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SEC 46

September 13, 2006

In response to your request, I am providing you with a summary of the methods we used to estimate radon exposures to Fernald workers. We have given NIOSH a report of our project, as well as a data file of the estimates. Dr. Hornung and I also are preparing manuscripts to submit to peer-reviewed journals. I've also attached the print outs of a set of slides that I used in reporting to the Fernald Workers' Medical Monitoring Program Advisory Group on July 13, 2006.

#### History of the radon sources at the FMPC site

The Feed Materials Production Center (FMPC) at Fernald, Ohio produced uranium metal products for use in Department of Energy defense programs. From 1953 until 1958, high grade pitchblende ore, i.e. rock containing very high concentrations of uranium and uranium daughter products, was used as the raw material for the production of the metal products. This ore was obtained from the Shinkolobwe mine in the Belgium Congo. Uranium was separated from the pitchblende ore by the use of the three-phase Purex process: digestion, extraction, and denitration. The aqueous raffinate or waste from this process was pumped into one of two large concrete silos for storage. These residues, which were assigned the code name "K-65", contain impurities from the Purex process and all of the daughter products of uranium including  $^{226}\text{Ra}$  (the parent nuclide of  $^{222}\text{Ra}$ ) and  $^{232}\text{Th}$ . The K-65 material was slurried from the refinery (Plant 2/3) through pipes into one of two storage silos, K-65 Silo #2, located near the western boundary of the production area.

The radium containing material in K-65 Silo #1 came from another source. Prior to 1953, large amounts of radium bearing radioactive waste were shipped to the Fernald site from other DOE sites, especially Mallinckrodt Chemical Works in St. Louis, and eventually stored in the K-65 Silo #1. When the waste material first arrived, however, it was placed in sealed metal drums which were temporarily stored on a concrete pad near Plant 1, resulting in radon gas emissions inhaled by workers in this area. An internal FMPC memorandum indicates that 12,997 drums of K-65 material were received at the FMPC in the period September 25, 1951 to July 31, 1952. Other drums of K-65 material were stored in Plant 8 for long periods of time. Eventually the drummed material was transferred to Silo #1 when it was completed and ready for operation.

The K-65 silos, located on the western edge of the production area, were constructed in 1951 to 1952, and materials were added to the silos from July, 1952 to September, 1958. The silos had problems with deterioration almost since the time of construction. Significant cracking in the walls and seepage of the contents was noted from the 1950's. Because of these problems, periodic repairs and improvements

to the Silos were implemented from the 1960's through the 1980's. Two components were added to maintain structural stability. Embankments or berms (built in 1964 and enlarged in 1983) were built to relieve tensile stress in the vertical walls and to protect against deterioration of the concrete. The berms also acted as direct radiation shields. Placement of protective dome covers in 1970's, constructed of steel and plywood, blanketed the center of each silo. By themselves, the covers offered little if any barrier to the diffusion and ventilation of radon. Vents in these domes were sealed in 1979. In 1986, a waterproof protective membrane of liquid neoprene was added, and in 1987 a layer of polyurethane foam was placed on dome surface in an attempt to mitigate the migration of radon gas to the environment. No routine environmental monitoring for radon was performed prior to 1980.

A cohort mortality study of Fernald workers, conducted by Dr. Donna Cragle, reported a significant excess of lung cancer deaths among hourly workers (Standardized Mortality Ratio=1.26, 90% confidence interval 1.02, 1.54), with a borderline significant trend ( $p=0.08$ ) in increase in risk with external radiation dose. Surprisingly, there was no correlation with internal radiation dose, which would have been an expected finding. Because there was a radon point source, but only limited air monitoring data from recent years, a radon exposure reconstruction was necessary to obtain lung cancer risk estimates.

#### **Radon Exposure Estimation:**

In our grant application to NIOSH, we proposed to extend the K-65 Silo radon dispersion model, developed by RAC for the 10 kilometer community exposure domain, to the inside the fence line for the Fernald plant site. George Killough, the model's principal developer, assured us that the model would be accurate for worker locations. As a consultant on this project, he produced radon estimates for latitude and longitude coordinates supplied to him by our University of Cincinnati team. These sets of geographical coordinates represented the centroid of each of the buildings on the site occupied by workers. We did additional analyses to estimate the diurnal variation in exposure, significant at the site, as well as the decrease in emissions with the capping of the K-65 silos. For all specified locations, we calculated yearly night and day exposure estimates.

Similar to the community-based dose reconstruction, Killough's worker model was based upon distance from the K-65 silos, wind speed and direction, and the radon source term. Because of the diurnal variation, we requested that Killough develop radon estimates for each of the three shifts for each calendar year. This was an important difference from the original RAC model, but necessary for the application to worker exposure estimates, because it has been well-established that outdoor radon levels peak in the early hours of the morning when atmospheric conditions are usually calm. Therefore, the estimates that we provided for NIOSH are weighted by shift, either reported directly by the worker or assigned to workers based upon average shift distributions for other workers in their departments and time periods.

#### **Model validation**

Since the RAC model is a conceptual mathematical model developed from physical principles and is not based directly upon measured data, we attempted in two ways to investigate the accuracy of its estimates. First, we discovered radon measurement data that had not been used by the RAC group to calibrate their model. These were data collected by John Cardarelli for his Master's thesis during the period of March through September 1991. Data were collected both in the immediate vicinity of the K-65 silos and at various points around the production area. We then compared Cardarelli's measurements by shift and location to the closest year of our estimates, 1988. Since no remediation

was done between 1988 and 1991, we assumed that results should be comparable. The estimates were in reasonably good agreement to Cardarelli's measurements.

Our second attempt to validate the model was the use of CR-39 film attached to windows in selected buildings around the site. The CR-39 film measures alpha activity from the decay of Pb-210, a uranium decay product which was embedded in the glass over several decades. This assay, therefore, serves as a cumulative measure of radon and its decay products since each building was constructed. We assumed that the glass was the original window pane. We attempted to select windows that appeared to be weathered and were located high enough that they were less likely to have been broken. A pilot survey of 20 surface monitors was sent to Dr. Fewes at the HH Wills Physics laboratory in Bristol, England. Results indicated very small replicate error and good ability to measure radon alpha tracks (Po-210) on both low and high exposed buildings. In the pilot study, we were surprised to discover very high values for some of the glass. With NIOSH support, we expanded our sample size to 110 samples placed throughout the site on buildings selected for their proximity to both the K-65 silos and the plant 2/3 area where we found the initial high measurements.

Results of the larger survey indicated that glass sampled in the plant 2/3 area were substantially higher than glass closer to the K-65 silos. Once we became aware of these CR-39 findings, we conducted additional document searches and met with workers several times to develop a better understanding of this local source of radon. The Q-11 silos, close to the Plant 2/3 area, had been used in the period 1953-58 to store highly grade ore before it was processed in the Plant 2/3 complex. Although the amount of material was considerably less than that stored in the K-65 silos, the Q-11 silos were much closer to the production area and were not sealed. The hot raffinate by-product of the chemical separation of high grade uranium also was handled and temporarily stored in the Plant 2/3 area. In addition, drums of similar waste product from Mallinkrodt Chemical Works were stored on a pad near Plant 1, directly north of the Q-11 silos.

Because of the potential for much higher exposures to workers in the Plant 1 and Plant 2/3 area employed during the period 1952-58, we decided to develop a statistical prediction model with Q-11 as the point source. Input variables in this model included CR-39 Po-210 results by building, distance and direction from each building to the Q-11 silos, average wind speed and direction by year, and construction year for each building.

