Project Title: Measurement of Radon, Thoron, Isotopic Uranium and Thorium to Determine Occupational and Environmental Exposure and Risk at Fernald Feed Material Production Center

Covering Period: September 15, 1997 to March 31, 2004

Date of Report: July 1, 2004

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Award Number: DE-FG07-97ER62522 EMSP 74050

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Project Objectives:

All research objectives have been accomplished with the exception of measurements during the actual transport of radium from the Fernald silos which is due to take place in early 2005.

1. Objective: To develop a new and novel area and personal radon/thoron detector for both radon isotopes (^{222}Rn / ^{220}Rn) to better measure the exposure to low airborne concentrations of these gases at Fernald. These measurements are to be used to determine atmospheric dispersion and exposure to radon and thoron prior to and during retrieval and removal of the 4000 Ci of radium (^{226}Ra) in the two silos at Fernald.

Achievement: A Patent (6,744,055) was issued during the renewal phase of this contract on June 1, 2004 (submitted June 2000) for our miniature radon/ thoron personal and area detector. It can be worn or placed in the field. No similar detection system exists. These detectors have been deployed on top of the silos and around their perimeter for over 3 years. The radium has not as yet been removed from the silos and the focus of the last phase of the research is to obtain measurements using the detector to assess exposure with this state-of-the-art technique.

2. Objective: To develop a new miniature personal (or area) aerosol particle size sampler that will operate continuously for weeks or months of continuous air sampling under adverse conditions such as outdoors in dusty and freezing climactic conditions. Aerosol particle size is

the major determinant of lung dose and without a measurement of the inhaled aerosol particle size accurate lung dosimetry cannot be obtained. No DOE facility, with the exception of Fernald, is measuring the inhaled particle size spectrum for dosimetric purposes.

Achievement: A miniature particle size sampler (IMP Integrating Miniature Psize Sampler) was injection molded on July 8, 2004 during the renewal of this contract and is available at low cost to anyone from the molding company. Over 2 years of data were obtained on top of the radium silos and in 5 other locations around Fernald such as near the raffinate pits, using a prototype version similar to the new unit.

There is presently no other particle size detector that will yield an integrated size spectrum over a 1 to 2 month interval for detailed lung dosimetry,

History

Fernald has considerable radium and thorium-230 waste and was the largest storage facility in the United States for thorium-232. The site is almost completely remediated (July 2004) and the land will eventually be returned to the State of Ohio. The plant was located at a relatively remote location in Hamilton county Ohio (1050 acres) as all of the historic centers involved in the production or processing of nuclear material for weapons or nuclear reactors.

One of the primary operations at the Fernald site was separation of uranium from the naturally occurring uranium bearing ore. Natural uranium supports a chain of 13 radionuclides that includes several long lived members, namely radium-226 (1600 y), thorium-230 (8×10^4 y), and lead-210 (22 y). In the ore all the nuclides are in equilibrium, i.e., the specific radioactivity (Ci/gm) of each member is the same. Following the uranium extraction via a solvent, the radium was precipitated as solid radium barium sulfate, similar to a fine grained white sand. The viscous residue solution containing thorium-230 and lead-210 was disposed of in onsite raffinate pits.

The ^{226}Ra decays directly to ^{222}Rn and its short lived decay products. There is always some normal off gassing of radon from the silos and as the ^{226}Ra is transferred out of the silos the atmospheric concentrations and the dispersion of the radon should be determined. It is well documented that the actual lung dose and risk from radon is not directly from the gas, but from the deposition of its particulate short lived decay products on the lining of the bronchial airways (Harley et al. 1996, NCRP 1984, NAS 1999). The decay products attach to the natural aerosol particles and this particle size distribution determines the ultimate lung dose.

Because Fernald had also been a storage site for thorium-232, the potential for exposure to its gaseous decay product radon-220 (thoron) exists. Thorium-232 supports a decay chain of 10 radionuclides that include radon-220. We began to measure thoron because there was the potential for exposure.

