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**RADIATION HAZARDS OF URANIUM  
MINING AND MILLING**

by

**R.M. FRY**

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ABSTRACT

As well as the need to control the more conventional hazards associated with any mining and milling operation, special care must be exercised in the mining and milling of uranium because of the radioactive nature of the ore. Uranium ore, though only mildly radioactive, contains a complex mixture of radioactive isotopes, the behaviour of which during each phase of the mining and milling process, determines the controls that must be established to ensure safe working conditions.

This paper examines each of the radiological problems that arise in these processes and explains their scientific background. The radiation standards that should be applied and the procedures that should be adopted to minimise radiation exposure to all persons are set down in the Australian 'Code of Practice on Radiation Protection in the Mining and Milling of Radioactive Ores 1975'. If the requirements of this code are adhered to, the radiological safety of miners, mill workers and members of the public can be assured.

The major operational requirement is to ensure that exposure of miners over their working lives to radon and its daughter products does not lead to an unacceptable increase in their chance of contracting lung cancer. Studies on the incidence of lung cancer amongst underground uranium miners indicate that this risk will be small if lifetime exposures are kept below about 120 'working level months', even amongst underground miners who smoke cigarettes. The risk is much smaller again for miners who do not smoke. Other hazards that must be controlled are exposure of miners and mill workers to external radiation and to dusts containing long-lived radioactive alpha emitting isotopes. Finally, the solid waste products from the mill (the tailings) which contain most of the naturally occurring radioactivity, must be properly impounded and after closure of the mill, stabilized to ensure long-term containment. Access by the public to the stabilized tailings must be controlled and habitation within the controlled area prohibited.

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INHALATION; LUNGS; MILLING; MINERS; MINING; NEOPLASMS; RADIATION  
DOSES; RADIATION HAZARDS; RADON; URANIUM; WORKING CONDITIONS

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## HISTORY OF URANIUM MINING

Uranium is not an uncommon element. It is widely distributed in all kinds of rocks and is readily measurable in the sea. There are about 3 parts per million by weight in the Earth's crust (i.e. about 3 grams per tonne) and 3 parts in  $10^9$  in the sea, giving a total mass in the ocean of some  $4 \times 10^9$  tonnes. It is about 1000 times as abundant as gold and is as common as zinc.

Despite its abundance and its occurrence in a great number and variety of minerals, uranium is rarely found in concentrations sufficiently high to warrant mining. Although some research and development has been directed towards the extraction of uranium from low grade resources, such as the very extensive shale deposits in parts of the USA and Sweden, and from the sea, most mining today is confined to ores containing more than half a kilogram or so of uranium per tonne. Australia has a number of these rich ore deposits and at Nabarlek, where the average ore grade is some 17 kg U per tonne, we have one of the richest uranium deposits in the world. Average ore grades in the Alligator Rivers province are 2 to 3 kg U per tonne of ore.

Because of the ubiquity of uranium it is not surprising that it was handled, and even used, long before it was identified. Uranium compounds, especially its oxides, have been used in the glass and ceramics industry for centuries. Some glass produced in southern Italy in the first century A.D. has been found to be coloured by uranium, and it has been used extensively in the famous Bohemian glass and tile industry since at least the 15th century. Uranium salts can produce fine lustrous black ceramic glazes and soft yellowish-green to brilliant orange colours in ceramic glazes and in glass.

Bohemia features significantly in the history of uranium and indeed in the history of modern physics. The story begins in the 15th century with the mining of silver at Joachimsthal, now called Jachymov, a sizable town about 20 kilometres north of Karlsbad in Czechoslovakia. The silver was threaded through a hard black greasy-looking ore, called pitchblende (in 1727) because of its resemblance to pitch. This material was piled around the mines, essentially useless, though it had a certain curiosity value because it glowed in the dark giving out a faint greenish-blue light. Pieces of it were sold around Europe as a novelty, especially in Germany and France and it was such a piece that led Becquerel in 1896 to the discovery of radioactivity. It was from pitchblende from the Joachimsthal area that the German chemist Klaproth in 1789 first isolated the compound of uranium that led to its identification and from which, 100 years later, Marie and Pierre Curie separated the intensely radioactive element radium.

With the discovery of radium and its growing use in medicine, the mining of uranium took on a new dimension. Radium at one time fetched as much as \$200 000 per gram which, naturally enough, triggered off a frantic search for new deposits of its mother element, uranium. Joachimsthal was the sole supplier of radium for a decade or so but a series of discoveries, first in Colorado and Utah, then in the Congo and finally at Great Bear Lake in Canada, saw the price of radium drop in the mid-thirties to about \$20 000 per gram. At this time too, the demand for radium had essentially been satiated and the uranium-cum-radium industry was virtually dead. Uranium in 1940 was worth a dollar or two a pound. The revival of the uranium industry came of course with the discovery of nuclear

energy, first during the war with the manufacture of atomic bombs and later with the development of nuclear power.

### Mountain Sickness

Joachimsthal had not however disappeared from the uranium story. Perhaps the thing that did most to make the name familiar to the uranium mining industry was its association with that of its near German neighbour, Schneeberg, just across the Erz Mountains into old Saxony. It was studies on respiratory diseases occurring in the miners in this region that first led to the recognition of an association between underground uranium mining and lung cancer.

It had been known since the 15th century that a large proportion of miners in the Erz Mountains region contracted and died from a respiratory tract disease called locally 'Schneeberger Bergkrankheit' (Schneeberg mountain sickness). In 1879, the disease was recognised as a form of respiratory cancer but it was not correctly diagnosed as primary lung cancer until 1913. Autopsy studies in the twenties proved the identical nature of the disease in the Schneeberg and Joachimsthal miners and established that, of those who died, some 50% had lung cancer. Many of the rest died of silicosis, tuberculosis or a combination of these diseases which remained common in underground mining until well into the present century.

### The Cause of Mountain Sickness

The cause of the lung cancer was not known at first. Initially it was attributed to the inhalation of various components of the ores, notably arsenic and cobalt. In the early 1920s, radon, a radioactive gas always associated with uranium in its natural state, was suspected as a possible cause and, though the details of the explanation of just how it acts to induce cancer have been modified over the years, there is now little doubt that radon is basically responsible.

Apart from the dangers common to all forms of underground mining, excessive radon exposure is the major hazard in the underground mining of uranium. Now that the relationship between radon exposure and the induction of lung cancer is better understood, excessive exposure can be avoided. Before going on to explain how radon exposure can be controlled, it will be necessary to know something of the nature of radon - its source, what happens to it in a mine atmosphere, how it is thought to induce effects in the lung - and the level of risk associated with a given exposure. This will require some understanding of radioactivity and radioactive decay chains, and of the manner in which the radiations which are emitted by radioactive substances are thought to produce biological effects.

### RADIOACTIVITY

All elements which occur naturally are made up of a mixture of isotopes, i.e. atoms having identical chemical properties but differing in weight. Some of these isotopes are unstable and will break down, or disintegrate, of their own accord by emitting particles and radiation from the nucleus of the atom until a stable arrangement of nuclear particles is attained. In so doing, the atom changes into an isotope of another element. This process is called 'radioactive decay'.

The particles that are emitted from the nucleus are either alpha particles - positively charged nuclei of helium atoms - or beta particles, which are negatively charged electrons. During this decay process, electro-magnetic radiation - gamma rays or X-rays - are also emitted. Both the particles and the gamma rays are often collectively called radiation. These radiations are emitted with sufficient energy to knock electrons from the cloud of electrons which surround the nuclei of all atoms, a process called ionization. These radiations can therefore ionize the atoms of material through which they pass and are therefore called ionizing radiations.

It is the ionization of atoms in constituent molecules which is believed to be the cause of the injury produced in living cells when they are irradiated by ionizing radiation. The injury is thought to result from structural changes in certain critical molecules arising directly from ionizations occurring within the molecules themselves, or induced by reactions with chemical 'poisons' produced in the cellular material by the radiation.

#### The Laws of Radioactive Decay

The rate at which radioactive isotopes, often called radionuclides, decay varies enormously from one isotope to another; some decay in time scales measured in tiny fractions of a second, others must be measured in terms of billions of years. It is an interesting property of nature that no two radionuclides decay at exactly the same rate. There is also almost nothing one can do to change this rate of decay. It is not affected by temperature or pressure, it makes no difference whether the isotope is in a solid, liquid or gaseous state, and it remains essentially unchanged when the isotope is combined chemically with other elements.

It is important to understand clearly what is meant by the rate of decay of a radionuclide. The process of radioactive decay is such that in any assembly of atoms of a radioactive isotope, the number of disintegrations that occur per unit of time is directly proportional to the number of atoms present. That is, if one has, at a given instant of time,  $N$  atoms of a radionuclide, the rate at which the atoms are decaying is proportional to  $N$ . Thus, if the constant of proportionality is denoted by  $\lambda$ , we have

$$\text{Rate of decay of } N \text{ atoms} = \lambda N \text{ disintegrations per unit time,}$$

where  $\lambda$  is called the 'radioactive decay constant' of the radioactive isotope and, from what was said above, it has a different value for every radionuclide; and  $N\lambda$ , the rate of decay, is called the 'activity' of the assembly of  $N$  atoms.

Clearly, as the decay process proceeds, the number of atoms present of the original radionuclide progressively declines and the activity decreases correspondingly. This sort of process, where the rate at which an assembly of a certain size changes is proportional to that size, is common in nature and is called an 'exponential process'. The equation which gives the number of atoms remaining at any time  $t$ , can be deduced by a little mathematics (which need not be given here); it is



$$N = N_0 e^{-\lambda t}$$

where  $N_0$  is the number of atoms initially present at the beginning of the process, when  $t = 0$ , and  $e$  is the number (2.718...) which is the base of the system of natural logarithms.

Another term which is very commonly used to characterise the rate of decay of a radionuclide is its 'half-life'. This is the time required for a given initial number of atoms of a radioactive isotope to be reduced to half of that number; that is for  $N$  to become  $\frac{1}{2}N_0$ . There is obviously a close relationship between the half-life of a radionuclide and its decay constant. This relationship can be deduced from the above equation and is

$$T_{\frac{1}{2}} = \frac{\log_e 2}{\lambda} = \frac{0.693}{\lambda}$$

As suggested above, the half-lives of the various radionuclides range between millionths of seconds and billions of years. Clearly the only radioactive isotopes to be found naturally on Earth are those which have the half-lives greater than, or at least of the same order as, the age of the Earth (otherwise they would have decayed away to nothing); or those which are being continually replenished.

