



## Gamma Ray Shielding Properties and Natural Radioactivity of Some Rocks of Trabzon District, Türkiye



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**I**N THIS study, the gamma-ray attenuation properties of spilitic basalt, andesite, dolomite, limestone and syenite rocks obtained from Trabzon Province and its vicinity have been investigated. The gamma-ray mass attenuation coefficients and radioactivity concentrations of the rocks considered have been determined by using coaxial high purity germanium detector (HPGe) detector system. The capability of andesite by means of gamma-ray shielding for low energy gamma rays has been found highest when it is compared with those of basalt, dolomite, limestone and syenite rocks. In the places where the rocks were extracted, the activities of the natural radioactive elements (<sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K) and artificial radioactive elements (<sup>137</sup>Cs) have been determined by analyzing the natural radiation concentration on site and using gamma spectroscopic analysis to reveal the presence of radioactivity. The <sup>137</sup>Cs concentration has been found to be below the detection limit. The radioactivity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K have been found to be in acceptable levels. Furthermore, chemical element analyzes have been carried out to determine the content of these rocks precisely. The gamma-ray mass attenuation coefficients of andesite for low energy gamma rays have been found to be highest among those of other rocks considered in this work.

**Keywords:** Rock; Gamma-ray mass attenuation coefficient; Radionuclide analysis; Chemical analysis.

### 1. Introduction

Natural stones are one of the oldest building materials known and used by people. Architecture and art also use natural stones. In the world we live in, natural stones that provide aesthetics, durability and modernity to living spaces and structures have become a part of life. In line with the increasing interest and demand for these stones, natural stone production has increased at the same rate. Stones are divided into three main groups as magmatic, sedimentary and metamorphic stones. Basalt, andesite, syenite are magmatic rocks. Dolomite and limestone are among sedimentary rocks (Karahan 2018).

There are natural radioactive substances in the air, water and soil of the world we live in. Mankind

and other living beings have been living together for millions of years with cosmic rays coming from the universe and radiation emitted from natural radioactive materials on the earth. Natural radionuclides with very long half-lives such as <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the soil cause the soil to be radioactive. The largest contribution (about 99%) to the radioactivity from the <sup>238</sup>U decay chain comes from <sup>214</sup>Pb and <sup>214</sup>Bi, while it comes from <sup>228</sup>Ac and <sup>208</sup>Tl from the <sup>232</sup>Th decay chain. Natural radionuclides crumble into the soil and cause the natural radioactivity to increase. The natural radioactivity of a region varies according to the geological structure of the region. When looking at the structure of the earth, rock beds are found just

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below the soil layer of a certain thickness. Granite rocks spread over wide areas in some regions contain significant amount of thorium. Studies have shown that the measured gamma radiation levels are high in areas where such rocks are found (Canbazoğlu 2004; UNSCEAR 2000).

Radioactive substances, which are found in air, water, and all plant and animal foods, are taken into our body by respiratory and digestive ways, and they accumulate in various organs over time (UNSCEAR, 2008). Since we are affected by cosmic rays, natural and artificial radioactive materials in the earth, the human body is naturally exposed to both internal and external radiation (Othman et al., 2023). Therefore, it is important to determine radioactivity levels in air, water and soil.

Although radiation has negative effects on living things, nuclear techniques and radioisotopes provide great benefits to humanity in cancer treatment and in solving some biological problems in medicine and in some industrial applications (Cutler, 2020). Since we cannot remove the radiation risk from our lives, we need to take protective measures to minimize the radiation hazard. There are three basic ways to protect from radiation. These are distance, time and shielding. In these ways, shielding is the most important method of radiation protection. Shielding is based on placing material between the system to be protected and the radiation source (Akkurt et al., 2011). For this purpose, studies have been carried out on many materials in the world and are still in progress. Some of these are studies conducted to determine the attenuation coefficients of various rocks.

