© by PSP Volume 26 – No. 3/2017, pages 2087-2092 Fresenius Environmental Bulletin

A SURVEY ON ELEMENTAL DISTRIBUTIONS OF VOLCANIC TUFF QUARRIES IN TURKEY

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ABSTRACT

Volcanic tuff stones are widely used as coating materials for insulation and/or ornamental purposes in the construction sectors in Turkey. Some volcanic tuff quarries are close to the residential areas, water resources and agricultural lands. The study presents the results of analyses of twenty-eight elements in seventy-six volcanic tuff stone samples collected from seventeen quarries generally located Cappadocia region which is a popular tourist destination. Nine major oxides (SiO_2, CaO, TiO_2, Fe_2O_3, Al_2O_3, MgO, Na_2O, P_2O_5 and K_2O) and nineteen minor elements (V, Sc, Cr, Mn, Co, Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Ba, La, Ce, Pb, Th and U) were analyzed by using a wavelength dispersive X-ray fluorescence spectrometer. The highest average concentration of environmental polluting elements V, Cr, Mn, Co, Ni, Cu, Zn, Sr, Pb, Th and U analyzed in volcanic tuff samples are 117.7, 256.4, 1578.0, 71.1, 300.5, 60.2, 141.6, 2452.2, 99.7, 102.8 and 40.1 mg/kg, respectively.

KEYWORDS:
Volcanic tuff, Chemical composition, Environment, Polluting elements, Major oxides, WDXRF

INTRODUCTION

Our environment continues to be damaged or polluted by toxic metals, persistent organic pollutants, radionuclides and other hazardous materials. Especially pollutant discharges from mining and other sources have led to metal toxic elements accumulation in air, food, water and soil or land. Turkey has one third of the world's total natural stone (volcanic tuff, granite, marble, travertine, andesite and basalt) reserve and the diversity of the reserve is great with more than 400 different colours and patterns [1]. Steadily growing production of natural stones can cause several environmental problems such as water and soil pollution. Also, accumulation of slag of the natural stones can lead to environmental radiological problems through (a) its dispersal into atmosphere (b) its handling or disposal.

Volcanic tuff stones having a porous in different colours structure were consisted of consolidated volcanic ash, and large and small pieces ejected from vents during a volcanic eruption [2]. There are volcanic rocks originating from Paleozoic and probably older as well as Mesozoic, Tertiary and even Quaternary in Turkey [3]. Volcanic rocks of Tertiary and Quaternary age are widespread in Turkey and these volcanic rocks are formed in various volcanic provinces such as Nevşehir, Kayseri, Isparta, Afyon, Manisa, Eskişehir and Diyarbakır [4]. Volcanic tuff stones have various colours and appearances due to the component types and chemical properties of matrix [1]. Therefore, volcanic tuff stones are widely used as coating material of the interior and exterior surfaces of the buildings in Turkey. Also, they are used as structural material in the construction of inner and/or outer walls of masonry buildings especially in Cappadocia region because the masonry walls made of tuff stone is more durable than concrete and brick walls [1,2]. In addition to volcanic tuffs are used in the cement industry as an admixture for the production of pozzolanic cements [2].

There are several studies related to volcanic stones in the literature [1,2,4-11]. However, according to our literature survey, a comprehensive study related to the elemental distributions of the volcanic tuff quarries in Turkey has not yet been performed. Therefore, the aim of the study is to determine the elemental distributions of seventeen quarries located in Nevşehir, Kayseri, Isparta, Afyon, Manisa, Eskişehir and Diyarbakır. For this aim, the concentrations of nine major oxides (SiO_2, CaO, TiO_2, Fe_2O_3, Al_2O_3, MgO, Na_2O, P_2O_5 and K_2O) and nineteen minor elements (V, Sc, Cr, Mn, Co, Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Ba, La, Ce, Pb, Th and U) in seventy-six volcanic tuff stone samples collected from seventeen quarries were measured by using a wavelength-dispersive X-ray fluorescence (WDXRF) spectrometer.

TABLE 1
Place of quarry, sample code and sample colors.

Quarry code	N	Sample colours (number of samples)	Quarry place (Geographical region)
Q1	6	Yellow (2), black (1), cherry (1), dark gray (1), dusty-rose (1),	Kayseri-Tomarza (Central Anatolia)
Q2	6	Yellow (2), brown (1), black (1), dusty-rose (2)	Kayseri-Tomarza (Central Anatolia)
Q3	7	Cherry (1), red (1), brown (1), yellow (1), gray (1), dusty-rose (1), black (1)	Kayseri-Tomarza (Central Anatolia)
Q4	7	Gray (4), black (1), dusty-rose (1), brown (1)	Kayseri-Tomarza (Central Anatolia)
Q5	4	Yellow (3), brown (1)	Kayseri-Tomarza (Central Anatolia)
Q6	6	Yellow (1), black (1), gray (1), dusty-rose (1), fawn (1), dark brown (1)	Kayseri-Develi (Central Anatolia)
Q7	3	Gray (4)	Kayseri-Melikgazi (Central Anatolia)
Q8	3	Gray (2), yellow (1)	Kayseri (Central Anatolia)
Q9	6	Yellow (1), cherry (1), black (1), white (1), red (1), turquoise (1)	Nevşehir-Ürgüp (Central Anatolia)
Q10	5	Yellow (1), cherry (1), dusty-rose (1), light brown (1), black (1)	Nevşehir-Avanos road (Central Anatolia)
Q11	4	Yellow-white (1), white (1), dark yellow (1), Cappadocia rose (1)	Nevşehir-Aksaray road (Central Anatolia)
Q12	1	Brown (1)	Nevşehir-Ürgüp (Central Anatolia)
Q13	6	Fawn (2), dark brown (1), beige (1), brown (1), cream (1)	Isparta (Mediterranean)
Q14	6	Beige (1), yellow (1), gray (1), cream (1), dusty-rose (1), white (1)	Afyon (Aegean)
Q15	2	Brown (2)	Manisa (Aegean)
Q16	2	Rose-colored (2)	Eskişehir (Central Anatolia)
Q17	2	Maroon (1)	Diyarbakır (Southeast Anatolia)

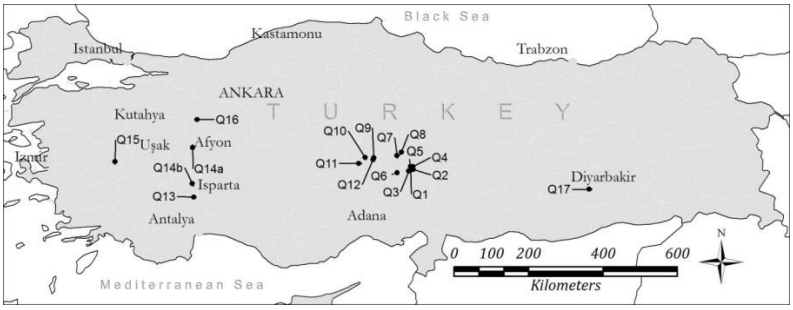

FIGURE 1
Simplified map showing the locations of volcanic tuff quarries.

MATERIALS AND METHODS

Sample gathering and sample preparation. **S**ampling sites were selected in seventeen quarries located in Central Anatolia, Eastern Anatolia, and Mediterranean and Aegean regions in Turkey (Fig. 1). Seventy-six volcanic tuff stone samples collected from these quarries were put in polyethylene bags, transported to the laboratory, catalogued and coded properly (Table 1). The samples were crushed and then dried in a temperature-controlled furnace at 75 ºC for 4 h to remove moisture. They were powdered and homogenized using agate grinders. 12 g of the sample was mixed with 3 g of cellulose in a mortar made of boron carbide for 5 min. The sample was pressed at 25 tons pressure placing into 40 mm diameter stainless steel pellet set for 1 minute. Powdered pellet was placed into sample containers made of stainless steel and then counted for 2 hours. The concentrations of elements in the sample were calculated as ppm (mg/kg).

