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THEMATIC ISSUE

Mineralogical and geochemical (stable C and O isotopes) variability of marbles from the Moldanubian Zone (Bohemian Massif, Czech Republic): implications for provenance studies

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Abstract Metacarbonates of the Moldanubian Zone (Bohemian Massif, Czech Republic) were studied to obtain qualitative and quantitative mineralogical-petrographic as well as stable isotopic data for the purpose of stone provenance studies, potentially applicable in material research studies of cultural heritage artefacts. Twenty-six samples from twelve different historical quarries, as well as two samples from historical artefacts, were analysed by both mineralogical-petrographic and geochemical methods including: polarizing microscopy, cathodoluminescence, scanning electron microscopy with microanalysis, petrographic image analysis, powder X-ray diffraction, and isotope ratio mass spectrometry. The petrographic characteristics allowed for the discrimination of groups of (1) calcitic marbles, (2) dolomitic marbles, and (3) carbonate–silicate rocks. These groups exhibit characteristic features such as (1) the presence/abundance of major rock-forming minerals, (2) grain geometric characteristics (specifically, mean carbonate grain size and index of grain size homogeneity), and (3) the presence of specific accessory phases. The content of non-carbonate minerals, some rock fabric parameters, as well as the carbon and oxygen isotope data exhibited significant variability, even within a single quarry in the case of some impure marbles and carbonate–silicate rocks. Although the carbon and oxygen isotopic ranges displayed overlaps among the quarries studied, the isotopic signatures throughout the Moldanubian Zone allowed for discrimination of a group of white calcitic marbles with high carbon and oxygen depletion, as well as white dolomite–calcitic marble with higher carbon isotope values when compared with other marble resources of the Bohemian Massif. A combination of the isotopic signature with detailed mineralogical-petrographic characteristics seems to provide sufficient information for discrimination of the Moldanubian marbles from one another. The provenance of the Vrchotovy Janovice artefact is very probably from the Rabí quarry, among the Moldanubian marbles. The provenance of the artefact from the Prague Klementinum was not definitively assigned; however, the Nehodiv quarry was considered its probable source locality.

Keywords Provenance potential · Spectrometry-microscopy · Isotopic-petrographic · Natural stone database · Quarry · Marble

Introduction

Since the Gothic period, numerous marble (polishable crystalline limestone) deposits have been exploited for dimension stone and other purposes (e.g. lime burning) from the Moldanubian Zone (MZ) (Bohemian Massif, Czech Republic) (Cžjžek 1851; Hanisch and Schmid 1901). Historical records provide evidence on the extraction of blocks of natural stones from several localities such as the quarries at Bohumilice, Český Krumlov, Jaroškov, Nezdice, Nehodiv, Ostružno, and Rabí (Hanisch and

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Schmid 1901; Hochstetter 1854; Kubíček 1929; Rybařík 1994). Stone varieties coming from these quarries were generally sold under the commercial trade name “šumavský mramor” (“Bohmerwaldmarmor”—i.e. Bohemian Forest marble) (Hanisch and Schmid 1901). Several important mediaeval castles, including Český Krumlov (a UNESCO world heritage site since 1992) and Rabí, were built from marbles extracted from approximately 1 km distant (Kukal et al. 2010). During the 20th century, marbles from other quarries were employed in the stone industry to be used (predominantly in Prague) for different polished products including: altars (Jaroškov and Ostružno quarries), cladding slabs (Bohumilice, Jaroškov, and Nehodiv quarries), stairs (Bohumilice and Jaroškov quarries), paving slabs (Bohumilice and Jaroškov quarries), as well as various other construction and/or decorative purposes (Nehodiv quarry) (Rybařík 1994). Blocks of Nehodiv marble were also exported to Germany (Cmuntová et al. 1970). During the 20th century, some of these marble deposits were re-examined as sources of raw materials for dimension stone production, but also as the raw material for crushed stone, inorganic fillers, and/or for lime burning (Cmuntová et al. 1970; Otová et al. 1981).

During the past two decades, extensive sampling and laboratory studies of many rock types, including limestones, have been conducted with the aim of establishing a complex database and collection (lithotheque) of local natural stones used in construction, architecture, and/or sculpture (Přikryl et al. 2002, 2004; Přikryl 2007). In the Bohemian Massif, marbles (crystalline limestones) often occur in the form of relatively small lenses, or deformed and folded tabular bodies, in complex metamorphic suites of silicate rocks (such as quartzites, amphibolites, graphitic schists, erlans). This complex geological situation is also manifested in the presence of varying amounts of non-carbonate phases. Most of the analytical studies on the provenance of marbles have been conducted by using relatively pure white marbles from the Mediterranean area, where the methodology based upon a combination of various mineralogical-petrographic and chemical criteria was originally established (e.g. Barbin et al. 1992; Herz 1992; Schmid et al. 1999; Attanasio et al. 2000; Polikreti and Maniatis 2002; Antonelli and Lazzarini 2015). However, adoption of the same analytical protocol for the study of marbles with higher and/or variable amounts of impurities remains a matter of dispute (Šťastná and Přikryl 2010).

In the present study, we have focused on several (12) classical marble localities situated within a single metamorphic terrain (MZ of the Bohemian Massif, Czech Republic). The objective of the present study was to provide detailed petrographic and stable isotopic data for potential stone provenance studies. Evaluations of the variability of the various quantitative petrographic parameters or stable isotopic data of marbles within this large metamorphic terrain present a major challenge. This study completes the previously published analytical data gathered from marbles coming from other areas of the Bohemian Massif including the Lugicum, Silesicum, Moravicum, Sedlčany-Krásná Hora metamorphic “Islet”, Kutná Hora, and Svratka crystalline complexes (Jehlička et al. 2009; Šťastná and Přikryl 2009, 2010) (Fig. 1). The methodology selected has previously been applied to some marble artefacts coming from the Czech Crown Lands (Šťastná et al. 2009, 2011, 2012). In addition to the analytical protocol used in our previous studies [the combination of qualitative and quantitative optical polarizing microscopy (PM), cathodoluminescence (CL), petrographic image analysis (PIA), and isotope ratio mass spectrometry (IRMS)], the range of analytical techniques was extended by using scanning electron microscopy with an energy-dispersive spectrometer (SEM-EDS) for a detailed characterization of the non-carbonate and accessory phases. Moreover, this technique was supplemented by powder X-ray diffraction (XRD) data of the insoluble residues. Application of the above-mentioned methods is also important from the point of view of the material restrictions from heritage objects (relatively small specimens necessary for the preparation of thin sections, and very small amounts of samples for the study of stable isotopes) and is advantageous in cases where there is a limited availability of materials. This methodology (except for the XRD of the insoluble residue) was applied to two artefacts coming from Central Bohemia (region of the MZ) with unclear provenances. Comparisons of Moldanubian marbles with the selected artefacts were aimed at supporting the reliable use of the present discrimination parameters in provenance studies.

Methodology

Microscopic techniques

Microscopic investigation of the samples consisted of conventional PM, PM coupled with CL, and SEM-EDS. For all of these techniques, uncovered polished thin sections were prepared from the rock samples. The PM and CL microscopy were carried out in the Laboratory of Microscopic Techniques (Charles University in Prague, Czech Republic). CL studies were performed using a CL 8200 MK4 cold cathodoluminescence stage (Cambridge Image Technology Ltd.) coupled with a LEICA DMLP optical microscope. The electron energy applied to the thin sections was 14–15 kV, and the beam current was operated at 320–350 µA. The cross-polarized images (XPL), as well as the luminescence colours of each

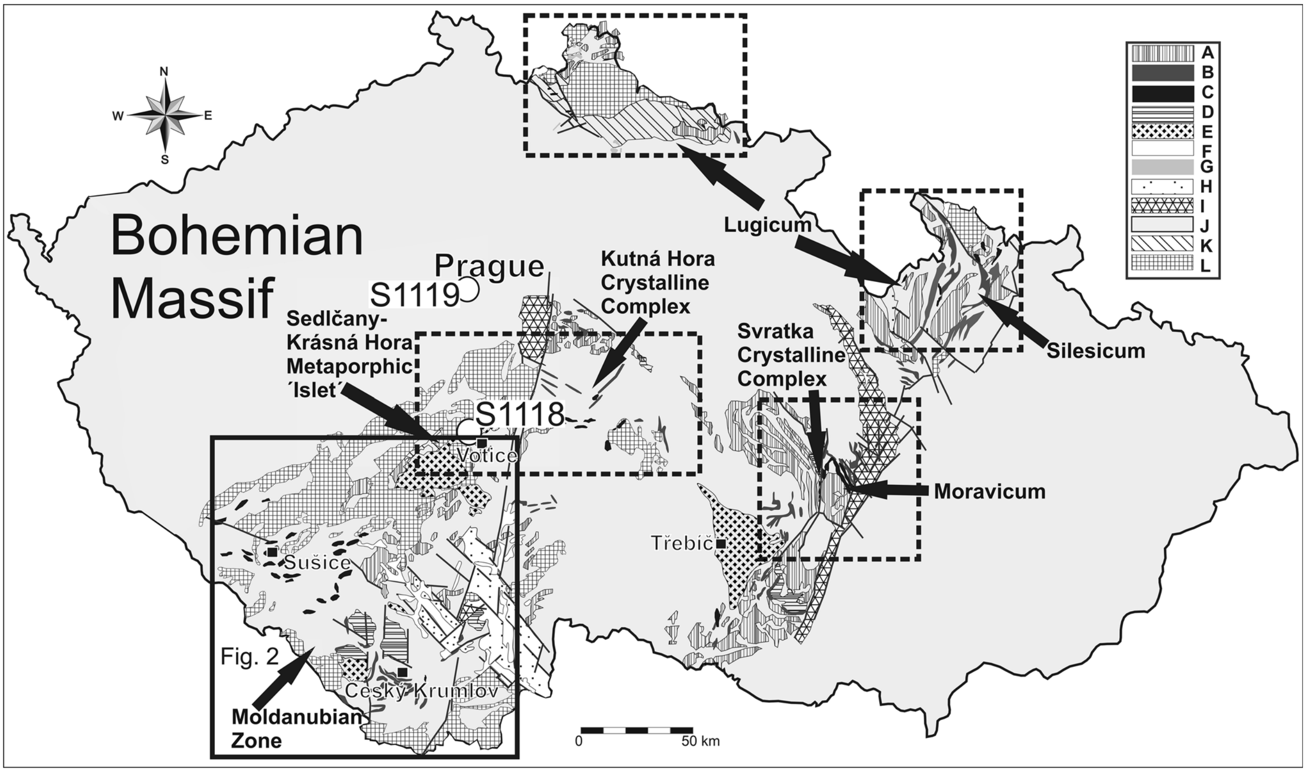


Fig. 1 A simplified geological map of areas studied in the present study (*rectangle with the solid line*) and previous studies (*rectangles with dashed lines*), and their positions in the Bohemian Massif (Czech Republic): *A* orthogneisses (Proterozoic–Palaeozoic); *B* amphibolites (Proterozoic–Palaeozoic); *C* crystalline limestones (Proterozoic–Palaeozoic); *D* granulites (Proterozoic–Palaeozoic); *E* durbachites (Palaeozoic); *F* sands, sandstones, clays, claystones (Neogene and Palaeogene); *G* volcanic rocks (Neogene and Palaeogene); *H* sandstones, marlstones, limestones (Cretaceous); *I* sandstones, siltstones, conglomerates, and other sedimentary rocks (Permian); *J* paragneisses, phyllites, granulites, migmatites, eclogites, and other metamorphic rocks (Proterozoic–Palaeozoic); *K* weakly metamorphosed metasediments (Palaeozoic); *L* granites (Variscan and Pre-variscan). The legend of the geological map is only valid for the areas within the rectangles. *Rectangle with the solid line* refers to the location of more detailed geological map for the Sušice-Votice and Český Krumlov areas (Fig. 2). Positions of the studied marble artefacts: *S1118* Vrchotovy Janovice Castle, *S1119* Prague Klementinum. Modified and completed after Šťastná et al. (2011)

