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Research Article

ENVIRONMENTAL TOXICITY CHARACTERISTICS OF NEMRUT VOLCANISM PYROCLASTIC, BITLIS/TURKEY CASE

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ABSTRACT

This study aims to determine and evaluate the toxicity characteristics of Ahlat stone and pumice, which are pyroclastic rocks specific to Nemrut Volcanism and are common construction materials in the region. There are several studies in the literature that deals with constructional properties, but environmental properties have not been evaluated yet. Behaviour of Pyroclastic rocks due to the environmental conditions are important because pyroclastic rocks contain heavy metals (lead, chromium arsenic, etc.) in their structures due to geological activities. Depending on environmental conditions, these heavy metals can be released from the rock structure and cause hazardous health problems in people who contact them. This situation is directly related to the stability of the rock structure. The TCLP procedure was applied in this study to determine the stability of the rock structure and to determine the potential effect of heavy metals released from the structure on human health and the results obtained were evaluated. The obtained XRF results show that the different colored examined Ahlat stones consist mainly of SiO_2, Al_2O_3, K_2O, and Fe_2O_3 compounds.TCLP results show that the materials are in stable structures, they are not affected by environmental conditions and the amount of heavy metal leakage from structures is very low. However, chromium and arsenic in volcanic areas can have a carcinogenic effect on children. For this reason, a comprehensive soil and water pollution monitoring and analysis study is recommended in the volcanic area for public health.
Keywords: Ahlat stone, TCLP, toxicity, pyroclastic rocks.

1. INTRODUCTION

The use of natural stones in the construction industry is increasing day by day and gaining importance. A number of natural stones are formed as a result of volcanic activity. Volcanic activity is the evidence of the dynamic behavior and intense crust movement in the layers of the earth. The materials formed as a result of fragmentation regardless of the type of eruption of volcanic activities and the origin of the grains is called pyroclastic rocks. Pyroclastic rocks are composed of fragments that result from various volcanic eruptions or as a result of a direct eruption. Pyroclastics are rocks formed as a result of the volcanic material coming out of the volcano chimney to a sedimentation environment by means of volcanic mechanism and wind, joining each other with a temperature of over 500-600°C, or as a result of bonding of glassy material pieces with conversion product minerals (such as zeolite). Such materials remain in the

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environment for a long time and can be affected by environmental factors [1-6]. Pyroclastic rocks, which is evaluated in the natural stone class, have been widely used and continue to be used in different application areas in many settlements of the world due to the ease of obtaining, their processability properties, their applicability to traditional and modern construction technologies, their economical costs, lightness and insulation properties [7-11]. Turkey is a very rich country in terms of natural stones because of its geological structure. Numerous natural stones have been used in the region for different kinds of purposes since ancient times. Among these stones, Ahlat, Ankara, Midyat, Nevşehir, Oltu, Pileki, Lüle, Sille stones, travertine and marble are some natural stones whose economic and cultural value have reached the national dimension [12]. The natural stone origins mentioned in the text are shown on the map in Figure 1. This map was produced using Generic Mapping Tools [13].

Figure 1. Location of some natural stones in Turkey

The quarries where the best examples of Ankara Stone are taken from Gölbaşı district of Ankara. This stone is a product of volcanic common in the Central Anatolia Region. As it is understood from its name, Midyat Stone is located in Midyat district of Mardin. It is produced from Midyat formation spread over a wide area in the South-eastern Anatolia Region. Although Nevsehir stone is of volcanic origin, it is produced in the region between Nevsehir-Urgup-Avanos. It is a product of Cappadocia Volcanic Province. The production of Oltu Stone is performed in Oltu district of Erzurum. Oltu Stone is characterized as very high-calorie coal and / or lignite. The geology of Oltu Stone is explained by the Olurdere Formation that contains it. Pileki stone production is carried out around Rize, in the district of Rize Iyidere. The stone is extracted from Pileki Cave as before. Sille Stone is produced in Sille, which is a historical and cultural settlement center 15 km away from Konya city centre. The Meerschaum, which is identified with the city where it is named, is also known as Eskişehir Stone. It is extracted from underground in three different regions of Eskişehir province. It is produced efficiently in the Yakaboyu region, around the village of İmişehir-Karatepe and in the Humid Region near Porsuk Dam. Ahlat stone is also a natural stone which is a product of Nemrut Volcano [12].

