Importance of radon studies in rural areas and correlation of indoor radon level with radon inventory

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Abstract: The measurement of indoor radon in villages is very important as 50% of the world populations live in villages, and rural houses normally have very low air exchange rates. This study has estimated the possible indoor radon level from five types of rocks, which are readily available to build houses in Eastern Singhbhum district of India. In the process to calculate this radon inventory, radium content and radon flux rate were measured in the selected rocks. Then calculated radon emanation factor was compared with the possible indoor radon concentration computed from the available air ventilation rate of typical Indian houses. It was found that granite generated low radon level and it can be a good alternative choice as building material instead of other regionally used rocks. This study also suggests that important geological parameters, which develop and increase porosity, can effectively increase radon inventory.

Keywords: rural environment; indoor radon; radon emanation; rocks.

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1 Introduction

Rocks are one of the most popular and ancient choices for preparing buildings in the world (e.g. normally used for flooring purposes and wall construction). Most commonly used rock in buildings is granite, but there are many other rocks, which are readily available and are being used for this purpose. It is important here to mention that selection of these rocks depends not only on their availability and geotechnical properties but also on the socio-economic condition of the area. In India, more than 70% of the population live in villages (Census India, 2011). Most common practice of building houses in villages is using local rocks and soils. Moreover, ventilation rate of common Indian houses is very low (mean ventilation rate of 1.73 per hour; Shaikh et al., 1992), which may induce more adverse situation.

All rocks are comprised of several major (abundance more than 1%) and numerous minor elements (abundance less than 1%). Radium is one such minor element in rocks. Radon (^{222}Rn) is decay product of radium (^{226}Ra) and its concentration inside any indoor environment is a major concern in the field of environmental radioactivity. Indoor radon concentration depends mainly on radon flux rate in a room and ventilation rate of a room (European Commission, 1999; UNSCEAR, 2000), while radon flux from any material results from two processes: (a) emanation or release of radon atoms in inter-granular pore spaces after their formation in grains, and (b) migration of produced radon atoms to open atmosphere (Porstendörfer, 1994). Therefore, cavity or opening in the matrix of the material plays an important role in radon build-up process and the fraction of radon atoms that can escape from grains to inter-granular pore spaces is called radon emanation fraction or emanation factor (f) (Porstendörfer, 1994; Nazaroff, 1992; Tanner, 1980). Matrix of a rock is controlled by its mineralogy and texture, and also by structural distribution of crystals and minerals.

Several studies have been undertaken to measure radon potential from different Indian building materials (Chauhan and Chakarvarti, 2002; Sahoo et al., 2007; Pereira et al., 2008). But most of the studies are confined to in situ soils and commercial building materials. Variation in mineralogical, textural and structural distribution of rocks not only results non-uniformity in the radon source distribution but also develops heterogeneity in available pore spaces (Rama and Moore, 1990; Satomi and Kruger, 1982; Semkow, 1991; Sasaki et al., 2004; Banerjee et al., 2011). Therefore, the fate of any radon emanation therefore depends on these variations.

The objective of this work is to calculate radon emanation rates in the selected rock types (generally used for the construction of Indian village houses) and to estimate the possible indoor radon level in typical Indian village houses made of those rocks in the study area. This study also explains the correlation between radon emanation rates and indoor radon level for these rocks. Moreover, the computed indoor radon level from selected rocks is then compared with the actual indoor radon concentration in the study area to recommend most suitable rocks for dwellings in this area.

2 Methodology

Rock samples for the present work were collected from East Singhbhum district of Jharkhand, eastern part of India (Figure 1). This area is geologically famous for wide range of rock types and also for uranium mineralisation (Saha, 1994). This area was selected for this study so that the elevated uranium concentration in the studied rocks ensures an increased precision level in all the measurements, while wide range of rock types in the investigated area can show variegated control on radon generation. Rock types selected for this study include granite, schist, quartzite, shally quartzite and metamorphosed shale (Figure 2).

Figure 1 Generalised map of India showing the study area

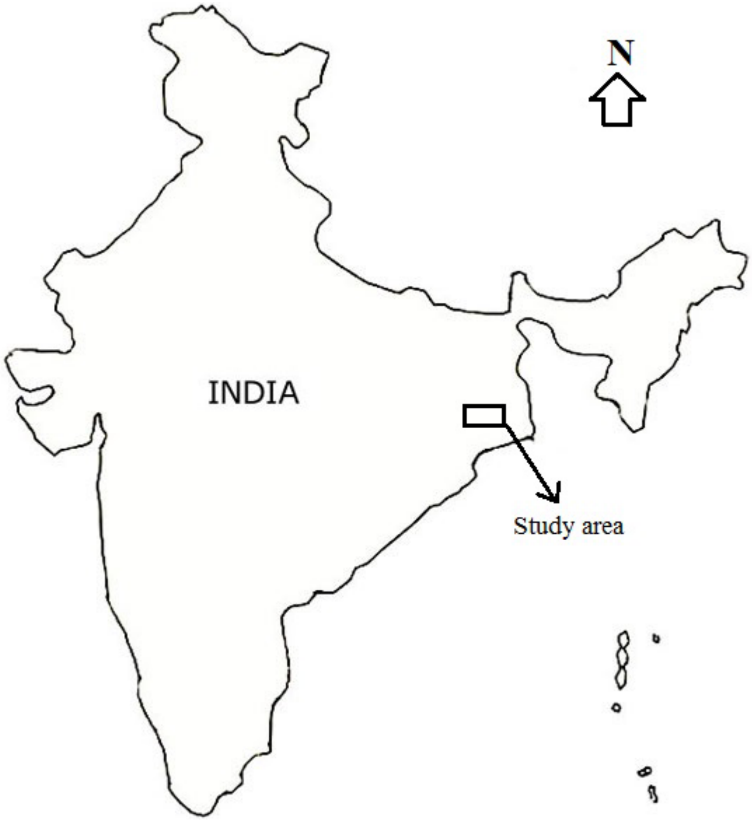
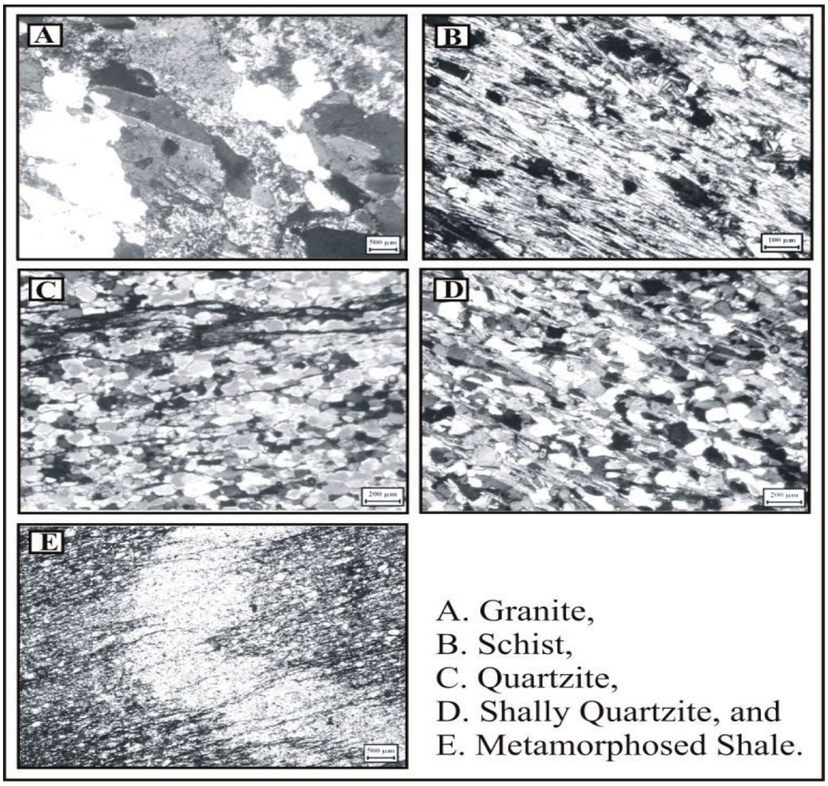


Figure 2 Photo-micrographs of the selected rock samples



2.1 Sampling and sample preparation

A total of 35 rock samples (one to 1.5 kg each) were collected from fresh rock exposures. Air-dried rock samples were first used for measuring radon flux rate. Samples were then crushed, grounded to fine powder and homogenised by passing through a 250 micron sieve accordingly. Then they were sealed in airtight plastic containers and left for more than one month, to allow secular equilibrium between ^{226}Ra and the noble gas radon (^{222}Rn) and its decay products, before counting by gamma ray spectrometry.

