

Radionuclide concentration ratios in Australian terrestrial wildlife and livestock: data compilation and analysis

M. P. Johansen · J. R. Twining

Received: 27 November 2009 / Accepted: 29 July 2010 / Published online: 15 August 2010
© Springer-Verlag 2010

Abstract Radionuclide concentrations in Australian terrestrial fauna, including indigenous kangaroos and lizards, as well as introduced sheep and water buffalo, are of interest when considering doses to human receptors and doses to the biota itself. Here, concentration ratio (CR) values for a variety of endemic and introduced Australian animals with a focus on wildlife and livestock inhabiting open rangeland are derived and reported. The CR values are based on U- and Th-series concentration data obtained from previous studies at mining sites and ^{241}Am and $^{239/240}\text{Pu}$ data from a former weapons testing site. Soil-to-muscle CR values of key natural-series radionuclides for grazing Australian kangaroo and sheep are one to two orders of magnitude higher than those of grazing cattle in North and South America, and for ^{210}Po , ^{230}Th , and ^{238}U are one to two orders of magnitude higher than the ERICA tool reference values. When comparing paired kangaroo and sheep CR values, results are linearly correlated ($r = 0.81$) for all tissue types. However, kidney and liver CR values for kangaroo are typically higher than those of sheep, particularly for ^{210}Pb , and ^{210}Po , with values in kangaroo liver more than an order of magnitude higher than those in sheep liver. Concentration ratios for organs are typically higher than those for muscle including those for ^{241}Am and $^{239/240}\text{Pu}$ in cooked kangaroo and rabbit

samples. This study provides CR values for Australian terrestrial wildlife and livestock and suggests higher accumulation rates for select radionuclides in semi-arid Australian conditions compared with those associated with temperate conditions.

Introduction

Accumulation of radionuclides in the tissues of wildlife and livestock is important when considering dose to consumers of these animal products (Ewers et al. 2003; Whicker and Shultz 1982). Radionuclide uptake is also important when considering dose to the biota itself, and there is a need for additional uptake data for wildlife and livestock suitable for general use in biota dose assessments (Howard et al. 2009b) and for specific use in parameterising dose assessment models (Beresford et al. 2008; Howard and Larsson 2008; USDOE 2002).

Rangeland locations in Australia provide opportunity for measuring uptake of radionuclides and their progeny in a variety of fauna. Introduced species, such as sheep, cohabit these areas with endemic species, such as kangaroo, and can share similar trophic position while having differing physiology. Published radionuclide uptake data of native Australian fauna in scientific journals are few (Martin et al. 1998; Read and Pickering 1999) and are unavailable for macropods such as kangaroo and wallaby, which dominate these environments and are an important food source in Australia. Far fewer studies have been conducted on uptake of natural-series radionuclides in wildlife and livestock in semi-arid climates generally than similar studies performed on domesticated animals in temperate climates such as in Europe (Calmon et al. 2009; Ewers et al. 2003; IAEA 1994).

This paper is based on a presentation made at the second meeting of the Wildlife Transfer Coefficient Handbook Working Group of the IAEA EMRAS II programme (held at the IAEA, Vienna, 22–24 July 2009).

M. P. Johansen (✉) · J. R. Twining (✉)
Australian Nuclear Science and Technology Organisation,
Locked Bag 2001, Kirrawee DC, NSW 2232, Australia
e-mail: mjo@ansto.gov.au

J. R. Twining
e-mail: jrt@ansto.gov.au

Radionuclide accumulation in terrestrial animals can be quantified as a concentration ratio (CR_t), which, as used in this paper for wildlife and livestock, primarily relates concentration of a radionuclide in the tissue of a receptor animal to the concentration in the soil of the animal's forage area. This use of the CR_t term is more specific than, but consistent with, the general CR definition of *the ratio of the activity density of a radionuclide in the receptor compartment to that in the donor compartment* (ICRU 2001). Soil-to-tissue CR_t values, in contrast with feed-to-tissue values, have been used particularly in studies where animals forage on a variety of diet items over their natural ranges and therefore it is impractical to provide the animals with controlled feed material (Martin et al. 1998) most especially it has been adopted for assessments of exposure of wildlife (e.g., Wood et al. 2009).

Soil-to-tissue CR_t values integrate multiple underlying processes such as soil-to-plant uptake, soil-to-plant adherence, forage selection by grazers, and gastrointestinal absorption processes, among others (Beresford et al. 2007; Howard et al. 2009a). However, the mobility and transfer of radionuclides within these underlying processes can have up to order-of-magnitude variation according to the physicochemical form of the radionuclides, soil types, climatic conditions, receptor species age, and physiology, among others (Linsalata et al. 1989; Martin et al. 1998; and Wood et al. 2009). The limitations of, and large degree of variation among, reported CR values, and other similar measures of accumulation of radionuclides and metals in fauna, have been described and discussed in previous publications including Beresford et al. (2000, 2007), Calmon et al. (2009), Higley and Bytwerk (2007), and Howard et al. (2009b). However, despite limitations, soil-to-animal CR values are applied in current dose assessment software as useful parameters that encompass variation within uptake processes (Beresford et al. 2008).

