



Differences of natural radioactivity and radon emanation fraction among constituent minerals of rock or soil

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ABSTRACT

We examined differences in the radioactive characteristics among the main minerals forming granite materials. Using a non-toxic high-density agent, minerals were separated from rock (granite-gneiss) and soil (weathered granite) samples. The natural radioactivity (²³⁸U and ²²⁶Ra) and radon emanation fraction of the minerals were then studied by gamma-ray spectrometry. The radon emanation fractions (27–43%) of the minerals from the soil were much higher than those (0.6–4.6%) of the rock minerals. Additionally, the emanation fractions differed greatly among the minerals separated from both the bulk rock and soil. These results were discussed in terms of the differences of surface area and radium distribution in the mineral grains. It was noticeable that a higher emanation fraction than expected for quartz was commonly observed in the rock and soil samples. We then estimated the contribution of each constituent mineral to the total radon exhalation from the bulk samples. The result depended not only on the radon emanation fraction, but also on the ²²⁶Ra activity and the mineral content. Furthermore, using the obtained data, we also discussed the effect of grain size on radon emanation and why this has been reported to vary markedly in previous studies.

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1. Introduction

Since terrestrial materials include radium (²²⁶Ra) originating from the decay of uranium (²³⁸U), all such materials release radon (²²²Rn) to varying degrees. Radon atoms, generated by the alpha decay of radium, recoil with an initial energy of 86.0 keV. Depending on their birthplace, recoil direction, etc. recoil atoms of radon can escape to pore spaces or stay in grains (Sakoda et al., 2009). Radon emanation fraction is defined as the number of radon atoms released per the number of radon atoms generated. This factor is known to be one of the most important parameters influencing indoor and outdoor radon concentrations. Many researchers have measured the radon emanation fraction of various environmental materials (Nazaroff et al., 1988). The United Nations Scientific Committee on the Effect of Atomic Radiation UNSCEAR (2000) reported that the emanation fractions of rock and soil typically range from 5% to 70% with a representative value of 20%. It has been also demonstrated that radon emanation is enhanced by increasing moisture content (Bossew, 2003; Strandén et al., 1984; Strong and Levins,

1982), temperature (Iskandar et al., 2004; Strandén et al., 1984) and specific surface area (i.e. decreasing grain size) (Barton and Ziemer, 1986; Garver and Baskaran, 2004; Kalkwarf et al., 1985; Somlai et al., 2008). On the contrary, other researches showed grain size having either an inverse effect or no effect (Breitner et al., 2008; Markkanen and Arvela, 1992; Sakoda et al., 2008a). Thus, more investigation is needed (see Section 4.2.1).

Rock and soil are composed of a variety of minerals. In spite of extensive existing data on the radon emanation, previous works have focused entirely on bulk rocks and soils rather than individual constituent minerals. Thus, whether the magnitude of the radon emanation fraction is related to types of rock or soil minerals is still not well understood. Having such data, the contribution of each mineral to radon exhalation from the ground surface could be better explored, eventually leading to an assessment of the most significant mineral in regard to human radon exposure.

In this study, we measured the specific surface areas, the ²³⁸U and ²²⁶Ra concentrations and the radon emanation fractions of minerals separated from rock and soil samples. Compared the results among the separated minerals, we discussed the radon emanation in terms of surface area and ²²⁶Ra distribution in mineral grains. We also evaluated the major contributors to radon release from the bulk samples.

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2. Materials and methods

2.1. Samples and mineral separation

Two granite-related samples with relatively high natural radioactivities were used in this study. One was crushed granite–gneiss rock collected in Badgastein, Austria, which had been used in a previous work (Sakoda et al., 2008b). The other was weathered granite soil collected in Okayama, Japan. Both were sieved into 250–500 μm grains after drying at 105 $^{\circ}\text{C}$ for 24 h. To identify their constituent minerals, the rock and soil samples were studied by X-ray diffraction (XRD) and inductively coupled plasma-atomic emission spectrometry (ICP-AES).

