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# Estimation of radon exposures to workers at the Fernald Feed Materials Production Center 1952–1988

RICHARD W. HORNUNG[a], SUSAN M. PINNEY[b], JEFFREY LODWICK[b], GEORGE G. KILLOUGH[c], DAVID E. BREWER[d] AND JAMES NASUTA[b]

[a]*Cincinnati Children's Environmental Health Center, Cincinnati Children's Hospital Medical Center, Cincinnati, Ohio, USA*
[b]*Department of Environmental Health, University of Cincinnati, Cincinnati, Ohio, USA*
[c]*Hendecagon Corporation, Oak Ridge, Tennessee, USA*
[d]*Institute for the Study of Health, University of Cincinnati Medical Center, Cincinnati, Ohio, USA*

The Feed Materials Production Center (FMPC) at Fernald, Ohio produced uranium metal products for use in Department of Energy defense programs. Radium-contaminated waste material was stored on-site in two K-65 silos on the west side of the facility and provided a source of $^{222}$Ra. The initial objective of this study was to estimate radon exposures to employees at FMPC working from 1952 to 1988. A modified Gaussian plume model was used to estimate exposures to workers. In an effort to validate these model-based estimates, we used 138 CR-39 film assays from window glass sampled in buildings throughout the site. Results from the CR-39 assays indicated a second substantial source of radon, the smaller Q-11 silos located in the production area. A response-surface regression analysis using a cubic spline model was fit to the CR-39 data to estimate $^{210}$Po surface activity levels at geographic coordinates throughout the facility. Knowledge of the age of the glass, the amount of contaminated waste in the Q-11 silos, and $^{210}$Po decay rates were used to estimate annual exposures to radon decay products (WLM: working level months). Estimated WLM levels associated with the Q-11 source term indicated that employees working in the vicinity during the period when they were filled with radium-contaminated waste (1952–1958) received substantially higher radon exposures than those from the K-65 source during this period. Results of the two models, corresponding to the K-65 and Q-11 sources, were combined to estimate WLM levels by year for each of the 7143 Fernald workers during the period 1952–1988. Estimated cumulative exposures to individual workers ranged from $<0.5$ to 751 WLM. Estimated radon exposures from this newly discovered source have important implications for future epidemiologic studies of lung cancer in workers at the Fernald facility.
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## Introduction

The Feed Materials Production Center (FMPC) at Fernald, Ohio produced uranium metal products for defense programs managed by the Department of Energy (DOE) and its predecessors. Radioactive waste from this process and waste shipped to Fernald from other DOE sites was stored in two silos on-site. These silos, code-named K-65 and located on the western edge of the production area, were constructed in 1951 to 1952. Waste 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 1950s. Because of these problems, periodic repairs and improvements to the silos were implemented from the 1960s through the 1980s.

Cragle et al. (1996) produced a report on the results of a cohort mortality study of white male workers at the FMPC site from 1951 to 1989. This study reported a statistically significant excess relative risk (RR) of 8.0 per Sv for lung cancer with a 10-year lag for external dose. No statistically significant relationship with lung cancer was detected for internal dose. These results were unexpected given the relatively low levels of external dose (mean = 14.8 mSv) and the lack of a dose–response with internal radiation, which generally has a stronger relationship to lung cancer. Internal dose, however, was estimated largely from urinalysis data measuring intake of soluble uranium compounds. A study of this same cohort by Ritz (1999) found a similar significant dose–response for the RR of lung cancer with external exposure, particularly among workers exposed after the age of 40 years (RR = 2.88, 95% CI = 1.29–6.44). Although the RR of lung cancer associated with internal dose (estimated from soluble uranium intake using urinalysis data) was elevated (RR = 1.92, 95% CI = 0.53–6.96), it again did not

1. Address all correspondence to: Dr. S.M. Pinney, Department of Environmental Health, University of Cincinnati, P.O. Box 670056, Cincinnati, Ohio 45167-0056, USA.
Tel.: +1 513 558-0684. Fax: +1 513 558-4240.
E-mail: susan.pinney@uc.edu
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reach the level of statistical significance. In 1998 a dose reconstruction study of residents surrounding the Fernald site, sponsored by the Centers for Disease Control and Prevention (CDC), was released (Killough et al., 1998). This study had initially been targeted at exposures to uranium and uranium compounds in air and water. However, the detailed examination of records and interviews with plant managers revealed that the K-65 silos on the west side of the plant contained waste, which was contaminated with a large quantity of $^{226}$Ra. This provided a continuous source of $^{222}$Rn through vents at the top of the silos and cracks in silo walls. The average volume of waste in the silos was estimated at over 200,000 ft$^3$ producing 170,000 Ci of radon from 1951 to 1988 (Killough et al., 1998).

The significant elevation in lung cancer mortality reported by Cragle et al. (1996) among Fernald workers, and the subsequent estimates of large quantities of radon emitted from the silos on-site, prompted the National Institute for Occupational Safety and Health (NIOSH) to fund a study to estimate radon exposures to Fernald workers from 1952 to 1988, the year when production ceased. This paper reports the results of this NIOSH-funded effort to estimate exposures to radon decay products for all Fernald workers first employed from 1952 through 1988. Accordingly, the initial objective of this research was the estimation of annual exposure rates for radon decay products measured in working levels (WL) for each individual worker employed for more than 3 months between 1952 and 1988. A WL is a measure of exposure rate to any short-lived $^{222}$Rn decay products. The term ''exposure rate'' is used throughout this paper to indicate activity in air of short-lived decay products of $^{222}$Rn measured in WL, which is the metric of choice for all epidemiologic lung cancer studies. One WL is defined as any combination of short-lived radon decay products in 1 liter of air resulting in the emission of $1.3 \times 10^5$ MeV of potential $\alpha$-energy. At 50% equilibrium, one WL is approximately equal to 7400 Bq/m$^3$ (200 pCi/l) of $^{222}$Rn (BEIR, 1988). In this study, a dynamic representation of radon decay products in the plume was used rather than a fixed equilibrium. Cumulative exposure to radon decay products is expressed as working level months (WLM), which is the product of WL and months of exposure.

## Methods

The original plan for estimation of radon exposure to Fernald workers involved the use of an atmospheric transport model that applied a modified form of the Gaussian dispersion model originally intended for estimation of exposure to residents living within a 10-km radius of the Fernald plant (Killough and Schmidt, 2000). This model was applied to estimate releases from waste material stored in the K-65 silos at the west side of the plant (Meyer et al., 1996).

The radon dispersion model was developed by Radiological Assessments Corporation (RAC) under contract to the CDC as part of the overall dose reconstruction that also included uranium and thorium doses from air and water transport. The model had been applied to the estimation of radon exposures to residents living near the facility. Details of the development of this model are given by Voillequé et al. (1995) and Killough et al. (1998) and are beyond the scope of this paper. We have, however, included an abbreviated description of the atmospheric dispersion model in Appendix A. The model was studied to determine what revisions would be required to make it applicable to the estimation of radon exposures to Fernald workers who were considerably nearer to the K-65 silos. The radon transport model involved emission rates from the silos, meteorological data, distance from the source, and decay rates of radon decay products. The output of the model was annual average exposure in either pCi/l or Bq/m$^3$ at any location specified by longitude and latitude coordinates. The original model was modified to produce estimates associated with the three shifts operated at the plant. The modification was based upon stratification of the hourly meteorological measurements into three periods corresponding to the three shifts worked by Fernald employees. This was necessary due to the substantial change in radon concentrations over a 24-h period. The early morning hours typically had higher concentrations of radon since wind speeds were generally lower and there was a higher potential for atmospheric inversions that trapped radon at ground level (Merrill and Akbar-Khanzadeh, 1998).

Since the model was capable of estimating annual radon concentrations for any location and each shift, the next step was to locate each worker's position within the facility and the shift that he worked. Personnel records did not contain information on location and shift worked, although these records did indicate codes for department and job title for periods of employment for each worker, and building for some. Preliminary studies indicated that, if this information were used to assign location, at least 10% of production workers would be misclassified for their primary location, and no consideration would be given to other buildings in which they worked. Therefore, a set of assumptions were developed to assign location for each year of employment. Location was rarely a single building, but usually a set of buildings, time weighted for use in exposure estimation.

Supplementary information on locations and shifts was available from questionnaires used in the Fernald Workers Medical Monitoring Program and through structured interviews funded through this study. Historical records and information obtained through personnel records, questionnaires, and interviews were used to develop the assumptions to assign shift and worker location for various combinations of building, department, and job title noted on the personnel record for each period of employment. The Fernald site roster originally obtained from NIOSH listed 7143 persons,

employed at the site during the years of operation from 1952 to 1988. Of the 7143 workers noted on the file, 2231 were known to be deceased and we were unable to obtain a mailing address for 1601 (some of whom were probably deceased). Mailed questionnaires were sent to the remaining 3464, and 2666 (77%) completed questionnaires were returned after postcard and phone reminders. Upon receipt of a completed questionnaire, the worker was contacted by phone to obtain additional information about job tasks, exposures, and incidents. Specific questions about shift worked (day, evening, night, or rotating) during different time periods of employment were included only about midway in the questionnaire administration and, therefore, data regarding shift work were available for only 1207 workers. Workers were asked specifically about plants or buildings in which they worked, and what proportion of their total workday they actually spent in that building. Questions on job tasks also elicited information about work performed at locations other than the primary assigned production or work area.

The worker locations (based on responses from 2666 workers) and shift assumptions (based on responses from 1207 workers) derived from the data were then applied to all 7143 workers on the plant roster, for each period of the worker's employment linked by building, department, job title, and year. For some workers, location for a given year was defined by the primary building in which each employee was working in that year, as noted on the personnel records. For others, the combined job title and department information was used to assign them to several buildings for each period of employment, with the time weighted using assumptions derived from the questionnaire and interview data. Some mobile workers (maintenance, security, etc.), who could not be assigned to a specific set of buildings, were assigned as a plant-wide average of the buildings in their general work area, weighted for the number of persons in those buildings during that year. Shift was assigned on the basis of standardized job titles. The questionnaire data regarding shift worked for specific job periods were used to determine a set of proportions for each job title or cluster of similar job titles. For example, based on all responses from chemical operators, we assigned 35% of their exposures to day shift, 26% to the second shift, 16% to the third shift, and 23% to rotating shifts. These proportions were used as weights for shift-specific exposure estimates.

After locations for each worker in each year and the shift worked were determined, the estimated exposure at that location for the shift worked was assigned. When a worker was assigned to multiple buildings in a given year, a time-weighted average exposure was calculated. The ultimate result was a matrix with rows corresponding to workers and columns corresponding to years from 1952 to 1988, where each cell $(i, j)$ was the estimated exposure rate in either $Bq/m^3$ or WL for worker $i$ and year $j$.

*Validation of model estimates*

The original Gaussian plume model was calibrated to outdoor measurements of radon made at various locations on the site during the years 1981 through 1987 (Killough and Schmidt, 2000), with estimated emission rates from the K-65 silos as input. Before 1979, when vent pipes were removed and the silo domes were capped, very few radon measurements were available. The RAC dose-reconstruction team made a thorough search for any historical measurements that could have been used to calibrate their model, but the era of highest exposures (1952–1979) was difficult to validate. We sought a way of validating our exposure estimates, particularly before 1979. Several case–control studies of radon-related lung cancer from exposures received in homes (Field, 2001; Steck et al., 2002) employed a method for estimating cumulative exposure. This method involved the use of CR-39 plastic film applied to glass in mirrors or framed pictures that were kept in each home that a case or control inhabited over their adult lifetime. The CR-39 film records tracks from $^{210}$Po $\alpha$-particles that decay from $^{210}$Pb which is a long-lived (22 years half-life) decay product of $^{222}$Rn imbedded in the glass. Counts of tracks from the CR-39 plastics can be used to estimate integrated radon exposure over long periods of time (Lively and Steck, 1993; Mahaffey et al., 1993).

Since CR-39 had been primarily used in laboratories and homes, a test of its feasibility in a uranium processing facility was necessary. Eight CR-39 films were obtained from Track Analysis Systems Ltd. (TASL) in Bristol, England. They were placed on the inside and outside of glass window panes in a building near the center of the production area and in a building near the eastern edge of the plant farther from the K-65 silos. Four replicates were made by placing films side-by-side on the same pane of glass. These films were exposed for approximately 2 weeks in November 1999 and then sealed and sent to TASL for analysis with no indication of where they had been placed. Upon receipt of the exposed CR-39 films, TASL etched the films in a solution of NaOH, calibrated them with a $^{252}$Cf fission source, and scanned them with an automated image analysis system. Surface activity of $^{210}$Po is measured with corrections for lower energy particles from $^{234}$U, $^{235}$U, and $^{238}$U and similar lower energy $\alpha$-particles (Fews and Henshaw, 1982).

Higher energy contamination ($^{214}$Po, $^{218}$Po, and $^{212}$Po) was eliminated from track counts and did not cross contaminate $^{210}$Po readings.[2] The actual measurements were made by etching and scanning TASTRAK $\alpha$-particle sensitive CR-39 plastic, followed by spectroscopic analysis of associated $\alpha$-tracks (Fews, 1992). When track density was extremely high, tracks were counted by eye using averages of 10 microscope image frames randomly distributed over the

[2]A.P. Fews. Report to NIOSH on the surface concentration of Po-210 on window glass of a Uranium processing plant, May (2001).

sample. Results for all samples were reported as total $^{210}$Po activity per unit area in Bq/m$^2$.

