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Radiological risk assessment of radionuclide concentrations and elemental analysis resulting from natural and synthetic zeolite samples in Saudi Arabia

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ABSTRACT

Since zeolite have been widely used for several industries, including paint, paper, fertilizer, building, textiles, and agriculture, it is important to measure the radionuclides concentrations to determine the health effect and to protect environment. To determine the activity concentrations and distribution of naturally occurring radionuclides, twenty-eight zeolite rock and soil samples were collected from Jabal Shamah in Jeddah, in the Makkah region of Saudi Arabia. Furthermore, two 500G artificial zeolite samples that Sigma Life Science brought from Germany were analyzed. The measurements were performed using gamma-ray spectrometry of NaI (Tl) detector. In zeolite rock samples, the mean activity concentrations of ^{226}Ra, ^{232}Th, and^{40}K were, respectively, 35.7± 1.8 Bq/kg, 29.3± 1.5 Bq/kg, and 282.9± 4.7 Bq/kg respectively while ^{226}Ra, ^{232}Th, and ^{40}K concentrations in zeolite soil samples were 27.9 ± 1.4 Bq/kg, 23.1 ± 1.2 Bq/kg, and 293.0 ± 14.7 Bq/kg, respectively. But for ^{226}Ra, ^{232}Th, and ^{40}K artificial zeolite samples, the activity concentrations were 82.2 ± 4.17 Bq/kg, 69.6 ± 3.52 Bq/kg, and 739.3 ± 37.0 Bq/kg, respectively.

The levels of radiation hazard metrics, including internal and external hazard, radium equivalent activity annual dose, and other recommended limits by the International Commission on Radiological Protection (ICRP), were also calculated and compared with others. Furthermore, the concentrations of the heavy elements and their oxides were measured using x-ray fluorescence (XRF); the findings were compared with literature descriptions of ranges from other regions of the world.

Keywords: Natural radioactivity levels, Radium equivalent, Activity annual dose, External and internal hazard, X-ray fluorescence (XRF), environment

1. INTRODUCTION

Since the creation of the Earth, naturally occurring radioactive elements have existed. There are still some radioelements in the Earth's crust that have extraordinarily long half-life and have been decomposing for many Years. These radio elements can be found in all-natural materials, such as soil, drinking water, eating, breathing, building stones, plants, rivers, the ocean, etc. Radioactive elements are found naturally in our muscles, bones, and tissue. We also get radiation from outer space, often known as cosmic radiation or cosmic rays, in addition to the radiation from these sources [1], [2]. Natural radioactivity produces 85% of the world's yearly average ionizing radiation in the form of gamma radiation [3]. Human health is at risk from increased exposure to these radio nuclides, which can lead to several illnesses, including malignancies of the kidney, liver, pancreas, lungs, and bones [4], [5]. The concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the soil differ because they are dependent on the kind of rock and the soil. Natural sources account for about 96% of the total radiation dose that humans receive, with man-made sources accounting for 4% [6], [7], [8]. External exposure to gamma radiation from terrestrial radionuclides (^{238}U, ^{232}Th, and ^{40}K) and internal exposure to radioactive gases (radon and its daughters) are the two primary ways that people are exposed to radioactive materials in the environment [9].

Zeolites are used in many different fields, such as gas detection, biotechnology, environmental remediation, catalysis, and medicine. Zeolites are found naturally and can also be created in a lab. The two main categories of zeolites are natural and manufactured [10]. There are also zeolites, which include about 65 different species of natural zeolites and roughly 100 different species of artificial zeolites. Naturally occurring zeolites typically contain the sodium ion, which is widely distributed in the environment, as a loosely bound counter ion. Natural zeolites have a wide range of industrial and environmental applications [11]. Natural zeolites are used as raw materials in the construction and paper industries. Agriculture employs them in addition to other applications [10].

When zeolite synthesis and synthetic materials became available, interest in these later operations grew because they offered several advantages, including:

(a) greater versatility because of the great variability of their structures,

(b) more open frameworks, appropriate for adsorption and catalysis,

and (c) quality, i.e., consistency in constitution.

While natural zeolites are the precursors of contemporary synthetic materials, it would be inaccurate to consider them only from a historical standpoint. In significant fields that are worth looking into and have bright futures, natural zeolites are still being researched and applied [12].

