Radiation Protection Dosimetry (2017), Vol. 175, No. 4, pp. 503–507 doi:10.1093/rpd/ncw379
Advance Access publication 17 January 2017

EXPOSURE TO RECYCLED URANIUM CONTAMINANTS IN GASEOUS DIFFUSION PLANTS

Jeri L. Anderson[1,*], A. Iulian Apostoaei[2], James H. Yiin[1] and Chih-Yu Tseng[1]
[1]Division of Surveillance, Hazard Evaluations and Field Studies (DSHEFS), National Institute for Occupational Safety and Health (NIOSH), Cincinnati, OH 45226, USA
[2]Oak Ridge Center for Risk Analysis, Inc., Oak Ridge, TN 37830, USA

*Corresponding author: JLAnderson@cdc.gov

Received 3 October 2016; revised 1 December 2016; accepted 8 December 2016

As part of an ongoing study of health effects in a pooled cohort of gaseous diffusion plant workers, organ dose from internal exposure to uranium was evaluated. Due to the introduction of recycled uranium into the plants, there was also potential for exposure to radiologically significant levels of ^{99}Tc, ^{237}Np and ^{238,239}Pu. In the evaluation of dose response, these radionuclide exposures could confound the effect of internal uranium. Using urine bioassay data for study subjects reported in facility records, intakes and absorbed dose to bone surface, red bone marrow and kidneys were estimated as these organs were associated with *a priori* outcomes of interest. Additionally, ^{99}Tc intakes and doses were calculated using a new systemic model for technetium and compared to intakes and doses calculated using the current model recommended by the International Commission on Radiological Protection. Organ absorbed doses for the transuranics were significant compared to uranium doses; however, ^{99}Tc doses calculated using the new systemic model were significant as well. Use of the new model resulted in an increase in ^{99}Tc-related absorbed organ dose of a factor of 8 (red bone marrow) to 30 (bone surface).

INTRODUCTION

The National Institute for Occupational Safety and Health (NIOSH) is currently studying a pooled cohort of workers (n = 29 303) at three US gaseous diffusion plants to evaluate the relationship between internal exposure to uranium and cause-specific mortality and cancer incidence. Because of the tendency of uranium to be taken up by bone marrow and kidneys, disease mortality and incidence of *a priori* interest were bone, hematopoietic and kidney cancer as well as non-malignant renal disease. As part of this study, radiation absorbed dose from internal exposure to uranium was estimated, as well as potential confounding exposures such as exposure to external ionizing radiation and non-radiological exposures. Another possible confounding exposure was to fission products and transuranic contaminants in recycled uranium from other nuclear weapons production facilities that was used as feed for the gaseous diffusion plants. This recycled uranium was the result of the industrial process of recovering uranium from irradiated reactor fuel by chemical separation in an effort to expand domestic sources of uranium for the US during the period of March 1952 to March 1999[(1)].

In addition to uranium, the main nuclides of radiological concern in facilities handling recycled uranium are plutonium (^{238}Pu and ^{239}Pu), neptunium (^{237}Np) and technetium (^{99}Tc). ^{238}Pu, ^{239}Pu and ^{237}Np are long-lived alpha-emitters with alpha energies of 5.2–5.5, 5.0–5.2 and 4.6–4.9 MeV, respectively. ^{99}Tc is primarily a beta-emitter with a half-life of 211 100 years and maximum beta energies of 0.204–0.294 MeV (average energy = 0.0565–0.0846 MeV).

The biokinetic models for plutonium, neptunium, and technetium are described by the International Commission on Radiological Protection (ICRP) in Publication 67[(2)]. About 50% of plutonium and 45% of neptunium is deposited in the skeleton after leaving the blood compartment, 1 and 1.5%, respectively, is deposited in the urinary path in the kidneys, and another 0.5% for both is deposited in kidney tissue where it is retained for a longer period of time. In the ICRP 67 model, technetium is primarily concentrated in the thyroid, gastrointestinal tract and liver. A new biokinetic model proposed by Leggett and Giussani[(3)] provides a more realistic description of the systemic behavior of technetium, showing uptake in the kidneys and bone in addition to other tissues.

The purpose of this work is to estimate intakes and organ absorbed doses from exposure to radioactive contaminants in recycled uranium for study subjects in the NIOSH pooled gaseous diffusion plant study. These data will be used in dose–response evaluation as part of the ongoing epidemiological analyses of cause-specific mortality and cancer incidence in the cohort.