lithotype, were photographed with a Canon digital camera. SEM-EDS was conducted at the Laboratory of Electron Microscopy and Microanalysis (Charles University in Prague). The thin sections were coated in a carbon atmosphere. The measurements were realized on a Tescan Vega instrument, with an energy-dispersive analytical system (Oxford Instruments LINK ISIS 300) under the following conditions: 0.8 nA (for back scattered electrons—BSE, EDS); 120 s counting time; and a 15 kV accelerating voltage. A 53 Minerals Standard Set #02753-AB (SPI Supplies) was used for the standard quantitative calibration.

Petrographic image analysis

PIA was implemented by using microphotographs obtained by PM and CL, following a procedure described in detail elsewhere (Přikryl 2001, 2006). The PIA procedure consisted of five successive steps:

1. Image acquisition (microphotographs of thin sections),
2. Outlining of the boundaries of individual mineral grains,
3. Recognition and verification of individual mineral phases,
4. Image measurement with relevant image analysing software (in our case using SigmaScan®Pro version 5.0.0 by SPSS Inc. 2016),
5. Data analysis.

Step (2) in this case was executed manually by an experienced operator—geologist/petrologist, because the fully automated image analysis had a limited capability for the recognition of individual grains.

About 200 carbonate grains were analysed for each thin section. From the basic geometric characteristics obtained during the measurement of individual grains (area A_i, length of major and minor axes, length of the perimeter L_P), the grain shape characteristics (indicated in Table 1) were computed.

Table 1 Definitions of the microfabric parameters, measured and calculated, using the petrographic image analysis (modified after Přikryl 2006)

Fabric parameter		Equation	Unit	Definition
Equivalent diameter (ED)		$\mathrm{ED} = \sqrt{\frac{4A_i}{\pi}}$	mm	Diameter of a circle occupying the same area as measured object (Petruk 1986; Přikryl 2006)
Aspect ratio (AR)		$\mathrm{AR} = \frac{D_{i(\max)}}{D_{i(\min)}}$	–	Elongation (ellipticity) of grains based on the ratio of major and minor axis lengths (Přikryl 2006)
Compactness	Area-perimeter ratio (APR)	$\mathrm{APR} = \frac{4\sqrt{A_i}}{L_p}$	–	2D shape compactness from area-perimeter ratios (Bogaert et al. 2000)
	Shape factor (SF)	$\mathrm{SF} = \frac{4 \cdot \pi \cdot A_i}{L_P^2}$	–	Roundness of grain expressed from shape (also called form) factor as a deviation of the shape of the studied object from a circle (Howarth and Rowlands 1987)
Index of grain size homogeneity (*t*)		$t = \frac{A_{\mathrm{avg}}}{\sqrt{\sum \left(A_i - A_{\mathrm{avg}}\right)^2}}$	–	Rock fabric parameter allowing distinction between homogranular and heterogranular fabrics (Dreyer 1973)

Insoluble residues

Study of the non-carbonate fraction is common in the case of sedimentary limestones, mostly for the purposes of petrogenetic studies (Flügel 2004; Critelli et al. 2007; Amendola et al. 2016), or for the evaluation of the hydraulicity of limestones employed as a raw material for natural hydraulic lime (Kozlovcev and Přikryl 2015). In the present study, the non-carbonate fraction was obtained as the so-called insoluble residue (IR) by treatment of crushed samples (6 g of each type) with 1 M HCl solution for 8 h. The samples were then centrifuged, washed in distilled water, filtered through a filter paper on a laboratory suction device, and dried to a constant weight. The weight loss was used for the determination of the total content of the non-carbonate fraction.

The insoluble residues obtained were then analysed by powder XRD using a PANalytical X'Pert Pro diffractometer equipped with a monochromator (PANalytical Co.) with X'Celerator multichannel detectors (Laboratory of XRD, Institute of Geochemistry, Mineralogy and Minerals Resources, Faculty of Science, Charles University in Prague). Measurement conditions were the following: Cu cathode α, 40 kV, 30 mA, measuring step 0.05°/200 s, angle 2.99–70° 2Θ. The resulting diffraction patterns were processed and evaluated by using X'Pert High Score 1.0d software and PDF-2 database (JCPDS-ICDD).

C and O stable isotopes

The isotopic composition of the carbonates (carbon and oxygen) was studied by mass spectrometry with a Finningan MAT 251 instrument. Extraction of CO_2 from carbonates was performed using the conventional reaction with 100% H_3PO_4 at 25°C under vacuum (methodology according to McCrea 1950). Samples for analysis were removed from macroscopically dominant portions of the metacarbonates (i.e. carbonate groundmass); secondary veins were not included. The overall analytical precision was ±0.1‰ for both carbon and oxygen isotope determination. Results were expressed in the usual δ notation relative to the international Pee Dee Belemnite (PDB) standard.

Samples and their geological setting

The examined marbles were collected from abandoned quarries located in the MZ, which constitutes a major crystalline unit of the Bohemian Massif, Czech Republic (Figs. 1, 2). The Bohemian Massif extends through Austria, Czech Republic, Germany, and Poland and is bounded by the MZ to the west, the Saxothuringicum to the north-west, the Lugicum to the north-east, and the Moravo-Silesicum to the south-east (e.g. Dallmeyer et al. 1995). The MZ is interpreted as a root zone of the Variscan belt, resulting from the collision of Gondwana with Laurasia (e.g. Schulmann et al. 2009). The MZ is internally divided into the following principal units: Monotonous Group, Varied Group, and Gföhl Unit (e.g. Fiala et al. 1995; Linnemann et al. 2008) (Fig. 2).

Marble layers are deposited along with calc–silicate rocks, amphibolites, orthogneisses, graphite gneisses, and quartzites included in the Varied Group, where the dominant rocks form biotite–sillimanite paragneisses (Chlupáč et al. 2002). According to recent studies, the age of the metasediments of the Varied Group has been suggested as Early to Mid-Palaeozoic (e.g. Pacltová 1994; Janoušek et al. 2008; Košler et al. 2014; Drábek and Stein 2015). Rocks of the Varied Group were affected by regional metamorphism in the amphibolite-granulite facies (~700–800°C, ~8–11 kbar), with later retrogression

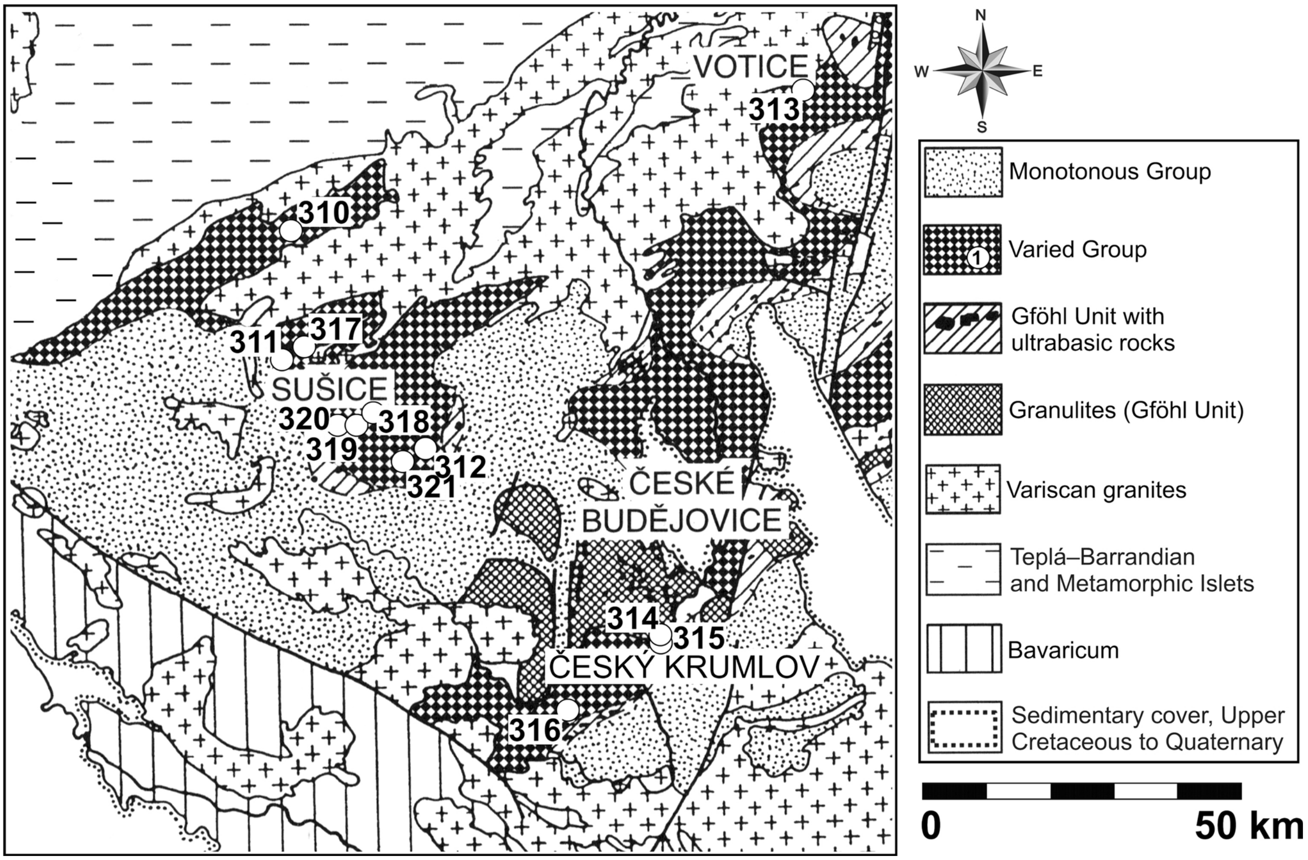


Fig. 2 A simplified geological map (adopted from Chlupáč et al. 2002) of the Sušice-Votice and Český Krumlov areas within the Moldanubian Zone, including the positions of the studied metacarbonate quarries: *310* Nehodiv, *311* Rabí, *312* Bohumilice, *313* Votice, *314* Vyšný, *315* Český Krumlov, *316* Bližná, *317* Hejná, *318* Soběšice, *319* Nezdice, *320* Ostružno, *321* Jaroškov

(~500–650°C, ~6 kbar) (Petrakakis 1997). Metacarbonate occurrences of the Varied Group are mainly scattered in the following areas: Sušice-Votice (including Plánice-Kasejovice, Sušice-Horažd'ovice, Strakonice, Nezdice-Soběšice, and Volyně-Vimperk zones) (Cmuntová et al. 1970; Žák et al. 1997), Český Krumlov (Fig. 1) (e.g. Houzar and Novák 1995, 2002; Drábek and Stein 2015), and Třebíč (e.g. Novák and Houzar 1996; Houzar and Leichmann 2003).