X-ray fluorescence analysis. Different spectrometers such as inductively coupled plasma-optical emission spectrometer (ICP-OES) were used for the determination of major, minor and trace elements in various samples such as foods and dust [12,13]. X-ray fluorescence (XRF) method, which bases on excitation of characteristic X-rays in a sample as a result of interaction of primary X-ray beam with sample atoms, is a well-respected and reliable method for determining elemental composition of the samples. The elemental composition analyses of volcanic tuff samples were performed using a WDXRF spectrometer (PANalytical Axios advanced model). The specifications of the spectrometer are given in Table 2. For the analysis of major elements, the spectrometer used Super IQ- Analysis Software was calibrated using advanced fundamental parameters algorithm. For the accurate analysis of trace elements Pro-Trace software was used.

TABLE 2
Specification of the X-ray fluorescence spectrometer.

Module	Specification
Software	Super IQ and 2- Pro-Trace
Power	4 kW
Tube	Rh anode, SST (Super Sharp Tubes) type, 160 mA Tube filters: brass, beryllium and aluminum of 0.2 and 0.7 mm
Crystal	7 crystal (PX10, Ge-111-C, PE002-C, PX1, PX4A, PX7, LiF220)
Collimator	3
Mask	6 different dimensions
Detector	Fluid and scintillation
Standard	IQ+ (16 standards) and Pro-Trace (26 standards)
Measurement system	Capability of 60 consecutive sample measurements

TABLE 3
Statistical data of concentrations of major oxides analyzed in all volcanic tuff samples and their average abundance in the earth's crust.

	Concentration (%)								
	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5
Average	55.50	11.50	4.20	1.00	2.50	4.60	2.00	0.90	0.20
SD*	10.40	2.50	3.40	1.00	2.50	1.70	1.40	0.90	0.20
SE*	1.20	0.30	0.40	0.10	0.30	0.20	0.20	0.10	0.02
Median	58.00	11.00	3.70	0.70	2.00	4.80	1.60	0.60	0.10
Min	22.00	5.00	0.60	0.10	0.10	0.10	0.30	0.10	0.01
Max	69.00	22.10	16.70	5.20	12.70	7.30	7.00	4.30	0.90
Skewness	-1.00	1.60	1.30	1.90	2.00	-0.80	1.40	4.20	0.90
Kurtosis	0.70	4.70	1.80	4.20	4.50	-0.10	1.90	1.70	1.80
Average abundance in the earth's crust	54.55	16.17	0.92	4.91	8.72	2.74	1.32	0.86	0.20

*SD: standard deviation; SE: standard error

RESULTS AND DISCUSSION

Seventy-six volcanic tuff stone samples collected from seventeen quarries in Turkey were analyzed using the WDXRF spectrometer for determining chemical compositions of the samples and elemental distributions of the quarries. A total of twenty-eight elements were detected in the samples. Some statistical data of the concentrations of major oxides and elements in the samples and comparison of the concentrations of these elements with their average abundance in the earth's crust taken from Yaroshevsky (2006) are given in Table 3 and Table 4, respectively [14]. As can be seen from Table 3, the average concentrations of Fe_2O_3, Na_2O and K_2O measured in all samples are higher than the average abundance of earth crust while the average concentrations of SiO_2, Al_2O_3, MgO, CaO, TiO_2 and P_2O_5 measured in the all samples are lower or equal than the average abundance of earth crust. From Table 4, the average concentrations of environmental pollutant elements V, Cr, Mn, Co, Ni, Cu, and Zn are lower than their average abundance of earth crust while the average concentrations of Ba, Zr and Sr are higher than their average abundance of earth crust while the average.