Mass attenuation coefficients of rocks collected from different parts of India (olivine basalt, green marble, jet black granite, teflon black granite, cuddapah limestone, white earth and pink marble) were investigated experimentally and theoretically. Mass attenuation coefficients of the samples were experimentally determined by using  $\gamma$ -rays with energy of 122 keV, 356 keV, 511 keV, 662 keV, 840 keV, 1275 keV, 1330 keV and theoretically calculated for both XCOM and MCNP5 codes. It shows that pink marble exhibits better protection than other rocks (Obaid et al., 2018). The attenuation coefficients for gamma-ray of feldspathic basalt, compact basalt, volcanic rock, dolerite, pink granite sandstone and concrete at 122, 356, 511, 662, 1170, 1275, 1330 keV energies has

been investigated experimentally and theoretically (WinXCom). The results obtained showed that feldspathic basalt, compact basalt, volcanic rock, dolerite and pink granite are more efficient than sandstone and concrete for gamma-ray protection applications (Obaid et al., 2018). Zaim and Hatipoğlu (2018) investigated the total linear attenuation and mass attenuation coefficients, half-value and one-tenth thicknesses for the Kırklareli province marble sample using the gamma energies of 661.7 keV, 1173.2 keV and 1332.5 keV. Measurements were made both experimentally (by using gamma scintillation detector NaI (TI)) and theoretically (XCOM) and compared with each other. The results obtained by measuring the marble discs were also compared with the results obtained from measuring the lime powder tablets. It is concluded that mass attenuation coefficient of limestone at these energies is higher than Kırkkale marble (Zaim and Hatipoğlu 2018). In another study, linear attenuation coefficients of 3 different Amasya Marbles were determined for  $\gamma$ -rays with energy of 511, 835 and 1274 keV. Measurements were made by using a gamma scintillation detector NaI (TI) and theoretical calculations were done with XCOM. It was determined that it has a better attenuation coefficient than basalt and andesite for these energies (Mavi et al., 2015). In another study, GATE software was used as the Monte Carlo code to calculate the gamma-ray attenuation coefficient of some granite samples at 662 keV, 1173.2 keV and 1332.5 keV photon energies. The simulated results of mass attenuation coefficients are compared with the experimental and theoretical data given in the previous study. A satisfactory agreement was observed between the GATE code and the XCOM (XCOM program which was developed by Berger and Hubbell for calculating mass attenuation coefficients or photon interaction cross-section for any element, compound or mixture at energies 1 keV to 100 GeV) results (Ozyurt et al., 2018). In addition to examining the radiation effect of natural rocks, there are studies on the determination of the attenuation coefficients of different elements, compounds, mixtures and concretes (Tufekci and Gokce, 2018; Sayyed et al., 2018; Unal et al., 2016; Akkurt et al., 2012; Phutthanet et al., 2018; Baltas, 2020; Erenler et al., 2020).

In the present study, the gamma-ray attenuation properties of spilitic basalt, andesite, dolomite, limestone and syenite rocks obtained from Trabzon

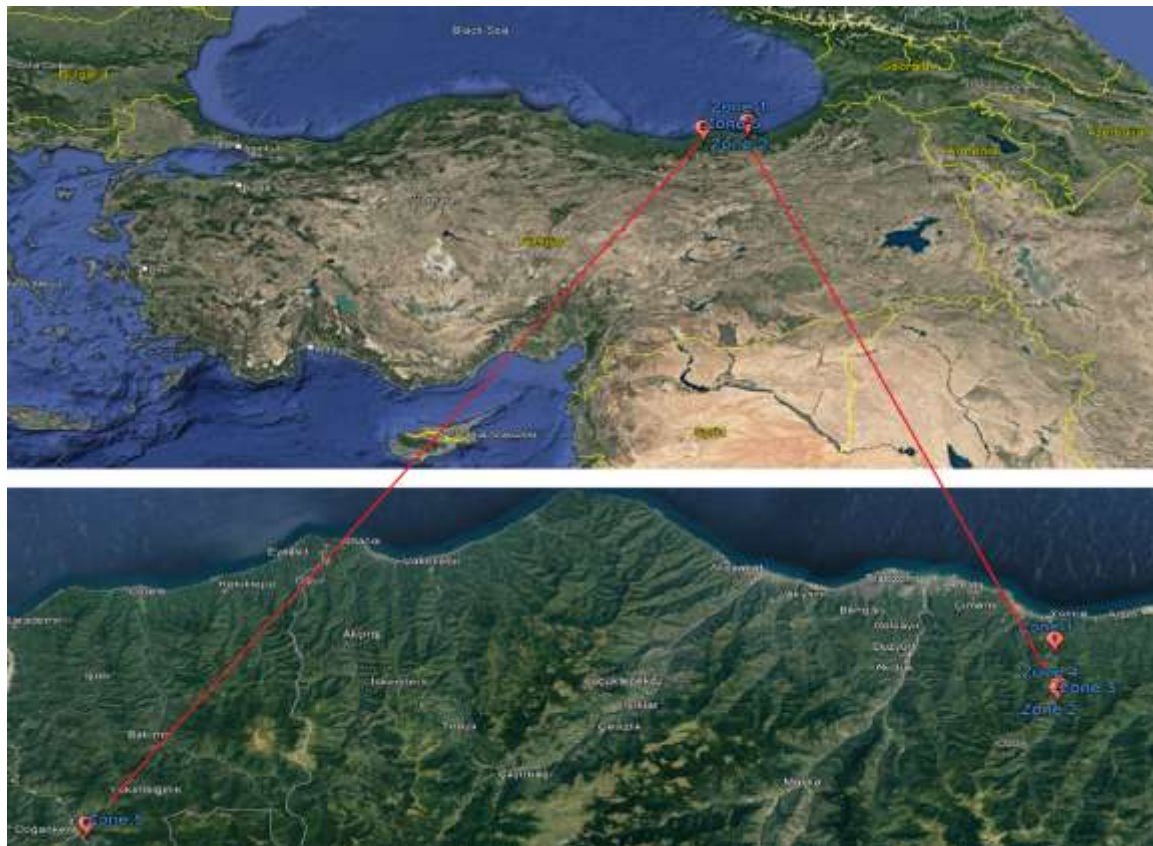
Province and its vicinity and the effect of these rocks on radiation shielding have been investigated. In addition to radionuclide measurements to determine the presence of radioactivity, chemical element analyzes have been carried out to determine the content of these rocks precisely, in order to examine the natural radiation concentration

