ASSESSING EXPOSURE TO RADON AND RADIATION FROM GRANITE COUNTERTOPS

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LIST OF ABBREVIATIONS AND ACRONYMS

^{208}Tl	thallium-208
^{212}Pb	lead-212
^{214}Bi	bismuth-214
^{214}Pb	lead-214
^{222}Rn	radon-222
^{226}Ra	radium-226
^{228}Ac	actinium-228
^{232}Th	thorium-232
^{234m}Pa	proctactinium-234m
^{234}Th	thorium-234
^{238}U	uranium-238
^{40}K	potassium-40
ACH	air changes per hour
ACI	Activity Concentration Index
Bq kg^{-1}	becquerels per kilogram
Bq m^{-2} h^{-1}	becquerels per square meter per hour
Bq m^{-3}	becquerels per cubic meter
CONTAM	multi-zone indoor air quality model
C_{sat}	saturated radon concentration
EC	European Commission
EMSL	EMSL Analytical, Inc.
EPA	U.S. Environmental Protection Agency
Flux	becquerels of radon released per square meter per hour
GM	Geiger-Müeller
h^{-1}	per hour
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
k	total radon loss
keV	kiloelectron volt
m^2	square meter
m^3	cubic meter
m^3 sec^{-1}	flow rate units of cubic meters per second
MIA	Marble Institute of America
mSv	millisievert
mSv a^{-1}	millisievert per annum (year)
NCRP	National Council on Radiation Protection & Measurements
NIST	National Institute of Standards and Technology
NRC	Nuclear Regulatory Commission
pCi L^{-1}	picocuries per liter
U.S.	United States
µRem	microRem
µRem h^{-1}	microRem per hour

1.0 EXECUTIVE SUMMARY

1.1 OVERVIEW

Environmental Health & Engineering, Inc. has completed a detailed evaluation of potential human health risks of naturally occurring radioactive materials in granite countertops. The extensive measurements and rigorous mathematical modeling conducted to date indicate that (i) external doses of ionizing radiation emitted from granite countertops are well below levels that would pose a health concern and (ii) contributions from granite countertops to radon levels in homes are lower than background levels of radon exposure typically found outdoors and indoors.

1.2 BACKGROUND AND OBJECTIVE

Both radon gas and low levels of ionizing radiation are natural constituents of the environment that we are exposed to every day, with most of our exposure arising from soil, cosmic and internal sources. The elements that produce radon gas and ionizing radiation are of course natural and are minor constituents of many common building materials such as concrete, brick, gypsum and natural stone. To address any potential health risk associated with the use of granite countertops in home settings, the Marble Institute of America (MIA) commissioned Environmental Health & Engineering, Inc. (EH&E) to design and carry out a series of experimental studies required to characterize radiation exposure and risk quantitatively.

The overarching question addressed by EH&E was whether granite countertops significantly increase exposures and doses of radiation in homes. A comprehensive study was designed to answer this question in a scientifically valid manner. The in-depth analysis focused on risks of exposure to radon gas and ionizing radiation associated with the majority of types of granite used as countertops in the United States.

1.3 METHODS

To achieve these aims, numerous measurements were made in a systematic manner from multiple points on many varieties of stone sold as granite countertop. The testing protocol included (i) determining pertinent gamma activity concentrations, (ii) measuring total alpha, beta, and gamma emissions, (iii) measuring total external radiation dose; and (iv) determining radon flux from a wide range of representative stones. Results of the measurements were input into widely-accepted mathematical models to estimate annual doses of ionizing radiation and exposure to radon for a number of typical, realistic residential scenarios. To characterize potential radiation risks of granite countertops, the estimates of dose and exposure were compared to a variety of health-based benchmarks for radiation safety published by authoritative organizations charged with protection of public health such as the National Council on Radiation Protection and Measurements (NCRP).

Over 400 samples from 115 varieties of stone were evaluated that comprise approximately 80 percent of the annual U.S. market share for granite countertops, based on the most recent sales data available. Thirty-nine different slabs of countertop, representing 27 unique varieties of stone, were evaluated for emissions of radiation and radon on a full-slab basis. To increase the scope of the analysis, radon emissions were determined for one to nine discrete samples for each of another 88 types of stone plus 24 of the stone types for which full-slab testing was conducted. This collection of stones was selected to represent the types of stone that make up the majority of granite countertop sales in the United States as well as three “varieties of concern,” based on descriptions in media reports. Three slabs for each “variety of concern” as well as for three randomly chosen varieties were analyzed in order to assess the amount of variability that can exist among slabs of the same type and to increase the likelihood of discovering any anomalies that may exist.

1.4 RESULTS

1.4.1 Radiation Emissions

Direct measurements demonstrated that the annual dose for full slabs positioned parallel to and 6 inches from a human receptor is less than 0.3 millisievert per year (mSv a^{-1}) for each of the stones tested. At this level, the European Commission recommends that building materials should be exempted from all restrictions regarding their radioactivity (EC 1999). The United States has not established an exemption level for building materials based on radioactivity to our knowledge. All of the measurements were also well below the 1.0 mSv a^{-1} dose limit for the general public recommended by the NCRP

(NCRP 1993). The dose measurements are summarized in Figure 1.1 in order of decreasing market share.

The results presented in the figure represent conservative and therefore health-protective estimates of dose because they assume that the entire body of a person is 6 inches away from and parallel to the full slab for 4 hours per day. In practice, granite countertops are in a horizontal position, while a person is typically seated or standing when near to a counter. Accounting for the distance and horizontal position of a stone countertop relative to a person would result in lower actual doses. A modeling analysis confirmed by direct measurements showed that actual radiation doses would in fact be lower than the values shown in Figure 1.1.

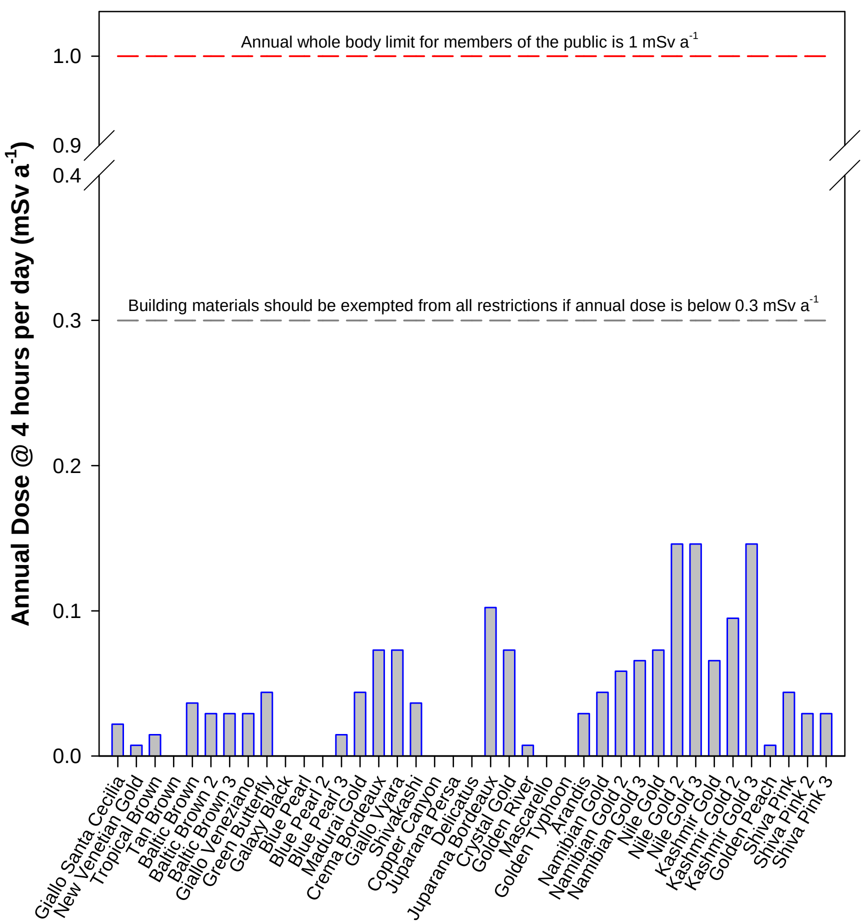

Figure 1.1 Annual dose (mSv a^{-1}) from gamma radiation for each variety of stone based on the median dose measured 6 inches from the surface of the slab and assuming that the full body of a person is parallel to the slab 4 hours per day for 365 days per year. Stone types are presented in order of decreasing market share.

1.4.2 Radon Emissions

The radon emission test results combined with the market share data indicate that the average predicted contribution to indoor radon from all stones tested is less than 0.01 picocuries per liter (pCi L^{-1}), a concentration that is well below both the average outdoor radon concentration in the United States of 0.4 pCi L^{-1} and the U.S. Environmental Protection Agency (EPA) guideline for remedial activities to be undertaken of 4.0 pCi L^{-1} (EPA 1993a).

Modeled concentrations of radon in indoor air that were estimated from the measurements of radon flux are shown in Figure 1.2. Emission rates, or flux, were measured in becquerel per square meter per hour (Bq m^{-2} h^{-1}) and then used to estimate contributions to radon in indoor air (pCi L^{-1}) accounting for ventilation and amount of granite countertop in the home. Similar to Figure 1.1, the results are presented in order of decreasing market share. The types of stone that constitute the majority of granite countertop sold in the United States are on the lower end of radon emissions and exhibit the least variability. This point is illustrated by the estimated radon concentrations for three slabs of the granite type known as Baltic Brown that can be found near the left end of the chart in Figure 1.2. Stone types on the higher end of radon emissions generally exhibited greater variability and account for less than 1% of the market share. The results for Nile Gold shown near the right end of Figure 1.2 are an example of a type of granite that exhibited greater variability among slabs.

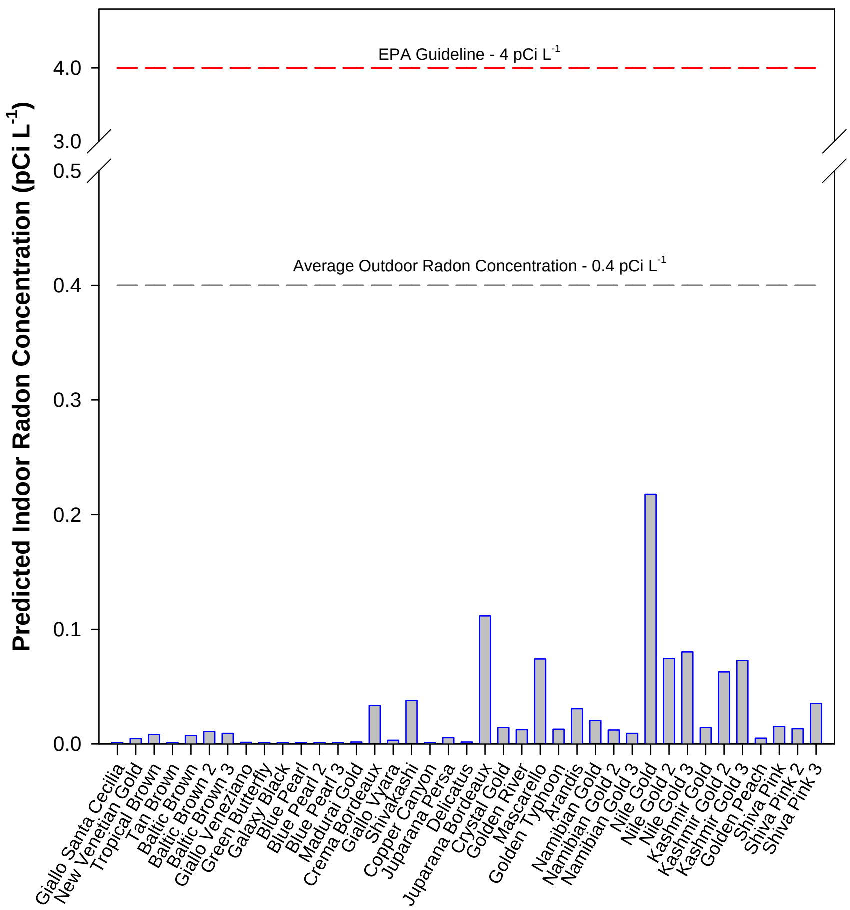

Figure 1.2 Full-Slab Testing—Estimated contribution to indoor radon concentrations by type of stone in order of decreasing market share.

The estimated contributions to indoor radon determined by testing of 213 discrete samples from 112 types of stone were all below background radon levels (Figure 1.3). All of the results for these stones fell within the range of radon flux and concentration values determined for whole slabs that are shown in Figure 1.2. Stone numbers and corresponding stone types are presented in the Appendix (Table A.1).

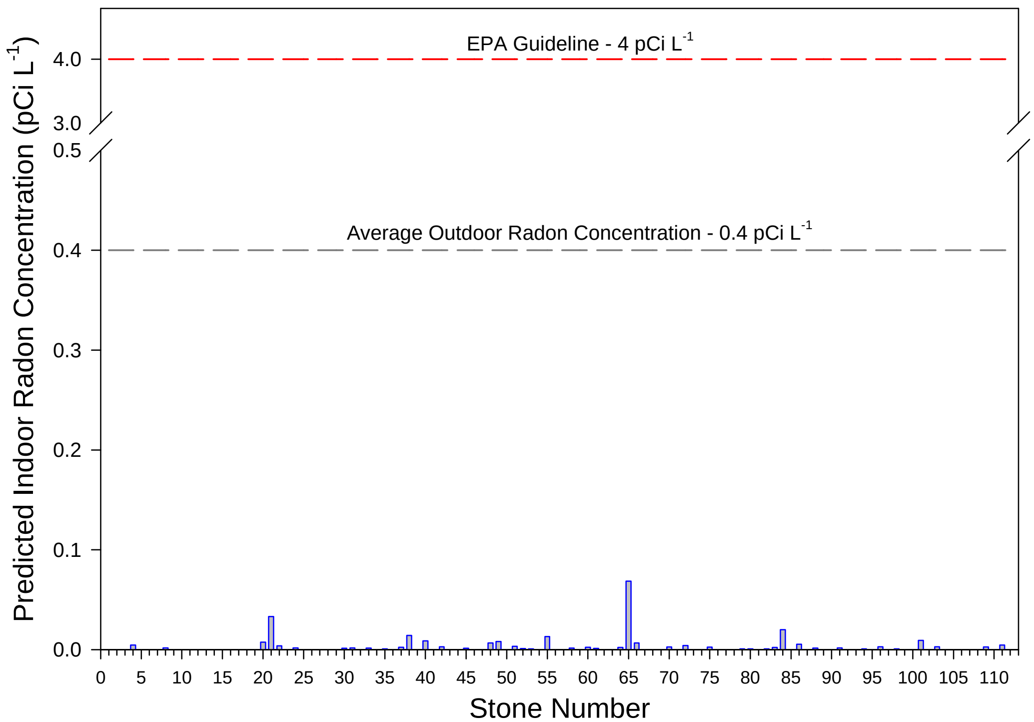

Figure 1.3 Discrete Sample Testing—Estimated contribution to radon concentrations in indoor air by type of stone in order of decreasing market share.

In summary, the analyses completed to date indicate that:

- Radon levels associated with emissions from granite countertops in homes are low in comparison to typical background levels of radon exposure. In other words, natural stone is a minor contributor to concentrations of radon gas within homes. These findings are consistent with an earlier review of the scientific literature that EH&E performed (EH&E 2008).

- External dose associated with radiation emissions for all of the slabs tested are well below health-protective guidelines, including the exemption limit of 0.3 mSv per year recommended by the European Commission. The United States has yet to establish an exemption level for building products based on radioactivity to our knowledge.

- A portion of stones used as countertops may contain limited areas that are enriched in radioactive materials relative to the remainder of the slab. The areas of enrichment in the stones evaluated for this study make up a small proportion of the stone, on the order of less than 10 percent of the surface area. Detailed measurements of these enriched areas showed that they make a negligible contribution to potential doses of ionizing radiation.

- Assessing exposure to radon and radiation requires accounting for duration and frequency of exposure, not just absolute magnitude. Additionally, careful consideration of several key parameters is warranted. For radon, measurements of radon flux from a countertop must account for variability across the countertop surface, the effect of any backing material on the stone, and diffusion through the slab. It is critical that ventilation is accounted for when estimating radon concentrations in indoor air from measurements of radon emissions from stones. For radiation, distance and geometry must be incorporated into dose assessments.

- While significant variability was observed across stone types, the stones at the lower end of radon emissions were found to account for the vast majority of sales and also exhibited little variability among slabs. The varieties of granite countertop that exhibited the greatest variability of radon flux among slabs represent a small fraction of the U.S. market.

2.0 INTRODUCTION

Both radon gas and low levels of ionizing radiation are natural constituents of the environment that we are exposed to every day, with most of our exposure coming from soil, cosmic and internal sources (Shapiro 2002). The elements that produce radon gas and various gamma emissions are also natural and are minor constituents of many common building materials such as concrete, brick, gypsum and granite stone. As such, structural building materials have long been recognized as a source of radon and radiation emissions in residential and other buildings (Ingersoll 1983; Mustonen 1984; NCRP 1987b; Stranden 1988; Khan, et al. 1992; Lee, et al. 2001; Sahoo, et al. 2007). Given the low emission rates of most building materials, their contribution to radiation and radon exposures are thought to be negligible relative to background sources and health-based standards (NCRP 1987a; NCRP 1987c; NCRP 1993).

Whether the same is true for stone countertops sold as granites, hereafter referred to as granite countertops, has recently become a topic of discussion. Recent reports in the popular media have raised questions about these countertops as a potential source of radiation and radon emissions that could then pose a risk to human health. Granite stone is a popular choice for kitchen and bathroom countertops in homes throughout the United States and abroad. These surfaces are typically referred to as "granite" but in fact can consist of a variety of stone types that includes granite and marble. Regardless of the specific type, all granite stones used as countertops are composed of several major minerals and many minor constituents, such as radium. As is the case with granite, and other materials used as structural building components, granite countertops can contain radium-226 (^{226}Ra), the precursor of radon-222 (^{222}Rn). ^{222}Rn is a radioactive gas that is the subject of health-based guidance published by the EPA (EPA 1996b).

Although studies of radiation and radon associated with granite countertops have yet to appear in the scientific literature, concentrations of radioactive elements (i.e., activity concentrations) and emissions of radon have been reported for approximately 500 samples of granite building materials. Activity concentrations of uranium-238 (^{238}U), thorium-232 (^{232}Th), potassium-40 (^{40}K), and ^{226}Ra have been measured in samples of granite obtained in Egypt, Saudi Arabia, Pakistan, South Korea, China, Brazil, Kenya, and Finland. Screening analyses of those samples indicated that the measured activity concentrations correspond to radiation hazard indices that are generally below U.S., European, and international guidelines and standards for external dose (Mustonen 1984; NCRP 1987b; Mustapha, et al. 1997; Chowdhury, et al. 1998; EC 1999; Lee, et al. 2001; IAEA 2003; Kumar, et al. 2003; Arafa 2004; Ahmed 2005; ICRP 2005; Al-Saleh and Al-Berzan 2007; El-Taher, et al. 2007; Lu, et al. 2007; Ghosh, et al. 2008; Mujahid, et al. 2008). Of note, a small portion of the granite samples had a radiation hazard index that exceeded the European Commission screening value for limited use as superficial building materials (external dose of 0.3 mSv a^{-1}). EC protocols would require an evaluation of potential external dose attributable to these granites for reasonably anticipated uses of the material (EC 1999).

Radon flux from samples of granite building materials have also been reported in the scientific literature. In those samples, the average flux was approximately 1.2 becquerels per square meter per hour (Bq m^{-2} h^{-1}) and ranged from 0.01 to 13.5 Bq m^{-2} h^{-1} (Khan, et al. 1992; Chao and Tung 1999; al-Jarallah 2001; al-Jarallah, et al. 2001; El-Dine, et al. 2001; Petropoulos, et al. 2002; Fazal ur, et al. 2003; Stoulos, et al. 2003; Sundar, et al. 2003; Arafa 2004; al-Jarallah, et al. 2005; Osmanlioglu 2006; Singh, et al. 2008; Sonkawade, et al. 2008). In a study in which the radon fluxes measured for those samples were applied to granite countertops in hypothetical homes with ventilation rates typical of U.S. homes, the predicted concentrations of radon in indoor air were low in relation to typical background concentrations of radon and relevant benchmarks for radon exposure (EH&E 2008).

Although radioactivity and radon flux associated with granite building materials has been evaluated in the literature, there is less information regarding the potential risks of radiation emissions from granite countertops and their potential effects, if any, on exposures and health. This may be in part because granite was not commonly used as a countertop at the time that the National Council on Radiation Protection (and related organizations) evaluated radiation emitted from consumer products (NCRP 1987c). Improving the knowledge base regarding the potential risks of granite countertops is important because of the prevalence of this material as a functional and decorative feature of homes.

To address this knowledge gap, EH&E undertook an extensive measurement-based study of radioactivity associated with granite countertops and estimated the potential exposures, doses, and health risks. In this study, EH&E characterized radiation exposures and doses for a diverse sample of granite countertops in multiple ways, including direct measurements of activity concentrations, external radiation exposure, and radon flux. Using these measurements and state-of-the-art modeling tools, radiation dose and radon exposure was estimated for the countertop samples. The exposure and dose results were compared to relevant benchmarks following risk characterization methods used by the EPA and others for building materials (NCRP 1993; EC 1999; EPA 2003; ICRP 2005).

3.0 MATERIALS AND METHODS

3.1 OVERVIEW

The MIA commissioned two studies that evaluated over 400 samples from 115 varieties of stone that comprise approximately 80 percent of the annual U.S. market share for granite countertops, based on the most recent sales data available from 13 of the largest granite distributors in the United States. EH&E was retained to evaluate thirty-nine different slabs of countertop, representing 27 unique varieties of stone that were estimated to constitute over 30% of the U.S. market, for emissions of radiation and radon on a full-slab basis. To increase the scope of the analysis, EMSL Analytical, Inc. (EMSL) was retained to determine radon emissions from one to nine discrete samples for each of another 88 types of stone plus 24 of the stone types for which full-slab testing was conducted. This report focuses on the full-slab testing conducted by EH&E but incorporates the results from EMSL testing and the scientific literature into the modeling for comparative analysis.

The collection of stones was selected to represent the types of stone that make up the majority of granite countertop sales in the United States as well as three varieties suggested in media reports. These three full-slabs of granite and three randomly chosen full-slab varieties were analyzed to assess the variability that can exist among slabs of the same type. Another objective of this study was to increase the likelihood of discovering any surface emission anomalies that may exist. Therefore, the granite types included in this study were intentionally selected from the potentially higher emitting stones.

Radiation measurements were taken on each full slab of granite countertop using three instruments: a Geiger-Müeller (GM) counter, a gamma ray meter, and a MicroRem Survey meter. This array of instruments was selected to obtain measures of total radioactivity, gamma ray emissions, and external dose rate required to evaluate potential doses and risks of ionizing radiation emitted from the slabs. In further full slab testing, radon flux was measured in six standardized locations on each slab as well as areas where elevated levels of gamma emissions were identified. To complete the characterization of slabs, a small piece was removed from two corners of each slab and analyzed for activity concentrations of common radioisotopes by gamma ray spectroscopy. Details of the sampling design for the evaluation of full slabs are provided in Section 3.2.

Measured activity concentrations and radon flux were input to mathematical models to obtain estimates of external dose and indoor radon levels. The scenarios modeled were conservative exposure scenarios (i.e., higher than anticipated real life) under conditions representative of residential building stock in the U.S. For radon, the modeling included flux measurements from the full slab evaluation as well as two other sources: (i) discrete samples of granite countertop analyzed by EMSL (EMSL 2008) and (ii) discrete samples of granite building materials reported in the scientific literature. Radon flux measurements presented in those additional reports were included in the modeling analysis to complement the findings of the full slab assessment outlined above. In recognition of the different types of granite and testing methods reflected in these sources of information, the modeling results are presented separately for the radon flux results available for full slabs of granite countertop (EH&E), discrete samples of granite countertop (EMSL), and discrete samples of granite building materials (published literature). Although differences in the samples and methods should be considered when comparing results among these three sources, the entire set of data is presented here to gather all of the relevant information available at this time into a single report. Details of the radiation dose and radon exposure modeling are described in Section 3.3.

Health risks associated with the radioactive constituents of granite countertops were estimated using both the measured and modeled findings together with conservative assumptions (i.e., health-protective) regarding human activity patterns. The magnitude of the risks were evaluated by comparing them to: (1) radiation screening criteria established by the European Commission; (2) dose limits for ionizing radiation recommended by the U.S. National Council for Radiation Protection and various international organizations; and (3) guidance on radon exposure from the U.S. Environmental Protection Agency, the European Commission and other organizations. Details of the methods used to characterize risks of radiation dose and radon exposure associated with granite countertops are presented in Section 3.4.

3.2 MEASUREMENTS

3.2.1 Sampling Design

All measurements were made using a standardized sampling approach to allow comparability between measurements and among slabs of granite.

A total of 39 slabs were tested on a full-slab basis, with each slab measuring approximately 10 feet by 5 feet by 1.25 inches. Each slab was tested for radiation and radon flux. As shown in Figure 3.1, radioactivity and external dose was measured at multiple positions on each slab. These positions were located along a grid, with fourteen sampling points, and at two additional points on the upper right and left corners. Radioactivity and radiation dose measurements were obtained at each of the 16 points while slabs were positioned vertically. In addition, radon emission and gamma ray measurements were made at six locations (indicated in red on Figure 3.1) when the slabs were placed in a horizontal position. These measurements at pre-determined locations were supplemented by screening the entirety of each slab using the GM counter to identify areas with greater than 500 counts per minute. These potentially enriched areas of radioactivity were subjected to the full suite of testing discussed above. At the conclusion of the measurements and screening, a 5 inch by 2.5 inch piece was cut from the upper left and right corners of each slab (Z1 and Z2) for activity concentration analysis.

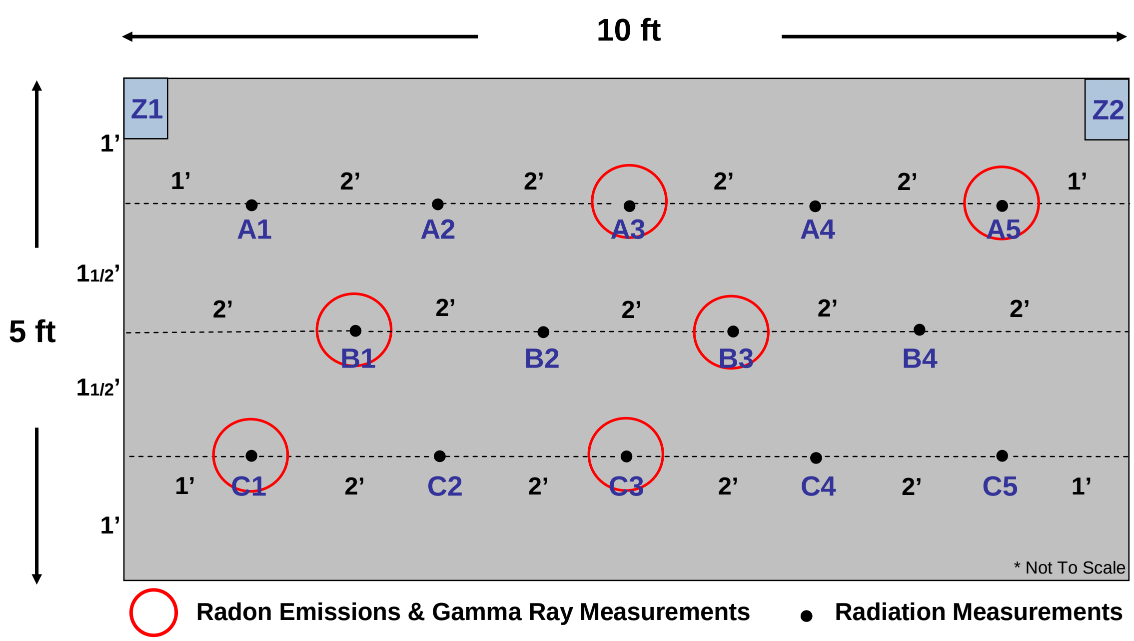

Figure 3.1 Slab Sample Locations for Radon and Radiation Testing

3.2.2 Radiation Measurement Methods

3.2.2.1 Radioactivity Measurements

Radioactivity measurements were made at each of the 16 points using a GM counter (Model 3, Ludlum Measurements Inc., Sweetwater, Texas), which detects alpha, beta and gamma radiation (Figure 3.1), measured as counts per minute. Of these emissions, only gamma rays play a significant role in external dose to a human body. Although there are significant limitations with the GM measurements they were taken as part of this study to permit comparison with reports of radiation exposure associated with granite countertops that have appeared in the popular media. The GM counter was also used as a rapid-screening tool to identify potentially enriched areas that required further investigation. Since any health hazards from granite countertop radiation would arise primarily from emitted gamma rays, radiation measurements of only the gamma rays were made by placing a one-inch thick alpha/beta absorber on the slab surface with the GM counter atop the absorber. GM measurements were also made immediately on the surface of the slab and one inch from the slab surface without the alpha/beta absorber.

To obtain more accurate measures of gamma ray emissions from granite countertops, a gamma ray scintillator (Model 44-10, Ludlum Measurements, Inc., Sweetwater, Texas) was used to measure gamma radiation at six locations (indicated in red on Figure 3.1). The gamma ray meter with a 2 inch by 2 inch sodium-iodide detector was used to obtain a comprehensive measurement of gamma ray emissions and to account for high energy emissions that are not reflected in the GM measurements.

3.2.2.2 Activity Concentration Measurements

A gamma spectroscopy system with a high purity germanium detector (Canberra Reverse Electrode High Purity Germanium System, Meriden, Connecticut) was used to measure the activity concentrations of selected isotopes (^{238}U, ^{232}Th, ^{40}K, ^{226}Ra) in the Z2 sample of each slab. The Z2 samples were kept whole to avoid any increase in the loss of radon daughter atoms. Each sample was placed in contact with the detector and counted for 30,000 seconds. The system was calibrated using a standard source of similar dimensions as the granite samples. The detector was shielded with lead to reduce any interference from background radiation.

The specific activities were averaged from the gamma-ray photopeaks at several energies. For each slab, the specific activity for the uranium and thorium series activities were determined using a weighted average of the daughter isotopes identified in the series. The ^{238}U series, which contains ^{226}Ra and ^{222}Rn, was determined using the ^{234}Th, proctactinium-234m (^{234m}Pa), lead-214 (^{214}Pb) and bismuth-214 (^{214}Bi) gamma ray lines. The thorium-232 (^{232}Th) series specific activity was determined using actimium-228 (^{228}Ac), lead-212 (^{212}Pb), and thallium-208 (^{208}Tl) gamma ray lines. The specific activity of the ^{40}K is determined directly using its own gamma ray line at 1461 kiloelectron volts (keV). The minimum detectable specific activity of ^{40}K was 14 becquerels per kilogram (Bq kg^{-1}), ^{232}Th was 2 Bq Kg^{-1} (based on ^{208}Tl) and 3.5 Bq kg^{-1} for ^{238}U (based on ^{214}Pb daughter specific activity).

3.2.2.3 Dose Measurements

The radiation dose rate (mSv h^{-1}) from external sources was measured on vertical slabs using a MicroRem Survey Meter (Bicron Radiation Measurement Products, Solon, Ohio). The MicroRem meter measures external dose and is the most appropriate device for rapid assessment of potential doses of ionizing radiation. Measurements were collected at the surface, six inches and 12 inches from the vertical slabs at each of the 16 pre-determined grid and corner locations. In addition, radiation dose was also measured for areas identified as potentially enriched. Radiation dose measurements were taken on vertically placed slabs to provide a conservative estimate of exposure compared to when stones are horizontal, as when used as countertops. When used as countertops and placed in a horizontal position, the increased distance from the source and non-uniform radiation plane lead to decreased radiation exposure.

Dose rates for each slab were calculated using the median of all measurements taken on the slab, corrected for background radiation. Corresponding annual doses were calculated using the slab-median dose rates and an assumed exposure duration of 1,460 hours per year (4 hours per day for 365 days per year).

3.2.2.4 Long-Term Radiation Dose Equivalents

Radiation dose to tissues was measured under pseudo-real world conditions for one granite countertop slab, selected based on surface dose rate readings of twice background. A mannequin was placed next to a granite countertop (5.75 feet x 2 feet plus a 4 inch backsplash) with both arms extended over the countertop for 30 days (Figure 3.2). Radiation film badges (Luxel+, Landauer Inc, Glenwood, Illinois) were placed on the mannequin at locations ranging from the fingertips to the torso.

Figure 3.2 Photograph of Mannequin Positioned for Long-term Dosimeter-based Radiation Testing

Radiation film badges were analyzed and reported as shallow and deep radiation dose equivalents, the units in which standards for allowable radiation doses are expressed. Deep dose equivalent applies to external whole body exposure at a tissue depth of one centimeter. Shallow dose equivalent applies to the external exposure to the skin or extremity at a tissue depth of 0.007 centimeters.

3.2.3 Radon Flux Measurements

For each of the 39 slabs, radon flux was determined simultaneously at the six locations shown in Figure 3.1 as well as potentially enriched areas identified with the GM counter.

During all sampling events for radon flux, slabs were placed in a horizontal position across supports approximately 3.5 feet above the ground. Seven of the granite slabs were selected for additional testing to characterize radon emissions from the unpolished, bottom surface of the granite.

Two types of quality assurance samples were obtained in the field throughout the period when radon flux measurements were made. First, repeated measurements were made from a single slab on alternate sampling days to provide a form of continuing calibration and information on precision of the method. Second, radon concentrations in air of the test room were measured on alternate days to control for potential variation in background levels of radon.

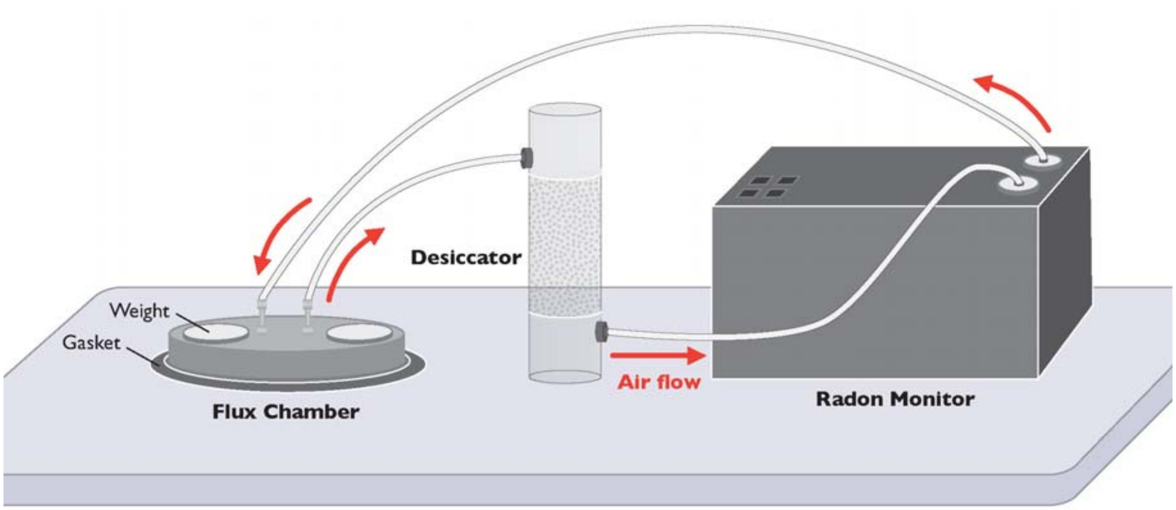

Figure 3.3 Schematic of the Radon Flux Measurement Setup

Radon flux was measured at each site using a flux chamber connected to a real-time radon monitor (Durridge Rad-7 Radon Monitors, Bedford, Massachusetts) via tubing, as depicted in Figure 3.3. The radon monitor was purged for at least 10 minutes prior to beginning the test. For each test, the flux chamber (volume of 0.0016 cubic meters [m^3] and surface area of 0.038 square meters [m^2]) was fit to a rubber gasket, placed on the granite surface at the pre-specified location, and weighted with 2.0 pounds to seal the gasket against the surface. The monitor and tubing added an additional 0.0013 m^3 to the flux chamber setup, bringing the total volume to 0.003 m^3. The radon monitor drew and returned air at a flow rate of 0.7 liters per minute, resulting in constant air pressure in the flux chamber. Air from the flux chamber was passed through a desiccation column to remove any moisture.

One-hour integrated radon concentrations were recorded every hour, for a minimum of 24 hours. These measurements were subsequently characterized as follows:

$$C_t = C_{sat}\left(1 - e^{-kt}\right)$$ **--- (Equation 3.1)**

where C_t is the radon concentration at time *t* (Bq m^{-3}), C_{sat} is the saturated radon concentration (Bq m^{-3}), *k* is total instantaneous loss rate for radon (h^{-1}), and *t* is the elapsed time (hours). Since granite slabs are porous, *k* is the sum of radioactive decay of radon, loss to diffusion through the slab, and loss due to any leakage between the sampling system and the granite slab. The parameters C_{sat} and *k* were estimated from Eq. 3.1 using an iterative curve-fitting procedure implemented with the NLIN procedure in SAS statistical software (SAS Institute, Cary, North Carolina).

Radon flux rates (Flux) were then calculated using the estimate of C_{sat} and *k*, based on the following equation:

$$F = \frac{C_{sat} \times V \times k}{SA}$$ **--- (Equation 3.2)**

where F is the emission rate or Flux (Bq m^{-2} h^{-1}), C_{sat} is the saturated Radon concentration (Bq m^{-3}), V is the total volume of sampling system (m^3), k is the total radon loss (h^{-1}), and SA is the surface area under the flux chamber (m^2).

For granite slabs that had potentially enriched areas identified (2 out of 26 stone types), flux rates were calculated using a weighted average approach based on the percent of surface area represented by each test chamber.

3.3 MODELS

Mathematical models were used to estimate radiation dose and indoor air concentrations. These models are publicly available and their performance is well documented. Measurements from the current study were used as inputs. For external dose, the output from the models was used to supplement radiation dose measurements and to characterize whether these measurements differ from estimated doses that may occur in real-world settings. Indoor air radon concentration models were used to provide an estimate of radon exposures, which take into account not only the radon flux but also the amount of countertop present and home ventilation rates.

3.3.1 Dose Modeling

The radiation dose rate (µRem h^{-1}) from countertops in a model kitchen was estimated for several types of granite countertop to provide an estimate of real-world dose from granite countertops and to provide additional information to aid in interpretation of the potential radiation dose rates that are described above. The computer software program MicroShield® (Version 7, Grove Software, Inc., Lynchburg, Virginia) was used to generate the estimates of dose using activity concentrations and dimensions of granite countertops as the primary model inputs. Model estimates obtained using vertical granite positions were compared to measured doses in the same vertical configuration (Section 4.2.1.1). The modeling results at six inches and the dose measurements collected at six inches were found to be in good agreement (median difference < 2.5 µRem h^{-1}), indicating that the model provides reasonably accurate estimates of dose associated with emissions from granite countertops.

Using the modeled dose rates, an annual dose for each height was calculated based on an individual spending one hour each day of the year at each of the four locations, for a total of four hours per day in the kitchen. This value is almost three times greater than the average amount of time reported spent on any given day in the kitchen in U.S. homes (EPA 1996a).

The layout of countertop in the model kitchen is illustrated in Figure 3.4. The countertop constitutes over 50 square feet of stone distributed across a primary counter, secondary counter, island, and backsplash behind the primary counter. Dose was modeled at four locations in the kitchen identified as D1 through D4 in the figure. The four dose point locations were chosen to represent locations where people may spend their time in the kitchen and ranged from direct contact to a maximum of six inches away from the primary counter or island. Further, dose was calculated at three heights (countertop height, 12 inches above the counter, and 24 inches above the counter), for a total of 12 dose estimates per model run.

In a set of model runs where activity concentrations were assumed to be homogeneous throughout the countertops in a kitchen, activity concentrations measured in the Z2 samples described earlier from nine of the slabs were input to the model. The samples were selected to represent the range of measured activity concentrations, including the highest, lowest, and seven intermediate values ranked according to an activity concentration index (ACI) recommended by the European Commission (EC 1999) and discussed in Section 3.4.1.

The potential impact of enriched areas of radioactivity on estimated doses was evaluated in another set of model runs. In this evaluation, two areas of enriched radioactivity were assumed to exist at locations proximate to dose location D1 and D3 (see Figure 3.4). The enriched areas were assumed to have a square footprint 2 inches by 2 inches in size. Since activity concentrations for these theoretical enriched areas were not specifically measured, the maximum recorded activity concentrations, obtained in a report sent to the Marble Institute of America of data reported from ARS International (Port Allen, Louisiana), were used as the activity concentration input. Both the assumed enriched area placement and activity concentration were selected to represent worst-case scenarios (e.g., maximum total annual dose for the four dose points).

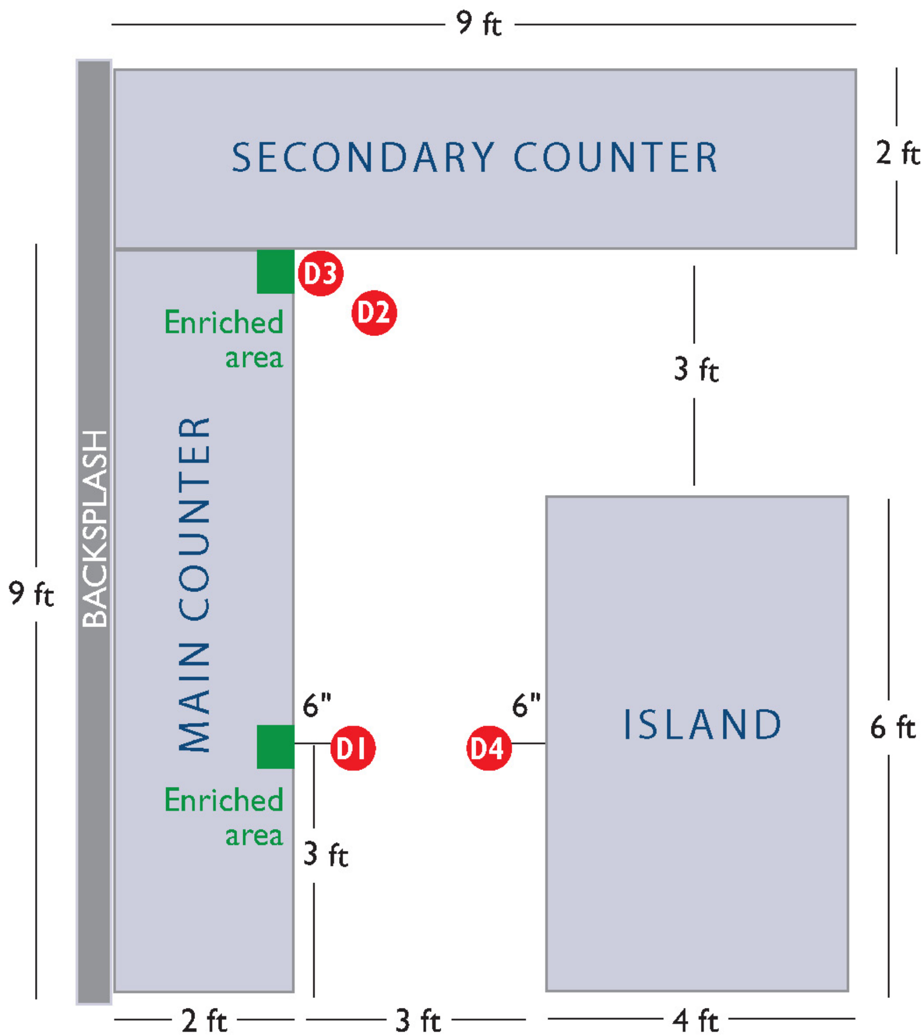

Figure 3.4 Layout of Kitchen, Stone Countertop and Dose-point Locations for Dose Modeling

3.3.2 Indoor Radon Concentration Model

3.3.2.1 Indoor Air Model (CONTAM) Description

The CONTAM multi-zone indoor air quality model (Walton and Dols 2003) developed by the National Institute of Standards and Technology (NIST) was used to estimate indoor radon concentrations following a previously published methodology (Myatt, et al. 2008). Briefly, airflow among indoor and outdoor zones of the building (i.e., rooms and ambient air) in CONTAM occurs via flow paths such as doors, windows, and cracks. Inter-zonal flow is based on the empirical power law relationship between airflow and the pressure difference across a flow path. Simulation of a mechanical ventilation system in CONTAM also induces circulation of air in CONTAM. After airflow among zones is established, mass balance equations are used to calculate pollutant concentrations based on the sources and sinks in each zone.

The model was run under four scenarios—two home ventilation conditions for each of two residential building templates developed by NIST. The home templates represent a newer, two-story detached home (post-1990) and an older, single-story detached home (pre-1940) (Table 3.1).

3.3.2.2 CONTAM Model Inputs

Radon flux values were entered into the CONTAM model to represent emissions in the kitchen and bathrooms. The emission rates used covered the full range of emission rates measured in this study, as well as values reported by EMSL laboratories and others in the scientific literature. Radon flux was modeled with emissions from the top, bottom, and sides of each piece of granite countertop assumed to be present within a home.

Due to limited published data on granite countertop usage in homes, two months of sales data obtained from a natural stone fabricator was used to determine appropriate inputs for the average surface amount of granite countertop in homes. The average installation calculated from these data consisted of 50 square feet and 18 square feet of granite countertop in kitchens and bathrooms, respectively. Countertop allocated to bathrooms was divided among the bathrooms in each template. The footprint of 68 square feet was extrapolated to a total surface area for radon flux of approximately 144 square feet accounting for potential emissions from the top, bottom, and edges of the countertop.

Table 3.1 CONTAM Model Template Information

Template	Age	Air Exchange Rate per Hour		No. of Floors	Square Footage			No. of Bath	Countertop Surface Area (square feet)	
		Natural	Forced Air		Home	Kitchen	Bathroom		Kitchen	Bathroom
DH72	before 1940	0.5	0.4	1	1,900	300	200	2	50	18
DH28	after 1990	0.2	0.2	2	3,000	200	370	3.5	50	18

3.3.2.3 The Role of Air Exchange and Ventilation

An important variable in modeling expected indoor air contaminant concentrations is the home ventilation rate, typically measured as air changes per hour (ACH). Air exchange rates in residential buildings vary considerably based on the type of construction, seasonal variability in weather, and geographic region (ASHRAE 2005).Typical air exchange rates for residential buildings in North America range from a seasonal average of about 0.2 ACH for tightly constructed homes to upwards of 2 ACH for loosely constructed housing (ASHRAE 2005). Additional studies have shown that an ACH of approximately 0.5 is a reasonable estimate of average seasonal air exchange rate for residences (Grimsrud, et al. 1982; Palmiter and Brown 1989; Ek, et al. 1990; Parker, et al. 1990). A value of 0.2 ACH was used for the newer home template and 0.4 – 0.5 ACH for the older home, which represent the lower bound and average of the typical seasonal average air exchange rates for residential buildings in North America (ASHRAE 2005).

3.4 RISK CHARACTERIZATION

3.4.1 Screening Evaluation

Measured activity concentrations were used to calculate an activity concentration index (ACI) for each of the granite slabs following a method recommended by the European Commission (EC). The ACI is typically used as a screening tool to identify materials of potential radioactivity and radon exposure concern (EC 1999). Consistent with other types of screening evaluations, the ACI is derived using measured activity concentrations assuming that a person is standing in the center of a rectangular room in which the material being evaluated lines the interior surface of the floor, ceiling, and all four walls (Markkanen 1995). This assumption is extremely conservative, given that the surface area of granite countertops in typical homes is only a small fraction of that for floors, ceilings and walls of a house. The ACI accounts for potential gamma radiation emitted by radium, thorium, and potassium in the slab and is calculated from the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K expressed in units of Bq kg^{-1} as shown in Equation 3.3.

$$ACI = \frac{C_{Ra}}{300\ Bq\ kg^{-1}} + \frac{C_{Th}}{200\ Bq\ kg^{-1}} + \frac{C_K}{3000\ Bq\ kg^{-1}}$$ **--- (Equation 3.3)**

where C_{Ra}, C_{Th} and C_k are the radium, thorium and potassium activity concentrations, respectively.

According to the EC (EC 1999), appropriate dose assessments should be performed for superficial building materials such as tiles and boards with an ACI>2, a value where the annual dose may exceed 0.3 mSv. Likewise, an ACI>6 is indicative of a potential dose in excess of 1.0 mSV per year, a level that warrants controls according to the EC (EC 1999). To evaluate the appropriateness of the six room surface ACI as a screening tool for granite countertops, the calculated ACI values for this studies granite slabs were compared to the measured and modeled doses for the same slabs.

3.4.2 External Dose

To characterize potential risks of external exposure to ionizing radiation emitted from granite countertops, the measurements and modeling of the external dose were compared to relevant and appropriate guidelines published by a number of authoritative national and international organizations.

The dose estimated from both the measurements and modeling used a conservative (i.e., health-protective), and potentially realistic exposure scenario. Annual dose calculated from measurements assumed that a person stood parallel to and 6 inches from the surface of each slab for 1,460 hours per year of exposure (4 hours per day). Doses estimated from the mathematical modeling included 365 hours per year (1 hour per day) when a person was in direct contact with enriched areas on a granite countertop such as might occur while preparing food or sitting at a counter. In accordance with the goal of obtaining health-protective estimates of dose, receptor locations with the maximum dose rates estimated with the model were used to characterize risks of external dose.

The guideline values used as benchmarks for external dose are summarized in Table 3.2. In the United States, the Nuclear Regulatory Commission (NRC) and Environmental

Protection Agency (EPA) also provide guidance on limits for external doses of radiation to the general public. Although similar to the values tabulated below, the radiation dose limits established by the NRC and EPA are not directly relevant to building materials. The NRC dose limit applies to radiation emitted from facilities licensed by the NRC (NRC 2007). The EPA dose limit applies to residual levels of radiation that remain following the remediation of a formerly contaminated site. The EPA limit is unique in that it takes into account the fact that remediated sites may introduce additional routes of exposure, "The cleanup levels to be specified include exposures from all potential pathways through all media (e.g., soil, ground water, surface water, sediment, air, structures, and biota)" (EPA 1997). Unlike the dose limits recommended by the NCRP, ICRP, IAEA, and EC, neither the NRC nor the EPA limits apply to consumer products or other sources of radiation that are common to residential or commercial settings.

Table 3.2 Limits for Annual Dose of Radiation (mSv a^{-1}) in Excess of Background Published by National and International Organizations

Limit	Organization	Note	Reference
1.0	NCRP	Recommended annual radiation dose limit for individual members of the public from all radiation sources other than natural background and the individual's medical care	(NCRP 1993)
1.0	ICRP	Annual radiation dose limit for situations having societal benefit, but without individual direct benefit, and there is no information, no training, and no individual assessment for the exposed individuals in normal situations	(ICRP 2005)
1.0	IAEA	Annual radiation dose limit for members of the public that are exposed to a given source or practice	(IAEA 2003)
0.3	EC	Building materials that contribute an annual dose of gamma radiation less than the limit should be exempt from all restrictions regarding their radioactivity	(EC 1999)
1.0	EC	Annual radiation doses from building materials that exceed the limit should be accepted only in some very exceptional cases	(EC 1999)

NCRP National Council on Radiation Protection and Measurements
ICRP International Commission on Radiological Protection
IAEA International Atomic Energy Agency
EC European Commission
mSv a^{-1} millisievert per year

3.4.3 Radon

To characterize the risk of exposure to radon in indoor air of residences as a result of emissions from granite countertops, indoor air concentrations estimated from the measured radon flux (see Section 4.2.2) were compared to relevant benchmarks that included exposure guidelines for radon, background levels of radon, and quantitative estimates of health risk. The U.S. Environmental Protection Agency, for example, has established an action level of 148 Bq m^{-3} (4.0 pCi L^{-1}) for radon in residential indoor air (EPA 1993b). Similarly, the ICRP recommends an action level for radon in indoor air of dwellings that is no lower than 200 Bq m^{-3} (5.4 pCi L^{-1}) (ICRP 2005). In specific reference to building materials and radon, the EC states that the amount of radium in building materials should be below a level where it is unlikely to be a major cause of radon concentrations in indoor air that exceed 200 Bq m^{-3} (EC 1999).

Normative levels of exposure to radon in air are another relevant benchmark to help interpret our measured results. With a flux from soil of approximately 65 Bq m^{-2} h^{-1}, concentrations of radon in outdoor air range from approximately 3.7 to 37 Bq m^{-3} (0.1 to 1 pCi L^{-1}) (NCRP 1987a) with a national average of 14.8 Bq m^{-3} (0.4 pCi L^{-1}) (EPA 1993a). Radon levels inside of homes are typically greater than levels in outdoor air. The average concentration of radon in indoor air of U.S. homes is reported be 48 Bq m^{-3} (1.3 pCi L^{-1}) with a median level of 26 Bq m^{-3} (0.7 pCi L^{-1}) (Marcinowski, et al. 1994). Sources of radon that account for the difference between levels in outdoor and indoor air include infiltration of radon gas from soil through foundations, volatilization from domestic water use, and emissions from building materials such as concrete, gypsum board, and granite (NCRP 1987a).

Risks of lung cancer associated with radon in indoor air are also a useful benchmark for interpreting the results of our exposure assessment. We calculated the estimated risk of lung cancer mortality from lifetime exposure to radon associated with emissions from granite countertops following methods detailed by the EPA (EPA 2003) and derived from an analysis of health risks of radon exposure among miners reported by the National Research Council (NRC 1998). Specifically, the excess cancer risk of lifetime exposure to radon was estimated from a unit risk factor of 1.6×10^{-4} per Bq m^{-3} (5.8×10^{-3} per pCi L^{-1}) (EPA 2003). The application of risk models developed from observations of miners to the general population requires low dose extrapolation and incorporates all of the attendant uncertainties (EPA 2003). Despite these uncertainties, the risks estimated with this method allow contributions of radon exposure anticipated for granite countertops to be compared to common sources of risk in the everyday environment.

4.0 RESULTS

4.1 MEASURED VALUES

4.1.1 Radiation and External Dose

4.1.1.1 Activity Concentration Analysis

The activity concentrations for each of the granite stones tested are reported in Figure 4.1. Of the 39 stones tested, 18 had hazard index levels at or higher than 6, which corresponds to a dose criteria of 6 mSv a^{-1} for superficial materials such as tile and boards (EC 1999) (see Section 4.3.1. for further discussion on the applicability of the hazard index). According to the EC document, a dose assessment should be performed if the hazard index exceeds 2. Therefore, the direct measurements and modeling of dose presented in the Methods section were used to conduct a dose assessment as described in the following sections.

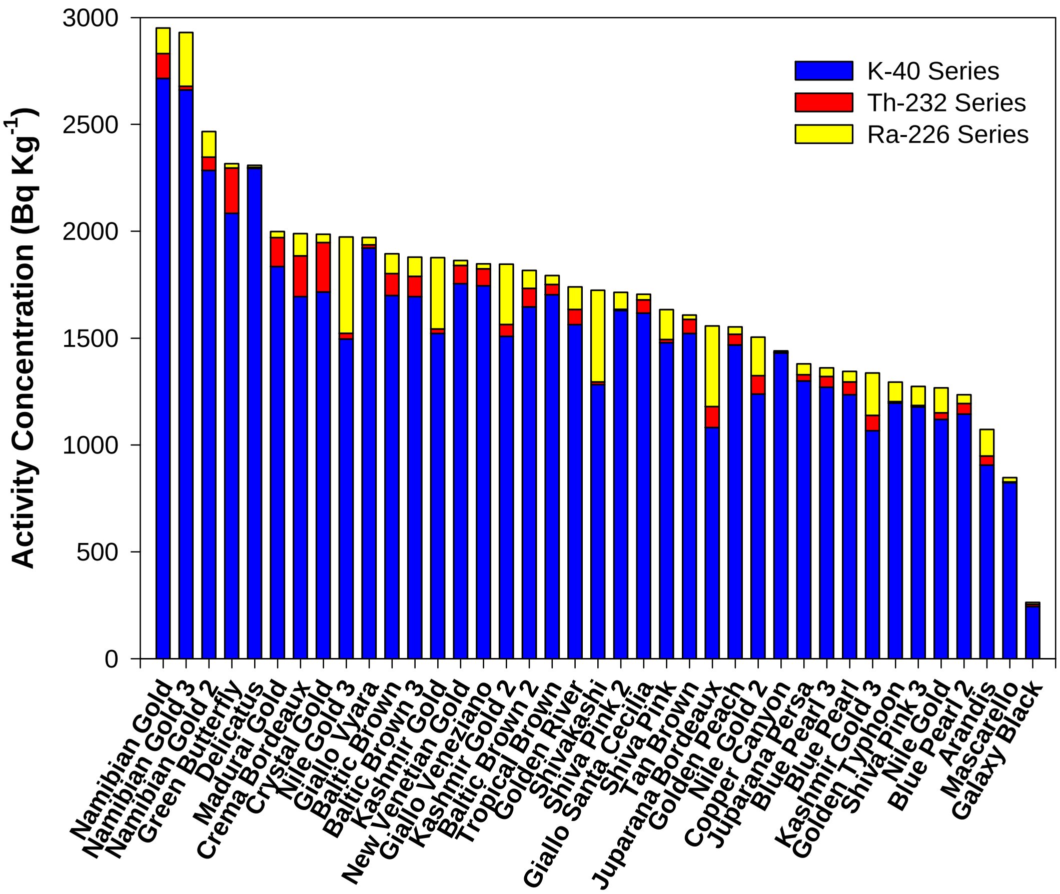

Figure 4.1 Activity Concentrations for the Ra-226, Th-232 and K-40 Series (Bq kg^{-1}) for Each of the 39 Stones

4.1.1.2 Gamma Ray Activity

Measurements obtained with the GM on the 39 full-slabs tested ranged from background to 3,000 counts per minute (cpm). The average and extremes of the readings encompass the range of GM data presented in the popular press for granite countertops. This finding indicates that the stones included in the present evaluation reflect the range of gross activity reported elsewhere for other samples of granite countertop, including those that have been implicated as being of concern in media reports.

As shown in Figure 4.2, GM readings were inversely correlated with the percentage of total external exposure attributable to gamma rays. This observation indicates that readings obtained from a GM are not a reliable indicator of dose from gamma rays, a key limitation for use of a GM as a screening tool for radiation dose assessments of granite countertops.

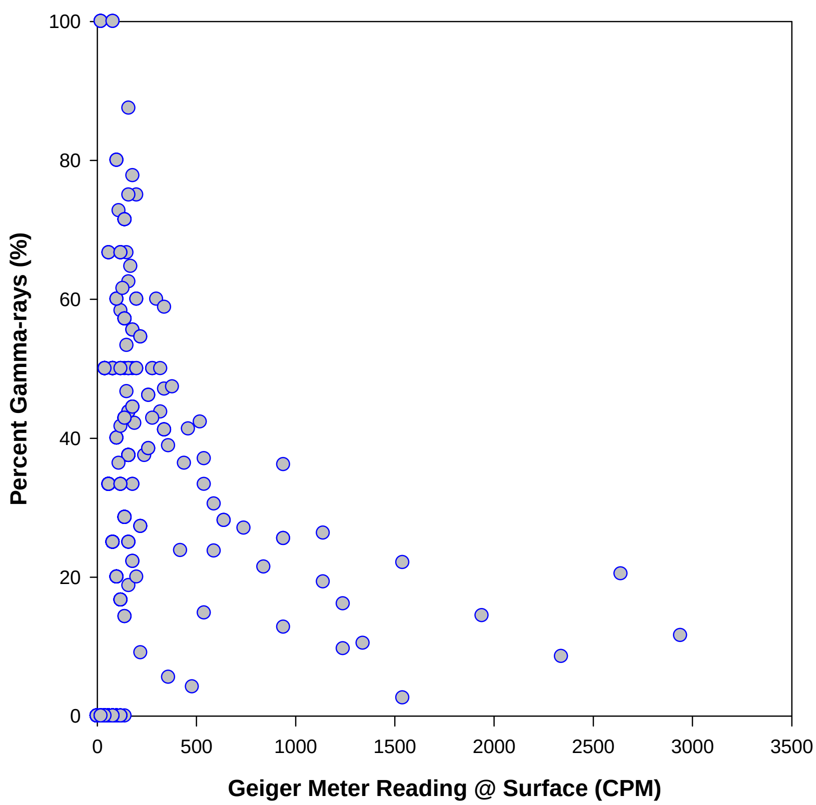

Figure 4.2 Geiger-Müeller Readings in Counts Per Minute and the Corresponding Percent of Radiation from Gamma-rays

4.1.1.3 External Dose Measurements

For each of the granite countertops tested, the median annual dose for granite countertops positioned vertically is less than 0.3 mSv, indicating that the granite countertops should be exempt from all restrictions regarding radioactivity, as recommended by the European Commission (EC 1999). The United States has not established an exemption level for building materials based on radioactivity (see Section 4.3.2 for details). Further, the measured doses are also well below the 1.0 mSv dose limit for the general public recommended by the NCRP (NCRP 1993).

The dose measurements are summarized in Figure 4.3 in order of decreasing market share. For six stone types, three different full slabs were tested to assess differences across slabs of the same type (denoted with a '*' in Figure 4.3).

These results represent a conservative, exposure-protective scenario because the doses were determined for exposures in which the countertop is positioned vertically, such that it would be parallel to a person standing in front of a wall of granite, at 6 inches from the granite. Since radiation dose is dependent on distance and position of the stone, these results represent a worst-case and unlikely scenario, as in practice, people typically sit or stand next to a granite countertop that lies perpendicular to their body.

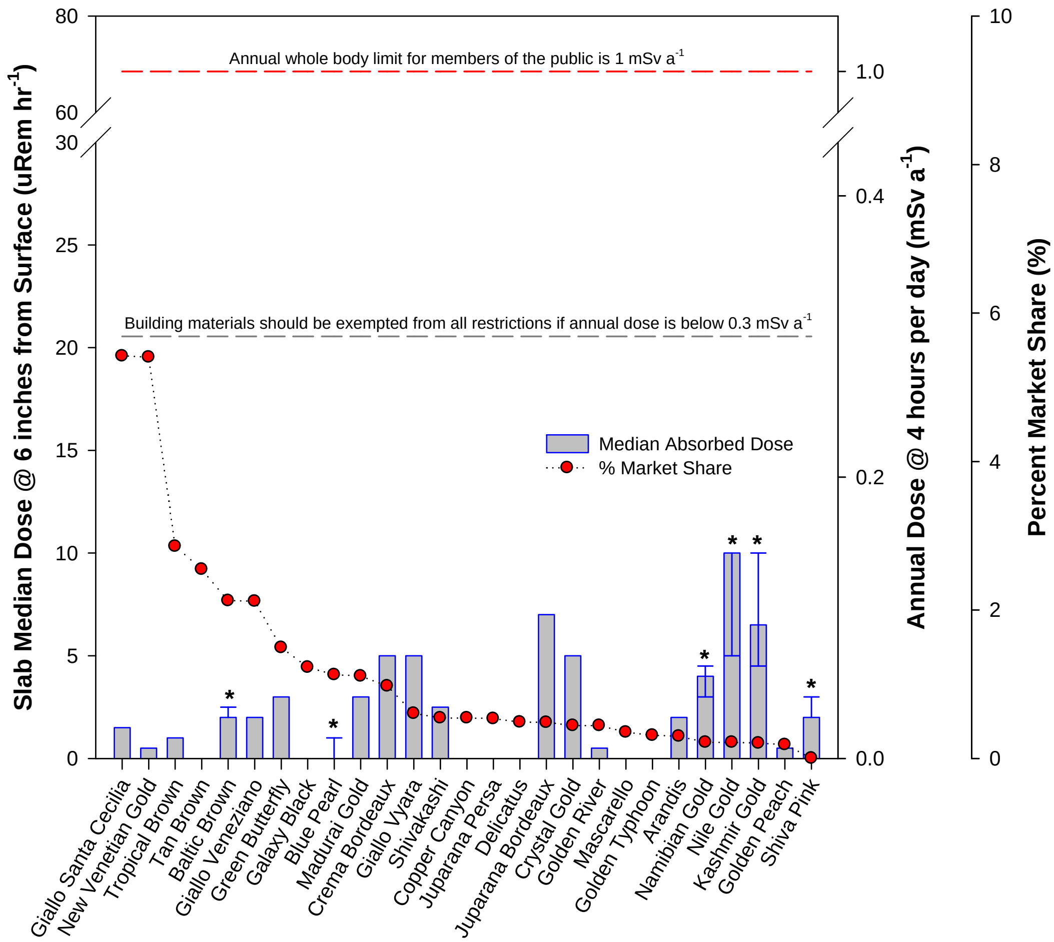

Figure 4.3 Distribution of annual doses (mSv per year) based on median parallel plane exposure measurements at 6 inches above the surface and assuming 4 hours per day of exposure for 365 days per year. Bars denoted with a star (*) represent the range of test results from three slabs of that stone type.

As is well understood in the health physics field, radiation dose was found to decrease with increasing distance from the countertop, as shown in Figure 4.4, where dose measurements taken at 3 distances from the counter (surface, 6 inches and 12 inches) for one slab, with two areas elevated above background, are presented. The results of the measurements collected at three distances from the stones show that radiation dose significantly decreases with increasing distance, becoming indistinguishable from background at 12 inches.

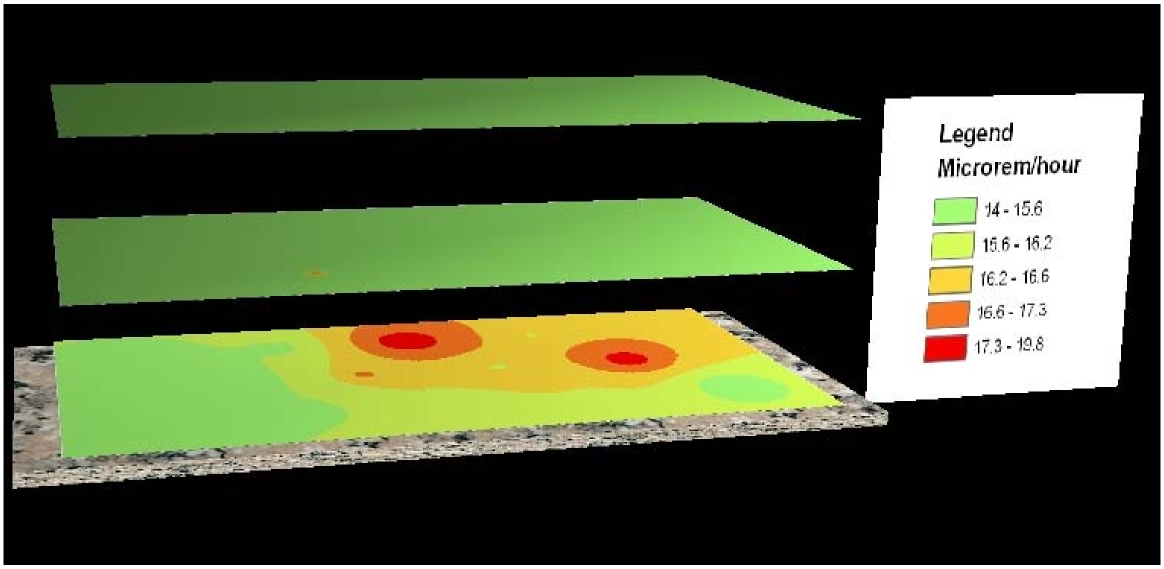

Figure 4.4 Three-dimensional Figure Demonstrating the Reduced Radiation Dose Rate for One Granite Slab at the Surface and Distances of 6 and 12 inches

4.1.1.4 Long-term Radiation Dose Equivalents

Radiation dose equivalents, a measure of radiation doses to tissues, were measured on a mannequin standing in front of a pre-selected granite slab continuously, for 30 days (720 consecutive hours). The granite slab had a measured dose rate of 12 microRem per hour (µRem h^{-1}) on the surface. Figure 4.5 demonstrates that radiation dose rate significantly decreases with distance from the countertop, with measured dose rates on the mannequin in the range of 0 to 3 µRem h^{-1}, all less than the limit of detection of 5 µRem h^{-1}.

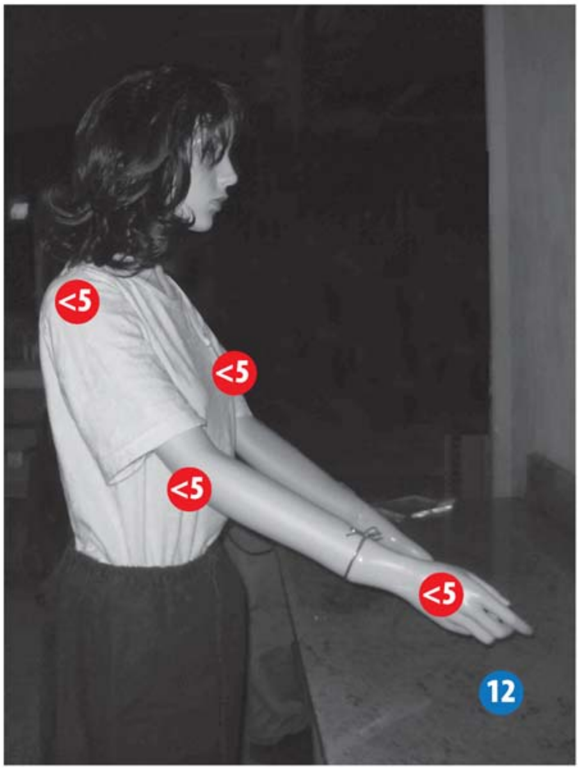

Figure 4.5 Results of the Long-term Radiation Badge Dosimeter Testing on a Mannequin, Reported in Dose Rates of μRem h^{-1}. The blue circle represents the dose rate on the counter surface. The red circles are measured dose rates on the body, and are all below the limit of detection (<5 μRem h^{-1})

4.1.2 Radon Flux

4.1.2.1 Full-Slab Testing

Radon flux measurements for the 39 granite slabs (27 unique types) are shown in Figure 4.6, with results presented in order of decreasing market share. The types of granite that constitute the majority of stones sold in the United States are on the lower end of radon emissions and exhibit the least variability. This point is illustrated by the estimated radon concentrations for three slabs of the granite known as Baltic Brown that can be found near the left end of the chart in Figure 4.6. Stone types on the higher end of radon emissions generally exhibited greater variability and account for less than 1% of the market share. The results for Nile Gold shown near the right end of Figure 4.6 are an example of a type of granite that exhibited greater variability among slabs.

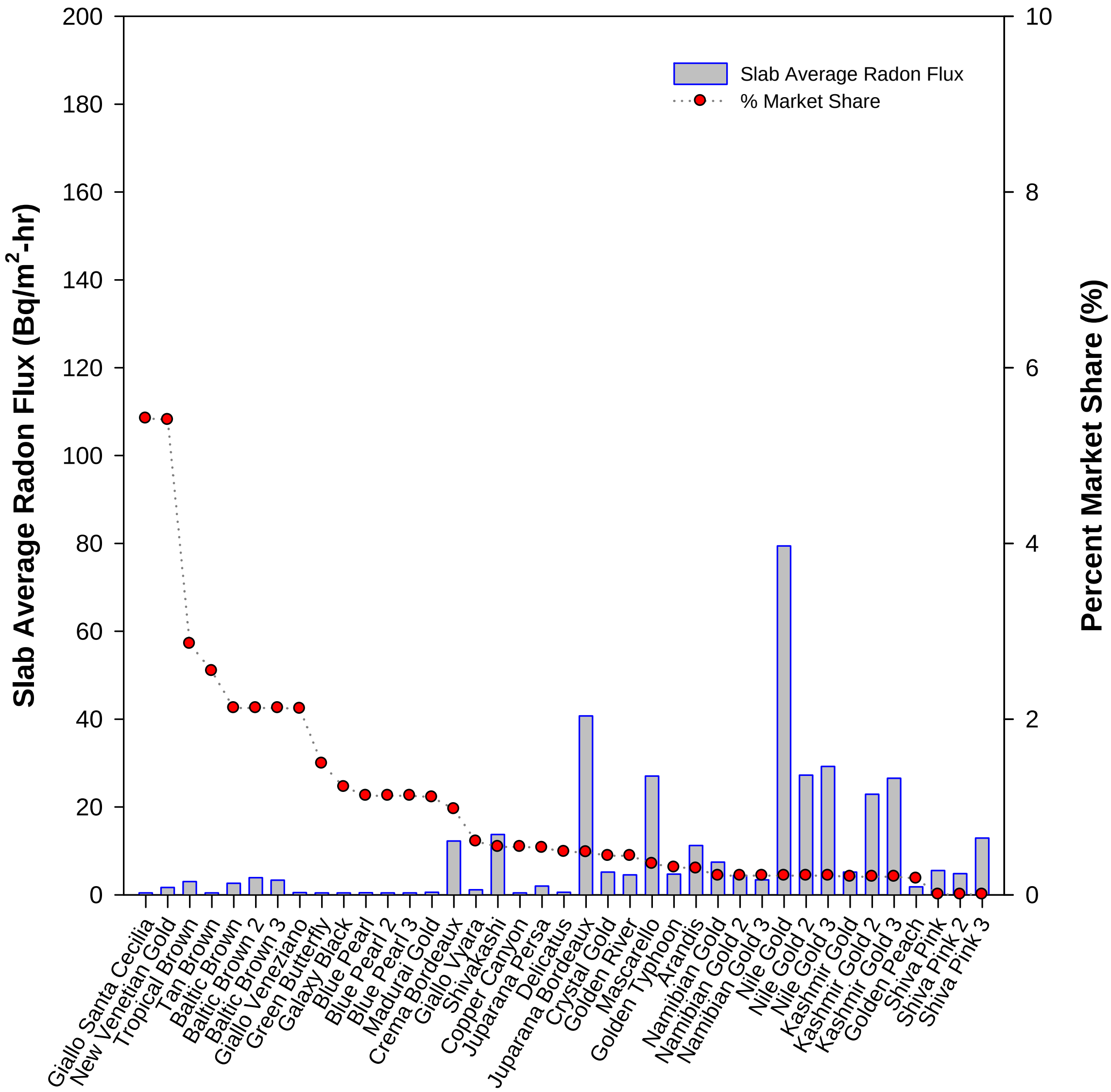

Figure 4.6 Radon Flux Measurements on Full-slab Samples, in Order of Decreasing Market Share

4.1.2.2 Discrete Sample Testing By Other Investigators

Radon flux reported in other studies of granite are summarized in this section along with estimates of the corresponding indoor radon concentrations that were developed using the approach described previously (see Section 3.3.2). The results of these other studies were divided into two groups. One group is composed of measurements of radon flux from discrete samples of granite countertops that were performed by (EMSL 2008). The other group is measurements of radon flux from discrete samples of granite building materials, including stones intended for exterior and interior use, that were compiled from publications in the scientific literature.

