

## ORIGINAL ARTICLE

## Exposure assessment for a cohort of workers at a former uranium processing facility

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Exposure was assessed for a cohort of 6409 workers at a former uranium processing facility as part of a mortality study. Workers at the facility had potential for exposure to a wide variety of radiological and chemical agents including uranium, thorium, radon, external ionizing radiation, acid mists, asbestos, and various solvents. Organ dose from internal exposure to uranium was assessed, along with dose from external ionizing radiation and exposure to radon. Qualitative assessment of exposure to thorium, acid mists, asbestos, coal dust, welding fumes, and other chemicals was also performed. Mean cumulative organ dose from internal uranium exposure ranged from 1.1 mGy (lung) to 6.7  $\mu$ Gy (pancreas). Mean cumulative external ionizing radiation dose was 13.4 mGy. Mean cumulative radon exposure was 26 working level months (WLMs). The chemical agents to which the largest numbers of study subjects were exposed were acid mists, machining fluids, and a tributyl phosphate/kerosene mixture used in the refining process.

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## INTRODUCTION

The National Institute for Occupational Safety and Health (NIOSH) is examining the mortality experience in nuclear workers at the Fernald Feed Materials Production Center (FMPC). Workers at FMPC had potential for exposure to a variety of radiological and chemical agents including various compounds of uranium, thorium, radon, external ionizing radiation, acid mists, asbestos, a variety of solvents, and other chemicals. Several studies have been done of this group of workers with inconsistent results. An early study of exposure and non-malignant respiratory disease<sup>1</sup> showed no evidence of increased risk. A later study of uranium dust exposure and lung cancer<sup>2</sup> also showed no evidence of increased risk. A more recent study showed an excess of lung cancer deaths among hourly workers and a positive dose response between external ionizing radiation and lung cancer mortality.<sup>3</sup> This same study also showed an increase in stomach cancer for salaried workers and evidence of a relationship between internal radiation dose and death from non-malignant respiratory disease. None of these studies included assessment of exposure to radon, acid mists, or other radiological and chemical agents present at the FMPC. The current study design improves upon previous studies through added follow-up and more detailed exposure assessment. In particular, this paper describes the methods used to assess exposure to uranium, external ionizing radiation, radon, thorium, acid mists, asbestos, solvents, and other agents present at the facility for 6409 workers who were hired before 31 December 1985 and followed through 31 December 2004.

The primary mission of the FMPC was to produce high-purity uranium metal products in the form of derbies, ingots, billets, and fuel cores for other sites in the U.S. nuclear weapons complex. This was a large-scale, integrated facility designed to convert

uranium ore into uranium metal using a series of chemical and metallurgical processes. The facility, which takes up ~136 acres, was built on a 1050-acre parcel of land ~18 miles northwest of Cincinnati. The first plant (Pilot Plant) began operation in October 1951 and all main production plants were fully operational by 1954. The FMPC ceased uranium production in July 1989 and initiated site closure and environmental restoration activities. The site received formal acceptance of completion of cleanup from DOE on 1 January 2007. Table 1 describes the main production plants and processes at the facility.<sup>4</sup>

The FMPC stored radioactive solid residues from ore, ore concentrates, and precipitates previously processed elsewhere. These residues included radium-bearing materials known as Q-11 and K-65. The K-65 material, for example, contained significant quantities of radium (~0.19 MBq/g) that posed both an external radiation hazard and an internal hazard from <sup>222</sup>Rn and other short-lived progeny. Onsite storage of K-65 material in 55-gallon drums began in 1951. In 1952 through 1958, the material was transferred to large storage tanks (silos) at the site.<sup>5</sup> The site also maintained long-term storage facilities for a variety of thorium materials for the DOE.<sup>4</sup> These thorium materials were also potential sources of internal exposure and external ionizing radiation exposure.

## METHODS

Cohort entry was restricted to workers with at least 30 days of employment by National Lead of Ohio (NLO) between 1951 and 1985. The study roster was developed from three primary records sources, namely and in order of priority: (1) employment information in the NLO Employee Database (NLOEMP), (2) a database (Master 5) that was first populated using information from a previous epidemiologic study<sup>3</sup> and later expanded

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using FMPC employment records, and (3) the radiation exposure database (HIS20) that was obtained from the site.

### Internal Exposure to Uranium

The primary exposure of concern in the epidemiological analysis is internally deposited uranium. Therefore, the aim of the exposure assessment was to quantify the absorbed dose to target tissue resulting from uranium uptake. The main source of exposure information was urine bioassay samples routinely collected by FMPC dosimetrists beginning in August of 1952. By the end of that year, 245 urine samples had been analyzed by the bioassay group and by the end of 1957, >40,000 urine samples had been analyzed.<sup>6</sup> All employees who worked in the process areas were expected to submit routine samples at 3-month intervals. The remaining employees were required to submit samples semiannually.

