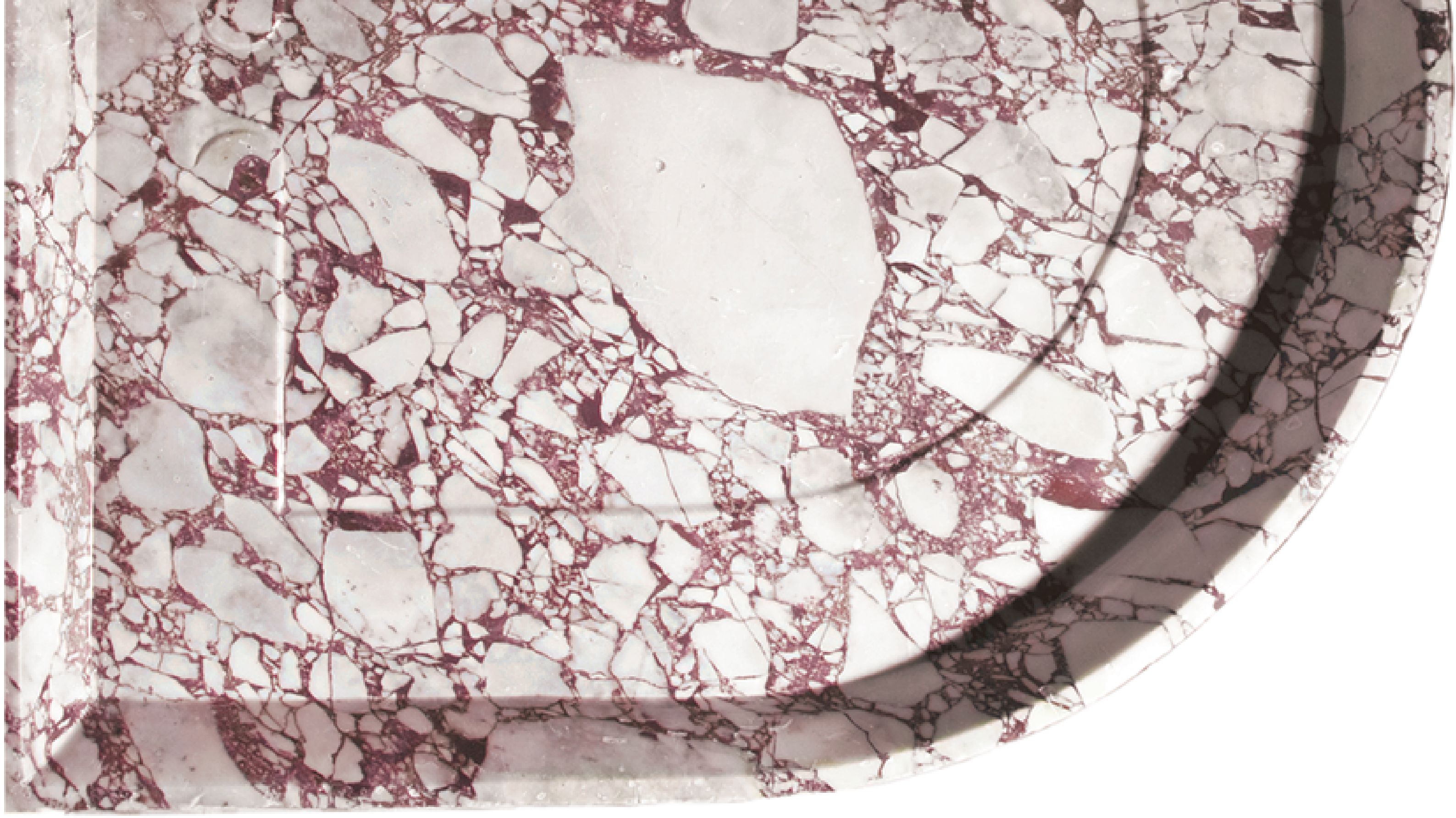
Unraveling the Carrara – Göktepe Entanglement

Prochaska, Walter; Attanasio, Donato; Bruno, Matthias



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UNRAVELING THE CARRARA – GÖKTEPE ENTANGLEMENT

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Abstract

Systematic investigations of imperial portraits from the first century AD up to late antiquity revealed a dramatic change in the portrait marbles starting approximately in Trajanic times. By the beginning of the 2nd century a so far unknown marble of exceptional quality suddenly arrived in Rome and became the portrait marble par excellence, which has hitherto been taken for Carrara marble. The isotope composition alone does not discriminate sufficiently, but there are several other characteristics to enable unambiguous archaeometric identification. Trace element contents, namely low Fe, Mn and the exceptional high Sr numbers, discriminate the Göktepe perfectly against Carrara marbles. Characteristic differences in the trace mineral contents of these marbles also exist.
The analytical data of the Göktepe and Carrara marbles presented in this paper allow an unambiguous discrimination of these marbles, those from Göktepe being in fact the most important portrait marbles of Roman Antiquity.

