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Use of Historical Uranium Air Sampling Data to Estimate Worker Exposure Potential to Airborne Radioactive Particulate in a Uranium Processing Facility

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Historical industrial hygiene monitoring records from a uranium processing plant were collected and analyzed to characterize exposure potential to airborne radioactive particulate. More than 2,100 samples were collected during the period of 1954–1968. The data was organized by job title, plant number, and year of measurement. Laboratory analysis of air samples indicated a wide range of potential exposures to the alpha-emitting particulate. Logarithmic transformation of the data was necessary to approximate Gaussian distributions. Geometric Mean (GM) values were used as the measure of central tendency within years. GM values ranged from 23–49 disintegrations per minute per cubic meter of air sampled (dpm/m^3) with the years 1963 and 1964 being significantly higher than other years (ANOVA: $p < 0.05$). When comparing exposure potential across plants, GM ranged from 20–68 dpm/m^3, with plants 5 and 8 being significantly higher than the others (ANOVA: $p < 0.05$). Exposure potential for specific job titles across the plants varied widely. GM for clerks was the lowest (11 dpm/m^3) while furnace operators were the highest (235 dpm/m^3). Other job titles with potentially high exposures were chemical operators, forklift operators, machine operators, and furnace operators. This analysis indicates the magnitude and distributions of worker exposure to alpha-emitting airborne particulate. Additional analysis and epidemiologic studies are planned for this facility.

Keywords Uranium, Radiation, Airborne, Particulate, Sampling, Inhalation, Exposure Assessment

From 1952 to 1989, a raw material production center located in the midwestern United States processed uranium and thorium ores that were used in the development of nuclear weapons for the Department of Energy (DOE). Workers were potentially exposed to a variety of chemical and radiological agents including both external and internal ionizing radiation. Multiple studies conducted on the cohort have found increased mortality from some cancers and nonmalignant respiratory disease as well as for other health outcomes.[1–3] As part of an update mortality study by the National Institute for Occupational Safety and Health (NIOSH), additional air monitoring information has been collected from the site. The analysis of these data identifies jobs, plant locations, and years where exposure to airborne radioactive particulate occurred between 1952 and 1989.

BACKGROUND

The 1,050 acre site was constructed in 1951 and production began in 1952. The facility processed uranium ore concentrates and other recycled materials from stages of nuclear weapons production. Uranium oxides or ingots of uranium metal that were machined for use as fuel cores and target elements constituted the end products. All production ceased in July 1989.

