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Araştırma Makalesi Research Article

Multivariate Statistical Evaluation of Geochemical Properties of "Alanya Emperador Dark" Marbles

Ipek YAZICI[1], Mustafa Gurhan YALCIN[1*], Ozge OZER ATAKOGLU[1], Fusun YALCIN[2]

[1]*Department of Geological Engineering, Akdeniz University, 07058 Antalya, Turkey*
[2]*Department of Mathematics, Akdeniz University Faculty of Science, 07058 Antalya, Turkey*

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Abstract

The formation of marbles, which are regarded as natural stones, can be attributed to various rock origins and formation conditions. The study area is located in the Obaalacami village of Alanya district in the province of Antalya, which is located in the Taurus belt, and it covers an area of about 100 hectares. In this study, the major and trace element components of the "Emperador Dark" marbles found in the Alanya region were interpreted using geostatistical analyses. Geostatistical methods can be used to reveal and interpret the origin of marble and its formation conditions. In this context, X-ray fluorescence (XRF) spectrometry was employed to analyze samples collected from various locations within the marble quarry. The CaO values of the marble quarry were found to have a normal distribution, and this was interpreted using the histogram values together with the measures of central tendency such as the mode, median, and arithmetic mean values. The major and trace element contents of the "Emperador Dark" marbles, which meet the assumption of normality, were obtained using the Pearson correlation coefficient. The strong negative correlation of CaO, which played a major role in the formation of marbles, with SiO_2 and MgO was associated with the mineralogical composition that played a role in marble formation. These correlations were also tested by regression analysis, and it was proved that a statistically significant model had been obtained.

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Author ID (ORCID Number)

I. Yazici, 0000-0003-1405-8994
M. G. Yalcin, 0000-0002-8011-5371
O. Ozer Atakoglu, 0000-0003-2678-1194
F. Yalcin, 0000-0002-2669-1044

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1. INTRODUCTION

Marble is a metamorphic rock formed by limestone, which is one of the carbonate rocks, under metamorphism conditions (Tarinc et al., 2019a, b; Yazici et al., 2019; Ozer et al., 2020). Marble is an industrial product that was formed by carbonate rocks under physical and chemical conditions (Yalcin et al., 2016a, b, c; Tarinc et al., 2019a, b; Aydin et al., 2020; Ozer et al., 2020). Various methods of different types are used to reveal these conditions. Analyzing the correlation between the compositions of the major and trace elements that played a role in the marble formation is crucial for interpreting the origin and environment conditions.

According to the literature, the researchers tried to evaluate the geochemical properties of marble samples at a national and international scale using different types of methods. In the study on Zangalline marbles found in Iraq, the major and trace element behaviors were geochemically evaluated. Descriptive statistical values of major element oxides and trace elements were explained according to mineralogical characteristics (Mirza, 2019).

*Corresponding Author, e-mail: gurhanyalcin@akdeniz.edu.tr

In the study conducted to reveal the geochemical properties of the marble quarry in Akcay village, the geochemical data of the marble quarry were interpreted using different types of multivariate statistical methods. It was determined that mostly the CaO, P_2O_5, and Sr element played a role in the formation of marbles (Tarinc et al., 2019b).

In the study on Zambia marbles in the Zambezi belt in Africa, the geochemical contents of the marbles were analyzed. Inferences were made about the elemental chemistry that played a role in the formation of marbles using the correlations of major and trace elements (Munyanyiwa & Hanson, 1988).

In a study on the geochemistry of marbles that form outcrops in the Gongchangling iron deposits in China, the researchers tried to interpret the formation mechanism of the marble. In this study, the major and trace element contents were determined, and the anomalies of these features facilitated the interpretations about the formation, origin, and environmental conditions of marbles (Li et al., 2012).

The study area is located in the Alanya district of Antalya province, and it has a boundary with Alanya Bay on the east and Alanya peninsula on the West (Figure 1). In the literature, there is no study on the correlations of major and trace elements that are among the geochemical properties of "Emperador Dark" marbles. In this context, the elemental associations that contributed to the formation of the marble quarry were interpreted by revealing these correlations.