For disposal, the thorium-230 in the raffinate pits is dug and first taken to a large on-site soil dryer. The relocation of this material to Utah involves drying at high temperature to remove any volatile organic material in the waste. During the drying there is the potential for release and inhalation of the alpha emitter thorium-230, and a need to measure air concentrations and particle size. We have measurements during such an excursion from the dryers.

As ^{226}Ra is removed for ultimate burial in Nevada (or possibly onsite), it is necessary to accurately assess airborne concentrations, the dispersion of radon on and offsite and the particle size of the aerosolized radon decay products. As material is dug from the raffinate pits and dried there it is necessary to measure concentrations and aerosol particle size to assess dispersion and consequent lung dose.

Status:

In this report, all of the goals specified in the, were met or exceeded. The exception is the actual measurements of ^{222}Rn and ^{220}Rn during the transfer of ^{226}Ra from the silos to the holding tanks which is expected to take place from November 2004 to February 2005. The measurements prior to the transfer are complete.

OF NOTE

An important research conclusion obtained in this study is that ^{220}Rn (thoron) is present naturally in all atmospheres and historic published radon measurements are compromised unless thoron has been excluded from the gas measurement. Some enhanced thoron was observed near the pits at Fernald presumably due to the presence of ^{232}Th in the raffinate pits. Although ^{230}Th is the waste product following separation of uranium, some adventitious ^{232}Th was probably present in the ore and was carried through in the separation process.

With the exception of the detector developed for Fernald, there is presently no other instrument to measure both radon and thoron gas simultaneously, and in duplicate, in a miniature, passive personal or area monitor. The 4Leaf is now available to anyone, at low cost, from the molding company for any projects.

^{222}Rn (radon) and ^{220}Rn (thoron) Measurements

The miniature radon thoron detector was (4Leaf) was located on top of the silos as well as in 25 locations around the silos and in 40 locations around the Fernald facility for almost 4 years. The 4Leaf was modified during this time from a 3 chambered version to final form of a 4 chamber version to obtain duplicate measurements of each gas (radon and thoron) which permits the precision of the measurement to be calculated.

Figure 1 shows photos of the 4Leaf interior and exterior. This detector measures even very low outdoor environmental concentrations of both gases with proven accuracy. To assess thoron

bronchial dose, the decay products of thoron ^{212}Pb and ^{212}Po , are usually measured (Schery 1985, 1990, Schery and Zarcony 1985, Harley and Chittaporn, 2000, Harley et al. 2000), and ^{220}Rn concentrations are also estimated from these decay products.

Thoron gas itself is rarely measured, because of the difficulty in measuring an alpha particle emitting gas with a very short half life ($t_{1/2} = 55$ secs). The measurement of the two gases normally requires expensive, laboratory, real time instrumentation with various types of decay chambers to permit a difference in signal with and without the ^{220}Rn (Israel, 1964, NCRP, 1987). The NYU passive alpha track detector measures the integrated signal from the alpha decay of thoron gas, over any time period, depending upon the exposure assessment needs in an inexpensive miniature passive field detector.

The ^{222}Rn and ^{220}Rn personal monitor was originally designed to use alpha track detection film in 3 separate chambers using different entry diffusion barriers for signal differentiation between the two gases. A conducting foam, directly above each chamber excludes the solid decay products and permits only radon or thoron gas passage. The monitor shape is trilobed (similar to a cloverleaf) with a face dimension of 5 cm. across chambers and a thickness of 1 cm., weight 26 grams. It is molded from conducting ABS (CNI) plastic, i.e., plastic with embedded nickel coated carbon fibers. The nuclear track film used for detection of the alpha particles emitted within each chamber is a 9x9 mm square solid state nuclear track film (CR-39). The pristine film background is 5 to 15 alpha particle tracks depending upon the batch and the entire film area is counted.