It has been said that the number of disintegrations that occur per unit of time in a given quantity of a radioactive isotope is a measure of its activity. This is determined by counting, with an appropriate detector, the number of alpha, beta or gamma rays that are emitted from a known quantity of the isotope in a known period of time. The unit of activity currently used is the curie (abbreviation Ci), which is  $3.7 \times 10^{10}$  disintegrations per second. This rather peculiar number was adopted since, historically, radioactivity was often determined by comparing the activity in a sample with the activity in a known quantity of radium-226, and  $3.7 \times 10^{10}$  happens to be the number of disintegrations per second in one gram of radium-226. The curie is too large a number for discussing the levels of radioactivity that occur naturally; a millionth ( $10^{-6}$ ) of a curie, the microcurie (abbreviation  $\mu\text{Ci}$ ) or a million-millionth ( $10^{-12}$ ) of a curie, the picocurie (abbreviation  $\text{pCi}$ ) are more appropriate. One  $\text{pCi}$  is 0.037 disintegrations per second or 2.22 disintegrations per minute.

### Radioactive Decay Chains

It was stated that when an isotope decays it changes into the isotope of another element. Sometimes this new atom is stable, but in some cases it is not. If it is not, it too will decay radioactively, with its own characteristic half-life, turning into yet another kind of atom. This process may continue through a chain of isotopes, each parent atom when it 'dies', giving birth to a radioactive 'daughter' atom of a different element, until finally a stable nuclear configuration is reached, the final product being the stable isotope of some naturally occurring element.

### THE URANIUM-238 RADIOACTIVE DECAY CHAIN

All of these processes are to be seen in the complex series of progeny that result from the decay of the original atoms comprising the most abundant isotope of natural uranium, U-238. (Another uranium isotope, U-235, found in very small amounts in natural uranium, does not belong to the U-238 decay chain. Although of fundamental importance to the nuclear energy industry, it has no relevance to the hazard of natural uranium.)

A slightly simplified version of the uranium-238 decay series is illustrated in Figure 1; Table 1 lists some of the properties of the various radionuclides that go to make up this decay chain. Some of the minor alternative decay pathways that are possible have been omitted from this table but what remains is complicated enough, as the original uranium-238 nucleus transmutes through a series of 14 decay products, finally ending up as a stable isotope of lead, Pb-206, sometimes called radiogenic lead. The whole basis of the radiological hazards associated with the mining and milling of uranium is contained within this Table and Figure 1.

One can now see why radium (Ra-226) is always found with uranium. Though it has a half-life of only 1600 years, it is continually produced, through the series of the four preceding decay products, by the very long-lived prime parent U-238 (half-life  $4.5 \times 10^9$  years). Similarly, radon (Rn-222) is present in all uranium ores, being the immediate daughter of Ra-226 itself. Though its half-life is only 3.8 days, its supply is continually replenished by the decay of Ra-226.

The decay of radon is quickly followed by the decay of its four short-lived progeny, coming to a temporary slowing down with the 22 year half-life isotope of lead, Pb-210, which in the early days, when this complex series of transformations was being disentangled, was called radium D. Though Pb-210 has a longish half-life, its daughter radium E (bismuth-210) is short-lived (5 day half-life) and decays to radium F, the 138 day half-life isotope of polonium, Po-210. Thus, wherever radon has been, one is likely to find traces of Pb-210 and Po-210. This can sometimes be used to monitor the amount of radon, or of its short-lived daughters that have been taken into the body. For example, Pb-210 levels in bone have been found to be correlated with cumulative exposures to radon daughter products.

#### Radioactive Equilibrium Amongst Uranium-238 Daughters

Table 1 indicates what a complex mixture of isotopes is to be found in a sample of uranium ore. Provided the ore is old enough for all the daughter products to have had time to build up, and provided none of them has been leached out or otherwise removed from the ore, each of the 14 decay products will be present, and in fixed proportions which are determined by the half-lives of each individual isotope. The laws of radioactive decay are such that when equilibrium has been reached - the sort of equilibrium that is attained is called 'secular equilibrium' - the number of disintegrations occurring in a given time is the same for each daughter product. The number of atoms of each of the 13 radioactive decay products will build up and be kept at just the right number to ensure that each decay product is producing exactly the same number of disintegrations per second as is occurring amongst the atoms of the U-238. Another way of saying this is that the activities of each of the decay products are

equal; that is, the product of the number of atoms present of each radionuclide and its decay constant is the same for all of the decay products. In symbols, this means:

$$\lambda_0 N_0 = \lambda_1 N_1 = \lambda_2 N_2 = \dots \dots \lambda_{13} N_{13}$$

where  $N_0$  is the number of U-238 atoms present,  $N_1$  is the number of atoms of the first daughter, thorium-234,  $N_2$  is the number of atoms of the second decay product, Pa-234 (an isotope of protactinium), and so on down to  $N_{13}$  which is the number of polonium-210 atoms; and  $\lambda_0, \lambda_1, \lambda_2 \dots$  are the decay constants of U-238, Th-234, Pa-234 etc., and  $\lambda_{13}$  is the decay constant of Po-210. This is an important relationship. From it one can calculate for example, in a known mass of uranium ore, the associated amount of Th-230 or of Ra-226; the rate of evolution of Rn-222, and the activity of the radon daughters associated with a given quantity of Rn-222. The mass of the associated daughter products is extremely small. Thus in 1000 tonnes of ore containing 1 tonne of U (an average sort of commercial ore) there is only 0.3 Ci of Ra-226 which, in the case of Ra-226 (and only for this isotope), is also 0.3 g. One can begin to see the magnitude of the achievement of Marie and Pierre Curie in extracting measurable quantities of Ra-226 from uranium ore.

One final point needs to be made about the radiations that are emitted from a body of uranium ore. If all daughter products are present then all the radiations associated with each daughter product as listed in Table 1 will be emitted, though most of them will be absorbed within the ore. If a daughter product is removed, its associated radiations will of course disappear, and so too will the radiations associated with those subsequent daughter products which are dependent upon it to maintain them at equilibrium levels. The radiation will not reappear at full strength until there has been time to re-establish equilibrium. This is readily observable in the gamma radiation emitted from uranium at various stages in the mining and milling process. In the ground, all daughter products may be present and all the gamma rays listed in Table 1 emitted. If, however, the radon were removed, then after a brief period during which the already established short-lived radon daughters decayed, the gamma rays from radium B and radium C would disappear. Since these are responsible for the major part of the gamma emission from uranium ore, the removal of radon would substantially reduce the gamma dose rate from the orebody.

Again, if all isotopes other than uranium are removed from the ore, as happens during the milling process, the extracted uranium is initially practically free of gamma radiation. This will build up slightly over the ensuing months as Th-234 re-establishes itself (there will be a small contribution from Pa-234, the daughter of Th-234) but the long half-life of U-238 prevents the build-up of subsequent daughters. There will of course be traces of Ra-226, not removed in the extraction process, which will lead to the production of some radon and its gamma emitting daughters in the uranium concentrates.

#### HAZARDS OF URANIUM MINING

With this background, we can now turn to an examination of the radiological hazards that are associated with the mining and milling of

uranium. These hazards are determined by what happens to the daughter products during each phase of these operations. In the case of mining, attention is focused on radon.

### Radon

Radon is a gas. Though it is radioactive, it is chemically inert and belongs to the same family as helium, neon, argon, krypton and xenon - the so-called 'noble gases'. It is the heaviest of the noble gases and is 7.7 times denser than air.

Although radon itself is relatively short-lived, its parent radium has a long half-life and keeps the supply of radon in uranium ore continually replenished. If the radon gas cannot escape from the orebody, it and its daughter products remain in the ore and, in decaying, give out the complex mixture of alpha, beta and gamma radiations listed in Table 1. Since no orebody is completely impervious, some radon will diffuse out and, if the orebody is not too far from the surface, escape into the atmosphere. Uranium prospecting is sometimes carried out by measuring radon levels in the air spaces in the soil.

The actual volume of radon in an orebody is extremely small. One curie of radon occupies only  $6.6 \times 10^{-4}$  cubic centimetres at normal conditions of pressure and temperature. Thus in the 1000 tonnes of ore considered above, with 0.3 Ci of Ra-226, and therefore also 0.3 Ci of Rn-222, there is only about  $2 \times 10^{-4}$  cubic centimetres of radon. This is the volume of a sphere having a diameter somewhat smaller than that of the full stop at the end of this sentence.

### *Natural levels of radon in air*

Radon and its daughter products are measurable in the atmosphere at all times and all places, originating from trace amounts of uranium present in the Earth's crust. Its concentration varies from day to day, depending on meteorological conditions which affect not only the amount of mixing and dilution that occurs in the atmosphere, but also the rate of emanation of the gas from the Earth's surface. Radon levels will build up near the surface under still, inversion conditions when mixing is minimal.

Radon levels over the ocean are much less than over land because of the lower Ra-226 content of the ocean. For this reason, radon levels in the atmosphere at coastal sites are very dependent on whether the wind is blowing from the land or the sea. Concentrations of radon in ground level air out-of-doors vary between about 0.04 and 0.4 picocuries per litre; some mean annual values from different parts of the world are shown in Table 2.

Radon is never strictly in equilibrium with its short-lived decay products in surface air since they tend to attach themselves to aerosols which are removed by one means or another from the atmosphere, or to deposit directly onto vegetation and the Earth's surface. Near equilibrium conditions can obtain during stable atmospheric conditions or inside poorly ventilated buildings. Radon and its daughter products contribute the major part of the naturally occurring background radioactivity in the air of the lower atmosphere.

Radon levels inside buildings can often be much higher than in the outside air, particularly if it can accumulate in poorly ventilated spaces. Indoor concentrations depend upon the concentration in the outside air, the rate at which radon emanates from the walls and floor (which in turn is dependent upon the porosity and Ra-226 content of the building materials) and upon the ventilation rate in the room. Some measured Rn-222 concentrations inside dwellings are given in Table 3. Indoor levels can be a factor of ten or more higher than outside, and concentrations of up to 10 picocuries per litre have been reported in slag and adobe huts in the USSR.