The results of this prediction model were the input data for use by our health physicist, Jeff Lodwick, to calculate radon decay product exposure estimates in Working Levels (WL) from the Po-210 estimates. Through historical documents and interviews of retired workers, we determined that Q-11 and the drummed material on the Plant 1 storage pad were present only for the period 1952-58, for 4153 workers employed at the site during those years. Dr. Lodwick used knowledge of decay product half-lives and the number of years between CR-39 measurements and the period of interest, to calculate annual WL levels for each building within reasonable proximity of the Q-11 silos. Buildings at a large distance from the Q-11 silos were used as a calibration background for the other estimates and were assigned zero exposure from the Q-11 source. All of these background buildings demonstrated very low levels of Po-210 alpha tracks in the CR-39 measurements. Using locations at distant buildings as calibration background produced estimates of radon exposures solely from the Q-11 source, even though the CR-39 film would have also recorded radon released from the K-65 silos. The total yearly radon exposure for each worker was then obtained by summing the contributions from each source.

### **Assignment of worker locations and shift**

The most important factors for determining worker radon exposure for this cohort (N=7142) were location of the worker and shift worked (because of diurnal variation in radon levels). Personnel records did not include this information. Historical records and information from worker interviews and questionnaires (from a subset of the cohort) were used to assign worker location for various combinations of Plant, department, and job title noted on the personnel record. Shift was assigned on the basis of standardized job titles. Questionnaire data regarding shift worked for specific job periods, available for 1207 workers, were used to determine a set of probabilities of shift assignment for clusters of job titles. The worker's locations were linked with the matrices of yearly exposure estimates by location specific to shift, and estimates for each shift were weighted by the shift probabilities assigned to the worker. For the 1207 workers who were asked shift information on their questionnaires, we also calculated a second set of radon estimates using their self-reported information. Location and shift assignment have been validated using historical sampling records and other questionnaire data, not used in assumption development.

### **Radon estimates:**

As might be expected, the estimated radon estimates from the K-65 source generally declined over time with a dramatic drop after 1979 when the silos were capped. The distribution across workers within each year ranged from slightly skewed, generally following a lognormal distribution, to fairly symmetrical. The exposures from the Q-11 source, on the other hand, are essentially bi-modal. The workers who were located in buildings in close proximity to the Q-11 silos are estimated to have relatively high radon exposures, while workers in buildings farther from the source are exposed at much lower levels. Yearly mean day-shift worker exposure attributable to the K-65 source term ranged from 0.002 to 0.085 WL. By contrast, yearly mean exposure to workers on the day shift in the area of the Q-11 silos ranged from 1.3 WL to 7.8 WL from 1953-1958. Worker cumulative exposure from the K-65 source ranged from 0.0 – 51.8 WLM, median 1.5 WLM; cumulative exposure from the Q-11 source ranged from 0.0-700.1 WLM, median 2.6 WLM. The 90<sup>th</sup> percentile cumulative radon exposure from K-65 was 18.06 WLM and from Q-11 source was 31.52 WLM.

I hope this provides you with the information you need for your Special Exposure Cohort application. Please contact me if you have any questions.

Susan M. Pinney, PhD  
Associate Professor

## **Methods for Radon Exposure Estimates for Fernald Workers**

*Susan M. Pinney, PhD*  
Department of Environmental Health

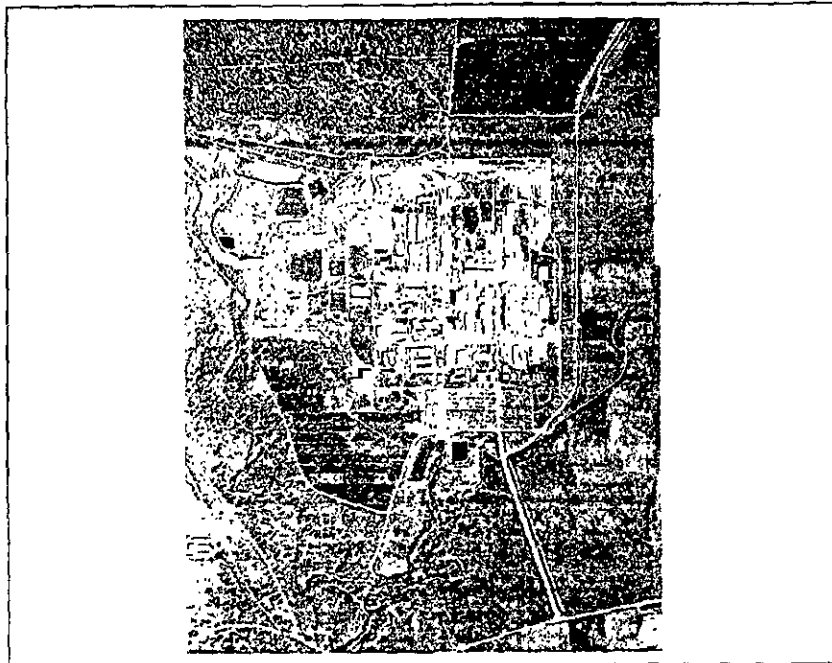
*Richard Hornung, DrPH*  
Institute for Health Policy and Health Services  
Research

**University of Cincinnati**

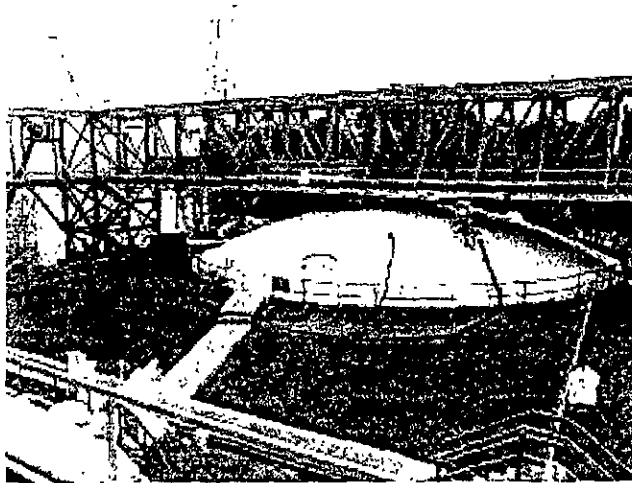
### **Events leading to research study**

- Excess relative risk for lung cancer was found in a mortality study of workers at the Fernald Feed Materials Plant near Cincinnati OH in 1996.
- A CDC dose reconstruction project for estimation of exposures to residents within 10 km of Fernald indicated relatively high radon levels off site.
- Sources of radon decay products were determined to be two K-65 silos located on the west side of the plant and used for storing waste materials including radium.
- NIOSH contracted with the University of Cincinnati to estimate radon exposures and cigarette smoking rates among Fernald workers for possible follow-up of the lung cancer mortality study.

- **STUDY OBJECTIVE:** Estimate annual and cumulative exposures to radon decay products among workers employed at Fernald site from 1952 to 1988.
- **STUDY METHODS (at beginning of study):**
  - Use deterministic mathematical model developed for the CDC dose reconstruction project to estimate radon levels across the site, arising from the K-65 silos.
  - Investigate and estimate radon exposures from secondary sources using CR-39 assay of window glass.
  - Assign locations of workers; assign a set of shift probabilities to workers.
  - Assign estimated annual radon levels measured in Working Levels (WL) to workers by their shift and working location in each year of employment.



## K-65 Silo - 2003



## Radon

- Naturally occurring decay product of  $^{226}\text{Radium}$ , the fifth daughter of  $^{238}\text{Uranium}$
- Both  $^{238}\text{Uranium}$  and  $^{226}\text{Radium}$  are found in most soils and rocks
- Radon decays with a half-life of 3.82 days into a series of short-lived radioisotopes that are collectively referred to as radon daughters, radon progeny and radon decay products.

## Uranium Decay Chain

isotope	Half-life	Radiation
Uranium-238	4,500,000,000 years	alpha
Thorium-234	24 days	beta, gamma
Protactinium-234m	1.2 minutes	beta, gamma
Uranium-234	250,000 years	alpha, gamma
Thorium-230	80,000 years	alpha, gamma
Radium-226	1,622 years	alpha, gamma
Radon-222	3.8 days	alpha
Polonium-218	3.05 minutes	alpha
Lead-214	26.8 minutes	beta, gamma
Astatine-218	2.0 seconds	alpha
Bismuth-214	19.7 minutes	beta, gamma
Polonium-214	0.000184 seconds	alpha, gamma
Thallium-210	1.3 minutes	beta, gamma
Lead-210	22 years	beta, gamma
Bismuth-210	5.0 days	beta
Polonium-210	138 days	alpha, gamma
Thallium-206	4.2 minutes	beta
Lead-206	Stable	none

## Health Effects of Radon

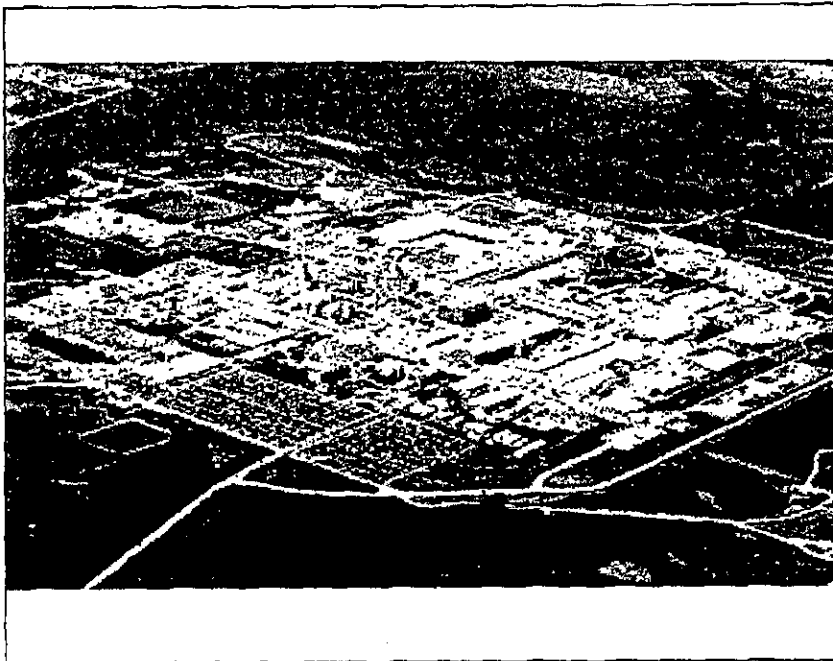
- **When radon progeny are inhaled and release alpha particles within the lungs, the cells lining the airways receive genetic damage, and ultimately lung cancer may result.**
- **Combined risk of smoking and radon – more than additive, less than multiplicative.**
- **Radon also known to increase risk of cataracts**



## Sources of Radon at Fernald

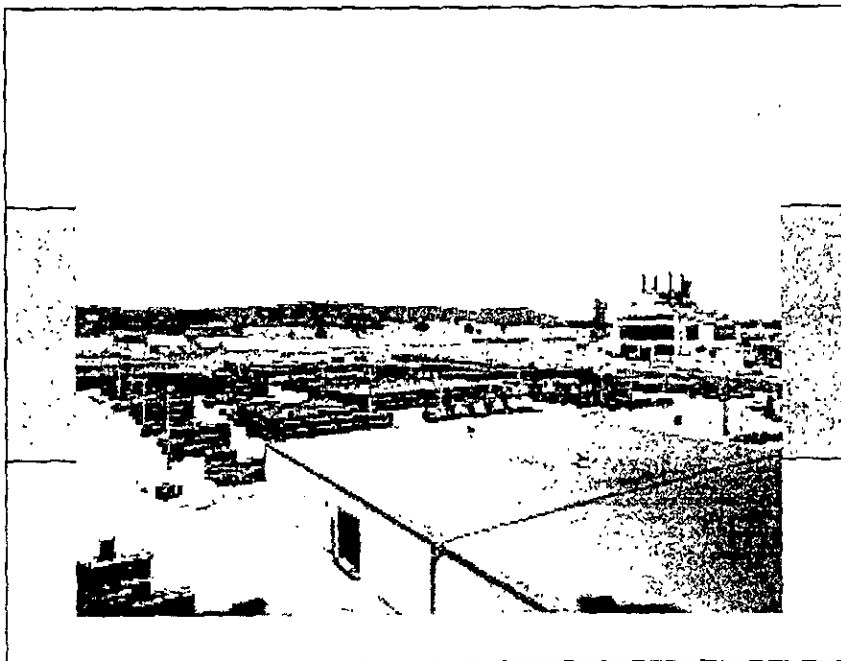
(At time of grant proposal)

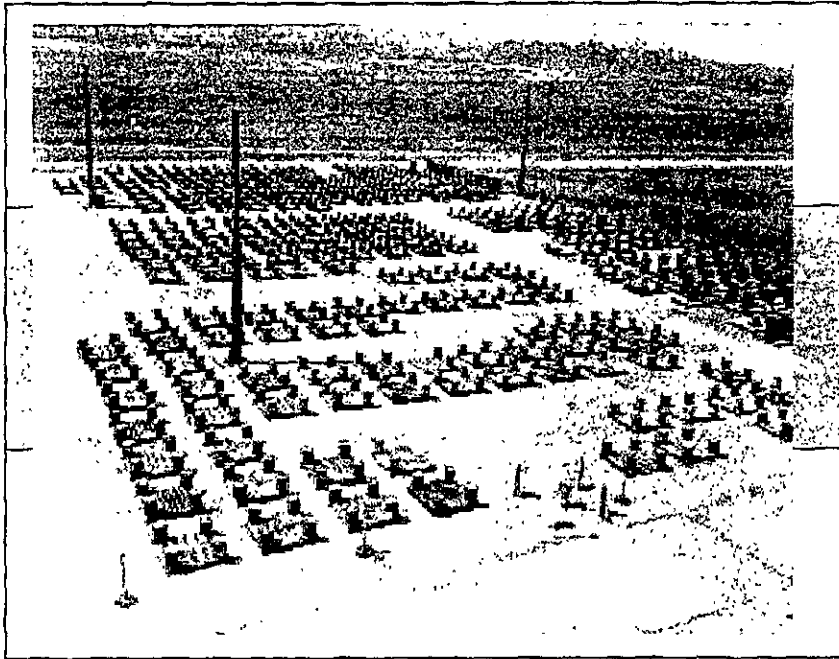
- **K-65 Silos – held raffinate from processing ores rich in uranium, which contained radium**
- **Radium bearing residues from Mallinckrodt Chemical Works and other DOE sites**
  - **Approximately 25,000 barrels – into Silo 1**
- **Processing of pitchblende ores at FMPC**
  - **During 1950's – residues into Silo 2**



## Radioactive Waste Shipped to Fernald

- Prior to 1953, large amounts of radium bearing radioactive waste were shipped to FMPC from other DOE sites, especially Mallinckrodt Chemical Works in St. Louis (12,997 drums)
- Drums stored on a concrete pad near Plant 1 until K-65 silos were completed
- Some drums of K-65 material also stored near Plant 8 for longer period of time