Duplicate measurements were made of ^{222}Rn and a single measurement of ^{220}Rn in the 3 chambers. As measurements were conducted it was clear that ^{220}Rn is detected in all locations whether at Fernald or not. The 3 chamber detector was modified to 4 chambers so that direct measurement of precision could also be obtained for the ^{220}Rn measurements. The 4 chamber detector is now used exclusively. A photo of both the 3 and 4 chamber detectors is shown in Figure 1. When measuring the total gas radon concentration, thoron concentration must always be known, otherwise the gas measurement is ambiguous. The newly fabricated 4 chamber version has been in use for about 2 years.

The unit displays no charge artifacts and thus the calibration is constant in all situations. The efficiency for ^{222}Rn is (0.009 Track per Bq m⁻³ day) (0.32 Track per pCi/L day) and the lower limit of detection is 220 Bq m⁻³ day (6 pCi/L day) using a video imaging system for counting the alpha particle tracks. The efficiency for thoron is 0.013Track per Bq m⁻³ day, (0.48 Track per pCi/L day) and the lower limit of detection is 1100 Bq m⁻³ day (30 pCi/L day). The difference in efficiency for the 2 gases is due to the change in volume of the detection chamber with the diffusion barrier in place. Each chamber has a conducting foam entrance filter to prevent entry of the decay products of either radon isotope, and an "O" ring seal to prevent leakage of thoron gas around the diffusion barrier. We have demonstrated using exposures to very high thoron concentrations that no thoron entry to the radon chamber takes place.

Various diffusion barriers were tested for their efficiency in permitting radon entry while inhibiting thoron entry. The most appropriate barrier film for our purposes is 3 mil aluminized Mylar, and this is now used in all the ^{222}Rn measurements.

After exposure in our radon and thoron test chambers, or in the field, the CR-39 alpha track film is etched in 6 N KOH overnight to reveal the alpha particle tracks as shallow pits. The tracks are scored either visually after enlarging the 9x9 mm area with a microfiche reader and printing to a standard "hard copy" paper image (about 23X), or by image analysis. The image analysis technique scores all tracks on the 9x9 mm film directly, using an Olympus Zoom microscope, and a Data Translation frame grabber and image analysis program (GlobalLab Image/2). Track counting is normally performed using image analysis with about 10 to 20% of samples also scored visually for QC. Pristine nuclear track film and exposed positive controls are etched with each batch of research or field samples for QC. Extensive quality control accompanies all measurements. The detectors are submitted to all intercomparison exercises both at the USDOE EML laboratory (EML 1997) and the commercial Bowser Morner radon intercomparisons. Our results have never differed from the intercomparison target value by more than 10%.

Results

Radon and thoron data

A 4Leaf has been on top of the silos since early 2000 and twenty five surround the radium containing silos at Fernald since May 2000. Most of the measurements show normal background radon concentration, however, downwind of the silos there is a somewhat elevated ^{222}Rn concentration.

The radon and thoron data are shown in Figure 2 and 3. Silo 1 is shown in Figure 4 where the detectors are located. The ^{222}Rn concentration on top averaged about 200 Bq m⁻³ over the period December 1999 March 2004. The ^{220}Rn (thoron) concentration averaged about 40 Bq m⁻³ during the same period, or about 25% of the total signal.

Measurements for quality control are made in the National Weather Service site in Central Park, Manhattan. The detectors are located at 10 cm. and 87 cm. above bare earth in two locations within the fence. These data are shown in Figure 5. In these measurements ^{220}Rn almost always exceeds the ^{222}Rn .

It is of significance that the detectors measured both radon and thoron simultaneously, at normal background concentrations. The thoron was in most cases a significant fraction of the total gas signal or exceeded the ^{222}Rn concentration. This suggests that historic measurements of "radon" made in homes with detectors capable of permitting thoron gas entry could have provided an effective ^{222}Rn concentration that was somewhat higher than actual due to the

confounding influence of ^{220}Rn . Because the dose from thoron decay products is about a factor of five less than that from radon per unit exposure, the health effects from survey data have surely been overestimated.

