Actinides in Deer Tissues at the Rocky Flats Environmental Technology Site

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Introduction

The Rocky Flats Environmental Technology Site (Rocky Flats), operated by the U.S. Department of Energy (DOE), is a former nuclear weapons research, development, and production facility located northwest of Denver, Colorado, USA. Historical site activities included the fabrication of components for nuclear weapons from plutonium, uranium, beryllium, and stainless steel, and support activities included chemical recovery and purification of recyclable transuranic actinides. In 1992, the mission of the Rocky Flats site changed from weapons production to environmental cleanup and closure. Cleanup and remediation is being completed by the DOE under oversight by the U.S. Environmental Protection Agency (EPA) and the Colorado Department of Public Health and Environment (CDPHE).

By mandate of the Rocky Flats National Wildlife Refuge Act of 2001 (P.L. 107-107), at site closure, portions of the site will become the Rocky Flats National Wildlife Refuge to be managed by the U.S. Fish and Wildlife Service (Service). Transfer of property is contingent on EPA certification that cleanup and closure activities have been completed, and that all monitoring and maintenance activities are operating properly and successfully.

The majority of the Rocky Flats site has remained undisturbed since its acquisition by the Federal Government and provides habitat for many wildlife species, including abundant populations of mule and white-tailed deer, and seasonal populations of elk [1]. According to the National Wildlife Refuge System Improvement Act of 1997 (P.L. 105-57), hunting is one of six wildlife-dependent priority public uses that must receive enhanced consideration in Service Refuge planning and management. Given that Rocky Flats ungulates have had access to actinide-contaminated areas [2], measurements of actinides in a range of tissues were needed to provide important information regarding potential human consumption risks, and resultant compatibility of incorporating hunting as a recreational use on the Refuge.

The Service conducted this study to determine concentrations of selected actinides in relevant Rocky Flats ungulate tissues. Further, these analytical results were used to carry out a series of conservative, risk-based calculations to define human risk associated with ingesting these tissues.

Methods

Study Area

The Rocky Flats Environmental Technology Site is a 6,240-acre property located approximately 16 miles northwest of Denver, Colorado, and is bordered by Boulder, Broomfield, and Jefferson counties (Figure 1). Vegetation communities at Rocky Flats include unique xeric tallgrass prairie and tall upland shrubland, along with riparian

woodland, riparian shrubland, wetlands, mesic mixed grassland, xeric needle and thread grassland, reclaimed mixed grassland and ponderosa pine woodland [3].

Field Collection

Deer tissues were collected on the Rocky Flats site on December 8th, 2002, during a Chronic Wasting Disease (CWD) study conducted by the Colorado Division of Wildlife. Twenty-six resident deer were culled to test for CWD, and at this time, USFWS biologists and one Rocky Flats ecologist harvested lung, liver, kidney, muscle and bone tissues from the carcasses. Control tissue samples were obtained on February 4th, 2004 from a mule deer killed by a vehicle at the Rocky Mountain Arsenal National Wildlife Refuge (RMA). All tissues were rinsed with distilled water to remove any surface contamination, and were subsequently individually weighed, labeled, and double bagged. Bulk tissues remained frozen in a secure, sealed freezer (-10°C) at the RMA until July 6th, 2004 at which point sub-samples were processed, packaged in ice, and shipped overnight to General Engineering Laboratories (GEL) in Charleston, SC for laboratory analyses. Additional sub-samples were re-sent on July 21st, 2004 due to laboratory error.

Actinide Analyses

As the primary edible portions of the deer, all muscle and liver tissues were analyzed for all actinide isotopes of concern (^{241}Am, ^{238}Pu, ^{239,240}Pu, ^{233,234}U, ^{235,236}U and ^{238}U). A subset of harvested lung, kidney, and bone tissues were analyzed for select actinides to obtain information regarding relative accumulation in non-edible tissues. In total, 90 tissue samples were analyzed for Plutonium isotopes; 27 muscle, 27 liver, 6 kidney

(composite), 15 lung, and 15 bone samples. Seventy-five sets of Americium isotopic analyses were completed; 27 muscle, 27 liver, 6 kidney (composite), and 15 lung samples. Uranium analyses were conducted on 75 samples; 27 muscle, 27 liver, 6 kidney (composite), and 15 lung samples.