#### *Radon levels in underground mines*

Not surprisingly, radon concentrations can reach very much higher levels than these in poorly ventilated underground uranium mines. No one knows what the air concentrations of radon were in the mines at Schneeberg and Jachymov. The mines were extensive and unventilated and some estimates have put the average levels to which the miners were exposed before the hazard was recognised, as high as 15 000 picocuries per litre. Measurements in the late 1930s, when some effort had been made to reduce concentrations by forced ventilation, ranged between 50 and 5000 picocuries per litre.

Median radon levels measured in the 1940s in some US mines which had only natural draught ventilation were 2500 picocuries per litre in Colorado and 5000 picocuries per litre in Utah.

It should be pointed out that high radon levels can arise in non-uranium underground mines. Average concentrations in excess of 1000 picocuries per litre have been measured in ventilated areas in a Newfoundland mine producing fluor spar, where uranium was present in only trace amounts in the ore. The radon entered the mine dissolved in ground water.

#### Relation between Radon Exposure and Lung Cancer

Miners working for lengthy periods in each of the mines mentioned above have shown a significantly increased incidence of lung cancer. If radon and its daughter products are in some way responsible for this, and this is now generally accepted, clearly, lengthy exposure to air concentrations of the order of 1000 picocuries per litre is unacceptable. The question is, what is the maximum integrated exposure (air concentration times length of exposure) that will not lead to an unacceptable increase in the risk of a miner getting lung cancer?

Since the primary standards of radiation protection are expressed as allowable doses of radiation, the unit of which is the rem (see glossary), the conventional approach to the problem is to calculate the radiation dose to the organ at risk, in this case the lung. The recommended maximum permissible dose rate to the lungs of radiation workers is 15 rem per year. One would like therefore to be able to calculate the limiting concentration of radon that would lead to a lung dose of 15 rem per year in exposed workers. Such calculations have been tried but, in the light of present knowledge, cannot give a satisfactory answer. Therefore, observations on the incidence of lung cancer in miners, as a function of exposure, have been used to determine acceptable maximum limits of exposure. Although calculation of dose is impracticable, the attempts have been instructive, and therefore both the theoretical and the empirical approaches are described.

*Principles of dose calculations*

Attempts to calculate the dose to lung tissue due to the inhalation of radon and its daughters have been unsuccessful because of the great complexity of the physical and biological processes involved. They have however contributed much to our understanding of these processes, provided guidance to areas requiring additional research and enabled the relative importance of many of the different physical and biological parameters to be assessed.

Perhaps the most important outcome of these theoretical assessments and associated experimental work has been the realization that it is the inhalation of the short-lived daughter products of radon, that is the radium A, B, C and C', rather than the gas itself that contributes most of the lung dose. These daughter products will tend to attach themselves to available surfaces such as dust particles in the atmosphere or become nuclei for the condensation of water vapour. Some will remain free or unattached radioactive atoms.

When air containing these daughter products is inhaled, the free atoms are deposited most efficiently in the upper respiratory tract, and the attached atoms, which are for the most part associated with dust particles of breathable size, tend to be deposited deeper in the lung. Because the half-life of these isotopes is short compared with processes that remove inhaled particulates from the lung, practically all of the alpha and much of the beta radiation emitted during the decay process is absorbed by lung tissue. This absorbed radiation energy may kill some cells, though not in significant numbers and, more importantly, may induce latent damage in cells which will eventually cause them to become malignant. The most heavily irradiated region is the cells lining the bronchial tubes which is the area where lung cancer in uranium miners originates.

The radon itself, being an inert gas, and having a long half-life relative to the time a breath of air remains in the lung, tends to be exhaled without depositing a significant amount of radiation energy in the lung. Some radon will dissolve in body fluids and get into the blood stream where it, and those daughter products formed by its decay inside the body, can lead to the irradiation of various organs, but these doses are insignificant relative to the dose to the lung from the inhaled daughter products.

A successful radiation dose model must therefore take account of the size distribution and chemical and physical properties of the aerosol particles and the unattached atoms; the state of radioactive equilibrium of the attached daughters; the deposition pattern of these particles in the lung, taking account of the structure of the lung and such parameters as breathing rate, depth of breathing, nose versus mouth breathing etc.; the physiological mechanisms which remove particulates from the lung; the tissue structure of the lining of the bronchi including any matter, such as the mucous layer, that may absorb radiation before it reaches the sensitive cells. And having calculated the distribution of deposited radiation energy in the relevant tissue, one is still left with the problem of relating this to the chance that the irradiated tissue will become malignant. Faced with these difficulties, it is not surprising that no completely satisfactory dose model has been produced.

*The concept of the 'Working Level' as a measure of radon concentration*

With the short-lived daughter products playing such an important role it is the level of these in the atmosphere rather than the concentration of radon that should be measured and controlled to provide protection to miners. A measure of radon activity in the air (say in picocuries per litre) is not an index of the hazard of exposure unless the state of equilibrium with its daughter products is also known. A concept has recently been devised to give a measure of what one really wants to know, the total radiation energy associated with the short-lived daughter products in a given quantity of air that is potentially available to irradiate the critical lung cells if the daughters are inhaled. The most important contributors to the dose to the cells lining the bronchial tubes are the two alpha particle emitting daughters radium A and radium C'. The dose contribution from the beta particles and gamma rays emitted by radium B and radium C is negligible, whilst the long-lived daughters, radium D, radium E and radium F are eliminated from the bronchi before any significant number of disintegrations of these isotopes has occurred.

Let us consider 100 picocuries of radon (that is a quantity giving 222 disintegrations per minute) in equilibrium with its short-lived daughters; that is, we have in association with the radon, 100 picocuries each of RaA, RaB, RaC and RaC'. We wish to know the total amount of alpha energy emitted during decay of the atoms which go to make up 100 picocuries each of RaA, RaB, RaC and RaC'. First we must calculate the number of atoms of each daughter product associated with the activity of 222 disintegrations per minute. This is obtained from the relationship established on page 3, namely that the activity of N atoms is given by  $N\lambda$ . Thus for RaA, whose half-life is 3.05 minutes and whose decay constant,  $\lambda$ , is

therefore  $\frac{0.693}{3.05} = 0.227$  per minute, we have

$$N \times 0.227 = 222 \text{ disintegrations per minute}$$

whence,

$$N = 977 \text{ atoms.}$$

Similarly, using the respective half-lives of the other daughter products, we find that there are 8500 atoms of RaB in 100 picocuries of RaB, 6310 atoms of RaC in 100 picocuries of RaC and 0.0009 atoms of RaC' in 100 picocuries of RaC'.

Each atom of RaA when it decays to RaB gives out 6.00 MeV (For definition of MeV see the footnote to Table 1.) of alpha energy (see Table 1) and this particular RaB atom will eventually become a RaC' atom giving out a 7.69 MeV alpha particle. Thus each of the 977 atoms of RaA is eventually associated with the emission of a total of (6.00 + 7.69) MeV of alpha energy. Similarly, each of the 8580 atoms of RaB will eventually become a RaC' atom and emit a 7.69 MeV alpha particle, and so too will each initial RaC atom, and of course each initial RaC' atom, though there are so few of the latter present that their contribution is literally zero. The total amount of energy eventually emitted from all of the daughter atoms can thus be calculated and summed. This calculation is set out in Table 4

where it can be seen that the short-lived decay products in equilibrium with 100 picocuries of radon eventually release a total of  $1.3 \times 10^5$  MeV of alpha energy. It is interesting to note the seeming paradox that most of the energy - 90% of it - comes from the two non-alpha particle emitting daughters RaB and RaC. This is because of their relatively long half-lives and therefore the correspondingly larger number of atoms of each that must be present to give their 100 picocuries worth of activity.

The assumption is now made; any combination of RaA, RaB, RaC and RaC', regardless of its state of radioactive equilibrium, which will result in an ultimate emission of  $1.3 \times 10^5$  MeV of alpha energy has the same potential hazard as the equilibrium combination, if distributed in the same volume of air. If the volume of air is taken as one litre, a new unit, the 'Working Level' (WL) emerges.

Thus one WL is defined as 'any combination of the short-lived decay products of radon (RaA, RaB, RaC and RaC') in one litre of air which will result in the ultimate emission by them of  $1.3 \times 10^5$  MeV of alpha particle energy'.

Note that 1 WL is equivalent to 100 picocuries per litre of radon only in the one special case when the radon is in equilibrium with its short-lived decay products. To express a radon concentration, measured in picocuries per litre, in working levels, it is necessary to know the state of disequilibrium between the radon and its short-lived daughters under the conditions obtaining at the time.

Thus we can write that, in general,

$$\text{Concentration in WL} = F \times \left( \frac{\text{Concentration in picocuries per litre}}{100} \right),$$

where F is a factor, variously called the 'disequilibrium factor' or 'working level ratio', which is a measure of the lack of equilibrium between the radon and its short-lived daughters. F is very variable in practice and rarely has a value of one. Because of the half-lives of RaB and RaC it would take about three hours for initially fresh radon to come into equilibrium with its short-lived daughters. Any process which removes the daughter products, particularly movement and dilution of the air, will ensure that F remains less than one. In a space where the ventilation rate was as low as one air change per hour, F would be kept to about 0.7 and would drop to about 0.4 at two air changes per hour.

A WL is a measure of a concentration. To obtain an accumulated or integrated exposure, one must multiply by the time of exposure. Integrated exposures are usually measured in 'Working Level Months' (WLM). Breathing air with a concentration of 1 WL of radon daughters for the working hours in a month (taken as 170 hours) results in an exposure of 1 WLM.

Finally, a unit for rate of exposure is required. A person who accumulated 1 WLM of exposure each month would be exposed at the rate of 12 WLM per year. The unit WLM per year is commonly used to measure exposure rate. An exposure rate of 4 WLM per year is important in what follows. This exposure rate will result from working continuously for 170 hours per month in an atmosphere with a daughter product concentration of 0.33 WL.



