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Whole-organism concentration ratios in wildlife inhabiting Australian uranium mining environments

Gillian A. Hirth [a, *], Mathew P. Johansen [b], Julia G. Carpenter [a], Andreas Bollhöfer [c, 1], Nicholas A. Beresford [d]

[a] Australian Radiation Protection and Nuclear Safety Agency (ARPANSA), 619 Lower Plenty Rd, Yallambie, 3085, Victoria, Australia
[b] Australian Nuclear Science and Technology Organisation, Locked Bag 2001, Kirrawee, DC, NSW 2232, Australia
[c] Environmental Research Institute of the Supervising Scientist, GPO Box 461, Darwin, 0801 Northern Territory, Australia
[d] Natural Environment Research Council - Centre for Ecology & Hydrology, Bailrigg, Lancaster LA1 4AP, United Kingdom

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ABSTRACT

Wildlife concentration ratios for ^{226}Ra, ^{210}Pb, ^{210}Po and isotopes of Th and U from soil, water, and sediments were evaluated for a range of Australian uranium mining environments. Whole-organism concentration ratios ($CR_{wo\text{-}media}$) were developed for 271 radionuclide-organism pairs within the terrestrial and freshwater wildlife groups. Australian wildlife often has distinct physiological attributes, such as the lower metabolic rates of macropod marsupials as compared with placental mammals. In addition, the Australian $CRs_{wo\text{-}media}$ originate from tropical and semi-arid climates, rather than from the temperate-dominated climates of Europe and North America from which most (>90%) of internationally available $CR_{wo\text{-}media}$ values originate. When compared, the Australian and non-Australian CRs are significantly different for some wildlife categories (e.g. grasses, mammals) but not others (e.g. shrubs). Where differences exist, the Australian values were higher, suggesting that site-, or region-specific $CRs_{wo\text{-}media}$ should be used in detailed Australian assessments. However, in screening studies, use of the international mean values in the Wildlife Transfer Database (WTD) appears to be appropriate, as long as the values used encompass the Australian 95th percentile values. Gaps in the Australian datasets include a lack of marine parameters, and no CR data are available for freshwater phytoplankton, zooplankton, insects, insect larvae or amphibians; for terrestrial environments, there are no data for amphibians, annelids, ferns, fungi or lichens & bryophytes. The new Australian specific parameters will aide in evaluating remediation plans and ongoing operations at mining and waste sites within Australia. They have also substantially bolstered the body of U- and Th-series $CR_{wo\text{-}media}$ data for use internationally.

1. Introduction

Evaluation of radiation doses to wildlife is required for environmental impact assessments conducted at mining sites involving naturally occurring radioactive material (NORM). Whole-organism concentration ratios ($CR_{wo\text{-}media}$) are essential in these assessments (if site-specific data are unavailable) as they relate radionuclide activity concentration of the whole-organism (wo) to that of the organism's host environmental medium (media) (Howard et al., 2013; Beresford, 2010). Some standard models for calculating dose rates to wildlife (e.g., ERICA Tool) utilise summarised $CR_{wo\text{-}media}$ values from the Wildlife Transfer Database (WTD, http://www.wildlifetransferdatabase.org/), which was developed through recent work within the International Atomic Energy Agency (IAEA) (Copplestone et al., 2013). Subsequently, the WTD has been updated (see Beresford et al., 2014 and Brown et al., 2016) and we refer to this updated version of the database as 'WTD 2013'.

Major uranium (U) deposits, as well as former and currently operating mines exist in Australia (Fig. 1). However, relatively few data from these sites were included in the pre-2013 WTD due to lack of published $CR_{wo\text{-}media}$ values from Australia. These sites reflect varied environmental conditions (Hirth, 2014) as well as a range of organism types that have been under-represented in the WTD 2013 (e.g. reptiles (Wood et al., 2010)). Within the WTD 2013, data related to U mining sites are numerous for some radionuclide-

* Corresponding author.
E-mail address: Gillian.Hirth@arpansa.gov.au (G.A. Hirth).
[1] Present address: Bundesamt für Strahlenschutz (BfS), Rosastrasse 9, 79098 Freiburg, Germany.