The internal coherence of the Varied Group (largely identical with the terms Drosendorf or Český Krumlov) is uncertain (see e.g. Suk 1983; Chlupáč 1992; Finger et al. 2007). According to the traditional lithostratigraphic concept, the Varied Group can be further subdivided into the Český Krumlov and Sušice (Sušice-Votice) Units (Fig. 2) based on distinct lithology and geographical distributions (Kodym and Suk 1958; Jenček and Vajner 1968).

The Český Krumlov Unit encompasses marbles (± dolomite) with graphitic layers and lenses (formerly exploited), rare calc–silicate rocks, amphibolites, and quartzites. For the Sušice Unit, metacarbonates (marbles, dolomites, and calc–silicate rocks) are infrequently accompanied by non-exploitable graphite; amphibolites and quartzites are almost absent (Cháb et al. 2010). A specific lithotype of carbonatite-like marbles has been described in varied sequences of different geological units (MZ and Moravo-Silesicum), which indicates that these crystalline complexes (Český Krumlov, Vratěnín, Vranov-Olešnice, Velké Vrbno Units) could originally have been an identical unit (Houzar and Novák 2002).

The studied metacarbonate samples came from quarries of the Varied Group (Sušice and Český Krumlov Units) (Fig. 2). Twenty-six specimens (rock types) were collected from 12 abandoned quarries of the MZ in the Šumava/Bohemian Forest region (Fig. 2; Table 2). The number of specimens for each locality was based on the macroscopically observable heterogeneity on the exposed quarry faces. Metacarbonates including marbles (>50 vol% of carbonate minerals) and carbonate–silicate rocks (5–50 vol% of carbonate minerals) (Rosen et al. 2007) were sampled in the form of blocks weighing from 10 to 20 kg. For each rock type, 1–5 thin sections were prepared, depending on the macroscopic homogeneity and variability of the rock fabric (foliation, grain size, and shape). These

Table 2 Locations and macroscopic descriptions of studied samples from the Moldanubian Zone and artefacts

Sample code (no. of samples)	Latitude (N)	Longitude (E)	Locality	Moldanubian Zone (Bohemian Massif)	Rock appearance description
310 (3)	49°24′34″	13°33′6″	Nehodiv (wall quarry)	Sušice Varied Unit	Greyish white with grey bands
311 (3)	49°16′22″	13°36′33″	Rabí (wall quarry)	Sušice Varied Unit	Greyish to yellowish white, fragmenting, massive or banded
312 (5)	49°5′30″	13°47′43″	Bohumilice (wall outcrops of the railway cutting)	Sušice Varied Unit	Greyish white to greenish grey with green and brown bands
313 (1)	49°38′25″	14°38′58″	Votice (wall quarry)	Sušice Varied Unit	Dark grey
314 (2)	48°49′55″	14°17′54″	Vyšný (wall quarry)	Český Krumlov Varied Unit	Greyish white to dark grey, massive or banded
315 (1)	48°49′1″	14°18′22″	Český Krumlov (wall quarry)	Český Krumlov Varied Unit	Grey with dark grey/black bands
316 (2)	48°43′49″	14°6′25″	Bližná (wall quarry)	Český Krumlov Varied Unit	Grey, massive or banded
317 (2)	49°17′26″	13°40′36″	Hejná (wall quarry)	Sušice Varied Unit	Greyish white, with yellowish green veins or brown bands
318 (2)	49°12′19″	13°40′4″	Soběšice (small shaft quarry)	Sušice Varied Unit	Greyish white
319 (3)	49°10′50″	13°36′36″	Nezdice (wall quarry)	Sušice Varied Unit	Greyish or bluish white with brown veins or white needles
320 (1)	49°10′16″	13°35′23″	Ostružno (shaft quarry)	Sušice Varied Unit	Greyish white, translucent
321 (1)	49°6′44″	13°40′35″	Jaroškov (filled and recultivated quarry)	Sušice Varied Unit	White with brown stains

Artefact code (no. of samples)	Latitude (N)	Longitude (E)	Locality	Artefact appearance description
S1118 (1)	49°40′13″	14°34′67″	Vrchotovy Janovice	Sculpture of monkey; greyish to yellowish white; weathered; dark green to grey minerals of cm-scale and minor grey bands
S1119 (1)	50°05′12″	14°25′1″	Prague	Sculpture of cupid; greyish white; weathered; minor grey bands

thin sections were oriented perpendicular to the foliation, if present. For the remaining rocks, the orientation for the cuts was random. Most of the studied marbles exhibited a light-to-medium grey colour with common variations of various hues, thus providing a cm-scale banded appearance (Fig. 3; Table 2). Pure white or multi-coloured marbles made up only a minor part in the studied set (Fig. 3).

Two historical marble artefacts with unknown provenances were studied: a fragment of a sculpture of a monkey (transported from unknown location and of unknown age), constituting part of the fountain from the court of Vrchotovy Janovice Castle (Fig. 4a); and a fragment of a sculpture of Cupid, part of the J. Stepling memorial (emplaced in 1780), located in the outer area of the Prague Klementinum (Fig. 4b). The geographical positions of both artefacts are depicted in Fig. 1. The marble from the monkey sculpture was greyish to yellowish white, with medium-to-coarse carbonate grain size and macroscopically visible non-carbonate minerals (Table 2). The sculpture of Cupid displayed a greyish white colour with weak banding, and carbonates with various medium-to-coarse grain sizes on macroscopic observation (Table 2).

Results

Petrographic characteristics

Basic subdivision of studied materials

The mineralogical-petrographic characteristics of the studied marbles and carbonate–silicate rocks exhibited significant variability. They differed in their mineralogical composition (type of mineral phases and their content) (Table 3), as well as in the microfabric (microstructural parameters) (Table 4). Based on these characteristics, the entire set of studied samples can be subdivided into the following groups: (1) very fine- to medium-grained calcitic marbles, (2) fine-grained marbles containing more than 10 vol% of dolomite, and (3) fine-grained carbonate–silicate rocks (i.e. rock with carbonate content below 50 vol%).

Calcitic marbles

Very fine- to medium-grained calcitic marbles were composed of dominant calcite and variable amounts of non-

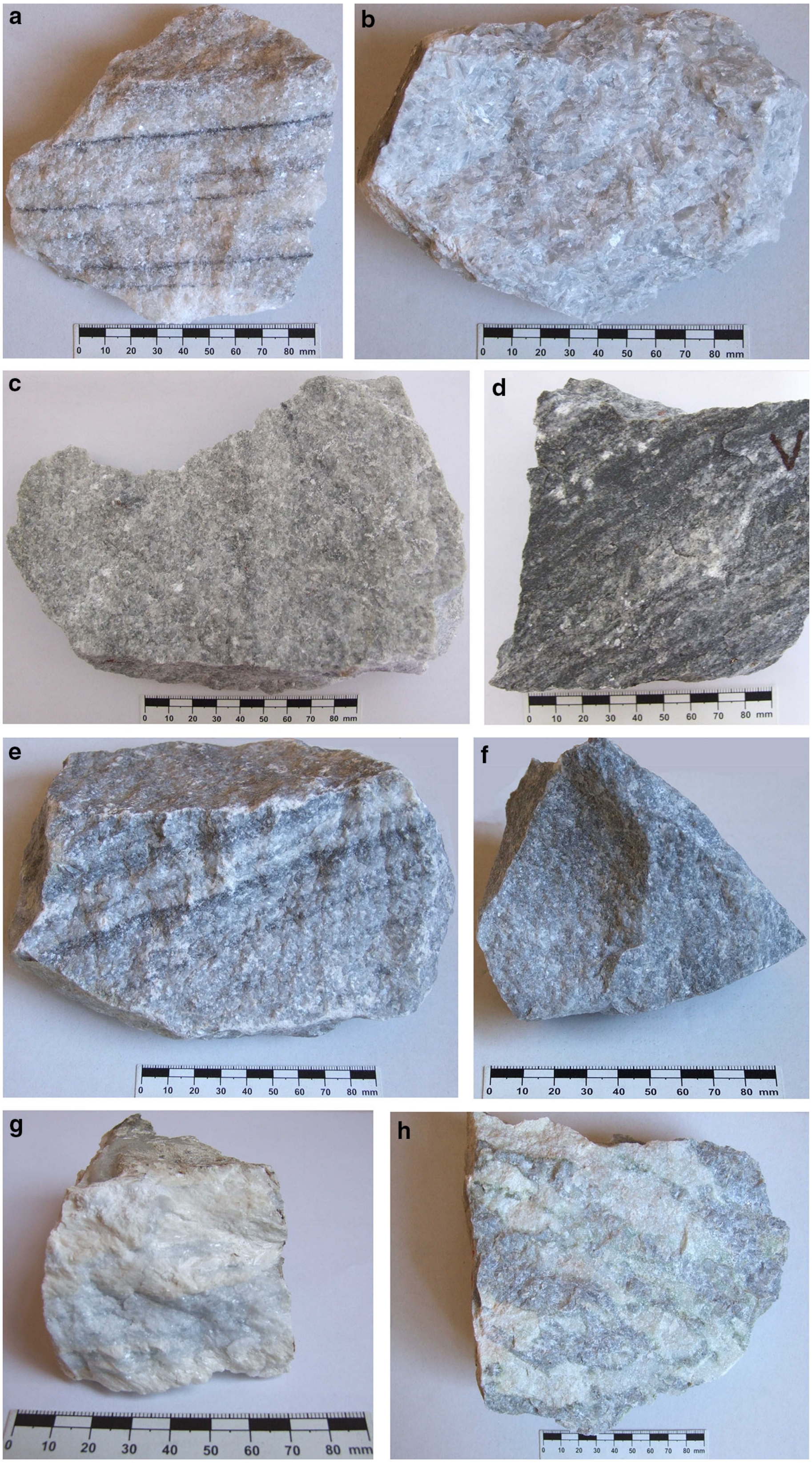


Fig. 3 Macroscopic photographs of selected metacarbonate samples from quarries of the Moldanubian Zone. **a** Nehodiv (310C), **b** Rabí (311A), **c** Vyšný (314A), **d** Vyšný (314B), **e** Bližná (316A), **f** Bližná (316B), **g** Nezdice (319C), **h** Bohumilice (312C)

Fig. 4 Macroscopic photographs of the two marble artefacts. **a** Sculpture of a monkey (Vrchotovy Janovice Castle) (S1118), **b** sculpture of Cupid (Prague Klementinum) (S1119)

carbonate phases; with silicate minerals, sulphides, and oxides dominant (Fig. 5; Table 3). Additionally, graphite was present, forming the dark grey colour of some of the calcitic marbles (marbles from the Votice, Vyšný, and Český Krumlov quarries). The median IR content commonly ranged from 2 to 9 wt%, with the exception of the impure calcitic marbles from the Vyšný and Votice quarries where IR reached 30 and 33 wt%, respectively (Table 3).