All samples were single-void urine samples that, with the exception of the pre-employment and re-employment samples, were collected at the start of the shift.<sup>6</sup> Incident and special samples were also collected at the plant but were not used in the dose assessment. The urine samples were analyzed by a fluorophotometric method that was sensitive to uranium concentrations from ~5 to 1000  $\mu\text{g/l}$  and a precision of  $\pm 10\%$ .<sup>7</sup> The lower limit of detection was 1  $\mu\text{g/l}$  ( $10 \pm 3 \mu\text{g/l}$  at 95% confidence).<sup>8</sup> Urine samples in the 1990s were analyzed using kinetic phosphorescence analysis (KPA) that was reported to have a lower limit of detection of 0.007  $\mu\text{g/l}$ .<sup>9</sup> Urine samples in the late 1990s through 2004 were analyzed using ICP-MS that has an instrument detection limit of ~0.0001  $\mu\text{g/l}$ .<sup>10</sup>

Historical records on plant bioassay monitoring practices were retrieved along with an electronic database (HIS20) containing >250,000 bioassay samples. The program had a coding system for urine samples that was used to designate the reason for the urine sample and provide information about the time at which the sample was collected and/or submitted.

To prepare bioassay data for use in intake and dose estimation, the individual urine samples were adjusted for background urinary uranium levels and any contamination due to laboratory processing using uranium concentration in pre-employment urine samples. The single-void urine samples were also normalized to 24-h excretion assuming a 24-h excretion of 1.6l, and converted from mass per day to activity per day by assuming the central value of uranium isotopic enrichment was that of natural uranium (i.e., specific activity of 0.025 Bq/ $\mu\text{g}$ ). Most of the uranium at FMPC was at or near normal isotopic abundance, having an enrichment of 0.2–1.5% (H. West Memorandum to N. Ingle. Technical Basis for Decision Made on Uranium Internal Dosimetry of FMPC Employees. Oak Ridge, TN, USA: Oak Ridge Institute for Science and Education; 12 April 1994) with very small amounts having higher enrichments.

All exposure to uranium was assumed to be chronic. Most of these study subjects worked in multiple plants during the course of their employment and were exposed to a wide variety of uranium compounds. As only one chronic intake was assigned to each study subject, it was assumed that Type M absorption (e.g., compounds such as  $\text{UO}_3$  and  $\text{UF}_6$ ) best described the compounds to which these workers were exposed.<sup>11</sup> The weighted mean particle size of uranium aerosols for the facility was determined by air monitoring to be 7.9- $\mu\text{m}$  activity median aerodynamic diameter (AMAD).<sup>12</sup> The default ICRP AMAD closest to this value is 10  $\mu\text{m}$ , which was assumed for this intake assessment.

InDEP (Internal Dose Evaluation Program; developed under a contract with NIOSH by SENES Oak Ridge, Center for Risk Analysis, Oak Ridge, TN, USA) was used to evaluate the urinalysis data. InDEP calculates intakes from bioassay data (e.g., urinalyses) using least-squares regression techniques. InDEP was then used to calculate organ doses for six organs (lung, liver, kidney, red bone marrow, lower large intestine, and pancreas) selected *a priori* based on expected outcomes from exposure to uranium. The InDEP program operates in a batch mode and can analyze exposure for hundreds of individuals simultaneously. InDEP was verified and validated by comparing calculations performed in InDEP with calculations performed in the IMBA Expert (IMBA Expert ORAU-Edition, v. 4.0.9, by JABASoft and the U.K. Health Protection Agency, May 2005) computer program. The comparison showed that intakes and doses calculated using both programs were comparable. A more in-depth demonstration of the

capabilities of InDEP will be available in a forthcoming publication (in production).

The normalization of the single-void urine samples to 24-h activity introduced large uncertainty due to biological variability, and hence the bioassay data were fit using the uniform logarithmic error method of least-squares regression. The data were uniformly weighted assuming a geometric SD of 1.8.<sup>13,14</sup>

### External Ionizing Radiation

The FMPC operated a personal monitoring program beginning with film dosimetry in 1951, which was replaced by thermoluminescent dosimetry in 1985. Film badges were issued to all employees following the integration of dosimetry with the security badge in 1952; however, badges were required to be processed for only those individuals with a potential to exceed 10% of the allowable dose during the monitoring period. Film badge exchange and processing were performed weekly before October 1956, bi-weekly before January 1959, and then monthly thereafter. The minimum detectable exposure using film dosimeters was ~0.3 mGy; however, measurements recorded before 1981 were reported in 1-mGy increments.

