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Measurements of ^{234}U, ^{238}U and ^{230}Th in Excreta of Uranium Mill Crushermen

Prepared by D. R. Fisher, P. O. Jackson, G. G. Brodacynski, R. I. Scherpelz

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ABSTRACT

Pacific Northwest Laboratory conducted a research program to measure uranium and thorium levels in excreta of uranium mill crushermen who are routinely exposed to airborne uranium ore dust. The purpose of this work was to determine whether ^{230}Th was preferentially retained over either ^{234}U or ^{238}U in the body.

Urine and fecal samples were obtained from fourteen active crushermen with long histories of exposure to uranium ore dust, plus four retired crushermen and three control individuals for comparison. Radiochemical procedures were used to separate out the uranium and thorium fractions, which were then electroplated on stainless steel discs and assayed by alpha spectrometry.

Significantly greater activity levels of ^{234}U and ^{238}U were measured in both urine and fecal samples obtained from uranium mill crushermen, indicating that uranium in the inhaled ore dust was cleared from the body with a shorter biological half-time than the daughter product ^{230}Th. The measurements also indicated that uranium and thorium separate *in vivo* and have distinctly different metabolic pathways and transfer rates in the body. The appropriateness of current ICRP retention and clearance parameters for ^{230}Th in ore dust is questioned.

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ACKNOWLEDGMENTS

The authors gratefully acknowledge the cooperation and assistance of the uranium mill crushermen, retired crushermen, and other persons who provided excreta specimens for this study. We also wish to thank the managers, radiation safety officers, and foremen at the uranium mills who helped us by coordinating visits and discussions with their employees, and by distributing and retrieving the sample collection kits according to schedule. By previous agreement, the names of the above individuals and companies will remain confidential.

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We sincerely appreciate the cooperation, administrative support, and technical review by Dr. Stephen A. McGuire, the contract officer from the Occupational Radiation Protection Branch of the U.S. Nuclear Regulatory Commission.

MEASUREMENTS OF ^{234}U, ^{238}U, and ^{230}Th IN EXCRETA OF URANIUM MILL CRUSHERMEN[a]

INTRODUCTION

Uranium mill crushermen are routinely exposed to uranium ore dust in the crusher building. This report presents results of a series of measurements of ^{234}U, ^{238}U, and ^{230}Th in urine and fecal samples obtained from crushermen. Similar measurements were also made on the excreta of a smaller number of retired crusher workers, and control individuals who live near but who do not work at uranium mills.

Radiometric analyses of excreta samples were performed to:

- measure the rate at which the radionuclides were being excreted from the body,
- determine the excretion pathways involved,
- estimate the body burdens, and
- estimate the annual inhalation exposure to the radionuclides by the crushermen.

The measurement results were compared to excretion rates predicted by the International Commission on Radiological Protection's (ICRP) metabolic models. The levels of ^{234}U, ^{238}U, and ^{230}Th in the excreta of active crushermen were also compared with levels measured in the excreta of retired crushermen and control subjects.

The purpose of this study was to determine whether the measurements of thorium and uranium in excreta would indicate higher body retention of ^{230}Th than of either ^{234}U or ^{238}U. It was intended that the results of this work would provide additional information for assessing the acceptability of the special maximum permissible concentration (MPC) for airborne uranium ore dust given in Title 10, Part 20 of the Code of Federal Regulations (CFR).

(a) Prepared for the U.S. Nuclear Regulatory Commission under a Related Services Agreement with the U.S. Department of Energy under Contract No. DE-AC06-76RLO 1830.

BACKGROUND

The radionuclides ^{234}U, ^{238}U, and ^{230}Th are long-lived alpha emitters belonging to the naturally existing ^{238}U decay series, and are common to the rock dust generated during uranium ore crushing, transfer, and sampling operations at uranium mills. In general, the specific activity of uranium ores is low, and radiation exposures received by crusher workers due to external penetrating radiation or inhaled alpha- and beta-emitting dust particles are usually maintained well below currently established guidelines for radiation protection. Nonetheless, it is desirable that all worker exposures to radioactive materials be kept as low as is reasonably achievable (National Council on Radiation Protection and Measurements (NCRP) 1971; ICRP 1977b).

Workers are protected by federal regulations which limit the permissible concentration of radioactive materials in the air that is breathed. Operational guidelines have been derived in terms of maximum permissible concentrations (MPCs) which, during a 40-hour-per-week occupational exposure, should not permit the dose to certain important body organs from inhalation of radioactive materials to exceed maximum permissible levels. Current MPCs for individual radionuclides in air and water and for restricted (occupational) areas and unrestricted areas (such as those which are accessible by members of the general public) are given in Title 10, Part 20 of the Code of Federal Regulations (CFR). Separate values are provided for soluble (in body fluids) and insoluble forms of the radionuclides. The estimated degree of hazard associated with exposure to radionuclides is reflected in these values; the lower the MPC, the more hazardous the radionuclide. Among members of the ^{238}U decay series, the radionuclide with the lowest MPC is ^{230}Th.

Uranium ore dust is a good example of a mixture where more than one radionuclide is present in air or water. For regulatory purposes, operational limits for this case can be derived according to the rule of mixtures (ICRP 1977a):

$$\frac{1}{L_m} = \sum_i \frac{P_i}{L_i} \qquad (1)$$

where

L_m = the MPC for the average activity concentration of the mixture
P_i = the activity fraction of nuclide i
L_i = the limit given for nuclide i.

A similar formula is given in 10 CFR Part 20 (Note 5 to Appendix B), along with rules for determining MPCs for mixtures with unknown concentrations or unidentified radionuclides.

Since 1960, the Code of Federal Regulations has contained an additional special provision for the limiting value of a mixture of radionuclides consisting of ^{238}U and its daughters in uranium ore dust prior to the chemical separation of uranium from the ore at the mill (Statement of Considerations 1960). The operational limits for airborne uranium ore dust according to this provision are currently given in Note 4 of Appendix B (10 CFR Part 20) as 10^{-10} μCi/mℓ gross alpha activity and 5×10^{-11} μg/m^3 natural uranium in air. The use of these special MPCs has been favored by the uranium industry, since it is difficult to chemically separate the uranium series radionuclides and measure the activity of each radionuclide individually.

The special MPCs for uranium ore dust were originally derived on the assumption that the long-lived alpha emitters in the uranium series down through radium (^{238}U, ^{234}U, ^{230}Th, and ^{226}Ra) were present in the aerosol in secular equilibrium, and that upon entering the body they were similarly metabolized. However, later studies indicated that ^{226}Ra may exist in disequilibrium from ^{238}U and ^{230}Th in the ore dust (Sill 1977; Jackson and Thomas 1980), and that the ^{230}Th separated readily *in vivo* from ore dust inhaled by rats, beagles, and hamsters (Stuart and Beasley 1965; Stuart and Jackson 1974). Compared to uranium, thorium was found to be preferentially retained in the lungs and thoracic lymph nodes of beagles (Stuart and Jackson 1974, 1975; Stuart and Beasley 1967). Although inhaled thorium oxide exhibited relatively slow clearance from the lung (Ballou and Hursh 1972; Albert 1966;

Stuart and Jackson 1975), with time it relocated to the skeleton and tracheobronchial lymph nodes (Wrenn et al. 1981; Stuart and Jackson 1975).

If the maximum permissible concentrations of individual radionuclides in ore dust were considered separately, the MPC derived from the rule of mixtures (Equation 1) would be lower by approximately a factor of three than the special MPC for uranium ore dust. Since some studies indicate that the thorium daughter may separate from the ore and be selectively retained in lung, lymph, and skeleton, the technical basis for the special MPC for uranium ore dust may be questionable.

The present study was undertaken to determine the degree of accumulation of uranium and thorium in body tissues of uranium millworkers. This was accomplished by using indirect bioassay techniques, and by comparing the bioassay results with current ICRP models of the metabolism of inhaled radioactive materials.

SUMMARY AND CONCLUSIONS

1. Wide variations were found in uranium and thorium (^{230}Th) levels measured in the urine and feces of active uranium mill crushermen. Workers are exposed to uranium ore dust under many different occupational conditions, and variation is expected among individuals in the uptake, metabolism, and clearance of radionuclides in the uranium ore dust.

2. The amounts of ^{234}U or ^{238}U measured in urine and fecal samples of active crushermen and control volunteers were significantly greater than the amounts of the daughter product ^{230}Th. This disequilibrium was statistically significant on each sampling day.

3. The observation that uranium excretion predominated over ^{230}Th excretion leads to the conclusion that ^{230}Th is preferentially retained in the body and is cleared with a longer biological half-time than either ^{234}U or ^{238}U. The uranium and thorium components in respirable ore dust appeared to undergo a physical separation in the body following lung deposition, to follow distinctly separate metabolic routes, and to have different transfer rates between various organs of the body.

4. The uranium and thorium fractions in the metabolized ore dust were excreted principally in the feces. More than 97% of the excreted ^{230}Th and more than 75% of the excreted uranium were associated with feces. This was found to be true for both long-term and short-term clearance components of the inhaled or ingested materials.

5. Active crushermen excreted significantly higher levels of ^{234}U or ^{238}U than did retired workers or controls. Significantly higher levels of ^{230}Th were also excreted in feces by active crushermen (except in day 4 samples, which were not statistically different).

6. Levels of ^{230}Th measured in urine of active crushermen were similar to levels measured in retired workers. Thus, urinalysis does not appear to be a suitable method for monitoring worker exposure to ^{230}Th.

7. Significantly more ^{230}Th was measured in urine of retired crushermen than in urine of controls. In contrast, levels of ^{234}U or ^{238}in urine of retired workers were not statistically different from levels measured in urine of controls. These results show additional evidence for a very long clearance half-time for systemic ^{230}Th relative to uranium.

8. The results of measurements on excreta samples for ^{234}U, ^{238}U, and ^{230}Th in ore dust were compared with excretion rates predicted by a combined metabolic model based on the ICRP model and its transfer rate coefficients for internal dosimetry. A comparison of radionuclide levels in daily samples to levels in daily urine samples showed that both the uranium and thorium components of ore dust follow excretion patterns that are characteristic of inhaled Class Y materials.

9. The ICRP metabolic model was used to estimate body burdens, inhalation rates, and annual intakes of ^{238}U and ^{230}Th for active crushermen. This study indicates, however, that the ICRP parameters for thorium may not be appropriate when applied to inhalation of ore dust. Further study of thorium retention and metabolism is needed.

METHODS

RECRUITMENT OF VOLUNTEERS

A review was made of all operating uranium mills in the western United States for the purpose of selecting mills from which volunteer crusher operators might be obtained for this study. Selection of mills was made on the basis of the following criteria:

- number of workers with a minimum of five years' experience in ore crushing operations
- facilities with indoor rock-crushing machinery
- dry crushing and grinding operations
- willingness of mill management to cooperate with the study.

Seven uranium mills were then visited for the purpose of introducing this project to mill management and safety directors, and to obtain volunteer crushermen to participate in the excreta collection phase of the study. Three additional mills in two states refused to allow site visits. For the most part, the mill managers who permitted site visits were also willing to assist with the study, although mixed reactions were observed. At each facility visited, plant tours were permitted to observe operating conditions.

One of the objectives of this work was to determine the concentrations of uranium and thorium in excreta of "maximally exposed" crushermen. Particular care was taken in the selection of crushermén and ore station pickers for participation in the study. We looked for workers with at least five years' experience as crusher operators who had not been assigned to the yellowcake precipitation, drying, or loading area of the mill, and who also spent a considerable fraction of their working day inside the crusher building.

Workers who agreed to participate and who fulfilled the minimum requirements of the study were difficult to locate. At four of the seven mills visited, no workers would volunteer. At other mills, some workers who initially agreed to participate later declined. Ultimately, a total of 14 crushermen at three different mills participated fully in the study. Both acid

and alkaline leach processes were represented. Also, these mills had some of the dustiest crushing circuits observed in the industry. Thus, it is possible that the workers selected for this study are representative of some of the highest exposures currently to be found.

A typical crusher building worker is pictured in Figure 1. Respirators are not normally worn by workers in mill rock-crushing buildings.