Additionally, commercially produced zeolites are made for specific applications. Medical monitoring, gas collection, heavy metal removal, industrial process control, and environmental mitigation are just a few of the many uses for them as chemical sensors. Synthetic zeolites have attracted the attention of scientists and researchers due to their significant versatility and adaptability. There are further

benefits to using synthetic zeolites over natural ones. For instance, controlling certain processes, such as the control of N_2O emissions, is best accomplished with artificial zeolite.

Moreover, synthetic zeolite has a better adsorption capacity for the removal of various hazardous metal ions, including Cr^{3+}, Ni^{2+}, Zn^{2+}, Cu^{2+}, and Cd^{2+}, and improved radioactive waste removal capabilities when compared to natural zeolite. Furthermore, naturally occurring zeolites have a smaller pore volume than synthetic zeolites [10]. In recent years, there has been a growing interest in the effects of natural radioactivity.

The effective average annual dosage of radiation exposure from natural sources was determined to be 2.4 msv y^{-1} per person. The intrinsic radioactivity in rock and soil must be measured in order to calculate the amount that any radioactive discharge alters the natural background activity over time. Any radioactive emissions into the environment must be closely monitored for the protection of the ecosystem. The source of external exposure is terrestrial radio nuclides.

By all spectrometric measurements, the external radiation field's three sources the ^{40}K, the gamma-emitting radionuclides in the ^{226}R which comes from ^{238}U series and ^{232}Th series, and the gamma-emitting radionuclides approximately equalize the quantity of gamma radiation incident on humans in normal indoor and outdoor environments [13], [14].

The present project aims to:

1- measured the concentrations of different types of elements and their oxide using XRF from zeolite samples like Aluminum, iron, silicon.... etc.

2- measured natural radioactivity level of (^{226}Ra, ^{232}Th and ^{40}K) in samples of zeolite used in Saudi Arabia by using γ-ray spectrometry.

3- Calculate the radiological parameters (radium equivalent activity Raeq, level index Iγr, external hazard index Hex and absorbed dose rate) which is related to the external γ-dose rate.

4- Compare the results of concentration levels and radiation hazards with similar studies carried out in other countries.

2. MATERIALS AND METHODS

2.1 Location of Study Area

This study was carried out at Jabal Shamah in is situated in the western region of Saudi Arabia in Jeddah in the Makkah region and Jabal Shamah is located on coordinate (20°46'52"N 39°40'18"E), which is 100 km south of south-eastern Jeddah, is a rich source of natural zeolites are shown in Fig.1 , it is extended from Northwest Saudi Arabia to Jordon and Syria. It is a section of the Arabian Plate's lava fields [15].

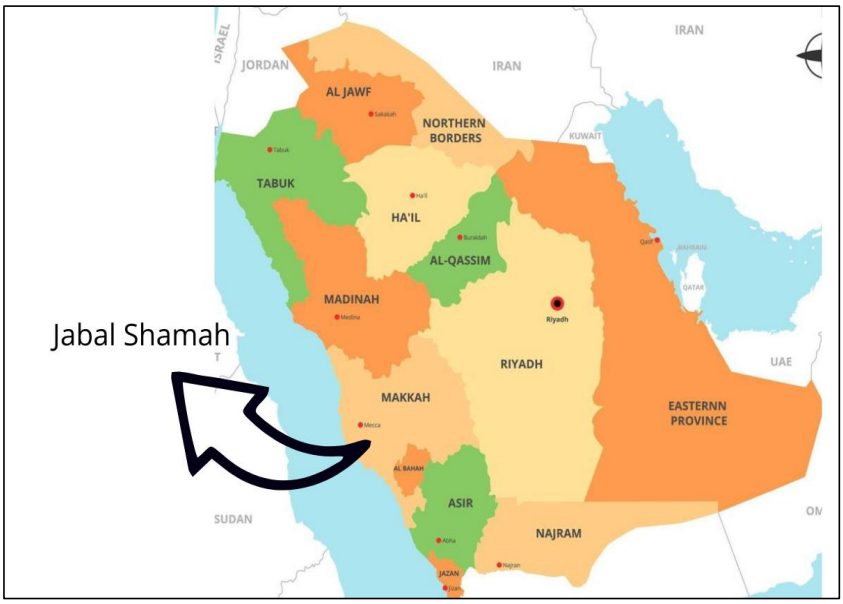

Figure1. Location of Jabal Shamah in Saudi Arabia.