## 2. Experimental

Spilitic basalt, andesite, dolomite, limestone and syenite rock samples were taken within the scope of field studies for Trabzon province Yomra district and Giresun province Tirebolu district, which are our study areas. The study area and sampling points are shown on the map given in Figure 1. While collecting rock samples, attention was paid to the fact that a rock sample taken from a region could

on-site and reveal the presence of radioactivity. Thus, the radiological and chemical structure of spilitic basalt, andesite, dolomite, limestone and syenite rocks obtained from Trabzon Province and its close vicinity have been determined. Such a study has not been conducted before for these rock types in the considered region.

best represent the geological structure of that region and not be brought to the region later. The samples taken from the surfaces of the rocks with the help of a hammer were placed in clean, closable nylon bags and brought to the laboratory.



**Fig. 1. Localization of sampling area.**

Basalt, limestone, syenite, andesite and dolomite samples collected were crushed and pounded with the help of crushers in Mining Engineering Department of Karadeniz Technical University and passed through a 80 mesh sieve for homogeneity. For chemical analysis, attenuation test and radionuclide analysis, the samples must be ground and powdered. In order to form absorption discs,

parts with a mass of approximately 1.5 g were taken from each sample and discs with a diameter of 12 mm were formed under 5 bar pressure with a hydraulic press machine. It was observed that the discs formed were strong and did not break down due to the structure of the samples. For radionuclide analysis, rock samples, which were crushed and pounded, were placed in plastic boxes with a

diameter of 6 cm and a height of 5 cm, prepared in accordance with the experimental geometry, and the boxes were kept tightly closed for 1 month. Thus, the radioactive balance between  $^{238}\text{U}$  and  $^{232}\text{Th}$  products was achieved and the samples were made ready for counting.

AMATEK-ORTEC-GEM25P4-76 model high purity coaxial Ge detector (HPGe) in KTU Department of Physics was used for determining both the gamma-ray mass attenuation coefficients and the radionuclide concentrations of the samples. The detector system which is used in the analysis of radioactive materials emitting gamma-rays of different intensity and energy, consists of a radiation detector, liquid nitrogen-based cooling mechanism, electronic system and amplifiers that detect the generated signals. This detector has a resolution of 1.7 keV at 1.33 MeV and a relative efficiency of 33%.