Radon flux results for 112 varieties of granite countertop measured by EMSL are summarized in Figure 4.7. The stone numbers and corresponding type of granite represented in the figure are cross-referenced in Appendix A (Table A.1). The varieties of granite included in the samples tested by EMSL account for approximately 80% of the sales of granite countertops in the U.S. Details of the testing procedure are available elsewhere (EMSL 2008). The average radon fluxes for types of granite reported by EMSL are encompassed within the range of fluxes observed for the full-slab testing (see Figure 4.6).

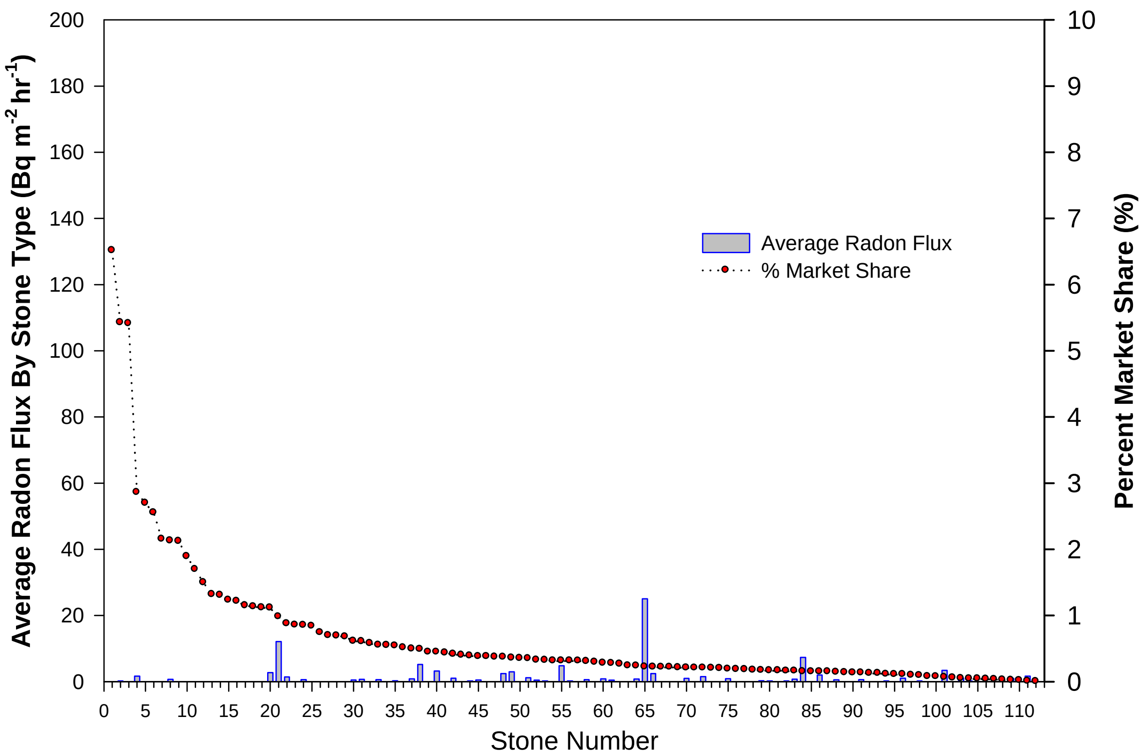

Figure 4.7 EMSL Analytical, Inc. Sample Testing—Measured Flux (Bq m^{-2} h^{-1}) by Decreasing Market Share (See Appendix Table A.1 for Corresponding Stone Type)

Radon flux measurements compiled from 14 peer-reviewed studies representing over 500 granite samples collected around the world are summarized in Figure 4.8 (Khan, et al. 1992; Chao and Tung 1999; al-Jarallah 2001; al-Jarallah, et al. 2001; El-Dine, et al. 2001; Petropoulos, et al. 2002; Fazal ur, et al. 2003; Stoulos, et al. 2003; Sundar, et al.

2003; Arafa 2004; al-Jarallah, et al. 2005; Osmanlioglu 2006; Singh, et al. 2008; Sonkawade, et al. 2008). These reports do not include detailed information on the types of granite that were evaluated, thus the flux measurements cannot be linked to specific types of granite sold as countertop. Moreover, descriptions contained in these papers indicate that the samples include granite intended for uses other than countertops in residences. The range of radon flux for granite reported in the published literature is well within the rates observed for full slabs and samples of granite countertops that are reported here. Thus, radon flux measured in this diverse cross-section of granite indicates that the samples of granite countertop evaluated in the present study contain emission rates not observed in prior studies.

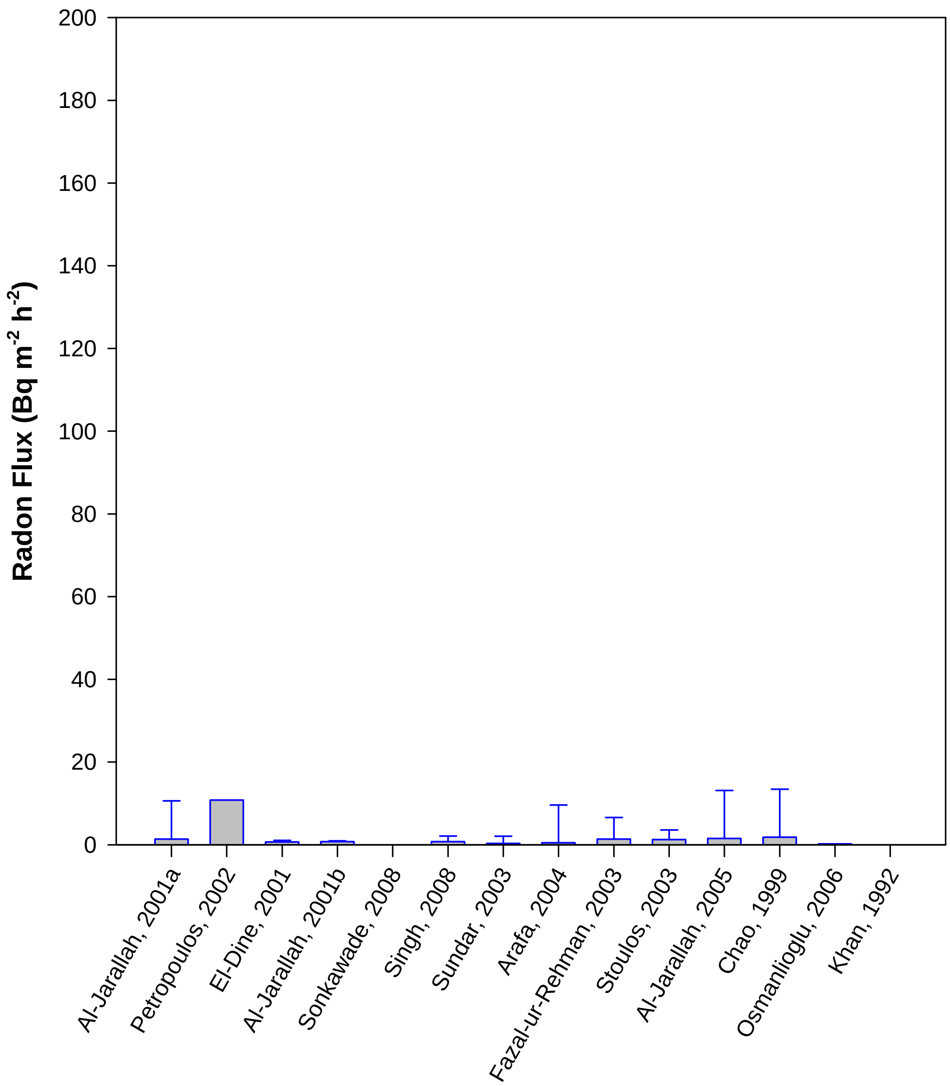

Figure 4.8 Peer-reviewed Journal Results—Average and Maximum Measured Flux (Bq m^{-2} h^{-1}) from 14 Peer-reviewed Studies

4.1.2.3 Effect of Backing and Polishing on Radon Emissions

Seven granite slabs were selected for additional testing to characterize radon emissions from the unpolished, bottom surface of the granite. Two of these stones had a fiberglass mesh backing on the bottom surface, commonly used for reinforcement. An important observation that could potentially affect estimates of radon contribution in a home for these granite slabs with a fiberglass mesh backing was that radon emissions from bottom surfaces with backings were low, even though these two slabs were on the high end of the distribution for radon emissions on the top surface. Additional testing would be required to determine if mesh backing helps mitigate the overall flux of radon from granite countertops.

Emissions of radon were similar between the polished and unpolished surfaces of the six granite slabs selected for this analysis. Further research would need to be conducted to determine if emission rates could vary between the two surfaces, perhaps as a result of differences in surface area and porosity from the polishing.

The average radon emission results in Figure 4.6 incorporate the underside results when measured. For several granite slabs that had backing on the underside but no corresponding measurements, the results presented did not have any correction applied and are presumed to provide an overestimate of emissions.

4.2 MODEL RESULTS

4.2.1 Modeled Dose

The effective dose equivalent rate modeled with MicroShield ranged from 0.005 mSv a^{-1} for the Galaxy Black stone to 0.06 mSv a^{-1} for the Namibian Gold stone type. The highest dose rate is more than 15 times lower than the 1 mSv a^{-1} criteria (NCRP 1993) and five times lower than the 0.3 mSv a^{-1} exemption criteria for building materials set forth by the EC (EC 1999). All estimated doses were also more than 10 times lower than measured doses on slabs in a vertical position. The lower modeled doses demonstrate the importance of accounting for distance and geometry when estimating dose from countertops which are in a horizontal position relative to a person (e.g., perpendicular exposure plane).

Table 4.1 Modeling of Exposure Rate and Effective Dose in a Model Kitchen and an Assumed High Exposure Scenario

Granite Type	Effective Dose Equivalent Rate (mSv a^{-1})*
Arandis	0.031
Baltic Brown	0.040
Galaxy Black	0.005
GreenButterfly	0.056
Mascarello	0.011
Namibian Gold	0.060
New Venetian Gold	0.034
Shiva Pink2	0.026
Shivakashi	0.036

mSv a^{-1} millisievert per annum (year)

* Effective dose equivalent rate based on exposures at each of four dose locations for one hour per day for 365 days.

When the model results that incorporated enriched areas were used to estimate annual doses (again assuming four hours were spent in the kitchen each day for 365 days, with two hours each day spent directly in front of two enriched areas) the maximum annual dose increased slightly from 0.06 mSv a^{-1} to 0.17 mSv a^{-1} and remained well below the 0.3 mSv a^{-1} exemption level set forth by the EC. These results indicate that annual doses remain within safe levels even when radiation "hot spots" are located near the countertop edge.

4.2.2 Radon Exposure Modeling

4.2.2.1 Estimated Indoor Radon Concentrations

Measured radon fluxes (Figure 4.6) were used to estimate indoor radon concentrations using the NIST indoor air model, CONTAM, assuming 50 square feet of granite countertop in the kitchen, 18 square feet of countertop divided among the bathrooms, and a total radon emitting surface area (including the top, bottom and edges of the granite slab) of approximately 144 square feet.

The predicted whole-house average radon concentrations for the four scenarios (old and new home, with both run under two ventilation rate conditions) are plotted for a range of typical radon emissions (radon flux rates) in Figure 4.9. The newer home (template

DH28) had higher predicted radon concentrations compared to the older home (template DH72), which can be explained by the lower air changes per hour in the newer home (0.2 ACH v. 0.4 – 0.5 ACH). .

As expected, estimated radon concentrations in the source rooms (kitchen, bathroom [not shown]) were higher than the corresponding whole house concentrations. The effect was greater for the natural ventilation scenario compared to the forced air scenario because forced air ventilation circulated air throughout the home and provided a relatively homogeneous distribution of radon in the home. This distribution of air throughout the home under the forced air ventilation scenario also increased whole-house radon concentration estimates (solid lines v. dotted lines in Figure 4.9). Annual whole-house average concentration has been reported by the EPA to be a reasonable estimate of annual exposure, since people spend the majority of their time at home in rooms other than the kitchen and bathrooms (EPA 1996a).