2.2 Samples Collection and Preparation

In order to measure the natural radioactivity in zeolite rock, soil and artificial sample (30 samples) of Jabal Shamah in Saudi Arabia in Jeddah province as given Table 1. The samples were ground and sifted using sieves. The samples were put in polyethylene bottles of 200 mL volume. The polyethylene bottles were completely closed for more than one month to enable radioactive equilibrium to ensure that radon was removed [16][17]. Gamma-spectrometric measurements were performed with NaI (Tl) detector. A gamma-ray energy spectrum is created when these emissions are identified using energy-calibrated gamma ray sources such as Co-60 and Cs-137. The measuring time for gamma-ray spectra ranged was (6h). In order to determine the background distribution due to naturally occurring radionuclides in the environment around the detector, an empty polystyrene container was counted in the same manner as the samples. After measurement and subtraction of the background, the activity concentration was calculated. The values of e and η for each isotope used to calculate the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K are shown in Table 2. The activity concentrations of the natural and artificial radionuclides in the measured samples were computed using the following relation. To ensure that in order to compute radionuclide activity, the secular equilibrium between the parents and daughters in the samples determines the concentration for each gamma-ray photo-peak. [18] [19][20].

$$A = \frac{N_P \times 100}{e \times \eta \times m} \quad (1)$$

Where: Np: is the net count rate (cps), e : is the abundance of the γ-peak in a radionuclide
m: is the mass of sample(kg), η: is the measured efficiency for each gamma-ray peak observed for the same number of channels.

Table 1: Description of the area under study and samples distribution

Samples Location	**Sample zeolite**	**Samples Number**
Jabal Shamah in Jeddah in the Makkah region	Rock	12
	Soil	16
	artificial	2

Table 2: Gamma rays and their related isotopes used to calculate the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K.

Nuclide	Isotopes	Energy (KeV)	Measured efficiency for each gamma-ray peak (η)(%)	Abundance of γ-peak in a radionuclide (e)
^{226}Ra	^{214}Pb	351.90	37	0.21
	^{214}Bi	609.30	46	0.12
	^{214}Bi	1120.30	15	0.08
	^{214}Bi	1764.5	15.9	0.067
^{232}Th	^{228}Ac	911.10	29	0.09
	^{212}Pb	238.60	44	0.25
^{40}K	^{40}K	1460	11	0.07

3. RESULT AND DISCUSSION

3.1. Activity Concentrations of ^{226}Ra, ^{232}Th and ^{40}K

The radioactivity concentrations of the radioactive isotopes ^{226}Ra, ^{232}Th, and ^{40}K for zeolite samples, and their concentrations were determined using gamma ray spectroscopy of NaI (Tl) detector. The concentrations of radioactive isotopes in rock and soil with comparison with others are shown in Table3. It was found that the average concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the present work were lower than in other studies [21].While in artificial the average concentrations of radioactive isotopes of ^{226}Ra, ^{232}Th and ^{40}K were 82.2±4.17 Bq/kg, 69.9 ± 3.52 Bq/kg and 739.3 ± 37.0 Bq/kg respectively. The uncertainty in the average value is calculated using the standard deviation method.
It appears in all measured samples that the activity of ^{40}K is higher than that of ^{226}Ra and ^{232}Th [22].The average concentrations of radioactivity for ^{226}Ra, ^{232}Th, and ^{40}K (Bq/kg) in zeolite samples from Jeddah Province are shown in Figure 2.

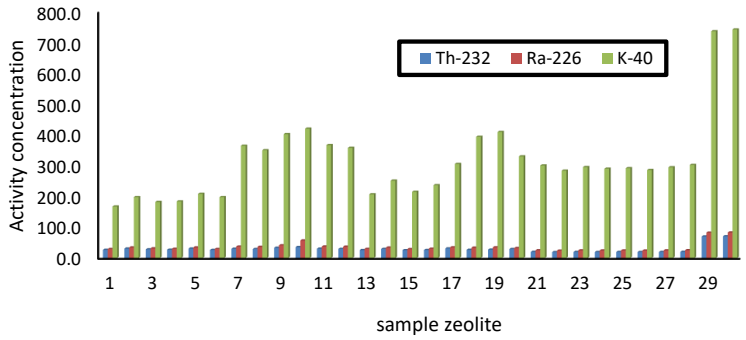

Figure 2. aveareg activity of Ra-226, Th-232 and K-40 in zeolite from Jabal Shamah in Jeddah in the Makkah region.