Keywords
Göktepe marbles, Sr-content, trace element analysis

1. Introduction

As a consequence of the discovery of the Göktepe quarries in the Aphrodisias region some years ago a re-evaluation of the use and provenance of portrait marbles, especially all through imperial times, became inevitable. Soon after this discovery, it became evident that this marble is very similar to the famous and widely used Carrara marbles. Existence of a new unknown marble type had not been recognized and the marbles from the yet unknown Göktepe site were commonly mistaken for Carrara. Systematic search and investigation of a big number of artefacts displayed in many museums soon uncovered the prominent use of the Göktepe marbles. This quarrying area supplied the most important portrait marble in Roman imperial times. By the beginning of the 2nd century a so far unknown marble of exceptional quality suddenly arrived in Rome and became the portrait marble par excellence. The provenance analyses of the marbles of 163 imperial portraits revealed that the preponderance of Parian Lychnites and Carrara marbles in early imperial times abruptly gave way to the dominant use of Göktepe marbles for imperial portraits starting in Trajanic/Hadrianic times. A detailed investigation of the diachronic use of the marbles of the imperial portraits and a comprehensive reference of the use of the Göktepe marbles in general is given elsewhere (Attanasio *et al.*, this volume). For the description of the Göktepe quarries and their location and characteristics we refer to previous publications on this topic.[1]

Because of the overwhelming importance of these two types of marbles and the fact that they can easily be confused with one another, the fundamental characteristics of the Göktepe marbles in comparison to those of Carrara will be explained below. We assume that the specific material characteristics of the Carrara marbles are well known and therefore this paper focuses on the petrographic and chemical characteristics of the Göktepe marbles, their similarities to and differences from Carrara marble and the discussion of the cause of the Carrara - Göktepe entanglement.

An essential prerequisite for marble provenance analysis is the access to and the availability of a databank for the variables analyzed. Numerous stable isotope data for different regions and quarries were published in the past[2]. However, systematic data on other variables for further discrimination such as trace element chemistry are only sporadically available. Our databank comprises approximately 3000 quarry samples and includes the numeric data for stable isotopes, trace elements, EPR data, the results of inclusion fluid chemistry and MGS (maximum

1 ATTANASIO *et al.* 2008; 2009.

2 The most comprehensive published collection of isotope data is given by ATTANASIO *et al.* 2006.

	DS	$MgCO_3$	Fe ppm	Mn ppm	Sr ppm	Li/Na	Cl/Na	K/Na	F/Na	Br/Na	I/Na	SO_4/Na	$\delta^{18}O_{(PDB)}$	$\delta^{13}C_{(PDB)}$	MGS
Carrara															
median	2550	1,60	100	27	158	1,64	1711	306	8,6	4,7	17,0	561,8	-1,87	2,12	0,80
mean	3086	1,63	152	28	165	1,71	1649	395	11,5	6,5	20,4	1011,3	-1,87	2,14	0,81
SD (σ)	1881	0,27	166	9	23	0,90	289	264	12,3	8,2	14,6	2113,0	0,53	0,15	0,19
Göktepe															
median	3877	0,74	39	6	626	0,39	1511	132	16,4	2,2	2,4	235,4	-3,06	2,46	0,68
mean	6176	0,79	41	5	647	0,45	1564	205	19,0	2,4	2,8	818,9	-3,37	1,80	0,66
SD (σ)	5698	0,20	10	4	178	0,26	353	234	13,1	1,2	2,1	3130,8	0,97	1,50	0,17
Göktepe low Sr															
median	1939	0,95	93	11	130	0,75	2086	625	13,0	2,8	4,2	439,7	-3,14	2,08	0,40
mean	3305	1,76	102	9	137	1,00	2016	573	19,1	3,9	11,0	657,7	-3,13	1,73	0,45
SD (σ)	3386	1,78	48	8	37	0,65	470	281	17,8	4,0	14,2	463,7	1,06	1,00	0,30

Table 1. The average numbers and standard deviation of the analytical data

grain-size) numbers. Furthermore, a collection of microscopic thin-sections of the classical marbles is available.

In the following we refer to three groups of samples from our databank; all three groups deal exclusively with white marbles. The marbles from Carrara are all grouped in one set and no intra-site discrimination (although possible to some extent) will be discussed here. Here we deal only with white Carrara marbles and we do not include "Carrara Bardiglio" in these considerations. Secondly the Göktepe samples from the ancient quarry sites are being grouped as "Göktepe" and these are usually high Sr-marbles. Finally a set of samples collected outside the site at distances ranging from 15 to 0.9 km from the ancient quarries are grouped as "low Sr-Göktepe. It is important to note that no evidence of ancient exploitation or use of these regional marbles could be found. The only remarkable exceptions are the low Sr marbles extracted from the small ancient quarry identified as "2C" (Attanasio *et al.* 2015). Here black as well as white marbles were mined; however, we refer only to the white marbles in this paper. Recent studies of the marbles of the Esquiline Group sculptures proved the at least limited use of this type of Göktepe marble.[3]

2. The methods applied

During the last few years the problems arising when the provenance of white marbles is investigated by one single method, like stable isotope analysis, have been discussed in detail.[4] A detailed description of the analytical procedures is given in these papers and therefore only a summary of the procedures is listed below. The obvious consequence from the complexity of the characteristics of marbles and their overlap is to apply a combination of analytical methods followed by a statistical evaluation of the results.

In this work, ample space is given to the characterization of the petrographic descriptions of the marbles. The mean values of the analytical data are presented in table 1.

