

Chapter 4

ENVIRONMENTAL RADIATION

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4.1 INTRODUCTION

Environmental radiation permeates space, air soil, and water around us. It has been naturally present since the birth of Earth about 4.65 billion years ago in a massive supernova explosion creating the heavy elements on earth and the other rocky planets Mars, Venus and Mercury. Consequently, life has evolved in the ashes and remnants of this explosion which were gravitationally grabbed in space by the sun into our solar system in an environment which has significant levels of ionizing radiation.

The natural background radiation is low intensity radiation that is a part of our natural environment. We are continually being bombarded with neutrinos from the sun. Part of it is from constituents in the soil such as the common heavy metals of thorium and uranium and their daughter nuclides such as radium and radon. These are continuously transforming through the process of radioactive decay giving up emissions of alpha, beta and gamma radiation. Although not very abundant, they are widely spread and minerals containing them are practically everywhere.

Radiation originates from outer space as cosmic radiation, the ground as terrestrial radiation such as radon²²², and even from within our own bodies like carbon¹⁴ and potassium⁴⁰.

In the case of K⁴⁰ in living matter, with a 1.28 billion years half life, it emits a beta particle and becomes a stable member of the calcium family of isotopes.

For C¹⁴ in living matter, with a half life of 5730 years it emits a beta particle and becomes a stable member of the nitrogen family of isotopes:



In smoking, the Rn²²² decay product of radon Pb²¹⁰ (T_{1/2} = 22.3 y) exists in solid form in the particulate matter of tobacco leaves and consequently in smoke. It decays into Po²¹⁸, Po²¹⁴, then Po²¹⁰ (T_{1/2} = 138.38 d) which emits an energetic alpha particle with 5.3 MeV of energy

Radiation is present in the air we breathe, the food we eat, the water we drink, and in the construction materials used to build our dwellings. Certain foods such as bananas, coconuts and Brazil nuts naturally contain higher levels of radiation than other foods. Brick and stone homes have higher natural radiation levels than homes made of other building materials such as wood. The USA's Capitol, and Central Station in New York are largely constructed of granite, and contain higher levels of natural radiation than most homes.

Cosmic rays from outer space also bombard the earth with streams of highly energetic particles, and high energy particles from the solar wind continuously fall on the

earth and increase in intensity during solar flares and at the peaks of the 11 or 22 years solar cycle.

4.2 RADIATION EXPOSURE

Naturally occurring background radiation is the main source of radiation effective dose for most humans. Levels typically range from about 0.359 to 0.940 centiSievert, [cSv] or rem per person per year but can be reach more than 5 [cSv/(person.yr)].

Levels of natural or background radiation can vary greatly from one location to the next. For example, people residing in Colorado, USA, are exposed to more natural radiation than residents of the east or west coast because Colorado is reached by more cosmic radiation at a high altitude of one mile above sea level at Denver, its capital, and more terrestrial radiation from soils rich in naturally occurring uranium and thorium. Furthermore, a significant fraction of our natural exposure is due to radon, a low boiling point gas from the Earth's crust from the decay chain of thorium and uranium that is present in the air we breathe.

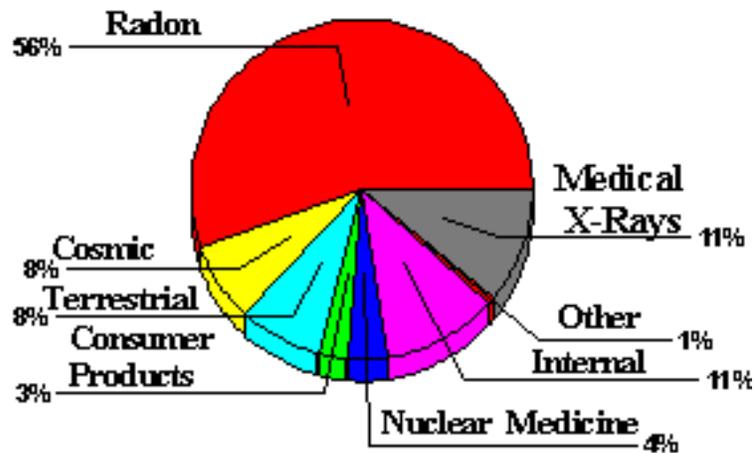


Fig. 1: Percentage contributions to the per capita annual effective dose equivalent.

There exist about sixty naturally occurring and 1,500 different radioactive nuclides, mostly man made. They are categorized as to their origin as:

1. Primordial: existing from before the formation of the earth.
2. Cosmogenic: formed as a result of cosmic ray interactions with Earth's atmosphere.
3. Human made: enhanced or formed due to human activity.

Assuming an average indoor radon concentration of 1.3 picocurie per liter [pCi/L], radon and its decay products is considered as the primary contributor to the radiation dose.

Figure 1 shows an estimate of the percentage contributions from different sources. Estimates of these doses differ from one source to another based on different assumptions and models. Effective dose estimates differ for instance according to models considering whole body irradiation, against others that consider effective dose to the gonads, or

internal dose effects. Table 1 shows an effective dose range of 0.359 to 0.940 cSv or rem from the low to high background radiation areas. Most of the exposure comes from radon gas and its progeny nuclei. Without the radon contribution, the range would otherwise be from 0.109 to 0.690 cSv or rem.

Table 1: Effective doses from different sources of exposure.

Source of exposure	Effective dose, [cSv] or [rem]	
Natural background (world population)	Normal background areas	High background areas
Cosmic rays	0.038 / a	0.20 / a
Terrestrial rays	0.046 / a	0.43 / a
Radionuclides in tissue	0.025 / a	0.06 / a
<i>Subtotal</i>	0.109 / a	0.690 / a
Inhaled radionuclides	0.250 / a	
<i>Total</i>	0.359-0.940 / a	
Medical diagnostics USA population	[cSv], [rem] per exam	
Skull	0.022	
Cervical spine	0.020	
Chest	0.008	
Cholioangiogram, liver	0.189	
Lumbar Spine	0.127	
Upper gastro intestinal series	0.244	
Abdomen (KUB)	0.056	
Barium enema	0.406	
Intravenous pyelogram	0.158	
Pelvis	0.044	
Hip	0.083	
Extremities	0.001	
Computed Tomography, (CT) scan, head or body	0.111	
Pediatic CT scan, abdomen	2.5, stomach dose	
Single screening mammogram	0.3, breast dose	
Space radiation exposure		
Astronaut, 3 day space shuttle mission	0.2–0.3	
Astronaut, 60 day space station mission	5.0	

Occupational exposure	
Average cumulative occupational dose in monitored radiation workers	Cumulative reported badge dose 2.0

4.3 HUMAN-MADE RADIATION

In 1895 x rays were discovered by Röntgen in Germany and have become since then a useful source of medical diagnosis and therapy. In 1896 radioactivity was discovered by Henry Becquerel in France. In 1934 it was discovered that radioisotopes can be made and their usage spread in hospitals laboratories and industries. In 1945 atomic devices were developed and tested originally in the atmosphere, underground, and then their testing was banned. The fission fragments and resulting from the atomic testing in 1950s still exist in the stratosphere and are descending to the Earth's surface as radioactive fallout.

Man made sources of radiation from medical, commercial, and industrial activities contribute 0.0672 cSv or rem to our annual radiation exposure. One of the largest of these sources of exposure is medical x-rays. Diagnostic medical procedures account for about 0.05 cSv or rem each year.

Other sources of technological radiation exposure include high flying airplanes, space travel, particle accelerators, and television and computer video screens. In addition, some consumer products such as tobacco, fertilizer, welding rods, kerosene lamps thorium mantles, luminous watch dials, and smoke detectors contribute another 0.0062 cSv or rem to our annual radiation exposure.

The additional man made radiation dosage is primarily as a result of the use of x rays in the search for decayed teeth, broken bones, lung lesions, swallowed objects, surgical instruments forgotten inside the body, and other useful or useless procedures. For instance, x rays are sometimes administered to investigate ankle sprains to eliminate the possibility of bone breaks, which otherwise could lead to an undesirable liability insurance situation.

Ionizing radiation is generated in a range of useful applications and medical, commercial and industrial activities. The most familiar and, in national terms, the largest of these sources of exposure is medical x-rays.

Natural and artificial radiations are not different in kind or effect. The breakdown of the effective doses from medical diagnosis applications is listed in Table 1.

4.4 HIGH BACKGROUND RADIATION AREAS, HBRAs

Background radiation levels are from a combination of terrestrial radiation from isotopes such as K^{40} , Th^{232} , Ra^{226} and cosmic radiation. They are occurring in air, water, soil, minerals, building materials and food supplies. About 60 radio nuclides exist in nature. The absorbed dose rate level is fairly constant over the world, being of the order of 8-15 [μ rad/hr]. About 82 percent of the radiation is from natural sources, particularly radon gas.

There are some areas with sizable populations that have high background radiation levels. These areas are primarily located in areas such as Guarapari, Brazil,

South-West France, the Kerala Coast, India, Ramsar, Iran and China. The higher radiation levels are due to high concentrations of radioactive minerals in soil. One such mineral, Monazite, is a highly insoluble rare earth mineral that occurs in beach sand together with the mineral Ilmenite, which gives the sands a characteristic black color. The principal radionuclides in Monazite are from the Th²³⁰ decay series, but there is also some Ra²²⁶ from the uranium decay chain. Monazite sands are used in the manufacture of industrial abrasives such as grinding wheels and sand paper.

Certain areas of southwest India in the Travancore-Cochin state along the coastline are characterized by patches of radioactive sand with the highest thorium content in the world at 33 percent. The highest known level of background radiation affecting a substantial population is in Kerala and Madras States in India where some 140,000 people receive effective dose rates which average over 1.5 cSv or rem per year from gamma radiation in addition to a similar dose from radon gas.

Comparable levels occur in Brazil and Sudan, with average effective dose rates up to about 4 cSv/yr or rem/yr to many people. The source is environmental radiation from deposits of thorium and uranium, with radon and its decay products nuclides. The Brazilian coastal sands contain radioactive monazite, zircon, thorianite, and niobate – tantalate, as well as non-radioactive ilmenite, rutile, pyrochlore and cassiterite.

Several places are known in Iran, India and Europe where natural background radiation gives an annual per capita effective dose rates of more than 5 cSv or rems and up to 26 cSv or rems at Ramsar in Iran. Lifetime effective doses from natural radiation range up to several hundred cSv or rems. There is no evidence of increased cancers or other health problems arising from these high natural levels.

In the Nile Delta, villages were built on Monazite alluvial deposits containing rare earth metals and thorium in the vicinity of the cities of Edku and Rosetta, East of Alexandria.

Monazite beach black sand at the village of Abou Khasba near Rosetta, Egypt, contains the elements Sc, Cr, Fe, Co, Zn, Cs, La, Eu, Sm, Hf, Pa, and Np, using activation analysis [4]. The detection of Pa and Np in the process of activation analysis suggests the presence of Th and U.

In Brazil, the Monazite black sand deposits are found along certain beaches. At Guarapari in Espirito Santo, Brazil people sun bathe over and rub it on their bodies ascribing to it a medicinal value in the cure of arthritis or rheumatism. The external radiation dose rate levels on these black sands range up to 5 mrad/hr, which is equivalent to 400 times the normal background radiation level in the USA. Some of the major streets of the surrounding cities have radiation dose rate levels as high as 0.13 mrad/hr, which is more than 10 times the normal background level.

High environmental radiation areas exist in Brazil in the state of Minas Gerais near Rio de Janeiro. The village of Guarapari with a population of 6,000 persons was tested with dosimeters disguised as religious medallions. The average gamma ray dose to the population was found to be 6 times the world's average with some exposures 30 times this amount. In that area is a large rare earths ore deposits that form the Morro do Ferro hill in Minas Gerais that rises about 250 meters above the surrounding area. An ore body near the top of the hill is close to the surface, and contains an estimated 30,000 tons of thorium and 100,000 tons of rare earth elements. The radiation dose rate levels near the top of the hill are 1 to 2 mrad/hr over an area of about 30,000 m². The ferns found there

have absorbed so much Ra^{228} , that they will produce an autoradiograph if placed on photographic paper with a radiation dose rate of 3 mrad/hr.

On the Southwest coast of India along the 570 km coastline of Kyrala, the Monazite deposits are larger than those in Brazil. The dose from external radiation is, on average, similar to the doses reported in Brazil, 500-600 mrad/yr, but individual doses up to 3,260 mrad/yr have been reported.

An area in China, has dose rates that are about 300-400 mrad/yr (3-4 mGy/yr). This is also from monazite that contains thorium, uranium and radium.

In areas of high natural background radiation, an increased frequency of chromosome aberrations has been noted. The increases are consistent with those seen in radiation workers and in persons exposed at high dose levels, although the magnitudes of the increases are somewhat higher than predicted. No increase in the frequency of cancer documented in populations residing in areas of high natural background radiation.