2.2 Estimation of radium (^{226}Ra) content by HPGe gamma ray spectrometry

Radium (^{226}Ra) in rock samples was measured by using high resolution gamma ray spectrometers consisting of coaxial HPGe detector (EG & G, Ortec) of 30% and 10% relative efficiencies at the Radiochemistry Division, Variable Energy Cyclotron Centre, BARC, Kolkata, India. The detectors were coupled to a PC-based 4K multichannel analyser and an ADC (Ortec Model 92X Spectrum Master), with appropriate software (Winmca – Maestro) for data acquisition and analysis. Energy peaks used for estimating ^{226}Ra were 186.1 keV from ^{226}Ra and 609.4 keV from ^{214}Bi. The correction for interference of the photo-peak 185 keV of ^{235}U on 186.1 keV peak of ^{226}Ra was undertaken during each calculation of ^{226}Ra.

2.3 Measurement of radon mass flux rate

Most flux measurements are based on the accumulation technique (Rama and Moore, 1984; Greeman and Rose, 1996). In this technique, sample material is placed in a closed container and radon concentration is measured by either passive alpha-track detector or online alpha counters (Ferry et al., 2002). To avoid the effects of back diffusion, leakage, etc., present flux measurements were undertaken using an online alpha detector (Aldenkamp et al., 1992; Mayya, 2004).

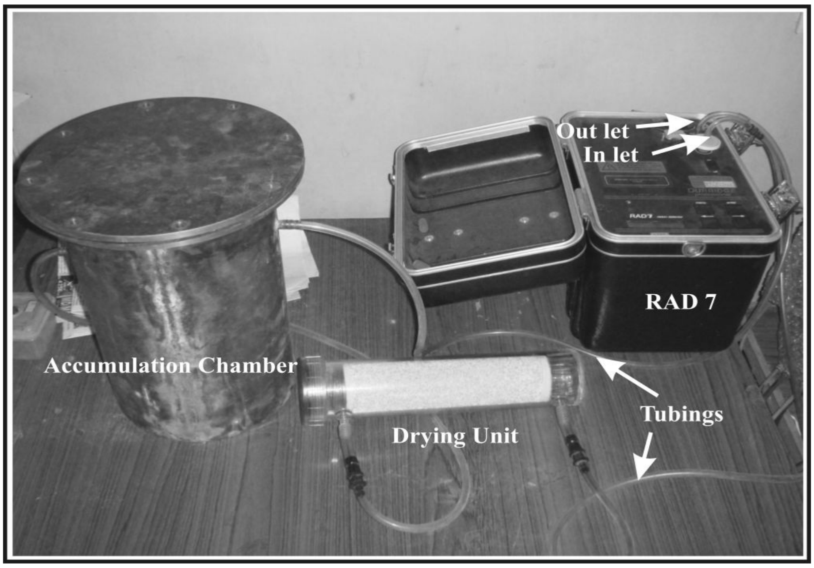
Mass flux set-up for present study consists of a cylindrical accumulation chamber (diameter of 20 cm and height of 25 cm) which was coupled with an online alpha detector (RAD7). Each block of rock samples was placed inside the chamber and flux system was sealed airtight (Figure 3). ^{222}Rn build up inside the sampling chamber was recorded by the RAD7 assuming that the total ^{222}Rn atoms at the pore volume generated from grains of the sample matrix have also reached the sampling volume (v) of the chamber. Each sample was kept for continuous measurement for 18 hrs (Stranden, 1983). The radon flux was measured using the following growth kinetic equation:

$$C(t) = \frac{J_m M}{v\lambda_e}[1 - \mathrm{Exp}(-\lambda_e t)] + C_0 \mathrm{Exp}(-\lambda_e t) \tag{1}$$

where C_0 = ^{222}Rn concentration in the chamber volume at t = 0 (Bqm^{-3}); $C(t)$ = ^{222}Rn concentration in the chamber volume at t (Bqm^{-3}); J_m = mass flux rate of ^{222}Rn from the sample (Bq kg^{-1} h^{-1}); M = total mass of the sample (kg); v = effective volume of chamber (volume of chamber − volume of sample) (m^3); λ_e = effective decay constant of ^{222}Rn, which is the sum of leak rate (if existing), back diffusion rate into sample matrix and radioactive decay constant of ^{222}Rn (h^{-1}); and t= measurement time (h).

The mass flux rate of ^{222}Rn (J_m) of rocks is determined by fitting the observed build-up of ^{222}Rn concentration inside the chamber volume to the exponential growth curve given in equation (1).

Figure 3 Radon mass flux chamber along with the RAD7



2.4 Determination of radon emanation factor

Based on the work of Sahoo et al. (2007), it was assumed that ^{222}Rn atoms that reached the pore volume of the sample mass exhaled totally into the chamber volume of the set-up. Then the ^{222}Rn mass flux rate (J_m) can be written as:

$$J_m = Qf\lambda \tag{2}$$

where Q is the radium content (Bq kg^{-1}) of the sample, f is the ^{222}Rn emanation factor of the sample and λ is the decay constant of ^{222}Rn (h^{-1}). Substituting the value for J_m in equation (2) derived through equation (1), the ^{222}Rn emanation factor 'f' was obtained by using the ^{226}Ra content of the material.

2.5 Indoor air radon level

A typical room in Indian villages has size of 10 × 10 sq. foot room with eight foot ceilings and has a ventilation rate of 39190.6 L/h of living space per hour (800 ft^3 × 1.73 per hour or 22653.5 L × 1.73 per hour), where mean ventilation rates in Indian village houses is 1.73 per hour (Shaikh et al., 1992). The concentration of indoor radon-level per kg of rocks in unit litre of indoor air volume (Bq/kg/L) can be inferred from the equation below (after Environmental Health and Engineering Report [Environmental Health and Engineering, 2013]).

$$\text{Indoor radon level} = \frac{\text{Radon flux rate}}{\text{Ventilation rate}}.$$

3 Results and discussion

3.1 Distribution of radium content, radon flux rate and radon emanation factor in rocks

All experimental and calculated results from five selected rocks are summarised in Table 1 and are described below:

Table 1 Distribution of radium content, radon flux and radon emanation in the selected rocks

Rock type	*Radium content (Bq/kg)*	*Radon flux rate (Bq/kg/h)*	*Radon emanation factor (%)*	*Indoor radon level (Bq/kg/L)* (calculated from radon flux rate)*	
Quartzite 1	172.06 ± 14.3	0.48 ± 2.88E-02	37	1.23E-05	Average value 0.53E-05
Quartzite 2	60.85 ± 5.1	0.18 ± 0.48E-02	39	0.46E-05	
Quartzite 3	131.21 ± 10.9	0.02 ± 0.12E-02	2	0.05E-05	
Quartzite 4	161.6 ± 8.1	0.48 ± 2.90E-02	39	1.22E-05	

Table 1 Distribution of radium content, radon flux and radon emanation in the selected rocks (continued)

