RADON PROGENY INHALATION STUDY

AS APPLICABLE TO URANIUM MINING

MASTER

SPECIAL REPORT

ATOMIC ENERGY CONTRACT

AT-(11-1)-1500

**COLORADO STATE UNIVERSITY
FORT COLLINS, COLORADO**

August 1974

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Fate of Inhaled Radon Progeny

by

Donald R. Alexander

Special Report on U.S. Atomic Energy
Commission Contract No. AT (11-1)-1500

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

For research entitled
Radon Progeny Inhalation Study
As Applicable to Uranium Mining Operations

Directed by Principal Investigator
Keith J. Schiager

Department of Radiology and Radiation Biology
Colorado State University, Ft. Collins, Colorado

1974

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ABSTRACT

A shadow shield whole body counter and linear scanning techniques were used to determine the deposition and translocation of inhaled radon (^{220}Rn) progeny in volunteer experimental subjects.

The inhalation atmosphere was produced by combining nebulizer produced NaCl aerosol in a size range from $0.033\ \mu\text{m}$ to $0.088\ \mu\text{m}$ with ^{220}Rn progeny generated from a ^{228}Th solution. Total deposition percentages of 68% and 36% were determined for $0.033\ \mu\text{m}$ and $0.073\ \mu\text{m}$ aerosol respectively. An average of 54% of the deposited aerosol was found in the pulmonary region.

Movement of deposited radioactivity from the respiratory tract is clearly shown to be a function of particle size. Aerosols in a size range from $0.068\ \mu\text{m}$ to $0.088\ \mu\text{m}$ were shown to have a biological half life of approximately 20 hours in the pulmonary region while the $0.033\ \mu\text{m}$ aerosol had a biological half life of nearly 60 hours for the same region. A distinct difference in biological half life was shown between the pulmonary and tracheobronchial regions.

Donald R. Alexander
Department of Radiology and
Radiation Biology
Colorado State University
Fort Collins, Colorado 80523
August 1974

ACKNOWLEDGMENTS

This work was performed under Contract AT(11-1)-1500 between the U.S. Atomic Energy Commission and the Colorado State University Department of Radiology and Radiation Biology. Dr. Keith J. Schiager, the principal investigator of the project served as my major professor and advisor during the course of the work. His help and friendship are greatly appreciated. Dr. Edward L. Gillette, Dr. Hilding G. Olson and Dr. F. Ward Whicker constituted the remainder of my graduate committee, their efforts in my behalf are appreciated.

I am indebted to Stephanie Baker and Robert Fox for the technical assistance which proved to be so valuable during the course of the work. A. William Alldredge, Stephen T. Bard, Robert A. Fox, Don Paine and Keith J. Schiager consented to serve as experimental subjects, their willingness and cooperation cannot be measured and my sincere appreciation is infinite.

I am very grateful for the help and understanding given me by Dr. Max Zelle and Dr. James Johnson during the last hours of the work.

Finally, and certainly the work could not have been accomplished without the understanding and sacrifice on the part of my family. To them I dedicate this work.

Donald R. Alexander

TABLE OF CONTENTS

<u>Chapter</u>		<u>Page</u>
	LIST OF TABLES	vii
	LIST OF FIGURES.....	viii
I	INTRODUCTION.....	1
II	OVERVIEW OF THE PROBLEM.....	3
	Radiation and Carcinogenesis of the Lung	3
	Lung Models and Deposition of Inhaled Radioactivity.....	4
	Particle Size and Lung Deposition.....	6
III	EXPERIMENTAL PROCEDURES AND MATERIALS	10
	Laboratory Exposure Chamber.....	11
	Source of Radioactivity	11
	Build up and Decay of Thoron Progeny.....	17
	Determination of ThB (^{212}Pb) and ThC (^{212}Bi)	20
	Determination of Working Level Equivalent for ThB and ThC.....	24
	Aerosol Generation.....	26
	Aerosol Growth Kinetics.....	29
	Particle Size Analysis.....	33
	Unattached Fraction.....	36
	Assessment of Potential Hazard to the Individual.....	40
	Growth and Decay of Thoron Progeny <u>in situ</u>	41
	Working Level Exposures of the Experimental Subject	43
	<u>In Vivo</u> Measurements of Inhaled Activity.....	43

IV	DEPOSITION OF INHALED RADIOACTIVITY.....	50
	Regional Deposition	54
V	TRANSLOCATION OF DEPOSITED ACTIVITY.....	56
	Effect of Particle Size.....	57
VI	SUMMARY AND CONCLUSIONS	62
	<u>In Vivo</u> Detection.....	63
	Deposition of Inhaled Radioactivity	64
	Translocation of Thoron Progeny	65
	Assessing the Hazard to the Experimental Subject	67

LIST OF TABLES

<u>Table</u>		<u>Page</u>
1	Uranium Decay Series.....	13
2	Thorium Decay Series.....	14
3	Average Inhalation Concentrations of Thoron Progeny	19
4	Aerosol Characteristics.....	38
5	Average Chamber Conditions and Exposures for the Experimental Subjects.....	44
6	Summary of Chamber Conditions for Each Experiment.....	51
7	Total and Regional Deposition Value for Four Experimental Subjects.....	55
8	Summary of Translocation Values	68
9	Derivation of Exposure Limit for Inhaled Thoron Decay Products	69

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Respiratory tract deposition as a function of particle mass median aerodynamic diameter.....	8
2	Simplified portion of the Uranium-238 decay chain showing radon progeny of importance in the assessment of potential hazard to the uranium miner...	15
3	Simplified portion of the Thorium-232 decay chain showing the thoron progeny used in the study "Fate of Inhaled Radon Progeny"	16
4	Growth and decay of Thoron progeny from an initially pure source of Thoron (^{222}Rn).....	18
5	Graph of equation (3') showing the buildup and decay of Thorium-C in the exposure chamber when the ratio of ThB: ThC is 3: 1	25
6	Output characteristics of several compressed air nebulizers as a function of solute concentration	28
7	Decrease in particle count per cm^3 as a function of time for two aerosols generated in the exposure chamber	30
8	Particulate volume $1/n$ as a function of time in the exposure chamber where n represents the number of particles per cubic centimeter	32
9	Aerosol penetration through the diffusion battery as a function of particle size, for various flow rates in l/min.	35
10	Growth of aerosol in the exposure chamber as a function of time post generation	37
11	Buildup and decay of ^{208}Tl from <u>in situ</u> decay of ThB and ThC; ThB: ThC = 3: 1	42
12	Position of the subject to be counted, using the shadow shield whole body counter. Top figure illustrates the position of experimental subject and counter in end view. The bottom figure illustrates a side view of the detector and the anatomical positions assumed	45

<u>Figure</u>		<u>Page</u>
13	Response curve obtained by scanning a fixed thorium-228 source located in the plywood phantom....	47
14	Typical linear scan of experimental subject after smoothing of the data.....	53
15	Fraction of initially deposited activity of the 0.033 μm and 0.068 μm aerosol deposited in the tracheobronchial region of the respiratory tract as a function of time post exposure measured in hours	58
16	Fraction of initially deposited activity of the 0.033 μm and 0.068 μm aerosol deposited in the pulmonary region of the respiratory tract as a function of time post exposure measured in hours	59
17	Fraction of initially deposited activity in the pulmonary region for a 0.088 μm aerosol as a function of time post exposure measured in hours	61
18	Consent statement and exposure record	71

Chapter I

INTRODUCTION

Over the last several decades considerable concern and activity has been generated over the importance of the inhalation of toxic particulate matter. Much of this concern centers around the worker in industries engaged in the mining and milling of uranium. The earliest recognized hazard stems from the high incidence of respiratory tract malignancies prevalent among uranium miners. At the same time, the general public is exposed to low levels of carcinogenic aerosols, both radioactive and non-radioactive (Nelson, 1970).

A majority of the lung carcinomas originate in the bronchial region, and it is felt that this is the region of greatest deposition of submicron aerosols. A recent study (Schlesinger and Lippmann, 1972) involving deposition of particulate matter in the respiratory system was done using a model of the tracheobronchial tree. While this approach contributes significantly to the understanding of the dynamics of particle deposition, the model and its description of the aerodynamic behavior of particles only approximates the in vivo case. The lack of experimental data describing the in vivo deposition of inhaled radioactivity justifies this experimental work.

This work had as its objectives:

- (1) To determine the relationship between radon progeny carrier aerosol sizes and the deposition of the radon progeny in the respiratory tract.

(2) To determine the relative deposition of radon progeny in the ciliated, versus the non-ciliated regions of the respiratory tract.

(3) To determine the relative translocation and clearance rates of radon progeny from the ciliated and non-ciliated lung regions.

In a word, the "fate" of inhaled radon progeny implies the deposition and translocation of radon progeny within the respiratory tract.

Chapter II

OVERVIEW OF THE PROBLEM

Radiation and Carcinogenesis of the Lung

"The deterioration of man's relationship with his environment is being documented by an increasing number of alarming findings; among them is a clear link between the induction of cancer and the air we breathe." (Hanna, et al., 1970)

Even before the above statement was made much work had gone into the establishment of a connection between lung cancer and the inhalation of air containing toxic carcinogens. As summarized in hearings before the Joint Committee on Atomic Energy and the Congress of the United States (USAEC, 1967), three pertinent findings relating lung cancer and inhaled radioactivity were issued. They are: 1) there exists an excess of lung cancer among uranium miners; 2) that this excess of lung cancer is due at least in part to exposure to high concentrations of radon and its short lived daughter products within the uranium mines; and 3) the risk of lung cancer increases with increasing exposure.

The earliest practical experience with the occurrence of lung cancer originated with the high incidence of pulmonary disease among the miners of the Joachimsthal and Schneeberg mines of central Europe. With the advent of the uranium mining and milling industry in the United States during the 1940's and 1950's it had become clear that increased deaths due to lung cancer among uranium miners were associated with the exposure to excessively high concentrations of radon daughters existing in the mines at that time (USAEC, 1967; NIOSH, 1971). Details of radiation

exposure to persons working in radon contaminated atmospheres were described in the early 1950's (NIOSH, 1971; Bale and Shapiro, 1956). The crux of the problem centers around the deposition of progeny of radon in the respiratory tract with particular attention being paid to that amount of activity deposited in the tracheobronchial region. This region appears to be the principle site for the occurrence of bronchogenic cancer. One study done by Saccamanno, et al., (1964) concerned itself with the analysis of data obtained from all males in a 15 county area in Western Colorado and Eastern Utah, who were diagnosed as having cancer of the lung between January 1, 1948 and December 31, 1963. Statistical data was collected from as many available sources as possible, i.e. relatives, hospitals, vital statistics offices, physicians and the U.S. Public Health Service. As much information as possible was collected concerning the subjects' occupation, smoking habits, location of cancer and cancer types, etc. At the final analysis it was concluded that those people who had worked in the uranium mines had a significantly higher proportion of the small undifferentiated cancer cells than did a comparable group of non-miners.

Lung Models and Deposition of Inhaled Radioactivity

"When air containing radon or thoron is breathed some of the decay products are deposited on the surface of the respiratory systems, and the radiation from this active deposit then gives a dose to the epithelium." This statement, taken from the work of Chamberlain and Dyson (1956), lays the foundation for research related to inhalation, deposition, and dose to the epithelial lining of the respiratory tract.

The literature abounds with work seeking to answer the questions related to the fate of inhaled radon progeny; (Walsh, 1970); suffice it here to point out the principle areas of most discussion.

According to the ICRP Task Group on Lung Dynamics (ICRP, 1966); the aerodynamic properties of the dust; the physiology of respiration; and the anatomy of the respiratory tract govern dust deposition. These interrelationships have been well studied, demonstrated by experiments, and supported by theoretical treatments to conclude that dust deposition can be reasonably predicted in both quantitative and qualitative terms.

The ICRP Task Group on Lung Dynamics (ICRP, 1966) has taken the Findeisen-Landahl lung model (Findeisen, 1935; Landahl, 1950), and simplified the model to arrive at three well defined anatomical regions described as follows.

1. The nasopharynx, (N-P) --Beginning with the anterior nares and extending through the anterior pharynx, back and down through the posterior pharynx to the level of the larynx or epiglottis.
2. The tracheobronchial, (T-B) --Consisting of the trachea and the bronchial tree down to and including the terminal bronchioles.
3. The third compartment is referred to as the pulmonary, (P); this compartment consists of several structures; the respiratory bronchioles, alveolar ducts, atria, alveoli, and the alveolar sacs.

The nasopharynx, (N-P) and the tracheobronchial, (T-B) make up what is medically referred to as the "upper respiratory tract" while the pulmonary, (P) region constitutes the "lower respiratory passages"

(ICRP, 1966). Some experimental work, for example, Altschuler, et al., (1964) and Jacobi (1964) has substantiated that the Findeisen-Landahl lung model is adequate for the purpose of dose calculations; yet there is a measure of disagreement as Hague and Collinson (1967) considered the Landahl model to be antiquated and not based upon sufficient experimental data. Instead they chose the model of Weibel (1963) which was more extensive and according to Weibel, "based on more experimental data with greater accuracy in the dimensions of the proximal airways." A thorough comparison of the anatomical models may be found in Walsh (1970). Suffice it to say at this point; that while the estimated deposition of inhaled radioactive material, and the estimated dose to the respiratory tract depend upon which anatomical model is chosen; a clearer picture of the "fate" of inhaled radioactivity would be enhanced by conducting experimental inhalation exposures.

Particle Size and Lung Deposition

Greater differences, than those of anatomical modeling, exist in the determination of particle sizes and their subsequent deposition in the respiratory tract. The mechanisms of deposition appear to be impaction, sedimentation and diffusion, with the first occurring primarily in the N-P region and in areas where a change in direction of air flow would cause particles to impact on the walls of the air ways. Diffusion of particles from an air stream moving in a cylindrical tube has been investigated and mathematically formulated by Gormley and Kennedy (1949). The equations describing the fraction of particles penetrating the cylindrical tube are used (1) to predict the deposition of particulate matter which diffuses to the wall of the bronchial tubes and (2) as will be shown later, a means of evaluating the aerodynamic diameter of the inhaled aerosol.

While the former application provides a means of determining deposition in a model it cannot describe accurately the dynamic character associated with particle deposition in the respiratory tract. Previous studies by Chamberlain and Dyson (1956), Hague and Collinson (1967), and Altshuler, et al., (1964) combined anatomical modeling and the Gormley-Kennedy equation to predict particle deposition in the respiratory tract and estimate the dose to lung tissue from inhalation of radon and progeny.

The deposition and clearance of airborne particles in the lung as stated by P.E. Morrow (1970) is "... the outcome of the intrinsic instability of aerosols and the spatial and temporal relationships which develop between the respiratory structures and aerosol particles during respiration." The physical dimension of the particle is one such intrinsic parameter which relates directly to deposition. The larger particles tend to deposit by impaction and sedimentation while the smaller ones deposit by diffusion. The depth of penetration of an inhaled particle into the respiratory tract is a function of the size and density of the aerosol (Lippman and Albert, 1969). In addition the clearance mechanisms associated with dissolution, phagocytosis and direct penetration through the alveolar membrane may be directly affected by particle size (Hatch and Gross, 1964).

The ICRP Task Group on Lung Dynamics, (ICRP, 1966) has proposed that the evaluation of an inhalation hazard should be initiated by examining the relation between the deposition pattern of a given aerosol and some parameter of the aerosol. Figure 1, from the ICRP, (1966) illustrates the fraction of an inhaled aerosol deposited as a

Figure 1: Respiratory tract deposition as a function of particle mass median aerodynamic diameter (ICRP, 1966).

function of the mass median aerodynamic diameter (MMAD)* for the three anatomical regions. The probability of deposition for particles in a size range less than $0.1 \mu\text{m}$ diameter increases to a maximum at $0.01 \mu\text{m}$ diameter for the pulmonary region and the tracheobronchial region; below this value the deposition probability decreases in these regions (Morrow, 1970). At the same time the deposition of the aerosol with aerodynamic diameter less than $0.1 \mu\text{m}$ diameter increases in the nasopharynx region. Below approximately $0.1 \mu\text{m}$ diameter, deposition is predominately by diffusion, consequently the $0.01 \mu\text{m}$ diameter particles diffuse quickly to the walls of the respiratory tract. It therefore becomes increasingly necessary to generate quantitative data with which to resolve the questions pertaining to deposition and dose resulting from inhalation of extremely small radioactive aerosols.

* The mass median aerodynamic diameter is the median diameter or geometric mean of the log-normal distribution of particles as a function of the mass or volume of the particle (Raabe, 1970).

Chapter III

EXPERIMENTAL PROCEDURES AND MATERIALS

Previous work carried out as a portion of the Radon Progeny Inhalation Study as Applicable to Uranium Miners (AEC Contract (11-1)-1500) by members of the Radiology and Radiation Biology Department at Colorado State University, placed emphasis on the physical characterization of the aerosol particulates and their association with the radioactivity in the uranium mine. These studies dealt with the health and safety precautions for the uranium miner and methods of measuring and evaluating long-term doses received by the miner as a consequence of his employment in the industry.

The technique utilized in this work was actually developed in 1968-69 when whole body measurements were made on uranium miners to locate sites of radioactivity deposited in the respiratory tract (Radon Progeny Inhalation Study, COO-1500-3, 1967). At that time, a series of exposures was carried out on volunteer subjects to characterize the deposition of radioactivity in the lung. This work, following as a logical extension of the prior experiments, combined two aspects of the inhalation study:

- (1) To generate an aerosol particulate in the size range normally found in the uranium mine, and
- (2) to combine the aerosol with a radioactive material and expose human volunteer subjects to this combination in the form of a breathing atmosphere.

Following inhalation of the atmosphere the subject was gamma counted in a shadow shield whole body counter to locate regions of

deposition of the sub-micron sized radioactive aerosol. Counting was continued over a period of time to determine the translocation rate of deposited activity from the respiratory tract.

Laboratory Exposure Chamber

Earlier portions of the Radon Progeny Inhalation Study simulated a working uranium mine atmosphere in a laboratory inhalation chamber (Holleman, 1968).

The chamber, 4.3 m long by 1.4 m wide and 2.0 m high was constructed of sheet metal and covered on the outside with two inches of polystyrene foam insulation to assist in maintaining uniform temperature in the chamber. A large, 4' x 4' glass window in one wall of the chamber allows the experimenter to observe the experimental subjects during the course of the experiment. Two Plexiglas strips, 48" x 6" in the wall of the chamber were drilled with 2.5" diameter ports through which the inhalation atmosphere was generated and sampled. Entrance to the chamber is gained through a double door airlock which reduces loss of the atmospheric conditions generated in the chamber.

Source of Radioactivity

The source of radioactivity for the experiment was 1.9 year half life thorium 228, in the form of a thorium nitrate solution. The solution was contained in a gas de-emanation apparatus which allowed the removal of the thoron, (radon 220), present in equilibrium with thorium 228 by bubbling the solution with a stream of dry nitrogen. A series of gas dispersion frits and traps prevented the parent solution from becoming entrained with the nitrogen and entering the chamber. A final absolute filter placed in the generation line was used to insure against any thorium

entering the chamber. Alpha spectroscopy of filter samples of the atmosphere within the chamber showed the air to be free of thorium 228.

Thoron (^{220}Rn), the source gas for the inhalation experiment, decays with a 55 second half life to thorium A (^{216}Po) which decays with a 0.15 second half life to thorium B (^{212}Pb). The thorium B in turn becomes the source of radioactivity for the inhalation. The airborne radioactivity present in the environs of the uranium mine originates with the emanation of radon (^{222}Rn) released at the face of the uranium bearing deposit as the deposit is mined. The source of the airborne radioactivity for the experiment, as previously stated, is the "emanation thoron" (Tn or ^{220}Rn). Tables 1 and 2 from the Radiological Health Handbook (1970) give the decay scheme for uranium 238, precursor of radon; and for thorium 232; the precursor of thoron. Figures 2 and 3 illustrate the radon and thoron progeny of interest in a simplified manner. The use of ^{220}Rn as an analog for ^{222}Rn for generation of the inhalation atmosphere was justified by the following considerations:

- (1) The controlling daughter of the thoron progeny chain, ^{212}Pb , decays with a half life of 10.64 hours; this results in an effective half life for the inhaled progeny of about 10 hours, while the effective half life of the ^{222}Rn progeny chain is approximately 30 to 40 minutes; consequently the experimenter has a longer time period in which to make measurements on the experimental subjects.
- (2) The decay chain has a series of gamma rays with sufficient energies to allow for external detection. In particular the 2.62 MeV gamma ray from thorium C" (^{208}Tl) occurs with sufficient frequency to be easily detectable, and in an energy

Table 1
Uranium Decay Series

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities		
			α	β	γ
$^{238}_{92}\text{U}$	Uranium I	$4.51 \times 10^9 \text{ y}$	4.15 (25%) 4.20 (75%)	---	---
$^{234}_{90}\text{Th}$	Uranium X_1	24.1d	---	0.103 (21%) 0.193 (79%)	0.063c† (3.5%) 0.093c (4%)
$^{234}_{91}\text{Pa}^m$	Uranium X_2	1.17m	---	2.29 (98%)	0.765 (0.10%) 1.001 (0.60%)
99.87% $^{234}_{91}\text{Pa}$ 0.13% $^{234}_{91}\text{Pa}$	Uranium Z	6.75h	---	0.53 (66%) 1.13 (13%)	0.100 (50%) 0.70 (24%) 0.90 (70%)
$^{234}_{92}\text{U}$	Uranium II	$2.47 \times 10^5 \text{ y}$	4.72 (28%) 4.77 (72%)	---	0.053 (0.2%)
$^{230}_{90}\text{Th}$	Ionium	$8.0 \times 10^4 \text{ y}$	4.62 (24%) 4.68 (76%)	---	0.068 (0.6%) 0.142 (0.07%)
$^{226}_{88}\text{Ra}$	Radium	1602y	4.60 (6%) 4.78 (95%)	---	0.186 (4%)
$^{222}_{86}\text{Rn}$	Emanation Radon (Rn)	3.823d	5.49 (100%)	---	0.510 (0.07%)
$^{218}_{84}\text{Po}$	Radium A	3.05m	6.00 (~100%)	0.33 (~0.019%)	---
99.98% $^{214}_{82}\text{Pb}$ 0.02% $^{218}_{85}\text{At}$	Radium B	26.8m	---	0.65 (50%) 0.71 (40%) 0.98 (6%)	0.295 (19%) 0.352 (36%)
$^{218}_{85}\text{At}$	Astatine	~2s	6.65 (6%) 6.70 (94%)	? (~0.1%)	---
$^{214}_{83}\text{Bi}$	Radium C	19.7m	5.45 (0.012%) 5.51 (0.008%)	1.0 (23%) 1.51 (40%) 3.26 (19%)	0.609 (47%) 1.120 (17%) 1.764 (17%)
99.98% $^{214}_{84}\text{Po}$ 0.02% $^{210}_{81}\text{Tl}$	Radium C'	164 μs	7.69 (100%)	---	0.799 (0.014%)
$^{210}_{81}\text{Tl}$	Radium C''	1.3m	---	1.3 (25%) 1.9 (56%) 2.3 (19%)	0.296 (80%) 0.795 (100%) 1.31 (21%)
$^{210}_{82}\text{Pb}$	Radium D	21y	3.72 (.000002%)	0.016 (85%) 0.061 (15%)	0.047 (4%)
$^{210}_{83}\text{Bi}$	Radium E	5.01d	4.65 (.00007%) 4.69 (.00005%)	1.161 (~100%)	---
~100% $^{210}_{84}\text{Po}$.00013% $^{206}_{81}\text{Tl}$	Radium F	138.4d	5.305 (100%)	---	0.803 (0.0011%)
$^{206}_{81}\text{Tl}$	Radium E''	4.19m	---	1.571 (100%)	---
$^{206}_{82}\text{Pb}$	Radium G	Stable	---	---	---