Gamma-ray counts were performed using the GamaVision program.  $^{238}\text{U}$  degradation product  $^{214}\text{Pb}$  (295.2 keV),  $^{214}\text{Pb}$  (352 keV),  $^{214}\text{Bi}$  (609.4 keV);  $^{232}\text{Th}$  degradation product  $^{212}\text{Pb}$  (238.6 keV),  $^{208}\text{Tl}$  (583.1 keV),  $^{228}\text{Ac}$  (911.1 keV),  $^{137}\text{Cs}$  (661.6 keV),  $^{40}\text{K}$  (1460 keV) peaks were determined. While determining the areas of the peaks, the smallest error and the largest area were marked, and then it was determined which radionuclide belongs to the energy values seen in the peaks. In the gamma spectrum of radionuclide measurement,  $^{238}\text{U}$  degradation product  $^{214}\text{Pb}$  (295.2 keV),  $^{214}\text{Pb}$  (352 keV),  $^{214}\text{Bi}$  (609.4 keV); the degradation product of  $^{232}\text{Th}$ ,  $^{212}\text{Pb}$  (238.6 keV),  $^{208}\text{Tl}$  (583.1),  $^{228}\text{Ac}$  (911.1 keV) and  $^{40}\text{K}$  (1460 keV) peaks were determined and activity values were calculated by using the equation (Duran et. al., 2022):

$$A = N/(m\varepsilon Pt) \quad (1)$$

where  $A$  is the activity,  $N$  is the net count area which is obtained from substituting of background area from total area,  $m$  is the net mass of the

sample,  $\varepsilon$  is full peak efficiency. Full energy peak efficiency was determined using Standard Reference Material (IAEA-375) prepared by International Atomic Energy Agency. Decay corrections were performed to the sampling date.  $P$  is transition probability of gamma decay,  $t$  is the counting time. After the determination of the activities, the absorbed doses ( $D$ ) were calculated by using the equation (Duran et. al., 2022):

$$D \left( \frac{\text{nGy}}{\text{s}} \right) = (0.462x^{238}\text{U}) + (0.604x^{232}\text{Th}) + (0.0417x^{40}\text{K}) \quad (2)$$

and the annual effective dose equivalents ( $AEDE$ ) were calculated by using the following equation (Duran et. al., 2022):

$$AEDE \left( \frac{\mu\text{Sv}}{\text{y}} \right) \text{ Absorbed Dose Rate} \times 0.7 \text{ SvGy}^{-1} \times \text{Occupancy Factor} \times \text{Time}. \quad (3)$$

In this equation, the environmental gamma dose conversion factor does not change in measurements made both indoors and outside, and is taken as 0.7 Sv/Gy. The occupancy factor is the time people are exposed to these rays. In this study, occupation factor was taken as 0.8 for indoor and 0.2 for outside, considering that people spend 20% of their time outdoors and 80% indoors. Time is the number of hours in a year (8760 s/y). The detector used for radioactivity measurements has been also used for gamma-ray attenuation measurement of samples. For the gamma-ray attenuation measurements, collimator, radioactive source and absorber (sample) also are required. The used system in the present study is shown in Figure 2.



Fig. 2. The system used for gamma ray attenuation measurements.

The main reason for using the collimator is to obtain a capillary gamma-ray beam from the radioactive source showing isotropic gamma-ray emission and direct it to the detector. Thus, by placing the sample to be examined on this beam

line, the gamma-ray is weakened and the attenuation capability of the sample can be determined from there. The geometry of the collimator and detector system used in our experiments is shown in Figure 3.