Five retired crushermen also agreed to serve as volunteer subjects, although one later declined due to poor health (Hodgkins disease). Retired

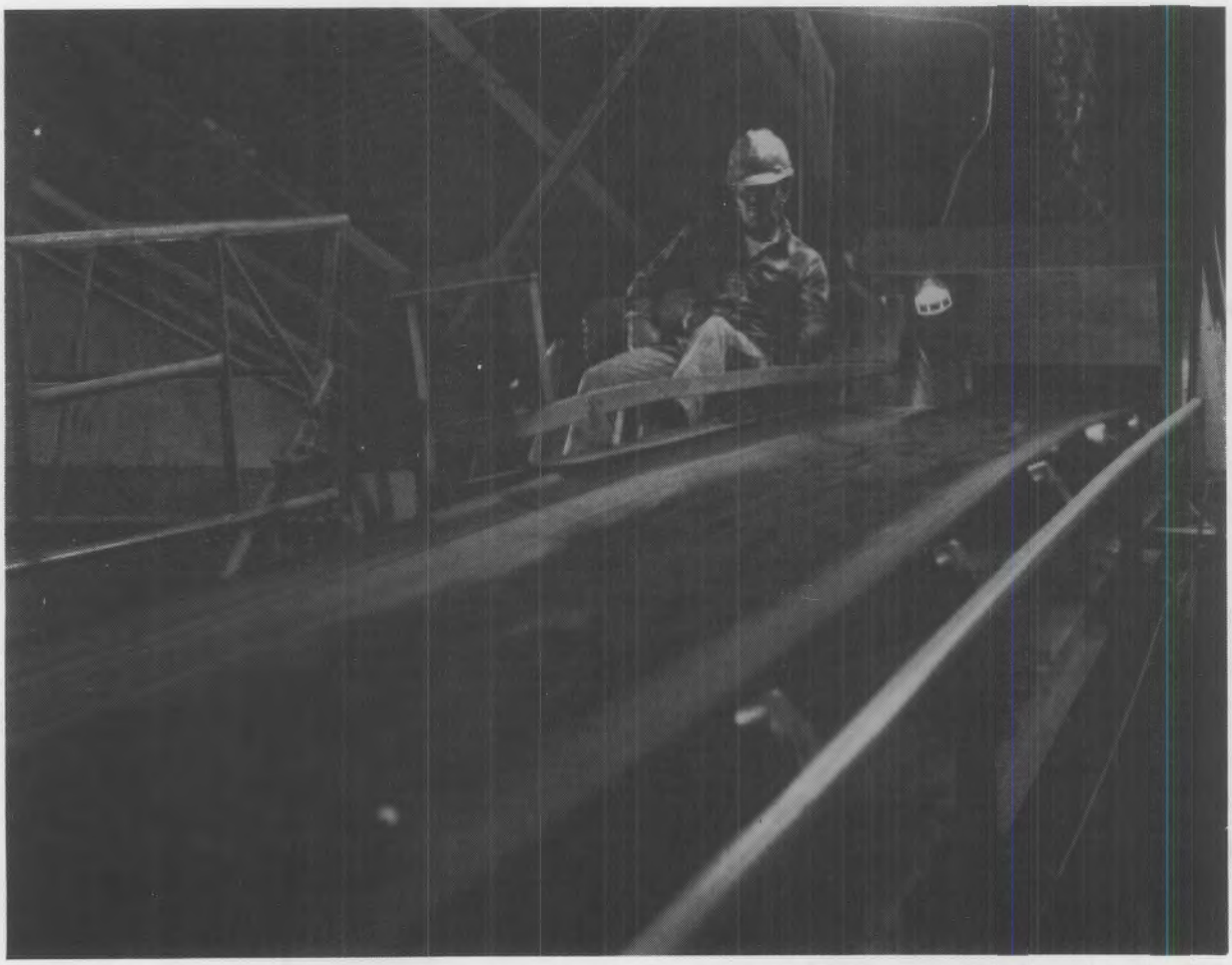

FIGURE 1. A "Station Picker" at the Top of the Ore Conveyor in the Mill Crusher Building Removes Wood Splinters, Wire, and Other Debris from the Crushed Ore Which Might Interfere With Ore Processing

workers were more responsive to the study than were active millworkers. Three members of the general public, who lived in the same communities as the millworkers but who did not themselves work at the uranium mills, were enlisted to serve as control subjects.

COLLECTION OF EXCRETA SAMPLES

New one-liter, screw-cap polyethylene bottles were used for the collection of urine. Prior to collection of the urine samples, the bottles were thoroughly cleaned and their internal surfaces were leached by filling with 4M HNO_3, which was allowed to stand in the bottles overnight. The bottles were then rinsed with distilled water and drained thoroughly.

Sample collection kits were prepared and shipped to volunteer subjects. The contents of each three-day sampling kit are shown in Figure 2. Detailed instructions were enclosed with each kit. Sample collection kits sent to retired workers and controls subjects contained sufficient supplies for the packaging of a two-day sampling. Care was taken to provide extra packing, double bagging, and plastic gloves for prevention of sample contamination by dust containing uranium or thorium. Duplicate kits were sent to workers who failed the first time to follow the sample collection schedule or correct procedures.

Excreta specimens were collected by the volunteer at home according to the instructions given him. Workers were sampled on days 1, 3 and 4 during the four-day-off period at the completion of the monthly A-B-C-D shift schedule. Retired workers and control individuals were instructed to obtain their excreta specimens on any two consecutive days. One fecal sample and a maximum volume of one liter of urine were collected each day.

Fecal samples were collected using a commode specimen collection system.(a) These containers consist of a snap-cap, quart-sized cup attached to a plastic winged collar (Figure 2) that is placed between the toilet seat and the bowl.

(a) Sage Products, Elk Grove Village, Illinois.

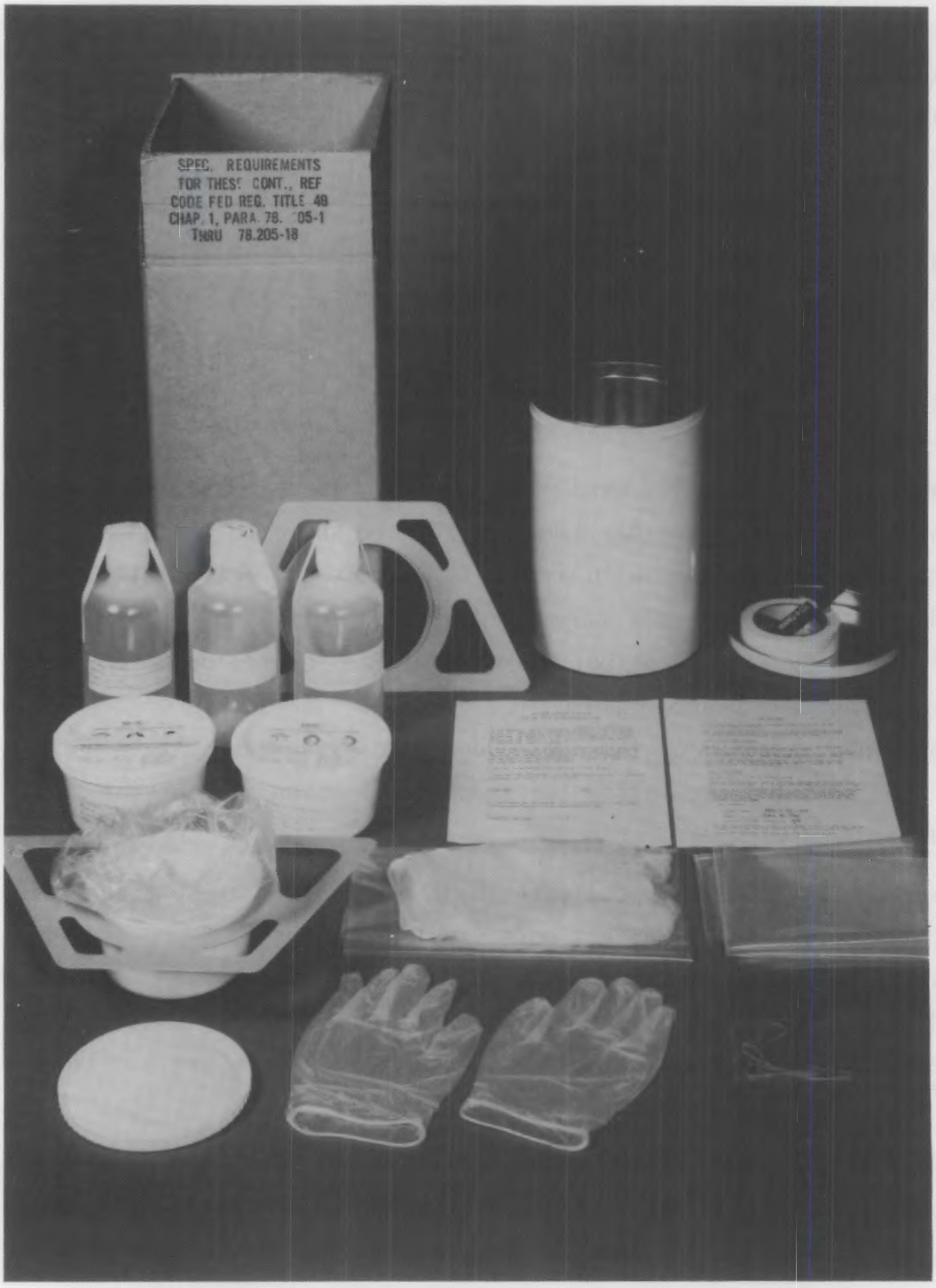

FIGURE 2. Contents of the Three-Day Excreta Sample Collection Kit

The cups are lined with two plastic bags, one inside the other. The bags were new, but not pre-cleaned prior to use. Volunteers were instructed to void feces into the inner bag only, and while wearing disposable plastic gloves, to seal the inner bag with a rubber band. The outer bag was pulled over the inner bag with the specimen, and then sealed. The double-bagged specimen was then placed back into the commode collector cup, and the lid was snapped on. After the winged collar was removed, the cup was placed in a metal cannister which was then taped shut.

The volunteers were instructed to clearly label their specimen collection containers using the marking pen provided in the sample collection kit. Sample identification included the date the sample was obtained, the volunteer's name, and an estimate of the fraction of the total daily fecal or urine excretion represented by the sample. The filled urine bottles and fecal specimen canisters were then packed inside a heavy cardboard carton and shipped back to our laboratory.

ANALYSIS OF EXCRETA

Sample Dissolution

Urine. The volume of urine in each bottle was measured and the samples were transferred to borosilicate glass beakers. Iron and phosphate carriers, nitric acid (HNO_3), and hydrogen peroxide (H_2O_2) were added to the urine in quantities proportional to sample volume. These additives provided for complete exchange of carrier ions with the uranium and thorium in the sample, and prevented hydrolysis during sample evaporation. A constant aliquot of calcium carrier and the radiotracers ^{232}U and ^{228}Th were added, and the urine was heated to near-boiling for gradual evaporation to dryness.

The salts in the sample were oxidized by digestion with HNO_3 and perchloric acid ($HClO_4$) to break down organic residues. When the $HClO_4$ began to fume, the beaker was covered and the sample was refluxed for two hours. The salts were then dissolved in dilute HNO_3 by heating the mixture until a clear solution was obtained. This process destroyed the turbid solutions which otherwise often formed during dilution of the sample.

Feces. Fecal samples were weighed upon receipt at the laboratory. The inner plastic bag with the sample inside was then transferred to a borosilicate glass beaker in a fume hood, and the tare weight of the outer bag and cup was determined. The inner bag was then opened, and 100 mℓ of 0.1 M HNO_3 was added. Low acid strength minimized the possibility of a potentially hazardous and violent oxidation reaction with the sample during subsequent ashing and drying procedures. Calcium, iron, and phosphate carriers plus ^{232}U and ^{228}Th radiotracers were then added to the sample. The nitric acid kept the ions and tracers in an unhydrolized form, and provided sufficient volume for exchange with suspended constituents in the sample.

The beaker was placed in a 90°C muffle furnace within a fume hood, and the samples were slowly evaporated without boiling. The furnace temperature was then raised to 200°C, 250°C, 400°C, and 450°C incrementally to prevent spattering or ignition of combustible gasses which evolved from the sample. The sample was then ashed to 450°C overnight.