METHODS

Individual bioassay data consisting of activity concentrations of ^{238}Pu, ^{239}Pu, ^{237}Np and ^{99}Tc or gross beta were abstracted from electronic databases obtained

Published by Oxford University Press 2017. This work is written by (a) US Government employee(s) and is in the public domain in the US.

Downloaded from https://academic.oup.com/rpd/article-abstract/175/4/503/2910616/Exposure-to-Recycled-Uranium-Contaminants-in
by NCIPC user
on 13 September 2017

from the Oak Ridge Gaseous Diffusion Plant (also known as K-25), the Portsmouth Gaseous Diffusion Plant, and the Paducah Gaseous Diffusion Plant by NIOSH as part of previous studies. Bioassay data sets were selected for study subjects who had at least one reported positive (i.e. greater than zero) sample. Additionally, historical plant records were examined to ascertain bioassay program procedures, analytical methods, and detection levels for the radionuclides of interest.

Urine samples determined to be collected over a period of <24-h (e.g. spot samples) and recorded as activity concentration were normalized to 24-h excretion by multiplying by the ICRP Reference Man urine excretion volume of 1.6 L d^{-1}. Urine data for ^{99}Tc activity were generally reported as gross beta activity especially in samples that were analyzed prior to the 1990s. Urinalysis data for each of the facilities were examined to estimate a factor that would be used to reduce the gross beta activity to more closely approximate the ^{99}Tc concentration in the urine.

For study subjects with no bioassay records, urine radionuclide concentration data were imputed using methods described in Anderson *et al.*[4]. Briefly, levels of ^{99}Tc, ^{238}Pu, ^{239}Pu and ^{237}Np radioactivity in urine associated with specific departments were averaged to determine a department-specific average activity concentration for each of the radionuclides. Then, for each study subject's work history records, the average concentration for the radionuclide for the corresponding department number was assigned as a bioassay sample using the end date of that work history record as the assumed collection date of the imputed urine sample. This method is based on the assumption that a steady state of urine excretion was reached at the end of the study subject's work history record. For study subjects with reported bioassay data, intakes and organ doses calculated using imputation methods were compared to intakes and doses calculated using reported bioassay data.

Many of the urine samples had activity levels reported as zero or as a facility administrative or detection level. Therefore, it was necessary to estimate a substitution value for the left-censored bioassay data because the bioassay data were assumed to have lognormally distributed measurement error for the purpose of intake calculations. This substitution value was assumed to be proportional to the facility detection limit or administrative limit and the fraction of bioassay data censored in the individual's bioassay data set for ^{99}Tc[5]. Because of the infrequent monitoring for transuranics in the cohort, the substitution value was assumed to be proportional to the facility detection and the fraction of data censored in the combined data set for a department. The relationship between the substitution value (S) and the facility detection/administrative limit (L) and the fraction of the individual's bioassay data that is censored (f) is described by the following equation:

$$S = L(1-f)$$

Individual intakes and absorbed dose to the bone surface, red marrow and kidney were calculated using the Internal Dose Evaluation Program (InDEP; Oak Ridge Center for Risk Analysis, Inc. Oak Ridge, TN, v. 4.2, 2016), using the ICRP Human Respiratory Tract Model of ICRP Publication 66 and assuming a chronic intake of an aerosol with a default 5-μm activity median aerodynamic diameter (AMAD) particle size[6]. For ^{238}Pu, ^{239}Pu and ^{237}Np, the absorption from the lung was assumed to be the ICRP default Type M (moderate absorption or moderately insoluble)[7] whereas ^{99}Tc was assumed to be in the form of pertechnetate (TcO_4^-) and was assigned Type F (fast absorption or soluble). For technetium, intakes and organ doses were calculated using both the current ICRP biokinetic model and the new model proposed by Leggett and Giussani[3]. The period of chronic intake was assumed to start on one March 1952 (the approximate date of the introduction of recycled uranium into the fuel cycle) or the individual's date of hire, whichever is later, and end on the date of employment termination.

RESULTS AND DISCUSSION

Workers were monitored for exposure to these various radionuclides, however, the individual bioassay data records collected as part of previous NIOSH studies indicate that *in vitro* bioassay monitoring was sporadic compared to that done for uranium at each of the facilities. Facility records indicate that analyses for transuranics consisted of isotopic analysis using ion-exchange separation techniques and alpha spectroscopy and ^{99}Tc analyses was performed by gross beta proportional counting and then liquid scintillation counting in later years (1990s). It is unclear from the plant records whether any separation chemistry was used for ^{99}Tc in bioassay samples.

