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Actinides in Deer Tissues at the Rocky Flats Environmental Technology Site

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ABSTRACT

Limited hunting of deer at the future Rocky Flats National Wildlife Refuge has been proposed in U.S. Fish and Wildlife planning documents as a compatible wildlife-dependent public use. Historically, Rocky Flats site activities resulted in the contamination of surface environmental media with actinides, including isotopes of americium, plutonium, and uranium. In this study, measurements of actinides [Americium-241 (^{241}Am); Plutonium-238 (^{238}Pu); Plutonium-239,240 ($^{239,240}\text{Pu}$); uranium-233,244 ($^{233,234}\text{U}$); uranium-235,236 ($^{235,236}\text{U}$); and uranium-238 (^{238}U)] were completed on select liver, muscle, lung, bone, and kidney tissue samples harvested from resident Rocky Flats deer ($N = 26$) and control deer ($N = 1$). In total, only 17 of the more than 450 individual isotopic analyses conducted on Rocky Flats deer tissue samples measured actinide concentrations above method detection limits. Of these 17 detects, only 2 analyses, with analytical uncertainty values added, exceeded threshold values calculated around a 1×10^{-6} risk level (isotopic americium, 0.01 pCi/g; isotopic plutonium, 0.02 pCi/g; isotopic uranium, 0.2 pCi/g). Subsequent, conservative risk calculations suggest minimal human risk associated with ingestion of these edible deer tissues. The maximum calculated risk level in this study (4.73×10^{-6}) is at the low end of the U.S. Environmental Protection Agency's acceptable risk range.

Keywords: Actinides Tissue concentrations Refuge management Ungulates Human risk

INTRODUCTION

The Rocky Flats Environmental Technology Site (Rocky Flats), operated by the U.S. Department of Energy (USDOE), 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 USDOE under oversight by the U.S. Environmental Protection Agency (USEPA) and the Colorado Department of Public Health and Environment.

By mandate of the Rocky Flats National Wildlife Refuge Act of 2001 [Pub. L. No. 107-107, 115 Stat. 102 (2001)] 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 (USFWS). Transfer of property is contingent on USEPA 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 (Kaiser-Hill 2001). According to the National Wildlife Refuge System Improvement Act of 1997 [Pub. L. No. 105-57, 111 Stat. 1252 (1997)], the 6 wildlife-dependent priority public uses that must receive enhanced consideration in USFWS Refuge planning and

management are hunting, fishing, wildlife observation, photography, interpretation, and environmental education. Future Rocky Flats Refuge lands will provide unique access for the disabled and youth to hunt in a controlled environment within close proximity to the Denver metropolitan area. Given that Rocky Flats ungulates have had access to actinide-contaminated areas (Symonds and Alldredge 1992), 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 USFWS conducted this study to determine concentrations of selected actinides in relevant Rocky Flats ungulate tissues. Bone and kidney tissue samples were obtained because actinides are known to accumulate at higher concentrations in these organs (Whicker and Schultz 1982). Lung tissues were evaluated to assess actinide exposure to deer via the inhalation pathway. Finally, liver and muscle tissues were investigated because they are the tissues of the organism that are typically consumed by humans following a successful hunt. Analytical results from these edible tissues were used to carry out a series of conservative risk-based calculations to define human risk associated with ingesting these tissues.

STUDY AREA

The Rocky Flats Environmental Technology Site is a 6,240-acre property located approximately 16 miles northwest of Denver, Colorado, USA, 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 (USFWS 2004).

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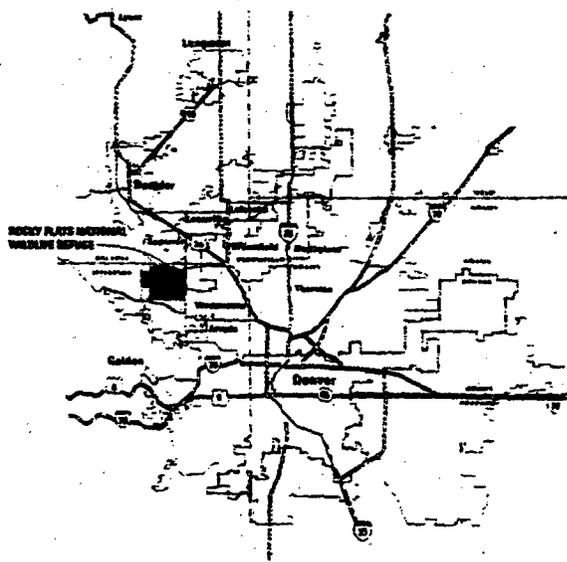


Figure 1. Location of the future Rocky Flats National Wildlife Refuge and surrounding communities.

METHODS

Field collection

Deer tissues were collected on the Rocky Flats site on 8 December 2002, during a chronic wasting disease study conducted by the Colorado Division of Wildlife. Twenty-six resident deer (24 mule, 1 whitetail, and 1 hybrid) were culled to test for chronic wasting disease and, at that time, USFWS biologists and 1 Rocky Flats ecologist harvested lung, liver, kidney, muscle, and bone tissues from the carcasses. Control tissue samples were obtained on 4 February 2004 from a mule deer killed by a vehicle at the Rocky Mountain Arsenal National Wildlife Refuge. Although the Rocky Mountain Arsenal was once a chemical weapons manufacturing center, a lack of historical radionuclide use on the site makes it an appropriate location for collection of reference deer tissues. The opportunistic harvest and subsequent analysis of tissues from this animal functioned, in conjunction with historical reference samples, as a qualitative indicator of regional, background radionuclide concentrations in deer tissues. It was decided that additional deer would not be culled for the expressed purpose of verifying low, "background" actinide concentrations. All tissues were rinsed with distilled water to remove any surface contamination and individually weighed, labeled, and double-bagged.

Bulk tissues remained frozen in a secure, sealed freezer (-10°C) at the Rocky Mountain Arsenal until 6 July 2004; at which point, subsamples were processed, packaged in ice, and shipped overnight to General Engineering Laboratories in Charleston, South Carolina, USA, for laboratory analyses. Although the normal contract sample-holding time for radiological samples is 180 d, the shortest half-life of the actinides of interest (^{241}Am , ^{238}Pu , $^{239,240}\text{Pu}$, $^{233,234}\text{U}$, $^{235,236}\text{U}$, and ^{238}U) is 88 y (^{238}Pu). No appreciable loss of activity would have occurred during the 18 months between the time of collection and the time of isotopic analyses. Additional subsamples were resent on 21 July 2004 because of 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 , $^{239,240}\text{Pu}$, $^{233,234}\text{U}$, $^{235,236}\text{U}$, and ^{238}U). A subset of harvested lung, kidney, and bone tissues was analyzed for select actinides to obtain information regarding relative accumulation in nonedible 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 USDOE Environmental Measurements Laboratory Methods Manual and uses similar principles of radiochemical separation and counting (USDOE 1997). All samples were digested, if necessary, and aliquoted. Transuranic elements were scavenged by coprecipitation with iron hydroxide; 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 coprecipitation 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 α emissions.

These analyses were performed according to General Engineering Laboratories 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 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

To ensure 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:

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

Radioactivity (Bq) =

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

Tissue Concentration ($\text{pCi} \cdot \text{g}^{-1}$) =

$$= \left[\frac{\text{Radioactivity (Bq)}}{\text{Edible Tissue Mass (kg)}} \right] \cdot \left(\frac{1 \text{ kg}}{1,000 \text{ g}} \right) \cdot \left(\frac{27 \text{ pCi}}{1 \text{ Bq}} \right) \quad (3)$$

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: