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EMANATION FROM GRANITE COUNTERTOPS

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ABSTRACT

Emanation of radon (^{222}Rn) from granite countertops was measured using continuous and integrating radon monitors, and in air collected from encapsulated pieces of the countertops. A majority of the countertops emitted a measurable amount of radon. Although the granites contained the precursors, no thoron (^{220}Rn) emanation was detected. Gamma-ray spectroscopy was used to determine amounts of radium (^{222}Rn precursor) and other radionuclides in the granites. There was a correlation (r^2=0.50) between the measured radon emanation and the radium content of the granites. Based on gamma-ray spectroscopy results, radon-emanation losses from the granite samples averaged 26% of the total radium concentrations. Although some granite countertops emitted substantial amounts of radon (range= <0.1 - 2.3 pCi/L), the contribution to indoor radon concentrations was estimated to average 0.6 pCi/L.

INTRODUCTION

Radon (^{222}Rn) is a gaseous decay product of radium, a naturally occurring radionuclide found in all rocks and soils. Several extensive epidemiological studies have linked inhalation of the radioactive decay products of radon to an increased risk of lung cancer. As radon contributes half of the radiation dose received by the public from all sources, over 20,000 lung-cancer deaths are attributed to radon annually in the United States (*1*). A major fraction of indoor radon typically enters homes at the soil-foundation interface, and while reports of radon exhalation from building materials have been published, little information currently exists on the contribution from granite countertops to indoor radon levels.

There was considerable controversy regarding radon emanation from granite countertops following the publication of an article (*2*) suggesting that the emanation could be significant. The author correctly noted that (i) little information existed in the literature regarding radon emanation from granite countertops, and (ii) granite countertops are likely to emit radon. The latter is intuitive, as it is well-known that granites contain radium (^{226}Ra), the parent of ^{222}Rn. A rebuttal (3) claimed that a typical granite countertop emits $<10^{-6}$ pCi/L (or <1 decay per year) of radon. Due to the health implications of such a discrepancy, the goal of the present study was to measure radon emanation from, and the ^{226}Ra content of, granite countertops, using radioanalytical techniques.

EXPERIMENTAL

Eight tiles of granite and/or marble (collectively called granite in this study) were obtained from a local company that cuts and installs granite slabs as kitchen countertops and fireplace hearths. The granite pieces were >1 ft^2 (930 cm^2) each, and were remnants of installations. All were ~3.1 cm thick, except for granite $^{\#}$1 (2 cm). The colors of the granites ranged from black to whitish hues (Fig. 1). While the individual origins of these granites are unknown, one yellowish sample ($^{\#}$6) was labeled “Amazon Gold”. In addition to the granites brought to the laboratory, five granite countertops and hearths installed in homes were measured in-situ using the integrating monitor.

Integrating monitor

Determinations of radon flux from the granites were conducted using chambers containing a charged Teflon disc (electret). These passive radon measurement devices are commercially available (Rad Elec Inc., Frederick, MD), have been standardized for radon flux measurements, and have been described elsewhere (4). During use of the device, radon enters the 960-mL chamber (surface area of 182 cm^2) through an attached Tyvek sheet in contact with the granite, and it exits through filtered vents. Figure 2 is a photograph of the flux chamber loaded with an electret. The chamber was taped to the granite so as to eliminate air leakage during the measurements, which were conducted at ~24-hr intervals for each granite. Electrets containing an initial charge of >500 V were used for all flux measurements. Duplicate measurements were conducted 2 months apart for each granite. The voltage reader was standardized using electrets exposed at a calibration chamber, and performance was monitored using reference electrets. Aluminum foil placed between the flux chamber and the granite provided the background discharge rate due to environmental gamma. The seven aluminum-foil background measurements had an average discharge of 0.51 ± 0.10 V/h.