Table 2

Thorium Decay Series

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{232}_{90}\text{Th}$	Thorium	$1.41 \times 10^{10} \text{ y}$	3.95 (24%) 4.01 (76%)	---	---
$^{228}_{88}\text{Ra}$	Mesothorium I	6.7 y	---	0.055 (100%)	---
$^{228}_{89}\text{Ac}$	Mesothorium II	6.13 h	---	1.18 (35%) 1.75 (12%) 2.09 (12%)	0.34c# (15%) 0.908 (25%) 0.96c (20%)
$^{228}_{90}\text{Th}$	Radiothorium	1.910 y	5.34 (28%) 5.43 (71%)	---	0.084 (1.6%) 0.214 (0.3%)
$^{228}_{88}\text{Ra}$	Thorium X	3.64 d	5.45 (6%) 5.68 (94%)	---	0.241 (3.7%)
$^{220}_{86}\text{Rn}$	Emanation Thoron (Tn)	55 s	6.29 (100%)	---	0.55 (0.07%)
$^{216}_{84}\text{Po}$	Thorium A	0.15 s	6.78 (100%)	---	---
$^{212}_{82}\text{Pb}$	Thorium B	10.64 h	---	0.346 (81%) 0.586 (14%)	0.239 (47%) 0.300 (3.2%)
$^{212}_{83}\text{Bi}$	Thorium C	60.6 m	6.05 (25%) 6.09 (10%)	1.55 (5%) 2.26 (55%)	0.040 (2%) 0.727 (7%) 1.620 (1.8%)
$^{212}_{84}\text{Po}$	Thorium C'	304 ns	6.78 (100%)	---	---
$^{208}_{81}\text{Tl}$	Thorium C''	3.10 m	---	1.28 (25%) 1.52 (21%) 1.80 (50%)	0.511 (23%) 0.583 (86%) 0.860 (12%) 2.614 (100%)
$^{208}_{82}\text{Pb}$	Thorium D	Stable	---	---	---

Figure 2: Simplified portion of the Uranium-238 decay chain showing radon progeny of importance in the assessment of potential hazard to the uranium miner.

Figure 3: Simplified portion of the Thorium-232 decay chain showing the thoron progeny used in the study "Fate of Inhaled Radon Progeny".

range away from interference from other gamma rays occurring naturally or in tandem with the thoron progeny. It should be emphasized that while the ThB and ThC will be deposited in the respiratory tract. The in vivo detection procedure will utilize the in situ decay of ^{208}Tl .

- (3) The thoron (^{220}Rn) progeny, ThB, ThC and ThC" match, chemically and physically, the short lived radon (^{222}Rn) progeny of the uranium mine and therefore provide the investigator with comparable physical, chemical and biological behavior.
- (4) Finally, the absence of a long-lived daughter, such as ^{210}Pb , 22 year half life, in the thorium chain prevents any potential hazard, to the subjects, resulting from accumulation of long-lived radioactivity within the body.

Build up and Decay of Thoron Progeny

After the laboratory inhalation chamber was charged (10-20 minutes at 3-5 l/min) a decay time of 10 minutes was allowed for the thoron-thorium A series to decay and for the ingrowth of ThB. Figure 4 shows the theoretical growth and decay of ThB and ThC from an initially pure source of thoron. The figure indicates that equilibrium between the ThB and ThC-ThC" occurs in 5-6 hours. Trial experiments performed in the exposure chamber to determine the concentration of ^{212}Pb and ^{212}Bi in the chamber indicated that at such time when the concentration of ^{212}Pb was a maximum the ratio of ^{212}Pb to ^{212}Bi was 3:1. Table 3 summarizes the average air concentrations of ThB (^{212}Pb) and ThC (^{212}Bi) in the exposure chamber during inhalation.

Figure 4: Growth and decay of Thoron progeny from an initially pure source of Thoron (^{220}Rn).

Table 3

Average Inhalation Concentrations of Thoron Progeny

Experiment No.	Air Concentration pCi/l	
	ThB	ThC
1	649	237
2	650	245
3	358	115
4	873	375

Determination of ThB (^{212}Pb) and ThC (^{212}Bi)

Concentrations of ^{212}Pb and ^{212}Bi present in the exposure chamber were determined using alpha spectroscopy of air samples drawn during the course of the experiment. For the purpose of determining the air concentrations of ^{212}Pb and ^{212}Bi the decay scheme for ^{232}Th , Figure 3, may be simplified without loss of generality (Thomas, 1971). The following assumptions may be accepted:

- (1) The decay of ThA decaying via astatine (about 0.01%), may be ignored.
- (2) Thoron (Tn) may be considered to decay directly to ThB, due to the short half life, 0.15 sec., of ThA.
- (3) ThC may be considered to decay directly, by alpha emission to ThD (stable lead), with 35.0% of the alphas at 6.06 MeV and 65.0% at 8.78 MeV.

The simplified decay chain for thoron (Tn) may be written as:

The appropriate decay constants are:

$$\begin{aligned}\lambda_B(\text{ThB}) &= 0.0654 \text{ hr}^{-1} \\ \lambda_C(\text{ThC}) &= 0.6874 \text{ hr}^{-1}\end{aligned}$$

The development of equations for determination of air concentrations for ThB and ThC follows the pattern of Martz, et al. (1969) and Duggan (1973).

Let

N_i = Number of atoms of the i^{th} nuclide

C_i = Air concentration of i^{th} nuclide

Q = Sampling flow rate

i = 0, 1, 2, 3 for Thoron, Tn, ThB, ThC, ThD

If an atmosphere containing a mixture of thoron and thoron progeny is sampled by drawing air through a membrane filter at a flow rate Q for a time T the equation

$$\frac{dN_i}{dt} = \lambda_{i-1}N_{i-1} - \lambda_i N_i + C_i Q$$

describes the collection of the i^{th} nuclide on the filter. For ThB and ThC;

$$\frac{dN_1}{dt} = \lambda_0 N_0 - \lambda_1 N_1 + C_1 Q \quad (1)$$

and

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 + C_2 Q \quad (2)$$

If the chamber, initially charged with thoron ($T_{1/2} = 55s$) is allowed to stand for 10-20 minutes before sampling, the term $\lambda_0 N_0$ in equation (1) is zero and

$$\frac{dN_1}{dt} = C_1 Q - \lambda_1 N_1$$

The solution of this equation is straightforward, resulting in

$$N_1 = \frac{C_1 Q}{\lambda_1} (1 - e^{-\lambda_1 t}) \quad (3)$$

Similarly equation (2) may be solved in straightforward fashion to yield

$$N_2 = \frac{C_1 + C_2}{\lambda_2} (1 - e^{-\lambda_2 t}) Q - \frac{C_1}{\lambda_2 \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) Q \quad (4)$$

Rearranging the terms of equation (4) gives

$$N_2 = \frac{C_1 \lambda_1 Q}{\lambda_2 \lambda_1} \left[\frac{(1 - e^{-\lambda_1 t})}{\lambda_1} - \frac{(1 - e^{-\lambda_2 t})}{\lambda_2} \right] + \frac{C_2 Q}{\lambda_2} (1 - e^{-\lambda_2 t})$$

Let $N_i(T)$ = number of atoms of the i^{th} nuclide on the filter at the end of sampling, then

$$N_1(T) = \frac{C_1 Q}{\lambda_1} (1 - e^{-\lambda_1 T})$$

and

$$N_2(T) = \frac{C_1 \lambda_1 Q}{\lambda_2 \lambda_1} \left[\frac{1 - e^{-\lambda_1 T}}{\lambda_1} - \frac{1 - e^{-\lambda_2 T}}{\lambda_2} \right] + \frac{C_2 Q}{\lambda_2} (1 - e^{-\lambda_2 T})$$

At some time, t , later

$$N_1 = N_1(T) e^{-\lambda_1 t}$$

and

$$N_2 = \frac{\lambda_1 N_1(T)}{\lambda_2 - \lambda_1} \left[\frac{-\lambda_1^t}{e} - e^{-\lambda_2^t} \right] + N_2(T) e^{-\lambda_2^t}.$$

The activity on the filter at the later time (t) is given by:

$$N_1 \lambda_1 = C_1 Q (1 - e^{-\lambda_1^T}) e^{-\lambda_1^t},$$

and

$$N_2 \lambda_2 = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} (e^{-\lambda_1^t} - e^{-\lambda_2^t}) N_1(T) + \lambda_1 N_2(T) e^{-\lambda_2^t}.$$

For simplicity the following terms are defined

λ_B, λ_C = decay constants for ThB and ThC

$N_1(T)$ = B_o

$N_2(T)$ = C_o

C_B, C_C = Atoms/liter of ThB and ThC respectively in the air sampled

A_B, A_C = pCi/l of ThB and ThC in the air sampled.

Summarizing:

$$B_o = \frac{C_B Q}{\lambda_B} \left(1 - e^{-\lambda_B^T} \right), \text{ and} \quad (1')$$

$$C_o = \frac{C_B \lambda_B Q}{\lambda_C - \lambda_B} \left[\frac{1 - e^{-\lambda_B^T}}{\lambda_B} - \frac{1 - e^{-\lambda_C^T}}{\lambda_C} \right] + \frac{C_C Q}{\lambda_C} \left(1 - e^{-\lambda_C^T} \right). \quad (2')$$

At some time t after sampling the activity of ThC on the filter is given by:

$$\lambda_C C_C = \frac{\lambda_B \lambda_C}{\lambda_C - \lambda_B} \left[e^{-\lambda_B t} - e^{-\lambda_C t} \right] B_0 + \lambda_C e^{-\lambda_C t} C_0, \quad (3')$$

therefore the desired concentrations and activities may be determined by counting the air sample filters immediately following a sampling time T and again at some time t greater than 300 min (Duggan, 1973). The first measurement gives directly the concentration of ThC activity on the filter. The second measurement allows the calculation of the activity of ThB on the filter. Therefore, concentrations of ThB and ThC in the air may be determined.

Equation (3') represented graphically in Figure 5 with $B_0 : C_0 = 3:1$ and a sampling time of 5 minutes, (typical conditions existing in the inhalation chamber and sampling time for determination of ThB and ThC), shows the increase in alpha activity, $\lambda_C C_C$, until transient equilibrium is reached at some time greater than 300 minutes post sampling (Duggan, 1973).

All air samples were alpha counted using silicon surface barrier detectors and a TMC 400-channel pulse height analyzer.

Determination of Working Level Equivalent for ThB and ThC

The working level (WL) unit of exposure to inhaled radioactivity given by American National Standards Institute (ANSI N13.8, 1973) is defined to be "Any combination of radon daughters in one liter of air that will ultimately release 1.3×10^5 MeV of alpha energy during decay to ^{210}Pb (RaD)." There is no equivalent definition for thoron daughters but the same principle may be applied and the potential alpha energy from ThB

Figure 5: Graph of equation (3') showing the buildup and decay of Thorium-C in the exposure chamber when the ratio of ThB:ThC is 3: 1.

and ThC determined and compared with the definition. For each ThB atom and ThC atom present in the air, the complete decay to stable lead (Pb-208) results in the ultimate emission of 7.8008 MeV of alpha energy. Therefore the working level value from ThB and ThC may be determined from

$$\begin{aligned} \text{WL} &= \frac{(7.80) (C_B + C_C) \text{ MeV}}{1.3 \times 10^5 \frac{\text{MeV}}{\text{WL}}} \\ &= 6.00 \times 10^{-5} (C_B + C_C) \end{aligned}$$

C_B, C_C = Air concentrations of ThB and ThC atoms/liter

Aerosol Generation

From the standpoint of deposition of radioactivity in the lung, the aerodynamic diameter of the inhaled radioactive aerosol is the most important parameter (Mercer, et al., 1965). For the purpose of this work to be fully realized, careful attention was paid to the generation and measurement of a suitable solid aerosol for attachment to the radioactivity. There are a number of techniques commonly used for the generation of aerosols, including dispersion from dry powders, atomization of solution and suspensions using compressed air and ultrasonic nebulizers, hot wire techniques and exploding wire production (Raabe, 1970; Stein, et al., 1973; Mercer, et al., 1968; Mercer, et al., 1965). By far the simplest and most convenient method is the generation of the aerosol using a common compressed air nebulizer. The output characteristics of the compressed air nebulizer have been extensively investigated (Mercer, et al., 1968; Raabe, 1970) and the fundamental production parameter relating aerosol size with nebulizer solution concentration is

$$D_p = D_s \left(\frac{c}{\rho} \right)^{1/3}$$

where

- D_p = Mass median aerodynamic diameter (μm) (10^{-6} meters)
 D_s = Volume median diameter (μm)
 c = Solute concentration (g/ml)
 ρ = Dry particle density (g/cm^3)

The volume median diameter of the nebulizer produced droplets is a function of the pressure drop across the output orifice. The nebulizer generates droplets by shattering a liquid stream with fast moving air. The liquid is drawn into the stream of air by the sudden drop in pressure created by the air from the orifice. Evaporation of the liquid occurs rapidly resulting in the formation of the desired aerosol. The effect of solute concentration on dry particle size is shown in Figure 6 for several compressed air nebulizers (Mercer, et al., 1968).