Once the beaker had cooled, concentrated HNO_3 and 30% H_2O_2 were added and boiled with the ashed sample to destroy traces of carbon and easily oxidized organic material. Final destruction of organic matter and dissolution of insoluble species in the sample were accomplished by boiling with a mixture of HNO_3, $HClO_4$, hydrochloric acid (HCl), and hydrofluoric acid (HF) in teflon beakers until the $HClO_4$ fumed. Treatment with HF was repeated to volatilize residues of silica and solubilize difficult elements such as titanium. The residue was then dissolved in dilute nitric acid.

Radiochemical Separations

The radiochemical separations which were used to purify the uranium and thorium fractions from the excreta samples are variations of the isotope dilution method previously reported for separating these elements from ore dust filters and tissue samples (Jackson and Thomas 1980; Beasley 1965).

The dissolved excreta samples were wet-ashed, and the uranium and thorium fractions were co-precipitated on calcium ammonium phosphate and ferric hydroxide. Since substrate ions were present in high concentration in the urine and fecal samples, the large precipitate made extraction separation

techniques such as those previously described by Jackson and Thomas (1980) somewhat unmanageable. To reduce the quantity of precipitate and to increase the efficiency of the solvent extraction purification, the following procedural changes were instituted.

First, the method of Toribara and Koval (1967) for co-precipitating thorium on ferric hydroxide at low (~4.0) pH was employed. This step was repeated to remove the bulk of the calcium ammonium phosphate, which is the predominant constituent of the initial precipitate. However, uranium was only partially (30% to 60%) carried on this precipitate, and an additional step was required to separate the uranium. A ferric hydroxide precipitation in the presence of carbonate ions (Galkin and Sudarikov 1964) was used to hold the uranium carbonate complex in the supernate solution while the thorium was carried on the precipitate. For urine specimens, the latter step was sufficient for removing the bulk of the precipitate. For fecal specimens, it was necessary to repeat the ferric hydroxide precipitation at low pH once or twice before separating out ^{230}Th (Figure 3).

The second change which was required to increase the efficiency of solvent extraction was the dry ashing of fecal samples. Jackson and Thomas (1980) avoided this step because ^{210}Po was being measured, and the ashing procedure would cause ^{210}Po to be lost by volatilization.

Since uranium is not effectively separated from the bulk of the ions in the supernate of the precipitation steps, the volume of liquid is much too great for the tertiary amine extraction method (Beasley 1965) to be effective. Uranium was therefore separated from the material by evaporation to dryness and dilution of the material using 7M HCl. The uranium fraction was then removed by passing the diluted solution through a strong anion exchange resin (Figure 4). Most of the iron originally present in the sample had previously been separated from the uranium during the carbonate precipitation steps. Elution with ammonium iodide dissolved in 10M HCl removed the remainder of the ferrous ion. Some fecal samples tended to salt out on the ion exchange resin unless generous volumes of HCl were used to dilute the material. When this occurred, it was necessary to clean the outlet of the separation column to maintain

FIGURE 3. Co-Precipitation of Thorium on Ferric Hydroxide at Low pH

FIGURE 4. Separation of Uranium Using Ion Exchange Resins

adequate flow. The uranium was normally eluted free of salts using 0.1M HNO_3. In some cases it was necessary to repeat the ion exchange separation to obtain a uranium fraction sufficiently free of carrier for electrodeposition to proceed.

The thorium fraction which co-precipitated on ferric hydroxide was first purified by extraction into Alamine-336 in xylene from an aluminum nitrate-tetrapropyl ammonium nitrate salting solution (Figure 5). After back extraction with 7M HCl, the thorium was freed of any remaining traces of uranium and aluminum by co-precipitation on neodymium fluoride. Following volatilization of the fluoride (HF), the thorium was made carrier-free by extraction into thenoyltrifluoroacetone (TTA) in xylene, and back extraction into 2M HNO_3.

The final step involved electroplating the individual uranium and thorium fractions from a sulfuric acid medium onto stainless steel discs.

FIGURE 5. Thorium Fraction Purification by Extraction into Alamine-336 in Xylene

Alpha Spectrometry

The uranium or thorium content of each prepared sample was determined by measuring the alpha particle emissions of ^{238}U (4.15, 4.2 MeV), ^{234}U (4.72, 4.77 MeV), and ^{230}Th (4.62, 4.68 MeV) using a conventional multichannel alpha spectrometer. Silicon surface-barrier diode detectors were employed in the counting apparatus (Figure 6).

Recoveries of uranium and thorium were determined from measurement of ^{232}U (5.26, 5.32 MeV) and ^{228}Th (5.34, 5.43 MeV) tracers which were added to the raw excreta specimens. Thorium-228 decays to ^{224}Ra and its daughters within a relatively short time following electroplating of the sample, and the ingrowth of ^{224}Ra and daughter activity must be taken into account when determining the thorium recoveries. Since 5% to 6% of the alpha particle energies from ^{224}Ra overlap with the 5.43-MeV alpha particle energy from ^{228}Th, an appropriate correction factor was included in the calculations.

FIGURE 6. Measurement of Samples on Stainless Steel Discs Using a Multichannel Alpha Spectrometer

The alpha spectrometer was calibrated using a second ^{234}Pu standard on a stainless steel disc. The secondary standard was calibrated against a National Bureau of Standards radium-D,E,F source.

RESULTS

MEASUREMENTS OF URANIUM AND THORIUM IN EXCRETA

Each urine and fecal sample obtained from active crushermen, retired workers, or control volunteers was carefully assayed for its ^{234}U, ^{238}U, and ^{230}Th content. The results of these measurements are given in Table 1 (urine) and Table 2 (feces). The identification and work location of each study participant remains confidential. The symbol "W" refers to active millworkers, "R" refers to retired millworkers, and "C" refers to control individuals. The results are given as net activity measured in disintegrations per minute (dpm) above background, plus or minus an absolute standard deviation. The activities are reported as total quantity of uranium or thorium excreted per day. Daily excretions of ^{234}U, ^{238}U, and ^{230}Th were derived from the measurements made on the samples received and information provided by the study participants concerning the fraction of the total daily excretion represented by those samples. No creatinine analyses were performed to verify the estimated fractions for urine samples. The unit dpm can be readily converted to picocuries (pCi) by dividing by 2.22, or to Becquerel (Bq) by dividing by 60.

"Natural" uranium is a mixture of ^{238}U, ^{235}U, and ^{234}U in the ratio 0.9928/0.0072/0.000055, as typically found in uranium-bearing rock and soils. The natural uranium content of the study participants' urine samples was determined for comparison to existing regulatory guidelines. Only one urine sample exceeded 15 µg/ℓ, and that sample was found to contain ∿16.5 µg/ℓ natural uranium. The natural uranium content of each urine sample is presented in the Appendix, Table A.1.

The participants' daily fecal excretion of natural uranium was determined for comparison to other published results. The quantity of natural uranium excreted per day in feces is given in the Appendix, Table A.2.

RADIOCHEMICAL RECOVERIES

Radiochemical recoveries of tracers added to excreta samples were determined for each sample to evaluate the efficiency of the radiochemical separation

TABLE 1. Net Activity Measured in Urine Samples (dpm/day)

Subject	Day	^{234}U	^{238}U	^{230}Th
W-1	1	10.2 ± 0.30	9.68 ± 0.32	0.332 ± 0.094
	3	3.42 ± 0.13	3.28 ± 0.13	0.212 ± 0.064
	4	5.82 ± 0.61	6.08 ± 0.62	0.448 ± 0.102
W-2	1	3.06 ± 0.21	2.70 ± 0.19	0.290 ± 0.070
	3	1.10 ± 0.11	0.825 ± 0.112	0.345 ± 0.126
	4	1.80 ± 0.11	1.82 ± 0.11	0.092 ± 0.080
W-3	1	1.99 ± 0.15	1.87 ± 0.19	0.029 ± 0.063
	3	0.930 ± 0.097	0.805 ± 0.085	0.058 ± 0.043
	4	0.762 ± 0.089	0.596 ± 0.093	0.065 ± 0.077
W-4	1	22.3 ± 0.80	20.3 ± 0.70	0.570 ± 0.078
	3	12.5 ± 0.40	12.1 ± 0.40	0.423 ± 0.049
	4	8.80 ± 0.31	9.01 ± 0.31	0.309 ± 0.051
W-5	1	0.130 ± 0.068	0.039 ± 0.056	-0.002 ± 0.039
	3	0.206 ± 0.057	0.148 ± 0.049	0.008 ± 0.033
	4	0.186 ± 0.060	0.163 ± 0.052	0.104 ± 0.045
W-6	1			
	3	(a)	(a)	(a)
	4			
W-7	1	18.8 ± 1.0	19.0 ± 1.0	0.924 ± 0.178
	3	4.40 ± 0.21	4.27 ± 0.20	0.080 ± 0.038
	4	2.18 ± 0.14	2.47 ± 0.14	-0.002 ± 0.036
W-8	1			
	3	(a)	(a)	(a)
	4			
W-9	1	0.801 ± 0.070	0.673 ± 0.063	0.017 ± 0.033
	3	0.691 ± 0.056	0.693 ± 0.056	0.067 ± 0.041
	4	0.619 ± 0.062	0.569 ± 0.057	0.143 ± 0.062
W-10	1	1.77 ± 0.10	1.75 ± 0.10	0.246 ± 0.076
	3	1.41 ± 0.10	1.30 ± 0.10	0.053 ± 0.036
	4	2.82 ± 0.30	2.95 ± 0.30	0.240 ± 0.087
W-11	1	5.19 ± 0.84	5.20 ± 0.84	0.044 ± 0.102
	3	1.85 ± 0.08	2.43 ± 0.11	0.230 ± 0.105
	4	5.65 ± 0.28	5.73 ± 0.27	0.045 ± 0.114

(a) Total daily excretion not indicated on sample.

TABLE 1. (Continued)

Subject	Day	^{234}U	^{238}U	^{230}Th
W-12	1	11.1 ± 0.70	12.5 ± 0.80	0.360 ± 0.107
	3	2.54 ± 0.22	2.62 ± 0.22	0.119 ± 0.026
	4	1.44 ± 0.11	1.46 ± 0.11	0.068 ± 0.024
W-13	1	5.28 ± 0.23	4.79 ± 0.21	0.434 ± 0.081
	3	3.05 ± 0.12	3.05 ± 0.12	0.227 ± 0.035
	4	0.22 ± 0.03	0.183 ± 0.027	0.003 ± 0.020
W-14	1	1.52 ± 0.11	1.42 ± 0.11	0.269 ± 0.073
	3	1.44 ± 0.11	1.71 ± 0.12	0.254 ± 0.063
	4	0.794 ± 0.111	0.621 ± 0.099	0.150 ± 0.045
R-1	1	0.136 ± 0.078	0.084 ± 0.072	0.144 ± 0.047
	2	0.101 ± 0.040	0.111 ± 0.038	0.027 ± 0.033
R-2	1	0.059 ± 0.026	0.053 ± 0.025	0.002 ± 0.030
	2	0.047 ± 0.030	0.072 ± 0.025	0.022 ± 0.040
R-3	1	0.017 ± 0.058	0.049 ± 0.065	0.127 ± 0.037
	2	0.095 ± 0.033	0.068 ± 0.030	0.114 ± 0.032
R-4	1	0.315 ± 0.050	0.285 ± 0.047	0.375 ± 0.072
	2	0.322 ± 0.059	0.298 ± 0.056	0.609 ± 0.131
C-1	1	0.078 ± 0.071	0.095 ± 0.056	-0.030 ± 0.035
	2	0.115 ± 0.057	0.075 ± 0.037	0.021 ± 0.026
C-2	1	0.149 ± 0.045	0.155 ± 0.036	0.197 ± 0.032
	2	0.051 ± 0.038	0.010 ± 0.031	0.033 ± 0.033
C-3	1	0.101 ± 0.026	0.044 ± 0.024	-0.013 ± 0.020
	2	0.015 ± 0.054	0.089 ± 0.044	0.016 ± 0.031

TABLE 2. Net Activity Measured in Fecal Samples (dpm/day)