Petrographic methods: In general a sound investigation of the microfabric of the investigated marbles is desirable. While these investigations easily can be done on quarry samples, the tininess of many samples available from artefacts prevents a sound petrographic investigation. The rarely occurring plagioclase is characteristic, for example, of Carrara marbles, but the chance of finding one crystal in a small thin section of a few mm^2 is rather small. Also, according to petrographic standards, for grain size measurement a few hundred mineral grains should be counted. A widely used parameter is maximum grain size (MGS), which is either determined by petrographic microscopy or by a hand magnifier on polished surfaces. The characteristics of the grain boundaries and intergrowth of crystals are also evaluated.

Isotopic methods: This method is the state of the art and the most widely used approach in marble provenance analysis. A considerable advantage of this method is the very small amount of sample required. It has to be kept in mind, however, that the extremely small size of samples in the order of some mg taken from weathered or partly weathered surfaces of ancient artefacts entails the risk of wrong results because of appreciable modification of the isotope composition due to weathering of the

3 ATTANASIO *et al.* 2015.

4 PROCHASKA, GRILLO 2010; PROCHSKA, ATTANASIO 2012.

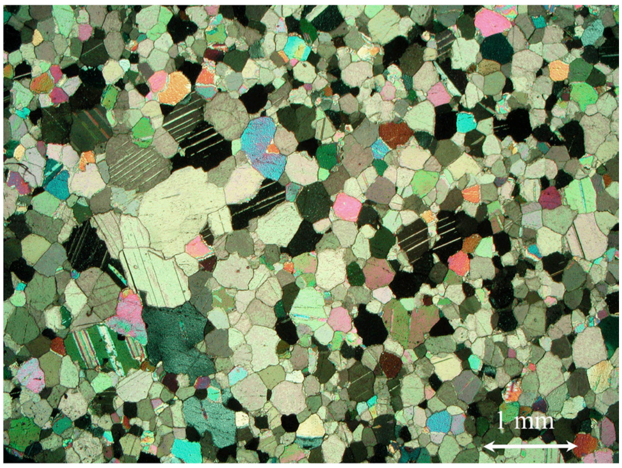

Fig. 1. Thin section of a fine-grained Göktepe marble with calcite groundmass of approx. 0.3 mm and patchy recrystallization (left hand side) with calcite grains of up to 0.8 mm

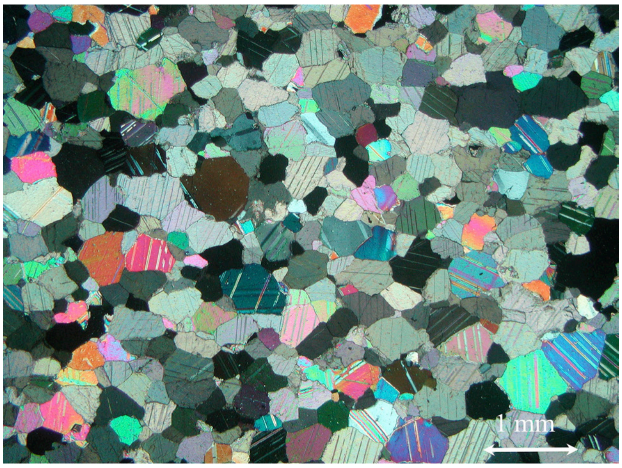

Fig. 2. Thin section of a fine-grained Carrara marble with a homoeoblastic calcite texture of approx. 0.7 mm MGS

corresponding surfaces. Furthermore, only in rare cases will the exclusive use of stable isotope analysis without combination with other methodological approaches provide satisfactory results.

Trace element chemistry: Additional variables can be obtained by chemical analysis of the marbles.

In this context it is important to mention that those elements that are incorporated into the carbonate lattice (Mg, Fe, Mn, Sr, and Zn) exhibit a fairly homogeneous and consistent distribution and can advantageously be used to discriminate different types of white marbles. As we will demonstrate below, the discrimination of the marbles from Carrara and from Göktepe can be achieved solely on the basis of their Sr content.

Analysis of fluid inclusions: This technique for characterizing marbles was developed to establish further analytical variables for a better discrimination of different marbles. This method can be used in concert with the established methods and enhance discrimination success when using multivariate statistical discrimination. The results from fluid inclusion investigations of carbonate rocks show that the fluid phase is usually relatively uniform with respect to its chemical composition. A series of chemical parameters (cations as well as anions) can be detected simultaneously by means of ion chromatography. During recent years, this method has been repeatedly applied for the discrimination of different marbles when other methods failed.[5]

5 PROCHASKA, GRILLO 2010; PROCHASKA 2013.

Evaluation of the data: The acquisition of a big number of variables requires statistical data processing. When combining the results from isotope analysis, trace element analysis and the analysis of the chemistry of inclusion fluids, we use multivariate discrimination analysis for data evaluation. The compositional fields of the marbles from a quarry or a given marble-producing site are usually presented as statistical ellipses (90 % ellipses, which means that 90 % of the samples of this population is within the ellipse).