External dosimetry measurements were recorded from hard copy records to electronic media and migrated to the site exposure database (HIS20). The HIS20 database was the primary source of external radiation exposure information and was the official record of dose at facility closure. Results were available in annual increments before 1981 and monthly thereafter. These data were the main source of external exposure information used in this exposure assessment. The study roster was also linked to DOE's Radiation Exposure Monitoring System (REMS) to abstract exposure data reported by other DOE facilities. HIS20 also provided information on previous exposures during employment elsewhere, such as the commercial nuclear power industry and the military.

External radiation exposure was examined using the unadjusted penetrating whole-body "dose" values reported in the available dosimetry records systems. External radiation dose did not include neutron dose. There was little potential for neutron exposure at the FMPC. Reported doses from each available monitoring period were accumulated to provide estimates of annual exposures. Annual values were then summed to determine the cumulative dose for each worker. These estimates were used to examine exposure patterns and provide for comparison with other cohorts of uranium and other nuclear workers. Additionally, discrepancies in monitoring data were identified by range checks, examination of distributions, and inspection of individual records.

### Radon

Radon exposure was assessed by combining the FMPC cohort work history created for this study with a radon exposure matrix developed previously by Hornung et al.<sup>15</sup> This radon exposure matrix was based on a Gaussian dispersion model coupled with a source term for the K-65 silos that was originally intended to estimate exposures to residents living near the FMPC. A detailed description of this atmospheric transport model and the development of the K-65 source term is described elsewhere.<sup>16–18</sup> Hornung et al.<sup>15</sup> modified this model by applying stratified meteorological measurements to annual average exposure at various locations at the facility. This resulted in radon exposure estimates that corresponded to work location and shift assignment (three shifts) for each worker at FMPC, thereby accounting for the 24-h variation in radon concentrations.<sup>15</sup>

The exposure matrix consisted of files containing assigned location codes and annual cumulative working level months (WLMs). (A working level (WL) is a measure of the concentration of radon progeny in 1 liter of air that results in the emission of  $1.3 \times 10^5$  MeV of alpha particle energy. A WLM is equal to 170 h of working at 1 WL.) Before combining with the cohort work history, annual cumulative WLM for each subject in the Hornung et al.<sup>15</sup> cohort was divided by the number of days the person worked in that year to get the daily exposure. This allowed for combining the currently updated work history which may have varied somewhat from the work history assigned by Hornung et al.<sup>15</sup>

## Thorium

Thorium exposure was assessed qualitatively for workers determined to have potential for exposure to thorium. Work history for each member of the cohort was examined to determine whether it coincided with thorium production activities. Also, plant historical documents and air and bioassay data were examined in an effort to determine the workers with the most potential for thorium exposure.

Several sources were used to ascertain workers with potential thorium exposure. An undated memo (R. Bravard and O. Boehler Memorandum to File. Thorium Bio-assay Investigation. Undated) listed the names of five chemical operators, badge numbers, job title, current plant location, and minimal information regarding thorium work. Another memo (R.H. Starkey memorandum to T.C. Mick, M.D. Thorium Workers. Cincinnati, OH, USA: National Lead Company of Ohio; 26 December 1967) from 26 December 1967 listed 41 names, badge numbers, and plant location. This list was part of an internal report that was intended to identify previous thorium workers for the purpose of *in vivo* bioassay. (M. Boback, personal communication to M. Rolfes. Division of Compensation and Analysis Support. National Institute for Occupational Safety and Health. 30 August 2007.) Electronic air sample data from the Pilot Plant and the Plant 9 Warehouse from May 1977 through December 1985 were also available.

Plant history and process information were also used. Thorium exposures were assumed to occur in the plants and during the time periods shown in Table 2. The Fernald work history for each of the workers

**Table 1.** Description of the main production plants and processes at the FMPC.

Plant	Dates of operation	Description
Pilot	1951–1989	Operating prototype of all phases of the FMPC uranium metal production process; UF <sub>6</sub> to UF <sub>4</sub> conversion producing dilute HF by-product; coating of metal-casting crucibles
1	1964–1979	Thorium production operations
	1953–1989	Sampling plant; weighing, sampling, classification, sorting of incoming raw material and residues from on-site operations
2/3	1953–1989	Ore-refinery plants; used to convert uranium feed material and recycled residue to high-purity UO <sub>3</sub> using nitric acid, kerosene, and tributyl phosphate
4	1968	Production of thorium nitrate crystals and thorium oxide
	1953–1989	Green salt plant; used to convert UO <sub>3</sub> to UO <sub>2</sub> by reduction with hydrogen; then further reaction with anhydrous HF to produce UF <sub>4</sub>
5	1953–1989	Metals production plant; conversion of UF <sub>4</sub> to U metal derbies by thermite reduction, which were then used to produce ingots
6	1952–1989	Metals fabrication plant; fabrication of ingots into finished uranium cores using machining fluids; core degreasing using trichloroethylene (later caustic); scrap pickling using nitric acid
7	1960–1963	Thorium oxidation in one furnace
	1954–1956	UF <sub>6</sub> reduction plant; conversion of UF <sub>6</sub> to UF <sub>4</sub> with HF as a by-product
8	1953–1989	Scrap recovery plant; used to convert U metal and metal-bearing waste from FMPC operations and off-site sources into U <sub>3</sub> O <sub>8</sub>
	1969–1971	Conversion of thorium residues to thorium hydroxide
9	1954–1989	Special products plant; casting of oversized enriched uranium ingots into billets
	1954–1956	Thorium production

Abbreviations: HF, hydrofluoric acid; UF<sub>6</sub>, uranium hexafluoride; UF<sub>4</sub>, uranium tetrafluoride; UO<sub>2</sub>, uranium dioxide; UO<sub>3</sub>, uranium trioxide; U<sub>3</sub>O<sub>8</sub>, triuranium octaoxide.

in the lists described above was examined to determine whether it coincided with the dates and plants in Table 2. Additionally, because Plant 9 was exclusively a thorium-producing plant from January 1954 through December 1955, any worker assigned to this plant during this time period was assumed to have potential for exposure to thorium.

## Chemicals and Other Non-Radiological Exposures

An exposure matrix was created to provide metrics for potential chemical exposure to Fernald workers. Work history records were used to create a file containing Division, Department, Plant/Building and Job Title information for all workers in the cohort over their working lifetime at FMPC. Process records concerning chemical hazards and process locations (i.e., processes and buildings) over time were used along with site-specific knowledge of the functions performed by the workers in these departments, plants, and jobs to assign potential for chemical exposure. For each work history segment, "Yes/No" assignments were made based on combinations of job title, work organization, and work location, with consideration of site process history for potential for exposure to nine agents/agent combinations: nitric acid/nitrogen dioxide (HNO<sub>3</sub>/NO<sub>2</sub>), machining fluids, hydrofluoric acid (HF), tributyl phosphate/kerosene (TBPK), vehicle exhaust, asbestos, trichloroethylene (TCE), coal dust, and welding fumes.

Cumulative exposure estimates were summarized using descriptive statistics. Multicollinearity among the large number of exposure variables was assessed using collinearity diagnostics in the REG procedure (SAS Software, version 9.2, Copyright (2002–2003), SAS Institute, SAS and all other SAS Institute, product or service names are registered trademarks or trademarks of SAS Institute, Cary, NC, USA). Linearity was considered noteworthy if the regression variance inflation factor (VIF) was > 10 or if the Condition Index exceeded 30. Relationships between the various exposures of interest were also evaluated using Pearson's correlation coefficients.

Temporal variations in radiation exposures were examined graphically using normalized values for annual external, internal uranium, and radon exposures. External doses were estimated from onsite dosimetry measurements (mGy). Patterns of annual internal uranium exposure were examined using estimates of daily uranium intake (in Bq/day) calculated from urine bioassay data. Radon exposures were estimated using the model described by Hornung et al.<sup>15</sup> Normalization for each exposure variable was accomplished by dividing the yearly average value by the overall average between the years 1952 and 2004.

## RESULTS

Table 3 shows the cohort demographics and provides a description of the bioassay data used in the radiation organ dose assessment.

Descriptive statistics for intakes and organ doses cumulated up to the date last observed for each study subject are shown in Table 4.

Onsite monitoring results for whole-body penetrating radiation were available for 6002 (94%) study subjects, resulting in a collective dose of 80.7 person-Gy. There were 310 workers with exposures recorded at other nuclear facilities. Only one worker was exposed exclusively at other facilities. The combined cumulative dose distribution ( $n = 6003$ ) was highly right skewed, with mean and median values of 13.4 and 2.0 mGy, respectively.

**Table 2.** Plants and time periods of potential exposure for which there was significant potential for exposure to thorium.

