Conf-9411105--4

PNL-SA-24340

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November 1994

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Presented at the
40th Conference on Bioassay, Analytical & Environmental Radiochemistry
November 13-17, 1994
Cincinnati, Ohio

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

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PRACTICAL ISSUES IN DISCRIMINATING BETWEEN ENVIRONMENTAL AND OCCUPATIONAL SOURCES IN A URANIUM URINALYSIS BIOASSAY PROGRAM

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INTRODUCTION

Workers at two Department of Energy facilities, the Pantex Plant in Texas and the Hanford Site in Washington, are potentially exposed to class Y depleted or natural uranium. Since trace amounts of uranium are naturally present in urine excretion, site bioassay programs must be able to discern occupational exposure from naturally occurring uranium exposure.

In mid-1985 Hanford established a 0.2-μg/d environmental screening level for elemental uranium in urine; the protocol was based on log-normal probability analysis of unexposed workers. A second study of background uranium levels commenced in 1990, and experiences in the field indicated that there seemed to be an excessive number of urine samples with uranium above the screening level and that the environmental screening level should be reviewed (Sula, Carbaugh, and Bihl, 1991). Due to unforeseen problems, that second study was terminated before the complete data could be obtained.

Natural uranium in rock (by weight, 99.27% ^{238}U, 0.72% ^{235}U, and 0.006% ^{234}U) has approximately equal activity concentrations of ^{238}U and ^{234}U. Earlier studies, summarized by the U.S. Environmental Protection Agency in 51 FR 32068, have indicated that ^{234}U (via ^{234}Th) has a greater environmental mobility than ^{238}U and may well have a higher concentration in ground water. By assuming that the ^{238}U to ^{234}U ratio in the urine of nonoccupationally exposed persons should reflect the ratio of environmental levels, significant occupational exposure to depleted uranium would shift that ratio in favor of ^{238}U, allowing use of the ratio as a co-indicator of occupational exposure in addition to the isotope-specific screening levels. This approach has been adopted by Pantex. The Pacific Northwest Laboratory is studying the feasibility of applying this method to the natural and recycled uranium mixtures encountered at Hanford. The Hanford data included in this report represent work-in-progress.

METHOD

To establish the Pantex environmental baseline against which samples could be compared, 75 urine samples were obtained over a three year period from site personnel not occupationally exposed to uranium. These samples were analyzed by radiochemistry for their ^{234}U and ^{238}U content. At Hanford, a study was commenced in the spring of 1994 with 20 nonoccupationally-exposed individuals providing simulated 24-hour urine samples and a concurrent sample of home

drinking water. Eight of these individuals will be resampled at three-month intervals to investigate the possibility of a temporal fluctuation in background levels. In addition to isotopic uranium, the Hanford samples were also analyzed for elemental uranium using kinetic phosphorimetry.

A lognormal probability analysis (Corley et al. 1981) was performed on the elemental uranium, ^{238}U and ^{234}U data. The n data points were ranked in order of increasing magnitude, and the probable fraction (F) calculated by:

$$PF = \frac{Rank-0.5}{n}$$

The percentile ranking for each datum was then calculated by:

$$Percentile = PF * 100$$

Lognormal plots of the data for elemental uranium, ^{238}U, and ^{234}U, were made using a plotting program (Figures 1, 2, and 3). A line was drawn through the data on the graph to extrapolate the data out to the 99.5 percentile. The ratio of ^{238}U to ^{234}U was calculated for each sample and analyzed using the same method (Figure 4). Selected percentiles and their corresponding urine values are shown in Tables 1 and 2.

TABLE 1. Background Urinary Excretion of Isotopic Uranium in Occupationally Unexposed Workers at Pantex

Percentile	Probability of Exceeding	Sample Activity ^{238}U (dpm/d)	Sample Activity ^{234}U (dpm/d)	Sample Activity Ratio (^{238}U : ^{234}U)
50	0.50	0.045	0.10	0.50
90	0.10	0.12	0.23	1.0
95	0.05	0.16	0.30	1.2
99	0.01	0.25	0.52	1.7
99.5	0.001	0.30	0..60	2.0

TABLE 2. Background Urinary Excretion of Uranium in Occupationally Unexposed Workers at Hanford

Percentile	Probability of Exceeding	U-Mass (μg/d)	Sample Activity ^{238}U (dpm/d)	Sample Activity ^{234}U (dpm/d)	Sample Activity Ratio (^{238}U : ^{234}U)
50	0.50	0.020	0.020	0.028	0.7
90	0.10	0.040	0.072	0.14	2.3
95	0.05	0.055	0.11	0.22	3.5
99	0.01	0.10	0.20	0.50	7.5
99.5	0.005	0.12	0.25	0.65	10

DISCUSSION

From the plots and tables, it is apparent that the two different sites exhibit substantially different lognormal distributions. This cautions against using data for one site as the basis for establishing background levels at another site.

