RADON PROGENY INHALATION STUDY

FINAL REPORT

On AEC Contract No. AT(11-1)-1500

June 1, 1965 - May 31, 1974

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ABSTRACT

This report summarizes the efforts and achievements made during a nine-year study of radon progeny inhalation by uranium miners. Major accomplishments were in the areas of characterization of radon progeny aerosols and in the development of equipment and methods for exposure evaluation. This report contains no technical data, as these were all presented in previous progress and special reports.

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INTRODUCTION

Contract No. AT(11-1)-1500, for the study of radon progeny inhalation exposures to uranium miners, was initiated on June 1, 1965 and terminated on May 31, 1974. During the contract period, the objectives of the study remained unchanged; the accomplishments on each of the three basic objectives are discussed below. Although the overall objectives stayed the same throughout the course of the study, the emphasis of the research varied considerably from year to year, depending primarily upon the urgent needs of the uranium mining industry and requests from the AEC for specific data.

FIRST OBJECTIVE: To investigate the relationships between the physical characteristics of radon progeny aerosols and the deposition and retention of these materials in the human respiratory tract.

Carrier-Particle Characterization

During the first three years of this contract numerous measurements of radon progeny aerosols were made in the Schwartzwald uranium mine near Golden, Colorado. Activity concentrations and ratios of the individual radon daughters were determined with an alpha spectrometry system. The size distributions of radon progeny aerosols were investigated by means of cascade impactors and diffusion tube samplers. Although those techniques were not sufficiently accurate to determine the complete size distribution curves, we were able to conclude that more than 95 percent of the radon progeny in the atmosphere of the Schwartzwald mine was carried on particles of less than 0.5- μ m diameter. Although the median size of the aerosols seemed to be related to the water content of the atmosphere, we subsequently determined that the size distribution was correlated with the concentration of condensation nuclei which was in turn influenced by the water content.

Total Respiratory Deposition

Total deposition of radon progeny in the respiratory tract was measured for a number of experimental subjects and working uranium miners by means of in vivo gamma-ray counting techniques. A mobile shadow-shield counter, mounted in a trailer, was used for the deposition measurements. Although an increased deposition in the nasal passages was observed for the smaller particle-size distributions, it was not possible with this technique to obtain quantitative determinations of deposition patterns in the tracheobronchial tree. In other experiments, the total deposition of radon progeny was measured by sampling the inhaled and exhaled air in controlled breathing experiments. The total deposition measured by these techniques agreed well with measurements made by George^{*}, but as with the in vivo counting techniques it was not possible to obtain the complete deposition pattern.

As a result of the measurements made in the Schwartzwalder mine, we concluded that the only way to obtain the quantitative deposition data that would be needed for calculations of dose to the respiratory tract would be by controlled inhalation exposures in a laboratory chamber. A 12-m³ aerosol exposure chamber was subsequently constructed and used for controlled exposures. It was also used routinely for calibration of radon and radon progeny air sampling equipment.

Early experiments, using ^{222}Rn and its progeny as the inhaled source, resulted in quantitative determinations of the total activity deposited in four general body regions: nasal cavities, lungs, stomach and gut. The influence of respiration rate and tidal volume on total deposition was also investigated. The dose to basal cells

* George, A. C., Deposition of radon daughters in humans exposed to uranium mine atmospheres. Technical memorandum, Health and Safety Laboratory, New York Operations Office, USAEC. 1968.

of the tertiary bronchioles was calculated to be 0.8 rad per working-level-month (WLM) of inhalation exposure (COO-1500-12).

Translocation Studies

An effort was made to determine the fractional deposition of radon progeny in the ciliated and non-ciliated portions of the respiratory tract by measurements of translocation or clearance rates. Unfortunately, the physical decay rate of the ^{222}Rn progeny was so much more rapid than the clearance rate that no reliable inferences could be made. Subsequently, experiments were conducted with ^{220}Rn progeny as the inhaled source. This proved to be a much more suitable source for controlled experiments because (1) the controlling 10.6-hour half-life of ^{212}Pb permitted measurements of in vivo distributions, sequentially, for periods of up to 48 hours following exposure, and (2) the 2.62-MeV gamma ray from ^{208}Tl provided an easily distinguishable emission for external detection.