OF NOTE

The 5 year average radon concentration on top of the silos is 200 Bq m^{-3} (5 pCi /L) and the average thoron concentration 40 Bq m^{-3} (1 pCi l^{-1}). When the head space radon collector was added in Fall of 2003, the radon on top of the silos dropped to 20 Bq m^{-3} (0.5 pCi l^{-1}) and the thoron to 30 Bq m^{-3} (0.8 pCi l^{-1}).

Particle Size Sampler

The Integrating Miniature Particle Size Sampler (IMPS)

There is presently no sampler available commercially or otherwise that will sample for up to 2 months in severe weather conditions and provide particle size distribution information on any inhaled aerosol particles even the natural aerosol at normal environmental concentrations. This personal sampler (4 cm. x 2 cm. diam./height, weight 55 gms.) has been developed for personal or area use specifically for Fernald. The pilot version used a commercial sampler head modified with 5 extra filtration stages. The modifications added a ZnS alpha phosphor inlet impaction stage, followed by four, very fine mesh stainless steel screen filters.

An exit Millipore filter collected any residual particles. A low flow pump (4-6 lpm) draws the atmosphere sampled through the sampler for periods of up to 2 months for an integrated particle size sample. Six units have sampled outdoors at the USDOE Fernald site for about 2 years. The locations are, on top of and between the two ^{226}Ra storage silos, near the raffinate waste pits, and several portable locations during the decontamination process. Radon and thoron measurements are always made simultaneously with the particle size measurements using the 4 chamber alpha track detector. Two background locations in New Jersey are monitored synoptically, and four detectors are located in the New York City National Weather Service site in Central Park for quality control.

The short lived ^{222}Rn decay products attach to the ambient aerosol, and ultimately decay to $^{210}\text{Pb}/^{210}\text{Po}$ which are used as the particle tracer. The ^{210}Po on the 6 filtration stages are alpha counted in our ultra low background scintillation counters (5 counts per day) and a deconvolution program estimates the particle size distribution from the measurements. Extensive calibrations were performed using standard latex particles, and freshly formed (unattached) ^{222}Rn decay products, to determine the deposition characteristics of the sampler over the size range from ultrafine ($< 1 \text{ nm}$) nanometer to micrometer sized particles. Three size modes are evident in all outdoor field samples, an ultrafine, a small sized (100-200 nm), and a micron sized mode.

The size distribution on top of the radium silos at Fernald and outdoors at a research home in New Jersey remains approximately the same over seasonal intervals at each site. The median diameter is the same at both locations, about 200 nm. However, the seasonal concentration of ^{210}Pb / ^{210}Po varies by a factor of 2 with the highest concentrations in winter, 1700 vs 600 uBq/m^3 at Fernald, and 650 versus 300 uBq/m^3 in New Jersey. The size distribution of the ultrafine or unattached fraction of the decay products (about 1 to 5 nm) also varies seasonally with larger median diameter occurring in colder season. The reason for these differences is likely due to the well known increase in the number of temperature inversions during winter.

The particle size sampler is shown in Figure 6 and on site at Fernald in Figure 7.

This particle size sampler is now available for anyone to purchase from the molding company. It is molded from conducting plastic so no charge artifacts are present. The new version has ring screen holders for up to 6 fine mesh screen filters and should give higher resolution to the particle size spectrum than the pilot version. We are attempting to encourage its use at DOE sites as there is presently no size distribution aerosol particle data being obtained. Without the size information it is not possible to estimate lung dose with accuracy.

The particle size distribution data measured by the sampler on top of (between) the 2 silos during 2004 is shown in Figure 8, and the particle size distribution data near the digging at raffinate pit 5 is shown in Figure 9. The comparison particle size distribution data at a (quality control) home in Northern New Jersey is shown in Figure 10.

