TID-12729

MASTER

CORRELATION OF URANIUM ALPHA SURFACE CONTAMINATION,
AIR-BORNE CONCENTRATIONS, AND URINARY EXCRETION RATES*

By

N. B. Schultz and A. F. Becher

Oak Ridge Gaseous Diffusion Plant
Union Carbide Corporation
Oak Ridge, Tennessee

Facsimile Price \$	1.60
Microfilm Price \$	.80
Available from the Office of Technical Services Department of Commerce Washington 25, D. C.	

A B S T R A C T

This paper presents statistical evaluations of field data including measurements of alpha activity on work surfaces, in the environmental air; and the results of urinalyses obtained from employees assigned to a UF_6 manufacturing facility. Two years are studied wherein personnel were subject to relatively low concentrations of both soluble and insoluble normal uranium compounds. Approximately 700 urine specimens were collected and 500 spot-air samples, 4000 shift-length air samples, and 24 surface surveys were made. Statistical correlations of these data are presented, and a method is proposed for estimating the exposure potential of employee groups working in environments having varying uranium concentrations.

This paper was submitted for publication in the open literature at least 6 months prior to the issuance date of this Microcard. Since the U.S.A.E.C. has no evidence that it has been published, the paper is being distributed in Microcard form as a preprint.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:
A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.
As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Prepared for Presentation at Health Physics Society Meeting
Las Vegas, Nevada
June 13 through June 16, 1961

*This document is based on work performed at the Oak Ridge Gaseous Diffusion Plant operated by Union Carbide Corporation for the United States Atomic Energy Commission.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CORRELATION OF URANIUM ALPHA SURFACE CONTAMINATION, AIR-BORNE CONCENTRATIONS, AND URINARY EXCRETION RATES

INTRODUCTION AND SUMMARY

On the premise that a study of a work population in varied uranium-processing facilities can be of value to the industry as a whole, this paper is presented to stimulate investigation of more practical means of assessing certain employee exposures than are presently available. Statistical evaluations of such routinely-collected data as the alpha activity levels in environmental air and on work surfaces are reviewed together with the results of urinalyses for the employees regularly assigned to these work locations. The data were routinely recorded during the operation of a uranium hexafluoride feed manufacturing and uranium recovery facilities at the Oak Ridge Gaseous Diffusion Plant wherein personnel were subject to relatively low concentrations of both soluble and insoluble uranium compounds with an isotopic balance varying from that of natural to those of product level enrichments in the 234 and 235 isotopes. The primary study covered a two-year period during which time approximately 500 spot-air samples, 4000 shift-length general air samples, 24 surface surveys were made of the work areas, and 700 urine specimens were collected. All of the data were treated to provide a common time interval which in this instance was a one-month cycle. The follow-up period included (a) these same employees during a subsequent 12-month interval and (b) other selected employee groups covering an 18-month period. There was no clinically detectable evidence of injury or long-term deposition of uranium in the group studied.

Correlation coefficients were statistically determined for the monthly average values of uranium alpha surface contamination levels, air-borne concentrations, and urinary excretion rates. Attention is particularly directed to the fact that, despite the many variables which can be introduced into actual field conditions as contrasted with a controlled laboratory study, there was a statistically significant, positive correlation between the average uranium alpha activity on the work surfaces and the average urinary excretion rate for the 70 employees initially studied; a similar relationship between the general air and the urinalysis data was not statistically significant at the 95% confidence level. Subsequent application of this premise to uranium sampling and recovery processes revealed that this interrelationship was reliable and permits prediction to within less than one order of magnitude of the potential average exposure for personnel assigned routinely to such uranium-processing operations as described, once the simply-determined uranium surface levels have been measured.

DISCUSSION

Although environmental exposures on repetitive jobs can be evaluated reasonably well by recognized air sampling techniques, they provide at best an estimate of "average" conditions and for widely separated field-type jobs of a non-routine nature become impracticable to administer routinely. It is therefore suggested, that at least for these latter types of jobs, surface measurement of uranium alpha activity provides a broad sampling

base for estimating the potential exposure. In this instance, whether the employee is exposed to "fall-out" of the uranium particulates which may be released to the environment or to the air-borne dusts stirred up by work activities and ventilation within the environment, is of course irrelevant.

The commonly used air sampling techniques also do not permit adjustment to be made for many other variables which would be involved if we were to attempt to duplicate precisely the air one breathes. These would have to recognize such factors as the turbulent effects produced by body movements or respiration as well as the environmental factors themselves. Neither do we have human data to permit reproducing the biological factors which influence the actual lung retention of inspired heterogeneous particles. Thus, even detailed monitoring and weighting of the environmental air activity levels does not provide a precise measurement.