The average concentration of toxic element Pb is about two times higher than the average abundance of earth crust. The average concentrations of earth elements Y, La and Ce are higher than the average abundance of earth crust while the average concentration of Sc is about two times lower than the average abundance of earth crust. The average concentration of radioactive element U is four times higher than the average abundance of earth crust while the average concentration of radioactive element Th is two times higher than the average abundance of earth crust. The average concentrations of oxide contents, expressed as weight percentage of oxides, for volcanic tuff quarries are reported in Table 5. The concentrations of SiO_2, CaO, TiO_2, Fe_2O_3, Al_2O_3, MgO, Na_2O, P_2O_5 and K_2O in all volcanic tuff quarries varied from 22.0 to 69.0 %, 0.1 to 12.7 %, 0.1 to 4.3 %, 0.6 to 16.7 %, 5.0 to 22.1 %, 0.1 to 5.2 %, 0.1 to 7.3 %, 0.01 to 0.92 % and 0.3 to 7.0 %, respectively.

As can be seen from Table 5, the highest average concentration of SiO_2 (66.7 %), CaO (7.6 %) and K_2O (6.6 %) were found in Q8, Q7 and Q5, respectively. The highest average concentration of TiO_2 (4.3 %) and Fe_2O_3 (16.7 %), Al_2O_3 (17.6 %) and Na_2O_3 (6.3 %) and MgO (3.5 %) and P_2O_5 (0.8 %) were found in Q17, Q11 and Q15, respectively. The average concentrations of elements for volcanic tuff quarries are given in Table 6.

TABLE 4
Statistical data of concentrations of minor elements analyzed in all volcanic tuff samples and their average abundance in the earth's crust.

	Concentration (mg/kg)																	
	Sc	Y	La	Ce	Ba	Zr	Nb	Rb	V	Cr	Mn	Co	Ni	Cu	Zn	Sr	Pb	Th	U
Average	4.65	32.94	59.45	95.31	671.81	290.68	24.44	139.94	42.44	11.88	581.51	11.66	15.04	14.10	51.28	401.17	28.08	29.86	10.21
SD	3.21	14.74	72.77	102.29	1255.26	130.94	19.54	101.17	42.26	38.55	331.80	12.61	38.40	13.76	26.31	687.00	33.00	25.00	11.00
SE	0.58	1.74	8.58	12.05	147.93	15.43	2.30	11.92	4.98	4.58	39.10	1.49	4.53	1.63	3.10	81.03	3.92	2.95	1.30
Median	3.65	32.94	37.50	65.26	529.00	335.50	16.76	134.50	29.24	2.45	593.50	8.02	5.00	10.57	48.03	194.50	13.45	23.64	6.15
Min	0.16	10.22	6.26	33.93	31.00	92.00	7.91	8.00	0.92	0.73	12.00	34.00	1.88	1.70	14.03	19.00	0.50	2.07	0.05
Max	11.93	78.53	323.61	519.87	10269.00	585.00	93.29	497.00	181.13	256.39	1578.00	71.06	300.49	63.53	141.64	3502.00	116.76	111.85	47.22
Skewness	1.08	0.97	2.99	3.00	6.79	0.18	2.12	1.75	1.66	5.49	0.74	2.77	6.28	1.83	1.29	3.05	1.73	2.12	2.05
Kurtosis	0.36	1.53	7.50	7.95	50.24	-0.54	3.64	3.61	2.40	31.03	0.45	8.94	44.53	3.53	1.82	9.08	1.41	4.13	3.17
Average abundance in the earth's crust	10.00	29.00	29.00	70.00	650.00	170.00	20.00	150.00	90.00	83.00	1000.00	18.00	58.00	47.00	83.00	340.00	16.00	13.00	2.50

TABLE 5
Average concentration of major oxides analyzed in volcanic tuff samples.