*Observations of lung cancer incidence in underground uranium miners*

An enhanced incidence of lung cancer has been observed in underground miners from a number of areas and attempts have been made to relate excess cancers to radon exposure. The most thorough study of this kind is one carried out by the US National Institute for Occupational Safety and Health (NIOSH) which dealt with the incidence of lung cancer in underground uranium miners from the Colorado Plateau. The sample consists of 3366 white and 780 non-white (99% Navajo Indians) underground uranium miners who had one or more months of underground uranium employment before 1st January 1964, and considers deaths from all causes in the period July 1950 through to September 1968.

A simplified summary of the results for white miners is given in Table 5. The total number of deaths in the white miner group was 437 compared with an expected number of 277 in an age-cigarette smoking matched control group. (Most of these miners were smokers; the interaction between smoking and radon exposure is discussed later.) The excess deaths were largely due to cancer of the respiratory tract and accidents. A total of 70 deaths from lung cancer were observed against an expected 12, and 126 violent deaths were recorded compared with 54 expected.

Estimates were made of the cumulative exposure of each miner in terms of WLM. These were based on nearly 43 000 measurements of radon daughter levels, in some 2500 mines, made between 1951 and 1968, and on the work record of each miner. Miners were divided into six cumulative exposure categories - < 120, 120-359, 360-839, 840-1799, 1800-3719 and 3720 WLM and over - and the excess lung cancer incidence in each category determined. The results are shown in Table 6.

No excess cancer was observed among miners exposed to less than 120 WLM. In the three groups to 1799 WLM, incidence of cancer was some five times the expected rate, although the risk appears to be constant over this range of exposure. In the 1800-3719 WLM category about a 15-fold excess was found, rising to a 25-fold increase for exposures exceeding 3720 WLM. Figure 2 shows the relationship between observed incidence of lung cancer expressed in terms of cases per person per year (the person years at risk are given in Table 6 for each exposure category) and cumulative exposure.

The relatively flat portion of exposure-response curve between 120 and 1800 WLM may be spurious and will probably disappear as more cases are added to the series. The group is still under study and additional respiratory cancer cases are continuing to appear. Between September 1968 and December 1971, a further 37 deaths from lung cancer were recorded, plus an additional eight cases that were still alive in February 1973. As well, an additional 22 cases of probable lung cancer - nine deaths and 13 still alive - exist among the 3366 sample. It can be seen from Table 6, where the total cases to 1971 are also tabulated, that the newly reported cases have occurred in the higher exposure categories. The statement that no excess cancers are observed among miners in the less than 120 WLM category still holds.

These more recent data were included in a review of the NIOSH study carried out in 1972 by an Advisory Committee on the Biological Effects of Ionizing Radiations (the BEIR Committee) set up by the US National Academy of Sciences and the National Research Council. The results of this review

are shown in Figure 3 where the excess number of lung cancer cases above expectation (expressed as cases per year per million persons exposed) is plotted against cumulative exposure to radon daughters. It can be seen that already the additional cases that have arisen since 1968 have tended to remove the relatively flat part of the curve previously observed and that a fairly clear linear relationship between exposure and lung cancer incidence is emerging, at least at the higher exposure levels.

This enables an estimate to be given of the risk associated with an exposure to radon daughters in this group of miners, most of whom were smokers. The slope of the straight line indicates that the risk is about 3.2 cases per year per million smoking miners exposed to 1 WLM. If one assumes that this risk continues on average for 30 years, the total number of excess cases expected in a population of  $10^6$  smoking miners after exposure to 1 WLM, is about 100. Thus if the exposure is limited to  $\frac{1}{3}$  WLM per year, the risk amongst miners who smoke is about 400 cases per million per year. This is an individual risk of getting lung cancer from the radon exposure of four in ten thousand per year which is the estimated risk associated with smoking an additional one to two cigarettes per day.

There appears to be no published analysis of the incidence of lung cancer in the USA uranium miners as a whole. A recent publication [C.W. Mays Health Physics 25:585, 1973] states that there has been a total of 235 known cases of respiratory cancer among American uranium miners. The total number of miners at risk is not given; it may be as high as 20 000.

Only the results of the NIOSH study relating to white miners have been discussed so far. Of the 780 non-white miners examined, total expected deaths (90) actually exceeded the observed deaths (72) owing to a significant deficit in heart disease among the miners. The sample is regarded as too small to enable conclusions to be drawn though it is believed that more recent data, which include additional lung cancer cases, indicate an incidence-exposure relationship similar to that for white miners after allowance is made for differences in smoking habits. These more recent data were also available to the BEIR committee and are plotted in Figure 3. Most of the Navajos were non-smokers, or smoked at most only the odd cigarette per day: the relative number of excess lung cancer cases is much less than among the more heavily smoking white miners.

#### *The effect of smoking*

It should be noted that although every attempt was made in the analysis to isolate the correlation between lung cancer incidence and radon daughter exposure alone, this has not been done with certainty. Cigarette smoking, diesel smoke and exposure to miscellaneous dusts may have contributed to the development of lung cancer, though as more studies are made of the incidence of disease in mine atmospheres where radon and its daughters are not present, it seems that diesel fumes and miscellaneous dusts are unlikely to be of more than minor significance.

The relationship between cigarette smoking and lung cancer induction by exposure to radon daughters is still not clear, but smoking alone cannot account for more than a fraction of the total number of cancers in uranium miners. Respiratory cancer rates among smoking and non-smoking uranium miners are six to nine times greater than among non-miners with similar smoking habits, and the highest lung cancer rates among miners are about ten times greater than the highest rates among very heavy smokers who are

also city dwellers.

In the NIOSH study, 71% of the white miners were smokers. However, all the miners who contracted lung cancer, but one, had been smokers, giving a 99% representation of smokers in the lung cancer group. This over-representation of smokers in the lung cancer group indicates some interaction between the effects of smoking and of radon daughter exposure. It is thought that smoking may act as a 'promoter'; that is, it accelerates the induction of the lung cancer after the first exposure to the carcinogen proper. This is supported by the observation that the latent period for the induction of cancer appears to be six or seven years less for smokers than for non-smokers. The average induction period amongst the Erz Mountains miners who did not smoke cigarettes was 20 to 21 years compared with an average of 14 years among US miners.

In summary, the results of the NIOSH study to date indicate that if the lifetime cumulative exposure to radon daughter products of miners, even those who smoke cigarettes, is kept below about 150 to 120 WLM, it is unlikely that there will be a detectable increase in the incidence of lung cancer. It is also becoming clear that the risk of inducing lung cancer by exposure to radon daughters is much reduced at all levels of exposure if miners do not smoke. The NIOSH study, which is being continued, is also examining the possibility that radiation exposure may have increased the incidence of respiratory diseases other than lung cancer in the miners. Some of the evidence suggests that this is possible; there may, for example, be an increased incidence of non-specific respiratory disease, but even the incomplete data are sufficient to show that there could be no such effect at accumulated exposures of less than 300 or 400 WLM. Since this level would not be reached if radon exposure were limited sufficiently to prevent an increased incidence of lung cancer, the possibility can be disregarded.

#### *Recommended radon exposure limits for uranium miners*

There is at present no internationally accepted limit for radon or radon daughter exposures for miners. The International Commission on Radiological Protection (ICRP) proposed in 1959 a value of 30 picocuries per litre for radon with its short-lived daughters assumed present to the extent they occur in unfiltered air.

Values of 100 picocuries per litre and 300 picocuries per litre for radon have been used from time to time, often with confusion as to the assumed state of equilibrium of the daughter products. Until July 1971, the USA had a maximum permissible exposure rate of 12 WLM per year. An Australian Mining Code dealing with uranium mining and milling, agreed to by an inter-State Conference in 1956, proposed a maximum permissible concentration of '100 picocuries per litre of radon or its daughter products RaA or RaC'.

Some existing exposure limits applicable to uranium mines are tabulated on page 15. In comparing the numbers in this listing it will help to remember that 100 picocuries per litre of radon in equilibrium with its short-lived daughters is 1 WL, and that continuous occupational exposure to this concentration will lead to an exposure rate of 12 WLM per year.

|              |  |
|--------------|--|
| ICRP         | 30 pCi/l for radon with its short-lived daughters 'assumed present to the extent they occur in unfiltered air'. (If equilibrium could be assumed this would be equivalent to 3.6 WLM/y).   |
| Belatom      | 300 pCi/l for radon in equilibrium with its short-lived daughters. (This is equivalent to 36 WLM/y).   |
| France       | 600 pCi/l for radon with an assumed disequilibrium ratio, $F = 0.5$ , making this equivalent to 300 pCi/l for radon disequilibrium with its short-lived daughters (Measurements in mines have shown $F$ to be nearer 0.2 than 0.5; this means the effective limit is 120 pCi/l for radon in equilibrium, or about 15 WLM/y). |
| Canada       |  |
| Ontario      | 8 WLM/y, reduced to 6 WLM/y in 1974.   |
| Newfoundland | 4 WLM/y.   |
| Saskatchewan | ICRP, 30 pCi/l as above.   |
| National     | Epidemiological study of 7000-8000 miners being undertaken to consider whether a reduction to 4 WLM/y is indicated.  |
| USA          | 12 WLM/y reduced to 4 WLM/y in 1971.   |
| South Africa | No legislative controls. Aim is to keep exposures below 12 WLM/y.  |

*The new Australian 'Code of Practice on Radiation Protection in the Mining and Milling of Radioactive Ores' recommends that exposure rates be kept below 4 WLM per year. This standard is therefore as restrictive as any operating anywhere in the world.*

#### Control of Radon Levels in Underground Mines

During the mining of uranium the ore is broken up, allowing radon to escape. It will also be diffusing continuously out of exposed ore faces at a rate depending on the type of rock, its porosity and permeability, the size of the particles in which the uranium is incorporated and the degree of weathering of the rock. Oddly enough, the rate of emanation does not seem to be related to the uranium content of the ore. Radon is also quite soluble in water so that any ground water that has been in contact with uranium ore can contain sizable amounts of radon. This is readily released when the water is exposed, especially if it is aerated by flowing, splashing etc. Radon daughter products grow from radon quite quickly so that control of daughter levels becomes essentially the control of radon levels.

Clearly the way to manage radon levels in a given area is to remove it as quickly as possible, i.e. to ventilate and to limit, as far as practicable,

the amount which enters. Influx of radon can be reduced by keeping exposed ore faces to a minimum, sealing off worked out stopes and maintaining them at a negative pressure, keeping all pit water enclosed and removing broken ore as soon as possible. Keeping the mine atmosphere under positive pressure has been tried without much success.