Rock type	*Radium content (Bq/kg)*	*Radon flux rate (Bq/kg/h)*	*Radon emanation factor (%)*	*Indoor radon level (Bq/kg/L)* (calculated from radon flux rate)*	
Quartzite 5	65.1 ± 3.3	0.15 ± 0.60E-02	30	0.38 E-05	
Quartzite 6	125.3 ± 6.3	0.07 ± 0.42E-02	7	0.18E-05	
Quartzite 7	139.5 ± 7.0	0.06 ± 0.36E-02	6	0.15E-05	
Mica schist 1	344.73 ± 28.6	0.70 ± 4.20E-02	27	1.79E-05	Average value 0.57E-05
Mica schist 2	135.87 ± 11.3	0.04 ± 0.20E-02	4	0.10E-05	
Mica schist 3	160.6 ± 13.3	0.04 ± 0.24E-02	3	0.10E-05	
Mica schist 4	243.57 ± 20.2	0.57 ± 3.42E-02	30	1.45E-05	
Mica schist 5	144.18 ± 12.0	0.10 ± 0.60E-02	9	0.26E-05	
Mica schist 6	121.25 ± 10.1	0.06 ± 0.60E-02	7	0.15E-05	
Mica schist 7	150.14 ± 12.5	0.26 ± 1.56E-02	23	0.66E-05	
Mica schist 8	304.59 ± 25.3	0.03 ± 0.18E-02	1	0.08E-05	
Metamorphosed shale 1	221.18 ± 18.4	0.11 ± 0.66E-02	7	0.28E-05	Average value 0.26E-05
Metamorphosed shale 2	239.34 ± 19.9	0.16 ± 0.96E-02	9	0.41E-05	
Metamorphosed shale 3	242.0 ± 12.1	0.10 ± 0.60E-02	5	0.26E-05	
Metamorphosed shale 4	249.1 ± 12.5	0.12 ± 0.72E-02	6	0.31E-05	
Metamorphosed shale 5	228.9 ± 11.4	0.08 ± 0.48E-02	5	0.20E-05	
Metamorphosed shale 6	251.0 ± 12.5	0.07 ± 0.42E-02	4	0.18E-05	
Metamorphosed shale 7	238.4 ± 11.9	0.06 ± 0.36E-02	3	0.15E-05	
Granite 1	70.64 ± 5.9	0.03 ± 0.18E-02	6	0.08E-05	Average value 0.08E-05
Granite 2	88.83 ± 7.4	0.07 ± 0.42E-02	10	0.18E-05	
Granite 3	92.5 ± 4.6	0.03 ± 0.18E-02	4	0.08E-05	

Table 1 Distribution of radium content, radon flux and radon emanation in the selected rocks (continued)

Rock type	*Radium content (Bq/kg)*	*Radon flux rate (Bq/kg/h)*	*Radon emanation factor (%)*	*Indoor radon level (Bq/kg/L)* (calculated from radon flux rate)*	
Granite 4	98.4 ± 4.9	0.02 ± 0.13E-02	2	0.05E-05	
Granite 5	95.3 ± 4.8	0.02 ± 0.12E-02	3	0.05E-05	
Granite 6	90.4 ± 4.5	0.02 ± 0.12E-02	3	0.05E-05	
Granite 7	95.7 ± 4.8	0.02 ± 0.11E-02	2	0.05E-05	
Shally quartzite 1	118.52 ± 9.8	0.06 ± 0.36E-02	7	0.15E-05	Average value 0.21E-05
Shally quartzite 2	183.04 ± 15.2	0.09 ± 0.54E-02	7	0.23E-05	
Shally quartzite 3	178.8 ± 8.9	0.06 ± 0.36E-02	4	0.15E-05	
Shally quartzite 4	175.5 ± 8.8	0.08 ± 0.48E-02	6	0.20E-05	
Shally quartzite 5	209.6 ± 10.5	0.10 ± 0.60E-02	6	0.26E-05	
Shally quartzite 6	186.1 ± 9.3	0.11 ± 0.66E-02	8	0.28E-05	

Note: *Indoor radon level is the representative radon level that can be developed in 1 litre of indoor air from 1 kg of the rock samples analysed

1 *Quartzite*: Seven samples were analysed from this rock type. Radon flux rate and radon emanation factor were found to be directly proportional to radium content in five samples, while in other two samples this relation was opposite.

