements have not yet been identified.

Analytical methods

Major element concentrations were determined in the Laboratory of Chemical Methods, Institute of Rock Structure and Mechanics, AS CR, Prague, using wet chemical analysis (ing. Vojtěch Chalupský). The concentrations of Sr, Nb, Zr and Y were measured by X-ray fluorescence in the Gematest Ltd., Prague (ing. V. Štrublová). Selected samples of the marbles were analyzed by instrumental neutron activation (INAA) for the rare earth elements (REE), Rb, Ba, Th, Ta, Hf, Sc and Cr in the Radioisotope Laboratory of the Institute of Nuclear Physics, AS CR, Prague (RNDr. Jaroslav Frána, CSc.). The accuracy is better than 5% for major elements and 2–10% for trace elements. Very high concentrations of U and Th in the sample MU-1 did not enable accurate determination of the individual REE´s using the INNA.

Metamorphism of carbonate rocks

Marbles (both OMC and MCC) from all regional units underwent a polyphase metamorphism in amphibolite facies. Several stages were recognized from textural relations and mineral assemblages of marbles (Novák et al. 2002).

The metamorphism M1 is characterized by simple equilibrium assemblages: Cal+Phl+Qtz, Dol+Cal+Tr and Cal+Tr+Phl. Large porphyroblast of gray amphibole I (tremolite) with abundant inclusions of graphite, quartz, calcite and rare dolomite were very likely produced by the reaction:

(1) Dol + Qtz = Tr + Cal + CO_2

Amphibole I was partly dissolved and overgrown by colorless amphibole II (tremolite to Na,Al-enriched tremolite). Locally, narrow rims of Na,Al-enriched amphibole III (magnesiohornblend, pargasite, edenite) were found around amphibole II. The Na,Al-enriched amphibole III was likely produced by the reactions:

(2) Ab + Dol + Qtz + H_2O = CaNaAl Amp + Cal + CO_2 or

(3) Ab + Tr = CaNaAl Amp + Cal + CO_2.

The metamorphism M2 is characterized by formation of diopside as discrete grains around amphibole III or overgrowths on amphibole II. It originated according to the reaction:

(4) Tr + Qtz + Cal = Di + H_2O + CO_2.

In the OMC from the Moldanubicum, rare serpentinized forsterite (Český Krumlov Unit, Vratěnín Unit), spinel and humite minerals (Český Krumlov Unit) were exceptionally found (Šarbach 1984, Pauliš – Lukšan 1992), nevertheless, these minerals are not known from the marble localities with the MCC.

The P-T conditions for the metamorphism M1 were derived for P_{fluid} = 6 kbar at $T_{min.}$ = 550 °C for X_{CO2} = 0.4 and $T_{max.}$ = 600 °C for X_{CO2} = 0.7, perhaps up to $T_{max.}$ < 630 °C for X_{CO2} = 0.9. These P-T conditions are similar to those suggested for marbles of the Český Krumlov Unit by Čížek (1985) and to those from the Drosendorf Unit in Austria (Högelsberger 1987).

Formation of diopside-bearing assemblages in siliceous marbles during metamorphism M2 corresponds to the temperature up to T > 630–660 °C, or decreasing pressure with the temperatures similar to those given above for the M1.

The mineral assemblages in marbles are rather regularly distributed with the marble bodies, no apparent differences between OMC and MCC were observed except overgrowths of Na,Al-enriched amphibole III, which seem to be more abundant in the MCC. Generally, the presence of forsterite, abundance of diopside and very rare spinel and humite minerals in the marbles from the Český Krumlov and Vratěnín Units (Moldanubicum) exhibit higher grade of metamorphism relative to marbles of the Olešnice and Vranov Units (Moravicum).

Chemical composition of marbles

Major and minor elements

The MCC exhibit highly variable contents of Si and Al varying from almost pure marbles with ~2 and 0.2 to silicate-rich rocks with 31.7 wt.% SiO_2 and 4.49 wt.% Al_2O_3, respectively; variable but high SiO_2/Al_2O_3 ratios 21.35–6.18 are typical. Silicate-poor marbles have low amounts of MgO up to ~2 wt.%, whereas silicate-rich portions exhibit up to 9.29 wt.% MgO. Minor concentrations of Fe, Na and K were found in silicate-rich rocks (Table 1). The associated OMC and MCC do not exhibit any apparent differences in the major elements, disregarding slightly lower SiO_2/Al_2O_3 ratio and slightly increased Na and K contents in the MCC.

