GAMMA ACTIVITY IN LAHORE: PAKISTANI BUILDING MATERIALS

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الخلاصـــة :

تَـمَّ بواسطة جهاز مقياس الطيف لأشعة جاما قياس الإشعاع الطبيعي الصادر من العناصر (^{232}Th ، ^{226}Ra ، ^{40}K) الموجودة بعينات مختلفة من الطوب الطيني اللـيِّن والرمل والأسمنت - مجـمَّـعـة أو مسحوقة - وشرائح الرخام ، والزجاج المستخدم في بناء المنازل بمدينة لاهور (باكستان) . وقد عُـيِّـن لهذه المواد النشاط المكافيء للراديوم وأيضاً أدلة الخطر . وقد كان أقصىٰ قيمة للنشاط المكافيء للراديوم هي (٣١٩ ± ١٠) بيكرل/كجم للمواد الـخَـزَفية ، وهذه أقل من القيمة العظمىٰ المقبولة (٣٧٠ بيكرل/كجم) . وكانت أقصىٰ قيمة لأدلة الخطر أقل من الواحد ، ماعدا بعض عيِّـنات الـخَـزَف (السيراميك) . ولهذا فإنَّ المواد المستخدمة في البناء المعماري في لاهور لاتشـكِّل مصدر خطورة من أشعة جاما .

وتَـمَّ نظرياً حساب معدل الجرعة المكافئة الناتجة عن أشعة جاما عند منتصف حجرة ذات حجم قياسي ، وأظهرت حساباتنا أنَّ معدل الجرعة المكافئة لأشعة جاما أصغر بنسبة (٢,٥) إذا كان البناء قد شُـيِّـد من الخرسانة بدلاً من الطوب الطيني .

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ABSTRACT

Natural radioactivity due to ^{40}K, ^{226}Ra, and ^{232}Th has been measured, using a HPGe (high purity germanium) based γ-ray spectrometer, in samples of unbaked clay bricks, sand, cement, aggregate or crush, marble chips, and glass used for the construction of dwellings in the city of Lahore (Pakistan). The radium equivalent activities and hazard indices have been determined for these materials. The maximum value of the radium equivalent activity is 319 ± 10 Bq kg^{-1} in ceramics, which is less than the normally accepted limit of 370 Bq kg^{-1}. The maximum values of hazard indices are also less than unity except for some ceramic samples. Therefore, the materials used for the construction of dwellings in Lahore do not pose a major γ-ray hazard.

The dose equivalent rate due to γ-rays at the centre of a standard size room is theoretically determined. Our calculations show that the γ-dose equivalent rate is smaller by a factor of 2.5 if the buildings are made from concrete rather than from clay bricks.

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INTRODUCTION

Natural radiation is the largest contributor to the collective dose received by the world population. The sources of the natural radiation are classified as external sources of extraterrestrial and terrestrial origin and internal sources due to naturally occurring radionuclides in the human body. Terrestrial radiation originates from the radionuclides present in the crust of the Earth, in building materials, and in air [1].

Building materials with relatively higher concentrations of terrestrial radionuclides have been reported in different countries of the world [2 – 4]. Several new building materials are being explored and introduced to make the building industry economical. The raw materials used in the fabrication of these building materials may sometimes contain higher concentrations of the naturally occurring radionuclides. Industrial byproducts and wastes, being cheaper and sometimes free of cost, are also used; if produced from mines, these may have higher concentration of terrestrial radionuclides than in normally used construction materials [5, 6]. Therefore, it is necessary to measure the activities in all the materials used for the construction of dwellings, to assess the extent of radiation hazard associated with them and to take precautionary measures if necessary.

The most commonly used materials for the construction of dwellings in the various cities of Pakistan are bricks, cement, gravel aggregate or crush, sand, marble chips, and ceramic tiles. In the present studies, the natural radioactivity in construction materials used in the city of Lahore has been measured using a HPGe based γ-ray spectrometer. This city is the capital of Punjab province and is the second biggest city of Pakistan. It is a historical and industrial city expanding at a great pace and requiring new construction. The building materials of different nature are used. Some of the construction materials are fabricated around and in the city while the others are transported from the nearby towns.