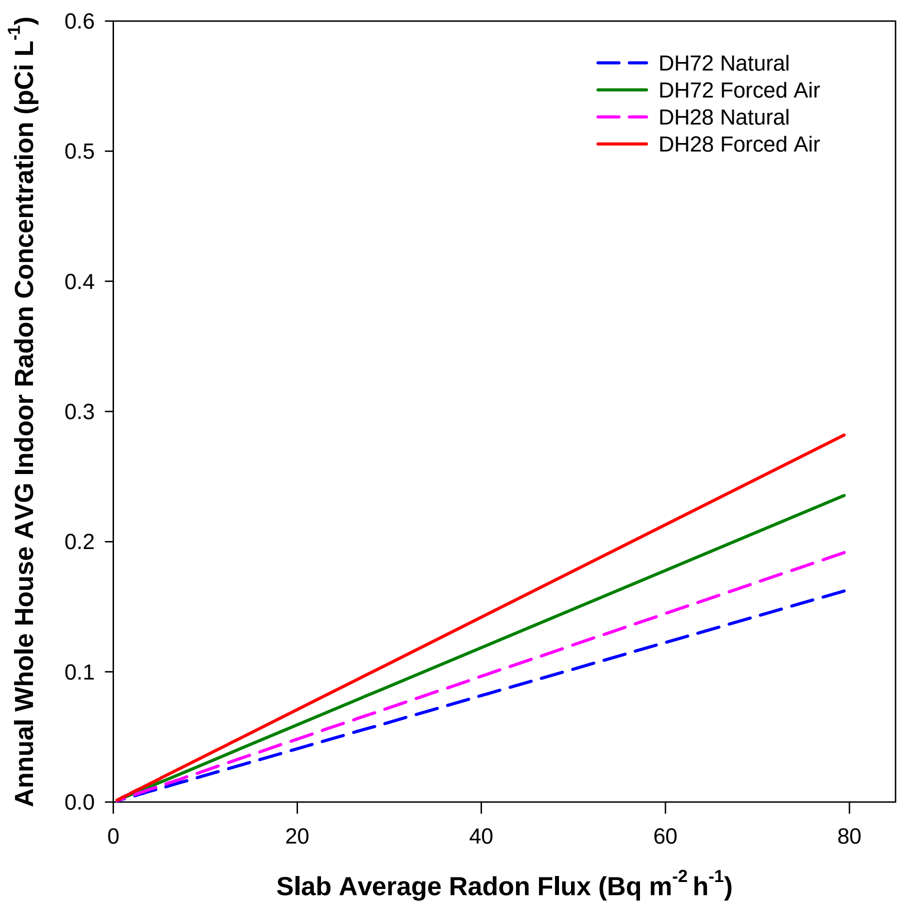

Figure 4.9 Predicted Indoor Radon Concentrations for a Range of Radon Emission Rates Under Four Housing Scenarios

4.2.2.2 Stone-type Specific Indoor Radon Contribution

The average results from the four modeling scenarios were combined with the specific radon flux measurements presented in Figure 4.6, Figure 4.7, and Figure 4.8 in order to predict the contribution to radon in indoor air (Figure 4.10, Figure 4.11, and Figure 4.12). The majority of granite slabs tested have measured radon emissions that correspond to predicted indoor radon concentrations less than 0.05 picocuries per liter (pCi L^{-1}). For the highest emitting stone in a tightly constructed house (worst case analysis for this study), the predicted whole-house annual average radon concentration is less than 0.3 pCi L^{-1}, a level similar to the average outdoor radon concentration in the U.S and well below the U.S. Environmental Protection Agency action level of 4.0 pCi L^{-1}. The average predicted contribution to indoor radon calculated from the results for individual slabs was 0.009 pCi L^{-1}. This result may be biased high because the types of granite subjected for full slab evaluation were selected to over-represent varieties with higher emission rates that were mentioned in media reports.

The six stone types for which radon flux was measured from three slabs are denoted with an asterisk (*) in Figure 4.10. The variability observed among the triplicate slabs is depicted with bars showing the range in slab average radon emissions. The granite types that constitute the majority of stones sold are on the lower end of radon emissions and exhibit the least variability while granite types with higher radon emissions have higher variability and account for less than 1% of the market share.

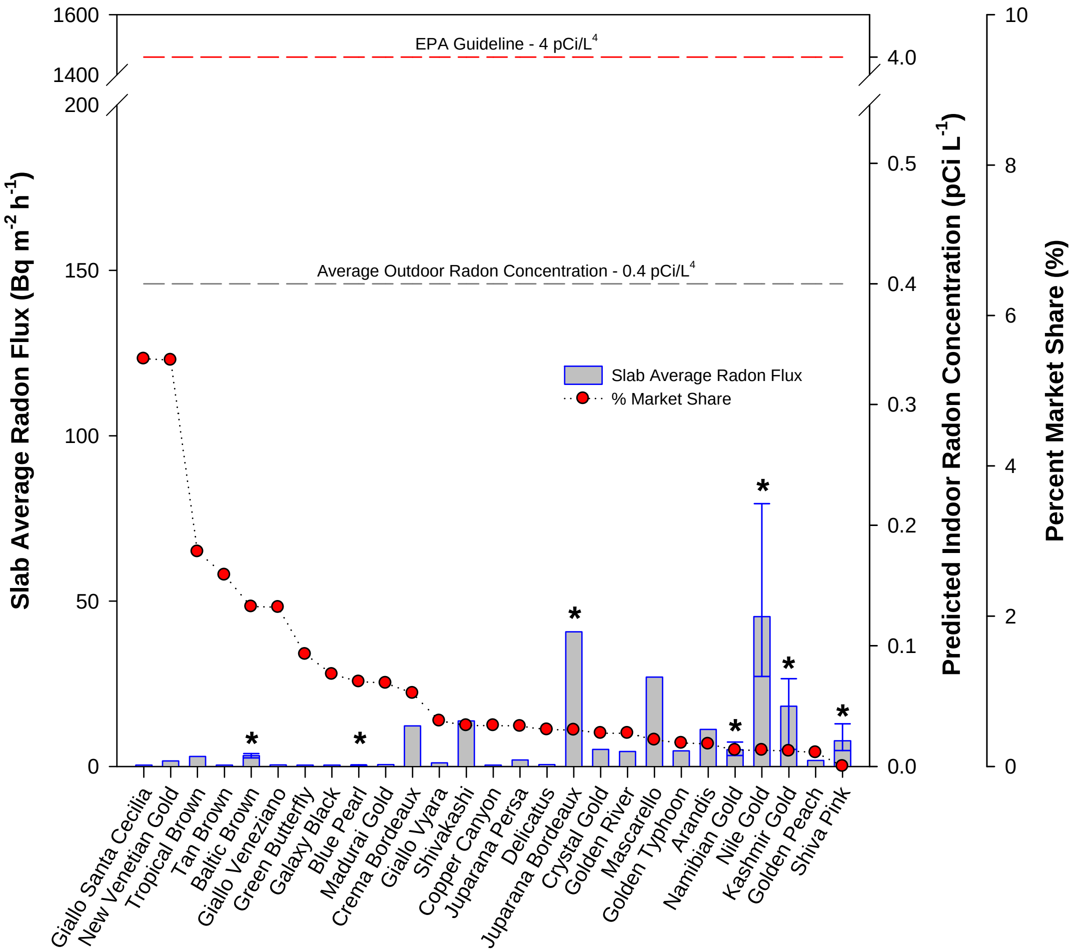

Figure 4.10 Radon Flux Measurements of Full Slabs. Predicted contribution to indoor radon concentrations by granite type, in order of decreasing market share. Bars denoted with a star (*) represent the range of test results from three slabs of that stone type.

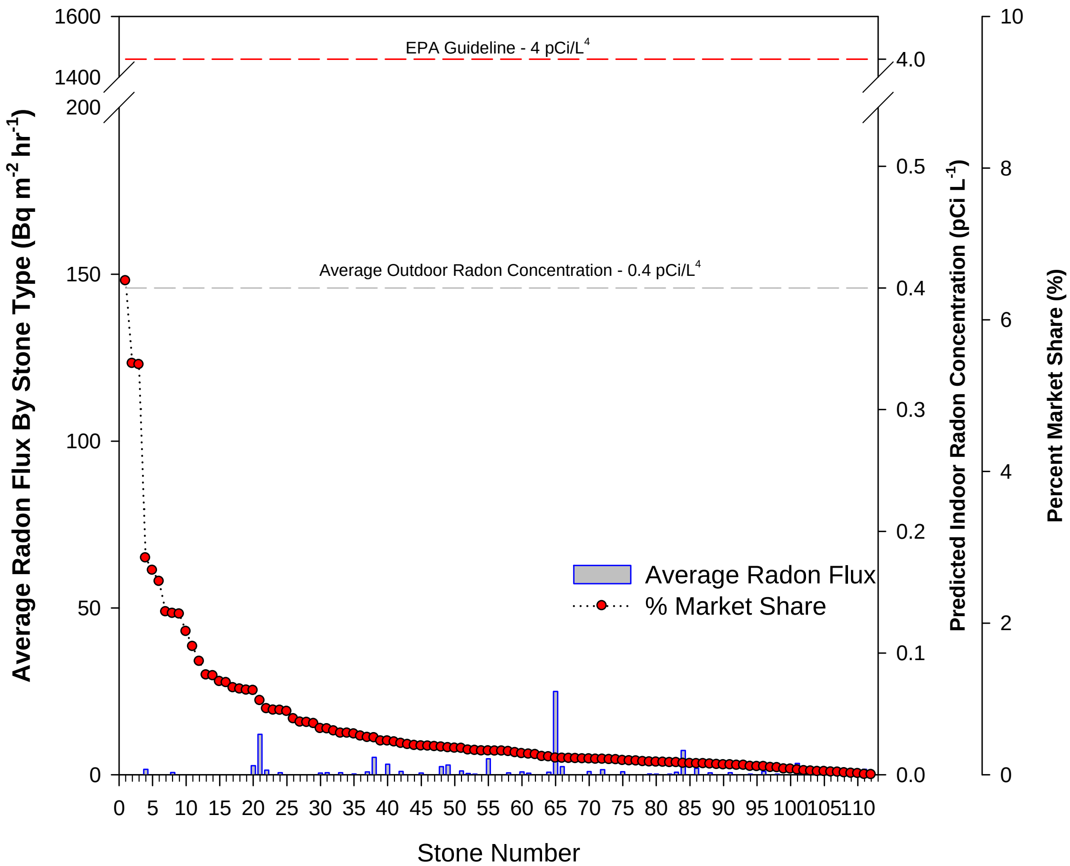

Figure 4.11 EMSL Analytical, Inc. Testing—Predicted Contribution to Indoor Radon Concentrations by Stone Type, in Order of Decreasing Market Share

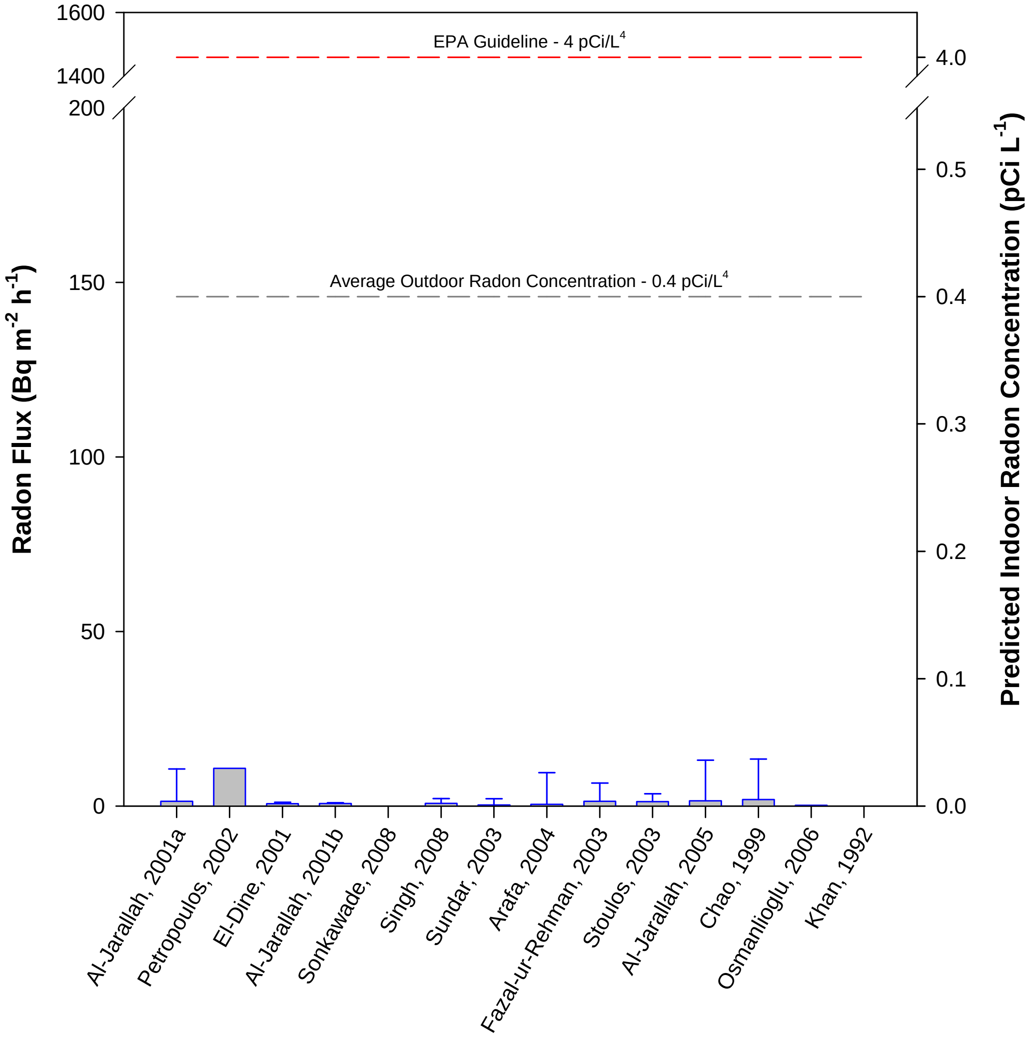

Figure 4.12 Peer-reviewed Journal Results—Predicted Average and Maximum Contribution to Indoor Radon Concentrations

4.3 RISK CHARACTERIZATION

Results from granite samples characterized in this study are representative of reported radiation and radon emission values and therefore constitute a valid sample for conducting an overall risk characterization for granite countertops. Measured and modeled values were used together with conservative assumptions (i.e., health-protective) of human activity patterns to examine whether granite used in homes as countertops poses a public health risk. As discussed previously, the magnitude of these risks were evaluated by comparisons to: (1) radiation screening criteria established by the European Commission; (2) dose limits for ionizing radiation recommended by the U.S. National Council for Radiation Protection and other international organizations; and

(3) guidance on radon exposure from the U.S. Environmental Protection Agency, the European Commission and other organizations.

4.3.1 Activity Concentration Index

Under the criteria established by the EC, all but one of the tested slabs had ACI values that call for a dose assessment based on typical granite countertop usages and scenarios. This finding runs counter to our measured and modeled doses (Section 4.1.1 and Section 4.2.1), which show that all slabs have an associated dose less than 0.3 mSv a^{-1} when used as a countertop in a home. These universally low doses should correspond to an ACI less than two for most of the granite slabs. The contradictory findings indicate that the ACI screening system is not appropriate for granite countertops, as ACI values of both 2 and 6 resulted in an almost 100% false positive rate in our samples. This is likely due to the very large surface area of granite (approximately 90 m^2 [970 square feet]) in the model room with dimensions of 4 m x 5 m x 2.8 m used by the EC. These findings indicate the need for a recalibration of the ACI screening system for granite countertops, as it would otherwise result in the performance of numerous unnecessary dose assessments for granite countertops.