Quartzite consists of recrystallised, tightly interlocked grains, mainly of quartz. Though this variety of metamorphosed rock appears and behaves like rigid bodies (Figure 3), porosity and discontinuities in quartzite can be developed due to presence of micro-scale fractures and in some cases by recrystallised flakes of mica which lie along the inter-granular boundaries (Spray, 1969; Whitten and Brooks, 1972; Batzle et al., 1980; Kranz, 1983). The positive correlation between radon flux and emanation factor can occur owing to these micro-scale features. On the contrary, the inverse relation can be attributed by the rigid, mica-less and less fractured variety of quartzite. It is important to mention here that quartzite has a good advantage because of its crude foliation plane. These weak planes help this rock to break easily into oriented blocks and increase its preference as building material than other rocks.

2 *Mica schist or schist*: Most of the samples among eight analysed rocks showed directly proportional relationship of radon gas generation and radium activity.

Schist is that group of metamorphic rock where lamellar flakes of mica and micaceous minerals are abundant. These flaky minerals have very close spatial arrangements called foliation plane, which increases inter-granular porosity in these rocks. Mica grains also have spaced cleavage planes, which again increase their porosity. These rocks are often associated with large grains of chlorite and garnet, which are intensively fractured by the effect of high pressure and temperature related to metamorphism. As these large minerals are formed well before the formation of foliation planes in schist, often they are surrounded by large amount of cavities resulting discontinuities in foliation planes (Spray, 1969; Whitten and Brooks, 1972). All of these factors increase porosity in schist (as channels of porous layers; Figure 3) and can attribute positive correlation of radon gas flux and radium activity.

3 *Metamorphosed shale*: All the seven samples analysed in this group of rock showed very low radon flux rates and emanation values. But all values were in positive correlation with their respective radium activities.

This rock group is another variety of metamorphic rock which contains very fine grained quartz (clay size) and mica. Though abundance and parallel arrangement of mica flakes make the rock foliated, very fine grains decrease the amount of porosity in them. Only the presence of fractures and micro-cracks can develop very low porosity in these rocks (Spray, 1969; Whitten and Brooks, 1972; Gavigilo et al., 2009). These low porosity values decrease the radon emanation as well as radon flux.

4 *Granite*: This rock is most widely studied rock in the context of radon flux and building materials. In present study, seven samples were analysed and they showed lowest radon flux rates and emanation factors among other analysed rocks. Radon flux and radium activity showed direct proportional relation in this rock too.

Granite is a massive igneous rock and it is very commonly used as building material and decorative material for its lucrative colour, lustre and many other attractive properties. In this rock, grains are arranged in interlocking pattern which results low porosity. Therefore, porosity in granite can be negligible except the presence of few micro-cracks and fractures in it (McWilliams, 1996; Tuğrul and Zarif, 1999; Sousa et al., 2005).

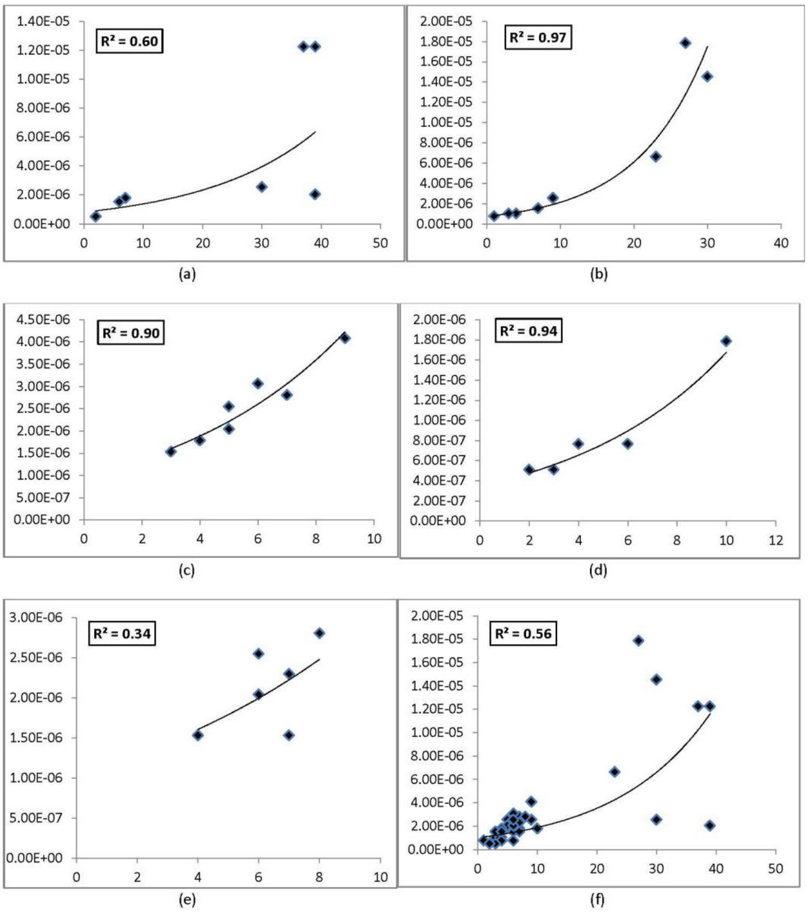
5 *Shally quartzite*: Six samples from this rock group were analysed and all of them indicated positive relation between radon flux and radium activity.