In addition to the use of CR-39 film for validation of the air dispersion model, a set of radon measurements were discovered that were not used to validate the model when it was developed to estimate off-site radon levels for residents near Fernald. These data were collected for 7 months from March to September 1991 by a student for a Master's degree thesis (Cardarelli, 1992). Samples were taken around the K-65 silos and in the vicinity of five buildings on the Fernald site. The protocol for the Master's thesis called for a survey using three monitors simultaneously: Pylon monitors, Radon Gas monitors (RGM II), and Femto-tech monitors, all three located at each measurement site. The Pylon monitor utilizes passive diffusion of radon gas into a scintillation chamber where $\alpha$-particles are counted. The RGM II utilizes a pump to draw air through a particulate filter into a scintillation chamber. The Femto-tech monitor is similar to the Pylon monitor in using passive diffusion, but it employs a pulsed ion-sensing chamber instead of a scintillation chamber. The data available for our validation consisted of the arithmetic mean (AM) of the three monitor results for each hour. The hourly means were then used to calculate geometric means (GMs) for each 8-h shift across the 7 months when sampling was done since the distribution of radon levels over time was right-skewed.

*A new radon source*

When the results of our pilot study were returned from TASL, two unexpected findings were revealed. Since these films were potentially exposed to a much higher background of $\alpha$-radiation than found in homes, replicate results were expected to have a high degree of variation. However, the measures at both locations agreed to within 15%, although only based on four replicates. Samples placed near the center of the plant closer to the K-65 silos were expected to have higher radon exposures by approximately a factor of 2. Instead, the measured $^{210}$Po levels in the production area were over eight times higher than the samples placed at the eastern edge of the plant (mean = 30.8 Bq/m$^2$ in the production area and 3.6 Bq/m$^2$ at the eastern edge). This indicated that there was likely to have been another source of radon near the production area.

Interviews with retired Fernald employees who had worked at the site since the beginning of production revealed that six smaller silos in the production area had been used in the 1950s to store high-grade ore obtained from the African Metals Corporation before processing. This ore had a very high content of $^{226}$Ra and therefore a high potential for emission of radon and its decay products. 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. In addition, drums of similar waste product from Mallinkrodt Chemical Works were stored on a pad directly north of the Q-11 silos. The RAC Task 2/3 report (Voilleque et al., 1995) noted this potential, but concluded that the smaller volume of the Q-11 silos would preclude them from being a major source of radon to the neighborhoods surrounding Fernald. However, the close proximity of the Q-11 silos to the main production area made a more detailed assessment using the CR-39 films, an important next step.

During October and November 2000, 110 CR-39 films were strategically placed to cover areas of the facility where most workers were assigned. Films were placed on both the exterior and interior of each pane of glass to estimate whether radon exposures were elevated inside buildings as well as in the ambient air outside. The films were exposed to the glass from 19 to 21 days (452–502 h), the exact time of exposure was recorded, and the films were then sealed and sent to TASL for analysis. On the basis of these results, 20 additional films were placed in December 2001 in the highest exposed areas to provide additional coverage and precision to our estimates in the production area.

When the CR-39 assay results were returned to the study team, the 138 sample results were used to estimate $^{210}$Po surface activity levels (Bq/m$^2$) at each location in the plant. Because of the need to estimate radon exposures at worker locations that were not directly measured, a two-dimensional response surface multiple regression model was developed to predict $^{210}$Po surface activity by grid coordinates and calendar year, both inside and outside buildings on the Fernald site. The model assumed that the primary exposure source was the Q-11 silos. Therefore, the distance and direction from this point to each building occupied by workers for any calendar year was calculated. Following the method used in the atmospheric dispersion model associated with the emanations from the K-65 silos (Killough et al., 1998), we calculated a term incorporating both wind speed and direction using meteorological data from an on-site tower. This was done by dividing the wind rose for the Fernald site into eight sectors and calculating a weight as shown below:

$$W_i = P_i/\mathrm{WS}_i$$

where $P_\mathrm{i}$ = proportion of time the wind is blowing toward the $i$th sector;
$\mathrm{WS}_i$ = average wind speed in m/sec in sector $i$, $i = 1$–8

The wind rose was centered at the location of the Q-11 silos. Each glass sampling location was located by latitude and longitude coordinates in one of the eight sectors and the corresponding weight was assigned. The primary term in the regression model was then the distance from the CR-39 sampling location to the Q-11 silos divided by the appropriate weight for that location. Other terms in the model were building construction year and whether the sample was taken inside or outside the building. A term for building construction year was necessary for inclusion in the

regression model because the amount of time the building glass was exposed to radon decay products was associated with the measured $^{210}$Po surface activity recorded on the CR-39 film. The model was fit using a restricted cubic spline model for weighted distance from the Q-11 source (Stone and Koo, 1985). The spline model was used to produce a smoothed nonlinear relationship between measured $^{210}$Po surface activity and weighted distance to each building. The spline function is a piecewise cubic polynomial fitted in six intervals of the weighted distance (defined by seven nodes), which can be estimated with six coefficients. The general form of the model was as below:

$$\ln(^{210}\mathrm{Po}) = \beta_0 + \beta_{1-6}(F(d)) + \beta_7 \mathrm{yr} + \beta_8 S + \varepsilon,$$

where $F(d)$ is a seven-node restricted cubic spline function of weighted distance, yr = year glass was installed, $S$ = an indicator of whether the sample was on the inside or outside of the window pane, and $\varepsilon$ is a random error term. The natural log of $^{210}$Po surface activity in Bq/m$^2$ was used as the dependent variable since these data were highly right-skewed. Owing to the potential for spatial correlation in such a response surface model, the residuals were examined for residual spatial autocorrelation that might remain after the covariate-adjusted weighted spline model was developed. All statistical analyses were conducted using SAS, version 9.1 (SAS Institute Inc., 2006).