Some non-carbonate and accessory minerals were clearly identified using CL analysis. Scapolite with blue CL was present in the calcitic marble from the Nehodiv quarry (Fig. 5a–c). Clinozoisite with intense green CL was detected in calcitic marbles from the Rabí (Fig. 5d–f) and Hejná quarries. One type of calcitic marble from the Nezdice quarry (sample No. 319C) was characterized by macroscopically visible wollastonite, which exhibited very intense yellow-green CL in the optical microscope (Fig. 5g–i). The diopside contained in the Rabí (Fig. 5f) and Votice marbles exhibited a very dull CL, probably because of its higher Fe content (FeO $>$ 1 wt% evaluated by SEM-EDS). The Hejná, Nezdice, and Vyšný marbles included diopside with an intense yellow-green or blue CL, as well as with a lower Fe content (FeO $<$ 1 wt% evaluated by SEM-EDS). The Votice marble contained monoclinic pyrrhotite with a ferromagnetic characteristic (Table 3).

Concerning the fabric parameters, the calcitic marbles exhibited significant variations in their grain size (Table 4; Fig. 6). The highest values of equivalent diameter (ED) were detected in the calcitic marbles from the Rabí and Hejná quarries. On the other hand, the calcitic marble from the studied set of rocks exhibiting the finest grain size came from the Votice quarry (Fig. 6). Fabric parameters describing the circularity and shape compactness of the carbonate grains displaying the highest aspect ratio (AR) value (and lowest values of area-perimeter ratio—APR, and shape factor—SF) of the calcitic marbles came from the Hejná quarry. The calcitic marble from the Český Krumlov quarry, with a more isometric shape of the carbonate grains, showed the lowest AR value (and highest values of APR and SF). The calcitic marbles exhibited a quasi-isotropic fabric, with no shape-preferred orientation (SPO), with the exception of the grey marbles from the Český Krumlov, Votice, and Vyšný quarries (Table 4). Marbles from the Nehodiv and Rabí quarries were predominantly structured by carbonate grains of uniform size (homeoblastic fabric, index of grain size homogeneity with t values up to 0.06). However, a heteroblastic fabric, with larger carbonate grains, occurring predominantly in very fine to fine-grained carbonate grains was found in the calcitic marbles from the Votice and Český Krumlov quarries (t value of 0.02) (Table 4). In the studied grey calcitic marbles, the marble sample from the Vyšný quarry demonstrated variability in the mineral assemblage and rock fabric within this single quarry. The various rock types from the Vyšný quarry exhibited distinctive rock fabric parameters, as well as varied mineral assemblages (Tables 3, 4). A massive type of marble from the Vyšný quarry (sample No. 314A) displayed a lower content of non-carbonate minerals (IR of 5 wt%), larger carbonate grains (ED of 0.5 mm), with a more homeoblastic fabric (t of 0.06), and shape-preferred orientation compared with the banded type of impure Vyšný marble (sample No. 314B), which was enriched in feldspars (IR of 30 wt%, ED of 0.1 mm, t of 0.03) (Tables 3, 4).

Table 3 Mineral assemblage and values (minimum-maximum, median) of insoluble residues (IR in percentage by weight, determined by treatment with 1 M HCl solution) of the studied rocks from the Moldanubian Zone and the marble samples of the artefacts

Colour	Group	Quarry (sample code)	IR (wt%)	Carbonates	Mineral assemblage
White	Calcitic marbles	Nehodiv (310A-C)	2.2–3.8, 2.9	Cal	Tr + Scp + Ms + Phl + Chl + Qz ± Ab ± Kfs ± Ttn + Ap + Zrn + Py + Gr
White		Rabí (311A-C)	2.4–10.2, 8.6	Cal	Di + Tr + Qz + Phl + Ab + Tr ± Ath ± Zo ± Chl + Zrn + Ttn + Ap + Py
White		Hejná (317A-B)	1.8–2.4, 2.1	Cal	Phl + Qz + Di + Tr + Chl + Pl(Ab$_{76}$, An$_{24}$) ± Zo + Ttn + Ap + Lm
White		Nezdice (319A-B)	1.2–5.5, 3.4	Cal	Phl + Tr + Ed + Di + Qz + Chl + Pl(Ab$_{52}$, An$_{48}$) + Ap + Py
White		Nezdice (319C)	22.8	Cal	Wo + Di + Qtz + Ap
Grey	Calcitic marbles	Votice (313)	33.0	Cal	Kfs + Di + Pl(Ab$_{17}$, An$_{79}$, Or$_{4}$) + Ms (Phl) + Po + Gr + Tr + Qz + Ap + Ttn
Grey		Vyšný (314A)	4.8	Cal + Dol	Phl + Qz + Tr + Py + Gr + Ms + Dol + Zo + Kfs + Ab + Chl + Ap
Grey		Vyšný (314B)	29.7	Cal	An + Kfs + Qz + Tr + Py + Gr + Ms + Phl + Di + Ttn + Tur
Grey		Český Krumlov (315)	2.1	Cal	Tr + Py + Gr + Ms(Phl) + Qz + Di + Kfs + Chl
White	Dolomite–calcitic marbles	Soběšice (318A-B)	0.9–1.2, 1.1	Cal + Dol	Tr + Qz + Phl ± Ath + Tlc + Chl + Ap ± Py ± Hem
White		Ostružno (320)	10.1	Cal + Dol	Tr + Qz + Di + Ed + Chl + Ap
White	Calcite–dolomitic marbles	Bohumilice (312A-B, D)	3.5–12.6, 8.1	Dol + Cal	Phl + Fo + Chl + Di + Atg + Ap ± Qz ± Sp ± Zrn
White	Dolomitic marbles	Jaroškov (321)	1.4	Dol + Cal	Chl + Phl + Ms + Ap + Py
Grey	Dolomite–calcitic marbles	Bližná (316A)	6.4	Cal + Dol	Phl + Tr + Di + Qz + Ap + Py + Gr
Grey	Calcite–dolomitic marbles	Bližná (316B)	10.4	Dol + Cal	Fo + Phl + Di + Lz + Tr + Ap + Py + Gr
Coloured	Carbonate–silicate rock	Bohumilice (312C, E)	60.2–67.3, 63.75	Cal + Dol	Di + Fo + Atg + Phl + Chl + Sp ± Qz ± Kfs ± Tr + Ap

Colour	Group	Artefact (sample code)	Age	Carbonates	Mineral assemblage
White	Calcitic marbles	Sculpture of monkey (S1118)	–	Cal	Qz + Pl(Ab$_{56}$, An$_{42}$, Or$_{2}$) + Ab Ms + Di + Ttn + Ap
White	Calcitic marbles	Sculpture of cupid (S1119)	1780	Cal	Qz + Di + Ap

Mineral phases (abbreviations after Whitney and Evans 2010): *Ab* albite, *An* anortite, *Ap* apatite, *Atg* antigorite, *Ath* anthophyllite, *Cal* calcite, *Chl* chlorite, *Di* diopside, *Dol* dolomite, *Ed* edenite, *Fo* forsterite, *Gr* graphite, *Hem* haematite, *Kfs* K-feldspar, *Lm* limonite, *Lz* lizardite, *Ms* muscovite, *Phl* phlogopite, *Pl* plagioclase, *Po* pyrrhotite, *Py* pyrite, *Qz* quartz, *Scp* scapolite, *Sp* sphalerite, *Tlc* talc, *Tr* tremolite, *Ttn* titanite, *Tur* tourmaline, *Wo* wollastonite, *Zo* clinozoisite, *Zrn* zircon

Table 4 Fabric parameters (minimum-maximum, median) of studied rocks from the Moldanubian Zone and marble samples of artefacts

Colour	Group	Quarry (sample code)	ED (mm)	AR	APR	SF	SPO	*t*
White	Calcitic marbles	Nehodiv (310A-C)	0.16–3.05, 0.71	1.01–3.08, 1.47	0.57–1.05, 0.90	0.25–0.87, 0.63	–	0.05–0.08, 0.06
White		Rabí (311A-C)	0.25–6.65, 1.13	1.03–3.34, 1.50	0.55–1.02, 0.85	0.24–0.82, 0.57	–	0.05–0.06, 0.06
White		Hejná (317A-B)	0.18–8.13, 1.07	0.01–3.94, 1.56	0.59–1.03, 0.84	0.27–0.83, 0.55	–	0.04
White		Nezdice (319A-B)	0.13–4.12, 0.60	1.01–3.19, 1.50	0.50–1.03, 0.83	0.20–0.83, 0.54	–	0.03–0.05, 0.04
White		Nezdice (319C)	0.11–3.07, 0.41	1.04–3.50, 1.58	0.41–1.04, 0.87	0.13–0.85, 0.60	–	0.04
Grey	Calcitic marbles	Votice (313)	0.01–0.41, 0.05	1.03–3.44, 1.50	0.71–1.05, 0.93	0.39–0.86, 0.68	+	0.02
Grey		Vyšný (314A)	0.10–1.61, 0.46	1.03–2.91, 1.54	0.61–1.03, 0.87	0.29–0.83, 0.59	+	0.06
Grey		Vyšný (314B)	0.01–0.79, 0.11	1.03–4.66, 1.46	0.64–1.06, 0.90	0.32–0.89, 0.64	–	0.03
Grey		Český Krumlov (315)	0.03–2.62, 0.14	1.03–3.03, 1.43	0.71–1.07, 0.95	0.40–0.90, 0.70	+	0.02
White	Dolomite–calcitic marbles	Soběšice (318A-B)	0.07–1.41, 0.29	1.00–4.17, 1.61	0.44–1.03, 0.82	0.15–0.83, 0.53	–	0.05–0.06, 0.05
White		Ostružno (320)	0.03–2.03, 0.12	1.03–2.86, 1.44	0.61–1.04, 0.91	0.30–0.86, 0.66	–	0.02
White	Calcite–dolomitic marbles	Bohumilice (312A, B, D)	0.03–0.90, 0.25	1.01–3.94, 1.46	0.61–1.05, 0.88	0.29–0.87, 0.61	–	0.05–0.09, 0.07
White	Dolomitic marbles	Jaroškov (321)	0.08–2.06, 0.28	1.04–2.85,1.44	0.52–1.03, 0.86	0.21–0.83, 0.59	–	0.03
Grey	Dolomite–calcitic marbles	Bližná (316A)	0.03–0.87, 0.20	1.06–3.51, 1.51	0.61–1.03, 0.88	0.30–0.83, 0.61	–	0.05
Grey	Calcite–dolomitic marbles	Bližná (316B)	0.05–0.97, 0.22	1.03–4.09, 1.53	0.56–1.01, 0.84	0.25–0.80, 0.55	–	0.05
Coloured	Carbonate–silicate rock	Bohumilice (312C, E)	0.04–0.77, 0.17	1.01–3.49, 1.47	0.56–1.02, 0.88	0.25–0.82, 0.60	–	0.05–0.07, 0.06