Quarry code	Average concentration (%)								
	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	P_2O_5
Q1	55.0	1.2	11.2	5.8	1.1	3.5	5.5	1.3	0.3
Q2	58.5	0.7	11.4	4.2	0.6	2.0	6.1	1.1	0.2
Q3	57.0	1.3	11.0	4.4	0.9	2.5	5.3	1.9	0.2
Q4	56.3	1.4	11.9	6.3	0.9	3.1	5.2	1.4	0.2
Q5	58.0	1.0	10.5	3.2	0.7	1.4	4.8	6.6	0.1
Q6	58.3	0.7	10.9	4.2	0.7	2.3	5.4	1.7	0.2
Q7	44.0	1.8	10.8	6.6	2.7	7.6	4.3	0.9	0.4
Q8	66.7	0.3	10.8	1.1	0.3	0.6	6.1	1.6	0.1
Q9	55.2	0.7	10.8	3.6	1.0	1.9	5.5	1.5	0.3
Q10	65.0	0.3	15.8	2.7	0.5	1.1	-*	1.7	0.1
Q11	65.5	0.2	17.6	0.7	0.2	1.4	6.3	0.9	0.1
Q12	51.0	0.3	9.0	1.4	1.7	1.0	1.5	3.7	0.0
Q13	49.7	0.7	11.7	3.6	1.4	2.6	3.5	4.3	0.2
Q14	56.5	0.1	9.3	2.7	0.2	0.7	3.0	3.1	0.0
Q15	33.0	3.4	11.0	10.0	3.5	6.6	2.7	0.9	0.8
Q16	52.5	0.2	9.0	1.4	1.6	0.9	1.4	3.9	0.0
Q17	22.0	4.3	5.0	16.7	2.6	6.5	0.5	0.3	0.7

* Not observed

TABLE 6
Average concentration of minor elements analyzed in volcanic tuff samples.

Quarry code	Average concentration (mg/kg)																		
	Sc	V	Cr	Mn	Co	Ni	Cu	Zn	Rb	Sr	Y	Zr	Nb	Ba	La	Ce	Pb	Th	U
Q1	-*	65.7	7.7	613.3	17.4	8.8	24.6	49.5	92.7	236.5	33.7	322.8	15.2	476.5	30.1	66.1	11.0	18.6	4.2
Q2	-*	36.3	2.3	538.3	9.4	5.6	14.5	45.6	96.0	222.8	35.3	349.8	16.1	583.3	35.0	74.6	13.0	20.2	5.9
Q3	6.5	27.5	4.2	571.4	13.6	6.8	5.5	42.6	97.0	208.6	33.3	324.9	15.6	463.1	38.4	62.8	11.8	18.7	6.1
Q4	-*	81.9	6.6	680.0	16.1	9.7	25.7	59.4	91.6	239.3	37.8	304.1	15.2	531.0	42.6	63.0	11.0	18.2	4.5
Q5	5.0	14.9	-*	429.0	5.7	3.6	-*	44.2	120.5	145.5	54.6	369.0	17.5	486.5	54.5	108.3	14.3	49.8	13.2
Q6	-*	39.9	2.6	515.2	8.6	4.2	12.7	49.4	115.7	191.2	29.7	320.7	16.0	529.8	38.0	69.0	13.3	20.3	4.3
Q7	7.3	73.7	75.0	779.3	22.3	65.6	15.2	60.6	49.8	366.8	26.5	219.3	11.7	327.0	34.1	52.3	10.7	10.3	4.9
Q8	-*	10.1	2.3	287.3	8.9	4.5	6.9	32.7	154.7	68.3	43.0	347.0	18.8	575.0	43.3	76.7	13.6	29.4	7.2
Q9	-*	46.9	4.3	447.8	10.3	8.4	17.4	40.8	114.3	188.8	32.9	360.5	17.0	549.7	37.4	70.1	12.6	21.6	5.7
Q10	-*	41.4	3.1	406.6	7.9	3.4	12.1	47.1	132.2	106.8	18.8	199.6	16.1	430.2	35.7	60.1	16.8	24.5	4.7
Q11	-*	15.4	2.6	146.0	7.5	2.2	7.3	19.4	93.8	67.0	11.8	110.3	13.6	132.5	35.6	50.3	16.5	24.4	4.6
Q12	2.6	10.1	-*	410.0	4.6	4.8	-*	28.6	174.0	204.0	12.3	104.0	13.3	637.0	37.2	54.4	20.8	23.2	9.5
Q13	1.5	35.5	8.3	797.8	6.4	17.0	15.7	106.8	209.3	2452.2	29.8	554.7	66.1	3140.8	295.3	422.8	99.7	102.8	40.1
Q14	3.3	4.4	-*	608.2	1.2	4.9	-*	33.5	408.8	33.0	59.6	102.7	41.6	55.8	26.4	43.4	98.1	47.4	24.9
Q15	4.8	117.7	3.5	1222.5	26.9	19.1	26.6	66.2	67.0	1243.5	25.2	250.0	92.8	952.0	79.1	126.7	2.8	10.3	8.6
Q16	3.3	10.0	4.4	587.5	1.5	16.0	-*	35.3	227.5	97.5	25.5	117.5	22.0	620.0	42.3	61.0	54.4	23.7	11.1
Q17	11.9	101.1	256.4	1578.0	71.1	300.5	60.2	141.6	24.0	597.0	18.9	225.0	27.7	94.0	32.6	68.6	4.6	4.3	4.6

