Journal of Environmental Radioactivity 162-163 (2016) 154–159

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Short communication

A database of radionuclide activity and metal concentrations for the Alligator Rivers Region uranium province

Che Doering [a,*], Andreas Bollhöfer [a,b]

[a] Environmental Research Institute of the Supervising Scientist (ERISS), GPO Box 461, Darwin, NT 0801, Australia
[b] Federal Office for Radiation Protection (Bundesamt für Strahlenschutz, BfS), 79098, Freiburg, Germany

ARTICLE INFO

Article history:
Received 1 March 2016
Received in revised form
26 May 2016
Accepted 27 May 2016

Keywords:
Radionuclides
Metals
Bush food
Wildlife
Database
Tropical radioecology

ABSTRACT

This paper presents a database of radionuclide activity and metal concentrations for the Alligator Rivers Region (ARR) uranium province in the Australian wet-dry tropics. The database contains 5060 sample records and 57,473 concentration values. The data are for animal, plant, soil, sediment and water samples collected by the Environmental Research Institute of the Supervising Scientist (ERISS) as part of its statutory role to undertake research and monitoring into the impacts of uranium mining on the environment of the ARR. Concentration values are provided in the database for 11 radionuclides (^{227}Ac, ^{40}K, ^{210}Pb, ^{210}Po, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, ^{238}U) and 26 metals (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Na, Ni, P, Pb, Rb, S, Sb, Se, Sr, Th, U, V, Zn). Potential uses of the database are discussed.

Crown Copyright © 2016 Published by Elsevier Ltd. All rights reserved.

1. Introduction

Environmental assessments of uranium mining sites can be data intensive because of the potentially large number of contaminant-pathway combinations that need to be characterised in order to evaluate exposure risks to people and the environment. This is the case for Ranger uranium mine in the Alligator Rivers Region (ARR) of northern Australia where, for more than three decades, the Environmental Research Institute of the Supervising Scientist (ERISS) has been undertaking research to generate data and independently assess the environmental impacts of uranium mining in the region (Van Dam et al., 2002). Data generated by ERISS on radionuclide activity and metal concentrations in various environmental compartments have recently been compiled into a database to facilitate their storage and analysis. This paper describes the development of the database and discusses some of its potential uses.

2. Regional context

The ARR is located in the wet-dry tropics of northern Australia (Fig. 1). It is an area rich in Aboriginal culture, biological diversity and uranium mineral deposits. Most of the ARR is Aboriginal owned land and around two thirds of it is World Heritage listed as Kakadu National Park. Ranger uranium mine is located in the ARR. It is Australia's largest uranium producer and one of the top uranium-producing mines worldwide. The mine is surrounded by Kakadu National Park but is technically not part of it. Mining at Ranger began in 1980 and is scheduled to cease by 2021. The site is to be remediated by 2026 such that it could be incorporated into Kakadu National Park (Commonwealth of Australia (1999)).

The statutory position of Supervising Scientist and an associated research institute (i.e. ERISS) were established in 1978 following an Australian Government inquiry into the environmental concerns surrounding uranium mining in the ARR (Fox et al., 1977). The Supervising Scientist is responsible for the environmental oversight of uranium mining in the ARR, and ERISS undertakes research into its environmental impacts. An important part of the research by ERISS has been to collect Aboriginal bush foods[1] from the environment and analyse them for radionuclides and metals. The initial reason for this has been to assess potential exposures to Aboriginal people from dietary intakes of radionuclides and metals, and

* Corresponding author.
E-mail address: che.doering@environment.gov.au (C. Doering).

[1] Bush foods are the edible tissues of wild plants and animals which are traditionally hunted and gathered by Aboriginal people for sustenance.

http://dx.doi.org/10.1016/j.jenvrad.2016.05.029
0265-931X/Crown Copyright © 2016 Published by Elsevier Ltd. All rights reserved.