Figure 1. *Site Location Map of The Study Area*

2. MATERIAL AND METHOD

Chemical analyses of 10 samples collected from various locations of the Emperador Dark marble quarry were carried out using X-ray fluorescence (XRF) method (Oz & Ozer, 2019). The correlation values of the major and trace elements, which were obtained according to the results of the analyses, were determined and interpreted using the SPSS23 software package.

3. RESULTS AND DISCUSSION

The results of the geochemical analysis of the samples are given in Table 1. The effect of the marble quarry on the formation mechanism of the major and trace elements was explained using descriptive statistical methods (Table 2).

Table 1. *Elemental Concentrations of The Dolomites Area (Yazici, 2020)*

Sample Code	MgO	Al_2O_3	SiO_2	CaO	K_2O	Fe_2O_3	SO_3	TiO_2	Sr	Zr	LOI
I1	4.49	0.18	0.64	50.10	0.05	0.09	0.01	0.06	0.01	0.33	44.00
I2	5.33	0.11	0.74	47.30	0.03	0.06	0.01	0.06	0.01	0.30	46.00
I3	6.36	0.41	0.93	46.80	0.03	0.07	0.06	0.05	0.01	0.33	44.90
I4	7.80	0.27	1.27	46.00	0.03	0.07	0.06	0.02	0.01	0.31	44.10
I5	9.64	0.47	1.21	44.90	0.03	0.05	0.05	0.02	0.01	0.33	43.30
I6	5.78	0.46	1.16	47.20	0.03	0.06	0.05	0.04	0.02	0.30	44.90
I7	4.03	0.21	0.74	47.50	0.03	0.07	0.06	0.05	0.01	0.28	47.00
I8	7.23	0.23	0.79	47.80	0.03	0.06	0.04	0.02	0.01	0.32	43.40
I9	5.91	0.19	0.77	48.20	0.05	0.08	0.06	0.08	0.01	0.35	44.30
I10	6.91	0.02	0.64	48.40	0.03	0.06	0.03	0.02	0.01	0.33	43.50

Table 2. *Descriptive Statistical Values of The Major and Trace Elements (Yazici, 2020)*

	MgO	Al_2O_3	SiO_2	CaO	K_2O	Fe_2O_3	SO_3	TiO_2	Sr	Zr
Mean	6.35	0.25	0.89	47.42	0.03	0.07	0.04	0.04	0.01	0.32
Median	6.14	0.22	0.78	47.40	0.03	0.07	0.05	0.04	0.01	0.32
Mode	4.03[a]	0.02[a]	0.64[a]	44.90[a]	0.03[a]	0.05[a]	0.01[a]	0.02[a]	0.01[a]	0.33
Std. Deviation	1.65	0.15	0.24	1.40	0.01	0.01	0.02	0.02	0.00	0.02
Variance	2.71	0.02	0.06	1.97	0.00	0.00	0.00	0.00	0.00	0.00
Skewness	0.60	0.22	0.70	0.08	1.46	0.99	-0.89	0.50	0.87	-0.05
Kurtosis	0.54	-0.84	-1.27	1.15	0.77	1.36	-0.71	-0.76	0.16	0.42
Range	5.61	0.45	0.63	5.20	0.03	0.04	0.05	0.06	0.01	0.07
a: Multiple modes exist. The smallest value is shown.										

