AN ABSTRACT OF THE THESIS OF

Krystina M. Tack for the degree of Master of Science in Radiation Health Physics presented on March 1, 2006.

Title: Determining the Bioavailability of Soil-Associated Radium Using In Vitro Methodology.

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Kathryn A. Higley

Soil that is contaminated with radioactive elements poses an exposure hazard to those whom may take up temporary or permanent residence on such a site. Of particular interest is the internal exposure from ingestion of this radioactive soil. Although most ingestion of soil is inadvertent, usually being attached to foodstuffs that are not properly cleaned, it is possible that a person might consume a larger quantity. Childhood soil ingestion from simple hand-to-mouth activities is one explanation for this larger intake, as well as geophagia (eating dirt) or pica (craving and eating non-food items). The assumption that any person might consume a "mouthful" of dirt is a rare but possible occurrence that, when analyzed, will help with decisions about safe contamination levels of soil.

Samples of soils contaminated with radium-226 were sent from an engineering and environmental firm to Oregon State University's Department of Nuclear

Engineering and Radiation Health Physics for assessment. The analysis of the samples was aimed at the determination of bioavailability and bioaccessibility of the radioactive species found in the soils. Subsequent site remediation actions for the New Jersey-based project would be partially dictated by the results of Oregon State University's testing.

Initially, the soils were tested for the presence of carbonates, for leachability of radioactivity in water and in acid, and for particle size distribution, i.e., soil type. Each of the eight samples was then subjected to a stomach/intestinal analogue to determine how much of the radioactivity would be transferred to solution upon human ingestion, (bioaccessibility). Mass balance and gamma spectrometry outputs for the soil samples before and after the digestion was one way the loss to solution was assessed. Another method to determine the loss of radioactivity to solution was to count aliquots of the digestive fluids in a high purity germanium detector, using a library of only radium isotopes and their progeny to locate peaks. The combination of results from mass balance and gamma spectrometry outputs allowed for OSU's researchers to determine the bioaccessibility of each soil's radioactive components. Using the determined bioaccessibility and previous animal models, the determination of bioavailability varied between the samples, from zero to 28% of the total initial radioactivity in the samples.

A hot particle estimation of the dose from the non-bioavailable portion of the samples yielded a high dose to a small number of cells. Assuming ingestion of the most radioactive sample, (Sum-03a), the amount of damaged (killed) tissue in each section of the gastrointestinal tract was estimated to be less than 0.0407 cm^3. This

small volume of tissue is not likely to result in evident damage as the healthy human gastrointestinal tract regenerates all surface cells approximately every six days and most items are resident in the digestive system for less than 48 hours.

Determining the Bioavailability of Soil-Associated Radium Using In Vitro Methodology

by

Krystina M. Tack

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I understand that my thesis will become part of the permanent collection of Oregon State University libraries. My signature below authorizes release of my thesis to any reader upon request.

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Krystina M. Tack, Author

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Jerry, Tish, Shelly and Angie

DETERMINING THE BIOAVAILABILITY OF SOIL-ASSOCIATED RADIUM USING IN VITRO METHODOLOGY

1. INTRODUCTION

Naturally occurring radium is found in almost all rock, water, soil, vegetation, and animals as it is formed by the decay of uranium and thorium. Industrial uses for uranium and radium can cause the natural level of radium to increase in certain areas. In this case, the radium is termed Technologically Enhanced Naturally Occurring Radioactive Material or TENORM. Some of the historical uses for radium in industries have been luminescent paints for gauges and dials, cancer treatment through seed implantation, as a flaw detector for metallic parts, and it was even added to the tips of lightning rods to increase the efficiency of ionizing the surrounding air. (EPA, 2005a)

Any process that concentrates uranium or thorium (the parent nuclides of radium) in a particular location will increase the levels of radium as well. Understanding the process that increased the level of radium in a location is not as important as protecting the people and the environment from the effects of this alpha emitting radiation. Decommissioning or clean-up of sites where there has been contamination of radium or other hazardous material is often funded by the Superfund program, a federal trust fund of $8.5 billion. (EPA, 2005b) This money is utilized for removing dangerous materials, backfilling the land, the proper disposal of those

materials, and restructuring the use of any site so that it will meet the public standards set by the Environmental Protection Agency (EPA).

Some places in the United States, such as New Jersey, have more than a few Superfund sites. New Jersey is the former site of many industrial processes that have helped severely contaminate its land, which explains why it is home for well over 100 Superfund sites. (EPA, 2005b) Radium contamination in New Jersey has been a problem for years, mostly in the soil and subsequently the drinking water. Because radium decays by alpha and gamma emission, its presence in the environment is not significant unless it is deposited internally (or it becomes radon gas, not part of this analysis). The range of alpha particles is no more than a few centimeters in air, but when they are deposited internally (as is the case with contaminated drinking water or ingestion as a solid), these alphas contribute significantly to the radiation dose of the consumer.

Ingestion of soil by humans is usually inadvertent, being attached to foodstuffs that have not been thoroughly cleaned or from soiled-hand transfer to the mouth. There are special instances wherein larger amounts of soil may be ingested such as small child hand-to-mouth activities or in the rare case of geophagia.[1] When this dirt is also contaminated with radionuclides, the internal dose from those emissions is of increased concern from a radiation protection standpoint. The level of radioactivity that is safe for public exposure must include the rare but significant consideration of potential ingestion of contaminated soil or rocks.

[1] Geophagia (or pica) is the eating of non-nutritive substances, such as dirt or clay at a developmental stage when the behavior is inappropriate. (4Therapy, 2005)

The radiation exposure threat posed by the ingestion of soil associated radium and its progeny was assessed in this analysis. By determining the amount of dissociated radioactive species from the solid soil, the amount of radioactivity available for systemic distribution can be estimated. With this knowledge and the need to adhere to limits for exposure to the general public, the safe level of contamination and land use for one New Jersey site was conservatively determined.

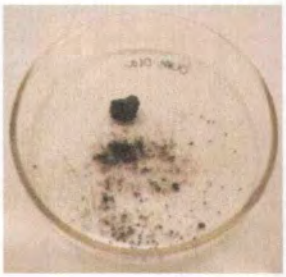
2. BACKGROUND

2.1 MACTEC SOILS

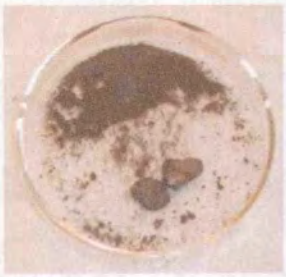
MACTEC is an engineering, environmental and remedial construction firm, originally started in 1975 to provide services to the electric utility and nuclear power plant construction industries. Today, MACTEC's mission is to help their clients manage risk while "...protect(ing) the environment by delivering value driven engineering, environmental and construction services that impact the world in which we live." (MACTEC, 2006) Approximately 2-5% of MACTEC's United States work is on Superfund sites.[2]

MACTEC contracted the Department of Radiation Health Physics at Oregon State University (OSU) to determine the potential hazard posed by ingestion of radioactively contaminated soils. These "soil" samples were collected by MACTEC at an undisclosed location in New Jersey. The samples contained dirt, sand, clay, organic matter, and even some rocks (See Figure 2.1). For ease of explanation, these samples will be referred to simply as soils from this point forward. The following images and listed total sample weights were recorded for each of the MACTEC labeled soils. The soils were later fractionated for analytical processes.

[2] Information via January 20, 2006 email correspondence with Vanessa Campbell from MACTEC @ VCAMPBELL@mactec.com.



Sum-01a – 0.846g



Sum-02a – 3.919g

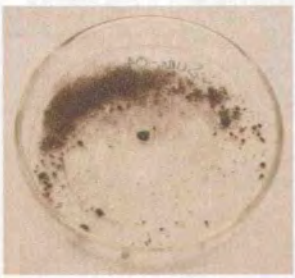
Sum-02b – 1.863g

Sum-02c – 1.870g

Sum-02d – 20.464g

Sum-02e – 0.719g

Sum-03a – 0.202g



Sum-04a – 0.704g

Note:
Samples are shown in 60 mm Petri dishes w/ total sample mass in grams

Figure 2.1 Preliminary images of soil samples from MACTEC.

2.2 GAMMA SPECTROMETRY

Radionuclides often emit gamma particles upon their decay, which can then be used to find and quantify them. When the samples arrived at OSU, it was already known that they were radioactive but the nuclides present were undisclosed. The use

of a high purity germanium gamma spectrometer revealed that the samples each contained radium-226 (^{226}Ra), which has a single gamma at 186 keV emitted with an abundance of 32.8%. When ^{226}Ra decays, its first daughter is radon-222 (^{222}Rn) which is an alpha emitting gas, but several daughter progeny have gammas for gamma spectrometry. Those used in this analysis were lead-214 (^{214}Pb) and bismuth-214 (^{214}Bi).

2.3 RADIUM-226 CONSIDERATIONS

Marie Curie noted upon her discovery of radium that it was "a new element with very curious properties." (IAEA, 1990a) It was found to be one million times more radioactive than uranium and Curie noted it to be chemically similar to barium[3]. ^{226}Ra and ^{228}Ra are the most abundant naturally occurring isotopes of radium, resulting from the decay of uranium-238 (^{238}U) and thorium-232 (^{232}Th), respectively. The heaviest of the alkaline earth group of metals, the name radium is most often taken to mean radium-226 as it is the most important isotope occurring in nature due to its long half life and naturally abundant parent (U-238). (IAEA, 1990b)

Radium is present in all foods and the average person in the United States consumes from 1.7 to 2.3 pCi of ^{226}Ra per day (from a tri-city study). (ICRP, 1975) There is a fairly extensive set of data for adult distribution and retention of ^{226}Ra in

[3] The unit Curie was actually defined by the Radiology Congress of 1910 to be what Curie called the "quantity of emanation in equilibrium with one gram of radium," which was later defined numerically to be 3.7 x 10^{10} disintegrations per second. (IAEA, 1990a)

humans due to its former use in many industrial processes. The behavior of radium in laboratory animals and the fact that barium performs similarly in the body allow for better estimations of body performance of ingested radium. Laboratory animal data can be extrapolated to humans, but one must understand that secretion of fluids into the GI tract is much higher in humans, which leads to less radium recycling, meaning a smaller net retention in humans. (ICRP, 1993)

The use of a gamma spectrometer to identify and quantify ^{226}Ra and its progeny is useful, but it is important to remember that alpha particles are the primary method of decay for radium and its progeny with abundances nearing 100%. ^{226}Ra decays by alpha emission to radon-222 (^{222}Rn) which is a gas. In an open environment, this is useful for dispersion of the progeny. However, if the ^{226}Ra decays to ^{222}Rn in an enclosed environment (as would be the case with ingestion), the energy for each alpha will be deposited in the tissue and following progeny will contribute to the dose. The gaseous progeny are also a consideration in the analytical testing making time stamping very important as the in-growth of progeny can alter the gamma spectral output.[4]

Table 2.1 was created to display the decay scheme for ^{226}Ra and also list the energies associated with those decays. The physical half life of ^{226}Ra is approximately 1600 years depending on the source of the information.[5] The biological half life of ^{226}Ra is only about 900 days, making the effective half life of ^{226}Ra in the body 900

[4] In this analysis, all samples were vented in a fume hood immediately prior to sealing and counting.
[5] The radiological half life of Ra-226 was found to be 1599 years, 1600 years and 1622years. The IAEA uses 1622 years and the ICRP uses 1600 years.

days. Approximately 80% of ingested radium is excreted with the feces, but the 20% that enters the blood stream is preferentially deposited in the skeleton. (EPA, 2005a) This is not surprising because radium is chemically similar to calcium. What should be recognized, however, is that the more calcium need an individual has, the more radium will be absorbed (i.e., infants and growing children will absorb more radium than adults who are not growing).