4.5 COSMIC RADIATION DOSES

Cosmic radiation can be divided into two types, primary and secondary. Primary cosmic radiation is made up of extremely high energy particles of energy up to the GeV energy level. They are mostly protons, with some larger particles such as Fe nuclei. A large percentage of it comes from outside of our solar system and is found throughout space. Some of the primary cosmic radiation is from our Sun, produced during solar flares.

The Earth's atmosphere and its magnetic field shield the Earth's surface from the primary cosmic rays, which are of concern for space missions. Little of the primary cosmic radiation penetrates to the Earth's surface, the vast majority of it interacts with the atmosphere. When it does interact, it produces the secondary cosmic radiation, which is what we actually see here on earth. These reactions produce other lower energy radiations in the form of photons, electrons, neutrons pions and muons that make it to the surface as cosmic ray showers.

The annual dose a person gets from cosmic radiation depends on the altitude. From cosmic radiation the USA, the average person will receive a dose of 27 mrem per year and this roughly doubles every 6,000 foot increase in elevation.

Typical cosmic radiation dose rates are: 4 $\mu\text{R/hr}$ in the northeastern USA close to sea level, 20 $\mu\text{R/hr}$ at 15,000 feet, and 300 $\mu\text{R/hr}$ at 55,000 feet. Thus the radiation dose increases with the altitude with a thinner shielding layer of the atmosphere.

There is only about a 10 percent decrease at sea level in cosmic radiation rates when going from pole to the equator, but at 55,000 feet the decrease is 75 percent. This is on account of the effect of the Earth's and the Sun's geomagnetic fields on the primary cosmic radiations.

Flying at high altitudes for long durations adds an extra radiation dose to the average annual dose as shown in Table 2.

Table 2: Cosmic ray radiation doses under normal solar conditions.

Route	Subsonic flight at 36,000 ft.		Supersonic flight at 62,000 ft.	
	Flight	Absorbed dose	Flight	Absorbed dose per

	duration	per round trip	duration	round trip
	[hrs]	[mrad]	[hrs]	[mrad]
Los Angeles-Paris	11.1	4.8	3.8	3.7
Chicago-Paris	8.3	3.6	2.8	2.6
New York-Paris	7.4	3.1	2.6	2.4
New York-London	7.0	2.9	2.4	2.2
Los Angeles-New York	5.2	1.9	1.9	1.3
Sydney-Acapulco	17.4	4.4	6.2	2.1

4.6 PRIMORDIAL RADIONUCLIDES

Primordial radionuclides were created in the nucleosynthesis process and supernovae events. The original nuclides and their decay products are typically long lived, with half lives often on the order of billions or millions of years. Initial radionuclides that exist for less than 30 half lives are not measurable.

Table 3: Occurrence of some primordial radionuclides in the lithosphere.

Nuclide	Half-life	Abundance	Specific Activity	Radiation
U ²³⁴	2.44x10 ⁵ a	0.0054 a/o U.		α, γ, SF
U ²³⁵	7.04x10 ⁸ a	0.72 a/o U		α, γ, SF
U ²³⁸	4.468x10 ⁹ a	99.2746 a/o U 0.5-4.7 ppm total uranium, common rocks. 2x10 ⁻⁶ [gm/gm soil]		α, γ, SF
Th ²³²	1.40x10 ¹⁰ a	100 a/o 1.6-20 ppm in common rock types crustal average: 10.7 ppm		α, γ, SF?
Ra ²²⁶	1600 a	2x10 ⁻¹² [gm/gm soil]	0.42 pCi/g in limestone 1.3 pCi/g in igneous rock	α, γ
Rn ²²²	3.824 d	noble gas annual average air concentration: 0.016 pCi/L to 0.75 pCi/L		α, γ

K ⁴⁰	1.28x10 ⁹ a	0.012 a/o K 3x10 ⁻⁴ [gm/gm soil]	soil concentration: 1-30 pCi/g	β, γ; ε, β ⁺
V ⁵⁰	>4x10 ¹⁴ a	0.2 ppm		γ
Rb ⁸⁷	4.8x10 ¹⁰ a	75 ppm		β
Cd ¹¹³	9x10 ¹⁵ a			β
In ¹¹⁵	5x10 ¹⁴ a	0.1 ppm		β
Te ¹²³	1.2x10 ¹⁵			α
La ¹³⁸	1.05x10 ¹¹ a	0.01 ppm		β, γ
Ce ¹⁴²	>5x10 ¹⁵ a			α
Nd ¹⁴⁴	2x10 ¹⁵ a			α
Nd ¹⁴⁵	>10 ¹⁷ a			α
Sm ¹⁴⁷	1.2x10 ¹¹ a	1 ppm		α
Sm ¹⁴⁸	8x10 ¹⁵ a			α
Sm ¹⁴⁹	>1x10 ¹⁶			α
Gd ¹⁵²	1.1x10 ¹⁴ a			α
Dy ¹⁵⁶	>1x10 ¹⁸ a			?
Hf ¹⁷⁴	2x10 ¹⁵ a			α
Ta ¹⁸⁰	>1.6x10 ¹³ a			ε, β?
Lu ¹⁷⁶	2.1x10 ¹⁰ a	0.01 ppm		β, γ
Re ¹⁸⁷	5x10 ¹⁰			β
Os ¹⁸⁶	2x10 ¹⁵			α
Pt ¹⁹⁰	6.1x10 ¹¹			α
Pb ²⁰⁴	1.4x10 ¹⁷			α
Bi ²⁰⁹	>2x10 ¹⁶			α
At ²¹⁵	0.1 ms			α, γ
At ²¹⁸	2 s			β
At ²¹⁹	0.9 m			β
Pu ²⁴⁴	8.3x10 ⁷ a			α, SF

4.7 COSMOGENIC RADIONUCLIDES

Cosmic radiation permeates all of space, the source being primarily outside of our solar system. The radiation occurs in many forms, from high speed heavy particles such as Fe to high energy photons, pions and muons. The upper atmosphere interacts with most of the cosmic radiation, and produces radioactive nuclides such as C¹⁴. They can have long half-lives, but the majority of them have shorter half-lives than the primordial nuclides. Their radiation is not very penetrating and they contribute little to the external exposure. However, in gaseous form like C¹⁴O₂, and HT³ they can be inhaled or ingested.

Beryllium¹⁰ with a half life of 1.6 million years is produced by cosmic rays and is detected in marine deposits. They are present in ferromanganese nodules dredged from a depth of 4,000 meters at 20 °N, 160 °W. From the activity of Be¹⁰, it was deduced that these nodules accumulated from the dissolved minerals in seawater at a rate of 1.8 +/- 0.5 mm per million years.

Table 4: Occurrence of cosmogenic radionuclides and activity density in air in the lower troposphere.

Nuclide	Half-life	Source	Specific Activity	Activity density [Trans/(min.m ³)]
C ¹⁴	5730 a	Cosmic-ray interactions, N ¹⁴ (n,p)C ¹⁴	6 [pCi/g] in organic material	4
T ³	12.33 a	Cosmic-ray interactions with N and O spallation from cosmic-rays, Li ⁶ (n, α)T ³ with cosmic ray showers	0.032 [pCi/kg]	10
Be ⁷	53.28 d	Cosmic-ray interactions with N and O	0.27 [pCi/kg]	1
Be ¹⁰	1.62x10 ⁶ a			10 ⁻⁷
Al ²⁶	7.3x10 ⁵ a			
Kr ⁸¹	2.1x10 ⁵ a			
Si ³²	700 a			2x10 ⁻⁶
Ar ³⁹	269 a			
Na ²²	2.6 a			10 ⁻⁴
S ³⁵	87.2 d			
Ar ³⁷	34.8 d			
P ³²	14.28 d			2x10 ⁻²
P ³³	25.3 d			1.5x10 ⁻²
Na ²⁴	15.02 hr			
S ³⁵	87 d			1.5x10 ⁻²
Si ³¹	2.62 hr			
Cl ³⁶	3x10 ⁵ a			3x10 ⁻⁸
Cl ³⁸	37.2 m			
Cl ³⁹	55.7 m			
Cl ^{34m}	32.2 m			

4.8 HUMAN MADE RADIONUCLIDES

Humans have added to the natural inventories. The amounts are small compared with the natural amounts. Due to the shorter half-lives of many of the nuclides, they have seen a marked decrease since the halting of above ground testing of nuclear weapons.

Table 5: Source of Human made radionuclides

Nuclide	Half-life	Source
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T ³	12.3 yr	weapons testing fission reactors nuclear fuel reprocessing nuclear weapons manufacturing
I ¹³¹	8.04 d	weapons testing fission reactors medical treatment of thyroid nodules
I ¹²⁹	1.57 x 10 ⁷ yr	weapons testing fission reactors
Cs ¹³⁷	30.17 yr	weapons testing fission reactors nuclear fuel reprocessing
Sr ⁹⁰	28.78 yr	weapons testing fission reactors nuclear fuel reprocessing
Tc ^{99m}	2.11 x 10 ⁵ yr	decay product of Mo ⁹⁹ used in medical diagnosis
Pu ²³⁹	2.41 x 10 ⁴ yr	bred by neutron capture in fertile U ²³⁸
U ²³³	1.592 x 10 ⁵ yr	bred by neutron capture in fertile Th ²³²

4.9 SOIL RADIOACTIVITY

Activity levels in soils vary greatly depending on soil type, mineral make up and density. The natural radioactivity in a square mile of soil to a depth of 1 foot is shown in Table 6.

Table 6: Natural radioactivity in a total volume of 7.894 x 10⁵ m³, with a soil density of 1.58 [gm/cm³].

Nuclide	Specific activity [pCi/gm]	Mass of nuclide	Activity found in volume of soil Ci
Uranium	0.7	2.2 metric tonnes	0.8
Thorium	1.1	12 metric tonnes	1.4
Potassium ⁴⁰	11	2 metric tonnes	13
Radium	1.3	1.7 g	1.7
Radon	0.17	11 µg	0.2
Total			>17

4.10 RADIONUCLIDES IN THE MARINE ENVIRONMENT

There is a considerable amount of radionuclides dissolved in the water of the world oceans. In the long term, uranium could be extracted from the ocean's water for energy production.

The total volume of water in the world oceans is about $1.3 \times 10^{18} \text{ m}^3$. The concentrations of some radionuclides are shown as well as their total activity.

Table 7: Occurrence of radionuclides in the water of the world's oceans.

Nuclide	Activity density [pCi/L]	Total activity Ci
Uranium	0.9	1.1×10^9
Potassium ⁴⁰	300	3.8×10^{11}
Tritium	0.016	2×10^7
Carbon ¹⁴	0.135	1.8×10^8
Rubidium ⁸⁷	28	3.6×10^{10}

4.11 RADIONUCLIDES IN FOOD AND WATER

The common radionuclides in food are K^{40} , Ra^{226} and U^{238} and their associated decay products. Brazil nuts are notorious for their 14,000 times content of radioactivity compared with common fruits. Cereals are also high as much as 500-600 times that of fruits. Fruits have the lowest content of natural radioactivity.

All muscle building foods such as meat contain potassium. The radioactive potassium K^{40} in our muscles provides 10-20 times the internal exposure of any of the other incorporated radionuclides such as Ra^{226} and C^{14} .

Table 8: Specific activities of radionuclides in food.

Food	Specific activity K^{40} [pCi/kg]	Specific activity Ra^{226} [pCi/kg]	Relative activity
Bananas	3,520	1	391
Brazil nuts	5,600	1,000-7,000	1,400
Carrots	3,400	0.6-2	378
White potatoes	3,400	1-2.5	378
Beer	390	-	43
Red meat	3,000	0.5	33
Lima bean	4,640	2-5	516
Drinking water	-	0-0.17	0.019
Cereals			60
Teas			40
Liver and kidney			15
Flours			14
Peanuts and peanut butter			12
Chocolates			8

Biscuits			2
Evaporated Milk			1-2
Fish			1-2
Cheese and eggs			0.9
Vegetables			0.7
Fruits			0.1

High natural radioactivity exists in the drinking water of several municipal water supplies and the well water from surface and deep aquifers in rural areas. The radioactivity was found to result from Ra²²⁶ which has been leached out of the soil, partly through chemical and bacterial action along with other common elements such as calcium, magnesium and mercury.

In Illinois, the deep wells at Joliet have 300 times more radium than Chicago's lake water.

Maine wells contain 3,000 times the radium concentration as the Potomac River in Washington DC. Some springs in Kansas, Colorado, and Jachymov in the Czech Republic, the concentration is 10,000 times greater. Such water is sometimes bottled and sold for profit to unsuspecting consumers as "mineral water", or "spring water." People seek relief from skin ailments and arthritis by swimming in the radioactive mineral springs of health spas.

Snake oil salesmen cures for all ills were ascribed to radium and thorium. It was later realized that radium resembles calcium so much chemically that it tends to accumulate in the human bones and at large concentration can cause serious damage.

4.12 RADIONUCLIDES IN THE HUMAN BODY, POTASSIUM⁴⁰

The occurrence of natural radionuclides in the body of an adult are shown in Table 9. K⁴⁰ is the most abundant radioisotope in the human body at 17 mg in a 70 kgs adult. It is a part of potassium as a constituent of the muscle system.

Table 9: Activity, daily intake and total amount of naturally occurring radionuclides in a 70 kgs adult.

Nuclide	Total Activity of Nuclide Found in the Body	Daily Intake of Nuclides	Total Mass of Nuclide Found in the Body
U	30 pCi	1.9 µg	90 µg
Th	3 pCi	3 µg	30 µg
K ⁴⁰	120 nCi	0.39 mg	17 mg
Ra	30 pCi	2.3 pg	31 pg
C ¹⁴	0.1 µCi	1.8 ng	22 ng
T ³	0.6 nCi	0.003 pg	0.06 pg
Po	1 nCi	~0.6 µg	0.2 pg

Natural potassium has three isotopes in nature:

$$\begin{aligned}
& {}_{19}\text{K}^{39}, 93.26 \text{ a/o} \\
& {}_{19}\text{K}^{40}, 0.012 \text{ a/o} \\
& {}_{19}\text{K}^{41}, 6.73 \text{ a/o}
\end{aligned}$$

Only K^{40} is radioactive with a long half life of 1.28×10^9 years. With a branching ratio of 89 percent it emits a negative electron to decay into Ca^{40} :



where $\bar{\nu}$ is an antineutrino associated for parity conservation in negative beta decay.

With a branching ration of 11 percent it undergoes an electron capture process with the emission of gamma photon turning it into the argon⁴⁰ isotope:



To determine the amount of K^{40} in the body of an individual, a phantom can be used. Phantoms (Fig. 2) are artificial bodies, approximately the size, shape and density of a human body, used for calibrating radiation counters such as a whole-body counter (Fig. 3). They are designed so that the radioisotopes they contain have similar distributions to those of the isotopes expected in the actual human body.

The phantom would contain a specified amount of the radioactive isotope such as K^{40} or I^{131} , as a reference value.

The number of nuclei of a radioactive isotope of natural abundance γ in g_{phantom} grams of a substance placed in a phantom is:

$$N = \gamma \frac{g_{\text{phantom}}}{M} A_v \quad (4)$$

The actual activity of the g_{phantom} grams is:

$$A = \lambda N = \frac{\ln 2}{T_{1/2}} N = \gamma \frac{g_{\text{phantom}}}{M} A_v \frac{\ln 2}{T_{1/2}} \quad (5)$$

If the reading from the counter is A_{phantom} and the background count is $A_{\text{background}}$, the net count from the phantom is:

$$A_{\text{phantom}} - A_{\text{background}}$$

If we measure in a tested individual an activity of A_{person} the net radiation is:

$$A_{\text{person}} - A_{\text{background}}$$

The ratio of the net measured activities is:

$$\frac{A_{person} - A_{background}}{A_{phantom} - A_{background}} = \frac{g_{person}}{g_{phantom}} \quad (6)$$

from which:

$$g_{person} = g_{phantom} \frac{A_{person} - A_{background}}{A_{phantom} - A_{background}} \quad (7)$$

As an example, let $g_{phantom} = 140$ grams of K be used in the phantom, the background level is 12,000 [trans/min], and the whole body counter reading of the phantom is 16,800 [trans/min].

If a person is counted in the whole body counter with a reading of 14,400 [trans/min], we can say that:

$$g_{person} = 140 \frac{14,400 - 12,000}{16,800 - 12,000} = 140 \frac{2,400}{4,800} = \frac{140}{2} = 70 \text{ grams}$$

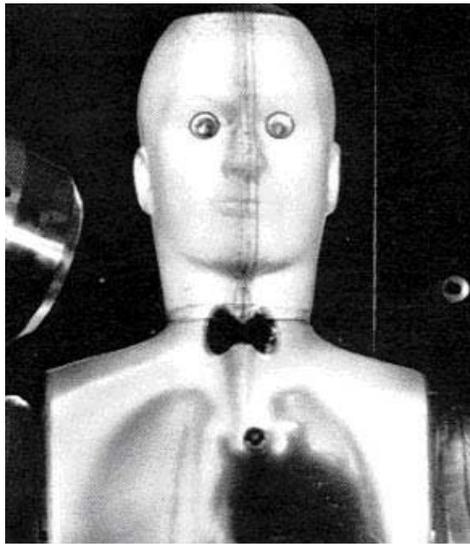


Fig. 2: Phantom for thyroid and I^{131} studies.



Fig. 3: Whole body counter for internal body radionuclides studies..

One can also calibrate the detector's efficiency as follows:

$$\eta_{counter} = \frac{A_{phantom}}{A_{\gamma}} = \frac{A_{phantom}}{f_{\gamma} \gamma \frac{g_{phantom}}{M} A_v \frac{\ln 2}{T_{1/2}}} \quad (8)$$

The number of nuclei of potassium in 140 grams of potassium is:

$$N' = \frac{g_{phantom}}{M} A_v = \frac{140}{40} 0.6021 \times 10^{24} = 2.11 \times 10^{24} \text{ nuclei}$$

The number of nuclei of K^{40} is:

$$N = \gamma \frac{g_{phantom}}{M} A_v = \frac{0.012}{100} \frac{140}{40} 0.6021 \times 10^{24} = 2.53 \times 10^{20} \text{ nuclei}$$

The activity from the K^{40} is:

$$\begin{aligned}
 A &= \lambda N = \frac{\ln 2}{T_{1/2}} N \\
 &= \frac{0.6931}{1.28 \times 10^9 \times 12 \times 30 \times 24 \times 60} 2.53 \times 10^{20} \\
 &= 2.64 \times 10^5 \left[\frac{\text{trans}}{\text{min}} \right]
 \end{aligned}$$

Fraction of these transformations emitting gamma rays:

$$A_\gamma = f_\gamma A = \frac{11}{100} 2.64 \times 10^5 = 2.91 \times 10^4$$

The efficiency of the detector is thus:

$$\eta_{\text{counter}} = \frac{A_{\text{phantom}}}{A_\gamma} = \frac{4,800}{2.91 \times 10^4} = 0.1649 = 16.49 \text{ percent}$$

Fatty tissues have a low potassium concentration than muscle tissue. It is thus possible to use K^{40} counting in the human body to determine the amount of lean tissue in any individual, and indirectly, the amount of fat. Estimates of the amount of fat based on specific gravity are not satisfactory. Variable and unmeasurable air spaces affect body density, and the immersion of a person in a water tank to determine the body's specific gravity is a clumsy and uncomfortable approach.

Variations in potassium content are found in persons with muscle diseases and malfunctions. A sharp drop in potassium content accompanies the profound muscle weakness, including the heart muscle that follows a diabetic coma. Administration of potassium produces improvement in the condition known as familial periodic paralysis.

Whole body counting of muscular dystrophy and myotonia atrophica patients shows there is a gradual and progressive decrease in body potassium.

4.13 RADIONUCLIDES IN CONSTRUCTION MATERIALS

The concentrations of uranium, thorium and potassium in construction materials are shown in Table 10.

Table 10: Radionuclides concentrations and specific activities in construction materials.

Material	Uranium		Thorium		Potassium	
	ppm	pCi/g	ppm	pCi/g	ppm	pCi/g
Granite	4.7	1.7	2	0.22	4.0	32
Sandstone	0.45	0.2	1.7	0.19	1.4	11.2
Cement	3.4	1.2	5.1	0.57	0.8	6.4
Limestone concrete	2.3	0.8	2.1	0.23	0.3	2.4

Sandstone concrete	0.8	0.3	2.1	0.23	1.3	10.4
Dry wallboard	1.0	0.4	3	0.32	0.3	2.4
By product gypsum	13.7	5.0	16.1	1.78	0.02	0.2
Natural gypsum	1.1	0.4	1.8	0.2	0.5	4
Wood	-	-	-	-	11.3	90
Clay Brick	8.2	3	10.8	1.2	2.3	18

By product gypsum is calcium sulfate from the ash from coal fired power plants and contains the radioactive substances left in the coal ash after burning it in power plants boilers.

4.14 URANIUM ORE MINERALS

INTRODUCTION

Uranium in small amounts is widely spread throughout the rocks of the Earth's crust, yet most of its occurrences are not useful for extraction. It is the major source of radium and radon gas in addition to thorium. The definition for an ore mineral as well as for an ore deposit depends upon its economical extractability and the future exploration, mining and metallurgical technology. A uranium mineral that is not considered an ore mineral today may become one in the future.

The main uranium ore minerals are pitchblende and uraninite which contain theoretically up to 85 per cent uranium but actually between 50 and 80 percent. Carnotite, torbernite, tyuyamunite, autunite, uranophane, and brannerite, contain about 45 to 60 percent.

In other minerals, uranium is an important but minor constituent of the minerals. Davidite, samarskite, and euxenite contain only 1 to 18 percent. The majority of uranium bearing minerals contain uranium in small or trace amounts as an accessory to other major constituents.

Most of the uranium minerals in pegmatites and placers are refractory where the uranium is present in combinations which are difficult to break down chemically in order to recover uranium. These minerals also usually occur scattered sparsely throughout the deposit so that recovery becomes difficult and expensive. Thus, even though some of the individual minerals may contain up to 50 per cent uranium, they are not considered as ore minerals.



Fig. 4: Uranium ore minerals.

PRIMARY URANIUM ORE MINERALS

Primary uranium minerals are found in veins or pegmatites. Flat lying deposits of pitchblende in sedimentary rocks have been discovered. The refractory primary uranium minerals are found in placers.

The primary uranium minerals are black or dark brown, dense and heavy, and have a shiny or pitch like luster. When exposed to weathering at or near the surface, they are sometimes altered to form bright colored secondary uranium minerals. There are three major primary uranium ore minerals, and the most important of these, uraninite and pitchblende, are really varieties of the same mineral.

URANINITE

Uraninite is combined UO_2 and UO_3 ; with 50-85 percent uranium oxide U_3O_8 . It is a naturally occurring uranium oxide with cubic or octahedral crystal form. It has a specific gravity of 8-10.5, a grayish-black color sometimes with a greenish cast and a hardness of 5-6 on the Moh's scale from 1 to ten, about the same as steel. Its streak is black. Its most widespread occurrence is in pegmatites, in which it is found in small amounts, throughout the world. It is an important constituent of all important primary deposits, occurring closely associated with its massive variety, pitchblende. In spite of its occurrence in pegmatites uraninite is not by itself a refractory mineral and should not be confused with the many complex uranium minerals also found in pegmatites.

Uraninite is the principal uranium-bearing mineral in the very low grade Witwatersrand and Orange Free State gold bearing conglomerates in South Africa, and the medium-grade uranium and copper-bearing carbonaceous slates at Rum Jungle, in

Northern Territory, Australia. In both of deposits uraninite occurs as finely disseminated crystals, usually invisible to the naked eye.

PITCHBLENDE

Pitchblende, like uraninite is combined UO_2 and UO_3 with 50-80 percent U_3O_8 . It is the massive variety of uraninite, without apparent crystal form, that occurs most abundantly in the rich primary vein deposits of uranium. It is the chief constituent of high grade uranium ores and has provided the largest part of all uranium produced throughout the world, forming the principal product of the Shinkolobwe mine, in the Congo; the Eldorado mine, Great Bear Lake, Northwest Territories, Canada; and the mines at Joachimsthal, Czechoslovakia.

Pitchblende is somewhat less dense than uraninite, having a specific gravity of between 6 and 9, but its other properties, with the exception of crystal form, are the same. It occurs as irregular masses often with a rounded, layered, botryoidal structure.

The principal occurrences of pitchblende are in primary hydrothermal vein deposits, usually of the mesothermal or medium temperature and pressure type, in igneous and metamorphic rocks and in flat lying bedded deposits in sedimentary rocks.

It is commonly associated with one or more of the primary ore minerals of iron, copper, cobalt, lead, silver, and bismuth; and the presence of these minerals in a mineral deposit is one indication of favorable conditions for pitchblende. It is usually accompanied also by bright colored secondary uranium minerals where subjected to weathering or other alteration. The commonly associated gangue, or non ore minerals in a vein or a deposit, are quartz and other silica minerals, carbonates, fluorite, barite, and hydrocarbons. Quartz, calcite, and dolomite are usually the most abundant. Pitchblende, in vein deposits, is most likely to be deposited in existing open spaces in rock formations, rather than by replacement of the rock itself, and the richest deposits occur where large open fractures were available for filling by the mineralizing solutions. There are no important pitchblende replacement deposits like those of copper, lead, zinc, and silver, where rock formations have been substantially replaced by ore through solution of the original constituents and deposition of the ore minerals.

Deposition of pitchblende is usually accompanied by strong alteration of the wall rock along the veins. The presence of hematite, a red iron oxide mineral, extending from the pitchblende a few inches to a few feet into the wall rock is the most characteristic feature. The formation of hematite has occurred in all of the major pitchblende vein deposits and in many of the deposits of minor importance. Other alteration features often associated with pitchblende deposition are the formation of kaolin, chlorite, sericite, and silica minerals in the wall rock. The alteration of the wall rock to form these minerals is known as kaolinization, chloritization, sericitization, and silification. Kaolinization causes the wall rock to become soft and clay-like, so that it may be easily gouged with a knife blade or even the fingernail. Chloritization and sericitization cause the rock to become a waxy or greasy green or gray, sometimes soft and flaky. Silification results in a hard flint-like texture.

In the flat lying deposits of pitchblende in sedimentary rocks, such as sandstones and conglomerates, the pitchblende is deposited between and around the grains of the rock and in available rock openings. The two most important examples are the copper-

uranium deposits in southern Utah and northern Arizona, in which pitchblende occurs with a variety of secondary uranium and copper minerals and copper and lead sulfides, and the deposits in Big Indian Wash near La Sal, Utah, in the central Colorado Plateau, where the pitchblende is associated with the vanadium mineral, vanoxite, and some secondary minerals, principally carnotite, tyuyamunite, and becquerelite. Pitchblende deposits in sandstone exist on the Laguna Indian reservation east of Grants, New Mexico, and significant pitchblende discoveries were made in the Black Hills, South Dakota, and the Wind River, Wyoming districts.

Pitchblende has also been found in smaller amounts disseminated in volcanic rocks in the southwestern United States, in some of the carnotite deposits of the Colorado Plateau, and in the deposits in limestone in the Grants district, New Mexico.

DAVIDITE

Davidite is a rare earth-iron-titanium oxide with 7-10 percent U_3O_8 . Davidite occurs at the old Radium Hill mine near Olary, South Australia, an early producer of small quantities of radium. Deposits near Tete in Mozambique. Davidite is a dark brown to black mineral with a glassy to submetallic luster. It has about the same hardness as pitchblende: 5-6 on the Mohr's scale. It has a low specific gravity of 4.5. It occurs most commonly in angular, irregular masses, sometimes with crystal outlines, but never in round, botryoidal shapes like pitchblende. When it is exposed to weathering, a thin yellow-green coating of carnotite or tyuyamunite may form on its surface. This is particularly true at Radium Hill, Australia.

Davidite is deposited in hydrothermal veins, presumably at a higher temperature and pressure than pitchblende. The veins have many of the characteristics of pegmatites. The associated vein minerals are ilmenite, hematite, biotite, mica, quartz, calcite, and pink feldspar. The rocks enclosing the veins at Radium Hill, Australia, are largely gneisses or schists, with chloritic and sericitic alteration near the veins. At Tete, Mozambique, davidite veins are found in more basic rocks like gabbro and anorthosite. Gabbro are rocks with a high iron, calcium, and magnesium content, as opposed to acidic rocks having a high sodium, potassium, and quartz content and which are the most common wall rocks of uranium vein deposits.

Davidite is almost never found as the "pure" mineral, but rather in complex intergrowths with ilmenite which has very similar physical properties and chemical composition.

SECONDARY URANIUM ORE MINERALS

Compared with the dull black, gray, and brown colors of the primary minerals, the secondary minerals present an array of bright yellow, orange, green, and all the shades of these colors. Some possess the property of fluorescence or the property of emitting light or glowing during exposure to ultraviolet light resulting in even more brilliant coloration.

They occur as earthy or powdery materials or as fine, delicate, needle-like or platy, flake-like crystals. They can be confused with other colorful minerals, such as malachite or copper carbonate, limonite or iron hydroxide, and sulfur.

The secondary uranium ore minerals deposits are numerous and widespread than those of the primary ore minerals. They have two major modes of occurrence:

1. In the weathered or oxidized zones of primary deposits they are formed by decomposition of the primary minerals in place.
2. As irregular, flat-lying deposits in sedimentary rocks, primarily sandstones. Also as conglomerates, shales, and limestones, formed by precipitation from solutions that may have carried the uranium some distance away from the original source.

The secondary uranium ore minerals occur frequently along with a large variety of other secondary uranium minerals, mainly the uranium phosphates, carbonates, sulfates, hydrous oxides and silicates, in what may be considered a third type of secondary mineral deposit. These have been referred to as oxidized secondary deposits or simply as oxidized deposits. Most of these deposits are oxidized vein deposits, the complete oxidation of the primary minerals in place making it difficult to prove the original primary character. They may be formed by ground-water solutions that have dissolved uranium from a broad area of slightly mineralized rocks and concentrated it by precipitation in veins and fracture zones. These deposits are numerous throughout arid and semi-arid regions, such as the western and southwestern USA, the west coast of South America, the Mediterranean area, and southern Russia.

The secondary minerals in the weathered zones of primary deposits have at some places contributed significant uranium production, particularly where weathering has been deep, as at Shinkolobwe in the Congo; at Urgeirica, Portugal; at Marysvale, Utah; and in some of the copper-uranium deposits of the southwestern USA.

The flat lying deposits in sedimentary rocks represent the most important occurrence of the secondary minerals, and the most important deposits of this type are the carnotite deposits of the Colorado Plateau area of Colorado, Utah, Arizona, and New Mexico, which have been radium, vanadium, and uranium producers since 1898.

Seventy five percent of the more than one hundred uranium minerals are secondary minerals, but of these only six may logically be considered ore minerals. Most of the others, many of them extremely rare, occur primarily as the weathering products in the oxidized zones of primary deposits, but some are found associated with the secondary ore minerals in deposits in sedimentary rocks. Unlike the primary uranium ore minerals, the secondary ore minerals seldom occur singly or only two to a deposit. They usually occur together in groups of several of both the ore and non-ore minerals, although, as in the case of the carnotite deposits, one mineral may be predominant. The dominant colors of the secondary uranium ore minerals are yellow and green, orange being confined primarily to the non-ore minerals.

CARNOTITE

Carnotite is $K_2O \cdot 2UO_3 \cdot V_2O_5 \cdot nH_2O$ with 50-55 percent U_3O_8 . It is a potassium uranium vanadate, the most important of the secondary uranium ore minerals, having provided about 90 percent of the uranium production from secondary deposits. It is a lemon-yellow mineral with an earthy luster, a yellow streak, and a specific gravity of about 4. It occurs most commonly in soft, powdery aggregates of finely crystalline material or in thin films or stains on rocks or other minerals. Its powdery nature gives the

impression of even greater softness than its Mohr's hardness scale rating of 2-3 would indicate. It can be easily scratched with the fingernail. Carnotite is not fluorescent.¹

The most noted occurrences of carnotite are in the Colorado Plateau area of the USA, where it was first identified in 1898 and has since provided the major domestic uranium production, on the western edge of the Black Hills, South Dakota, and in the Ferghana basin, Russia. It occurs in sandstones in flat-lying, irregular, partially bedded ore bodies of from a few tons to a few hundred thousands of tons in size. In the higher-grade deposits with more than one-third of 1 per cent U_3O_8 , the carnotite is present in sufficient quantity to color the rock a bright yellow; but in poorer deposits, particularly below 0.20 per cent U_3O_8 , it is often difficult to distinguish it from the sandstone itself. Its color is also often masked by iron staining or by the dark-colored vanadium minerals usually associated with it. Most carnotite deposits range in grade from 0.10 per cent to 0.50 percent U_3O_8 .

Although carnotite is the principal mineral in the carnotite deposits, over twenty other secondary uranium minerals are found associated with it. The most common of these is the secondary ore mineral, tyuyamunite. All of the other secondary ore minerals, torbernite, autunite, schroekingerite, and uranophane, have also been found in carnotite deposits. The other associated secondary minerals are the rare oxides, carbonates, arsenates, vanadates, phosphates and silicates. The most common non uranium minerals found associated with carnotite are the vanadium minerals, corvusite or hydrous-vanadium oxide, hewettite or calcium vanadium oxide, and roscoelite or vanadium mica-silicate. Minerals of the common metals, such as copper, lead, zinc, and manganese, have also been identified in carnotite deposits, as well as pitchblende and uraninite.

A general affinity of uranium for certain organic materials, which has had some effect on its deposition in almost all types of deposits, is perhaps most clearly displayed in the carnotite deposits of the Colorado Plateau area. In a large number of these deposits, the carnotite is intimately associated with silicified or carbonized wood fossil wood, and a variety of coal-like and asphaltic materials, all of which are good indicator substances for carnotite. In the Temple Mountain district, Utah, carnotite occurs in sandstones so impregnated with asphaltic material that the deposits are considered a special type and are called uraniferous asphaltite deposits. Elsewhere, fossil wood in the form of logs or accumulations of branches and twigs, locally called trash pockets, is the most common type of associated organic material.

Carnotite is present in varying amounts in nearly all of the other secondary uranium deposits and is the principal mineral in some deposits, like those at Jean and Erie near Las Vegas, Nevada, and near San Carlos, Chihuahua, Mexico. Carnotite is found also in small amounts in the oxidized zone of any primary uranium deposit containing even trace amounts of vanadium, for example, the davidite deposits at Tete, Mozambique, and at Radium Hill, South Australia, in the fluorite deposits of the Thomas Range, Utah, and other parts of the southwestern USA, and in places as thin stains and coatings at the outcroppings of the very low grade, uranium-bearing shale, phosphate, and lignite deposits.

TYUYAMUNITE

Tyuyamunite is $\text{CaO} \cdot 2\text{UO}_3 \cdot \text{V}_2\text{O}_5 \cdot n\text{H}_2\text{O}$ is 48-55 percent U_3O_8 . Tyuyamunite is closely related to carnotite as indicated by the chemical formula, which is the same except that calcium substitutes for the potassium of carnotite. The physical properties of tyuyamunite are the same except for a slightly more greenish color than carnotite and, in some cases, a very weak yellow green fluorescence not found in carnotite.

It is found in small amounts in almost any deposit or with any occurrence of carnotite. It is, as one would suspect, more abundant where there is an appreciable amount of calcium, usually in the form of calcite or limestone. Tyuyamunite first obtained importance as an ore mineral because of its occurrence in a deposit in southeastern Turkistan, near the town of Tyuya Muyun, for which it was named. It occurs there, and at other localities in the region, associated with other secondary uranium minerals, particularly carnotite and torbernite, in fractures in limestones, dolomites, and shales. It is also an important constituent of the deposits in limestone at Grants, New Mexico, and has been identified in the deposits at Big Indian Wash, Utah.

TORBERNITE AND METATORBERNITE

Torbernite and metatorbernite is $\text{CuO} \cdot 2\text{UO}_3 \cdot \text{P}_2\text{O}_5 \cdot n\text{H}_2\text{O}$ with 60 percent U_3O_8 . They are hydrous copper uranium phosphates, the only difference between the two being the number of water molecules present; their physical properties are identical. They have a bright emerald color, a pearly luster, hardness of 2-2 1/2 at about the same as the fingernail, and specific gravity of about 3.5 which is a little denser than quartz. They occur in flat, square, translucent crystals which usually fluoresce with a faint green color.

Torbernite and metatorbernite are the most common of the secondary uranium minerals that are found associated with primary deposits where oxidation has occurred. They are common in nearly all such deposits except pegmatites, which usually do not contain the necessary copper to form them. They are most noted for their abundance in the oxidized zones at Shinkolobwe, Joachimsthal, and in the copper-uranium deposits of Utah and Arizona. They have provided a substantial uranium production from the Urgeirica mine and nearby deposits in Portugal and from Marysvale, Utah, and they occur in the oxidized zone at Rum Jungle, Northern Territory, Australia. They occur with the other secondary uranium minerals in the oxidized secondary deposits whenever copper has been present in the depositing solutions or surrounding rocks. They are associated with tyuyamunite in Turkistan and with autunite at Bukhova, Bulgaria, and at Mt. Painter, South Australia. The principal non uranium minerals associated with torbernite are the clay minerals, limonite, quartz, pyrite, and the copper sulfides and carbonates.

AUTUNITE AND META AUTUNITE

Autunite and Meta autunite is $\text{CaO} \cdot 2\text{UO}_3 \cdot \text{P}_2\text{O}_5 \cdot n\text{H}_2\text{O}$ with 60 percent U_3O_8 . Reference to the chemical formula will show that these two minerals have the same composition as torbernite, with calcium substituting for copper. Because of this similarity, they are commonly found together.

The physical properties of autunite are similar to those of torbernite, except for its color, which is predominantly lemon or sulfur-yellow, although occasionally apple green,

and its brilliant yellow to greenish-yellow fluorescence in ultraviolet light. Autunite has a hardness of 2-2 1/2, is slightly denser than quartz with a specific gravity of 3.1, has a colorless to pale yellow or green streak, and occurs in small square, rectangular, or octagonal flat, translucent crystals or as thin coatings or stains on rock or other mineral surfaces.

Autunite is found in varying amounts in almost all deposits of the other secondary uranium minerals. It is an oxidation product of pitchblende and uraninite and most of the other primary minerals, and may also be derived from some other secondary minerals, like gummite and uranophane. As such it is an important constituent of the oxidized zones at Shinkolobwe and other important primary ore deposits and is a common secondary uranium mineral in most pegmatites. It is present in small amounts in many of the carnotite deposits of the Colorado Plateau area and in larger amounts in the tyuyaunite deposits of Turkistan.

