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ORIGINAL PAPER

Determination of radon exhalation from granite, dolerite and marbles decorative stones of the Azad Kashmir area, Pakistan

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Abstract Extensive export quality reserves of granite, dolerite and marbles which are used for interior decorations as wall facing, paving floors, kitchen counter tops, etc., are available in Azad Kashmir. Since these stones contain radium in trace amounts, therefore, its use as a building material may be a potential source of indoor radon. In order to assess health hazards due to the use of these stones as a building material, samples were collected from different mining sites. After processing, these samples were placed in plastic containers and box type radon detectors were installed in it at the height of 25 cm above the surface of the samples. The containers were then hermetically sealed. After 60 days of exposure to radon, CR-39 detectors were etched in 6 M NaOH at 70 °C for 9 h and measured track densities were related to radon concentration. Radon exhalation rate form the studied granites, marble and dolerite samples varied from 87 ± 26 to 353 ± 36 mBq m^{-2}h^{-1}, 79 ± 25 to 650 ± 42 mBq m^{-2}h^{-1} and 90 ± 26 to 324 ± 36 mBq m^{-2}h^{-1}, respectively. These decorative stones are therefore used in buildings and for export purposes as the observed radon exhalation values are smaller than that of the EPA recommended-action level.

Keywords Radium · CR-39 detectors · Radon exhalation · Decorative stones

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Introduction

Radon (^{222}Rn), a class A carcinogen, is a gaseous decay product of ^{226}Ra, a naturally occurring radionuclide found in varying amounts in almost all rocks and soils. Being a noble gas, it has greater ability to migrate freely through soil underlying the houses and get trapped within closed rooms rising to elevated levels (Matiullah et al. 1993; Rahman et al. 2007a). A sustained radon exposure has been associated with an increase in the risk of lung cancer as reported by Smith (1988), Jacobi (1988) and Lubin (1999). Radon exposure is known to be the second leading cause of lung cancer after cigarette smoking (BEIR VI "Health Effects of Exposure to Radon" 1999). Radon mainly enters the buildings from the soil-underneath the foundations, emanation from ground water use, building materials, and from the outdoor air (Bertrand et al. 1994; Durrani and Ilic 1997). Building material usually consists of bricks, cement, gravels, sand, tiles, marbles and granite. Uses of decorative stones (granite, marble dolerite) to make rooms stylish and impressive have long been seen. Granite, dolerite and marble are decorative stones used in stair floors and kitchen tops, in columns, arches balusters, steps, windowsills, walls, tombstones mausoleums, statuaries and novelties like table top, book ends, lamp bases and others.

These stone are mined from the earth's crust. The most elements in the earth's crust contain ^{238}U in trace amount, therefore, any material can be a potential radon emitter, and therefore, any building materials extracted from the earth's crust can potentially be radioactive. But the degree of threat posed by these construction materials varies, depending upon concentration of ^{238}U and ^{226}Ra contents (Al-Jarallah et al. 2001). Certain granites are part of uranium-rich bedrocks and used as building materials (Bertrand et al. 1994; Durrani and Ilic 1997). Granite is a form of igneous rock, which is composed primarily of Quartz, Alkalie and Feldspar. The decorative stones (granite, dolerite and marble) contain slightly higher amount of ^{238}U and ^{226}Ra contents as compared to other building materials and are thought to be sources of airborne radioactivity and external radiation from the decay series of uranium in buildings.

In the past several decades, radon exhalation rate from building materials has remained the subject of many studies (Abu-Jarad et al. 1980; Tufail et al. 2000; Rahman et al. 2007b, 2009, 2008; Ahmed et al. 1998; Matiullah et al. 2004). Since building materials are a significant source of indoor radon, therefore, remedial actions have to be taken in addition to the ventilation to reduce the concentration of indoor radon.

This work deals with determination of radon exhalation from the locally mined export quality decorative stones and is a continuation of our previous studies (Rafique et al. 2008, 2009, 2010). The main objective of this work is to set up radon baseline data and to find out whether or not these decorative stones are safe to be exported for use in construction of buildings.