3. The analytical results

3.1. The petrographic features

The macroscopic features of Göktepe marble are very similar to other high quality marbles used in antiquity and they very much resemble the characteristic Carrara marbles. The trained eye, however, becomes aware of the finer grain of the Göktepe marbles and the "ivory" feel of these marbles. Yet the average numerical MGS data are not too different, 0.8 mm for the Carrara and 0.68 mm for the Göktepe marbles. The reason is the homogenous grain-size of the Carrara marbles, slightly below 1 mm, whereas in the Göktepe marbles occasionally patchy recrystallized clusters of a few grains up to 1 mm can be observed in a polygonal groundmass of calcite crystals of approximately 0.3 mm (Figs. 1, 2). This paper generally deals with the white varieties of the Göktepe marbles, and yet it should be mentioned that there also occurs a black variety of very fine-grained Göktepe marble (grain-size below 0.1 mm) directly bordering on the white varieties (Fig. 3). As in the case of the Carrara Bardiglio marbles, the organic pigmentation and further

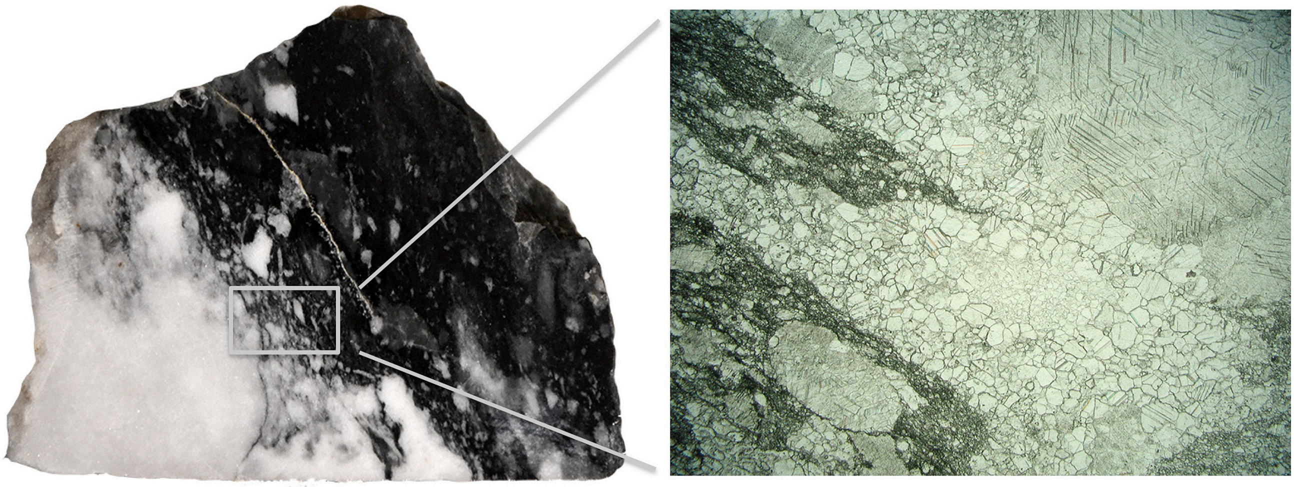

Fig. 3. Transition between the black and the white varieties of the Göktepe marbles. In the thin section (image length is 7 mm) on the right hand side the polygonal texture of the white marble sections can be recognized. The very fine-grained parts are the black varieties. In the right upper corner of the image a big relic of a shell fragment can be seen

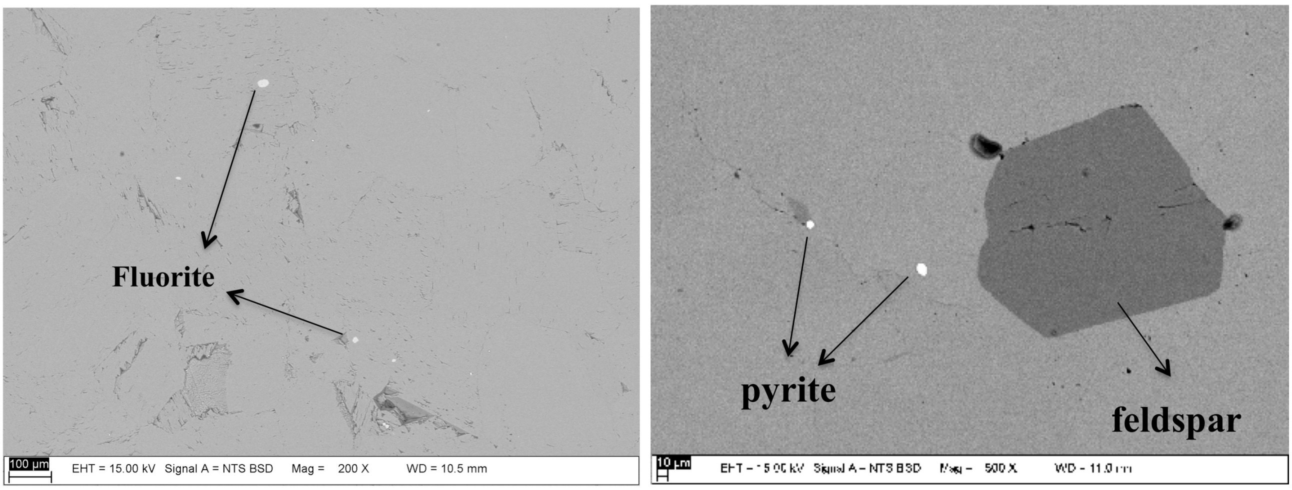

Fig. 4. SEM images of a Göktepe marble (left hand side) with only tiny fluorite crystals and a Carrara marble on the right hand side with small pyrite crystals and a larger feldspar crystal

impurities of clay minerals prevent the calcite crystals in this rock type from recrystallization in contrast to the white, very pure varieties. This black Göktepe marble also was used for very high-quality sculptures and artefacts which can be found in different museums.

