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Minero-petrographic and isotopic characterization of two antique marble quarries in the Denizli region (western Anatolia, Turkey)

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Abstract

The marbles are widely used rock types in the structures of today and ancient world. In this study, two ancient marble quarries named as Hierapolis and Domuzderesi in Denizli region, have been examined. The marble samples from these quarries can be grouped into three types, based on color, crystal size, crystal boundaries and foliation status. These groups are identified as i) white, ii) gray veined and iii) gray marbles. In terms of microscopic features, heteroblastic polygonal (white marbles in Hierapolis, gray-veined and gray marbles in Domuzderesi) and homeoblastic polygonal textures (white marbles in Domuzderesi) have been defined. The mineral compositions of all marble groups are quite similar. They are consisting predominantly of carbonate minerals (calcite, dolomite) and, as accessory minerals quartz, muscovite, pyroxene and ferric iron oxides. These results are also supported by the XRD studies. According to the geochemical analyses, protoliths of Hierapolis and Domuzderesi marbles are limestone. In Hierapolis marble quarry, the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values change between 0.63 - 3.52 ‰ and (-9.55) - (-1.21)‰, in turn. The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of Domuzderesi marble quarry range from (-1.44) ‰ and 3.41 ‰, -13.26 ‰ and (-5.3) ‰, respectively. Minero-petrographic, geochemical and C-O stable isotope results reveal that Hierapolis and Domuzderesi ancient quarries have similar characteristics which have originated from a same protolith.

Key words: Hierapolis; Domuzderesi; Denizli; Antique marble quarry; C-O stable isotopes.

Introduction

Denizli graben is 7-28 km wide and 62 km

long NE-SW trending collapse area. It was firstly defined by Westaway (1990; 1993) and is located at the intersection of three well-

identified major grabens, the Gediz, the Küçük Menderes and the Büyük Menderes grabens, in the western Turkey. Because Denizli graben has a strategic location which is the passageway between the Aegean, Middle Anatolia and the Mediterranean regions, it has hosted numerous ancient settlements in different time periods. The earliest known settlement dates back to the the Chalcolithic period (Şimşek, 2007).

Futhermore, there are important remains of ancient Greek and Roman cities like Hierapolis, Laodicea, Tripolis and Colossae. In these civilizations, natural stones such as marble and travertine were utilized extensively as building stones, artefacts, statues and sarcophagus. Marbles were commonly used for sculpture and as a building material during pre- and early historical periods and also in the Greek and Roman periods in addition to later times. In the Mediterranean region, there are many high-quality marble locations, which were used in antiquity and are still operated today. Minero-petrographic, geochemical and provenance analyses of these marbles have great importance for restoration of ancient artworks, monuments and buildings or for determining imitations.

Although there is a well-documented marble quarry history, the methods of utilization and diffusion, minero-petrographic, geochemical and isotopic characteristics of the marble in the Mediterranean region, two ancient marble quarries named as Hierapolis and Domuzderesi in Denizli region have not been investigated in detail to date. In this paper, we provided mineralogic, petrographic, major-trace elements compositions and C-O stable isotope analyses on marbles of ancient Hierapolis and Domuzderesi quarries to identify the minero-petrographic and geochemical characterizations. In addition, we also intended to provide informations for forthcoming restorations in three important archaeological sites in the Denizli region (Hierapolis, Laodicea and Tripolis).

Geological Setting of Denizli Region

Western Turkey, very rich in archaeological sites of different periods is situated in the Mediterranean region. Metamorphic rocks of the Menderes Massif and the Neogene sedimentary formations constitute dominant rock units of the Western Turkey. The Menderes Massif covers a large area in Western Turkey, and composes of various metamorphic rocks. The detailed stratigraphy of the Denizli and surrounding areas were given by Şimşek (1984) and can be summarized as follows. The Paleozoic Ortaköy formation and Lycian nappes are the oldest geological units in the study area. The Ortaköy formation is represented by metamorphic rocks mostly composed of schist and marble, occasionally amphibolite and quartzite. Middle Eocene Lycian nappes are characterized by ophiolitic units which consist of serpentinitized mafic-ultramafic rocks and allochthonous carbonate rocks. Both of these units are found on the northern and southern flanks of the Denizli Graben. The depression areas of Denizli graben is filled by fluvial, fluvio-lacustrine, terrestrial and travertine deposits, which are named as Kızılburun, Sazak, Kolonkaya, Ulubey formations and Asartepe formation. All these units unconformably overlie the Paleozoic Ortaköy formation and Lycian nappes. Early Middle Miocene Kızılburun formation comprises pebble stone, sandstone, marl, claystone, siltstone, clay, clayey-silty marl and sands and pebbles in thin stripes and lenses. Middle Miocene Sazak formation is harmoniously located on the Kızılburun formation. It is characterized by fluvio-lacustrine sediments which composed of claystone, siltstone, marl, mudstone, clayey limestone, gypsum and gypsiferous mudstone and shale. Middle-Late Miocene Kolonkaya formation consists of mudstone, siltstone, marl, alternating with sandstone, claystone, siltstone, black shale, marl, clayey limestone, conglomerate

lithologies. Late Miocene-Late Pliocene Ulubey formation conformably overlies the older Neogene units. It is represented by lacustrine limestone, claystone and marl levels. Limestone is the dominant rock lithology and contains silica nodules and fossils. Ulubey formation generally displays smooth, horizontal stratification. The Quaternary Asartepe formation, characterized by coarse-grained fluvial deposits, travertine and recent alluvium unconformably overlies the older geologic units in Denizli graben (Figure 1) (Şimşek, 1984; Okay, 1989; Konak et al., 1987; Sun, 1990; Westaway, 1993; Sözbilir, 1997;

2005; Gökğöz, 1998; Özkul et al., 2002; 2013; Koçyiğit, 2005; Kaymakçı, 2006; Alçıçek et al., 2007; Bozcu, 2009; Hançer, 2013; Koralay et al., 2013; Erten et al., 2014; Topal and Özkul, 2014).

Analytical techniques

The determination of scientific characterizations of the marbles was performed to the following steps and analyses:

- i) Mineralogical and petrographic parameters (i.e. mineral association, grain size and

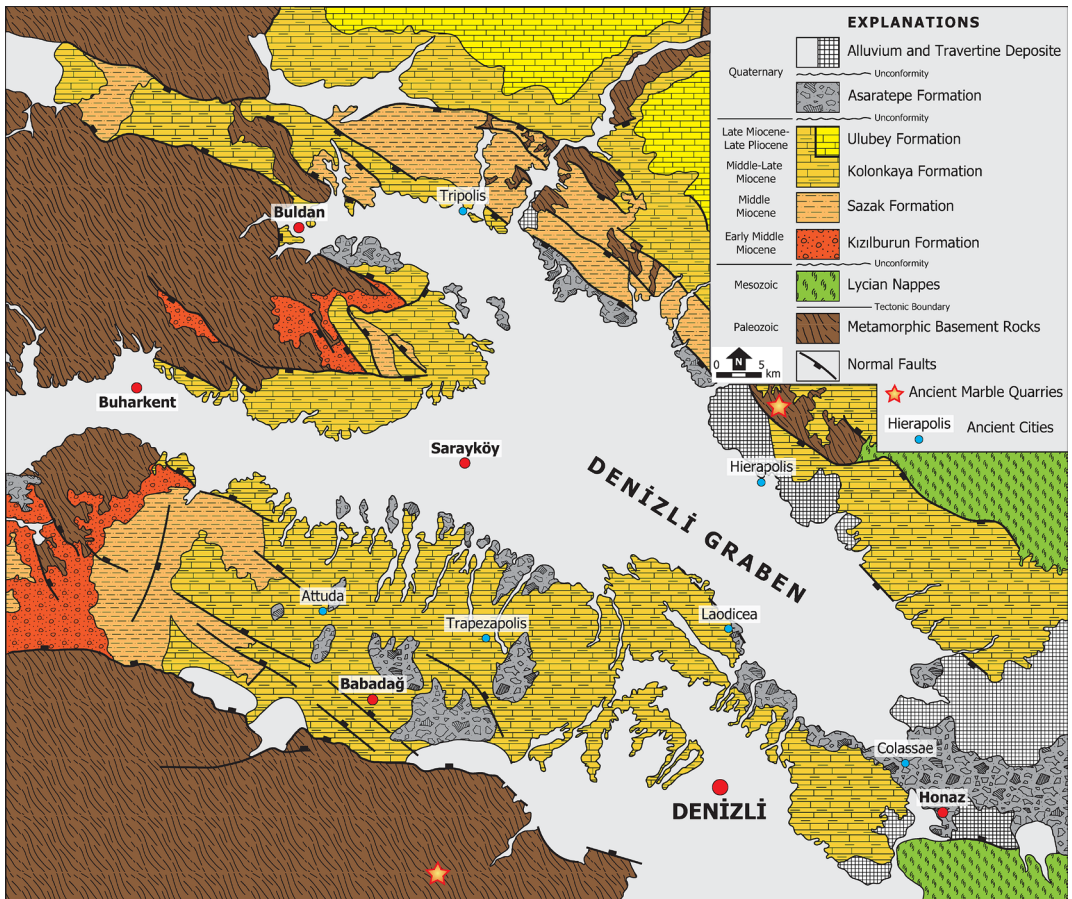


Figure 1. Geological map of the Denizli Graben.

structure, texture of samples) were examined under polarized light microscope using thin sections.