Although limitations on the use of human subjects for experimentation precluded the collection of extensive data, the results were adequate to permit reasonably good determinations of fractional deposition and clearance half-times in the pulmonary and tracheobronchial portions of the respiratory tract (COO-1500-24). Earlier data from the ^{222}Rn progeny studies were generally confirmed, as were values assumed by the ICRP Task Group on Lung Dynamics.*

SECOND OBJECTIVE: To investigate and develop practical field methods for measuring and evaluating inhalation exposures received by individual uranium miners.

Initial Bioassay Studies

During 1967 and 1968 a feasibility study was conducted to identify potential bioassay methods for evaluating exposures of uranium miners. Concentrations of lead-210 and polonium-210 were measured in urine,

* ICRP Task Group on Lung Dynamics, Deposition and retention models for internal dosimetry of the human respiratory tract, Health Physics 12:173-207, 1966.

feces, blood, and hair obtained from seventeen miners at the Schwartzwalder mine. These concentrations were correlated with the cumulative exposure of the miners to short-lived airborne radon progeny. Although the activity concentrations in urine and feces were quite high, useful correlations with exposure could not be found for these biological media. This failure to find a correlation was attributed to the high variability of concentrations in individual samples and the possibility of external contamination being introduced into the samples by careless collection techniques.

Definite correlations were found between activity concentrations in blood and hair and cumulative inhalation exposures. Facial whisker samples were eventually selected as the most promising bioassay medium on the basis of good correlation with exposure and ease of sample collection and processing. Although this feasibility study indicated potentially useful correlations, the results were not adequate for direct application to routine monitoring in the uranium mining industry (COO-1500-10).

Field Monitoring Studies

In order to evaluate the applicability of bioassay monitoring to a larger group of miners under a variety of mining conditions, a second field study was initiated in September 1968 in the Grants, New Mexico area. A two-year contract was obtained from the National Institutes of Environmental Health Sciences to provide additional support for this study.

Four mines in the Ambrosia Lake district near Grants, New Mexico were selected to provide a representative sampling of different environmental conditions underground and different company practices. In each of the four mines, ten miners were selected for participation in the study also on the basis of providing a representative sample of the various underground operations and duties. The bioassay data obtained from these 40 miners over a period of 16 months was correlated with their inhalation exposures as measured by air sampling techniques.

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The Ambrosia Lake study produced three important results:

1. There was essentially no correlation between the concentrations of ^{210}Po (RaF) found in whiskers with the actual exposures of miners to airborne radon progeny. This was true even when the data were averaged over periods as long as 480 days.
2. There were low, but positive, correlations between the concentrations of ^{210}Pb (RaD) in whiskers and the actual exposures of miners to airborne radon progeny. For 30-day averages, the correlation coefficients ranged from 0.21 to 0.24; for 480-day averages, the correlation coefficients were only slightly improved, ranging from 0.31 to 0.59.
3. A very basic and important problem, affecting any potential bioassay monitoring system for uranium miners, was identified and evaluated. This problem is the extreme variability in the sources and concentrations of ^{210}Pb in uranium mine atmospheres.

The conclusion from the bioassay studies was that for any bioassay system to be utilized for practical dosimetry of inhalation exposures by uranium miners, all of the sources and rates of ^{210}Pb intake would have to be accounted for. Since documentation of the sources of ^{210}Pb might require at least as much, if not more, air sampling effort than that needed for the direct determination of airborne radon progeny concentrations, the practical application of bioassay is dubious (COO-1500-21).

Integrating Radon Progeny Air Sampler

A personal air sampler was developed for use by uranium miners. It consisted of a battery powered air sampler and thermoluminescent dosimeters (TLD's) to integrate the radon progeny activity collected on a membrane filter. This sampling system was developed initially during the experimentation in the Schwartzwalder mine during 1967-68 (COO-1500-16). Subsequently, it was tested on a larger scale in conjunction with the bioassay field study at the Ambrosia Lake mines in New Mexico.