In an endeavor to enhance production of a submicron particle for use in the inhalation experiment, a variety of modifications have been attempted. The Dautrebande aerosol generator is one such modification where the stream of nebulized droplets is made to impinge on a series of baffles enroute from the nebulizer. The larger droplets impact on the baffles while the smaller ones are swept out of the generator. The resulting volume median diameter of the droplet is smaller, resulting in a smaller initial diameter of the aerosol. The total output of the nebulizer is also reduced; consequently the particle number is reduced. A procedure similar to the Dautrebande nebulizer was tried using a DeVilbiss nebulizer and an Anderson impactor. The nebulizer was attached via tygon tubing to the bottom on the Anderson impactor; the sieve plates of the impactor were reversed in order and the nebulizer output was forced through the

Figure 6: Output characteristics of several compressed air nebulizers as a function of solute concentration (Mercer, 1968).

impactor. The output of this arrangement resulted in a smaller initial particle size and reduced number of particles.

Copious production of ultrafine particles of nichrome resulted from the use of a laboratory heat gun. While this method of aerosol production was the most desirable in terms of particle size and number, the heat added to the exposure chamber was undesirable and consequently the method was abandoned.

Aerosol Growth Kinetics

The choice of using a DeVilbiss (model unknown) nebulizer and low concentrations of sodium chloride solution was a compromise between the factors (a) particle size, (b) stability of particle growth, (c) particle numbers and (d) exposure chamber conditions. Solutions of sodium chloride with 7.46×10^{-5} gm of NaCl per ml H_2O and 7.46×10^{-7} gm of NaCl per ml H_2O were atomized into the exposure chamber. Growth kinetics of the aerosol were observed by particle counting techniques utilizing the Gardner Type CN condensation nuclei counter. Figure 7 shows the change in particle numbers with time after initial generation for NaCl particles and nichrome particles. The rapid change in particle numbers is attributed to the continuous and spontaneous coagulation of the aerosols (Green and Lane, 1964), and was described by Whytlaw-Gray (1932) as following the expression

$$\frac{1}{n} = \frac{1}{n_0} + Kt$$

where

n = Number of particles per cm^3 at any time t ,

n_0 = The number of particles present immediately upon generation

Figure 7: Decrease in particle count per cm³ as a function of time for two aerosols generated in the exposure chamber.

and

K is the coagulation constant with units of $\text{cm}^3/\text{sec}/\text{particle}$.

If the reciprocal of the particle numbers is plotted as a function of time a relation

$$\tau = \tau_0 + Kt$$

where

$$\tau = \frac{1}{n}$$

$$\tau_0 = \frac{1}{n_0}$$

produces a straight line for nearly all cases of particle production (Green and Lane, 1964). The fact that the aerosols generated in this experiment did not produce such a straight line, Figure 8, demonstrates according to Fuchs (1964), the polydisperse nature of the population and the rapidly changing character of the aerosols themselves. The graph approaches linearity as the particle distribution becomes more monodisperse.

The quantity τ has a definite physical significance, representing the average volume of a single particle in the air. (Green and Lane, 1964). the quantity τ is called the particulate volume, and n is referred to as the particle number.

The rapidity of combination of the condensation nuclei with the thoron progeny may clearly be seen by determining the half time of coagulation for a particulate system. From the expression,

$$\tau = \tau_0 + Kt$$

when

$$\tau = 2\tau_0,$$

this implies

$$t = \frac{\tau_0}{K} \quad \text{or} \quad t = \frac{1}{Kn_0},$$

Figure 8: Particulate volume $1/n$ as a function of time in the exposure chamber where n represents the number of particles per cubic centimeter.

K is of the order of magnitude 10^{-8} to 10^{-9} cm^3/sec (Green and Lane, 1964); n_0 for the aerosol-activity system is of the order 10^5 particles/ cm^3 , therefore time, t , is of the order of 15-20 minutes which agrees with the experimentally determined half time loss of particles in the chamber.

Particle Size Analysis

As previously stated the most important physical parameters affecting the fate of inhaled radon progeny are the aerosol characteristics. A list of the important properties of this characterization include: aerodynamic size, physical density, chemical composition, solubility, heterogeneity, particle number and the fraction of the activity that remains unattached from the aerosol.

The aerodynamic diameter of the inhaled radioactive aerosol, following the suggestion of Rabbe (1972), is defined to be the Equivalent Unit Density Sphere* as determined using diffusion tube techniques. Methods for measuring atmospheric aerosols by means of diffusion batteries have gained respectability since the early work of Townsend (1900). The derivation of the equation for diffusion of particulate matter from air stream flowing through a cylindrical tube, by Gormley and Kennedy (1949), and its subsequent application in many inhalation studies, (Chamberlain and Dyron, 1956; Altshuler, et al., 1969; Martin and Jacobi, 1972) as well as numerous studies of measurement of radioactive aerosols (Thomas, 1955; Fuchs, et al., 1962; Sinclair, 1972; George and

* The diameter of a sphere with unit density having the same settling velocity as the particle under investigation.

Hinchliffe, 1972; James, et al., 1972; George, 1972; Thomas and Hinchliffe, 1972 and Spurny and Lodge, 1972) has substantiated the use of the diffusion technique. Diffusion tubes are particularly useful for measurements in the size range below $0.1 \mu\text{m}$, (Thomas, 1955; Schumann and Petrausch, 1971) and hence are applicable to the study at hand. The aerosol to be measured is drawn through a relatively long diffusion tube or diffusion battery under conditions of laminar flow. The fraction of particles penetrating the length of the tube (battery) is described by the Gormley-Kennedy equation:

$$F = 0.18 \exp(-7.314h) + 0.0975 \exp(-44.6h) + 0.0325 \exp(-114.0h) + \dots$$

$$\text{when } h \geq 0.0156$$

$$F = 1.0 - 4.07^{2/3} + 2.4h + 0.446h^{4/3} + \dots$$

$$\text{when } h < 0.0156$$

$$h = \pi DL/2Q$$

$$D = \text{Diffusion coefficient of particle (cm}^2/\text{sec)}$$

$$L = \text{Length of diffusion tube (cm)}$$

$$Q = \text{Flow rate (cm}^3/\text{sec)}$$

The equations were derived for monodisperse aerosol distributions which are not found in nature. However, in the analysis of an actual atmosphere the equations may be used to determine an effective diffusion coefficient for the radioactive aerosol; this in turn is related to the aerodynamic diameter of a unit density sphere.

Figure 9 illustrates the theoretical diffusion battery penetration vs. particle diameter for unit density spheres as a function of sampling flow rate, determined using the Gormley-Kennedy equations.

Figure 9: Aerosol penetration through the diffusion battery as a function of particle size, for various flow rates in l/min.

The "cluster-tube", (Sinclair, 1969), diffusion battery was constructed of a bundle of 125 17ga. hypodermic stainless steel tubes 1.067 mm ID and 60 cm in length. The tubes were encased in a 1 inch OD by 60 cm stainless steel tube and the interstitial spaces were filled with cement so that the aerosol passed only through the hypodermic tubes. The flow through a single tube was 1/125th of the flow through the battery. A female collar on one end allowed the direct coupling of a 1 inch membrane filter holder to the battery. While the battery may be operated at any number of flow rates, flow rates between 1.5 l/min and 3 l/min produced the most consistent results.

Figure 10 illustrates the change of particle size in the inhalation chamber as measured using the diffusion battery. The particle agglomeration from the initial generation occurs in a predictable fashion thereby making it possible to choose an exposure time post generation which gives the desired particle size for inhalation. During the course of the exposure, diffusion battery samples were taken repeatedly, i.e. as often as sampling times and initial counting times would allow. While it was not possible to determine the log-normal distribution of particle size it was possible to determine the effective activity median aerodynamic diameter (AMAD) and geometric standard deviation, σ_g . Table 4 lists the mean, standard deviation and particle concentration in the exposure chamber at the time of each exposure.

Unattached Fraction

A long standing concern related to aerosol characterization of the uranium mine atmosphere is that fraction of the radium-A (^{218}Po) activity which remains unattached, that is, that activity which fails to become attached to the larger aerosol particles in the air, and is described

Figure 10: Growth of aerosol in the exposure chamber as a function of time post generation.

Table 4
Aerosol Characteristics

Experiment No.	Effective AMAD* (microns)	Geometric Deviation σ_g	Particle Concentration No./cm ³ (10 ⁴)
1	.033	1.06	1.32 ± 0.21
2	.068	1.01	4.41 ± .95
3A	.088	1.28	5.77 ± 1.24
3B	.073	1.01	5.18 ± 1.20

* AMAD is defined as the activity median aerodynamic diameter.
 σ_g is the geometric standard deviation and is a measure
of uniformity of size.

by some as a free ion. The maximum permissible concentration in the air (MPC_a) for radon as specified by the International Commission on Radiation Protection, (ICRP, 1964), includes the fraction of equilibrium amount of radium A atoms which are unattached to condensation nuclei as a part of its formulation. The formula given by the ICRP (1964) is:

$$MPC_a = \frac{3 \times 10^3}{(1+1,000f)} \text{ pCi } ^{222}\text{Rn per liter of air where } f \text{ is the}$$

unattached fraction of radium A atoms. The diffusion coefficient of the unattached radium A atom has been determined to be between $0.034 \text{ cm}^2/\text{sec}$ and $0.054 \text{ cm}^2/\text{sec}$ (Raabe, 1969; Altshuler, et al., 1964). Moreover Raabe (1969) arrived at a diffusion coefficient for a cluster of six water molecules around a radium A atom (or a polonium oxide molecule), using kinetic theory, of $0.054 \text{ cm}^2/\text{sec}$. This corresponds to a unit density sphere with a diameter of approximately $0.001 \text{ } \mu\text{m}$ (Billings and Gussman, Inc., 1972).

A number of studies have been undertaken to measure the unattached fraction of radium A atoms in a real or simulated uranium mine atmosphere (Fusamura and Kurosawa, 1967; George and Hinchliffe, 1972; James, et al., 1972; Craft, et al., 1966; McLaughlin, 1972; Mercer, 1969 and George, 1972). The method chosen in this study which is most compatible with the sampling technique used in the experiment is based upon the collection efficiency of a wire screen (James, et al., 1972; George, 1972; Thomas and Hinchliffe, 1972). The procedure is simply to sample the desired atmosphere by drawing air through a wire screen (60 mesh) at an appropriate flow rate (about 2 l/min). Behind the screen, and separated from the screen by an "O" ring, a membrane filter, type GA, $0.45 \text{ } \mu\text{m}$ pore, is located. A reference sample is drawn using a membrane filter without

the screen. The unattached radium A atoms are collected with good efficiency, about 80% for 60 mesh screen (George, 1972), and the difference between the activity on the screen filter and the reference filter is a measure of the unattached fraction of activity. The zero efficiency for condensation nuclei and appreciable efficiency for free ions is taken as sufficient confirmation that the wire screen may be used to determine the unattached fraction of the activity in the air.