Table 3: Comparison of natural radioactivity concentration ($Bqkg^{-1}$) in the zeolite Samples for present study with previous study reported from different countries of the world.

Bq/kg	^{226}Ra		^{232}Th	^{40}K	REFERENCES
Zeolite (in Saudi Arabia)	Rock	35.7± 1.8	29.3 ± 1.5	282.9 ± 4.7	Present study
	Soil	27.9 ± 1.4	23.1 ± 1.2	293.0 ± 14.7	
Zeolite (in Turkey)	85 ± 4		129 ± 2	1030 ± 24	[16]
Zeolite (in Serbia)	64.8		36.5	820.3	[11]
Zeolite (in Turkey)	101.2		133.2	937.5	[11]
Zeolite (in Greece)	119		132.6	968.4	[11]

3.2. Radiological Parameters

3.2.1. Radium Equivalent Activities (Ra_{eq})

The radium equivalent activity (Ra_{eq}) is the radiation hazard index that is most widely used. Ra_{eq} [23] [22]. It is calculated using the estimates that the gamma-ray dose rates are produced by 370 Bq/kg of Ra, 259 Bq/kg of Th, and 4810 Bq/kg of K. Ra_{eq} is given by

$$Ra_{eq} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_K \quad (2)$$

The values calculated vary from 69.8 Bq/kg (sample 22) to 234.8 Bq/kg (sample 30) as shown in Fig 5.

These values are below the 370 Bq kg-1 permissible maximum value [22] .

3.2.2. Hazard Index (H)

The external hazard index for samples is given by the following equation [23] .

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

In addition to the external irradiation, radon and its short-lived compounds are also hazardous to the respiratory organs. The internal hazard index (H_{in}), which is determined by the following equation, measures the internal exposure to radon and its daughter products:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (4)$$

where A_K, A_{Ra} and A_{Th} are the activity concentrations (Bq kg^{-1}) of the specific radiation. As shown in Figure 3, all of which are below the crucial value of unity. In light of this, it can be said that there is no gamma radiation hazard health from the rock, soil, or artificial features based on the results of the radium equivalent activity and external hazard indices [23] [22].

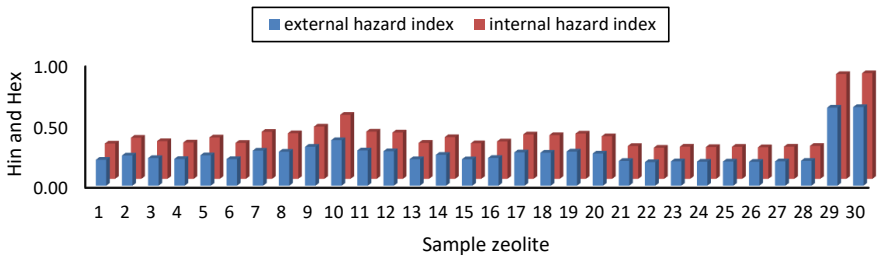

Figure 3: Average radiological hazards (H_{ex}, H_{in}) in rock, soil and artificial.

3.2.3. Representative Level Index

An additional indicator known as the representative level index, $I_{\gamma r}$, was used for assessing the hazards of radiation associated with the designated radionuclides, ^{226}Ra, ^{232}Th, and ^{40}K.For the zeolite samples that have been studied, $I_{\gamma r}$ was calculated using the following equation [22] .

$$I_{\gamma r} = (1 / 150)\, A_{Ra} + (1 / 100) A_{Th} + (1 / 1500) A_K \quad (5)$$

Where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively. The $I\gamma r$ calculated a mean value of 0.72 for rock, 0.61 for soil, and 1.74 for artificial. The average representative level index, $I_{\gamma r}$ are plotted in Figure 4.

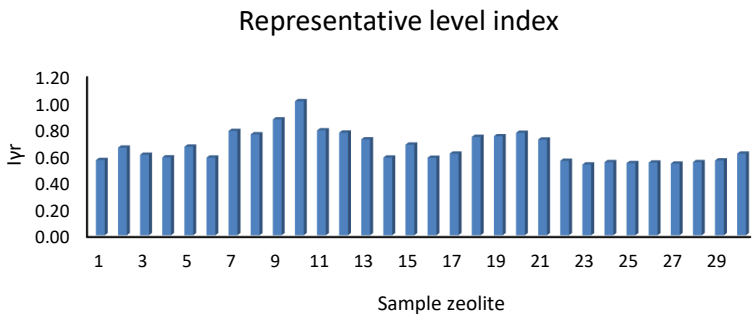

Figure 4: The average representative level index, $I_{\gamma r}$

3.2.4. Absorbed Dose

According to [24] criteria, the outdoor external dose (D_{out}) for a uniform distribution of radionuclides in air at 1 meter above the earth surface were determined. The radiation dose that is received outside due to the concentration of radio nuclides in environmental materials is expressed by the absorbed dose rate. Additionally, it represents the first major step in assessing health risk and is expressed [22] [25] .