The Pantex data led to the conclusion that the activity of ^{238}U in a sample and its ratio relative to ^{234}U were independent variables. The 95 percentile results for these two parameters were 0.16 dpm/d and 1.2 dpm/d, respectively; 5% of Pantex workers not occupationally exposed to uranium might exceed one or the other of these values. However, the likelihood of a worker not occupationally exposed simultaneously exceeding both values is only 0.0025, or one-in-four-hundred. These two values together are used as the environmental screening levels to initiate dose assessment actions. If the ^{238}U result is below 0.16 dpm/d, any observed activity is considered to be associated with environmental background. Results above 0.16 dpm/d are reviewed in light of the ^{238}U to ^{234}U ratio; if the ratio exceeds 1.2, then occupational intake is concluded, and a dose assessment performed.

The Hanford data are presenting a greater challenge. Preliminary indications based on the first 20 samples indicate that the screening level of 0.2 μg/d may be appropriate for most individuals. However, individuals not matching the lognormal distribution appear as fliers to the distribution, differing by as much as 10 to 30 times.

Comparing the urine excretion with the uranium concentration in drinking water would give mixed results. At levels where the lognormal distribution applies, the correlation between urine excretion and drinking water concentration is poor, with a factor of 30 variation in water concentration for the same urinary excretion.

Unlike the data from Pantex, the Hanford data for ^{238}U and ^{234}U radioactivity in urine samples did not show that ^{234}U excretion in unexposed workers exceeded ^{238}U excretion. Furthermore, use of the ^{238}U to ^{234}U ratio as a co-indicator of Hanford occupational exposure in addition to the isotope-specific screening levels does not appear feasible due to the preponderance of ^{238}U; the essentially natural composition of Hanford uranium cannot be readily distinguished by isotopic analysis from environmental sources.

Data obtained through the routine bioassay monitoring program for Hanford workers has provided some interesting insights. The arithmetic mean value for 414 pre-exposure baselines was 0.027 μg/d, significantly different than the mean value of 0.063 μg/d for 1597 post-baseline routine samples. This observation indicates that there was probably some low-level exposure occurring among the work force. For this comparison, no attempt was made to identify the nature of the exposure. However, for the time period of interest, most workers being sampled were considered as potentially exposed to chronic, low-level, occupational sources of soluble uranium. The 50 percentile value of the in-progress background study (0.020 μg/d) compares very favorably with the routine-monitoring baseline arithmetic mean (0.027 μg/d).

The routine-monitoring program investigates workers who exceed the 0.2 μg/d screening level, driven by bioassay goals for minimum detectable dose. The process of investigating these has been complicated, time-consuming, and expensive. Typically, no obvious occupational exposure is found. Follow-up samples on workers (one month or more after the high-routine) have often showed continued high levels, with no potential additional exposure. Where high results have continued, home water samples have been collected to identify drinking water as a possible environmental source. However, sampling home drinking water one month after a high-routine may not have any pertinence to the original high routine sample: some local municipalities switch their drinking water sources from unconfined to confined aquifers and have interties with other municipalities that get their drinking water from very low-concentration surface rivers. These conditions can easily imply an order-of-magnitude change in drinking water concentration. It is hoped that the current study will be able to help clarify the possible temporal variations.

CONCLUSION

Distinguishing between uranium excretion attributed to natural environmental sources and low-level occupational sources can be extremely difficult. If the background can be well-characterized relative to the occupational source material, then a urinalysis program may be effective. If the background cannot be readily distinguished from an occupational source, then a low-level bioassay urinalysis program may be extremely difficult to evaluate. Under such circumstances, primary emphasis may best be directed toward workplace monitoring programs such as personal air sampling to identify individual exposures.

ACKNOWLEDGEMENT

This work was supported by the U.S. Department of Energy under Contracts DE-AC06-76RLO and DE-AC04-91AL65030.

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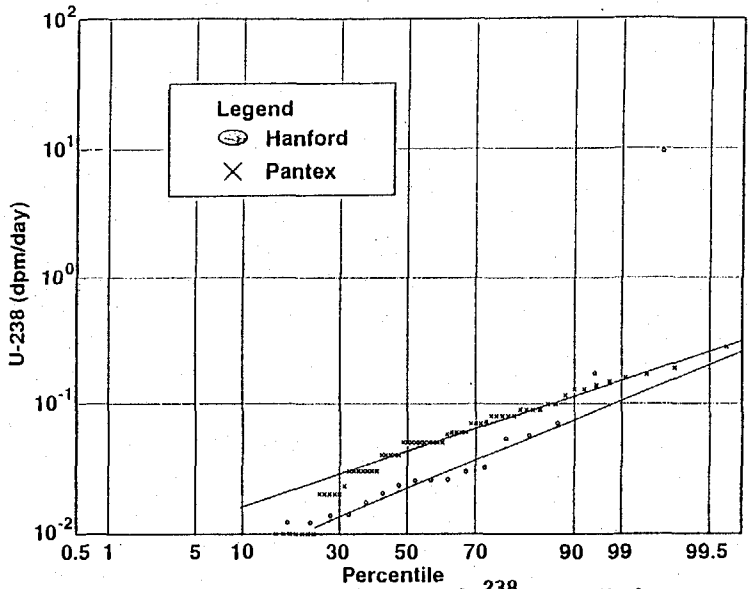