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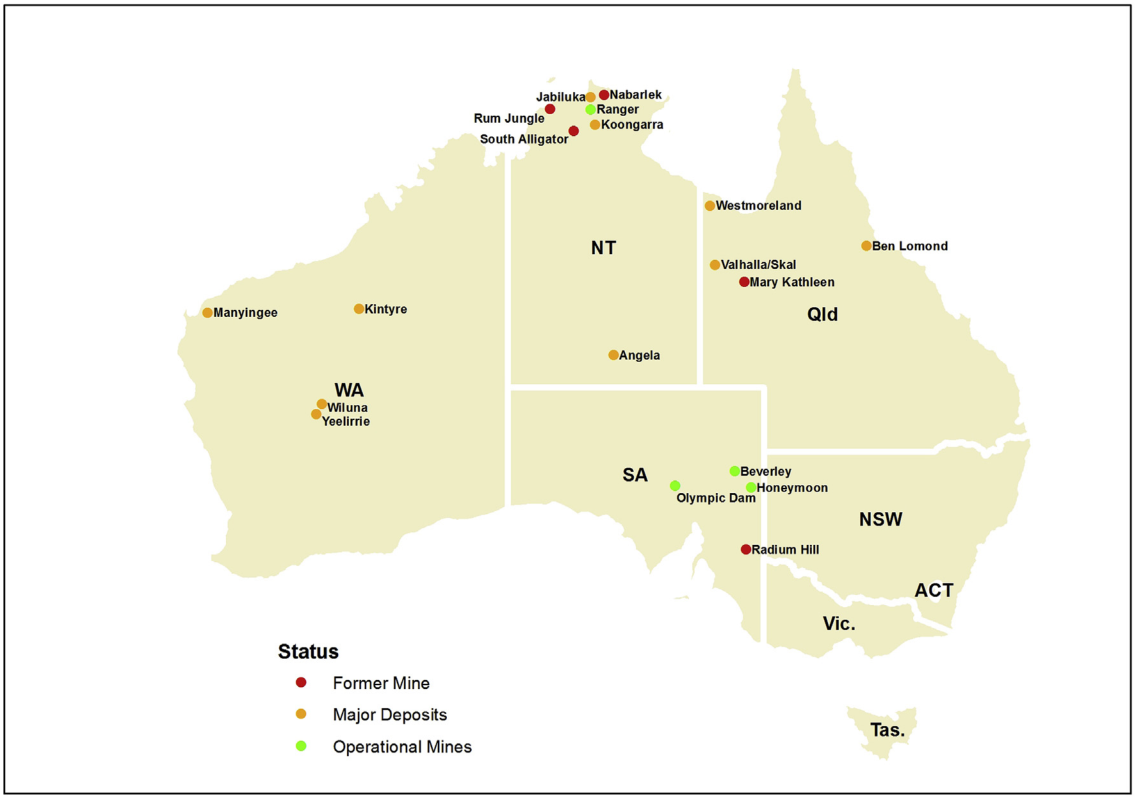

Fig. 1. Major Uranium deposits, operational mines and former mining operations in Australia.

wildlife categories (e.g. n = 569 $CR_{wo\text{-}soil}$ values for U uptake in terrestrial grasses), but sparse for others of importance (e.g. n = 1 for polonium (Po) in terrestrial reptiles). Transfer data for U, Po, and Ra are lacking for more than half of the wildlife categories of the WTD 2013, particularly for those which are not part of the human-ingestion food chain, which has been the focus of most past studies. Doering and Bollhöfer (2016a) however have since published some U-series $CR_{wo\text{-}soil}$ values for mammals and reptiles for the wet-dry tropics of northern Australia.

Some Australian wildlife display unique characteristics that may affect transfer. For example, the metabolic rate of macropod marsupials (e.g. kangaroo, wallaby) is typically approximately 70% of that of similar-sized placental mammals (Tyndale-Biscoe, 2001). Despite this lower metabolism, accumulation of U, Po, and lead (Pb) was greater in most organs of kangaroo as compared with co-located sheep (Johansen and Twining, 2010). Australian environments are also home to numerous reptiles, which fill a broad range of ecological trophic levels as well as providing an important traditional and current food source (Ryan et al., 2005; Martin et al., 1998). The major U deposits in Australia exist predominantly within arid/semi-arid (interior), or tropical/sub-tropical regions and have lower representation in the WTD, which is dominated by data from temperate climate regions (e.g. Europe and North America). These Australian attributes raise questions about the suitability of using the default $CR_{wo\text{-}media}$ values in standard biota dose models for Australian wildlife and environmental conditions. Although some $CR_{wo\text{-}media}$ values have been previously available for Australian wildlife, they have been mostly focused on transfer to tissues, instead of whole-organisms (Johansen and Twining, 2010), and on human ingestion pathways (Martin et al., 1998). A recently published comprehensive environmental dataset specific to the wet-dry tropics of Northern Australia and a tool for calculating $CRs_{wo\text{-}media}$ from those data (Doering and Bollhöfer, 2016b,c) can be used to determine $CR_{wo\text{-}media}$ after upscaling them to whole organism values using appropriate conversion factors (Yankovich et al., 2010). However, data from this data set have yet to be included in the WTD.

In this study, we develop and document a comprehensive set of $CR_{wo\text{-}media}$ values for wildlife inhabiting Australian U mining environments, in both arid and tropical areas. We test whether the new data for Australian conditions differ from the temperate-dominated non-Australian data within the WTD. We identify new data that may substantially improve the robustness of the transfer data within existing database categories and identify data gaps needing future attention.

2. Materials and methods

Most data were sourced from mining site operators and government agencies that provided reports on locations with current or former mining operations, as well as exploratory investigations of prospective mine sites. Some reports are formal environmental assessments, while others are commercial or government reports. Additional data are from scientific journal publications, as well as new unpublished data from site investigations conducted by the authors.

Most of the raw data accessed in this study were not in directly useable formats and required some form of conversion. Prior to ~1980 radionuclide activities were reported in pCi requiring conversion to Bq. A significant amount of data required activity concentrations to be converted from dry-, or ash-based data, to a fresh mass basis (Bq kg^{-1} FM). When available, the reported site-specific dry:fresh weight ratios were used. When no site-specific dry:fresh weight ratios were reported, reference values were used from the author's direct measurements on Australian organisms, or from Beresford et al. (2008) and Hosseini et al. (2008). For shrubs, the reported dry:fresh weight ratios for similar species from arid/desert regions of Australia (average dry:fresh weight ratios in shrub/grass foliage of 0.6) were similar to those used in the WTD for woody parts (0.5), but higher than those used for leaves/berries (0.1) (Beresford et al., 2008). This will potentially result in higher fresh weight $CRs_{wo\text{-}media}$ in more arid climates as they have comparatively low water content. The Australian studies reported that foliage was generally sampled for shrubs, trees and grasses so the value of 0.6 was considered appropriate for all Australian arid region plants.