Subject	Day	^{234}U	^{238}U	^{230}Th
W-1	1	886 ± 32	934 ± 33	516 ± 15
	3	176 ± 5.0	156 ± 4.0	63.0 ± 2.2
	4	57.6 ± 1.4	47.6 ± 1.2	14.6 ± 0.5
W-2	1	2253 ± 77	1930 ± 66	69.1 ± 3.0
	3	48.8 ± 1.6	37.5 ± 1.3	2.60 ± 0.45
	4	42.7 ± 8.8	31.8 ± 6.5	4.94 ± 0.30
W-3	1	155 ± 14	143 ± 13	38.4 ± 1.4
	3	145 ± 5.0	139 ± 5.0	22.3 ± 0.8
	4	20.2 ± 0.5	17.2 ± 0.5	6.49 ± 0.35
W-4	1	790 ± 24	756 ± 23	443 ± 15
	3	340 ± 9.0	320 ± 8.0	129 ± 5.0
	4	57.0 ± 7.0	50.9 ± 6.1	13.1 ± 0.5
W-5	1	2.44 ± 0.12	2.22 ± 0.13	1.26 ± 0.13
	3	10.9 ± 0.3	9.78 ± 0.30	5.80 ± 0.32
	4	11.6 ± 0.38	10.8 ± 0.40	8.94 ± 0.82
W-6	1	125 ± 4.0	125 ± 4.0	94.2 ± 3.1
	3	15.6 ± 0.7	16.3 ± 0.7	10.9 ± 0.5
	4	288 ± 7.0	289 ± 7.0	202 ± 7.0
W-7	1	1188 ± 62	1128 ± 60	916 ± 28
	3	356 ± 11.	346 ± 11	236 ± 6.0
	4	22.1 ± 0.6	19.7 ± 0.6	7.02 ± 0.32
W-8	1			
	3	(a)	(a)	(a)
	4			
W-9	1	172 ± 4.0	151 ± 4.0	76.2 ± 2.6
	3	45.3 ± 1.4	45.7 ± 1.4	24.0 ± 1.0
	4	24.7 ± 1.0	18.5 ± 0.7	7.24 ± 0.3
W-10	1	86.1 ± 2.5	76.9 ± 2.2	7.46 ± 1.02
	3	(b)	(b)	(b)
	4	13.1 ± 0.9	8.38 ± 0.62	3.73 ± 0.51
W-11	1	449 ± 15	417 ± 14	194 ± 6.0
	3	12.9 ± 0.4	10.7 ± 0.3	5.44 ± 0.24
	4	24.9 ± 1.3	13.5 ± 0.98	3.42 ± 0.20

(a) Total daily excretion not indicated on sample.
(b) No sample provided, empty container marked "none."

TABLE 2. (continued)

Subject	Day	^{234}U	^{238}U	^{230}Th
W-12	1	105 ± 20	97.8 ± 18.0	36.6 ± 1.3
	3	59.0 ± 2.2	53.8 ± 4.0	27.8 ± 1.0
	4	58.5 ± 1.5	53.1 ± 1.5	21.8 ± 1.5
W-13	1	163 ± 12	147 ± 10	16.9 ± 1.5
	3	34.2 ± 1.6	20.8 ± 1.1	1.22 ± 0.07
	4	29.4 ± 0.8	23.8 ± 0.7	3.54 ± 0.25
W-14	1	90.6 ± 2.2	81.6 ± 2.0	50.4 ± 1.7
	3	45.4 ± 1.2	37.6 ± 1.0	34.2 ± 1.1
	4	4.66 ± 0.16	2.79 ± 0.11	1.08 ± 0.09
R-1	1	17.7 ± 0.6	15.3 ± 0.5	8.94 ± 0.66
	2	22.7 ± 0.9	20.0 ± 0.8	12.3 ± 0.4
R-2	1	7.05 ± 0.86	5.37 ± 0.68	2.17 ± 0.13
	2	2.42 ± 0.19	2.21 ± 0.18	2.60 ± 0.17
R-3	1	(a)	(a)	(a)
	2			
R-4	1	96.1 ± 2.8	63.8 ± 1.9	0.332 ± 0.051
	2	60.8 ± 1.7	41.5 ± 1.2	0.577 ± 0.059
C-1	1	18.0 ± 2.1	13.2 ± 1.6	3.23 ± 0.76
	2	(a)	(a)	(a)
C-2	1	6.19 ± 0.33	5.17 ± 0.29	0.929 ± 0.310
	2	14.5 ± 2.4	11.7 ± 1.9	0.268 ± 0.300
C-3	1	15.1 ± 0.5	7.56 ± 0.30	0.662 ± 0.062
	2	14.7 ± 0.5	8.10 ± 0.32	1.09 ± 0.08

(a) Total daily excretion not indicated on sample.

methods. Initial measurements on a limited number of samples indicated radiochemical recoveries of 50% to 80% for uranium and thorium in urine samples. When recoveries of all samples were later reviewed, it was found that unusually low recoveries (<20%) occurred in approximately 15% of the fecal sample measurements for uranium and thorium and 15% of the urine sample measurements for thorium. The low recoveries observed correlated with batches of samples contributed by individual volunteers, and could have resulted from undetermined dietary preferences. The average and standard deviation of the radionuclide recovery percentages are shown in Table 3.

The recovery averages indicate that a considerable degree of matrix interference remained for the separation schemes employed. It was nonetheless possible to make statistically positive measurements of the concentrations of uranium and thorium in millworker excreta.

TABLE 3. Radionuclide Recovery Percentages for Uranium and Thorium Separations from Excreta Samples

Sample Type	Average Recovery (%)	
	Uranium	Thorium
Urine	52 ± 19	43 ± 22
Feces	36 ± 18	53 ± 27

DISCUSSION

URANIUM AND THORIUM LEVELS IN EXCRETA SAMPLES

The results of measurements on urine and fecal samples for uranium and thorium indicated the following:

- Workers were exposed to many different dust exposure conditions.
- Levels of ^{234}U were similar to levels of ^{238}U in excreta.
- Uranium and thorium were in significant disequilibrium in both urine and feces on all sampling days for each group (crushermen, retired workers, and controls).
- Uranium and thorium in respirable uranium ore dust have distinctly separate metabolic results and transfer rates between various organs of the body.
- Most of the uranium and thorium is cleared from the body in feces rather than urine. This is the case for long-term as well as short-term clearance components of inhaled or ingested materials.
- The excretion of uranium predominates over the excretion of thorium, indicating that ^{230}Th is preferentially retained in the body.

These findings are discussed in the following paragraphs.

Variability Among Workers

The measurement results (Tables 1 and 2) show large differences in the relative amounts of uranium and thorium in excreta from one worker to another. The wide variations may be explained by the fact that the uranium mill crushermen who participated in this study were exposed to many different levels of uranium ore dust under different working conditions. The cumulative amount of ore dust inhaled by individual crushermen is dependent upon length of service and job category, the dust level or dust suppression capabilities of the mill, and the ore type and quality (including percent moisture).

The $^{234}U/^{238}U$ Equilibrium in Excreta

The activities of ^{234}U measured in urine and feces of study participants were found to be similar to but not in exact equilibrium with activities of ^{238}U in the same samples. Among active crushermen, the $^{234}U/^{238}U$ ratio was found to average 1.12 in urine samples and 1.17 in fecal samples. The ratio was somewhat higher and considerably more variable in samples obtained from retired millworkers and control individuals. Among workers, the slight disequilibrium between ^{234}U and ^{238}U was statistically significant on day 1, but not on day 3 and day 4.

Previous studies of uranium mill aerosols showed ^{234}U and ^{238}U to be in approximate activity equilibrium (Sill 1977, Jackson and Thomas 1980). The measured $^{234}U/^{238}U$ ratios in excreta therefore demonstrate a slight preference for retention of ^{238}U over ^{234}U in the body. This effect may be explained by the valence chemistry of the uranium atoms: the parent ^{238}U is usually a tetravalent species in geologic formations, whereas its third-generation decay daughter ^{234}U could exist as a hexavalent species (which is a more soluble chemical state). Since biological processes for removal of uranium depend largely upon the solubility of the material, the valence states of the material could affect its retention in the body. The more soluble forms should be cleared more rapidly by natural processes. Thus, slightly greater amounts of ^{234}U could be expected in excreta samples.

Uranium/Thorium Disequilibrium in Excreta

The amounts of ^{234}U or ^{238}U found in urine and fecal samples of crushermen and control subjects were significantly greater than the amounts of the daughter product ^{230}Th. The disequilibrium between uranium and thorium is illustrated by Figure 7 (urinary excretion) and Figure 8 (fecal excretion). These two figures compare the mean daily excretion of ^{238}U and ^{230}Th by active crushermen, retired workers, and controls. (The activities of ^{234}U in excreta were similar to the acitivies of ^{238}U and are not shown separately in Figures 7 and 8.) The mean activities of ^{238}U and ^{230}Th excreted by retirees and controls for both sample collection days are shown in the figures, since no significant differences between day 1 and day 2 sample means were found. Error bars were not included in the figures because the data were not normalized.

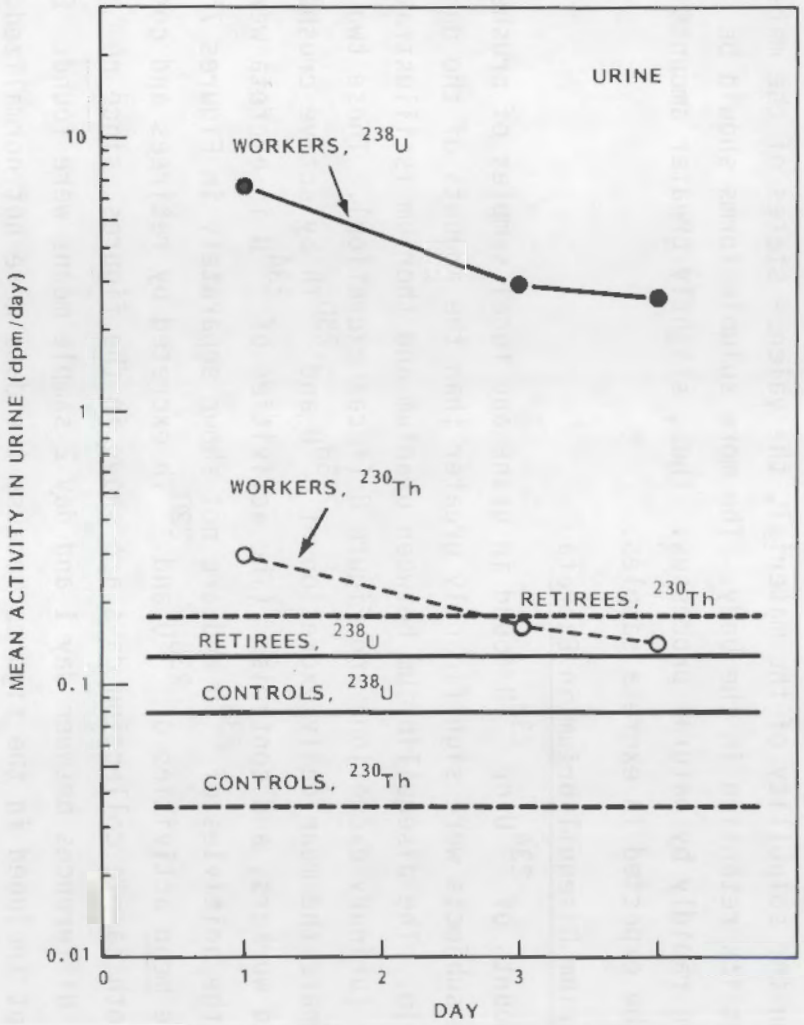

FIGURE 7. Mean Daily Urinary Excretion of ^{238}U and ^{230}Th by Active Crushermen, Retirees, and Control Subjects