Assessment of Potential Hazard to the Individual

Emphasis over the last few years has been to reduce the concentrations of radioactivity in the uranium mines to a level which produces the least possible risk to the miner. The most recent standard, ANSI 13.8 (1973), entitled "Radiation Protection in Uranium Mines" follows the standard set a few years ago of "...the cumulative exposure guides for radon daughters shall be 4 work level months per year." The standard further states, "In a few exceptional mines where thorium ore is present, it is possible that thoron daughter concentrations may be significant and the operator should be aware of this possible problem" (page 17). The "working level month" (WLM) is that special unit of cumulative exposure obtained by the product of working levels and the time of exposure, e.g. a miner working in a 0.1 WL atmosphere for 1 month (173 hours) will have a cumulative exposure of 0.1 WLM. The previously established precedent of maintaining experimental exposures at one-tenth of the occupational exposure has been adopted for this experiment. There are no epidemiological studies for thoron to provide evidence of an exposure-risk relationship, (Duggan, 1973). The potential hazard from inhalation of thoron and

thoron progeny has not been established but it may be a factor of 10 lower than the potential hazard from radon progeny (Harley and Pasternack, 1972).

Growth and Decay of Thoron Progeny in situ

Measurements of activity in the exposure chamber indicated that at the time of exposure air concentrations of ThB and ThC existed in a ratio of approximately 3:1. In addition, at the time of exposure, i.e. within 30 minutes of the end of thoron generation time, the ^{208}Tl (ThC") existed in such a low concentration, that any inhaled quantity of the nuclide would decay before the subject could be counted. As the ThB and ThC decay through ^{208}Tl , the 2.62 MeV gamma from the ^{208}Tl will be detected using the 4" by 5" sodium iodide detector. The early counts in the 2.62 MeV peak stem from the decay of the initial deposits of ThC. Later counts, indicating an increase in ^{208}Tl activity must be the resulting ingrowth from the ^{212}Pb deposited during inhalation. Figure 11 shows the ingrowth of ^{208}Tl when an initial deposit of 3:1 ThB to ThC occurs. Since ThC decays 35% of the time to ^{208}Tl the effective alpha decay constant for ThC may be derived using the branching ratio (Segre, 1965).

$$\begin{aligned}\frac{\lambda_B}{\lambda_A} &= \frac{65}{35} \\ \frac{\lambda_B + \lambda_A}{\lambda_A} &= \frac{65 + 35}{35} \\ \lambda_A &= \frac{(.6874)(35)}{100} \\ \lambda_A &= .2406\end{aligned}$$

Using the alpha decay constant of $.2406 \text{ hr}^{-1}$ for ^{212}Bi the appropriate growth and decay curve, Figure 11, may be generated for ^{208}Tl .

Figure 11: Build up and decay of ^{208}Tl from in situ decay of ThB and ThC; ThB:TC = 3:1.

Working Level Exposures of the Experimental Subject

Air samples taken during the course of each inhalation experiment were used to determine the working level exposure to the experimental subjects. Table 5 summarizes those exposures.

In Vivo Measurements of Inhaled Activity

In vivo detection of inhaled activity was performed using a shadow shield whole body counter and linear scanning techniques. The shadow shield counter, similar to the one developed and used by Hanford laboratories (Palmer and Roesch, 1965) as a mobile whole body counter, was constructed at Colorado State University and used in prior studies of deposition of radon progeny in uranium miners (Holleman and Schiager, 1967). Figure 12 illustrates the positioning of the subject to be counted under the counter. The subject reclines on a bed mechanism which can be moved beneath the 4" x 5" NaI (Tl) crystal at a constant rate*. The pulse-height analyzer used in this study was equipped with an MCS, i.e., multi-channel scaling mode of operation. The MCS mode stores all the counts detected in one channel for a pre-set time (dwell time) before moving to the next channel. Consequently counts recorded above one region of the body may be stored in one channel of the pulse-height analyzer. As the subject is moved beneath the crystal the counts are stored in succession, in successive channels of the pulse-height analyzer. The linear scan speed was adjusted with a variable speed drive mechanism so that the scan from the top of the head to the gut region was accomplished at $1.06 \pm 7\%$ cm per channel of MCS scan.**

* Total distance of approximately 67 cm in 50-60 minutes.

** MSC mode of operation of the Ortec 6220 multichannel analyzer with dwell time of 0.8 min obtained from external time source.

Table 5

Average Chamber Conditions and Exposures for the Experimental Subjects

Subject	Chamber Concentration (pCi/l)			Exposure Duration (min)	Exposure WLM
	ThB	ThC	Rn		
DRA	873	375	359	53	.585 *
AWA	873	375	359	53	.585 *
STB	650	245	236	50	.408
RAF	650	245	236	50	.408
DP	649	237	231	30	.244
KJS	649	237	231	30	.244

* Exposures in excess of the anticipated exposures are explained in Chapter VI under the section titled Assessing the Hazard to the Experimental Subject.

Figure 12: Position of the subject to be counted, using the shadow shield whole body counter. Top figure illustrates the position of experimental subject and counter in end view. The bottom figure illustrates a side view of the detector and the anatomical positions assumed.

Lead bricks were used to build a four-inch thick shield around the crystal photomultiplier tube to shield the detector from activity originating outside the system. A tapered hole lead collimator was used to effect a reasonable balance between sensitivity of the system and spatial resolution. Figure 13 shows the response curve obtained by scanning a point source located in a plywood phantom at the same scan speed used with the experimental subjects. For the scanning arrangement used in the experiment, the resolution is to a first approximation, proportional to the aperture diameter, while the sensitivity of detection is proportional to the square of the aperture diameter (IAEA, 1971). The resolution of the collimated detector, defined as the full width at half maximum, (FWHM), of the response curve was determined to be between 12-17 centimeters of linear scan (Brownell, 1958).

Care must be taken to maintain, as nearly as possible, constant geometry of the linear scans. The counting rate varies rapidly with depth of activity in the subjects (IAEA, 1971) and may vary as much as 8% for high energy gamma activity and up to 25% for low energy gamma-ray activity (Marinelli, et al., 1955; 1962).

The sensitivity of a single detector with a single-slit collimator varies inversely as the source-to-detector distance, as opposed to the inverse square of this distance (IAEA, 1971), for the cylindrical hole collimator used in this work.

Quantitative determination of activity deposited in the experimental subject is exceedingly difficult to obtain due to distribution of the small quantities inhaled over a comparatively large region, i.e. head, throat, lung and gut, use of a calibration phantom facilitates the estimation of deposition amounts. The activity in the experimental subject was compared

Figure 13: Response curve obtained by scanning a fixed thorium-228 source located in the plywood phantom.

with an activity distribution in a plywood phantom when both were counted using linear scanning techniques. The plywood phantom was fitted with sealed sources of low concentrations of thorium-228. When these discrete sources were scanned, a much sharper profile was obtained than when the experimental subject was scanned. Nevertheless, a comparison of activities is possible. When considering the quantitation of the results of a linear scan, it is necessary to consider not only the resolution and sensitivity of the detection system but also the variation of response to location of activity in different anatomical regions of the body (Tothill, 1971). The results of the linear scan are displayed as a plot of counts per channel as a function of distance or channel number. The experimenter, by knowing the scan speed and the dwell time per channel, may correlate the scan with an anatomical position of the experimental subject. A peak in the count rate occurs when the crystal passes over a region of increased activity. The area under the peak may be related to the activity in the region when compared with a similar scan over a known amount of activity in the phantom.

Care must be exercised to determine the contribution of activity located in an adjacent region of the subject to the count rate recorded when the detector is above another region. This problem was solved by repeating the linear scan procedure with a reference source located successively in each of the regions, N-P, T-B, pulmonary and gut. The resulting scans may then be used to formulate a matrix relating the count rate of one region with the activity in an adjacent region (Miller, 1964). In all, the measurement technique selected seeks to maximize the response to activity directly beneath the detector and minimize the

contribution of activity external to the subject and to that located in other regions of the subject.

Linear scans were run on the experimental subjects at intervals of two to three hours for 36 to 48 hours following exposure. The scans were used to determine the biological half time of activity deposited in the regions of interest, as well as the total and regional deposition values. In addition to the linear scanning, in vivo counting used in one experiment was performed using a scan and a stationary counting procedure. The stationary procedure was executed by locating the subject in fixed position below the crystal. Counting positions were defined and ten minute counts were made over each of the following.

- (1) The bridge of the nose, representing the N-P region
- (2) The sternal notch, representing the T-B region.
- (3) 10 cm right or left of the lower end of the sternum,
representing the pulmonary region.
- (4) Directly above the navel, representing the gut region and
- (5) With the subject lying on his stomach, the region above the
small of the back, representing the kidney.

Chapter IV

DEPOSITION OF INHALED RADIOACTIVITY

Deposition is the end result of the processes that determine what fraction of the inhaled airborne material will be caught in the respiratory track. Earlier works, particularly the work of Holleman, et al. (1968), have shown a definite relation between the respiratory rate and the percent of inhaled aerosol which is deposited in the respiratory tract. Deposition has been shown to increase almost linearly from lower to slightly higher than normal respiratory frequencies. In an endeavor to measure the deposition patterns in the experimental subject, the subject was asked to enter the exposure chamber where an atmosphere of thoron (^{220}Rn) progeny and sodium chloride aerosol had previously been generated (Chapter III). The subject was requested to breathe as normally as possible, either through the nose or mouth. In order to suppress the subject's conscious effort at breathing, each was given a small task to perform, e.g. measurement and recording the aerosol particle count using the Gardner Small Particle Detector or sampling the exposure atmosphere for the presence of radon-222. Table 6 shows the complete set of atmospheric conditions present in the chamber at the time of exposure. The experimental subjects were exposed for 30 or 50 minutes and following exposure changed clothes, washed any body area that might have surface contamination, (only hands and face) and presented himself to be counted in the shadow shield whole body counter.

In the analysis, the MCS linear scan counts per channel were smoothed using a seven point least squares smoothing routine, (Savitzky and Golay, 1964). The smoothed points were summed into 5 channel groups representing 2 minute or 4 minute time periods,

Table 6

Summary of Chamber Conditions for Each Experiment

Experiment No.	Radioactivity Concentrations			Particle Size μm	Relative Humidity	Temperature °C	Working Level	Working Level Month
	ThB	pCi/l ThC	Rn					
1	649	237	231	.033	46%	27°C	84.37	.244
2	650	245	236	.068	49%	20°C	84.64	.408
3	358	115	165	.088	51%	25°C	46.74	.225
4	873	375	359	.073	40%	25°C	114.63	.585

spanning $5.54 \pm .60$ cm or $5.09 \pm .12$ cm respectively. Figure 14 shows a typical scan following the data smoothing and summation routine. These data, representing the deposited ^{212}Pb as measured using the 2.62 MeV gamma from ^{208}Tl , were further corrected to account for the fact that the ingrowth of ^{208}Tl lags behind the decay of the ^{212}Pb . Figure 11 (Chapter III) indicates the growth and decay of the ^{208}Tl in situ.

The post-exposure count rates were used to determine the total and regional deposition values by comparison with linear scans of a plywood phantom with known amounts of thorium-228 nitrate solution located in discrete regions of the phantom, i.e. in the nose, tracheo-bronchial, pulmonary and gut regions. These data were compiled into a matrix following the manner of Miller, (1964) and used to determine that fraction of the airborne activity that was deposited in the respiratory tract. Earlier work done by Holleman (1968) indicated an average deposition fraction of 46% for aerosols with an effective diffusion coefficient of $1.3 \times 10^{-5} \text{ cm}^2/\text{sec}$ while the work of Booker, et al. (1969) found deposition fractions between 34 and 60% for thoron progeny combined with nichrome aerosol in a size range between $0.05 \mu\text{m}$ and $0.5 \mu\text{m}$. Deposition values near 100% were observed for activity combined with ambient room aerosols.