The major and trace element compositions of the samples collected from the studied marble quarry were examined by calculating the measures of central tendency (arithmetic mean, mode, and median), the measures of variability (range, variance, and standard deviation), and the measures of the shape of the distribution (kurtosis and skewness values). The geochemical compositions of the marble samples are listed as follows: CaO (50.10-44.90 wt%) > MgO (9.64-4.03 wt%) > SiO_2 (2.27-0.64 wt%) > Zr (0.35-0.28 wt%) > Al_2O_3 (0.47-0.02 wt%) > Fe_2O_3 (0.09-0.05 wt%) > SO_3 (0.06-0.01 wt%) > TiO_2 (0.08-0.02 wt%) > K_2O (0.05-0.03 wt%) > Sr (0.02-0.01 wt%). Considering the range values, the major element oxides of MgO and CaO were observed to have the largest range. The presence of high MgO in the marble quarry can be associated with the presence of dolomite (Mirza, 2019). Since marble is a metamorphic product, which underwent metamorphism, the major oxide of SiO_2 has played a significant role in the formation (Brownflow, 1996; Ahmad et al., 2014; Mirza, 2019; Ince et al., 2021; Yalcin et al., 2021). The standard deviation values of the major and trace elements were found to be smaller than their arithmetic mean values. This information led to the interpretation that the geochemical data of the marble samples were gathered around the mean value. On the other hand, the mode, median, and arithmetic mean values of the major element oxide of CaO, which contributed the most to the formation of marble samples, were observed to be very close to each other. This caused the distribution to have a skewness value of 0.08 and to display a normal distribution (Figure 2a). The mode, median, and arithmetic mean values of the major element oxide of MgO, which contributed the most to the formation of marble samples, were observed to be very close to each other. This caused the distribution to have a skewness value of 0.60 and to display a normal distribution (Figure 2b). The mode, median, and arithmetic mean values of the major element oxide of SiO_2, which contributed the most to the formation of marble samples, were observed to be very close to each other. This caused the distribution to have a skewness value of 0.70 and to display a normal distribution (Figure 2c). The mode, median, and arithmetic mean values of the trace element of Zr, which contributed the most to the formation of marble samples, were observed to be very close to each other. This caused the distribution to have a skewness value of -0.05 and to display a normal distribution (Figure 2d). The mode, median, and arithmetic mean values of the major element oxide of Al_2O_3, which contributed the most to the formation of marble samples, were observed to be very close to each other. This caused the distribution to have a skewness value of 0.22 and to display a normal distribution (Figure 2e). The mode, median, and arithmetic mean values of the major element oxide of Fe_2O_3, which contributed the most to the formation of marble samples, were observed to be very close to each other. This caused the distribution to have a skewness value of 0.99 and to display a normal distribution (Figure 2f). The mode, median, and arithmetic mean values of the major element oxide of TiO_2, which contributed the most to the formation of marble samples, were observed to be very close to each other. This caused the distribution to have a skewness value of -0.76 and to display a normal distribution (Figure 2g). The mode, median, and arithmetic mean values of the major element oxide of SO_3, which contributed the most to the formation of marble samples, were observed not to be very close to each other. This caused the distribution to have a skewness value of -0.89 and to display a normal distribution (Figure 2h). The mode, median, and arithmetic mean values of the major element oxide of K_2O, which contributed the most to the formation of marble samples, were observed not to be very close to each other. This caused the distribution to have a skewness value of 1.46 and to display a normal distribution (Figure 2i). The mode, median, and arithmetic mean values of the major element oxide of Sr, which contributed the most to the formation of marble samples, were observed not to be very close to each other. This caused the distribution to have a skewness value of 0.87 and to display a normal distribution (Figure 2j).

Before choosing the method to be applied to determine the correlation coefficients of the major and trace elements, the data were subjected to the normality test (Ozer et al., 2019; 2020; Yalcin et al., 2007; 2008; 2015; 2016a; 2019a, b; Yalcin & Ilhan, 2013; Atakoglu & Yalcin, 2021) (Table 3). In the analysis, the Shapiro-Wilks test is used if the sample size is less than 29, while the Kolmogorov-Smirnov test is preferred if the sample size is greater than 29 (Kalayci, 2010; Yalcin et al., 2013; Yalcin & Mert, 2018; Atakoglu et al., 2021). Therefore, the Shapiro-Wilk test was found to be the appropriate normality test for the marble samples (Table 3). In this context, the following hypotheses were established:

H_0: The data are normally distributed.

H_a: The data are not distributed normally.