- ii) Qualitative mineralogical composition of the marble samples was determined with X-ray powder diffraction (XRPD), using an Inel Equinox 1000 diffractometer (Instrumental condition: $\text{CoK}\alpha$ radiation obtained at 30 kV and 30 Ma, 10-100° 2 θ investigated range, 0.030° step). In this study, 19 marble samples were analyzed for their mineralogical compositions by XRPD. The analyzed samples were finely grounded by tungsten carbide crushing vessel. Then a few milligrams of the powder were placed on sample holder and put into the XRD machine.
- iii) Major oxide and some trace elements (V, Cr, Co, Rb, Sr, Y, Pb, Th) for the samples were analyzed by a Spectro XEPOS Polarized Energy Dispersive X-ray Fluorescence (PEDXRF) spectrometer at the Department of Geological Engineering at Pamukkale University. The Spectro XEPOS uses a 50 W Pd end window X-ray tube. The spectral resolution of this detector is 160 eV for Mn $\text{K}\alpha$. During the measurement the sample chamber is flushed with He. For the XEPOS XRF analyses, 41 marble samples were crushed in a tungsten carbide crushing vessel, and 6.25 g of powdered sample was mixed with 1.4 g of wax. The mixture was pressed at 20 N in an automatic press to obtain a pressed disc.
- iv) Trace elements (including Ni, Cu, Zr, Nb, Ba, La, Ce, Hf, Ta) for the samples were analyzed by a Spectro XLAB-2000 PEDX-ray fluorescence spectrometer at the Department of Geological Engineering at Ankara University. The instrumentation was equipped with a 400 W Rh end window tube and a Si(Li) detector with a resolution of 148 eV (1000 cps Mn $\text{K}\alpha$). The available targets were Al_2O_3 and B4C used as Barkla polarizer, an HOPG (High Oriented Pyrolytic Graphite)-crystal used as Bragg polarizer and Al, Mo, and Co used as secondary targets. The irradiation chamber was operated under a vacuum system. For the XLAB-2000 XRF analyses, 41 marble samples were crushed in a tungsten carbide crushing vessel, and 4 g of powdered sample was mixed with 0.9 g of wax. The mixture was pressed at 20 N in an automatic press to obtain a pressed disc.
- v) Loss on Ignition (LOI) of marble samples was determined by Protherm PLF 120/27 electric furnace at the Department of Geological Engineering at Pamukkale University. Two grams of each marble sample were used for the determination of LOI. The powders were ignited in the laboratory furnace for half an hour at 1050 °C and cooled down in a desiccator to reach the room temperature, and then the loss in weight was calculated and recorded as a percentage of the original sample weight.
- vi) Stable isotope analysis was performed by continuous flow-isotope ratio mass spectrometry at Iso-Analytical Limited, Crewe, UK using an ANCA-G gas purification module and 20-20 mass spectrometer (Europa Scientific Ltd, Crewe, UK). Sample, reference and control carbonates were weighed into Exetainer tubes (Labco, UK), flushed with 99.995% helium and converted to carbon dioxide by adding phosphoric acid by injection. The reaction containers were left overnight to allow complete conversion. The phosphoric acid used had been prepared for isotopic analysis in accordance with Coplen et al. (1983). The reference material used was IA-R022 (Iso-Analytical working standard calcium carbonate, $\delta^{13}\text{CV-PDB} = -28.63\text{‰}$ and $\delta^{18}\text{OV-PDB} = -22.69\text{‰}$). NBS-18 (carbonatite, $\delta^{13}\text{CV-PDB} = -5.01\text{‰}$ and $\delta^{18}\text{OV-PDB} = -23.2\text{‰}$) and NBS-19 (limestone, $\delta^{13}\text{CV-PDB} = +1.95\text{‰}$ and

$\delta^{18}\text{O}_{\text{V-PDB}} = -2.2\text{‰}$) were measured as control samples.

Minero-Petrographic Investigations

Hierapolis marble quarries

Hierapolis marble quarries are found on the northern flank of the Denizli Graben. They are located in Denizli M22-a2 topographic map. Hierapolis quarries were reported for the first time by Monna and Pensabene (1977) during a reconnaissance tour in Turkey in the mid-seventies. Attanasio and Pensabene (2002), Attanasio et al. (2006) and Şimşek (2007) gave brief informations about quarries in their books.

In this paper, three well-preserved ancient marble quarries were identified in detailed. Quarries are located about 3 km northeast of North Gate of the Hierapolis city (Figure 2a). Quarries were located on the hillside and their altitudes are ranging between 1 and 10 meters with respect to the valley floor. They were operated in the marbles of the Paleozoic Ortaköy formation. Chlorite-mica schists and phyllites can be seen as interbeds or lenses inside marbles. White, gray-veined and gray marbles have been exploited from Hierapolis quarries. In addition, red/brown/green-veined, red cemented brecciated marbles was also extracted from quarries (Figure 2 b,c). Marble

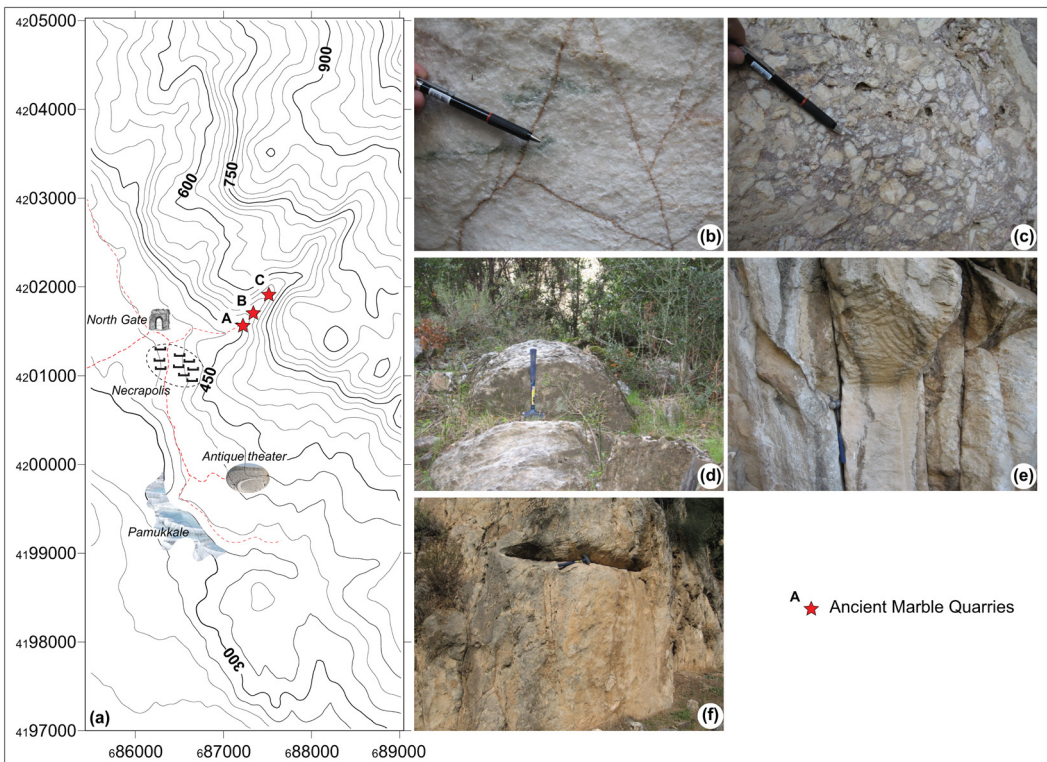


Figure 2. a) Topographic map of the Hierapolis quarries; b) red/brown/green-veined marbles; c) red cemented brecciated marbles; d) unfinished cylindrical marble blocks; e), f) the working faces bearing cutting marks left by the pickaxes and the pointed chisels.

productions were made with various sizes of quarries inside narrow valley. Extraction zone have northeast-southwest orientations and covers an area approximately 80 m long and 40 m wide. The quarries are partially filled by debris deposits generated during the shaping process. Unfinished cylindrical or rectangular marble blocks can be observed inside these debris deposits (Figure 2d). The working faces still bear nicely preserved cutting marks left by the pickaxes and the pointed chisels (Figure 2 e,f). Furthermore, there are small man-made cavities in the bedrock at regular intervals. The wooden and/or iron wedges should be nailed into these cavities in order to separate blocks from the main rock mass (Bingöl, 2004; Malacrino, 2010).

Petrographic properties of Hierapolis marble

Macroscopic and microscopic investigations revealed that the marble quarried at Hierapolis can be classified into three groups based on color, crystal size and foliation status. These groups are described as white, gray-veined and gray marbles. The differences and/or similarities between marble groups are summarized in Table 1.

The white marbles are quite homogeneous in color (Figure 3a). The components of white marble are seen by the naked eye, and the crystal surfaces display bright reflection in the daylight. Crystals vary from medium (2 mm) to coarse (> 5 mm) grained, and have generally straight boundary shapes. A specific characteristic of this marble, besides its whiteness, is its near complete lack of impurities. Crusting, sugaring and spills on the surface of white marbles, on the other hand, can be observed in some place of quarries. The geologic (i.e mineralogic and chemical composition of constituent materials, crystal shape and size, micro-crack) and climatic factors (i.e wind, acid rains, hot-cold cycles, freeze-thaw cycles) would cause degradation of the marbles.

The white marbles have mainly heteroblastic polygonal texture and consist of only carbonate

minerals (Figure 3b). Calcite is the dominant carbonate mineral. The lesser amount of dolomite was seen in some samples. The crystal boundaries are regular. In coarse calcite crystals, rhombohedral cleavage and polysynthetic twins are very characteristic (Figure 3b).

The gray-veined marbles are observed as alternating with white marbles. An irregular gray-colored veined structure on a white background is the most typical feature of this marbles (Figure 3c). The vein widths range from 0.05 to 0.1 cm. The gray-veined marbles have crystallized structure (0.1 to < 1 mm in size) that is visible to the naked eye or able to be seen through a simple hand lens.

Microscopically, the gray-veined marbles have heteroblastic mosaic texture and show mineral orientation. They consist of carbonate minerals and small amount of quartz, pyroxene and muscovite (Figure 3d). Calcite is the dominant mineral. It has subhedral to anhedral crystals shapes with sutured boundaries. Coarse calcite crystals have perfect cleavage and polysynthetic twinning. In some samples, deformed and kinked calcite crystals can be observed. Dolomite is the second abundant carbonate mineral. Quartz, pyroxene and muscovite are seen as rounded grains which are varying from 30 to 100 μm in size.

The gray marbles are brownish grey to dark brown in color. Their fresh surfaces are characteristically dark grey to blackish grey (Figure 3e). It is thin (0.1 mm) to medium (< 1 mm) grained marble and displays mineral orientation.

Mineralogically, calcite is the most abundant carbonate mineral. In addition, dolomite, quartz and pyroxene may be present in some samples. The gray marbles display heteroblastic mosaic textures under the microscope (Figure 3f). Metamorphic stress indicators such as lineated minerals, deformed and kinked calcite crystals, are characteristic in these marbles. Quartz and pyroxene are present as small single, rounded crystals.

Table 1. Some mineralogical and petrographic features of the marbles of Hierapolis and Domuzderesi quarries.

Macroscopic Properties		Microscopic Properties		Other Characteristics	
Colour	Crystal Size (mm)	Mineral Composition	Accessory Minerals		
Hierapolis Quarries	White	Calcite ± Dolomite	-	Heteroblastic Polygonal	The crusting, tearing and spills are characteristics on the surface of marble blocks because of the degradation. In coarse calcite crystals, rhombohedral cleavage and polysynthetic twins are quite characteristic.
	Gray-veined	Calcite > Dolomite	± Quartz ± Pyroxene ± Muscovite	Heteroblastic Mosaic	The gray colored irregular veins, are ranging from 0.05 to 0.1 cm, is the principal characteristic of this group marbles. It shows mineral orientation, In coarse calcite crystals, polysynthetic twins are characteristic.
	Gray	Thin to medium (0.1 - <1)	Calcite ± Dolomite	± Quartz ± Pyroxene	Heteroblastic Mosaic
Domuzderesi Quarries	White	Calcite ± Dolomite	± Quartz ± Pyroxene ± Muscovite	Homeoblastic Polygonal	The crusting, tearing and spills are characteristics on the surface of marble blocks because of the deterioration. In coarse calcite crystals, rhombohedral cleavage and polysynthetic twins are quite characteristic.
	Gray-veined	Calcite > Dolomite	± Quartz ± Pyroxene ± Muscovite	Heteroblastic Mosaic +	The gray colored, irregular veins, are ranging from 0.03 to 0.1 cm, is characteristic. In coarse calcite crystals, polysynthetic twins are characteristic.
	Gray	Thin to medium (0.1-2)	Calcite > Dolomite	± Quartz ± Pyroxene	Heteroblastic Mosaic +



Figure 3. Field and their microscopic photographs of the Hierapolis marbles; a) white marble; b) heteroblastic polygonal texture and perfect rhombohedral cleavage lines of carbonate minerals in white marble; c) gray-veined marble; d) heteroblastic mosaic texture in gray-veined marble and rounded quartz grains; e) gray marble; f) heteroblastic mosaic texture and lined carbonate minerals in gray marbles.