For K-25, both gross beta and ^{99}Tc results were reported for 2068 urine samples analyzed from 1989 to 1993, however, no sample-specific information was found regarding procedures used. Additionally, for one year during 1993, it appears that an intermediate analysis was performed on 192 urine samples resulting in three separate activities being reported, although all values were labeled as either 'beta' or '^{99}Tc.' Using the lowest value as the assumed ^{99}Tc activity, it was estimated that the gross beta activity (or the initial reported value) was an average factor of 3.2 (fifth–95th percentile: 1.1–4.5) greater than the ^{99}Tc activity (or the final reported value). For Paducah, according to records showing

Downloaded from https://academic.oup.com/rpd/article-abstract/175/4/503/2910616/Exposure-to-Recycled-Uranium-Contaminants-in
by NCIPC user
on 13 September 2017

that the analysis changed from gross beta (proportional counter) to liquid scintillation counting, combined measurements from both time periods showed a reduction by a factor of 8.9 (fifth–95th percentile: 2.8–23) in reported ^{99}Tc urinalysis results.

Table 1 shows a summary of the data available in the individual bioassay records for the three facilities. Approximately 10% of the combined cohort had reported bioassay data for ^{99}Tc and <0.1% had reported data for ^{238,239}Pu and ^{237}Np. Department numbers were found not to be associated with ^{237}Np measurements, so it was not possible to impute bioassay data for that radionuclide. At Portsmouth, routine *in vitro* bioassay monitoring was not done for transuranics; however, over 20 000 urine samples were analyzed for gross beta to estimate exposure to ^{99}Tc. Paducah and, to a lesser extent, K-25 also had relatively consistent monitoring for ^{99}Tc. Urinalysis data were imputed for an additional 34 and 9% of the cohort for ^{99}Tc and ^{239}Pu, respectively.

The distributions of intakes and cumulative organ absorbed doses (Tables 2 and 3, respectively) calculated from reported and imputed bioassay data were highly skewed and generally approximated a log-normal distribution. Organ doses from ^{99}Tc and ^{239}Pu exposures were highest at K-25 and lowest at the Paducah facility. For all three facilities, median organ doses from exposure to ^{99}Tc were lower and exposure to transuranics were higher than those from exposure to uranium. For example, median bone surface doses from uranium exposure varied from 0.68 to 1.8 mGy for the three facilities[4] compared to 0.14 to 0.55 mGy from exposure to ^{99}Tc and 9.6 to 74 mGy from exposure to plutonium. The lower technetium dose is due to more rapid biological clearance and lower energy beta radiation of ^{99}Tc compared to uranium, whereas the higher plutonium doses are the result of the bone-seeking tendency of plutonium and more energetic alpha radiation. Median doses to the red marrow and kidney from ^{99}Tc were similar to uranium and slightly higher for plutonium.

Use of the new systemic model for technetium resulted in significantly higher calculated intakes and doses for the organs of interest than those calculated using the current ICRP model. Intakes were increased by a factor of 1.5 and absorbed organ doses increased by a factor of 30, 8 and 15 for bone surfaces, red bone marrow, and kidneys, respectively. These values compare well with the ratio of predicted cumulative activities based on the proposed model and the ICRP reported by Leggett and Giussani[3] of 25 for bone and 11 for kidneys.

Table 4 shows the ratio of intakes and organ doses from contaminate radionuclide exposure to intakes and organ doses from uranium exposure. Calculated intakes of contaminate radionuclides for study subjects were, in general, poorly correlated with their calculated uranium intakes (Pearson correlation coefficients: −0.12 and 0.27 for ^{239}Pu at K-25 and Paducah, respectively, and 0.045, 0.33 and 0.93 for ^{99}Tc at Paducah, K-25 and Portsmouth, respectively). However, the extent of correlation appears to be dependent on the facility enrichment level estimated from uranium specific activity in urine samples. The urine ^{99}Tc becomes more correlated with urine uranium activity with increasing facility enrichment levels and ^{239}Pu becomes more correlated with uranium with decreasing facility enrichment levels. The Paducah facility was originally designed to produce feed (UF_6) for both the K-25 and Portsmouth facilities and produced uranium enriched to about 0.96%[8]. In the fluorination process of recycled uranium, transuranics tended to form non-volatile compounds resulting in concentration in the ash from the fluorination tower or to be trapped at the feed points of the gaseous diffusion plant cascade, both areas where there was a potential for worker exposure[9]. Alternatively, technetium is

Table 1. Average activity in urine samples analyzed for exposure assessment of the combined cohort. Activity measurements were converted to 24-h excretion.