One characteristic feature of this rock group is high abundance of mica flakes than in quartzite. This rock consists of recrystallised aggregates of quartz with randomly oriented mica flakes. Though presence of mica flakes tends to increase porosity, their random orientation decreases porosity in this rock (Spray, 1969; Whitten and Brooks, 1972; Yamasaki and Chigira, 2006).

3.2 Estimated indoor radon level from these rocks and its relation with radon emanation

Most important in any type of radiation study, particularly from building materials, is to estimate the possible indoor radon level from those materials. In the present study, this estimation was calculated by using the mean ventilation rate of common rural Indian houses (Shaikh et al., 1992). The typical room type (800 ft^2) in common Indian rural houses is used to estimate the expected indoor radon level per kilogram of these measured rocks per hour. It was observed that maximum indoor radon level was found in schist and quartzite, and lowest values came from granites. Then radon emanation factor was compared with the expected indoor radon level to understand the correlation between these two values in studied rocks (Figure 4). Good correlation (R^2 values more than 0.9) was observed in schist, metamorphosed shale and granite, but this correlation was not good in quartzite ($R^2 = 0.6$), whereas very poor correlation was observed in shally quartzite ($R^2 = 0.3$). As mentioned before, internal structures, micro-structures and textures (arrangement of grains) in rocks generally control radon flux from them; correlations found in present study also confirm these controls. Therefore, the good correlation in schist and metamorphosed shale can be easily related to the abundance and connectivity of foliation planes, mica cleavages and micro-fractures, which act as channels to transport the emanated radon gas from mineral grains. In quartzite, this correlation, which was not as good as in the previous two rocks, can be inferred to the poor abundance of these cavities although they are connected to each other, whereas fractures, micro-fractures and mica cleavage planes can generate cavities in shally quartzite, but their random distribution in this rock disrupts the transport of emanated radon gas to open atmosphere. In granites, this correlation was good, though it had the least porosity value among the studied rocks. Distribution of radium, which is supposed to be uniform within the matrix of granite, is the most important controlling factor in radon flux, while, in other studied rocks, distribution of radium is not uniform and it is rather concentrated in the mica-rich layers. This helps radon flux to have good correlation with possible indoor level in granite.

Figure 4 Correlation between radon emanation and indoor radon (see online version for colours)



(a) (b)

(c) (d)

(e) (f)

4 Conclusions

1 Good correlation was observed between radon emanation factor and expected indoor radon level in all rocks except quartzite and shally quartzite. Even in granite, this correlation was very good.

2 Average indoor radon level is this area is reported to be around 50 Bq/m^3 (Kumar and Tiwary, 2008), which is greater than the typical indoor radon level (30 Bq/m^3) in other parts of the world (Chambers, 2008). People living in this area generally prefer quartzite and shally quartzite for their houses. This study suggests that granite can be a good alternative choice for buildings instead of those two preferences.

3 In the study area and other rural areas of the world, where indoor radon has elevated levels, good precautionary measures are required to reduce it.

4 Mentioned geological parameters (foliation plane, fracture, micro-fracture, mineral cleavage) should be accounted for carefully as they may increase radon level apart from other parameters.

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