Table 3. *Normality Test of The Major and Trace Elements (Yazici, 2020)*

Tests of Normality						
	Kolmogorov-Smirnov[a]			**Shapiro-Wilk**		
	Statistic	df	**P-value**	Statistic	df	**P-value**
MgO	0.10	10	0.20*	0.97	10	0.90
Al_2O_3	0.16	10	0.20*	0.92	10	0.44
SiO_2	0.25	10	0.06	0.85	10	0.06
CaO	0.14	10	0.20*	0.97	10	0.92
K_2O	0.27	10	0.03	0.75	10	0
Fe_2O_3	0.13	10	0.20*	0.93	10	0.51
SO_3	0.24	10	0.09	0.83	10	0.03
TiO_2	0.20	10	0.20*	0.93	10	0.46
Sr	0.19	10	0.20*	0.92	10	0.37
Zr	0.17	10	0.20*	0.96	10	0.83
*: This is a lower bound of the true significance. a: Lilliefors Significance Correction						

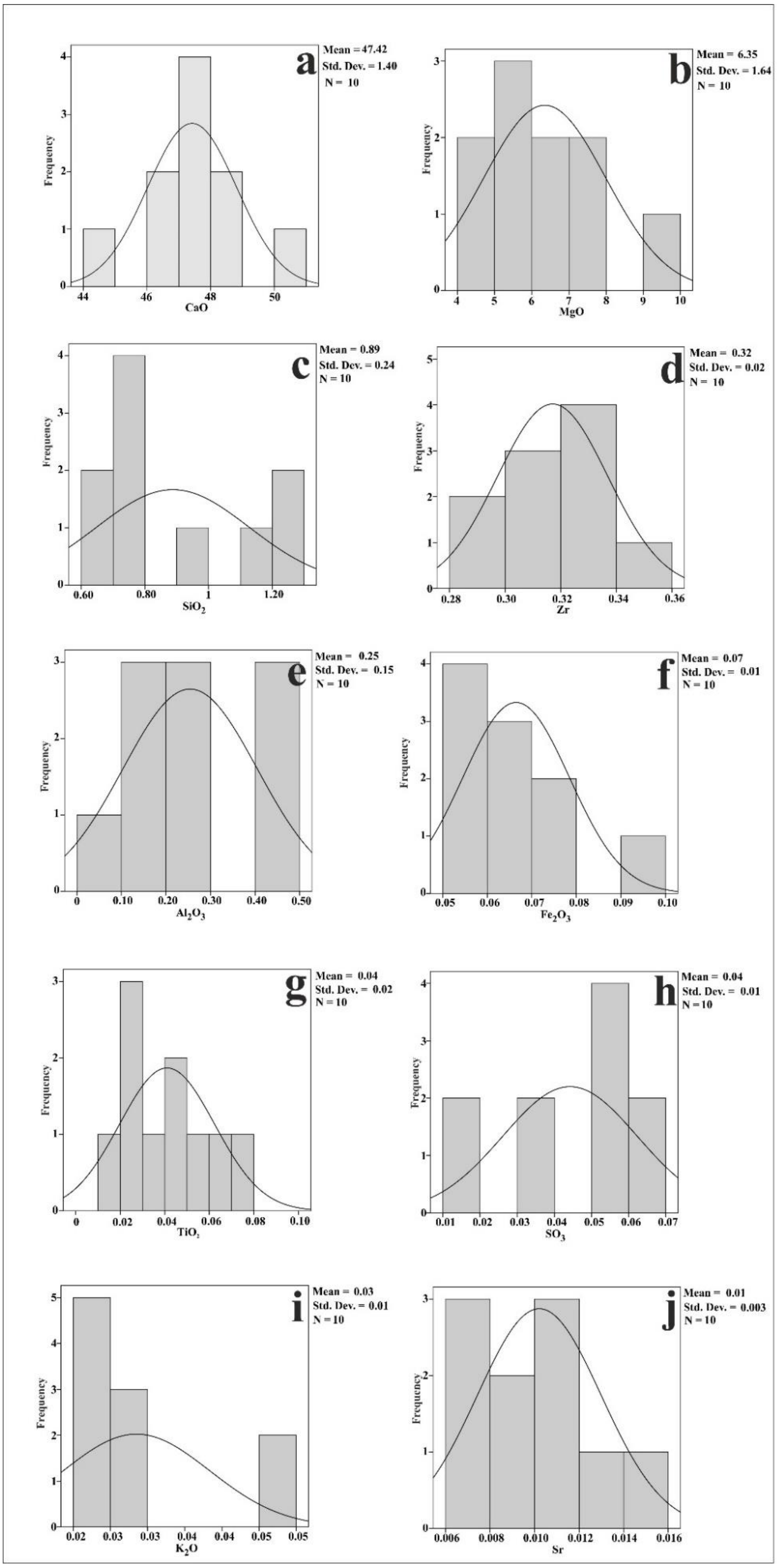

Figure 2. *Kurtosis Values of The Histograms of The Geochemical Data,*
a) *CaO,* ***b)*** *MgO,* ***c)*** *SiO_2,* ***d)*** *Zr,* ***e)*** *Al_2O_3,* ***f)*** *Fe_2O_3,* ***g)*** *TiO_2,* ***h)*** *SO_3,* ***i)*** *K_2O,* ***j)*** *Sr*

The H_0 hypothesis was found to be valid for the distributions of MgO, Al_2O_3, SiO_2, CaO, Fe_2O_3, TiO_2, Sr, and Zr at a significance level of 5%. The P-values of K_2O and SO_3 were found to be 0.004 and 0.038, respectively; then, the arithmetic mean and median values of the data were examined. The arithmetic mean and median values of K_2O were calculated as 0.335 and 0.03, respectively, while these values were found to be 0.0442 and 0.01 for SO_3. Considering these values, the normality assumption was approached intuitively, and the distributions of the major element oxides of K_2O and SO_3 were deemed to meet the H_0 hypothesis.

The correlation between the two variables was evaluated according to Table 4.

***Table 4**. Interpretation of The Correlation Coefficient (Kalayci, 2010)*

r	**relation**
0-0.25	very weak
0.26-0.49	weak
0.50-0.69	medium
0.70-0.89	high
0.90-1	very high

The major and trace elements meeting the normality assumption were analyzed using the Pearson correlation coefficient (Table 5).

***Table 5.** Correlations of The Major and Trace Elements (Yazici, 2020)*

	MgO	**Al_2O_3**	**SiO_2**	**CaO**	**K_2O**	**Fe_2O_3**	**SO_3**	**TiO_2**	**Sr**	**Zr**
MgO	1									
Al_2O_3	0.388	1								
SiO_2	0.632*	0.780**	1							
CaO	-0.709*	-0.624	-0.814**	1						
K_2O	-0.528	-0.415	-0.498	0.697*	1					
Fe_2O_3	-0.643*	-0.299	-0.451	0.707*	0.823**	1				
SO_3	0.244	0.536	0.518	-0.527	-0.305	-0.171	1			
TiO_2	-0.709*	-0.254	-0.494	0.561	0.822**	0.658*	-0.134	1		
Sr	-0.432	0.187	0.266	-0.011	-0.013	-0.014	-0.097	0.162	1	
Zr	0.334	-0.060	-0.139	0.179	0.400	0.170	0.050	0.263	-0.703*	1

There are moderate positive correlations between the major element oxides of MgO and SiO_2 (r=0.632*), CaO and K_2O (r=0.697*), and Fe_2O_3 and TiO_2 (r=0.658*) while there is a high and moderate positive correlation between the major element oxides of CaO and Fe_2O_3 (r=0.707*).

There are a high and strong positive correlation between the major element oxides of Al_2O_3 and SiO_2 (r=0.780**), K_2O and Fe_2O_3 (r=0.823**), and K_2O and TiO_2 (r=0.822**).

There is a moderate negative correlation between the major element oxides of MgO and Fe_2O_3 (-0.643*).

There are a high and moderate negative correlation between the major element oxides of MgO and CaO (r=-0.709*), MgO and TiO_2 (r=-0.709*), and Sr and Zr (r=-0.703*).

There is a high and strong negative correlation between the major element oxides of SiO_2 and CaO (r=0.814**).

According to the coefficient of determination (explainable variance) (R^2), 72% of the CaO values of the marbles can be explained by SiO_2 and MgO, or 72% of the SiO_2 and MgO can be explained by CaO (Table 6).

Moreover, in the study on Akcay marbles, the correlation between CaO and MgO was found to be r=-0.564** and the correlation between CaO and SiO_2 was calculated as r=-0.463*. On the other hand, the study on Zangalline marbles revealed that the negative correlation between CaO-SiO_2 increased with the increase in the metamorphism conditions during marble formation.

Table 6. *Coefficient of Determination (explainable variance) Table*

Model Summary[b]				
Model	R	**R Square**	Adjusted R Square	Std. Error of the Estimate
1	0.85[a]	**0.72**	0.64	0.83
a: Predictors: (Constant), SiO_2, MgO b: Dependent Variable: CaO				

The significant P-value in ANOVA reveals the error rate of the statistics (Yalcin & Unal, 2018; Ozer & Yalcin 2020; Ozer et al., 2019; 2020; Yalcin et al., 2020; Atakoglu et al., 2021). In the present study, the P-value value was found to be 0.01 (Table 7). Therefore, the model that expressed the correlation of CaO with SiO_2 and MgO found to be statistically significant.

Table 7. *Error Rate Table*

ANOVA[a]						
Model		Sum of Squares	df	Mean Square	F	**P-value**
1	Regression	12.84	2	6.42	9.22	**0.01[b]**
	Residual	4.87	7	0.69		
	Total	17.71	9			
a: Dependent Variable: CaO b: Predictors: (Constant), SiO_2, MgO						

Factor analysis was performed to extract and classify the connections between the geochemical data of the marble samples (Kalayci, 2010; Atakoglu et al., 2021). Using scree plot, it was determined how many factors the geochemical data were collected under (Figure 3) (Leventeli & Yalcin, 2021).

According to the scree plot, it was determined that the slope started to disappear between three/four points. In the explainable total variance table, where the cumulative variance ratio is given, it is clarified under how many factors the data are collected (Table 8).

For geochemical data, the presence of three factors greater than 1 is observed. Principal component matrix (PCA) analysis was performed on the data whose number of factors was determined. The geochemical contents that create the data collected under three factors were determined (Table 9).

