PRE- AND POST-MARKET MEASUREMENTS OF GAMMA RADIATION AND RADON EMANATION FROM A LARGE SAMPLE OF DECORATIVE GRANITES

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ABSTRACT

Reports of radiation from granite counter tops generated public interest in the potential exposures to external gamma radiation and internal exposure from radon decay products created by building materials. The gamma radiation, radionuclide content and surface dose rates were measured in 322 slabs of 254 named stone types (area ~50 ft^2 per slab) and 14 smaller samples (area ~1 ft^2). Average surface gamma dose rates ranged from 1 to 24 μrem h^{-1} with hot spots up to 160 μrem h^{-1}. Radon emanation, measured for 60 slabs of 24 named stone types, ranged from 1 to 400 pCi ft^{-2} h^{-1}. Emanation fractions ranged from roughly 5% to 50%. Individual sampling points on the slabs showed emanation rates as high as 1300 pCi ft^{-2} h^{-1}. Model calculations suggest that some home occupants might receive doses that warrant remedial actions when substantial areas of some types of granite are installed in small, minimally ventilated living spaces. Based on these results, pre-market screening protocols have been developed and are being used to remove potentially problematic slabs from inventory. Post-market screening measurements are being tested in a pilot study of 35 homes. The maximum result radon emanation measured from granites in 11 houses was 90 pCi ft^{-2} h^{-1}.

INTRODUCTION

Media reports of radiation from granite counter tops recently generated public interest in this issue. While any building material derived from rock has the potential to create radiation exposures, some natural decorative stone like granite can have elevated concentrations of naturally-occurring radioactive materials (NORM). The ^{238}U family, and to a lesser extent the ^{232}Th family, create two pathways for exposure. In addition to external radiation dose from gamma rays, these families have a member which is a radioactive gas that can escape from the rock matrix into the environment. These gases, often referred to as radon and thoron respectively, decay into chemically active decay products that can deliver internal radiation doses when inhaled.

Customers buying decorative granite, and industry that supplies them, want an answer to a deceptively simple question, "Is it safe?" There are a number of underlying issues that help formulate an answer to that question. These finer points may not be of interest to everyone but can help support the answer if the questioner asks for details. For example, while public health officials might measure the "safety" using the average impact of the radiation exposures for the general population, individuals are usually more concerned about their

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[1] This work was supported, in part, by a grant from the CSB/SJU faculty development program.

own specific exposure. A product that might be deemed relatively safe in general, might not be judged so by a specific consumer whose use of the product and radio-sensitivity are out of the ordinary. Thus, a blanket statement of a product's safety needs to include the judgment of sensitive, yet reasonable consumers.

The following guiding philosophy for pre-market screening was developed through discussions with the president of Cold Spring Granite Co.[2] (CSG). We wanted to find and implement a procedure to protect public from granite countertops that might generate radiation exposures deemed unsafe or higher than normally encountered in nature. We wanted a reasonable consumer to feel confident that the product was safe when used in a "normal" application. That desire required the development of a cost-effective method to identify products that may (or may be perceived to) cause an excess radiation risk to the public, to test the CSG current countertop inventory using the identification and assessment methods, to apply the identification methods to all incoming stock of exiting stone types, and to apply the assessment method to new types of stones that may be added to the product line.

Since there are no comprehensive federal (US) regulations for NORM exposure, the recommendations of national and international radiation protection organizations were adopted as the reference safety standards. Those recommendations are based on the effective annual dose to an individual from controllable sources (HPS, 2009). In this work, the dose was calculated for an individual in a high, but not maximal, exposure scenario. In addition, the dose from an average or typical installation was calculated to estimate of the public health impact of decorative granite.

At present, over a thousand named "granite" stones are being used in the US as countertops, desktops, flooring and wall tile. The radiation characteristics of only a few dozen of these stones have been reported in the literature (Kitto, 2005, 2008, 2009; Brodhead, 2008) or been released to the public (EH&E, 2008). Based on those early reports, it is clear that many of the high radiation granites show substantial variation in their radiation characteristics. With the diverse needs of the natural stone industry and consumers in mind, the goals of this project were to substantially expand the number of stones analyzed for radioactive and assessed for radiation exposure. That required the development of measurement methods and predictive models suitable for "in situ" assessment of the external gamma radiation dose and internal radon-related dose for stones in the supply system (pre-market) or in the home (post-market).

MATERIALS AND METHODS

Two distinct sets of measurements and protocols were needed to fit the needs, environments, and capabilities of the stone industry and consumers. A higher level of technical skill and resources are available in the industry for assessing radiation potential of stones that have been processed but not yet customized and installed. However, they may

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[2] Cold Spring Granite Company 17482 Granite West Road Cold Spring, MN 56320

have to perform the tests more quickly than measurements in the home because of the large number of stones that may need to be tested.

Pre-market measurements and protocols

Companies that either quarry, process or import decorative stone generally have a central facility where the material can be measured while being processed, either in the polish line or at the receiving dock or warehouse. This environment suggests that an effective assessment might rely on a series of measurements that escalate from quick, simple screens to more complete investigation of radionuclide content or radon flux depending on the results of the screen tests and/or previous experience with the particular type of stone. This is the measurement and assessment approach described in this paper.

Granite slabs and samples

Decorative granite for countertop installations are usually quarried in large blocks and then cut and processed into slabs that are roughly 5 to 6 ft wide by ~10 ft long. While most slabs are 3 cm thick, some are as thin as 2 cm. Decorative granite for floor or wall tiles are usually 1 to 2 cm thick. One side of the granite is polished after being coated with a liquid polymer which is sucked into cracks and crevices by applying a pressure differential across the slab. Stones that are likely to crack are often covered with a polymer net that is glued to the non-polished side. At least one slab from every one of the 254 stone types in the 2008-09 CSG inventory was screened for gamma dose rate and radionuclide content during the period October 2008 to May 2009.

Smaller samples from fourteen stone types which were selected based on their radiation properties were also analyzed. The polished surface area of these samples was approximately 1 ft^2. Two of these samples from the current CSG inventory (CK08 and LD08) were from the same stone types that had been analyzed twenty years earlier (Steck, 1988). These stones had shown the highest radiation potential of the seven analyzed in 1988. One of the samples (CK88) was the actual stone that had been measured in 1988. The non CSG samples, all quarried outside North America, had been sent by concerned individuals because they displayed high radioactivity during screening tests.

Gamma dose rate screening

Simple gamma ray counters, usually scintillators or G-M detectors, provide the quickest and easiest radiation measurement that can be made. However they do not always give an accurate estimate of the actual effective gamma dose rate because the energy dependant response of the detector is usually different than tissue. Nonetheless, if proper precautions and calibrations are used, they can be useful for pre-screening granite. We used two simple scintillation survey counters; the Ludlum 12S micro R meter[3], and the Polimaster PM1703 [4] as quick pre-screening devices. These detectors are shown on the right in Figure 1. The Ludlum is in the right rear. These pre-screening detectors may not give an accurate assessment of the effective dose rate from a mixed source of radionuclides like granite. The naturally occurring materials (NORM) in stones emit radiation from three primary families; the uranium-radon (U-Rn) family, the thorium-thoron (Th-Tn) family, and ^{40}K. The energy

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[3] Ludlum Measurements, Inc. 501 Oak Street Sweetwater, Texas 79556 USA

[4] POLIMASTER Ltd. 112 M. Bogdanovich str. Minsk, 220040, Republic of Belarus

spectrum from granite generated in most scintillation materials is quite different from the spectrum from the source (^{137}Cs) that is normally used to calibrate the detector's response. One solution to this problem is to use a scintillation material whose absorption is similar to tissue like the Thermo Scientific (TS) Micro Rem Tissue Equivalent Survey Meter[5] shown in Figure 1 on the front-left. Another solution is to actually measure the absorption spectrum in the detector and use software and probe characteristics to calculate the effective dose rate like the Canberra Inspector 1000(In1k)[6] shown in the left-rear of Figure 1.