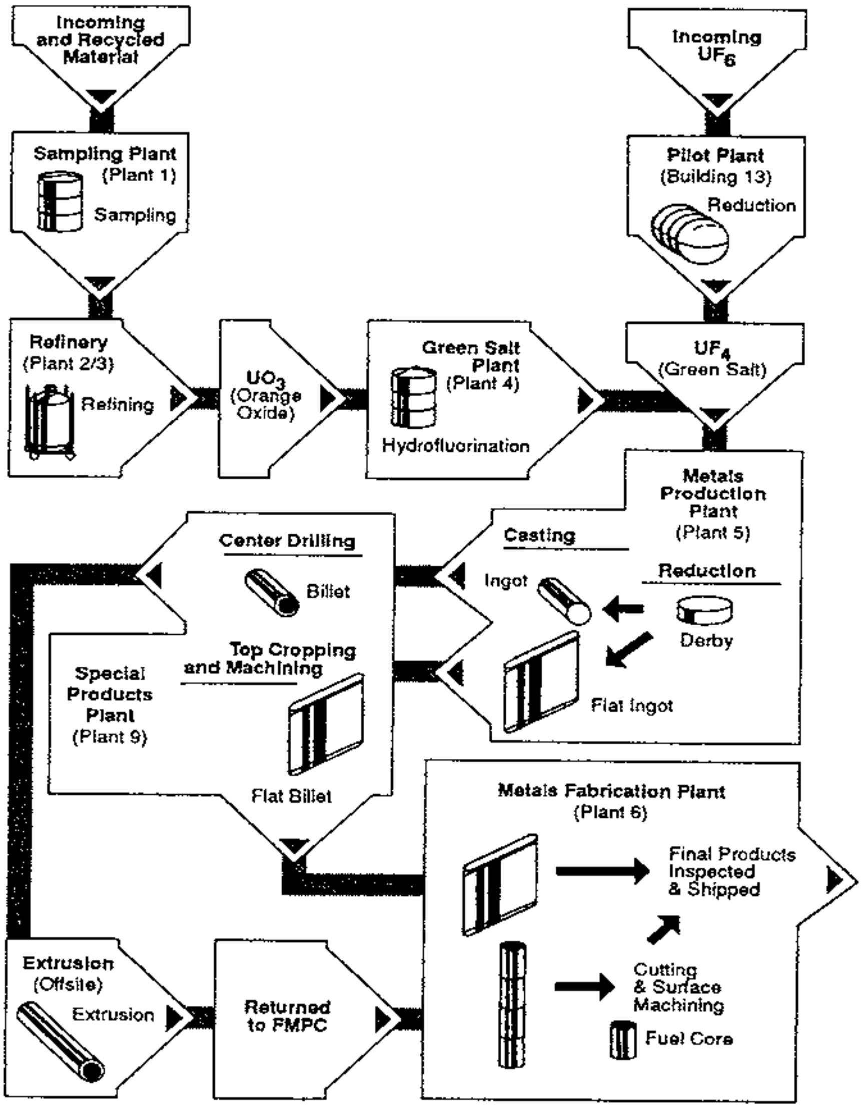
HEALTH EFFECTS OF IONIZING RADIATION

Epidemiologic studies to assess human health effects from ionizing radiation exposures primarily have been conducted in two distinct groups. The first group includes two populations with acute, high dose exposure: survivors of the atomic bombings at Hiroshima and Nagasaki, and patients treated with radiation. These studies provide strong evidence for the association between high-level, acute external exposure and subsequent mortality from breast, hematopoietic, and many other types of cancers.[4] The second group is comprised of uranium miners exposed to radon. These workers have experienced excessive mortality due to lung cancer.[5,6] Studies at various DOE sites (Savannah River, Hanford, and Oak Ridge) have also been conducted. Both the Savannah River and Oak Ridge study found an increase in leukemia deaths, while the Hanford study did not find such a relationship.[7–9]

Uranium adverse health effects stem from both its chemical and radiological properties and are difficult to separate. Lung cancer is associated with radiation, while effects on the kidney and nonmalignant respiratory disease are due to the chemical action of uranium.[10]

PROCESS DESCRIPTION

Key elements of the chemical and physical processes at the site during its production years are displayed in Figure 1. The processes consisted of dissolving ore concentrates and recycled materials in nitric acid to produce a uranyl nitrate solution which was extracted into an organic solvent. The purified uranyl nitrate was then extracted into water and thermally denitrated to produce uranium trioxide (UO_3), called orange oxide. The orange oxide was converted to uranium tetrafluoride (UF_4), commonly



Source: Fernald Environmental Management Project (FEMP), Annual Site Environmental Report, 1991. U.S. Department of Energy, Fernald Office. Contract #DE-AC05-86OR21600, December, 1992.

Note: FMPC = Feed Material Production Center.

FIGURE 1
Former site production process.

called green salt, and either shipped to a gaseous diffusion plant for enrichment or utilized on-site to manufacture uranium ingots via chemical reduction. The ingots were machined into different shapes depending on the end use.