It is an important constituent of the oxidized ores at Urgeirica, Portugal, and at Marysvale, Utah, and the most prominent mineral in the White Signal, New Mexico, district, at Mt. Painter, South Australia, and in the numerous low-grade secondary occurrences in the Mojave Desert and at other localities in southern California and Nevada. In addition, it frequently occurs as thin stains on fracture surfaces in granite and pegmatites in the Appalachian region of the eastern United States from Stone Mountain in Georgia to New England. The associated non uranium minerals are the same as for torbernite, except that the copper minerals may be absent.

URANOPHANE

Uranophane is $\text{CaO} \cdot 2\text{UO}_3 \cdot 2\text{SiO}_2 \cdot 6\text{H}_2\text{O}$ with 65 percent U_3O_8 . It is a hydrated calcium uranium silicate containing silica in place of the phosphate of autunite. It is slightly lighter in color and somewhat denser than autunite with a specific gravity of 3.85, and has a different crystalline form; it may occur as stains or coatings without apparent crystal form or as finely fibrous or radiating crystal aggregates.

The origin and occurrence of uranophane are very similar to autunite and torbernite. At least two of these three minerals are almost always found together, in proportions varying with availability of copper and phosphorus, uranophane becoming predominant where these two elements are scarce or absent. Although it has as broad a geographic occurrence as the other two, uranophane, with a few exceptions, is usually present in smaller quantities.

It is an important constituent of the secondary deposits in limestone near Grants, New Mexico, where it earned its reputation as an ore mineral, and in deposits in sandstone in southern Carbon County, Wyoming. It is also the most common secondary uranium mineral found in the noncommercial deposits in granite and pegmatites in the eastern USA. Its most noted occurrences of this type are at Stone Mountain, Georgia in granite, and at the Ruggles mine at Grafton, New Hampshire as pegmatite.

SCHROEKINGERITE

Schroekingerite is $\text{NaCa}_3(\text{UO}_2)(\text{CO}_3)_3(\text{SO}_4)\text{F} \cdot 10\text{H}_2\text{O}$ with 30 percent U_3O_8 content. It is a complex hydrated sulfate, carbonate, and fluoride of calcium, sodium, and

uranium. It has a yellow to greenish-yellow color with a pearly luster, a bright yellow-green fluorescence, and a paler yellow or greenish yellow streak. It is very soft at less than 1 on the Mohr's hardness scale, easily water soluble, and is the least dense of the uranium minerals at specific gravity of 2.5. It occurs as globular coatings on rock fracture surfaces or as small rounded masses composed of aggregates of flaky crystals distributed through soft rocks or soil.

Schroekingerite is the least important of the uranium ore minerals. It is a significant constituent of the secondary ores at Marysvale, Utah, and probably occurs in small amounts in the oxidized zones of most of the important primary deposits. The only known occurrence in which schroekingerite is the principal mineral is at Lost Creek near Wamsutter, Wyoming. It occurs there as small pellets distributed through clay beds at or near the ground surface over a considerable area to form a low grade uranium deposit.

4.15 ENVIRONMENTAL MONITORING AROUND NUCLEAR POWER PLANTS

Before a nuclear reactor is granted an operation license, health physicists make reference measurements of the natural radiation background. Under operation the effluent cooling water is checked for beta ray activity compared with the intake from local streams.

The atmosphere in a reactor building is passed on filters which are checked for the daughters of radium before its discharge. The surroundings of nuclear power plants are routinely monitored for the presence of environmental radionuclides.

As an example we consider the Wolf Creek generating station radiation environment in 1998.

The radionuclides detected in the environment included Ac^{228} , Be^7 , Co^{60} , Cs^{137} , T^3 , K^{40} , Ra^{226} , Sb^{125} , Sr^{89} , and Th^{228} . The isotopes Ac^{228} , Be^7 , K^{40} , and Th^{228} are naturally occurring radionuclides common to most sample types.

The isotopes Cs^{137} , T^3 , Sb^{125} , and Sr^{89} are fission products formed from reactor operation. The isotope Co^{60} is an activation product of the alloying element Co^{59} occurring in nature with a 100 percent abundance, in steel components.

A small background component of T^3 with a half-life of 12.3 years and Cs^{137} with a half-life of 30.1 years, activity is due to the nuclear weapons tests during the 1950 in the atmosphere.

Small amounts of T^3 are also produced naturally from cosmic ray interactions with water vapor in the upper atmosphere. Background levels of Cs^{137} and T^3 are accounted for in control samples and in pre operational data. These fission and activation product activity levels are not above expected regulated levels and are very small when compared with activity levels due to naturally occurring radionuclides.

The most significant radionuclide present in surface water samples collected was T^3 , a beta emitter. The highest T^3 concentration measured was 13,355 pCi/L. This maximum tritium concentration is 67 per cent of the National Primary Drinking Regulation Maximum Contaminant Level (MCL) of 20,000 pCi/L. The water from the water cooling lake is not used as a drinking water source. All other surface water, ground water, and drinking water samples collected in the environs indicated no radio nuclides present attributable to the operation of the power station.

Algae and sediment samples are excellent indicators for the monitoring of a fission and activation product buildup in the environment surrounding the power plant. Algae samples have been the best indicators for monitoring the seasonal fluctuations of fission and activation product levels in the water cooling lake. Sediment samples have been excellent indicators for the long term buildup of fission and activation product activity levels in the lake. The highest fission product activity other than tritium was 623 pCi/kg-dry of Cs¹³⁷ found in a bottom sediment sample. The highest activation product activity observed was 1,537 pci/Ci/kg-dry of Co⁶⁰ found in the same bottom sediment sample.



Fig. 5: Cooling lake around Wolf Creek power plant, Burlington, Kansas.

Airborne sample analysis showed that no radio nuclides attributable to the plant's operation were present above the lower limits of detection. The highest gross beta activity noted was 0.073 pCi/m³, due primarily to naturally occurring Rn²²² progeny, specifically the long-lived isotope Pb²¹⁰ with a half life of 22.3 years.

Sample analysis of terrestrial vegetation, soil, milk, grain, domestic meat, and aquatic vegetation samples collected in the environs of the plant indicated no radio nuclides present attributable to its operation. The isotope Cs¹³⁷ was identified in some soil samples and a pasturage sample. A control soil sample showed a maximum Cs¹³⁷ activity of 1,016 pCi/kg-dry, and a pasturage sample showed a Cs¹³⁷ activity of 107 pCi/kg-dry. The presence of ¹³⁷Cs is not attributed to the operation of the power plant.

Sample analysis of edible fish portions collected in the environs of the plant showed that no gamma emitting radio nuclides were present caused by the plant operation. All edible fish samples were analyzed for T³ in tissue fat and water. A total of twenty-seven fish was taken from the cooling lake. The highest T³ concentration in tissue was 18,349 pCi/kg-wet found in a composite sample of two Smallmouth Buffalo taken at the lake's ultimate heat sink. Using an ICRP 30 dose conversion factor for ingestion:

$$(h_{E,50}) = 6.40 \times 10^{-8} \text{ [mrem / (pCi T}^3 \text{ ingested)]},$$

a standard man consuming 21 kg/yr of fish containing 18,349 pCi/kg T³ would receive a Committed Effective Dose Equivalent (CEDE) of:

$$\begin{aligned}
 CEDE &= 18,349 \left[\frac{pCi}{kg} \right] \times 21 \left[\frac{kg}{yr} \right] \times 6.4 \times 10^{-8} \left[\frac{mrem}{pCi} \right] \\
 &= 0.0247 \left[\frac{mrem}{yr} \right]
 \end{aligned}
 \tag{9}$$

The projected yearly dose equivalent from tritium in fish is far below the 100 mrem/yr regulatory limit set for an individual member of the public.

Control samples taken on the Neosho River showed tritium activity ranging from 6,856 pCi/kg-wet to 10,329 pCi/kg-wet. These results are above the Low Effective Dose LED, of 1,200 pCi/kg-wet and are an indication of a problem with the analytical technique, scintillation fluorescence, chemiluminescence, cross-contamination, or a combination thereof in the Radiochemistry Laboratory.

Direct radiation monitoring sites showed no significant changes from pre operational data. The grand yearly average of all sites was 70 mR/yr, corresponding to 8 µR/h. The lowest direct radiation levels are found closest to the plant. The direct radiation levels on the plant baffle dikes at the 1,200 m exclusion area boundary are the lowest of any monitored site. The average baffle dike direct radiation level was 49 mR/y, corresponding to 6 µR/h. Limestone was used to construct the baffle dikes, and has a lower natural background radioactivity than the original soil present before the construction of the plant. This effect of construction on the terrestrial component of natural background radiation was noted on radiation surveys conducted around the site before bringing the initial fuel load on the site. The water from the plant also acts as an effective shield from terrestrial radiation that was present before lake filling.

4.16 RADON GAS DOSIMETRY

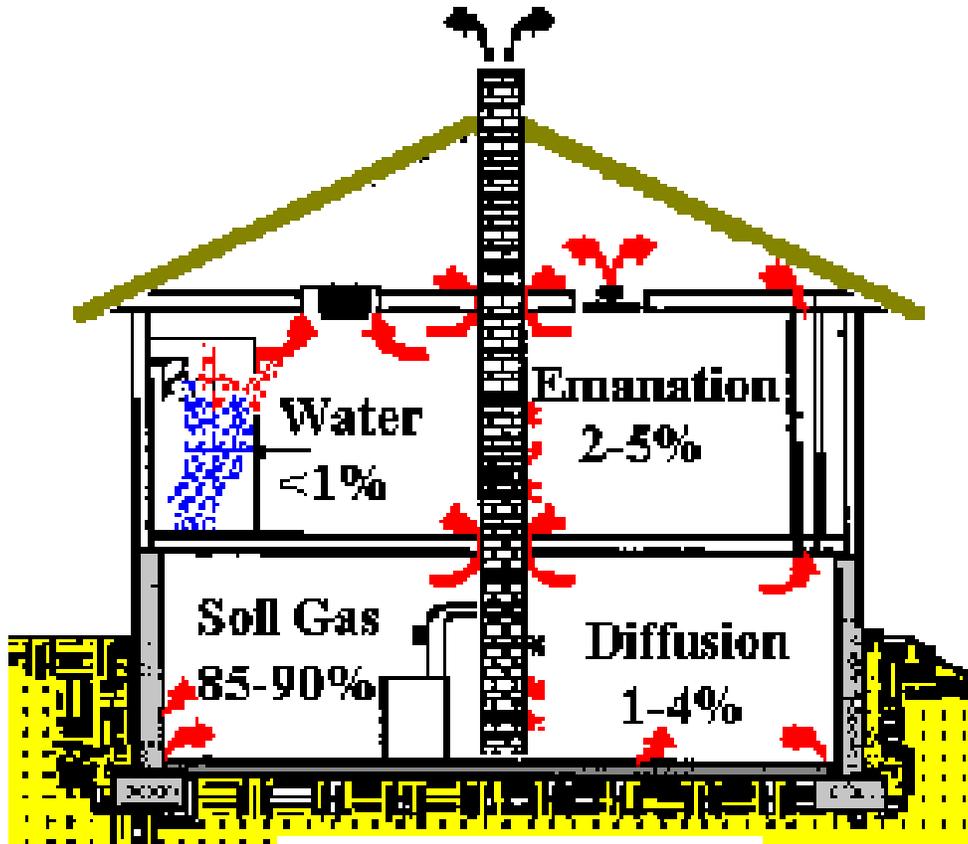


Fig. 6: Source of radon gas into a typical residence.

INTRODUCTION

Radon is the second leading cause of lung cancer in the USA with about 20,000 lung cancer deaths each year related to radon exposure. A radioactive, invisible, odorless gas that comes from the decay of naturally occurring uranium in the Earth's soil, radon can accumulate in homes at dangerous levels. Because families spend more time indoors during the winter months, January is an opportune time to test for radon.

Radon is a noble radioactive gas of natural origin. It is produced as a decay product of the disintegration of the uranium, thorium and radium located in the Earth's crust. Possessing a low boiling point, it is gaseous at normal temperatures, whereas the other decay products remain in solid at these temperatures. It constitutes the chief source of natural exposure to ionizing radiation. Radon exposure is omnipresent for the general public, but with variable concentrations, because radon mainly comes from granitic and volcanic subsoils as well as from certain construction materials.