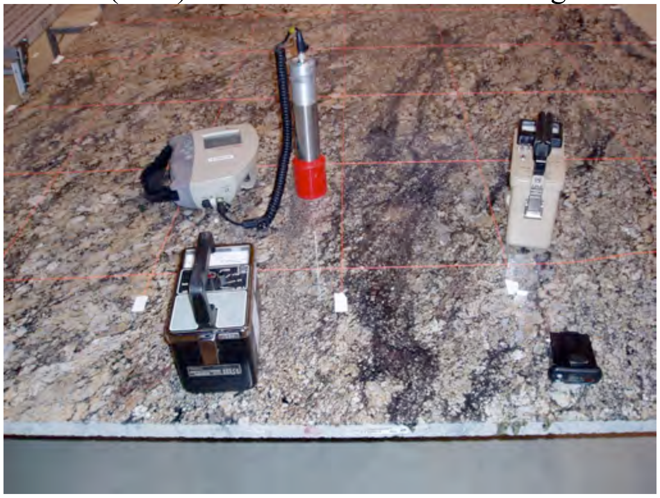

Fig 1 Scintillation-based gamma detectors used for pre-screening

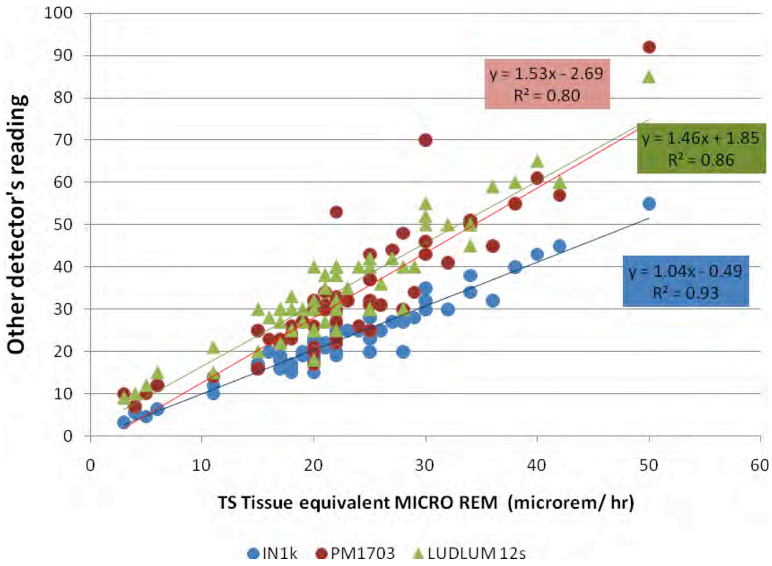

Fig 2 The relative response of three other instruments compared to the TS meter.

Figure 2 shows the relative response of these detectors when exposed side-by-side on the surface of the slab of Coral Gold granite shown in Figure 1. The correlation between various detectors is probably good enough for most field pre-screening tests. Some of the variation in response is due to the actual spatial variation of the gamma radiation within the area

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[5] Thermo Fisher Scientific Inc. 81 Wyman Street Waltham, MA 02454

[6] Canberra Industries, Inc. 800 Research Parkway, Meriden, CT 06450, U.S.A.

covered by the detector array. However, the slope of the response curves means that the readings from the Ludlum 12S and the PM1703 would need to be converted using their respective regression curves to get reasonable estimates of effective dose rates.

In this work, slabs were screened for gamma dose rate by placing either the TS or the IN1k in contact with a spot on the polished surface, waiting for roughly 12 s for the meter to respond before recording the reading. Since the detector on the surface "saw" an effective area of about 1 square foot, readings were taken on each slab at 50 locations in 1 foot intervals. Using this procedure, it took roughly 10 minutes to screen each slab. All dose rates reported in this paper are background corrected. In the slab warehouse the background was approximately 5 μrem h^{-1}(50 nSv h^{-1}).

Gamma dose rate spatial variation

The In1k was used to measure dose rates on the surface and in the space within 3 feet of two horizontal slabs; one slab that was radon rich and another slab that was thoron rich (shown in Figure 1). The grid points were separated by 1 foot from 0 to 3 feet in both horizontal and vertical directions except for a series of points 0.5ft above the surface and along the edges of the slabs. The grid point data was analyzed by SURFER® using a kriging procedure to give values at all grid points within 3 feet of the horizontal surface. These grid point values were analyzed to determine horizontal and vertical functions that represented the spatial variation. These functions were then used to integrate the dose over the space occupied by an individual whose exposure was being modeled.

Gamma radionuclide content

Spectroscopic screening measurements were made of the slabs using the In1k with a 2x2 stabilized NaI detector and Genie 2000 analysis software in an attempt to find a quick and inexpensive way to estimate the radon generating potential of granites. The hypothesis was that radon emanation was well enough correlated with the U family content in the material that it would serve as a surrogate for radon flux. Several approaches were used to calibrate the detection efficiency of the probe and geometry for field-grade (±20%) precision and accuracy in the radionuclide content averaged over the slab. The resolution of the probe limited the identification and analysis of many of the NORM peaks usually used in laboratory analysis. The highest energy peaks for each family were used to characterize the family's radionuclide content. Since these peaks belonged to radon and thoron progeny for the U and Th families, these results are labeled remnant radon, thoron progeny since some radon and thoron generated in the slab can escape before decaying to progeny. Given the short half-life of thoron, it is believed that little thoron escapes from deep in the granite so the thorium content determined from remnant thoron progeny should be a good approximation to the thorium family concentration. That situation may not hold for the radon progeny in all granites depending on the porosity, fracturing and coatings on the granite. The efficiency of probe-detector was calibrated with a multistep procedure. The detection efficiency was calculated for the radon progeny peak using a NIST-traceable radium standard at a variety of locations within a square foot surface location. Then the thorium peak efficiency was slightly adjusted by measuring sample CK88 whose radionuclide concentration had been determined by a NIST-standard calibrated HpGe spectrometer in 1988.

Gamma exposure models

In keeping with the radiation protection philosophy described above, the exposure of individuals to external gamma radiation was estimated using parameters that would lead to elevated, but not maximal doses. For example, one model assumed that the exposure to a seated home worker came from geometry where the highest point of radiation on the slab ("hot spot") was centered at the edge of his desk's work area. But the worker only spent 40 hours per week for 48 weeks at that desk. Thus, these estimates are only rough approximations and the uncertainty in their value is taken into account when comparing them to dose recommendations.

The actual exposure of individuals to external gamma radiation was calculated from a simplified model of the geometry of the person-source exposure conditions and the gamma dose field constructed from measured surface gamma dose rates as described above. Two exposure models were used to estimate the annual dose from countertops: (1) a kitchen worker who spent 4 hours per day within 3 feet of a horizontal countertop and (2) a home worker who spent 40 hours per week sitting near a desk top that had a hot spot at his work area. Two other models were used to assess the annual dose from floor and wall tiles in a room: (1) a worker standing on a granite floor for 8 hours per day, and (2) a sleeper lying 2 feet above the floor and 2 feet from a granite tile wall for 8 hours per day.

Radon emanation from slabs and samples

After all stone types had been screened for radionuclide content, those with the highest remnant radon progeny were selected for radon emanation measurements. Both the polished and unpolished sides were measured because they are both free to emanate in typical granite installations. A previous study had suggested that there might be different emanation rates from the different surfaces (Brodhead, 2008). Since a nearly vertical orientation was the most convenient way for slabs to be measured in the warehouse, emanation accumulators had to be designed to adhere to polished, netted, and rough surfaces. They also had to be radon leak proof. Figure 3 shows a typical set of emanation measurements being made on a polished and a netted surface.

Fig 3: Emanation measurements on slabs. The slab on the left is an example of a netting glued to the non-polished side.

At least seven locations were sampled on each slab; four or more on the polished side and three on the other side. At one location on the polished side, a continuous radon monitor[7]

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[7] Radon Scout SARAD GmbH Wiesbadener Straße 10 01159 Dresden GERMANY

(CRM) was enclosed by a 4.6L accumulator which covered 0.47 ft^2. At all the other locations, electret ion chambers [8](EIC) were enclosed by accumulators which were 3L stainless steel bowls which covered 0.43 ft^2. The sampled area was only about 5% of the total surface area of the slab. The smaller granite samples were measured with accumulators centered on the sample that covered 20 to 40% of the surface. A new integrated EIC-based radon flux monitor (RFM) was also used to measure the samples.