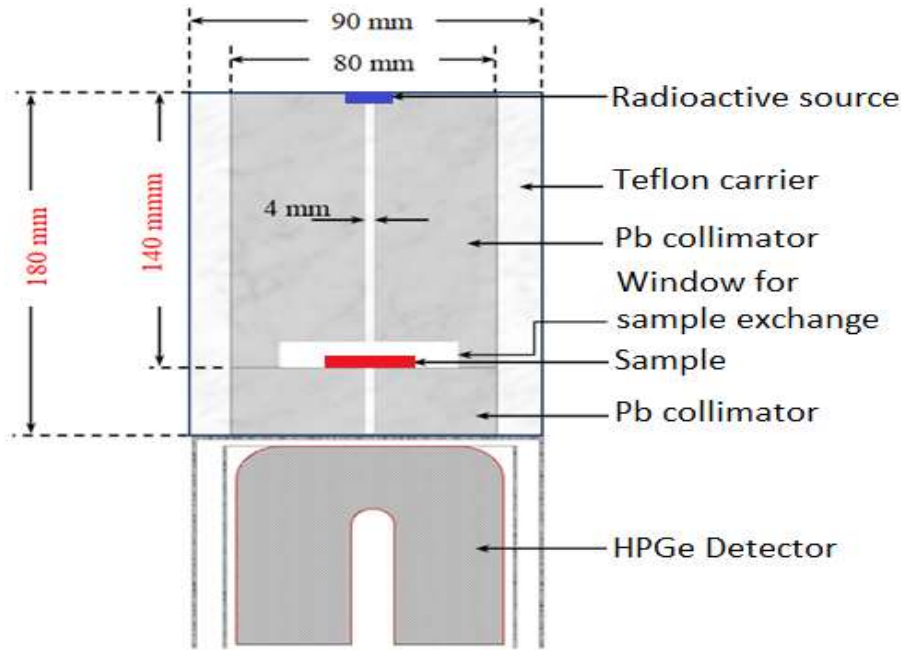


Fig. 3. Schematic representation of the collimator and detector system.

The radioactive source used for measurements is Eu-152. There are 14 gamma-rays emitted from the Eu-152 radioactive source and their energies are listed in Table 1.

TABLE 1. Gamma-ray energies emitted from the Eu-152 radioactive source.

Gamma	Energy (keV)
$\gamma_1$	121.7817±0.0003
$\gamma_2$	244.6974±0.0008
$\gamma_3$	344.2785±0.0012
$\gamma_4$	411.1165±0.0012
$\gamma_5$	443.965±0.003
$\gamma_6$	778.9045±0.0024
$\gamma_7$	867.380±0.003
$\gamma_8$	964.072±0.018
$\gamma_9$	1085.837±0.010
$\gamma_{10}$	1089.737±0.005
$\gamma_{11}$	1112.076±0.003
$\gamma_{12}$	1212.948±0.011
$\gamma_{13}$	1299.142±0.008
$\gamma_{14}$	1408.013±0.003

For the gamma-ray attenuation measurements, each count has been carried out for 3600 seconds. The gamma-ray spectrum obtained for the Eu-152 radioactive source in the attenuation setup (without sample) is shown in Figure 4. The 7 gamma energy peaks are shown in this figure. The other 7 peaks in the 121.76-1408.01 keV energy range are not considered because they are weak in the spectrum.

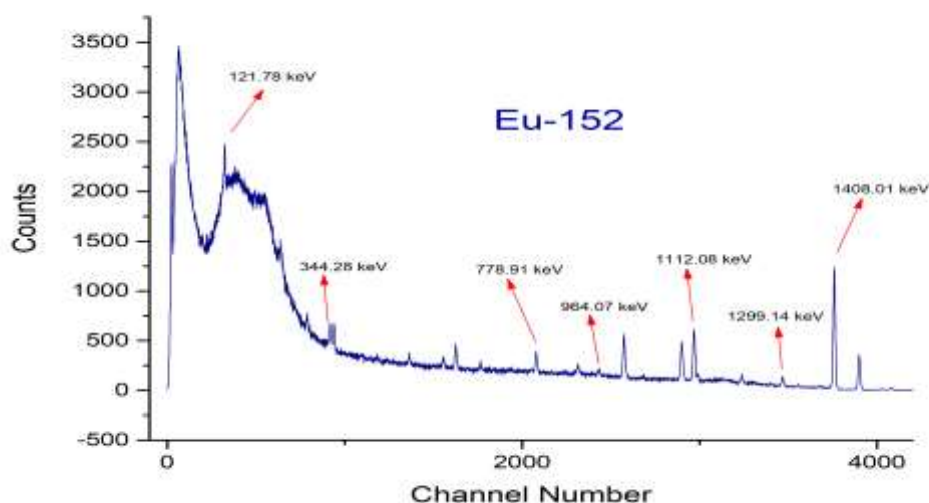


Fig. 4. Eu-152 gamma-ray spectrum obtained from the measuring setup.