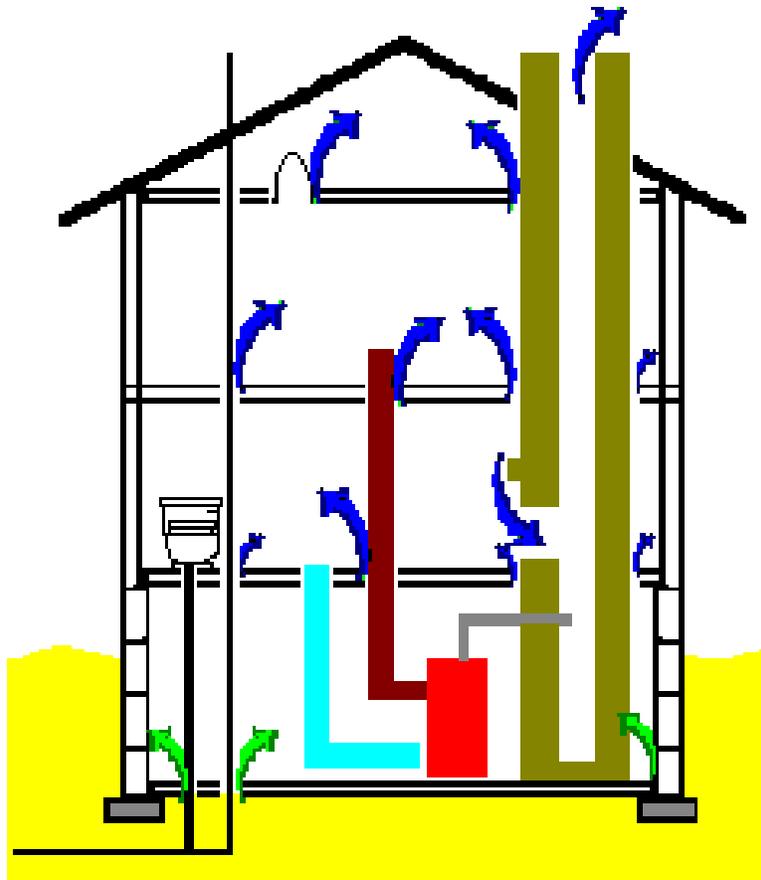


Fig. 7: Radon pathways into a home through the chimney effect.

In 1987, the International Center for Cancer Research (ICCR) in France declared radon to be carcinogenic for humans causing lung cancer based on the results of experimental animal studies and epidemiological studies among uranium miners. Results available at the time were related to very high exposure levels, and the extrapolation of this risk for the general public exposed to far lower levels in a domestic environment is raising numerous debates.

The sources of radon into a typical residence is shown in Fig. 6. These primarily result from soil gas with lower contributions from emanations from construction materials, diffusion from the soil, and from the water source if obtained from wells.

Because of a vacuum situation created by the Venturi effect and the chimney or stack effect, the radon gas is effectively being pumped from the ground, leading to concentration levels inside homes that could exceed the outdoors concentration, particularly in well insulated homes with insufficient air circulation (Fig. 7).

Today, the data for the annual exposure of uranium miners are closer to the concentration levels inhaled in specific homes, and current discussions bear more particularly on the interaction of radon with other components present in the mine environment but absent from housing, which might interact when a cancer develops, or on the possibility of a multiplicative action between radon and tobacco smoke.

Inside the most exposed housing with an activity density of more than 1,000 Bq/m³, annual exposure levels are close to or higher than the exposure limits accepted in

professional environments, and the results of the study of cohorts of uranium miners show an excess of lung cancer for these concentrations cumulative over 15-20 years of exposure.

Below an activity density level of 1,000 Bq/m³ the results currently available relating to environmental exposures are not always consistent and some uncertainties remain. The respective influences of the duration and the intensity of exposure are as yet not well known. It is not certain that an exposure to 400 Bq/m³ for 10 years presents the same risk as an exposure to 4,000 Bq/m³ during one year. Yet the importance and the urgency of the prevention message to be passed on to the public depend on the scale of the risk for a given level of exposure, taking into account age, gender, tobacco habits and other exposure factors.

PROPERTIES

Radon was discovered in 1900 by Friedrich Ernst Dorn in Germany. Named after the element radium, radon was called niton at first, from the Latin word "nitens" meaning "shining" but has been called radon since 1923. It is an essentially inert, colorless, odorless gas at ordinary temperatures. Its melting point is 202 °K and its boiling point is 211 °K. When cooled below the freezing point radon exhibits a brilliant phosphorescence which becomes yellow as the temperature is lowered and orange red at the temperature of liquid air.

The atomic radius is 1.34 Å (angstroms) and it is the heaviest known gas, being nine times denser than air. Because it is a single atom gas, unlike oxygen, O₂, which is diatomic, it easily penetrates many common materials like paper, leather, low density plastic in plastic bags, most paints, and building materials like gypsum board or sheetrock, concrete blocks, mortar, sheathing paper or tarpaper, wood paneling, and most insulation materials.

Radon is also fairly soluble in water and organic solvents. Although reaction with other compounds is comparatively rare, it is not completely inert and forms stable molecules with highly electronegative materials.

Radon is considered a noble gas that occurs in several isotopic forms. Only two are found in significant concentrations in the human environment: Rn²²², and Rn²²⁰. Rn²²² is a member of the radioactive decay chain of U²³⁸, and Rn²²⁰ is formed in the decay chain of Th²³². Rn²²² decays into a sequence of radionuclides called radon decay products, radon daughters, or radon progeny.

It is Rn²²² that most readily occurs in the environment. Atmospheric releases of Rn²²² results in the formation of decay products that are radioisotopes of heavy metals such as polonium, lead, and bismuth, that rapidly attach to other airborne materials such as dust and other materials facilitating inhalation.

OCCURRENCE AND USAGE

Radon is a noble gas. Only two of its isotopic forms are found in significant concentrations in the human environment: Rn²²² and Rn²²⁰. Their decay products are not gases and occur as unattached ions or atoms, condensation nuclei, or attached to particles. Rn²²² a decay product of U²³⁸ is commonly found in uranium and iron mines. Radon has

been used in some spas for presumed medical effects. Mineral water containing radium and consequently radon is marketed as natural and spring water possessing unspecified health effects. In addition, radon is used to initiate and influence chemical reactions and as a surface label in the study of surface reactions. It can be obtained by pumping the gases off a solution of a radium salt, sparking the gas mixture to combine the hydrogen and oxygen, removing the water and carbon dioxide by adsorption, and freezing out the radon.

PRODUCTION

Radon is not produced as a commercial product. Radon is a naturally occurring radioactive gas and comes from the radioactive decay of uranium and thorium. Most soils contain varying amounts of uranium and thorium. It is usually found in igneous rock and soil, but in some cases, rural well water may also be a source of radon. Municipal treatment eliminates most of it from treated municipal water supplies.

RADIATION EXPOSURE

The primary routes of potential human exposure to radon are inhalation and ingestion. Radon in the ground, groundwater, or building materials enters working and living spaces and disintegrates into its decay products. In comparison with levels in outdoor air, the concentrations of radon and its decay products to which humans are exposed in confined air spaces, particularly in underground work areas such as mines and buildings, are elevated. Although high concentrations of radon in groundwater may contribute to human exposure through ingestion, the radiation dose to the body due to inhalation of radon released from water is usually more important.

Concentrations of radon decay products measured in the air of underground mines throughout the world vary by several orders of magnitude. In countries for which data are available, concentrations of radon decay products in underground mines are now typically less than $1,000 \text{ Bq/m}^3$ of Rn or approx. 28 pCi/l. The average radon concentrations in houses are generally much lower than the average radon concentrations in underground ore mines.

Workers are exposed to radon in several occupations. Underground uranium and iron miners are exposed to the highest levels of radon and its decay products. Other underground workers and certain mineral processing workers may also be exposed to significant levels. Exhalation of radon from ordinary rock and soils and from radon rich water can cause significant radon concentrations in tunnels, power stations, caves, public baths, and spas.

Peripheral lymphocyte chromosomes from 80 underground uranium miners and 20 male controls in the Colorado plateau were studied. Taken into account were confounding factors such as cigarette smoking and diagnostic radiation. Groups that were increasingly exposed to radon and its decay products were selected. Significantly more chromosomal aberrations were observed among miners with atypical bronchial cell cytology, suspected carcinoma, or carcinoma in situ than among miners with regular or mildly atypical cells, as evaluated by sputum cell cytology.

The Environmental Protection Agency (EPA) and the Surgeons General's Office have urged widespread testing for radon. They estimated that as many as 20,000 lung cancer deaths are caused each year by radon. Next to smoking, radon is the second leading cause of lung cancer. The EPA says that nearly 1 in 3 homes checked in seven states and on three Indian reservations lands had screening levels over 4 pCi/L, the EPA's recommended action level for radon exposure.

Radon is a national environmental health problem. Elevated radon levels have been discovered in virtually every state. The EPA estimates that as many as 8 million homes throughout the country have elevated levels of radon. State surveys to date show that 1 out of 5 homes has elevated radon levels. Radon seeps into homes from the surrounding soil through cracks and other openings in the foundation. Indoor radon has been judged to be the most serious environmental carcinogen to which the general public is exposed and which the EPA must address. Based on current exposure and risk estimates, radon exposure in single-family houses may be a causal factor in as many as 20,000 of the total lung cancer fatalities which occur each year. The radon decay products Po^{218} and Pb^{214} , in solid form can attach to the surface of aerosols, dusts, and smoke particles which may be inhaled, and become deeply lodged or trapped in the lungs. Once lodged, they can radiate and penetrate the cells of mucous membranes, bronchi, and other pulmonary tissues.

Some scientific studies of radon exposure indicate that children may be more sensitive to radon than adults. This may be due to their higher respiration rate and their rapidly dividing cells, which may be more vulnerable to radiation damage.

ACTION LEVEL

All water and air contains some radon, though levels are normally much too low to be of concern. For many years it has been recommended that, if the concentration of radon in indoor air exceeds the Action Level (AL) of:

$$\begin{aligned} \text{AL} &= 4 \text{ [pCi/(liter of air)]} \\ &= 148.4 \text{ [Bq/(m}^3 \text{ of air)],} \end{aligned}$$

steps should be taken to reduce it. It is now recognized that some private water supplies contain levels of radon which should also be controlled. However, it is important to recognize that radon in water almost certainly presents a smaller public health hazard than radon in air.

NATIONAL HEALTH ADVISORY ON RADON

On January 13, 2005, USA Surgeon General Richard H. Carmona issued a National Health Advisory on radon. The advisory is meant to urge Americans to prevent this silent radioactive gas from seeping into their homes and building up to dangerous levels.

Indoor radon is the second leading cause of lung cancer in the USA, after smoking, and breathing it over prolonged periods can present a significant health risk to families all over the country. Yet, this threat is completely preventable, and does not

present freedom of choice issues like smoking or drinking. Radon can be detected with a simple test and fixed through well established venting techniques. Simple test kits can reveal the amount of radon in any building. Those with high levels can be fixed with simple and affordable venting techniques. According to the USA Environmental Protection Agency (EPA) estimates, one in every 15 homes nationwide has a high radon level at or above the recommended radon Action Level (AL) of 4 picoCuries (pCi/L) per liter of air.

WORKING LEVEL (WL) AND WORKING LEVEL MONTH (WLM)

The Working Level Month (WLM) is a measure of the exposure to airborne radon and its daughter nuclides. The Working Level (WL) is a measure of the atmospheric concentration of radon gas and its progeny. One WL is defined as:

$$\text{WL} \triangleq [\text{Combination of short lived radon daughters in 1 Liter volume of air that will result in the ultimate emission of } 1.3 \times 10^5 \text{ MeV of alpha particles energy}] \quad (10)$$

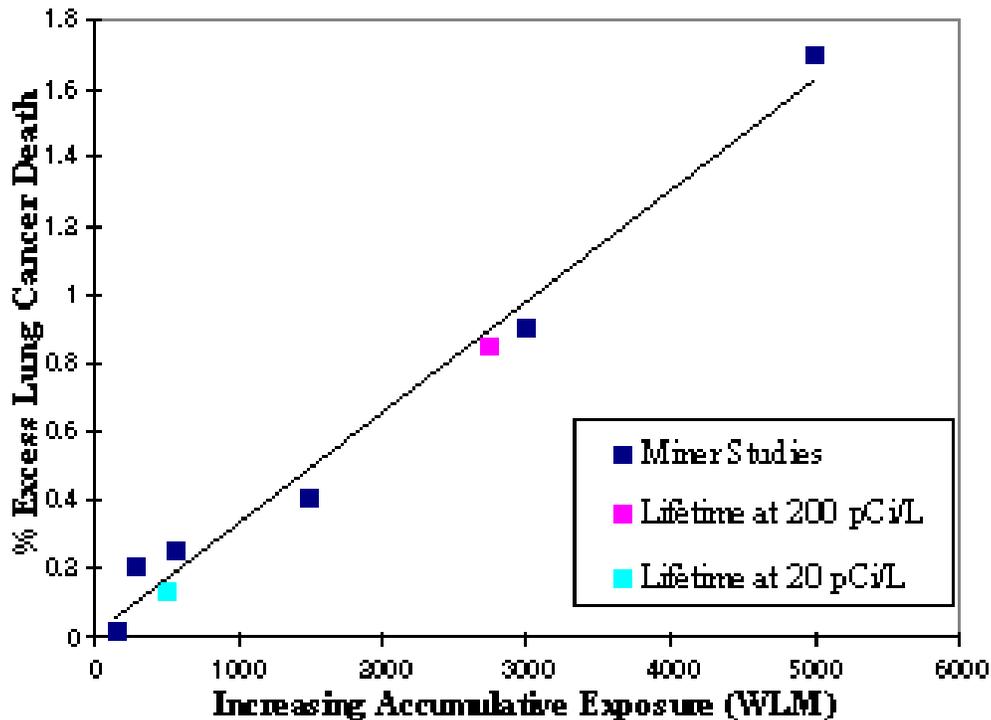


Fig. 8: Percent excess lung cancer risk from radon gas inhalation as a function of exposure in Working Level Months (WLMs).