Most of the reported activity concentration data were tissue-specific (typically muscle for game-animal species) as they were originally collected for the purpose of assessing ingestion doses to humans. For these, the $CR_{wo\text{-}media}$ values were calculated using tissue-to-whole organism ratios following standard approaches (Yankovich et al., 2010; Wood et al., 2010; Hosseini et al., 2008). However, for some species, suitable factors did not exist for converting activity concentrations from tissue data to a whole-organism basis (e.g. Th for some species) and these were therefore not included. The primary review on evaluating which data could be used to calculate $CR_{wo\text{-}media}$ values for wildlife inhabiting Australian uranium mining environments is reported in Hirth, 2014.

Available data were limited to the terrestrial and freshwater aquatic ecosystem categories recognised in the WTD. As many species may move between, or ingest diet items from, various ecosystem types during daily or seasonal routines, or over changing life stages, species are grouped here according to what was reported in the source study, or if unavailable, by their known dominant habitat type. For example the $CR_{wo\text{-}media}$ for goanna lizard *Varanus panoptes* is grouped here relative to freshwater as originally reported, although it forages in both aquatic and on the sediments/soils of floodplain areas (Martin et al., 1998). This has led to differing approaches to calculating uptake factors (Martin et al., 1998; Wood et al., 2010). In this instance, we have reported the values in Wood et al. (2010) as it was used in the WTD 2013.

The WTD 2013 provides summary tables for $CR_{wo\text{-}media}$ values for organism-radionuclide combinations across generic ecosystems (Howard et al., 2013). Most data reported here were incorporated into the WTD during the update that resulted in WTD 2013 (Beresford et al., 2014; Brown et al., 2016). The WTD accepts new data that pass quality assurance and fit-for-purpose screening and periodically updates online summary information (Copplestone et al., 2013). While most data reported here were included in the WTD 2013 update, a small number were not yet finalised at the time of the update, and some of the data reported here were excluded. The WTD excludes data from sites with high heavy metal concentrations at which non-linear transfer may be observed (Copplestone et al., 2013) or other unusual, highly site-specific conditions (Brown et al., 2016). Specifically, the Australian data from a major mine-tailings storage area, with acidic conditions reported in Read and Pickering (1999) were excluded from the WTD 2013. However, these atypical data, specific to acidic mine-tailings, are included in the present study, as a separately identified set, as they are representative of a type of waste configuration that is not uncommon at U mining and processing sites. Hence, as a separate set, these data are potentially useful for Australian and international readers assessing U-mining areas where mine tailings, or similar acidic wastes, are present.

The $CR_{wo\text{-}media}$ values reported here include those available in reports and journal manuscripts as of 2014 for the Ranger Uranium Mine area. Since that time, further $CR_{wo\text{-}media}$ values have been published and concentration ratios made available for that region as part of planning for rehabilitation of the mine (Doering and Bollhöfer, 2016a). Although not included here, additional $CR_{wo\text{-}media}$ values can be calculated from Doering and Bollhöfer (2016b,c) and will be submitted for addition to the WTD in the near future, to provide additional breadth in the number and types of species studied.

The Australian $CR_{wo\text{-}media}$ data were compared with non-Australian data from the WTD 2013 summary tables (using the non-parametric Mann–Whitney *U* test, two-tailed, at $p < 0.01$ and $p < 0.05$ as indicated) appropriate for non-normal distributions. Geometric means (GMs) and geometric mean standard deviations (GMSDs) were calculated by standard equations (Wood et al., 2013).

3. Results and discussion

The work undertaken has resulted in 271 new or revised $CR_{wo\text{-}media}$ values for ^{226}Ra, ^{210}Pb, ^{210}Po and isotopes of Th and U covering five terrestrial wildlife groups (grasses, shrubs, trees, reptiles and mammals) and six freshwater wildlife groups (algae, crustaceans, molluscs, fish, reptiles and vascular plants). The complete set of $CR_{wo\text{-}media}$ values are provided in the supplementary material (Table S1) including those data not yet submitted to the WTD. The new Australian data added substantially (and in some instances provides all of the data) for ^{226}Ra, ^{210}Po and isotopes of Th and U to the existing WTD data in the categories of freshwater algae, crustaceans, and reptiles, as well as in the category of terrestrial trees.

3.1. $CR_{WO\text{-}media}$ values measured from Australian terrestrial organisms

Table 1 presents the summary of the terrestrial $CR_{wo\text{-}media}$ values including the data specific to an acidic tailings retention site (TRS) from Read and Pickering (1999). These data were excluded from the WTD 2013 summary tables as they present outliers (predominantly for mammal and reptile samples) and because the TRS site reflects highly site-specific conditions. The reason for the variation in these results does not appear to be related to a difference in soil activity concentrations, which were similar between the control and the TRS site (of Read and Pickering, 1999). However there was a significant difference in the airborne dust activity concentrations with the TRS site reporting higher activity concentrations, (Pb ~1.5× higher, Po 3× higher; Th ~8× higher; and U ~4× higher). The enhanced exposure of reptile and mammals to these radionuclides via ingestion/inhalation of dust may be one explanation for this variation. The authors cited the acidic nature of the TRS site as a significant factor that increased the bioavailability of the NORM radionuclides.

Metabolic rates of macropod marsupials (e.g. kangaroo, wallaby) is typically approximately 70% of that of similar-sized placental mammals and this may be a factor that can affect transfer. A comparison of mammal data is presented in Fig. 2. The red kangaroo data is from an arid/desert region of South Australia and the water buffalo from the wet-dry tropical region of Northern Australia. The significant difference between the kangaroo and the water buffalo may be related to metabolism; however it may be the result of the significantly different climate and dietary habits of these organisms.