Geology of decorative stones in Azad Kashmir

Granite, dolerite and marble are decorative stones, which are mined in many places in the Azad Kashmir, Pakistan. The mining locations include Nauseri, Jura, Keran, Kel, Kundal Shahi and around Taobat areas in Neelum Valley as may be seen in Fig. 1.

Granite

Salt and pepper type granite is found in Jura, kundal shahi and along the Neelum Valley road in the district Muzaffarabad. Owing to its hardness and appealing colours, this rock is extensively being used as a decorative stone in buildings. Preliminary geological investigations have identified extensive reserves of granite, dolerite and marbles (Fig. 2).

The Mansehra type leucogranite outcrops are present in the studied area. It forms sheet like pluri-kilomtric bodies isoclinally folded together with the other geological units. Generally, the core of the sheet is massive or less deformed than the peripheral zone where it rather appears as a leucocratic orthogneiss. Its chemical and mineral compositions range from granite to granodiorite. The typical mineral assemblage includes quartz, plagioclase, potash feldspar, muscovite, biotite and accessory, garnet, and tourmaline (Schouppe et al. 1993).

Granite characteristically contains more than 70 % silica and relatively high soda and potash $No_2 + k_2o$ which ranges from 5 to 12 %, and MgO content is usually <1 %. Granite occurs exclusively as intrusive bodies, and may occur in almost any forms like dyke, Sills, plugs, basses, ring complexes. The leucogranite of the Neelum valley can be correlated with the 516 My old Mansehra granite of the western side of the Hazara Kashmir Syntaxis and with the 400–500 My old Shengus Gneiss of the Nanga Parbat area (Fig. 3).

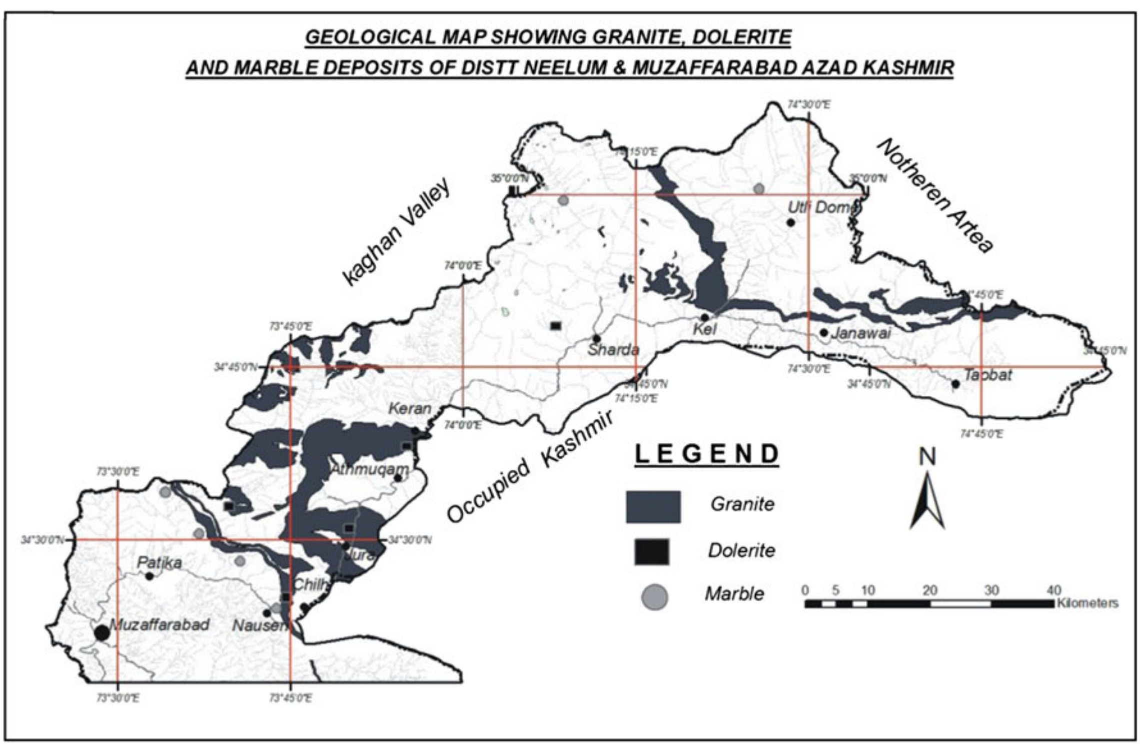

Fig. 1 Geological map showing granite, dolerite and marble reserves in districts Neelum and Muzaffarabad