NATURE OF MATERIALS AND SAMPLE COLLECTION

The materials used in this study have different origin and were collected by various means. Some of the materials were collected from the fabrication sites of the materials, some from the suppliers, and the others from the under construction sites. A brief description of the materials used in this study is as follows:

Bricks

Bricks are the major component of the building materials used in the city of Lahore. The bricks are prepared from soil and are baked in kilns. The fabrication sites lie in the geological region of Punjab Plains which consists of sediments deposited by the rivers flowing from Himalayas [7]. In and around Lahore recent alluvium (Quarternary) are deposited on the basement of igneous and metamorphic rocks. The alluvium consists of unconsolidated sand, silt, and minor amounts of clay deposited by the river Ravi and other small streams. As most of the strata has been deposited by the streams, the strata is mostly heterogeneous and individual strata has no continuity. Forty unbaked brick samples were collected from the fabrication sites in such a way so that each site is about one kilometer apart.

Sand

The city of Lahore is expanding on both banks of the river Ravi. During the winter season the quantity of water is reduced. Sand is dug out of the river and stored for construction purposes. The other sources of sand are small streams in the vicinity of the city. An other better quality sand is also available and is brought from the beds of river Chenab which is about 100 km towards the north. Therefore, the sand used in the city of Lahore belongs to various streams and the rivers Ravi and Chenab. The sand is composed of mainly quartz grains (more than 93%) and the remaining are aluminum minerals and silicates. The size of the grains ranges from 0.06 to 2 mm. The samples were collected from the suppliers and the under construction sites. Activity of twelve samples has been measured.

Cement

Cement is used for plastering of the house, making the concrete blocks for walls and lying the floors and ceilings of the dwellings. The dwellings built in the public sector have an extensive use of cement whereas the houses made in the private sector try to keep the quality of cement used as low as possible.

There are more than twenty cement factories in different parts of Pakistan but none in the premises of the city of Lahore. Therefore, the cements used in the city have different origins. Eighteen cement samples belonging to various factories were collected from the suppliers and under construction sites to determine the amount of radioactivity.

Gravel Aggregate or Crush

Gravel aggregate or crush is used in concrete for making concrete blocks, lying down the foundations and making lanterns. The sources of the gravel aggregate or crush samples used in this work are the suppliers and the under construction sites. Gravel aggregate or crush available at Lahore is composed of sandstone, limestone, and dolomite extracted from the Salt Range and Margalla formations located in the northern areas and hills near Chinout in Sarghoda division.

The crush is prepared from sandstone, limestone, and dolomite beds lying in different formations. Gravels are obtained from stream beds in the foothill zone of Indus Basin. The predominant rock type, being limestone, provides excellent parent material to yield good quality gravel which cannot be transported by natural agencies and is dumped in the stream beds and piedmonts. Purple sandstone, magnesian sandstone, and other sedimentary rocks are exposed in various parts of the Salt Range and are quite suitable for use as building or engineering stones [8, 9]. Sandstones are mainly composed of SiO_2 (upto 90%) and the remaining is CaO, MgO, Al_2O_3, Fe_2O_3, and silicates in variable concentrations.

Limestone and dolomite are available from various formations in Pakistan but the stone crush provided in the city of Lahore is mostly from the Salt Range and Margalla hills. The stone is extracted from the hills and is converted into small pieces by crushers. The chemical analysis of the limestone and dolomite is given in Table 1. Fourteen samples were collected and analyzed for radioactivity.

Marble Chips

In geology, a metamorphic rock composed of essentially a crystalline aggregate of calcite and/or dolomite is a marble. Calcite is a carbonate of calcium and dolomite is a carbonate of calcium and magnesium. They crystallize in the hexagonal system and have perfect rhombohedral cleavage. The chemical composition is almost in the same range as for limestone and dolomite, as shown in Table 1.

The marbles are crushed into chips 1–10 mm in size. These chips are then used in combination with cement and sand for lying down the floors in the house, and making the thick plasters in the bath rooms to protect them from water. The chips are transported from Azad Kashmir, the NWFP (North Western Frontier Province) and the Baluchistan provinces of Pakistan. Ten samples were collected from the suppliers of marble in the city of Lahore to determine the amount of radioactivity.

Ceramics

Ceramics in a house are used in the form of tiles in floors and bath rooms, sanitary fittings in kitchens and toilets. The other uses of ceramics in a house are the crockery and the decoration pieces. There are different factories of ceramics products in the city of Lahore and in the nearby towns. Twenty one samples were collected for radioactivity measurements from the factories located in the city of Lahore and those in the nearby towns.

Glass

Glass in a house is used as window panes which are mostly made of transparent glass. The glass is also used as crockery. The glass is manufactured in different factories existing in the city of Lahore. Thirteen glass samples were collected from these factories and were analyzed. The origin of the raw material for the glass is not known.

Table 1. Chemical Analysis in % of the Samples of Limestone and Dolomite Collected from Different Localities [8].

	CaO	MgO	Al_2O_3 Fe_2O_3	SiO_2	Loss on ignition
Limestone	43 – 55	0.04 – 4.40	0.24 – 1.52	0.22 – 1.98	56.5 – 37.1
Dolomite	26.32 – 31.43	16.80 – 21.02	0.22 – 7.36	0.06 – 11.68	56.6 – 28.51

EXPERIMENTAL METHODS

The samples were crushed into small pieces and dried in a microprocessor controlled furnace at 110°C until the weight of the sample became constant. The drying time varied from 6 hours to 20 hours. The samples were pulverized to powder form of grain size of about 1 mm. The powder was packed in the radon impermeable plastic containers. The volume of the powder was about 163 cm^3. The geometrical dimensions of the packed sample were kept identical to those of a reference calibration material (soil-6 from IAEA) which was also packed in a similar container under identical conditions. The samples were stored for more than 40 days to attain the secular equilibrium between ^{226}Ra and its daughter product ^{222}Rn [10].

The measurement technique consisted of HPGe based γ-ray spectrometry. The system was calibrated with soil-6. The details of the system and calibration procedure have been described elsewhere [10, 11]. The detection efficiency of the system 'η' was plotted as a function of γ-ray energy on the log-log graph paper and a polynomial of degree 2 was fitted to the experimental data, as follows:

$$\log\eta = 9.002 - 1.923\,(\log E) + 6.448 \times 10^{-2}\,(\log E)^2. \qquad (1)$$

Every sample was measured for 40,000 second. In this way only two samples could be measured in a day. Background was measured at weekends and average of the background was subtracted from the samples measured in that week. The activity analysis was done on a PC based MCA using the 'Intergamma' software obtained from Intertechnique, France.

RESULTS AND DISCUSSION

Specific activity due to the terrestrial radionuclides, ^{40}K, ^{226}Ra, and ^{232}Th, has been determined in the samples of construction materials. The minimum, maximum, and average values of every type of sample are represented in Table 2. As can be seen from this table, the largest of the average values due to ^{40}K is for bricks and the smallest for glass. It can also be seen from this table that the minimum and the maximum values of ^{40}K activity for cement, aggregate or crush, marble chips, glass, and ceramics show a large variations from the average values. This spread in the values is because most of the materials belong to different geological origins. The specific activity due to ^{226}Ra is largest for ceramics and is smallest for the marble chips. The minimum and the maximum values of this quantity for aggregate or crush, marble chips, and glass also show a large spread about the average values. The reason of this spread is once again the difference in the origin of raw materials for the products under consideration.

The ceramics show the largest and the aggregate or crush samples show the smallest value of the specific activity due to ^{232}Th. The minimum and the maximum values of this quantity for aggregate or crush, marble chips, and ceramics also show a large spread with respect to the average values. The reason of this spread is once again associated with the raw materials.

The sum of the activities due to ^{40}K, ^{226}Ra, and ^{232}Th is also given in the Table 2 as total activity. It is clear from this table that the average value of this quantity is largest for the bricks whereas it is smallest for glass samples. The large variations in these values are due to different composition and origin of the raw materials used for fabrication of bricks and glass.

The ratio of the specific activities due to ^{232}Th and ^{226}Ra have also been determined, and are shown in Table 3. This table shows that the bricks and sand used in this area contain more ^{232}Th than ^{226}Ra whereas the other materials, being of diversified origin, do not have a clear trend.

The sum of the activities do not provide an exact indication of the radiation hazard associated with the materials. A common index is defined in terms of radium equivalent activity (Ra_{eq}) as given by the following equation [12]:

$$Ra_{eq} = A^{Ra} + \frac{10}{7}A^{Th} + \frac{10}{130}A^{K}, \qquad (2)$$

where, A^{Ra}, A^{Th}, and A^{K} are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K respectively. The assumptions used in Equation 2 are that 10 Bq kg^{-1} of ^{226}Ra, 7 Bq kg^{-1} of ^{232}Th, and 130 Bq kg^{-1} of ^{40}K produce an equal γ-ray dose [12]. For the safe utilization of materials, the annual limit on the exposure is 1.5 mGy which corresponds to a value of 370 Bq kg^{-1} for radium equivalent concentration [13].

The determined minimum, maximum, and average values of radium equivalent activity are given in Table 3 for the materials under investigation. For the estimation of radiological consequences instead of comparing the average values the maximum value is taken into account. Considering the radium equivalent activity, the maximum value of 319 Bq kg^{-1} is for ceramics but it does not exceed the limit of 370 Bq kg^{-1}. Therefore the radiation hazard associated with these materials except for ceramics is of minor nature.

The other quantities indicating the radiation hazard are External and Internal hazard indices; H_{ex} and H_{in}, respectively. These are defined [12, 13] as follows:

$$H_{ex} = \frac{A^{Ra}}{370} + \frac{A^{Th}}{259} + \frac{A^{K}}{4810} \tag{3}$$

$$H_{in} = \frac{A^{Ra}}{185} + \frac{A^{Th}}{259} + \frac{A^{K}}{4810} . \tag{4}$$

For safe use, these indices must be less than unity. The calculated values of these indices for our measurements are given in Table 3. As can be seen from this table the values of these indices are less than unity except for some samples of ceramics where the internal hazard indices exceed the value of unity. Therefore these materials can be used in construction without any significant hazard.

We have already measured the specific γ-ray activities of some materials used for the construction of dwellings in the capital of Pakistan, Islamabad, and in its twin city Rawalpindi [14]. The average values of specific activities for bricks, sand, cement, and aggregate used in the city of Lahore are compared in Figure 1 with those used in the cities of Islamabad and Rawalpindi, for the sum of activities due to ^{40}K, ^{226}Ra, and ^{232}Th. The values of the sum of the activities in the cement samples are similar in both the cities as the suppliers of cement are same in both the regions. The values of the sum of the activities in the samples of bricks, sand and aggregate used in the city of Lahore is more than in the materials

Table 2. Gamma Ray Activities due to ^{40}K, ^{226}Ra, and ^{232}Th, and Sum of These Activities Measured in the Construction Material Samples Collected from the City of Lahore.

No. of samples	Material	Value	^{40}K (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	Total (Bq kg^{-1})
40	Bricks	Minimum	633 ± 29	40 ± 3	57 ± 3	740 ± 30
		Maximum	1024 ± 39	59 ± 3	73 ± 4	1148 ± 40
		Average	790 ± 05	50 ± 1	64 ± 1	903 ± 6
12	Sand	Minimum	562 ± 27	23 ± 3	36 ± 3	625 ± 30
		Maximum	729 ± 35	44 ± 4	69 ± 3	838 ± 35
		Average	633 ± 9	35 ± 1	51 ± 1	720 ± 9
18	Cement	Minimum	74 ± 23	21 ± 3	16 ± 2	140 ± 23
		Maximum	425 ± 21	60 ± 4	51 ± 4	482 ± 21
		Average	216 ± 5	41 ± 1	28 ± 1	285 ± 6
14	Aggregate Crush	Minimum	18 ± 14	14 ± 2	12 ± 1	47 ± 13
		Maximum	1015 ± 27	63 ± 3	98 ± 4	1171 ± 27
		Average	175 ± 5	30 ± 1	16 ± 1	221 ± 5
10	Marble Chips	Minimum	21 ± 14	6 ± 3	5 ± 2	38 ± 14
		Maximum	1036 ± 27	35 ± 3	64 ± 3	1135 ± 27
		Average	248 ± 6	16 ± 1	20 ± 1	284 ± 6
21	Ceramics	Minimum	144 ± 24	63 ± 3	10 ± 5	341 ± 25
		Maximum	834 ± 32	124 ± 5	125 ± 4	964 ± 32
		Average	403 ± 6	83 ± 1	86 ± 1	573 ± 6
13	Glass	Minimum	26 ± 20	13 ± 2	11 ± 3	65 ± 20
		Maximum	223 ± 21	63 ± 7	31 ± 3	264 ± 21
		Average	68 ± 6	24 ± 1	22 ± 1	114 ± 6

Note: The S.D. in the average value is the propagation error.

used in the city of Islamabad/Rawalpindi. These higher values of activity in the materials used in Lahore are due to the different origin of the materials. For example the aggregate or crush used in Islamabad/Rawalpindi is from the Margalla hills but in Lahore crush is also provided from other sources such as from the hills near Chinout which have different geological formations than Margalla hills.

The radium equivalent index for different materials for the cities of Lahore and Islamabad/Rawalpindi is compared in Figure 2. This figure shows that the values of this quantity are more for all materials used in Lahore except that of cement. The results of cement are comparable within the statistical errors as these should be since the supplies in both the cities are from the same sources. For other materials since the origins of the material are different, the radium equivalent values are also different.

In Pakistan, gamma-ray activities are known only for the materials used in the cities of Islamabad/Rawalpindi and Lahore. The values of radium equivalent activities for the materials of Islamabad/Rawalpindi and Lahore have been averaged and given as the representative value for Pakistan. The determined value of Ra_{eq} is compared with those available in literature [15–19] for other countries of the world in Figure 3. This figure shows that Pakistan values of Ra_{eq} within the experimental and statistical errors are comparable with many other countries. The values of Ra_{eq} for the bricks of Malaysia and those for the sand, cement, and concrete of Bangladesh are higher than the respective materials under comparison. The higher values are probably due to the geological origin of the materials.

In Pakistan, most of the buildings in public sector are constructed from concrete which is a mixture of

Table 3. Activity Ratio of ^{232}Th to ^{226}Ra, Radium Equivalent Activity (Ra_{eq}), External Hazard Index, and Internal Hazard Index Calculated for the Construction Material Samples Collected from the City of Lahore.

Material	Value	$\frac{^{232}Th}{^{226}Ra}$	Ra_{eq} (Bq kg^{-1})	H_{ex}	H_{in}
Bricks	Minimum	1.1 ± 0.1	178 ± 6	0.48 ± 0.02	0.59 ± 0.02
	Maximum	1.5 ± 0.2	237 ± 6	0.64 ± 0.02	0.80 ± 0.02
	Average	1.3 ± 0.1	202 ± 1	0.55 ± 0.00	0.68 ± 0.00
Sand	Minimum	1.3 ± 0.1	118 ± 6	0.32 ± 0.02	0.38 ± 0.02
	Maximum	1.6 ± 0.3	192 ± 8	0.52 ± 0.02	0.64 ± 0.03
	Average	1.5 ± 0.1	158 ± 2	0.43 ± 0.01	0.52 ± 0.01
Cement	Minimum	0.4 ± 0.1	74 ± 6	0.20 ± 0.02	0.28 ± 0.02
	Maximum	1.9 ± 0.3	116 ± 7	0.31 ± 0.02	0.47 ± 0.02
	Average	0.8 ± 0.1	98 ± 1	0.26 ± 0.00	0.37 ± 0.01
Aggregate Crush	Minimum	0.1 ± 0.1	31 ± 3	0.09 ± 0.01	0.15 ± 0.01
	Maximum	1.7 ± 0.1	276 ± 7	0.75 ± 0.02	0.90 ± 0.03
	Average	0.5 ± 0.1	66 ± 1	0.18 ± 0.00	0.26 ± 0.00
Marble Chips	Minimum	0.3 ± 0.1	20 ± 3	0.05 ± 0.01	0.07 ± 0.02
	Maximum	2.3 ± 0.3	207 ± 6	0.56 ± 0.02	0.65 ± 0.02
	Average	1.1 ± 0.1	64 ± 1	0.17 ± 0.00	0.22 ± 0.01
Ceramics	Minimum	0.1 ± 0.1	127 ± 8	0.34 ± 0.02	0.59 ± 0.03
	Maximum	1.5 ± 0.1	319 ± 10	0.86 ± 0.03	1.20 ± 0.04
	Average	1.1 ± 0.1	237 ± 2	0.64 ± 0.00	0.87 ± 0.01
Glass	Minimum	0.4 ± 0.1	34 ± 5	0.09 ± 0.01	0.13 ± 0.02
	Maximum	1.9 ± 0.3	108 ± 8	0.29 ± 0.02	0.46 ± 0.04
	Average	1.1 ± 0.1	61 ± 2	0.17 ± 0.00	0.23 ± 0.01

cement, sand, and aggregate in 1:2:4 ratio by volume and the water to cement ratio is 0.5 by weight. In the private sector the buildings are normally constructed by fixing the clay bricks with mud or cement – sand mixtures and plastering them with sand-cement mixtures. The volume ratio for mud, cement – sand mixture and bricks in such a construction is 1:1.5:22. Therefore for practical purposes the walls are very approximately made of bricks.

Assuming the composition of the concrete as given above, the calculated activities of ^{40}K, ^{226}Ra, and ^{232}Th in concrete are 240.1, 26.7, and 21.1 Bq kg^{-1} respectively. The activities of these radionuclides in water are not known and have been neglected.

For a room of size 4 m × 5 m × 2.8 m and wall thickness 0.2 m, the γ-dose equivalent rate $\dot{H}$ (mSv y^{-1}) at the centre of the room assuming that 80% of the total time is spent indoors is given by [20]:

$$\dot{H} = 4.9 \times 10^{-3} \sum C_j A_j \,, \qquad (5)$$

where C_j are the coefficients of γ-dose equivalent rate (nGy h^{-1}) for different radioisotopes having A_j (Bq kg^{-1}) concentrations. The coefficients for the calculation of γ-dose equivalent rate based on a Monte Carlo approach by Koblinger [21] and on a Mesh adaptive approach by Mirza *et al.* [22] are given in Table 4.

The γ-dose equivalent rate for the wall made of bricks fixed with mud and for the walls made from concrete are calculated using Equation (5) and are given in Table 4. It is clear from this table that the γ-dose equivalent rate if the wall are constructed with concrete is smaller by a factor of approximately 2.5 than that for the walls made of bricks. This trend that walls made of bricks give a higher γ-dose equivalent rate than those made from concrete, is also true for other countries for which the data is available (Figure 3).

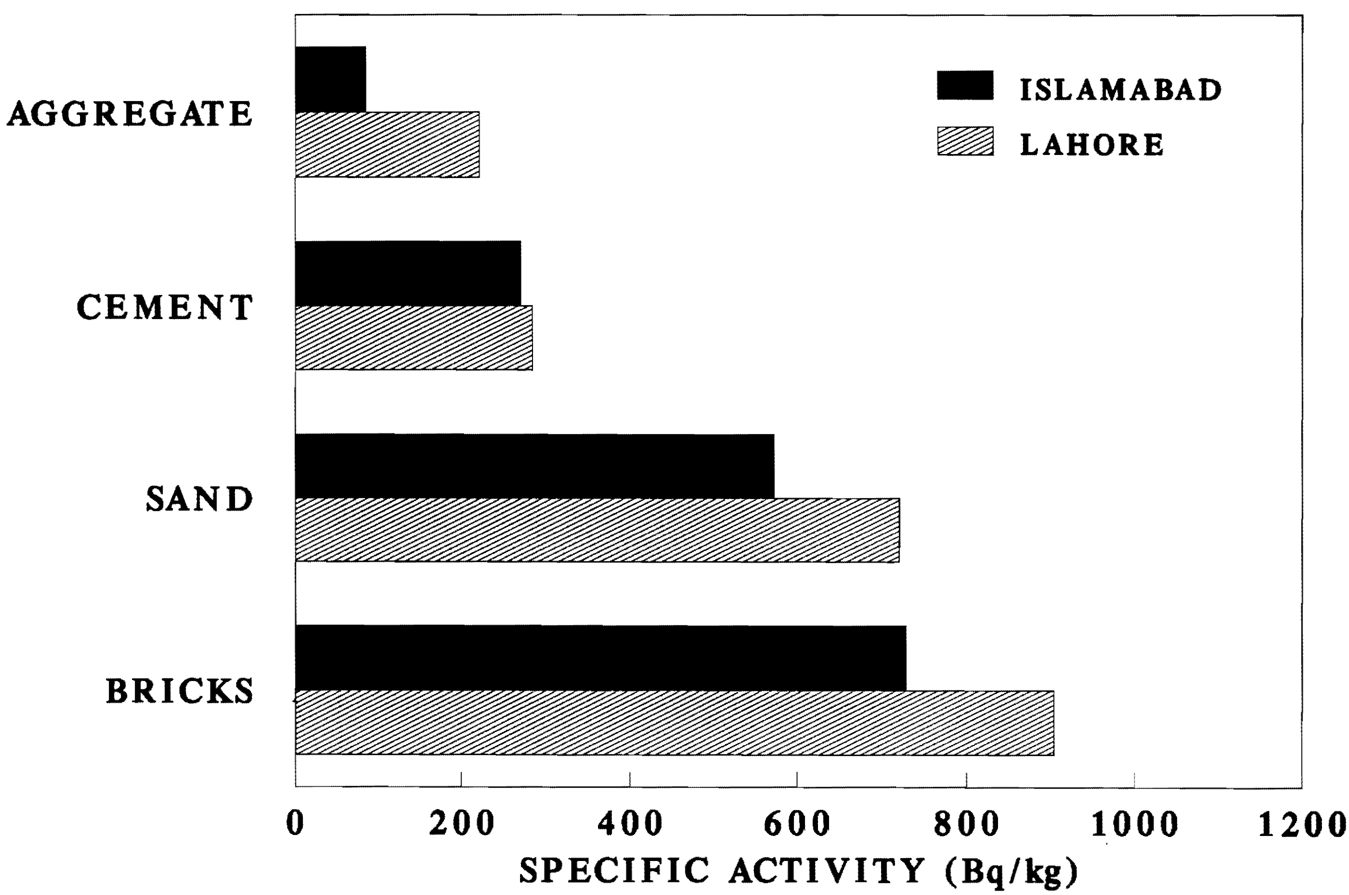

Figure 1. Comparison of Sum of the γ-Activities due to ^{40}K, ^{226}Ra, and ^{232}Th in Construction Materials Used in the City of Lahore with Those Used in the Twin Cities of Islamabad and Rawalpindi.