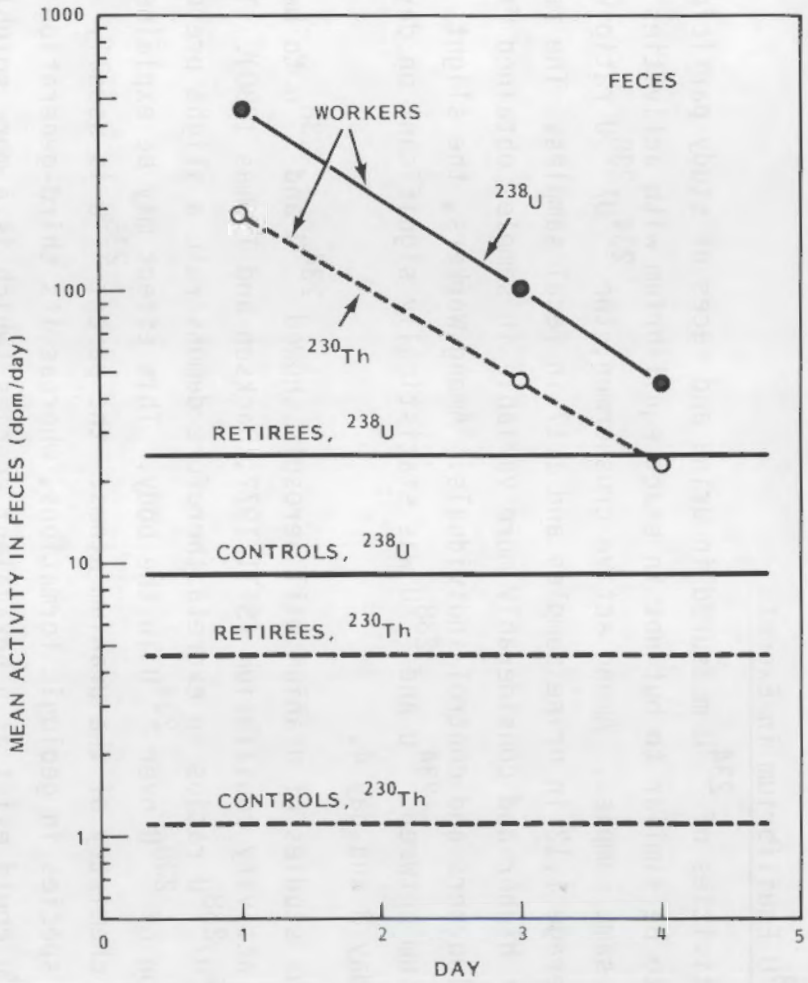

FIGURE 8. Mean Daily Fecal Excretion of ^{238}U and ^{230}Th by Active Crushermen, Retirees, and Control Subjects

The uranium and thorium in excreta of active crushermen result from both chronic (long-term) and acute (recent) inhalations of low-level uranium ore dust. The day 1 average represents the sum of the long-term clearance plus the short-term clearance of recently inhaled materials. The rapidly cleared ore dust component is no longer dominant in excreta samples after a few days away from work, and the urinary and fecal excretion of uranium and thorium is primarily due to longer-term clearance processes. With longer periods of time away from work, the excretion of uranium and thorium by crushermen should approach the levels characteristic of retired crushermen. Control individuals with no unusual exposures to uranium ore dust demonstrated even lower levels of uranium and thorium in excreta (Figures 7 and 8).

Following the inhalation of uranium ore dust, the thorium and uranium fractions apparently undergo a physical separation. Uranium and thorium then exhibit distinctly separate metabolic routes and transfer rates between various organs of the body. This action is evidenced by the large differences in the uranium and thorium activities measured in the excreta samples.

Since uranium and thorium were found to be in disequilibrium in both urine and feces on all sampling days for each group in the study, we also considered the possibility that intakes of uranium were higher than intakes of ^{230}Th. Among controls, the excretion of ^{238}U was approximately twice as great in urine and nine times as great in feces as the excretion of ^{230}Th. A portion of the excess uranium detected in the excreta of control volunteers may have been due to ingestion of uranium in foodstuffs and drinking water which was low in ^{230}Th (see also ICRP 1975 for a discussion of the dietary intakes of uranium and thorium by reference population groups). All of the control volunteers did in fact reside near major uranium mining activities, and it is probable that the local potable water contained dissolved uranium but very little ^{230}Th. Thus, a portion of the uranium/thorium disequilibrium in each study group may be explained by the disproportionate ingestion of these materials.

Uranium millworkers may also be exposed to yellowcake dust--either from direct exposure near the yellowcake precipitation, drying, and packaging areas of the mill, or from inhalation of yellowcake dust released during dryer stack emissions. The yellowcake is depleted in ^{230}Th because thorium is removed

during the uranium refinement process. However, none of the active crushermen who participated in this study had previously been assigned to the yellowcake areas of the mill, and the crushing circuit and yellowcake areas are physically separated such that exposures to yellowcake dust would appear to be unlikely. The feces from one worker (W-2, day 1) did show an unusually high uranium/thorium disequilibrium, which could be attributable to accidental or intentional contamination by yellowcake (see Table 8, footnote "a").

Fecal Versus Urinary Excretion Routes

Uranium and thorium were much more prevalent in fecal samples than in urine, confirming the fact that fecal excretion is the major pathway for clearance of both elements (as ore dust) from the body.

The ratio of uranium in feces to uranium in urine was highly variable among active crushermen. On the average, only about 1% to 5% of the excreted uranium was associated with urine. A smaller fraction of the inhaled thorium was associated with urinary excretion: on the average, only 0.1% to 1% of the excreted ^{230}Th was measured in urine samples. For both uranium and thorium, the ratio of material in feces to material in urine decreased with time away from the job. This effect was due to rapid clearance of a portion of the insoluble dust from the respiratory tract into the gastrointestinal system.

A comparison of Figures 7 and 8 shows that only a small fraction of uranium and thorium excreted by retired workers and control subjects was associated with the urine clearance pathway. It appears that most of the uranium and thorium deposited in the body is slowly cleared by fecal excretion rather than urinary excretion. This mode of long-term clearance could presumably be accomplished by biliary secretions of the liver which dump into the intestinal tract. However, no such provision is given in models currently used for radiation protection dosimetry.

Body Retention of ^{230}Th

The observation that uranium excretion predominates over the excretion of thorium leads to the conclusion that ^{230}Th is preferentially retained in the body and is cleared with a longer biological half-time than is uranium. Excreta analysis does not provide information about the location of materials deposited in the body.

Baseline Levels of Uranium and ^{230}Th in Excreta

A baseline value for the daily urinary excretion of natural uranium can be derived from the results given in Appendix Table A.1. The mean daily urinary excretion of natural uranium by the three control individuals was 0.11 µg/day. This rate is in good agreement with published values for reference man of 0.05 to 0.5 µg/day (ICRP 1975).

A summary of the levels of natural uranium measured in fecal samples of study participants is given in Appendix Table A.2. The mean daily fecal excretion of natural uranium by the three control subjects was 15.1 µg/day, which is about an order of magnitude higher than published values for reference man of 1.4 to 1.8 µg/day (ICRP 1975). As already indicated, the increased levels of natural uranium in excreta of control subjects may be attributable to ingestion of dissolved uranium in local water supplies.

Tables 1 and 2 (main text) provide an indication of the baseline values for excretion of ^{230}Th in urine and feces by non-uranium workers. The mean daily excretion of ^{230}Th by control subjects was 0.037 dpm/day (0.017 pCi/day) in urine, and 1.24 dpm/day (0.56 pCi/day) in feces.

DIFFERENCES AMONG WORKERS, RETIREES, AND CONTROLS

Statistical tests were performed to see if the mean ^{238}U and ^{230}Th activities measured in excreta of workers, retirees, and controls were different, and to see if there were any statistical differences within groups between days on which the excreta samples were obtained. A one-tailed, two-group t-test was used to identify significant differences, if any, in the mean activities measured in the excreta of the study participants.

Activities Measured in Urine

Thorium levels of urine of active crushermen were not statistically different from levels measured in urine of retired crushermen (Figure 7). Urinalysis therefore does not appear to be a suitable method of monitoring worker exposure to ^{230}Th in ore dust. However, significantly more ^{230}Th was measured in urine of retired millworkers than in urine of control subjects

($p < 0.10$). These findings show evidence for long-term systemic clearance of ^{230}Th from retired workers previously exposed for long periods of time to airborne uranium ore dust.

Significantly greater levels of uranium were measured in urine of active crushermen than in urine of either retired workers or control subjects ($p < 0.05$). In contrast, levels of uranium in urine of retired workers were not statistically different from uranium levels measured in urine of controls. These findings provide additional evidence that the uranium incorporated into the body following long-term chronic exposure to ore dust is cleared from the body with a shorter biological half-time than that of ^{230}Th.

Activities Measured in Feces

A two-group t-test showed that significantly greater levels of uranium and thorium were measured in feces of both active and retired crushermen than in feces of control subjects ($p < 0.01$), as demonstrated in Figure 8. Also, significantly greater levels of uranium and thorium were measured in feces of active crushermen than in feces of retired crushermen ($p < 0.1$), except for the day 4 sample collection which was not significantly different. This information shows that the uranium and thorium activities in feces are the result of both short-term and long-term clearance of uranium and thorium derived from inhaled ore dust. The long-term fecal elimination of these materials may result from either clearance of insoluble uranium and thorium from the lungs via the mucous pathway to the gastrointestinal system, or clearance of solubilized material from the bloodstream by the liver and bile duct into the intestines.

Statistical Differences Among Sampling Days

A paired t-test design was used to compare the mean sample measurement results for each group from one sample collection day to the next. Day 1 excreta samples from retired workers and control subjects were not significantly different from day 2 excreta samples. However, among active crushermen, significant differences were observed in the uranium and thorium content of excreta samples obtained on day 1 compared with either day 3 or day 4 samples ($p < 0.05$). Levels of uranium and thorium in day 3 samples were generally

higher than in day 4 samples. Due to the wide variation of individual measurements, the sample means for day 3 were not statistically different than the sample means for day 4.

ESTIMATION OF URANIUM AND THORIUM BODY BURDENS

Urine and fecal bioassay provides an indirect method for estimating:

(1) an individual's exposure to radionuclides, and

(2) residual body burden.

To relate the results of excreta analysis to initial exposure and organ or body burdens, an appropriate biological model is required. The model should describe rates at which the inhaled activity is transferred from the lung to other body compartments or organs until the activity is eventually excreted from the body. The excretion rates predicted by the model can be compared to levels actually measured.

The ICRP metabolic model (ICRP 1979) was chosen for estimating depositions of inhaled ore dust and excretion rates of ^{234}U, ^{238}U, and ^{230}Th from the body. Results of the excreta measurements were then used to back-calculate the individual body burdens and average inhalation exposures to uranium ore dust for each worker who participated in the study.

Metabolic Pathway Modeling

The deposition and translocation of inhaled substances are dependent upon factors such as particle size distribution of the aerosol and its chemical form, and natural clearance mechanisms in the body. Insoluble materials may become lodged on alveolar walls of the lungs or be transported up the bronchial airways by mucous and swallowed. Soluble materials may dissolve in lung fluids and be transferred to the bloodstream, or they may be phagocytized by alveolar macrophages and discarded into the lymphatic system.

Radionuclides present in extracellular fluids are transported by the bloodstream to the kidneys where they are filtered out and excreted in the urine. Thus, urinalysis provides an indication of the systemic movement of materials from the body.

Radioactive materials present in feces may have entered the body by either inhalation or ingestion. Ingested materials pass directly into the intestinal system, where a small amount may be absorbed into the bloodstream. The remaining material is then excreted in the feces.

The ICRP dosimetric model for internally deposited radionuclides (ICRP 1979) was used to estimate intake, deposition, retention, and clearance of inhaled uranium ore dust. The current ICRP dosimetric model includes a modified version of the Task Group on Lung Dynamics' (1966) lung model and Eve's (1966) model for transport of materials through the gastrointestinal (GI) tract. The other organs of interest for this study are the kidney (which constitutes the pathway for urinary excretion), and a compartment designated "all other body tissues." The essential elements of this metabolic model for radiation dosimetry are shown schematically in Figure 9. We shall hereafter refer to this diagram as the "combined metabolic model."

Solubility Classification for Uranium Ore Dust. The ICRP method for dosimetry employs a solubility classification scheme which predicts the fate of inhaled radioactive materials in the body. Three lung clearance categories are involved: D, W, and Y, which correspond to biological clearance half-times from the lung of 0 to 10 days, 11 to 100 days, and more than 100 days, respectively, from the lung (ICRP 1979). The D, W, or Y classification determines which set of transport fractions and rates describe translocation of the material from one anatomical compartment to another.