Facility (total *n* of study subjects)	Number of study subjects with urine sample data	Average number of urine samples per study subject	Activity concentration per study subject[a] (Mean ± SD) (Bq d^{-1})
K-25 (n = 16 978)			
^{239}Pu	16	5	0.011 ± 0.020
^{99}Tc	1613	6	41 ± 190
Paducah (n = 5390)			
^{238}Pu	12	1	0.0036 ± 0.0083
^{239}Pu	11	2	0.0045 ± 0.0089
^{237}Np	11	4	0.00024 ± 0.000080
^{99}Tc	614	26	9.3 ± 8.8
Portsmouth (n = 6935)			
^{99}Tc	401	51	23 ± 52

[a]Data distributions are skewed.

Downloaded from https://academic.oup.com/rpd/article-abstract/175/4/503/2910616/Exposure-to-Recycled-Uranium-Contaminants-in
by NCIPC user
on 13 September 2017

Table 2. Calculated intakes (median [95th percentile] among all study subjects) from reported and reported + imputed bioassay data for each contaminant radionuclide. No bioassay data were imputed for ^{237}Np.

	Count/% imputed	Reported data (Bq d^{-1})	Reported + imputed data (Bq d^{-1})
^{99}Tc[a]			
K25	11166/86	140 [300 000]	160 [340]
Paducah	654/6.1	19 [110]	20 [150]
Portsmouth	672/40	15 [280]	40 [490]
^{99}Tc—new model[b]			
K25	11166/86	280 [2 300 000]	240 [4400]
Paducah	654/6.1	30 [1300]	33 [750]
Portsmouth	672/40	23 [510]	60 [730]
^{239}Pu			
K25	2721/99	0.56 [2.0]	0.52 [8.5]
Paducah	19/42	0.039 [2.8]	0.24 [2.5]
^{238}Pu			
Paducah	13/7.7	0.034 [2.4]	0.034 [2.4]
^{237}Np			
Paducah	11/NA	0.012 [0.034]	0.012 [0.034]

[a]Technetium systemic model from ICRP Publication 67.
[b]New technetium systemic model by Leggett and Giussani.

Table 3. Cumulative organ absorbed doses (median [95th percentile]) for each contaminant radionuclide by study facility. No organ doses were imputed for ^{237}Np.

	Bone surface (mGy)	Red marrow (mGy)	Kidney (mGy)
^{99}Tc[a]			
K-25	0.018 [0.076]	0.018 [0.076]	0.018 [0.076]
Paducah	0.0048 [0.042]	0.0048 [0.042]	0.0048 [0.042]
Portsmouth	0.0068 [0.091]	0.0068 [0.091]	0.0068 [0.091]
^{99}Tc—new model[b]			
K25	0.55 [20]	0.15 [5.7]	0.28 [11]
Paducah	0.14 [2.4]	0.041 [0.66]	0.078 [1.3]
Portsmouth	0.21 [2.9]	0.055 [0.74]	0.11 [1.4]
^{239}Pu			
K-25	73 [910]	4.0 [46]	0.35 [4.0]
Paducah	14 [380]	0.79 [24]	0.070 [2.2]
^{238}Pu			
Paducah	9.6 [458]	0.57 [28]	0.051 [2.6]
^{237}Np			
Paducah	0.68 [3.9]	0.038 [0.19]	0.0038 [0.018]

[a]Technetium systemic model from ICRP Publication 67.
[b]New technetium systemic model by Leggett and Giussani accounts for observed preferential uptake of technetium in bone and kidneys.

more easily volatilized during the fluorination process and more likely to be introduced into the cascade. Also, because of its lower molecular weight, technetium would travel further through the cascade and end up in the enriched product. Additionally, the majority of the recycled uranium processed by Portsmouth came from the feed manufacturing facilities at both the Paducah and K25 facilities[10].

There are several sources of uncertainty in the intake and dose calculations including those resulting from various assumptions made on the physical and chemical form of the compounds to which the study subjects were exposed. Recommended default parameters were assumed for particle size and solubility (absorption type), however, significant uncertainty is introduced in the conversion of activity in spot samples (i.e. concentration) to 24-h excretion. Another large source of uncertainty in the technetium intake and dose calculations is due to the lack of information on the chemical preparation used for the technetium bioassay samples and any potential interference from other radionuclides in the urine samples.

