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Accumulation of plutonium in mammalian wildlife tissues following dispersal by accidental-release tests

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ABSTRACT

We examined the distribution of plutonium (Pu) in the tissues of mammalian wildlife inhabiting the relatively undisturbed, semi-arid former Taranaki weapons test site, Maralinga, Australia. The accumulation of absorbed Pu was highest in the skeleton (83% ± 6%), followed by muscle (10% ± 9%), liver (6% ± 6%), kidneys (0.6% ± 0.4%), and blood (0.2%). Pu activity concentrations in lung tissues were elevated relative to the body average. Foetal transfer was higher in the wildlife data than in previous laboratory studies. The amount of Pu in the gastrointestinal tract was highly elevated relative to that absorbed within the body, potentially increasing transfer of Pu to wildlife and human consumers that may ingest gastrointestinal tract organs. The Pu distribution in the Maralinga mammalian wildlife generally aligns with previous studies related to environmental exposure (e.g. Pu in humans from worldwide fallout), but contrasts with the partitioning models that have traditionally been used for human worker-protection purposes (approximately equal deposition in bone and liver) which appear to under-predict the skeletal accumulation in environmental exposure conditions.

1. Introduction

The internalisation of Pu and its subsequent sequestration in mammalian organs has been the topic of numerous studies and reports, mainly related to the occupational exposures of humans working with plutonium compounds (Durbin, 1975; Leggett et al., 2005; Suslova et al., 2002, 2012; Thomas et al., 1984). Absorption and translocation models have been developed for Pu in humans (ICRP, 1990, 1993, 1994) and non-human mammals such as canines (Polig et al., 2000). Development and calibration of these models requires sequestration data and the International Commission on Radiological Protection (ICRP) conducted extensive reviews of mammalian data which resulted in recommended values of approximately equal (45%) Pu partitioning in bone and liver, later modified to 45–50% in bone, 30–45% in liver, and 10% in other tissues and early excreta (ICRP, 1972, 1986, 1990, 1993). These reviews initially relied almost exclusively on laboratory animal data, often with high exposure concentrations (ICRP, 1972). The subsequent reviews included data on humans who had been exposed to the relatively low concentrations of Pu in the general worldwide fallout from weapons testing. Thomas et al. (1984) suggested higher accumulation of absorbed Pu in bone (70% of absorbed body Pu) than in liver (30%) when including environmental exposure, and also estimated accumulation of 1% kidney, 1% spleen, 0.2% lung, as well as 0.8% in excreta (total exceeds 100% due to method derivation).

Although often reported as simplified percentages, the data on partitioning of Pu in mammal tissues exhibit large variation due to: i) different biokinetic controls such as variable exposure times

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(Durbin, 1975; ICRP, 1972, 1986; Thomas et al., 1984); ii) variation in life stage, (e.g. higher skeletal deposition of Pu occurs in the developing bones of juveniles) (ICRP, 1990, 1993); iii) the physico-chemical form of Pu (Durbin, 1975; ICRP, 1986); iv) differences in the route of entry of the Pu into the organism (e.g., gut absorption, lung absorption, injection, intratracheal) (ICRP, 1986); and v) the quantity of introduced Pu (Durbin, 1975; ICRP, 1986; Park, 1986).

Given the large variation in data, the prediction of Pu sequestration in mammalian wildlife in environmental settings may not be, with confidence, directly extrapolated from the bulk of the data obtained from past laboratory experiments (despite these being animal-based). In these experiments, the evaluation periods were often short, exposures were mostly acute, the entry was often by injection or similar direct method, and the concentrations that were administered were often high. These constraints were consistent with the ICRP interest in the protection of workers at nuclear facilities. However, they do not match well with most wildlife exposure conditions where animals living in contaminated areas can be exposed routinely over long time-periods (chronic *vs.* acute), the activity concentrations are comparatively low, and entry routes are primarily by inhalation and ingestion. These environmental exposure conditions, typical for wildlife, are also characteristic of impact assessments for human exposure in the non-radiological worker settings (e.g. contamination of agricultural areas). However, due to sparse data, the ICRP partitioning values have also been used in assessments on human exposure to Pu in agricultural and general environmental settings (Daniels et al., 2007) and for derivation of wildlife transfer parameters (Beresford et al., 2008a).