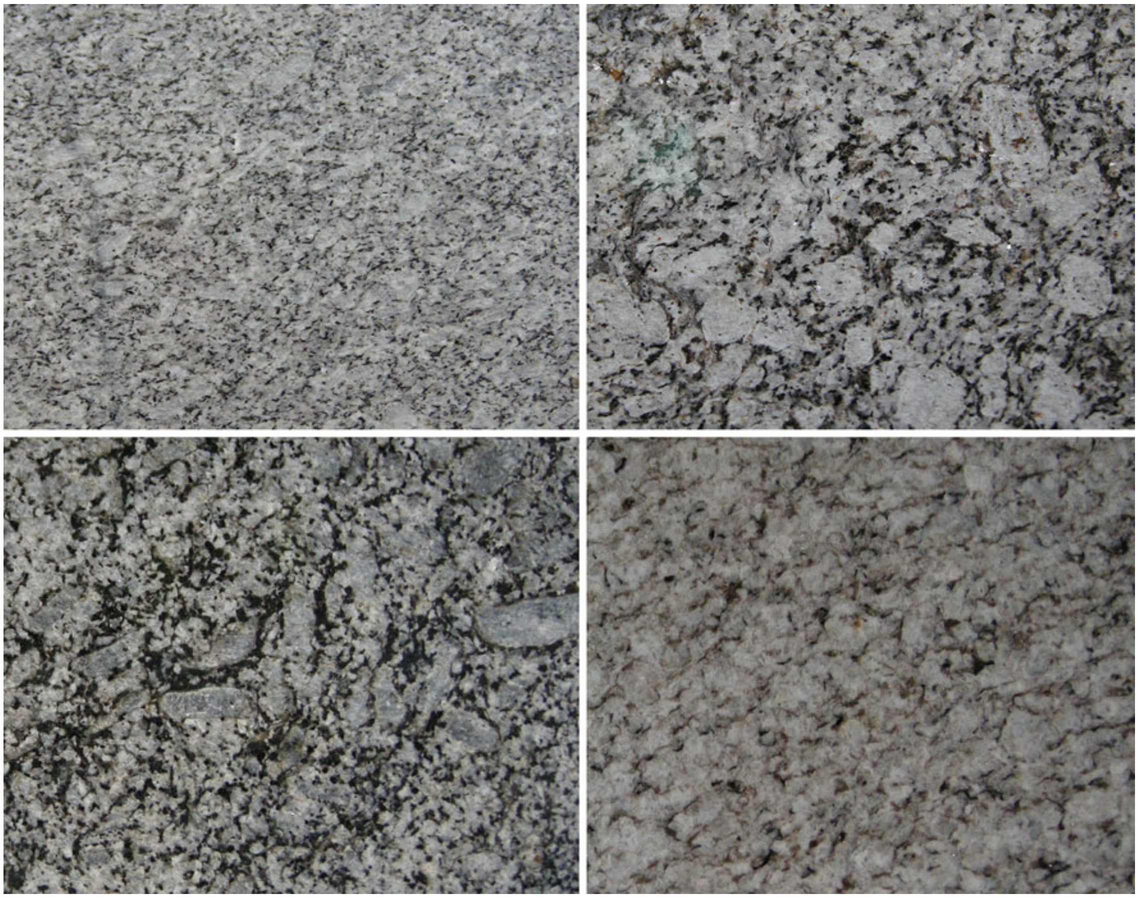

Fig. 2 Types of granite stones found in Azad Kashmir, Pakistan

Dolerite

It is medium-grained basic igneous rock, is mineralogical and chemically same as gabbro and basalt. Dolerite occurs mainly as dykes, sills and plugs, which are often old volcanic rocks. Mineralogically it is glassy basic igneous rock. The essential minerals are calcic plagioclase, pyroxene, with or without olivine; magnetite is an important accessory, whilst quartz hornblende and hypersthenes are some time present in significant amount. Basalts/dolerites contain low SiO2 content. (45–50 %), having high content of FeO, MgO and CaO whilst Na_2o and k_2o are low.