Trace elements

The MCC are moderately to strongly enriched in Nb, Mo, REE, Zr and Sr (Table 1): Nb ≤ 393, Mo ≤ 28, total REE ≤ 124, Zr ≤ 4212 and Sr ≤ 1063 (all in ppm). The MCC are characterized by the assemblage of elements Nb+Mo (±Zr+REE). Behavior of Zr, Mo, Y, U, Th, REE is quite irregular. We found only sporadically apparent correlations between the individual elements, e.g., positive correlations Nb-Ca a Zr-Si, Mo-Nb and negative correlations Zr-Nb and U-Nb, but, some Nb,Zr-enriched marbles have the concentrations of Mo below the detection limit. Figure 4 illustrates composition of all marbles normalized on PML = Phanerozoic marine limestones (Condie et al. 1991). Slightly elevated contents of Nb (up to 8 ppm) and Y (up to 15 ppm) were also observed in dolomite marble from Šléglov, Velké Vrbno Unit, Silesicum (unpubl. data of the authors). The concentrations of the trace elements in associated OMC are very low and comparable to the ordinary marbles elsewhere in the studied regional units.

Isotopic compositions of C and O

The $\delta^{13}C$ values in the studied MCC range between 1.92 and –3.44 ‰ PDB and $\delta^{18}O$ between –6.87 and –12.66 ‰ PDB (23.83 and 19.10 ‰ SMOW). Compared to the carbonatite-like marbles from Bližná (Drábek et al. 1999), REE-rich graphite-free marble from Muckov is close to ordinary marbles or to so-called transitional marbles as defined by Drábek et al. (1999) as well as the MCC from Vranov, Olešnice and Korolupy. Differences in isotopic compositions are likely related to variable concentrations of graphite and silicates (cf. Nabelek 1991).

Discussion

The mineral assemblages and major element chemistry of MCC and OMC are very similar, whereas, the trace element signatures are different and variable. This suggests that at least two independent sources (and/or processes) controlled chemical composition of marbles. The dominant one, which caused major chemistry, seems to be sedimentary admixture of dominant clastic quartz and minor clay minerals reflected in high SiO_2/Al_2O_3 ratio and abundant tremolite (diopside) relative to minor phlogopite and Na,Al-enriched amphiboles. The second source may be detritus from relatively primitive volcanic rocks (cf. Drábek et al. 1999, Patočka 1991, Willimský 2001), which only in part influenced major chemistry (slightly decreased SiO_2/Al_2O_3 ratio), but controlled the trace element signature. However, why the admixture of detritus caused only minor change in major chemistry but high increase of trace elements remains an open question.

The abundance of scapolite and tourmaline in some marbles (Kadounová 1987, Kříbek 1988, Kříbek et al. 1997) and in associated metapelites (Sekanina 1965, Patočka 1991) may suggest participation of the fluids with evaporitic geochemical signature mobilized from host rock sequences (cf. Drábek et al. 1999). Scapolite-bearing rocks and albite-dolomite rocks from the Olešnice Unit (Sekanina 1965) may suggest evaporitic origin of trace elements.

Comparing the Bližná marbles (ordinary marbles, transitional marbles and carbonatite-like marbles – Drábek et al. 1999) and marbles from the studied localities, the OMC exhibit similar characteristics including major and trace chemistry and isotopes of C and O relative to the Bližná ordinary marbles. The MCC are similar to the so-called

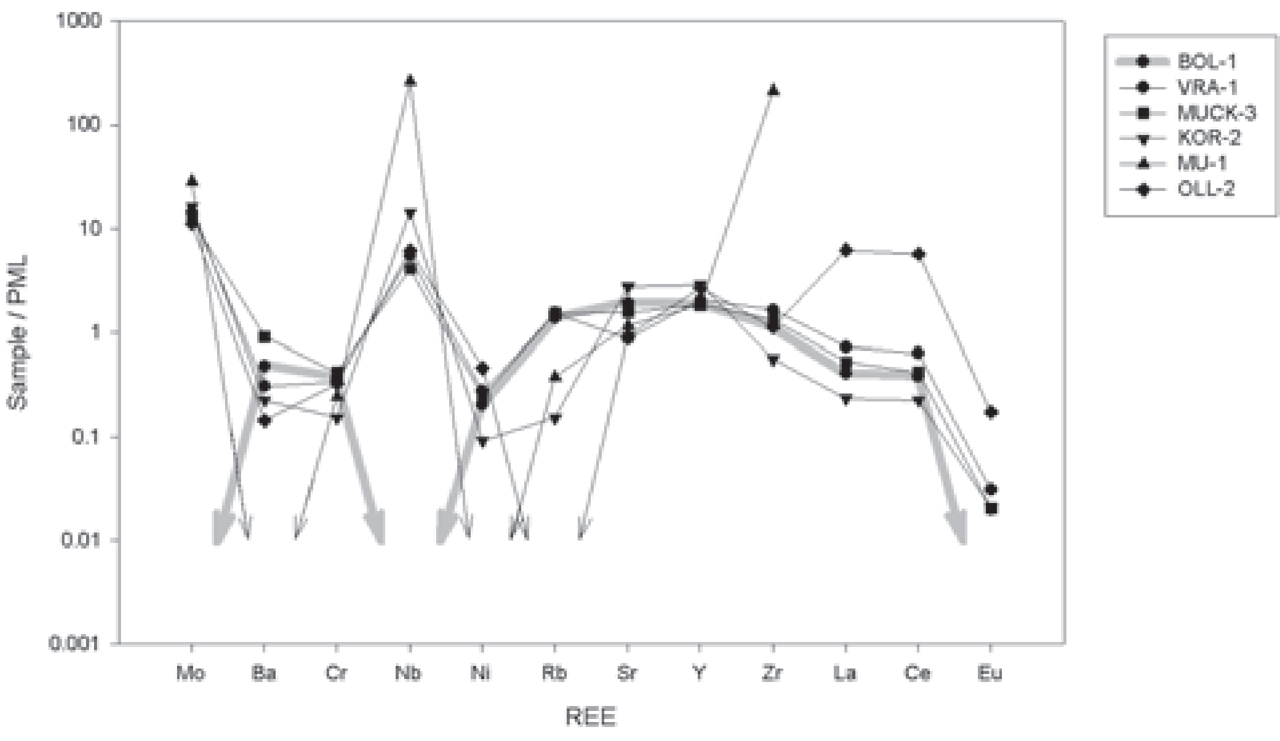

Fig. 4 Normalized element distributions in average marbles and carbonatite-like marbles. Normalization values of PML = Phanerozoic marine limestone according to Condie et al. (1991), average Mo content of continental crust from Taylor – McLennan (1985). The sample BOL-1 represents an ordinary marble.

T a b l e 1 Representative chemical compositions of carbonatite-like marbles from variegated units of the Bohemian Massif.

Unit	Český Krumlov	Český Krumlov	Vratěnín	Vranov	Olešnice
Sample	MU-1	MUCK-3	KOR-2	VRA-1	OLL-2
Locality	Muckov	Muckov	Korolupy	Vranov	Olešnice
wt %					
SiO_2	1.94	21.03	1.73	19.05	12.17
TiO_2	0.06	0.07	0.03	0.05	0.05
Al_2O_3	0.21	1.22	0.28	0.97	0.57
Fe_2O_3	0.12	0.10	0.15	0.04	0.10
FeO	0.17	0.50	0.20	0.58	0.75
MnO	0.006	0.030	0.015	0.022	0.019
MgO	2.20	8.58	1.84	9.29	4.87
CaO	52.50	40.17	52.29	39.69	41.87
Na_2O	0.02	0.23	0.28	0.07	0.40
K_2O	0.17	0.64	0.05	0.51	0.06
P_2O_5	0.05	0.03	0.03	0.05	0.03
H_2O^-	0.10	0.08	0.09	0.14	0.06
L.O.I.	41.61	26.62	42.42	28.89	38.27
Total	99.156	99.3	99.405	99.352	99.219
F	0.11	0.39	0.07	0.32	0.04
Cl	0.01	0.01	0.02	0.08	0.03
ppm					
As	b.d.	0.09	0.24	0.14	0.15
Ba	b.d.	75.32	18.29	25.17	12.21
Co	0.34	1.14	0.82	1.53	1.00
Cr	3.65	6.14	2.23	5.01	4.76
Cs	0.40	1.65	0.25	1.81	0.07
Cu	18	16	26	16	18
Hf	0.08	0.26	0.10	0.27	0.20
Mo	28	12	16	14	11
Nb	393	6	21	8	9
Ni	b.d.	3.65	1.35	4.10	6.73
Rb	7.39	29.74	3.05	29.70	b.d.
Sb	0.25	b.d.	0.02	b.d.	0.03
Sc	0.29	1.06	0.43	1.32	1.33
Sr	455	606	1063	341	363
Ta	58.81	0.08	0.30	0.05	0.06
Th	10.71	0.78	0.33	0.74	0.42
U	461.06	0.99	0.28	0.45	0.14
Y	9	9	14	10	13
Zn	b.d.	9	5	28	6
Zr	4214	26	11	32	22
La		2.53	1.13	3.55	30.18
Ce		4.08	2.20	6.22	55.46
Pr		b.d.	b.d.	b.d.	b.d.
Nd	123.07	2.85	1.06	3.00	23.67
Sm		0.36	0.20	0.52	3.52
Eu	0.21	0.09	0.04	0.13	0.83
Gd		b.d.	0.28	0.58	2.80
Tb	0.24	0.04	0.02	0.06	0.35
Dy		b.d.	b.d.	b.d.	b.d.
Ho		0.12	0.08	0.12	0.33
Er		b.d.	b.d.	b.d.	b.d.
Tm		0.02	0.02	0.03	0.09
Yb	0.54	0.14	0.08	0.21	0.47
Lu	0.08	0.02	0.01	0.03	0.06
Total REE	124.14	10.25	5.14	14.45	117.76
(La+Ce)/(Yb+Lu)		40.90	36.90	40.90	163.10
Cr/Th	0.34	7.87	6.76	6.77	11.33
$\delta^{13}C$ (‰ PDB)	1.92	–0.89	0.68	–3.44	–2.77
$\delta^{18}O$ (‰ PDB)	–6.78	–7.26	–12.66	–11.54	–10.80
$\delta^{18}O$ (‰ SMOW)	23.83	23.43	17.86	19.01	19.78