The site was organized into 9 major plants, each with a different function. At the beginning of the production stream, the Sampling Plant (Plant 1) received ores and recycled materials. After crushing, grinding, and classifying, the feed materials were digested in nitric acid and sent to the Refinery (Plants 2 and 3). The uranyl nitrate slurry was pumped into columns for extraction into an organic solvent (tributyl phosphate and kerosene). This solution was then re-extracted with aqueous sodium carbonate and transferred to Plant 4 where UO_3 was reduced in the presence of hydrogen to UO_2 and then converted to UF_4 (green salt) via hydrofluorination. The green salt was transferred to Plant 5 (Metals Production Plant) where thermal reduction in a closed pot lined with magnesium fluoride (MgF_2) was used to produce uranium metal. The result of this reaction was a metal derby. The derbies entered Plant 6 (Metals Fabrication Plant) where they were machined into final form with furnaces, lathes, straighteners, and stampers. Plant 7 was used to convert UF_6 to UF_4 by reaction with anhydrous hydrogen fluoride in tandem with the Pilot Plant. However, this plant only operated from 1954–1956 and was later used for storage. Scrap from the Plant 6 operation entered Plant 8 (Scrap Recovery), where uranium chips were heated to remove impurities and then recycled to Plants 2 and 3. Plant 9 (Special Products) was used for machining and casting of uranium metal pieces that were larger in size than those produced by Plant 6. Also, Plant 9 produced thorium metal, much in the same way uranium was produced.

SUMMARY OF RELEVANT LITERATURE

Since the beginning of production at the site, respirable uranium particulate was an air contaminant and potential health hazard in various work areas.[11] Inhaled particles had the potential for causing lung cancer as did the uranium oxide fume.[12] Other exposure sources included hot-rolling ingots that oxidized to generate scale and dust, crushing of ores and handling dry oxides, metal shaping and machining operations, dissolution processes, and maintenance activities.

Early health monitoring of workers has been documented in both site records and the scientific literature.[11] Originally, uranium urine monitoring and uranium particulate air samples were evaluated to determine exposure empirically. Assuming chronic exposure, a urine uranium level of 0.03 mg/L would result from exposure to 50 $\mu g/m^3$ of uranium which was equivalent to approximately 70 disintegrations per minute of natural uranium per cubic meter of air sampled (dpm/m^3). The last value was used as a company-enforced maximum permissible air activity level.

Another study examined the relationship between airborne particle dimensions and urinary excretion of uranium at four different operations within the facility: filling drums with UF_4, blending and filling reduction vessels with UF_4, operating remelt furnaces, and operating lathes that shape rolled uranium blanks.[13] Exposure was primarily to particles in the 0.4 μm (micron) range. This particle size was associated with metallurgical dusts and fumes. Once again, agreement (r = 0.53) was noted between urinary levels of uranium and air sampling data.[13]

A study conducted by Breslin et al., in the late 1960s compared air sampling methods at the site. Breathing zone and general area samples (n = 500), collected side by side, were in agreement (r = 0.94).[14] The agreement among samples was attributed to the numerous machines that served as contaminant generators. A more recent study of personal air samples collected in a similar uranium facility found that the distribution of the uranium concentration in air was skewed and approximated a lognormal model.[15]

In 1983, a morbidity study of 4,101 employees hired at this site between 1952 and 1972 was conducted to investigate the relationship between exposure to uranium and development of nonmalignant respiratory disease.[1] Qualitative exposure rankings were developed by industrial hygienists, plant foremen, and engineers to assign exposure risk classes for each job and plant area. Exposure group rankings ranged from none to heavy (0–4, respectively). The study indicated that the relative risk of nonmalignant respiratory disease such as bronchitis, emphysema, asthma, influenza, pneumonia, and acute respiratory infection proportionately increased with increasing cumulative uranium exposure.

A later epidemiologic study that included this site examined the relationship between uranium dust and lung cancer mortality among workers employed in four uranium processing or fabrication operations located in Missouri, Ohio, and Tennessee.[16] The length of time from first exposure to the completion of the study was approximately 50 years. Odds ratios for lung cancer mortality were not found to increase with increasing cumulative dose estimates. Additional analyses, which included cumulative external doses; exposures to thorium, radium, and radon; and study facility characteristics also did not reveal any clear association between exposure and increased risk. However, it was noted that dose misclassification and potential exposures to other lung carcinogens may have precluded these conclusions.