If a material of thickness  $x$  is placed in the path of a beam of gamma radiations, the intensity of the beam will be attenuated according to Beer-Lambert's law:

$$I = I_0 e^{-\mu t} \quad (4)$$

where  $I_0$  and  $I$  are the unattenuated and attenuated photon intensities, respectively, and  $\mu$  (in units of  $\text{cm}^{-1}$ ) is the linear attenuation coefficient of the material. A coefficient more accurately characterizing a given material is density-independent mass attenuation coefficients  $\mu/\rho$  (in units of  $\text{cm}^2/\text{g}$ ):

$$I = I_0 e^{-\left(\frac{\mu}{\rho}\right)\rho x} = I_0 e^{-\left(\frac{\mu}{\rho}\right)d} \quad (5)$$

where  $d$  is mass per unit area (in units of  $\text{g}/\text{cm}^2$ ) and  $\mu/\rho$  is mass attenuation coefficient in units of  $\text{cm}^2/\text{g}$  (Erenler et al., 2020).

Chemical analysis of samples has been carried out in order to determine the chemical content of the test samples and the amount of elements

The measurement results obtained for gamma-ray attenuation have been analyzed by using GammaVision program. The area of each gamma energy peak was calculated and it was determined how many gamma ray counts were performed at the relevant energy.

contained in it. In this study, X-ray fluorescence spectrometer (XRF), one of the chemical analysis methods, has been used. The chemical contents of the test samples obtained after the grinding process have been determined with the help of the XRF tester.

The samples have been prepared as pellets without using any additives after drying at  $105^\circ\text{C}$  for 1 day. In accordance with the sample amount, the sample has been placed in a 30 mm sample holder and semi-quantitatively studied in the oxide form in the Boron-Uranium range. The results have been prepared as both oxide and metal.

#### Statistical analysis:

In order to determine the uncertainties in the gamma-ray mass attenuation coefficient at the relevant energy, first of all, the area under the relevant energy peaks in the gamma spectrum was calculated by making a Gaussian fit using the GamaVision (Version 6.07) program. Then, taking these uncertainties into account, the uncertainties in the mass attenuation coefficient according to Equation 5 were calculated with EXCEL computer software. In addition, Gnuplot software was used for the graphics presented in this study.

### 3. Results and Discussion

The mass attenuation coefficients are determined by performing transmission experiment. Mass attenuation coefficients obtained for basalt, limestone, syenite, andesite and dolomite by analyzing of the peaks with 121.78, 344.28, 778.91, 964.07, 1112.08, 1299.12 and 1408.01 keV energy in the gamma spectrum of each sample are listed in Table 2.

Theoretical values for the mass attenuation coefficients are calculated by XCOM program

which was developed by Berger and Hubbell for calculating mass attenuation coefficients or photon interaction cross-section for any element, compound or mixture at energies 1 keV to 100 GeV. The determined gamma-ray mass attenuation coefficients of samples are shown in Figure 5 as a function of gamma-ray energy together with theoretical results obtained from NIST-XCOM database (Photon Cross Section Database) (Hubbell and Seltzer, 2004).

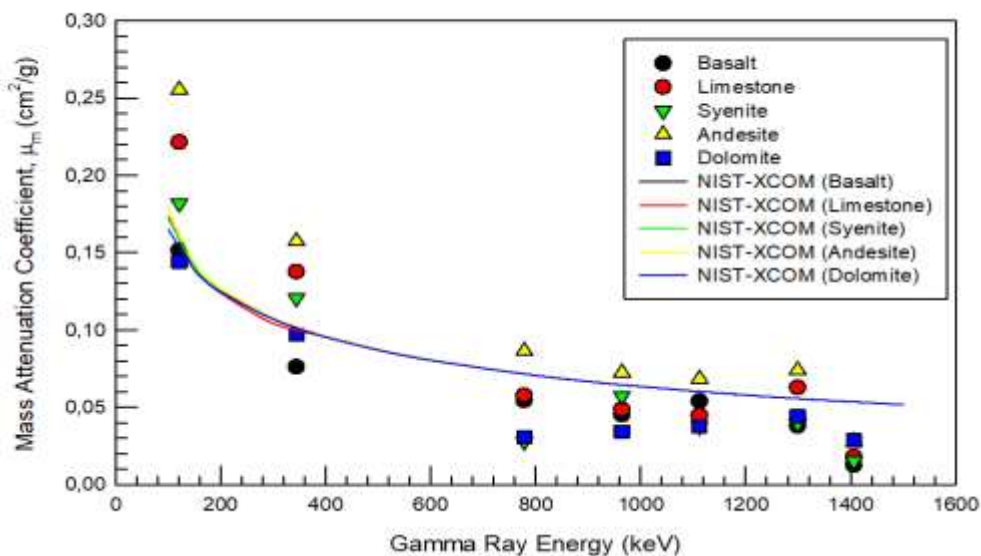
**TABLE 2. Mass attenuation coefficients of the samples in units of cm<sup>2</sup>/g for gamma-rays at specific energies.**