The ventilation of a uranium mine requires careful planning. There must be no recirculation (radon daughters will be building up as the radon is flushed along in the air stream), stagnant areas must be eliminated, contaminated air must be kept away from miners, and seals and bulkheads must be kept under continual maintenance to prevent leakage of contaminated air into working areas.

The levels of radon daughters can also be controlled to some extent by extracting them from the mine atmosphere either by filters or electrostatic precipitators. Similarly, respirators containing filters which will remove daughter products can be worn or, in extreme circumstances, self-contained breathing apparatus can be supplied. However, it is a maxim of industrial hygiene that all operations are to be planned to avoid the habitual use of respiratory protective devices.

Essential to the control of mine atmosphere quality is of course the measurement of radon daughter concentration. Many methods are available for doing this but all depend basically upon drawing a known volume of air through a filter to extract the daughter products, whose activity is then determined with some form of radiation counter. Sufficiently accurate survey measurements at concentrations below 0.3 WL can be made in the field in ten minutes or so.

#### Control of Radon Exposure in Open Cut Mining

Radon daughter exposure is less likely to be a problem in open cut mines. Natural ventilation will keep air concentrations well below acceptable working levels except perhaps for those ores giving very high radon emanation rates and under very still atmospheric conditions. These conditions occur mostly at night and mixing will begin as soon as the sun comes up, diluting radon which may have accumulated in the bottom of the open cut.

Some recent measurements made by the AAEC in the Alligator Rivers area have shown that quite high levels of radon daughters can build up under still conditions at the bottom of deep narrow trenches. The highest value recorded was 23 WL in a trench 5 m deep and 1 m wide under still conditions in the early evening. The levels in this trench, which was cut through ore of above average grade, were very variable and were reduced by a factor of 50 or more when atmospheric mixing was established. Conditions in such a narrow trench are not representative of conditions that will obtain at the bottom of a large open cut.

Even if relatively high levels of radon (i.e. high relative to a third of a working level or so) were to build up overnight there are a number of options available to the mine manager to ensure that miners are not exposed to unacceptably high concentrations. For example:

- (a) Mining operations can be delayed until some time after sunrise when radon levels will have been diluted.

- (b) The cabins of the mining equipment can be sealed and supplied with bottled air and maintained at a slight positive pressure to ensure no ingress of the pit atmosphere.
- (c) The cabins may be supplied with air from which the radon daughter products have been removed by filtration.

#### Other Radiological Health Hazards in Uranium Mining

There are two remaining potential radiation hazards in uranium mining that must be assessed and controlled to ensure safe working conditions for miners - exposure to external radiation and the inhalation of dust containing uranium and its long-lived daughter products. Control of these hazards is also dealt with in the Australian Code of Practice on Radiation Protection in the Mining and Milling of Radioactive Ores.

##### *External radiation*

As discussed above, the gamma rays emitted from a uranium orebody will vary depending on the state of equilibrium of some of the decay products of radon, a long way down the chain from the parent uranium-238. In general, the control of gamma exposure is not a problem in uranium mining, maximum dose rates in working areas of mines being of the order of a millirem per hour (mrem/h) and less. Uranium miners are classified as radiation workers and are therefore subject to a yearly dose limit of 5 rem. With a 40 hour working week, this is equivalent to an average limiting dose rate of 2.5 mrem/h. A recent survey of nine mines in the USA reported dose rates ranging from 0.1 to a maximum of 2.6 mrem/h with mean dose rates lying between 0.2 and 0.7 mrem/h.

External dose rates may however be quite high close to deposits of massive pitchblende. A value of 20 mrem/h was recorded near such an orebody at the El Sherana mine in the South Alligator River area. Even with such rich ore, the average dose rate in work areas in high dose rate regions was only 2.5 mrem/h. In such relatively high dose rate areas, weekly or monthly accumulated doses must be kept within prescribed limits by limiting the time a miner spends in these areas. All miners who work in places where they could receive more than three tenths of the maximum permissible dose must be carefully monitored and accurate records must be kept of their external exposure.

##### *Inhalation of long-lived radioactive elements*

A uranium mine atmosphere will contain dust particles incorporating the early daughters of uranium-238, the only ones of any radiological significance being the long-lived alpha emitters, uranium-238 itself, uranium-234, thorium-230 and radium-226. Measured levels of these isotopes are usually found to be well below the maximum permissible concentrations recommended by the International Commission on Radiological Protection. In fact, it is often the case that other constituents of the ore are more toxic than these radioactive components. This is particularly so for free silica, and reducing dust loadings to maintain safe conditions for silica exposure will in general ensure that the levels of the long-lived alpha emitters are well within prescribed limits.

### HAZARDS OF URANIUM MILLING

The milling of uranium is a relatively straightforward chemical operation which gives rise to only minor radiological hazards for mill workers. In this process the ore is crushed and ground and the uranium dissolved, either by an alkaline leach or with sulphuric acid. Only uranium is dissolved from the ore, leaving almost all of its decay products in the residue of sand-like material (tailings) which is despatched to a waste retention system or tailings dam. Radon will be released during the grinding and dissolution stage so that its short-lived daughter products will disappear; the first two decay products of uranium, thorium-230 and protactinium-234, will decay relatively rapidly (with a half-life of 24 days) after separation. What passes out in the tailings therefore will be the thorium-230, radium-226 and possibly some of the lead-210, together with its two decay products, bismuth-210 and polonium-210. These last three will fairly quickly be re-established because nearly all of the original radium-226 will now be in the tailings and radon-222 will grow in with its half-life of 3.8 days.

The dissolved uranium - it is now essentially the pair uranium-238 and uranium-235 (though the amount of uranium-234 is minuscule) and the uranium-235 - is purified and concentrated in a solvent extraction unit, precipitated with ammonia, separated, calcined to yellowcake ( $U_3O_8$ ), pulverized and packed into drums. The net result, radiologically, is that the original uranium decay chain has been separated into two fractions with the most radioactive part now in the tailings. The end product yellowcake, is only mildly radioactive.

The extraction process is not of course perfect and traces of radium-226 will remain in the mill product. The radium-226 content of yellowcake produced by the acid leach process varies from about 0.001% to 0.2% of the concentration of the radium in the natural uranium; that is, radium in yellowcake may range from 3 to 600 pCi per gram.

#### External Radiation

External gamma radiation levels in a mill treating average grade ores are generally low and less than 1 mrem/h one foot from critical items of the plant, though higher levels would be expected when high grade ores are handled. Levels in general areas are less than 0.1 mrem/h. Beta radiation levels can be 1 to 2 mrem/h close to certain plant items.

The highest radiation levels are to be found in the area where the drums of yellowcake are stored, the dose rate near a pile of drums being 2 to 3 mrem/h. Again, cumulative external doses may be controlled by limiting access to, and occupancy of, the higher dose rate areas.

#### Radon and Radioactive Dusts

Radon and dust levels within the plant are controlled by ventilating the operations that are a source of these materials and scrubbing and filtering the effluents before release to the atmosphere through a stack.

The efficiency of removal of the dusts and radon daughters, and the height of the stack must be such that those materials that are released (and radon gas will be amongst them) are sufficiently diluted not to raise ground level concentrations within the works and off site above specified

limits. Off site dose limits where people may be living will be those appropriate to members of the public and are set a factor of ten lower than those applicable to workers. Assessment of the degree of dispersion that will obtain requires a knowledge of the meteorology of the area and exposure of members of the public should be kept as far below the dose limits as can reasonably be achieved.

Complete enclosure of those sections where the packing and sampling of yellowcake is carried out is most important and automatic operation is to be preferred. If this is done and all operations adequately ventilated, good housekeeping on a continuous basis - vacuuming, washing down, mopping and polishing - is all that is then required to ensure safe working conditions.

These conditions would now be required in any new plant, but they did not always obtain in the past. It used to be a common practice to pay little heed to dust production in the packing area, protection of operators against the inhalation of yellowcake being provided mainly by respirators. Despite this, mill workers do not appear to have suffered any deleterious effects from chronic exposure to yellowcake. Certainly no excess respiratory cancer has been reported among US uranium mill operators.

There is a suggestion in one recent analysis of mortality amongst a group of 662 mill operators who were working in US mills in 1950, of an excess of cancer and leukaemia arising in lymphatic and hematopoietic tissue. Five such cases were seen against an expectation of two though the overall mortality of the mill workers was the same as expected. It is not known if the difference is significant, since the numbers are small, and NIOSH has undertaken an enlarged study of mill workers to determine whether the difference is a real one. If it is shown to be significant, the excess might be attributable to the long-lived isotopes of either uranium or thorium which are present in mill and mine dusts. In any case, the possibility emphasizes the need for proper ventilation and dust control, especially when ore is being crushed and handled. The report refers only to mill operations in the early 1950s, a period when occupational hygiene was less strictly regulated than now.

#### The Tailings Retention System

Almost all of the radium-226 and thorium-230 originally in the uranium ore is discharged in the tailings. Since the mass of the tailings is virtually the same as the mass of the original ore, the concentration of these two isotopes in the tailings will also be about the same as that in the original ore. The mass of tailings left after extracting 100 000 tonnes of uranium from an average grade orebody (0.1% uranium) will be about a hundred million tonnes and will contain 30 000 curies of radium-226 and 30 000 curies of thorium-230. It should be emphasized that this is the same situation that existed before the ore was mined except that now the whole orebody (less the originally contained uranium), is piled on the surface in a ground-up form and the radium-226 and thorium-230 are more readily available for leaching.

The radioactivity in the tailings will remain indefinitely on any human time scale. The long half-life thorium-230 (80 000 years) will ensure that the supply of radium-226 is maintained, and as long as radium is there, radon will be continually produced. This gives rise to two potential sources of irradiation of members of the public - escaping radon, which will



be carried along with the wind for considerable distances, though being progressively diluted all the way; and gamma radiation from radon daughter products remaining in the tailings. The latter will lead to an external gamma radiation field in the vicinity of the tailings with dose rates somewhat above normal background levels.

There are two other ways in which the tailings could lead to the irradiation of people. The thorium, radium and the longer-lived radon daughter products lead-210 and polonium-210 may be subject to leaching from the tailings, possibly resulting in the contamination of surface and ground waters. Finally, the tailings themselves could be blown about or otherwise dispersed in the environment.