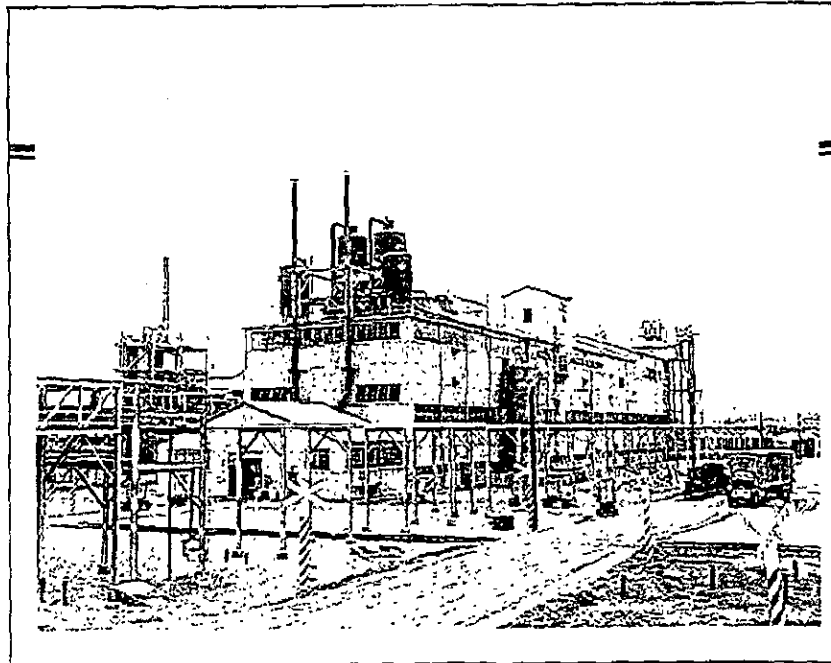


## **Pitchblende Ore Processed at Fernald from 1952-1958**

- Rock containing very high concentrations of uranium and uranium daughter products (including radium).
- Received at FMPC from mine in the Belgian Congo (Shinkolobwe mine).
- Code name Q-11; Stored in Q-11 ore silos near Plant 1

## Pitchblende Ore Processing

- Uranium separated from ore by 3-phase Purex process: digestion, extraction and denitration
- Aqueous raffinate or waste contained all daughter products from uranium and impurities including  $^{226}\text{Radium}$  (the parent nuclide of  $^{222}\text{Radon}$ )
- Assigned code name K-65
- Pumped into large concrete silo for storage (K-65 silo)



## CDC Fernald Dosimetry Reconstruction Radon Release Estimates

Period	Radon Release Estimate
1952-1953	K-65 Silos      1900 Ci
	Drummed waste    720 Ci
1953-1958	K-65 Silos      4900 Ci/yr
1959-1979	K-65 Silos      6200 Ci/yr
1980-1987	K-65 Silos      940 Ci/yr
1988	K-65 Silos      230 Ci

### CDC/RAC Deterministic Model

- Model was developed to estimate radon levels outside the Fernald plant site
- Model was based on physical and mathematical principles, such as emission rates from K-65 silos, plume dispersion from a point source, meteorological conditions, and engineering controls
- Data were only used to calibrate the model
- Output of model was 24-hr average exposures to residents within 10-km radius of K-65 silos

## Location of Workers

- **Since radon exposure resulted in dispersion of radon gas from a source, location of workers was a key parameter in exposure estimation**
- **7143 persons ever worked at the site**
- **2158 were participants of the Fernald Worker Medical Monitoring Program, from whom we have collected detailed occupational history information (questionnaires and interviews)**

## Location Assignment Method

- **Personnel records did not include location, except for production workers assigned to a Plant**
- **General personnel records misclassified location for approximately 1/5 of workers for whom location could be inferred from the records**

## Location Assignment Method

- **Obtained file of job history (multiple record per person file) from NIOSH— each record contained Plant, Department and Job Title information for periods of employment, with beginning and end dates.**
- **For some workers, no work location could be inferred from information in personnel records:**
  - **Mechanical Division (electricians, carpenters, machinists, etc.)**
  - **Laboratory Division**
  - **Inventory and time clerks who worked in offices in the production plants, and**
  - **Workers in the Security division**

## Work Location Assignment

- **We developed a series of assumptions regarding work locations for combinations of plant, department and job title, using records such as FWMMP occupational history questionnaire and interview information, union contracts, and position descriptions.**
- **Each record of the worker's job history was then assigned a location code, indicating one or several work locations for that time period.**

### Worker Assigned to a Specific Building

ID: 34076

**Hire Date**      **Term Date**  
 11/17/1954      01/17/1957

Date	Plant	Dept Code	Job Title	Location Code
11/17/1954	2	Prod	Laborer	Plant 2/3
09/26/1955	3	Prod	Chemical Operator Helper	Plant 2/3
04/02/1956	3	Prod	Chemical Operator	Plant 2/3
11/09/1956	3	Prod	Laborer	Plant 2/3

### Worker Assigned a Combination Code

ID: 37444

**Hire Date**      **Term Date**  
 08/01/1952      01/01/1983

Date	Plant	Dept Code	Job Title	Location Code
08/01/1952	Anal	Tech	Technologist B	ANDBldg15,P
10/01/1955	Anal	Tech	Technologist II	ANDBldg15,P
08/27/1956	Anal	Tech	Section Leader I	Bldg15+P
09/21/1959	Anal	Tech	Section Leader II	Bldg15+P
05/01/1972	Anal	Tech	Technologist III	ANDBldg15,P



### **Work Location Code – linked to multiple exposure estimate locations**

- **“Single location” code - Plant 2/3**
  - 66% Building 2A – Ore Refinery Plant
  - 10% Building 2D- Metal dissolver building
  - 7% each Buildings 3C, 3E and 3H – Control Tower, Hot Raffinate Building, Refinery Sump
  - 3% Building 39A – Incinerator Building
  
- **Combination code – ANDBLDG15,P**
  - 60% Building 15a - Laboratory
  - 40% P [Production Area]– many buildings in the production area

### **Overall Area Codes for Some Workers**

- Some job titles known to work at a variety of areas throughout plant site
- For other job titles, insufficient information to assign them to a specific location.
  
- Developed “area” codes
- Multiple exposure estimation sites linked to each area, weighted by number of employees, with specific or combination location assignment, assigned to that location in that calendar year.