Nemrut Mountain is located within the borders of the province of Bitlis, on the important crossing points of the Eastern Anatolia Region. Tatvan, Ahlat and Güroymak districts of Bitlis are the closest settlements to Nemrut Mountain. Ahlat and pumice stones have come to the fore as important pyroclastic rocks for med as a result of Nemrut Volcano's explosion over time. Both construction materials continue to be widely used for different purposes in the province and nearby settlements. While Ahlat Stone can be in different colors, there is no color change in pumice.

As a result of geological activity (volcanic eruption, earthquakes, landslides, etc.), some basic and trace elements can be included in the environmental circulation and hence within the structure of pyroclastic materials. Depending on the size of the volcanic eruptions, large amounts of ash and pyroclastic materials emerge. These structures do not only contain nutrients (nitrogen, phosphorus, potassium, calcium, etc.), they also contain toxic metals and metalloids (arsenic, mercury, etc.). Metals are resistant to environmental fragmentation, and when they exceed certain threshold values, they can have an ecotoxic effect and create environmental hazards [1]. For this reason, it is important to determine the amount of metal in the pyroclastic materials used as construction materials, to determine how much these metals can leak from the structure of these materials and to evaluate this situation in terms of environment and human health.

The toxicity characteristics of Ahlat and pumice stones, which belong to Nemrut Volcano within the borders of Bitlis and which are widely used as contruction materials in the region, were tried to be determined in the scope of the study. For this purpose, it has been tried to determine whether these construction materials are stable in terms of heavy metals by applying the Toxicity Characteristic Leaching Procedure (TCLP) for pumice samples in addition to Ahlat stones of different colors obtained from quarries. Composite compositions of color change were determined by applying X-Ray Fluorescence Spectroscopy (XRF) experiment to different color Ahlat stones. It is tried to be determined whether the result values for Ahlat stones are affected by the color change.

2. NEMRUT VOLCANO AND NEMRUT PYROCLASTICS

Nemrut Mountain is one of the largest of the quaternary volcanoes in Eastern Anatolia. It is a characteristic stratovolcano with large and small lakes, hot water and gas outlets in its 35 km caldera. The dimensions of the caldera are approximately 7.5 x 6.5 km and its vertical walls rise up to 2900 m. The Nemrut volcano has started to produce volcanic products from the early period of the Quaternary, and various pyroclastics have been formed that spread over large areas through a cleft eruption with the first volcanic activity [14]. It is asserted that pyroclastic material with a volume of about 100 km came out in this phase. These are observed from place to place as ignimbrites. Later, basic lavas came out and spread around. After this phase, intermediate and acidic lavas were formed and trachyandesite, trachyte, rhyolite and green colored obsidians were formed. Afterward, the Nemrut caldera was formed, following this tuff, ignimbrite, trachyte and trachyandesitic lavas and gray-black obsidian were formed in both the caldera and around the caldera. Nemrut volcanics cover all older units angular-unconformably. Nemrut Mountain, which forms the unit itself in the west of Lake Van, was used as the name [15-17]. Nemrut Volcano is an active volcano although it is asleep today. With the rise of the region due to the active tectonism in the Van Lake Basin, the fact that the rocks are resistant to weathering and abrasion also caused the formation of a highly inclined surface shape around the lake. It is known that lava flows occurred in 1441, 1597 and 1692 [18-21]. The location and tectonic structure of the Nemrut volcano is shown in Figure 2.