* Not observed

The concentration of V, Cr, Mn, Co, Ni, Cu, Zn, Sr and Pb environmental polluting elements detected in all volcanic tuff quarries varied from 0.9 to 181.1 mg/kg, 2.0 to 256.4 mg/kg, 12.0 to 1578.0 mg/kg, 0.3 to 71.1 mg/kg, 1.9 to 300.5 mg/kg, 1.7 to 63.5 mg/kg, 14.0 to 141.6 mg/kg, 19.0 to 3502.0 mg/kg and 0.5 to 116.8 mg/kg, respectively. The concentration of Th and U radioactive elements detected in all volcanic tuff quarries varied from 2.1 to 111.9 mg/kg and 0.1 to 47.2 mg/kg, respectively. It can be seen from Table 6 that the highest average concentration of Cr, Mn, Co, Ni, Cu, and Zn were detected in volcanic tuff samples from Q17. The highest average concentration of V was found in Q15 while the highest average concentration of Sr, Pb, Th and U were found in Q13. Cr and Cu were not observed in volcanic tuff samples from Q5, Q12, Q14 and Q5, Q12, Q14 and Q16, respectively. The concentration of Sc, Y, La and Ce rare earth elements detected in all volcanic tuff samples varied from 0.2 to 11.9 mg/kg, 10.2 to 78.5 mg/kg, 6.3 to 323.6 mg/kg and 33,9 to 519.9 mg/kg, respectively. From Table 6, the highest concentration of Sc (11.9 mg/kg) and Y (59.6 mg/kg) were analyzed for Q17 and Q14, respectively. The highest average concentration of La and Ce were found in Q13. Sc was not observed in volcanic tuff samples from Q1, Q2, Q4, Q6, Q8, Q9, Q10 and Q11.

CONCLUSIONS

Elemental analysis of 76 volcanic tuff stone samples collected from different 17 quarries located in Central Anatolia, Eastern Anatolia, and Mediterranean and Aegean regions in Turkey were

performed using WDXRF technique. SiO_2, CaO, TiO_2, Fe_2O_3, Al_2O_3, MgO, Na_2O, P_2O_5, K_2O, V, Sc, Cr, Mn, Co, Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Ba, La, Ce, Pb, Th and U were detected in the volcanic tuff stone samples. The average concentrations of toxic element Pb measured in the samples from quarry of Q10, Q11, Q12, Q13, Q14 and Q16 are higher than their average abundance in the earth's crust. The average concentrations of radioactive element U measured in the samples from all quarries are higher than the average abundance in the earth's crust. The average concentrations of radioactive element Th measured in the samples from all quarries are higher than the average abundance in the earth's crust except for Q7, Q15 and Q17. The average concentrations of environmental polluting elements measured in the samples from Q17 are higher than their average abundance in the earth's crust.

ACKNOWLEDGEMENT

This study is carried out within the framework of a master thesis conducted at Kastamonu University and Research Project 114Y042 supported by The Scientific and Technological Research Council of Turkey (TÜBİTAK). The authors would like to thank Turkish Atomic Energy Authority (TAEK) and TÜBİTAK.

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Received: **02.09.2016**
Accepted: **07.02.2017**

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