## Overall Area Code - Example

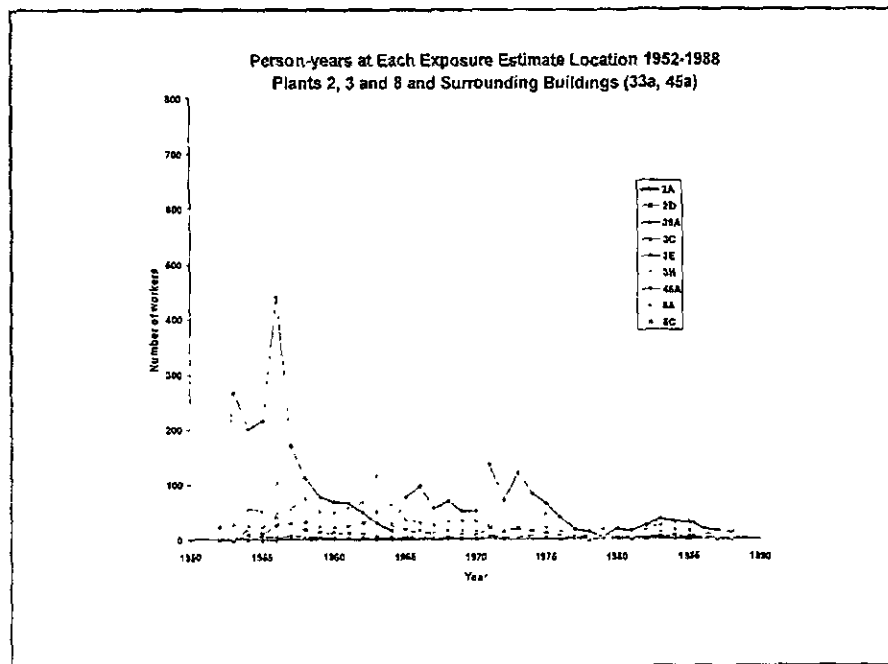
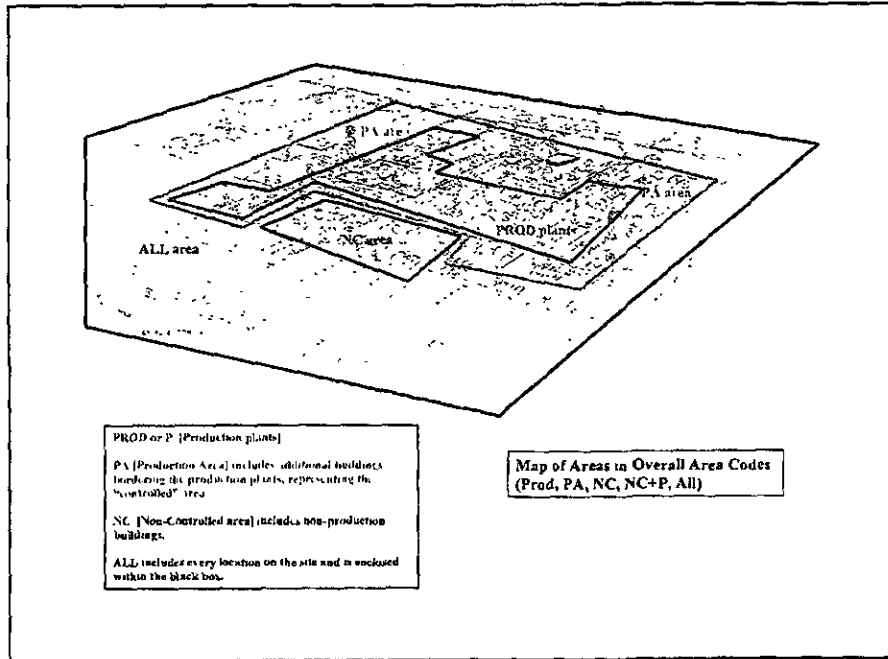
- Overall Area Code – ALL
- 30 location components – with exposure value specific to each calendar year, and weighting, specific to each year

### Worker Assigned to an Area Location

ID: 36831

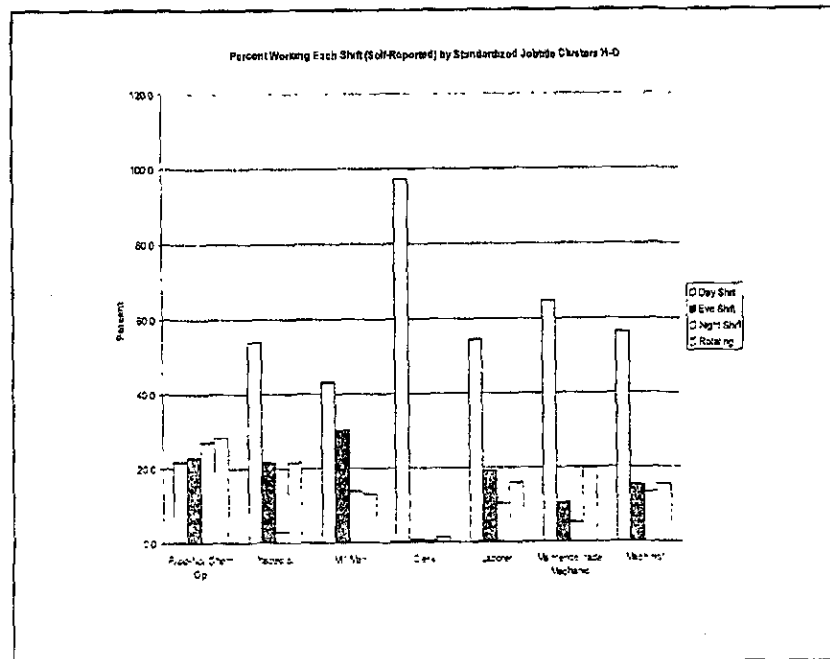
**Hire Date**      **Term Date**  
10/01/1951      01/01/1989

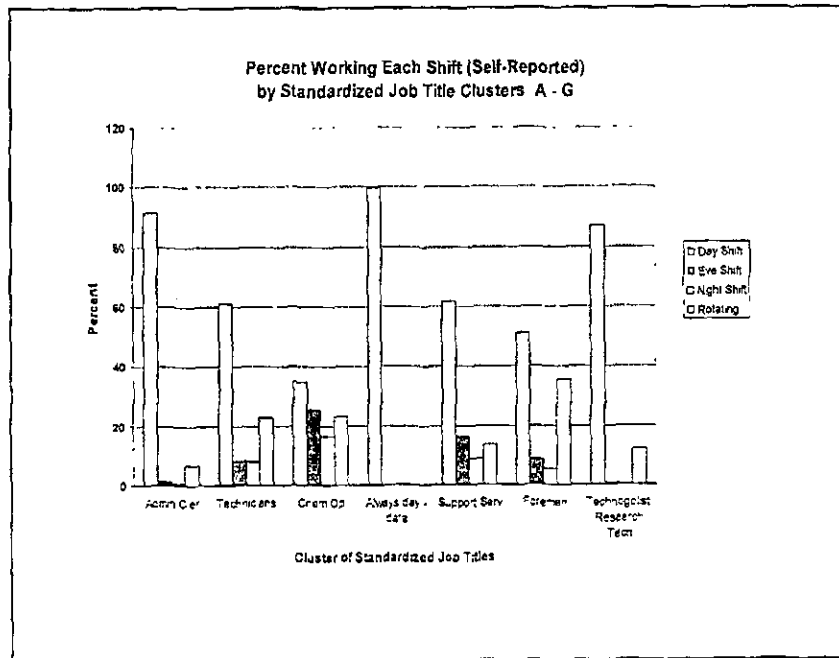
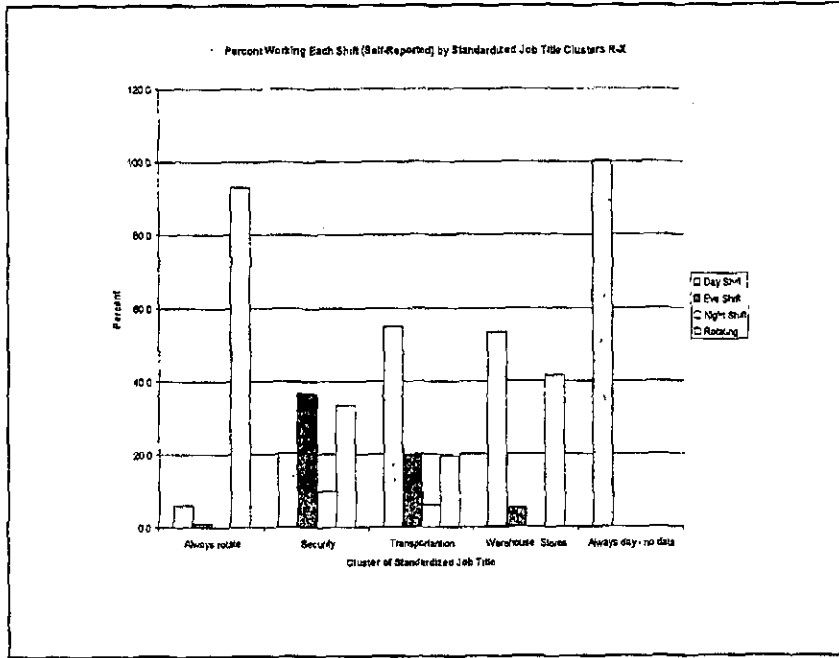
Date	Plant	Dept Code	Job Title	Location Code
10/01/1951		Sec	Security Police	All
11/27/1975		Sec	Security Police Officer	All
01/16/1978		Sec	Police Sergeant	All