Marble

Marble is used as a dimension stone for facing walls, paving floors and in the manufacturing of terrazzo tiles. Several marble occurrences have been discovered in the districts Muzaffarabad and Neelum. The thickness of these marble ranges from 3 to 25 m and the strike extension is 200 m to 17 km. Significant deposits of snow white to appealing shades of yellowish green marbles have been discovered in the Nauseri, Treri, Jhugian areas in the

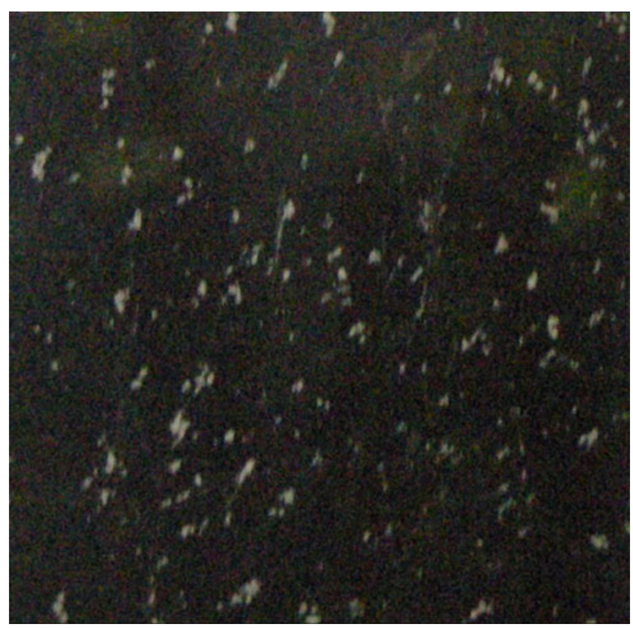

Fig. 3 Dolerite a decorative stone found in Azad Kashmir, Pakistan

district Muzaffarabad and Dhanwan area in the Kotli district. Geological reserves of these deposits are more than 20 million tons. The Nauseri marble, which is 17 km in length, is pure white in colour with occasional grey bands and fine grained. It takes good polish and is being mined by private sector. Other marble beds located in the area have very low quality and thickness.

Materials and methods

In order to carry out the present work, 'can' technique was used to measure the radon exhalation from the most commonly used decorative stones, namely granite, dolerite and marbles. In this regard, 25 samples of granite, ten samples of marbles and 15 samples of dolerite were collected from different mining locations in the Azad Kashmir, Pakistan. All the samples were crushed and dried for 4 h in an oven at 110 °C. These samples (each weighting 500 g) were then put into plastic cans of volume 8.55×10^3 cm^3. CR-39 based box type detectors were installed in cans at height of 25 cm from the surface of the samples. The cans were then hermetically sealed and the detectors were exposed to radon for 60 days. After the exposure, CR-39 detectors were etched in 6 M NaOH at 70 °C for 9 h and tracks were counted under an optical microscope. After the background correction, track densities were related to the radon concentrations (Bq m^{-3}) using a calibration factor of 0.0092 tracks cm^{-2} h^{-1} = 1 Bq m^{-3} of ^{222}Rn (Khan et al. 1991). Experimental arrangements concerning measurement of radon exhalation are shown in the Fig. 4.