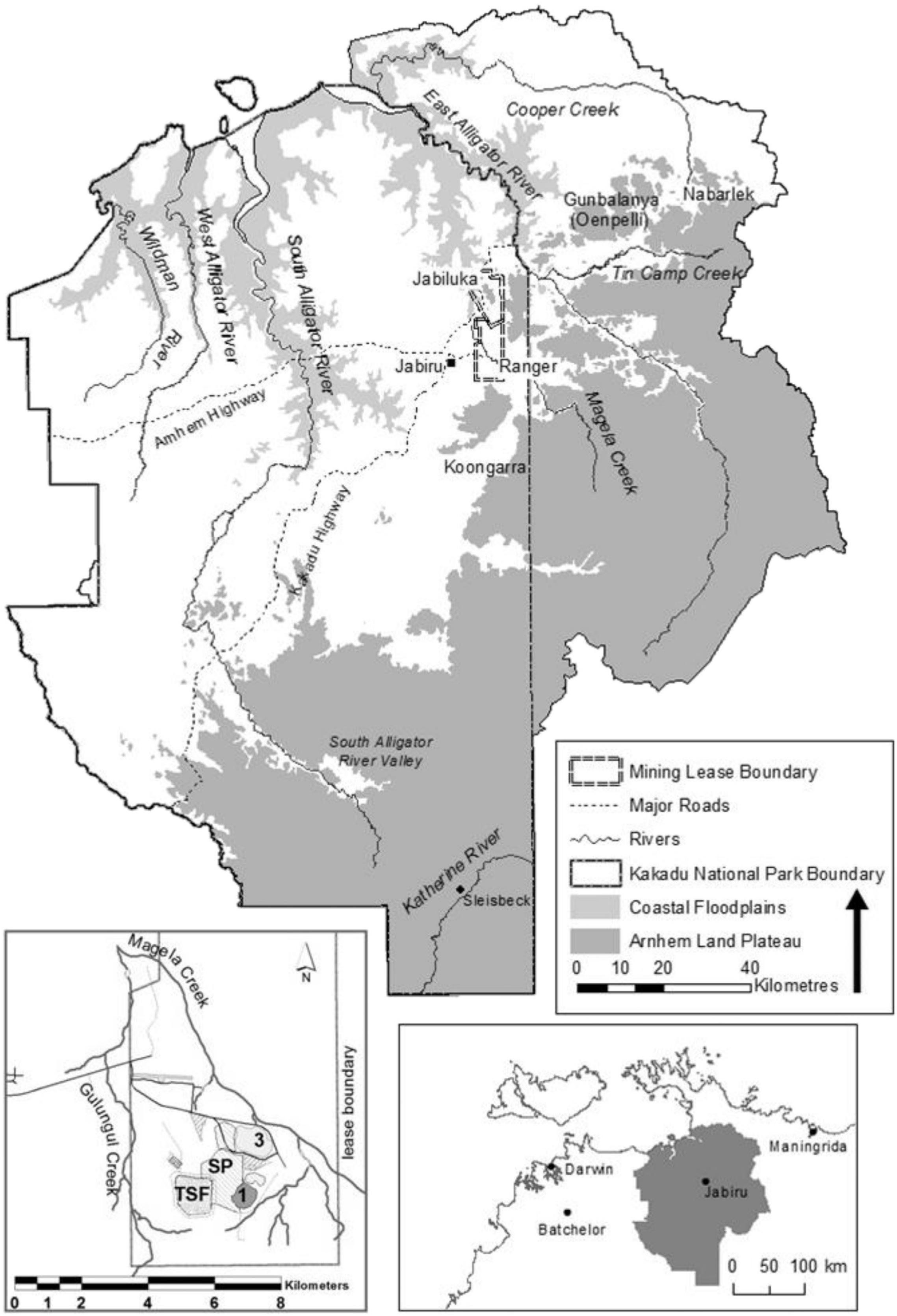

Fig. 1. Alligator Rivers Region map showing Kakadu National Park and the location of Ranger uranium mine. The inset at the bottom left shows an expanded view of the mine and surrounding creeks (TSF = tailings storage facility, SP = stockpiles, 1 = pit 1, 3 = pit 3). The inset at the bottom right shows the location of the Alligator Rivers Region in the broader geographical context of Australia's 'Top End'.

provide assurance that bush foods sourced from Kakadu National Park and its wetlands remain fit for consumption, particularly in areas downstream of uranium mining sites. More recently, the research has been broadened to also collect and analyse biota tissue samples not usually eaten as bush foods, but which could be of potential importance to estimate exposures to wildlife.