TABLE 2.1 ^{226}Ra and its progeny with decay mode, energy and associated gammas with their respective abundances.

Nuclide	Half Life	Decay Mode	Energy (MeV)	Associated Gamma Energy in keV (%abund.)
Radium-226	1622 y	α	4.78	186.2 (32.8%)
Radon-222	3.825 d	α	5.49	510 (0.07%)
Polonium-218	3.05 m	α	6.00	510
Lead-214	26.8 m	β	0.67	295 (19%), 352 (36%)
Bismuth-214	19.7 m	β	3.27	609, 1120, 1764 (each 17%)
Polonium-214	167.7 μs	α	7.69	799 (0.014%)
Lead-210	22.3 y	β	0.017	46.5 (4%)
Bismuth-210	5.0d	β	1.161	266.2, 305.2
Polonium-210	138.4 d	α	5.305	-
Lead-206	stable	-	-	-

(IAEA, 1990a)

2.4 OVERVIEW OF METHODOLOGY

An analytical plan was constructed with the initial task of determining the radionuclide species present in the eight soil samples using gamma spectroscopy. The

type of soil was determined by particle size distribution, where the percentage of various sized particles was matched with a published soil description. Soil pH (as well as soil type) is a good predictor of soil performance in the environment, so a simple water-soil slurry analysis of the pH was completed. Gross analyses of both acid and water leachability were then completed in an effort to estimate possible solubility in the digestive tract of humans.

A representative soil was selected to test for the presence of carbonates or calcite, which could predict behavior of soils in the environment or in the digestive tract. An analogue of the human digestive tract was created using a hybrid of previous methodologies. Each of the eight samples was put through "digestion" in this *in vitro* model. Gamma spectroscopy before the soil was digested, after digestion and after drying the sample yielded the portion of the nuclides that were transferred to solution, i.e., bioavailable. Finally, a sample calculation was carried out to predict that dose to portions of the digestive tract from unabsorbed (non-bioavailable) radium as it would leave the body with the feces.

2.5 PARTICLE SIZE ANALYSIS

Assessment of the soils first involved particle size analysis to determine the type of soil. The method utilized was one set forth by the United States Department of Agriculture (USDA) Soil Survey Laboratory Methods Manual. (USDA, 2004) By

determining the size distribution of the particles in the soils, the type, texture, structure, and to some degree the chemistry of the soil is known.

2.6 SOIL PH

The second step of the soil sample analysis would determine the soil pH which is the negative log of the hydrogen ion concentration and is represented on a scale from zero to fourteen. A low number for pH means that there are a high number of hydrogen ions and a soil is said to be acidic. Conversely, a high pH indicates a lesser number of hydrogen ions and the soil is termed basic or alkaline. Knowing the soil's pH helps determine its absorptive qualities in a human. The following is a list of pH descriptions and ranges for some common substances:

Extremely acidic [< 4.5] – lemon (2.5), stomach acid (2.0)

Slightly acidic [6.1 – 6.5] – salmon (6.2), cow's milk (6.5)

Neutral [6.6 – 7.3] – saliva (6.6–7.3), blood (7.3)

Moderately alkaline [7.9 – 8.4] – sodium bicarbonate (8.4)

Very strongly alkaline [>9.1] – milk of magnesia (10.5), ammonia (11.1), lime (12)

Acidic soils tend to have minerals and nutrients that are more soluble or available than those soils that are neutral or slightly alkaline. (SUNY-ESF, 2006)

2.7 CARBONATES AND SOLUBILITY

The next step in sample analysis was to determine the presence of carbonates. The simple test consists of adding hydrochloric acid drop-wise into a dried soil sample and watching for bubbling. If there were carbonates in the samples, these soils would be more easily dissolved by acids and therefore more soluble in the stomach environment of a human.

Gross solubility determination was another quick test done prior to more destructive testing of the soil samples. The radioactivity transferred to solution in both the deionized water slurry and the hydrochloric acid (HCl) slurry was said to be the soluble component of each sample. The degree of solubility is a major factor in the accessibility of substances in the gastrointestinal tract. In essence, if it can be transferred into solution, it can more readily be absorbed in the small intestine of a human.

2.8 SIMULATING THE HUMAN DIGESTIVE TRACT

After determining the type of soil the samples contained, the pH of the dry soil, the potential solubility of those soils in the environment (water slurry) and the estimated gross solubility in the stomach (HCl slurry), the final step was to subject the samples to an artificial human digestive tract model. Very limited work had been done previously to determine radionuclide release in a modeled human GI tract. Two studies were analyzed, NRPB W-17: "*The Availability of Soil-Associated*

Radionuclides for Uptake after Inadvertent Ingestion by Humans," (NRPB, 2002) and *"The Bioaccessibility of Selected Radionuclides and Heavy Metals: An Investigation of Bioaccessibility, Bioavailability and Natural Soil Characteristics."* (Ellickson, 2001) A hybrid assessment methodology was created using components from both works.

In vitro modeling of the human digestive tract must achieve nearly physiologic conditions to validate the data gleaned from such work. The mechanical destruction of the samples that would occur in the mouth from the teeth and tongue should be done first, followed by submersion of such a sample in a gastric (stomach) environment. Consumed objects such as food are usually transported from the mouth to the stomach in about 5 seconds. (Ward, 2005) A healthy human stomach has a pH of approximately 2 and food can be kept in this organ for up to three hours. The average mixed meal is present in the stomach for about 1.5 hours. (Gastro, 2006)

Very little absorption of nutrients occurs in the stomach, but the substance being digested is turned into a semi-fluid paste called chyme. This chyme is moved a little at a time into the small intestine (SI). The emptying rate of the stomach is dependent on the type of food (substance) being digested. Liquids will pass through the stomach fairly quickly, but solids remain until they are wholly mixed with gastric fluids and are transported to the small intestine. In the small intestine, foods that are high in fat may remain as long as six hours, while proteins are moved though in three

hours, and carbohydrates are moved out of the stomach and through the small intestine at a faster rate than proteins. (Shier, 2004)

Following the stomach phase of analysis, the model should then simulate the digestion that would take place in the intestine, specifically the small intestine. The small intestine of the human is a more basic environment with a pH that is just higher than neutral, around 7.5. This organ is the most important component in the human digestive tract as it is the site of almost all nutrient absorption.

The small intestine is lined with villi that increase the total internal surface area to 250 square meters—the size of a tennis court. (About.com, 2006) It then comes as no surprise that very little material that can be absorbed will reach the other end (of the small intestine). Absorption of some ions like those of sodium, chloride, potassium, nitrate and bicarbonate happens easily in the small intestine. However, the ions of magnesium, sulfate and calcium are poorly absorbed. (Shier, 2004) The transport time for substances through the adult small intestine is, on average, four hours. This time is dependent on the content of the diet and the overall health of the individual.

In a human, the small intestine empties into the large intestine, (called the large intestine because its internal diameter is about double that of the small intestine). (ICRP, 1975) For purposes of modeling the digestive tract, this is an unimportant step because "The large intestine has little or no digestive function." (Shier, 2004) Absorption of water and electrolytes does occur in the proximal half of the large

intestine, reabsorbing 90% of the water that enters it by osmosis. Active transport moves ions such as sodium through the walls of the large intestine. After reabsorbing the water in the large intestine, all undigested nutrients and substances are removed from the body with the feces.

Simulation of the human digestive tract in a laboratory setting is a useful tool to evaluate the effects of ingestion of any substance. Drugs must undergo solubility testing in an artificial gastrointestinal (GI) environment to determine if they are released in the proper environment to be effective. Hazardous material ingestion by humans such as the potential consumption of the MACTEC soils can only be modeled artificially for obvious reasons. Knowing if a substance is transferred to solution and knowing the absorptive tendencies of the human digestive tract can predict the risk posed by potential ingestion.

2.9 BIOACCESSIBILITY

The Food and Drug Administration's Center for Drug Evaluation and Research released guidelines for drug testing that assess the behavior of a drug in the digestive tract using *in vitro* methodology. (USDHHS, 1997) The three basic steps to move from oral administration to absorption were generally applied to this analysis as follows:

1. The radionuclide must be released from the soil
2. The radionuclide must be dissolved or solubilized under physiologic conditions
3. The radionuclide must then be permeable across the gastrointestinal tract

Because the first two steps, release and dissolution/solubilization, are critical to the systemic distribution of any substance, *in vitro* analysis is used as a relevant predictor of *in vivo* performance. The *in vitro* conditions for batch testing of drug performance (USDHHS, 1997) are the same as those used in this study to determine solubility of radionuclides. The fraction of any contaminant that is soluble in GI fluids (and therefore would be available for gastrointestinal mucosal transport) is the definition of bioaccessibility. (Contaminated-Land, 2006)

2.10 BIOAVAILABILITY

Bioavailability is literally the amount of the total material that reaches systemic circulation, i.e., if a substance was administered intravenously, the bioavailability would be 100%. If the administration of a substance is by ingestion, this percentage can be significantly lower due to the single pass metabolism of humans. Permeability through the gastrointestinal tract is not easily tested *in vitro* and most estimations rely on the extrapolation of animal data to human populations. These animal models can be inaccurate predictions of human performance as can be seen in Kristie Ellickson's work at Rutgers in 2001. The mean lead oral bioavailability in rats was found to be 0.7% with only an average of 59% total recovery of the administered lead.[6] When the rat study is compared with the *in vitro* bioaccessibility study, the mean bioaccessible

[6] Total recovery refers to the summation of all lead in tissues and excrement. In the mentioned study, 41% of the lead was not recovered after administration to the rats.

lead was much higher at 10.7% and the average recovery was nearly 75%. (Ellickson, 2001)

In the present analysis, use of a laboratory method to predict *in vivo* performance of ^{226}Ra in humans will be used to determine bioavailability. Bioaccessibility is the cornerstone for predicting bioavailability in such a study, since what is truly being measured is how soluble the radionuclide contaminated soils are in the digestive analogue. Assuming that the bioaccessibility is equal to the bioavailability allows for more conservative calculations of the dose to humans and removes the need to correlate rat performance to human performance.

3. LITERATURE REVIEW

3.1 ELLICKSON AT RUTGERS

Two previous works on bioavailability and bioaccessibility of soil associated components were chosen to contribute to this analysis. The first work titled *"The Bioaccessibility of Selected Radionuclides and Heavy Metals: An Investigation of Bioaccessibility, Bioavailability and Natural Soil Characteristics,"* (Ellickson, 2001), was completed by Kristie Ellickson at Rutgers in May of 2001. In this work, *in vivo* animal models were used to compare against *in vitro* results. This large dissertation was re-issued without the accompanying *in vivo* analysis in 2002 as a journal article for the Health Physics Society entitled *"The Bioaccessibility of Low Level Radionuclides from Two Savannah River Site Soils."* (Ellickson, 2002)

The dissertation, (*in vivo / in vitro*), analyzed twelve male Sprague-Dawley rats that were fed contaminated soil, four that were vehicle control, and one that was fed a NIST standard reference soil[7]. They were then sacrificed on day one, day two, day three and day four post ingestion. The two elements analyzed in the rat study were arsenic (As) and lead (Pb). Since ^{226}Ra decays to ^{214}Pb and ^{210}Pb, this is partially applicable to the current study. ^{226}Ra was not analyzed in the rat study by Ellickson; however it was part of the *in vitro* portion of the work.

[7] National Institute of Standards and Technology (NIST). Standard Reference Material 2710 Montana Soil Highly Elevated Trace Element Concentrations.

The *in vitro* analysis of the NIST soil sample showed that the bioaccessibility of lead, measured by solubility in human GI analogueue, was approximately 10.7%. (Ellickson, 2001) The results of the *in vivo* analysis became the bioavailability component of Ellickson's work. The sacrificed rat that consumed the NIST soil sample produced a result of 0.7% bioavailability. (Ellickson, 2001)

3.2 NRPB-W17

The second work that contributed to the current analytical method was the National Radiological Protection Board (NRPB) report W17 titled "*The Availability of Soil-Associated Radionuclides for Uptake after Inadvertent Ingestion by Humans.*" (NRPB, 2002) In this report, authors Shaw and Green recognized the inaccuracy of extrapolating animal data to human populations; thus there is no *in vivo* component to this work. Shaw and Green's NRPB *in vitro* enzymolysis procedure is similar to that used in Ellickson's dissertation, with the following list of changes:

- Ellickson's saliva step is omitted in the NRPB version
- The stomach fluid analogueue is a 1% (w/v) in the NRPB version. Ellickson's was 0.32% (w/v)
- The NRPB intestinal analogueue is a 50:50 enzyme to bile salt mix in a 0.3M sodium bicarbonate solution. Ellickson's was simply a 0.2M sodium bicarbonate solution

NRPB report W17 utilized the same references as did Ellickson (Ruby and Davis, 1996). The NRPB version had a well described prescriptive for reproducibility and was selected by this author due to time constraints and its similarity to the MACTEC/OSU study (soil only, no animal model). The NRPB study utilized sequential filtration down to 0.45μm to constitute the difference between the solid and liquid phases. The NRPB counted the liquid phase in a Marinelli Beaker in a High Purity Germanium Counter, and the solid phase was counted (after drying to a constant weight) in a Petri dish.

The data supporting the details for the NRPB report made it an ideal candidate for replication in the MACTEC/OSU analysis. The times for the stomach and intestinal phases were selected based on the ICRP retention times for the standard man from ICRP-2.[8] The procedure was well-documented and the conservative estimation of availability by determining accessibility served the goals of the project most directly.

3.3 ICRP PUBLICATION 23: REPORT OF THE TASK GROUP ON REFERENCE MAN

Absorption data on ^{226}Ra salts that are orally administered is mostly the product of rat studies. The ICRP recognizes that the rate of absorption varies from 80% in immature rats to only about 5% in adult rats and that it is increased by starvation. Radium that is found present in the feces is the portion that is unabsorbed.

[8] ICRP-2's "Standard Man" was superseded by ICRP-23's "Reference Man" in 1975. The residence times in the stomach and small intestines are listed in ICRP-30's literature review of this document.

There is one human study referenced by the ICRP in which a man took in 50 μg of elemental radium by mouth. The man's fecal loss was 27 μg on day five and up to 33 μg by day six. In this single human observation, it is recognized that some loss could be due to the emanation of radon but conservatively the absorbed amount of ^{226}Ra was considered equal to the retention percentage which was 25-35%. (ICRP, 1975a)

The components of the digestive system for the reference man are covered with great detail. The length of the adult male intestinal tract is approximately 660 cm, the small intestines being 500 cm of this total. The female intestines are figured at around 94% of the total length of the male intestinal tract. The surface area of the male large intestine is stated to be 3460 cm^2 with a range from 2800 cm^2 to 3996 cm^2. (ICRP, 1975b) The total weight of the empty gastrointestinal tract for the reference adult male is 1200 g, and the female is 1100 g. The weight of the contents of the GI tract is 1005 g. The Reference Man suggested total weight of feces lost per day is 135 g, but the dry weight is approximately 20% of the wet weight. (ICRP, 1975c) These data are useful when estimating the dose to a person from non-absorbed ^{226}Ra after ingestion.

3.4 ICRP PUBLICATION 30: LIMITS FOR INTAKES OF RADIONUCLIDES BY WORKERS

ICRP 30 published the Gastrointestinal Tract Model, and it lists the mean residence time and the associated removal constants, λ, for substances within the GI tract as portions of a 24 hour day. (ICRP, 1979) They are listed below in Table 3.1.

Table 3.1 Residence times and removal constants for GI Tract

Portion of GI Tract	Weight (g)	Residence (day)	λ (day-1)
Stomach (ST)	150	1/24	24
Small Intestine (SI)	640	4/24	6
Upper Large Intestine (ULI)	210	13/24	1.8
Lower Large Intestine (LLI)	160	24/24	1

(ICRP, 1979)

Using these values and understanding what amount of radioactivity was consumed allows for dose estimates for ingesting individuals. For oral administration of ^{226}Ra, each of the nuclides in the decay scheme deposits approximately 20% of its decay energy in the blood stream or organs, (i.e., the f_1 value is 0.20).[9] The f_1 value for children is not discussed in this work.[10]

3.5 ICRP PUBLICATION NUMBER 60: 1990 RECOMMENDATIONS OF THE INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION

Tissue weighting factors (w_t) make it possible to assess risk from non-uniform radiation, using the assumption that "The probability of a detrimental effect in any tissue is proportional to the dose to that tissue."[11] (Cember, 1996) The w_t values are chosen to be independent of the type or energy of the incident radiation. The use of these w_t values makes dose calculations reflect distribution of radiation within the

[9] It was previously thought that the fraction of Ra-226 that would be transferred to blood from the gastrointestinal tract (f_1) was 0.3. (ICRP-2, 1959)

[10] The f_1 value for infants is 0.6 and the f_1 value for children over age 3 is 0.2. (ICRP, 1995)

[11] Examples: If a person gets a 65 mSv dose equivalent to the thyroid, (w_t thyroid = 0.05) then the effective dose equivalent, H_E=(65 mSv)(0.05) = 3.25 mSv. (Cember, 1996)

body. Table 3.2 lists the tissue weighting factors for the GI tract recommended by the ICRP.

Table 3.2 ICRP-60 Tissue Weighting Factors (w_t)

GI Section	w_t
Stomach (ST)	0.12
Small Intestine (SI)	0.025
Upper Large Intestine (ULI)	0.025
Lower Large Intestine (LLI)	0.12

3.6 ICRP PUBLICATION NUMBER 67: AGE-DEPENDENT DOSES TO MEMBERS OF THE PUBLIC FROM INTAKE OF RADIONUCLIDES: PART 2 INGESTION DOSE COEFFICIENTS

In 1993, a biokinetic model for the alkaline earth metals and lead was published by the International Commission on Radiation Protection (ICRP). The model depicts the transfer of radium throughout various body systems after it has entered the blood (plasma). Preferentially, radium from the blood deposits on the bone surfaces and can migrate into areas of the bone that have a lesser exchange rate with the blood. This is significant because although much of the radium is deposited on the bone surface, it will be recycled back into the blood within a few days. Only that migrated portion will stay in the bone and deliver its decay energy. When the radium returns to the plasma, it is assumed to follow the same transfer parameters to bone, soft tissue, urine and feces.[12] ICRP-67 notes that:

[12] It should be noted that the feces to urine ratio of radium excretion is 36:1 (i.e., primary removal is with the feces).

"In the fully mature human, skeletal retention may decrease from one-quarter or more of injected activity in the first day or two after injection to less than 10% at 1 month. Thereafter, skeletal retention gradually decreases to a level of about 0.5%-1% by 25 years. Limited data for humans indicate that soft tissue radium may represent 20% or more of total body radium during the first several weeks after exposure but probably represents a much smaller percentage at times remote from exposure" (ICRP, 1993)

3.7 ICRP PUBLICATION NUMBER 72: AGE-DEPENDENT DOSES TO MEMBERS OF THE PUBLIC FROM INTAKE OF RADIONUCLIDES: PART 5 COMPILATION OF INGESTION AND INHALATION DOSE COEFFICIENTS

The fraction of a material in the body that is bioavailable or distributed systemically is expressed as an f_1 value.[13] In 1995, the ICRP compiled tables to add ^{226}Ra f_1 values for children of various ages and committed effective dose per unit intake for 3 month old infants, 1 year olds, 5 year olds, 10 and 15 year old children as well as adults (those over the age of 25 years old). These are calculated for acute intakes but can be used for protection purposes for chronic intakes by summing all intakes in a one year period. Table 3.3 lists the above-mentioned f_1 values and the committed effective dose per unit intake, $e(\tau)$, (in units of Sv/Bq).[14]

[13] An f_1 value of 0.2 would mean 20% of the material taken into the body (in this case by ingestion) would be distributed throughout the body.

[14] Sv is the abbreviation for Sievert, which is the unit for the dose equivalent, (1 J/kg or 100 rem). Bq is the abbreviation for Bequerel, which is a unit of activity equal to 1disintegration per second (dps).

Table 3.3 Age dependent doses from ingestion of Ra-226

Age	f_1	$e(\tau)$ (Sv/Bq)
< 1 year	0.6	4.70E-06
> 1 year	0.2	9.60E-07
5 years	0.2	6.20E-07
10 years	0.2	8.00E-07
15 years	0.2	1.50E-06
> 25 years	0.2	2.80E-07

(ICRP, 1995)

4. MATERIALS

4.1 HIGH PURITY GERMANIUM DETECTION SYSTEM

Gamma spectra were assessed using a lead shielded D Spec Analyzer HPGe system with Gamma Vision software. The system efficiency is 27% and the resolution is 1.87 keV. Initially, a standard library was used to identify that the soil samples contained radium-226. After that point, a library was created to recognize only those peaks of interest; radium-224, radium-226, bismuth-214, and lead-214. The nuclide library is presented in Appendix C.

4.2 NALGENE® FILTRATION UNIT

After the gastrointestinal simulation was complete, the entire sample was filtered once with a disposable culture filtration unit. The unit has a permanent 0.8 μm filter and a 1 μm gross pre-filter was placed on top. Vacuum suction was applied using a standard laboratory pump. Figure 4.1 displays an image of one 500 mL capacity unit.

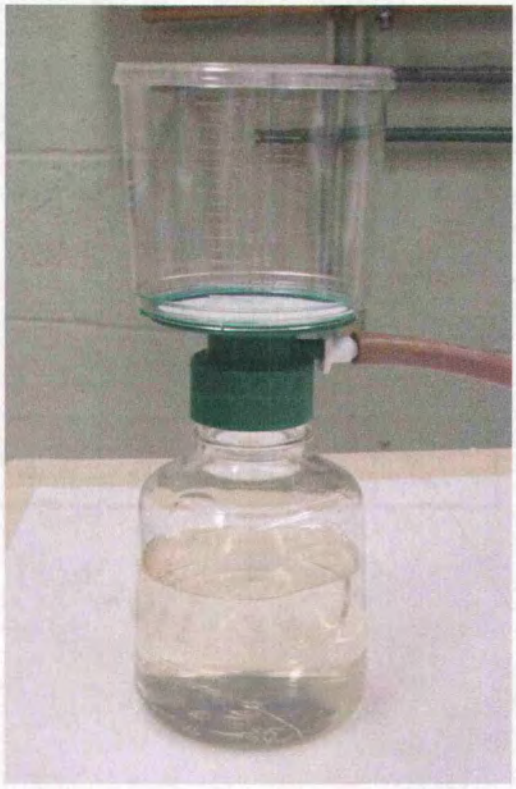


Figure 4.1 Filtration unit with 0.8 μm filter.

4.3 PH INDICATOR STRIPS

The pH was to be determined at various points in the stomach and intestinal digestions (see Figure 4.2). Selected ranges of pH indicator strips were chosen to be precise, yet not jeopardize potential loss of soil associated with fluid that might occur with use of a pH meter. The ranges utilized were:

- pH= 0.0 to 6.0 (to assess stomach phase pH and correct preparation)
- pH= 2.0 to 9.0 (to assess combined stomach and intestinal phase pH)
- pH= 6.5 to 10.0 (to assess the intestinal phase pH, prior to use)

Figure 4.2 Digestive solutions with their respective pH range indicators

4.4 KONTES CYTOSTIR® CULTURE VESSEL WITH HOT PLATE AND DIGITAL THERMOMETER

The simulated gastric and intestinal phases took place in apparatus that mechanically degrades the sample while maintaining a constant physiologic temperature of 37 °C. The vessel chosen was a three-necked glass culture vessel with a magnetic stir paddle (see Figure 4.3). The hot plate was temperature-regulated to stay within ±2 degrees of 37 °C.

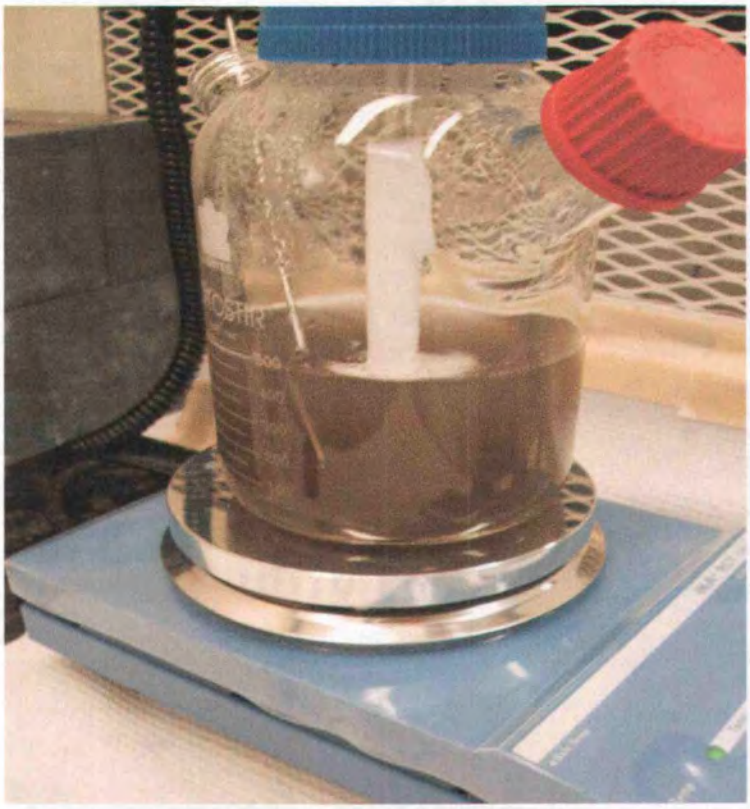


Figure 4.3 One liter culture vessel digesting a soil sample.

4.5 FISHER ISOTEMP® OVEN 200 SERIES

The Fisher Isotemp furnace was used to dry all soil samples prior to analysis as well as after digestion to dry the solid phase and reduce the liquid phase by evaporation. The furnace was set to 72 °C,[15] and samples were dried until they reached a constant weight.

[15] This temperature was selected to mimic the conditions in Ellickson's work at Rutgers in which samples were dried at 72 °C due to safety concerns and reduced risk of dust suspension.

4.6 OHAUS EXPLORER BALANCE

A single closed box balance was utilized for all assessed sample weights. Weighing papers and weighing boats were used for not only samples, but also the dry gastric and intestinal ingredients. The balance was calibrated immediately prior to this laboratory work and reads to the nearest one-thousandth of a gram (0.001).

4.7 COUNTING VIALS

The four dram vials or "poly vials" that were utilized to maintain constant geometry between samples are irradiation vials for use in the TRIGA reactor at OSU. Shown below is a vial with a standard pencil for size comparison (Figure 4.4).

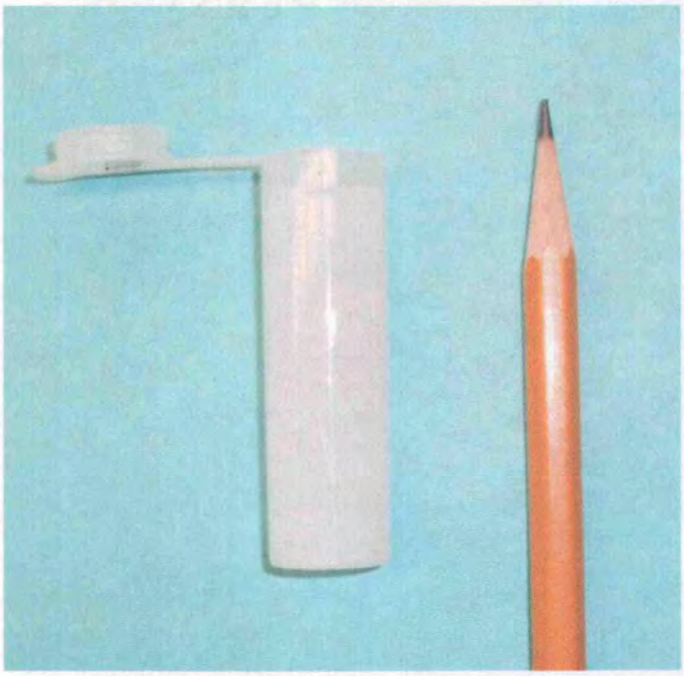


Figure 4.4 Four dram vial with size comparison.

4.8 OTHER MATERIALS

Many standard laboratory materials were used in this analysis. The following is a list of those necessary items to complete this type of analysis:

- Glassware including watch glasses, various sizes of beakers, volumetric flasks, graduated cylinders and Petri dishes
- Disposable pipettes
- Centrifuge (small, tabletop) with tubes and counter weight
- Digital timer
- Chemicals (see Appendix A)
- Waste containers (radioactive and chemical)

5. METHODS

5.1 RECORD KEEPING

Throughout the entire analytical process, all documentation was kept in a single carbon copy lab manual. Pictures of each step of the process, including all materials, were recorded with a digital camera.

5.2 PARTICLE SIZE

The particle size distribution was determined by an independent researcher at Oregon State University. The result of that analysis is presented in this work, but the specific methodology and materials are not listed. (Bytwerk, 2006) Particle size is an important aspect of bioavailability as it can dictate intestinal transport, but it was not necessary to know the type of soil in order to complete the digestion.

Ideally, soils would have been sieved to particle sizes below 250 μm to simulate the size of particles likely to stick to a person (or child's) hand and be inadvertently ingested (Ellickson, Rutgers 2001). Because some of the samples are simply "rocks" and of limited quantity, it was decided that small and large particles alike would be subjected to the simulated gastric and intestinal environments. This would provide a more conservative estimate of dose from inadvertent ingestion

because those larger particles would then be used to further calculate the potential dose delivered as they are moved through the system and out with the feces.

5.3 LABORATORY METHODOLOGY

The methodology is mostly taken from that of the NRPB-W17 report and some from Ellickson's work at Rutgers. However, the assessment done at Oregon State University was looking at ^{226}Ra and its progeny. The digestive portion of the study is no different than prescribed by the NRPB, yet the determination of the presence of ^{226}Ra is solely by gamma spectrometry (Ellickson and NRPB both utilized LSC for Sr-90, and the NRPB study used a gas flow beta counter for Y-90).

The simulated stomach fluid consisted of a mixture of pepsin and hydrochloric acid solution and was scaled down from the NRPB methodology to 200 ml to accommodate the very small sample sizes. The pH was monitored during the hour long stomach phase and was maintained at or below a pH of 2.5.

At the completion of the hour long stomach phase, the simulated intestinal fluid was added. The intestinal fluid consisted of a mixture of pancreatin, α-amylase, and bile salts and was dissolved in a 0.3M sodium bicarbonate solution. The pH was monitored during the two hour long intestinal phase and was maintained above a pH of 7.5.

5.4 STOMACH MODEL

The synthetic fluid produced as the stomach analogueue was taken directly from the NRPB methodology and is presented in Appendix B.

5.5 INTESTINAL MODEL

The synthetic fluid produced as an intestinal analogue was modified from the US Pharmacopoeia by NRPB and taken directly from the NRPB methodology. The fluid utilized sodium bicarbonate which would yield the greatest solubility of metals and would provide a more conservative estimate of bioaccessibility. (Appendix B)

5.6 AGITATION METHODS

The method of *in vitro* agitation chosen was based on equipment available on site. For the simulated digestion, a culture vessel at physiologic temperature with a magnetic stirrer was utilized (see Figure 4.3). This method was considered a reliable analogue to the mechanical degradation of products that would normally happen within the human digestive tract.

5.7 DETERMINATION OF SOLUBILITY

Previous studies had removed portions of solution after both the stomach phase and after the intestinal phase and analyzed these aliquots. However, determination of solubility of the samples after BOTH the stomach and intestinal phases was considered by this researcher to yield the most applicable data. It should be noted that most all of the absorption that occurs in the human digestive tract is through the small intestine, not in the stomach. Solubility of radionuclides from the soil samples was determined only at the completion of both the stomach and intestinal phases of *in vitro* digestion.

5.8 FILTRATION

After digestion, the simulated digestive solution with the associated soil sample was filtered using a Nalgene disposable vacuum filtration unit with two associated filters. The first filter was a 1-2 μm pore size pre-filter and the second was a fixed cellulose nitrate filter with pore size of 0.8 μm. Filtration of each sample took approximately one hour. Due to the small sample sizes, it was decided that sequential filtration would result in a potentially significant loss of each sample and mass balance would be a less reliable value. With the filtration methods utilized in this study, total mass lost was very small for each sample and was attributed to solubilization in the digestive model—not to soil orphaned on experimental apparatus.

The NRPB study utilized filtration down to 0.45 μm particles, but it was decided that to yield a more conservative estimate of solubilization, the modified method used by this researcher would differentiate solution from solid at 0.8 μm particles. The following is a list of standard sizes of common substances for comparison.

- Human hair – 200 μm
- Ragweed pollen, red blood cells – 20 μm
- E Coli, Staphylococcus – 2 μm

Particles in a range of sizes can be absorbed in the small intestine and can be affected by the presence of food, lipids, and proteins as well as the health of the digestive tract. By assuming a slightly larger bioavailable particle size, the most conservative estimate of systemic distribution and therefore dose could be attained.

5.9 TOTAL MASS BALANCE

Soil samples were weighed prior to digestion. After each digestion, the simulated digestive tract solution with the associated soil sample was filtered using a disposable filtration unit fitted with a permanent filter. The filter was dried overnight in a Fisher Isotemp Oven at 72 °C. A 5 ml portion of the solution was weighed and removed for gamma spectroscopy while the rest of the approximately 400 ml was placed in the oven for evaporation to reduce the volume to 100 ml. After evaporation, another 5 ml aliquot was removed for counting and stored for future gamma

spectrometry, if necessary. The initial mass and the dried filter mass were used to assess total mass lost to solution. The soil (or rock) mass left on the filters was then used to calculate the dose to the intestinal tract as it would pass through the small and large intestines, through the colon and out with the feces.

5.10 GAMMA SPECTROSCOPY

Every sample was counted upon initial receipt by Oregon State University's Department of Nuclear Engineering and Radiation Health Physics. The calibration for the High Purity Germanium Counter was done using a standard source of ^{125}Sb (antimony) ^{154}Eu (europium) and ^{155}Eu.[16] (Appendix C). A general library of peaks determined that the samples contained ^{226}Ra and its progeny (see Appendix C). A library was created utilizing these energies for recognition for all subsequent counting. The samples were counted again after fractionation, prior to digestion. From this point forward, all counts were done in four (4) dram polyethylene vials (poly vials) at the same shelf position above the detector to maintain constant geometry. After digestion, a 5 ml aliquot of fluid was counted and preserved for future counting. The dried filters and soil were also counted as well as another 5 ml aliquot of digestive analogueue after it was evaporated. All sealed vials were retained for future counting to assess progeny to yield better counting statistics.

[16] NIST 19459 was the mixed nuclide standard source was used to calibrate the HPGe at various shelf heights within the detector. The range of gamma energies is from 86.5 keV for ^{155}Eu to 1274.4 keV for ^{154}Eu. See Appendix C.

6. RESULTS

6.1 PARTICLE SIZE DISTRIBUTION (SOIL TYPE)

In work done by David Bytwerk for Oregon State University (Bytwerk, 2006), it was determined that the soil type was sandy loam. Because of small sample sizes, only three of the eight samples were analyzed for particle size determination as the method is destructive to the sample.

6.2 SOIL PH

A 1:1 soil to water slurry was created. (UGCAES, 2006) Indicator papers were used to assess the pH of the soil. The pH of the slurry was found to be 7[17], and the pH of the water alone was also 7.[18]

6.3 CARBONATES / CALCITE TEST

This analysis used an aliquot of Sum-02d which was 0.329 grams of the finest particles. It was anticipated this portion would have the greatest surface area for a potential reaction (bubbling). The sample was placed on a large watch glass and

[17] The color-change indicators used to assess the pH read to the nearest twentieth, i.e., the color change indicated a darker color than a pH of 6.8 and a lighter color than that of a pH of 7.2.
[18] The soil pH can be altered by storage, drying and handling. Ideally, the soil pH would have been assessed at the site prior to removal and transport of the samples.

hydrochloric acid (HCl) (0.25N) was added drop-wise to observe the production of bubbles (which would indicate the presence of carbonates). No bubbling was observed nor delayed production of bubbles. It was concluded that this sample did not contain carbonates or calcite (non-carbonate sample).

6.4 SOLUBILITY IN WATER

This analysis used an aliquot of Sum-02d which was 0.392 grams of the finest particles, again in an effort to be conservative about solubility. The sample was weighed, placed in a centrifuge tube, and deionized water was added, (20:1 ratio, water to soil), based on the recommendations of the Field Leach Test USGS. (USGS, 2005a) The tube was vigorously shaken for five (5) minutes and then centrifuged for several minutes. The sample was then placed in a properly calibrated HPGe detector (gamma spectrometer) and counted for 1200 seconds (in the centrifuge tube). The water from the sample was removed with a transfer pipette to a second centrifuge tube. The tube was refilled or q.s.'d[19] to the initial volume and counted again in the centrifuge tube to conserve geometry.

There was no detectable ^{226}Ra activity in the water. The activity of progeny was assessed as well. For ^{214}Pb, the activity in the water represented only 0.29% solubility in water as well as for ^{214}Bi, which was approximately 0.45% soluble in

[19] The abbreviation q.s. is a Latin term, quantum sufficit, or as much as suffices.

water. The conclusion of these results is that the representative sample is relatively insoluble in water.

6.5 SOLUBILITY IN ACID

An aliquot from Sum-02d that was 0.308 g of the finest particles (anticipated most soluble, to be conservative), was placed in a centrifuge tube with a sufficient amount of 0.25N HCl (approximately 5 ml). The sample was shaken vigorously for five (5) minutes and then centrifuged for several minutes. The entire sample and centrifuge tube was then placed in a properly calibrated HPGe detector (gamma spectrometer) and counted for 1200 seconds. The acid from the sample was removed with a transfer pipette to a second centrifuge tube. The tube was q.s.'d to the initial volume (with acid) and counted again.

To discern if the activity in the sample was soluble in acid (HCl) the amounts of ^{226}Ra as well as ^{214}Pb and ^{214}Bi were observed. ^{226}Ra was found to be approximately 8.85% soluble in HCl, ^{214}Pb was approximately 4.4% soluble and ^{214}Bi was approximately 6.4% soluble in acid. The conclusion of these results establish that the representative sample is moderately soluble in acid (HCl).

6.6 IN VITRO DIGESTION

The HPGe gamma spectroscopy reports can be found in Appendix D. They are grouped in order and labeled with the sample name and "Initial Solid," "Initial Liquid," "Evaporated Liquid," or "Final Solid."

The following subsections list the sample by name given by MACTEC, beginning with SUM, a two digit code and a letter, (A through E). The size (mass) of the analyzed sample is listed, along with the mass deficit, the percentage of total mass lost to solution and the concentration of the initial sample. Each sample has its own table of radioanalytical results.

6.6.1 SUM-01A

Sample Size: 0.841 g

Mass lost to Model / Solution: 0.009 g

Percent of Mass Lost: 1.070%

Activity Ra-226 per gram of Soil: 6.178 μCi/g

Table.6.1 Radioanalytical results for sample Sum-01a

Sum-01a	Ra-226 (μCi)	Pb-214 (μCi)	Bi-214 (μCi)
Initial Solid	5.1959	3.7535	3.0064
Initial Liquid	0.0013	0.0002	0.0001
Evaporated Liquid	0.0019	0.0001	0.0001
Final Solid**	5.3407	3.8278	2.9996
% Radioactivity Lost in Model	-2.7868	-1.9795	0.2262

**Uncertainty in counting: 0.4183% for Ra-226, with a standard deviation of 2.988%

6.6.2 SUM-02A

Sample Size: 1.632 g

Mass lost to Model / Solution: 0.117 g

Percent of Mass Lost: 7.169%

Activity Ra-226 per gram of Soil: 0.720 μCi/g

Table.6.2 Radioanalytical results for sample Sum-02a

Sum-02a	Ra-226 (μCi)	Pb-214 (μCi)	Bi-214 (μCi)
Initial Solid	1.1751	0.7576	0.6017
Initial Liquid	0.0129	0.0001	0.0001
Evaporated Liquid	0.0011	0.0001	0.0001
Final Solid**	1.1419	0.6290	0.4956
Percent Radioactivity Lost in Model	2.8253	16.9672	17.6334

**Uncertainty in counting: 0.8255% for Ra-226, with a standard deviation of 3.072%

6.6.3 SUM-02B

Sample Size: 0.717 g

Mass lost to Model / Solution: 0.062 g

Percent of Mass Lost: 8.647%

Activity Ra-226 per gram of Soil: 0.537 μCi/g

Table.6.3 Radioanalytical results for sample Sum-02b

Sum-02b	Ra-226 (μCi)	Pb-214 (μCi)	Bi-214 (μCi)
Initial Solid	0.3849	0.1506	0.1215
Initial Liquid	0.0015	0.0001	0.0002
Evaporated Liquid	0.0016	0.0001	0.0001
Final Solid**	0.3092	0.1014	0.0825
% Radioactivity Lost in Model	19.6690	32.6761	32.0938

**Uncertainty in counting: 1.206% for Ra-226, with a standard deviation of 3.195%

6.6.4 SUM-02C

Sample Size: 1.248 g

Mass lost to Model / Solution: 0.114 g

Percent of Mass Lost: 9.135%

Activity Ra-226 per gram of Soil: 0.494 μCi/g

Table.6.4 Radioanalytical results for sample Sum-02c

Sum-02c	Ra-226 (μCi)	Pb-214 (μCi)	Bi-214 (μCi)
Initial Solid	0.6169	0.1989	0.1586
Initial Liquid	0.0014	0.0001	0.0001
Evaporated Liquid	0.0013	0.0001	0.0001
Final Solid**	0.5298	0.1285	0.1030
% Radioactivity Lost in Model	14.1257	35.4148	35.0508

**Uncertainty in counting: 0.7245% for Ra-226, with a standard deviation of 3.046%

6.6.5 SUM-02D

Sample Size: 3.025 g

Mass lost to Model / Solution: 0.115 g

Percent of Mass Lost: 3.802 %

Activity Ra-226 per gram of Soil: 0.481 μCi/g

Table.6.5 Radioanalytical results for sample Sum-02d

Sum-02d	Ra-226 (μCi)	Pb-214 (μCi)	Bi-214 (μCi)
Initial Solid	1.4551	1.2467	1.1575
Initial Liquid	0.0014	0.0001	0.0002
Evaporated Liquid	0.0014	0.0001	0.0001
Final Solid**	1.5195	1.2318	1.1609
% Radioactivity Lost in Model	-4.4258	1.1952	-0.2937

**Uncertainty in counting: 0.9097% for Ra-226, with a standard deviation of 3.096%

6.6.6. SUM-02E

Sample Size: 0.715 g

Mass lost to Model / Solution: -0.016 g*

Percent of Mass Lost: -2.238%*

Activity Ra-226 per gram of Soil: 1.176 μCi/g

*This sample was a rock with very few small broken pieces. The negative numbers can be attributed to fluctuations in atmospheric pressure causing changes in the mass balance values.

Table.6.6 Radioanalytical results for sample Sum-02e

Sum-02e	Ra-226 (µCi)	Pb-214 (µCi)	Bi-214 (µCi)
Initial Solid	0.8408	6.5096	4.0117
Initial Liquid	0.0018	0.0001	0.0002
Evaporated Liquid	0.0126	0.0001	0.0002
Final Solid**	0.6018	6.1804	4.9682
% Radioactivity Lost in Model	28.4246	5.0571	-23.8428

**Uncertainty in counting: 3.757% for Ra-226, with a standard deviation of 4.782%

6.6.7 SUM-03A

Sample Size: 0.198 g

Mass lost to Model / Solution: -0.007 g*

Percent of Mass Lost: -3.535%*

Activity Ra-226 per gram of Soil: 34.089 µCi/g

*This sample was very small and contained grainy soil and some rocks, which upon visualization appeared to be completely retained on the filter, (no change in digestive solution appearance). The negative values can be attributed to fluctuations in atmospheric pressure causing changes in the mass balance values.

Table.6.7 Radioanalytical results for sample Sum-03a

Sum-03a	Ra-226 (µCi)	Pb-214 (µCi)	Bi-214 (µCi)
Initial Solid	6.7497	4.3351	3.5009
Initial Liquid	0.0123	0.0002	0.0002
Evaporated Liquid	0.0596	0.0003	0.0002
Final Solid**	5.4787	2.7908	2.2383
% Radioactivity Lost in Model	18.8305	35.6232	36.0650

**Uncertainty in counting: 0.3718% for Ra-226, with a standard deviation of 2.982%

6.6.8 SUM-04A

Sample Size: 0.692 g

Mass lost to Model / Solution: 0.036 g

Percent of Mass Lost: 5.202%

Activity Ra-226 per gram of Soil: 2.493 μCi/g

Table.6.8 Radioanalytical results for sample Sum-04a

Sum-04a	Ra-226 (μCi)	Pb-214 (μCi)	Bi-214 (μCi)
Initial Solid	1.7253	1.2450	1.0191
Initial Liquid	0.0013	0.0001	0.0002
Evaporated Liquid	0.0011	0.0001	0.0001
Final Solid**	1.6889	1.2049	0.9519
% Radioactivity Lost in Model	2.1098	3.2209	6.5950

**Uncertainty in counting: 0.4909% for Ra-226, with a standard deviation of 2.999%

7. DISCUSSION

7.1 PARTICLE SIZE DISTRIBUTION (SOIL TYPE)

Particle size analysis is used in soil science to assess soil texture. The sandy loam soil is one that contains 30% or more medium sand, less than 30% fine sand and less than 25% very coarse sand. (Agronomy, 1986) All the samples in the analysis fit this description. Sandy loam soils tend to hold less nutrients but be more permeable and aerated than higher clay content soils. (ISU, 2006)

7.2 CARBONATES / CALCITE TEST

Because of the small sample sizes, one soil (Sum-02d) was tested as a representative for all eight samples. Because there was no bubbling seen, there are no carbonates in the sample. This makes the samples less degradable by acids and the expectation is that the lack of carbonates will also lower the bioavailability.

7.3 SOLUBILITY IN WATER

As with previous testing, only the soil with the largest amount was tested (Sum-02d). The inability of the radioactivity to be transferred to water was determined by gamma spectroscopy. Although solubility in water is not a good

predictor of solubility in the gastrointestinal tract, leachability in the environment is a concern. If the sample had been soluble in water, another route of ingestion (drinking water) would pose a significant risk for dose to the public.

Previous studies have used the solubility of radium in water as a predictor of the availability for plant uptake. The assumption with water extraction of radium is that the water extracts the neutral and negatively charged radium complexes with organic ligands. (IAEA, 1990c) However, the works all previously used 5:1 ratios of water to soil, but with the present analysis, a 20:1 ratio was used in an effort to mimic the USGS Field Leach Test. Thus, the apparent insolubility in water of the sample is not necessarily an indication that there are no neutral or negatively charged radium complexes with organic ligands.

7.4 SOLUBILITY IN ACID

A quick test for solubility in acid before simulated digestion predicted some possible bioavailability in acidic environments. Approximately 8% of the ^{226}Ra in the sample was transferred to solution. Direct correlation between solubility in HCl and the digestive tract was not possible as the sample that was tested, Sum-02d, was later digested and had essentially no bioavailable radioactivity.

In previous work, the leaching of radium with 0.5M HCl was used as a method of determining the radium that was not associated with primary mineral particles.

(IAEA, 1990c) In this analysis, 0.25N HCl (0.25M HCl) was used.[20] Thus, it is not possible to state with confidence that approximately 8% of the sample ^{226}Ra was unassociated with primary mineral particles.

7.5 MASS LOST IN THE MODEL

Mass balance was attained by weighing the initial sample and then reweighing that sample after filtration and drying to a constant weight. The soil after filtration was associated with the filters as well as the portion of the gastrointestinal model that was too large to pass through the filter. A blank stomach and intestinal model was passed through a filter, dried, and weighed to allow for more accurate subtraction for soil mass balance with each sample.

For all samples tested, the highest percent of mass lost to the model was no greater than 9.2% (Sum-02c). This mass was calculated using the following simple formula:

$$\frac{InitalMass - FinalMass}{InitialMass} \times 100 = \%MassLost$$

The term "lost in the model" accounts for all losses, not just those that are into solution. Losses could be resultant of soil adhering to the culture vessel, the filtration apparatus or lost in transfer to the counting vials. However, the percentage lost to the

[20] Molarity = moles substance / L solution. Normality refers to multiple chemical functionalities. HCl only has one acidic proton; thus 0.25N HCl = 0.25M HCl.

model can be conservatively stated to be that portion of the soil that was transferred to solution and is therefore bioavailable.

A slightly negative value for mass lost was seen with two of the soil samples (Sum-02e and Sum-03a). For Sum-02e, the sample was simply a rock which the digestion did not appear to degrade. The fluctuation in weight could be attributed to atmospheric pressure changes due to the small appearance in weight gain (+0.016g post digestion) or to simple error in measurement. Sample Sum-03a also had a slight increase in weight from initial to final assessment. The apparent gain was only +0.007 g but the sample was the smallest of all eight, weighing in at a mere 0.198 g. With such small masses, miniscule variations can significantly impact the results.[21]

7.6 RADIOACTIVITY LOST IN THE MODEL

Again, the term "lost in the model" accounts for all losses, not just those that are into solution. In the case of radioactivity, apparent losses can be due to slight fluctuations in counting statistics or slight changes in geometry of samples within the counting vials (i.e. addition of filters to final solid soil). As is true with mass balance, that portion of the radioactivity that is changed from the initial solid to the final solid is conservatively stated to be bioavailable.

The soil samples were counted prior to digestion in the simulated gastrointestinal model and after the solids were filtered and dried. Five milliliter

[21] The Ohaus balance measures to 0.001 g (one thousandth of a gram).

aliquots were also counted immediately after digestion (5 ml of 400 ml total), and another five milliliter aliquot was counted for each sample after reduction of the total volume to ¼ the post digestion amount, or 100 ml. It should be noted that all samples were counted in the same geometry to prevent any fluctuations due to geometrical changes. Count times were consistent as well at 1200 seconds live time[22].

The percentage of radioactivity lost was calculated using the following formula:

$$1-\left(\frac{Final\ ^{226}Ra\ Activity}{Initial\ ^{226}Ra\ Activity}\right)\times 100 = \%RadioactivityLost$$

For all samples counted, the highest percent of radioactivity lost in the model was no greater than 28.42%, (Sum-02e). The percent radioactivity lost was calculated using the gamma spectroscopy measured amount of Radium-226 in the initial and final solids. The uncertainty listed is for ^{226}Ra from the gamma spectroscopy of the final solid. The highest uncertainty was 3.757% for Sum-02e, which was nearly a four-fold increase in uncertainty from the other seven samples.

The uncertainties listed are for ^{226}Ra in the final solid. Uncertainty is affected by dead time which is increased by high count rates. For sample Sum-02e, the uncertainty is high because the dead time was 46%. Because this sample was counted after many other samples had been entirely analyzed, it was decided that for comparison the shelf position should not be changed (even thought the sample had a high count rate). It was important to be able to compare the samples to each other, as

[22] Live time is the clock time plus the dead time, i.e., the time the detector is "live" and counting events. Real time is simply clock time.

well as each step in the analysis to an initial and final quantity of radioactivity. The aliquots of liquid after previously digested samples had very low activities and placing those further from the detector's face would decrease the likelihood of detecting any activity at all. Thus, a caveat for the uncertainty related to the greatest amount of bioavailable radioactivity must be noted.

7.7 ORTEC'S UNCERTAINTY IN COUNTING

The listed uncertainty values in results section 6.6 was for ^{226}Ra and were calculated by the Gamma Vision software on the HPGe system. Uncertainty is listed as "Uncertainty Counting" and "1 Sigma Total" in the reports from each gamma spectral analysis, (Appendix D). The counting uncertainty is the uncertainty of the peak area due to statistical uncertainty and is calculated as follows:

$$\text{Net area error} = \sqrt{(Gross\ Area\ Error)^2 + (Background\ Error)^2}$$

The background error must be calculated independently because the uncertainty of the channels used to calculate the end points and the ratio of the number of channels in the peak to the number of channels used to calculate the background. In simple terms, a long (time) background is acquired and some portion of it is used to differentiate background from sample activity. This is represented by the following equation:

$$\text{Background Error} = \left(\frac{(Bkg\ Area)(Peak\ Width}{(Width\ Low\ Avg) + (Width\ High\ Avg)} \right)^{1/2} \quad \text{(Ortec, 2002)}$$

The term "1 Sigma Total" is the total of all the random and systemic errors in each factor used to produce the final nuclide concentration. Random uncertainties in this analysis were counting and additional uncertainties. The systemic uncertainties in this analysis were nuclide uncertainty from the library, efficiency fitting uncertainty from calibration, and calibration source uncertainty. Geometry correction can also contribute to systemic uncertainty but was not used in this analysis. These values can be found on the first two pages of each spectral report in Appendix D.

7.8 CORRELATION BETWEEN CONCENTRATION OF RADIUM-226 AND BIOAVAILABILITY

To calculate the concentration of ^{226}Ra in the samples, the initial solid activity of ^{226}Ra was divided by the mass of the sample. For example:

$$\text{Sum-01a} = \frac{\textit{Initial Solid Activity}}{\textit{Sample Weight}} = \frac{5.195\ \mu Ci}{0.841\ g} = 6.782\ \frac{\mu Ci}{g}$$

Table 7.1 lists the sample name, concentration (as calculated above) and radioactivity lost to solution.

Table 7.1 Sample name, concentration and % bioavailability.

Sample Name	Ra-226 Concentration (μCi/g)	% Bioavailable
Sum-01a	6.1782	-2.79
Sum-02a	0.720	2.83
Sum-02b	0.537	19.67
Sum-02c	0.494	14.13
Sum-02d	0.481	-4.43
Sum-02e	1.176	28.42
Sum-03a	34.089	18.83
Sum-04a	2.493	2.11

Table 7.1 was used to create Figure 7.1 using Microsoft® Excel. It should be noted that the y-axis in Figure 7.1 is a dual function, having values of percentage for the dotted bars, which indicate bioavailability of ^{226}Ra, and values of concentration in μCi/g for the left hatched bars. There are two bars for each sample, one dotted on the left and one left hatched, immediately to the right of the dotted bar. The dotted area bar is absent for samples Sum-01a and Sum-02d as those samples had zero percent bioavailability.