Continuous monitor

Continuous radon measurements were collected at 1-hr intervals using an AB-5 (Pylon Electronic Development Co. Ltd., Ottawa, Canada) passive radon detector (PRD) interfaced to a personal computer. The PRD is an alpha-scintillation counter with an absolute efficiency of 74% for ^{222}Rn and its short-lived alpha-emitting progeny. The PRD was calibrated in a radon chamber prior to use (1.1 cpm per pCi/L). For this study, the PRD was placed vertically on top of a radon flux chamber, with the PRD inlet located where the electret would normally be placed. The junction between the PRD and radon chamber was sealed with tape to eliminate air infiltration, and measurements were conducted for >20 hr for each granite. Radon emanated from the granite, passed through the Tyvek sheet of the flux chamber, migrated to the top of the chamber, and diffused through the filter of the PRD. Therefore, no contribution from short-lived ^{220}Rn (55-sec half-life) is possible. A photograph of the setup is shown in Figure 3. Duplicate measurements were conducted for each granite. Aluminum foil placed between the flux chamber and the granite provided the system background, which averaged 0.56 ± 0.11 cpm.

Isotopic gamma

Concentrations of isotopes comprising the ^{238}U and ^{232}Th decay series were determined in the granite samples using gamma-ray spectroscopy measurements. Cleaved pieces (~0.5 kg) of the granites were measured for 1000 min each using a 20% efficient Ge(Li) detector in an ultra-low-background shield. The cleaved pieces were not ground, pulverized, or otherwise altered. Data were acquired with a multiplexed system coupled to an Ethernet network as described elsewhere (5). Spectra were collected and analyzed using the Genie Spectroscopy System. Detection limits for the isotopes in the decay series were <0.01 pCi/g. Quality control was ensured by the use of standards traceable to the National Institute of Standards and Technology, successful participation in external proficiency-testing studies, and conformity to National Environmental Laboratory Accreditation Program standards.

Lucas cells

The small cleaved pieces of granite used for the gamma-ray measurements were placed inside separate glass desiccators (~4 L volume), to allow the radon (3.8-day half-life) to ingrow to secular equilibrium from the ^{226}Ra in the granites. After at least 15 days (i.e., >93% ingrowth), a small portion (125 mL) of the radon was transferred from the desiccator to an evacuated ZnS-coated scintillation chamber ("Lucas cell"). The transfer was completed using a

2-cm piece of tubing to connect an evacuated Lucas cell to the vent nipple of the desiccator (Fig. 4). Filled cells were measured within 1 min of the transfer, to estimate ^{220}Rn (thoron) emanation from the granite pieces. After allowing ingrowth of the radon decay products (>3 hr), we measured each Lucas cell at least 6 times over a 1-week period, for 100 mins each, using an alpha-scintillation counter (Model SC-5; Randam Electronic Inc., Cincinnati, OH). The radon concentrations were determined by a least-squares fitting of the decay-corrected measurements. All chamber backgrounds were <0.8 cpm, and the absolute efficiency of the detector was 76% (2.28 cpm/dpm for ^{222}Rn and its short-lived alpha-emitting progeny).

RESULTS AND DISCUSSION

Integrating monitor

Duplicate measurements of radon flux from the granites showed close agreement (Fig. 5). The electret discharge rates determined from the 24-hr measurements ranged from 0.4 to 19 V/hr, and were all, with one exception, at least twice the discharge rate observed for the aluminum-foil background (0.5V/hr), thereby indicating that radon release from granite is prevalent. The granite countertops and hearths that were installed in five homes had emanations that provided discharge rates ranging from 0.6 to 3.9 V/hr. For an 8-hr period, a 2.5 V/h decrease is measurable with a precision of ~10% (4.).

As the flux chamber had been calibrated, the flux of radon from the granites could be quantified. The measured fluxes, shown by the right ordinate in Figure 5, have been converted to commonly used units in Table 1; they ranged from 75 to 6230 pCi/m^2-hr. In comparison, 32 granites used in Saudi Arabia were reported (6) to emit up to 286 pCi/m^2-hr, while granites used in the United States emanated up to 42 pCi/m^2-hr (7). The greater fluxes measured in the current study, relative to previously reported values, indicate that the granites are quite variable, and/or that the integrating monitor was sensitive to thoron emanating from the surfaces.