On the other hand, it is not only logical to assume that those areas having the greatest uranium alpha activity on the work surfaces are areas where the alpha air activity is also the highest, but this has been demonstrated to be the case in previous studies made at the ORGDP,^{1, 2} and is further substantiated by the results of this study. Suffice it to say, the surface contamination levels can be rather simply and accurately obtained as will be discussed in more detail later.

METHODS AND EQUIPMENT

Population Studied

Personnel in the original study group were employees assigned to operations and maintenance activities in the uranium hexafluoride feed manufacturing plant at ORGDP. The environment results in exposure to relatively low concentrations of both soluble and insoluble compounds of natural and U-235 depleted uranium. The operation was a continuous one employing four shifts of operations personnel. During the study period, the men worked rotating eight-hour shifts with a duty cycle of seven days, and rest cycles varying from 48 to 60 hours.

The primary study was made of data which had been routinely collected during an arbitrarily selected two-year period beginning July 1, 1957, and ending June 30, 1959. For purposes of comparison, a subsequent 12-months follow-up of this segment of the plant population was made, along with a tabulation for an 18-month period beginning January 1, 1959, and ending June 30, 1960, involving employees exposed to soluble uranium in a UF_6 sampling facility and the similar experience for employees exposed to both soluble and insoluble uranium compounds as well as uranyl nitrate solutions in an equipment-cleaning and uranium recovery facility. Both of these latter groups were exposed to uranium enriched in both the U-234 and U-235 isotopes at varying levels.

Description of Operations (Photos 1 - 4)

The feed manufacturing operations are located in a single, large, relatively open area of approximately 43,000 ft², and involve the reduction of UO_3 with hydrogen to UO_2 , the hydrofluorination of UO_2 to UF_4 , and the fluorination of UF_4 to UF_6 . The gaseous uranium hexafluoride when released to

the atmosphere hydrolyzes, the reaction products being HF and UO_2F_2 . The particle size of the latter varies widely from the submicron range to large agglomerates, depending upon several factors, such as ambient temperature, relative humidity, and the concentration of the UF_6 at the time of the release. In the recovery facility, uranium-bearing materials undergo organic solvent extraction and calcining operations, while in the sampling facility, gaseous UF_6 is withdrawn from shipping cylinders.

Urinalysis Program

A total of 70 employees were included in the original program. The urinalysis sampling frequency for both the chemical operators and the maintenance mechanics was bi-monthly and for the foremen, quarterly. The urine samples were obtained on a random or "spot" basis, with an average of about 30 samples collected each month. Urine samples were taken at the plant dispensary daily and a portion of each of the groups were sampled each week. Samples were submitted at any convenient hour during the shift on the date that the dispensary visit was scheduled. Urine analyses were made for total uranium by the fluorophotometric method, while alpha activity was determined by electrodeposition and then counting in a low-background proportional counter. Results of the 728 urinary samples analyzed are shown in Table 1. There was no clinically detectable evidence of injury as a result of these exposures and, in addition, there was no indication of long-term deposition exhibited by any of these employees.

Air Sampling

Operational spot-air samples were taken by the health physics surveyor on a monthly basis at randomly selected times. An established building grid system was used which provided sample points on ten-foot centers; one-third of the grid was covered each month and the procedure was repeated during each successive three-month cycle. No special efforts were made to include or exclude areas of maximum or minimum alpha air activity, and it may be noted that no releases of significance involving uranium materials were recorded. A Staplex "Hi-Volume" portable air sampler was used with a Whatman 41 filter paper 9 cm in diameter, and samples were obtained at face levels with a flow rate of about 15 ft^3 per minute for approximately ten minutes duration. A total of 473 such samples were obtained, the results of which are shown in Table 2.*

"Long-term" or eight-hour shift-length air samples taken during the first 18 months of the study period were collected simultaneously by five vacuum-pump type samplers located through the area; the samplers were run continuously with the filter papers being changed manually by operations personnel at the end of each shift. The filter paper samples were then collected and counted in a low-background proportional counter; the overall counting and collection efficiency of this method was found to be about 30%. During the last six months of the study period, locally developed

---

*The curie, as used in this report, is the classical unit which represents an activity of 3.7×10^{10} disintegrations per second.

continuous air samplers were used. The collection is performed automatically by the device, which subsequently counts and records both the 30-minute peak levels and the eight-hour shift averages. A total of 3963 long-term air samples were collected, and these data are shown in Table 3.