Successful management of the tailings must minimize exposure via each of these pathways and ensure that any such exposures that may occur will be within prescribed safe limits.

Some numbers will help to put the potential radiation exposure from tailings in perspective.

#### *Radon emission from tailings*

The concentration of radium-226 in tailings in the example previously considered, is 300 picocuries per gram. Each atom of radium-226 as it decays turns into an atom of radon-222, so the rate of radon production equals the rate of radium decay. The latter is determined by the half-life of radium-226 so that the rate of radon production can be readily calculated. The answer is, one gram of radium-226 (this is also 1 curie of radium-226) produces 2 microcuries of radon-222 per second. Thus the radon production rate in tailings containing 300 picocuries of radium-226 per gram is 0.0006 picocuries of radon per gram per second. The density of dry tailings is about 1.6 grams per cubic centimetre which means that the production rate of radon per unit volume is about 1000 picocuries per cubic metre per second.

Not all of the radon produced in the mound of tailings is released. Some will be retained in the tailings themselves, and some will decay before it can diffuse out of the tailings pile into the atmosphere. It has been found experimentally that only about a quarter of the radon produced is released from the tailings particles, and that effectively only the radon released in the first metre depth or so of tailings will find its way into the atmosphere. That is, a quarter of all the radon produced in each cubic metre of tailings at the surface of the pile will be released to atmosphere through each square metre of surface. The emanation rate from the pile will thus be about 250 picocuries per square metre per second.

The natural rate of release of radon-222 from the Earth's surface depends very much on the radium-226 content of the soil, its permeability and meteorological conditions (rain, wind speed, pressure). Average emanation rates in non-uranium bearing areas are between 1 and 2 picocuries per square metre per second. However at any one place it may vary with time over a range of ten and it can be higher by a factor of 100 over uranium bearing deposits than over normal soil. The emanation rate from the tailings pile will thus be some 200 to 300 times the natural emission rates in non-uranium areas but of the same order as that occurring over uranium orebodies.

The numbers used above to estimate radon emanation rates from tailings are illustrative only, though they are of the right order. Reported values for the radium-226 content of tailings range from 100 to 1000 picocuries per gram, the higher values being associated with the tailings coming from richer orebodies. Emanation rates would be expected to be related to the radium content of the tailings though a simple proportional relationship is unlikely to obtain. Tailings actually consist of two parts, a coarse-grain sandy fraction and the 'slimes' which come from the clay-like material in the original ore. Although the slimes comprise only about a quarter of the weight of the tailings, they contain about three quarters of the total radioactivity so that the amount of radon that escapes from a tailings pile will depend to some extent on the relative disposition of the slimes and sandy fraction.

The released radon will be quickly diluted in the atmosphere. Under average meteorological conditions dilution by a factor of 200 to 300 will occur within a few hundred metres downwind of the tailings.

During operation of the mill, the tailings will be covered by a layer of water which will reduce the release of radon to the atmosphere by a factor of about 25. At the close of the mill, and after the tailings dry out, radon release will increase to its maximum amount unless inhibited in some way. It is common practice now to stabilize tailings piles by planting them with grasses, sometimes after covering them with a layer of rock and soil. This will reduce emanation by a factor of four or so, depending on the thickness of the soil cover.

#### *External radiation*

Gamma radiation from a pile of tailings is in general not a serious problem. Radiation levels one metre from the pile surface tend to be less than 1 mrem per hour and average around 0.5 mrem per hour though 'hot spots' with much higher dose rates have been reported. As with radon emanation, higher surface dose rates are to be expected over the tailings from higher grade ores. Also if the tailings are managed so that separation of the slimes and the sandy fraction can occur, radiation levels will be higher over areas where the slimes predominate than over areas where the coarser material has deposited. Thus surface dose rates as high as 20 mrem per hour have been measured near the retaining wall of the tailings area associated with the old South Alligator uranium operation in the Northern Territory. The ore treated at this mill was of high grade and management of the tailings was such that the slimes tended to collect at the retaining wall. The dose rate drops to 2 mrem per hour over the sandy fraction. Because of these high radiation levels, access to tailings piles must be controlled and occupation of these areas prohibited.

The dose rate falls quickly with distance and is at normal background levels (0.01 mrem per hour) within about 50 metres of the pile face. A 60 cm covering of packed earth would also reduce gamma ray exposure over a tailings pile to near background levels.

#### *Exposure to radioactive dust from tailings*

The layer of water covering the tailings during the life-time of the mill, and stabilization of the tailings pile after the mill has closed will ensure that air-borne dust is minimized. Measurements around inactive,

unstabilized tailings piles in the USA show long-lived alpha air concentrations well below permissible levels within two to three hundred metres of the pile. Proper stabilization of the tailings at the completion of operations should ensure that release of tailings as dust is eliminated altogether.

#### *Seepage of radioactivity from tailings*

Seepage from the tailings retention system will be limited by design of the tailings dam to a standard that will ensure that during operation, contamination of surface and underground waters remains below prescribed limits. These limits will be set so that, having regard to all the pathways by which released radioactive material could eventually be ingested by man - by drinking contaminated water, eating fish or animals that may have themselves ingested the material, or eating vegetation that may have been grown in contaminated areas - doses to members of the public will be kept acceptably low. Seepage will also be controlled to ensure that the release of non-radioactive elements in the tailings (e.g. heavy metals) is below levels that could lead to damage to the environment.

After closure of the mill and the drying out of the tailings, seepage will be much reduced, the only remaining source of seepage being rain which falls directly on the piles, or runoff which may percolate through the tailings and leach out radioactive materials. This is minimized by ensuring that runoff is channelled around the dam, and by inhibiting as far as possible chemical processes within the tailings which could convert the radioactive material into a more readily leachable form. Some bacteria can do this so that attempts should be made to modify the environment within the tailings so that it is inimical to their growth. This is a special problem with ores containing significant quantities of sulphides (e.g. pyrite)

#### *Long-term management of tailings*

It is clear that although the concentration of radioactive materials in tailings is no greater than it was in the original ore, they pose a sufficient environmental and public health problem to require that they be kept under long-term surveillance. For tailings which have been accumulated above the ground, a number of requirements must be instituted to ensure that they remain a minimal exposure hazard to the public after milling operations have ceased. These requirements must include the following:

- . The tailings dam should be stabilized to preclude wind and water erosion. This may be done by grading the tailings pile, covering it with rock, earth and topsoil, and seeding it. Even more effective stabilization might be achieved if vegetation could be encouraged to root directly in the tailings.
- . Stabilized areas should be protected against runoff from surrounding areas.
- . Access by the public to stabilized areas should be controlled and no habitation permitted within the controlled area.
- . The covered area should not be subdivided, and neither private roads nor trails or rights-of-way should be permitted across the area.

- . Removal or use of the tailings should be controlled.
- . A continuing surveillance and monitoring program should be established to assess environmental levels of radioactive materials and to ensure that the dam continues to perform its proper function.

These restrictions must be binding on the mill operator for as long as he retains lease of the land, and on successive owners.

Most of these problems would disappear if the tailings could be returned to where they came from. This is not always practicable but is being done in some places, particularly in underground mines. Backfilling of opencut mines with tailings is also being assessed, the major problem being to ensure that enhanced contamination of ground or surface waters does not result.

#### CONCLUSION

The purpose of this paper has been to provide a background which will enable the radiological problems that can arise in the mining and milling of uranium to be understood. The radiation standards that should be applied and the procedures that should be adopted to minimize radiation exposure of workers and members of the public living in the vicinity of the mine and mill are set down in the Australian 'Code of Practice on Radiation Protection in the Mining and Milling of Radioactive Ores 1975'. If the requirements of this code are adhered to, the radiological safety of miners, mill workers and members of the public can be assured.

#### FURTHER READING

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## GLOSSARY

In this paper many terms are defined on first occurrence. However it may be helpful to the reader not familiar with nuclear and other terms used to have the following additional definitions.\*

|                         |  |
|-------------------------|--|
| background level        | The naturally occurring radiation level to which all persons are subject.  |
| carcinogen              | A substance which induces cancer in a living organism.   |
| cardio-vascular disease | Disease of the heart and blood vessel system.  |
| epidemiological study   | A study of the incidence of disease in a population.   |
| hematopoietic           | Relating to blood formation.   |
| irradiation             | Exposure to ionizing radiation.  |
| lymphatic               | Relating to the body's lymph system.   |
| monitor                 | A device used to measure the level of <i>ionizing radiation</i> or quantity of radioactive material.   |
| neoplasm                | New growth, or tumour.   |
| noble gases             | The gases, helium, neon, argon, krypton, xenon and radon-222, which are inert to all the usual chemical reactions.   |
| particulate             | Having particle form. Can be used to describe matter (in powder form) or radioactive particles.  |
| radiogenic              | Caused by radioactive decay.   |
| renal disease           | Disease of the kidney.   |
| rem                     | A unit of radiation dose equivalent, the product of absorbed dose, quality factor and other modifying factors necessary to obtain an evaluation of the effects of irradiation received by exposed persons, so that the different characteristics of the exposure are taken into account; whence <i>millirem</i> etc. |
| stoppage                | An enlarged part of the mine workings from which ore has been withdrawn.   |
| yellowcake              | The uranium oxide concentrate produced by a uranium treatment plant.   |

Sources

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United Kingdom Atomic Energy Authority : Glossary of Atomic Terms, Eighth Edition, 1974, UKAEA.