The primary objective of the present study is to provide CR_t values for endemic and introduced fauna at a range of Australian locations having readily measurable radionuclide soil concentrations. The natural series ^{210}Pb , ^{210}Po , ^{226}Ra , ^{230}Th , and U , as well as weapons-associated ^{241}Am and $^{239/240}\text{Pu}$ are investigated. This paper makes use of data from a variety of sources and particularly focuses on radionuclide uptake at sites, such as prospective uranium mining sites, where wildlife and livestock habitually feed on open rangeland environments. Concentration ratios are derived for endemic fauna such as kangaroo, which have substantially unique characteristics and thus potentially different uptake rates, for comparison with those from more frequently studied species such as the standard reference animals and plants (ICRP 2003). Additional CR_t value data, particularly for infrequently studied Australian species, as well as for more common grazing species such

as sheep in semi-arid conditions, are expected to contribute to current efforts to improve the breadth of radioecological databases (Howard et al. 2009a, b) and to improve available empirical parameters for use in biota dose assessment software such as ERICA (Beresford et al. 2008) and RESRAD-BIOTA (USDOE 2002).

Materials and methods

Data sources

Data used in this paper originated from reports generated for Australian locations that, at the time of study, included four prospective uranium mining sites (Beverley and Honeymoon sites, South Australia; Manyingee and Yeelirrie sites, Western Australia), riparian and floodplain locations downstream of the Ranger uranium mining site, NT, one location within the waste area of an active uranium mining site (Olympic Dam, South Australia), and one former, 1950s-era weapons testing site (Maralinga, South Australia) (Fig. 1). These sites have semi-arid climates (~ 250 – 400 mm annual rainfall averages), except for the Ranger NT location which has a tropical-monsoonal climate. The present study primarily made use of Australian government reports (Ellis 1979; Giles et al. 1990; Williams 1978) and commercial reports (Williams 1980, 1981; Lawson and Williams 1985). Select data from two journal publications, Martin et al. (1998) and Read and Pickering (1999) have been used in this study to provide an inclusive listing of Australian CR values. The search for data was



Fig. 1 Locations at which samples were collected to derive Australian terrestrial soil-to-animal CR_t values reported here. The Maralinga site is a former nuclear weapons testing area and the remainder are in the vicinity of operational or prospective uranium mines

intended to be comprehensive of studies performed in Australia prior to 2009 on radionuclide uptake in wildlife and livestock.

The source data typically included radionuclide concentration data on animals considered as potential food for humans. Data for kangaroos (*Megaleia rufa*, *Macropus giganteus*), lizards including Sand Goanna (*Varanus gouldi*), and witchetty grubs (*Endoxyla leucomochla*) were reported in reference documents as representing food sources for indigenous peoples or other potential consumers of wildlife. Mammal tissue types included muscle (flesh) as representing the predominant tissue likely to be ingested, and also various organs (heart, kidney, liver) that have the potential of being ingested. Kangaroo meat is processed and sold commercially as a food product in Australia as well as in other countries. Most samples considered in this study were uncooked, however, some results for cooked samples were provided from Maralinga with sampling methods designed to consider traditional food preparation methods (Giles et al. 1990).

Sampling methods, as described in the data source documents, typically included the collection of about 1 kg of tissue per animal for larger species such as kangaroo and sheep with data representing individual animals. Where noted in the source documents, the sampled animals were typically adults of either sex. Samples of smaller species, such as rabbit and lizard, were typically treated as individual animals. Muscle and viscera samples were sealed in plastic bags and frozen during transport. Samples were weighed fresh, dried (typically at 60°C), and ashed. Sample analysis was typically completed by gamma and alpha spectrometry on digested ash samples and in one case by neutron activation analysis. Soil sample depths ranged from 0–1.0 to 0–10 cm below the ground surface.

Derivation of CR_t values

A general definition for concentration ratio, CR, is the ratio of the activity density of a radionuclide in the receptor compartment to that in the donor compartment (ICRU 2001). In the present study, the receptor compartments are animal tissues (muscle, viscera, etc.) measured on a fresh (wet) mass basis, and concentration ratios with respect to such tissues are denoted by CR_t . The donor media is primarily the soil of the animal's forage area measured on a dry mass basis. However, in one table, CR_t values relative to range forage (dry mass basis) are presented. Data subsets herein, with sufficient numbers of CR_t values, appear to generally align with a log-normal distribution pattern. However, summary values based on a normal distribution (mean and standard deviation) are reported here to be consistent with current database input requirements and with other literature reports. One-way ANOVA tests are used in the present study when comparing different CR_t data sets.

The process used for deriving the CR_t values in this study included assessment of the source reports to ensure they provided descriptions of site location, sampling design and methods, and laboratory analysis methods sufficient to permit replication. Values of CR_t were derived if the original soil and animal sample data were available from the same report (in all but one case, the soil and animal samples were taken during the same sampling activity) and if the soil samples appeared to represent typical soils within the expected local forage ranges of the animals sampled (see discussion below on uncertainty). In deriving the feed-to-muscle CR_t values, data were used if the vegetation samples were specifically selected as representing site livestock forage at the same location as the animal and soil samples. For example, one study describes the vegetation samples included as the “preferred animal fodder for cattle, sheep, and kangaroo” (Lowson and Williams 1985).

Key uncertainty factors

Derivation of soil-to-animal CR_t values assumes that the soil sample data are representative of the area over which the receptor animal ranges. Although estimates of the size of forage ranges are available from general literature, actual measurement and delineation of the forage ranges did not occur in any of the source studies. Therefore, in considering data from the source studies, while the soil samples were representative of conditions at the study sites, the wildlife sampled may have ranged further than the soil sampling area. In general, the representativeness of the soil samples relative to forage areas is expected to be inversely proportional to the size of the forage area with, for example, a soil sample from a forage area of a square metre being more representative than a soil sample from a forage area of 1000's of square metres. Although dingo and goanna CR_t values are included here, they are expected to have a relatively high level of uncertainty as their predation ranges can be large and are not delineated in the source studies. Where available, the authors used site descriptions of soil and animal sampling locations to determine which of the available soil sample sets best represented the animal forage ranges.