Under the microscope, both Carrara and Göktepe marbles show typical polygonal textures with straight grain-boundaries; however, the Göktepe marbles develop the above described patchy recrystallizations. To distinguish between these two marbles solely on a petrographic basis can be extremely difficult. The polygonal textures with straight grain-boundaries in both marbles indicate a good recrystallization under equilibrium condition and practically no post-crystalline deformation.

Despite the very similar petrographic/textural features of the two marbles with respect to the calcite fabric, there are clear and crucial differences in the nature of the trace minerals. However, both marbles are generally very pure and the few trace minerals can only be safely determined by scanning electron microscopy. The only trace mineral in the Göktepe marbles is fluorite occurring in tiny crystals of a few µm. Accessory minerals in the Carrara marbles are ubiquitous, small apatite grains, pyrite and sporadically occurring larger feldspar crystals (Fig. 4).

The grade of metamorphism: The purity of the white Göktepe marble and especially the lack of critical trace minerals prevent a sound estimate of the metamorphic grade. However, the organic pigmentation of the black variety allows the estimation of the maximum temperatures the rock underwent during metamorphic crystallization. Investigations by Raman spectroscopy indicate that the organic colouring substance is semi-graphite (Fig. 5). This marble suffered a maximum

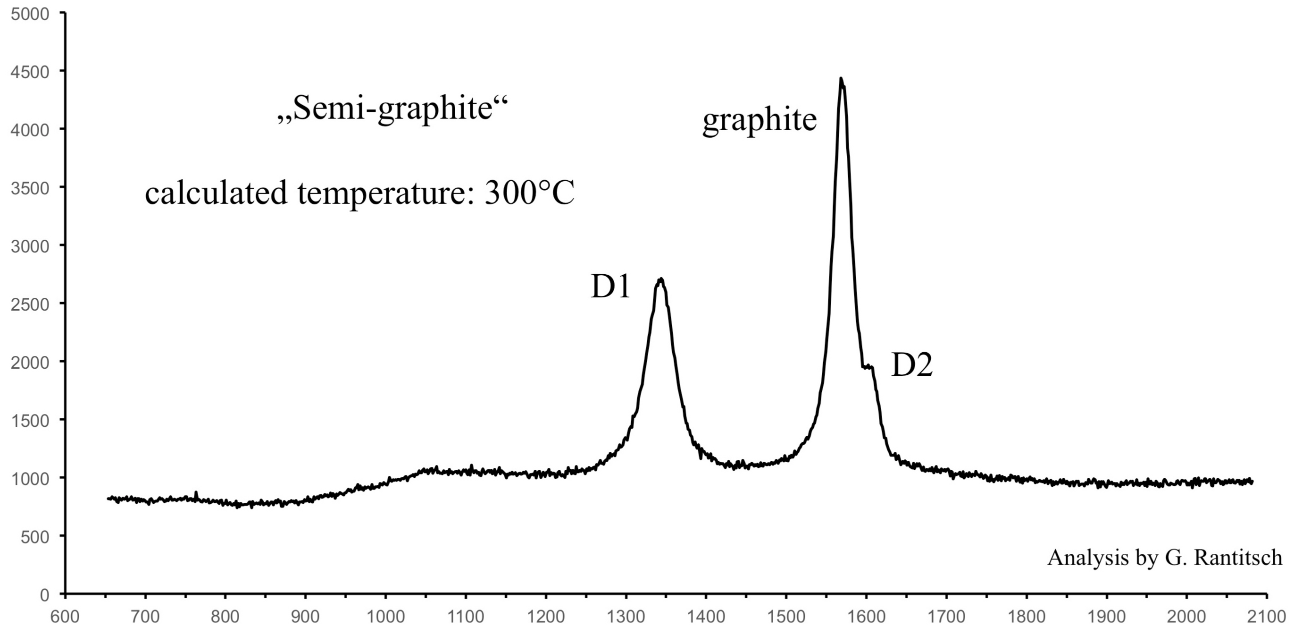

Fig. 5. Raman spectrum of the organic residue of a black Göktepe marble. The degree of recrystallization is very low as indicated by the pronounced D1 and D2 peaks and the not very sharp graphite peak