The estimated $^{210}$Po levels obtained from the multiple regression model were then used to predict the exposure rate to radon decay products in WL for each worker location and calendar year. This was done by assuming that the estimated $^{210}$Po surface activity in the year of the assay was equal to the product of total WL for each year, the deposition rate, and the decay of $^{210}$Pb since the year of deposition:

$$[^{210}\mathrm{Po}]_{i,2000} = \sum_{t=t_{0,i}}^{2000} \eta_i [\mathrm{WL}_{\mathrm{K65},i,t} + \mathrm{WL}_{\mathrm{Q11},i,t} + \mathrm{WL}_{\mathrm{bg}}] \mathrm{e}^{-\lambda(2000-t)}$$

where, $[^{210}\mathrm{Po}]_{i,2000}$ is activity of $^{210}$Po embedded in glass surface (Bq/m$^2$) for building $i$ at the time of CR-39 analysis (2000), $\mathrm{WL}_{\mathrm{K65},i,t}$ is the average WL for year $t$ and building $i$ due to estimated K-65 radon, $\mathrm{WL}_{\mathrm{Q11},i,t}$ is the average WL for year $t$ and building $i$ due to the Q-11 source, $\mathrm{WL}_{\mathrm{bg}}$ is the average background radon, which is assumed here to be 0.003 WL, $\lambda$ is the decay rate of $^{210}$Pb ($3.11 \times 10^{-2}$ per year), $t_{0,i}$ is the year that building $i$ was constructed (i.e. glass exposure began), $\eta_i$ is the rate at which $^{210}$Pb (expressed as $^{210}$Po surface activity, $(\mathrm{Bq/m^2})_{\mathrm{Po-210}}$) became embedded into the glass surface per WL of exposure, $(\mathrm{Bq/m^2})_{\mathrm{Po-210}}/\mathrm{WL}$, for building $i$.

$\eta_i$ is estimated using the following equation:

$$\eta_i = \frac{[^{210}\mathrm{Po}]_{i,2000}}{\sum_{t=t_{0,i}}^{2000} [\mathrm{WL}_{\mathrm{K65},i,t} + \mathrm{WL}_{\mathrm{bg}}] \mathrm{e}^{-\lambda(2000-t)}}$$

$\eta_i$ was estimated for buildings where Q-11 contribution to glass radon exposure was assumed to be negligible. Values of $\eta_i$ for each sampling location were averaged to get an average deposition rate $\eta$ in $(\mathrm{Bq/m^2})_{\mathrm{Po-210}}/\mathrm{WL}$. This approach to the estimation of the deposition rate was necessary due to the special characteristics of our study. In previous studies using CR-39 plastics to predict historical radon levels in houses, the deposition rate was estimated using radon chambers (Lively and Steck, 1993). This was not applicable to our situation when there was an open-air source with deposition occurring over 1000 m in the distance.

The average deposition rate was applied to the locations assumed to be affected by Q-11 radon to estimate yearly WL for each year $t$ at building $i$ using the following equation:

$$\sum_{t=t_{0,i}}^{2000} (\mathrm{WL}_{\mathrm{Q11},i,t} e^{-\lambda(2000-t)}) = \frac{[^{210}\mathrm{Po}]_{i,2000}}{\eta} - \sum_{t=t_{0,i}}^{2000} (\mathrm{WL}_{\mathrm{K65},i,t} + \mathrm{WL}_{\mathrm{bg}})\, \mathrm{e}^{-\lambda(2000-t)}$$

For any specific year $t_k$

$$\mathrm{WL}_{\mathrm{Q11,i,t_k}}\, \mathrm{e}^{-\lambda(2000-t_k)} = \omega_{t_k,i} \sum_{t=t_{0,i}}^{2000} \mathrm{WL}_{Q11,i,t}\, \mathrm{e}^{-\lambda(2000-t)}$$

$$\mathrm{WL}_{\mathrm{Q11,i,t_k}} = \frac{\omega_{t_k,i} \left( ([^{210}Po]_{i,2000}/\eta) - \sum_{t=t_{0,i}}^{2000} (\mathrm{WL}_{K65,i,t} + \mathrm{WL}_{bg}) \mathrm{e}^{-\lambda(2000-t)} \right)}{\mathrm{e}^{-\lambda(2000-t_k)}}$$

where $\omega_{t_k}$ is a dimensionless weighting factor for the normalized mass of radium ($m_{t_k}$) estimated to be in the Q-11 silos in year $t_k$ to the total mass of radium processed through the Q-11 silos, and for the ratio of the fraction of $^{210}$Po that is left from year $t_k$ to the total amount of $^{210}$Po that is left after decay. The term $\omega_{t_k}$ is based on the product of the mass of material reported to be in the silos (Q-11 ore or metal oxide material) and the reported radium concentrations of the respective material type in the silos and from the decay of $^{210}$Pb. The reported mass of material was taken from Fernald reports, internal memos, and graphs of material volume at various years from 1952 to 1960 (Voillequé et al., 1995). This factor was necessary to weight for the Q-11 contribution ($M_{t_k}$) and radioactive decay for year $t_k$ of exposure relative to other years of exposure.

$$\omega_{t_k,i} = \frac{M_{t_k} \mathrm{e}^{-\lambda(2000-t_k)}}{\sum_{t=t_{0,i}}^{2000} M_t \mathrm{e}^{-\lambda(2000-t)}}$$

$$M_{t_k,i} = \frac{m_{t_k}}{\sum_{t=t_{0,i}}^{2000} m_t}$$

The estimates of mass stored in the Q-11 silos were taken from internal reports indicating the weight in pounds at the end of each year from 1952 to 1958. The Q-11 silos were not sealed and therefore the emission rate of radon was assumed

**Table 1.** GMs and AMs and SD for cumulative radon exposures (WLM) for individual workers by calendar period and source.

| | | Source: K-65 | | | | | Source: Q-11 | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Calendar period | No. of workers | GM | GSD | AM | SD | Range | GM | GSD | AM | SD | Range |
| 1952 | 1406 | 0.24 | 1.58 | 0.17 | 0.13 | 0.001–0.70 | 0.34 | 7.77 | 3.34 | 7.14 | 0–45.4 |
| 1953 | 1969 | 0.59 | 1.97 | 0.62 | 0.40 | 0.001–1.93 | 0.86 | 9.03 | 5.63 | 11.84 | 0–92.8 |
| 1954 | 2695 | 0.68 | 1.82 | 0.69 | 0.40 | 0.004–2.05 | 0.51 | 6.89 | 5.06 | 16.17 | 0–92.2 |
| 1955 | 2967 | 0.81 | 1.65 | 0.79 | 0.36 | 0.003–2.03 | 0.69 | 8.41 | 7.38 | 23.32 | 0–133.5 |
| 1956 | 3223 | 0.82 | 1.61 | 0.80 | 0.35 | 0.003–1.99 | 1.23 | 12.30 | 10.99 | 22.98 | 0–133.9 |
| 1957 | 3012 | 0.76 | 1.73 | 0.76 | 0.37 | 0.002–1.99 | 0.73 | 8.94 | 8.14 | 24.12 | 0–130.0 |
| 1958 | 2646 | 0.84 | 1.54 | 0.81 | 0.33 | 0.002–1.97 | 0.51 | 7.17 | 6.75 | 22.91 | 0–113.8 |
| 1959 | 2647 | 0.93 | 1.65 | 0.93 | 0.42 | 0.001–2.44 | 0.02 | 1.65 | 0.05 | 0.20 | 0–1.18 |
| 1960–1964 | 2906 | 0.95 | 1.64 | 0.95 | 0.42 | 0.002–2.43 | 0.02 | 1.45 | 0.03 | 0.14 | 0–1.18 |
| 1965–1969 | 2173 | 0.90 | 1.73 | 0.91 | 0.43 | 0.002–2.20 | 0.01 | 1.01 | 0.01 | 0.01 | 0–0.01 |
| 1970–1974 | 1275 | 0.94 | 1.74 | 0.97 | 0.46 | 0.002–2.19 | 0.01 | 1.01 | 0.01 | 0.01 | 0–0.01 |
| 1975–1979 | 832 | 0.99 | 1.60 | 0.99 | 0.42 | 0.001–2.17 | 0.01 | 1.01 | 0.01 | 0.01 | 0–0.01 |
| 1980–1984 | 1401 | 0.07 | 1.18 | 0.07 | 0.02 | 0.001–0.20 | 0.01 | 1.01 | 0.01 | 0.01 | 0–0.01 |
| 1984–1988 | 2021 | 0.06 | 1.21 | 0.06 | 0.03 | 0.001–0.19 | 0.01 | 1.01 | 0.01 | 0.01 | 0–0.01 |

AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation; WLM, working level months.

to be directly proportional to the mass of contaminated waste in the silo in any given year. The total amount of Q-11 material processed was approximately 2424 ton ($2.2 \times 10^9$ g). Between 1958 and 1962, hot metal oxide was stored in the Q-11 silos, but this material produced less than 1% of the $^{226}$Ra emissions of the Q-11 material.[3] After 1962, there was a residual of approximately 8 ton ($7.3 \times 10^6$ g) of metal oxide that remained until the silos were removed in 1990.[4]

## Results

The air dispersion model for K-65 radon emissions was applied to coordinates at the centroid of each building on the Fernald site where workers had been assigned. The results were generated in pCi/m$^3$ units on an annual basis for each of the three shifts. The estimated radon exposures were highest from 1952 to 1979, the year when the K-65 silos were capped. After that time, the estimated levels dropped by a factor of 3.8 for the day shift, 7.6 for the evening shift, and 19.5 for the night shift. The differences in the degree of reduced exposure were due to an assumed ''solar pumping'' effect during daylight hours after 1979. This was due to heating of the silos causing pressure to rise and release greater amounts of radon, in daylight hours after 1979 through cracks in the capped silo domes. These values were linked to the individual work histories to produce yearly and total cumulative estimates of radon exposure for each worker in WLM units. Table 1 shows the GM, geometric standard deviation (GSD), AM, and SD, and range of cumulative annual radon exposures (WLM) estimated for individual workers by calendar period from the K-65 source. Typical annual exposure levels as characterized by the GM ranged from 0.24 to 0.99 WLM until 1979 when the K-65 silos were sealed. At that time, the average exposures dropped by approximately an order of magnitude. The maximally exposed workers from the K-65 source during any period received slightly more than 2 WLM until the silos were capped.

Figure 1 presents a comparison of the dispersion model estimates for 1988 with the radon levels measured in 1991 reported in the Cardarelli thesis, by shift and location. The meteorological data used for the model predictions (a composite joint frequency table of 1986–1991 hourly data recorded on site) gives a somewhat wider separation of predictions for the three shifts at each building than the corresponding measurements show. However, all but two model predictions lie within the indicated 5th to 95th percentile interval of the measurement data. If the error variability among buildings is judged by the intermediate second shift value, all five building predictions are within a factor of two of the corresponding second shift observations (although some predictions for other shifts lie outside the factor of two interval). The scatter in the on-site calibration data for the model (Figure A2 in Appendix) would not lead one to expect better overall agreement than this.

### *CR-39 assay analysis*

The 138 CR-39 assays from window glass around the Fernald site were used to develop a weighted response surface model using a restricted cubic spline function. The $R^2$ for the spline model was 0.53, which indicates that the multiple regression model using this smoothing function explained 53% of the variability in the measured $^{210}$Po surface activity levels. We used the model to smooth multiple measurements

[3]1962 memo by J.J. Costa.
[4]memo: ''Removal Site Evaluation, Plant 1 Ore Silos,'' August (1991).