b.d. = below detection

transitional marbles from Bližná in trace chemistry and concentrations of Nb, Mo, Zr and REE. The highest concentrations of the relevant elements found in the MCC from Muckov, situated about 3 km of Bližná, are lower than those in carbonatite-like marbles. However, isotopic compositions of O and C in the MCC from locality Muckov are comparable to the ordinary marbles in Bližná. Generally, the MCC from studied localities exhibit geochemical features very similar to those of the Bližná carbonatite-like marble, nevertheless, the concentrations of relevant elements are lower (except the locality Muckov).

The MCC were exclusively found in calcite marbles from the variegated units located commonly in the eastern part of the Bohemian Massif – Vranov, Vratěnín and Olešnice Units (and perhaps also Velké Vrbno Unit in Silesicum), respectively, and in the Český Krumlov Unit (Drábek et al. 1986, 1999, Veselovský et al. 1987). The host volcanosedimentary sequences exhibit very similar lithology in all the units characterized by: (i) graphite-rich rocks closely associated with marbles, (ii) abundant tourmaline-bearing metapelites with intercalations of quartzite, calc-silicate rocks and metabasites, (iii) marbles with large porphyroblasts of gray tremolite, (iv) marbles with carbonatite-like geochemical signature, and (v) marbles with Na,Al-enriched amphiboles with edenite and pargasite substitutions (Novák et al. 2002). These rock sequences with MCC are also typically associated with Proterozoic orthogneisses (Fiala et al. 1995, Klöztli et al. 1999). Consequently, it seems that these lithologically very similar sequences, currently classified into distinct geological units (Moldanubicum, Moravicum), may represent originally an identical unit (cf. Finger – Steyrer 1995, Fritz 1996). They are located between principal geological units – Silesicum and Lugicum in the north or Moravicum (Brunovistulicum) and Moldanubicum in the south, and between Gföhl Unit and Proterozoic Světlík orthogneiss in southern Bohemia. This idea is also supported by the study of marbles in the eastern part of the Bohemian Massif and namely tremolite-bearing marbles (see Houzar – Novák 1995, Houzar et al. 2000).

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Mramory geochemicky podobné karbonatitům z pestrých jednotek Českého masivu a jejich geologický význam

V některých pestrých jednotkách jihovýchodního okraje Českého masivu se vzácně vyskytují metamorfované karbonátové horniny s anomálním chemickým složením připomínajícím karbonatity (MCC). Jsou představovány polohami mramorů se zvýšeným obsahem Nb a Mo, místy také Zr, REE, U, Th a Y, které se vyskytují v tělesech mramorů s normálním chemickým složením. Zdrojem stopových prvků byla nejspíše příměs materiálu z primitivních vulkanických hornin (alkalické bazalty a/nebo karbonatity) nebo fluida derivovaná z okolních metaevaporitů. Mineralogicky jde o převážně kalcitické mramory s kolísajícím obsahem silikátů – tremolitu, ale i edenitického nebo pargasitického amfibolu, dále flogopitu, diopsidu, skapolitu a plagioklasu; s akcesorickým pyritem, grafitem, titanitem, apatitem a turmalínem. Nejtypičtějším mramorem tohoto typu je již dříve popsaný „karbonatitoid“ z Bližné (Veselovský et al 1988, Drábek et al. 1999) a z Muckova, kterým se svým složením blíží MCC z Korolup, Vranova a Olešnice-Lamberka. Nálezy MCC v litologicky podobných metamorfovaných sekvencích, zařazovaných obvykle k různým geologickým jednotkám (moldanubikum, moravikum, silesikum), naznačují, že českokrumlovská pestrá skupina moldanubika, litologicky velmi podobná olešnická a vranovská jednotka v moraviku a pravděpodobně i velkovrbenská jednotka v silesiku mohou reprezentovat identickou jednotku v Českém masivu (srovnej např. Finger – Steyrer 1995 a Fritz 1996).