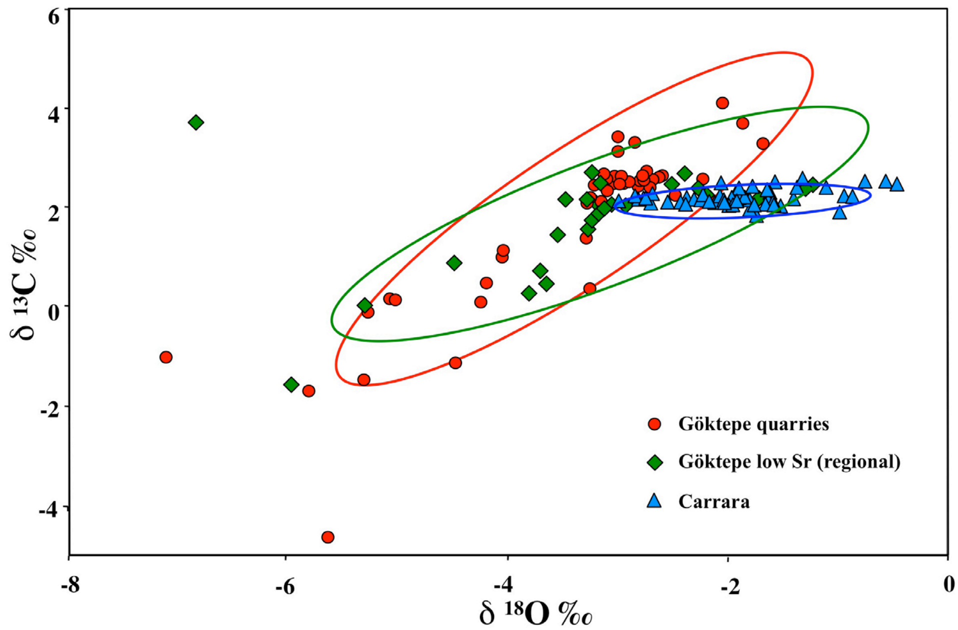

Fig. 6.
Stable isotope diagram of the 3 investigated types of marbles. Clearly the data fields overlap to a high degree thus inhibiting a precise provenance determination on that basis

metamorphic temperature of approximately 300° C and is, in petrographic terminology, at the transition of limestone to marble. Consequently, relics of fossils can still be observed in the black Göktepe marbles. For the Carrara marbles a slightly higher temperature of metamorphism (325 ± 30°C) was reported[6].

6 OESTERLING *et al.* 2007.

3.2. The isotopic composition

The results of the isotope analyses of the three groups of marbles are displayed in fig 6. As can be seen in table 1 and fig. 2 the isotope composition alone does not discriminate sufficiently enough between the two data fields of the Göktepe and Carrara marbles. The C-isotopes of the Carrara marbles are in a very small range and also the scatter in the O-isotopes is limited, resulting in a very consistent data field with a compact core field without any considerable outliers.

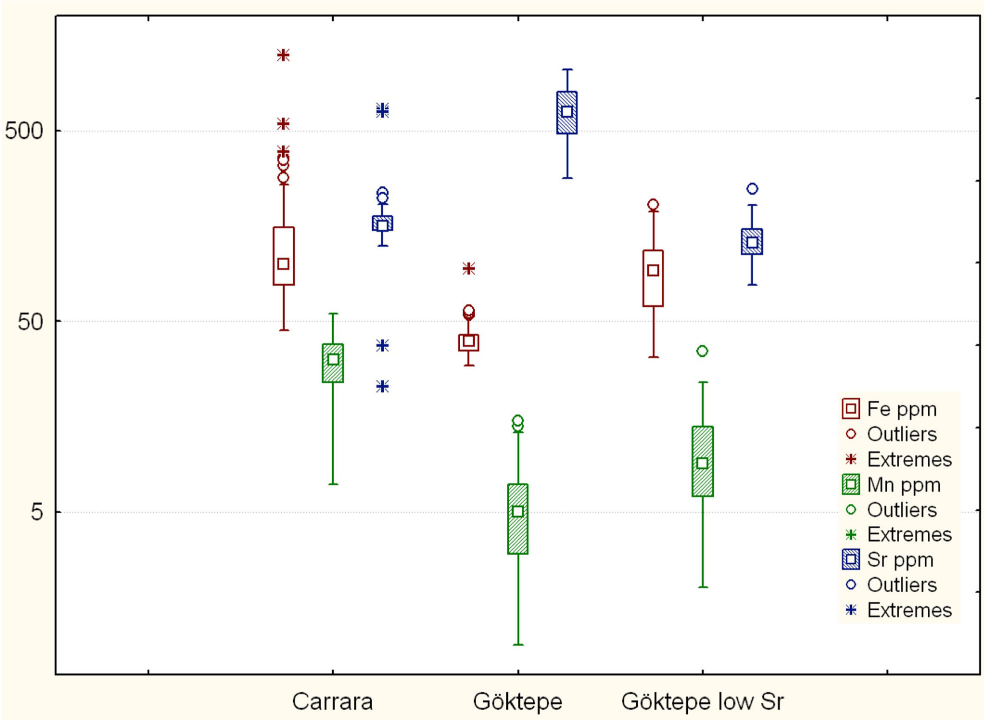

Fig. 7.
Boxplot for the Fe, Mn, and Sr contents of the three marble types under investigation. While the low Sr-Göktepe marbles are not too different in their composition, the Göktepe quarry samples clearly can be differentiated

The Göktepe projection points show a concise core field in both O- and C-composition, but there are also a series of outliers towards lighter isotopes that result in a relatively big 90 % ellipse. The core field of the Göktepe marbles is shifted more than 1‰ towards lighter O-isotopes compared to the Carrara core-field whereas the C-isotopc composition is relatively similar.

The regional, low Sr-Göktepe marbles show a wide scatter in the isotope diagram and largely overlap with both the Göktepe quarry samples and the Carrara marbles.