Colour	Group	Artefact (sample code)	ED (mm)	AR	APR	SF	SPO	*t*
White	Calcitic marbles	S1118	0.63	1.50	0.89	0.62	–	0.04
White	Calcitic marbles	S1119	0.90	1.49	0.90	0.64	–	0.07

AR aspect ratio, *ED* equivalent diameter, *APR* area/perimeter ratio, *SF* shape factor, *SPO* shape-preferred orientation, *t* index of grain size homogeneity

Marbles with increased dolomite content

Fine-grained marbles containing more than 10 vol% of dolomite were composed of varying proportions of calcite and dolomite (10–50 vol% dolomite: Bližná-316A, Soběšice, and Ostružno; 50–90 vol% dolomite: Bližná-316B, Bohumilice-312A, B, D; 90–100 vol% dolomite: Jaroškov), as well as varying amounts of non-carbonate accessory minerals (Table 3). Graphite was present in the grey marble from the Bližná quarry. The median IR content of dolomite–calcitic, calcite–dolomitic, and dolomitic marbles ranged from 1 to 10 wt% (Table 3). The CL fabric of the carbonates of these marbles was characterized by homogeneous calcite (orange CL) and isolated dolomite grains (dark red to reddish brown CL). One type of marble from the Bližná quarry (sample No. 316B) included characteristic non-carbonate minerals with distinctive CL colours (dark red CL of forsterite, and intense yellow-green CL of diopside) (Fig. 4j–l). The identification of forsterite and dolomite (with a similar CL colour) was achieved by their characteristic high (forsterite) or low (dolomite) relief in thin section by PM. Diopside with a lower Fe content (FeO $<$ 1 wt% by SEM-EDS) was analysed from the Bližná marble, as well as from the Bohumilice (yellow-green CL) and Ostružno (blue CL) marbles.

Marbles from the Bližná, Soběšice, and Jaroškov quarries displayed conformable grain size and grain shape characteristics (Table 4). The highest ED, AR, and *t* values (as well as the lowest values of APR and SF) were detected in the marble from the Soběšice quarry. A heteroblastic fabric (*t* value of 0.02) with larger carbonate grains occurring in prevalent fine-grained carbonate grains (median ED value of 0.1), along with a circular shape of the carbonate grains (highest values of APR and SF), was found in the dolomite–

calcitic marble from the Ostružno quarry (Table 4). No SPO was found in the group of marbles including > 10 vol% dolomite. Specimens from the Bližná quarry exhibited equivalent fabric parameters (Table 4), but contained varied mineral phases (Table 3). The banded type from the Bližná quarry (sample No. 316 A) exhibited a greater content of phlogopite and tremolite compared with the massive type (sample No. 316 B), which additionally contained forsterite and lizardite (Fig. 4j–l).

Carbonate–silicate rocks

Fine-grained carbonate–silicate rocks from the Bohumilice quarry constitute a separate group because two out of five sampled rock types (sample Nos. 312C, E) exhibited a prevalence of non-carbonate minerals (IR 60–67wt%) over the carbonate minerals (Table 3). The remaining three rock types (312A–B, D) from the Bohumilice quarry can be classified as calcite–dolomitic marbles with a low content of the non-carbonate fraction (IR 4–13 wt%). The carbonate–silicate rocks were predominantly composed of silicates (e.g. diopside, forsterite, serpentine minerals, and phlogopite), sulphides (sphalerite), and a relatively low proportion of carbonates (vol% of calcite > vol% of dolomite). Calcite–dolomitic varieties of the Bohumilice marbles were characterized by the dominance of dolomite over calcite, and minor amounts of phlogopite, forsterite, diopside, and other accessory minerals (see Table 3). Carbonate–silicate rocks and marbles from the Bohumilice quarry exhibited a fine grain size (median ED of 0.2 mm) (Fig. 6), almost uniform in size (homeoblastic fabric, median *t* value of 0.07), and no SPO (Table 4).

Artefacts

The mineralogical-petrographic characteristics of the studied artefacts are described in Tables 3 and 4. Marble samples of both artefacts, besides the predominant calcite, also contained non-carbonate minerals (such as diopside) either with a dull CL and higher Fe content (in the case of the artefact from Vrchotovy Janovice) (Fig. 7a–c) or with an intense yellow-green CL and higher Mg content (in the case of the artefact from the Prague Klementinum) (Fig. 7d–f). Other accessory minerals such as albite, apatite, muscovite, quartz, and titanite exhibited a dull CL (Fig. 7c, f). The marble samples of the studied artefacts exhibited calcite with small grain size (median ED of 0.6 and 0.9 mm) (Table 4; Fig. 6). The marble of the artefact from Vrchotovy Janovice exhibited carbonate grains with a heteroblastic fabric (median *t* value of 0.04) in contrast to the homeoblastic fabric of the marble of the artefact from the Prague Klementinum (median *t* value of 0.07) (Table 4).

C and O stable isotope geochemistry

The δ^{13}C and δ^{18}O data of the groundmass of the studied samples from the MZ ranged from −5 to +4‰ (PDB) and from −5 to −16‰ (PDB), respectively (Table 5).

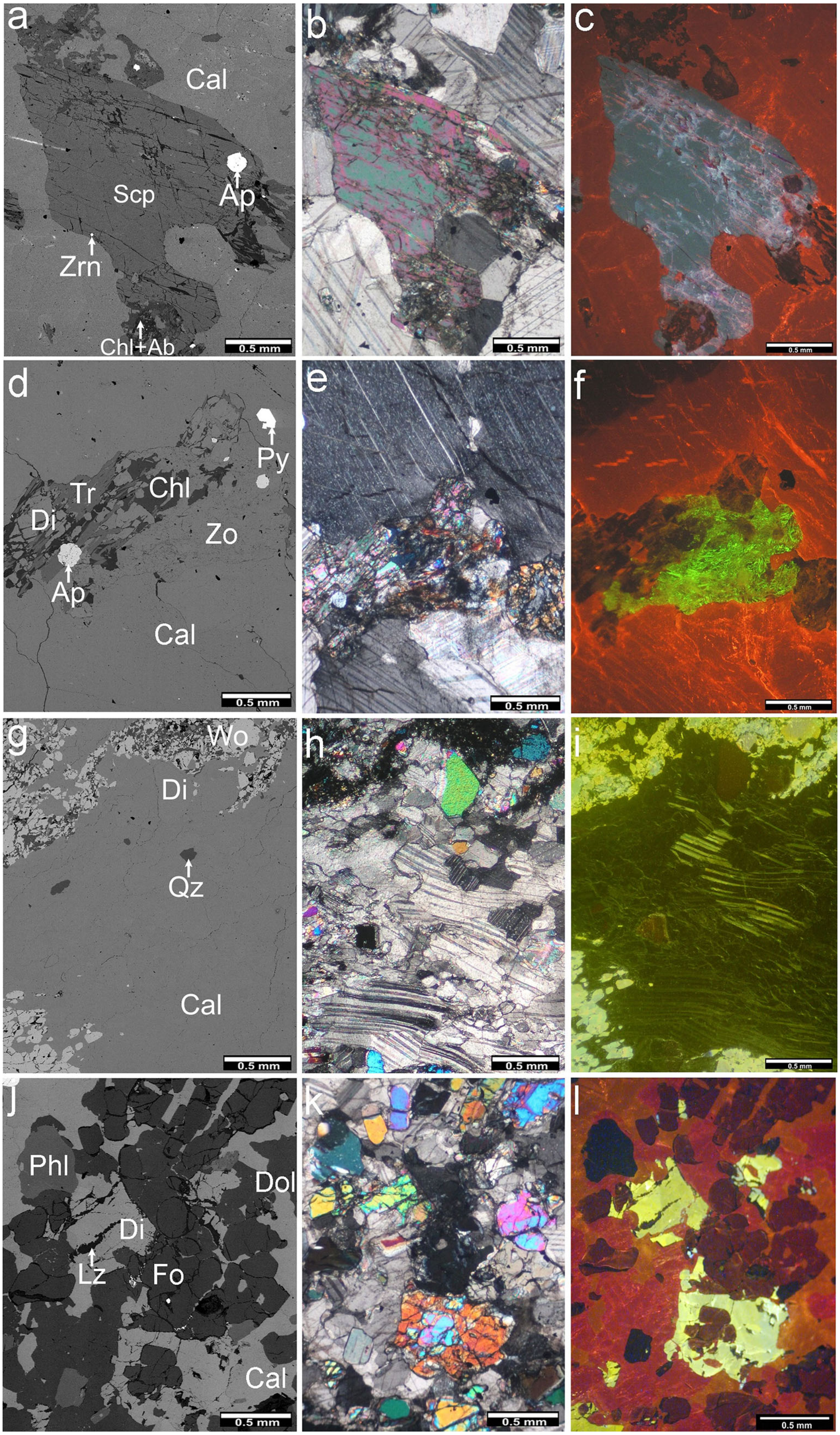
The general isotopic range of non-metamorphosed Palaeozoic limestones ranges from −1 ± 1 to +4 ± 2‰ (PDB) for δ^{13}C and from −8 to 0‰ (PDB) for δ^{18}O (Veizer et al. 1999). The lower δ^{18}O values of most of the metacarbonates studied probably indicate some interaction with metamorphic or magmatic fluids during the regional metamorphism of the MZ. Additionally, some of the studied marbles exhibited lower δ^{13}C values, which might be explained by decarbonation processes (Valley 1986). Variable δ^{13}C and δ^{18}O values of carbonates within marble types of a single locality were found for the Nezdice and Bohumilice marbles (Table 5, Fig. 8a, c, d). The shifts to lower C and O isotopic values occurred in silicate-enriched types of marbles (i.e. the wollastonite-rich type of Nezdice marble and the carbonate–silicate rocks from the Bohumilice quarry). Uniform C and O isotope data of the groundmass (δ^{13}C ~ −4 to −2‰, and δ^{18}O ~ −10 to −15‰, PDB) of the studied samples are typical for some types of Moldanubian marbles, which had all experienced the same metamorphic conditions and are geographically close to one another (i.e. Hejná, Nezdice, Ostružno, Rabí marbles). On the other hand, the Nehodiv and Soběšice marbles exhibited higher groundmass values of δ^{13}C (~ +2 to +4‰, PDB) compared to marbles in the nearby vicinity (Fig. 8a, c). Grey marbles from the Bližná, Český Krumlov, Votice, and Vyšný quarries exhibited a wide range of C and O isotope values (δ^{13}C ~ −2 to +3‰, and δ^{18}O ~ −5 to −13‰, PDB). Generally, these graphitic marbles displayed slightly higher δ^{13}C and δ^{18}O values than the white and coloured marbles studied, with the exception of the groundmass values of Bohumilice, Nehodiv, and Soběšice marbles. The C isotopic signature of graphitic marbles is, among other things, related to the low content of graphite included in the studied samples.