Surface Contamination Levels

A complete survey of the work surfaces within the building was made routinely each month by a health physics surveyor. This normally required about seven work days which were randomly selected during the month. Both the survey technique and work area under survey were standardized, and the surveyor assignment was unchanged during the study period. For purposes of comparing the relative health problems in various plant locations as well as to provide a rather simple method of evaluating the effectiveness of control measures, the surface contamination levels at ORGDP are expressed as "Contamination Indices." (Table 4) As can be seen from the table, these are weighted values based on the fraction of the work surface contaminated and the degree to which it is contaminated. This Contamination Index numerically approximates the average transferable activity expressed in alpha counts/min/100 cm² which is obtained by a standard smear technique and which is based on a counting efficiency of 20% utilizing a Samson survey meter. The maximum contamination index recorded in the study period was 1202, which represents about 40 µg/cm² of transferable normal uranium, and the average value was 687 or about 23 µg/cm². Deposits of UO₂, U₃O₈, and UF₄ at normal enrichments are visible on work surfaces and clothing at levels greater than about 1000 alpha c/min/100 cm², while accumulations of uranyl fluoride or the oxides enriched in U-235 are not visible even at much higher concentrations.

RESPIRATOR USAGE

Employee usage of respirators, where job conditions required their use, was also checked during the monthly health physics survey. From a total of 166 such individual checks, it was found that 41% of the employees assigned were wearing respirators.

STATISTICAL ANALYSES

Under the study conditions, arithmetic means of 20 µg/l excretion rate and 2×10^{-11} µc/cc general air activity are shown. If these data are extrapolated from a 41% respirator usage condition to a zero usage value, assuming the respirators to be 100% effective, then at the maximum permissible concentration of 7×10^{-11} µc/cc for soluble natural uranium in air, a corresponding urinary excretion rate of 120 µg/l is suggested.

The problem of measuring a trend by a straight line is undoubtedly familiar to all and consists of determining the position of a line which will follow a so-called average path with respect to the scattered points. Various methods of statistical analyses were used in this study and included the Least Squares Method,³ the Quadrant Sum Test for Association,⁴ and the Pearson Product-Moment Form.³ The monthly averages of surface levels, air concentrations, and urinary excretion rates shown previously were plotted for statistical evaluation. The lines of regression, the 95% confidence levels, and the coefficients of correlation are shown in Figures 1 - 7. The coefficient of correlation reflects the frequency that variations in one set of data are

directly attributable to variations in the second set of data. For example, a value of 1.00 represents complete mutual relationship, while values above 0.14 are "statistically significant" at the 95% confidence level, since these are "too large to be interpreted as due to accidents of sampling from unrelated populations." Attention is particularly called to the statistically significant, positive correlation between the average uranium surface levels and the average urinary excretion rates of employees working in the environment (Figure 1). Statistical analysis revealed that similar relationships between the spot-air to urinalysis and general air to urinalysis data of this study were not significant at the 95% confidence level, and suggested that the measurement of air-borne activities reflects greater uncertainties than the measurement of surface contamination levels. (Figures 2 and 3) The study data further substantiate earlier findings previously mentioned relative to the positive correlation between surface contamination levels and air-borne activities. (Figures 5 and 6) Subsequent investigations over an additional 12 months experience for the feed plant employees, as well as an 18 months sampling of two other groups exposed primarily to enriched soluble and insoluble air-borne compounds, revealed similar correlation between the surface contamination levels and the employee uranium excretion rates (Figure 7).

CONCLUSIONS

Despite the many variables which might be introduced by such factors as randomly collected urinary spot samples, respirator usage on certain jobs or operations, as well as area evacuation during known periods of high air activity, there was a positive correlation between the average alpha activity levels on work surfaces and the average urinary excretion rates for the groups studied. These data also suggest that at maximum permissible concentration in air for soluble natural uranium, the corresponding urinary excretion rate would be about 120 $\mu\text{g}/\text{l}$.

It is recognized that controlling the average exposure of a group of individuals to a value below the maximum permissible concentration may not assure that each individual's exposure is below this limit. However, the inter-relationship mentioned above would suggest that potential exposure estimates can be made to within at least one order of magnitude, such precision comparing favorably with the various techniques of air sampling as employed today.