3.3. Trace element and fluid inclusion composition

As mentioned above, trace element analysis focused on those elements that are bound to the calcite lattice and substitute basically for Ca. These elements (Fe, Mn, and Sr) are not related to sporadically and inhomogeneously occurring trace minerals like e.g. the rare earth elements which usually are bound to apatite and consequently these trace elements show a relatively homogenous distribution within the marbles. The most striking and obvious difference in the chemical composition is the exceptional high Sr content of the Göktepe marbles (283/1039 ppm) compared to Carrara (124/237 ppm). In our databank of approximately 3000 marble samples only the marbles from Cap de Garde in Algeria and some Alpine marbles exhibit similar high Sr contents; however, these marbles are very coarse-grained and cannot be confused with the marbles discussed here. Elevated Sr contents can be found in some Carrara Bardiglio marbles, but also here the petrographic features are clearly different from the marbles considered in this paper and are not susceptible to being confused which each other.

There is also a significant difference in the Fe and Mn contents between the Carrara and the Göktepe marbles to the effect that the latter shows distinctly lower numbers of these elements due to its very high purity. As can be seen in the boxplot in fig. 7, a perfect discrimination between the Carrara and the Göktepe marbles can already be achieved by the use of these three trace elements alone.

The regional low Sr-Göktepe marbles resist discrimination from Carrara marbles much more because of their similar Fe and Sr contents. Mn is considerably lower than in the Carrara marbles and is therefore of some use for discrimination.

However the composition of the inclusion fluids offers the chance to improve the discernibility of this group from the Carrara marbles as well as from the Göktepe quarry samples. In particular, the elemental ratios (normalized to Na) of Li, K, Br, and I can be used in this case when applying a statistical approach, as will be shown below.

4. Evaluation of the analytical data

Trace element contents, namely the low Fe, Mn and the exceptional high Sr numbers, discriminate the Göktepe marbles perfectly from Carrara marbles. The high Sr contents alone discriminate them very well from other fine-grained marbles and are therefore the most important analytical parameter for the discrimination of these two marbles. As stated above, more effort and

		Carrara	Göktepe	Göktepe low Sr	total
no of cases	Carrara	64	0	1	65
	Göktepe	0	56	0	56
	Göktepe low Sr	1	0	24	25
%	Carrara	98,5	0,0	1,5	100
	Göktepe	0,0	100,0	0,0	100
	Göktepe low Sr	4,0	0,0	96,0	100

Table 2.
The degree of the reassignment of the corresponding samples to their group using the variables MGS, Mg, Fe, Mn, Sr, DS, Li/Na, Cl/Na, K/Na, Br/Na, I/Na, $\delta^{18}O$ ‰ and $\delta^{13}C$ ‰

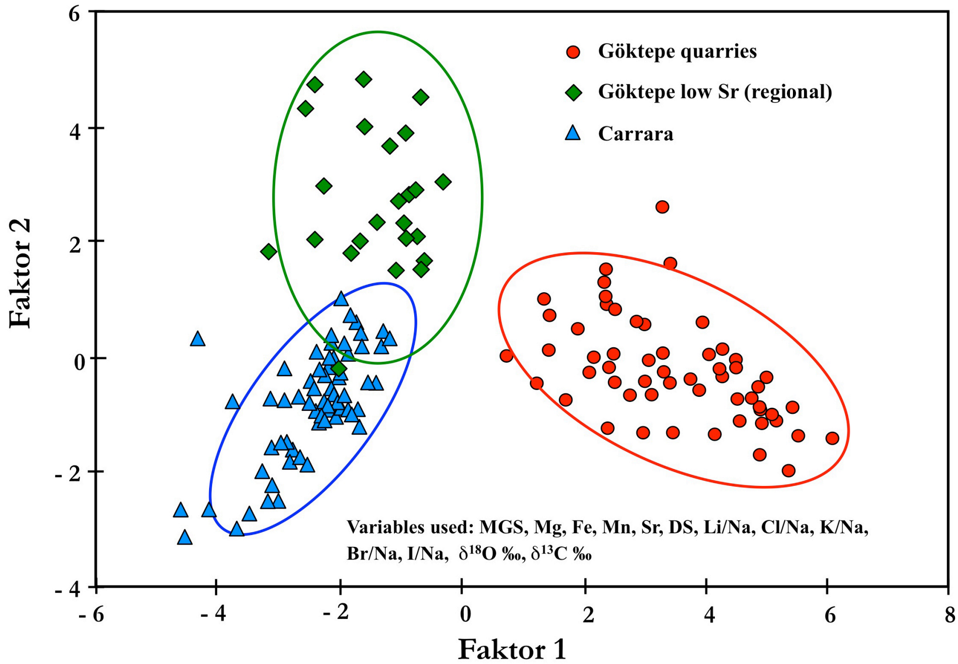

Fig. 8.
Multivariate diagram of the two most powerful canonical functions of the results of the discriminant analysis. The data fields are displayed as 90 % ellipses and the three marble types considered are largely separated