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The TLD air sampler proved to be accurate and reliable as long as the air flow was properly maintained. It proved to be impractical as a personal sampler for uranium miners for two reasons: (1) innumerable mechanical problems with the battery-powered pumps when worn in underground mining environments, and (2) lack of cooperation on the part of the miners in the wearing of the samplers. Because of these problems, few useful data were obtained from the field monitoring study.

The TLD samplers proved to be extremely valuable, however, in another project of interest to, but not directly sponsored by, the AEC. This project was the evaluation of radon progeny concentrations in buildings in Grand Junction, Colorado, where uranium mill tailings had been used in construction. For this use, the TLD samplers were provided with line-powered air pumps and placed in nearly tamper-proof housings. This sampling unit has been described in the open literature*. The fact that the TLD method for making integrated measurements of airborne radon progeny concentrations works extremely well, under semi-controlled conditions, indicates its potential use as an area dosimeter in uranium mines.

Alpha Spectrometry

Under this contract, alpha spectrometry was developed as a basic calibration system for determining airborne radon progeny concentrations and converting the measurements to units of Working Levels (WL-1500-7). The method was used, with portable field equipment, to make comparisons of the accuracies of other, simpler field measurement systems. It has also been used regularly, in conjunction with the CSU radon/aerosol chamber, for calibrating the TLD air samplers used by the Colorado Department of Health and the U.S. Environmental Protection Agency, as well as those used by CSU.

*Schiager, K. J., Integrating radon progeny air sampler, Amer. Indust. Hyg. Assoc. J., 35:165-174, March 1974.

Direct-Reading Working Level Survey Meter

A direct-reading working level meter was developed through the prototype and testing stage under this contract. The initial testing and calibration results were very encouraging and the last revision to this contract provided funding for construction of a final prototype instrument suitable for field testing and patent applications. The construction of the electronic portion of the final instrument was subcontracted to the Lawrence Berkeley Laboratory (LBL). The air pump portion of the instrument was to be, and has been, constructed by the principal investigator. Due to commitment of personnel to projects having "higher priorities", LBL has not completed the electronic package as of the date of this report. However, the instrument is expected to be completed in the very near future. It will then be field tested, most probably at uranium mines near Grants, NM. Applications for patents will then be filed, through the AEC, on both the air pump unit and on the sample collection - detection - electronics portion of the system. It is also anticipated that a description and evaluation of the instrument will be published in the open literature.

THIRD OBJECTIVE: To investigate and evaluate methods for controlling exposures of uranium miners to airborne radioactive materials.

Respiratory Protection

The effectiveness of filter type respirators against radon progeny inhalation was investigated during 1967 and 1968. The results of that investigation were published in the open literature (COO-1500-8) as well as in a detailed special project report (COO-1500-9). It was found that half-mask respirators equipped with a suitable filtration medium provided a high degree of protection (up to 95%) when used properly. The most effective filter medium was found to be resin-impregnated wool matting which at that time was commercially available only from a manufacturer in England.

Following the publication of our study, similar filters were made available by at least two U.S. manufacturers. However, the adequacy of this type of protection depends more upon the proper use of the respirator than it does upon the filter material. Consequently, we did not recommend that respirators be used as a basic control measure but, rather, that they be made available for temporary use by individuals who were required to enter areas of high radon progeny concentrations.

Radon Barriers

A separate but related research project was conducted under a grant from the Environmental Protection Agency. This project, begun in February 1970, involved the investigation of potential modifications to buildings to prevent or reduce the diffusion of radon into the structure through concrete walls or floors. The experimental methods and analytical techniques used in that study were primarily developed under this AEC contract. Some of the control measures found to be effective in buildings could also be applicable in uranium mines.

SUMMARY

Substantial quantities of data on both the physical and biological behavior of radon progeny were obtained during the tenure of the Radon Progeny Inhalation Study. A variety of new methods of exposure evaluation and control were developed and investigated, with some failures and some successes. The results of the study have been reported widely enough to allow other interested researchers to avoid the same errors, but also to benefit from the new and practical information.

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SUMMARY OF FEDERAL SUPPORT

<u>Agency</u>	<u>Cumulative Total</u>
U. S. Atomic Energy Commission, Contract No. AT(11-1)-1500, 1 June 1965 - 31 May 1974:	\$ 326,182
National Institute of Environmental Health Sciences, USPHS, Contract No. PH-43-68-1326, 27 June 1968 - 27 October 1970:	\$ 59,140

LIST OF GRADUATE STUDENTS TRAINED AND DEGREES RECEIVED

Name of Student	Period of Support	Dates of Degrees Awarded	
		M.S.	Ph.D
Martz, Dowell E.	7/1/66 - 6/30/68	-	6/68
Flack, Edwin D.	7/1/67 - 12/31/68	12/68	
Gotchy, Reginald L.	9/1/67 - 7/31/68	8/66	6/68
Holleman, Dan F.	9/1/68 - 1/31/69	6/66	12/68
McCurdy, David E.	9/1/68 - 6/15/69	8/66	6/69
Doerges, John E.	9/1/68 - 8/31/71	-	
Ali, Syed H.	2/16/69 - 8/31/69	8/69	
Beethe, Ronald L.	4/1/69 - 6/30/70	6/7;	
Meier, Charles H.	9/1/69 - 3/31/70	Dropped from program	
Corriveau, Ovide J.	4/1/70 - 8/30/70	Dropped from program	
Alexander, Donald R.	6/1/70 - 7/31/73	-	8/74
Savignac, Noel F.	10/1/70 - 1/31/72	8/68	6/74
Morris, Jerry W.	7/15/71 - 10/31/71		
Till, John E.	9/1/71 - 12/31/71	10/72	
Throckmorton, Carl R.	9/1/71 - 11/30/71	-	8/73

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USAEC Contract No. AT(11-1)-1500

Colorado State University, Ft. Collins, Colo.

LIST OF PUBLICATIONS

Report No. COO-1500-

- 1 Dahl, A. H. and K. J. Schiager, Radon decay products inhalation study. First annual progress report, for the period June 1 - December 31, 1965. February 1966.
- 2 Schiager, K. J., Inhalation hazards of radon decay products in uranium mines. Nuclear News, 9, No. 6, 21-23, June 1966.
- 3 Dahl, A. H. and K. J. Schiager, Radon progeny inhalation study. Second annual report, covering calendar year 1966. February 1967.
- 4 Schiager, K. J., Statement presented before the Joint Committee on Atomic Energy, June 8, 1967, in Radiation Exposure of Uranium Miners. Hearings before the JCAE, May-August, 1967, U. S. Government Printing Office, Washington, D. C., Part I, 381-388, 1967.
- 5 Schiager, K. J. and A. H. Dahl, Radon progeny inhalation study. Third annual progress report, covering year 1967. February 1968.
- 6 Schiager, K. J. and A. H. Dahl, Radon progeny inhalation study. Third year summary report. February 1968.
- 7 Martz, D. E., D. F. Holleman, D. E. McCurdy and K. J. Schiager, Analysis of atmospheric concentrations of RaA, RaB, and RaC by alpha spectroscopy. Health Physics, 17:131-138, July 1969.
- 8 Martz, D. E. and K. J. Schiager, Protection against radon progeny inhalation using filter type respirators. Health Physics, 17:219-228, August 1969.
- 9 Martz, D. E., Respiratory protection for uranium miners. (Ph.D. Dissertation). Colorado State University, Fort Collins, Colorado. June 1968.
- 10 Gotchy, R. L., A survey of bioassays for uranium miners. (Ph.D. Dissertation). Colorado State University, Fort Collins, Colorado. June 1968.
- 11 Holleman, D. F., D. E. Martz and K. J. Schiager, Total respiratory deposition of radon daughters from inhalation of uranium mine atmospheres. Health Physics, 17:187-192, August 1969.