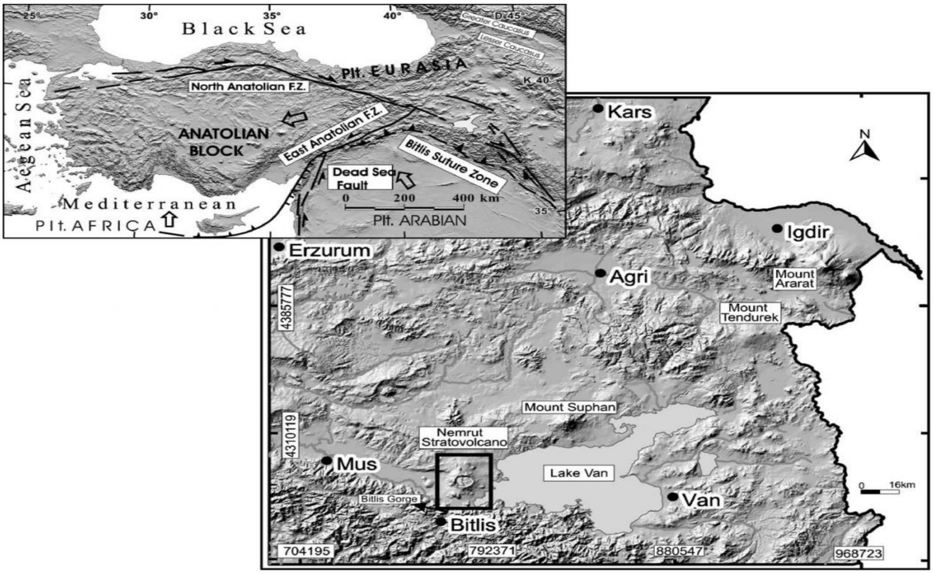

Figure 2. Location and tectonic structure of Nemrut volcano [19]

Ahlat stone, one of the pyroclastics which belong to Nemrut Volcanism, has been used extensively in mosques, minarets, cupolas, bridges, houses and tombstones, which are invaluable structures in terms of cultural and historical value in the district and continue to be used today. There are different color options of Ahlat stone and different choices can be made between these colors in accordance with the requests of the building owner. These stones, which are partially soft when extracted from the quarries, are cut into different sizes and harden as a result of interaction with the air over time. The shaping, lightness and high level of insulation properties of the Ahlat stone made it possible to use for constructions [5, 22-25]. Different usage areas of Ahlat stone are given in Figure 3.

Figure 3. The usage of Ahlat stone in different structures

Another product of Nemrut Volcano is pumice stone. Pumice material is a good example of light natural aggregates formed as a result of volcanic movements. It is a very porous glassy structure and it is formed by the gas output from the structure during the rapid cooling of the magma. It contains a lot of aluminum, iron and silica (SiO_2) in its structure. It has advantages such as low density, high heat and sound insulation, elasticity against seismic load and high plaster retention [26, 27]. As the pumice grain grows, specific weight of the grain decreases. As the grain sizes increase, the pore percentage increases. Pore percentage of pumice increases as it moves away from the volcano chimney. The excessive pore number and low specific weight have made the use of pumice for insulation purposes widespread. It is easy to grind as it is volcanic glass. It can be in different colors such as white, yellowish, gray, gray-brown and dull red. Pumice is widely used in different sectors [28-30].

3. MATERIAL AND METHOD

In the study, Toxicity Characteristic Leaching Procedure (TCLP), ICP analysis and X-Ray Fluorescence Spectroscopy (XRF) experiments were carried out for Ahlat and pumice stones of different colors.

3.1. Analysis Used

TCLP is a well-established technique created by the United States Environmental Protection Agency (USEPA) for the determination and assessment of toxicity [31]. It is designed to determine the mobility of organic and inorganic pollutants in liquid, solid or multi-phase wastes. Hazardous and non-hazardous wastes are distinguished through this procedure by simulating the leakage of pollutants in storage areas. However, there are studies indicating that the mobilization of heavy metals is negatively affected due to the high pH of the solution obtained as a result of the process. According to these studies there are limitations of the method in the evaluation of cemented wastes. In addition, it was observed that wastes that were not classified as hazardous at the time of storage due to pH fluctuations in the storage area may become dangerous over time [32]. The TCLP procedure created by the United States Environmental Protection Agency (USEPA) was used to determine the toxicity characteristics of the materials used as construction materials within the scope of the study [33]. The experimental study was carried out as stated in Bayraktar et al. (2015) [34] and elutes obtained were included in the ICP analysis. In terms of TCLP procedure, all samples complied with solution 1 and after the 18 hours reaction time, final pH values were determined as <5 for all samples. The eluates obtained as a result of the TCLP procedure were first filtered through a 0.45 μm membrane filter, then analyzed according to the EPA200.8 method in the existing ICP-MS device at Ağrı Ibrahim Çeçen University Science and Tehnology Application and Research Center [35-37]. XRF analysis is used for quantitative analysis (in the range of % or ppm) of elements with atomic numbers between 9 and 92 (Sodium to Uranium) in solid, liquid and powder samples. X-ray spectroscopy determines the chemical elements in the structure of a sample and the quantities of these elements by measuring the characteristic x-rays emitted from the atoms in the structure. This analysis is frequently used in different fields such as environment, geology, mineralogy and food [38-41]. X - Rays Fluorescence Spectrometry (XRF) analyzes were performed with PANalytical B.V. XRF device located in Bulent Ecevit University Science and Technology Application Center. Device is controlled by Epsilon 5 software [38-39].

3.2. Obtaining Samples

As part of the study, three different Ahlat stones and pumice samples which are products of Nemrut Volcano and which are obtained from the quarries located within the borders of the Ahlat district of Bitlis province were taken into consideration. The views of the construction materials used in the study are given in Figure 4.

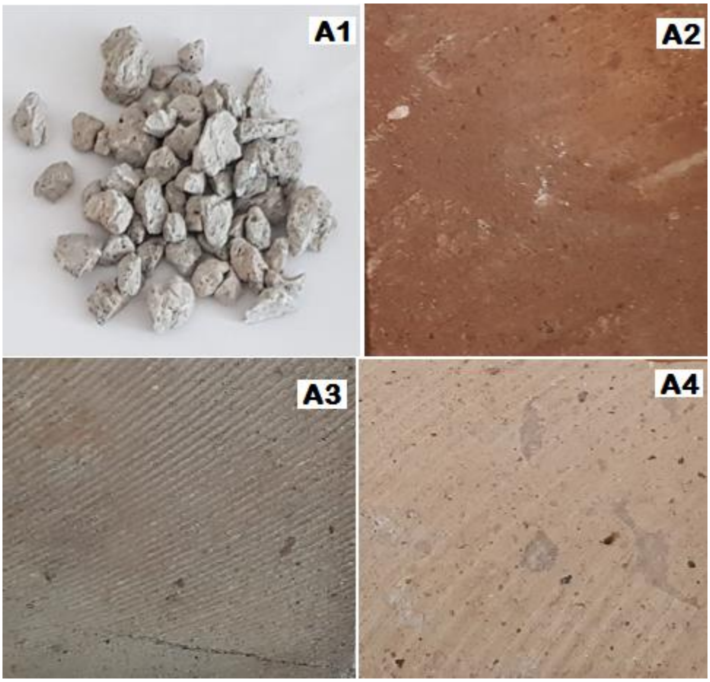

Figure 4. Samples taken into consideration in the study; A1: pumice; A2: Ahlat Stone (red), A3: Ahlat Stone (black), A4: Ahlat Stone (white)

4. TEST RESULTS

The XRF analysis results of different colored Ahlat stones are given in Table 1. XRF results show that all 3 different samples are mainly composed of silicon aluminum, potassium and sodium oxides. Results of the comparison of the analyzed construction materials with TCLP analysis and the TCLP limits are given in Table 2. The obtained TCLP results are well below the limit values given by USEPA. This shows that the examined material has a very stable structure and is not affected by changes in environmental conditions. Due to the limited amount of metals examined with the TCLP procedure, in order to expand the evaluation, the materials were accepted to be the wastes desired to be stored and the results were compared with the limits given for the storage of wastes. The obtained results are below the limit values for the storage of waste in both the US and Turkey. This shows that the materials do not pose a risk in terms of the metals examined under extreme environmental conditions.