3. Methods

3.1. Data compilation and conversions

The compilation of data focussed on information for single samples and did not include summary statistics. Data were compiled from reports and files of various formats produced by ERISS and collaborating scientists over many years. The data were entered into a MS Excel™ database to facilitate conversions and quality control processes. Table 1 describes the data entry fields.

The data were converted to a standard format in the database. All concentration values for biota were converted to a fresh mass basis with units of Bq kg^{-1} (radionuclides) or mg kg^{-1} (metals). The dry mass fraction of the sample was used to convert concentration values originally given on a dry mass basis. If the sample dry mass fraction was missing, then tissue arithmetic means calculated by wildlife group were used. Sediment and soil concentration values were entered on a dry mass basis with units converted to Bq kg^{-1} (radionuclides) or mg kg^{-1} (metals). Water concentration values were converted to units of Bq l^{-1} (radionuclides) or mg l^{-1} (metals).

Uncertainties have been included for radionuclide activity concentration values and generally represent one standard deviation based on counting statistics from alpha or gamma spectrometric measurement. No uncertainties were available for metal concentration values, but are estimated to be no more than about 10% based on the measurement methods used, typically ICP-MS. Radionuclide activity and metal concentration values originally reported as less than the detection limit have been included in the database and are preceded by the '<' symbol.

Radionuclide measurements were made by ERISS following standard methods developed for alpha spectrometry (Martin and Hancock, 2004; Medley et al., 2005) and gamma spectrometry (Murray et al., 1987; Marten, 1992; Esparon and Pfitzner, 2010). Detectors were routinely calibrated using standard reference materials, and detector backgrounds were also measured routinely. Metals measurements were made by external service providers whose quality control procedures included measurement of analytical blanks and standard materials with each batch of samples. Replicate samples were also generally included with each batch analysed to check on the reproducibility of results.

3.2. Quality control

Primary quality control of the compiled data was done by visual methods. Time-series plots of concentration values for each radionuclide and metal were generated by sample type and location to identify unrealistic values and obvious errors in transcription or unit conversions. These plots were also used to check that collection dates were realistic (i.e. within the time period from ~1980 to 2015). Sample geospatial coordinates were plotted in Google Earth™ to ensure that all locations were in the correct geographical region. Any issues identified were investigated and corrected.

4. Results

The database is included in the supplementary material in MS Excel™ file format and contains all reference details of where the data have been published, summarised or described. These

Table 1
Parameters included in the database.

Parameter
Database ID
Sample ID
Ecosystem (Freshwater or Terrestrial)
Sample type (Animal, Plant, Sediment, Soil or Water)
Common name (English)
Scientific name (Latin)
Bush food group (categorisation based on the Aboriginal model diet developed by Ryan et al. (2008a))
Wildlife group (categorisation based on the Wildlife Transfer Database (Copplestone et al., 2013) and used by IAEA (2014))
Compartment (specific tissue or media fraction)
Composite count (number of individuals pooled to form single sample for analysis)
Dry mass fraction
Sex
Mussel age (y) (age of freshwater mussels; included as the concentration of some radionuclides (e.g. ^{226}Ra) and metals (e.g. Pb) is age dependent (Bollhöfer, 2012; Bollhöfer et al., 2011))
Soil depth (cm)
Family code (identifier for linking different compartments from the same parent sample)
Collection date (day/month/year)
Location
Status ('Perturbed' indicates the location was significantly impacted by uranium mining or is an area with high natural background; 'Unperturbed' indicates it is not)
Easting
Northing
Zone (52S or 53S)
Datum (WGS84)
Geocoding (1 = coordinates given in reference; 2 = coordinates estimated in Google Earth™ from map or location description given in reference)
Radionuclide activity concentrations (^{227}Ac, ^{40}K, ^{210}Pb, ^{210}Po[a], ^{226}Ra, ^{228}Ra, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U and/or ^{238}U in units of Bq kg^{-1} fresh mass (biota), Bq kg^{-1} dry mass (soil and sediment) and Bq l^{-1} (water))
Metal concentrations (sAl, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, Mg, Mn, Na, Ni, P, Pb, Rb, S, Sb, Se, Sr, Th, U, V and/or Zn in units of mg kg^{-1} fresh mass (biota), mg kg^{-1} dry mass (soil and sediment) and mg l^{-1} (water))
Reference
Notes

[a] Only values corrected for ingrowth from the decay of ^{210}Pb have been entered in the database.