Plant	Time period of potential exposure
Pilot plant	Jan 1954–Dec 1955; Jan 1965–Dec 1979
Plant 2/3	Jan 1968–Dec 1968
Plant 6	Jan 1960–Jun 1963
Plant 8	Jan 1969–Dec 1971
Plant 9	Jan 1954–Dec 1955

**Table 3.** Cohort demographics and description of bioassay data used in the organ dose assessment.

Item	Quantity
No. of study subjects with employment > 30 days	6409
No. of male study subjects	5462 (85%)
No. of white study subjects	6053 (95%)
Mean employment duration in years	9.2 (median 5.3)
No. of study subjects with uranium urinalysis data	6011 (94%)
No. of female study subjects with uranium urinalysis data	848 (90%)
Total no. of urine samples available for cohort	223,520
Mean no. of urine samples per male study subject	37 (median = 17; range 1–517)
Mean no. of urine samples per female study subject	15 (median = 5; range 1–185)
Mean ± SD of activity in urine samples for each study subject (Bq/day)	0.39 ± 1.1
No. of study subjects with > 1 urine samples with reported uranium concentration ≤ 0 <sup>a</sup>	2239
Mean percentage of samples with reported uranium concentration ≤ 0 for each study subject	9.6% (IQ range 2.4–9.8%)
No. of study subjects with > 1 urine samples with adjusted uranium concentration ≤ 0	5391
Mean percentage of samples with adjusted uranium concentration ≤ 0 for each study subject	52% (IQ range 29–72%)

Abbreviation: IQ, interquartile range.

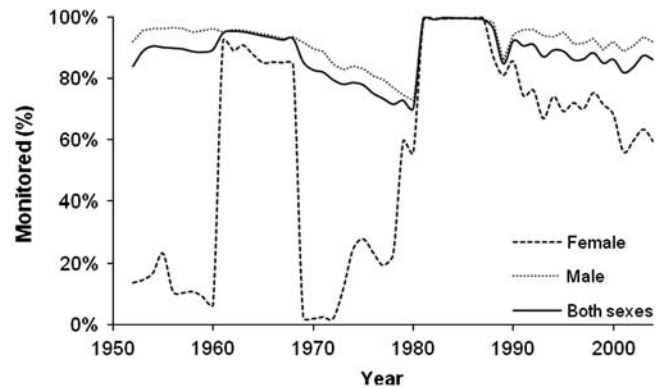
<sup>a</sup>Urine bioassay data obtained from Fernald had some sample uranium concentrations reported as < 0 in the late 1990s due to bioassay laboratory adjustments for assumed urinary background levels of uranium.

**Table 4.** Daily intake (in Bq/day) and cumulative organ doses (in mGy) calculated using InDEP (n = 5998).

	Mean	SD	Median	25th percentile	75th percentile
Intake	2.6	15	0.045	0.0018	0.81
Lung	1.1	3.3	0.053	0.0023	0.59
Kidney	0.093	0.29	0.0044	0.00019	0.051
Red bone marrow	0.032	0.10	0.0015	0.000061	0.018
Liver	0.032	0.1	0.0015	0.000054	0.018
Lower large intestine	0.011	0.036	0.00054	0.000020	0.0064
Pancreas	0.0067	0.022	0.00030	0.0000090	0.0035

Excluding those workers with zero dose (n = 1916), cumulative whole-body dose averaged 20 mGy (median 6.1 mGy; range 0.030–327 mGy).

Incomplete personal monitoring by gender was also observed. Between the years 1951–1960 and 1968–1978, monitoring practices did not include routine exposure monitoring of women because they typically held jobs that had a low potential for exposure that could exceed the allowable limit. Moreover, 100% coverage within a year was not achieved for either sex in any year, suggesting that some positions held by employees were



**Figure 1.** Percent of workers with at least one badge result with the year by sex.

exempt from monitoring throughout facility life regardless of gender (Figure 1).

All study subjects (n = 6409) were assumed to be exposed to radon. Cumulative radon exposure estimates were lognormally distributed with a mean and median of 26 and 3.9 cumulative WLMs, respectively.

A total of 150 workers were determined to be exposed to thorium at Fernald. Cumulative time spent as a thorium worker averaged 1.5 years, with a median of 1.2 years (interquartile range 0.58–1.3 years). Duration as a thorium worker is correlated with work in the Pilot Plant ( $r_{\text{Pearson}} = 0.771$ ) and weakly correlated with work in Plant 2/3 ( $r_{\text{Pearson}} = 0.592$ ) and with external dose ( $r_{\text{Pearson}} = 0.550$ ).

The final product of the chemical exposure assessment was a matrix in which the potential for exposure was addressed for each work history period for all workers (n = 6409), resulting in a job exposure matrix containing 27,372 records. For each record (i.e., the work history period for an individual worker) the matrix identified one process plant (if applicable), plus any of the other nine chemical agents with potential for exposure. The work history data contained relatively complete plant information for Production Division employees. Building/plant data for other workers were generally not available and these workers are included as “Unidentified Building/Plant.” Tables 5 and 6 show the distribution of workers and time spent in each of the assigned job exposure categories.