The flux can be calculated by the ingrowth of the radon in an accumulator measured by the CRM using equation 1:

$$C(Rn) = \frac{(FA)}{Vk}(1 - e^{-kT}) \tag{1}$$

where:

T is the accumulation time

F is the radon flux

A is the area of opening of the accumulator

V is the air volume of the accumulator

k *is* the effective loss rate of radon (The loss can come from decay, $7.55\text{x}10^{-3}$ h^{-1} , or leaks)

C(Rn) is the radon concentration at any accumulation time of T

Since accurate flux results require a known effective loss rate, numerous experiments were done to find a system that created a good, reproducible seal between the accumulator and the various surfaces on the slabs. While adhesive clay was acceptable for polished sides, the rough and netted sides required a quick-setting, adhesive along with the clay to hold and seal the accumulator on the surface. Figure 4 shows an ingrowth curve fit with a weighted least squares fit to determine the effective loss rate of radon for a leaky accumulator.

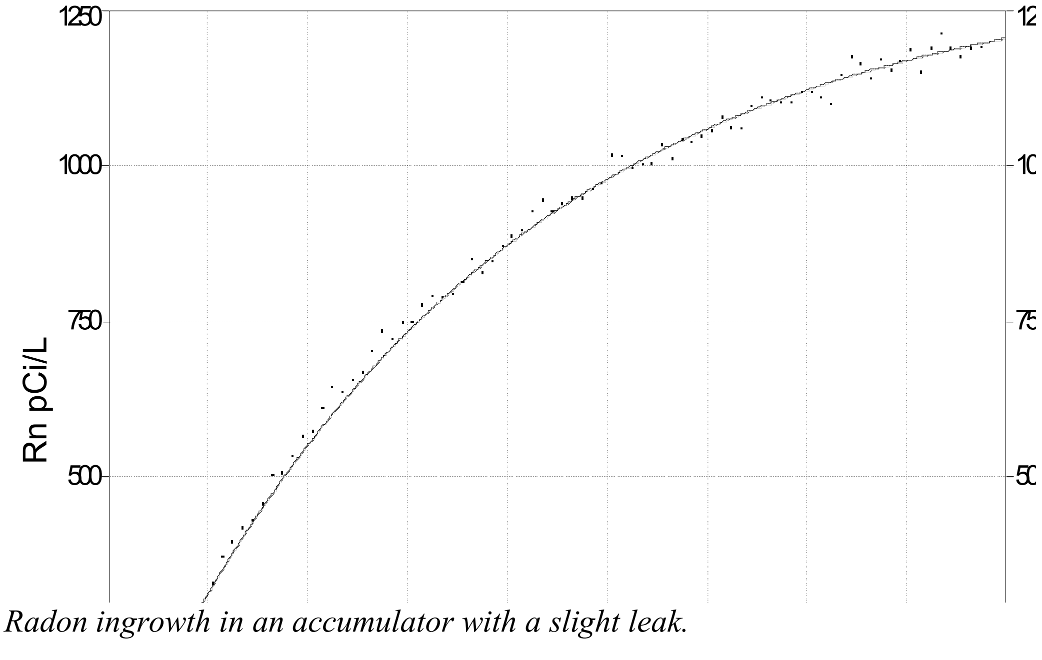

Fig 4 Radon ingrowth in an accumulator with a slight leak.

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[8] RAD ELEC Rad Elec, Inc. 5716-A Industry Lane Frederick, Maryland 21704

Once the effective loss rate is known, then the radon concentrations during the initial ingrowth can be corrected for loss and fitted to a straight line whose slope can be used to calculate the flux. Figure 5 shows a typical 22 hour ingrowth of a CRM-stainless steel bowl on the polished surface of a granite slab.

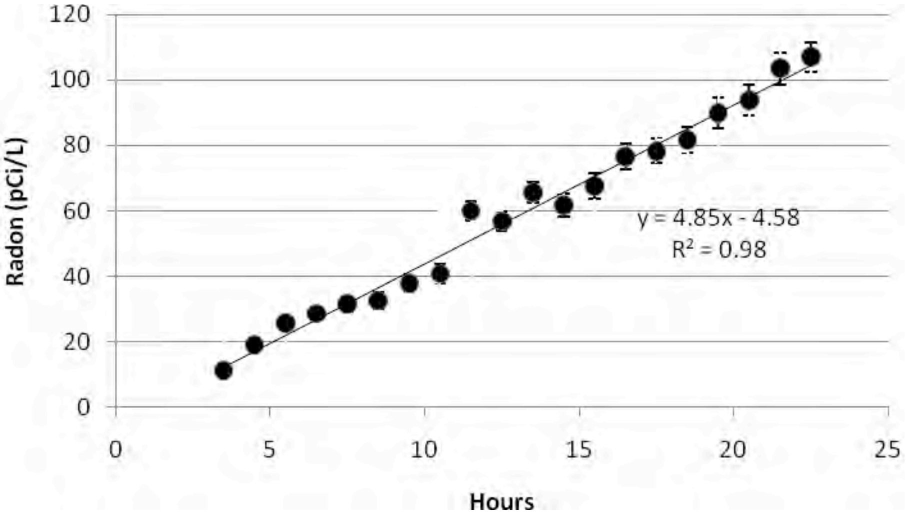

Figure 5: Typical ingrowth in a 5L accumulator attached to granite with the Radon Scout monitor. Slope used to determine emanation rate

Smaller samples can be measured more easily than slabs as the accumulator can enclose the sample and measure the emanation from all surfaces simultaneously. Granite sample emanations were measured in a well sealed 24 L aluminum case. Bowl style accumulators were used to measure the emanation from each side of each sample.

For integrating detectors like EICs or ATDs in accumulators, the flux can be calculated from the average radon using equation 2.

$$< C(Rn) >= \frac{(FA)}{Vk}\left[1-\left(\frac{1-e^{-kT}}{kT}\right)\right] \qquad (2)$$

where:

T is the accumulation time

F is the radon flux

A is the area of opening of the accumulator

V is the air volume of the accumulator

k is the effective loss rate of radon.

The loss can come from decay (7.55E-03 h^{-1}) or leaks

<C(Rn)> is the average radon concentration during the accumulation time of T

Each accumulator system was calibrated using NIST emanation sources (SRM 4974-8 and 4971-3) (Kotrappa 2005 Volkovitski, 2006). Additional details of the performance and calibration of the accumulator systems can be found in another paper from this Symposium (Kotrappa 2009A).

Radon Exposure and dose models

The high exposure scenario, called the conservative scenario, uses realistic estimates of the parameters like living space area, occupancy factor and ventilation rate. A simple model was used to estimate the radon concentration generated by granite used in a variety of ways indoors. The model assumed complete mixing of the source in the living spaces. The other important model parameters were taken from the EPA exposure factors handbook (EPA, 1997) to create a realistic but conservative estimate of exposures in a small (640 ft^2) minimally ventilated (0.35 ach) living space. The annual radon exposure was converted to effective dose using the dose conversion factors from UNSCEAR 2006 and typical high occupancy rates. The conversion was roughly 100 mrem for a year's exposure to 1 pCi L^{-1}. The annual dose was also calculated for a "more typical" modern home with an area of 3000 ft^2 and a ventilation rate of 0.2 ach to assess the public heath impact of granite in more common situations.

Post-market home measurements and protocols

The instruments and procedures used to assess the radiation impact of granite already installed in homes have to require less technical skill and be more cost effective than those in the pre-market environment. Simpler gamma dosimeters and emanation measurement systems were developed and tested so that "skilled" homeowners could deploy the detectors and make simple measurements.

A pilot study of a small number of selected volunteered homes is underway, testing procedures and measurement methods to see if they are practical and useful for assessing post-market radiation exposure from granite. Thirty-five homeowners, mostly from Minnesota, volunteered online to have their homes and granites tested for radon. The selection criteria were that the home was "small" and "tight" and had a substantial installation of "exotic granite". The measurement protocol included conventional radon in air measurements in the room where the granite was present, a room that was "remote" from any granite, and a basement, if the home had one. AirChek [9] short-term test kits were exposed for 4 days under closed house conditions. Landauer RADTRAK[10] detectors were simultaneously deployed for a 90 day (or longer) exposure under normal living conditions. A newly developed radon emanation measurement system was used to measure the radon emanation *in situ* at two locations on each type of granite in the home. The emanation system consisted of a high sensitivity radon-thoron discriminating track detector inside a radon-retaining tent that covered 1.8 ft^2. The track registration detector has been described earlier (Steck, 2006). The radon flux from an integrating detector like this can be obtained from the average radon concentration measured by the detector and equation 2.