Radon exhalation rate measurement

Like other natural stones, granite, marble and dolerite decorative stones also contain trace amounts of naturally occurring radionuclide like ^{238}U, ^{226}Ra, etc. Radium decays into ^{222}Rn by emitting α–particle. ^{222}Rn that

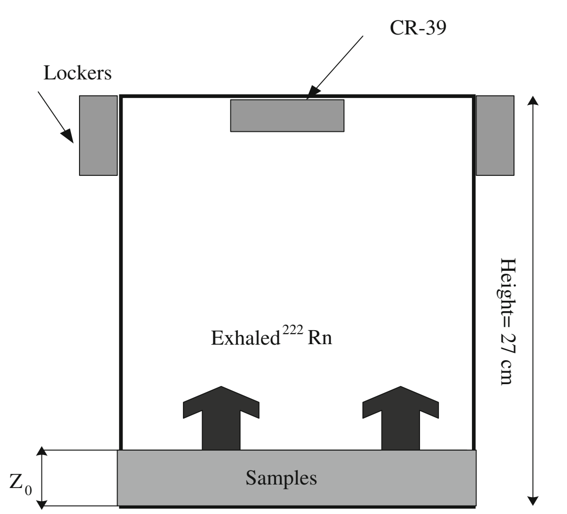

Fig. 4 Experimental set up for measuring radon exhalation rate

escapes from the mineral grain enters into the pore spaces and is transported to the indoor environment (Semkow 1990). Most of the radon produced remains within the grain and only a small fraction of it escapes to the pore spaces. This escaped fraction of radon to the pore spaces is called emanation coefficient. Radon emanation depends on numbers of factors (1) ^{226}Ra distribution and its concentration in the grain, (2) grain size, (3) water contents in pore spaces, (4) porosity, etc. (Sasaki et al. 2004; Duenas et al. 1997). Only a fraction of the atoms produced by emanation reaches the surface of the soil and is called exhalation.

Diffusion and convection mechanisms are responsible for the transport of radon in any medium. In current study, we have measured radon exhalation for each sample of decorative stone using the methodology adopted by Rehman et al. (2006b). For this purpose, radon concentration was experimentally found using CR-39 plastic track detectors. The following relation was used to determine radon exhalation rate:

$$F_0 = \frac{C(t)[\omega A + \lambda V]}{A\left[1 - e^{-\left(\frac{\omega A}{V} + \lambda\right)t}\right]}, \quad (1)$$

and for taking into account the back diffusion factor, the corrected value of radon exhalation rate is given by the following expression:

$$F = F_0 - \omega C, \quad (2)$$

where

A is the surface area of the sample (cm^2), V is the volume of void space in closed chamber, t is the radon accumulation time in the closed chamber. $\omega = \varepsilon\lambda Z_0$ is known as a back diffusion constant for given material, Z_0 is the thickness of sample in sealed chamber, $C(t)$ is the concentration of ^{222}Rn just on the surface of sample, which has to be exhaled from the surface of sample to void space of chamber,

$$F_0 = R\rho_b\lambda E Z_0,$$

where

λ is the ^{222}Rn decay constant (h^{-1}), ρ_b is bulk density of the sample (kgm^{-3}), E is the sum of fractional emanation coefficient of ^{222}Rn in air, water and adsorbed phase ($E_{air} + E_{water} + E_{solid}$), R is the concentration of ^{226}Ra (Bq Kg^{-1}).

Putting the value of $C(t)$ in Eq. (1), exhalation rate, F_0, was determined. In a closed chamber that contains a sample, ^{222}Rn concentration increases with the passage of time from zero to its maximum value. After reaching its maximum value, back diffusion of radon also takes place, which reduces the ^{222}Rn concentration by a factor ω in the chamber. Therefore, exhalation rate F, corrected for back diffusion, was determined using Eq. (2).

Table 1 Radon exhalation rate from Granite samples collected from the Neelum valley, Jhelum valley and Muzaffarabad city