This corresponds to:

$$1\text{WL} = 100 \left[\frac{\text{pCi of Rn}^{222} \text{ in equilibrium with its daughters}}{\text{Liter}} \right] \quad (11)$$

$$= 3,700 \left[\frac{\text{Bq}}{\text{m}^3} \right]$$

One WLM becomes the exposure resulting from inhalation of air with 1 WL concentration over a period of 1 working month or $8.5 \times 4 \times 5 = 170$ hours. One WLM exposure results in an alpha particles dose to the lungs of:

$$1 \text{ WLM} = 1 \text{ rad} = 1 \text{ cGy} \quad (12)$$

The relationship between the Action Level and Working Level is:

$$1 \text{ AL} = 4 \text{ pCi/l} = (4/100) \text{ WL} = 0.04 \text{ WL}.$$

RADON TESTING

Radon gas in the indoor air of America's homes poses a serious health risk. More than 20,000 Americans die of radon-related lung cancer every year. Millions of homes have an elevated radon level. Smokers' risk of lung cancer is much higher.

Homes are advised to be tested for radon every two years, and retested any time people move, make structural changes to a home, or occupy a previously unused level of a house. If a radon Action Level of 4 pCi/l or more is detected, steps must be taken to remedy the problem as soon as possible.

The following steps are recommended for the short term radon test. If the result is 4 pCi/L or higher a follow up test must be taken. The follow up test could be either a long-term test or a second short-term test. For a better understanding of the year round average radon level, a long term test must be taken.

The higher the initial short-term test result, the more certain one can be that a short-term rather than a long-term follow up test must be carried out. If the first short-term test result is several times the action level, for example, about 10 pCi/l or higher, one should take a second short-term test immediately. If a long-term test was taken as a follow up, the home must be fixed if the long term test result is 4 pCi/l or more.

If the follow up was a second short-term test, the higher the short-term results, the more certain one can be that the home must be fixed. Fixing the home must be considered if the average of the first and second test is 4 pCi/l or higher.



Fig. 9: Radon short term, long term and water test kits.

HUMAN EXPOSURE TO RADON AND ITS DECAY PRODUCTS

The concentration of radon in the air is higher in underground galleries than in houses and depends on ventilation and airing conditions as well as on the life-style of the occupants. The respiratory tract is radon's chief way of penetration into the human organism. Following inhalation, radon is rapidly exhaled because, as a noble gas, it has little affinity with the biological milieu, particularly the lungs.

Its radioactive decay products act in the opposite way. They are deposited as solids all along the pulmonary aerial tracts according to a distribution that is related to their granulometry. The chief descendants have a radioactive half life of the order of a few minutes, which limits their action to the lung tissues close to the deposit site, particularly the cells of the bronchial epithelium. More serious irradiation occurs by the alpha emitting decay products of Rn^{222} which possess higher ionization, such as Po^{210} which emits a powerful 5.3 MeV alpha particle with a half life of 138.38 days as it decays to the stable Pb^{206} isotope.

In the dusty atmosphere of mines, radon gas is in equilibrium with its daughter decay product Po^{218} . The concentrations of the three successive progeny: Pb^{214} , Bi^{214} and Po^{214} have been found in the mines' atmospheres at half the equilibrium values.

Analyzing the radon related health risk is a complex endeavor because the cumulated exposure is the result of a variable concentration of the air in a given place, weighted by the residence time at that place.

MINERS RADON EXPOSURE

Because of their professional exposure, pit workers and iron and uranium miners were among the first population groups to be subjected to epidemiological monitoring in order to evaluate the risk of cancer related to radon inhalation. The studies effectively carried out are cohort studies, that is to say studies that follow miner populations from the day they are hired by the mine, with a systematic and individual recording of their exposure and a health supervision program even after they leave their employer.

All the studies presented reveal higher cancer induced mortality when compared with a control-population not subjected to professional exposure. The studies have concluded that there exists indeed an excess mortality due to lung cancer and have shown

conclusively that the excess risk relative to lung cancer increases in a linear fashion according to the cumulated exposure to radon and its descendants. The results of these studies are shown in Fig. 10.

COEFFICIENT OF EXCESS RELATIVE RISK PER UNIT EXPOSURE (CERRUE)

Some studies suggest an inverse effect to the rate of exposure: with an identical cumulated exposure, a low rate seeming to induce a higher risk than the same cumulated exposure over a shorter period. These results have not been confirmed by other human or animal studies.

In the first publication relative to French uranium miners in 1993, a majority of the exposures to radon and its descendants were at low annual levels, and the duration of exposure was on average 14 years. An excess mortality due to lung cancer was shown and the increase in the risk of this cancer with cumulated exposure follows a linear relationship making it possible to estimate the slope called the Coefficient of Excess Relative Risk per Unit of Exposure (CERRUE):

$$\begin{aligned} \text{CERRUE} &= (0.3 / 1,000) \text{ [percent excess lung cancer deaths/WLM]} \\ &= 3 \times 10^{-6} \text{ [excess lung cancer deaths/cGy]} \end{aligned}$$

An extended study of this cohort and an additional study of a second population of French miners made it possible to make a new estimate of this coefficient which stands at between 0.35 and 0.8 percent per unit of exposure, depending on the study under consideration.

The results are close to the coefficient estimated from the international joint study. This study groups 11 cohorts of miners and was coordinated by the National Cancer Institute in the USA. It made possible the calculation of the risk based on 2620 cases of death by lung cancer observed in all these studies. The relative excess risk so obtained is 0.49 percent per WLM.

The analysis confirms the plausibility of the linear relationship and shows that the excess risk per unit of exposure decreases according to the age reached and to the time lapse since exposure or since the end of exposure. In addition, in those studies where data were available for tobacco consumption, the radon-tobacco interaction was also studied. Results show that this interaction is more than additive. It is in fact multiplicative or sub multiplicative. A joint international analysis studying the risk of cancers other than lung cancer has not shown, for any other localization, an increase in the risk as a function of cumulative radon exposure.

STUDIES OF THE CANCER RISK RELATIVE TO DOMESTIC EXPOSURE TO RADON

The risk of lung cancer relative to domestic exposure to radon was initially estimated by extrapolating the results observed in the professional environment to the general population.

These estimates were done by committees of international experts. However this approach has its limitations:

1. The absence of data for women among the cohorts of miners;
2. The uncertainties as to the effects of the dose rate;
3. Insufficient control of the confusion factors present in the mines and absent from houses such as gamma radiation and uranium dusts. On this last point in particular, exposure to gamma radiation and uranium dusts in mines has not been recorded in most countries. Other products such as arsenic and diesel fumes may exist in certain mines.

ECOLOGICAL STUDIES

Ecological-type studies have been carried out to verify the existence of a link between domestic exposure to radon and lung cancer at a geographical level, by comparing the rates of bronchial-pulmonary cancer in regions with a high and a low exposure to radon. The data used are, at the level of a same region, the average of the measurements of domestic exposure to radon and the average rate of death by cancer.

Such studies have been strongly criticized. Persons dying in a given region, and therefore forming the basis of the calculation of the annual death rate may not necessarily have lived for a very long time in that region, whereas the latency time for bronchial cancer after a given exposure is between 10 and 20 years. In addition, the average of the radon measurements at the level of a region is not an exact exposure indicator because intra-regional variability is often higher than inter-regional variability.

The chief criticism leveled at these studies is that of a poor accounting of one important risk factor, tobacco, as compared to a relatively low risk, radon. Indeed, tobacco consumption multiplies the risk of lung cancer by a factor of 5 to 10 among smokers as compared to non-smokers, whereas in the regions where the domestic concentration of radon is higher than the current recommendations given by the European Community (200 to 400 Bq/m³) the expected risk increase would be of the order of 1.2 to 1.5. The major role played by tobacco may explain the inverse relationship between radon and the bronchial cancer rate observed in certain ecological studies, in particular the Cohen study in the United States.

Ecological studies therefore do not provide, in cases of domestic exposure to radon, precise information sufficient to establish an estimate of the risk based on cumulated exposure.

CONTROL CASE STUDIES

Studies of control-cases among the general population have been set up, particularly during the last 10 years, with the aim of verifying in the field the results obtained in the context of professional exposure. Moreover, they aim to take into account co-factors such as active or passive tobacco consumption, something which has only rarely been done at the level of miners' cohorts, and they will also make it possible to study the risk for the female population.

The goal of these studies is to question persons suffering from lung cancer and "control" persons, in order to reconstitute a history of the houses occupied over a period of at least 30 years before inclusion and to measure the concentration of radon in these

dwellings. The minimum duration of 30 years was retained based on the studies of miners' cohorts showing that the risk diminishes over time since exposure. Distant exposures intervene less in the risk of cancer than more recent exposures.

Studies in Sweden, show a significant increase of the relative risk according to the level of domestic exposure. The relative risks are the following: 1.0; 1.1; 1.3; 1.8 (for, respectively: $\frac{3}{4}$ -50 Bq/m³, 50-80 Bq/m³, 80-140 Bq/m³, 140-400 Bq/m³, >400 Bq/m³). With constant tobacco consumption, this trend persists but in a non-significant way. It is worth noting that the excess is very high (relative risk = 32.5) for smokers of more than 10 cigarettes/day, living in houses where radon concentrations are higher than 400 Bq/m³.

Other studies, most notably a Canadian study in Winnipeg and also a Finnish study, do not highlight any positive trend of the relative risk with cumulated exposure to radon. A study in Missouri, where the cases are limited to women who were former smokers or non-smokers shows an absence of risk relative to radon after adjustment for former or passive tobacco consumption. A joint analysis, taking up the basic data from the 3 studies and limited to women (n = 1000), equally does not show a positive trend relative to cumulated exposure to radon.

CARCINOGENICITY

Radon's primary hazard is caused from inhalation of the gas and its highly radioactive heavy metallic decay products (Polonium, Lead, and Bismuth) which tend to collect on dust in the air. The problem arises when these elements stick to the delicate cells lining the passageways leading into the lungs.

There is sufficient evidence for the carcinogenicity of radon and its isotopic forms, Rn²²² and Rn²²⁰, in experimental animals. When administered by inhalation, preceded by a single exposure to cerium hydroxide dust, radon induced pulmonary adenomas, adeno carcinomas, invasive mixed adenosquamous carcinomas, and squamous cell carcinomas in male rats. Extrapulmonary metastases occurred in only one animal. Most or all of the tumors were believed to be bronchiolar or bronchio alveolar in origin. Radon decay products in combination with uranium ore dust induced a progression of activity from single basal cell hyperplasia in bronchioles to malignant tumors in male hamsters when exposed by inhalation. Lung tumors observed were adenomas, adeno carcinomas, and squamous cell carcinomas; bronchiolar and alveolar metaplasia, adenomatous lesions, fibrosis, and interstitial pneumonia were also observed.

When administered by inhalation in combination with decay products, uranium-ore dust, and cigarette smoke, radon induced nasal carcinomas, epidermoid carcinomas, bronchio alveolar carcinomas, and fibro sarcoma were observed in dogs of both sexes. In general, a significant increase was observed in respiratory tract tumors in rats and dogs in comparison with unexposed animals. A dose- response relationship was noted in experiments with rats in which radon was tested. In most instances, tumors at sites other than the lung were not reported, but in one study, mention was made of tumors of the upper lip and urinary tract in rats.

Reports exist about sufficient evidence for the carcinogenicity of radon and its decay products in humans. Increased incidences of lung cancer have been reported from numerous epidemiologic studies of groups occupationally exposed to high doses of radon, especially underground hard rock miners. These include particularly uranium

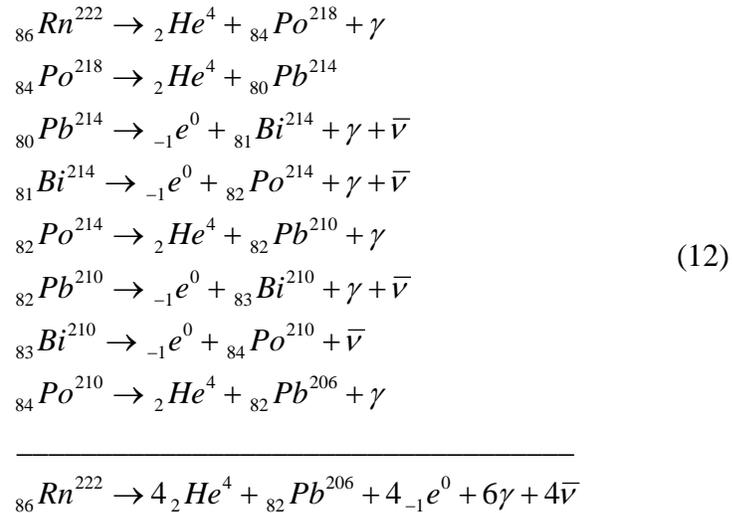
miners, but also groups of iron ore and other metal miners, and one group of fluorspar miners.