Domuzderesi (Yeşilköy) marble quarries

Domuzderesi marble quarries which are firstly defined by Monna and Pensabene (1977) are found on the southern flank of the Denizli Graben. In the southern flank of Denizli Graben, there are several tectonic units forming a nappe stack. These tectonic units are Menderes massif, Denizli unit, Tavas unit, Ortaca Mount unit, Göbecik Tepe unit, Honaz shale and Honaz ophiolite. Domuzderesi ancient quarries were opened in the Denizli unit which is made up of marble, banded marble, cherty marble, chlorite-graphite schist, phyllite and metaquartzite (Okay, 1989 and references therein). Due to

metamorphism and deformation, medium to large scale folds are highly characteristic in Denizli unit.

Domuzderesi quarries are found about 10 km southwest of Yeşilköy village (Figure 4a). Although there is rugged topography, it is easy to reach Domuzderesi quarries by using gravel and dirt road. Attanasio (2003) shortly reported these quarries in his book in which no less than 32 ancient quarries and the well preserved remains of a Roman transport road were noted (Figure 4b). In addition to dense scrub and forest cover, production of aggregates in the last 20 decades was made from this area; unfortunately, these

have led to the extinction of the ancient quarries. Therefore, three well-preserved ancient marble quarries could be defined on southwest slope of Kuzuluk hill (Figure 4a). White, gray-veined and gray marbles are described in quarries A, B, C and D. In the upper section of quarries, chlorite schist, graphitic schist and quartzite lenses can be observed. Quarries A, B and C are positioned at a height of 25-80 meters from the current stream bed (Figure 4c). The total extraction zone has northeast-southwest orientation and covers an area approximately 100 m long and 80 m wide. The working faces still bear nicely preserved cutting marks left by the pickaxes and the pointed chisels (Figure 4 d,e). Tool marks on the extraction surfaces of Quarry D is not clear with respect to quarries A, B and C. There are some debris deposits which were generated during the shaping process in front of the quarries. Unfinished cylindrical

or rectangular marble blocks can be observed inside debris deposits and also bed of stream (Figure 4f).

According to the macroscopic and microscopic investigations, the marbles quarried at Domuzderesi are white, gray-veined and gray in color (Figure 5 a,b,c,d,e,f). Their minero-petrographic properties are given in Table 1. Domuzderesi marble types are almost identical to marbles quarried at Hierapolis in terms of minero-petrographic properties

Maximum Grain Size (MGS) measurement

In archaeometric studies, one of the most common used parameters in marble provenance analyses is the maximum grain size (MGS). Many authors have stated that MGS measurement, when combined with analysis results such as XRPD, XRF, EPR and C-O stable isotopes, is highly helpful for distinguishing

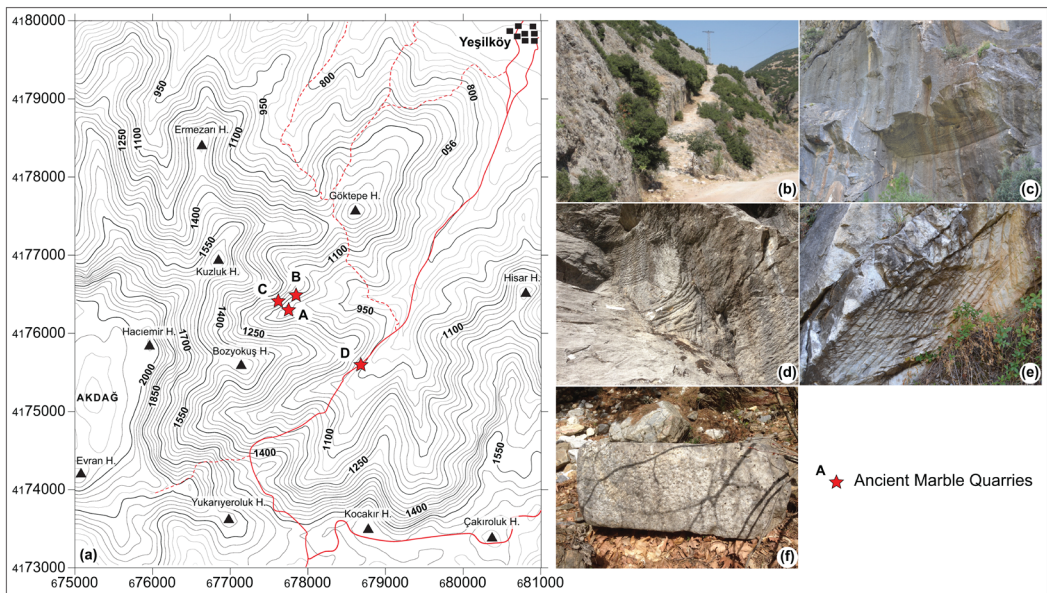


Figure 4. a) Topographic map of the Domuzderesi quarries; b) the well preserved remains of a Roman transport road; c), d), e) the working faces bearing cutting marks left by the pickaxes and the pointed chisels; f) Unfinished rectangular marble block in the bed of stream.

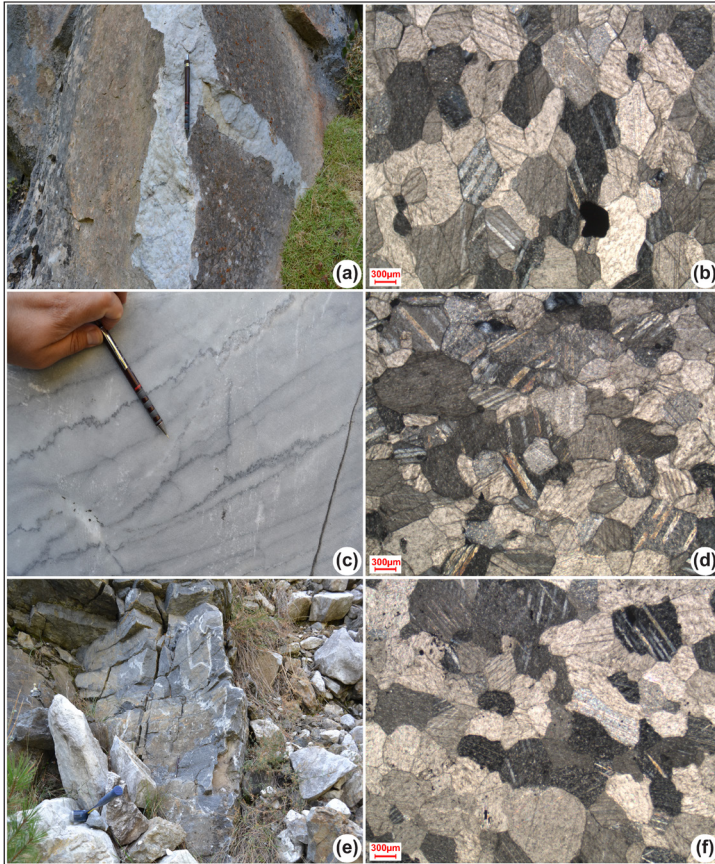


Figure 5. Field and microscopic photographs of the Domuzderesi marbles, a) white marble; b) homeoblastic polygonal texture and perfect polysynthetic twinning of carbonate minerals in white marble; c) gray-veined marble; d) heteroblastic mosaic texture in gray marble; e) gray marble; f) heteroblastic mosaic texture in gray marbles.

marble provenance (Hinterlechner-Ravnik and Moine, 1977; Gorgoni et al., 2002; Polikreti and Maniatis, 2000; Jarc et al., 2010; Barone et al., 2013). Attanasio et al. (2006) published the most comprehensive database of MGS for Mediterranean region. MGS is generally based on the microscopic examination of the thin sections. Since large numbers of samples needed to be measured, about 4000 MGS values (1500 for Hierapolis, 2500 for Domuzderesi marbles) were determined to compare each

other. The MGS measurements of the Hierapolis and Domuzderesi marbles are given in Figure 6, and the ranges in each group are showed in the statistical boxplots. The MGS of white marbles range between 318 and 3804 μm in the Hierapolis quarries, 287 and 1830 μm in the Domuzderesi quarries. The MGS values of gray-veined marbles ranging from 268 to 711 for Hierapolis quarries, 269 and 4154 for Domuzderesi quarries. The MGS of gray marbles is between 205 and 1319, 305 and

1961, respectively. According to Figure 6, the median values of marbles are very close to each other. In addition, there are many outlier values in Figure 6. These differences in MGS are probably related to the metamorphic history of investigated areas.

X-ray powder diffraction (XRPD) investigation

Although optic microscopic investigation is

always the first step to characterize the mineral and rock species, it is difficult to achieve a quantitative characterization of mineral phases. One of the most common methods used to determine of mineral phases of a rock (e.g. minerals, inorganic compounds) is powder diffractometry. Only some milligrams of the sampled material are required and it is particularly suitable when only powder samples are available.

Quarries / Marble Types		Maximum Grain Size (MGS) (µm)			
		Min.	Max.	Median	St. Dev.
Hierapolis Quarries	White	318	3804	953	508
	Gray-veined	268	711	555	119
	Gray	205	1319	542	182
Domuzderesi Quarries	White	287	1830	759	303
	Gray-veined	269	4154	856	476
	Gray	305	1961	845	303

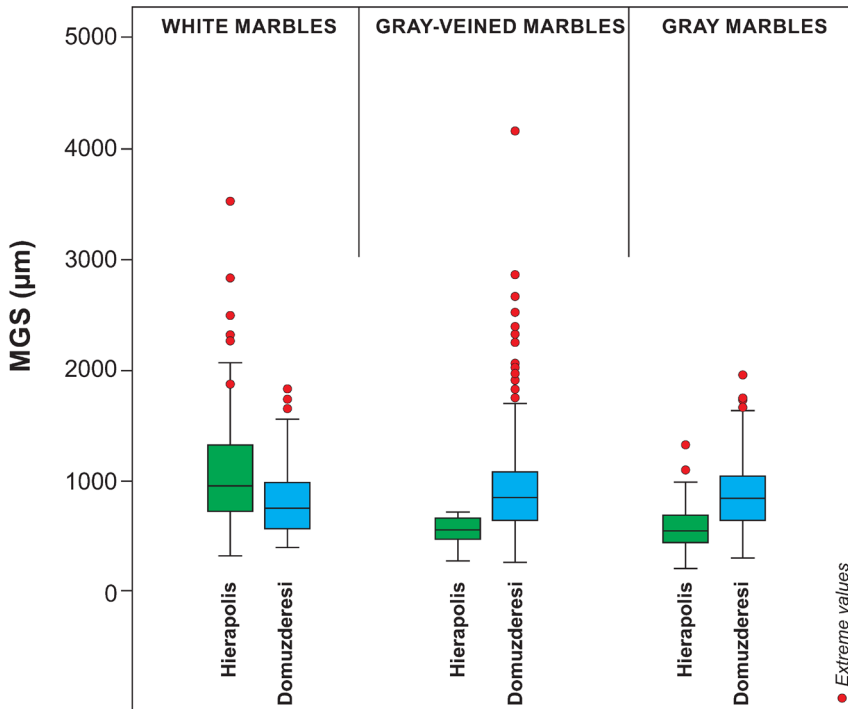


Figure 6. Boxplots of the MGS variable in the Hierapolis and Domuzderesi marble groups.