In a similarly structured study, researchers compared the mortality of cohort members at this site with external population rates (U.S. population) and also evaluated their mortality pattern using internal comparisons.[2] The workers were further stratified into hourly and salaried workers and standardized mortality ratios (SMRs) were computed. A "healthy worker effect" was reported for only the salaried workers (all cause SMR = 0.71). In the analysis of salaried workers, an excess of stomach cancer (SMR = 2.61, 90% confidence interval [CI] = 1.22, and 5.14) was found. In hourly workers, statistically significant excesses of lung cancer (SMR = 1.26, CI = 1.02, and 1.54), all cancers (SMR = 1.21, CI = 1.07, and 1.37), and motor vehicle accidents (SMR = 1.59, CI = 1.14, and 2.15) were reported.

Overall mortality among hourly workers was not consistent with a "healthy worker effect" (SMR = 0.95, CI = 0.88, and 1.01). Also, no relationships were found between lung cancer and internal dose.

In a more recent study of this site, data from the Comprehensive Epidemiology Data Resource (CEDR) was utilized to study patterns of cancer mortality in a cohort of 4,014 uranium-processing workers.[3] Results indicated that workers at the site exhibited significant increases in lung, solid, and total cancers as a result of increasing exposure to ionizing radiation. The study also noted the need to address issues related to exposure assessment (e.g. use and interpretation of available exposure data such as radioactive particulate exposure monitoring results).

A retrospective cohort study was conducted on 2,514 white male workers employed at Mallinckrodt Chemical Works (MCW) facility in St. Louis, Missouri between 1942 and 1966.[17] This facility is similar to the site studied here in that its primary function was to process uranium. A unique feature of MCW is that from 1946–1955, the facility processed pitchblend ore from the Belgian Congo, which was up to 60% uranium. Other facilities did not process such ore so the potential for radiation exposure was much greater than at other uranium processing facilities.

Despite the similarities in ore processing techniques at this site and the MCW facilities, results of the mortality studies are not consistent. A notable finding in the Dupree et al. (2000) study are excess deaths from chronic nephritis (SMR = 1.88, 95% CI = 0.75, and 3.81). No evidence of excess was found for lung cancer (SMR = 1.02, CI = 0.83, and 1.24) nor for stomach cancer (SMR = 0.38, CI = 0.12, and 0.89). A weak dose-response relationship based on 10 cases was found between external radiation dose and kidney cancer. No dose-response relationship was found between external radiation dose and lung cancer at the MCW facility as it was for this site.[17]

Results of occupational studies of nuclear workers have been inconsistent in characterizing the relationship between lung cancer mortality and occupational exposures. Published results from combined worker studies did not demonstrate a trend for increasing lung cancer with external radiation dose.[18] However, other site-specific studies have demonstrated a dose-response relationship with external radiation and lung cancer mortality.[3,19–21] The work environments for all of these studies contain exposures to potential lung carcinogens such as airborne radioactive particulates, internal radiation (including radon), asbestos, and chemicals.[2] Because of these multiple exposures the association with lung cancer remains unclear.

METHODS

Historical uranium monitoring data in numerous exposure survey reports were obtained from the site-operated records archive. The surveys were completed in the early years of facility operation by health and safety personnel to determine exposure levels to airborne radioactive particulates. Each survey lasted approximately one month and included air samples for different plants, job titles, and tasks. Samples were collected on Whatman number 41 paper filters using a high volume sampling pump. Sampling times depended on the duration of the tasks and ranged from a few minutes to approximately one-half hour. All filter samples were sent to the on-site laboratory for counting by either alpha scintillation or alpha proportional counter. Results of the laboratory analysis were reported as alpha disintegrations per minute per cubic meter (dpm/m^3) of air sampled. Multiple samples for different job titles and plants were collected over the years. The apparent purpose of the air sampling program was to determine whether excessive exposures were occurring. A distribution of plant exposure summary reports appears in Table I.