Strong evidence for exposure response relationships has been obtained from several studies, in spite of uncertainties that affect estimates of the exposure of the study populations to radon decay products. Several small case-control studies of lung cancer have suggested a higher risk among individuals living in houses known or presumed to have higher levels of radon and its decay products than among individuals with lower presumed exposure in houses. The evidence on the interaction of radon and its decay products with cigarette smoking with regard to lung cancer does not lead to a simple conclusion.

The data from the largest study are consistent with a multiplicative or submultiplicative model of synergisms and reject an additive model. In many studies of miners and in one of presumed domestic exposure, small cell cancers accounted for a greater proportion than expected of the lung cancer cases. In one population of uranium miners, this proportion has been declining with the passage of time. Because of the limited scale of epidemiologic studies of nonoccupational exposure to radon decay products available at the time reviews were made, quantification of risk has been based only on data of miners' experience.

RADON AND TOBACCO SMOKE

Tobacco smoke contains a naturally occurring radioactive element, Po^{210} . More of this element is found in the lung and other soft tissues of smokers than those of non smokers. It results as a decay product from gaseous Rn^{222} , and is deposited as a solid on tobacco leaves:



Po^{210} emits an energetic 5.3 MeV alpha particle with a short half life of 138.38 days, implying a high activity. It eventually decays into the stable isotope Pb^{206} .

Pb^{210} is the isotope decaying into Po^{210} . It is found in lung tissue and in the bones of cigarette smokers at twice the concentration normally present in the non smoking population.

Pb^{210} in the particulate matter of smoke is the primary source of the increased concentrations of both these radio elements in body tissues. These concentrations may increase by 8-30 percent the internal radiation dose to the bones of cigarettes smokers over that received by mankind as a whole.

The inhalation of these two isotopes in the particulate matter of cigarettes smoke delivers to the average smoker an effective dose rate of 8 cSv or rems per year to the basal cells of the bronchial tissue.

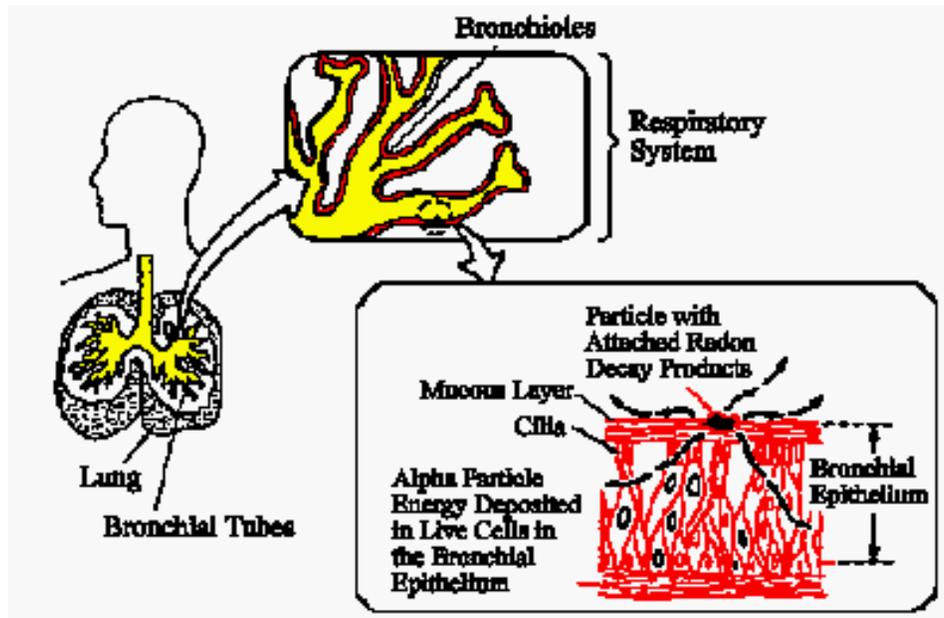


Fig. 10: Deposition of radon decay product radiation in the lung epithelium tissue.

A study of 383 cases of lung cancer in uranium miners shows evidence that alpha radiation from radon gas at exposure levels above 465 working level months (WLM) is a strong contributor to the development of lung cancer. Cigarette smoking plays the most significant role in causing lung tumor; this is also noticed in non miners who smoke cigarettes. A synergistic or additive effect of these two carcinogens is strongly suggested. The data indicate that small cell tumors develop in younger nonsmoking miners exposed to radon levels above 465 WLM. Lung cancers develop in smoking miners at lower levels of radon exposure than in nonsmoking miners. Based on an average mining experience of 15 years, there is substantial evidence that the present maximum allowable limit of 0.3 working levels (WL), or 4 working level months (WLM) per year, is safe, representing a margin of safety of approximately 10:1. A comparison of these data with the radon levels in some homes, averaging in the neighborhood of 0.025 WL, would indicate that health risks at these levels are negligible. It is suggested that 20 picocuries/liter, which equals 0.10 WL, be the maximum allowable level in homes.

4.17 RADIUM AND THORIUM ACCIDENTAL INTAKES

Men and women in the USA and Switzerland were exposed to radium toxicity. These were the painters of luminous dials in the watch industry around 1922. To thin out their painting brushes they engaged in the practice of licking it with their tongues, unaware of the toxicity of the radium containing paint.

Case records show the body burden of people who accidentally injected radium or thorium from different sources, is shown in Table 11.

Table 11: Body burden of cases of accidental injection of Ra²²⁶ and Th²³².

Case	Body burden [Bq]	Date of birth
Drank 210 bottles of Radithor in 1927.	63,640	1900
Drank about 78 ounces of Radium Water in 1932.	32,780	1897
Worked as radium chemist since 1946.	14,800	1925
Radium chemist for 7 years.	7,000	1922
Two injections of Thorotrast contrasting agent for x rays diagnosis.	3,300	1898
Radium dial painter in watch factory, 1918-1921.	72,500	1898
Radial dial painter for 4 ½ months , 1924.	1,924	1902

4.18 CESIUM AND STRONTIUM FROM NUCLEAR TESTING AND ACCIDENTS

A new radioactive contaminant has been introduced to the environment as Cs¹³⁷. It was discovered accidentally while counting K⁴⁰ when its 0.6616 MeV gamma ray showed up in the counting process. It was correlated to the nuclear testing in the atmosphere in the 1950s. Cs¹³⁷ has a half life of 30.17 years and once introduced into the body, it remains there for a lifetime. In the periodic table it mimics potassium, thus in soils poor in potassium, its intake is even larger in plants, and hence to humans through the food supply particularly beef and dairy products.

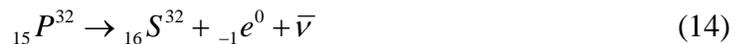
Later, the Chernobyl accident also released Cs¹³⁷ to its environment.

Strontium⁹⁰ another fission product which mimics calcium has also been detected from nuclear testing. Its half life is 29 years. Being a beta emitter, and not emitting any gammas, it is harder to detect.

In the case of reactor accidents at nuclear facilities neutrons could conceivably activate gold or silver teeth fillings, but a reaction of interest is the one producing P³² from S³² in human tissue, for instance human hair:



With a half life of 14.28 days decays through negative beta, without gammas emissions, back to S³²:



Like Sr^{90} , P^{32} is a pure beta emitter without gamma emission, and is consequently hard to detect. Counters that pick up the secondary bremsstrahlung radiation are needed in both cases.

At cyclotron facilities a hazard from Zn^{65} with a half life of 243.8 days exists. In the Marshall Islands at Rongelap Atoll people accumulated Zn^{65} in their bodies as a result of contamination of crabs and other food items by the fallout from the March 1954 testing at the Pacific Proving Ground.

At experimental reactor facilities a hazard exists from the inhalation of the three isotopes of iodine:

$$\begin{aligned} {}_{53}\text{I}^{131}, T_{1/2} &= 8.401 d \\ {}_{53}\text{I}^{133}, T_{1/2} &= 20.800 hr \\ {}_{53}\text{I}^{135}, T_{1/2} &= 6.585 hr \end{aligned}$$

In a nuclear criticality accident, the hazard would be from neutron bombardment of Na^{23} in the human body and the production of Na^{24} from the reaction:



Na^{24} emits two powerful gamma rays with energies of 1.38 and 2.76 MeV, with a half life of 15.02 hrs. Since a given neutron dose converts a known proportion of Na^{23} into Na^{24} , it is possible to determine how much neutron exposure a worker has received by obtaining his body count of radioactive Na^{24} .

4.19 RISK ASSESSMENT FOR RADON IN INDOOR AIR

The Environmental Protection Agency (EPA) indoor radon program promotes voluntary public actions to reduce the risks from indoor radon. EPA and the USA Surgeon General recommend that people do a simple home test and if high levels of radon are confirmed, reduce those high levels with straight-forward techniques.

Most inhaled radon is rapidly exhaled, but the inhaled decay products readily deposit in the lung, where they irradiate sensitive cells in the airways increasing the risk of lung cancer. EPA updated the Agency's estimates of lung cancer risks from indoor radon based on the National Academy of Sciences' (NAS) latest report on radon, the Biological Effects of Ionizing Radiation (BEIR) VI Report (1999). This report is the most comprehensive review of scientific data gathered on radon and builds on and updates the findings of the previous NAS BEIR IV Report (1988). NAS concluded that the findings of BEIR VI showed that if homeowners have not yet tested their homes for radon and remedied them if the levels are elevated, they are advised to do so.

Table 12: The lifetime risk of lung cancer death per person from radon exposure in homes from EPA risk assessment.

Radon activity density [pCi/L]	Never Smokers	Current Smokers^c	General Population
20	36 out of 1,000	26 out of 100	11 out of 100
10	18 out of 1,000	15 out of 100	56 out of 1,000
8	15 out of 1,000	12 out of 100	45 out of 1,000
4	73 out of 10,000	62 out of 1,000	23 out of 1,000
2	37 out of 10,000	32 out of 1,000	12 out of 1,000
1.25	23 out of 10,000	20 out of 1,000	73 out of 10,000
0.4	73 out of 100,000	64 out of 10,000	23 out of 10,000

The NAS BEIR VI Report confirmed EPA's long-held position that radon is the second leading cause of lung cancer and a serious public health problem. NAS estimated that radon causes about 20,000 lung cancer deaths each year. The report found that even very small exposures to radon can result in lung cancer and concluded that no evidence exists that shows a threshold of exposure below which radon levels are harmless. The report also concludes that many smokers will get lung cancer due to their radon exposure who otherwise would not have gotten lung cancer. This is because of the synergistic relationship between radon and cigarette smoking in causing lung cancer.

To update the EPA's previous risk estimates, EPA worked closely with the Agency's Science Advisory Board (SAB), an independent panel of scientific experts, to determine how best to apply the various risk models developed by the BEIR VI committee. EPA incorporated the SAB's advice and recommendations for modifying and extending the methods and approaches used in BEIR VI and constructed a single model yielding results midway between the results obtained using the two models preferred by the BEIR VI committee. These adjustments did not result in significant changes to the BEIR VI risk estimates.

EPA's updated calculation of a best estimate of annual lung cancer deaths from radon is about 21,000 (with an uncertainty range of 8,000 to 45,000) and is consistent with the estimates of the BEIR VI Report. [EPA's previous best estimate of annual lung cancer deaths from inhaled radon was based on the earlier BEIR IV Report and was about 14,000 (with an uncertainty range of 7,000 to 30,000).]

The SAB-endorsed modifications included applying the Agency's definition of excess risk that includes all radon-induced lung cancer deaths, rather than excluding premature deaths caused by radon in people who would otherwise have eventually died of lung cancer. EPA also used more detailed smoking prevalence data and more recent mortality data to calculate risks than were used by the BEIR VI committee. EPA also calculated numerical estimates of the risk per unit exposure [lung cancer deaths per working level month (WLM)], whereas BEIR VI estimated the fractional increase in lung cancers due to radon.

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EXERCISES

1. Radon²²² as a daughter in the decay chain of uranium is gaseous at room temperature. It is an inert or noble gas that does not interact chemically in the body. However it decays into Pb²¹⁰ which attaches itself to vegetation such as tobacco leaves as a solid and subsequently decays into Po²¹⁰ which emits an energetic alpha particle with 5.3 MeV of energy. The inhalation of these two isotopes in the particulate matter of cigarettes smoke delivers to the average smoker 8 rems per year to the basal cells of the bronchial tissue. The "cancer dose" is the total radiation dose that if spread through a population would cause one additional cancer death and is considered to be approximately 2,000 rems. Calculate the ensuing radiological risk in cancer deaths per year in a population of one million smokers.
2. Calculate the masses in metric tonnes of the radioactive elements in the world oceans from the data about their activity densities. If consuming 1.25 gm of U²³⁵ per day produces 1 MWth of power, calculate the amount of energy that can be produced from the uranium in the world oceans.
3. Express the recommended Action Level (AL) for radon gas in units of Bq/m³ of air, and pCi/Liter of air.
4. Express the Working Level (WL) for radon gas in units of Bq/m³ of air, and pCi/Liter of air.