Table 1. XRF Analysis Results of Ahlat Stone Samples

Compound	Unit	Sample		
		A2	A3	A4
Na_2O	%	3.322	3.019	2.876
MgO		0.530	0.466	0.266
Al_2O_3		22.182	21.589	20.404
SiO_2		58.868	62.37	64.398
Cl		0.0109	0.0580	0.0969
K_2O		6.607	5.923	5.898
CaO		1.437	ND*	ND
TiO_2		0.336	0.457	0.319
Fe_2O_3		6.409	5.757	5.329
Rb_2O		0.026	ND	ND
ZrO_2		0.128	0.321	0.298
BaO		0.144	ND	ND
ZnO		ND	0.040	0.032
CeO_2		ND	ND	0.084
Total		100.0	100.0	100.0

*Not detected

Table 2. TCLP analysis results of Ahlat stones and pumice samples and comparison with limit values

Elements	Amount leached to TCLP liquid				TCLP	EPA land disposal restriction limit values (mg/L) [34, 36]	Regulation on Landfilling of Wastes (RLW 2010)		
	mg/L				Limit		Landfilling criteria for inert wastes	Landfilling criteria for non-hazardous wastes	Landfilling criteria for hazardous wastes
	A1	A2	A3	A4	mg/L		mg/L		
Be	0.0010	0.0003	0.0007	0.0016					
B	0.2680	0.0270	0.0552	0.0474					
Na	71.2200	72	72	72					
Mg	1.8510	2.2910	2.5120	1.7370					
Al	0.4449	0.5228	0.5470	0.4090					
K	4.7390	3.6910	5.5000	5.6850					
Ti	0.0069	0.0024	0.0044	0.0056					
V	ND	ND	ND	ND					
Cr	0.0093	0.0129	0.0142	0.0165	5	0.6	0.05	1	7
Mn	0.1535	0.0736	0.1086	0.0707					
Fe	0.3457	0.3096	0.3230	0.2980					
Co	0.0013	0.0025	0.0110	0.0010					
Ni	0.0094	0.0079	0.0138	0.0093		11	0.04	1	4
Cu	0.0074	0.0074	0.0088	0.0131					
Zn	0.1553	0.0412	0.0617	0.1016		4.3	0.4	5	20
As	0.0011	0.0016	0.0015	0.0020	5		0.05	0.2	2.5
Se	0.0002	0.0002	0.0007	0.0006			0.01	0.05	0.7
Sr	0.1471	0.1133	0.5500	0.3010					
Mo	0.0006	0.0013	0.0016	0.0015			0.05	1	3
Cd	0.0008	0.0005	0.0007	0.0013	1	0.11	0,004	0.1	0.5
Sn	0.0070	ND	0.0058	0.0066					
Sb	0.0002	0.0001	0.0001	0.0003			0.006	0.07	0.5
Ba	0.0336	0.0648	0.0763	0.0231			2	10	30
W	0.0001	0.0001	0.0001	0.0001					
Hg	0.0007	0.0003	0.0002	0.0002	0.2		0.001	0.02	0.2
Pb	0.0022	0.0017	0.0022	0.0062	5	0.75	0.05	1	5
Bi	0.0000	ND	0.0000	ND					
Si	22.2033	16.7700	17.5300	16.1500					
Ca	50.6233	34.7300	85.2900	27.8600					

The amount of metal leaking from Ahlat Stone samples was compared with the metal content in the structure within the context of the study and given in Table 3, Table 4 and Table 5. When the A2 coded red Ahlat Stone is examined, it is understood that the most important metals leaking from the structure are Na (13.5%), Ca (8.1%) and Mg (3.2%), and the transition of other metals to the liquid is negligible.

Table 3. Leakage amounts of elements in A2 coded Ahlat stone

Elements	%	g in kg	mg in kg Stone	mg in kg TCLP liquid	% Leakage
Na	3.168	31.680	31680	4277	13.5
Mg	0.426	4.260	4260	136	3.2
Al	18.092	180.920	180920	31	0.02
Si	49.832	498.320	498320	996	0.2
K	12.852	128.520	128520	219	0.17
Ca	2.546	25.460	25460	2063	8.1
Ti	0.533	5.330	5330	0.14	0.003
Fe	12.038	120.380	120380	18	0.01
Ba	0.211	2.110	2110	3.85	0.18

As a result of exposition of the A3 coded black Ahlat stone to TCLP procedure, it can be seen from Table 4 that the most leaking elements of the structure are Na (15%) and Mg (4%) and no significant leakage could be detected for other elements.