The C and O isotopic values of both of the studied artefacts are depicted in Table 5. The artefacts mainly differed in δ^{13}C values (Fig. 8a). A higher value of δ^{13}C (~ 2‰, PDB) was measured in the sample from the Prague Klementinum, in comparison with a lower δ^{13}C value (~ −4‰, PDB) in the artefact from Vrchotovy Janovice (Fig. 8a).

Discussion

Distinct petrographic features of studied MZ marbles

Macroscopically, the metacarbonates from the MZ are white, grey, and/or multi-coloured rocks. However, the


a
Cal
Scp
Ap
Zrn
Chl+Ab
0.5 mm
b
0.5 mm
c
0.5 mm
d
Py
Tr
Chl
Di
Zo
Ap
Cal
0.5 mm
e
0.5 mm
f
0.5 mm
g
Wo
Di
Qz
Cal
0.5 mm
h
0.5 mm
i
0.5 mm
j
Phl
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Cal
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◀ **Fig. 5** Electron (backscattered images **a**, **d**, **g**, **j**), polarizing (crossed-polarized images **b**, **e**, **h**, **k**), and cathodoluminescence (CL images **c**, **f**, **i**, **j**) microscopy of selected marble samples from quarries of the Moldanubian Zone. Nehodiv—310B (**a–c**); Rabí—311B (**d–f**); Nezdice—319C (**g–i**); Bližná—316B (**j–l**). Mineral phases (abbreviations after Whitney and Evans 2010): *Ab* albite, *Ap* apatite, *Cal* calcite, *Di* diopside, *Dol* dolomite, *Fo* forsterite, *Chl* chlorite, *Lz* lizardite, *Py* pyrite, *Phl* phlogopite, *Qz* quartz, *Scp* scapolite, *Tr* tremolite, *Wo* wollastonite, *Zo* clinozoisite, *Zrn* zircon

studied metacarbonates were barely distinguishable in terms of their macroscopic characteristics alone. The results of the microscopic analysis as well as those from CL and electron microscopy supported a division between the calcitic marbles (Nehodiv, Rabí, Hejná, Nezdice—white; Votice, Vyšný, Český Krumlov—grey) and dolomitic marbles (Soběšice, Ostružno, Jaroškov—white; Bližná—grey) from the carbonate–silicate rocks (Bohumilice—multi-coloured). Despite these distinct features, the mineralogical-petrographic characteristics (including mineral content and fabric parameters) of the studied rocks exhibited significant variability, even within a single quarry (metacarbonates from the Bližná, Bohumilice, Nezdice, and Vyšný quarries). The dolomite–calcitic and calcite–dolomitic marbles (banded and massive types) from the Bližná quarry, which varied in their mineral content, represented the so-called ordinary metacarbonate rocks as described by Drábek and Stein (2015). Edenite, the calcium-rich amphibole which was characteristic for carbonatite-like marbles from Bližná and the eastern part of the MZ (Drábek and Stein 2015; Houzar and Novák 2002), was found as an accessory mineral in the calcitic marble from Nezdice, as well as in the dolomite–calcitic marble from Ostružno.

In some cases, the presence of a wide range of non-carbonate minerals can be helpful in provenance determinations (see Table 3). There were more impure types (with higher IR content) in the group of calcitic marbles compared to the dolomitic marbles. Selected non-carbonate minerals were found to be useful for the discrimination of the white calcitic marbles from the MZ. The Nehodiv marble was distinguished by the content of scapolite and graphite, as well as the absence of diopside. In one type of Nezdice marble, another mineral phase, wollastonite, was distinctive among the studied marbles. However, the presence of scapolite was often described in the marbles and calc–silicate rocks of the Varied Group (e.g. in the surroundings of Český Krumlov and Třebíč), where wollastonite was also identified (Houzar and Leichmann 2003; Houzar and Novák 2002; Křìbek et al. 1997; Novák et al. 2012). Clinozoisite and titanite were typical for marbles from the Hejná and Rabí quarries, which also differed in

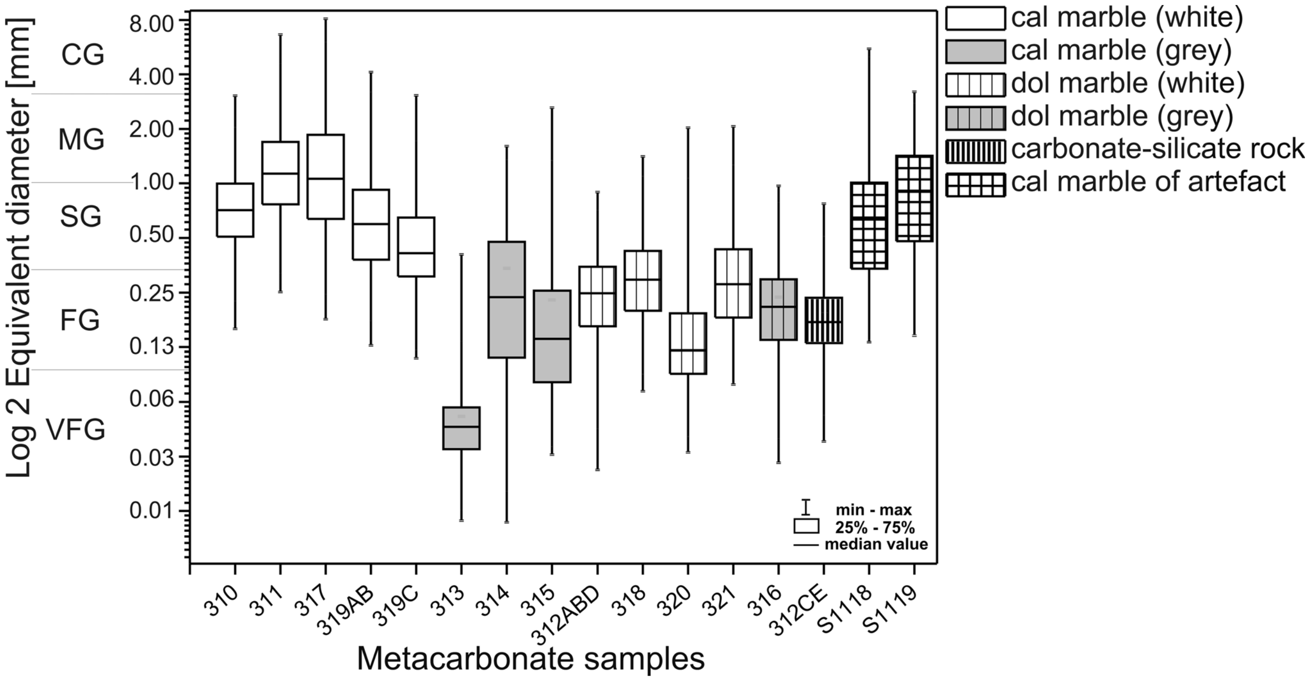

Fig. 6 Statistical evaluation of the equivalent diameter of carbonate grains in the studied metacarbonate samples of the Moldanubian Zone and marble samples of artefacts. Sample codes: *310* Nehodiv, *311* Rabí, *312(A–E)* Bohumilice, *313* Votice, *314* Vyšný, *315* Český Krumlov, *316* Bližná, *317* Hejná, *318* Soběšice, *319(A–C)* Nezdice, *320* Ostružno, *321* Jaroškov, *S1118* artefact sample of a monkey (Vrchotovy Janovice Castle), *S1119* artefact sample of Cupid (Prague Klementinum). Abbreviations of mineral phases and the grain size classification: *cal* calcitic, *CG* coarse grain size (10–3.3 mm); *dol* dolomitic, *MG* medium grain size (3.3–1 mm); *SG* small grain size (1–0.33 mm); *FG* fine grain size (0.33–0.1 mm); *VGS* very fine grain size (0.1–0.01 mm)

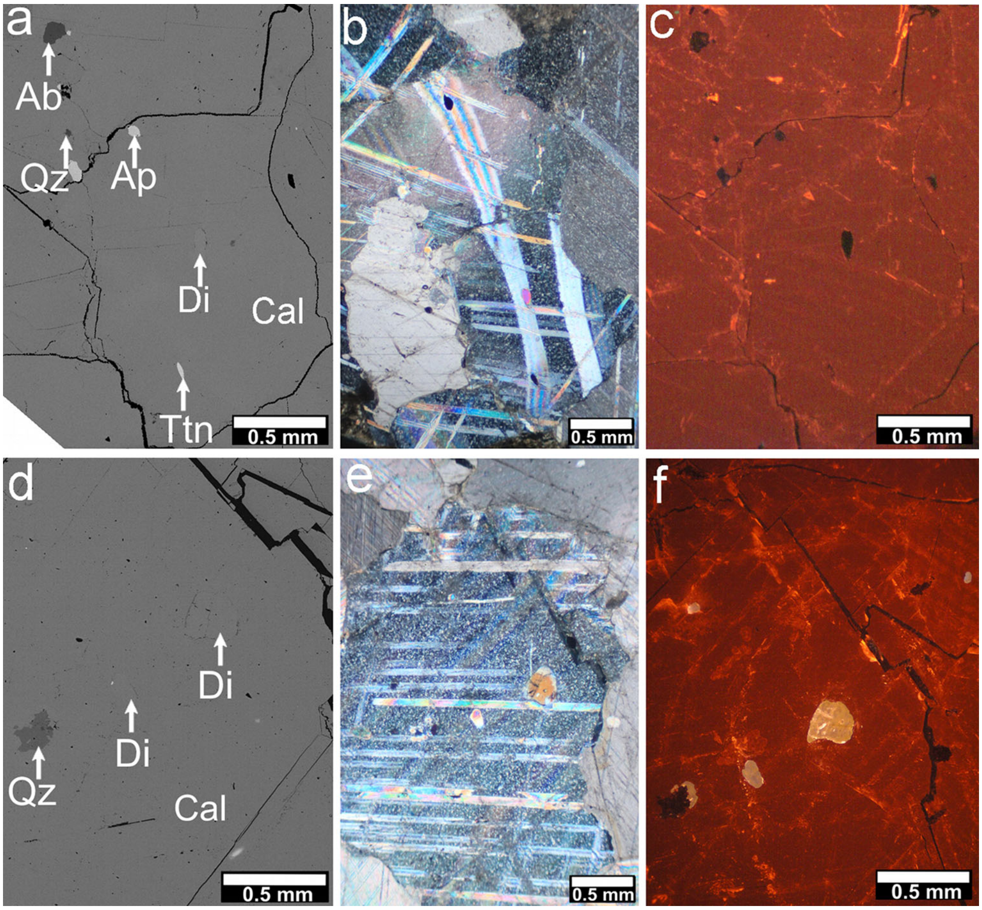

Fig. 7 Electron (backscattered images **a**, **d**), polarizing (crossed-polarized images **b**, **e**), and cathodoluminescence (CL images **c**, **f**) microscopy of marble artefacts. Marble sample of monkey, Vrchotovy Janovice Castle—S1118 (**a**–**c**); marble sample of Cupid, Prague Klementinum—S1119 (**d**–**f**). Mineral phases (abbreviations after Whitney and Evans 2010): *Ab* albite, *Ap* apatite, *Cal* calcite, *Di* diopside, *Qz* quartz, *Ttn* titanite