The ICRP (1966), gives theoretical values between 85% and 55% for particles in a size range from $0.01 \mu\text{m}$ to $0.1 \mu\text{m}$ respectively. The value of 68% for $.033 \mu\text{m}$ particles is higher than the one obtained by Holleman (1968) and more nearly approaches the value given by the ICRP, Task Group on Lung Dynamics (1966). Total deposition following inhalation of a $0.073 \mu\text{m}$ aerosol was determined to be 36% for subjects using a combination of nose and mouth breathing. This agrees closely

Figure 14: Typical linear scan of experimental subject after smoothing of the data.

with the work of Holleman for the same respiratory pattern; the value is lower than that of the ICRP Task Group but does follow the trend of a decrease in total deposition in this aerosol size range. While six experimental subjects were exposed, only the data from four were suitable for analysis due to a malfunction in the timing circuit used to control the MCS mode. Table 7 summarizes the total and regional deposition of the 4 experimental subjects.

Regional Deposition

The regional deposition of thoron progeny in the respiratory tract was determined as a percent of the total deposited activity found in each of the four previously mentioned anatomical regions. A significant fraction of the activity was determined to be present in the stomach-gut region. The longer inhalation period, (30 to 60 minutes), allowed a portion of the activity to be swallowed and subsequently detected in the stomach-gut region. The relation between the distance scanned per unit of time and the locations of the anatomical region of interest provided the necessary parameter for determining the activity in a region, i.e. the counts recorded over a finite length of linear scan is a measure of the activity deposited in that anatomical region. Again a matrix solution technique was used to distinguish the activity in one region from the contribution outside the region.

A seemingly clear relation exists between the particle size and total deposition of inhaled radioactive aerosol. The ranges of regional deposition percentages for the anatomical regions measured follow the general pattern as given by the ICRP (1966). They are 2.7 - 12.4%, 7.4 - 20.4%, 46.2 - 61.0% for the nasopharynx, tracheobronchial and pulmonary regions respectively. The ICRP does not report any deposition values for the gut region.

Table 7

Total and Regional Deposition Value for Four Experimental Subjects

Subject	Particle Size (microns)	Total Deposition (%)	Regional Deposition %				Inhalation Mode *
			N-P	T-B	P	Gut	
DRA	.073	36	4.2	18.6	52.4	24.6	N-M
AWA	.073	36	2.7	7.4	61.0	29.3	N-M
DP	.033	68	12.4	20.4	46.2	20.4	N
KJS	.033	68	9.6	15.0	55.0	20.2	M

*Inhalation Mode:

N-M - combination of nose and mouth

N - nose

M -mouth

Chapter V

TRANSLOCATION OF DEPOSITED ACTIVITY

Measurement of translocation of deposited radioactivity in the respiratory tract presents the severest test of the in vivo detection method. Following deposition, the activity may be subjected to one of several dynamic mechanisms ultimately at work to remove foreign particles from the lining of the respiratory tract. The mechanisms primarily in operation are the mucous escalator system, phagocytic action, and dissolution of the aerosol followed by adsorption into the blood or lymph system. This movement, coupled with the small deposition activities and poor counting geometry of the detection method was considered by Veall (1969), as sufficient to negate any attempt at accurately measuring translocation rates. More sophisticated analysis using convolution techniques have been used to unfold the linear scan (Abu-Shumays and Marinelli, 1971; Iinuma and Nagai, 1967). Results as accurate as $\pm 3\%$ have been obtained when all the parameters, i.e. crystal resolution, activity distribution, detector collimation and statistical fluctuations were properly chosen (Clemente, et al., 1970). In vivo studies do not allow control of all of the parameters, but sufficient control can be exercised to allow quantitative measurement of the translocation of deposited radioactive aerosols from the respiratory system.

The linear scan data, following smoothing and distribution corrections, (Chapter III), were fitted with exponential regression curves in order to determine the effective decay constant for the deposited activity (Albert and Arnett, 1955). The effective decay constant was used to determine the biological decay constant from the relationship

$\lambda_{\text{effective}} = \lambda_{\text{biological}} + \lambda_{\text{physical}}$. It becomes apparent as one evaluates the data that while a reliable measure of translocation of activity from the respiratory tract may be made by determining the effective decay constant, the use of the effective half life to determine the biological half life is plagued by the inherent difficulty of comparing the reciprocals of two numbers whose difference is small. Since,

$$\lambda_{\text{eff}} = \lambda_{\text{physical}} + \lambda_{\text{biological}} \quad \text{where} \quad \lambda = \frac{\ln 2}{T_{\frac{1}{2}}}$$

this implies

$$\frac{\ln 2}{T_{\text{eff}}} = \frac{\ln 2}{T_{\text{physical}}} + \frac{\ln 2}{T_{\text{biological}}}$$

Therefore,

$$\frac{1}{T_{\text{effective}}} = \frac{1}{T_{\text{physical}}} + \frac{1}{T_{\text{biological}}}$$

or

$$\frac{1}{T_{\text{biological}}} = \frac{1}{T_{\text{effective}}} - \frac{1}{T_{\text{physical}}}$$

which allows the determination of biological half life.

The physical half life of the controlling nuclide, ^{212}Pb , is 10.64 hours; regression analysis resulted in a range of effective half lives from a low of 6.89 hours to a high of 9.41 hours. The closer the effective half life is to 10.64 hours the larger the biological half life appears to be. Consequently small differences in the measurements of the effective half life result in exaggerated values of the biological half life.

Effect of Particle Size

The relation of particle size to effective removal of radioactivity from the respiratory tract is shown in Figures 15 and 16 where the least

Figure 15: Fraction of initially deposited activity of the 0.033 μm and 0.073 μm aerosol deposited in the tracheobronchial region of the respiratory tract as a function of time post exposure measured in hours.

Figure 16: Fraction of initially deposited activity of the 0.033 μm and 0.073 μm aerosol deposited in the pulmonary region of the respiratory tract as a function of time post exposure measured in hours.

square regression lines, significant at an alpha level of less than 0.05, indicate a difference in λ for the tracheobronchial region and the pulmonary region respectively. For the pulmonary region, particles with λ of 0.0772 hr^{-1} give an effective half life of 8.98 hours and biological half life of 57.56 hours. The effective half life of aerosols with $0.073 \mu\text{m}$ AMAD and λ of 0.1006 hr^{-1} is 6.89 hours yielding a biological half life of 19.57 hours. Figure 17 shows a comparable value resulting from a third experiment in which only values from the pulmonary region were obtained. These data indicate an effective half life of 6.94 hours and biological half life of 20.00 hours from particles with $0.088 \mu\text{m}$ AMAD. Again the regression is significant at the alpha level of 0.05.

Regression lines depicting translocation in the T-B region yielded comparable results with the larger particles ($0.073 \mu\text{m}$ AMAD) clearing the region with an effective half-time of 8.35 hours as compared with the 9.41 hour half-time of the $0.033 \mu\text{m}$ particles. Biological half-times compare at 38.73 hours and 81.09 hours respectively.

The linear scan technique used to determine the translocation times failed to reveal a rapid component of removal of activity from the anatomical regions. In addition, the removal of activity deposited in the nasopharynx region is at such a high rate as to be undetectable using this measurement technique. Regression analysis of the fraction of activity seen in the gut region for experimental subjects DRA and AWA, inhaling an aerosol with AMAD $0.073 \mu\text{m}$, gave an effective decay half life of 7.21 hours and biological half life of 22.40 hours. In contrast to these values, the regression analysis of experimental subjects DP and KJS, inhaling an aerosol with AMAD $0.033 \mu\text{m}$, gave an effective half life in excess of 10.64 hours.

Figure 17: Fraction of initially deposited activity in the pulmonary region for a 0.088 μm aerosol as a function of time post exposure measured in hours.

Chapter VI

SUMMARY AND CONCLUSIONS

An inhalation atmosphere was generated using a compressed air nebulizer technique to produce NaCl particles with a minimum initial diameter of $.007 \mu\text{m}$ as determined from the effective diffusion coefficient of $1.06 \times 10^{-3} \text{ cm}^2/\text{sec}$. These data were determined by mixing the aerosol with a radioactive atmosphere generated using a thorium 228 solution in the form of thorium nitrate in a gas de-emanation flask. Radon 220 (emanation thoron) was produced in the inhalation chamber by bubbling the solution with dry nitrogen from a high pressure nitrogen tank. The radon progeny, ^{212}Pb and ^{212}Bi , resulting from the decay of the radon 220 and polonium 216 in the chamber agglomerate rapidly with the sodium chloride aerosol. The aerosol size and radioactivity were determined using a "cluster-type" diffusion battery to measure the effective diffusion coefficient from which the activity median aerodynamic diameter, (AMAD), of the aerosol was determined. Particle growth of the aerosol from the initial size distribution followed a well defined pattern (Figure 10, Chapter III) which made it possible to choose a post generation inhalation time when the inhalation atmosphere would contain the desired particle size.

The fraction of the ThB and the ThC in the chamber remaining in the unattached state was measured using the wire screen method of A.C. George (1972). For the duration of the experiment, which included trial runs for generating the radioactive aerosol, checking the detection equipment, and performing the experiment, the values of the unattached fraction ranged from less than 0.1% to a high of 15%. The variability in any single experimental trial, over a period of several hours measurement time was from 25% to near 100%. The average unattached fraction in the

particle size range .020 - .040 μm was $.056 \pm 74\%$ and in the range .068 - .088 μm was $.036 \pm 58\%$. The ThB and the ThC concentrations were determined by measuring the activity on a 0.45 μm pore filter through which a sample of the inhalation chamber air was drawn. Activity measurements were made by alpha spectroscopy using silicon surface barrier detectors. The ratio of ThB to ThC in the chamber was determined to be of the order of 3:1 at the time of each inhalation experiment.

In Vivo Detection

In vivo detection of inhaled radioactivity was carried out using a shadow shield whole body counter equipped with a 5" by 4" NaI(Tl) detector. The 2.62 MeV gamma emitted from ThC" was used to measure the deposition and translocation of the ThB and ThC deposited in the respiratory tract during the inhalation experiment. The data collected using a linear scanning technique and multi-channel scaling on the Ortec analyzer was corrected for build up and decay of the ThC" and distribution of activity in the subject. These data were used to determine total deposition in the subject and regional deposition in the anatomical regions, nasopharynx, tracheobronchial, pulmonary and gut.

In vitro radioactivity measurements as normally performed in the laboratory consist of a comparison of the activity in a sample with the activity in a standard of the same geometry. It is therefore comparatively easy to make such comparisons.

In contrast in vivo detection of radioactivity in an experimental subject becomes a more difficult problem since the radiation field from the subject cannot be duplicated in a phantom. Consequently it becomes more difficult to insure that the sample and standard are counted using the same geometry. In addition, translocation of deposited radioactivity

with time is impossible to simulate in the phantom, consequently static measurements are used when a dynamic problem is in actuality the case. As one increases resolution of the detector the sensitivity of detection will inherently decrease, and some compromise between the two must be reached. Inherently a three-dimensional activity distribution is being evaluated using a two-dimensional technique. Nevertheless, the linear scan measurements utilized in this work yielded successful evaluations of total body deposition and translocation of inhaled thoron progeny. Knowledge of the anatomy of the respiratory tract makes it possible to relate a scan position with a respiratory tract location and aided in simulating as nearly as possible a calibration phantom.