The locations for emanation measurements on the granite surface were selected after the top surface was slowly scanned for gamma activity using the PM1703 meter. Emanation measurements were made at the location of the highest gamma reading and an "average" gamma location. The emanation systems effective loss rate (k) was measured using the Radon Scout and proved to be reasonably consistent. The track generation rate from radon

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[9] Air Chek, Inc. 1936 Butler Bridge Rd Mills River, NC 28759

[10] Landauer 2 Science Road Glenwood, Illinois 60425

was calibrated with a NIST SRM4794 source. The response of the thoron detector to thoron emanation has not yet been calibrated.

RESULTS

Pre-market summaries of slab measurements

Table 1 provides a summary of the pre-market slab measurement protocols.

Table 1: Summary of pre-market slab screening measurements and procedures

MEASUREMENT	INSTRUMENTS	Samples and time
Gamma Dose Screen **(GDS)**	TS microrem meter or Canberra Inspector 1000	322 slabs of 254 stone types (complete current inventory of stone types) at 50 sample locations separated by ~1 foot for about 12 s per location
Gamma Radon Progeny Analysis **(GRA)**	Canberra Inspector 1000+ Genie 2000 software	322 slabs of 254 stone types at 50 sample locations separated by ~1 foot for about 12 s per location; 5 minute post measurement analysis
Radon Surface Emanation **(RSE)**	RAD ELEC EIC in 3L accumulator or Radon Scout CRM in 5L accumulator	60 slabs of 24 stone types* at7 or more locations (4 or more on front, 3 on back) 3 to 4 square feet sampled for 24 hour

*22 of the stone types were suspected to have high radon potential based on screening tests and media reports

Table 2 gives the statistical summary of the gamma dose rate, remnant radionuclide concentrations, and radon emanation averaged across the slabs.

Table 2 : Statistical parameters for slab-average measurement results

MEASUREMENT (N)	Median	Range	Slab variation
Gamma dose rate(322)	4.3 μrem h^{-1} (43 nSv h^{-1})	1 to 24 μrem h^{-1} (10 to 240 nSv h^{-1})	10% median 0 to 75% range
Remnant radon progeny (322)	84 Bq kg^{-1} (2.3 pCi g^{-1})	<10 to 2300 Bq kg^{-1} (0.3 to 62 pCi g^{-1})	
Remnant thoron Progeny (322)	48 Bq kg^{-1} (1.3 pCi g^{-1})	<10 to 1300 Bq kg^{-1} (0.3 to 35 pCi g^{-1})	
Radon Surface Emanation (60)	62 pCi $ft^{-2}h^{-1}$ (24 Bq m^{-2} h^{-1})	3 to 300 pCi $ft^{-2}h^{-1}$ (1.2 to 120 Bq m^{-2} h^{-1})	36% median 0 to 120% range
Emanation fraction(60)	22%	3 to 59%	

Pre-market gamma radiation detail

Gamma Dose Screen (GDS)

Figure 6 shows the gamma radiation dose rates averaged across individual slabs. Individual sampling points ranged up to a maximum of 80 μrem h^{-1} (background subtracted).

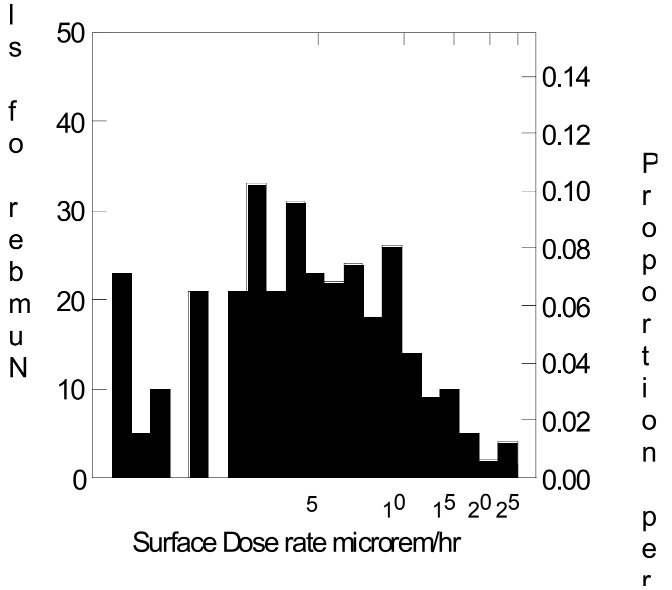

Fig 6 Measured gamma radiation dose rate averaged across the slab surface. (The number of observations per bar is shown in the left vertical scale while the fraction of the total observations is shown on the left vertical scale.)

Gamma Radon Progeny Analysis (GRA)

Figure 7 shows the average progeny concentration that remains in the slab for both radon and thoron. For the slabs as a group, remnant radon progeny averaged 210 Bq kg^{-1} (5.7 pCi g^{-1}) and ranged up to 2300 Bq kg^{-1}(62 pCi g^{-1}). Remnant thoron progeny averaged 90 Bq kg^{-1} (2.4 pCi g^{-1})and ranged up to 1300 Bq kg^{-1}(35 pCi g^{-1}).

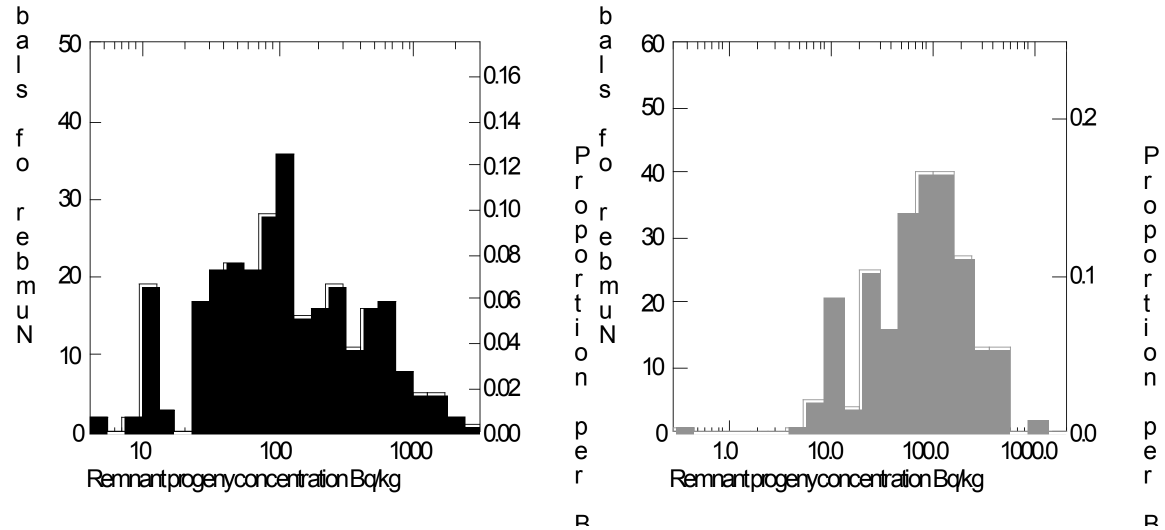

Fig 7 Remnant progeny concentrations measured in slabs: radon (left) and thoron (right).

Figure 8 shows the fraction of the dose created by each NORM family using the coefficients for the contributions to dose by family found in the European Commission report 112 (EC 1999) and the individual concentrations measured for individual slabs. It is worthwhile to note the substantial number of slabs where radon progeny make up the bulk of the dose and the substantial number where the radon progeny contributes little. The average contribution by family are: U/Rn 33 %, Th/Tn 24 %, K40: 43 %.