There was little evidence of multicollinearity in the suite of exposure variables examined. Of the 18 variables tested, only nitric acid exposure had a VIF exceeding 10 (VIF = 11.25). Although there were no Condition Index values exceeding 30, examination of the eigenvalues suggested that there were two sets of collinear relationships among exposure variables. These relationships involved correlations between the chemical exposure variables HF and HNO<sub>3</sub>/NO<sub>2</sub> ( $r_{\text{Pearson}} = 0.716$ ), HF and TBPK ( $r_{\text{Pearson}} = 0.856$ ), and HNO<sub>3</sub>/NO<sub>2</sub> and TBPK ( $r_{\text{Pearson}} = 0.728$ ).

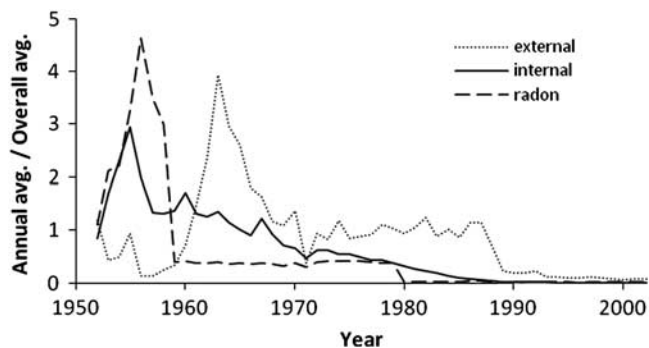
Inspection of annual exposures found temporal variation in yearly average radiation exposures for each exposure variable (Figure 2). Over half (60%) of the collective dose from external exposures occurred during operations in the 1960s. Annual average external doses also peaked during this time, with a mean of 2.5 mGy per year and ranging between 1.0 mGy (1960) and 5.0 mGy (1963) per year. Starting in 1970, average exposures remained relatively steady until sharply decreasing following cessation of uranium production operations in the late 1980s. The mean exposure since 1990 was < 0.20 mGy per year. Radon exposures were most pronounced during active ore processing before 1958. Over 80% of the collective WLMs were accumulated between the years 1951 and 1958, followed by a steep reduction in radon exposures evident in 1979 from steps taken by the FMPC to reduce environmental releases from silos 1 and 2.

**Table 5.** Number of workers (*n*) ever employed in and cumulative duration (in years) in process plants and buildings.

Building	<i>n</i>	Mean ± SD	Median	25th percentile	75th percentile
Pilot plant	321	3.9 ± 5.2	1.5	0.54	5.2
Plant 1	229	3.0 ± 4.2	1.0	0.27	4.6
Plant 2/3	465	3.8 ± 4.5	1.9	0.71	5.8
Plant 4	299	3.7 ± 5.0	1.4	0.56	5.0
Plant 5	908	4.4 ± 4.9	2.7	0.79	6.2
Plant 6	1051	6.1 ± 7.3	2.8	0.66	9.4
Plant 7	46	1.2 ± 0.86	1.0	0.62	1.6
Plant 8	221	4.9 ± 4.8	2.9	0.91	8.2
Plant 9	569	2.9 ± 4.2	1.4	0.44	3.7
Unidentified Plant/building	5121	8.0 ± 9.4	3.9	1.0	12
Mixed plants	63	2.7 ± 3.0	1.6	0.69	3.3

**Table 6.** Number of workers (*n*) and cumulative duration (in years) potentially exposed to agents of interest.

Exposure agent	<i>n</i>	Mean ± SD	Median	25th percentile	75th percentile
HNO <sub>3</sub> /NO <sub>2</sub>	2900	6.2 ± 7.7	3.0	0.75	9.0
Machining fluids	2057	5.8 ± 7.4	2.5	0.59	8.1
HF	2015	5.7 ± 7.2	2.6	0.67	8.4
TBPK	1870	5.5 ± 7.0	2.4	0.53	8.0
Vehicle exhaust	255	9.9 ± 9.1	8.3	2.3	14
Asbestos	225	9.2 ± 8.6	5.8	2.2	16
TCE	134	6.1 ± 7.6	3.4	0.38	10
Coal dust	91	10.5 ± 9.7	8.1	1.8	16
Welding fumes	70	8.3 ± 8.1	5.2	2.3	13

**Figure 2.** Temporal patterns in radiation exposure at the FMPC between 1952 and 2004.

## DISCUSSION

Mean estimates of absorbed dose to the lung for this study ( $1.1 \pm 3.3$  mGy) were slightly lower than estimates found for other studies. For example, Guseva Canu et al.<sup>19</sup> estimated a mean lung dose of 4.5 mGy (90 mSv assuming  $w_R = 20$ ) for 30 workers at a French nuclear fuel facility. They assumed a moderate absorption type (Type M), and an AMAD of  $5 \mu\text{m}$  unless specific values of the inhaled aerosol parameters were available in facility medical records.