Deposition of Inhaled Radioactivity

Measurements were made of total and regional deposition of thoron progeny in the respiratory system and gut of four experimental subjects. The results indicated an inverse relation between total deposition and particle size. Sixty-eight percent of the activity inhaled with AMAD of $0.033 \mu\text{m}$ was retained in the body while 36% of the activity associated with particles with AMAD of $0.073 \mu\text{m}$ was deposited in the body. The regional deposition values, indicate an average greater deposition percent for smaller particles in the tracheobronchial and nasopharynx region and slightly lower deposition percentages in the pulmonary and gut regions. Significant fractions of the inhaled activity appeared in the gut regions, this may be attributed to the activity deposited in the upper respiratory tract being swallowed, at the same time the activity deposited in the ciliated portions of the respiratory tract is being transported upward and swallowed. The lower deposition values of 4.2 percent and

2.7 percent in the N-P region for the two subjects using a combination of nose and mouth respiration may indicate a preference for mouth breathing as the experimental subjects carried out the task assigned to them during the course of the experiment. The one subject using nose breathing demonstrated a higher deposition value in the upper respiratory tract than did the subject using mouth breathing alone. Average regional deposition values for all four subjects of 7.23 ± 4.5 , 15.35 ± 5.76 , 53.65 ± 6.13 and 23.63 ± 4.29 for the four anatomical regions, nasopharynx, tracheobronchial, pulmonary and gut, indicate the general pattern of deposition predicted by the ICRP, (1966).

Translocation of Thoron Progeny

This work has measured the translocation of thoron progeny deposited in the respiratory tract for four separate individuals involved in three experiments using three different particle sizes. The size and nature of the aerosol used directly effects the translocation of the radioactivity from the respiratory tract. The effective clearance rates (physical + biological) of from 6.89 to 9.41 hours indicate biological rates of 19.57 to 81.09 hours, with the larger particles clearing at a faster rate than the smaller ones. The clearance rates for the pulmonary region are faster by a factor of almost three for the $0.073 \mu\text{m}$ particles over the $0.33 \mu\text{m}$ particles. Measured translocation of activity in the tracheobronchial region is clouded by the fact that the activity in that region may be continually replaced by activity from lower lung regions. Consequently the measured effective half-times of 8.35 hours and 9.41 hours may reflect an in situ physical decay. The regression on tracheobronchial data seems to indicate that the larger particles are cleared faster than the smaller ones, which is consistent with the regression on the pulmonary

data and the gut data. It does not appear to be feasible to assume that the mass of material deposited in the tracheobronchial region would place such a load on the mucosa as to cause the mucous escalator to be less effective (Hatch and Gross, 1964), yet the indication is there that the activity does remain in the tracheobronchial region for a longer period of time.

The data obtained for the gut tends to support the data obtained for the pulmonary region for the $0.073 \mu\text{m}$ aerosol. If we consider the pulmonary data to represent one end of the system and the gut the other, with the tracheobronchial region in the middle it would appear that measurements in the T-B region do indeed reflect more physical decay than translocation. With this thought in mind it appears plausible to conclude, that the development of carcinomas in the tracheobronchial region is enhanced as much by the passage of radioactive material through the region, as by deposition of particulate matter in the region.

It is possible that the clearance of radioactivity from the pulmonary region is enhanced by other mechanisms of the deep lung region that are not at work in the ciliated region. Specifically the mechanism of dissolution and adsorption would appear to be an important factor since the aerosol used is highly soluble in body tissue fluid. Hurst, et al., (1969) reported that ^{212}Pb attached to natural aerosol was cleared from the lung with an estimated half-time of 6.5 hours. The Hurst, et al. measurement relates only to total disappearance of activity from the lung and was measured using stationary counting over the chest. Although not specifically stated as such the 6.5 hour effective half-time would yield a biological clearance half time of 16.7 hours which compares favorably with the half times determined for the aerosols used in this study.

It should be noted that due to the method used to determine biological rates, i.e. from the effective decay constant, small differences in effective half times result in larger differences in calculated biological rates. Table 8 summarizes the effective decay constants and half-times for the aerosols of the experiment.

Assessing the Hazard to the Experimental Subject

The total lung dose to the worker in a uranium mine or mill has been estimated to be 0.8 rads/WLM (Holleman, 1968) associated with inhalation of the average equilibrium concentrations of 2700 pCi/WL of radium-A and 14,000 pCi/WL of radium C' as determined in an active uranium mining complex. The occupational dose received from this exposure was determined by Holleman (1968) to be 9.6 rem/year, the recommendation of the ICRP (1964) is 15 rem/year to the lung.

There have been few studies of the risk involved in inhalation of concentrations of thoron (^{220}Rn) and thoron progeny (e.g. ^{212}Pb and ^{212}Bi). Of the works done, Jacobi (1972) and Duggan (1973), it has been concluded that an acceptable level of thoron may justifiably be ten times as great as the acceptable level for radon. A comparison of the dose estimates for radon-222 and radon-220 along with a derivation of the exposure limits for inhaled radon-220 decay products is shown in Table 9, these estimates were used to derive what was considered by this investigator to be reasonable and safe limits of exposure for the experimental subjects.

Pre and post experiment cytological examinations of the sputum of the experimental subjects were performed by Dr. Geno Saccomanno of St. Mary's Hospital, Grand Junction, Colorado, with no abnormal cell differentiation appearing in the experimental subjects. The experimental procedure was reviewed by the Colorado State University Committee on

Table 8
Summary of Translocation Values

	Decay Constant hr^{-1}	Effective Half Life hr	Biological Half Life hr	Effective Particle Size (μm)
Pulmonary	0.772	8.98	57.56	.033
	.1006	6.89	19.57	0.073
	.0999	6.94	20.00	0.088
Tracheobronchial	.0737	9.41	81.09	.033
	.0830	8.35	38.73	0.073
Gut	.0961	7.21	22.40	0.073

Table 9
Derivation of Exposure Limit for Inhaled Thoron Decay Products

	^{222}Rn (radon)	^{220}Rn (thoron)
Range of Dose Estimates (rad/WLM)	0.07 - 1.0 ^a 0.23 - 1.6 ^c	0.004 - 0.1 ^a 0.06 ^b
CSU Best Estimate (rad/WLM)	0.8 ^d	0.02 ^d
Occupational Exposure Limit (WLM/year)	4	none
Occupational Dose Limit for Lung (rem/year)	$\left(0.8 \frac{\text{WLM}}{\text{rad}}\right) \left(3 \frac{\text{rem}}{\text{rad}}\right) \left(4 \frac{\text{WLM}}{\text{year}}\right) = 9.6$	
	ICRP: 15	15
Calculated WLM Limit	N/A	25 ^e
Limit for Experimental Subject (WLM/year)	0.4 ^f	2.5 ^f

- a. Mean dose per WLM to the tracheobronchial region and pulmonary region estimated from inhalation of radon and thoron in varying aerosol concentrations (Jacobi, 1972).
- b. Continuous inhalation of thoron at 100 pCi/l in an occupational exposure area (Duggan, 1973).
- c. The dose calculated depends upon the lung model assumed and reference atmosphere selected. The dose calculations reflected in these values are a result of using the reference atmosphere of Altshuler and lung models of Landahl and Weibel (Harley and Pasternack, 1972).
- d. Based on work done at CSU (Holleman, et al., 1969, Holleman, 1968).
- e. The ICRP recommendation of 15 rem/year occupational dose was used to estimate a WLM limit for thoron exposure.

Dose in rem = dose in rad x Quality Factor

A quality factor of three (3) has been accepted for particles in the lung (NIOSH, 1971)

$$\frac{15 \text{ rem/year}}{(3 \text{ rem/rad}) (0.2 \text{ rad/WLM})} = 25 \frac{\text{WLM}}{\text{year}}$$

- f. Based upon policy of CSU Radiation Safety Committee to limit the dose to experimental subjects to 10% of the occupational dose limit.

Human Research and again at the state level by a sub committee of the Colorado Department of Health, Radiation Hygiene Section. A copy of the consent statement and exposure record which was used to document the exposure is reproduced as Figure 18.

One experiment resulted in an exposure to the subjects of 0.585 WLM which was 46% over the desired level. The discrepancy came about as a result of underestimating the thoron generated in the chamber. In view of the low percentage of radioactivity deposited in the respiratory tract in this experiment, as compared with the radioactivity deposited in the respiratory tract from the experiment when the exposure level was 0.244, the over exposure certainly would produce no increased hazard.

CONSENT STATEMENT AND EXPOSURE RECORD
for experimental inhalation of radon and
radon daughter products.

Full Name of Subject: _____

Soc. Sec. No.: _____ Date of Birth: _____

I hereby consent to serve as a subject in the RADON PROGENY INHALATION STUDY.

The nature and general purpose of the experimental procedure and the known risks involved have been explained to me by Dr. Keith J. Schiager. He is authorized to proceed on the understanding that I may terminate my service as a subject in this research at any time I so desire.

I understand that the potential risk of inhalation of radon (^{222}Rn) or thoron (^{220}Rn) and their decay products is the induction of lung cancer. I also understand that the maximum exposure involved in this experiment is one-tenth of the annual occupational exposure limit, and that the available epidemiological evidence indicates that the risk of lung cancer induction is minimal even for cumulative exposures for 30 years at the occupational exposure limit.

I understand also that it is not possible to identify all potential risks in an experimental procedure, and I believe that reasonable safeguards have been taken to minimize both the known and the potential but unknown risks.

Subject's signature: _____ Date: _____

Witnessed by: _____ Signature: _____
(typed name and position)

EXPOSURE RECORD

Isotope	Chamber Concentration (pCi/l)	Exposure Duration (min)	Exposure (W.L.-hr)	Exposure (Rads)	Percent M.P.E.*	Accumulated %M.P.E.	Date	Investigators Initial

Signature of Investigator

Signature of Principal Investigator

Figure 18: Consent statement and exposure record.

BIBLIOGRAPHY

- Albert, Roy E. and Lawrence C. Arnett, "Clearance of Radioactive Dust from the Human Lung". AMA Archives of Industrial Health. 1956.
- Altshuler, B., N. Nelson and M. Kuschner, "Estimation of the Lung Dose from the Inhalation of Radon and Daughters". Health Physics. V. 10. pp. 1137-1161. 1964
- ANSI. N 13.8. (American National Standards Institute, "Radiation Protection in Uranium Mines". New York. 1973.
- Bale, W.F. and J.V. Shapiro, "Radiation Dosage to the Lungs from Radon and its Daughter Products". Proceedings of the U. N. International Conference on Peaceful Uses of Atomic Energy. V. 13. pp. 233-236. United Nations. New York. 1956.
- Billings and Gussman Inc., "Tables for Use in Aerosol Physics: Unit Density Spheres". Waltham, Mass. 1971.
- Booker, D.V., A.C. Chamberlain, D. Newton and A.N.B. Scott, "Uptake of Radioactive Lead Following Inhalation and Injection". British Journal of Radiology. V. 42. pp. 457-466. 1969.
- Brieger, Heinrich, "The Fate of Inhaled Particles". AMA Archives of Environmental Health. V. 6. pp. 57-60. 1963.
- Brownell, G.L., "The Theory of Radioisotope Scanning". International Journal of Applied Radiation and Isotopes. V. 3. pp. 181-192. 1958.
- Chamberlain, A.C. and E.D. Dyson, "The Dose to the Trachea and Bronchi from the Decay Products of Radon and Thoron". British Journal of Radiology. V. 29. pp. 317-325. 1956.
- Clemente, G.F., L.D. Marinelli and I.K. Abu-Shumays, "Regularization Unfolding in Low Gamma Activity Measurements". International Journal of Applied Radiation and Isotopes. V. 21. pp. 307-318. 1970.
- Craft, Bob F., James L. Oser and F. Warren Norris, "A Method for Determining Relative Amounts of combined and uncombined Radon Daughter Activity in Underground Uranium Mines". American Industrial Hygiene Association Journal. V. 27. pp. 154-159. 1966.