TABLE I
Distribution of plant exposure surveys (1954–1968)

Plant no.	No. of surveys	No. of samples	Years covered
1	12	152	1955, 1958–1968
2/3	13	158	1958–1962, 1965–1968
4	12	188	1954, 1958–1968
5	10	428	1958–1968
6	20	667	1958–1968
8	12	193	1958–1968
9	9	306	1959, 1960, 1962–1968
Laundry	12	61	1960–1968
Total	100	2153	

Both Breathing Zone (BZ) and General Area (GA) sampling data were available in the records. BZ samples were collected by holding the sampling device in the immediate vicinity of the worker's head, in front of the shoulder area. The GA samples were collected at various areas within a plant. All samples were documented and linked to a particular job title, year, and plant number. Both BZ and GA samples were used to calculate a daily weighted average. These values were then used to calculate summary statistics that were incorporated into monthly reports for each plant. This method of data analysis was commonly practiced during the operation of the facility.[13,14]

Job title, plant number, year, and exposure value data were abstracted and entered into an electronic database. The database contains uranium air sampling results between 1954 and 1968 with more than 2,100 sample measurements across 8 different plants.

Over the history of this site, numerous titles for similar jobs were used and appeared on the air sampling forms and in the roster information for the cohort. Similar job titles were collapsed into more generalized groups. For example, job titles such as forklift truck operator and lift truck operator were combined, and the data from the two categories were pooled for analysis. The condensed job dictionary was constructed for specific jobs and associated with major duties performed (Table II). Overall,

TABLE II
Job titles, plants, and description of tasks

Job title	Plant no.	Description of task(s)
Chemical operator	1, 2/3, 4, 5, 6, 8, 9	Operates equipment in the production of uranium compounds or metal
Clerk	1, 2/3, 4, 5, 6, 8, 9, Laundry	Processes paperwork, records, files documents
Cobbler	Laundry	Repairs and maintains worker uniforms
Derby operator	5, 8	Loads reaction vessels and removes finished metal derby product
Filter operator	8	Inspects and maintains filtration system
Foreman	1, 2/3, 4, 5, 6, 8, 9, Laundry	Supervises production/operations within an area
Forklift operator	1, 2/3, 4, 5, 6, 8, 9	Operates forklift truck within a plant area
Furnace operator	4, 5, 6, 8, 9	Operates furnaces used to produce metal products
Inspector	5, 6, 8, 9	Inspects products for tolerance/compliance specifications
Laundry operator	Laundry	Responsible for cleaning worker uniforms
Machine operator	1, 4, 5, 6, 8, 9	Operates a variety of machines used to measure chemical intermediates; also shapes metal products (e.g. lathe, grinder, press, etc.)
Maintenance	4, 6	Repairs and maintains equipment within all plants
Saw operator	5, 6, 9	Operates different types of metal cutting saws and saw blade sharpener
Superintendent	1, 2/3, 4, 5, 6, 8, 9	Oversees daily operation of plant

14 generalized job titles were used across the 8 plants. Maintenance workers who primarily worked in Plants 4 and 6 also provided occasional service to equipment located in other plants. No exposure reports were located for the Pilot plant.

RESULTS

A preliminary evaluation of the data set indicated that the distribution was skewed to the right, so a log transformation of all data was performed. This transformation resulted in the data set taking on a more Gaussian distribution (data not shown). The Geometric Mean (GM) value was chosen as the measure of central tendency. The dataset was first analyzed by one-way analysis of variance (ANOVA) followed by Tukey's multiple comparison test whenever the data met the assumptions of the ANOVA procedure (normality of distribution and homogeneous variances). If the assumptions for executing an ANOVA procedure were not met, a nonparametric multiple comparison test (Kruskal-Wallis Rank Sums) was employed to detect significant differences.[22]

Figure 2 indicates average exposure levels by year for pooled data from all plants and all job titles. GM ranged from 23.1–49.3 dpm/m^3. The years 1963 and 1964 were significantly higher than all other years except 1955, 1956, and 1959 ($p < 0.05$). One year (1954) was excluded from this analysis because only Plant 4 was surveyed and the data set was small ($n = 28$) in comparison to the other years.