The primary constituent minerals were separated from the raw samples, based on a heavy liquid separation method using sodium polytungstate (SPT; $3\text{Na}_2\text{WO}_4 \cdot 9\text{WO}_3 \cdot \text{H}_2\text{O}$). SPT is superior to more commonly known heavy liquids that have been used in the past because it is non-toxic, ecologically safe, simple to handle, and easy to recover almost fully (Munsterman and Kerstholt, 1996; Six et al., 1999). Based on the densities of minerals specified by XRD and ICP-AES, SPT was first made to produce specific gravities of interest by mixing with distilled water. A few grams of the sample were placed into a centrifuge tube (40 ml) containing a few tens of milliliters of SPT. The mixture was then centrifuged at $110 \times g$ and 20 $^{\circ}\text{C}$ for a few minutes to shorten the separation process substantially. Minerals heavier than the SPT were deposited while lighter minerals floated. Each of the minerals was taken from the centrifuge tube and washed ultrasonically. The process was repeated until the quantities needed for the experiments were obtained. The separated minerals were studied by XRD and ICP-AES to ensure that the separation was appropriately accomplished.

2.2. Measurement of specific surface area

By the one-point BET method, the specific surface areas of the samples were measured employing a Yuasa Ionics MONOSORB. Each dried sample was treated in a mixed-gas flow (N_2 30% and He 70%; flow rate 70 ml/min) at 150 $^{\circ}\text{C}$ for 15 min. The sample was then cooled to liquid nitrogen temperature (–196 $^{\circ}\text{C}$), and the grain surfaces of the sample were made to adsorb nitrogen gas. Once at equilibrium, the sample was heated to desorb the nitrogen. The amount of the adsorbed nitrogen was measured using a thermal conductivity detector, and the specific surface area of the sample was determined. The detection limit is 0.01 m^2g^{-1} in this system.

2.3. Measurement of ^{238}U and ^{226}Ra concentrations

The radioactive concentrations of ^{238}U and ^{226}Ra in the samples were determined by gamma-ray spectrometry. Each dried sample of several tens of grams was enclosed in a plastic container (100 ml). The container was sealed hermetically with an epoxy resin adhesive to prevent radon from leaking out. After the radioactive equilibrium between ^{226}Ra and its progeny had been reached (approximately 30 days), gamma rays were measured using a high-purity germanium detector (GMX-15200, SEIKO EG&G, Japan). Gamma rays emitted from $^{234\text{m}}\text{Pa}$ (1001 keV) were used for ^{238}U analysis, and those from ^{214}Pb (295 and 352 keV) and ^{214}Bi (609 keV) were used for ^{226}Ra analysis.

The peak area S of gamma rays was computed using a nonlinear least-squares fitting program adopting Gaussian peak shapes. The specific radioactivity A (Bq g^{-1}) of ^{238}U and ^{226}Ra was calculated as

$$A = \left(\frac{S}{t} - \frac{S_b}{t_b} \right) \frac{1}{\varepsilon_{\text{gamma}}} \cdot \frac{1}{\varepsilon_{\text{eff}}} \cdot \frac{1}{W}, \quad (1)$$

where S_b is the peak area of background gamma rays, t and t_b (s) are the measurement times of the sample and background, respectively, $\varepsilon_{\text{gamma}}$ is the branching ratio of a gamma ray (Firestone and Shirley, 1996), ε_{eff} is the detection efficiency of the detector and W (g) is the sample weight.

2.4. Measurement of radon emanation fraction

The radon emanation fraction was investigated by gamma-ray spectrometry simultaneously with the ^{238}U and ^{226}Ra measurements. After each dried sample was closed in the plastic container, gamma rays emitted from ^{214}Pb and ^{214}Bi were measured several times before and after establishing the radioactive equilibrium between ^{226}Ra and its progeny. It was assumed that the radon activities were always equal to those of its progenies.