The short-term dissolution of uranium ore dust in simulated lung fluids was studied by Kalkwarf (1979). His work showed that the different radionuclides common to uranium ore dust dissolve at different rates. From the results of this work, Kalkwarf assigned the uranium in the ore dust to the W classification and ^{230}Th to the Y classification. However, the short term of the study allowed for the possibility that components of the ore dust with considerably longer biological clearance half-times were present. The chemical forms of the uranium fractions of the ore dust were not specified. Earlier, Cooke and Holt (1974) performed similar studies on various uranium compounds and identified uranium trioxide (UO_3) as a class W compound, and uranium dioxide (UO_2) and

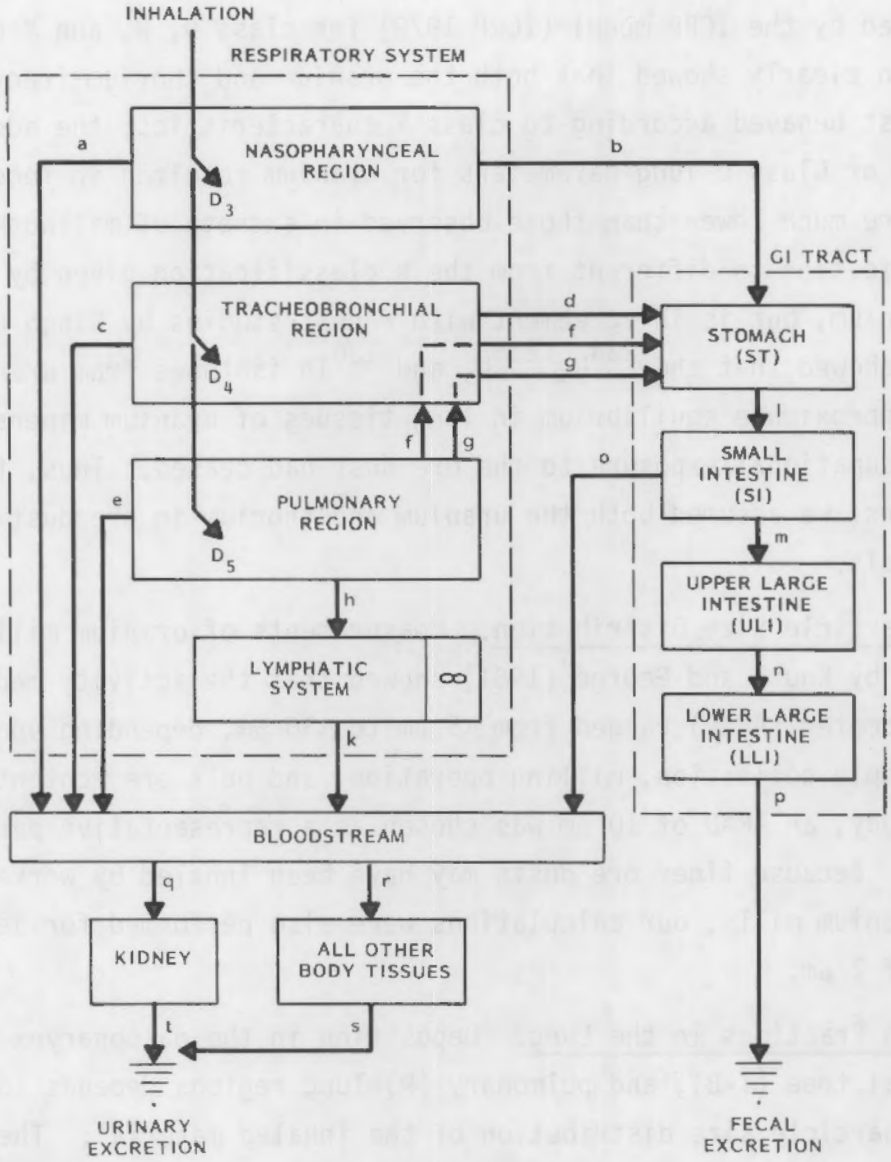

FIGURE 9. Schematic Diagram of the Combined Metabolic Model. The model includes the respiratory system, GI tract, bloodstream (transfer compartment), kidney, and "all other body tissues."

uranium octoxide (U_3O_8) as class Y compounds. However, Cooke and Holt did not investigate the solubility of uranium in its natural state as ore dust.

We studied the solubility characteristics of uranium ore dust by looking closely at the ratio of the amounts of uranium or thorium in feces of crushermen to the amounts in urine. We then compared these feces/urine ratios with

ratios predicted by the ICRP model (ICRP 1979) for class D, W, and Y compounds. This comparison clearly showed that both the uranium and thorium fractions in uranium ore dust behaved according to class Y characteristics; the adoption of either Class W or Class D lung parameters for uranium resulted in feces/urine ratios that were much lower than those observed in excreta of millworkers. This Y classification is different from the W classification given by Kalkwarf (1979) for uranium, but is in agreement with recent studies by Singh et al. (1981), which showed that the ^{234}U, ^{238}U, and ^{230}Th isotopes from uranium ore dust were in approximate equilibrium in lung tissues of uranium miners many years after occupational exposure to the ore dust had ceased. Thus, for our own calculations, we assumed both the uranium and thorium in ore dust to be class Y materials.

Aerosol Particle Size Distribution. Measurements of uranium mill ore dust particle sizes by Knuth and George (1981) showed that the activity median aerodynamic diameter (AMAD) ranged from ~5 μm to ~30 μm, depending upon location of the sample collection, milling operation, and bulk ore content. For the present study, an AMAD of 10 μm was chosen as a representative particle size diameter. Because finer ore dusts may have been inhaled by workers at some of the uranium mills, our calculations were also performed for aerosols with an AMAD of 2 μm.

Deposition Fractions in the Lung. Deposition in the nasopharynx (N-P), tracheobronchial tree (T-B), and pulmonary (P) lung regions depends to a large degree on the particle size distribution of the inhaled material. The deposition fractions given in Table 4 were derived for uranium ore dust of 2 μm and 10 μm AMAD using the ICRP approach.

Lung Clearance and Organ Transfer Rates. The ICRP lung model assumes that 1% of the class Y material deposited in the N-P region will be absorbed into the blood with a biological clearance half-time of 0.01 day (15 minutes). The remaining 99% will be cleared into the GI tract with a clearance half-time of 0.4 day (10 hours). Of the inhaled material which deposits in the T-B lung region, 1% will be absorbed into the blood with a clearance half-time of

TABLE 4. Deposition Fractions for Ore Dust Aerosols of 2 μm or 10 μm AMAD in the Respiratory System (Figure 9). Derived from ICRP (1979).

Region	Deposition Fraction 2 μm	10 μm
Nasopharyngeal (D_3)	0.49	0.85
Tracheobronchial (D_4)	0.08	0.08
Pulmonary (D_5)	0.18	0.05

0.01 day (15 minutes), and 99% will be carried into the GI tract with a clearance half-time of 0.2 day (5 hours). Material deposited in the deep lung (P) is assumed to translocate as follows: 40% will clear into the GI tract with a half-time of 1 day and another 40% will clear with a half-time of 500 days, 5% will dissolve into the bloodstream with a half-time of 500 days, and 15% will clear into the lymphatic tissues with a half-time of 500 days. Ninety percent of the material which enters the lymphatic system will be cleared from the system with a biological half-time of 1,000 days, and 10% is assumed to remain permanently in the lymph tissues (ICRP 1979).

Since thorium oxide in ore dust was shown to behave somewhat differently from uranium (Stuart and Jackson 1974, 1975), we made the following adjustments to the ICRP metabolic data: thorium was assigned an infinite biological retention in the thoracic lymph nodes, and a longer clearance half-time (4 years) was assumed for clearance of thorium from the pulmonary region of the lung to the bloodstream than for clearance of uranium (500 days). These corrections resulted in thorium concentrations in lymph and lung tissue relative to uranium which were consistent with the results of the animal studies by Stuart and Jackson (1974).

Transfer Through the GI System. Ingested material or material cleared from the lungs and swallowed is transported through four compartments of the GI tract (Figure 9). The transfer rates and fractions are specific to each radionuclide. The metabolic data for uranium and thorium were adopted from the ICRP (1979).

A small fraction of the uranium and thorium in the contents of the gastrointestinal system is absorbed into body fluids as the material passes through the small intestines. This fraction is understood to be 0.002 for uranium, and 0.0002 for thorium (ICRP 1979). Thus, very little absorption of either uranium or thorium takes place in the GI tract. Material cleared from the GI tract is assumed to pass directly into the feces.

Other Tissue Compartments. The circulatory system transfers material from the respiratory system and the small intestine to other tissues and organs of the body. We have grouped these tissues together (except for the kidneys) for convenience. The kidneys are part of the urinary excretion pathway, and are therefore represented as a separate compartment in the combined metabolic model.

The other body tissues deliver material back into the bloodstream for filtration and excretion by the kidneys. Rate constants for these pathways are generally unavailable. For simplification of the mathematics, the combined metabolic model assumes that materials from other body tissues are cleared directly from the body, bypassing the kidneys. The same convention is also used by the ICRP (1979) and others (Johnson and Carver 1981), and the reader is referred to the latter publication for a more complete discussion of this problem with the model.

Compartment Transfer Rates for the Combined Metabolic Model. Clearance half-times and fractions used in the combined metabolic model for uranium and thorium are summarized in Table 5.

Body Burden Calculations

The combined metabolic model was used to determine excretion rates and total body burdens of uranium and thorium in a standard reference man with a constant occupational exposure to uranium ore dust. The results of these calculations were later applied in the estimation of body burdens and inhalation exposures of workers who participated in the study. The necessary physiologic data for the reference man were obtained from ICRP Publication 23 (ICRP 1975). The mathematical formulations of Johnson and Carver (1981) served as a guide for the calculations. The recurrence formula of Scherpelz and Desrosiers

TABLE 5. Uranium and Thorium Clearance Half-times T (days), and Fractions f, Used in the Combined Metabolic Model. Adapted from ICRP (1979)

Compartment	Pathway[a]	Uranium		Thorium	
		T	f	T	f
Nasopharyngeal	a	0.01	0.01	0.01	0.01
	b	0.4	0.99	0.4	0.99
Tracheobronchial	c	0.01	0.01	0.01	0.01
	d	0.2	0.99	0.2	0.99
Pulmonary	e	500	0.05	1460	0.05
	f	1.0	0.4	1.0	0.4
	g	500	0.4	500	0.4
	h	500	0.15	500	0.15
Lymphatic	k	1000	0.9	∞	1.0
Stomach	l	0.029	1.0	0.029	1.0
Small intestine	m	0.115	0.998	0.115	0.9998
	o	57.6	0.002	577	0.0002
Upper large intestine	n	0.385	1.0	0.385	1.0
Lower large intestine	p	0.693	1.0	0.693	1.0
Bloodstream	q	0.5	0.657	0.5	0.1
	r	0.5	0.343	0.5	0.9
All other body tissues	s	6	0.349	700	0.222
		20	0.582	8000	0.778
		1500	0.0015		
		5000	0.067		
Kidney	t	0.5	0.816	0.5	1.0
		6	0.183		
		1500	0.0008		

(a) See Figure 9.

(1981) was used for solving the complex exponential equations suggested by Johnson and Carver. A constant inhalation rate of 1 pCi/day of either ^{238}U or ^{230}Th over a period of from 100 to 15,000 working days was assumed for the reference man. The calculations were performed for ore dust aerosols with particle sizes of 2 μm AMAD and 10 μm AMAD. The results of these calculations are given in Table 6 (Urinary Excretion Rates) and Table 7 (Fecal Excretion Rates). The total cumulative body burdens of ^{238}U or ^{230}Th are shown in Figure 10 for the two different particle sizes.

The combined metabolic model predicts that the ^{230}Th body burden will be an order of magnitude or more greater than the body burden of ^{234}U or ^{238}U following chronic inhalation of uranium ore dust. Equilibrium levels of uranium and thorium are not attained in the body within 15,000 days (approximately 60 work-years) of chronic exposure to ore dust; rather, the body burdens continue to increase with length of time spent on the job (Figure 10).