Sampling data were also pooled according to plant for all years to determine where plant(s) exposure levels were significantly different, regardless of job title. Figure 3 illustrates the

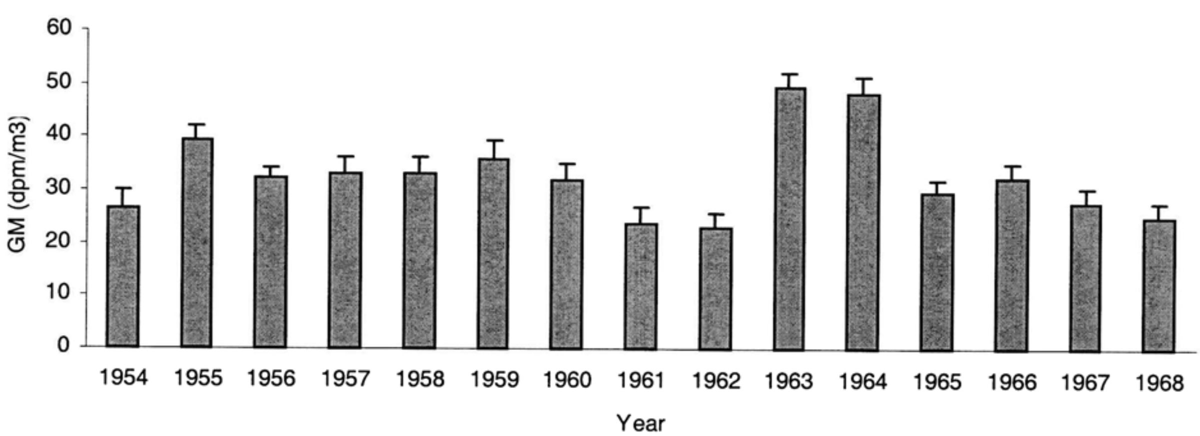


FIGURE 2
Exposures according to years: all jobs and plants combined (dpm/m^3) (error bars = GSD).

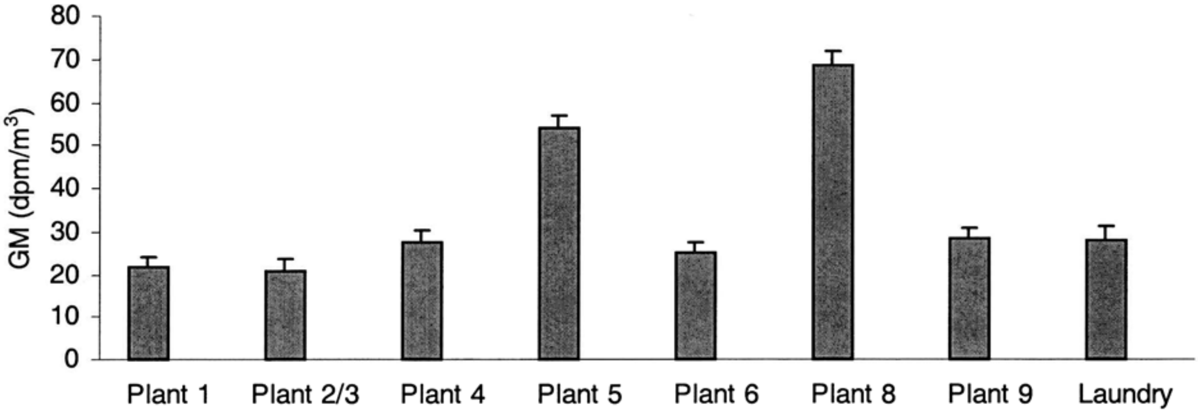


FIGURE 3
Exposures according to plant: all jobs and years combined (dpm/m^3) (error bars = GSD).

exposure values on a plant-by-plant basis (20.5–68.2 dpm/m^3). Plants 5 and 8 were found to be an environment that contained significantly higher levels of radioactive particulate than the other plants studied (ANOVA and K-W; $p < 0.01$). Further, the variance estimates for all samples combined in each of the remaining plants were relatively uniform (GSD < 3), as were the GMs (20.5–28.3 dpm/m^3).

Figure 4 presents average exposures for job titles that existed in at least 5 plants for all the years under study: chemical operator, clerk, foreman, forklift operator, furnace operator, machine operator, and superintendent. Significantly higher exposure estimates were observed for chemical operators, forklift operators, furnace operators, and machine operators who worked in Plants 5 and 8 than the same job title in other plants ($p < 0.003$), and for machine operators in Plant 4 ($p < 0.001$). The highest exposure estimate was for furnace operators in Plant 8 (235 dpm/m^3), while the lowest involved a clerk in Plant 1 (11 dpm/m^3).