Also, as previously mentioned categorisation of biota as either terrestrial or freshwater depending on where it lives may influence how $CRs_{wo\text{-}media}$ are utilised when undertaking an assessment (Stark et al., 2015). The goanna was one Australian species identified, classified in some instances as terrestrial (*Varanus gouldii*, the sand goanna or Gould's monitor) and others as freshwater (*V. panoptes*, the Argus monitor). While both species are terrestrial they have overlapping habitats in some regions of Australia with

Table 1
Summary of terrestrial $CR_{wo\text{-}media}$ values from Australian uranium mining areas. Additional data rows (shown for comparison and not included in general statistics) are values specific to acidic mine-tailing sites (Read and Pickering, 1999) and from the Ranger Uranium mine area (Doering and Bollhöfer, 2016a).

Terrestrial $CR_{wo\text{-}soil}$								
Wildlife group	Radionuclide	AM	AMSD	GM	GMSD	n[1] number of specimens[a]	n[2] number of species[b]	Reference[c]
Grasses	Pb-210	8.4E-01	6.3E-01	6.5E-01	2.1E+00	38	5	61, 271, 458
	Pb-210	*1.1E+00*	*6.2E-01*			*10*	*1*	*450* [d]
	Po-210	4.9E-01	2.0E-01	4.4E-01	1.6E+00	16	4	61
	Po-210	*2.8E-01*	*2.8E-01*			*10*	*2*	*450*
	Ra-226	1.3E+00	2.2E+00	4.1E-01	4.2E+00	6	4	61, 458
	Th-230	2.1E-01	1.4E-01	1.8E-01	1.7E+00	16	4	61
	Th-230	*7.3E-01*				*5*	*1*	*450*
	U-238	2.7E-01	3.4E-01	1.3E-02	3.9E+00	5	4	61, 458
	U-238	*1.7E+00*				*5*	*1*	*450*
Mammal	Pb-210	1.2E-01	2.2E-01	8.5E-03	1.6E+01	20	4	423, 429, Hirth 2014[e]
	Pb-210	1.7E-03				1	1	Doering & Bollhöfer 2016a[f]
	Po-210	2.0E-01	3.8E-01	5.9E-02	6.0E+00	26	6	61, 423, 429, 509, Hirth 2014
	Po-210	7.5E-02	9.7E-02			2	2	Doering and Bollhöfer 2016a
	Po-210	*5.2E-01*	*4.6E-01*	*7.7E-02*	*2.6E+01*	*3*	*1*	*450*
	Ra-226	4.4E-01	7.3E-01	6.7E-02	1.6E+01	25	4	423, 429, 458, 509, Hirth 2014
	Ra-226	2.1E-01	2.0E-01			6	2	Doering and Bollhöfer 2016a
	Th-230	2.0E-03	1.2E-03			2	2	Doering and Bollhöfer 2016a
	Th-230	*1.5E-02*	*2.1E-02*			*2*	*1*	*450*
	U	2.0E-03	6.2E-04			2	2	Doering and Bollhöfer 2016a
	U	2.4E-02	5.1E-02	4.9E-03	5.9E+00	33	7	61, 423, 429, 458, 509, Hirth 2014
	U	*2.1E-01*	*3.0E-01*			*2*	*1*	*450*
Reptile	Pb-210	1.51E+00				1	1	Hirth 2014
	Pb-210	2.4E-02	1.9E-02			3	2	Doering & Bollhöfer 2016a
	Pb-210	*7.8E-01*	*6.9E-01*	*1.9E-01*	*1.7E+01*	*5*	*2*	*450*
	Po-210	1.4E-01				1	1	Hirth 2014
	Po-210	1.1E-01	6.1E-02			2	1	Doering and Bollhöfer 2016a
	Po-210	*4.4E+00*	*7.7E+00*	*1.1E+00*	*7.7E+00*	*9*	*4*	*450*
	Ra-226	9.0E-01				1	1	Hirth 2014
Reptile	Ra-226	4.6E-01	3.0E-01			5	2	Doering and Bollhöfer 2016a
	Th-230	*1.6E-01*	*2.9E-01*	*1.0E-02*	*1.6E+01*	*8*	*5*	*450*
	Th-230	2.5E-01				1	1	Hirth 2014
	Th-230	1.1E-03	1.1E-03			3	2	Doering and Bollhöfer 2016a
	U	*1.5E+00*	*2.1E+00*	*1.4E-01*	*2.9E+01*	*11*	*7*	*450*
	U	2.4E-03	1.2E-03			4	2	Doering and Bollhöfer 2016a
Shrub	Pb-210	4.1E-01	1.8E-01	3.7E-01	1.5E+00	31	16	61, 560, Hirth 2014
	Pb-210	*1.5E+00*	*8.1E-01*			*25*	*3*	*450*
	Po-210	2.9E-01	6.8E-02	2.9E-01	1.2E+00	14	4	61, Hirth 2014
	Po-210	*7.4E-01*	*3.8E-01*	*6.8E-01*	*1.5E+00*	*15*	*3*	*450*
	Ra-226	2.2E-01	3.4E-01	9.8E-02	4.3E+00	18	14	61, Hirth 2014
	Th-230	1.6E-01	1.3E-01	1.2E-01	2.1E+00	22	5	61, Hirth 2014
	Th-230	*7.3E-01*	*4.7E-01*	*6.5E-01*	*1.6E+00*	*15*	*3*	*450*
	U	7.9E-02	1.0E-01	4.1E-02	3.3E+00	35	12	61, 560, Hirth 2014
	U	*1.9E+00*	*1.6E+00*	*1.4E+00*	*2.3E+00*	*15*	*3*	*450*
Tree	Pb-210	3.9E+01				17	2	271, Hirth 2014
	Po-210	6.0E-01				15	1	271, Hirth 2014
	Ra-226	1.7E-01				21	2	271, Hirth 2014
	Th-230	4.0E-02				15	1	271, Hirth 2014

[a] n^1 = number of specimens/measurements reported.
[b] n^2 = number of species, or pooled-species samples.
[c] References numbers correspond to WTD reference IDs and include: 61=(Williams, 1981), 271=(Davy and O'Brien, 1975), 423=(Lowson and Williams, 1985), 429= (Martin et al., 1998), 450=(Read and Pickering, 1999), 458=(Williams, 1978), 502= (Bollhöfer et al., 2011), 503= (Hancock, 1994), 504=(Johnston, 1987), 505=(Johnston et al., 1984), 507=(Martin et al., 1995), 508=(Ryan et al., 2008), 509=(Ryan et al., 2009), 560=(Williams, 1980).
[d] Italic text in highlighted rows represents values specific to acidic mine-tailing sites reported in reference number 450, Read and Pickering, 1999. Data excluded from WTD.
[e] Data from numerous commercial and government reports in (Hirth, 2014; see Appendix 1 Supplementary material, Table S1).
[f] Data from recently published work for wet-dry tropics of Australia (Doering and Bollhöfer, 2016a) that has not yet been submitted to the WTD.

different dietary habits. The *V. panoptes* is reported to live in the riparian zone of creeks and rivers and have a more significant aquatic food source than the *V. gouldii* (Martin et al., 1995). In the WTD 2013 the *V. panoptes* has been included as a freshwater $CR_{wo\text{-}water}$ reported in Wood et al., 2010. Upon review of the original source data for this organism (Martin et al., 1995) we were able to also determine the terrestrial $CR_{wo\text{-}soil}$ for the *V. panoptes* enabling comparison with other goanna $CRs_{wo\text{-}media}$ that have been reported in this study.

Fig. 3 shows the comparison of the $CR_{wo\text{-}soil}$ values for these goanna, the *V. panoptes* coming from the wet-dry tropics of Northern Australia with a reported aquatic dietary component of ~30% (Martin et al., 1995), but a primarily terrestrial existence. The *V. gouldii* was sampled from the arid-desert region of Western Australia with an entirely terrestrial diet and habitat. The Po $CR_{wo\text{-}soil}$ shows little difference between these two goanna however all other $CR_{wo\text{-}soil}$ values show ~ three orders of magnitude difference (for Th, Pb and Ra). Whilst these two reported goanna represent a

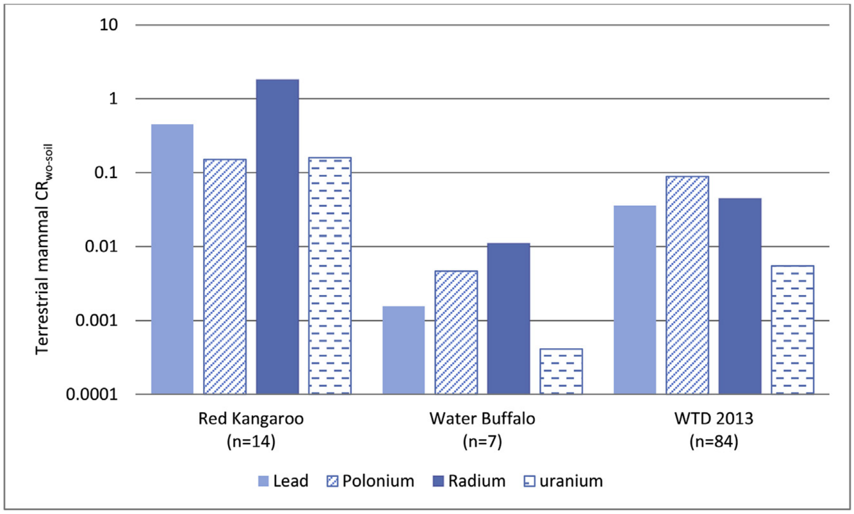

Fig. 2. Comparison of arithmetic mean terrestrial mammal CR$_{wo-soil}$ for red kangaroo (arid-desert), water buffalo (wet-dry tropics) and generic mammal value from WTD 2013. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

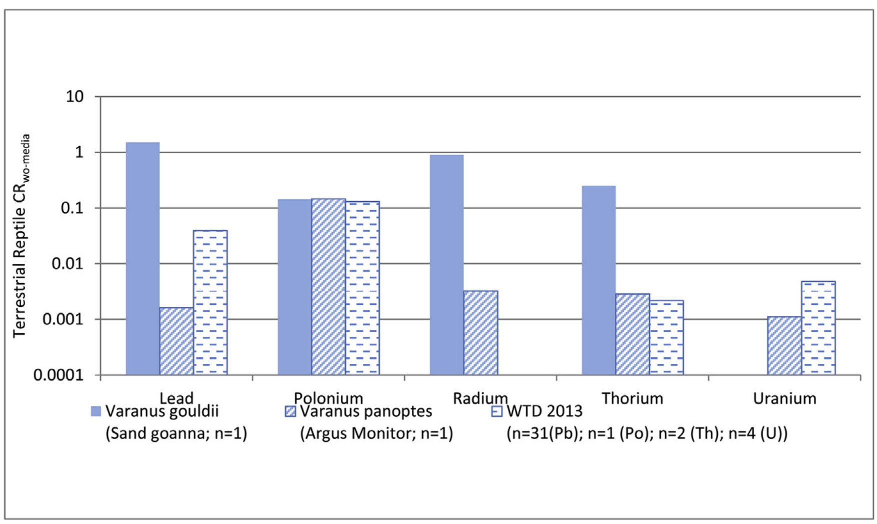

Fig. 3. Comparison of arithmetic mean terrestrial reptile CR$_{wo-soil}$ values for two species of goanna *Varanus panoptes* and *Varanus gouldii* and the generic reptile value from WTD 2013 (note: the WTD 2013 do not include any Australian reptile data).