Tables 6 and 7 show that the fecal elimination rates of ^{238}U and ^{230}Th from the body remain fairly constant with increasing length of exposure to constant levels of ore dust, whereas the urinary excretion rates increase with length of exposure. Thus, the ratio of the amount of uranium or thorium excreted per day in the feces to the amount in the urine decreases with length of occupational exposure time (Figure 11). A check of the ratios of daily fecal excretions of uranium (day 1, Table 2) to daily urinary excretions (day 1, Table 1) by participants in the study showed good agreement between the actual measurements and the ratios predicted by the combined metabolic model. The actual feces/urine ratios for ^{230}Th were generally higher, suggesting that the retention of ^{230}Th could be greater than that predicted by the model.

The length of occupational exposure to uranium ore dust was obtained for each worker in the study. The calculated excretion rates for the reference man were then compared to the actual measured radionuclide levels in the worker's daily excreta. The ratio of the activity measured in the worker's excreta to the amount predicted for the reference man was then multiplied by the predicted body burden in the reference man to produce an estimate of the actual body burden in the individual worker. The derived body burden results for individual

TABLE 6. Urinary Excretion Rates of ^{238}U and ^{230}Th Following Chronic Inhalation of 1 pCi/Day of Either Nuclide in Ore Dust. Two aerosol particles sizes (2 μm and 10 μm AMAD) are considered.

Length of Occupational Exposure (days)	Urinary Excretion Rate (pCi/day)			
	^{238}U		^{230}Th	
	2 μm	10 μm	2 μm	10 μm
100	0.0067	0.0093	0.0008	0.0012
500	0.0119	0.0109	0.0015	0.0020
1,000	0.0180	0.0126	0.0023	0.0028
5,000	0.0370	0.0179	0.0068	0.0061
10,000	0.0387	0.0184	0.0098	0.0082
15,000	0.0389	0.0185	0.0115	0.0094

TABLE 7. Fecal Excretion Rates of ^{238}U and ^{230}Th Following Chronic Inhalation of 1 pCi/Day of Either Nuclide in Ore Dust. Two aerosol particles sizes (2 μm and 10 μm AMAD) are considered.

Length of Occupational Exposure (days)	Fecal Excretion Rate (pCi/day)			
	^{238}U		^{230}Th	
	2 μm	10 μm	2 μm	10 μm
100	0.644	0.941	0.645	0.943
500	0.671	0.949	0.672	0.951
1,000	0.689	0.954	0.690	0.956
5,000	0.707	0.959	0.708	0.961
10,000	0.707	0.959	0.708	0.961
15,000	0.707	0.959	0.708	0.961

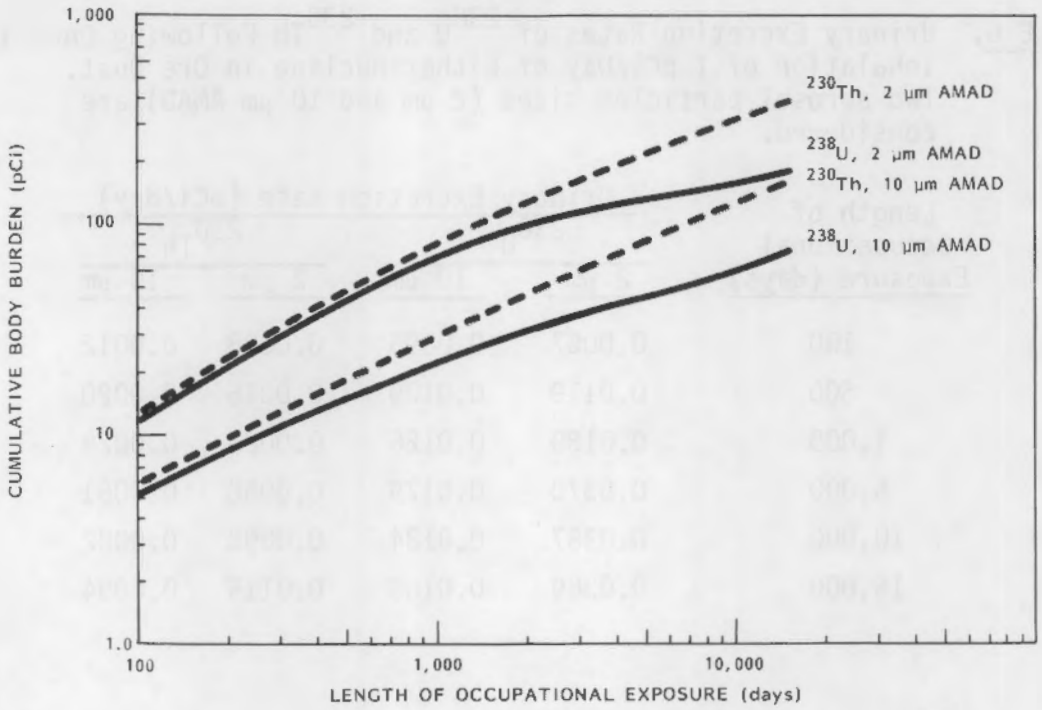

FIGURE 10. Total Cumulative Body Burden (pCi) as Thorium ^{230}Th and Uranium as ^{234}U and ^{238}U Following Chronic Occupational Inhalation of 1 pCi/day of the Nuclides Present in 2 µm and 10 µm AMAD Uranium Ore Dust Aerosols

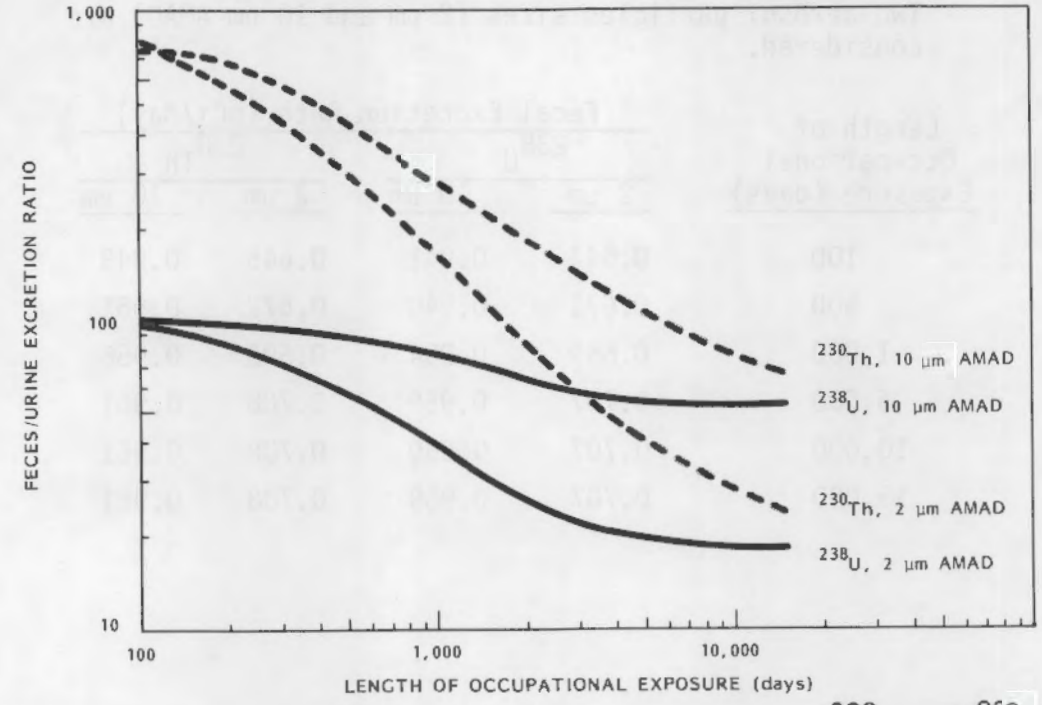

FIGURE 11. Ratio of Fecal to Urinary Excretion of ^{238}U and ^{230}Th for the Reference Worker. Ratios were derived for aerosols of 2 µm and 10 µm AMAD

workers who participated in the study are given in Table 8. The estimated ^{238}U body burdens in uranium mill crushermen ranged from approximately 0.1 nCi to 33 nCi, and the estimated ^{230}Th burdens ranged from 0.1 nCi to 29 nCi.

Inhalation Exposure Calculations

Estimates of the average inhalation exposure that could have resulted in the actual excretion rates measured among crushermen were derived by comparing the actual results with the predictions of the combined metabolic model. The duration of exposure was adjusted in each case to agree with the exposure history of the worker. The average inhalation rates so derived for each active crusherman are given in Table 9.

The derived average inhalation rates also provide a convenient check on the methods used to obtain them. The rates for inhalation of ^{238}U can be compared to inhalation rates for ^{230}Th to see if the values from the table are near equilibrium, since an assumption was made that ^{238}U and ^{230}Th were in approximate secular equilibrium in the ore dust inhaled by the workers. It can be seen from Table 9 that the derived inhalation rates of ^{238}U are generally within a factor of two of the rates derived for ^{230}Th. The most notable exceptions are subjects W-2, W-3, W-10, and W-13. Considering the possible sources of error (sample collection and potential contamination, radiochemical separation and alpha spectrometry, assumptions regarding the separate metabolism of uranium and thorium in the worker's body, and the assumed occupational exposure times), agreement within a factor of two appears to be respectable.

Derived Annual Intakes of ^{238}U and ^{230}Th for Study Participants

The ICRP recently recommended annual limits for intakes (ALIs) of many different radionuclides by workers (ICRP 1979). The recommended limits for class Y ^{238}U and ^{230}Th are 2×10^3 Bq (54 nCi) and 6×10^2 Bq (16 nCi), respectively. The recommended limits apply to aerosols with a 1 μm AMAD particle size. These limits are conservative for cases in which the particle size AMAD is greater than 1 μm, and a correction may be calculated. However, if the AMAD is unknown, it is recommended by ICRP that the limits based on an AMAD of 1 μm be used (ICRP 1979).

TABLE 8. Estimated ^{238}U and ^{230}Th Body Burdens in Uranium Mill Crushermen (Study Participants) Based on the Combined Metabolic Model and Measured Levels in Day 1 Excreta. Two aerosol particle sizes (2 μm and 10 μm AMAD) were considered.

Subject	Length of Occupational Exposure (Days)	Body Burden (nCi)			
		^{238}U		^{230}Th	
		2 μm	10 μm	2 μm	10 μm
W-1	625	33	7.5	19	5.1
W-2	1,500	110[a]	24.6[a]	4.6	1.2
W-3	1,500	8.1	1.8	2.6	0.7
W-4	750	29	6.5	19	4.9
W-5	3,000	0.1	0.04	0.1	0.03
W-6	750	4.9	1.1	4.0	1.0
W-7	500	34	7.6	29	7.8
W-8	5,250	(b)	(b)	(b)	(b)
W-9	1,500	8.6	1.9	5.1	1.4
W-10	500	2.3	0.5	0.2	0.06
W-11	625	15	3.3	7.1	1.9
W-12	1,125	4.8	1.1	2.0	0.5
W-13	1,000	6.8	1.5	0.9	0.2
W-14	1,000	3.8	0.8	2.6	0.7

(a) Fecal sample probably spiked with a small amount of yellowcake dust. Yellowcake is normally depleted in thorium. The uranium/thorium ratio measured for this worker's fecal sample on day 1 (see Table 2) suggests that yellowcake dust was added, either accidentally or intentionally. The worker was resampled at a later date, and a normal uranium/thorium ratio was measured in the fecal sample.

(b) Total daily excretion not indicated on sample.

TABLE 9. Derived Average Inhalation Rates of ^{238}U and ^{230}Th for Active Uranium Mill Crushermen, Based on Measured Excretion Rates and the Combined Metabolic Model for Two Aerosol Sizes

	Derived Inhalation Rates (pCi/day)			
	^{238}U		^{230}Th	
Subject	2 μm	10 μm	2 μm	10 μm
W-1	620	442	343	244
W-2	1245[a]	909[a]	45	32
W-3	92	67	25	18
W-4	499	357	293	210
W-5	1.4	1.0	0.8	0.6
W-6	83	59	62	45
W-7	757	535	615	434
W-8	(b)	(b)	(b)	(b)
W-9	97	71	49	36
W-10	52	37	5.0	3.5
W-11	277	198	129	92
W-12	64	46	24	17
W-13	96	69	11	8.0
W-14	53	39	33	24

(a) Fecal sample probably spiked with a small amount of yellowcake dust. Yellowcake is normally depleted in thorium. The uranium/thorium ratio measured for this worker's fecal sample on day 1 (see Table 2) suggests that yellowcake dust was added, either accidentally or intentionally. The worker was resampled at a later date, and a normal uranium/thorium ratio was measured in the fecal sample.