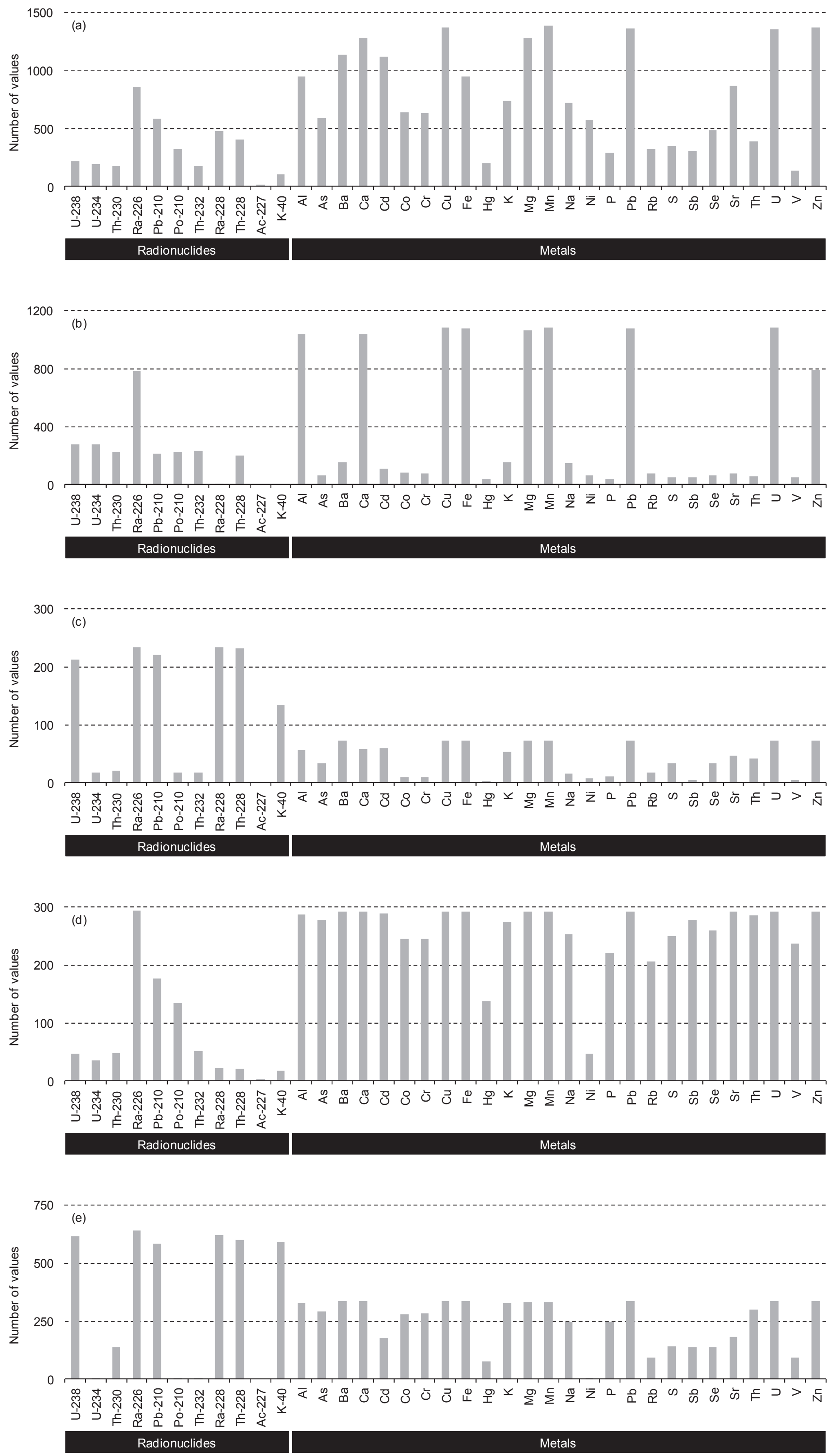

Fig. 2. Snapshots of data availability: (a) freshwater biota; (b) water; (c) sediment; (d) terrestrial biota; (e) soil.

reference details are not repeated in this paper.