Indoor radon levels can be estimated for a piece of granite emitting equally from two surfaces (Table 1) using Eq. 1.

$$\text{pCi/L (room)} = \frac{\text{emanation (pCi/m}^2\text{-hr) * granite area (m}^2\text{)}}{\text{air-exchange (room/hr)* volume (L/room)}} \qquad (1)$$

While four of the analyzed granite samples emitted >1000 pCi/m^2-hr, the contributions from a 3-m^2 granite countertop (both sides) to the radon concentrations in room air averaged 0.6 pCi/L for the eight granites, when a typical air-exchange rate (ACH; 0.5 h^{-1}) and room size (12’x12’x8’ or 32.6 m^3) are taken into account. This contribution is similar to that (0.4 pCi/L) reported by Steck (7). However, granites $^{\#}$1 and $^{\#}$5 are estimated to contribute 1.1 and 2.3 pCi/L, respectively, to the indoor air. The granites installed in the five homes are estimated to contribute 0.1 to 0.4 pCi/L to the indoor air. Based on the results of this analytical technique, the contribution of granites to indoor-air radon concentrations will often be <1 pCi/L.

Continuous monitor

In addition to the integrating monitor to measure radon flux from the granite surfaces, we used a continuous radon monitor (PRD) to quantify emanation from the surface of the granites. While the detector background was 33 ± 7 counts/hr (cph), average emissions from the granites ranged from 34 to 274 cph. As shown in Figure 6, two of the granites ($^{\#}$3 and $^{\#}$8) emitted little or no measurable radon. The four initial hourly measurements were not included in calculations, to allow ingrowth (or decay) of radon decay products in the PRD, as secular equilibrium is established. The need for a 4-hr ingrowth period is most obvious for granite $^{\#}$6 (Fig. 6).

The PRD is calibrated for concentration measurements, so units of flux were not determined. However, an indoor radon concentration can be estimated for a 3 m^2 piece of the granites using the following:

$$\text{pCi/L (room)} = \frac{\text{emanation (pCi/L)} * (3\ \text{m}^2/\text{chamber-area ratio}) * 2\ \text{sides}}{\text{air-exchange (room/hr)} * \text{volume (L/room)}}$$

When interfaced to the granites, the flux chamber and PRD accumulated from <0.01 to 3.5 pCi/L. Therefore, the radon emanations into the chamber (surface area =182 cm^2) ranged from ~0 to 190 pCi/L (average =50) for 1 m^2 of the granites (right ordinate of Fig. 6). Although the granites can emanate a significant quantity of radon, the contribution of the granites to the radon levels in the room air is estimated to be <0.1 pCi/L, when the air-exchange rate (0.5 h^{-1}) and room volume (32.6 m^3) are taken into account. Note that the rankings of the granites with regards to their radon emissions are somewhat similar for the integrating and continuous monitors.

Isotopic gamma

As the Ge(Li) detector was not calibrated for the irregular shape of the cleaved granite pieces, exact determinations of isotopic activity were not possible. However, estimates of radioactivity were made using a similar standardized geometry. Also, use of a single geometry allows an intercomparison of the granites' results. As shown in Figure 7, most of the granites contain 0.1-1 pCi/g of ^{238}U decay products, a range similar to that found in typical soils and reported elsewhere (7). One granite sample ($^{\#}7$) contained a concentration of the ^{238}U decay products that was roughly an order of magnitude greater. The similarity in activities of isotopes comprising the decay series suggests that radioactive equilibrium exists in the granites, with the exception of some ^{222}Rn losses. The difference between concentrations of ^{226}Ra and ^{214}Bi (or

^{214}Pb) measured in the granite pieces is indicative of the radon emanation. According to this approach, 16-51% (average=28%) of the radon appears to emanate from the granites. However, as ^{235}U produces a gamma-ray (185.7 keV) with an energy indistinguishable from ^{226}Ra (186.0 keV), the actual emanation may be slightly less. Since natural ^{235}U activity is only ~5% of the ^{238}U activity, and since many of the analyzed granites contained little or no measurable ^{235}U (based on 143-keV gamma-ray), any contribution from ^{235}U to the 186-keV photopeak is insignificant. The average radon emanation from 12 granite and marble samples has been reported (6) to be 21% of the total radium concentration. Granites #6 and # 7 contained the greatest ^{226}Ra concentrations and, as shown below, emanated the greatest amount of radon in the desiccators. The linear correlation coefficient (r^2) between emanated radon (measured with Lucas cells) and radium content was 0.50. An estimate of the indoor radon concentration for a piece of granite emitting equally from two surfaces can be made using the following:

$$\text{pCi/L(room)} = \frac{\text{density (g/cm}^3\text{)* }^{226}\text{Ra conc. (pCi/g)*granite volume (cm}^3\text{)*emanating factor (\%)}}{\text{air-exchange (room/hr)* volume (L/room)}}$$