- 12 Holleman, D. F., Radiation dosimetry for the respiratory tract of uranium miners. (Ph.D. Dissertation). Colorado State University, Fort Collins, Colorado, December 1968.
- 13 Gotchy, R. L. and K. J. Schiager, Bioassay methods for estimating current exposures to short-lived radon progeny. Health Physics, 17:199-218, August 1969.
- 14 McCurdy, D. E., K. J. Schiager and E. D. Flack, Thermoluminescent dosimetry for personal monitoring of uranium miners. Health Physics, 17:415-422, September 1969.
- 15 Schiager, K. J. and A. H. Dahl, Radon progeny inhalation study. Fourth annual progress report, covering the calendar year 1968. February 1969.
- 16 McCurdy, D. E., Thermoluminescent dosimetry for personal monitoring of uranium miners. (Ph.D. Dissertation). Colorado State University, Fort Collins, Colorado, June 1969.
- 17 Schiager, K. J., Radon progeny inhalation study. Fifth annual progress report, covering the calendar year 1969. February 1970.
- 18 Schiager, K. J., A field study of personal air sampling and bioassay techniques for monitoring radon progeny inhalation exposures of uranium miners. Second Congress of the International Radiation Protection Association, Paper No. 124. Brighton, England, May 3-8, 1970.
- 19 Schiager, K. J., Radon progeny inhalation study. Sixth annual progress report, for the period January, 1970 through July, 1971, August 1971.
- 20 Schiager, K. J., Six year summary - radon progeny inhalation study. August 1971.
- 21 Schiager, K. J. and N. F. Savignac, Radiation monitoring of uranium miners: a comparison of bioassay, TLD, and the Kuznetz determinations of current exposures. Special report, May 1972.
- 22 Schiager, K. J., A preliminary report on a direct reading, working level survey meter. March 1973.
- 23 Savignac, N. F. and K. J. Schiager, Uranium miner bioassay systems: lead-210 in whiskers, Health Physics 26:555-565, June 1974.
- 24 Alexander, D. R., Fate of inhaled radon progeny. (Ph.D. Dissertation). Colorado State University, Fort Collins, Colorado. August 1974.
- 25 Schiager, K. J., Radon progeny inhalation study. Final Report. October 1974.

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LIST OF ORAL PRESENTATIONS

- 1 Schiager, K. J., D. E. Martz and D. E. McCurdy, Air samples - mass or volume units? Presented at the American Industrial Hygiene Conference, Chicago, Illinois, May 1967.
- 2 Gotchy, R. L. and K. J. Schiager, Experimental bioassay program for uranium miners. Presented at the annual meeting of the Health Physics Society, Washington, D. C., June 1967.
- 3 Schiager, K. J., Environmental working hazards in uranium mines. Seminar talk presented at the National Center for Radiological Health, USPHS, Rockville, Md., October 20, 1967. (This paper was to be published in Radiological Health Data and Reports, but when it was finally planned for inclusion in July, 1971, it was so out of date that it was withdrawn.)
- 4 Martz, D. E. and K. J. Schiager, Respiratory protection against radon progeny: an evaluation of devices by in-vivo gamma-ray counting. Presented at the American Industrial Hygiene Conference, St. Louis, Mo., May 1968.
- 5 McCurdy, D. E. and K. J. Schiager, Aerosol characterization of radon progeny in mine atmospheres. Presented at the American Industrial Hygiene Conference, St. Louis, Mo., May 1968.
- 6 Uranium mining health physics and Mobile in-vivo analyzer. Two exhibits displayed and explained at the annual meeting of the Health Physics Society, Denver, Colorado, June 1968.
- 7 Schiager, K. J., Comments on: Radiation standards for uranium mining, 41 CFR 50-204.321. Presented before the Hearing Examiner, Bureau of Labor Standards, U. S. Dept. of Labor, Washington, D. C., November 21, 1968.
- 8 Schiager, K. J., Monitoring of airborne radon progeny with thermoluminescent dosimeters. Presented at the Nuclear Science Symposium sponsored by the Institute of Electrical and Electronics Engineers, San Francisco, Calif., October 1969.
- 9 Schiager, K. J., Uptake of Pb-210 and Po-210 in whiskers of uranium miners. Presented at an informal meeting on the biology and ecology of polonium and radiolead held at the Institute of Cancer Research, Sutton, Surrey, England, May 1970.