Additional uncertainty is introduced due to variation in soil sampling practices among the source reports, with sampling depths ranging from 1 to 10 cm, the latter being the sampling depth recommended for the current wildlife transfer database (<http://www.wildlifetransferdatabase.org>). Soil sampling depth is more relevant when considering fallout or aeri ally dispersed radionuclides, as was the case at the Maralinga site (0–2.5 cm) when compared to natural-series radionuclides at the other sites.

Some of the variation in this study's CR_t values may be due to additional pathways that are disassociated with soil-to-animal pathways such as ingestion of dissolved

radionuclides from various types of stock-watering wells or springs. The source reports are inconsistent in presentation of site water data and therefore such data are not used in this paper although water ingestion may have been a significant source of radionuclide uptake in sampled animals. In addition, source data do not allow for addressing variation in uptake rates relative to receptor gender, age, or relative to the concentrations of already accumulated radionuclides within the receptor tissue.

The original source reports also vary in their reported minimum detection activities (MDAs). Some data are reported as less than the MDA (<MDA) used during laboratory analysis. In deriving the CR_t values for this paper, the <MDA values are included only when the MDAs are less than the average value of the data set with the intent being that the <MDA value represents a valid non-detection useful for descriptive statistics.

The existence of multiple sources of variation points to the need for additional study to better determine which factors are most important for uptake in wildlife and livestock and to better quantify measurable means of reducing uncertainty. Uncertainty in CR_t s may be reduced through

further study of effects associated with pathway, gender, ecosystem type, and taxonomic differences.

Results and discussion

Natural-series radionuclides

Of the CR_t values derived from U- and Th-series radionuclide data, the mean soil-to-muscle values for ^{210}Po are up to two orders of magnitude higher than those for ^{210}Pb , ^{226}Ra , ^{230}Th , and U (Table 1). For example, ^{210}Po values in kangaroo are significantly higher ($p < 0.01$) by nearly two orders of magnitude than the other radionuclides measured. Elevated ^{210}Po levels in the organs of grazers and foragers at locations having high natural-series radionuclide concentrations have been previously observed in cattle (Smith-Briggs 1984), and sheep (Bunzl and Kracke 1984). Uranium CR_t values indicate the least amount of accumulation among the radionuclides considered.

During this study, fewer data were available for derivation of feed-to-muscle CR_t values (Table 1) compared with

Table 1 Summary of Australian mammal soil-to-muscle and feed-to-muscle, CR_t values for U- and Th-series radionuclides

		^{210}Pb	^{210}Po	^{226}Ra	$^{230}\text{Th}^a$	U ^b
Soil-to-muscle						
Fox ^c	<i>Vulpes vulpes</i>		9.5, 2.4 (8–11), 2	<2.8		3.4, 2.0 (2–5), 2
Kangaroo ^{c,d,e}	<i>Megaleia rufa</i> , <i>Macropus giganteus</i>	2.6, 3.2 (1–9), 6	220, 320 (2–740), 5	4.6, 8.9 (<0.01–24), 9	3.9 (na), 1	1.8, 1.9 (0.1–5), 9
Pig ^f	<i>Sus scrofa</i>	0.2 (na), 3	50 (na), 3	0.4 (na), 3	0.2 (na), 3	0.2 (na), 3
Rabbit ^c	<i>Oryctolagus cuniculus</i>			<1		2.6, 0.6 (2–3), 3
Sheep ^{c,d,e,g}	<i>Ovis aries</i>	2.0, 1.6 (<0.3–6), 14	73, 110 (<1–270), 5	5.1, 8.8 (<0.01–24), 11	4.7, 4.7 (1–14), 7	1.0, 1.0 (<0.04–3), 11
Water buffalo ^f	<i>Bubalus bubalis</i>	0.2 (na), 1	2 (na), 2	0.3 (na), 3	0.03 (na), 3	0.2 (na), 3
Feed-to-muscle						
Kangaroo ^{c,d,e}	<i>Megaleia rufa</i> , <i>Macropus giganteus</i>	13, 19 (<0.01–50), 6	6.7, 3.1 (3.6–11), 5	1.8, 3.7 (<0.1–12), 9	1.2	12, 15 (0.6–36), 9
Sheep ^{c,d,e}	<i>Ovis aries</i>	4.4, 9.0 (<0.1–24), 7	5.2, 4.7 (0.9–12), 5	6.2, 13 (<0.1–42), 9	5.6, 4.9 (0.9–11), 4	11, 12 (0.3–36), 9

Values are derived from raw data provided in the references listed in the footnotes (except for Martin et al. 1998), and comprise means, and if available, standard deviations, ranges, and numbers of samples; all CR_t values $\times 10^{-3}$

^a Th is ^{230}Th , except for values from Martin et al. (1998) which are the average of ^{230}Th and ^{232}Th

^b U is ^{238}U for reference Lawson and Williams (1985), U-total for references Williams (1978, 1981), and the average of ^{234}U and ^{238}U for Martin et al. (1998)

^c Williams (1981), Beverly, SA Australia, soil $n = 4$

^d Lawson and Williams (1985), Manyingee, WA Australia, soil $n = 12$

^e Williams (1978), Yeelirrie, WA Australia, soil $n = 12$

^f Martin et al. (1998), Ranger, NT Australia

^g Williams (1980), Honeymoon, SA Australia, soil $n = 6$

the soil-to-muscle values because fewer studies included flora sampling specifically targeting animal forage. Of the source studies, feed-to-muscle CR_t values were able to be derived for sheep and kangaroo from three locations (Beverly, South Australia; Manyingee and Yeelirrie, Western Australia). The results indicate that in contrast with the soil-to-muscle CR_t values, the feed-to-muscle values for ²¹⁰Po are not elevated relative to the other radionuclides (*p* > 0.05). These data suggest that the dominant pathway segment contributing to the elevated ²¹⁰Po levels in animal tissue, as discussed above, is not in the feed-to-animal transfer, but rather in the accumulation of elevated ²¹⁰Po in the feed plants. This is consistent with previous observations of elevated ²¹⁰Po adsorbed onto the surfaces of plants following atmospheric deposition (Vandenhove et al. 2009). Also in contrast, the sheep feed-to-muscle values for U are elevated relative to the other radionuclide values (*p* < 0.01), suggesting that U in particular has a relatively higher rate of assimilation via ingestion of forage material. However, the data do not provide for assessing the contribution of alternative pathways such as ingestion of water, dust inhalation, or direct soil ingestion.