Based on the range of measured densities (2.60-2.70 g/cm^3), the individual slab thicknesses (2-3.1 cm), and the ^{226}Ra content of the measured granites, a 3-m^2 granite countertop will contain 900 to 590,000 pCi of ^{226}Ra. Therefore the ^{222}Rn released by a 3-m^2 granite countertop into a room (32.6 m^3 and 0.5 ACH), will produce an average indoor radon level of 2 pCi/L, and a range of ~0 to 6 pCi/L for the eight analyzed granites, based of the emanation factors determined by the disequilibrium between ^{226}Ra and its progeny for the individual granites (16-51%). Three granite samples are estimated to produce >2 pCi/L in the room.

Concentrations of the gamma-ray emitting isotopes from the ^{232}Th decay series, ^{40}K, and ^{235}U are given in Figure 8. Granites #1 and # 8 contain the largest concentration of isotopes from the ^{232}Th decay series. Unlike the ^{238}U decay series, the isotopes comprising the ^{232}Th decay series have identical concentrations (i.e., radioactive equilibrium) in the granites, implying that an undetectable amount of ^{220}Rn emanated from the granites. This is expected, due to the short half-life (55 s) of the ^{220}Rn atom. Concentrations of ^{40}K were 1 to 100 times greater than those of ^{226}Ra, while the ^{235}U concentrations are typically <0.1 pCi/g. In a study of 29 granite countertops, Steck (7) reported averaged ^{222}Rn, ^{220}Rn, and ^{40}K concentrations of 1.5, 1.9, and 30.7 pCi/g, respectively. In the present study, the measured concentrations averaged 2.2, 1.5, and 29.6 pCi/g, respectively.

Lucas cells

Concentrations of radon that emanated from the granite pieces inside the desiccators ranged from <0.1 to 9.7 pCi/L. Six of the granites emanated <1 pCi/L of radon into the enclosed space. Two samples ($^{\#}$6 and $^{\#}$7) produced nearly 10-fold greater concentrations of radon (Table 2) and, as shown above, contained the greatest ^{226}Ra concentrations. The 8 pieces emitted from ~0 to 22 pCi/L for each kg of granite. An estimate of the indoor radon concentration from the granites can be made using the following:

$$\text{pCi/L (room)} = \frac{\text{emanation (pCi/L-kg) * countertop mass (kg)}}{\text{air-exchange (room/hr)* volume (L/room)}}$$

Based on the mass of the average granite countertop (~250 kg), the granites could be expected to contribute $\leq$0.3 pCi/L to a room (0.5 ACH and 32.6 m^3). Therefore, based on this method, radon emanation from the granites is expected to contribute little to the indoor concentration.

CONCLUSIONS

Radon emanation from eight pieces of granite countertops, and from five installed countertops, was determined. The former were also analyzed for radium and other gamma-ray emitters. Although most of the granites contained a substantial quantity of ^{222}Rn and ^{226}Ra, and although the various analytical techniques confirm that a portion of the ^{222}Rn emanates from the surface, the contribution to indoor radon concentrations is not likely to be a health concern in most cases.While the contribution to indoor levels was estimated to be insignificant for the encapsulated-granite and continuous- monitor methods, the integrating monitor and isotopic gamma analyses indicated that some granite countertops have the potential to significantly influence indoor radon levels.

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FIGURE CAPTIONS

Fig.1. Photograph of some of the analyzed granites.

Fig. 2. Photograph of the flux chamber (integrating monitor) loaded with an electret.