Highest and lowest exposure estimates within each plant regardless of year are presented in Table III. Machine operators in Plants 1 and 4 were significantly more exposed than all other job titles within those plants. In Plants 2 and 3, chemical operators had significantly higher exposures than all other job titles. Derby operators were significantly more exposed than others in Plants 5 and 9, while saw operators in Plant 6 were significantly more exposed. Furnace operators in Plant 8 were found to be significantly more exposed than others, while laundry operators were found to have significantly elevated exposures relative to

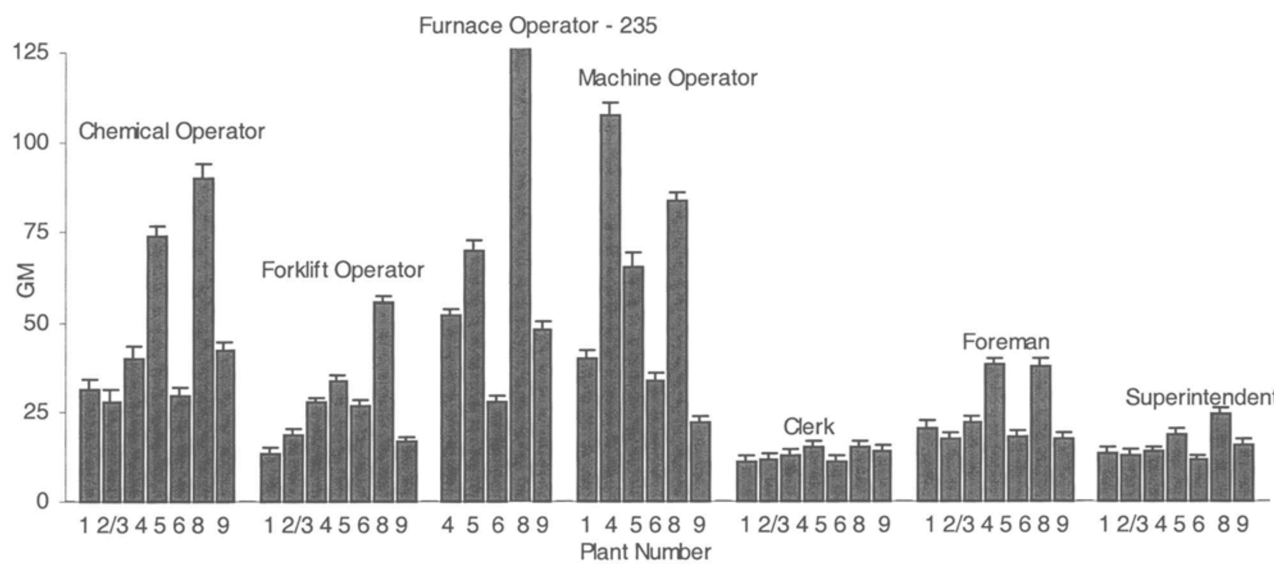


FIGURE 4
Exposures according to job titles across plants: (dpm/m^3) (error bars = GSD).

TABLE III
Highest and lowest exposures based on job titles within a plant (all years) (dpm/m^3)

Plant no.	Job title	n	GM	GSD	Minimum	Maximum
1	Machine operator (high)	14	40.0	2.1	10.0	160.0
	Clerk (low)	37	10.8	1.8	2.8	32.9
2/3	Chemical operator (high)	11	27.5	3.6	3.5	1680.0
	Clerk (low)	31	11.6	1.6	7.0	35.0
4	Machine operator (high)	5	107.8	3.2	32.9	448.0
	Clerk (low)	45	12.6	1.6	7.0	40.0
5	Derby operator (high)	44	142.1	2.7	30.0	1600.0
	Clerk (low)	59	15.1	1.7	7.0	63.0
6	Saw operator (high)	6	73.0	3.1	18.9	343.0
	Clerk (low)	71	11.1	1.6	3.5	63.7
8	Furnace operator (high)	51	234.7	2.5	42.0	1330.0
	Clerk (low)	19	15.3	1.7	7.0	40.0
9	Derby operator (high)	6	107.7	1.6	49.0	215.6
	Clerk (low)	22	14.1	1.8	5.6	40.0
Laundry	Laundry operator (high)	36	54.2	3.0	10.0	350.0
	Foreman (low)	9	9.6	1.2	7.0	14.0