Median estimates of absorbed bone marrow dose for this study (0.0015 mGy) were about a factor of 10 lower than the median estimates calculated for a cohort of uranium enrichment plant workers (mean 0.02 mGy).<sup>20</sup> This was partly due to the assumption of a more rapid absorption type (Type F) and an AMAD of  $5 \mu\text{m}$ .<sup>20</sup>

Natural sources of uranium in the body from intakes by ingestion and inhalation results in annual absorbed doses to a reference male of  $0.86 \mu\text{Gy}$  to the lung,  $0.39 \mu\text{Gy}$  to the kidneys,  $0.14 \mu\text{Gy}$  to the liver, and  $0.13 \mu\text{Gy}$  to the red bone marrow.<sup>21</sup> After 50 years of exposure at this level, cumulative doses to the lung, kidneys, liver, and red bone marrow would be  $\sim 0.043$ ,  $0.019$ ,  $0.0069$ , and  $0.0067$  mGy, respectively.

Mean cumulative external dose at FMPC (13.4 mGy) was slightly larger than that estimated for the Y-12 uranium facility by Watkins et al.<sup>22</sup> (mean 8.7 mGy) and Checkoway et al.<sup>23</sup> (mean 9.6 mGy). Dupree-Ellis et al.<sup>24</sup> estimated a mean cumulative external dose of 48 mGy for the Mallinckrodt uranium processing facility. The larger external dose for Mallinckrodt was likely due to exposure to radium and progeny from the Belgian Congo pitchblende ore that was processed at that facility in the 1940s and early 1950s. The processing residues from this facility were eventually stored at FMPC.

The temporal trend in exposure is consistent with U.S. weapons production schedules and changes in radiation protection standards. Uranium production at the FMPC peaked in the mid-1960s, about the time that Atomic Energy Commission (AEC) complex-wide improvements to radiation protection were instituted. Exposures rapidly decreased to a plateau between the 1970s and 1980s, which is consistent with steady uranium production during that time. A steep reduction is evident with the cessation of production in 1989. Given these observations, it is likely that workers hired before 1970 will be the most informative in dose-response analyses. Continued follow-up of exposure information will most likely have negligible impact on cumulative doses.

A downward bias in external doses may have occurred because of incomplete monitoring, especially for women. However, it is likely that workers who were not monitored for external dose held positions that had a low probability of exposure. For example, a review of the job titles for the unmonitored group revealed that most workers were assigned to administrative positions in locations outside of the production area where true doses from external exposure to occupational sources were relatively small compared with production workers. Given that females comprise a small fraction of the cohort study and that unmonitored workers appear to have held jobs with low exposure potential, it is unlikely that a large bias exists in population dose.

The long-term storage of radium-bearing wastes in the K-65 silos presented FMPC workers with a rather unique potential for significant occupational exposure to radon. Assuming a WLM-to-lung dose conversion factor of 4 mGy per WLM,<sup>25</sup> the mean cumulative lung dose from radon is  $\sim 100$  mGy, which is significantly larger than lung dose from either external exposure or internally deposited uranium, on average. The International Agency for Research on Cancer classifies radon as a human carcinogen<sup>26</sup> because of evidence of an association between radon and lung cancer summarized in the BEIR VI report.<sup>27</sup> Recent studies have suggested that other health outcomes such as leukemia<sup>28,29</sup> and cardiovascular disease<sup>30,31</sup> may also be linked to radon exposure, although results are equivocal. The average cumulative radon exposure estimated for the FMPC cohort (26 WLMs) is comparable to exposures (i.e., within a factor of 3) in some uranium miner studies reporting excess lung cancer risk.<sup>32–36</sup> The most compelling evidence of radon-related health effects stems from studies of early uranium miners who had estimated average cumulative exposures a magnitude greater than that of FMPC workers.<sup>30,37</sup>

Among production division workers, the largest number of workers was assigned to Plant 6, where they appear to have spent the most time. The fewest number of production division workers spent the briefest amount of time in Plant 7, which was only operational for ~2 years. The category for Unidentified Buildings/Plants was assigned the most workers and included all workers lacking a Building/Plant designation at some point in their work history record. This group includes individuals who spent most of their time in the process area including most maintenance personnel, a small group of production laborers, individuals from departments who supported production work and made periodic visits to the production area or worked in the laboratories, and individuals who provided administrative and other support in the non-process areas of the site. Approximately 63% of the cohort had their entire work history assigned exclusively to this category.