A set of U- and Th-series data are available from the Olympic Dam, South Australia, site (Read and Pickering 1999). However, those values are not included in Table 1 as they represent a distinctly different set of conditions at a small mining waste disposal site subject to repeated exposure of windborne acid spray (0.7–1.0 pH). The CR_t values derived from data on the single mammal sample (*Mus domesticus*) are 0.75, 0.063, and 0.65 for ²¹⁰Po, ²³⁰Th, and ²³⁸U, respectively. Additional data are available for small lizards including elevated derived average CR_t values of 1.7, 22, 0.8, and 6.0 for ²¹⁰Pb, ²¹⁰Po, ²³⁰Th, and ²³⁸U, respectively, in two body samples of *Ctenophorus nuchalis*.

These CR_t values are elevated by approximately three orders of magnitude above those from published non-waste site areas with differences likely associated with low pH conditions that can increase solubility of the subject radionuclides leading to potential increases in bioavailability and reduced soil concentrations due to enhanced leaching.

Consistent with the discussion on uncertainty above, data in Table 1 indicate standard deviations that are large relative to the mean values, and value ranges can vary up to 3 orders of magnitude given the same radionuclide and receptor. The CR_t values for ²¹⁰Po have the largest variation (SD = 320) and U the smallest (SD = 0.6). However, the instances where variation is small, such as for U in fox and rabbit, are likely associated with small sample sizes rather than variation associated with site conditions or uptake processes.

Former weapons testing site transuranics

Soil and animal tissue data for ²⁴¹Am and ^{239/240}Pu are available from the Maralinga former weapons testing site (Table 2). The ²⁴¹Am and ^{239/240}Pu CR_t data sets are not significantly different (*p* > 0.05) and are highly correlated (*r* = 0.99). Values of CR_t of ²⁴¹Am and ^{239/240}Pu for uncooked rabbit muscle are of the same order of magnitude compared to those used in ERICA (whole-body) while available (cooked) rabbit muscle values are one order of magnitude higher. The cooked tissue data reported for the Maralinga site are intended to represent traditional indigenous methods of food preparation where contact of meat with soil or ash may occur (Table 2). The limited rabbit data set indicates significant differences (*p* < 0.05) between cooked and uncooked CR_t values suggesting some traditional cooking methods (food preparation on the ground surface, cooking in ashes or sand pits) can introduce additional

Table 2 Summary of soil-to-animal CR_t data from the Maralinga site, South Australia

			²⁴¹ Am	^{239/240} Pu
Dingo	<i>Canis lupus dingo</i>	Viscera ^a		1.34
		Muscle and bone ^a		<0.01
Goanna	<i>Varanus gouldi</i>	Whole ^b	1	0.7, 0.0 (0.7–0.7), 2
Kangaroo	<i>Megaleia rufa</i>	Cooked muscle ^b	2.5, 3.7 (0.5–12), 10	2.3, 3.1 (0.4–9), 10
		Cooked viscera ^b	28, 36 (2–110), 10	31, 45 (1–130), 10
Lizard	<i>Pogona minor, A. cristatus, and C. nuchates</i>	Whole ^b	5.6, 3.5 (3–9.6), 3	3.8, 2.3 (1.7–7.7), 3
Rabbit	<i>Oryctolagus cuniculus</i>	Muscle ^b	13, 28 (0.08–83), 8	40, 122 (0.05–410), 11
		Cooked muscle ^b	51, 170 (1–710), 17	120, 450 (0.5–1,900), 17
		Viscera ^a		0.72
		Muscle and bone ^a		0.02
Witchetty grub	<i>Endoxyla leaucomochla</i>	Whole ^b	2.2, 3.9 (0.2–10), 6	1.6, 2.6 (0.1–6.9), 6

Values are means, and if available standard deviations, ranges, and numbers of samples; all CR_t values × 10⁻³

^a Ellis (1979)

^b Giles et al. (1990)

radionuclide contamination. For cooked muscle, mean values of CR_r of ^{241}Am and $^{239/240}\text{Pu}$ for rabbit are generally an order of magnitude higher than those for kangaroo muscle. This suggests greater dust contamination of rabbits during food preparation due to their larger surface area to volume ratio as one possibility among other potential causes.

Although living below the ground surface, witchetty grubs have whole-body CR values that are not particularly elevated relative to soil concentrations. This is consistent with expected limited transport of ^{241}Am and $^{239/240}\text{Pu}$ into subsurface layers of soil due to adsorption on near-surface soils and apparent limited transfer into the roots of the witchetty bush (*Acacia kempeana*) within which the grubs live and feed. Low CR_r values are also consistent with the particulate nature of the Pu contamination at the Maralinga site that is expected to be less available for direct assimilation in animal tissues (Haywood and Smith 1990).