4.3.2 Comparisons with Governmental Criteria

4.3.2.1 External Exposure to Ionizing Radiation

The measurements and modeling described above demonstrate that the slabs of granite countertop material included in this study are a very minor source of radiation dose within a home and present a negligible risk to human health. All of the dose rates determined from the measurements and modeling were well below the standard for a radiation dose attributable to a specific source of 1 mSV a^{-1} established by the NCRP, ICRP, and IAEA (NCRP 1993; IAEA 2003; ICRP 2005).

Additionally, dose associated with radiation emissions for all of the slabs tested are below the exemption limit for building materials of 0.3 mSv per year recommended by the European Commission (EC 1999). The United States has yet to establish an exemption level for building products based on radioactivity. The NRC and EPA site/soil cleanup standards are not appropriate for use in assessing stone countertops. Those standards are specifically intended for soil and combine the potential radiation dose that could be received through many pathways, including direct radiation, inhalation of dust, consumption of food grown in contaminated soil, and drinking contaminated water and therefore are not applicable when talking about solid stone countertops. As with any risk assessment, a specific scenario must be selected to represent ways that the area might be used after the cleanup so that proper assumptions can be employed to allow comparison with applicable standards. The same type of approach must be used in assessing the unique type of exposure scenarios that can occur in using stone countertops.

Even under a set of high exposure assumptions about potential exposure to radiation emissions from a granite countertop, the maximum estimate of dose derived from dosimeter measurements in our study is approximately 6 times below the dose criterion of 1 mSv a^{-1} established by the NCRP, ICRP, and IAEA. This conservative scenario was selected as the basis of our primary estimates of dose in order to yield results that are highly unlikely to underestimate actual upper end doses that could be possible for these slabs.

In addition to considering the conservatism of our primary estimates of dose, it is also important to note that the estimates incorporate variation in radioactivity among slabs and within a slab. The latter is of special interest considering that areas in some slabs were identified approximately 0.6 m^2 in size where gamma ray activity was as much as 6 times greater than the average for the slab. These enriched areas were found to comprise less than 10 percent, and more commonly less than 1 percent, of the overall radiation dose attributable to a slab because they only occurred in isolated spots of the slabs evaluated. The implications of this observation are important for characterizing risks of radiation exposure from granite countertops. For instance, even though a small area of high radioactivity on a granite countertop discovered during a radiological survey may become the focus of the surveyor, these locations are not indicative of an environmental hazard and were not an important contributor to radiation dose for the slabs included in this study.

The conservatism of the primary dose estimates was confirmed by modeling that was completed to account for a more realistic spatial orientation between a person and a granite countertop (see Section 4.2.1). The maximum dose obtained from modeling was approximately 16 times less than an annual effective dose of 1 mSv a^{-1}. This estimate is also conservative in that it maintained the assumption that a person was either in contact or within 6 inches of the counter, 4 hours a day, for 365 days per year. Modeling of hypothetical stones with two enriched areas also confirmed that the slabs evaluated in this study are not a source of radiation exposure that is meaningful in relation to the health based exposure limits discussed previously..

4.3.2.2 Radon

As described in Section 4.2.2, the concentration of radon in indoor air estimated from the average of the measured radon fluxes is approximately 400 times below the EPA action level for radon inside of homes (4 pCi L^{-1} compared to 0.01 pCi L^{-1}). Similarly, the indoor radon level predicted from the maximum flux of radon determined from a whole slab was approximately 17 times less than the EPA action level.

4.3.3 Normative Values

4.3.3.1 Ionizing Radiation

In addition to being well below government criteria, exposure to radiation estimated to arise from granite countertops is also low in comparison to background sources of radiation. An average person receives a total annual dose equivalent of approximately 3 mSv a^{-1} according to the NCRP (NCRP 1987b).

Radiation from the sun and other cosmic sources accounts for approximately 10% of the total dose (NCRP 1987b). Radiation doses from cosmic sources vary substantially with altitude. For example, the dose equivalent in cities at high elevation like Denver is approximately 0.5 mSv a^{-1}. In comparison, the dose received from cosmic sources in cities at sea level like San Diego are approximately 0.25 mSv a^{-1}, about 50% lower. The conservative estimates of dose associated with granite countertops that we estimated for a model kitchen (see Table 4.1) are approximately 0.03 mSv a^{-1} and only about one-eighth of the change in dose that would result for a person who relocates from a coastal area to the Rocky Mountains.

People are also exposed to radiation from cosmic sources while traveling in airplanes. The NCRP has estimated that a person receives a dose equivalent of approximately 0.005 mSv per hour while in transit aboard a plane (NCRP 1987b). The dose received during a single transcontinental flight of 5 to 6 hours is therefore roughly equal to the conservative estimates of dose attributable to emissions from a granite countertop over an entire year.

Our estimates of dose equivalent from external exposure to radiation associated with granite countertops are also well within the range of doses attributed to a wide range of consumer products. For instance, familiar building materials such as concrete and gypsum board have been reported to produce a dose of about 0.04 mSv per year for a typical person (NCRP 1987c). The radiation dose from decay products of radon that is naturally present in domestic water supplies is also estimated to be in the same range (NCRP 1987c).

4.3.3.2 Radon

As noted earlier, radon gas is a natural constituent of outdoor and indoor air. The flux of radon from soil is approximately 65 Bq m^{-2} h^{-1} (1.8 pCi L^{-1}) (NCRP 1987a) which is greater than the slab average flux observed for all but two of the stones that we evaluated on a full-slab basis. Concentrations of radon in outdoor air produced by emissions of radon from soil range from approximately 3.7 to 37 Bq m^{-3} (0.1 to 1 pCi L^{-1}) (NCRP 1987a) with a national average of 14.8 Bq m^{-3} (0.4 pCi L^{-1}) (EPA 1993a). The average concentration of radon in indoor air attributable to emissions from granite countertops was conservatively estimated to be approximately 0.3 Bq m^{-3} (0.009 pCi L^{-1}), approximately 40 times lower than the typical level of radon in outdoor air of the U.S.

Radon levels inside of homes are typically greater than levels in outdoor air. The average concentration of radon in indoor air of U.S. homes is reported be 1.3 pCi L^{-1} with a median level of 0.7 pCi L^{-1} (Marcinowski, et al. 1994). Approximately 6% of homes in America are estimated to have radon concentrations in indoor that exceed the action level of 148 Bq m^{-3} (4 pCi L^{-1}) established by the EPA (Marcinowski, et al. 1994), with some areas of the country having 30-40% of homes above this threshold (Steck, et al.

1999). Based on the results of our study, a typical granite countertop would be a negligible source of radon for homes that exceed the EPA action level.

Sources of radon that account for the difference between levels in outdoor and indoor air include infiltration of radon gas from soil through foundations, volatilization from domestic water use, and emissions from building materials such as concrete, gypsum board, and granite (NCRP 1987a). Information on radon emissions was compiled for concrete, gypsum and brick reported in the scientific literature to facilitate a comparison with emissions from granite countertops observed in this study.

The average radon emissions, calculated based from over 300 measurements of these common building materials, are presented in Table 4.2 along with the average results for granite countertops from this study (Ingersoll 1983; Mustonen 1984; Chao and Tung 1999; Petropoulos, et al. 2002; Stoulos, et al. 2003; Maged and Ashraf 2005; de Jong, et al. 2006; Kobeissi, et al. 2008; Ngachin, et al. 2008; Sonkawade, et al. 2008). While the average radon emission rates for these building materials are similar to granite countertops, it is important to consider the mass of material in a home that may be emitting radon. As demonstrated in Table 4.2, the surface area potentially emitting radon may be 40 times higher for gypsum than for granite countertops, and 10 times higher for concrete. For a home constructed of concrete with gypsum board walls, emissions of radon from granite countertops would be a minor fraction of total radon emissions for the residence.

Table 4.2 Comparison of Average Radon Emissions (Bq m^{-2} h^{-1}) and Amounts Used in Typical Homes for Building Materials (concrete, gypsum, and brick) and Granite Countertops from this Study

Material		Use	Flux (Bq m^{-2} h^{-1})	Estimated Surface Area (m^2)
Granite	Full Slab	Kitchen + Bathroom Countertop	3.3	14
	Discrete		0.6	
Gypsum		Wall	4.1	554
		Ceiling		276
Concrete		Floor	8.0	138

Bq m^{-2} h^{-1} becquerel per square meter per hour
m^2 square meter

4.3.4 Estimated Health Risks

The EPA has estimated that the average lifetime risk of lung cancer mortality associated with radon in indoor air is 1.6×10^{-4} per Bq m^{-3} (5.8 x 10-3 per pCi L^{-1}) of radon exposure (EPA 2003). Based on the national average concentration of radon in indoor air of 48.1 Bq m^{-3} (1.3 pCi L^{-1}) (Marcinowski, et al. 1994), the average American has a 7.5×10^{-3} risk of fatal lung cancer as a result of exposure to background levels of radon exposure. In other words, the typical American faces a 0.75% chance of dying from lung cancer as a result of background levels of radon at home. In comparison, the additional risk associated with lifetime exposure to radon emitted from a typical slab of granite countertop observed in our study is 5.2×10^{-5} (0.005%). This means that the change in risk from having a typical granite countertop in a home would change from 0.75% to 0.755%.

This level of risk is less than many familiar events or activities in our everyday environment. For instance, the lifetime risk of dying from falling out of bed based on mortality data compiled by the CDC for 1999-2005 is 1.4×10^{-4}, approximately 3 times higher than the risk posed by a typical installation of granite countertop based on our measurements (CDC 2008). Two other unlikely, yet familiar, causes of death with a similar likelihood are falling by tripping, slipping, and stumbling (1.4×10^{-4}) and exposure to forces in nature such as extreme temperature, powerful storms, and lightning (3.5×10^{-4}).

5.0 CONCLUSIONS

EH&E has completed a detailed evaluation of potential human health risks of naturally occurring radioactive materials in granite countertops. The extensive measurements and rigorous mathematical modeling conducted to date indicate that (i) external doses of ionizing radiation emitted from granite countertops are well below levels that would pose a health concern and (ii) contributions from granite countertops to radon levels in homes are lower than background levels of radon exposure typically found outdoors and indoors.

- Radon levels associated with emissions from granite countertops in homes are low in comparison to typical background levels of radon exposure. In other words, natural stone is a minor contributor to concentrations of radon gas within homes. These findings are consistent with an earlier review of the scientific literature that EH&E performed (EH&E 2008).

- External dose associated with radiation emissions for all of the slabs tested are well below health-protective guidelines, including the exemption limit of 0.3 mSv per year recommended by the European Commission. The United States has yet to establish an exemption level for building products based on radioactivity to our knowledge.

- A portion of stones used as countertops may contain limited areas that are enriched in radioactive materials relative to the remainder of the slab. The areas of enrichment in the stones evaluated for this study make up a small proportion of the stone, on the order of less than 10 percent of the surface area. Detailed measurements of these enriched areas showed that they make a negligible contribution to potential doses of ionizing radiation.

- Assessing exposure to radon and radiation requires accounting for duration and frequency of exposure, not just absolute magnitude. Additionally, careful consideration of several key parameters is warranted. For radon, measurements of radon flux from a countertop must account for variability across the countertop surface, the effect of any backing material on the stone, and diffusion through the slab. It is critical that ventilation is accounted for when estimating radon concentrations in indoor air from measurements of radon emissions from stones. For radiation, distance and geometry must be incorporated into dose assessments.

- While significant variability was observed across stone types, the stones at the lower end of radon emissions were found to account for the vast majority of sales and also exhibited little variability among slabs. The varieties of granite countertop that exhibited the greatest variability of radon flux among slabs represent a small fraction of the U.S. market.

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APPENDIX A
STONE NUMBERS AND CORRESPONDING STONE TYPE

Table A.1 List of Stone Numbers and Corresponding Stone Type from Figures 4.7 and 4.9

Stone No.	Stone Type	Stone No.	Stone Type	Stone No.	Stone Type
1	Verde Ubatuba	39	Golden Crystal	77	Zimbabwe Black
2	Santa Cecilia	40	Yellow River	78	Yellow Bamboo
3	New Venetian Gold	41	Bianco Romano	79	Ivory Coast
4	Tropical Brown	42	Typhoon Green	80	African Ivory
5	Giallo Ornamental	43	Key West Gold	81	Coffee Brown
6	Tan Brown	44	Dakota Mahogany	82	Magma Gold
7	Premium Black	45	Amber Fantasy	83	Wild Sea
8	Baltic Brown	46	Emerald Pearl	84	Sage Brush
9	Giallo Veneziano	47	Yellow Star	85	Colonial Gold
10	Giallo Portofina	48	Juparana Golden	86	Rosewood
11	Suede	49	Mascarello	87	African Fantasy
12	Verde Butterfly	50	Calacatta	88	Delerium
13	New Caledonia	51	Espirito Santo	89	Cambrian Black
14	Verde San Francisco	52	Sucuri	90	Peach Red
15	Galaxy Black	53	Coral	91	Gold Coast
16	Sapphire Blue	54	Blues in the Night	92	Santa Fe Brown
17	Giallo Fiorito	55	Typhoon Bordeaux	93	Paradiso Dark
18	Blue Pearl	56	Giallo Arctic	94	Red Montana
19	Tropic Green	57	Volga Blue	95	Solarius
20	Madura Gold	58	Autumn Harmony	96	Castor Blue
21	Crema Bordeaux	59	Azul Bahia	97	Steel Gray
22	Bianco Antico	60	Carioca Gold	98	Juparana Lorean
23	Impala Black	61	Pauline Grey	99	Sodalite Blue
24	Crema Caramel	62	Juparana Fantastico	100	Rosa Beta
25	Giallo Napolean	63	Nepson Gold	101	Desert Dream
26	Black Pearl	64	Gold Leaf	102	Virginia Mist
27	Giallo Vicenza	65	Niagara Gold	103	Shiva Cream
28	Pokarna Green	66	Juparana Tier	104	Almond Mauve
29	Luna Pearl	67	Peacock	105	Nordic Black
30	Vyara	68	Red Dragon	106	Jurassic Green
31	Azul Platino	69	Costa Smeralda	107	African Tapestry
32	Bainbrook Brown	70	Kashmir Gold	108	Orion
33	Shivakashi	71	Labrador Antique	109	Ivory Gold
34	Copper Canyon	72	Seafoam Green	110	Juparana Tier Bordeaux
35	Juparana Persa	73	Marinace Green	111	Gabarone
36	Verde Peacock	74	Juparana Colombo	112	Zambezi
37	Delicatus	75	Golden Beach		
38	Bordeaux	76	Magma		