In addition to being affected by exposure conditions, the uptake and sequestration of Pu in mammals is known to vary according to the physico-chemical form of the Pu, which, in environmental systems, is related to its source and the manner of its dispersal (Salbu, 2001). For example, Pu uptake in the same fish species was higher by orders of magnitude when the Pu was released directly from processing facilities compared to that of atmospheric fallout (Johansen et al., 2013). Studies on exposure to atmospheric fallout have generated some data on Pu partitioning (McInroy et al., 1979; Singh et al., 1983; Takizawa, 1995). Fewer data relate to accidental releases of Pu (e.g. Chernobyl, Ukraine; Mayak, Russia; Palomares, Spain), and at sites where accidental release scenarios were simulated (e.g. Nevada Test Site, United States; Semipalatinsk, Kazakhstan; and Maralinga, Australia). Some of these releases have dispersed Pu over large areas, potentially affecting a range of environmental receptors. The Pu from these sources is characteristically different from that of fallout as it was dispersed at lower temperatures (relative to nuclear detonation) and is often in particulate oxide forms that have low weathering and mobility rates, and therefore may provide for ongoing, persistent exposure in affected areas for many thousands of years (ARL, 1986; Cooper et al., 1994; Johansen et al., 2014). Particulate forms of Pu that are generated and dispersed in relatively low-temperature explosions would be the most likely form released from any future accidental releases associated with the nuclear fuel cycle. The fuel cycle comprises most of the inventory of worldwide Pu which was ~1900 metric tonnes in 2010 and growing by 70–90 metric tonnes per year (Ewing et al., 2010).

The main objective of this study is to better quantify the distribution of Pu in the tissues of wildlife mammals to support assessment of potential impacts to environmental receptors, and to add to the sparse Pu sequestration data for mammals in general under environmental exposure conditions. We test whether such sequestration data compares well with that from previous studies (e.g. ICRP reviews) and assess past and current data for underlying differences.

2. Materials and methods

2.1. Site description and Pu contamination history

The former weapons testing site of Taranaki, Maralinga, is located on the southern edge of the Great Victoria Desert, South Australia, where semi-arid conditions exist with sparse and erratic rainfall (annual mean of 224 mm; MARTAC, 2003). Monthly mean temperatures range from 13 to 25 °C. Summer daytime temperatures commonly exceed 40 °C. Further details of the Taranaki site, including study maps, climate, and physiography are provided in ANSTO (1990), Johansen et al. (2014) and MARTAC (2003).

At the Taranaki test site, approximately 22 kg of Pu were dispersed in twelve “safety trial” tests carried out in 1961–63. Dispersion involved burning, high-explosive detonations, and partially-critical tests which released sufficient energy from vertical jets firing upward to lift plutonium-contaminated debris up to heights of more than 750 m (Burns et al., 1994). In down-wind study areas, particles of submicron to >150 μm diameter sizes were deposited. Although particle spatial distributions are highly variable, measurements within the study deposition plume indicate the presence of 64 particles per kilogram soil based on a screening level of 0.1 Bq of ^{241}Am (ARL, 1990), and approximately one particle >150 μm diameter per 50 kg of 0–10 cm soil (Johansen et al., 2014). The particles contain variable amounts of Pu (mean of ~16%) as PuO_2, uranium isotopes, and a range of metals with iron (Fe) and chromium (Cr) being prominent (ARL, 1990; Burns, 1986; Burns et al., 1990). Particles were found to be friable and were variably soluble in 0.16 M hydrochloric acid (1–96 % soluble over 40 days), but not soluble in simulated lung fluid of neutral pH (Cooper et al., 1994). However, absorption of Taranaki ^{239}Pu into the blood of mammals was previously demonstrated in laboratory rats by Stradling et al. (1992), with blood receiving 5.1% ± 0.07% at the end of one year after a single lung intratracheal instillation.

2.2. Sampling and analysis

Sampling and analysis is described in Johansen et al. (2014) and summarised here. Samples were gathered at two time periods: 1) approximately 25 years after Pu deposition, termed here “1980s period” (ANSTO, 1998; ARL, 1986, 1988, 1990), and 2) approximately 50-years post deposition (2010–2012). Sampling was focused on the Northwest Plume (NW plume) at the Taranaki site and was mainly at distances from 2 to 3 km from the location of the original firing pad (Supplemental Fig. 1). Sampling was in relatively undisturbed sites outside of the zone that was remediated in the late 1990s. In these locations, the soil Pu activity concentrations have remained relatively constant over time, as have the uptake rates in mammals (Johansen et al., 2014).

A total of 98 organ/tissue samples were evaluated, primarily from the placental herbivore *Oryctolagus cuniculus* (European rabbit; n = 12) and the marsupial herbivore *Macropus rufus* (red kangaroo; n = 3), with some limited data also from the placental omnivore *Pseudomys hermannsburgensis* (sandy inland mouse; n = 2).

All specimens from the 2010–12 period (*O. cuniculus* and *P. hermannsburgensis*) were gathered using live trap techniques approved under animal ethics protocols, followed by either release, or selective anaesthesia and euthanasia of target specimens. Muscle subsamples were typically retrieved from inside of the thigh muscle mass. Samples of the whole femur and vertebrae served to capture both trabecular and cortical bone tissue types, and were

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