FIGURE 1. Background ^{238}U in Urine

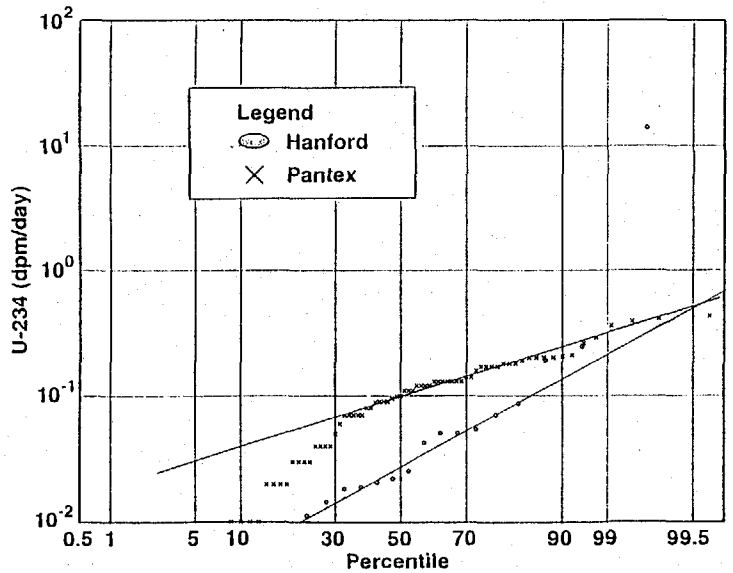

FIGURE 2. Background ^{234}U in Urine

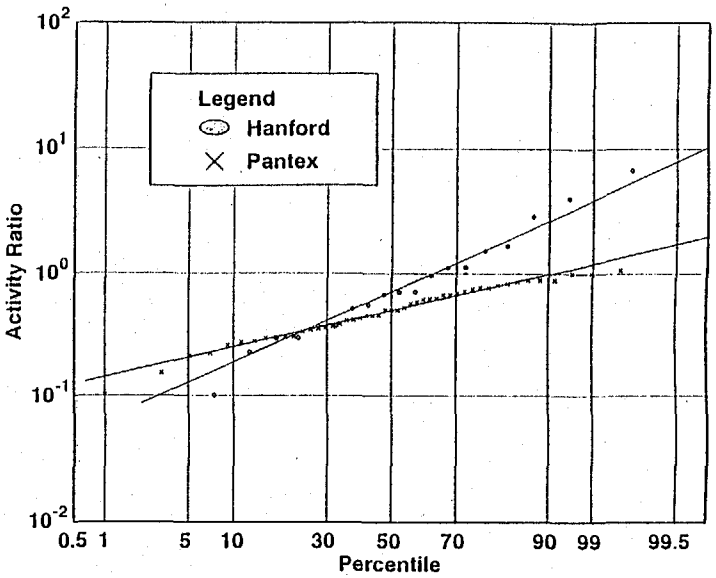

FIGURE 3. Background ^{238}U to ^{234}U Activity Ratio in Urine

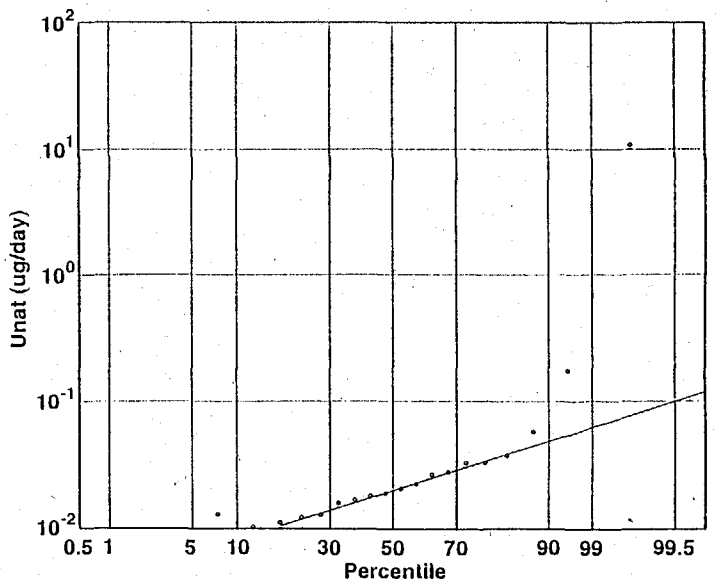

FIGURE 4. Background Elemental Uranium in Urine at Hanford