their larger (medium) carbonate grain size, compared to other white calcitic marbles. The presence of diopside with a higher Fe content, and the very dull CL in the Rabí marble, could be useful for discrimination of the Rabí and Hejná marbles, which have similar grain sizes. Concerning the distinctive mineral phases of the grey calcitic marbles, ferromagnetic pyrrhotite was only detected in the Votice marble. Tourmaline and clinozoisite were typical accessory minerals in the Vyšný marble. In the white dolomitic marble group, the presence of selected non-carbonate minerals produced distinctive features within a single locality; for example, sphalerite and forsterite (in the Bohumilice marble) and talc (in the Soběšice marble). Accessory minerals such as apatite, quartz, and phlogopite were detected in most of the studied rock types and thus presented a low potential for a provenance determination. Tremolite, pyrite, and chlorite are also poor provenance indicators because they represent phases frequently contained in metacarbonates from the Bohemian Forest region (Cmuntová et al. 1970).

Distinct petrographic characteristics of MZ marbles compared to other terrains of the Bohemian Massif

Comparing the distinct mineralogical-petrographic characteristics of calcitic white Moldanubian marbles and calcitic white marbles coming from other geological units of the Bohemian Massif, the Rabí and Hejná marble exhibited a similar carbonate grain size (mean carbonate grain size >1 mm); as did the Velké Kunětice, Supíkovice, Staré Hradisko, and Žulová marbles (coming from the Silesicum) (Šťastná et al. 2009). The differences in their contents of non-carbonate minerals were possible to be used for provenance determinations. Examples include: clinozoisite (only in the Moldanubian marbles), diopside (absent in the Velké Kunětice and Supíkovice marbles), and graphite (in the Staré Hradisko and Žulová marbles). Concerning white calcitic marble with a smaller carbonate grain size (mean carbonate grain size <1 mm), the Nehodiv and Nezdice marbles are comparable to the Nedvědice, Ujčov (coming from the Svratka Crystalline Complex), Velká Morava (coming from

Table 5 C and O isotope values of metacarbonates from the studied quarries of the Moldanubian Zone and marble samples of the artefacts

Quarry (sample code)	No. of samples	$\delta^{13}C$ (‰, PDB)	$\delta^{18}O$ (‰, PDB)
Nehodiv (310A–C)	3	1.61 to 2.46	−9.91 to −13.76
Rabí (311A–C)	3	−2.62 to −3.71	−14.47 to −14.72
Bohumilice (312A–B, D)	3	−0.26 to −1.02	−9.26 to −11.08
Bohumilice (312C, E)	2	−5.18 to −5.53	−14.11 to −15.08
Votice (313)	1	−1.32	−12.87
Vyšný (314A–B)	2	−1.33 to −1.60	−7.28 to −7.80
Český Krumlov (315)	1	2.68	−9.84
Bližná (316A–B)	2	0.16 to 2.30	−7.20 to −4.60
Hejná (317A–B)	2	−3.96 to −4.29	−10.42 to −11.26
Soběšice (318A–B)	2	3.40 to 3.64	−11.26 to −11.49
Nezdice (319A–B)	2	−3.79 to −4.17	−10.24 to −11.71
Nezdice (319C)	1	−5.24	−16.39
Ostružno (320)	1	−3.71	−11.32
Jaroškov (321)	1	−2.22	−11.90
Artefact (sample code)	**No. of samples**	**$\delta^{13}C$ (‰, PDB)**	**$\delta^{18}O$ (‰, PDB)**
S1118	1	−4.11	−13.30
S1119	1	2.14	−14.71

the Lugicum), as well as the Lipová-Na Pomezí and Vápenná marbles (coming from the Silesicum). Scapolite (only in the Nehodiv marble) and wollastonite (in one type of Nezdice marble, as well as in marbles coming from the Svratka Crystalline Complex) in this case were useful for discriminations. The calcitic group of Moldanubian grey graphitic marbles was indistinguishable from marbles from Branná and Horní Lipová (coming from the Silesicum), and Lysice (coming from the Moravicum). The monoclinic pyrrhotite (in the Horní Lipová, Skoupý, and Votice marbles) (Šťastná et al. 2009), clinozoisite, and tourmaline (only in Moldanubian marbles) could serve for provenance determinations. The dolomitic group of Moldanubian marbles (along with the studied quarries in Table 3, also including the Český Šternberk quarry described in Šťastná et al. 2009), are comparable with marbles from Bohdaneč (coming from the Kutná Hora Crystalline Complex), as well as from Raspenava and Strážné (coming from the Lugicum) (Šťastná et al. 2009). The Jaroškov marble is typical, with predominant dolomite in this group. Edenite (only in the Ostružno marble), forsterite (in the Bližná, Bohumilice, and Raspenava marbles), magnetite (only in the Raspenava marble), talc (only in the Soběšice and Bohdaneč marbles), and sphalerite (only in the Bohumilice marbles) could be efficient non-carbonate minerals for the discrimination of dolomitic marbles in the Bohemian Massif.

Comparison of C and O stable isotopes

Considering the stable isotopes of C and O in marbles from the MZ as a separate parameter, some overlap of the quarries was detected. One overlapping group (with a lower $\delta^{13}C$ signature) consisted of white calcitic marbles from the Hejná and Nezdice quarries (Fig. 8a). The Rabí marble could also be considered as falling into this overlapping group when considering all types of inclusions in the Nezdice marble (Fig. 8a). The higher variability of C and O isotopic values within a single quarry, enlarging the isotopic range of a source locality, was connected by the analysis of some marbles and carbonate–silicate rocks with higher contents of non-carbonates (Fig. 8a, d). Similar isotopic trends of impure marbles of the MZ have been described elsewhere (Žák and Sztacho 1994; Žák et al. 1997). C and O isotopic values of graphitic marbles within the studied areas were discussed by Čížek et al. (1984) and Drábek et al. (1999). Additionally, another group of dolomitic marbles coming from the Bohumilice, Jaroškov, and Ostružno quarries could barely be discriminated among based only on their C and O isotopic values (Fig. 8c).

Looking at the data on a large regional scale, marbles from the Hejná, Nezdice, Ostružno, Rabí ($\delta^{13}C$ ~−4‰, PDB), and Soběšice ($\delta^{13}C$ ~+4‰, PDB) quarries exhibited unique C isotopic values in comparison with other marble quarries from different geological units of the Bohemian Massif, Czech Republic (Fig. 8). Regarding overlapping ranges of the Moldanubian and other marbles of the Bohemian Massif, the Nehodiv marble displayed C and O isotope values of the groundmass analogous to values of the Supíkovice marble (coming from the Silesicum) (Fig. 8a). The graphitic (grey) calcitic marbles displayed the largest overlaps. C and O isotope values of the Vyšný marble were similar to the isotopic range of marbles