The database contains 5060 sample records split between the terrestrial and freshwater ecosystems as follows: terrestrial biota (n = 339); soil (n = 800); freshwater biota (n = 1815); water (n = 1859); and sediment (n = 247). There were considerably more sample records for the freshwater ecosystem than the terrestrial ecosystem. The aquatic pathway has previously been identified as the most important pathway potentially affecting ecosystem health and exposures to people during the operational phase of mining in the ARR (Martin, 2000; Martin et al., 1998; van Dam et al., 2002), with the freshwater ecosystem being more heavily sampled as a consequence. Fish (n = 1084) and freshwater mussels (n = 553) in particular have been the focus of routine bioaccumulation monitoring programs (see, e.g., Brazier et al., 2009). The large number of sample records for water reflects the comprehensive water quality monitoring program conducted by ERISS in the creeks surrounding Ranger uranium mine during the wet season. The number of sample records available for the terrestrial ecosystem was limited by the opportunistic nature of sampling by ERISS. Sampling of terrestrial biota has primarily involved taking tissues from fresh roadkill and feral animals culled by Kakadu National Park rangers, and also accompanying samples from Aboriginal people caught on the occasional hunting and gathering trip. Soil samples were collected with biota samples and also as part of environmental characterisation studies.

The database has 57,473 concentration values across 11 radionuclides and 26 metals. Fig. 2 gives snapshots of data availability by ecosystem. Radionuclide data were most numerous for ^{226}Ra across both ecosystems and all sample types. Previous estimates of radiation doses to Aboriginal people resulting from a potential release of wastewaters from Ranger uranium mine have shown that ^{226}Ra in bush foods sourced from the freshwater ecosystem, especially freshwater mussels, would be the dominant exposure pathway (Martin, 2000; Martin et al., 1998). The current operational guideline value for radiological water quality downstream of Ranger uranium mine is consequently based on ^{226}Ra (Bollhöfer et al., 2016). Metal data for biota samples (freshwater and terrestrial) were relatively numerous for most elements as these samples were usually analysed for a standard set of metals. Metal data for water samples were most numerous for key water quality indicators (Al, Ca, Cu, Fe, Mg, Mn, Pb, U and Zn) routinely measured in creek monitoring samples during the wet season. Soil metal data were relatively numerous for most elements.

A small number of database records belonged to samples collected outside the ARR, but still within the same wet-dry tropical climate area of northern Australia. These included samples from semi-natural environments in Darwin, from remediated uranium mining sites at Rum Jungle near the township of Batchelor and from natural environments close to Maningrida in central Arnhem Land (Fig. 1). The species collected from these sites were the same as those found in the ARR, and the data were considered to be of general relevance to the ARR.

The volume of data contained in the database is too large to attempt any analyses of them in this paper. However, some thoughts and examples on possible uses are discussed below.

5. Discussion

The database represents a significant data resource for the ARR and the north Australian tropics in general and, to our knowledge, is the largest of its kind for tropical environments worldwide. It has been developed to put the radionuclide activity and metal concentration data of ERISS into a standard format, and to facilitate their quick access and analysis. Most of the individual sample data included in the database have previously not been published or made available elsewhere.

We envisage that the database will be heavily used to derive radionuclide and metal concentration ratios for bush foods and wildlife for use in environmental assessments of Ranger uranium mine, especially in the lead up to its decommissioning and remediation when closure criteria will need to be developed. In fact such assessments using concentration ratios derived from the database have already started (Doering and Bollhöfer, 2016).

A number of previous studies by ERISS have reported radionuclide activity concentrations and concentration ratios for bush foods (e.g. Martin et al., 1998; Ryan et al., 2005) and freshwater mussels in particular (Bollhöfer et al., 2011; Ryan et al., 2008b). However, the database can provide improved estimates of these parameters because of more sample data and the possibility for integrated analysis across multiple studies due to the general consistency over time of the measurement techniques used by ERISS. There are now also data available for a broader range of analytes, species and tissue types than before.

The data also have relevance to other research applications. For instance, the influence of group II metals on ^{226}Ra uptake in wild fruits from the ARR has been investigated using the data (Medley et al., 2013; Medley and Bollhöfer, 2016). We hope that including the compiled data in the supplementary material to this paper also encourages their broader use, such as within international data compilations and for model testing.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2016.05.029.