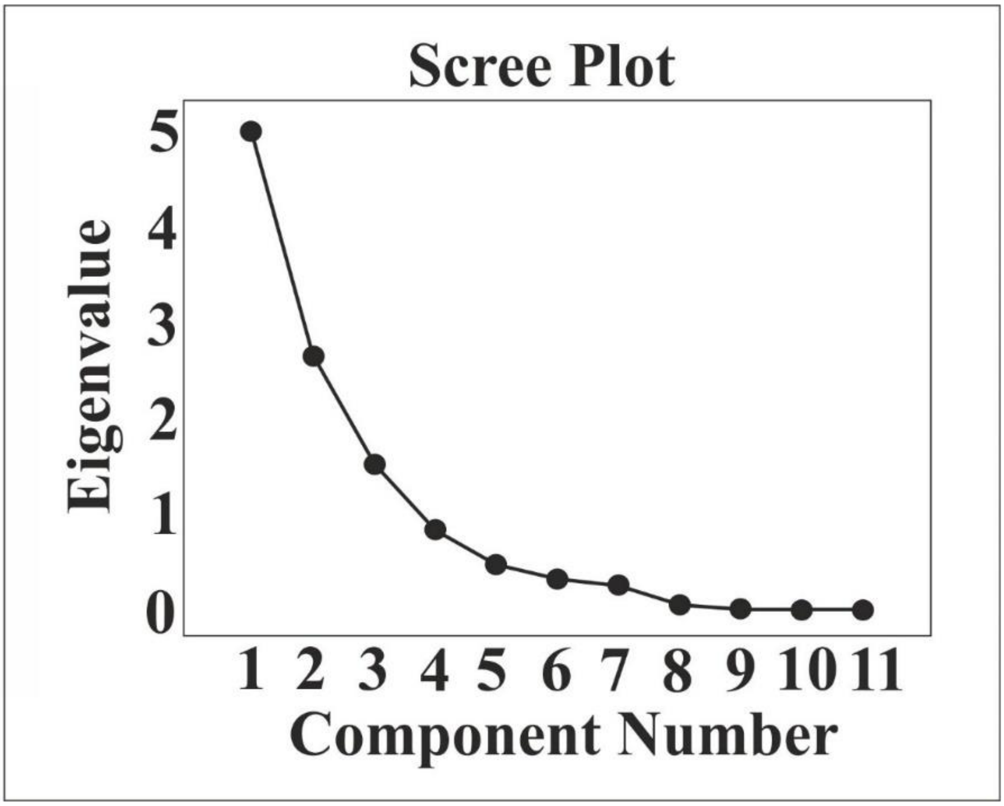

Figure 3. *Scree Plot of The Geochemical Data*

Table 8. *Determination of The Number of Factors Based on Eigenvalue Statistics of Marbles*

Total Variance Explained						
Component	Initial Eigenvalues			Extraction Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	**Cumulative %**
1	4.92	44.77	44.77	4.92	44.77	**44.77**
2	2.64	24.06	68.84	2.64	24.06	**68.84**
3	1.50	13.68	82.52	1.50	13.68	**82.52**
4	.82	7.52	90.04			
5	.46	4.23	94.28			
6	.31	2.86	97.15			
7	.25	2.28	99.43			
8	.05	.49	99.92			
9	.00	.07	100			
10	3.065E-16	2.787E-15	100			
11	-3.256E-16	-2.960E-15	100			
Extraction Method: Principal Component Analysis.						

Table 9. *Component Matrix Table of The Geochemical Data*

Component Matrix[a]			
	Component		
	1	2	3
CaO	0.90	-0.16	-0.13
K_2O	0.83	-0.21	0.37
MgO	-0.83	-0.50	-0.05
SiO_2	-0.82	0.18	0.36
TiO_2	0.79	0.09	0.47
Fe_2O_3	0.79	-0.05	0.38
Al_2O_3	-0.66	0.20	0.54
SO_3	-0.49	0.11	0.63
Zr	0.11	-0.88	0.36
Sr	0.06	0.84	0.02
LOI	0.31	0.84	0.07
% of Variance	**44.77**	**24.06**	**13.68**

Extraction Method: Principal Component Analysis
a: 3 components extracted.

The geochemical contents that create the three factors are shown with the principal component matrix (PCA) graph (Figure 4).

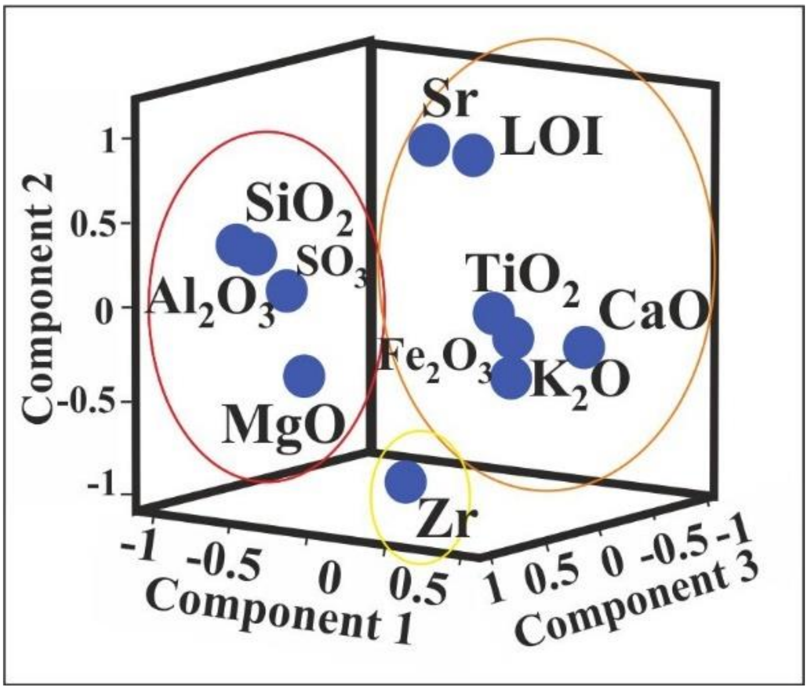

Figure 4. *Component Plot in The Varimax-Rotated Space:*
Component 1 (44.77%), Component 2 (24.06%) and Component 3 (13.68%)

Hierarchical classification of the geochemical contents of the marble samples was made in Figure 5.