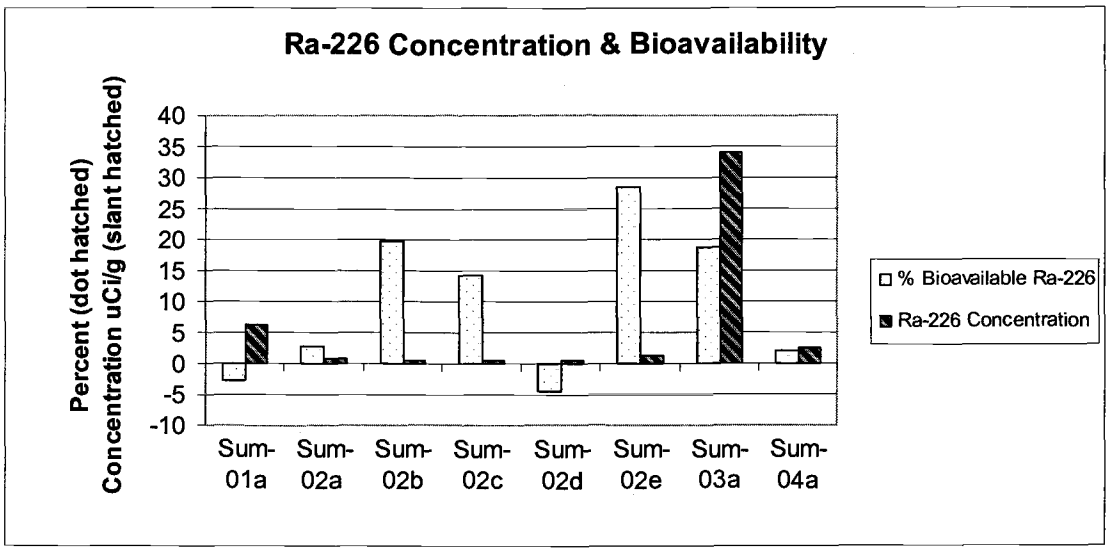


Figure 7.1 Radium-226 concentration does not predict bioavailability.

Figure 7.1 was generated solely to illustrate the inability to correlate concentration with bioavailability of ^{226}Ra. The reasons for this are at least two-fold. First, the physical form of the sample can play a large part in the ability of a human digestive system (or analogue of that system) to break it down. For instance, the entire sample Sum-02e was one rock that was not broken down at all in the simulated digestive system. The second reason that concentration is not a predictor of bioavailability is that the chemical form of the ^{226}Ra will strongly dictate the ability of the digestive system to solubilize and absorb it.

^{226}Ra chemistry is said to follow alkaline earth chemistry very closely. Radium is present in all compounds as Ra^{+2}. When radium is present as a sulfate ($RaSO_4$) it is insoluble.[23] But, if radium is present as a hydroxide ($Ra(OH)_2$) it is very

[23] $RaSO_4$ behaves as $BaSO_4$, which is very insoluble. (Cyberspace, 2006)

soluble.[24] Since radium is a Group II element, the following trends apply to solubility of radium compounds:

- Hydroxides (OH) become MORE soluble traveling down the Group IIA elements in the periodic table
- Sulfates (SO_4) become LESS soluble traveling down Group IIA
- Carbonates (CO_2) become LESS soluble traveling down Group IIA

^{226}Ra in food and water may be more readily absorbed by the GI tract than ingestion of radium compounds alone. ICRP-67 notes that GI absorption of ^{226}Ra may be about 15-21% when incorporated into food or water, and normal elderly humans who ingested mock radium dial paint ($RaSO_4$) absorbed about 20% on average. (ICRP, 1993)

7.9 CORRELATION BETWEEN MASS AND RADIUM-226 BIOAVAILABILITIES

Table 7.2 was used to create Figure 7.2 below, again using Microsoft® Excel. The brick-hatched bars represent the percentage of soil that was lost to solution and the solid grey bars represent the percent of the ^{226}Ra lost to solution. There are two bars for each sample, the solid grey on the left representing the percentage of ^{226}Ra lost to solution, and the brick-hatched bar, immediately to its right, displaying the

[24] $Ra(OH)_2$ behaves as $Ba(OH)_2$, which is very soluble. (Chemguide, 2006)

percentage of mass lost to solution. Samples Sum-02e and Sum-03a had no mass lost so there is no bar representing the percentage of mass lost.[25]

Table 7.2 Sample name, percentage of mass lost versus radioactivity lost

Sample Name	% Mass Lost	% Radioactivity Lost
Sum-01a	1.07	-2.79
Sum-02a	7.169	2.83
Sum-02b	8.647	19.67
Sum-02c	9.147	14.13
Sum-02d	3.802	-4.43
Sum-02e	-2.238	28.42
Sum-03a	-3.535	18.83
Sum-04a	5.202	2.11

[25] Sum-02e was a rock and Sum-03a was a very small sample less than 0.2 grams.

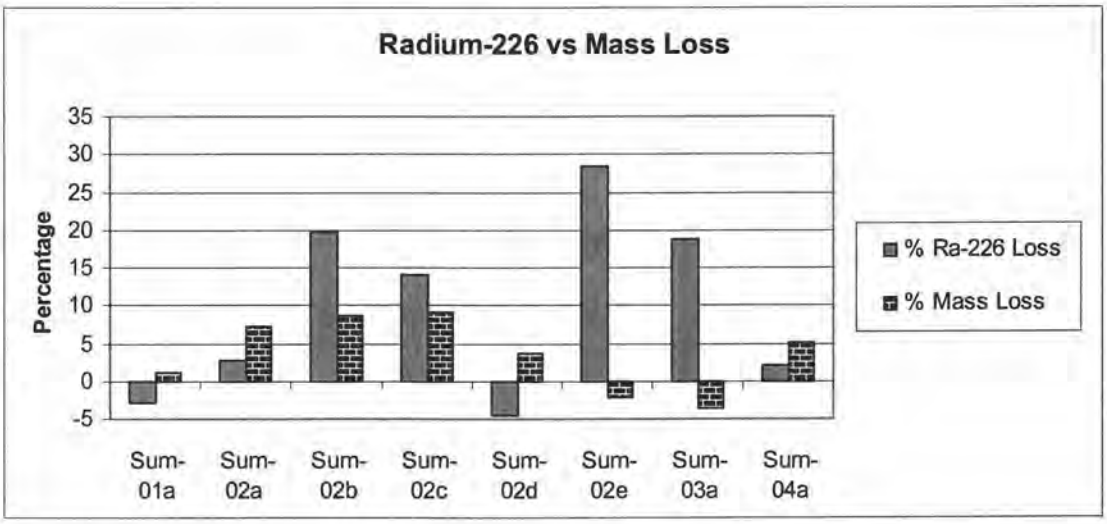


Figure 7.2 Comparison of ^{226}Ra lost versus mass lost for each of eight samples

Graphically, Figure 7.2 illustrates that the amount of mass lost is not indicative of the amount of ^{226}Ra lost in the digestive analogue. The ^{226}Ra lost from sample Sum-02e had a high associated uncertainty and is likely due to self-shielding within the sample itself, (a rock). Even after discounting Sum-02e's results, there can be no parallel drawn between how much soil was lost to solution and how much ^{226}Ra was transferred to solution.

7.10 DOSIMETRY CALCULATIONS

The committed effective dose equivalent for a person ingesting these soils can be calculated using the data in Table 3.3. The $e(\tau)$ is the committed tissue equivalent

dose per unit intake, where τ is the period in years over which the dose is calculated.[26] The age of the person is significant as metabolic data for radium changes with growth fluctuations.

The committed effective dose accounts for both the portion of ^{226}Ra that makes it to circulation and the dose delivered as the ^{226}Ra leaves the GI tract.[27] It is difficult to estimate the dose to the four compartments of the GI tract from that portion of the ingested activity that is evacuated with the feces (usually about 80% of the total ingested ^{226}Ra). However, if the activity is treated as a hot particle or many hot particles, the dose to the immediate surrounding tissue can be assessed. The following assumptions and equations have been used to calculate doses to portions of the GI tract:

- Hot particles in the GI tract are assumed to enter each GI compartment and be embedded in the tissue for the entire residence time in that compartment
    - Isotropic emission of alphas, but 100% absorption (conservative)
- ^{226}Ra average energy per disintegration 4.76 MeV (combination 4.777 MeV alpha, 4.591 MeV alpha and 186 keV gamma)
- Range of 4.76 MeV alpha in air
    - $R_{(cm)} = 1.24\ (4.76\text{MeV}) - 2.62 = 3.303$ cm
- Range of 4.76 MeV alpha in tissue
    - $R_{\text{tissue}} = \dfrac{R_{air} \times \rho_{air}}{\rho_{tissue}} = 4.271\text{E-}03$ cm in tissue

[26] e(τ) is calculated over 50 years for adults and over (70 – age) for children. (ICRP, 1996)
[27] This is the f_1 value or the bioavailable portion of Ra-226 from this analysis.

- Volume of hemisphere of tissue
    - $2/3\pi R^3$, where R is range in tissue in cm = 1.6322E-07 cm^3
- Weight of tissue
    - (Volume of tissue) x (density of tissue) = 1.6322E-07 g
- Assume 1 Bq (1 disintegration / second)
- Energy deposited per hour
    - $1\frac{dis.}{\sec} \times 4.76\frac{MeV}{dis.} \times 1.602E^{-13}\frac{J}{MeV} \times 3600\frac{\sec}{hr} = 2.746\text{E-}09$ Joule / hour
- Joules / mass in kg
    - 16.82 Gy per hour (from 1 Bq hot particle)

This is a simplistic assumption which neglects progeny, gammas and betas, but can be used as an initial estimate of the dose from the alphas of ^{226}Ra as the activity leaves the body. The dose estimate of 16.82 Grays per hour seems very large at first glance, but it should be noted that this dose is over a very small mass of tissue, (0.16μg). In his book, Eric Hall notes that "killing a small number of cells in a tissue matters very little; visible damage is evident only if a large enough portion of the cells are killed and removed from the tissue." (Hall, 2000) The volume that alphas affect is very small, ($1.6E^{-4}$ mm^3), and there is approximately 3264 cells in this tiny volume[28], versus an approximate 10 trillion cells in the whole body. (OSUSEP, 2006)

[28] There are approximately 20 million cells in a cubic millimeter. (OSUSEP, 2006)

Hall goes on to say that a hypothetical dose of 10 Gy targets dividing cells, but does not have any appreciable effect on the already differentiated, functioning cells. This is, of course, in reference to a whole body dose of 10 Gy, whereas the above considered dose is to a very small amount of tissue. The crypt cells (that are killed by whole body doses above 10 Gy) are the sensitive cells in the GI tract and are responsible for repopulating the GI tract with microvillus. If the doses calculated above are delivered to the lining of the intestines (SI, ULI or LLI) the cells killed would be replaced in a matter of days because they are regularly replaced after they are sloughed off and rubbed away by normal use. (Hall, 2000) The crypt cells would be unaffected and no long term effects would be noted from the short transit time through the intestinal tract.

The stomach is not widely discussed in radiation literature, as the focus for GI syndrome or the absorption of nutrients and radiation takes place in the small intestine. It is important to analyze what the effect these high doses would have on small amounts of tissue in the stomach. If the damage to a small amount of cells were likened to that damage from gastric ulcers, the symptoms would be similar (i.e., indigestion, nausea or a burning sensation in the upper abdomen). However, the cause of common ulcers is a bacterium called Helicobacter Pylori (H. Pylori), and the treatment is with antibiotics. (NetDoctor, 2006)

It is possible for a person to develop ulcers in the stomach from treatment with non-steroidal anti-inflammatories or NSAIDS, and it is more applicable to liken this sort of ulcer to one that might be caused by a hot particle in the stomach. If an ulcer-

type lesion in the stomach developed from killing of cells by hot particles, the treatment and prognosis would vary. If the ulcer were bleeding (peptic ulcer)[29], it might be necessary for a surgical procedure to stop the bleeding if it has perforated the gastric wall. (BUPA, 2006) If the ulcer were not severe, the only treatment option for the hot particle ulceration would be waiting, avoiding alcohol and smoking and losing weight if overweight. (BUPA, 2006)

If sample Sum-03a were ingested and it was assumed that the total activity of 6.74 μCi was in 1 Bq particles spread over each compartment of the GI tract, the total affected tissue in each compartment would be 0.0407 cm^3.[30] Even though the time spent in each section of the GI tract is longer than the stomach time of 1 hour, once the cells are dead, the additional dose is superfluous. Thus, in each section, killing just less than half a cubic centimeter of tissue is not likely to result in evident damage as the healthy GI tract will regenerate all surface cells in approximately 6 days. (About, 2006)

[29] Peptic ulcers are found in the duodenum, just below the stomach and are usually 1-2 cm in diameter. (BUPA, 2006)

[30] Calculated as: (6.74 μCi) x (3.7E04 Bq/ μCi) x (1.6322E-07 cm3 tissue/Bq) = total tissue affected in each compartment.