Analytical methodology was derived from a source method from the U.S. Department of Homeland Security Environmental Measurements Laboratory Methods Manual HASL 300 U-04-RC, and uses similar principles of radiochemical separation and counting [4]. All samples were digested, if necessary, and aliquoted. Transuranic elements were scavenged by co-precipitation with iron hydroxide, and the resultant precipitates were dissolved, and separation of elements was accomplished through the use of extraction chromatography and ion exchange resins. Elements were then prepared for measurement of radioactive isotopes by co-precipitation with Neodymium fluoride. Neodymium fluoride precipitates were trapped on filters, mounted on stainless steel disks and placed in a partially evacuated chamber for the measurement of isotopic alpha emissions.

These analyses were performed according to GEL method-specific quality control requirements, including proper instrument calibration, and the use of method blanks, matrix spikes, sample duplicates, and tracer recovery.

Detection limits for analyses were needed that were lower than standard soil and water radiochemistry methods, in order to detect actinide concentrations typical of tissue

samples. To reach these levels, the laboratory used large sample sizes and longer count times. Calculations used to determine appropriate detection limits are presented below.

Calculation of reportable limits based on potential human risks

In order to assure that detection limits were set sufficiently low to detect tissue concentrations of potential human concern via an ingestion pathway, the following calculations were carried out for each actinide isotope:

$$(1) \quad Effective\ Dose\ Equivalent\ (Sv) = \frac{Risk\ Level}{Risk\ Coefficient\ (1/Sv)}$$

$$(2) \quad Radioactivity\ (Bq) = \frac{Effective\ Dose\ Equivalent\ (Sv)}{Effective\ Dose\ Equivalent/Unit\ Intake\ (Sv \cdot Bq^{-1})}$$

$$(3) \quad Tissue\ Concentration\ (pCi \cdot g^{-1}) = \frac{Radioactivity\ (Bq)}{Edible\ Tissue\ Mass\ (kg)} \times \frac{1\ kg}{1000\ g} \times \frac{27\ pCi}{1\ Bq}$$

Input values and resultant dose calculations are presented in Table 1.

Following these calculations, maximum detection limits required for analytical analyses were established as follows: Isotopic Americium, 0.001 pCi/g; Isotopic Plutonium, 0.002 pCi/g; Isotopic Uranium, 0.02 pCi/g. These calculated values also serve as Report Thresholds (RT) for this study (Table 3).

Results

Contaminant analyses

In total, of the more than 450 individual isotopic analyses that were conducted on Rocky Flats deer tissue samples, 17 resulted in actinide concentrations measured above method detection limits (Table 2).

^{241}Am was detected in select lung, muscle, and kidney tissues of Rocky Flats deer, and was also detected in kidney and liver tissues of the control deer from the Rocky Mountain Arsenal. Both measured isotopes of plutonium (^{238}Pu and ^{239,240}Pu) were detected only in select bone samples from Rocky Flats deer. Uranium isotopes (^{233,234}U, ^{235,236}U and ^{238}U) were detected in select liver and muscle samples of Rocky Flats deer, and were also detected in liver tissue of the control deer.

Radiological Risk Assessment

In order to predict potential radiological risk resulting from ingestion of edible deer tissues from Rocky Flats, a highly conservative estimate of risk was conducted. Uncertainty values were calculated as a function of counting efficiency error, peak area error, isotopic abundance error, systematic error and sample calculated activity. Samples which yielded detectable quantities of any radioisotope were organized (Table 3), and experimental uncertainty values were added to measured results to produce a high-end estimate of tissue concentration. These values were compared with calculated report thresholds. Out of a total of 454 isotopic analyses, two (2) yielded detectable

concentrations of an actinide isotope of concern that, with the uncertainty value added, exceeded the pre-calculated report threshold (Table 3).

All liver and muscle tissues which yielded detects were utilized to back-calculate risk values associated with the ingestion of these tissues. As detailed above, uncertainty values were added to reported detection values to give a high-end estimate of tissue actinide concentrations. Bone, lung, and kidney tissues were not included in this analysis because they are not considered edible tissues for the purposes of this study. The same calculations that were used in the methods for setting detection limits were utilized in this analysis. A value of 2.3 kg was used as an approximate weight of edible organ meat (liver) from a 60 kg deer, while a value of 28 kg was utilized for the edible weight of muscle tissues. On the basis of these conservative assumptions, the actinide risk associated with consuming the edible organ meats of an adult deer on Rocky Flats is presented in Table 3. The highest risk calculated in this exercise was attributable to ^{241}Am in the muscle tissue of deer 38-189-53, with tissue concentrations translating to a 6.76×10^{-8} risk level. This level of risk corresponds with a 1:14,700,000 increased chance of cancer resulting from ingestion of 28 kg of muscle tissue. If this same individual consumed this same amount of deer tissue yearly, throughout his / her lifetime (70 yr), this would result in a 4.73×10^{-6} risk level, or a 1:210,000 increased chance of cancer. This risk level falls within the U.S. EPA's acceptable risk range of 1×10^{-4} and 1×10^{-6} [5].