very limited number of samples (n = 1 for each) they are very similar species sampled from very different climatic regions. The distinct variation in the CR$_{wo-soil}$ values indicates that habitat and diet may play a significant role in CR$_{wo-media}$ and reinforces the importance of having site specific data when detailed radiological assessments are required, in addition to a clear understanding of the biota and their behavioural and dietary habits.

3.2. CR$_{wo-media}$ values measured from Australian freshwater organisms

Table 2 presents the summary of the freshwater CR$_{wo-media}$ values. CR$_{wo-water}$ values for Ra in molluscs were found to range over four orders of magnitude and showed both seasonal and site (between and within) variability, highlighting the importance of understanding specific site (geochemistry and season) and wildlife (age) information (Bollhöfer et al., 2011). While most CR$_{wo-media}$ values for vascular plants submitted to the WTD reflect CR$_{wo-sediment}$ values rather than CR$_{wo-water}$ values they were not recommended in the international handbooks for use as they are likely to be highly specific to the site from which the data were derived (Copplestone et al., 2013). CRs$_{wo-media}$ for both sediment and water transfer to Australian vascular plants are included in the summary tables for information, and again these values demonstrate that transfer is likely to be highly site-specific, incorporating transfer processes from sediment to water and from water to biota as discussed in Copplestone et al., 2013.

3.3. Comparing Australian and non-Australian CR$_{wo-media}$ values

Most of the Australian CR$_{wo-media}$ values did not present as outliers when compared to the mean (arithmetic) summary values from the WTD 2013 (see Fig. 4 for U and Figs. S1–S4 in the supplementary material for Po, Ra, Pb and Th). The importance of this comparison is that it suggests the WTD 2013 values are adequate for use in screening assessments, as long as the values used encompass the Australian 95th percentile values. It does, however, raise the question as to the extent of the difference between the

Table 2
Summary of freshwater $CR_{wo\text{-}media}$ values from Australian uranium mining areas. Additional data that were not included in the general statistics for fish and crocodile from the Alligator Rivers Region are presented in the Supplementary Material Table S1 (see Reference ID 'E' Conway et al., 1974).

Freshwater $CR_{wo\text{-}water}$								
Wildlife group	Radionuclide	AM	AMSD	GM	GMSD	n[1] number of specimens[a]	n[2] number of species[b]	Reference[c]
Algae	Ra-226	1.3E+03				1	1	61
	U	2.8E+02				1	1	61
Crustacean	Pb-210	3.9E+01	4.7E+01			5	1	507
	Po-210	1.2E+03	5.0E+02			5	1	507
	Ra-226	2.7E+02	4.4E+02			5	1	507
	U-238	1.5E+02	3.1E+02			5	1	507
Fish	Pb-210	7.0E+01	6.4E+01	5.0E+01	2.3E+00	20	8	507
	Po-210	5.6E+03	1.2E+04	5.8E+02	7.1E+00	38	12	507
	Ra-226	9.4E+02	1.3E+03	4.7E+02	3.4E+00	35	12	271, 507
	Th-230	1.4E+02	7.4E+01	1.2E+02	1.7E+00	8	3	507
	U-238	1.9E+02	2.1E+02	1.0E+02	3.4E+00	16	8	507
Mollusc - bivalve	Pb-210	2.4E+04	2.2E+04			8	1	505, 508
	Po-210	2.3E+04	4.3E+03			4	1	504, 508
	Ra-226	7.1E+04	4.7E+04	5.9E+04	1.9E+00	14	1	502, 505, 508
Reptile	Pb	5.9E+02	5.9E+01			3	2	487
	Po	4.5E+03	2.4E+03	3.9E+03	1.8E+00	7	4	487
	Ra	1.6E+03	1.9E+03	6.6E+02	4.8E+00	8	4	271, 487
	Th	8.0E+02	5.7E+02	6.4E+02	2.0E+00	7	4	487
	U	9.6E+01	6.9E+01	8.0E+01	1.8E+00	7	4	487
Vascular plant	Ra-226	1.1E+02	8.0E+01			7	1	61
$CR_{wo\text{-}water}$	U	1.4E+01	2.0 + 01			5	1	61
Vascular plant	Pb-210	4.8E-03	4.3E-03			11	1	325, 503
$CR_{wo\text{-}sediment}$	Po-210	3.9E-03	5.1E-05			11	1	325, 503
	Ra-226	4.7E-03	3.6E-03	3.9E-03	1.8E+00	11	1	325, 503
	Th-232	2.1E-03	2.6E-03	1.2E-03	2.8E+00	17	1	325, 503
	U-238	2.1E-03	2.1E-03	1.6E-03	2.1E+00	17	1	325, 503

[a] n[1] = number of specimens/measurements reported.
[b] n[2] = number of species, or pooled-species samples.
[c] References numbers correspond to WTD reference IDs and include: 61=(Williams, 1981), 271=(Davy and O'Brien, 1975), 325=(Pettersson et al., 1993), 487=(Wood et al., 2010), 502=(Bollhöfer et al., 2011), 503=(Hancock, 1994), 504=(Johnston, 1987), 505=(Johnston et al., 1984), 507=(Martin et al., 1995), 508=(Ryan et al., 2008).

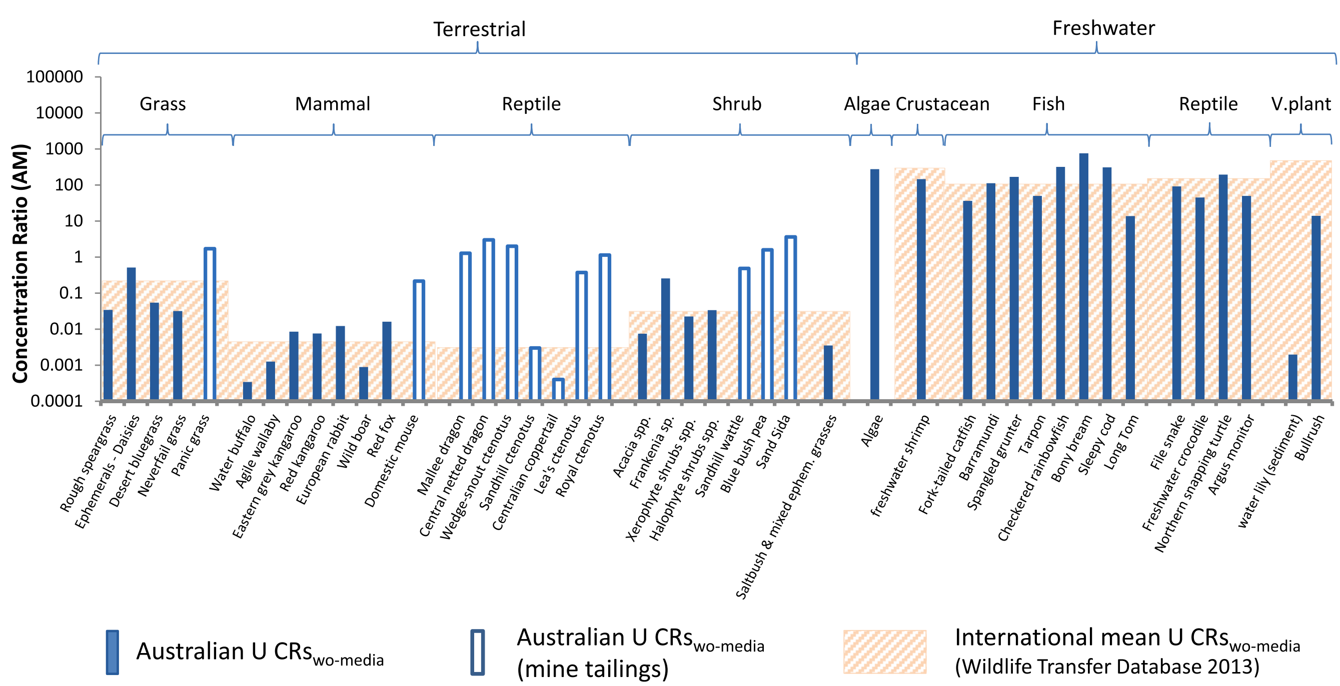

Fig. 4. Comparison of Australian and international arithmetic mean $CR_{wo\text{-}media}$ values for uranium for terrestrial and freshwater organisms.

$CR_{wo\text{-}media}$ values from the semi-arid/tropical regions of Australia and those from temperate regions. In the WTD 2013, the non-Australian data were dominated by temperate climates (approximately 92% of data record entries in the WTD 2013 for Pb, Po, Ra, Th, and U), with most of the remaining 8% sourced from Australia.

In Fig. 5, the WTD 2013 means (arithmetic) have been recalculated with the Australian data removed, which has allowed the mean Australian and non-Australian $CRs_{wo\text{-}media}$ to be compared. In

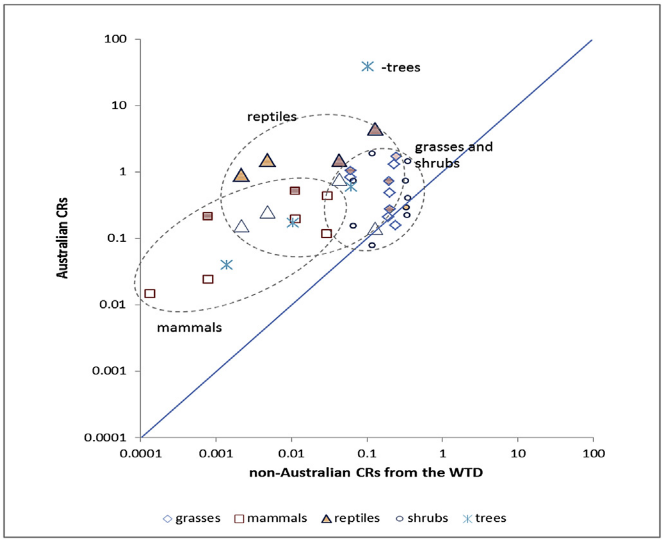

Fig. 5. Terrestrial Australian $CR_{wo\text{-}media}$ values versus non-Australian $CR_{wo\text{-}media}$ values from the WTD 2013. Values used are arithmetic mean $CR_{wo\text{-}media}$ for Pb, Po, Ra, Th, and U radionuclides for the wildlife categories indicated. Closed-pattern symbols represent acidic mine-tailing conditions.