Fig. 3. Photograph of the continuous monitor (PRD) interfaced with the flux chamber.

Fig. 4. Photograph of granite pieces in desiccators, Lucas cells, and alpha-scintillation counter.

Fig. 5. Results of replicate measurements of radon flux from the granites using integrating monitors are in close agreement.

Fig. 6. Emanation from many of the granite surfaces are above background.

Fig. 7. Concentrations of the gamma-ray emitting isotopes from the ^{238}U decay series measured in the granite samples.

Fig. 8. Concentrations of the gamma-ray emitting isotopes from the ^{232}Th decay series, ^{40}K, and ^{235}U measured in the granite samples.

Table 1. Results of radon flux measurements from granites, and estimated contributions to indoor radon levels.[a]

Sample	Radon flux (pCi/m^2-hr)	Contribution (pCi/L)
Granite 1	6230	2.29
Granite 2	224	0.08
Granite 3	82	<0.01
Granite 4	1290	0.47
Granite 5	3290	1.21
Granite 6	1180	0.43
Granite 7	370	0.13
Granite 8	262	0.09

[a] Assuming 6 m^2 surface area ($3\ m^2$/side), 0.5 room air-change/ hr, and a 32.6 m^3 (12' x 12' x 8') room.

Table. 2. Nuclides used for gamma-ray measurements of granites.

U-238 decay series		Th-232 decay series	
Isotope	Energy (keV)	Isotope	Energy (keV)
Pb-210	46.5	Ac-228	911.1
Th-234 (U-238)	63.3	Pb-212	238.6
Ra-226	186.0	Tl-208	583.1
Pb-214	295.2		
Bi-214	609.3	Additional isotopes	
		U-235	143.8
		K-40	1460.8

Table. 3. Information regarding granites stored in desiccators.

Sample	Air conc. (pCi/L)	% error[a]	pCi/L /kg	sample wt (g)
Granite 1	0.09	58	0.36	254
Granite 2	0.21	41	0.74	485
Granite 3	ND			652
Granite 4	0.71	20	1.52	469
Granite 5	0.32	24	1.76	346
Granite 6	9.66	4	21.8	442
Granite 7	7.82	6	12.1	646
Granite 8	ND			639

[a] One sigma counting error.

ND = not detected above background.

Figure 1.

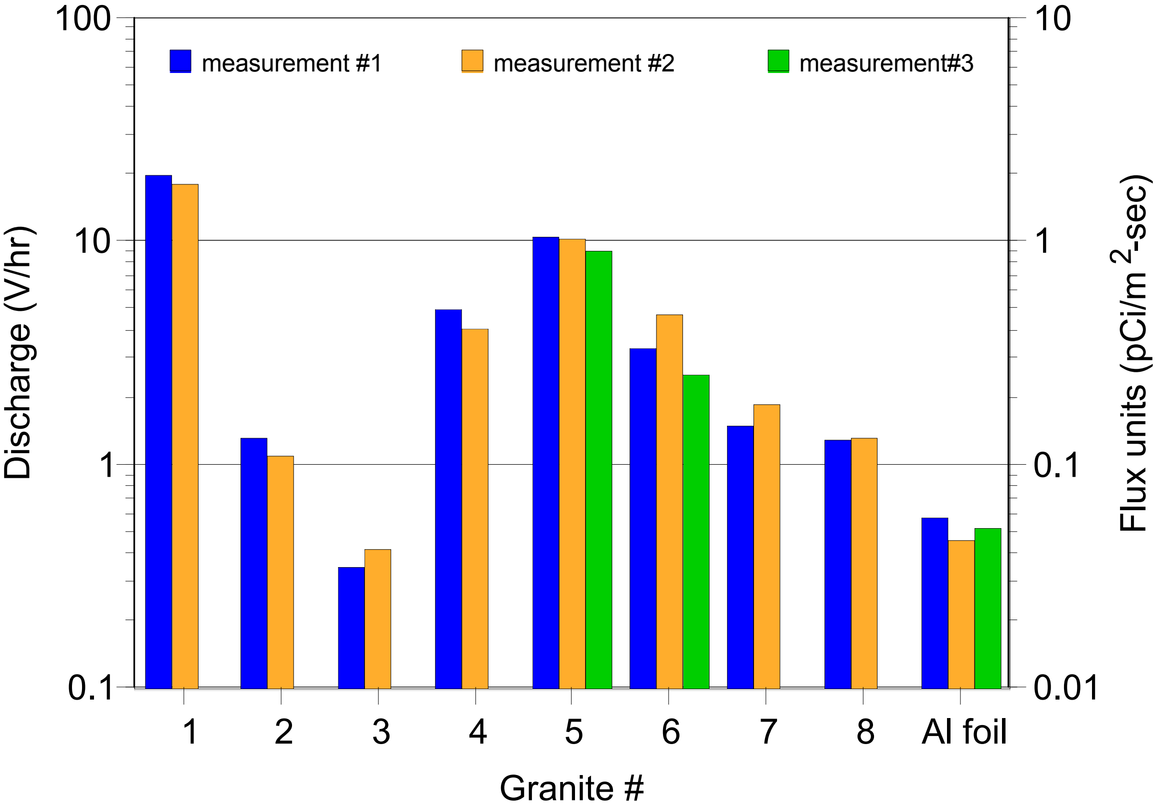
Figure 1.