Sample type	Location	Exposure time (days)	(Radon concentration ± error) (Bq m^{-3})	Radon exhalation rate (F ± error) (mBq m^{-2} h^{-1})
Granite 1	Leswa	60	174 ± 8	353 ± 36
Granite 2	Leswa	60	92 ± 10	186 ± 31
Granite 3	Jhugian	60	110 ± 10	223 ± 32
Granite 4	Islampura	60	115 ± 9	233 ± 33
Granite 5	Islampura	60	136 ± 9	276 ± 34
Granite 6	Jhugian	60	123 ± 9	249 ± 33
Granite 7	Jhugian	60	102 ± 10	207 ± 32
Granite 8	Keran	60	118 ± 9	239 ± 33
Granite 9	Taobat	60	108 ± 10	218 ± 32
Granite 10	Leswa	60	154 ± 9	311 ± 35
Granite 11	Keran	60	111 ± 10	224 ± 32
Granite 12	Jura	60	124 ± 9	252 ± 33
Granite 13	Jhugian	60	122 ± 9	247 ± 33
Granite 14	Jhugian	60	109 ± 10	221 ± 32
Granite 15	Khundal Shahi	60	141 ± 8	287 ± 34
Granite 16	Jura	60	85 ± 11	172 ± 30
Granite 17	Segam	60	80 ± 11	163 ± 30
Granite 18	Khundal Shahi	60	83 ± 11	167 ± 30
Granite 19	Khundal Shahi	60	55 ± 13	112 ± 27
Granite 20	Khundal Shahi	60	67 ± 12	135 ± 29
Granite 21	Leswa	60	78 ± 11	159 ± 30
Granite 22	Jura	60	43 ± 15	87 ± 26
Granite 23	Jura	60	44 ± 15	89 ± 26
Granite 24	Jura	60	58 ± 13	119 ± 28
Granite 25	Jura	60	81 ± 11	165 ± 30

A.M	G.M	S.D	G.S.D
A.M, G.M, S.D and G.S.D values of radon concentration from granite samples (Bq m^{-3})			
101	95	34	1.36
A.M, G.M, S.D and G.S.D values of radon exhalation rate from granite samples (mBq m^{-2} h^{-1})			
204	192	68	1.36

Results and discussion

Radon exhalation from decorative stones may contribute towards the significant radiation doses. In this regards, studies have been carried out in different parts of the world and extensive data are available in the literature. As mentioned earlier, vast deposits of the above-mentioned export quality decorative stones have been found in Azad Kashmir. In order to determine radon exhalation from these stones and hence dose delivered to dwellers, granite, marble and dolerite samples were collected from the district Muzaffarabad and Neelum. Places of collection of granite samples included Leswa, Jhugian, Islampura, Keran, Taobat, Jura, Khundal Shahi and Segam (see Fig. 1).

Table 1 shows measured values of radon exhalation rates from granite samples. The radon exhalation rate is seen to vary from 87 ± 26 to 353 ± 36 mBq m^{-2} h^{-1}. A minimum value for radon exhalation rate is found for granite sample collected from Jura, whilst maximum radon exhalation rates are found in sample collected from Leswa. Arithmetic mean, geometric mean, standard deviation and geometric standard deviation of radon concentrations for the studied granites samples are 101, 95, 34 and 1.36, respectively. No significant difference has been observed in radon exhalation rate from the studied samples.

Table 2 shows radon exhalation rate from marble samples, which were collected from Tereri, Nasuri and Jhugian locations in district Neelum, Azad Kashmir. Radon exhalation rate from marble samples varies from 79 ± 25 to 651 ± 42 mBq m^{-2} h^{-1}. A minimum value for radon exhalation was observed from a sample, which was collected from Nuseri, whilst maximum radon exhalation rate are found for the sample collected from Treri. The arithmetic mean, geometric mean, standard deviation and geometric standard deviation of radon concentrations reported for marble samples are 88, 70, 85 and 1.37, respectively.

Table 3 shows radon exhalation from dolerite samples, which were collected from different locations in district

Table 2 Radon exhalation rate from Marble and Dolerite samples collected from the Azad Kashmir, Pakistan

Sample type	Location	Exposure time (days)	(Radon concentration ± error) (Bq m^{-3})	Radon exhalation rate (F ± error) (mBq m^{-2} h^{-1})
Marble 1	Nuseri	60	45 ± 15	92 ± 30
Marble 2	Treri	60	60 ± 13	122 ± 26
Marble 3	Jhugian	60	66 ± 12	134 ± 24
Marble 4	Jhugian	60	51 ± 14	103 ± 28
Marble 5	Treri	60	55 ± 14	111 ± 28
Marble 6	Jhugian	60	66 ± 12	134 ± 24
Marble 7	Jhugian	60	57 ± 13	116 ± 26
Marble 8	Nuseri	60	39 ± 16	79 ± 32
Marble 9	Treri	60	321 ± 6	651 ± 12
Marble 10	Jhugian	60	122 ± 9	247 ± 18