The change in total radon concentration C (Bq m^{-3}) per time t (s) in the solid and gas phases in the closed container can be expressed as

$$\frac{dC}{dt} = \frac{E}{V} - \lambda C, \quad (2)$$

where E (Bq s^{-1}) is the radon exhalation rate from the sample, V (m^3) is the container volume and λ (s^{-1}) is the decay constant of radon. Solving Eq. (2), the growth of radon is given by

$$C(t) = \frac{E}{\lambda V} \{1 - e^{-\lambda t}\} + C_0 e^{-\lambda t}, \quad (3)$$

where C_0 (Bq m^{-3}) is the initial radon concentration. It should be assumed that at $t=0$, all radon atoms exist in the solid phase, namely in mineral grains. Eq. (3) can be interpreted as

$$N(t) = N_{\text{eq}} \{1 - e^{-\lambda t}\} + N_0 e^{-\lambda t}, \quad (4)$$

where N , N_{eq} and N_0 (s^{-1}) are the net count rates of gamma rays at time t (day), $t > 30$ and $t=0$, respectively. The values of N_{eq} and N_0 were derived by fitting a series of counts N to Eq. (4). Fig. 1 shows an example of measured data and fitting curves. It is clear that the fitting was successful. Finally, the radon emanation fraction F (%) was calculated as

$$F = \frac{N_{\text{eq}} - N_0}{N_{\text{eq}}} \times 100. \quad (5)$$

The detection limit of F depends on the obtained total counts of gamma rays, which are closely related to ^{214}Pb and ^{214}Bi activities

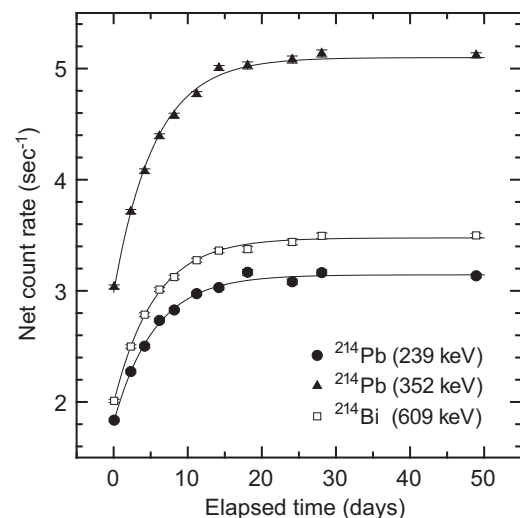


Fig. 1. Example of the growth of the radioactivity of radon progeny in a closed sample. The sample is weathered granite soil (bulk). Solid lines show the fitting curves.

of the samples and the measurement time. In the present experimental condition, the highest and lowest detection limits of F , which were estimated according to Currie's equation (Currie, 1968), are 3.5% for quartz from the rock and 1.5% for muscovite from the rock, respectively. The detection limits for the other minerals are between 1.5% and 3.5%.

3. Results

Table 1 presents the minerals separated from the raw rock and soil samples, their specific surface areas, ^{238}U and ^{226}Ra concentrations and radon emanation fractions. The rock sample was composed of quartz (Qu; SiO_2) and muscovite (Mu; $\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$). The surface areas of the muscovite grains were about twice those of quartz. The soil sample was mainly made of three minerals, quartz, microcline (Mi; KAlSi_3O_8) and goethite (Go; $\text{FeO}(\text{OH})$). Albite ($\text{NaAlSi}_3\text{O}_8$) and kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) may have also been present, but they were ignored because the amounts obtained were too small to be examined. The surface areas of microcline and goethite were 14 and 43 times larger than those of quartz, respectively.

For the minerals from the rock samples, the ^{238}U and ^{226}Ra activities in the muscovite were much higher than those in the quartz. However, the radon emanation fractions of the quartz were eight times higher than those of the muscovite. For the soil minerals, the radioactivities were much different: goethite, microcline and quartz in descending order of radioactivity. The radon emanation fractions of the goethite and the quartz were similar, but the microcline's fraction was much lower.