Figure 2.

Figure 3.

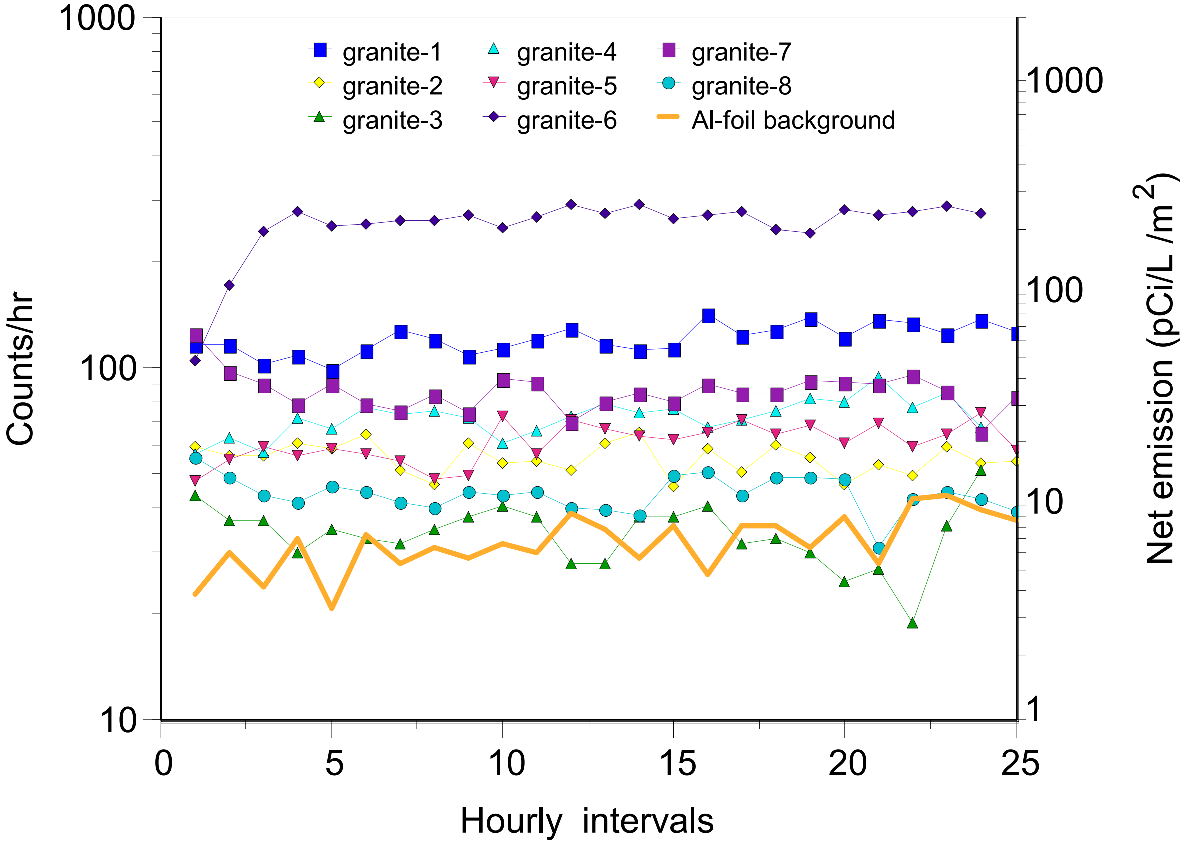
Figure 4.