A.M	G.M	S.D	G.S.D
A.M, G.M, S.D and G.S.D values of radon concentration from marble samples (Bq m^{-3})			
88	70	85	1.37
A.M, G.M, S.D and G.S.D values of radon exhalation rate from marble samples (mBq m^{-2} h^{-1})			
195	154	175	1.4

Table 3 Radon exhalation rate from Dolerite samples collected from the Azad Kashmir, Pakistan

Sample type	Location	Exposure time (days)	(Radon concentration ± error) (Bq m^{-3})	Radon exhalation rate (F ± error) (mBq m^{-2} h^{-1})
Dolerite 1	Keran	60	148 ± 8	300 ± 16
Dolerite 2	Jura	60	80 ± 11	162 ± 23
Dolerite 3	Jura	60	44 ± 15	89 ± 30
Dolerite 4	Jhugian	60	84 ± 11	170 ± 23
Dolerite 5	Khundal Shahi	60	75 ± 12	152 ± 24
Dolerite 6	Neelum Valley	60	86 ± 11	174 ± 23
Dolerite 7	Khundal Shahi	60	72 ± 12	146 ± 24
Dolerite 8	Neelum Valley	60	57 ± 13	116 ± 27
Dolerite 9	Jhugian	60	60 ± 13	122 ± 27
Dolerite 10	Jura	60	55 ± 14	111 ± 28
Dolerite 11	Khundal Shahi	60	59 ± 13	120 ± 26
Dolerite 12	Neelum Valley	60	160 ± 8	324 ± 16
Dolerite 13	Segam	60	98 ± 10	199 ± 21
Dolerite 14	Leswa	60	62 ± 13	126 ± 26
Dolerite 15	Khundal Shahi	60	61 ± 13	124 ± 26

A.M	G.M	S.D	G.S.D
A.M, G.M, S.D and G.S.D values of radon concentration from dolerite samples (Bq m^{-3})			
80	75	33	1.25
A.M, G.M, S.D and G.S.D values of radon exhalation rate from dolerite samples (mBq m^{-2} h^{-1})			
162	152	67	1.25

Neelum (see Fig. 1). Radon exhalation rate from dolerite samples ranges from 89 ± 30 to 324 ± 16 mBq m^{-2} h^{-1}. Minimum values of radon exhalation rate have been observed from a sample, collected from Jura, whilst maximum radon exhalation rate has been reported for sample #12 collected from Neelum valley. Arithmetic mean, geometric mean, standard deviation and geometric standard deviation of radon concentrations reported for dolerite sample are 80, 75, 33 and 1.25, respectively.

Comparison of A.M and G.M values of radon exhalation rate from decorative stones is shown in Fig. 5. In this figure, granite is seen to contribute more than those of marble and dolerite samples. On the other hand, marble samples

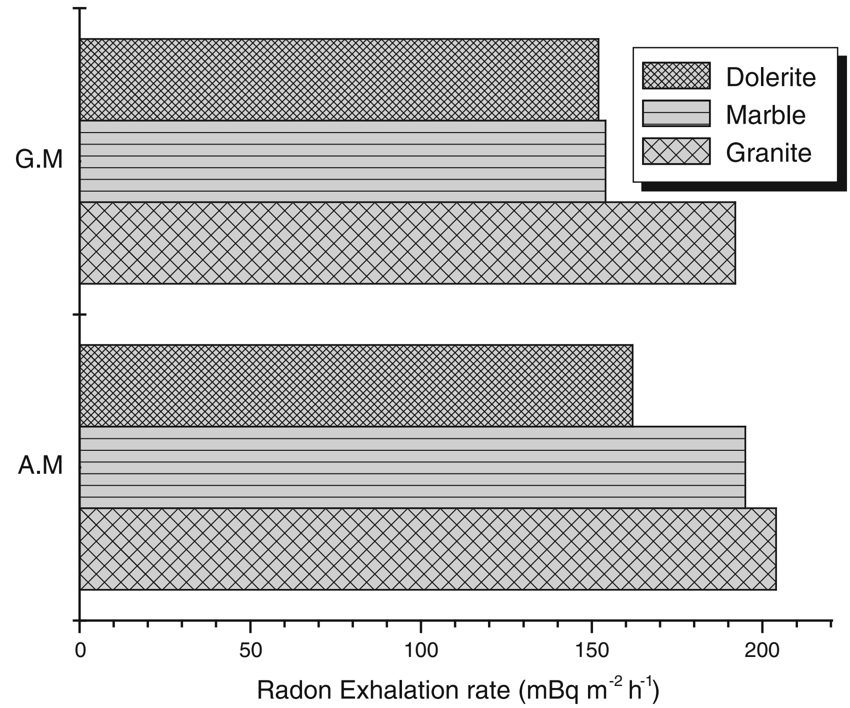

Fig. 5 Comparison of A.M and G.M values of radon exhalation rates for the studied decorative stones

Table 4 Comparison of current data of radon exhalation rate with the values reported for other countries of the world

Reference	Location	Sample type	(Average radon exhalation rate ± SD) (mBq m^{-2} h^{-1})
Amrani and Cherouati (1999)	Algeria	Granite	47–89
		Marble	35–66
Maged and Ashraf (2005)	Egypt	Marble	608
		Granite	1412
Al-Jarallah et al. (2001)	Saudi Arabia	Granite	720
Faheem et al. (2008)	Pakistan	Marble	120 ± 13
Oufni (2003)	Morocco	Quaternary samples	3–145
Shweikani and Hushari (2005)	Syria	Soil	72,000–32,400,000
Rahman et al. (2007)	NWFP, Pakistan	Sand, soil bricks	(261, 265, 292)
UNSCEAR (2000)	World average radon exhalation rate		57,600
Current study	Azad Kashmir, Pakistan	Granite marble dolerite	87 ± 26 to 353 ± 36, 79 ± 25 to 650 ± 42, 90 ± 26 to 324 ± 36

SD standard deviation

[a] unit used is (m Bq^{-1} kg^{-1} h^{-1})

have broader range of radon exhalation values. Dolerite stones have the lowest radon exhalation rate.

Referring to Table 4, significant variation is seen in the radon exhalation rate from building materials reported from different countries of the world. A lowest value of radon exhalation rate (i.e. 3.145 mBq m^{-2} h^{-1}) has been reported for quaternary samples [Morocco] (Oufni 2003). For soil samples, very high values ranging 72,000–32,400,000 mBq m^{-2} h^{-1} are reported for Syria by Shweikani and Hushari (2005), and Amrani and Cherouati (1999) have reported radon exhalation rate varying from 47 to 89 mBq m^{-2} h^{-1} and 35 to 66 mBq m^{-2} h^{-1} for granite and marbles (Algeria), respectively. For Saudi Arabia, Al-Jarallah et al. (2001) has reported average radon exhalation rate of 720 mBq m^{-2} h^{-1} for granite. Radon exhalation rates for granite, marbles and dolerite observed in the current study range from 87 ± 26 to 353 ± 36, 79 ± 32 to 651 ± 12, and 89 ± 30 to 324 ± 16 which are less than those reported by UNSCEAR (2000). Radon exhalation rates observed in the present study are well below the world average of 57,600 mBq m^{-2} h^{-1} and hence do not pose any health hazards to the residents of studied area. Moreover, as for radon exhalation is concerned, these stones are safe for exportation.

Conclusion

Radon exhalation rate from granite, dolerite and marbles decorative stones of local origin has been determined with an aim to assess the contribution of individual material to the total indoor radon exposure of the inhabitants of Azad Kashmir and to check whether or not these stones are safe to be exported. Results obtained from the current study show that radon exhalation rates from granite have relatively higher values as compared to those of marble and dolerite stone. In general, radon exhalation rate from the investigated decorative stones is well below the EPA recommended-action level. Therefore, the studied decorative stones are safe to be used for home interior decoration purposes as well as for exportation.

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