this comparison, most (83%) of the Australian data plot above the 1:1 line, with 45% of the Australian mean $CRs_{wo\text{-}media}$ greater than one order of magnitude above the non-Australian $CRs_{wo\text{-}media}$. The Australian $CR_{wo\text{-}media}$ values for grasses and mammals were significantly higher than non-Australian $CRs_{wo\text{-}media}$ (Mann–Whitney *U* test, two-tailed, at $p < 0.01$), although the values for shrubs were not. For this *U* test comparison, data were insufficient for trees and reptiles (the test did not include the mine-tailings data, which is generally elevated further (see Fig. 4 for example)). With or without the mine-tailings data, the apparent elevation of many Australian $CR_{wo\text{-}media}$ data suggests that use of site-specific, regional, or Australian-specific data is appropriate and beneficial at Australian sites where dose rates approach benchmarks and a more thorough evaluation is needed.

In addition to reflecting climate/environmental exposure conditions, the uptake of actinides in wildlife is known to vary according to its physico-chemical form (ICRP, 1986; Kohen and Limbach, 2005), which, in environmental systems, is related to its source and the manner of its dispersal (Salbu, 2001). Most of the data reported here represent naturally occurring forms, typically mineral soils that have been highly weathered over long time periods in environmental conditions. The elevated data from the acidic mine tailings appear to be influenced by speciation changes (e.g., increased mobility of U when oxidized from U(IV) to U(VI) in the presence of sulfidic minerals). Additional data were obtained from the former Taranaki nuclear weapons test site at Maralinga, South Australia, where the soil includes natural U, as well as contamination of processed (enriched) U that was dispersed during high-explosive (non-nuclear) events (Ikeda-Ohno et al., 2016). The Taranaki U $CR_{wo\text{-}media}$ values (2.0E-3 to 4.0E-3) for *Oryctolagus cuniculus* (European rabbit) were similar to mammal CRs from the Ranger mine, but two orders of magnitude lower than the acidic mine tailings values (Table 1). This suggests that the processing and release effects on the weapons U has not led to the elevated transfer to mammals (as seen at the acidic mine tailing sites). Such processing and release effects have been seen to impact uptake for other actinides at Maralinga, as well as other sites (e,g, plutonium; Johansen et al., 2016, Johansen et al. 2014). Further study of sites involving a range of processing and release conditions would be necessary for more complete comparison of the influence of physico-chemical form of U on transfer to wildlife.

3.4. Data gaps

There were no marine data available from Australia related to NORM extraction activities relevant to U or Th and their decay products in NORM scale issues associated with subsea oil and gas extraction. Drainage from some Australian mining sites in coastal rivers has contamination potential extending to coastal areas (e.g. Finniss River contaminations from Rum Jungle mine (Davy and O'Brien, 1975)). In Australia, some NORM producing industries (e.g. gas and petroleum extraction) have been subject to radiological environmental assessments as part of environmental planning approval processes (for example PTTEP Australasia, 2014). These assessments have largely relied upon generic transfer data that are based on a small number of environmental measurements from different environmental settings. As a substantial amount of subsea gas and oil extraction in Australian coastal shelf waters has the tendency for NORM-scale to accrete on subsea infrastructure followed by the need for decommissioning and disposal, there exists an emerging and growing need for parameters on NORM transfer to marine organisms in Australian waters.

4. Conclusion

The study resulted in 271 new or revised $CR_{wo\text{-}media}$ values from Australia covering terrestrial and freshwater wildlife groups that are now available for use in assessing radiological transfer at U mining sites and potentially other NORM-contaminated environments. In comparing with the WTD 2013 mean values (which include the Australian $CRs_{wo\text{-}media}$), the general Australian data did not present significant outliers, suggesting that the WTD summarised values are generally appropriate for use in screening level assessments within Australia in the absence of any site-specific data.

However, in this study we separated the Australian $CR_{wo\text{-}media}$ data from the non-Australian data, predominantly from temperate climates, for the same WTD categories, and found that most of the Australian $CRs_{wo\text{-}media}$ (83%) were higher than the non-Australian data, with significant differences in most cases where data was sufficient to allow comparison. In this paper, we report an additional $CR_{wo\text{-}media}$ data set representing mine-tailings where acidic conditions likely increase radionuclide mobilisation (Read and Pickering, 1999), and lead to elevated $CRs_{wo\text{-}media}$ in most categories. When these mine-tailing $CRs_{wo\text{-}media}$ were included, 45% of the mean (arithmetic) Australian $CRs_{wo\text{-}media}$ were elevated more than one order of magnitude above the non-Australian mean CRs. This apparent elevation of many Australian $CR_{wo\text{-}media}$ data suggests that the use of site-specific, regional, or Australian-specific data is required at Australian sites where dose rates approach benchmarks, or in instances when a thorough evaluation is appropriate. This agrees with the recommendation made by Wood et al. (2013): that summarised $CR_{wo\text{-}media}$ values are to be used with caution above screening level assessments given their inherent uncertainty.

Gaps in the Australian datasets remain with respect to wildlife groups as presented in the WTD 2013; for freshwater environments, there are no data for phytoplankton, zooplankton, insects, insect larvae or amphibians and, for terrestrial environments, there are no data for amphibians, annelids, ferns, fungi, lichens & bryophytes. There were no marine data available from Australia related to gas and NORM extraction activities and we recommend such data are required. These gaps reflect that most of the existing data had been collected in support of human ingestion dose assessments rather than for assessing impacts on the environment.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2017.04.007.

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