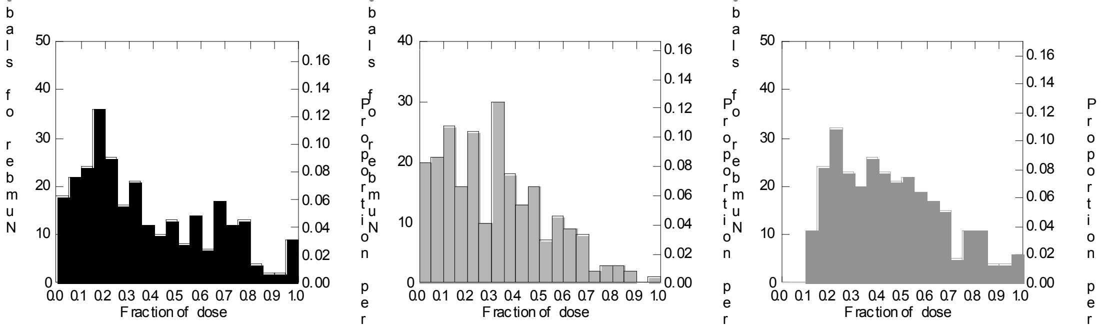

Fig 8 Relative contributions to surface dose from NORM families ^{238}U, ^{232}Th, and ^{40}K

Pre-market gamma dose rate spatial variation and exposure

Figure 9 shows the results of an analysis of the measured spatial variation of the background subtracted gamma dose rate around a slab of Coral Gold.

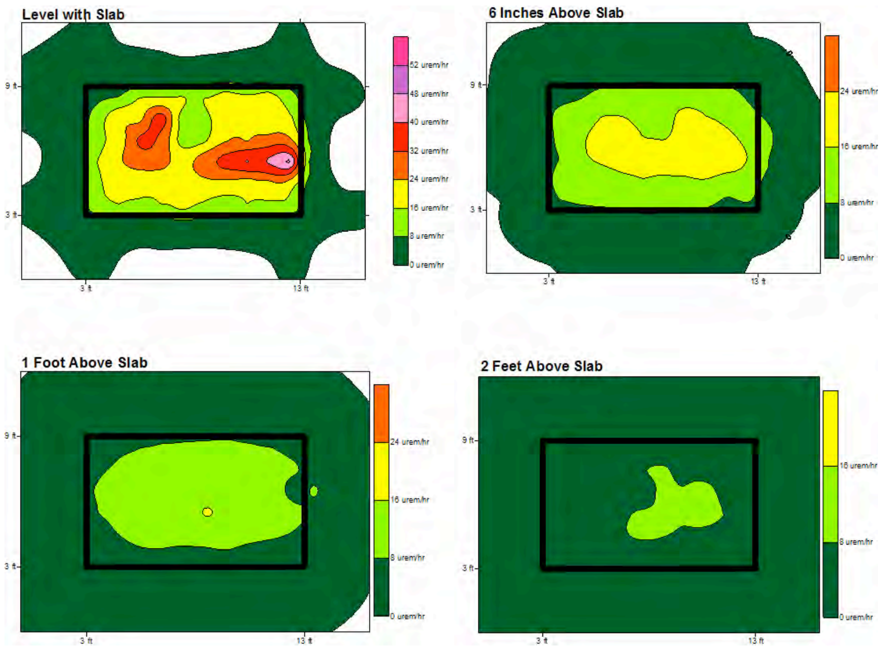

Fig 9 Gamma dose rate contour maps of the in the vicinity of the Coral Gold slab (Figure 1)

Preliminary exposure analysis using this distribution as a template suggests that a countertop would have to have an average surface gamma dose rate of 100 μrem h^{-1} above background to exceed the exposure recommendations for the kitchen worker. In the desk worker exposure scenario, the hot spot would have to exceed 60 μrem h^{-1}. In the case of exposure to floor and wall tiling installations, the safe average surface dose rates are lower; 33 μrem h^{-1}

for standing-floor scenario, 29 μrem h^{-1} for the sleeping – floor scenario, and 13 μrem h^{-1} for the sleeping-wall plus floor scenario.

Pre-market Radon Surface Emanation (RSE)

The median value for the slab-average radon emanation from the 60 slabs measured was 24 Bq m^{-2} h^{-1} (62 pCi ft^{-2} h^{-1}). Fluxes ranged from 1 to 117 Bq m^{-2} h^{-1}(3 to 300 pCi $ft^{-2}h^{-1}$). Figure 10 shows the flux distribution of the sample set.

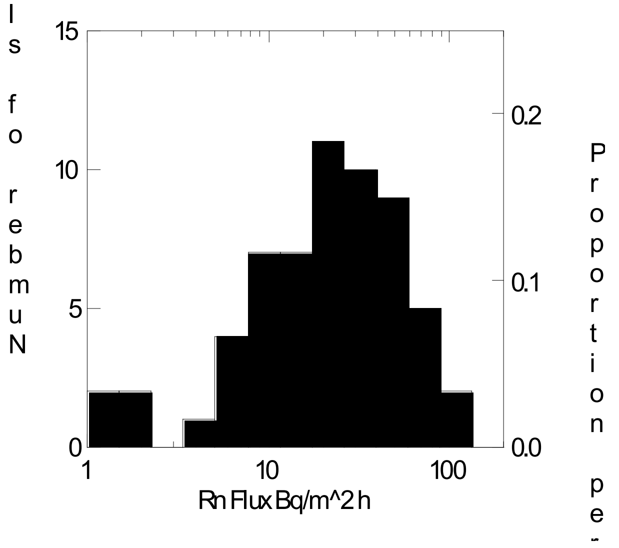

Fig 10 Slab average radon flux distribution for the 60 slabs

The radon emanation fraction can be calculated from the measured radon flux and the remnant radon progeny determined by gamma spectroscopy as described in the Methods section. The emanation fraction distribution had a median value of 22% with a range from 3 to 59%. For individual slabs, the flux variation across the polished side showed a median variation of 60% as did the variation across the unpolished side. But the flux often was quite different from the polished to the unpolished side. In particular most slabs that had netting "glued" to the unpolished side usually showed fluxes below the detection limits. But 4 out of 22 such slabs had almost equal flux on both sides. Similarly, while most slabs with untreated (rough) backs had about equal emanation from each side, in 5 out of 38 cases the polished had a higher flux than the rough side and in 3 cases the opposite was true.

A small study of the flux dependence using combinations of 1 cm thick tiles suggests that the flux scales better with mass than surface area up to a thickness of 5 cm.

The distribution of annual effective radon-related doses from the 60 slab sample is shown for the conservative scenario in Figure 11for three different kinds of installations. The conservative model's median dose for these types of granites is 17, 30 and 40 mrem for the countertop, floor, and floor plus wall installations while the maximum dose is 83, 150 and 200 mrem respectively.

In an exposure scenario that is believed to be more representative of typical modern homes, those same doses are lower as illustrated in Figure 12. The median doses in these cases are

6, 11 and 30 mrem for the countertop, floor, and floor plus wall installations while the maxima are 30, 57, and 150 mrem respectively.

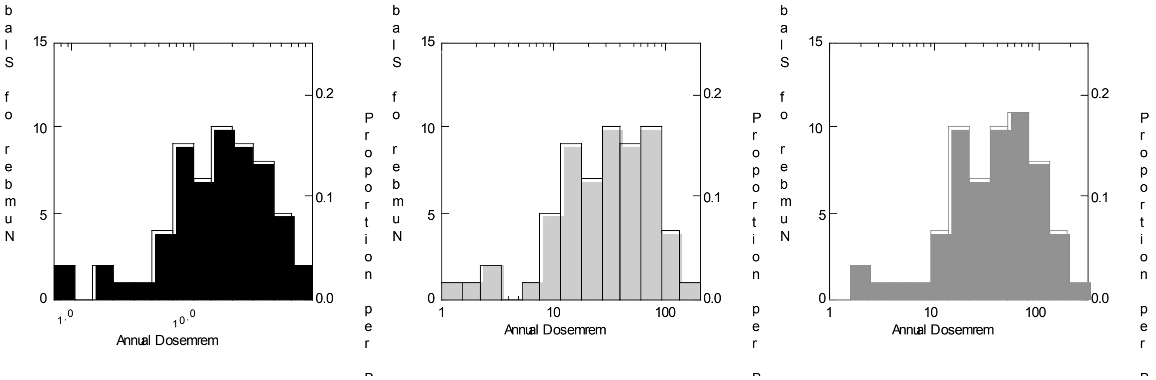

Fig 11 Effective annual doses due to radon from countertops, floor, and floor and walls in the conservative exposure case

