

URANIUM MINING PROBLEM AS A POSSIBLE OAK RIDGE NATIONAL  
LABORATORY HEALTH PHYSICS RESEARCH PROGRAM\*

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Dr. W. S. Snyder and I were requested to testify before the Joint Committee on Atomic Energy on July 26, 1967, on the subject of maximum permissible exposure of miners in the United States to radon and its daughter products. Perhaps we were chosen for this assignment because, beginning with the establishment of the Internal Dose Committees of the NCRP and ICRP and to the present time, I have been chairman and Snyder has been secretary of both of these committees. The purpose of these committees at the national and international levels, respectively, is to set maximum permissible body burdens and maximum permissible concentrations, MPC, in air, water and food for the approximately 500 radionuclides of interest. Dr. W. F. Bale, a member of this Advisory Committee to the AEC Division of Biology and Medicine whom we are pleased to have with us this morning, first indicated in 1951 that the principal risk to  $^{222}\text{Rn}$  (3.82 days) was not from this noble gas itself or the daughter products it produces in the air passages of the lungs but rather from the daughter products  $^{218}\text{Po}$  (3.05 min.),  $^{214}\text{Pb}$  (26.8 min.) and  $^{214}\text{Bi}$  (19.7 min.) that are inhaled. Since this time, much research has been done and many papers have been published relating to the appropriate MPC or WL (working level) values for Rn and its daughter products.

The reason for the current Congressional interest in the uranium mining problem is underscored by the announcement of Dr. L. J. Gehrig, Acting Surgeon General, at the recent Congressional hearings. He said that of the 10,000 uranium miners in the United States, 98 have died of lung cancer and that projections indicate there may be a total of 530 deaths due to lung cancer in this group that has already been exposed. It is unfortunate, however, that at the present time certain very vital and essential pieces of information needed in the evaluation of this problem and in assessing this hazard are not available. Because of our long continued interest in this problem, because of our concern for the inhalation exposure to uranium by a large Oak Ridge occupational group, in consideration of our responsibility for our own employees who are handling and exposed

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to the world's largest quantities of separated transplutonium elements and in view of certain limited but unique interests and talents of some of our employees, we are proposing that we offer to the AEC our assistance in the uranium mining problem.

The present MPC value for  $^{222}\text{Rn}$  is based primarily on the work of Chamberlain and Dyson (BRIT. J. RADIOI. 29, 342, 317, June 1956) who, using a fabricated bronchial model, found experimentally that 18% of the  $^{218}\text{Po}$  ions were retained in the lower portion of the model. They found further that essentially all the radiation dose was delivered by the unattached ions of  $^{218}\text{Po}$  because in the large bronchial tube only the very small ions ( $10^{-8}$  to  $10^{-7}$  cm) had sufficient mobility to reach the tube walls. On this basis and using the characteristics of the standard man and limiting the dose rate to the critical lung tissue to 15 rem/yr, the Internal Dose Committees of NCRP and ICRP under our guidance set the MPC for  $^{222}\text{Rn}$  by the equation,

$$\text{MPC} = \frac{3 \times 10^{-6}}{1 + 1000f} \cdot$$

In this equation, it was assumed that the first daughter of  $^{222}\text{Rn}$  (i. e.  $^{218}\text{Po}$ ) is in 100% equilibrium with the  $^{222}\text{Rn}$  and  $f$  is the percent of unattachment. Since  $f$  had been reported to be about 0.1 for the typical situation, the value of the equation became

$$\text{MPC} = 3 \times 10^{-8} \mu\text{c } ^{222}\text{Rn/cc air}.$$

In view of the fact that the so-called working level, WL, was defined in terms of the  $\alpha$ -energy deposited by all the daughters of  $^{222}\text{Rn}$  ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ ) in 100% equilibrium down to  $^{210}\text{Pb}$  (19.4 yr), the above value of  $3 \times 10^{-8}$  corresponded to 0.3 WL.

Haque and Collinson (HEALTH PHYS. 13, 431, 1967) more recently have indicated from a theoretical treatment of the problem that perhaps Chamberlain and Dyson should have used a longer tube with at least five bifurcations to evaluate in their experiments the bronchial region of maximum ion deposition. Haque and Collinson's calculations would indicate values of MPC based on the dose rate of 15 rem/yr at the fifth bifurcation as follows:

$$\text{MPC} = 10^{-9} \mu\text{c } ^{222}\text{Rn/cc air if the critical tissue is at a depth of } 30 \mu$$

or

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$$\text{MPC} = 3 \times 10^{-9} \mu\text{c } ^{222}\text{Rn}/\text{cc air if the critical tissue is at a depth of } 50 \mu.$$

Thus, the present MPC, although one-third the WL, may be too large by a factor of 10 to 30. Similarly, it may be too low if most of the  $\alpha$ -energy is lost in the mucal blanket which passes over the critical region of the segmental bronchi.