CONCLUSIONS

The materials used for the construction of dwellings in the city of Lahore show values of radium equivalent activity less than 370 Bq kg^{-1} and internal and external hazard indices less than unity, except for some samples of ceramics. It is concluded that these materials do not pose a major source of radiation hazard. The comparison of the results for Lahore and Islamabad/Rawalpindi shows that materials used in Lahore have more activity than in the materials used in Islamabad/Rawalpindi.

The γ-dose equivalent rate at the center of a standard size room made from concrete is smaller by a factor of 2.5 than if the room is constructed from clay bricks.

Table 4. External Dose Equivalent Rates from Different Room Models.

Method	Coefficients of specific dose rate ($nGy\,h^{-1}$ per $Bq\,kg^{-1}$)			Dose equivalent rate ($mSv\,y^{-1}$)	
	^{226}Ra	^{232}Th	^{40}K	Concrete houses	Brick houses
Monte Carlo [21]	1.05	1.18	0.09	0.37	0.97
Mesh adaptive [22]	1.2	1.29	0.1	0.41	1.1

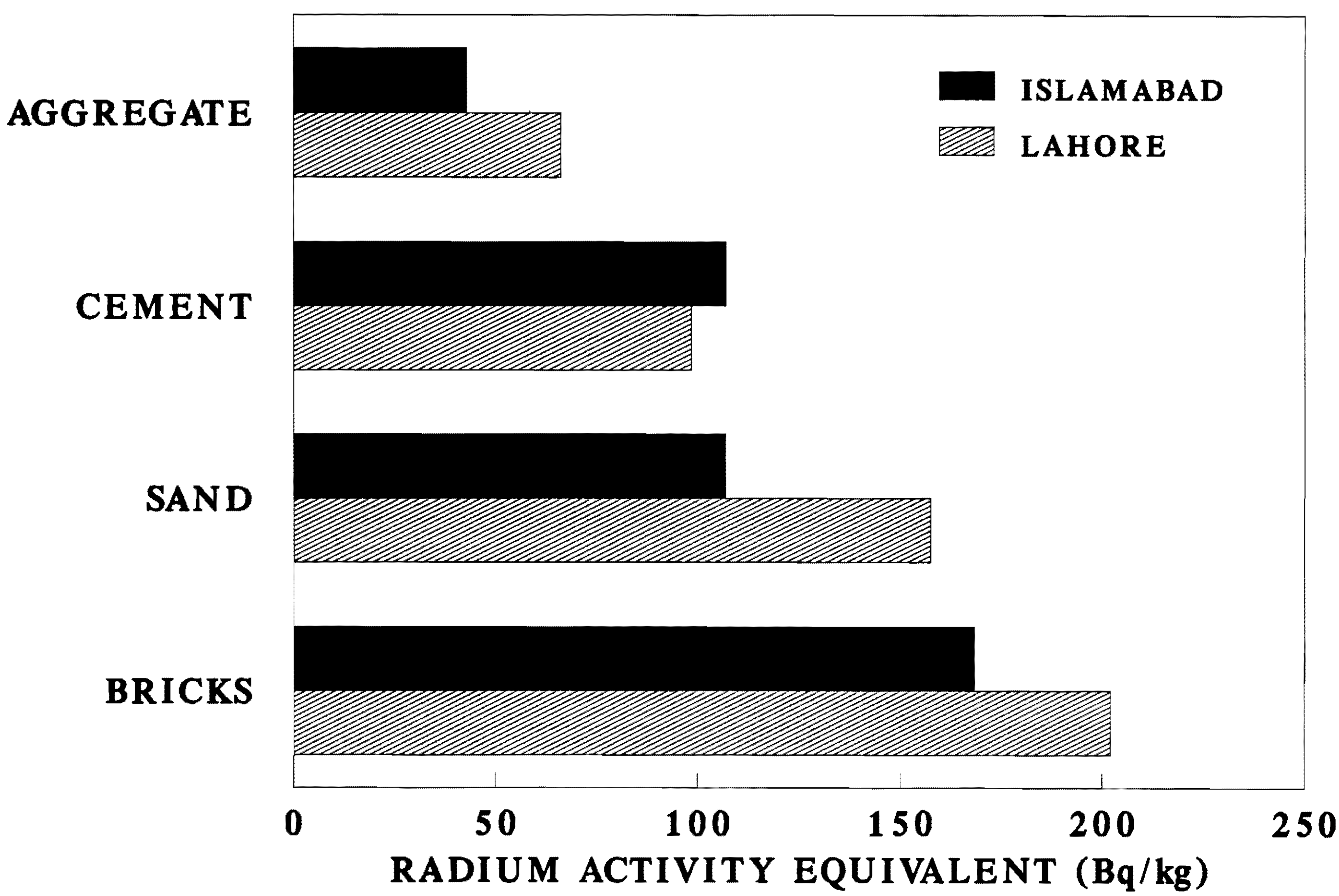

Figure 2. Comparison of Radium Equivalent Activities in the Construction Materials Used in the City of Lahore with Those in the Twin Cities of Islamabad and Rawalpindi.