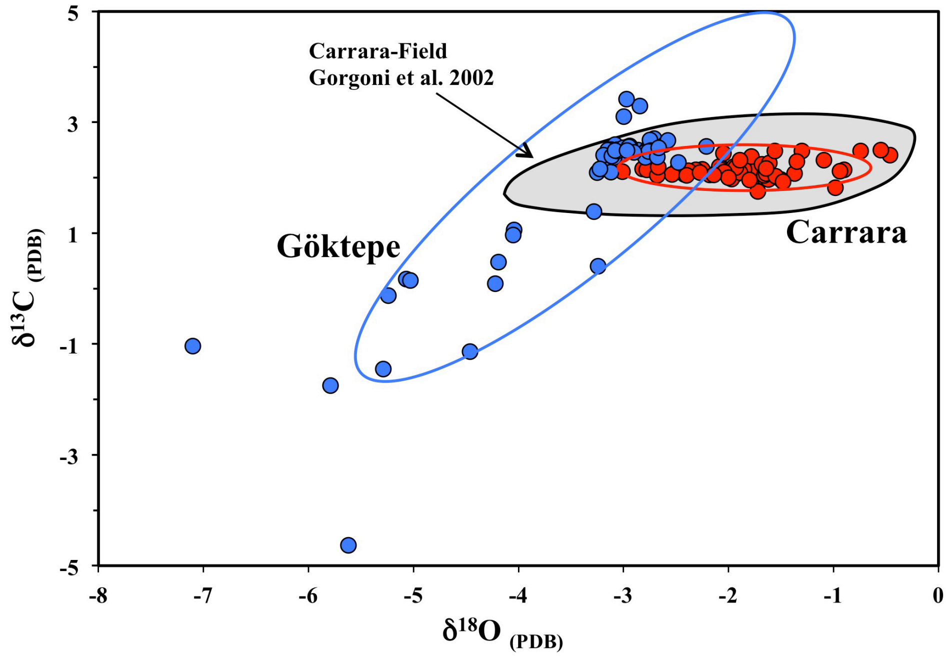

Fig. 9.
The widely used data field of Gorgoni *et al.* 2002 covers both core fields of the Göktepe and the Carrara marbles in the stable isotope diagram and thus the 2 different types of marbles cannot be discriminated on that basis

the use of more variables and a statistical, multivariate discrimination has to be used to safely set apart the low Sr-regional Göktepe marbles from the two other groups.

In table 2, the overall result of the statistical evaluation is presented. The best result for the correct reassignment of the analyzed samples to their corresponding group is obtained when a large number of variables for the multivariate calculation is used. These variables are MGS, Mg, Fe, Mn, Sr, DS, Li/Na, Cl/Na, K/Na, Br/Na, I/Na, $d^{18}O$ ‰, $d^{13}C$ ‰. The success rate for the reassignment is 100 % for the Göktepe quarry samples and in both other cases only one sample each is misclassified. In the bivariate diagram in fig. 8 the two most powerful canonical functions of the results of the discriminant analysis are graphically displayed. The data-fields (90 % ellipses) of the three considered marble types are largely separated.

5. Discussion and conclusions

The discovery of the Göktepe marbles some years ago unveiled the existence of a so far unknown marble of utmost importance which had not been previously recognized as an important type of portrait marble of its own. The production of this fine-grained white marble of highest quality used almost exclusively for portraits is attested from the beginning of the 2nd century until advanced late antiquity.

During recent years the general acceptance of this crucial role of the Göktepe marbles and even the existence of this marble as a prime portrait marble made slow progress. This inevitably raises the question of why a marble of that importance was not recognized for decades. While most of the provenance analyses involving these marbles were so far conducted by one research group the possibility of a provenance from the Göktepe marbles for Roman artefacts is more and more considered in recent publications by different scientists[7].

Innumerable investigations have been conducted on ancient marble quarries throughout the ancient world and a wide series of analytical methods have been applied. By contrast precise scientific investigations of artefacts like portraits and sculptures are much more rare. This is due to the fact that non-destructive analytical methods suitable for an established provenance analysis are, so far, not available. For serious geochemical investigations a certain amount of rock samples has to be available for analysis. Acquiring permission for the sampling of high quality artefacts, e.g. in museums, often meets considerable problems and the provenance estimation of a marble is often based solely on visual examination. Therefore the macroscopic resemblance of the Carrara marbles with those from Göktepe is the basic reason for the Carrara – Göktepe entanglement.

These circumstances also prevent in most cases a thorough petrographic investigation of the marbles of artefacts for in this case the sample size should be in the range of 1 cm. Because of the above-described textural similarities of the Carrara and Göktepe marbles, discrimination with a petrographic microscope is often difficult. However, in the case of the availability of a scanning electron microscope a trustworthy discrimination is possible.

A certain mitigation of the problem of taking large samples seemed to be at hand with the introduction of stable isotope analysis in marble provenance analysis. This was the only approach that was intensely applied during the last decades and where databanks were available. The extremely small amount of sample necessary, a few mg, scratched from a hidden surface seemed to be tolerable but problems with sample homogeneity and contamination cannot be controlled easily in this case. In this context it should be mentioned that the isotopic core fields of the Carrara marbles are indeed different from those of Göktepe, and yet their data fields overlap considerably as shown in figs. 6 and 9. The widely used isotope field for Carrara marbles published by Gorgoni *et al.* 2002[8] comprises a large area and includes both core fields and even a cursory discrimination is not possible on that basis (Fig. 9).

The petrographic and chemical features of the Göktepe and Carrara marbles presented in this paper allow an unambiguous discrimination of these two types of marbles. If a provenance investigation can be reduced to the differentiation between Carrara and Göktepe marbles and other fine-grained marbles can be eliminated by a different argumentation, a trace element analysis alone will result in an unambiguous result and will help in unravelling the Carrara-Göktepe entanglement.

7 LAPUENTE *et al.* 2012; PENSABENE *et al.* 2015.

8 GORGONI *et al.* 2002

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