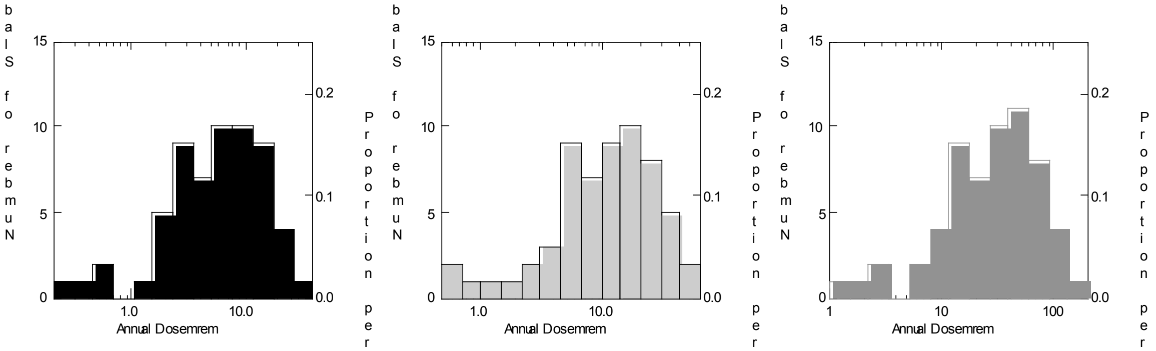

Fig 12 Effective annual doses due to radon from countertops, floor, and floor and walls in a more typical exposure case.

Pre-market small sample results

The smaller area granite samples show a wider range of those same radiation characteristics since most of them were selected because they had elevated gamma emissions. Two of the stone types (CK and LD) were selected because they are more representative of the characteristics of the majority of granites. Table 3 summarizes the major radiation characteristics of these 14 samples. The first three samples from the same slab. The next two are samples from the same slab of another stone type.

Table 3: radiation properties of granite samples

Sample[§]	Surface dose rate nSvh^{-1} (µrem h^{-1})	^{222}Rn emanation[†] Bq m^{-2} h^{-1} (pCi ft^{-2} h^{-1})	^{222}Rn emanation[†] %	^{222}Rn progeny[*] Bq kg^{-1} (pCi/g)	^{220}Rn progeny [*] Bq kg^{-1} (pCi/g)
FS09[‡]	1600 (160)	125 (320)	2	17000 (460)	30 (1)
FS08[‡]	200 (20)	120 (300)	17	1900 (51)	300 (8)
FSMK[‡]	160 (16)	60 (155)	12	1600 (43)	270 (7)
JBMK[‡]	1400 (140)	160 (420)	11	5800 (157)	900 (24)
JBSR[‡]	600 (60)	170 (440)	10	5000 (135)	500 (14)
JDJF	400 (40)	60 (150)	7	2500 (68)	500 (14)
NG08	400 (40)	38 (96)	2	7000 (190)	200 (5)
CB08	200 (20)	50 (130)	8	2000 (54)	200 (5)
JB08A[‡]	160 (16)	35 (90)	8	1400 (38)	200 (5)
JB08[‡]	100 (10)	30 (76)	11	800 (22)	200 (5)
SUMK	60 (6)	6 (16)	14	140 (4)	570 (15)
LD08	40(4)	4 (11)	16	110 (3)	100 (3)
CK08	30(3)	6 (16)	25	60 (2)	70 (2)
CK88	30(3)	0.5 (1.4)	3	70 (2)	70 (2)

[§] Cut from slabs or floor tiles; area ~ 0.1 m^2 (1 ft^2) x 2 or 3 cm thick

[†] Average of both polished and unpolished sides

[‡] Separate samples from the same slab

[*] Remnant concentration in an open, one square foot sample, measured at the center

Table 4 shows the radon flux from some of these samples as measured by different emanation systems. While a more thorough discussion of the implications of these measurements can be found in another paper in these Proceedings, this data illustrates the differences in flux between the sides of the different stones (Kotrappa, 2009A). Both the CB sample and the FS samples (same slab) show atypical emanation in that there is a significant difference in emanation from polished to the rough side of the slab. The JB (Juparana Bordeaux) samples are from the same stone type but different slabs. Even though they are both netted on the back side, the emanation is low from the netted side in JB08 but equivalent to the polished side in the case of JBMK.

Table 4 Comparative radon flux results (in pCi ft^{-2} h^{-1}) from both sides of selected samples using a variety of measurement systems.

Method	Enclosure +CRM	5L Bowl +CRM	3L Bowl +EIC	EIC Radon Flux Monitor
Sample/surface				
CK08				
polished		16	11	18
rough		13	21	21
all surfaces	16	15*	16*	20*
CB08				
polished		100	90	70
rough		260	250	270
all surfaces	155	180	170	170
FS08				
polished		30	30	25
rough		450	510	525
all surfaces	310	240	270	275
FSMK				
polished		42	46	52
rough		215	230	370
all surfaces	160	129	138	211
JB08				
polished		120	105	160
net		1	10	8
all surfaces	80	61	58	84
JBMK				
polished		495	430	829
net		365	290	280
all surfaces	415	430	360	555

* *The cells shaded grey are averages of the two cells above.*

Post-market radon and radon emanatation in homes

Only short-term radon measurements are currently available for the complete set of 35 upper Midwest homes. To date, the radon emanation from granites in 11 houses have been measured. The average emanation rate was 10 pCi ft^{-2} h^{-1} (4 Bq m^{-2} h^{-1}), the median was 5 pCi ft^{-2} h^{-1} (2 Bq m^{-2} h^{-1})and the maximum was 90 pCi ft^{-2} h^{-1} (35 Bq m^{-2} h^{-1}).

Table 5 Short-term average airborne radon concentrations in homes with decorative granite.

Location	*Average Radon (pCi/L)*	*Median Radon (pCi/L)*
Rooms with granite	2.7	2.0
Rooms remote from granite	2.3	1.6
Basements	4.6	3.8

DISCUSSION

The central focus of this work was to find practical methods of identifying granite installations that had the potential to generate radiation doses above recommended levels. In light of the significant variation and uncertainties associated with the factors that control NORM dose assessment to individuals, conservative scenarios of the effects of granite installations that produced effective doses less than 25 to 50 mrem yr^{-1} were deemed acceptably safe (HPS 2009). This choice helped frame acceptable measurable surrogates and models for the dose for the external gamma and internal radon progeny exposure pathways. Thus, the radiation protection goals are converted to finding easily measured surrogates that can be used in verifiable dose construction models.

External gamma dose

For external gamma radiation, one candidate for a practical surrogate is the radionuclide concentration distribution of the granite installation. The gamma dose field surrounding an installation can be calculated from the measured radionuclide concentration distribution in the installation and a spatial source - transport model that calculates the dose field in the vicinity of the material. The European Commission (EC, 1999) takes this approach by using the average concentration of the NORM families in a material to construct a dose index. The index is actually the predicted annual effective dose (in mSv) for an individual in the center of a room constructed of the NORM material. For thin claddings of the material, an index of 2 (corresponding to a dose of 30 mrem yr^{-1}) is supposed to trigger a detailed dose assessment of the actual geometry and use of the material. Figure 12, which shows the distribution of the index calculated for the material in CSG slabs shows that a significant number exceed 2 and the maximum index is above 9. Of course, most decorative granite installations will have lower doses because they involve less material than complete surface coating, and in the case of counter tops, have different exposure geometry. An investigation using this approach for the granites measured in this work is ongoing.

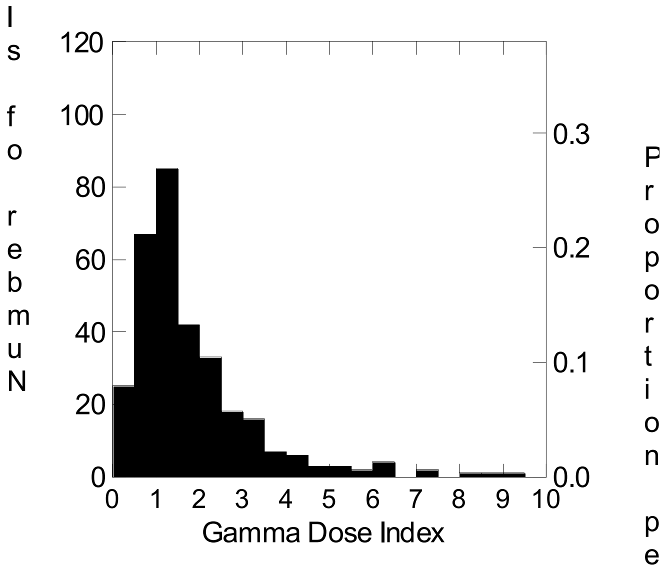

Fig 12: Gamma dose index calculated from NORM concentrations measured in 300 slabs. (The dose index corresponds to the annual dose (in mSv) under a hypothetical exposure scenario.)