Figure 5.



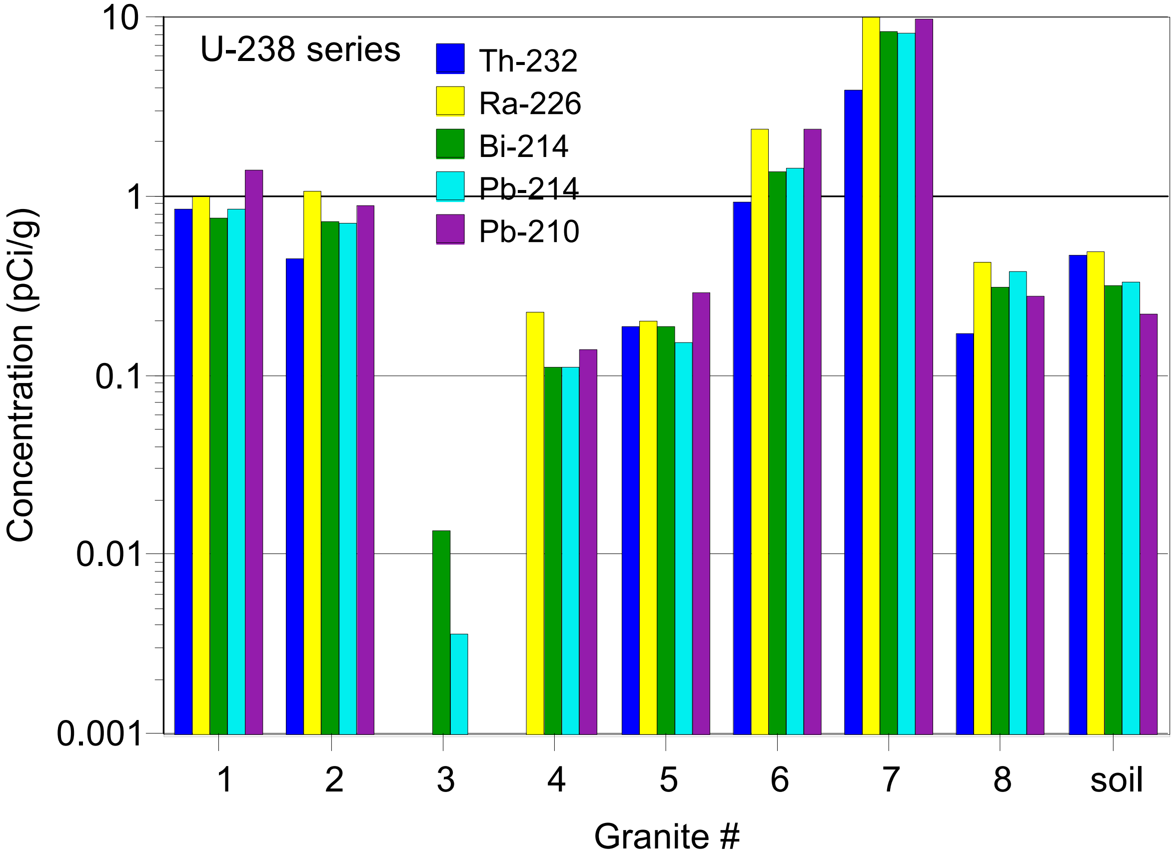
measurement #1
measurement #2
measurement#3
Discharge (V/hr)
100
10
1
0.1
Flux units (pCi/m^2-sec)
10
1
0.1
0.01
1
2
3
4
5
6
7
8
Al foil
Granite #

Figure 6.