Currently, mineral content of less than 1 w% can be detected quantitatively. In addition, the use of XRPD is important in order to determine the content of dolomite.

According to the XRPD analyses, calcite \pm dolomite (in white marbles); calcite \pm dolomite (in gray-veined marbles) and calcite \pm quartz (in gray marbles) are detected (Figure 7). All marbles are predominantly composed of calcite, and lesser amounts of dolomite and quartz.

Geochemical analysis

Major and trace element analyzes (Tables 2, 3, 4) were conducted to highlight the differences or similarities in major and trace element trends between each quarries. The analyzed data for quarries are shown in the box-plot diagrams (Figure 8) largely used in archaeometric studies in order to illustrate the differences and/or similarities between multiple data (including

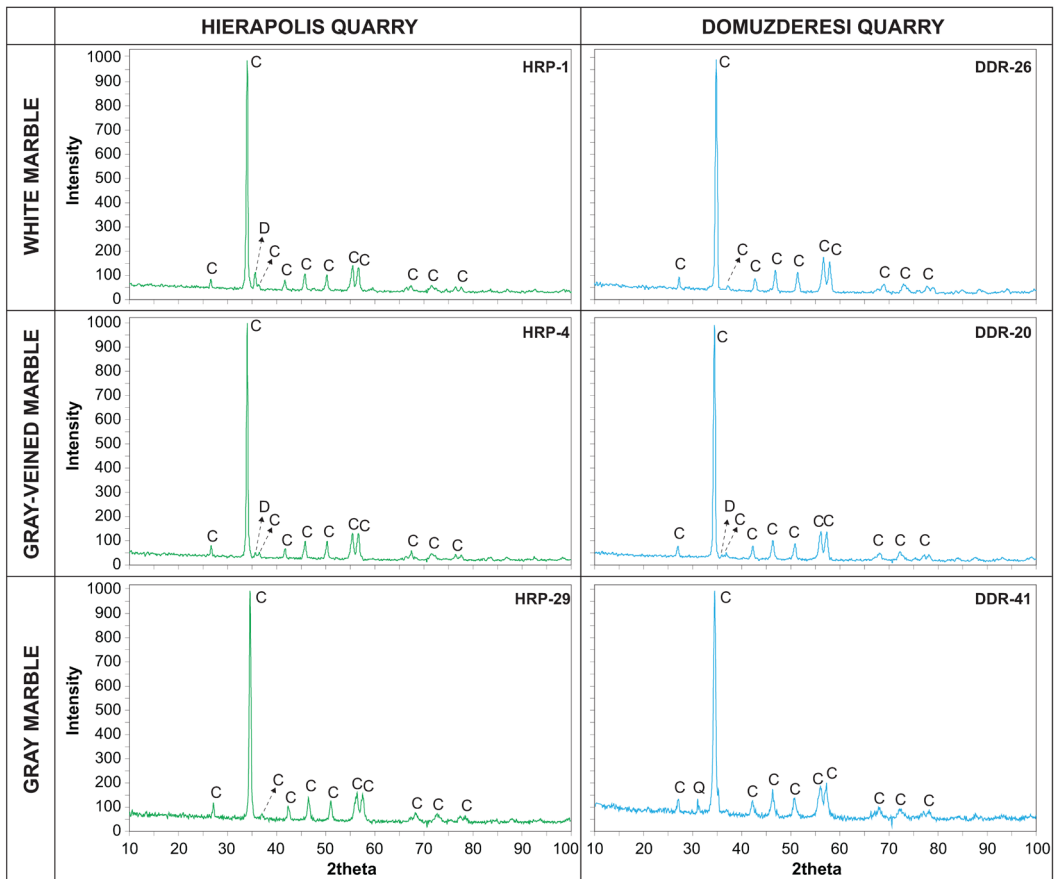


Figure 7. XRPD patterns of selected samples of white, gray-veined and gray marbles taken from the Hierapolis and Domuzderesi quarries (C = calcite, D = dolomite, Q = quartz).

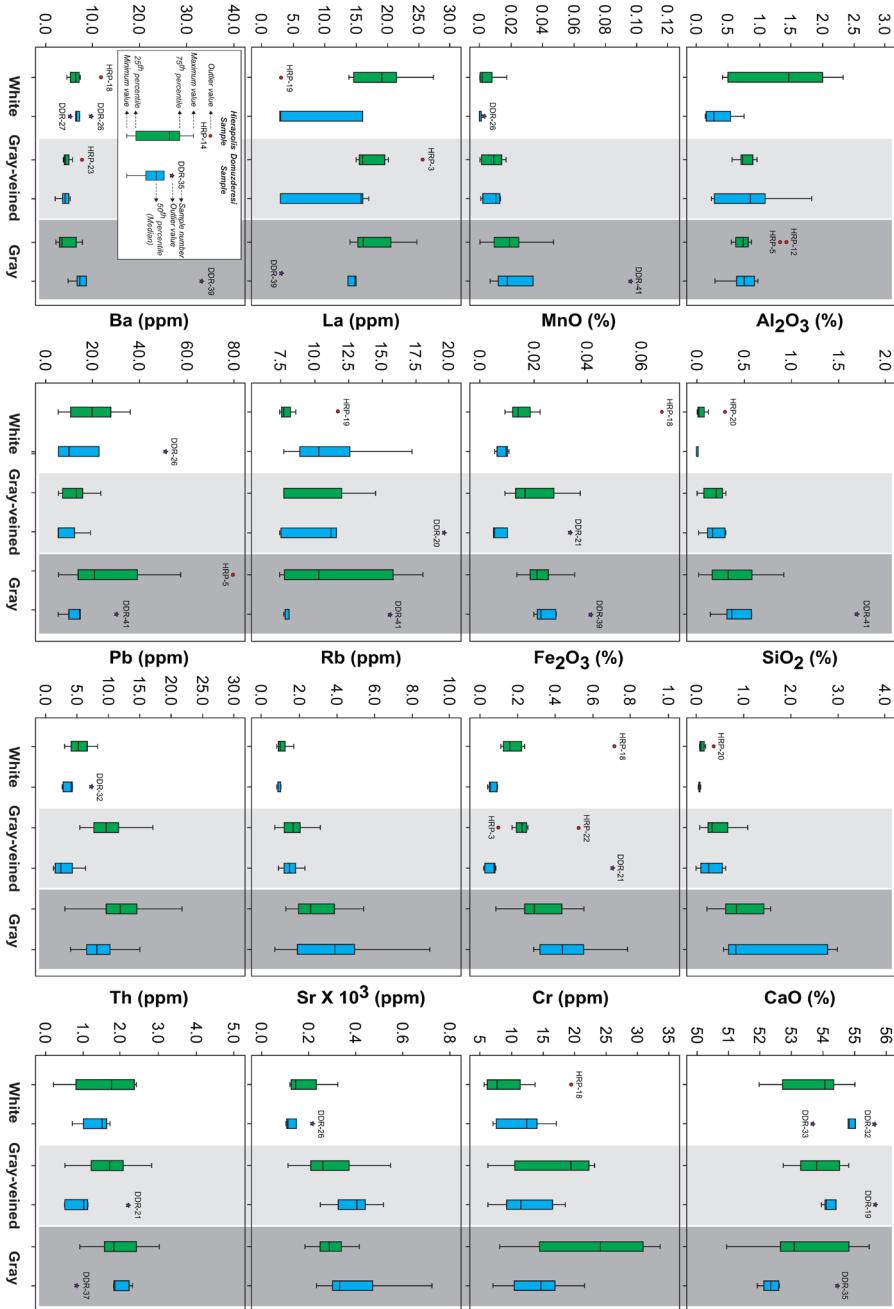


Figure 8. Boxplots of the analyzed major oxides and trace elements in the Hierapolis and Domuzderesi marble groups.

Table 2. Major oxide, trace and some rare earth element (REE) analyses of the white marbles of Hierapolis and Domuzderesi quarries.