Another candidate for a practical external dose surrogate is the measured gamma dose rate distribution on the slab's surface. These measurements can be used with a spatial dose field model derived from spatial variation measurements of the dose field near granite slabs. Figure 9 (above) illustrates that the dose falls off rapidly away from the slab vertically and horizontally, especially in the plane of the slab. Two slabs' dose rate fields have been studied so far. This approach is currently being verified on a third slab. Preliminary results suggest that none of the 300 granite slab materials measured would exceed recommended dose limits when used as a countertop or as flooring. Only two slabs had hot spots that would exceeded the limit when used as a desktop, but 14 granite slabs would have exceeded the limit if a bedroom floor and walls had been clad with that granite. However, since floor and wall tiles are usually one third the thicknesses of slabs, none of those materials in thinner tiles would have exceeded the recommendations. But, had the selected granite samples listed Table 3 been used as surface tiles, two materials would have exceeded the limit for floor installations and 4 would have exceeded the limit for floor plus walls.

Internal radon-related dose

For the internal dose from radon progeny, the "safe" reference effective dose can be nearer the upper end of the range described above, roughly 40 to 50 mrem y^{-1}. That range corresponds to the dose expected from the lowest practically-achievable indoor radon concentrations, those equivalent to outdoor air concentrations. Of the 60 slabs that were measured for radon emanation, 8 would exceed the reference value for countertop installations, 25 for floor installations, and 34 for floor plus wall installations. Of the smaller granite samples, 2, 5 and 6 of the 14 would exceed the reference levels for the different installations respectively.

From an operations perspective, it would be helpful if the external dose surrogate could be used for the internal dose as well. Unfortunately, neither surface dose rate nor radionuclide concentration is well enough correlated with the radon emanation rate to serve as a universal surrogate for internal dose. The radon that escapes from a slab depends on the parent nuclide (radium) content and the fraction of radon generated in the slab that can escape. This latter characteristic depends on the location of the radium bearing minerals in the slab and the physical porosity or fracturing on the slab. These characteristics can lead to large differences in radon emanation fractions from stone type-to-type and in some cases from slab-to-slab within the same type. Figure 13 illustrates one type of spatial variation where the radium bearing minerals are near the surface concentrated in a visible small spot. These "gamma hot spots" often, but not always, corresponded to radon flux "gushers".

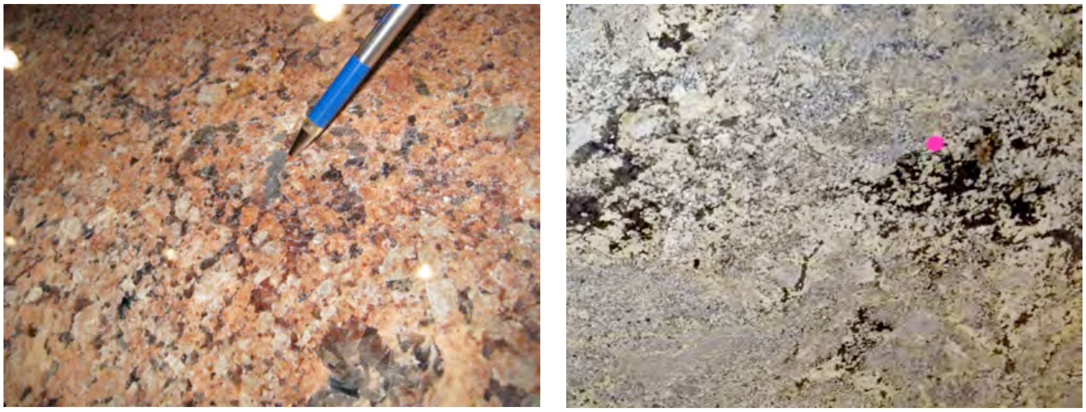

Fig 13: Small "hot spots" of mineralization; grey mineral in Juparana Bordeaux on left, and yellow mineral in Niagara Gold on right

Gamma dose as a radon flux surrogate

Figure 14 shows the surface gamma dose rate and radon flux at the ~450 points sampled on the 60 slabs measured for emanation. Recall that these slabs were pre-selected because they were high in remnant radon progeny, not on total gamma dose rate. The orange line shows the maximum radon flux that would produce internal doses at the recommended level for a counter top application in our conservative scenario (~ 60 Bq m^{-2} h^{-1}or 150 pCi ft^{-2} h^{-1})). Besides the poor correlation, surface dose rates would not make a good diagnostic statistic as plenty of examples of high emanation and low gamma dose as well as low emanation and high gamma dose are evident. For example if you chose a gamma dose rate trigger of 30 µrem h^{-1} for the hot spots, 8 areas would be classified falsely as "unsafe" and 36 would be classified falsely as "safe"

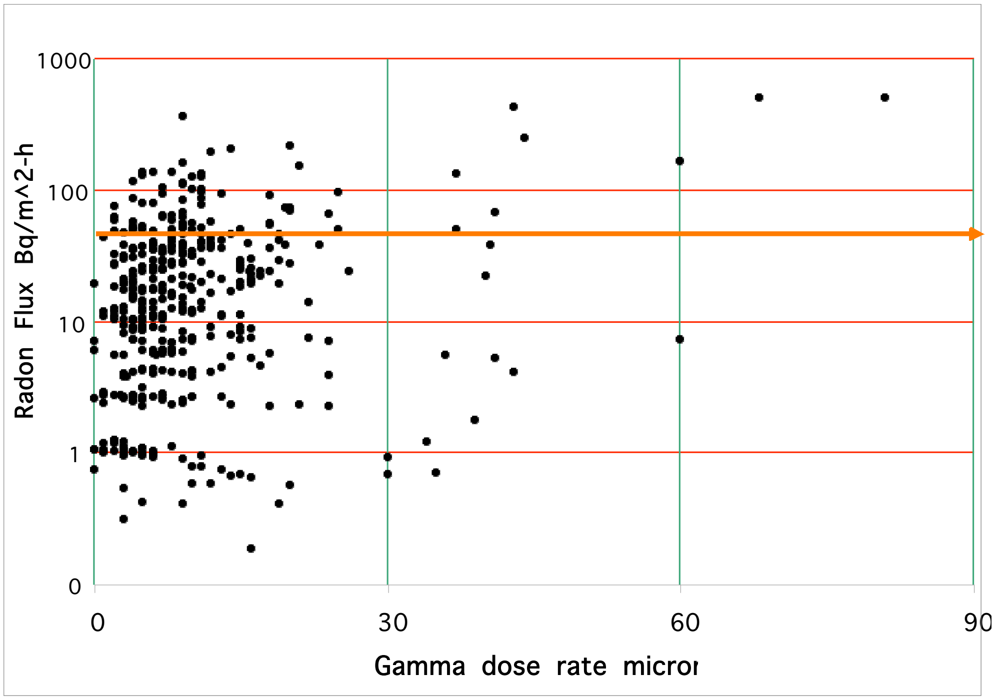

Fig 14 Radon flux and radon emanation at over 400 points on 60 slabs and 14 samples

Other combinations of gamma dose and radon flux, such as the hot spot gamma dose versus slab average emanation shown in Figure 15, suggest that surface gamma dose alone is not an adequate radon flux surrogate. For countertop applications (orange line), a gamma dose rate trigger of 30 μrem h^{-1}, causes 2 false positives and 3 false negatives in the sample of 60. The trigger would have to be lowered to ~ 10 μrem $h^{-1}$to eliminate false negatives. But that trigger would create almost 25 slabs to be falsely classified as "unsafe". The situation is exacerbated when more extensive granite applications are analyzed. The yellow and violet lines on Figure 15 show the flux limits that would cause radon-related dose to exceed the recommendation in the case of floor tile, and floor plus wall tile exposure scenarios.