$$D_{out} = 0.462\, A_{Ra} + 0.604\, A_{Th} + 0.0417 A_K \quad (6)$$

Equation (7) can be used to assess the indoor gamma dose (D_{in}) that is provided by ^{226}Ra, ^{232}Th and ^{40}K [25] .

$$D_{in} = 0.92\, A_{Ra} + 1.1\, A_{Th} + 0.08 A_K \quad (7)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively, in the samples.

as given in Table 4, where the upper limit is observed in artificial and the lower limit in soil zeolite. This information is calculated from the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, both of which are lower than the limits as recommended by the [26]. Except for artificial, it is above the permissible limit [25]. Figure 5 displays the samples' determined total absorbed dose and radium equivalent activity.

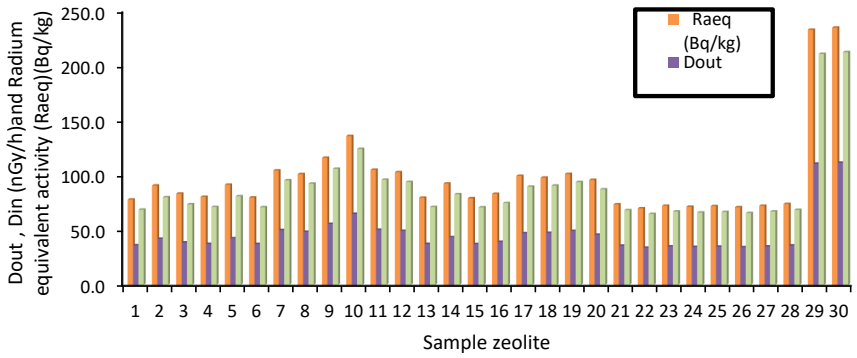

Figure 5: average activity radium equivalent (Bq/Kg), outdoor and indoor absorbed dose of rock, soil and artificial zeolite.

3.2.5. The Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent (AEDE) was calculated from the absorbed dose using an outdoor occupancy factor of 0.2 and 0.8 for indoors, and a dose conversion factor of 0.7 Sv Gy^{-1} is given by [25] [24] [19][22]

$$E_{effout}\left(\frac{\text{mSv}}{\text{y}}\right) = (\text{AEDE})\text{out door} = 1.4 \times 10^{-3} D_{out} \quad (8)$$

$$E_{effin}\left(\frac{\text{mSv}}{\text{y}}\right) = (\text{AEDE})\text{indoor} = 3.068 \times 10^{-3} D_{in} \quad (9)$$

The annual effective dose equivalent is shown in Figure 6 for the location under study, from outdoor and indoor as given in Table 4. The comparable global average is 0.41 mSvy^{-1}, of which outdoor exposure accounts for 0.07 mSvy^{-1} and 0.34 mSvy^{-1} from indoor exposure [13].

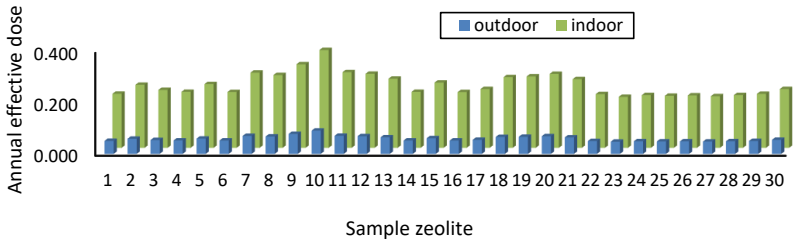

Figure 6: Average activity (Annual effective dose) in rock, soil and artificial.