(b) Total daily excretion not indicated on sample.

The average annual intakes of uranium and thorium for workers participating in the present study were derived by multiplying the values given in Table 9 by 250 working days per working year and are shown in Table 10. These intakes may be compared with ICRP recommended ALIs. The derived average annual intakes for four of the workers (W-1, W-4, W-7, and W-11) exceed ICRP recommended limits. However, this analysis is based on limited excreta sampling and a number of uncertain assumptions implicit in the biological modeling. Air sampling and time-weighted exposure studies would be necessary to determine whether excessive levels of natural uranium ore dust were being inhaled by any worker.

APPLICABILITY OF CURRENT MODELS FOR THORIUM METABOLISM

A note of caution is added to the present discussion concerning derived body burdens, inhalation rates, and annual intakes for thorium based on ICRP clearance half-times and fractions. Several of the findings from this study indicate that the biological parameters assumed for thorium metabolism (ICRP 1979) may overestimate the true excretion rates and underestimate the true body retention:

1. The ^{230}Th feces/urine ratios for excreta of crushermen were generally higher than the ratios predicted by the metabolic model.
2. The model predicts that the fecal excretions of ^{238}U will closely resemble the fecal excretions of ^{230}Th (see Table 7). However, the fecal measurements showed that the average daily excretion of ^{230}Th was roughly a factor of three less than the excretion of ^{238}U (see Figure 8).
3. Similarly, the urinary excretions of ^{230}Th observed for crushermen (Figure 7) were much lower relative to ^{238}U than those predicted by the model.

These findings suggest that the ICRP biological parameters for thorium may not be appropriate, and that further study of thorium metabolism and excretion patterns is needed.

TABLE 10. Derived Annual Intakes of ^{238}U and ^{230}Th for Active Uranium Mill Crushermen, Based on Measured Excretion Rates and the Combined Model for Two Aerosol Sizes

	Derived Annual Intake (nCi/yr)			
	^{238}U		^{230}Th	
Subject	2 μm	10 μm	2 μm	10 μm
W-1	155	111	86	61
W-2	311[a]	227[a]	11	8.1
W-3	23	17	6.2	4.5
W-4	125	89	73	52
W-5	0.36	0.26	0.20	0.15
W-6	21	15	15	11
W-7	189	134	153	108
W-8	(b)	(b)	(b)	(b)
W-9	24	18	12	9.0
W-10	13	9.1	1.2	0.9
W-11	70	49	32	23
W-12	16	12	6.0	4.3
W-13	24	17	2.8	2.0
W-14	13	10	8.2	6.0

(a) Fecal sample probably spiked with a small amount of yellowcake dust. Yellowcake is normally depleted in thorium. The uranium/thorium ratio measured for this worker's fecal sample on day 1 (see Table 2) suggests that yellowcake dust was added, either accidentally or intentionally. The worker was resampled at a later date, and a normal uranium/thorium ratio was measured in the fecal sample.

(b) Total daily excretion not indicated on sample.

PROBLEMS ASSOCIATED WITH FECAL BIOASSAY

This work shows that fecal excretion is the predominant exit pathway for clearance of radionuclides in uranium ore dust from the bodies of uranium mill crushermen. Fecal excretion accounted for approximately 95-99% of the total excretion. To study the behavior of uranium and thorium inhaled by crushermen, it was therefore important to collect fecal specimens as well as urine specimens. There are, however, special problems associated with the collection and processing of fecal material from workers.

Psychological Aversion

As we attempted to recruit volunteers for the study, we encountered a great deal of reluctance on the part of workers to provide both urine and stool specimens. Workers associate a stigma with the process of collecting and surrendering excreta specimens. The psychological aversion to fecal bioassay was reinforced by the repugnant sight and odor generated by samples. Workers voiced their strong dislike for any bioassay program involving collection of their own waste. Our request for voluntary participation in the study was met by embarrassment and disgust by the workers. Since the sample collection program relied totally on voluntary participation, an incentive cash honorarium was instituted to attract volunteers. Even so, the enlistment of subjects turned out to be a greater challenge than originally anticipated.

We carefully explained to the workers that the sample collection kit was simple, convenient to use, and effective for containing the specimen and its odors. The scientific value of the samples and the importance of the study were also explained to the workers before they were given an opportunity to volunteer as subjects. We expected that many of the crushermen would volunteer out of curiosity, or for medical reasons to see if a determination could be made that might be beneficial to their health. However, very few of the active crushermen reflected this attitude or interest. Those who did agree to participate were probably motivated to do so primarily by the cash honorarium that was promised.

We observed a correlation between the level of education of the contact, and willingness to participate in the study. In general, the more educated

workers and controls were more inclined to participate "for the advancement of science," whereas the lesser educated workers and controls either refused, or participated solely for the cash honorarium.

Retired mill workers were usually eager to participate. Retirees were more concerned about their own health and the possible health effects of uranium ore dust inhalation than were the active millworkers. Only one retiree (out of five interviewed) declined to participate, giving his current ill health as the reason.

The workers were approached both individually and in small groups. Individual encounters were more successful because the group tended to respond in a block; if one worker objected, then the others were likely to follow suit.

Most uranium millworkers understand that regulations require their employer to conduct a urinalysis program among certain job classifications and that participation may be a condition of their employment. However, none of the workers had ever been asked for a bowel specimen prior to this time. As a result, some workers suspected that we represented their company and were informants, or that we were not providing all the reasons for the special collection program. A few workers expressed concern that the analytical results might reveal levels of radioactivity that could indicate poor work habits (such as not using a respirator, or general carelessness), which might cause them to lose their jobs.

Other reasons were given by those who refused to participate in the sampling program. One worker mentioned concern about his irregularity. Another worker was concerned about having to take the sample kit to his home where small children could tamper with it. Several were embarrassed to bring their kits back to work after the four-day-off period, and did so reluctantly. Several workers refused to return the completed kit to their workplace, and were permitted to ship the container directly back to our laboratory.

Three or four workers participated one day and then refused to continue further because they disliked having to collect a bowel movement and experience the strong odors while bagging the material. Since participation was voluntary, those workers had little to lose but the honorarium.

Difficulties Following Instructions

One contact person at each mill was extremely helpful in coordinating the distribution and return of kits on our behalf. Most volunteers followed the sample collection instructions, although a few samples were mislabeled or packaged incorrectly for shipment back to our laboratory. At least two of the volunteers could not read and required other persons to read and re-read the sample collection instructions to them. Others may have declined to participate for the same reason.

Certain workers were not recommended by their supervisors for participation in the study either because of a previous history of not following instructions for collection of urine samples, or for reasons of gross uncleanliness.

Lack of Experimental Control

Since the sample collection kits were intended for use at home, we lost some degree of experimental control over the quality of the sample collection procedure. We trusted that the instructions were sufficiently clear and that the volunteers were sufficiently dependable to provide us with reliable samples. However, this may not always have been the case. It is difficult to guarantee that the sample was the volunteer's, or that the sample was actually obtained on the designated day.

Sample Transport Problems

Excreta samples were not sterilized prior to shipment back to the laboratory. However, sufficient packing material was included with the sample kit to protect the shipment from rough handling in transit. If a container had broken open enroute to the laboratory, serious repercussions from the carrier might have followed. Fortunately no such difficulties were encountered in the study.

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APPENDIX

CONCENTRATION OF NATURAL URANIUM IN EXCRETA SAMPLES

TABLE A.1. Concentration of Natural Uranium Measured in Urine Samples of Study Participants

Subject	Day	Natural Uranium (μg/ℓ)	Natural Uranium (pCi/ℓ)
W-1	1	6.27	4.37
	3	2.70	1.87
	4	4.07	2.82
W-2	1	2.19	1.52
	3	0.539	0.374
	4	0.964	0.669
W-3	1	1.91	1.33
	3	0.955	0.663
	4	0.752	0.522
W-4	1	11.19	7.76
	3	9.94	6.89
	4	7.62	5.28
W-5	1	0.193	0.184
	3	0.331	0.229
	4	0.320	0.222
W-6	1	11.10	7.70
	3	9.51	6.60
	4	8.24	5.72
W-7	1	12.38	8.59
	3	16.47	11.42
	4	4.41	3.06
W-8	1	3.31	2.29
	3	1.55	1.07
	4	1.22	0.85
W-9	1	0.783	0.543
	3	1.16	0.80
	4	1.11	0.77
W-10	1	1.32	0.92
	3	2.36	1.64
	4	1.30	0.90
W-11	1	3.88	2.70
	3	3.72	2.58
	4	2.56	1.77

TABLE A.1. (Continued)

Subject	Day	Natural Uranium (µg/ℓ)	Natural Uranium (pCi/ℓ)
W-12	1	4.62	3.21
	3	9.26	6.43
	4	2.05	1.42
W-13	1	5.69	3.50
	3	4.14	2.89
	4	0.343	0.238
W-14	1	1.95	1.35
	3	1.81	1.26
	4	0.752	0.522
R-1	1	0.084	0.058
	2	0.107	0.074
R-2	1	0.079	0.055
	2	0.075	0.052
R-3	1	0.054	0.038
	2	0.135	0.094
R-4	1	0.293	0.203
	2	0.268	0.186
C-1	1	0.069	0.203
	2	0.268	0.186
C-2	1	0.069	0.048
	2	0.108	0.075
C-3	1	0.120	0.083
	2	0.050	0.035

TABLE A.2. Daily Fecal Excretion of Natural Uranium by Study Participants

Subject	Day	Natural Uranium (μg/day)	Natural Uranium (pCi/day)
W-1	1	1210	839
	3	220	153
	4	70	49
W-2	1	2780	1930
	3	57	39
	4	50	35
W-3	1	198	137
	3	188	130
	4	25	17
W-4	1	1030	715
	3	438	304
	4	72	50
W-5	1	3.1	2.1
	3	14	9.5
	4	15	10
W-6	1	166	115
	3	21	15
	4	383	266
W-7	1	1540	1070
	3	466	323
	4	28	19
W-8	1	(a)	(a)
	3	(a)	(a)
	4	(a)	(a)
W-9	1	214	148
	3	60	42
	4	29	20

(a) Total daily excretion not indicated on sample.

TABLE A.2. (Continued)

Subject	Day	Natural Uranium (µg/day)	Natural Uranium (pCi/day)
W-10	1	108	75
	3	(b)	(b)
	4	14	9.7
W-11	1	575	399
	3	16	11
	4	25	18
W-12	1	135	94
	3	75	52
	4	74	51
W-13	1	206	143
	3	36	25
	4	35	24
W-14	1	114	79
	3	55	38
	4	5.0	3.5
R-1	1	22	15
	2	28	19
R-2	1	8.2	5.7
	2	3.1	2.1
R-3	1	(a)	(a)
	2	(a)	(a)
R-4	1	106	73
	2	68	47
C-1	1	21	15
	2	(a)	(a)
C-2	1	7.5	5.2
	2	17	12
C-3	1	15	10
	2	15	10

(a) Total daily excretion not indicated on sample.
(b) No sample provided, empty container marked "none."

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Pacific Northwest Laboratory conducted a research program to measure uranium and thorium levels in excreta of uranium mill crushermen who are routinely exposed to airborne uranium ore dust. The purpose of this work was to determine whether ^{230}Th was preferentially retained over either ^{234}U or ^{238}U in the body.

Urine and fecal samples were obtained from fourteen active crushermen with long histories of exposure to uranium ore dust, plus four retired crushmen and three control individuals for comparison. Radiochemical procedures were used to separate out the uranium and thorium fractions, which were then electroplated on stainless steel discs and assayed by alpha spectrometry.

Significantly greater activity levels of ^{234}U and ^{238}U were measured in both urine and fecal samples obtained from uranium mill crushermen, indicating that uranium in the inhaled ore dust was cleared from the body with a shorter biological half-time than the daughter product ^{230}Th. The measurements also indicated that uranium and thorium separate in vivo and have distinctly different metabolic pathways and transfer rates in the body. The appropriateness of current ICRP retention and clearance parameters for ^{230}Th in ore dust is questioned.

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