Acknowledgment: The authors wish to acknowledge the valuable contributions of Dr. J. R. Knight and J. F. Hudson for their evaluations of the voluminous data covered in this study. Access to the field notes and data compiled by B. F. Shinn is also acknowledged as are the many helpful comments and suggestions made by members of the ORGDP Health and Safety Staff and the Process Engineering Department.

REFERENCES

1. Bailey, J. C., and Rohr, R. C., Air-borne Contamination Resulting From Transferable Contamination on Surfaces, K-1008, November 24, 1953.
2. Becher, A. F., The Development of Surface Alpha Contamination Limits, HASL-58, October 15 - 17, 1958.
3. Brumbaugh and Kellog, Business Statistics, Chicago, Illinois, Irwin, 1946.
4. Waldfogle, E. A., Statistical Short Cuts, MIM-956, 1954.

Industrial Relations Division
Oak Ridge Gaseous Diffusion Plant
May 9, 1961

Table 1
Urinary Uranium Concentrations

<u>Number of Samples</u>	<u>Concentrations ($\mu\text{g/l}$)</u>		
	<u>Minimum</u>	<u>Average</u>	<u>Maximum</u>
728	8	20	1,747
<u>$< 20 \mu\text{g/l}$</u>	<u>$> 20 \mu\text{g/l}$</u>	<u>$> 100 \mu\text{g/l}$</u>	
85 %	15 %	2 %	

Table 2
Spot-Air Alpha Activity

<u>Number of Samples</u>	<u>Concentrations ($\mu\text{c/cc}$)</u>		
	<u>Minimum</u>	<u>Average</u>	<u>Maximum</u>
473	0.8×10^{-12}	7.1×10^{-11}	4.7×10^{-9}
<u>$< 7 \times 10^{-11} \mu\text{c/cc}^*$</u>	<u>$> 7 \times 10^{-11} \mu\text{c/cc}^*$</u>	<u>$> 7 \times 10^{-10} \mu\text{c/cc}^{**}$</u>	
88 %	12 %	1 %	

Table 3
Long-Term Air-Borne Alpha Activity

<u>Number of Samples</u>	<u>Concentrations ($\mu\text{c/cc}$)</u>		
	<u>Minimum</u>	<u>Average</u>	<u>Maximum</u>
3,963	1.0×10^{-12}	2.0×10^{-11}	2.4×10^{-9}
<u>$< 7 \times 10^{-11} \mu\text{c/cc}^*$</u>	<u>$> 7 \times 10^{-11} \mu\text{c/cc}^*$</u>	<u>$> 7 \times 10^{-10} \mu\text{c/cc}^{**}$</u>	
95 %	5 %	1 %	

*NCRP MPC_a for soluble natural uranium, occupational exposure.

**Ten times NCRP MPC_a for soluble natural uranium, occupational exposure.

Table 4

Contamination Index

The alpha Contamination Index is a weighted value which reflects both the degree and the extent of the contamination found. Essentially, it is the product of the Area Factor and a weighting factor which depends on the contamination intensity, summed up over the entire surface. For convenience at ORGDP, the contamination intensities for transferable and surface contamination are separated, each being divided into three classes with appropriate weighting factors; the ranges of these classes are given below.

Weighting Factors For Contamination Ranges					
Surface			Transferable		
Class	(c/min/100 cm ²)	Factor	Class	(c/min/100 cm ²)	Factor
I	500 - 2000	1	I	100 - 500	1
II	2000 - 10,000	5	II	500 - 2000	5
III	>10,000	20	III	> 2000	20

In determining the over-all Contamination Index for any location, the Contamination Indices for the three classes in that location are summed by the following relation:

$$C. I. = \frac{(A_I + 5A_{II} + 20A_{III}) \text{ Surface} + (A_I + 5A_{II} + 20A_{III}) \text{ Transferable}}{\text{Total Floor Area} \times 10^{-2}}$$

Figure 1

- 8 -

Uranium Surface Levels and Urinary Excretion Rates
(Feed Manufacturing Facility)

Figure 2

Spot-Air Activity and Urinary Excretion Rates

Figure 3

- 10 -

Shift-Length Air Activity and Urinary Excretion Rates

Figure 4

Shift-Length Air Activity and Spot Air Activity

Figure 5

Spot-Air Activity and Surface Contamination Levels

Figure 6

Shift-Length Air Activity and Surface Contamination Levels

Uranium Surface Levels and Urinary Excretion Rates
(Uranium Recovery, UF₆ Sampling, and Follow-up of Feed Manufacturing
Testing "Fit" to Figure 1)