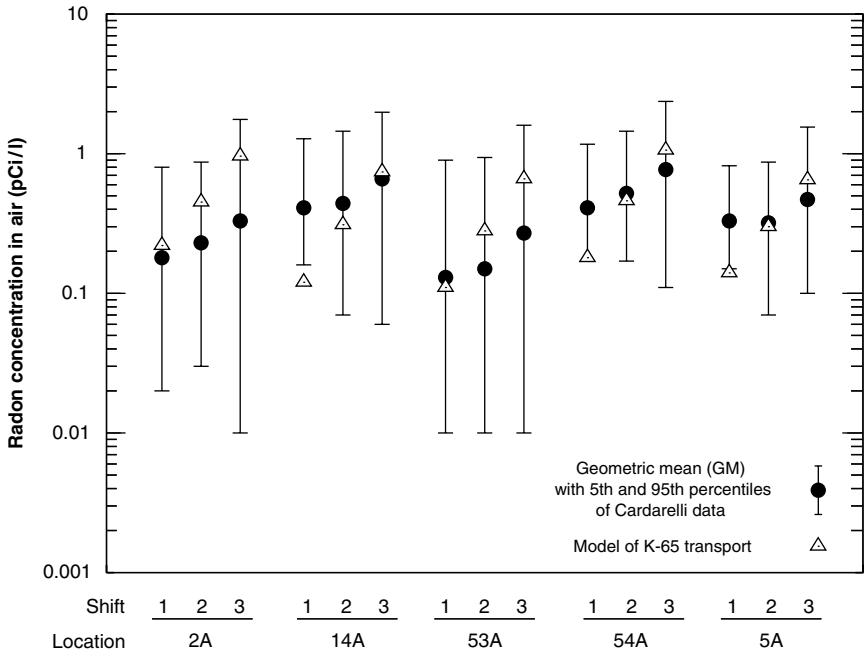


**Figure 1.** The figure indicates the comparison of Gaussian plume model estimates with radon measurements (pCi/l).

on the same building and to predict $^{210}$Po levels for buildings not measured based on their distance and direction from the Q-11 silos. Examination of possible residual spatial autocorrelation revealed that the correlation of residuals to their nearest neighbor in our distance metric was very low and nonsignificant ($r = 0.10$, $P = 0.25$). We, therefore, concluded that residuals could be considered spatially independent with no apparent bias in model coefficients. The estimates for the inside and outside $^{210}$Po levels in each building were calculated as GMs by exponentiating the predicted mean of the log-transformed data. We also produced estimates of the AMs by using the relationship: $\mathrm{AM} = \exp(\mu + 0.5\sigma^2)$, where $\mu$ and $\sigma^2$ are the mean and variance of the logs, respectively. The difference between inside and outside measurements was not statistically significant ($P = 0.16$), although the inside measurements were on average 32% lower than that of the outside measurements. This was initially somewhat puzzling since indoor radon levels in houses are usually higher than outside levels. Upon investigation with Fernald workers, we were told that windows and large doors in these buildings were often left open. This would cause a high degree of ventilation indoors which lowers radon levels. Since the source is thought to be the Q-11 silos external to the buildings, it became understandable that outside glass surfaces would be embedded with as much or more Pb-210. This also provided additional assurance that the radon source was the Q-11 silos and not the result of production activities inside the buildings. Inside estimates were used since most workers were exposed on the interior of each building. Although GMs are more appropriate descriptors for lognormal data, AMs are generally more appropriate for calculating cumulative exposures used in epidemiologic studies (Seixas et al., 1988), which is the intended use for these estimates. Using the same method as employed for the K-65 source estimates, these values were linked to the individual work histories to produce a cumulative estimate of radon exposure for each worker in WLM units. Table 1 presents the GM, GSD, AM, and SD, and range of cumulative annual radon exposures (WLM) estimated for individual workers by calendar period from the Q-11 source. The principal contribution of the Q-11 silos to the radon exposure for Fernald workers ended in 1958 when the silos were substantially emptied. A much smaller amount of hot metal oxide waste material was stored in the Q-11 silos from 1958 until 1962 after which less than 1% of the waste remained until the silos were removed in 1990. Inspection of Table 1 indicates that before 1958 radon exposures to workers in the production area near the Q-11 silos were dominated by this source compared to the K-65 source. For workers employed in buildings in that area, radon levels attributed to Q-11 ranged from 0.91 to 15.25 WL from 1952–1958 during the day shift and from 4.0 to 66.4 WL during the third shift. These estimated exposures are approximately 100-fold greater than radon levels attributed to the K-65 silos during that period. Figure 2 shows a contour plot of the response surface analysis for estimated $^{210}$Po levels from the Q-11 source. Buildings near the periphery of the plant were assigned no exposure from the Q-11 source because CR-39 assay results were at background levels and since these buildings were needed to calibrate results for buildings in the production area near the Q-11 silos.

After estimates of radon levels were obtained individually for the K-65 and Q-11 sources, the resulting levels were summed to obtain a total radon exposure level for each worker in each year that he was employed. As mentioned earlier, radon levels from 1959 to 1988 were assumed to be primarily attributable to the emanations from the K-65 silos. Table 2 presents the distribution of estimated cumulative radon levels in WLM from both the sources from 1952 to 1988 for all Fernald workers. The mean cumulative radon exposure for all 7143 workers was 24 WLM, the median was 2.9 WLM, and the exposures ranged from 0 to 751 WLM. The distribution is highly right-skewed with the highest decile of radon-exposed workers ($n = 715$) having a mean exposure of 167 WLM ranging from 46 to 751 WLM. Cumulative radon exposures in this decile came largely from workers exposed during the 1952–1958 period in the buildings near the Q-11 silos, with a mean contribution of over 76% from this source.

## Discussion

The original objective of this exposure assessment was to use a modification of the Gaussian plume model (K-65 silos as the source), developed originally for estimation of radon exposure to residents near Fernald, to estimate exposure to Fernald workers. The rationale was that radon exposures to workers inside the fenceline would be substantially higher than those to residents up to 10 km from the source, and possibly at epidemiologically relevant levels. Since very few measurements were made before 1979 when the silos were capped, we sought a method for validating the air dispersion model. The deployment of CR-39 films on window glass across the facility revealed an important second source of radon located near the production area. To our knowledge, the CR-39 assays have never been used before in an industrial setting. Replicate samples and the pattern in the levels of $^{210}$Po measured by the films throughout the plant indicated that CR-39 assays were reasonably reliable measures of cumulative radon concentrations during the periods when window glass was exposed. The laboratory conducting the assays expressed confidence that the lower energy $\alpha$-particles from isotopes of uranium ($^{234}$U, $^{235}$U, and $^{238}$U) could be accurately discriminated from the higher energy tracks associated with $^{210}$Po.[5] However, it is not possible to validate these assays directly since very few actual radon measurements were made in the early years of facility operation.

Upon further investigation and interviews with retired employees, likely sources of these elevated $^{210}$Po measures were the Q-11 silos near the production area where the highest measures were recorded. The accuracy of the response surface model used to smooth the $^{210}$Po measures across the two-dimensional plant grid depended to some extent on the location of the source. However, a detailed examination of plant records and process locations suggests that the only other possible radon source was a waste drum storage pad near the Q-11 silos. Minor shifts in the source location, if the drum storage contributed to the radon emissions, would have correspondingly small changes in estimated $^{210}$Po concentrations throughout the facility.

Another source of uncertainty in our cumulative radon estimates was the method used to calculate radon concentrations occurring up to 48 years earlier. These calculations involved assumptions that $^{210}$Po measurements at buildings distant from the Q-11 source (approximately 1000 meters) were attributable only to K-65 silo emissions and background radon. Results for these distant buildings were then used to calibrate our estimates. $^{210}$Po measurements in distant buildings were similar to background measurements (natural background plus K-65 emissions), which provided some assurance that they were relatively unaffected by the Q-11 source.

Because it is not possible to quantify the effect of these uncertainties on our calculations, the degree of bias or lack of precision that may be present in our cumulative radon estimates cannot be estimated. The multiplicative SE (GSD) in the weighted multiple regression model was slightly less than 3.0, suggesting that these estimates would have an approximate uncertainty of a factor of 6, where uncertainty is calculated as the ratio of the 95th percentile to the median.

Despite the uncertainties in these estimates, it seems clear that a significant source of radon existed other than the K-65 silos. Interviews with retirees and historical documents confirm that the six Q-11 silos contained waste contaminated with $^{226}$Ra. They also indicate that the Q-11 silos contained a substantial source of radon until they were emptied near the end of 1958.

This source of radon exposure primarily affects the 2576 workers who were first employed between 1952 and 1958 and who worked in the production area in the vicinity of the Q-11 silos. Their mean cumulative exposure from both the Q-11 and K-65 sources was 59.7 WLM with a range of 0.03 to 751 WLM. These cumulative exposures to radon decay products are in a similar range to those of several epidemiologic studies of underground uranium miners. For example, the studies of Canadian, French, Swedish, and Australian uranium miners reported mean radon exposures ranging from 7.6 to 80.6 WLM, and each of these studies showed a positive exposure–response relationship between radon and lung cancer (Lubin et al., 1994). These results have an important implication with respect to future studies of this population. A possible future epidemiologic study of lung cancer among Fernald workers may resolve the apparent paradox in earlier studies showing effects of external radiation rather than a significant relationship with internal

[5]P. Fews. Personal communication, February (2002).