3.2.6 Excess Lifetime Cancer Risk (ELCR)

calculated Excess lifetime cancer risk (ELCR) has been calculated by the following equation [25] [22].

$$(ELCR) = (AEDE) \times LE \times RF \quad (10)$$

Where: (AEDE): are the annual effective dose, LE: is the life expectancy which depends upon the average life of the individual of that region (66 years), RF (Sv^{-1}): is the risk factor per sievert, which is supposed to be 0.05 [27]. Table 4 shows the (ELCR) of indoor and outdoor exposure for rock, soil, and artificial. every sample's overall in (rock, soil) was determined to be lower than the global average of 1.45×10^{-3}. For zeolite artificial is above the permissible limit, where it poses a risk to the environment and people's health [25].

Table 4: The values of outdoor indoor and total excess lifetime cancer risk, indoor–outdoor absorbed dose, indoor, and outdoor annual effective dose

Radiation Risk	**D_{out} (nGy/h)**	**D_{in} (nGy/h)**	**E_{out} (mSv/y)**	**E_{in} (mSv/y)**	**$(ELCR)_{out}$ $\times10^{-3}$**	**$(ELCR)_{in}$ $\times10^{-3}$**	**Total (ELCR) $\times10^{-3}$**
Rock Average	**46.0**	**87.6**	**0.064**	**0.27**	**0.21**	**0.89**	**1.10**
Soil Average	**39.1**	**74.5**	**0.055**	**0.23**	**0.18**	**0.75**	**0.93**
Artificial Average	**111.0**	**211.6**	**0.1554**	**0.649**	**0.513**	**2.143**	**2.656**

3.2.7 Chemical Analysis by Energy Dispersive X-ray Fluorescence (EDXRF)

Using energy dispersive x-ray fluorescence analysis (ARL Quant'X EDXRF) for 30 elements, as indicated in (Table 5,6). The XRF results were compared in zeolite natural with other countries Table 6, and it became clear that the results are consistent with studies.

Table 5: Chemical Composition of Artificial Zeolite

Chemical composition (%)	SiO2	Al2O3	Fe2O3	BaO	SrO	Bi2O3	Cs2O	ZnO	RuO4	Y2O3	ZrO2	Ag2O
Artificial zeolite (Present study)	958.7	629.66	17.84	59.64	1.5	2.72	6.48	100	2.15	1.01	0.62	0.373

Table 6: Comparison of present Study of The XRF for zeolite natural with Others:

Sample	Chemical composition (%)								
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	Ref.
Saudi Arabia (Jabal Shamah)	56.70	16.65	9.53	8.31	1.67	0.89	3.31	1.60	Present study
Italy	51.68	18.16	3.565	4.01	1.18	1.85	5.74	0.46	[28]
Austria	67.25	11.91	1.43	2.68	1.01	0.76	2.83	-	[29]
Saudi Arabia (Harrat Shama)	66.96	15.17	1.06	3.80	1.53	0.16	2.20	0.08	[30]
Chinese	65.52	9.89	1.04	3.17	0.61	2.31	0.88	0.21	[31]
Turkey	70.90	12.40	1.21	2.54	0.83	0.28	4.46	0.09	[32]
Slovakian	61.36	14.73	3.64	1.79	2.76	-	-	-	[33]
China	76.38	11.23	1.69	5.62	1.28	3.80	-	-	[34]
Malaysia	73.40	12.50	1.88	5.28	0.98	1.06	4.47	-	[35]

From Table 8 we observe that the amount of SiO_2 in all samples was the highest value. Then the value of Al_2O_3.

CONCLUSION

When assessing the possible health risks, it's critical to ascertain the background radiation level. Gamma spectrometry was used to assess the radioactive content of 30 zeolite samples that were gathered from Jabal Shamah in Jeddah, in the Makkah region of Saudi Arabia. Zeolite samples activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K. The study's overall findings demonstrated that the observed levels fall short of global values. The permissible limit value for the total absorbed dosage rate for zeolite natural (rock. soil) was 59, both of which were below the lower requirements. as well as D_{in} 84 $nGyh^{-1}$, both of which are nearer the limit than recommended. Both the annual effective gamma doses and lifetime cancer risks

were below the global average. The Ra_{eq} activity value was found to be less than 370 $Bqkg^{-1}$, and the external hazard indices were found to be within the allowed level of unity. Because of this, the research area continues to be exposed to normal radiation levels, reducing the environmental and public health risks associated with the radioactivity of the rock and soil. However, these data can provide a general picture of the area being studied and can be used as a benchmark for future assessments and evaluations of possible radioactive risks to public health in this province. For artificial zeolite, it is beyond the permitted limit in every sample, endangering both human health and the environment.

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