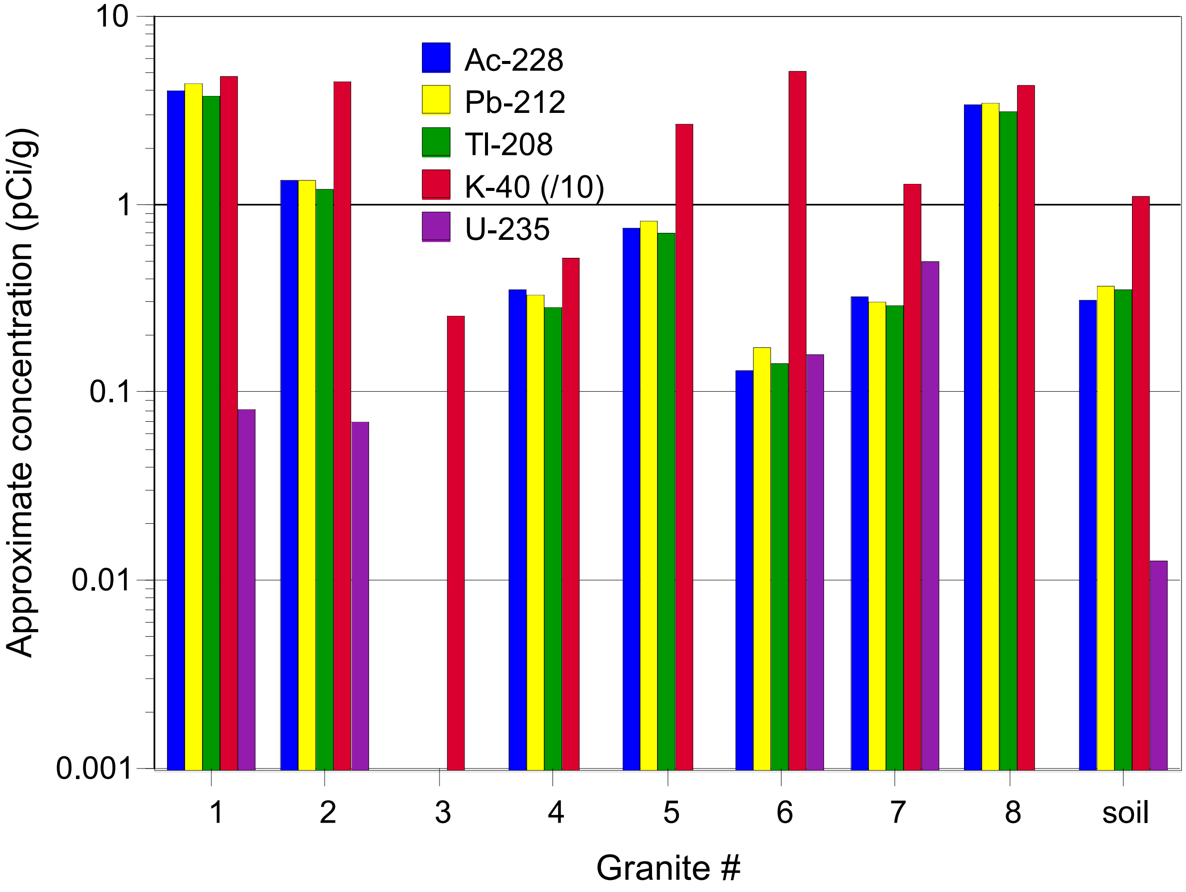
1000
100
10
Counts/hr
1000
100
10
1
Net emission (pCi/L /m^2)
0
5
10
15
20
25
Hourly intervals
granite-1
granite-2
granite-3
granite-4
granite-5
granite-6
granite-7
granite-8
Al-foil background

Figure 7.



U-238 series
Th-232
Ra-226
Bi-214
Pb-214
Pb-210
Concentration (pCi/g)
10
1
0.1
0.01
0.001
1
2
3
4
5
6
7
8
soil
Granite #

Figure 8.


Ac-228
Pb-212
Tl-208
K-40 (/10)
U-235
Approximate concentration (pCi/g)
10
1
0.1
0.01
0.001
1
2
3
4
5
6
7
8
soil
Granite #