Table 4. Leakage amounts of elements in A3 coded Ahlat stone content

Elements	%	g in kg	mg in kg Stone	mg in kg TCLP liquid	% Leakage
Na	2.853	28.530	28530	4277	15.0
Mg	0.370	3.700	3700	149	4.0
Al	17.772	177.720	177720	32	0.02
Si	54.143	541.430	541430	1041	0.19
K	12.091	120.910	120910	327	0.27
Ti	0.796	7.960	7960	0.26	0.0003
Fe	11.221	112.210	112210	19	0.02

As a result of the interaction of A4 coded white Ahlat stone with TCLP fluid, it is seen from Table 5 that the most leaking metals from the structure are Na and Mg, similar to A2 and A3, and leakage from other metals is negligible.

Table 5. Leakage amounts of elements in A4 code Ahlat stone

Elements	%	g in kg	mg in kg Stone	mg in kg TCLP liquid	% Leakage
Na	2.694	26.940	26940	4277	15.9
Mg	0.208	2.080	2080	149	7.2
Al	16.819	168.190	168190	32	0.02
Si	56.127	561.270	561270	1041	0.19
K	12.236	122.360	122360	327	0.27
Ti	0.509	5.090	5090	0.26	0.01
Fe	10.474	104.740	104740	19	0.02
Zn	0.079	0.789	789	6.03	0.76

As a result of the study, it is seen that the metals do not pose a danger to the environment and public health. However, it is stated in the literature that the radiation effect is an environmental factor that should be taken into consideration in the buildings where Ahlat stone is used [42].

In the study related to soil pollution around Tianchi Volcano in North Korea, it is stated that phosphate holding capacity and volcanic glasses are quite high as in volcanic soils. It was determined that Fe, K, Na, Mn, Pb, Zn, As and Sn concentrations were high in the soil near Tianchi, and Ca, Mg, Ti, Cu, Cr, Ni, Ba, Co and Sr concentrations were high in the study area from west to north. Although metal levels in volcanic ash and soil are considered safe, it has been observed that Zn concentration can increase up to 4 times compared to the background value. Although the ecological risk is low in the study area, it is especially stated that care should be taken in terms of the carcinogenic effects of Cr and as on children [1].

5. DISCUSSION AND CONCLUSION

Ignimbrites consisting of lava of Nemrut Mountain, which is one of the important volcanoes of our country and located within the borders of Bitlis province, are commonly found in this region. These stones are generally used as building materials in the region. The unit volume weight of the Ahlat stone takes very low values. This reduces the amount of load that will occur in the structure. The compressive and tensile strength of Ahlat stone is very low. However, it is a good thermal insulation material due to its hollow structure. Another product obtained from Nemrut volcano is pumice. Pumice, which has a significant amount of reserves in the Bitlis region, is widely used as a wall material for insulation purposes. Pumice produced from pumice in Bitlis province is also widely used in other provinces in the region. Currently, there are many facilities that produce pumice and Ahlat stones. Ahlat Stone and pumice are one of the important economic incomes of the region. In this context, these two building materials are very useful and important for the region.

Within the context of the study, the characteristics of Ahlat stone and pumice, which are specific to Nemrut Volcanism, and which are formed as a result of volcanic activity, were examined using the TCLP procedure. TCLP is one of the USEPA procedure for the determination and assessment of toxicity. It is designed to determine the mobility of organic and inorganic pollutants in liquid, solid or multi-phase wastes. In this study it was used to determine mobility of elements exist in stone samples.

Volcanic activity causes various metals in the underground to move towards the earth and mix with the natural stone structure formed by eruption. The stability of such materials is important in terms of creating environmental and public health hazard through the interaction of metals in the contents of materials with the environment. XRF results indicated that the different colored examined Ahlat stones consist mainly of SiO_2, Al_2O_3, K_2O, and Fe_2O_3 compounds. According to TCLP results Ahlat stone and pumice samples are stable structures. The studied elements are kept in this stable structure and there is no significant oscillation outside the structure due to environmental effects. This shows that the metals in the content will not cause any environmental and public health damage. However, studies in the literature show that concentrations of some metals may increase as a result of geological activity in volcanic areas. It is stated that especially with the increase of Cr and As, carcinogenic effects can be seen in children exposed to them. In this context, it is recommended to conduct a comprehensive monitoring and analysis study in terms of metals in volcanic areas and construction materials obtained from these areas and to evaluate the effects on the environment and public health.

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