Source documents indicate that the residual ^{241}Am and $^{239/240}\text{Pu}$ contamination at Maralinga included particulates of various sizes which may have been accessible for ingestion by macro fauna, but not necessarily bioavailable for gut transfer (Giles et al. 1990). The Maralinga studies were performed following small-scale remediation efforts and prior to a 1990s remediation effort in which surface material, including particulate contamination, was mechanically gathered from large areas and buried in covered trenches.

Variation among species

Variation of uptake among species can be related to size, metabolic rates, variable gastrointestinal absorption rates, potentially poor correlation between varying territorial ranges and the soil samples collected, among other species-specific differences (Beresford et al. 2007; Calmon et al. 2009; Higley and Bytwerk 2007; Howard et al. 2009a). Consistent with previous studies, variation among individuals exceeds differences among species. However, causal factors affecting uncertainty were not quantified in the data source studies used for this paper.

Relative to comparable data from other continents, accumulation of Ra, Th, and U in Australian grazers is elevated by one to two orders of magnitude (Fig. 2) compared with that of cattle in Brazil, South America and in United States, North America (Linsalata et al. 1989, 1991). Compared to the closest matching whole-body CR values available in ERICA and RESRAD BIOTA tools, the present study's ^{210}Po data for muscle in kangaroo and sheep are up to two orders of magnitude higher than the model default values. In addition, the present study's kangaroo CR_r values for ^{230}Th , and ^{238}U are more than one order of magnitude higher than the defaults available in ERICA. In contrast, the present study's values for ^{210}Pb and ^{226}Ra are up to one order of magnitude less than the

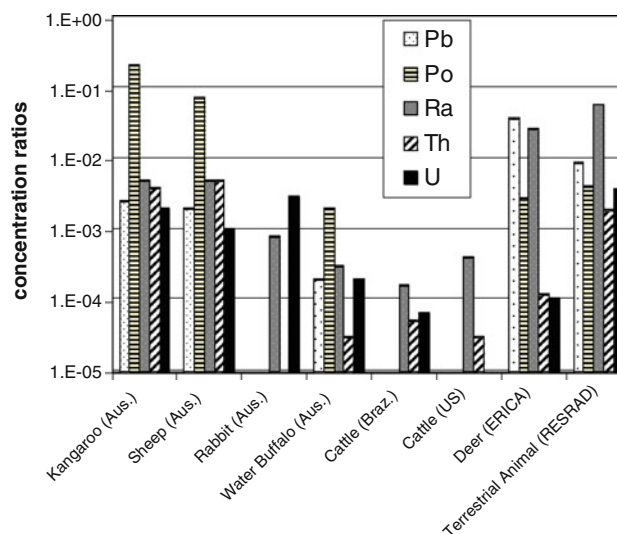


Fig. 2 Comparison of mean CR_r values (soil-to-muscle) for natural-series radionuclides from this study (kangaroo, sheep, rabbit), Martin et al. (1998) (water buffalo), Linsalata et al. (1989, 1991) (cattle), and reference CR whole-body values from the ERICA and RESRAD BIOTA software tools, respectively. Note that in contrast to the data in the accompanying tables, the data in this figure have not been multiplied by 10^3

defaults in ERICA and RESRAD BIOTA. Sparse data do not support derivation of whole-body CR s that would allow for one-to-one comparison between study data and that of ERICA and RESRAD BIOTA. However, the whole-body CR s are generally expected to be greater than the soil-to-muscle CR_r values for the set of radionuclides reported in this study due to accumulation of elevated levels of radionuclides in organs and, in some cases, in bone. Therefore, a direct comparison of whole-body CR s is expected to indicate higher values for the present study's data relative to the ERICA and RESRAD defaults than shown in Fig. 2. Muscle and bone proportions are estimated to average 50 and 10% of whole-body mass for kangaroo, and 33 and 9% for Australian range sheep (not including skin/fur and tail) (Hopwood et al. 1976).

For wildlife and livestock, radionuclide uptake primarily occurs following ingestion of soil, water and feed, as well as following inhalation of dust and gases (IAEA 1994; Whicker and Shultz 1982). For example, grazing sheep can uptake the ^{137}Cs associated with the soil that is adhered to range feed (Beresford et al. 2002; Hinton et al. 1995). Uptake and soil-to-plant adhesion rates can vary depending on factors such as season (Baeza et al. 2001), weather, and grazing density (Hinton et al. 1995) that have the effect of altering percentages of vegetative ground cover. It follows that CR_r values may be elevated in arid and semi-arid climates where higher proportions of bare ground surface can lead to greater availability of soil-sorbed radionuclides for transfer to forage through rainsplash or dust suspension

processes. Future studies that examine climate-related factors such as percentage of ground cover and variation in forage vegetation types could provide improved CR_t value estimates for application in dose assessment models and biota dose evaluations in general.

The present study's data for predators (dingo, goanna, fox) provide evidence of accumulation of radionuclides including uptake of ^{239/240}Pu in dingo viscera, and uptake of ²¹⁰Po, ²²⁶Ra, and U in fox. This accumulation occurred despite expected large forage areas of predators relative to the size of the contamination area. The CR_t values in predators are generally not significantly different (*p* > 0.05) to CR_t values of their probable prey (e.g., fox and rabbit Table 1, dingo and kangaroo Table 2, goanna and smaller lizards Table 2). However, the number of sample replicates for predators is few, and additional data would be needed to determine more representative CR_t values for predators relative to their prey at Australian locations.

Data presented for reptile species in this paper show similar whole-body CR values for Pu as presented by Wood et al. (2010) for lizard and snake species. The CR value presented by Wood et al. for Am is *circa* one order of magnitude higher than that derived here. However, the Wood et al. Am data are dominated by measurements on samples from coastal sand dune ecosystems which receive inputs from marine discharges of radionuclides.