## Work Shift – Assumption Development

- Specific questions on work shift included in FWMMP questionnaire modules
- Compiled responses of workers who completed questionnaires
- Used these data to develop assumptions about probability of working evening or night shift for each Jobtitle



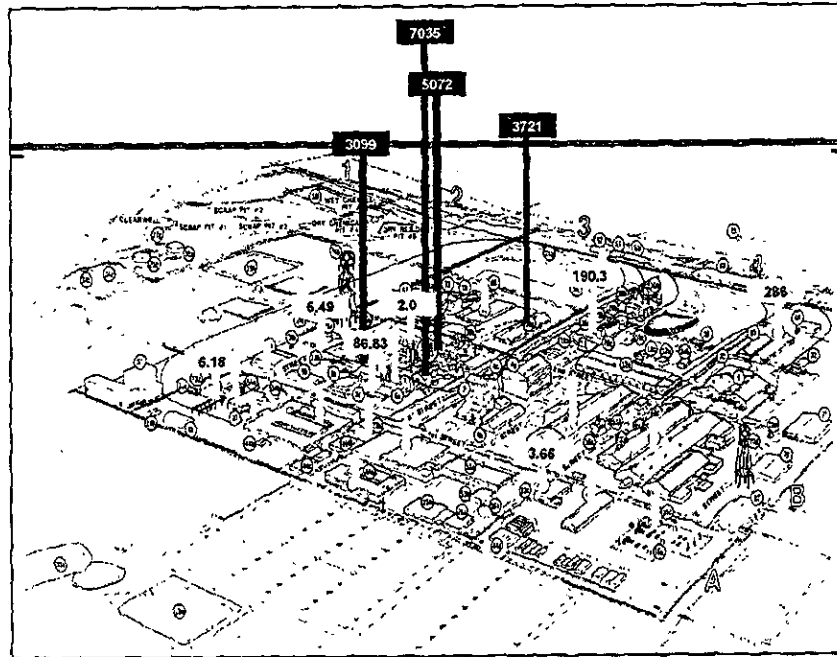


## CR-39 Assay

- CR-39 is a plastic film taped to window glass for about 3 weeks in buildings on site to record polonium-210 alpha activity, a primary radon decay product.
- Glass in place throughout the plant history serves as a cumulative radon dosimeter.
- CR-39 assay was developed at the University of Bristol and used in case-control studies of lung cancer and radon in homes, but never in an industrial, highly contaminated setting.

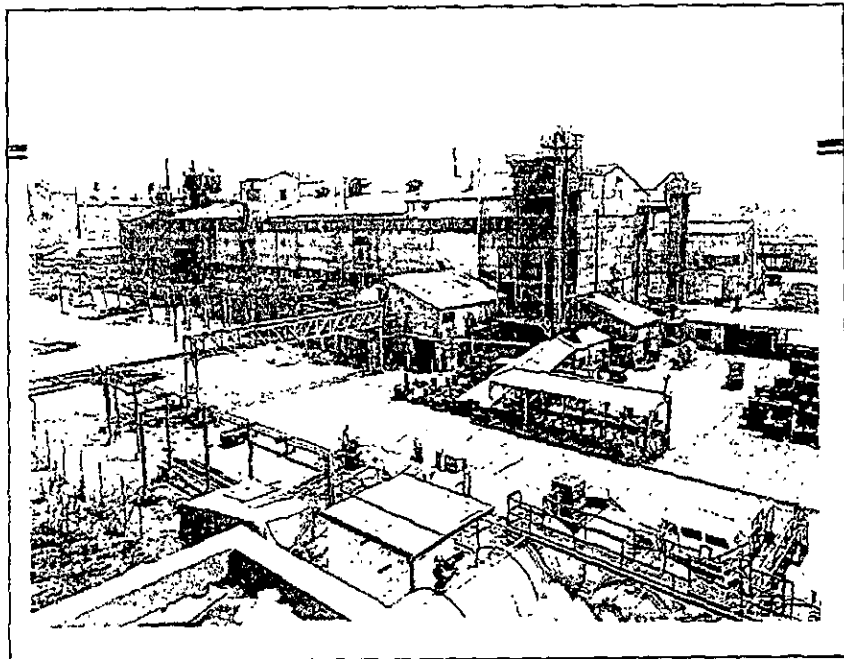
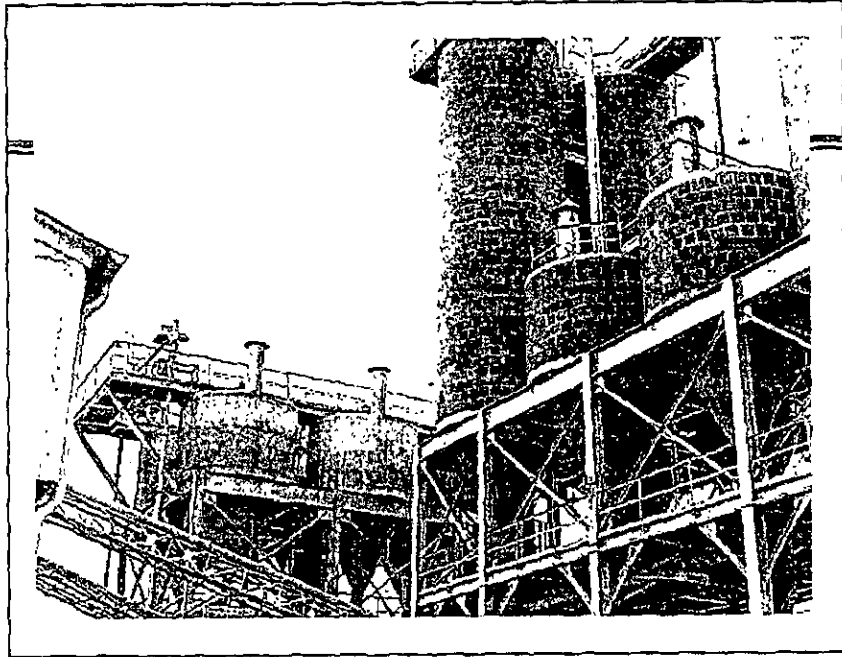
## CR-39 Assay

- Decided to use CR-39 assay to validate estimates from mathematical deterministic model.
- Asked NIOSH for funding
- Pilot study – 20 samples
- Main study – 80 samples
- Additional study – another 31 samples
- 131 CR-39 plastics were placed on both inside and outside of glass panes in buildings throughout the plant.