Discussion / Conclusions

The extremely low levels of actinides present in ungulate tissues at Rocky Flats are likely the result of low levels across a majority of the site's surface soils, very low soil to plant actinide transfer rates [6,7], and low gastrointestinal adsorption rates [8]. Though actinide specific, terrestrial animal assimilation of actinides from the gastrointestinal tract is assumed to be less than 0.01% [9].

Historical studies of actinide levels in deer tissues at Rocky Flats, although limited in both number and scope, have yielded similar results to those detailed in this study. The findings from these studies are presented in Table 4. In summary, plutonium analyses conducted on tissues from eight (8) Rocky Flats Region deer demonstrated tissue plutonium (^{238}Pu and ^{239,240}Pu) concentrations near or below detection limits [10]. Similarly, a total of 12 tissue samples from Rocky Flats deer were analyzed for plutonium isotopes in 1992, and all tissues had activities below detection limits [2].

The risk levels presented in Table 3 are the result of highly conservative assumptions and calculations, and as such, likely overestimate the risk associated with ingestion of Rocky Flats deer tissues. For instance, a hunting program at the future Rocky Flats National Wildlife Refuge would be highly controlled, limiting take of animals to a few weekends, and a few individuals, per year [3]. Further, estimates of the consumable mass of an average deer used in this study are higher than predicted in comprehensive investigations of mule deer carcasses [11]. Finally, as analytical results are only slightly higher than detection limits, it is probable that several could be considered non-detects, given the

equal magnitude of the uncertainty. The control samples in this study qualitatively support this possibility, as tissues obtained from a site not contaminated with actinides yield detectable concentrations of select actinides of the same magnitude as samples from the Rocky Flats site (Table 2). Analysis of control samples in a historical study of Rocky Flats deer tissue actinide concentrations demonstrated similar results, with 2 of 9 tissue analyses indicating detectable concentrations of $Pu^{239,240}$ in deer from uncontaminated regions (Table 4). The uptake of background concentrations of actinides stemming from fallout deposition following weapons testing could serve as an alternative explanation for the presence of detectable concentrations of actinides in off-site deer [12].

As presented, these risk levels are functional as indicators of maximum risk likely to result from human consumption of deer tissues from the Rocky Flats site under future management scenarios. Cleanup activities at Rocky Flats will continue until Site closure, at which point all surface soils will meet stringent, human-risk-based standards, as specified in the Final Rocky Flats Cleanup Agreement of 1996. As the cleanup at Rocky Flats concludes, levels of actinides in deer tissues may decrease, as contaminated surface soils will have been removed, resulting in fewer actinides available on the exterior surfaces of plants and/or assimilated into the plant itself.

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Figure 1: Location of the future Rocky Flats National Wildlife Refuge and surrounding communities.

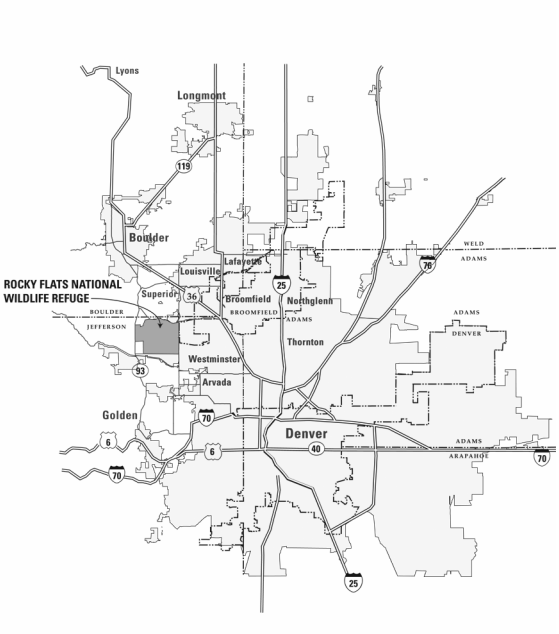

Table 1: Calculation of tissue actinide concentrations necessary for a 10^{-6} additional cancer risk, following human ingestion of all edible tissues from an average sized Rocky Flats deer. Detection limits are set approximately an order of magnitude lower than this calculated value.