The species evaluated in Tables 1 and 2 include seven mammals, two of which are endemic (kangaroo and dingo), four endemic reptiles (Goanna and three lizards), and one endemic insect (witchetty grub). Differences of CR_t values among mammal species, endemic and introduced, are not significant (*p* > 0.05) for the U- and Th-series values in Table 1. Differences among species are similarly not significant (*p* > 0.05) for the weapons-associated radionuclide CR_t values for uncooked tissue in Table 2. However, the lack of differences among species may result primarily from small sample sizes and associated high variation and suggests further study is required to better quantify any differences that may exist. Willey (2010) studied inter-taxa variation among plant species relative to uptake of Cs, Sr, Co, Cl, and Ru and observed patterns of phylogenetic variation in bioaccumulation. Jeffree et al. (2010), working with marine fish, have found similar phylogenetic differences. Further data are required for Australian species to assess this possibility for endemic organisms as well as in comparison with biota from other regions.

Kangaroo and sheep tissue types

Kangaroo and sheep samples were collected at three locations (Beverly, Yeelirrie, and Manyingee) allowing for comparison of relative uptake of ²¹⁰Pb, ²¹⁰Po, ²²⁶Ra, and U radionuclides among a range of tissue types (Table 3).

Table 3 Natural-series CR_t values (soil-to-animal) for kangaroo and sheep tissue paired at sampling sites in South (SA), and Western (WA) Australia

	²¹⁰ Pb		²¹⁰ Po		²²⁶ Ra		U	
	Kangaroo	Sheep	Kangaroo	Sheep	Kangaroo	Sheep	Kangaroo	Sheep
Beverly SA								
Muscle			4.0, 2.0 (2.3–6.2), 3	4.1, 2.7 (<1.2–6.6), 3	<0.9	8.3, 14 (<0.007–24), 3	2.2, 1.1 (1.0–2.9), 3	1.9, 1.0 (<1.0–2.9), 3
Kidney			360, 150 (270–540), 3	110, 114 (100–120), 2	<1.5	<3.5	5.1, 7.2 (1.0–13), 3	2.9, 2.7 (<1.0–4.8), 2
Liver			520, 318 (300–890), 3	34, 22 (18–50), 2	<1.1	12, 17 (<0.6–24), 2	1.6, 0.6 (1.0–1.9), 3	5.3, 6.1 (<1.0–9.6), 2
Yeelirrie WA								
Muscle	3.4, 3.8 (0.75–9.0), 4	2.1, 1.3 (0.7–4.4), 6			0.13, 0.22 (0.01–0.46), 4	0.16, 0.15 (0.02–0.41), 6	0.17, 0.16 (<0.09–0.41), 4	0.17, 0.12 (<0.04–0.33), 6
Kidney	150, 170 (43–340), 3	18, 6.5 (12–28), 6			1.0, 1.2 (<0.2–2.4), 4	0.81, 0.79 (<0.19–1.4), 6	1.3, 1.5 (0.42–3.0), 4	0.17, 0.13 (<0.07–0.37), 6
Liver	132, 180 (29–400), 4	11, 9.6 (4.1–28), 6			2.3, 3.1 (<0.16–6.7), 4	0.22, 0.18 (0.03–0.48), 6	<0.14	0.28, 0.18 (<0.13–0.50), 6
Bone	54, 49 (13–120), 4	49, 40(15–110), 6			180, 280 (30–600), 4	44, 38 (20–94), 6	<2.6	11, 20 (<1.8–52), 6
Manyingee WA								
Muscle	<1.1	3.0, 3.7 (<0.4–5.6), 2	533, 282 (327–740), 2	175, 127 (85–265), 2	20, 6.4 (<15–24), 2	<15	4.5, 0.8 (4.0–5.0), 2	2.0, 0.01 (1.9–2.0), 2
Viscera	<7.0	1.6, 1.7 (<0.4–2.8), 2	12, 12 (<4–21), 2	335, 120 (250–420), 2	30, 22 (<15–46), 2	27, 17 (<15–40), 2	6.3, 3.8 (3.6–9.0), 2	7.5, 3.6 (5.0–10), 2

Values are means, and if available standard deviations, ranges, and numbers of samples; all CR_t values × 10⁻³

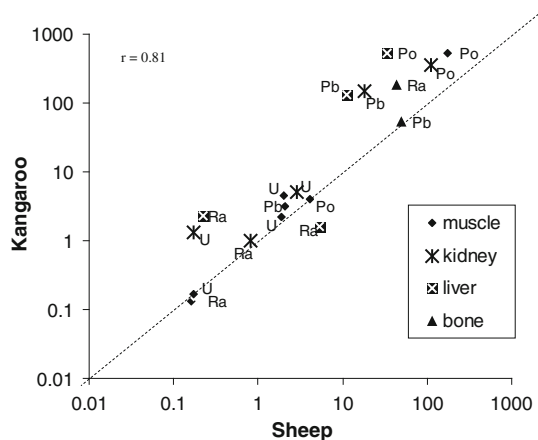


Fig. 3 Comparison of kangaroo and sheep CR_f values data pairs of tissue (muscle, liver, kidney, and bone) sampled at Beverley, South Australia; Manyingee and Yeelirrie, Western Australia; all values $\times 10^{-3}$

When considering all tissue types, kangaroo and sheep CR_f values are linearly correlated ($r = 0.81$, Fig. 3) over a range of 5 orders of magnitude. For kidney and liver, kangaroo data indicate consistently higher CR_f values than sheep, particularly for ^{210}Pb , and ^{210}Po with values in kangaroo liver up to an order of magnitude higher than those in sheep liver. Relative to muscle across all four radionuclides, sheep CR_f values average 68% of kangaroo values. Values of ^{226}Ra are elevated in bone compared with other tissue in both species. Similar observations of elevated ^{226}Ra in bone were previously made for cattle (Linsalata et al. 1991) and are consistent with expected accumulation of a divalent nutrient along with calcium in bone. Basal metabolic rates for marsupials are generally about 70% of those for comparable placentals; however, food habits and activity are sometimes strong enough to mask phylogenetic differences (Tyndale-Boscoe 2001). In addition, the lower metabolic rates imply slower elimination rates that may contribute to the higher accumulation in kangaroos when compared to sheep.

Comparison of tissue types for the same species indicate kidney and liver values are consistently higher than muscle values with order of magnitude differences in ^{210}Pb and ^{210}Po CR_f values. Among radionuclides, the rank of CR_f values for muscle and organs is generally $\text{Po} > \text{Pb} > \text{U} > \text{Ra}$ for both animal species (Fig. 3).

Conclusions

This study provides empirical radionuclide CR_f values for endemic and introduced species inhabiting Australian rangelands. The CR_f values reported here are derived from data obtained from previous studies at seven locations in Australia representing primarily mining sites having

U- and Th-series radionuclides in soil, and one former weapons testing site having ^{241}Am and $^{239/240}\text{Pu}$ in soil. Soil-to-muscle CR_f values of natural-series radionuclides for Australian kangaroo and sheep are one to two orders of magnitude higher than those for cattle in Brazil and in the United States. Soil-to-muscle CR_f values in Australian kangaroo and sheep for ^{210}Po , ^{230}Th , and ^{238}U are one to two orders of magnitude higher than whole-body ERICA tool reference values. Comparing among radionuclides, values of ^{210}Po in muscle and organ samples are consistently higher than values for the other natural-series radionuclides. Kangaroo organs typically have higher CR_f values than those of sheep, particularly for ^{210}Pb and ^{210}Po . Values for kangaroo muscle and sheep muscle are linearly positively correlated ($r = 0.81$) for paired samples gathered at three semi-arid sites with rangeland conditions.

In addition to CR_f values relative to soil, feed-to-tissue CR_f values are derived at rangeland locations. In contrast with the soil-to-muscle CR_f values, the feed-to-muscle values for ^{210}Po are not elevated relative to the other radionuclides suggesting the dominant pathway segment contributing to the elevated ^{210}Po levels in animal tissue is the accumulation of elevated ^{210}Po in the feed plants rather than the feed-to-animal transfer.

This study responds to a general need for better quantification of CR values and provides the first published set of CR_f values for kangaroo, sheep, dingo, and other fauna in semi-arid Australian conditions. Some data sets used are limited in scope and replication numbers, and the study highlights the need for additional data to more fully quantify these CR s. Additional data would better define CR values in general as well as providing for comparison between predator–prey values and for comparison of accumulation rates in semi-arid Australian conditions with those of other climates.

Acknowledgments Research for this paper was performed within the Radioactive Waste Science Project (project leader: Tim Payne) and the Nuclear Methods in Earth Systems Project (project leader: Henk Heijnis), within the Institute for Environmental Research, Australian Nuclear Science and Technology Organisation (ANSTO). Alexander R. Williams was the principle investigator on three of the original reference studies for this paper. Paul Martin and colleagues at the Environmental Research Institute of the Supervising Scientist (ERISS) provided a key reference for this study that includes data on aquatic as well as terrestrial species near the Ranger mine site, NT, Australia. We thank the principles and contractors of Paladin Energy Ltd and Uranium One Pty Ltd for allowing publication of commercially acquired data for animals which contributes to a very limited international set. Manuscript drafts were reviewed by Dr. Che Doering and Dr. Karina Meredith.

References

- Baeza A, Paniagua J, Rufo M, Guillén J, Sterling A (2001) Seasonal variations in radionuclide transfer in a Mediterranean grazing-land ecosystem. *J Environ Radioact* 55:283–302

- Beresford NA, Barnett CL, Mayes RW (2000) Radiocaesium variability within sheep flocks: determination of ^{137}Cs intake in free-ranging sheep. *Radiat Environ Biophys* 39:207–212
- Beresford NA, Barnett CL, Coward PA, Howard BJ, Mayes RW (2002) A simple method for the estimation of the bioavailability of radiocaesium from herbage contaminated by adherent soil. *J Environ Radioact* 63:77–84
- Beresford NA, Barnett CL, Wright SM, Howard BJ, Crout NMJ (2007) Factors contributing to radiocaesium variability in upland sheep flocks in west Cumbria (United Kingdom). *J Environ Radioact* 98:50–68
- Beresford NA, Barnett CL, Howard BJ, Scott WA, Brown JE, Copplestone D (2008) Derivation of transfer parameters for use within the ERICA Tool and the default concentration ratios for terrestrial biota. *J Environ Radioact* 99:1393–1407
- Bunzl K, Kracke W (1984) Distribution of ^{210}Pb , ^{210}Po , stable lead and fallout ^{137}Cs in soil, plants and moorland sheep of a heath. *Sci Tot Environ* 39:143–159
- Calmon P, Thiry Y, Zibold G, Rantavaara A, Fesenko S (2009) Transfer parameter values in temperate forest ecosystems: a review. *J Environ Radioact* 100:757–766
- Ellis WR (1979) A survey of the Maralinga atomic weapons testing range for residual plutonium contamination. AAEC/E476, Australian Atomic Energy Commission, Research Establishment, Lucas Heights
- Ewers LW, Ham GJ, Wilkins BT (2003) Review of the transfer of naturally occurring radionuclides to terrestrial plants and domestic animals. National Radiation Protection Board, Chilton, NRPB-W49, London UK
- Giles MS, Domel RU, Jeffrey RA, Twining JR, and Williams AR (1990) Final report to the technical assessment group for the Maralinga rehabilitation project, study no. 2 (Radioecology), Technical assessment group study program reports, Department of Primary Industries and Energy
- Haywood SM, Smith J (1990) Assessment of the potential radiological impact of residual contamination in the Maralinga and Emu areas. National Radiation Protection Board, Chilton, NRPB-R237, London UK
- Higley KA, Bytwerk DP (2007) Generic approaches to transfer. *J Environ Radioact* 98:4–23
- Hinton TG, Stoll JM, Tobler L (1995) Soil contamination of plant surfaces from grazing and rainfall interactions. *J Environ Radioact* 29:11–23
- Hopwood PR, Hilmi M, Butterfield RM (1976) A comparative study of carcass composition of kangaroo and sheep. *Aust J Zool* 24:1–6
- Howard BJ, Larsson CM (2008) The ERICA Integrated Approach and its contribution to protection of the environment from ionising radiation. *J Environ Radioact* 99:1361–1363
- Howard BJ, Beresford NA, Barnett CL, Fesenko S (2009a) Quantifying the transfer of radionuclides to food products from domestic farm animals. *J Environ Radioact* 100:767–773
- Howard BJ, Beresford NA, Barnett CL, Fesenko S (2009b) Radionuclide transfer to animal products: revised recommended transfer coefficient values. *J Environ Radioact* 100:263–273
- International Atomic Energy Agency (IAEA) (1994) Handbook of parameter values for the prediction of radionuclide transfer in temperate environments. Technical report no. 364. IAEA, Vienna
- International Commission on Radiation Units & Measurements (ICRU) (2001) Quantities, units and terms in radioecology (Report 65). *J ICRU* vol 1, No. 2
- International Commission on Radiological Protection (ICRP) (2003) A framework for assessing the impact of ionising radiation on non-human species—ICRP publication 91. Valentin J (ed) *Annals of the ICRP*, vol 33, No. 3
- Jeffrey RA, Oberhansli F, Teyssie J-L (2010) Phylogenetic consistencies among chondrichthyan and teleost fishes in their bioaccumulation of multiple trace elements from seawater. *Sci Tot Environ*. Available online doi:10.1016/j.scitotenv.2010.04.015
- Linsalata P, Morse R, Ford H, Eisenbud M, Franca EP, de Castro MB, Lobao N, Sachett I, Carlos M (1989) Transport pathways of Th, U, Ra and La from soil to cattle tissues. *J Environ Radioact* 10:115–140
- Linsalata P, Morse R, Ford H, Eisenbud M (1991) Th, U, Ra and rare earth element distributions in farm animal tissues from an elevated natural radiation background environment. *J Environ Radioact* 14:233–257
- Lowson RT, Williams AR (1985) A baseline radioecological survey, Manyingee uranium prospect, Western Australia; AAEC/C47, Australian Atomic Energy Commission, Research Establishment, Lucas Heights Research Laboratories
- Martin P, Hancock GJ, Johnston A, Murray AS (1998) Natural-series radionuclides in traditional North Australian aboriginal foods. *J Environ Radioact* 40(1):37–58
- Read J, Pickering R (1999) Ecological and toxicological effects of exposure to an acid, radioactive tailings storage. *Environ Monit Assess* 54:69–85
- Smith-Briggs JL (1984) Measurements of natural radioactivity in cattle liver, soil and grass. Proceedings of the 4th symposium on the determination of radionuclides in environmental and biological materials
- Tyndale-Boscoe CH (2001) Australian marsupials—to cherish and to hold. *Reprod Fertil Dev* 13:477–485
- USDOE (2002) RESRAD-BIOTA: a tool for implementing a graded approach to biota dose evaluation-ISCORS technical report 2004-02; DOE/EH-0676. United States Department of Energy, Washington, DC, USA
- Vandenhove H, Olyslaegers G, Sanzharova N, Shubina O, Reed E, Shang Z, Velasco H (2009) Proposal for new best estimates of the soil-to-plant transfer. *J Environ Radioact* 100:721–732
- Whicker FW, Shultz V (1982) Radioecology: nuclear energy and the environment. CRC Press, Boca Raton, Florida, USA
- Wiley NJ (2010) Phylogeny can be used to make useful predictions of soil-plant transfer factors for radionuclides. *Radiat Environ Biophys* (this issue) doi:10.1007/s00411-010-0320-2
- Williams AR (1978) The distribution of some naturally occurring elements in the environment of the Yeelirrie Uranium Deposit, Western Australia; Three baseline studies in the environment of the uranium deposit at Yeelirrie; AAEC/E447. Australian Atomic Energy Commission, Research Establishment, Lucas Heights
- Williams AR (1980) Some background radiological data for the proposed Honeymoon Uranium Development, South Australia; AAEC/C3. Australian Atomic Energy Commission, Research Establishment, Lucas Heights
- Williams AR (1981) Background radiological data for the proposed Beverly Uranium Development, South Australia; AAEC/C22. Australian Atomic Energy Commission, Research Establishment, Lucas Heights
- Wood MD, Leah RT, Jones SR, Copplestone D (2009) Radionuclide transfer to invertebrates and small mammals in a coastal sand dune ecosystem. *Sci Tot Environ* 407:4062–4074
- Wood MD, Beresford NA, Semenov DV, Yankovich TL, Copplestone D (2010) Radionuclide transfer to reptiles. *Radiat Environ Biophys* (this issue) doi:10.1007/s00411-010-0321-1