Sample No	HRP-1	HRP-2	HRP-7	HRP-9	HRP-18	HRP-19	HRP-20	HRP-28	DDR-26	DDR-27	DDR-28	DDR-32	DDR-33
Na ₂ O	0.41	0.32	0.33	0.36	0.43	0.28	0.33	0.29	0.35	0.30	0.26	0.37	0.32
MgO	2.31	1.51	2.33	0.51	1.41	0.48	0.41	1.69	0.15	0.28	0.14	0.53	0.75
Al ₂ O ₃	0.01	0.02	-	0.03	0.12	-	0.29	-	-	-	0.01	-	-
SiO ₂	0.11	0.10	0.07	0.14	0.20	0.07	0.36	0.08	0.08	0.07	0.08	0.07	0.07
P ₂ O ₅	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
K ₂ O	0.04	0.04	0.03	0.03	0.06	0.02	0.05	0.04	0.03	0.03	0.03	0.03	0.03
CaO	52.96	54.53	51.96	55.01	54.02	54.08	54.14	52.48	55.03	54.82	54.80	55.62	53.65
TiO ₂	-	-	-	-	0.02	-	0.01	-	-	-	-	-	-
MnO	0.01	0.01	0.01	0.01	0.07	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01
Fe ₂ O ₃ *	0.20	0.13	0.24	0.12	0.71	0.14	0.18	0.11	0.04	0.05	0.05	0.09	0.09
LOI*	44.76	44.66	44.81	44.29	44.59	44.83	44.54	45.14	43.89	44.02	44.43	42.48	44.89
Total	100.86	101.37	99.82	100.55	101.67	99.98	100.38	99.89	99.63	99.62	99.84	99.26	99.85
V	8.9	10.8	7.0	8.7	14.3	6.0	10.5	8.3	11.3	5.2	5.4	5.8	6.3
Cr	6.6	13.7	5.6	9.0	19.4	6.7	8.6	5.6	14.0	17.1	7.6	12.5	7.0
Co	21.7	21.1	13.8	20.3	15.4	3.0	27.2	17.9	3.0	3.0	16.0	3.0	16.0
Ni	3.9	4.1	3.9	4.3	4.3	4.3	4.3	4.0	4.2	4.5	4.3	4.2	3.9
Cu	1.6	1.5	1.5	1.6	1.6	1.7	1.5	1.4	1.7	2.0	1.5	1.5	1.5
Rb	1.1	0.9	0.8	1.1	1.7	0.9	1.4	0.9	0.8	1.0	1.0	0.9	0.9
Sr	322.3	153.8	126.7	119.1	211.4	250.7	123.8	135.9	215.7	102.7	146.4	105.7	111.1
Y	6.7	4.5	6.8	4.6	11.7	5.8	7.3	6.0	9.4	5.3	7.1	6.5	6.4
Zr	6.6	6.2	10.1	6.2	6.6	7.3	6.3	5.2	9.8	6.0	6.1	5.6	5.8
Nb	6.0	3.8	3.9	4.2	7.4	4.1	4.0	3.9	4.1	3.7	4.1	3.6	3.6
Ba	36.0	26.1	15.9	29.3	16.3	5.5	5.4	23.4	51.1	10.0	5.5	22.7	5.5
La	8.6	7.5	7.6	7.8	7.8	11.8	7.6	7.4	8.9	7.7	12.6	10.3	17.2
Ce	17.8	14.5	10.0	16.1	16.1	10.0	15.9	11.2	10.0	10.0	12.9	9.7	23.9
Hf	4.1	4.2	4.8	4.8	4.3	5.0	4.8	4.4	4.7	5.2	4.6	4.4	4.6
Ta	3.4	3.5	3.7	3.6	3.4	3.7	3.8	3.4	3.0	3.8	3.8	3.5	3.3
Pb	8.2	6.3	3.0	3.9	6.9	6.0	4.2	4.3	2.6	4.2	2.8	7.1	4.1
Th	2.0	1.5	2.4	2.4	2.3	0.4	1.2	0.2	1.5	1.7	1.0	1.6	0.7
Mg/Ca (x10 ⁻²)	3.7	2.3	3.8	0.8	2.2	0.8	0.6	2.7	0.2	0.4	0.2	0.8	1.2
Mn/Sr	0.3	0.7	0.6	0.9	2.5	0.4	1.4	0.5	0.2	0.8	0.3	0.8	0.7
Sr/Ca (x10 ⁻³)	0.9	0.4	0.3	0.3	0.5	0.6	0.3	0.4	0.5	0.3	0.4	0.3	0.3

Fe₂O₃ = Total Fe₂O₃, LOI* = Loss On Ignition*

Table 3. Major oxide (%), trace (ppm) and some rare earth element (REE) analyses of the gray-veined marbles of Hierapolis and Domuzderesi quarries.

Sample No	HRP-3	HRP-4	HRP-10	HRP-21	HRP-22	HRP-23	HRP-24	DDR-15	DDR-19	DDR-20	DDR-21	DDR-22
Na ₂ O	0.33	0.34	0.36	0.29	0.31	0.26	0.29	0.38	0.33	0.31	0.35	0.34
MgO	0.70	0.70	0.56	0.84	0.95	0.72	0.94	0.84	0.28	0.22	1.09	1.82
Al ₂ O ₃	-	0.07	0.24	0.07	0.30	0.30	0.20	0.30	0.17	0.02	0.31	0.11
SiO ₂	0.08	0.31	0.72	0.19	1.09	0.62	0.35	0.55	0.28	0.10	0.63	-
P ₂ O ₅	0.05	0.05	0.05	0.05	0.07	0.08	0.05	-	0.05	0.05	0.05	-
K ₂ O	0.03	0.05	0.06	0.04	0.06	0.10	0.06	0.08	0.05	0.03	0.08	0.04
CaO	54.82	54.55	53.78	54.50	52.73	53.43	53.15	54.06	55.65	54.41	53.95	54.11
TiO ₂	-	-	0.01	-	0.01	0.02	0.01	0.01	0.01	-	0.01	-
MnO	0.01	0.01	0.04	0.02	0.03	0.02	0.02	0.01	-	0.01	0.03	-
Fe ₂ O ₃ *	0.10	0.17	0.22	0.22	0.53	0.25	0.24	0.08	0.02	0.02	0.70	0.08
LOI*	44.28	44.15	44.42	44.53	43.41	44.12	44.98	43.56	42.21	44.19	43.10	43.81
Total	100.39	100.41	100.46	100.76	99.50	99.92	100.28	99.94	99.06	99.36	100.31	100.38
V	8.8	8.7	15.8	10.4	17.2	18.5	12.9	17.3	9.4	9.1	13.5	6.4
Cr	6.2	12.4	23.2	8.6	19.3	21.5	23.1	18.5	16.5	6.2	9.2	11.4
Co	25.6	20.1	16.0	16.0	15.0	18.9	15.0	16.1	17.0	16.0	3.0	3.0
Ni	4.2	4.4	3.9	4.3	4.3	3.7	4.2	4.3	4.1	4.6	3.9	4.1
Cu	1.7	1.6	1.5	1.4	1.7	1.3	1.7	1.6	1.7	1.5	1.5	1.6
Rb	1.1	0.7	2.1	1.3	1.7	3.1	2.0	1.8	1.2	0.9	2.3	1.5
Sr	111.1	545.0	250.4	287.6	452.9	164.9	261.7	324.9	438.7	404.0	246.7	1515.0
Y	5.7	4.1	4.0	3.9	4.0	7.7	3.8	4.9	4.1	3.6	5.2	2.0
Zr	10.6	12.6	6.4	7.4	8.8	9.0	7.6	8.2	8.4	7.9	12.8	13.0
Nb	4.2	6.0	3.8	4.0	3.8	5.0	3.9	3.8	4.1	3.7	3.7	3.7
Ba	23.5	14.0	17.5	5.5	13.1	9.0	5.6	19.1	12.3	5.4	5.4	5.5
La	12.7	7.7	11.3	7.7	7.7	14.5	7.7	11.6	11.2	19.6	7.5	7.4
Ce	12.6	10.0	14.1	10.0	11.0	12.4	16.7	13.6	16.9	10.0	10.0	10.0
Hf	4.7	4.9	4.5	4.6	4.9	4.4	5.1	4.8	6.5	4.7	4.9	5.1
Ta	3.4	3.9	3.6	3.9	3.6	2.8	4.1	3.8	5.4	3.9	3.6	3.8
Pb	5.4	17.0	13.1	9.0	10.0	9.5	6.3	4.2	2.4	1.2	6.3	1.5
Th	1.0	1.4	2.8	1.9	1.7	0.5	2.2	0.5	1.1	1.0	2.2	0.5
Mg/Ca (x10 ⁻²)	1.1	1.1	0.9	1.3	1.5	1.1	1.5	1.3	-	0.3	1.7	-
Mn/Sr	0.7	0.1	1.1	0.4	0.6	1.0	0.5	0.2	0.1	0.1	1.1	-
Sr/Ca (x10 ⁻³)	0.3	1.4	0.7	0.7	1.2	0.4	0.7	0.8	1.1	1.0	0.6	3.9

Fe₂O₃* = Total Fe₂O₃, LOI* = Loss On Ignition

Table 4. Major oxide (%), trace (ppm) and some rare earth element (REE) analyses of the gray marbles of Hierapolis and Domuzdereesi quarries.

Sample No	HRP-5	HRP-6	HRP-8	HRP-11	HRP-12	HRP-13	HRP-14	HRP-15	HRP-16	HRP-17	HRP-29	DDR-35	DDR-37	DDR-39	DDR-41	DDR-43
Na ₂ O	0.40	0.27	0.33	0.40	0.32	0.29	0.27	0.37	0.32	0.30	0.26	0.33	0.25	0.34	0.37	0.29
MgO	1.34	0.56	0.73	0.68	1.43	0.61	0.63	0.75	0.55	0.87	0.76	0.63	0.97	0.29	0.91	0.75
Al ₂ O ₃	0.22	0.92	0.15	0.75	0.45	0.17	0.36	0.72	0.02	0.33	0.14	0.14	0.37	0.58	1.69	0.32
SiO ₂	0.44	1.59	0.54	1.48	1.58	0.71	0.84	1.39	0.23	1.27	0.80	0.58	2.98	0.84	2.78	0.69
P ₂ O ₅	0.05	0.06	0.06	0.07	0.07	0.07	0.07	0.07	0.05	0.08	0.08	0.05	0.04	0.12	0.07	0.08
K ₂ O	0.07	0.17	0.06	0.13	0.12	0.06	0.10	0.12	0.03	0.08	0.08	0.04	0.13	0.11	0.26	0.06
CaO	55.46	51.61	55.25	53.09	50.92	55.09	52.73	52.64	54.56	52.65	53.96	54.45	51.90	52.58	52.10	52.34
TiO ₂	0.02	0.05	-	0.04	0.02	0.01	0.02	0.03	-	0.02	0.01	0.01	0.03	0.02	0.10	0.01
MnO	0.02	0.04	0.02	0.03	0.02	0.02	0.02	0.02	0.02	0.03	0.01	0.02	0.03	0.04	0.02	0.02
Fe ₂ O ₃ *	0.29	0.45	0.25	0.55	0.42	0.21	0.24	0.39	0.24	0.52	0.08	0.32	0.28	0.55	0.78	0.43
LOI*	42.65	43.64	43.08	43.49	43.85	43.49	43.79	43.50	44.93	43.19	44.35	43.35	43.58	43.87	41.60	44.73
Total	100.95	99.36	100.47	100.71	99.22	100.73	99.06	99.99	100.94	99.34	100.53	99.91	100.57	99.34	100.67	99.72
V	23.6	31.1	33.0	35.4	15.9	24.7	21.3	17.3	12.0	16.5	13.6	9.7	12.6	35.8	27.9	16.2
Cr	32.1	30.1	23.7	33.6	31.7	26.9	24.1	15.7	13.1	9.7	8.1	7.0	16.9	10.4	21.6	14.6
Co	18.6	22.9	24.6	15.0	14.0	16.0	15.0	15.5	19.9	21.1	16.0	14.8	15.0	3.0	15.0	13.7
Ni	4.2	3.9	3.9	3.9	3.7	4.2	4.7	3.9	4.4	3.9	4.5	4.3	3.5	3.8	4.1	4.0
Cu	1.7	1.6	1.4	6.8	1.5	1.5	1.9	1.1	1.5	1.6	3.1	1.6	1.6	1.6	1.5	1.5
Rb	2.0	5.4	1.9	4.0	4.2	1.9	3.3	3.7	1.3	2.6	2.3	0.7	4.9	3.9	8.9	1.9
Sr	296.9	246.5	413.5	285.5	312.1	249.3	251.3	220.8	182.8	364.2	411.0	469.1	1032.0	721.2	299.6	329.6
Y	3.4	6.0	2.2	7.5	3.5	2.6	3.4	4.2	2.2	6.9	7.7	4.8	6.7	32.9	8.6	7.2
Zr	7.4	6.0	7.4	13.7	7.5	6.8	7.7	8.7	6.7	8.6	8.5	8.4	28.4	9.7	21.8	7.3
Nb	4.1	3.5	3.6	3.9	4.0	3.5	4.3	3.7	4.1	4.3	4.2	3.9	3.8	4.1	4.3	3.9
Ba	79.6	37.3	9.0	23.3	11.9	57.4	40.8	15.8	20.8	5.5	17.4	5.4	14.8	14.4	30.2	9.9
La	10.3	7.4	7.6	17.8	9.7	15.6	7.8	16.0	7.7	11.3	18.0	8.1	7.8	7.7	15.6	7.8
Ce	11.6	12.0	10.0	31.1	10.0	11.1	11.1	24.2	12.6	11.0	11.0	12.1	12.0	28.1	16.8	10.0
Hf	4.8	4.4	4.9	5.2	4.6	5.0	5.4	4.5	5.0	4.6	5.1	4.9	4.6	5.1	4.7	4.6
Ta	3.8	4.4	3.9	4.6	3.9	3.9	4.3	4.0	3.6	3.6	4.4	3.7	3.7	3.6	3.7	3.5
Pb	13.8	21.6	9.9	18.9	11.8	9.3	15.1	13.2	8.8	10.4	3.0	3.9	6.5	14.9	10.2	8.1
Th	1.6	3.0	1.5	2.5	2.6	1.8	0.9	1.8	1.7	2.3	1.0	1.8	0.8	2.2	2.3	1.8
Mg/Ca (x10 ⁻²)	2.0	0.9	1.1	1.1	2.4	0.9	1.0	1.2	0.8	1.4	1.2	1.0	1.6	0.5	1.5	1.2
Mn/Sr	0.5	1.1	0.3	0.7	0.6	0.6	0.7	0.6	0.9	0.7	0.3	0.4	0.2	0.4	0.5	0.5
Sr/Ca (x10 ⁻³)	0.7	0.7	1.0	0.8	0.9	0.6	0.7	0.6	0.5	1.0	1.1	1.2	2.8	1.9	0.8	0.9