- Drew, Robert T. and Morton Lippmann, "Calibration of Test Atmospheres for Instrument Calibration" in *Air Sampline Instruments for Evaluation of Atmospheric Contaminants*. 4th Edition. American Conference of Governmental Industrial Hygienists. Ohio. 1972
- Duggan, M.J., "Some Aspects of the Hazard from Airborne Thoron". *Health Physics*. V. 24. pp. 301-310. 1973.
- Federal Radiation Council, "Guidance for the Control of Radiation Hazards in Uranium Mining". Report #8, Revised. U.S. Government Printing Office. Washington, D.C. 1967.
- Findeisen, W., "Über das Absetzen Kleiner in der Luft Suspendierten Teilchen in der Menschlichen Lunge bei der Atmung". *Archives Ges. Physiology*. p. 236. 1935.
- Fuchs, N.A. and I.B. Stechkina, "Resistance of a Gaseous Medium to the Motion of a Spherical Particle of a Size Comparable to the Mean Free Path of the Gas Molecule". *Faraday Society of London. Transactions*. V. 58. p. 1942. 1962.
- Fuchs, N.A., I.B. Stechkina and V.I. Starosselskii, "On the Determination of Particle Size Distribution in Polydisperse Aerosols by the Diffusion Method". *British Journal of Applied Physics*. V. 13. p. 280. 1962.
- Fuchs, N.A. and A.G. Sutugin, "Generation and Use of Monodisperse Aerosols" in *Aerosol Science*. C.N. Davies (Editor). Academic Press. London. pp. 1-30. 1966.
- Fusamura N. and R. Kurosawa, "Determination of f-Value in Uranium Mine Air" in *Assessment of Airborne Radioactivity in Nuclear Operations*. International Atomic Energy Agency. Vienna. pp. 213-227. 1967.
- George, A.C. and L. Hinchliffe, "Measurement of Uncombined Radon Daughters in Uranium Mines". *Health Physics*. V. 23. No. 6. 1972.
- George, Andreas C., "Measurement of the Uncombined Fraction of Radon Daughters with Wire Screens". *Health Physics*. V. 23. pp. 390-391. 1972.
- Gormley, P.G. and M. Kennedy, "Diffusion From a Stream Flowing Through a Cylindrical Tube". *Proceedings of the Royal Irish Academy*. V. 52. pp. 163-169. 1949.
- Green, H.L. and W.R. Lane, "Particulate Clouds: Dust, Smokes and Mists". D. Van Nostrand Company. 2nd Edition. Princeton. 1964.

- Hague, A.K.M.M. and A.J.L. Collinson, "Radiation Dose to the Respiratory System Due to Radon and its Daughter Products". Health Physics. V. 13. pp. 431-443. 1967.
- Hanna, M.G. Jr., P. Nettesheim and J.R. Gilbert, "Inhalation Carcinogenesis". Preface to the AEC Symposium Series #18. U.S. Atomic Energy Commission. 1970.
- Harley, N.H. and B.S. Pasternak, "Alpha Absorption Measurements Applied to Lung Dose from Radon Daughters". Health Physics. V. 23. pp. 771-782. 1972.
- Hatch, Theodore F. and Paul Gross, "Pulmonary Deposition and Retention of Inhaled Aerosols". Academic Press. New York. 1964.
- Holleman, D.F. and K.J. Schiager, "In Vivo Counting Techniques" in Radon Progeny Inhalation Study. Third Annual Progress Report on Atomic Energy Commission Contract AT(11-1)-1500. Colorado State University. Fort Collins, Colorado. pp. 7-1 - 7-6. 1967.
- Holleman, D.F., "Radiation Dosimetry for the Respiratory Tract of Uranium Miners" (Ph.D. Dissertation). Colorado State University. Fort Collins, Colorado. 1968.
- Hurst, J.B., A. Schraub, E.L. Sattler and H.P. Hofmann, "Fate of ^{212}Pb Inhaled by Human Subjects". Health Physics. V. 16. pp. 257-267. 1969.
- IAEA, "Measurement of Radioactivity in Body Organs". International Journal of Applied Radiation and Isotopes. V. 22. pp. 385-398. 1971.
- ICRP, "Permissible Dose for Internal Radiation". International Commission on Radiation Protection. Publication #6. Pergamon Press. 1964.
- ICRP, "Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract". Task Group on Lung Dynamics. Health Physics. V. 12. pp. 173-207. 1966.
- Iinuma, T.A. and T. Nagai, "Image Restoration in Radioisotope Imaging Systems" in Physics in Medicine and Biology. V. 12. pp. 501-509. 1967.
- Jacobi, W., "The Dose to the Human Respiratory Tract by Inhalation of Short-Lived ^{222}Rn and ^{220}Rn Decay Products". Health Physics. V. 10. pp. 1163-1175. 1964.

- James, A.C., G.F. Bradford and D.M. Howell, "Collection of Unattached RaA Atoms Using a Wire Gauze". *Aerosol Science*. V. 3. pp. 243-254. 1972.
- Landahl, H.D., "Removal of Airborne Droplets by the Human Respiratory Tract". *The Lung. Bulletin of Mathematical Biophysics*. V. 12. pp. 43-56. 1950.
- Lippman, Morton and Roy E. Albert, "The Effect of Particle Size on the Regional Deposition of Inhaled Aerosols in the Human Respiratory Tract". *American Industrial Hygiene Association Journal*. V. 30. pp. 257-275. 1969.
- McLaughlin, J.P., "The Attachment of Radon Daughter Products to Condensation Nuclei". *Proceedings of the Royal Irish Academy*. V. 72. Section A. No. 4. pp. 51-70. 1972.
- Marinelli, L.D., C.E. Miller, P.F. Gustafson and R.E. Rowland, "The Quantitative Determination of Gamma Ray Emitting Elements in Living Persons". *The American Journal of Reontgenology: Radium Therapy and Nuclear Medicine*. V. 73. No. 4. pp. 661-671. 1955.
- Marinelli, L.D., C.E. Miller, H.A. May and J.E. Rose, "Low Level Gamma Ray Scintillation Spectrometry: Experimental Requirements and Biomedical Applications". *Advances in Biological and Medical Physics*. V. 8. pp. 81-160. 1962.
- Martz, D.E., D.F. Holleman, D.E. McCurday and K.J. Schiager, "Analysis of Atmospheric Concentrations of RaA, RaB and RaC by Alpha Spectroscopy". *Health Physics*. V. 17. pp. 131-138. 1969.
- Mercer, T.T., R.F. Goddard, R.L. Flores, "Output Characteristics of Several Commercial Nebulizers". *Annals of Allergy*. V. 23. pp. 314-326. 1965.
- Mercer, T.T., M.I. Tillery and H.Y. Chow, "Operating Characteristics of Some Compressed-Air Nebulizers". *American Industrial Hygiene Association Journal*. V. 29. No. 1. pp. 66-78. 1968.
- Mercer, T.T. and William A. Stowe, "Deposition of Unattached Radon Products in an Impactor Stage". *Health Physics*. V. 17. pp. 259-264. 1969.
- Miller, C.E., "A New Technique for Determining the Distribution of Radium and Thorium in Living Persons" in *Assessment of Radioactivity in Man*. IAEA Symposium. V. 1. IAEA, Vienna. pp. 67-77. 1964.

- Morken, D.A., "Personal Communication to K.J. Schiager". December 17, 1968.
- Morrow, P.E., "Models for the Study of Particle Retention and Elimination in the Lung" in Inhalation Carcinogenesis. U.S. Atomic Energy Commission Symposium Series #18. 1970.
- Nelson, Norton, "Inhalation Carcinogenesis in Man: Environmental and Occupational Hazards" in Inhalation Carcinogenesis. U.S. Atomic Energy Commission Symposium Series # 18. 1970.
- NIOSH, "Radon Daughter Exposure and Respiratory Cancer Quantitative and Temporal Aspects". National Institute for Occupational Safety and Health. National Institute of Environmental Health Sciences. Joint Monograph #1. 1971.
- Palmer, H.E. and W.C. Roesch, "A Shadow Shield Whole-Body Counter". Health Physics. V. 11. pp. 1213-1219. 1965.
- Radiological Health Handbook. U.S. Department of Health, Education and Welfare. Public Health Service. 1970.
- Radon Progeny Inhalation Study. Second Annual Progress Report. COO-1500-3. United States Atomic Energy Commission. Contract AT(11-1)-1500. Colorado State University. Fort Collins, Colorado. 1967.
- Raabe, Otto G., "Concerning the Interactions that Occur Between Radon Decay Products and Aerosols". Health Physics. V. 17. pp. 177-185. 1969.
- Raabe, Otto G., "Generation and Characterization of Aerosols" in Inhalation Carcinogenesis. U.S. Atomic Energy Commission Symposium Series # 18. 1970.
- Raabe, Otto G., "Instruments and Methods for Characterizing Radioactive Aerosols". IEEE Transactions on Nuclear Science. V. NS-19. No. 1. pp. 64-75. 1972.
- Saccomanno, G., V.E. Archer and R.P. Saunders, "Lung Cancer of Uranium Miners on the Colorado Plateau". Health Physics. V. 10. pp. 1195-1201. 1964.
- Savitzky, Abraham and Marcel J.E. Golay, "Smoothing and Differentiation of Data by Simplified Least Squares Procedures". Analytical Chemistry. V. 36. No. 8. pp. 1627-1639. 1964.

- Schlesinger, Richard B. and Morton Lippmann, "Particle Deposition in Casts of the Human Upper Trachobronchial Tree". American Industrial Hygiene Association Journal. V. 33. No. 4. pp. 237-251. 1972.
- Schumann, G. and D. Petrasch, "Measurement of Aerosols in the Size Range Below 0.1 μ m". Journal of Aerosols Science. V. 2. pp. 151-159. 1971.
- Segre, Emilio, "Nuclei and Particles". W.A. Benjamin, Inc. New York. 1965.
- Siek, Robert D. and J. Byron Baird, "Uranium Mill Tailings in Grand Junction, Colorado". Health Physics Society Newsletter. V. 1. No. 3. 1972.
- Sinclair, David, "Measurement and Production of Submicron Aerosols". Proceedings 7th International Conference on Condensation and Ice Nuclei. pp. 132-137. 1969.
- Sinclair, David, "A Portable Diffusion Battery: It's Application to Measuring Aerosol Size Characteristics". American Industrial Hygiene Association Journal. V. 33. No. 11. pp. 729-735. 1972.
- Spurny, K.R. and J.P. Lodge, Jr., "A Note on the Measurement of Radioactive Aerosols". Journal of Aerosol Science. V. 3. pp. 407-409. 1972.
- Stein, R.L., W.H. Ryback and A.W. Sparks, "Deposition of Aerosol in a Plastic Chamber". Journal of Colloid and Interface Science. V. 42. No. 2. pp. 441-447. 1973.
- Thomas, Jess W., "The Diffusion Battery for Aerosol Particle Size Determination". Journal of Colloid Science. V. 10. pp. 246-255. 1955.
- Thomas, Jess. W., "Thoron Determination by the Two Filter Method". Health and Safety Laboratory Technical Memorandum. No. 71 1. U.S. Atomic Energy Commission. 1971.
- Thomas, Jess W. and Lawrence E. Hinchliffe, "Filtration of 0.001 μ m Particles by Wire Screens". Journal of Aerosol Science. V. 3. pp. 387-393. 1972.
- Tothill, P., "Techniques Based on Analysis of Profile Scanning Data". Measurement of Radioactivity in Body Organs. International Atomic Energy Agency Report 131. Vienna, Austria. pp. 43-53. 1971.

Townsend, J.S., "The Diffusion of Ions Into Gases". Philosophical Transactions of the Royal Society of London. Series A. V. 193. pp. 129-158. 1900.

USAEC, "Radiation Exposure of Uranium Miners". Summary analysis of hearings before the subcommittee on Research, Development and Radiation. Joint Committee on Atomic Energy Congress of the United States. 1967.

Veall, N., "Measurement of Radioisotope Uptake in Body Organs". Introductory survey with special reference to the use of external standard in Measurement of Radioactivity in Body Organs. International Atomic Energy Agency. pp. 1-10. 1971.

Walsh, P.J., "Radiation Does to the Respiratory Tract of Uranium Miners". A review of the literature. Environmental Research. V. 3. pp. 14-36. 1970.

Weibel, E.R., "Morphometry of the Human Lung". Springer, Berlin. 1963.

Whytlaw-Gray, R. and H.S. Patterson, "Smoke". Edward Arnold. London. 1932.