Isotope	Risk Level	Risk Coefficient* (1/Sv)	EDE per unit intake** ($Sv \cdot Bq^{-1}$)	Edible Tissue *** (kg)	Dose Calculation ($pCi \cdot g^{-1}$)	Detection Limit ($pCi \cdot g^{-1}$)
^{239}Pu	1×10^{-6}	0.073	9.56×10^{-7}	30.3	0.0128	0.002
^{241}Am	1×10^{-6}	0.073	9.84×10^{-7}	30.3	0.0124	0.001
^{234}U	1×10^{-6}	0.073	7.66×10^{-8}	30.3	0.1594	0.02
^{238}U	1×10^{-6}	0.073	6.88×10^{-8}	30.3	0.1774	0.02

* The Risk Coefficient is needed to convert risk to effective dose. Value obtained from 1990 recommendations of the International Commission on Radiological Protection [13].

** The sum of the effective dose equivalents to various tissues of the body, each multiplied by its weighting factor. Effective dose equivalent per unit intake provides an estimate of the lifetime radiation dose to an individual from radioactive material taken into the body through either inhalation or ingestion [14]. Values in the table are based on the most conservative, gastrointestinal absorption assumptions.

*** An average deer was assumed to weigh 60 kg, of which approximately 28 kg is edible muscle tissue and 2.3 kg is edible organ meat (liver).

Table 2: Comparison of measured concentrations of ^{241}Am, ^{238}Pu, $^{239,240}Pu$, $^{233,234}U$, $^{235,236}U$ and ^{238}U in lung, liver, muscle, bone and kidney tissues of deer sampled at Rocky Flats and the Rocky Mountain Arsenal NWR (control). Data are expressed as pCi/g wet tissue. The reported mean value is calculated as the arithmetic mean of all detects for a specific isotope within that tissue.

		^{241}Am		^{238}Pu		$^{239,240}Pu$		$^{233,234}U$		$^{235,236}U$		^{238}U	
Tissue	**Statistic**	**RF**	**Control**	**RF**	**Control**	**RF**	**Control**	**RF**	**Control**	**RF**	**Control**	**RF**	**Control**
Lung	**Mean**	0.000358	Bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
	n	2/14	0/1	0/14	0/1	0/14	0/1	0/14	0/1	0/14	0/1	0/14	0/1
	Max	0.000468	Bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
Liver	**Mean**	bdl	0.000384	bdl	bdl	bdl	bdl	0.00673	bdl	0.00218	0.00192	bdl	bdl
	n	0/26	1/1	0/26	0/1	0/26	0/1	3/26	0/1	1/26	1/1	0/26	0/1
	Max	bdl	0.000384	bdl	bdl	bdl	bdl	0.0125	bdl	0.00218	0.00192	bdl	bdl
Muscle	**Mean**	0.000307	Bdl	bdl	bdl	bdl	bdl	0.0033	bdl	bdl	bdl	0.00409	bdl
	n	4/26	0/1	0/26	0/1	0/26	0/1	1/26	0/1	0/26	0/1	1/26	0/1
	Max	0.000458	Bdl	bdl	bdl	bdl	bdl	0.0033	bdl	bdl	bdl	0.00409	bdl
Bone	**Mean**	n/a	n/a	0.000623	bdl	0.000624	bdl	n/a	n/a	n/a	n/a	n/a	n/a
	n	n/a	n/a	2/14	0/1	2/14	0/1	n/a	n/a	n/a	n/a	n/a	n/a
	Max	n/a	n/a	0.000642	bdl	0.000773	bdl	n/a	n/a	n/a	n/a	n/a	n/a
Kidney	**Mean**	0.000983	0.000252	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl
	n	1/5	1/1	0/5	0/1	0/5	0/1	0/4	0/1	0/4	0/1	0/4	0/1
	Max	0.000983	0.000252	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl	bdl

n = number of detects / total number of samples analyzed
bdl = below detection limits
n/a = not analyzed

Table 3: Comparison of Rocky Flats deer tissue samples having actinides measurements above detection limits with calculated Report Thresholds.