Downloaded from https://academic.oup.com/rpd/article-abstract/175/4/503/2910616/Exposure-to-Recycled-Uranium-Contaminants-in
by NCIPC user
on 13 September 2017

Table 4. Ratio of contaminant radionuclide to uranium for intakes (activity) and organ absorbed doses calculated using reported bioassay data.

	Intakes	Bone surface	Red marrow	Kidney
^{99}Tc[a]				
K-25	160	0.011	0.092	0.026
Paducah	34	0.0025	0.020	0.0060
Portsmouth	85	0.0061	0.052	0.014
^{99}Tc—new model[b]				
K25	330	0.45	1.0	0.55
Paducah	56	0.077	0.17	0.10
Portsmouth	130	0.18	0.42	0.22
^{239}Pu				
K-25	0.43	30	13	0.35
Paducah	0.062	4.0	1.8	0.048
^{238}Pu				
Paducah	0.070	4.2	2.0	0.052
^{237}Np				
Paducah	0.015	0.76	0.31	0.0086

[a]Technetium systemic model from ICRP Publication 67.
[b]New technetium systemic model by Leggett and Giussani.

CONCLUSIONS

This is the first study in which organ dose from contaminant radionuclides in recycled uranium was estimated in support of an epidemiological study of uranium-exposed workers. Intakes and absorbed organ doses from exposure to ^{99}Tc, ^{238,239}Pu and ^{237}Np were estimated for a combined cohort of 29 303 workers at three US gaseous diffusion plants. Although urinalysis data for these radionuclide nuclides were available for <10% of the combined cohort, using imputation methods organ doses from exposure to ^{99}Tc were estimated for 66% of the K-25 cohort, 12% of the Paducah cohort and 9.7% of the Portsmouth cohort. For ^{239}Pu exposure, organ doses were estimated for about 16% of the K-25 cohort. These data will be used to evaluate confounding in an upcoming study modeling dose response for selected outcomes in the combined uranium-exposed cohort.

ACKNOWLEDGEMENTS

This study was made possible by the cooperation and support of the US Department of Energy and its employees and contractors. The authors thank Dr David Tollerud of the University of Louisville and Dr Richard Hornung of the University of Cincinnati for assistance with Paducah work history and exposure data. The findings and conclusions in this report are those of the authors and do not necessarily represent the views of the National Institute for Occupational Safety and Health.

FUNDING

This work was supported in part through an agreement between the US Department of Energy and the National Institute for Occupational Safety and Health.

REFERENCES

1. U.S. Department of Energy. *A preliminary review of the flow and characteristics of recycled uranium throughout the DOE complex 1952-1999*: Project Overview and Field Site Reports. F-001-001 (2001).
2. International Commission on Radiological Protection. *Age-dependent doses to members of the public from intake of radionuclides: Part 2. Ingestion dose coefficients*. ICRP Publication 67 (1993).
3. Leggett, R. and Giussani, A. *A biokinetic model for systemic technetium in adult humans*. J. Radiol. Prot. **35**, 297–315 (2015).
4. Anderson, J. L., Apostoaei, A. I., Yiin, J. H., Fleming, D. A., Tseng, C. Y. and Chen, P. H. *Internal exposure to uranium in a pooled cohort of gaseous diffusion plant workers*. Radiat. Prot. Dosim. **168**, 471–7 (2016).
5. Anderson, J. L. and Apostoaei, A. I. *Method for analyzing left-censored bioassay data in large cohort studies*. J. Expo. Sci. Environ. Epidemiol. (2015) 10.1038/jes.2015.36.
6. International Commission on Radiological Protection. *Human Respiratory Tract Model for Radiological Protection*. ICRP Publication 66 (1994).
7. International Commission on Radiological Protection. *Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 4 Inhalation Dose Coefficients*. ICRP Publication 71 (1996).
8. U.S. Atomic Energy Commission. *AEC Gaseous Diffusion Plant Operations*. ORO-658 (1968).
9. U.S. Department of Energy. Recycled Uranium Mass Balance Project: Oak Ridge Gaseous Diffusion Plant Site Report (2000).
10. U.S. Department of Energy. Recycled Uranium Mass Balance Project: Portsmouth Ohio Site Report (2000).

Downloaded from https://academic.oup.com/rpd/article-abstract/175/4/503/2910616/Exposure-to-Recycled-Uranium-Contaminants-in
by NCIPC user
on 13 September 2017