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TABLE 1  
THE URANIUM RADIOACTIVE DECAY SERIES

| Isotope          | Symbol | Historical name        | Half-life                | Radiation        | Alpha energy (MeV)† | Remarks  |
|------------------|--------|------------------------|--------------------------|------------------|---------------------|--|
| Uranium-238      | U-238  | Uranium I              | $4.5 \times 10^9$ y      | $\alpha$         |                     | $\beta$ rays in uranium ore come from these      |
| Uranium-234      | Th-234 | Uranium X <sub>1</sub> | 24.1 days                | $\beta \gamma$ } |                     |  |
| Protactinium-234 | Pa-234 | Uranium X <sub>2</sub> | 1.18 min                 | $\beta \gamma$ } |                     |  |
| Uranium-234      | U-234  | Uranium II             | $2.50 \times 10^5$ y     | $\alpha \gamma$  |                     | end up in tailings                               |
| Uranium-230      | Th-230 | Ionium                 | $7.6 \times 10^4$ y      | $\alpha$ }       |                     |  |
| Uranium-226      | Ra-226 | Radium                 | 1620 y                   | $\alpha$ }       |                     |  |
| Rn-222           | Rn-222 | Radon                  | 3.82 days                | $\alpha$         | 5.49                | gas  |
| Po-218           | Po-218 | Radium A*              | 3.05 min                 | $\alpha$         | 6.00                | collects on dust in mine                         |
| Pb-214           | Pb-214 | Radium B               | 26.8 min                 | $\beta \gamma$ } | 7.69                | $\beta$ and $\gamma$ rays in ore come from these |
| Bi-214           | Bi-214 | Radium C               | 19.7 min                 | $\beta \gamma$ } |                     |  |
| Po-214           | Po-214 | Radium C'*             | $2.7 \times 10^{-6}$ min | $\alpha$         |                     |  |
| Pb-210           | Pb-210 | Radium D               | 22.0 y                   | $\beta \gamma$ } |                     | May be used to monitor Rn-222 exposure           |
| Bi-210           | Bi-210 | Radium E               | 5.0 days                 | $\beta$          | 5.30                |  |
| Po-210           | Po-210 | Radium F               | 138.4 days               | $\alpha$         |                     |  |
| Pb-206           | Pb-206 | Radium G               | Stable                   |                  |                     |  |

*Ra-A and Ra-C' constitute the alpha hazard in exposure to Rn-222 and its daughters.*

*eV stands for million electron volts. It is a measure of the energy with which radiations are emitted from radioactive isotopes. A 5 MeV alpha-particle will penetrate some 40 microns of soft tissue.*

TABLE 2  
MEAN RADON-222 CONCENTRATIONS IN EQUILIBRIUM WITH  
SHORT-LIVED DECAY PRODUCTS IN GROUND-LEVEL AIR

| Site                        | Radon-222 activity,<br>picocuries per litre |
|-----------------------------|---|
| Czechoslovakia - Bratislava | 0.14  |
| Hungary - Budapest          | 0.07  |
| USA - Chicago               | 0.03  |
| - Cincinnati                | 0.26  |
| - Coastal sites             | 0.002                                       |
| Brazil - Rio de Janeiro     | 0.05  |
| Japan                       | 0.09  |
| Philippines                 | 0.004                                       |
| North Atlantic Ocean        | 0.006                                       |
| South Pacific Ocean         | 0.002                                       |
| South Pole                  | 0.0005                                      |



TABLE 3

RADON-222 CONCENTRATIONS IN DWELLINGS

| Location | Type of building and building material | Radon-222 concentration indoors, picocuries per litre | Comments                 |
|----------|--|---|--------------------------|
| en       | Houses - Wood                          | 0.3 - 0.9   | 4 air changes per hour   |
|          | - Brick                                | 0.3 - 2.1   |                          |
|          | - Concrete                             | 0.3 - 4.5   |                          |
|          | Houses - Brick                         | 0.12 - 4.3  |                          |
|          | - Concrete                             | 0.4   |                          |
|          | - Adobe                                | 0.3 - 10.0  |                          |
|          | - Slag                                 | 4.0 - 8.0   |                          |
|          | House                                  | 0.06 - 0.31   | Adequate ventilation     |
|          | House                                  | 0.2 - 0.7   | Poor ventilation         |
|          | Industrial premises                    | 0.005 - 1.2   | Poor ventilation         |
|          | Office buildings                       | 0.06 - 0.35   | Air conditioned          |
|          | House - Wood                           | 0.005 - 0.23  | 2-6 air changes per hour |
|          | - Concrete                             |   |                          |
|          | - Basement                             | 0.1 - 0.94  | 1-3 air changes per hour |
|          | - Concrete                             | 0.03 - 4.8  |                          |

TABLE 4

THE RELEVANT DATA USED TO DERIVE THE WORKING LEVEL

| Nuclide | Alpha energy (MeV)* | Half-life              | No. atoms in 100 picocuries | Ultimate alpha energy per atom | Total ultimate alpha energy (MeV/100 picocuries) |
|---------|---------------------|------------------------|-----------------------------|--------------------------------|--|
| RaA     | 6.00                | 3.05 m                 | 977                         | 6.00 + 7.69                    | $0.134 \times 10^5$                              |
| RaB     | 0.00                | 26.8 m                 | 8580                        | 7.69                           | $0.660 \times 10^5$                              |
| RaC     | 0.00                | 19.7 m                 | 6310                        | 7.69                           | $0.485 \times 10^5$                              |
| RaC'    | 7.69                | $2.7 \times 10^{-6}$ m | 0.0009                      | 7.69                           | $0.000 \times 10^5$                              |
|         |                     |                        |                             | Total                          | $1.279 \times 10^5$                              |
|         |                     |                        |                             | Round up to                    | $1.3 \times 10^5$                                |

\* MeV = million electron volts

TABLE 5  
EXPECTED AND OBSERVED DEATHS BY CAUSE AMONG  
3366 WHITE UNDERGROUND URANIUM MINERS,  
JULY 1950 TO SEPTEMBER 1968

| Cause of death                  | Number of deaths |          |
|---------------------------------|------------------|----------|
|                                 | Expected         | Observed |
| Tuberculosis, all forms         | 3.65             | 13*      |
| Malignant neoplasms             |                  |          |
| Respiratory                     | 11.71            | 70*      |
| Other sites                     | 29.45            | 38       |
| Cardiovascular-renal<br>disease | 117.77           | 115      |
| Violent deaths                  |                  |          |
| Motor vehicle accidents         | 19.45            | 32*      |
| Suicides and homicides          | 14.77            | 15       |
| Other violent deaths            | 19.42            | 79*      |
| All other causes                | 60.30            | 72       |
| Unknown causes                  | -                | 3        |
| Total                           | 276.58           | 437      |

\* *In these cases the increased number of observed deaths over what was expected is a real increase and not due to statistical errors in sampling*

TABLE 6  
EXPECTED AND OBSERVED RESPIRATORY CANCER DEATHS IN RELATION TO  
CUMULATIVE RADON DAUGHTER EXPOSURE MEASURED IN WORKING LEVEL  
MONTHS (WLM) AMONG 3366 WHITE UNDERGROUND URANIUM MINERS  
JULY 1950 TO SEPTEMBER 1968. OBSERVED DEATHS AT  
DECEMBER 1971 ARE ALSO GIVEN

| Estimated Cumulative<br>WLM | Person years<br>at risk | Number of deaths<br>to 1968 |          | Number of deaths<br>to 1971 |
|-----------------------------|-------------------------|-----------------------------|----------|-----------------------------|
|                             |                         | Expected                    | Observed | Observed*                   |
| 120                         | 8516                    | 1.81                        | 1        | 2                           |
| 120 - 359                   | 9355                    | 2.57                        | 12       | 15                          |
| 360 - 839                   | 9046                    | 2.95                        | 14       | 21                          |
| 840 - 1799                  | 6607                    | 2.52                        | 12       | 26                          |
| 1800 - 3719                 | 3455                    | 1.43                        | 21       | 34                          |
| 3720 and over               | 978                     | 0.42                        | 10       | 17                          |
| Total                       |                         | 11.71                       | 70       | 115                         |

\* *These observed deaths cannot be compared with the expected deaths in column 3, nor can they be related to the person years at risk, in column 2.*

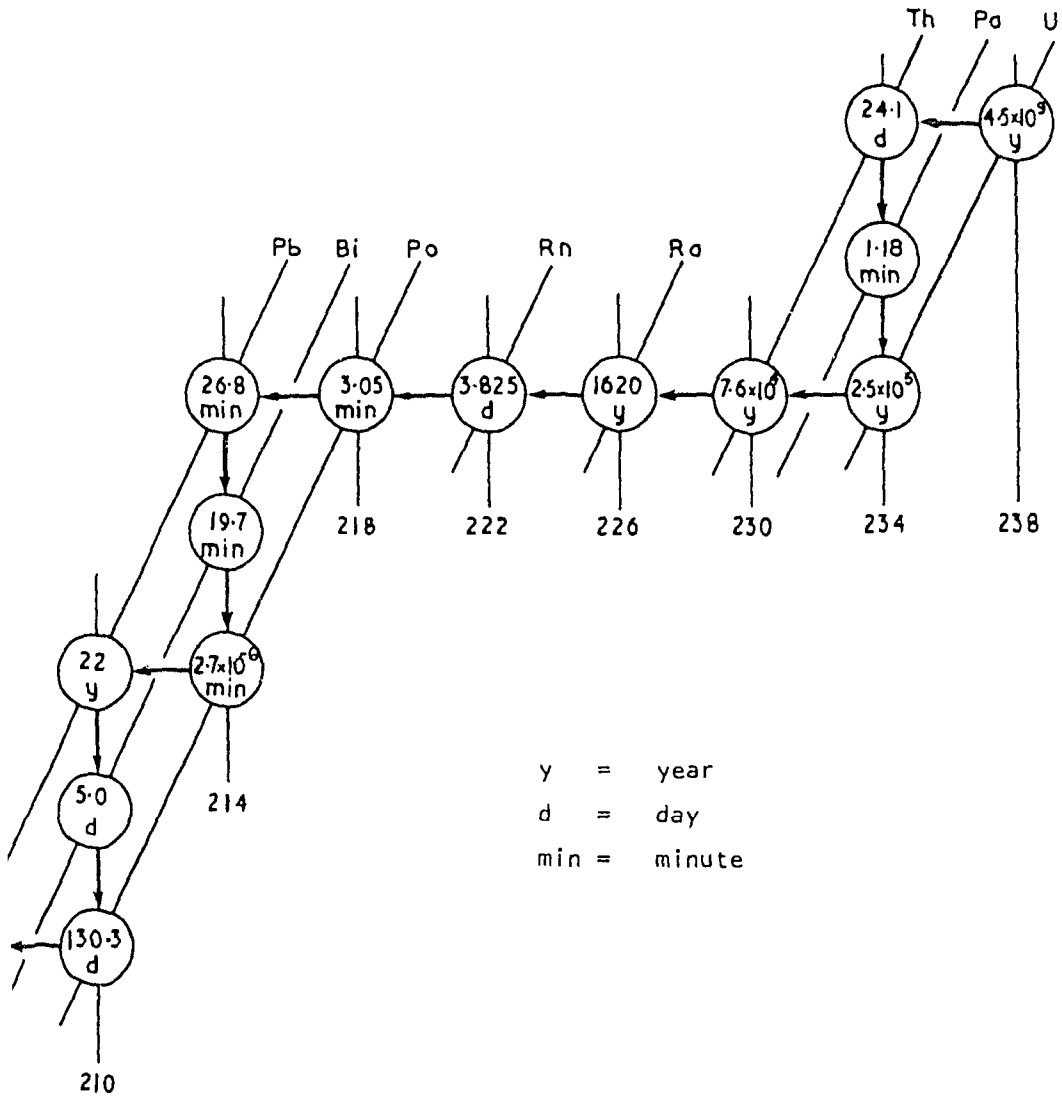


FIGURE 1

ACTIVE DECAY CHAIN OF U-238 THROUGH TO STABLE LEAD. EMISSION OF AN ALPHA PARTICLE LEADS TO THE ISOTOPE TO THE LEFT; EMISSION OF A BETA PARTICLE LEADS TO THE ISOTOPE IMMEDIATELY BELOW. HALF-LIVES ARE GIVEN IN THE CIRCLES. ISOTOPES OF THE SAME ELEMENT LIE ON THE SAME SLOPING LINE.

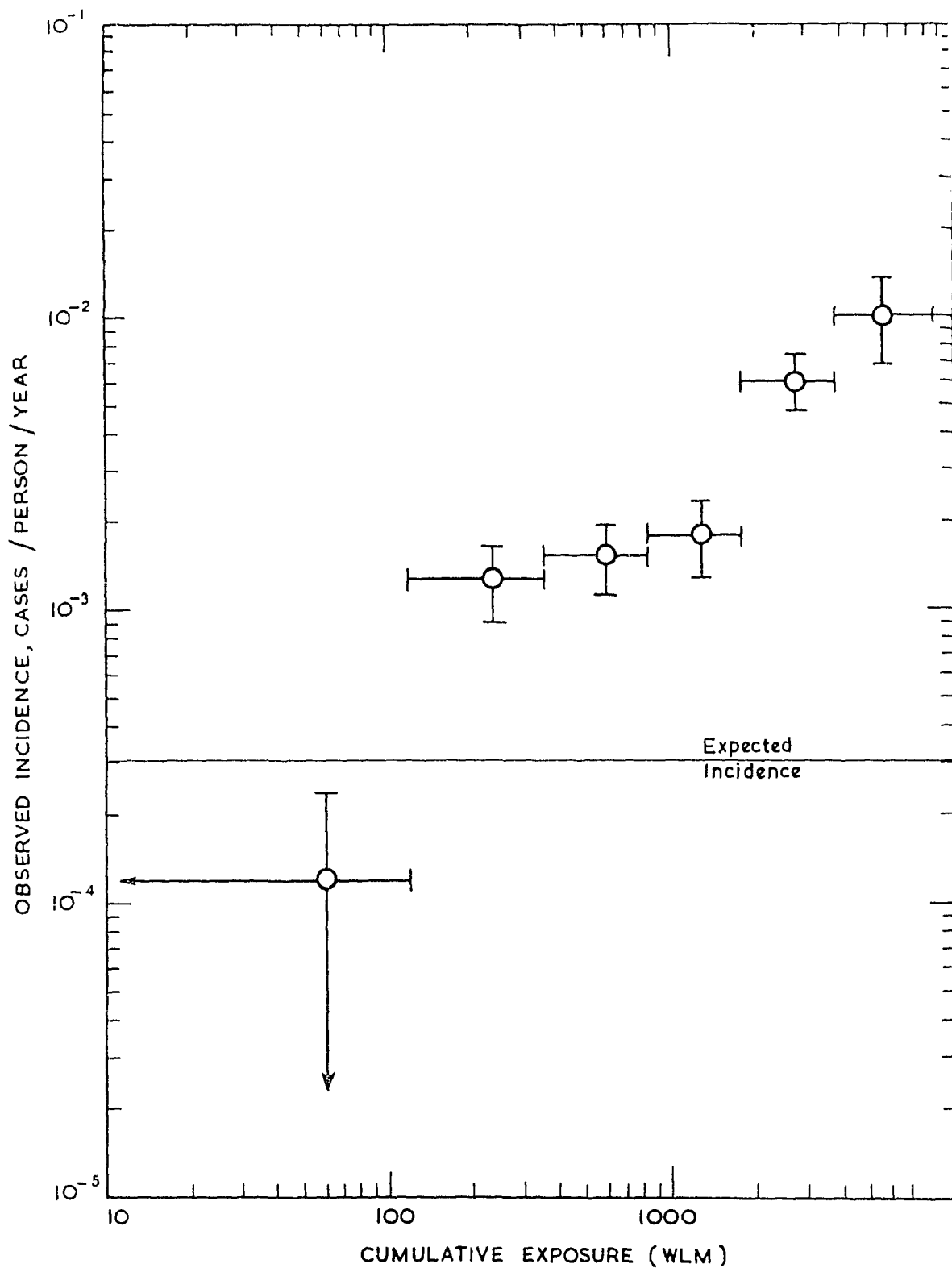


FIGURE 2

OBSERVED INCIDENCE OF LUNG CANCER IN A GROUP OF 3366 WHITE UNDERGROUND US URANIUM MINERS AS A FUNCTION OF THEIR CUMULATIVE EXPOSURE TO RADON DAUGHTER PRODUCTS, MEASURED IN WORKING LEVEL MONTHS (WLM). EXPECTED INCIDENCE IN THIS GROUP, HAD THERE BEEN NO EXPOSURE TO RADON, IS ABOUT  $3 \times 10^{-4}$  CASES PER PERSON PER YEAR.

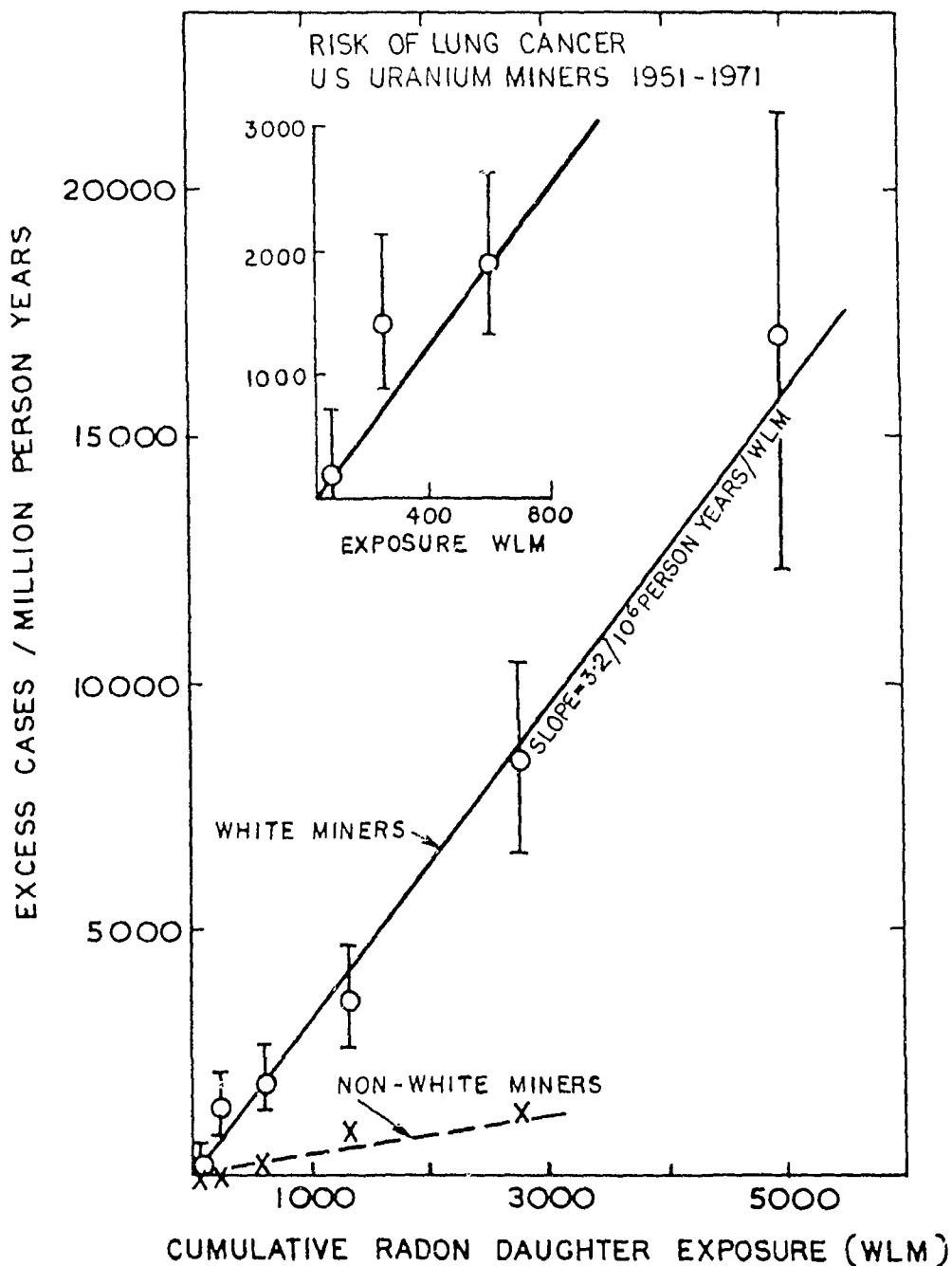


FIGURE 3

EXPOSURE-RESPONSE DATA FOR LUNG CANCER IN U.S. URANIUM MINERS (AFTER BEIR REPORT).

ERROR BARS FOR WHITE MINERS INDICATE THE STATISTICAL CONFIDENCE THAT MIGHT BE PLACED IN THE OBSERVED NUMBER OF EXCESS CASES.

INSERT: LOWEST EXPOSURE RANGE FOR WHITE MINERS.