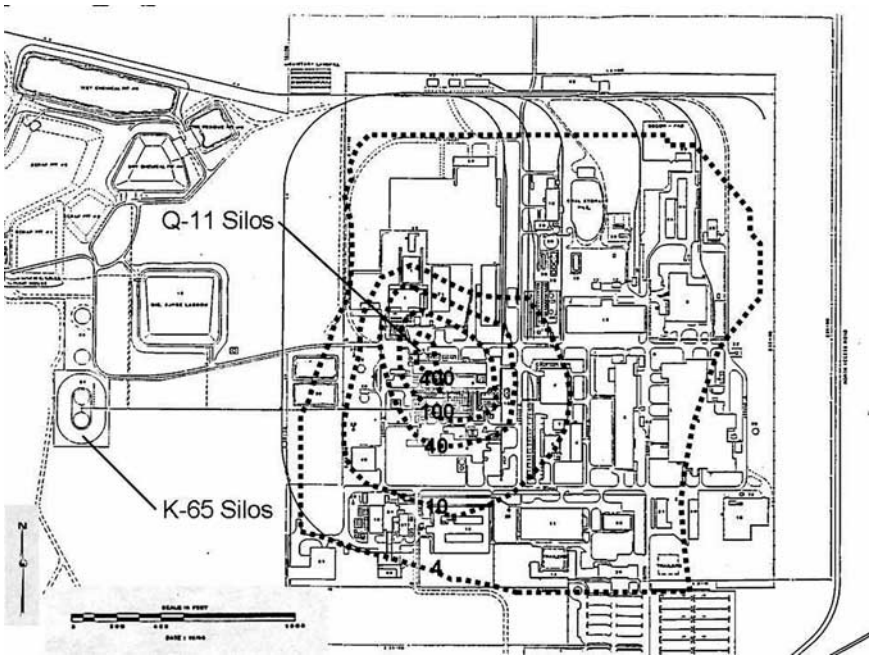


**Figure 2.** The figure indicates the contour plot of estimated $^{210}$Po levels for CR-39 assays from Q-11 source (Bq/m$^2$).

**Table 2.** Distribution of total cumulative exposure (1952–1988) to radon decay products from K-65 and Q-11 sources.

| WLM | No. of workers | Percentage (%) |
|---|---|---|
| <0.5 | 2288 | 32.0 |
| 0.5–5 | 1708 | 23.9 |
| 5–10 | 599 | 8.4 |
| 10–20 | 833 | 11.7 |
| 20–40 | 852 | 11.9 |
| 40–80 | 528 | 7.4 |
| >80 | 335 | 4.7 |

WLM, working level months.
$N = 7143$; mean = 24.2; median = 2.9; and range = 0.001–751.

emitters. In the event that an epidemiologic study is conducted to update the results of Cragle et al.'s (1996) study, the additional 10 years or more of vital status follow-up and the individual worker estimates of radon exposure enhance the probability of detecting a significant exposure–response relationship between internal radiation and lung cancer. These results may also partially explain the unexpected dose–response with external radiation found in the Cragle et al.'s (1996) study. The external radiation doses measured by personal dosimeters would likely be correlated with radon exposures since the highest external radiation exposures were associated with workers in the production area near the Q-11 silos.

In summary, the initial intent of this study was to adapt the Gaussian plume model from the K-65 silos, originally developed to estimate radon exposures to residents living within 10 km of the Fernald facility, for use in estimating radon exposures to Fernald workers employed from 1952 until 1988 when operations ceased. In an effort to validate this model, we identified an unexpected second significant source of radon exposure using CR-39 film applied to window glass throughout the facility. The second source was determined to be the Q-11 silos near the production area in the center of the facility. This source dominates the total radon exposure from both sources during the period 1952–1958 for workers in the vicinity of the Q-11 silos. The cumulative exposure to radon decay products from this previously unrecognized radon source has important implications for future epidemiologic studies of workers at the Fernald site.

## Acknowledgements

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## Appendix A

*Radon transport model from K-65 silos to receptor locations*

This appendix presents a brief summary of the models applied to the release and atmospheric transport of radon and progeny from the K-65 silos. The background for these models is extensive and complex, and space limitations preclude the inclusion of details here. The models are discussed more fully in the references. Please note that the SI units given in this appendix are intended to be a generic aid to the reader and may not necessarily be the units of corresponding quantities that were used in the simulations or presented in the results.

The Fernald Dosimetry Reconstruction Program, sponsored by the CDC, developed models of release history and atmospheric transport for radionuclides that were released from the former FMPC. Components of these models were calibrated to measurements that had been taken on the FMPC site and in the vicinity. Models specific to radon and its progeny were applied to releases from the K-65 silos and their atmospheric transport, and to the estimation of consequent air concentrations of these radionuclides at remote locations and dose (or WL) for exposed individuals at those locations (Killough et al., 1998).

The K-65 source term model is an empirical representation of the annual release of radon from the K-65 silos from 1952 through 1988. It was calibrated to a variety of data, primarily (1) the concentration of radon in the silo headspace gas, measured in 1987, (2) measurements of $\gamma$-ray exposure on the external silo dome surfaces before and after they were sealed, and (3) a series of temperature and pressure readings in the silo headspace gas, taken in 1987. It is a stochastic model, with some parameters derived as probability distributions, which propagate into the annual release estimates. Detailed information about the K-65 source term model and the ratio of decay products at the point of release is given in Appendix Q of Killough et al. (1998), with references to the earlier report of Voilleque et al. (1995) for some details.

Killough and Schmidt (2000) carried out an uncertainty analysis for off-site estimates of WLM, using a transport model for radon and its decay products that is based on its counterpart from the CDC dose reconstruction. Because of space limitations, however, they used a simplified surrogate for the K-65 source term, with statistical properties that approximated those of the more complicated authentic K-65 source term model. The transport model used for the study described in this paper is adapted from the transport model of Killough and Schmidt (2000), but it is coupled with the original K-65 source term developed for the CDC dose reconstruction study, as described in Appendix Q of Killough et al. (1998) and in Voilleque et al. (1995).

The atmospheric transport model for radon consists of a ground-level circular-source Gaussian plume (with horizontal and vertical dispersion parameters, $\sigma_y$ and $\sigma_z$, associated with Pasquill-Gifford stability categories A–F):

$$D(x,R) = \frac{\chi u}{Qf} = \frac{\sqrt{2}}{\pi^{3/2}R^2} \times \int_{-R}^{\min(R,x)} \frac{2P(\sqrt{R^2-\xi^2}/\sigma_y(x-\xi))-1}{\sigma_z(x-\xi)}\,\mathrm{d}\xi \quad \text{(A1)}$$

The quantity $D$ ($m^{-2}$) is the centerline diffusion of the plume and depends only on the downwind distance $x$ (m) from the center of the source area, the stability category (A–F)

through the horizontal and vertical stability functions $\sigma_y$ and $\sigma_z$ (m), and the radius $R$ (m) of the circular source area. The centerline ground-level concentration is $\chi$ (Bq/m$^3$), $u$ is the mean wind speed (m/s), $Q$ is the release rate at the source (Bq/s), and $f$ is the fraction of the time that the wind blows in the source-to-receptor direction. The function Pin the integrand is the standard normal cumulative distribution function.