$Fe_2O_3^* = Total Fe_2O_3$, $LOI^* = Loss On Ignition$

MGS, geochemical analyse results, C-O isotopes and EPR etc.). In general, white, gray-veined and gray marbles of two quarries show similar compositional spreads in terms of major oxide and trace elements. It is clear from the Figure 8 that the Al_2O_3 , SiO_2 , TiO_2 , Fe_2O_3 , Rb, Sr and Pb values of white marbles are lower than gray marbles. However, MnO, Co, Pb and Th contents of Hierapolis marbles are higher than the Domuzderesi marbles. In terms of other elements (MgO, Al_2O_3 , SiO_2 , CaO, TiO_2 , Fe_2O_3 , Cr, La, Rb, Sr, Y, Ba and Th), there are negligible differences between the Hierapolis and Domuzderesi ancient quarries.

Plots of Mg/Ca versus SiO_2 , Ca(%) versus Mg(%) and Sr versus Mg/Ca are useful for identifying protolith of marbles (Melezhik et al., 2008) (Figure 9). All marbles are placed on the limestone field (Figure 9 a,b,c). In addition, SiO_2 values of some gray-veined marbles are higher than %1 (Figure 9a). This can be associated with individual quartz grains which were determined during the petrographic examinations of marbles. According to these diagrams, the protolith of Hierapolis and Domuzderesi marbles should be limestone.

Average Phanerozoic Limestone (APL) (Condie et al., 1991) normalized multi-element variation diagram was prepared to compare the trace element contents of the white, gray-veined and gray marbles (Figure 10 a,b,c). On an APL-normalized multi-element variation diagram, the elemental patterns of marbles display slightly enrichment in Y, La, Ce and Nb elements. Although they are close to the APL values, there are clear decreases in the content of Rb, Mn, Ni, Ti, Pb, Zr, Fe, Ba, Al and Sr (Figure 10 a,b,c).

As a result, the geochemical results suggest that the marbles of Hierapolis and Domuzderesi quarries have similar geochemical compositions.

Discrimination Function Analysis (DA) on marble quarries

As the geochemical analysis results are quite

similar in two marble quarries, discriminant function analysis (DA) was performed to distinguish marble groups. The main goal of DA is to find a component(s) that groups differ on and produce classification functions. As a result of DA, discriminant functions are obtained in order to define the relationships across groups. We can also visualize how the two functions discriminate between groups by plotting the individual scores for the discriminant functions. DA is one of the most widely used statistical analyses in Earth science (Attanasio, 2003). Because marble samples were not systematically collected from Hierapolis and Domuzderesi quarries, geochemical analyses used for the DA, have been considered as non-parametric and non-homogeneous data in terms of statistical science. The "SPSS" statistical software package was used for the DA and Kruskal-Wallis H test was chosen. The test results are presented in Table 5 in which the value of p is less than 0.05 in 95% confidence intervals. According to this assumption, there were 27 elements, 16 of which (Al_2O_3 , SiO_2 , P_2O_5 , K_2O , TiO_2 , MnO, Fe_2O_3 , V, Cr, Co, Rb, Sr, Y, Zr, Ta, Pb) were found to be significant. Eigenvalues, % variance, % cumulative eigenvalue and canonical correlation coefficients, which are calculated from DA, are shown in Table 6. The functions 1 and 2 have high canonical correlation coefficients. Therefore, a binary diagram in which the functions 1 and 2 were used was designed to see the relationship between marble quarries (Figure 11). The marble types of Hierapolis and Domuzderesi quarries are successfully separated in this diagram. In terms of chemical composition, white and gray-veined marbles are highly similar to each other; however, gray marbles display slightly differences compared to white and gray-veined marbles.

C-O isotopes of Marble Quarries

C-O isotope analyses are widely utilized in

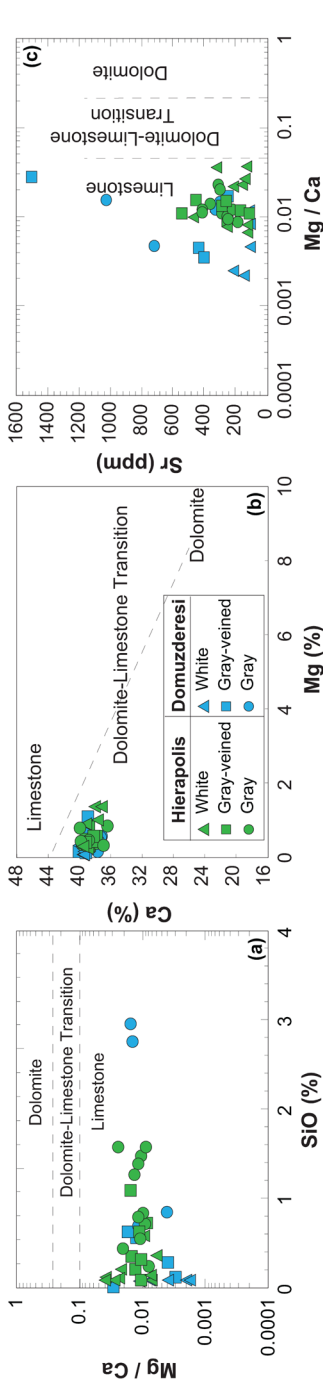


Figure 9. Chemical nomenclature diagrams for the Hierapolis and Domuzderesi marble groups a) the (Mg/Ca)-SiO₂ diagram, b) Ca - Mg diagram, c) the Sr - Mg/Ca diagram.

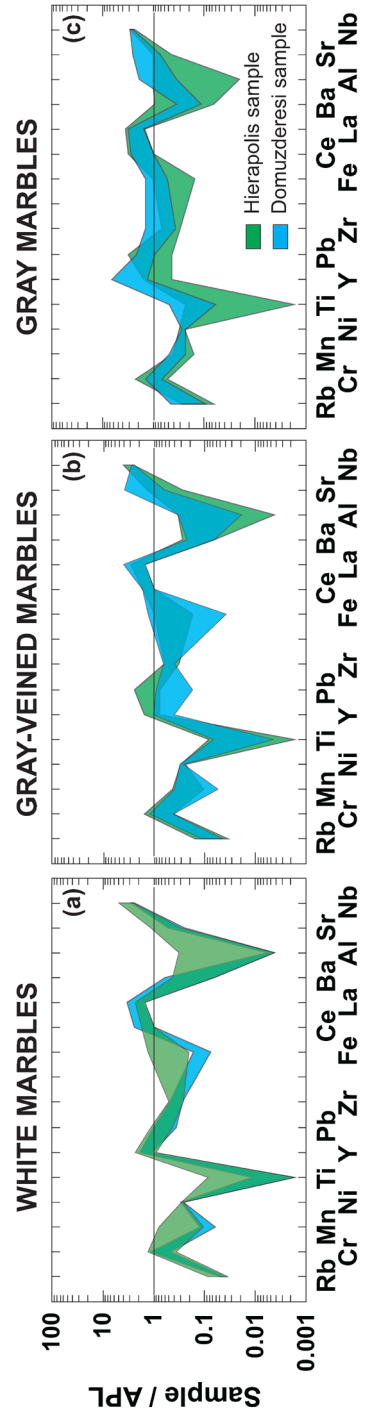


Figure 10. Average Phanerozoic Limestone (APL) (Condie et al., 1991) normalized elemental patterns of a) white, b) gray-veined, c) gray marbles.

Table 5. Kruskal-Wallis H test results of marbles from Hierapolis and Domuzderesi quarries.

Elements	Chi-Square	p value	Elements	Chi-Square	P value
Na ₂ O	3.56	.6136	Cu	0.54	.9908
MgO	7.96	.1587	Rb	20.21	.0011
Al₂O₃	23.32	.0003	Sr	19.78	.0014
SiO₂	27.18	.0001	Y	14.15	.0147
P₂O₅	18.13	.0028	Zr	15.81	.0074
K₂O	21.93	.0005	Nb	6.91	.2277
CaO	9.72	.0834	Ba	7.25	.2025
TiO₂	16.45	.0057	La	4.83	.4372
MnO	17.02	.0045	Ce	2.79	.7325
Fe₂O₃	20.11	.0012	Hf	6.51	.2599
V	23.65	.0003	Ta	15.35	.0090
Cr	13.41	.0199	Pb	22.44	.0004
Co	11.45	.0431	Th	5.78	.3281
Ni	3.01	.6980			

determining provenance of classical marbles. Craig and Craig (1972), for the first time, studied about carbon and oxygen isotopes on archaeological site. In the following years, Manfra (1975), German et al. (1980) and Herz (1985, 1987, 1992) developed the isotope database of marbles. The most cited study among them belongs to Herz (1987). In his study, Herz generated extensive and detailed C and O isotope database for white marbles from all the major marble quarries in the classical lands of the eastern Mediterranean (Italy, Greece, Turkey and Tunisia).

Hierapolis and Domuzderesi marbles have been subdivided into three different groups in terms of macroscopic and microscopic observations and geochemical investigations.