4. Discussion

4.1. Relationship between natural radionuclide and mineral type

The concentrations of ^{238}U and ^{226}Ra were found to be much different among the separated minerals. Silicate minerals (quartz, muscovite and microcline) and other types (goethite) should be dealt with individually to explain this result. Silicate minerals are considered first. When magma crystallizes into granite, it makes microcline first, then muscovite, and then quartz. Uranium is a large-ion lithophile element that bonds easily to silicates. Therefore, ^{238}U is expected to be abundant in a mineral that arises from the magma early. As a result of the decay of ^{238}U and its progenies (^{234}Th , ^{234}Pa , ^{234}U and ^{230}Th), early appearing minerals can be

expected to be rich in ^{226}Ra . This perspective is consistent with the result that the radioactivities of the muscovite and microcline were greater than those of the quartz. In contrast to the silicate minerals, goethite, a hydroxide mineral, might not be originally radioactive. Grains of goethite, the specific surface areas of which were much larger than those of the other minerals, may acquire highly mobile radionuclides such as ^{238}U , ^{234}U and ^{226}Ra by adsorption and/or sorption. Alternatively, it could also be thought that much smaller amounts than detected by XRD of accessory heavy minerals with high uranium activities, such as zircon and monazite, (Pavlidou et al., 2006; Plant and Saunders, 1996), coexisted with the goethite sample (4.28 g cm^{-3}).

Radioactive disequilibrium between ^{238}U and ^{226}Ra was seen in the samples derived from soil (Table 1). The ratio of ^{226}Ra to ^{238}U ranged from 2.10 to 3.50. The disequilibrium is attributed to excessive adsorptions of ^{234}U and ^{226}Ra and/or dissolution of ^{238}U . On the other hand, the $^{226}\text{Ra}/^{238}\text{U}$ ratio was found to be unity in the rock minerals. This may represent a minor degree of weathering to maintain freshness.

The distribution of ^{226}Ra within grains, which affects radon emanation (Sakoda et al., 2009; Morawska and Phillips, 1993), is deduced from the present results. From the above discussion, it may be reasonably understood that ^{226}Ra atoms in the soil minerals are enriched near the grain surfaces: goethite having the highest concentration, then microcline and then quartz. As long as only $^{226}\text{Ra}/^{238}\text{U}$ ratios are considered, microcline seems to include the most concentrated ^{226}Ra atoms near the surface. However, we acknowledged that goethite may acquire particularly great amounts of radionuclides by adsorption and sorption. On the other hand, ^{226}Ra may be uniformly distributed throughout the rock minerals, considering that they were not weathered.

4.2. Radon emanation and exhalation

4.2.1. Relationship between radon emanation fraction and mineral type

The radon emanation fractions (27–43%) of the soil minerals were much higher than those (0.6–4.6%) of the rock minerals. This supports the suggestion that radon emanation is dependent on the degree of weathering (Sakoda et al., 2008b). Weathering actions are closely related to specific surface area and ^{226}Ra distribution, as described above. Fig. 2 shows the radon emanation fraction as a function of specific surface area. The radon emanation fraction did not increase simply with increasing

Table 1
Specific surface area, ^{238}U and ^{226}Ra concentrations, and radon emanation fractions of the samples.