8. CONCLUSION

The bioavailability of ^{226}Ra in these samples was determined to be from zero to 28% using an *in vitro* methodology. It was also found that solubilities of ^{226}Ra from soils in water and HCl were not good predictors of bioavailability. The mass balance provided a lost fraction of total weight, but it could not be used to predict radionuclide bioavailability in gastrointestinal environments. Also, no correlation could be established between concentration of ^{226}Ra in soils and bioavailability.

The results from this analysis agree with the ICRP-67 Task Group estimation of 15-21% GI absorption of ingested ^{226}Ra. The f_1 value of 0.2 that is used for adults reflects not only the portion that is able to pass through the small intestine, but the demand for that compound in the body. Because radium behaves like barium and calcium in the body, growing children will absorb more than adults because of their nutritional demand for calcium. This is reflected in the increased f_1 values for infants and children.

The estimation of dose to a small amount of tissue from a potential hot particle is seemingly very high at 16.82 Gy per hour per Bq. The effects from this dose are seen in the immediate killing of only 3,264 cells per 1 Bq hot particle. The regeneration rate in the GI tract is very high and this miniscule loss of cells from an acute intake would likely go unnoticed, even with activities as high as those seen in sample Sum-03a (the highest activity sample). The dose to tissue from that portion of

the ingested Radium-226 that is non-bioavailable is very large but the impact is very low due to the tiny amount of tissue affected.

Future directions for this type of analysis should involve chemical speciation prior to simulated digestion and again after digestion. The chemical species may indicate performance of larger particles within the GI tract (i.e. particles of larger size might be found more readily accessible than has been hypothesized). Understanding chemical performance along with nutrient demand may further impact remediation efforts aimed at reducing risk of exposure via ingestion of ^{226}Ra.

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APPENDICES

APPENDIX A

DIRECTIONS FOR CREATING STOMACH AND INTESTINAL ANALOGUES

Mix the Saline HCl Solution as follows:

- Dissolve 17.56 g of Sodium Chloride in
- One (1) liter of Distilled Water and add
- 3.48 ml of concentrated HCl and
- Dilute to 2 liters with Distilled Water

To make Simulated Gastric Fluid

- Dissolve 4 g of Pepsin in
- 400 ml of the Saline HCl (from above)
- Stir on a magnetic stirrer

Mix the 0.3 M Sodium Bicarbonate as follows:

- Dissolve 50.4 g of Sodium Bicarbonate in
- One (1) liter of Distilled Water then
- Dilute to 2 liters with Distilled Water

To Make Simulated Intestinal Fluid:

- Dissolve 2.24 g of Pancreatin and
- 0.76 g of α-Amylase and
- 0.3 g of Bile Salts in
- 400 ml of the 0.3M Sodium Bicarbonate
- Stir on a magnetic stirrer

APPENDIX B

LABORATORY DIRECTIVE

1. Weigh empty "poly vial" and record weight to nearest .001 gram
2. Add sample, label poly vial on 2 sides, seal poly vial and weigh again, record weight
3. RECORD THE TIME
4. Calculate net weight (g)
5. Count sample in HPGe in the second position for 1200 seconds—watch dead time (INITIAL SOLID)
6. Prepare simulated stomach acid (watch temp and keep in stirring vessel)
7. Add entire soil sample, washing the sides of the poly vial with HCl to remove soil remnants
8. Timer should be set to count exactly ONE HOUR from the time soil was added
9. The pH should be tested periodically to ensure that pH is less than 3.5 (add HCl if necessary to lower pH)
10. The temperature should remain at 37 °C
11. Prepare intestinal phase, raise temperature
12. When timer sounds, add intestinal phase to stirring vessel of stomach acid and soil
13. Timer should be set to count exactly TWO HOURS from the time the intestinal component was added
14. The pH should be tested periodically to ensure that it is approximately 7.5
15. The temperature should remain at 37 °C
16. Upon completion of the intestinal step, carefully remove the stirring apparatus and the temperature probe from the vessel (make sure there is no sample stuck to them)
17. The liquid should then be slowly put through a vacuum filtration system (0.8 µm pore size) *gross 1-2 µm pre-filters can be changed if clogging occurs
18. The liquid phase should remain mixed—if it seems to be separating—mix well
19. Use a pipette to remove the top 5 ml of each sample and place each in pre-weighed poly vial
20. RECORD THE TIME, seal and label poly vial
21. Count poly vial for 1200 sec, recording the time (INITIAL LIQUID)
22. Save poly vials for possible counting later
23. Place the remaining liquid phase, uncovered for evaporation in the Isotemp® oven at 75 °C
24. RECORD THE TIME evaporation was started (evaporation should continue until the sample size is reduced to ¼ the initial volume)
25. Place the solid phase and associated filtration apparatus in the Isotemp® oven for drying

26. The solid phase will take approximately 24 hours to dry, but the weight should be assessed to confirm that it is constant before removing the sample from the apparatus
27. When the solid phase is at a constant weight (assessed only on a room temperature sample as weight can fluctuate with changing temperature), prepare to remove the filters from the filtration apparatus
28. In a fume hood, use a razor blade to cut the permanent filter from the apparatus and place the filter and all associated soil in a pre-weighed Petri dish for net weight.
29. Weigh the dry solid sample with the filters and record the weight
30. Return to the hood and use surgical tweezers to place the sample in a pre-weighed poly vial for counting
31. Seal the poly vial, wipe for potential removable contamination, label the sample name on two sides, and weigh the sample, recording the weight
32. RECORD THE TIME the sample was sealed
33. Count poly vial for 1200 sec in the same geometry previously used (FINAL SOLID)
34. When the evaporation is complete (typically takes 48 hours) remove the final liquid from the oven
35. Allow the sample to cool to room temperature
36. Use a disposable pipette to remove 5 mL of liquid and place in a poly vial
37. Seal the poly vial, wipe to remove contamination, label with the sample name
38. RECORD THE TIME the sample was sealed
39. Count poly vial for 1200 sec in the same geometry previously used (EVAPORATED LIQUID)

APPENDIX C

- Library of Radium Isotopes and Progeny for Gamma Spectral Analysis

- Certificate of Calibration Mixed Nuclides Standard Source

GammaVision Nuclide Library RaLib.Lib
Created: 10/26/2005 4:35:08 PM
Edited: 10/26/2005 4:37:53 PM

Ra-226	1600 Yrs.	5.0000	--------
185.99keV	3.28%		G-------
Ra-224	3.66 Days	5.0000	--------
241.00keV	3.9%		G-------
Bi-214	19.9 Min.	5.0000	--------
609.32keV	46.09%		G-------
1764.51keV	15.92%		G-------
1120.28keV	15.04%		G-------
1238.11keV	5.916%		G-------
2204.12keV	4.993%		G-------
768.36keV	4.885%		G-------
1377.65keV	4.02%		G-------
934.05keV	3.165%		G-------
1729.60keV	3.047%		G-------
1407.98keV	2.477%		G-------
1509.19keV	2.192%		G-------
1847.44keV	2.123%		G-------
1155.19keV	1.69%		G-------
665.45keV	1.563%		G-------
2447.71keV	1.553%		G-------
1280.96keV	1.474%		G-------
1401.50keV	1.386%		G-------
806.17keV	1.228%		G-------
2118.54keV	1.209%		G-------
1661.28keV	1.15%		G-------
Pb-214	26.8 Min.	5.0000	--------
351.99keV	37.1%		G-------
295.22keV	19.2%		G-------
10.80keV	13.6%		G-------
77.11keV	10.7%		G-------
241.92keV	7.47%		G-------
74.81keV	6.33%		G-------
87.20keV	3.7%		G-------
53.20keV	1.1%		G-------
785.95keV	1.09%		G-------
89.80keV	1.03%		G-------

Nuclide Flags	Peak Flags
T = Thermal Neutron Activation	G = Gamma Ray
F = Fast Neutron Activation	X = X-Ray
I = Fission Product	P = Positron Decay
N = Naturally Occcurring Isotope	S = Single-Escape
P = Photon Reaction	D = Double-Escape
C = Charged Particle Reaction	K = Key Line
M = No MDA Calculation	A = Not In Average
A = Activity Not In Total	

CERTIFICATE OF CALIBRATION
MIXED NUCLIDES STANDARD SOURCE

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Radionuclide A:	Sb-125	Customer:	OREGON ST. UNIV. RADATION CENTER
Radionuclide B:	Eu-154	P.O. No.:	RIP021
Radionuclide C:	Eu-155	Catalog No.:	GF-ML
Half Life (Sb-125):	2.759 ± 0.002 years	Reference Date:	June 1 1999
Half Life (Eu-154):	8.588 ± 0.008 years	Source No.:	661-21
Half Life (Eu-155):	4.846 ± 0.137 years		

Contained Radioactivity

Sb-125:	0.5506	μCi	Eu-155:	0.5881	μCi
Eu-154:	0.7231	μCi	Total Activity:	1.862	μCi

Description of Source

a. Capsule type:	D
b. Nature of active deposit:	Evaporated metallic salts
c. Active diameter/volume:	5 mm
d. Backing:	Epoxy
e. Cover:	Acrylic

Radioimpurities

None Detected

Method of Calibration

The source was assayed by gamma spectrometry using the 427.9 keV gamma ray of Sb-125 (0.297 gammas/decay), the 1274.4 keV gamma ray of Eu-154 (0.350 gammas/decay) and the 86.5 keV gamma ray of Eu-155 (0.311 gammas/decay)

Uncertainty of Measurement	Sb-125	Eu-154	Eu-155
a. Systematic uncertainty:	3.0%	3.0%	3.0%
b. Random uncertainty in assay:	0.6%	0.7%	0.6%
c. Random uncertainty in weighing:	0.0%	0.0%	0.0%
d. Total uncertainty at the 99% confidence level:	3.1%	0.0%	0.0%

NIST Traceability

This calibration is traceable to the National Institute of Standards and Technology.

Notes

1. Nuclear data were taken from IAEA-TECDOC-619, 1991.
2. IPL participates in an NIST measurement assurance program to establish and maintain implicit traceability for a number of nuclides, based on the blind assay (and later NIST certification) of Standard Reference Materials. (As in NRC Regulatorty Guide 4.15).

Daniel James Van Dalsem
QUALITY CONTROL

11 May 99
Date Signed

ISOTOPE PRODUCTS LABORATORIES
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IPL Ref No. 661-21

APPENDIX D

HIGH PURITY GERMANIUM SPECTROSCOPY REPORTS

Selected reports are included on the cd-rom found on the back cover of this document