### Detective Work

- Inspection of CR-39 data shows very high levels near plant 2/3
- Interviews with Fernald HP's and retired workers identified a potential source
- Q-11 silos near plant 2/3 had been used to store highly radioactive material for later processing
- Several other secondary sources were identified in the same area
- Search of documents reveals that Q-11 silos were filled from 1952-58



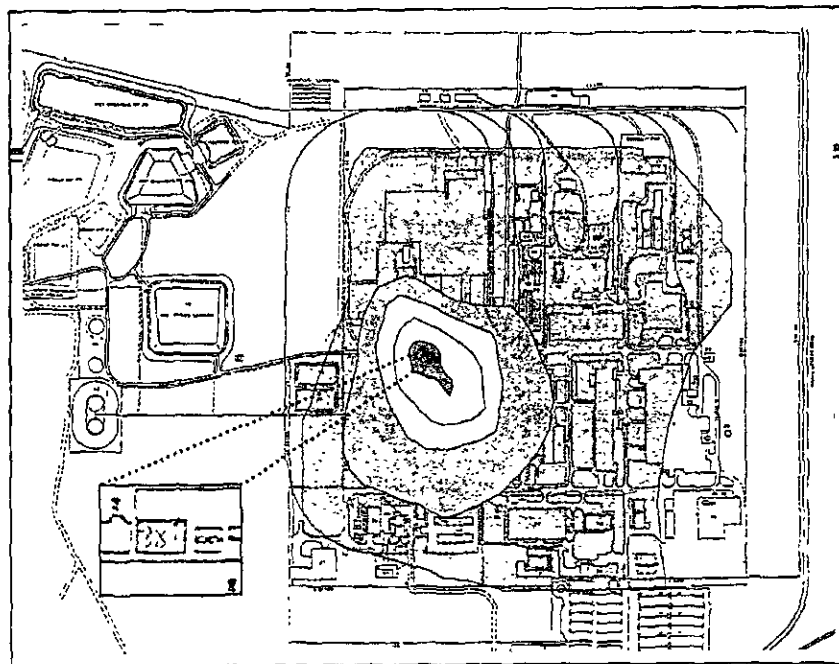
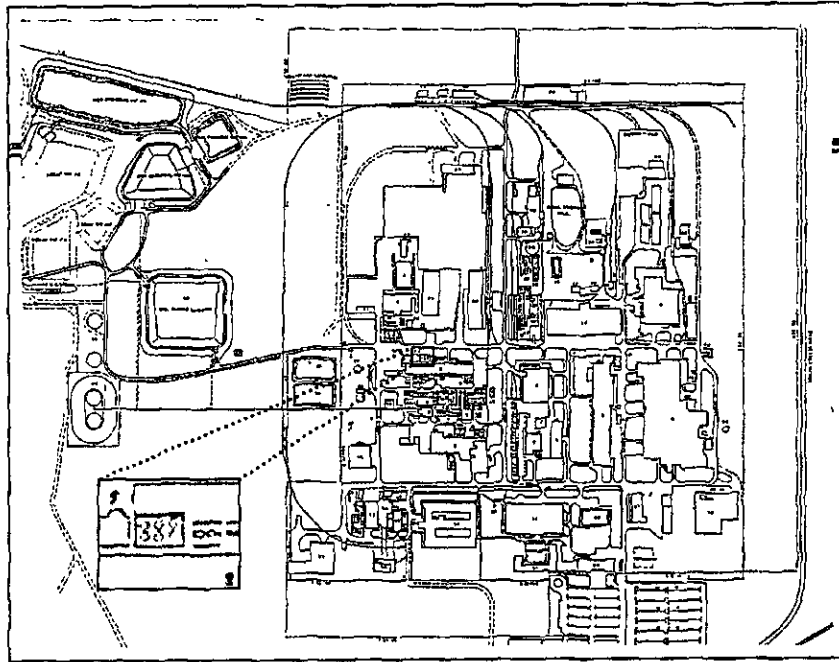


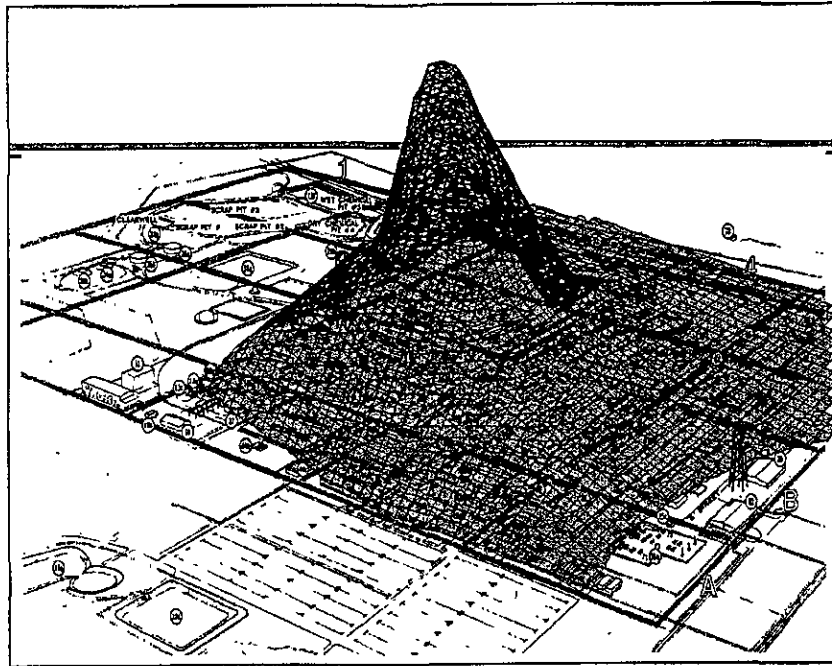
### Analysis of CR-39 Data

- Po-210 measures were linked to lat/long coordinates of the building
- Weighted multiple regression model was used to estimate Po-210 levels for each building where workers were assigned
- Weights were a function of wind speed and direction from Q-11 silos as the point source
- Other factors included in the model were building construction year and inside/outside location of film

### Model Results

- Polynomial and restricted cubic spline models both fit log Po-210 data reasonably well ( $R^2 = 0.50-0.53$ )
- 24% / year decrease for later construction years
- Po-210 levels are 60% less on inside of buildings
- Buildings upwind and at a distance from Q-11 silos were estimated to have exposures one to two orders of magnitude lower than Q-11 area
- Po-210 results were used by the study HP to calculate Rn exposures in WL by year for each bldg