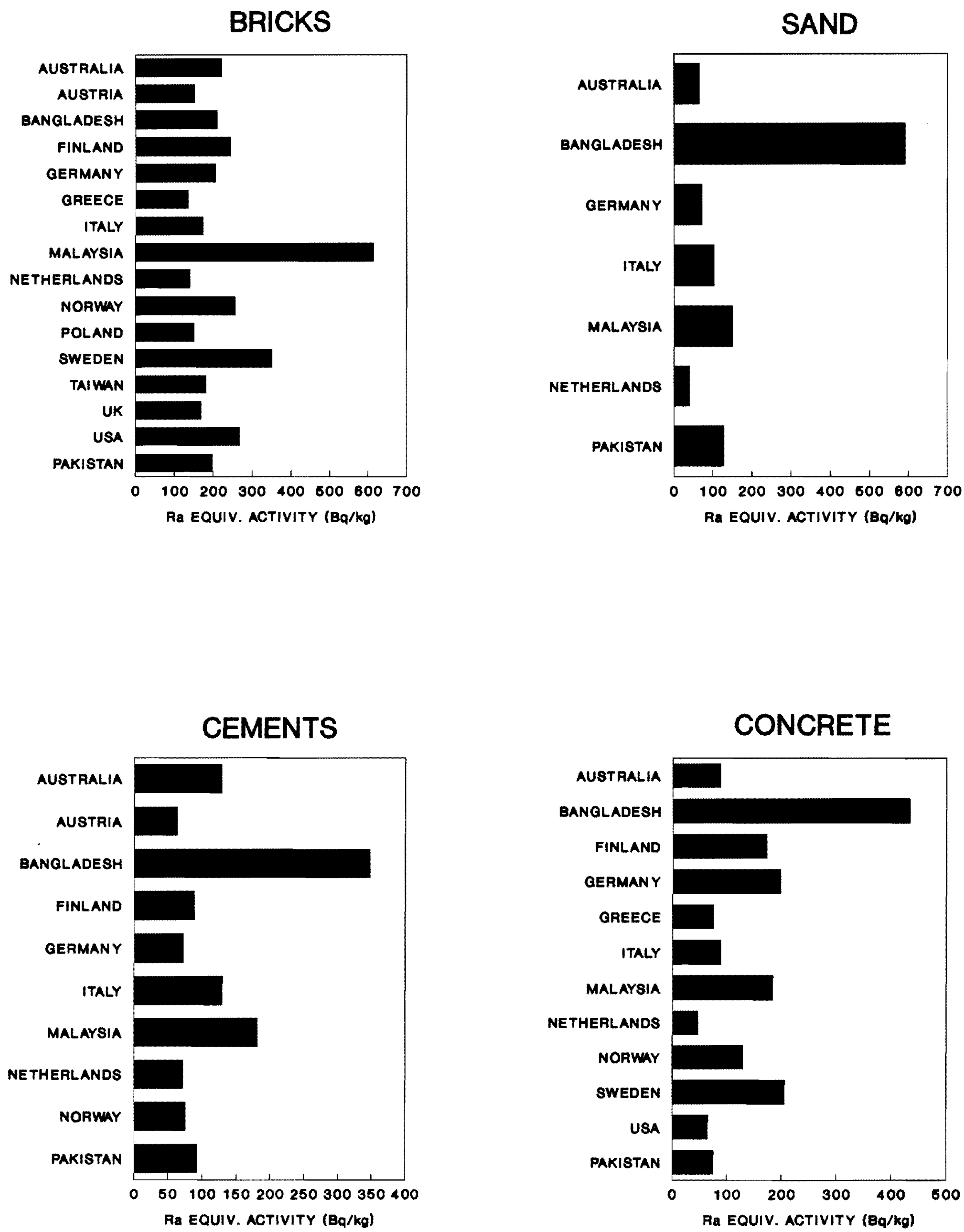

Figure 3. The Mutual Comparison of Radium Equivalent Activities in the Construction Materials Used in Different Countries of the World.

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