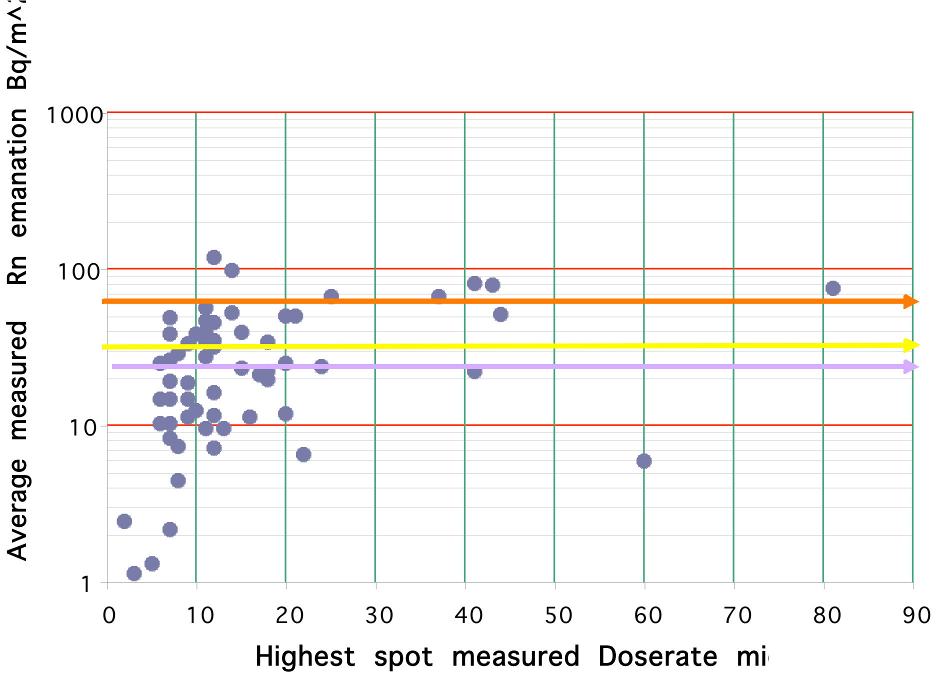

Fig 15 Slab average radon flux and maximum surface gamma dose for 60 slabs. The orange, yellow, and violet lines show the limits for "safe" countertop, floor, and floor plus wall applications.

Figure 8 (results section above) suggests one reason for the failure of surface gamma dose rate as a good surrogate for radon exposure; the remnant radon progeny contribution to the gamma dose has a significant number of low and high fractional contributions to the surface gamma dose rate.

Remnant radon progeny concentration as a radon flux surrogate

It has been suggested that remnant radon progeny content in the slab would be better correlated with radon emanation. Advanced portable gamma spectrometers like the Canberra IN1k, can determine the radionuclide content of NORM materials in a slab with elevated concentrations within a few minutes. These instruments are affordable and require only modest technical skill once they are calibrated so they are a reasonable alternative for large companies compared to the expense of a facility for measuring slab radon emanation.

Figure 16 shows the relationship between the slab average remnant radon progeny and the radon flux along with flux limits for various dose-exposure scenarios. While the correlation between radon flux and remnant radon progeny is better than surface dose rate, there are still enough variations to require additional measurements for some stone types. For countertop applications, slabs with remnant radon concentrations below about 300 Bq kg^{-1} would fall consistently in the "safe for counter tops" category. Figure 7 (results) shows that most of the CSG-inventory slabs (75%) meet this condition. The utility of this surrogate can be extended when radon emanation measurements are made on a number of samples of a particular stone which have fluxes near the limits. The trigger limit can be lowered for most stones whose emanation fraction has been repeatedly measured because their emanation fraction is likely to be smaller than the 50% used in setting the *a priori* limit.

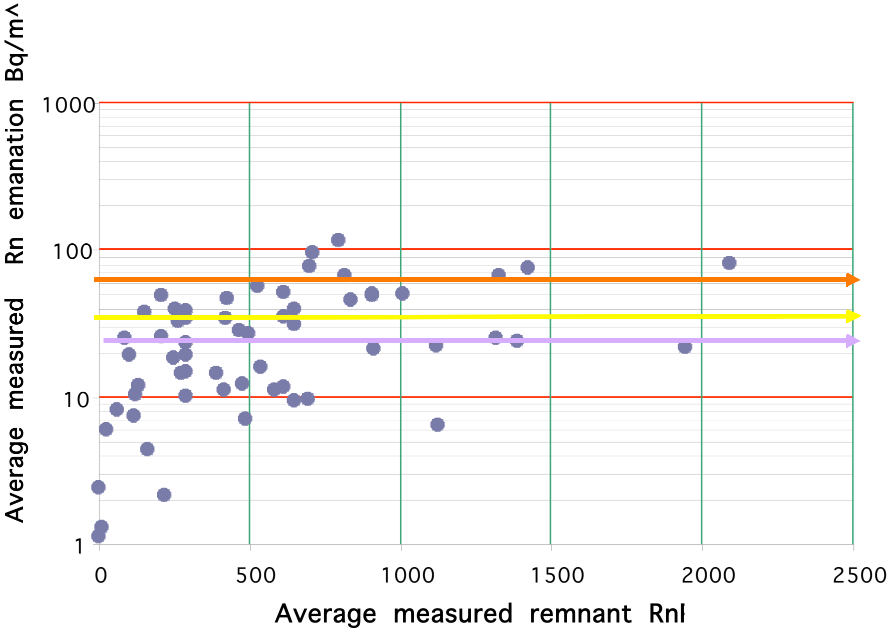

Fig 16 Slab average radon flux and average remnant radon progeny for 60 slabs. The orange, yellow, and violet lines show the limits for "safe" countertop, floor, and floor plus wall applications.

The radon progeny trigger level for limiting concentrations in floor tile, and floor plus wall tile exposure scenarios would be 150 and 110 Bq kg^{-1} respectively. Clearly many more stones destined for surface tile applications require radon emanation measurements.

Practical pre-market screening protocols

The pattern of flux values, remnant radon progeny concentrations and their variations can be combined in a classification scheme to screen existing and new inventory at the processing, distribution or fabrication level. The key elements are multiple measurements of the radionuclide content of a number slabs per stone type repeated over time as the quarried material is taken from different sections of the deposit. An historical database of radon progeny concentrations and radon emanation fractions for potentially troublesome stones would allow for more efficient and effective screening of inventory. Those stones with consistently low remnant radon progeny (exact value would depend on their use in the home and prior emanation measurements) can be sampled less frequently and less intensively. These slabs would have a trigger value on the surface gamma dose which, if exceeded, could indicate that remnant radon progeny was above the trigger level for emanation tests. When the dose trigger value is exceeded, the remnant radon progeny needs to be measured, and if the measured remnant progeny exceeds the emanation trigger level, radon emanation measurements need to be made or the slab kept out of inventory.

In this work, which focused primarily on slabs used as counter tops, the radon flux was measured for three slabs of each stone type that had remnant radon progeny greater than 300 Bq kg^{-1}. These measurements helped establish the average value and variation of the emanation fraction for each stone type. Those characteristics were used to calculate a trigger value for the remnant radon progeny of future measurements on that stone type. If the new measurement exceeded the trigger value, then the radon flux had to be measured for that slab or it would be rejected from inclusion in the inventory. This pre-market screening protocol is now being used by Cold Spring Granite to insure the safety of their counter top inventory.

CONCLUSIONS

Most decorative granites create acceptably low radiation exposures in most home installations. However, some stones should not be used in large scale installations in small living spaces with low ventilation because they generate enough radon to create doses in excess of those recommended by radiation protection organizations for controllable radiation sources. Those stone types that may create excessive internal radiation dose cannot be identified based on gamma measurements alone. In particular, the surface gamma dose rate which is easily measured with survey instruments is ineffective in screening out all potentially troublesome stones and falsely identifies others as troublesome. However, a system of increasingly sophisticated screening measurements and trigger values combined with more extensive analysis of the radon generating potential and radionuclide content of stone types can effectively insure the safety of decorative granite made available to the public.

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