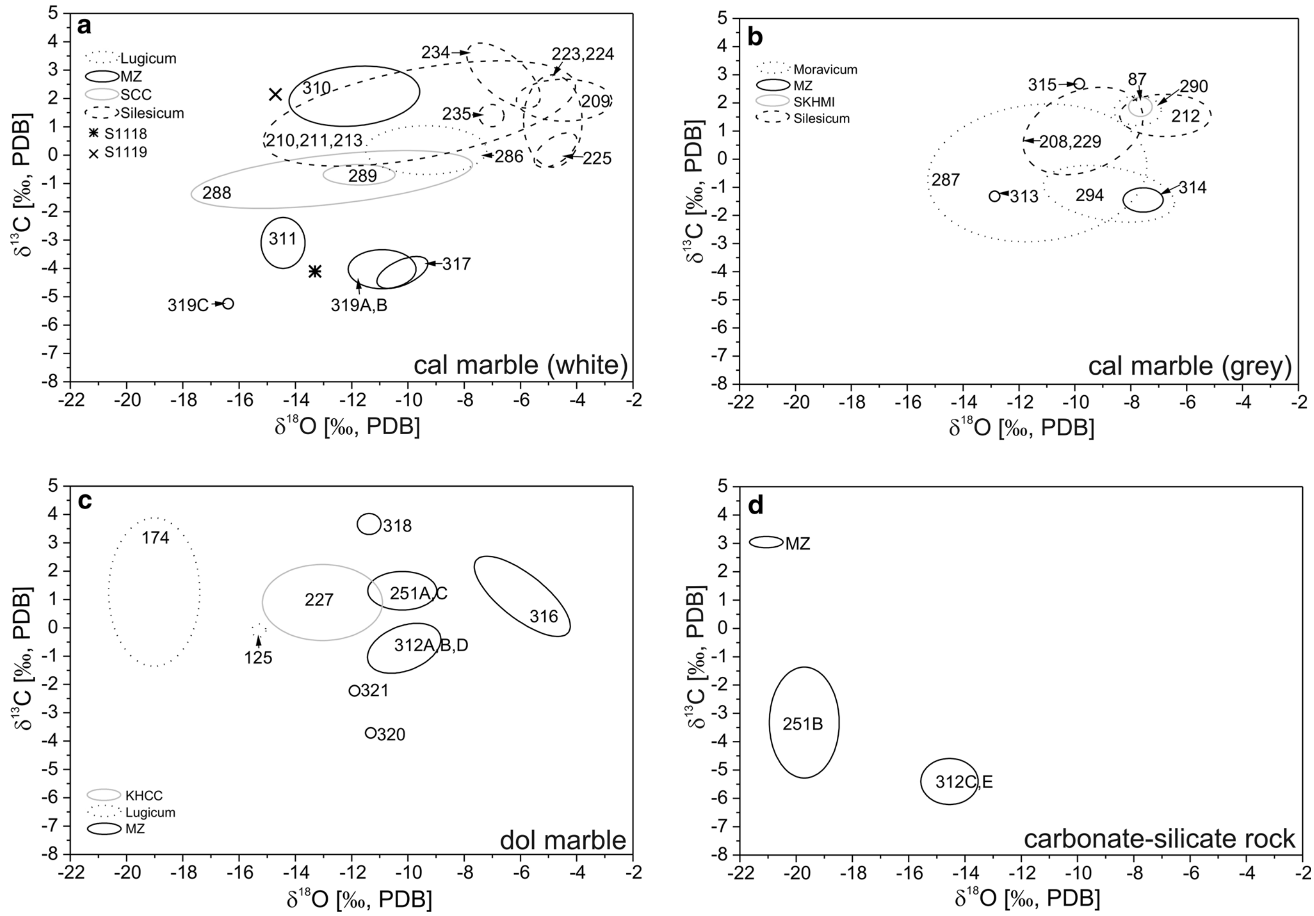

Fig. 8 Comparison of C and O stable isotope data of studied metacarbonate samples from the Moldanubian Zone and different geological units of the Bohemian Massif (Czech Republic) including marble samples of the artefacts. *Each ellipse* or *circle* represents envelopes enclosing all isotope values of a single quarry. *Ellipses* and *circles* are discriminated using different *grey shades* and *dot/dashed lines* according to which geological unit of the Bohemian Massif they belong to (data adopted from Šťastná et al. 2009). **a** White calcitic marbles, **b** grey calcitic marbles, **c** dolomitic marbles, **d** carbonate–silicate rocks. Kutná Hora Crystalline Complex (KHCC): *227* Bohdaneč; Lugicum: *125* Strážné, *174* Raspenava, *286* Velká Morava; Moldanubian Zone (MZ): *251* Český Šternberk, *310* Nehodiv, *311* Rabí, *312* Bohumilice, *313* Votice, *314* Vyšný, *315* Český Krumlov, *316* Bližná, *317* Hejná, *318* Soběšice, *319* Nezdice, *320* Ostružno, *321* Jaroškov; Moravicum: *287* Tišnov-Dřínová, *290* Tišnov-Květnice, *294* Lysice; Silesicum: 208, *229* Branná, *209* Velké Kunětice, *210* Supíkovice, 212 Horní Lipová, *234* Lipová-Na Pomezí, *223*, *224* Staré Hradisko, *225* Žulová, *235* Vápenná; Svratka Crystalline Complex (SCC): *288* Nedvědice, *289* Ujčov; Sedlčany-Krásná Hora Metamorphic Islet (SKHMI): *87* Skoupý

coming from the Tišnov-Dřínová and Lysice quarries in the Moravicum (Fig. 8b). The value of the Votice marble fell within the isotopic range of the Tišnov-Dřínová marble (Fig. 8b). Český Krumlov marble can barely be distinguished from the Branná, Horní Lipová (coming from the Silesicum), Tišnov (coming from the Moravicum), and Skoupý (coming from the Sedlčany-Krasná Hora Metamorphic Islet) marbles based on the C and O isotopic data alone (Fig. 8b).The studied dolomitic marbles did not show any direct overlaps with the ranges of the other geological units. Furthermore, based upon their isotopic signatures alone, the Bohumilice marble could barely be discerned from the Bohdaneč marble (coming from the Kutná Hora Crystalline Complex), or from the Český Šternberk marble (MZ, data published in Šťastná et al. 2009) (Fig. 8c).

Potential of the applied methods for provenance studies: application to artefacts

General usefulness of methods

The same sample (i.e. thin section) can be analysed by different successive techniques (PM, PIA, CL, SEM-EDS), which brings specific advantages to the examination of specimens from artefacts, from which only limited sampling is generally allowed (Šťastná and Přikryl 2010). Only a very small sample (~100 μg) is necessary for isotope measurement by mass spectrometry. On the other hand, the high material loss due to the leaching of carbonates (up to 99 wt%, see Table 3) for IR hardly makes this method applicable to historical artefacts (Šťastná and

Přikryl 2009). The fact that a sample obtained from an artefact can include less (or no) accessory minerals compared with the source rock is also connected with the small sample volume available for analysis. In this case, the quantitative rock fabric parameters of carbonate grains are useful distinctive features. ED and *t* were revealed as the most discriminating parameters from the selected microstructural parameters (see Table 4; Fig. 6). On the other hand, the fabric parameters describing the shape of carbonate grains (AR, APR, and SF) of the samples studied displayed similar values. A majority of the marbles exhibited a quasi-isotropic fabric with no SPO. Three of the grey calcitic marbles from the Český Krumlov, Votice, and Vyšný quarries displayed layering parallel to the foliation. Concerning the distribution of the C and O stable isotopes, the results of IRMS exhibited some unique distinctions; i.e. the Nehodiv marble from the remainder of the white calcitic Moldanubian marbles (Fig. 8a), and the Soběšice marble from the remainder of the dolomitic (Fig. 8c) Moldanubian marbles.

Testing the usefulness on artefacts

Two samples of calcitic marble from (1) a sculpture of a monkey (Vrchotovy Janovice Castle), and (2) a sculpture of Cupid (Klementinum, Prague) were examined by the methodology presented herein for the purpose of provenance determination. Marble from the first artefact can be assigned to the Moldanubian marbles on account of its low C and O isotopic values (Table 5; Fig. 8a). Marbles from the Nezdice and Rabí quarries were identified as the possible sources based on the values of the C and O stable isotopes, fabric parameters of the carbonate grains, and the mineralogical composition. The Nezdice marble exhibited carbonate grain size parameters, including values of ED and *t*, in closer agreement to the studied artefact compared to the Rabí marble (Table 4; Fig. 6). On the other hand, the mineral assemblage of this artefact is more conformable to the Rabí marble (inclusions of albite, titanite, and non-luminescent diopside with a higher content of Fe) (Table 3; Fig. 5f vs. 7c). Marble of the second artefact from the Prague Klementinum displayed rock fabric parameters of the carbonate grains (Table 4) as well as C and O isotope values (Table 5; Fig. 8a) similar to those of the Nehodiv marble. However, the artefact sample included a mineral assemblage (diopside with yellow-green CL) which did not correspond with that in the Nehodiv marble (absence of diopside, CL of calcite with a lower intensity compared to the sample from the artefact) (Fig. 5c vs. 7f). Looking at comparable mineralogical-petrographic (similar grain size parameters and CL of calcite; see Šťastná et al. 2009) and isotopic data (Fig. 8a) from other geological units of the Bohemian Massif, the marble from the Supíkovice (Silesicum unit) exhibited similar characteristics. However, as with the Nehodiv marble, diopside is not present in the Supíkovice marble. Diopside with a yellow-green CL (Šťastná and Přikryl 2010), and comparable values of C and O stable isotopes (Fig. 8c), was identified in the Český Šternberk marble; however, it is dolomitic, as well as more fine grained, compared to the artefact from the Prague Klementinum. We cannot exclude as an alternative that the locality of the second artefact as yet remains unknown.

Conclusions

The present study focused on the examination of qualitative and quantitative microscopic, mineralogical, and microstructural characteristics, as well as upon analyses of the isotopic composition of marbles from a single metamorphic terrain in order to identify likely provenance locations. PM and SEM-EDS characterized the mineralogical composition of the studied rocks. The qualitative CL data provided a useful tool to discern carbonates and identify the non-carbonate phases. Carbonates represented the dominant phases in the studied rocks and the content of IR were low (median IR of 1–10 wt%) with the exception of calcitic marbles from Nezdice (IR of 23 wt%), Votice (IR of 33 wt%), and Vyšný (IR of 30 wt%); also the carbonate–silicate rocks from the Bohumilice quarry (IR to 67 wt%). Some mineral phases associated with calcitic marbles (scapolite, tourmaline, and wollastonite), and some associated with metacarbonates including dolomite (talc and sphalerite), occurred within a single quarry. In the various quarries, diopside exhibited different CL (blue, yellow-green, or dull). However, displaying a wide range of accessory minerals is a common feature for the marbles of the Moldanubian Zone.

From the quantitative microstructural characteristics, carbonate grain size parameters (equivalent diameter, index of grain size homogeneity) proved to be useful as discriminating parameters. The median value of the equivalent diameter of the studied calcitic marbles ranged from 0.1 to 1 mm; metacarbonates containing more than 10 vol% of dolomite displayed median values ranging from 0.1 to 0.3 mm for the equivalent diameter. However, the quantitative values of the aspect ratio, area-perimeter ratio, and shape factor (which described carbonate grain shape and compactness) gave almost the same results for different rock types. A marked shape-preferred orientation was recognized in three types of grey calcitic marbles from the Český Krumlov, Votice, and Vyšný quarries.

The C and O isotopic values of metacarbonates within the Moldanubian Zone exhibited high depletion ($\delta^{13}C$ between −3 and −5‰, PDB; and $\delta^{18}O$ between −10 and −16‰, PDB) for the Hejná, Nezdice, Rabí (calcitic), and Ostružno (dolomite–calcitic) marbles. This range of isotopic values is globally unique compared with similar rocks from other marble quarries in the Bohemian Massif. The $\delta^{13}C$ values of the Soběšice marble are higher ($\delta^{13}C$ around +4‰, PDB) than the rest of the Moldanubian marbles, as well as marbles from other parts of the Bohemian Massif. Focusing on the variability of isotopic compositions of Moldanubian metacarbonates, they displayed some overlaps between quarry areas and at times displayed a wide spread of values within a single quarry (as observed in the case of the Bohumilice and Nezdice marble).

A similar research methodology was applied on marble samples from two artefacts. The Rabí marble is the most probable source locality of the artefact studied from Vrchotovy Janovice. In this case, the isotopic signature (low $\delta^{13}C$ value), carbonate grain size, and the presence of non-carbonate mineral phases (albite, diopside and titanite) of this artefact were used to fingerprint it. The Nehodiv marble was discussed as the potential source of the artefact from the Prague Klementinum. The fabric parameters of its carbonate grains, as well as the C and O isotopic values, were in agreement with the Nehodiv marble; however, the mineral assemblage and cathodoluminescence were dissimilar.

The results presented make up part of a quantitative database that can be employed for provenance studies of specimens obtained from various artefacts. The present study shows that a combination of mineralogical-petrographic characteristics along with C and O stable isotopes provides the fundamental approach necessary for reliable material research of marbles coming from metamorphic terrains with a complex geological history, such as the Moldanubian Zone of the Bohemian Massif.

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