Abbreviations: dpm/m^3—alpha disintegrations per minute/cubic meter of air. n—number of measurements. GM—Geometric Mean Value. GSD—Geometric Standard Deviation.

other workers in the laundry area. All of the differences for these comparisons were statistically significant (ANOVA or K-W: $p < 0.0001$). Once again, clerks were the least exposed in all plants. GM exposure values for the highest exposure job titles ranged from 27.5 dpm/m^3 (machine operator, Plants 2 and 3) to 235 dpm/m^3 (furnace operator, Plant 8).

DISCUSSION AND CONCLUSIONS

The results of this study indicate that personnel involved in the processing and fabricating of nuclear materials at this site were potentially exposed to a wide range of airborne uranium particulate levels. The inhalation of alpha-emitting particulate could have arisen from numerous operations within each plant. Some of these operations included the sampling and crushing of ore, mechanical and manual handling of dry uranium salts and oxides, and the shaping or machining of metallic uranium. Inhalation of alpha-emitting particulate has been shown to increase the risk of lung cancers. For inhalation of soluble uranium compounds, the radiological risk is relatively low, yet nephrotoxic effects are a concern.(16) However, these air sampling data did not indicate which uranium compounds were present in the air. For comparison, the lowest average value measured (clerks at 11 dpm/m^3) are orders of magnitude higher than the U.S. average airborne uranium concentration (3×10^{-4} dpm/m^3).(23)

Workers in Plant 5 and Plant 8 were potentially exposed to significantly higher levels of alpha-emitting airborne particulate. These exposures were very likely due to the machining operations and scrap recovery where large amounts of particulate were generated. Also, these plants were locations where uranium would sometimes spontaneously combust.

When data were analyzed by year of exposure only, worker exposure for 1963 and 1964 were significantly higher regardless of plant and job title. During this time, the facility production volume was at its highest.(24)

Plant clerks were the least exposed group of workers due to the lesser amount of time spent in plant environments with higher levels of airborne particulate. On occasion, clerks may have had the need to enter the production area which was known to contain a large amount of airborne particulate. However, the data did not allow the analysis of exposure on a task-by-task basis.

Throughout the history of the site, the facility utilized an airborne radioactivity level exposure limit of 70 dpm/m^3. The surveys conducted by health and safety personnel compared actual exposures to this upper limit. This analysis of the sampling data indicate that many job titles exceeded the limit, on average, over the period when these samples were collected (1954–1968): chemical operator (Plant 5, 8); furnace operator (Plant 5, 8); machine operator (Plant 4, 8); derby operator (Plant 5, 9); and saw operator (Plant 6). Actual worker exposures may have been reduced through appropriate use of Personal Protective Equipment (PPE) for control of these potential exposures at the site. In future analyses, critical information on PPE type and usage, compound solubility, and average particle size will be utilized to estimate bioavailability of uranium from inhalable and respirable particulate.

Since different enrichment levels of ^{235}U existed at the site, it is not possible to equate dpm/m^3 to mg/m^3 of uranium for all samples. As mentioned earlier, the solubility of uranium changed as the material was chemically or mechanically processed. Various uranium oxides and fluorides, as well as the base metal, were likely to be present in different ratios in many samples. Additional information will be collected from the site so that plant, job, and time-specific uranium exposure estimates may be developed for use in future analysis.

This data analysis provides an indication of the magnitude and distribution of worker exposure to alpha-emitting particulate within the facility. In the future, other historical data from the site will be used to examine the relationship between uranium levels observed in worker urine samples and the airborne particulate data for that specific job title. If sufficient information is located, models will also be developed for use in occupational epidemiologic studies to estimate organ doses that may have resulted from airborne radioactive materials at the site.

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