Total of 39 samples (21 samples from Hierapolis, 18 samples from Domuzderesi quarry) were analyzed for $\delta^{13}\text{C}$ - $\delta^{18}\text{O}$ isotope composition, and the results are summarized in Table 7.

For Hierapolis quarries, $\delta^{13}\text{C}$ values range from 1.78 to 3.52‰ (average 2.68‰, n = 7) in white marbles, from 0.63 to 3.08‰ (average 2.14‰, n = 7) in gray-veined marbles and from 0.90 to 3.12‰ (average 2.63‰, n = 7) in gray marbles. For $\delta^{18}\text{O}$ values ranging from (-9.35) to (-7.32)‰ (average -8.16‰, n = 7), (-9.55) to (-5.76)‰ (average -7.04‰, n = 7) and (-7.24) to (-1.21)‰ (average -5.55‰, n = 7), respectively. In Domuzderesi quarries, $\delta^{13}\text{C}$ values change between 0.32 and 2.51‰ (average 1.72‰, n = 6) in white marbles, (-1.44)-2.03‰ (average 0.22 ‰, n = 6) in gray-veined marbles and

Table 6. Eigenvalues, % variance, % cumulative eigenvalue and canonical correlation coefficients values which are calculated from DA.

Function	Eigenvalue	% of Variance	Cumulative %	Canonical Correlation
1	10.123	61.3	61.3	.954
2	2.372	14.4	75.7	.839
3	2.117	12.8	88.5	.824
4	1.127	6.8	95.4	.728
5	0.766	4.6	100.0	.658

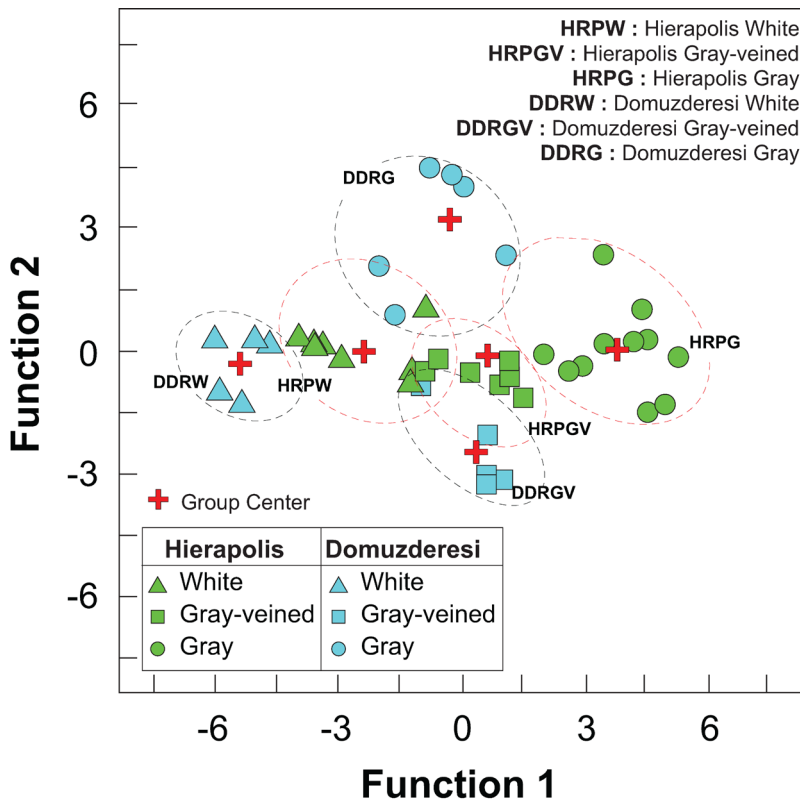


Figure 11. Discriminant function graphic showing the relationship between the marbles of Hierapolis and Domuzderesi quarries.

1.64-3.41‰ (average 2.67‰, n = 6) in gray marbles. For $\delta^{18}\text{O}$ values vary between (-8.70)-(-7.10)‰ (average -7.94‰, n = 6), (-9.42)-(-7.00)‰ (average -7.98‰, n = 6), and (-13.26)-(-5.30)‰ (average -7.78‰, n = 6), respectively.

Generally, the C and O isotope values of Hierapolis and Domuzderesi marbles display similar compositional spread compared with the each other. In addition, marbles have relatively negative value of their isotopic oxygen ratio. Furthermore, gray marbles show wide range of O isotopic composition. All marbles are also plotted on the conventional $\delta^{13}\text{C} - \delta^{18}\text{O}$ isotope diagram (Figure 12) in which Hierapolis and Domuzderesi marbles fall close to each other, thereby showing a congenerical isotopic composition.

Conclusions

The studied quarries are located at Denizli province, which is situated in western part of Turkey which was named by archaeologists as “Phrygia region” in the Asia Minor. Phrygia region is highly significant in terms of archaeology and geology sciences. According to the archaeology, there are many remains of protohistoric civilizations throughout the province (Figure 13). Hierapolis, Laodicea and Colossae are notably significant ancient cities among these civilizations, as they have many civilization buildings, such as temples, stadiums, theaters, health complexes and advanced infrastructure (e.g., canalization) systems. Natural stones such as marble, travertine and sandstone have been widely used in these buildings. For the reason mentioned above, Phrygia region can truly be called a complete “Archaeological Park”. In terms of geoscience, Phrygia region is tectonically one of the most active regions. Many cities since ancient times were damaged or destroyed by repeated tectonic activities (earthquakes) in the Phrygia

region (Altunel and Barka, 1996; Hancock and Altunel, 1997; Hancock et al., 1999; 2000; Akyüz and Altunel, 2001; Kaymakçı, 2006; Piccardi, 2007; Hançer, 2013; Topal and Özkul, 2014). In long time period, people accepted to live with earthquake phenomenon. After that, they chose to repair their cities instead of move to another place. In the process of repair and reconstruction, people extracted building stones from both current and new quarries. Today, signs of repairing and reconstruction processes are clarified in archaeometry studies conducted in the ancient cities.

In this context, we provide information about mineralogy, petrography, geochemistry and C, O isotopes of marbles collected from the Hierapolis and Domuzderesi ancient quarries. All findings can be summarized as follows;

- i) The marble samples from Hierapolis and Domuzderesi quarries have been classified into three groups, based on color, crystal size, crystal boundaries and foliation status. These groups are identified as white, gray-veined and gray marbles.
- ii) In terms of microscopic features, heteroblastic polygonal (white marbles in Hierapolis, gray-veined and gray marbles in Domuzderesi) and homeoblastic polygonal textures (white marbles in Domuzderesi) have been defined.
- iii) Microscopic investigations revealed that mineral composition of marble groups in two quarries are quite similar. They are consisting predominantly of carbonate minerals (calcite, dolomite) and, as accessory minerals quartz, muscovite, pyroxene and ferric iron oxides. According to the XRPD investigation, calcite ± dolomite (in white), calcite ± dolomite (in gray-veined) and calcite (in gray) have been recognized in Hierapolis marbles. In Domuzderesi marbles, calcite (in white), calcite ± dolomite (in gray-veined) and calcite ± quartz (in gray) have been detected.

Table 7. The results of C-O isotopic analysis of marbles from Hierapolis and Domuzderesi quarries.

	HIERAPOLIS MARBLES			DOMUZDERESI MARBLES		
	Sample No	$\delta^{13}\text{C}_{\text{V-PDB}} (\text{‰})$	$\delta^{18}\text{O}_{\text{V-PDB}} (\text{‰})$	Sample No	$\delta^{13}\text{C}_{\text{V-PDB}} (\text{‰})$	$\delta^{18}\text{O}_{\text{V-PDB}} (\text{‰})$
WHITE MARBLES	HRP-1	3.22	-8.62	DDR-25	1.19	-7.83
	HRP-2	2.57	-8.16	DDR-26	0.32	-8.70
	HRP-7	3.52	-7.32	DDR-27	2.54	-8.35
	HRP-9	2.15	-8.22	DDR-28	1.49	-7.10
	HRP-18	2.09	-9.35	DDR-32	2.51	-8.39
	HRP-19	1.78	-7.82	DDR-33	2.25	-7.27
	HRP-28	3.45	-7.65			
GRAY-VEINED MARBLES	HRP-3	0.69	-7.67	DDR-15	-1.44	-8.53
	HRP-4	0.63	-9.55	DDR-16	1.03	-7.30
	HRP-10	2.66	-5.76	DDR-19	-0.05	-7.83
	HRP-21	2.87	-6.40	DDR-20	0.10	-7.82
	HRP-22	2.66	-6.93	DDR-21	-0.37	-9.42
	HRP-23	2.40	-6.97	DDR-22	2.03	-7.00
	HRP-24	3.08	-6.00			
GRAY MARBLES	HRP-5	3.02	-7.24	DDR-35	2.41	-5.67
	HRP-8	3.06	-4.53	DDR-37	2.66	-13.26
	HRP-11	2.41	-6.19	DDR-39	1.64	-9.51
	HRP-12	3.11	-6.37	DDR-41	3.14	-6.39
	HRP-15	3.12	-6.49	DDR-42	3.41	-5.30
	HRP-16	2.77	-6.84	DDR-43	2.73	-6.52
	HRP-29	0.90	-1.21			

iv) In general, white, gray-veined and gray marbles from Hierapolis and Domuzderesi quarries show similar compositional spreads in terms of major oxide and trace elements. Average Phanerozoic Limestone normalized multi-element variation diagram, the elemental patterns of Hireapolis and Domuzderesi marbles are highly similar to

each other. In addition, low Mg/Ca ratio (< 0.1) and high Ca value (> %34) of marbles supports a limestone protoliths.

v) The C and O isotope values of Hierapolis and Domuzderesi marbles are highly close to each other. Furthermore, they are characterized by negative isotopic oxygen ratio. On the $\delta^{13}\text{C} - \delta^{18}\text{O}$ isotope diagram,

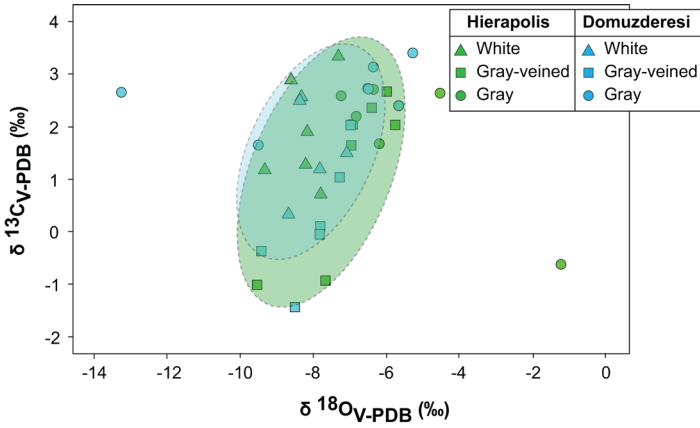


Figure 12. Distributions of the Hierapolis and Domuzdersi marbles on the conventional $\delta^{13}\text{C} - \delta^{18}\text{O}$ isotope diagram.