Sample	Mineral	Chemical composition	Specific surface area ($\text{m}^2 \text{g}^{-1}$) ^c	Radioactive concentration ^d			Radon emanation fraction (%) ^d
				^{238}U (Bq g^{-1})	^{226}Ra (Bq g^{-1})	$^{226}\text{Ra}/^{238}\text{U}$ (–)	
Granite–gneiss rock	Bulk ^a	–	0.08	6.54 ± 0.95	6.58 ± 0.25	1.01 ± 0.15	2.6 ± 0.9
	Quartz	SiO_2	0.05	0.91 ± 0.07	0.80 ± 0.03	0.87 ± 0.08	4.6 ± 0.5
	Muscovite	$\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$	0.11	12.78 ± 0.31	11.94 ± 0.41	0.93 ± 0.04	ND ^e
Weathered granite soil	Bulk ^a	–	16.18	3.35 ± 0.52	7.05 ± 0.26	2.10 ± 0.33	41.1 ± 3.1
	Quartz	SiO_2	2.00	0.63 ± 0.14	1.48 ± 0.14	2.36 ± 0.57	40.1 ± 2.6
	Microcline	KAlSi_3O_8	27.91	6.13 ± 1.01	21.46 ± 2.11	3.50 ± 0.67	27.3 ± 4.9
	Goethite	$\text{FeO}(\text{OH})$	86.40	29.97 ± 1.27	68.52 ± 6.38	2.29 ± 0.23	42.9 ± 6.6
	Albite ^b	$\text{NaAlSi}_3\text{O}_8$	–	–	–	–	–
	Kaolinite ^b	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$	–	–	–	–	–

^a Raw sample (not separated).

^b Amounts needed for the experiments could not be obtained, although the minerals may be present.

^c The value is the mean of three repeated measurements for each sample. The repeatability was 3% for the rock samples and 0.3% for the soil samples.

^d The error represents one standard deviation among at least three samples.

^e ND means "not detectable". The detection limit for the muscovite sample was 1.5%.

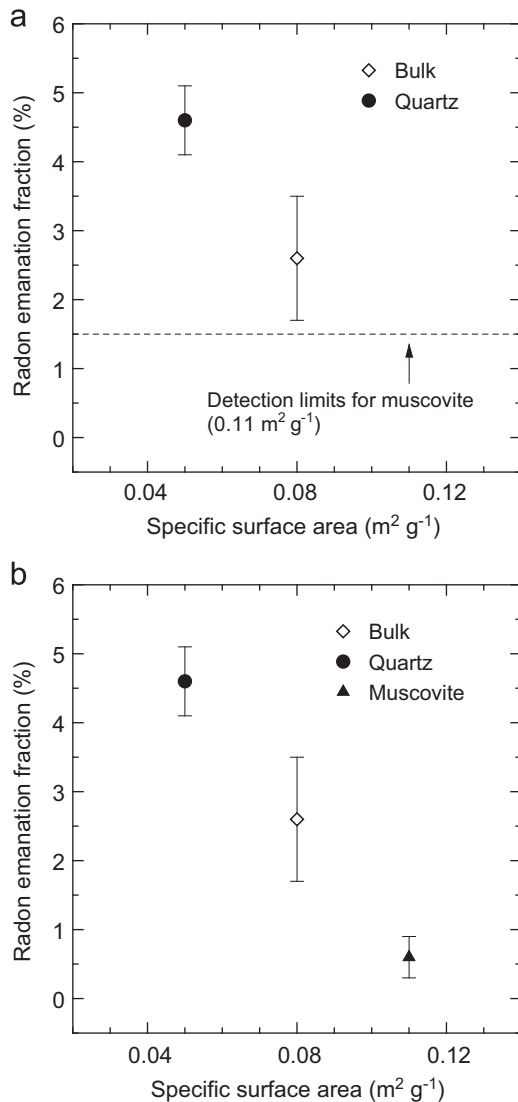


Fig. 2. Relationship between specific surface area and radon emanation fraction. (a) Granite-gneiss rock. (b) Weathered granite soil. In (a), the radon emanation fraction of muscovite is below the detection limit of 1.5% and is not plotted.

surface area. This might be due to differences in ²²⁶Ra distribution and/or the nature of the samples.

Alpha recoil is thought to play a major role in the radon emanation phenomenon, as mentioned in Section 1. Because the recoil range of radon is extremely short (e.g. 34 nm in quartz (Ziegler et al., 1985)), only radon generated within 34 nm of the surface has the possibility of being emanated. Taking into account the factors regarding surface area and ²²⁶Ra distribution that were discussed in Section 4.1, the radon emanation fraction trend of the minerals could be expected to be Go > Mi > Qu. In fact, the trend was Go ≈ Qu > Mi. The radon emanation fraction of the rock minerals, however, is expected to depend on specific surface area, assuming a uniform distribution of ²²⁶Ra. The measured value of the quartz was higher than those of the muscovite, regardless of its small surface area. These findings might suggest that quartz releases radon relatively easily. The reason cannot be explained at this time and remains to be investigated.