The above discussion points up what I believe to be a great deficiency in our present knowledge; viz., where is the critical tissue in which these small cell, undifferentiated carcinomas have their origin? Autopsies on miners who have died of this disease are of no avail because this is a very rapidly growing malignancy and the average duration of disability from the time of first diagnosis to death is only two and a half months. By then, metastasis is so widespread that one cannot say whether the malignancy began at the first or the tenth bifurcation. Thus, it is of importance that many autopsies be carried out on uranium miners who have died of causes other than the typical lung carcinoma in the hope that eventually a few cases of early development of the malignancy may be discovered.

The Health Physics Division does not propose doing these human autopsies but, in the meantime, while these data are slowly accumulating from studies of the miners in Colorado and Utah, we propose three types of experiments which should provide more confidence in what is considered to be an acceptable MPC or WL value. These would include (1) the study and evaluation of mathematical models (to be discussed shortly by Snyder), (2) the study and evaluation of experimental plastic models extending down below the fifth bifurcation of the lung model, and (3) the study of a few dogs to confirm our model findings. Finally, we propose a fourth program for the development of more suitable instruments to evaluate the various parameters of significance in determining the MPC or WL in the uranium mines.

A brief outline of each of the proposed programs follows:

1. A Study and Evaluation of Mathematical Models (to be discussed by W. S. Snyder)

We have had many years experience developing and evaluating such models for the many radionuclides of interest to the NCRP and the ICRP. This would be a normal extension of our present program in the revision of the Internal Dose Handbooks of NCRP and ICRP.

2. A Study and Evaluation of Experimental Plastic Models

Haque and Collinson have indicated by their calculations that the fifth bifurcation rather than the first is the location of the critical tissue. Their theoretical work

should be checked experimentally with a plastic or rubberized model extending the laboratory studies of Chamberlain and Dyson. Concurrently, the Haque and Collinson calculations should be extended. They assumed laminar flow and certainly eddy currents must be taken into account at the bifurcations and during the periods when the direction of air flow in the bronchi is being reversed. Perhaps also some means can be found to simulate the flow of a mucal blanket within this plastic model under study. The alpha dose would be measured at various locations and depths in this plastic model by means of instrument techniques described below. Members of the Health Physics Division have had several years of experience in the development and use of various techniques for the production of submicroscopic particles of various size distribution and with other specified and controlled characteristics. Studies would be made with the use of these particles as carriers and with various mixtures of unattached ions of the several daughter products of  $^{222}\text{Rn}$ . These studies should lead to the identification of the portion of the bronchial tree receiving the greatest dose under specified exposure conditions. They should determine the  $\alpha$ -dose at various depths in the critical tissue which is undergoing mitosis. They should indicate the relative importance of each of the daughter products relative to the significant dose and determine whether unattachment is a parameter of paramount importance. Also, these studies should indicate which of the daughter products of  $^{222}\text{Rn}$  are important in delivering dose to the critical lung tissue. At present, the instruments used in assessing the radiation hazard in the uranium mines simply measure the total  $\alpha$ -radiation per unit volume of air per unit time. They do not identify the relative dose contribution by each of the daughters nor do they indicate anything about the fraction of the ions that are unattached. The importance of a knowledge of these parameters is illustrated by the following examples:

$10^{-7}$   $\mu\text{c}/\text{cc}$   $^{222}\text{Rn}$  with 100% equilibrium of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  and  $f = 1$  corresponds to 1 WL and 30 MPC

$10^{-7}$   $\mu\text{c}/\text{cc}$   $^{222}\text{Rn}$  with 100% equilibrium of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  and  $f = 0.1$  corresponds to 1 WL and 3 MPC

$10^{-7}$   $\mu\text{c}/\text{cc}$   $^{222}\text{Rn}$  with 10% equilibrium of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  and  $f = 0.03$  corresponds to 0.1 WL and 0.1 MPC

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$10^{-7}$   $\mu\text{c/cc}$   $^{222}\text{Rn}$  with 100% equilibrium of  $^{218}\text{Po}$  and 10% equilibrium of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  and  $f = 1$  corresponds to 0.55 WL at  $t = 0$ , 0.18 WL at  $t = 1$  hr, 0.2 WL at  $t = 4$  hr and to 30 MPC at any value of  $t$

Until experiments such as these are completed, it is impossible to say whether either the WL or the MPC relates properly to the radiation hazard of the uranium miners. In any case, we can say now with certainty that either or both the WL and the MPC as now defined and measured in the mines do not properly assess the lung hazard of the uranium miners.