Figure 13. Protohistoric civilizations throughout the Denzli region.

Hierapolis and Domuzderesi marbles fall close to each other, thereby showing a congenerical isotopic composition. Based on these isotopic compositions, Hierapolis and Domuzderesi marbles belong to similar geological units in Denizli region.

- vi) Mineropetrographic, geochemical and C-O stable isotope results reveal that Hierapolis and Domuzderesi ancient quarries have similar characteristics which have originated from a same protolith.

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References

- Akyüz S. and Altunel E. (2001) - Geological and archaeological evidence for post-Roman earthquake surface faulting at Cibyra, SW Turkey. *Geodynamica Acta*, 14, 95-101.
- Alçıçek H., Varol B. and Özkul M. (2007) - Sedimentary facies, depositional environments and palaeogeographic evolution of the Neogene Denizli Basin of SW Anatolia, Turkey. *Sedimentary Geology*, 202, 596-637.
- Altunel E. and Barka A. (1996) - Evaluation of archaeoseismic damages at Hierapolis. *Geological Bulletin of Turkey*, 39, 65-74 (in Turkish).
- Attanasio D. and Pensabene P. (2002) - I marmi del teatro di Hierapolis, in Hierapolis IV, 69-85.
- Attanasio D. (2003) - Ancient white marbles: analysis and identification by paramagnetic resonance spectroscopy. *L'Erma di Bretschneider-Rome*, 263 pp.
- Attanasio D., Brilli M. and Ogle N. (2006) - Turkish Marble Quarries: Western Anatolia. In *The Isotopic Signature of Classical Marbles*, L'Erma di Bretschneider-Rome, 336 pp.
- Barone G., Bruno N., Giuffrida A., Mazzoleni P., Raneri S. (2013) - Archaeometric investigation of a Late Roman marble statue from Kaucana (RG) with considerations on the diffusion of Thasos marble in Sicily. *Periodico di Mineralogia*, 82, 2, 313-329.
- Bingöl O., (2004) - Arkeolojik Mimaride Taş (in Turkish). *Magnesia Kazıları Arkeoloji, Kültür Sanat, Dizisi: 1*, Homer Kitabevi ve Yayıncılık Ltd. Şti., 181 pp.
- Bozcu M. (2009) - Geology of Neogene basins of Buldan-Sarıcaova region and their importance in Western Anatolia neotectonics. *International Journal of Earth Sciences*, 99(4), 851-861.
- Condie KC., Wilks M., Rosen DM. and Zlobin V. (1991) - Geochemistry of metasediments from the Precambrian Hapschan Series, Eastern Anabar Shield, Siberia. *Precambrian Research*, 50, 37-47.
- Coplen TB., Kendall C. and Hopple J. (1983) - Comparison of stable isotope reference samples. *Nature*, 302, 236-238.
- Erten H., Şen Ş. and Görmüş M. (2014) - Middle and late Miocene Cricetidae (Rodentia, Mammalia) from Denizli Basin (southwestern Turkey) and a new species of Megacricetodon. *Journal of Paleontology*, 88, 3, 504-518.
- Germann K., Holzmann G. and Winkler FJ. (1980) - Determination of marble provenance: limits of isotope analysis. *Archaeometry*, 22, 99-106.
- Gorgoni C., Lazzarini L., Pallante P. and Turi B. (2002) - An updated and detailed mineropetrographic and C-O stable isotopic reference database for the main Mediterranean marbles used in antiquity in ASMOSIA V, interdisciplinary studies on ancient stone: proceedings of the 5th ASMOSIA Conference, Boston, 12-15 June 1998 (eds. J. Herrmann, N. Herz and R. Newman), 115-131, Archetype, London.
- Gökgöz A. (1998) - Geochemistry of the Kızıldere-Tekkehamam-Buldun-Pamukkale geothermal fields, Turkey. In: Georgsson L.S. (ed.). *Geothermal Training in Iceland 1998*, United Nations University Geothermal Training Programme, Reykjavik, Iceland, 115-156.
- Hancock PL. and Altunel E. (1997) - Faulted

- archaeological relics at Hierapolis, Turkey. *Journal of Geodynamics*, 24, 21-36.
- Hancock P.L., Chalmers R.M.L., Altunel E. and Çakir Z. (1999) - Travertines: using travertines in active fault studies. *Journal of Structural Geology* 21, 903-916.
- Hancock P.L., Chalmers R.M.L., Altunel E., Çakir Z. and Becher-Hancock A. (2000) - Creation and destruction of travertine monumental stone by earthquake faulting at Hierapolis, Turkey. In: McGuire, W.G., Griffiths, D.R., Hancock, P.L., Stewart, I.S. (Eds.), *The Archaeology of Geological Catastrophes: The Geological Society, London. Special Publications*, 171, 1-14.
- Hançer M. (2013) - Study of the Structural Evolution of the Babadağ-Honaz and Pamukkale Fault Zones and the Related Earthquake Risk Potential of the Buldan Region in SW Anatolia, East of the Mediterranean. *Journal of Earth Science*, 24, 3, 397-409.
- Craig H. and Craig V. (1972) - Greek Marbles: Determination of Provenance by Isotopic Analysis. *Science*, 176, 401-403.
- Herz N. (1985) - Isotopic analysis of marble, in *Archaeological Geology*. ed. G. Rapp, Jr., and J.A. Gifford (New Haven: Yale University Press), 331-351.
- Herz N. (1987) - Carbon and Oxygen Isotopic Ratios: A Data Base for Classical Greek and Roman Marble. *Archaeometry*, 29, 35-43.
- Herz N. (1992) - Provenance determination of neolithic to classical Mediterranean marbles by stable isotopes. *Archaeometry*, 34, 185-194.
- Hinterlechner-Ravnik A. and Moine B. (1977) - Geochemical characteristics of the metamorphic rocks of the Pohorje Mountains. *Geologija*, 20, 107-140.
- Jarc S., Maniatis Y., Dotsika E., Tambakopoulos D. and Zupancic N. (2010) - Scientific Characterization of the Pohorje Marbles, Slovenia. *Archaeometry*, 52, 177-190.
- Kaymakçı N. (2006) - Kinematic development and paleostress analysis of the Denizli Basin (Western Turkey): implications of spatial variation of relative paleostress magnitudes and orientations. *Journal of Asian Earth Sciences*, 27, 207-222.
- Koçyiğit A. (2005) - The Denizli graben-horst system and the eastern limit of western Anatolian continental extension: basin-fill, structure, deformational mode, throw amount and episodic evolutionary history, SW Turkey. *Geodinamica Acta*, 18, 167-208.
- Konak N., Akdeniz N. and Öztürk E.M. (1987) - Geology of the south of Menderes Massif. Guide Book for the Field Excursion along Western Anatolia, IGCP project no 5, 42-53.
- Koralay T., Kadioğlu Y.K. and Jiang S.Y. (2013) - Determination of Tourmaline Composition in Pegmatite from Buldan - Denizli (Western Anatolia-Turkey) using XRD, XRF and Confocal Raman Spectroscopy. *Spectroscopy Letters*, 46, 499-506.
- Malacrino C.G. (2010) - Constructing the Ancient World: Architectural Techniques of the Greeks and Romans. J. Paul Getty Museum-Los Angeles, 216 pp.
- Manfra L., Masi U. and Turi B. (1975) - Carbon and Oxygen isotope ratios of marbles from some ancient quarries of Western Anatolia and their archeological significance. *Archaeometry*, 17, 215-221.
- Melezhik V.A., Bingen B., Fallick A.F., Gorokhov I.M., Kuznetsov A.B., Sandstad J.S., Solli A., Bjerkgaard T., Henderson I., Boyd R., Jamal D. and Moniz A. (2008) - Isotope chemostratigraphy of marbles in northeastern Mozambique apparent depositional ages and regional implications. *Precambrian Research*, 162, 540-558.
- Monna D. and Pensabene P. (1977) - Marmi dell'Asia Minore, Consiglio Nazionale Delle Ricerche, Scienze Sussidiarie dell'Archeologia-Rome, 273 pp.
- Okay İ.A. (1989) - Geology of the Menderes Massif and the Lycian Nappes South of Denizli, Western Taurides. *Bulletin of the Mineral Research and Exploration*, 109, 37-51.
- Özkul M., Varol B. and Alçiçek M.C. (2002) - Depositional environments and petrography of the Denizli travertines. *Bulletin of the Mineral Research and Exploration*, 125, 13-29.
- Özkul M., Kele S., Gökgöz A., Shen C.C., Jones B., Baykara M.O., Förizs I., Németh T., Chang Y.W. and Alçiçek M.C. (2013) - Comparison of the Quaternary travertine sites in the Denizli extensional basin based on their depositional and geochemical data. *Sedimentary Geology*, 294, 179-204.
- Piccardi L. (2007) - The AD 60 Denizli Basin earthquake and the apparition of Archangel Michael at Colossae (Aegean Turkey). In Myths and Geology, Piccardi L. and Masse W.B. (eds). Geological Society of London, Special Publications, 273, 95-105.
- Polikreti K. and Maniatis Y. (2000) - The characterisation and discrimination of Parian marble in the Aegean region. In D.U. SCILARDI and D. KATSONOPOULOU (eds), *PARIA LITHOS*, Proceedings of 1st International Conference on the Archaeology of Paros and the Cyclades, Paros, October 2-5, 1997, Athens, 575-584.
- Sun S. (1990) - Denizli-Uşak Arasının Jeolojisi ve Linyit Olanakları (in Turkish). *Bulletin of Mineral*

- Research and Exploration Institute of Turkey (MTA), Scientific Report No: 9985.
- Sözbilir H. (1997) - Stratigraphy and sedimentology of the Tertiary sequences in the northeastern Denizli Province (Southwest Turkey). Ph.D. Thesis, Dokuz Eylül University Izmir-Turkey.
- Sözbilir H. (2005) - Oligo-Miocene extension in the Lycian orogen: evidence from the Lycian molasse basin, SW Turkey. *Geodinamica Acta*, 18,255-282.
- Şimşek Ş. (1984) - Denizli-Kızıldere-Tekkehamam-Tosunlar-Buldan-Yenice Alanının Jeolojisi ve Jeotermal Enerji Olanakları (in Turkish). Mineral Research and Exploration Institute of Turkey (MTA), Scientific Report No: 7846.
- Şimşek, C. (2007) - Laodikeia : Laodikeia ad. Lycum (in Turkish), Ege Yayınları, 384 pp.
- Topal S. and Özkul M. (2014) - Soft-Sediment Deformation Structures Interpreted as Seismites in the Kolankaya Formation, Denizli Basin (SW Turkey). *The Scientific World Journal*, Article ID 352654, 13pp.
- Westaway, R. (1990) - Block rotation in western Turkey, 1st Observational evidence. *Journal of Geophysical Research*, 95, 19, 857 -19, 884.
- Westaway R. (1993) - Neogene evolution of the Denizli region of western Turkey. *Journal of Structural Geology*, 15, 37-53.

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