OF NOTE

We have located particle size spectrum samplers on top of the silos, near the raffinate pits and at 4 other locations around Fernald. The particle size at Fernald consists mainly of 3 components, an ultrafine size of about 2-3 nanometers comprising 10 percent of the distribution, a 200 to 300 nm size comprising about 80 percent of the aerosol, and a 5000 to 10000 nm component comprising 10 percent of the aerosol spectrum.

Modeling of Radon Dispersion at Fernald

The EPA dispersion code ISC is being used to assess contour information around the silos. The qualitative agreement between our actual measurements around the silos and the ISC dispersion projections is quite good. The ISC code depends upon the available wind information and we used data from the National Climatic Center in Cincinnati.

Figures 11 and 12 show the wind vector and radon contours derived from our estimates of the silo exhalation rate for August to October 2000.

Summary:

Both the 4Leaf radon /thoron detector and the particle size sampler are now available from the molding company for any interested investigator. These instruments provide measurements not previously possible. Even very low background concentrations of the radon isotopes can be measured along with particle size data concerning what is being inhaled. The long lived radon decay products are used as aerosol tracers in this study but the particle size detector (IMP) has wide application to other aerosols. Without the particle size information for the inhaled species it is not possible to estimate lung dose with accuracy. The IMP sampler is of consequence for any pollutant not only for natural radioactivity but for any radiological situation such as in a nuclear reactor facility, or for other non radioactive or biological species. The method of measurement of the material on the filtration stages can be changed to accommodate the species of interest. Bioaerosols, viruses, asbestos or nuisance dust etc. can be sampled over integrated or short term to provide data on the desired pollutant.

This present contract is scheduled to terminate in September 2004. The ^{226}Ra in the silos was to have been relocated at the Nevada Test Site by this time and the data base expected to be complete. However, The State of Nevada issued an injunction to prevent shipment of the ^{226}Ra . The ultimate repository of the ^{226}Ra is not clear at this time but it may be located in the extensive burial pits at Fernald..

There are virtually no large sources, such as the Fernald silos, emitting a radioactive alpha emitting gaseous product making detailed studies of dispersion models possible. In our case the source term at the silos is measured and the environmental radon/ thoron samples provide the actual data to test the models.

Patent:

A patent application for the 3 chamber radon thoron detector was submitted by NYU in June 2000 with later modification for the 4 chamber version. The Patent Office awarded the patent on June 1, 2004. *A Personal Radon and Thoron Detector*, Number 6,744,055

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Figure 1. Radon thoron detector (4Leaf) Exterior and Interior

Figure 2. ²²²Rn (radon) Measurements on top of silos 1 and 2.

Figure 3. ^{220}Rn (thoron) Measurements on top of Silos 1 and 2.

Figure 4. Radium containing Silo 1. Weather shack containing samplers at top.

Figure 5. Radon and Thoron Measurements at, National Weather Service Station in New York City used as our Quality Control Site

Figure 6. Integrating Miniature Particle Size Sampler (IMP)

Figure 7. Integrating Miniature Particle Size Sampler (IMP) Near Raffinate Pit 5 at Fernald. IMP at right in Weather Shack.

Figure 8. Particle Size Distribution and Cumulative Fraction on top of Silos 1 and 2, December 2003 to March 2004.

Figure 9. Particle Size Distribution and Cumulative Fraction Nearby to Digging at Raffinate Pit 5, December 2003 to May 2004.

Figure 10. Particle Size Distribution and Cumulative Fraction used as a Quality Control Home. Northern New Jersey March to June 2002.

Figure 11. Wind Vectors for Fernald (Cincinnati Data) August to October 2000.

Figure 12. Radon Contour using EPA Model ISP for August to October 2000. Silos Located in Middle of Plot.