### 3. The Study of a Few Dogs to Confirm Model Findings

This study would follow completion of the two above studies and would be carried out in order to confirm the results with animals. The study probably would not involve altogether more than five dogs. The dogs would be used with inhalation exposure chambers in studies similar to those we conducted at Y-12 some years ago. No pathological studies are contemplated and the animals would be used simply as engineering devices to confirm the theoretical and plastic model studies. The principal instruments for dosimetry would be LiF meters and plastic foils as discussed in the following section. Hopefully, some of these meters could be implanted at various locations in the bronchial tree of anesthetized animals during periods of exposure. We would study the  $^{222}\text{Rn}$  and daughter product retention and dose at various locations in the bronchial tree and at various tissue depths for various particle size distributions and for various fractions of uncombined daughter products. Some of these studies would be conducted with cigarette smoke present to determine the influence it might have on dose distribution.

### 4. The Development of Instruments to Evaluate Properly the Characteristics of $^{222}\text{Rn}$ and Its Daughter Products That Relate to Lung Malignancies Among the Uranium Miners

Our radiation protection efforts on behalf of occupational workers during the past quarter of a century as they relate to operations in "hot" cells and maintenance in areas of high potential for inhalation of airborne,  $\alpha$ -emitting radionuclides have indicated that dust sampling devices must be worn near the nostrils of a

worker in order to properly evaluate the hazard. Many of the estimates of the radiation hazards from airborne,  $\alpha$ -emitting radionuclides in the uranium mines are based on spot checks with air samplers fixed for given time periods in what are considered to be typical working areas. Most of these sampling instruments measure the total number (or energy) of  $\alpha$ -particles emitted per unit volume of air per unit time as a function of the time following a given air sampling period. A few devices such as the radon film badge have been developed to be worn by the miners. At the Congressional hearings, W. S. Johnson of Eberline Instrument Corporation indicated how he proposed to modify this radon film badge so that it might measure the radon daughter activity. He suggested the retention of the daughters by filter deposition, impaction or direct plate-out and placing the naked but light protected, nuclear track emulsion film a few mm distant so that it would be a target for the  $\alpha$ -radiation from the daughter products. This device would be essentially the same as that being studied and mentioned at the hearings by Dr. K. J. Schiager of Colorado State University except in this case the sensing element is a small, Teflon disk containing lithium fluoride (Li F). In each case, use would be made of a small, constant speed, battery-operated, motor-driven pump to draw a known volume of air through the meter.

Our proposal of a simple personnel air monitor would be similar to that suggested above by Johnson and Schiager except we would make use of certain plastic foils as the sensing element. In fact, we would make comparative studies of the advantages in using these foils as against the Li F technique. We are fortunate that Dr. Klaus Becker of Germany and one of the original developers of many of the techniques used in both of these systems (Li F and plastic foil) has recently joined our Health Physics Division and has already started some of these preliminary studies. Most of these studies are at present being directed to the basic problems associated with the formation of  $\alpha$ -tracks in plastic film and the results of these studies probably will lead to improvements in this new system of dosimetry. He has found that foils of cellulose nitrate and cellulose triacetate are most sensitive of materials so far studied. The  $\alpha$ -particles make holes in the foils which are made visible for counting by an etching procedure. The use of dyes and/or

automatic flying spot counting techniques can be used for rapid counting. One interesting possible application of this dosimetry development is to use plastic models (in Study 2 above) made of materials which would serve as their own  $\alpha$ -dosimeter. Some of the advantages of this system are as follows:

	<u>Plastic Film</u>	<u>Li F Meters</u>	<u>Film</u>
(1) Problems due to light sensitivity	none	few	serious
(2) Track fading due to high humidity	none	none	serious
(3) Fading at temperatures below 60°C	none	slight	serious
(4) Darkroom requirements	none	none	serious
(5) Possibilities for rapid scanning	good	good	poor
(6) Sensitivity to $\alpha$	good	intermediate	good
(7) Retention of record after reading	permanent	record lost	permanent
(8) Cost per unit	low	intermediate	intermediate
(9) Cost for automation	intermediate	low	prohibitive
(10) Adaptability of meters to dosimetry inside body	good	intermediate	poor
(11) Background error	none	intermediate	intermediate