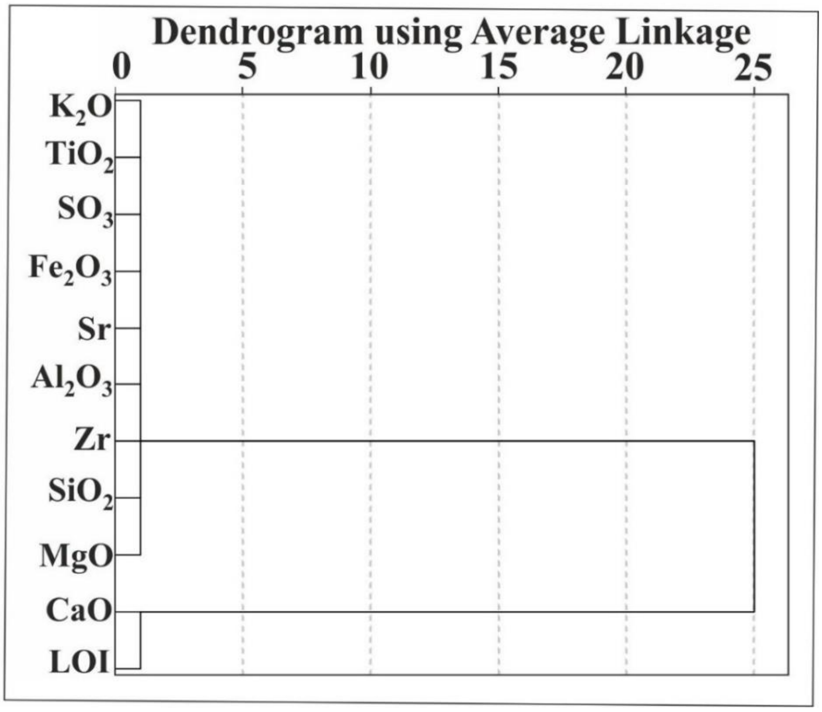

Figure 5. *Hierarchical Clustering of The Geochemical Data (Yazici, 2020)*

4. CONCLUSION

The data obtained were interpreted using their descriptive statistical data (measures of central tendency, measures of variability, and measures of the shape of the distribution). CaO, MgO, and SiO_2 were found to be the major element oxides that play the most significant role in the formation mechanism of marble samples.

The marbles vary in terms of their major and trace element compositions due to the metamorphism reactions they underwent during their formation. The studies conducted within this scope revealed negative correlations of CaO with SiO_2 and MgO, which are the major element oxides. Considering these correlations, the negative correlation of the major element oxide of CaO, which played the most role in the formation of "Emperador Dark" marbles, with SiO_2 and MgO could be associated with calcite and dolomite minerals, which were thought to decrease under metamorphism conditions. Moreover, the high positive correlation (0.780**) between Al_2O_3 and SiO_2 was associated with the presence of clay and mica group minerals in the environment.

According to factor analysis, the first factor i s 44.77% of the geochemical data; it was found that the second factor explained 68.84% of the geochemical data and the third factor explained 82.52% of the geochemical data. The geochemical data constituting the first factor are CaO, K_2O, TiO_2 and Fe_2O_3; It was determined that the geochemical data constituting the second factor were Sr and LOI and the geochemical data constituting the third factor were SiO_2, Al_2O_3, SO_3 and Zr. It has been determined that MgO is not found in its formation in all three factors. According to the hierarchical classification analysis, Fe_2O_3, Sr, Al_2O_3, Zr, SiO_2 and MgO formed the first cluster, CaO and LOI formed the second, and K_2O, which was not strongly selective, CaO ve Zr. The CaO and LOI cluster, which was extracted by hierarchical classification analysis and constituted the second group, was interpreted as an indirect indicator of CO_3 in the environment. in this context, the LOI value should be highest when the sample is pure calcitic.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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