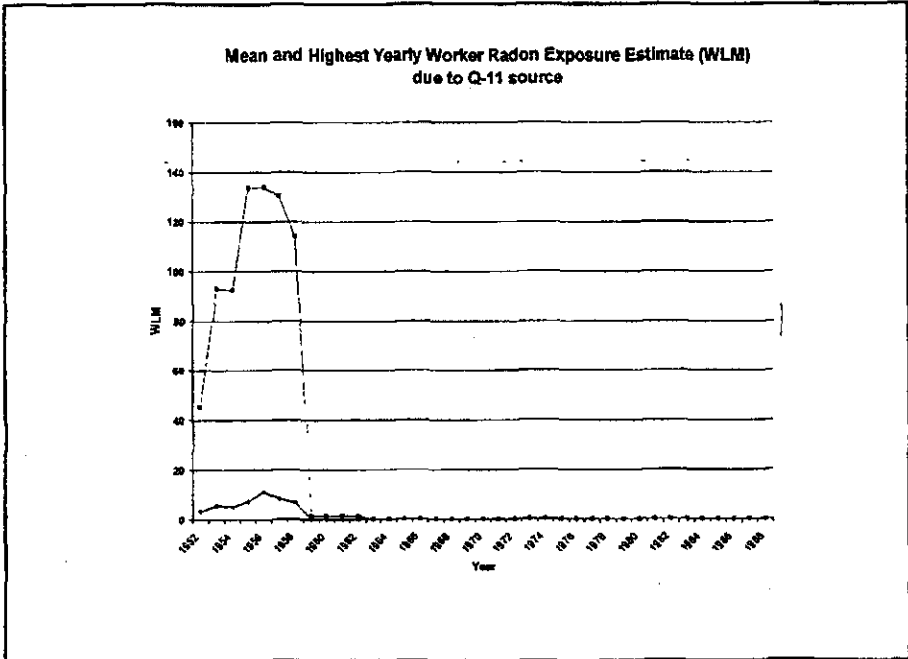
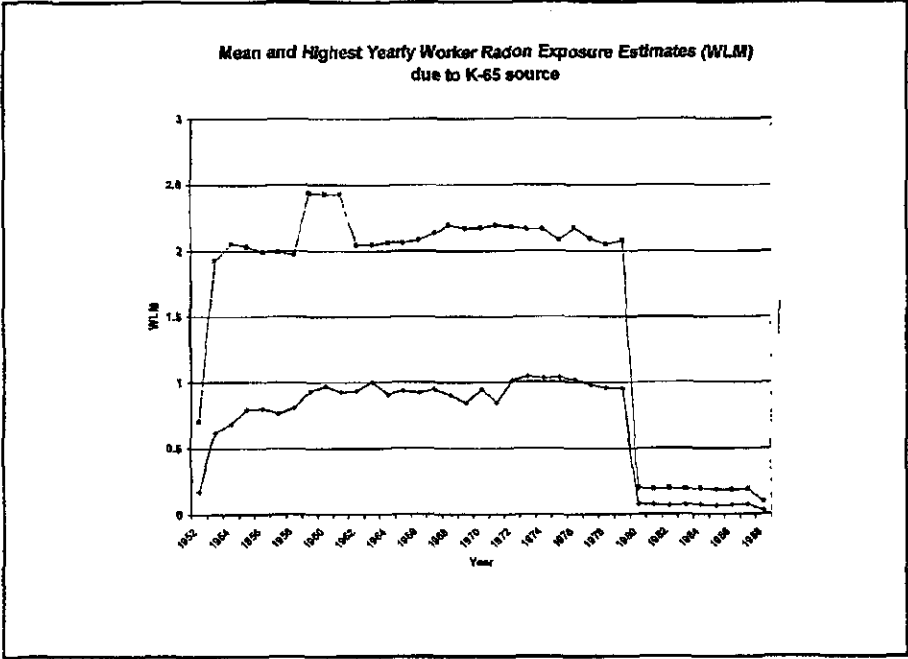


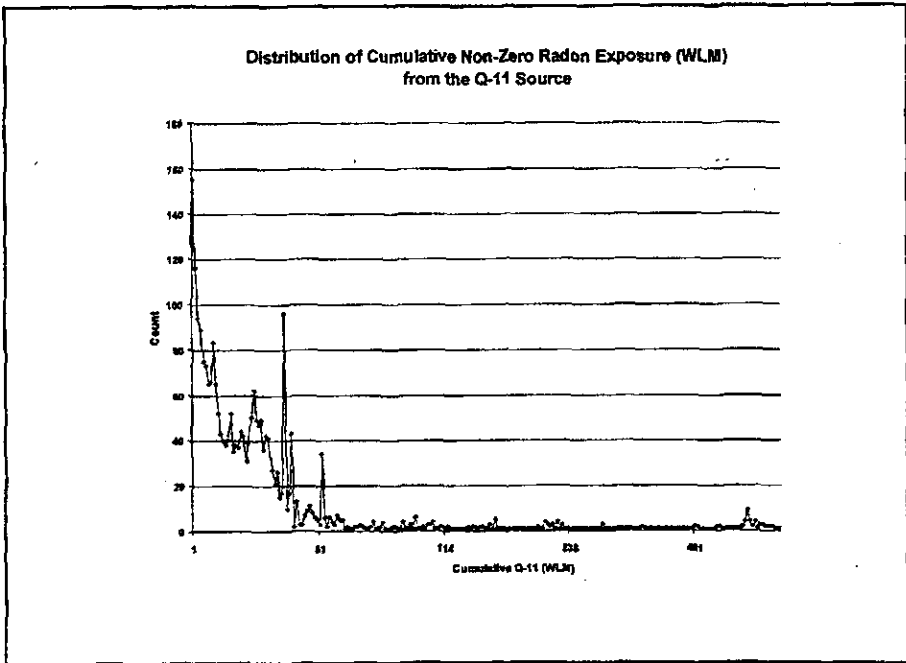
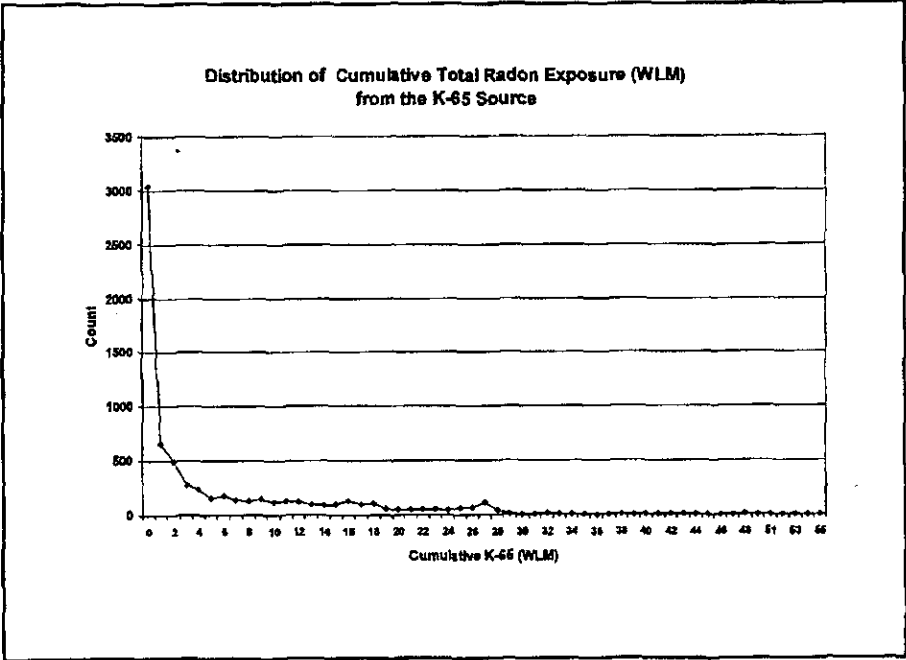
## Exposure Estimation Method

- Estimate radon exposure at selected site locations
  - Specific to calendar year
  - Specific to time of day (shift)

$$\text{K65 exposure} + \text{Q11 exposure} = \text{Total Radon Exposure}$$

- Place workers at site locations for each calendar year
- Assign a probability of working day, evening, night or rotating shift
- Link radon estimates with annual location assignments for each worker in the cohort





## Summary

- Deterministic model for K-65 source agrees reasonably well with the 1991 radon measurements (validation study using Carderelli data)
- CR-39 assays indicate a source of radon other than K-65, probably Q-11 silos, stored raffinate, local processing activities
- Final estimates assign high radon exposures to workers in the plant 2/3 area who were employed from 1952 to 1958
- The final product of this study is a matrix of radon exposures for each Fernald worker by year
- Exposures may be used in a future epi study or for compensation purposes under a current DOL / NIOSH program

## Value of FWMMP Occupational History Questionnaire and Interview Information

- **Use of the FWMMP occupational history questionnaire and interview information allowed us to:**
  - » Much more precisely assign worker location; workers assigned to multiple buildings rather than a single location
  - » Assign a probability of having worked the night shift, when exposures were 2-4 times higher
  - » Verify and validate our location and shift assignments
- Resulted in:
  - » Wider range of exposures and higher upper bound of the range
  - » For compensation: Worker assigned to multiple locations (each with a different exposure estimate) and shift (separate matrix for night shift)