The diffusion function of Eq. (A1) was multiplied by a two-parameter calibration factor $\mathrm{CF} = K \quad (x/R)^{\beta}$ (Figure A1), and the product was calibrated by regression to an aggregate of two sets of background-adjusted radon measurements taken on-site in the 1980s at various distances from the K-65 silos and at the site boundary (Figure A2). The K-65 source term model estimates for the years appropriate to the data were used in the calibration. Thus, uncertainty in the K-65 source term propagates into the distributions of the calibration parameters $K$ and $\beta$. This calibration also involved complexities related to (1) differences in daytime and nighttime release rates from the silos and (2) unstable air associated with daytime and stable air associated with nighttime. The unstable air contributes to vertical mixing of the radon, resulting in lower ground-level radon concentrations, whereas the stable nighttime air leads to higher ground-level concentrations of radon near a ground-level source. However, the concentration data are not broken down into daytime and nighttime components but are represented as 24-h averages. These asymmetries were considered in the calibration. We refer the reader to Killough et al. (1998) and Killough and Schmidt (2000) for details.

Meteorological data for the calibration and simulations were represented as an annual joint frequency table (wind speed, wind direction, and Pasquill-Gifford stability category). A composite table was derived from hourly measurements taken during the period 1987–1991 at an instrumented tower on the site (sampling was available for a few months of 1986, but none for previous years). Further discussion of this data set may be found on pages 51–54 of Appendix M of Killough et al. (1998). Applying this 5-year composite joint frequency table to specific years (particularly those before 1987) gives rise to uncertainty associated with year-to-year meteorological variability. This component of uncertainty was estimated by studying its counterpart in a long run (1951–1991) of meteorological data taken at the Cincinnati Airport. The approach is described in Killough et al. (1998), Appendix M, beginning on page M-54.

The atmospheric transport model uses the kinetics of the $^{222}$Rn decay chain and the source-to-receptor transit time to

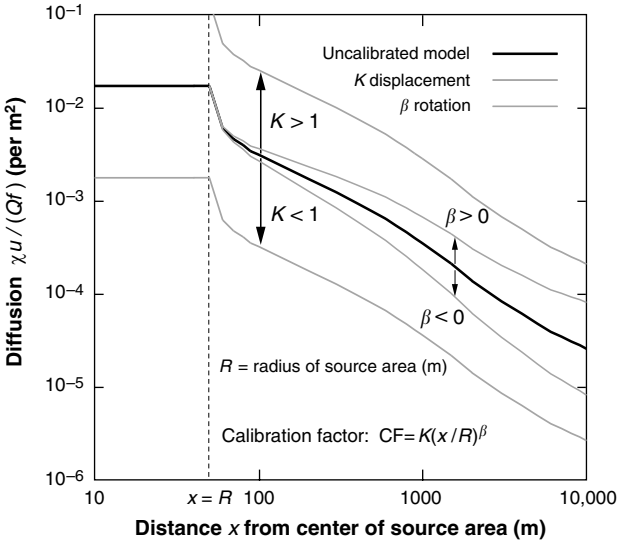


**Figure A1.** The figure indicates the geometry of the calibration factor for fitting the atmospheric diffusion curve to background-adjusted atmospheric radon measurements taken on the FMPC site during the 1980s. When the equation $y = K(x/R)^{\beta} D(x, R)$, $x \geq R$, is plotted with logarithmic axes, the parameters $K$ and $\beta$ control vertical displacement and shear rotations, respectively, of the graph of the diffusion function $D(x,R) = \chi u/(Qf)$ (see text for meanings of symbols). A regression procedure uses the radon data and the K-65 source term model for years appropriate to the data to estimate the joint distribution of ($K$, $\beta$).

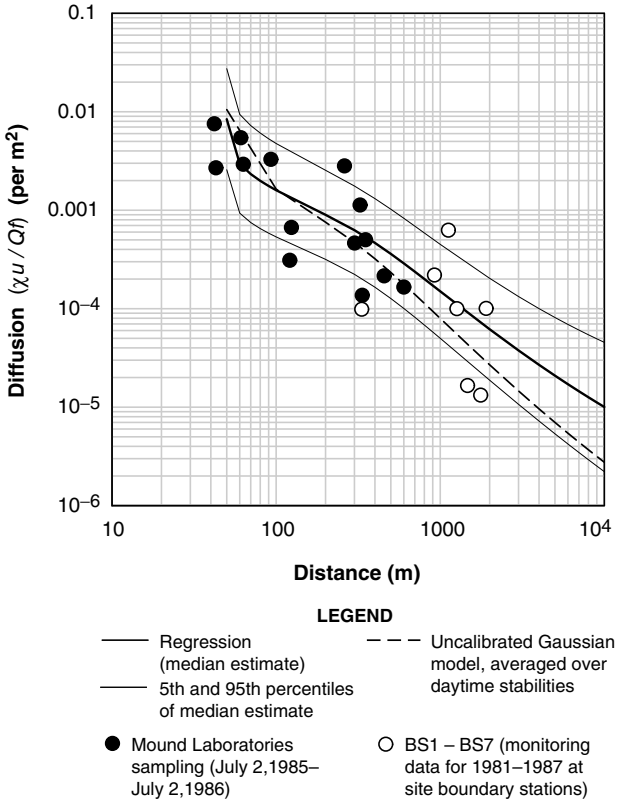


**Figure A2.** The figure indicates the radon air monitoring data, background-adjusted and converted to daytime diffusion estimates, and fitted by a regression procedure with the Gaussian circular area-source model $y = K(x/R)^{\beta} D(x, R)$. The regression curve (black line) and a corresponding uncalibrated model curve (dashed line) are also shown. This figure is an illustration, restricted to daytime parameter.

estimate concentrations of $^{218}$Po, $^{214}$Pb, and $^{214}$Bi in air at the downwind receptor location. The resultant concentrations are combined to calculate the WL estimate

$$\begin{aligned} WL = & 2.78\times10^{-5}[^{218}Po] + 1.37\times10^{-4}[^{214}Pb] \\ & + 1.01\times10^{-4}[^{214}Bi], \end{aligned} \quad (A2)$$

where the quantities in square brackets are expressed in Bq/m$^3$ of air (converted from the pCi/m version given by NCRP 1984, Eq. (7-1), p. 84). We note that the in corresponding equation (Eq. (6)) of Killough and Schmidt (2000), the coefficients shown are incorrect, but the calculations reported there were performed with the correct equation.