Chemicals chosen for assessment were those that were used in bulk quantities at the FMPC and those that had potential health consequences of interest. The chemicals to which the largest numbers of study subjects were exposed were HNO<sub>3</sub>/NO<sub>2</sub> (45%), machining fluids (32%), HF (31%), and TBP (29%). All four of these chemical agents were part of the uranium production process and, therefore, workers with exposure to these chemical agents typically also had internal uranium exposure. A relatively small percentage of the workforce was exposed to asbestos, vehicle exhaust, TCE, coal dust, and welding fumes. One of these chemicals, TCE, was part of the uranium production process and, therefore, workers with exposure to TCE also had internal uranium exposure. The remaining chemical agents (i.e., asbestos, vehicle exhaust, coal dust, and welding fumes) were primarily associated with site support activities, generally with a lesser potential for internal uranium exposure.

The evidence of correlation between some chemical exposure variables was not surprising given that the derivation of these variables shared similar assumptions. Interestingly, there was no evidence of correlation between radiation exposure variables (i.e., uranium intake, external radiation dose, and radon) or between radiation and chemical exposures. The lack of correlation between radiation variables is attributed primarily to temporal variation in production rates and protection criteria. Cumulative external radiation doses were not correlated with duration of employment, which suggest that external radiation exposures may have been more episodic. The lack of measurement data for chemical exposure necessitated that these exposures be derived using employment duration, which by design, followed a pattern of chronic exposure.

Recycled uranium introduced as feed contained minute quantities of transuranics such as <sup>237</sup>Np, <sup>239</sup>Pu, and <sup>241</sup>Am (Boback M.W. and Wing J.F. Memorandum to J.A. Quigley. Health Hazards of Recycle Material. Cincinnati, OH, USA: National Lead Company of Ohio; 24 March 1965); however, these contaminants were present in concentrations that were parts per billion of uranium, and doses from these radionuclides were not included in this exposure assessment. Although there is the possibility that doses were underestimated by this exclusion, omission of these doses was not expected to introduce any significant bias because the majority of doses are assumed to be due to uranium intake.

Use of biokinetic and dosimetric models to interpret bioassay data in calculating internal dose involves making assumptions regarding date of intake, particle size (AMAD), and absorption type. This becomes increasingly cumbersome when analyzing data for a large cohort. Particle size and absorption type vary with the stage in the uranium metal fabrication process, and thus the assumption of a single particle size distribution and absorption type for the entire cohort introduces a large amount of shared uncertainty in addition to the uncertainty inherent in the models.

Only qualitative (Yes/No) chemical exposure assessment was performed as only limited non-radioactive chemical exposure monitoring data were available for the study period. Owing to

changes in prime contractor at the site, several sources of work history information needed to be assembled to cover the study years. Imputation of data, resolution of discrepancies among the work history databases, and errors or data missing from the original databases could all have resulted in some misclassification.

General process history information was available for the entire study period. This information was used to identify chemicals, locations, and time periods of chemical use, which were used to match with study subjects for exposure potential assignment. Dates associated with the process information were typically general in nature, and locations of process activities were typically general in nature, as were many job titles, and therefore misclassification could have resulted. Furthermore, work history locations were not available for maintenance personnel, resulting in additional potential for misclassification. Site-specific knowledge among the assessors served to mitigate the potential for misclassification.

To reduce exposure misclassification, confounding, and other sources of bias, epidemiological studies of the health effects from ionizing radiation exposures should consider all sources of exposure including concomitant exposures to hazardous chemicals in the workplace. For this study, exposures to internally deposited uranium and external ionizing radiation were assessed using uranium urinalysis data and personal monitoring information that were available for 94% of the cohort. Historical records, employment histories, and industrial hygiene records were used to characterize concomitant exposures to several hazardous chemicals and radon. Estimates of these exposures were determined for each study subject among 6409 workers employed at the FMPC between 1951 and 2005. These data comprise the most in-depth analysis of exposures in this group to date, which improves upon the information used in previous studies of this and similarly exposed cohorts.

Overall, mean internal and external ionizing radiation doses were relatively low compared with other uranium plants operating in a similar timeframe. The assumption of a 10- $\mu$ m AMAD particle size based on plant air monitoring data resulted in slightly less internal deposition of uranium and lower organ dose. Additionally, most of the uranium was of normal isotopic abundance, with a small amount having an enrichment of 0.2–1.5%. The application of these assumptions to the estimation of dose for the entire cohort resulted in uncertainty in individual dose estimates as absorption type and particle size varies by plant and process. Additionally, the urinalysis data used for calculation of intakes were based on single-void samples, which introduced a large component of biological variability.

## CONFLICT OF INTEREST

The authors declare no conflict of interest.

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## Disclaimer

The findings and conclusions in this report are those of the authors and do not necessarily represent the views of the National Institute for Occupational Safety and Health.

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