Energy (keV)	Basalt (cm <sup>2</sup> /g)	Limestone (cm <sup>2</sup> /g)	Syenite (cm <sup>2</sup> /g)	Andesite (cm <sup>2</sup> /g)	Dolomite (cm <sup>2</sup> /g)
121.78	0.1516±0.0032	0.2215±0.0031	0.1819±0.0035	0.2554±0.0031	0.1442±0.0033
344.28	0.0761±0.0062	0.1377±0.0068	0.1206±0.0063	0.1574±0.0026	0.0971±0.0064
778.91	0.0544±0.0069	0.0576±0.0069	0.0278±0.0075	0.0863±0.0068	0.0307±0.0074
964.07	0.0451±0.0027	0.0487±0.0027	0.0573±0.0029	0.0723±0.0028	0.0343±0.0029
1112.08	0.0538±0.0021	0.0447±0.0021	0.0376±0.0023	0.0684±0.0022	0.0375±0.0022
1299.12	0.0380±0.0119	0.0628±0.0124	0.0395±0.0137	0.0740±0.0132	0.0440±0.0139
1408.01	0.0126±0.0009	0.0179±0.0009	0.0153±0.0010	0.0279±0.0009	0.0286±0.0009

The quoted errors are the standard deviations.

It is clearly seen from Table 2 and Figure 5 that mass attenuation coefficient ( $\mu_m$ ) decreases in increasing gamma energies for basalt, limestone, syenite, andesite and dolomite. There is not much change in  $\mu_m$  values in the gamma-ray energy range of 900-1300 keV. For low gamma-ray energies such as 121.78 keV and 344.28 keV andesite and limestone have mass attenuation coefficients 0.255

cm<sup>2</sup>/g and 0.222 cm<sup>2</sup>/g, respectively and these are relatively higher mass attenuation coefficients when they are compared with those of basalt, syenite and dolomite. In particular usage of andesite as to be radiation protective materials for low energy gamma-rays is seem to most favorable among the other stones considered in this work.



**Fig. 5. Determined mass attenuation coefficients of stone samples.**

Mass attenuation coefficients of Feldspathic basalt-Compact basalt, Olivine Basalt (Obaid et al., 2018) for 122 Kev energy were determined as 0.182 cm<sup>2</sup>/g, 0.181 cm<sup>2</sup>/g, 0.1726 cm<sup>2</sup>/g, respectively. In this study, mass attenuation coefficients at the same energy for Basalt, Limestone, Syenite, Andesite, Dolomite rocks were experimentally determined as 0.152 cm<sup>2</sup>/g, 0.222 cm<sup>2</sup>/g, 0.182 cm<sup>2</sup>/g, 0.255 cm<sup>2</sup>/g, 0.144 cm<sup>2</sup>/g, respectively. It is seen that mass attenuation coefficients for 122 keV energy of the basalt samples extracted in the Trabzon region are lower than the Feldspathic basalt-Compact basalt-Olivine Basalt (Obaid et al., 2018) samples. In this study, it is seen that mass attenuation coefficients of limestone and andesite, whose mass

attenuation coefficients were determined, are higher than other rocks.

As a result of gamma spectroscopic analysis of rock samples, the activities of <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K natural radioactive elements and <sup>137</sup>Cs artificial radioactive element have been determined by using Eq. (2). Annual effective dose values have been calculated by using the absorbed dose value by using Eq. (2) and Eq. (3) for each sample. These values are listed in Table 3. <sup>137</sup>Cs activity is not listed in the table because it is below the detection limit for all samples considered in this work.

**TABLE 3. The results of radionuclide analysis for rock samples. <sup>238</sup>U-<sup>232</sup>Th-<sup>40</sup>K activity values of rock samples, absorbed dose (D) and annual effective dose equivalents (AEDE).**

Rock	<sup>238</sup> U (Bq/kg)	<sup>232</sup> Th (Bq/kg)	<sup>40</sup> K (Bq/kg)	D (nGy/s)	AEDE (Sv/y)	
Basalt	16	6.31	650	38.31	47	This Study
	12	15	230			*
Limestone	10	12.3	160	18.72	23	This Study
	25	8	270			*
Syenite	25.6	17	42	23.85	29	This Study
Andesite	40,5	19	205	38.73	47.5	This Study
Dolomite	27	21	230	34.74	42.6	This Study
Granite	35	70	1200			*
Soil	35	30	400			*

\* UNSCEAR, 2000: United Nations Scientific Committee on the Effect of Atomic Radiation to the General Assembly, Sources, Effects and Risk of Ionizing Radiation, United Nations, New York, USA; 2000.