Deer ID	Tissue Type	Actinide Isotope	Result (pCi/g)	Uncertainty (pCi/g)	Result + Uncert. (pCi/g)	RT (pCi/g)	Above RT?	Calculated Risk Level
Rocky Flats Tissues								
38-189-34	Bone	Pu-238	0.000603	0.000682	0.001285	0.002	**No**	---
38-189-39	Bone	Pu-238	0.000642	0.000629	0.001271	0.002	**No**	---
38-189-39	Bone	Pu-239,240	0.000475	0.000538	0.001013	0.002	**No**	---
38-189-44	Bone	Pu-239,240	0.000773	0.000678	0.001451	0.002	**No**	---
38-189-39	Lung	Am-241	0.000468	0.000375	0.000843	0.001	**No**	---
38-189-40	Lung	Am-241	0.000247	0.000280	0.000527	0.001	**No**	---
38-189-K5*	Kidney	Am-241	0.000983	0.000574	0.001557	0.001	**Yes**	---
38-189-31**	Liver	U-233,234	0.00302	0.00224	0.00526	0.020	**No**	2.51×10^{-9}
38-189-32**	Liver	U-233,234	0.00469	0.00261	0.0073	0.020	**No**	3.48×10^{-9}
38-189-52	Liver	U-233,234	0.0125	0.00926	0.02176	0.020	**Yes**	1.04×10^{-8}
38-189-54	Liver	U-235,236	0.00218	0.00191	0.00409	0.020	**No**	1.83×10^{-9}
38-189-48	Muscle	Am-241	0.000244	0.000276	0.00052	0.001	**No**	3.87×10^{-8}
38-189-49	Muscle	Am-241	0.000258	0.000292	0.00055	0.001	**No**	4.10×10^{-8}
38-189-50	Muscle	Am-241	0.000267	0.000302	0.000569	0.001	**No**	4.24×10^{-8}
38-189-53	Muscle	Am-241	0.000458	0.000449	0.000907	0.001	**No**	6.76×10^{-8}
38-189-41	Muscle	U-233,234	0.00330	0.00374	0.00704	0.020	**No**	4.08×10^{-8}
38-189-34	Muscle	U-238	0.00409	0.00400	0.00809	0.020	**No**	4.21×10^{-8}
Control Tissue								
38-189-K6	Kidney	Am-241	0.000252	0.000285	0.000537	0.001	**No**	---
38-189-60	Liver	Am-241	0.000384	0.000435	0.000819	0.001	**No**	5.01×10^{-9}
38-189-60	Liver	U-235,236	0.00192	0.00188	0.0038	0.02	**No**	1.70×10^{-9}

*** Kidney sample 38-189-K5 is a composite sample, with kidney samples from deer 38-189-44, 38-189-45, 38-189-48, 38-189-49, and 38-189-54.**
****Tissues were involved in a laboratory fire during the ashing process, therefore, the reported values should be considered estimates of concentration. The other two samples involved in the fire (38-189-47 and 38-189-53) were non-detects.**

Table 4: Historical measurements of actinide concentrations in Rocky Flats deer tissue samples.

Study	Tissue	Actinide	n*	Max ($pCi \cdot g^{-1}$)
Hiatt, 1977	Lung	Pu-238	1/7	0.0146
		Pu-239,240	5/7	0.0150
	Liver	Pu-238	0/6	bdl**
		Pu-239,240	0/6	bdl**
	Bone (metacarpal)	Pu-238	0/7	bdl**
		Pu-239,240	1/7	0.0150
	Muscle	Pu-238	0/1	bdl**
		Pu-239,240	0/1	bdl**
	Control***	Pu-238	0/9	bdl**
		Pu-239,240	2/9	0.0191 (liver)
Symonds, 1992	Lung	Pu-238	0/3	bdl**
		Pu-239,240	0/3	bdl**
	Liver	Pu-238	0/4	bdl**
		Pu-239,240	0/4	bdl**
	Bone (Rib)	Pu-238	0/4	bdl**
		Pu-239,240	0/4	bdl**

***- n = number of detects / total number of samples analyzed**
****- bdl = below detection limits**
*****- Control tissues in this study were sampled from 1 lung, 3 livers, and 5 bones from 5 control deer outside of the Rocky Flats area. Detects occurred in the liver tissue sample of deer C1, and in the bone tissue sample of deer C4.**