As presented in Section 1, some papers have shown that radon emanation was not proportional to specific surface area (Breitner et al., 2008; Sakoda et al., 2008a; Markkanen and Arvela, 1992). Reviewing the previous data, we noticed that this effect was observed mostly for samples made not of a single compound

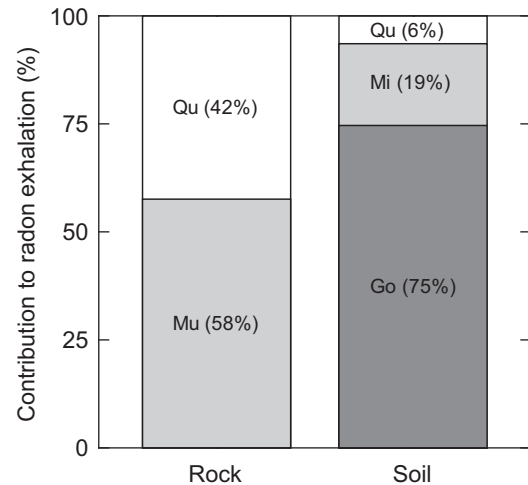


Fig. 3. Contribution of the constituent minerals to radon exhalation from bulk granite-gneiss rock (rock) and bulk weathered granite soil (soil). Qu, Mu, Mi and Go represent quartz, muscovite, microcline and goethite, respectively.

(e.g. mineral), but of mixed compounds (e.g. rock and soil). Physical and chemical resistance to weathering is different among minerals, for example, quartz is the strongest mineral. Hence, compositions of soil minerals sieved into fine fractions are likely to be quite different from coarsely sieved fractions. Further, it should be also remembered that different rock or soil minerals provide varied the radon emanation fractions (Table 1). On the other hand, based on simple model calculations, we indicated that grain size, depending on moisture content, could not affect the radon emanation for a different reason (Sakoda et al., 2009). In conclusion, the comparison of the radon emanation fractions among bulk soils made by dividing samples into several grain-size classes would not be important.

4.2.2. Contribution of individual minerals to radon exhalation from bulk samples

The radon mass exhalation rate E_m (Bq g⁻¹ s⁻¹) was calculated as

$$E_m = RF\lambda, \quad (6)$$

where R (Bq g⁻¹) is the concentration of ²²⁶Ra. Multiplying the mass exhalation rate by the weight percent of the mineral in the bulk, one can estimate its expected contribution. Fig. 3 shows the calculated radon exhalation contributions of the constituent minerals. The quartz granules in the rock and soil samples were found to have high radon emanation fractions (Table 1 and Fig. 2). However, one should be careful not to assume that quartz is the best contributor to radon release from the bulk samples to air. Our results show clearly that the contribution of quartz was slightly lower than that of muscovite in the rock and was the lowest in the soil. This reflects the lower radioactivity of quartz in each bulk sample.

5. Conclusions

We attempted to measure the natural radioactivities and radon emanation fractions of individual minerals separated from rock and soil samples. It was found that the soil samples were capable of releasing radon more readily than the rock samples. It was notable that high emanation fractions of radon from quartz were observed in spite of its grains' small surface areas. We also calculated how much each mineral contributes to the radon exhalation from bulk samples. Somewhat counterintuitive results

were observed depending not only on the radon emanation fraction, but also on ^{226}Ra concentrations and ratios of mineral contents.

Future studies to generalize radon emanation fraction trends for many different minerals would be beneficial. Based on the geology of a particular region, it would allow us to predict, without detailed emanation tests, what minerals are largely responsible for radon exposure to human.

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