X-ray fluorescence spectrometry (XRF), one of the chemical analysis methods, has been used to determine the amount of elements contained in the rock samples. As a result of the measurements, the elemental analysis results of the rock samples considered in this work are given in Table 4. As seen in Table 4, Syenite is predominantly silicium oxide, limestone calcium oxide, andesite silicon oxide, basalt silicon oxide, dolomite calcium oxide.

#### 4. Conclusions

In this study, the gamma ray attenuation properties of spilitic basalt, andesite, dolomite, limestone and syenite rocks obtained from Trabzon Province and its vicinity and the effect of these rocks on radiation shielding have been investigated. The gamma-ray

mass attenuation coefficients of andesite for low energy gamma rays have been found to be highest among those of other rocks considered in this work. It reveals that usage of andesite rocks for radiation shielding purposes can be suggested when we compared with other rocks basalt, dolomite, limestone and syenite. In addition, in the places where the rocks were extracted, radionuclide measurements were made in order to examine the natural radiation concentration on site and to reveal the presence or absence of radioactivity. It is observed that the scale with the highest <sup>238</sup>U value among them is andesite.



**TABLE 4. Chemical analysis results of Syenite, Limestone, Andesite, Basalt and Dolomite samples.**

Element weight%	Syenite	Limestone	Andesite	Basalt	Dolomite
O	53.1	49.9	50.5	52.5	49.7
Si	16.8	4.57	31.8	18.5	1.11
Fe	8.10	1.28	1.15	7.37	0.289
Al	6.92	0.996	8.37	6.61	0.466
Mg	6.46	0.152	0.0379	5.55	1.51
Ca	5.31	33.1	0.104	6.07	36.3
C	1.51	8.99	0.957	1.62	10.3
Ti	1.03	0.0516	0.0603	0.892	0.0187
Na	0.643	0.188	5.85	0.350	0.0282
P	0.0938	0.0305	-	0.104	-
Sr	0.0233	0.0403	-	0.0269	0.0183
S	0.0220	0.0106	-	0.0443	0.0447
Cl	0.0155	-	-	-	0.0100
K	0.0151	0.324	-	-	-
Cr	0.0118	-	-	0.0136	-
Mn	-	0.334	0.0304	0.165	0.0112
K	-	-	1.13	0.175	0.138
Zr	-	-	0.0155	-	-
Zn	-	-	0.0074	-	-
Oxide weight%	Syenite	Limestone	Andesite	Basalt	Dolomite
SiO <sub>2</sub>	41.0	10.3	69.0	44.2	2.48
Al <sub>2</sub> O <sub>3</sub>	14.7	1.98	16.0	13.7	0.919
Fe <sub>2</sub> O <sub>3</sub>	14.3	2.05	1.68	12.6	0.457
MgO	11.8	0.265	0.0635	9.97	2.60
CaO	8.80	50.3	0.148	9.82	54.6
CO <sub>2</sub>	6.13	33.7	3.55	6.46	38.5
TiO <sub>2</sub>	2.08	0.0962	0.103	1.75	0.0344
Na <sub>2</sub> O	0.943	0.266	7.95	0.509	0.0396
P <sub>2</sub> O <sub>5</sub>	0.252	0.0741	-	0.273	-
SrO	0.0352	0.0536	-	0.0392	0.0239
K <sub>2</sub> O	0.0214	0.417	1.39	0.242	0.176
Cr <sub>2</sub> O <sub>3</sub>	0.0210	-	-	0.0235	-
ZrO <sub>2</sub>	-	-	0.0214	-	-
ZnO	0.0122	-	0.0095	-	-
MnO	-	0.482	0.0401	0.254	0.0160
SO <sub>3</sub>	-	0.0279	-	0.127	0.117
Cl	0.0182	-	-	-	0.0106

(-) Below the detection limit.

#### Conflicts of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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