References

Bollhöfer, A., 2012. Stable lead isotope ratios and metals in freshwater mussels from a uranium mining environment in Australia's wet-dry tropics. Appl. Geochem. 27, 171–185.
Bollhöfer, A., Brazier, J., Humphrey, C., Ryan, B., Esparon, A., 2011. A study of radium bioaccumulation in freshwater mussels, *Velesunio angasi*, in the Magela Creek catchment, Northern Territory, Australia. J. Environ. Radioact. 102, 964–974.
Bollhöfer, A., Doering, C., Medley, P., 2016. A Revised Operational Limit for the Water 226Ra Activity Concentration Downstream of Ranger Mine. Internal Report 643. Supervising Scientist, Darwin (Unpublished paper).
Brazier, J., Bollhöfer, A., Ryan, B., Humphrey, C., 2009. Bioaccumulation in fish and freshwater mussels from Mudginberri Billabong. In: ERISS Research Summary 2007–2008. Supervising Scientist Report 200. Supervising Scientist, Darwin, pp. 53–56.
Commonwealth of Australia, 1999. Environmental Requirements of the Commonwealth of Australia for the Operation of Ranger Uranium Mine. Available from: www.environment.gov.au.
Copplestone, D., Beresford, N.A., Brown, J.E., Yankovich, T., 2013. An international database of radionuclide concentration ratios for wildlife: development and uses. J. Environ. Radioact. 126, 288–298.
Doering, C., Bollhöfer, A., 2016. A soil radiological quality guideline value for wildlife-based protection in uranium mine rehabilitation. J. Environ. Radioact. 151, 522–529.
Esparon, A., Pfitzner, J., 2010. Visual Gamma 2007, Gamma Analysis Manual. Internal Report 539. Supervising Scientist, Darwin.
Fox, R.W., Kelleher, G.G., Kerr, C.B., 1977. Ranger uranium Environmental Inquiry Second Report. Australian Government publishing service, Canberra.
IAEA, 2014. Handbook of Parameter Values for the Prediction of Radionuclide Transfer to Wildlife. Technical Report Series No. 479. International Atomic Energy Agency, Vienna.
Marten, R., 1992. Procedures for Routine Analysis of Naturally Occurring Radionuclides in Environmental Samples by Gamma-ray Spectrometry with HPGe Detectors. Internal Report 76, Supervising Scientist for the Alligator Rivers Region. Canberra.
Martin, P., 2000. Radiological Impact Assessment of Uranium Mining and Milling. PhD Thesis. Queensland University of Technology, Brisbane.
Martin, P., Hancock, G.J., 2004. Routine Analysis of Naturally Occurring Radionuclides in Environmental Samples by Alpha-particle Spectrometry. Supervising Scientist Report 180. Supervising Scientist, Darwin.
Martin, P., Hancock, G.J., Johnston, A., Murray, A.S., 1998. Natural-series radionuclides in traditional north Australian aboriginal foods. J. Environ. Radioact. 40, 37–58.
Medley, P., Bollhöfer, A., Iles, M., Ryan, B., Martin, P., 2005. Barium Sulphate Method

for 226Ra Analysis by Alpha Spectrometry. Internal Report 501. Supervising Scientist, Darwin.

Medley, P., Bollhöfer, A., Parry, D., Martin, P., 2013. Radium concentration factors in passionfruit (*Passiflora foetida*) from the Alligator Rivers region, northern territory, Australia. J. Environ. Radioact. 126, 137–146.

Medley, P., Bollhöfer, A., 2016. Influence of group II metals on Radium-226 concentration ratios in the native green plum (*Buchanania obovata*) from the Alligator Rivers Region, Northern Territory, Australia. J. Environ. Radioact. 151, 551–557.

Murray, A.S., Marten, R., Johnston, A., Martin, P., 1987. Analysis for naturally occurring radionuclides at environmental concentrations by gamma spectrometry. J. Radioanalytical Nucl. Chem. 115, 263–288.

Ryan, B., Martin, P., Iles, M., 2005. Uranium-series radionuclides in native fruits and vegetables of northern Australia. J. Radioanalytical Nucl. Chem. 264 (2), 407–412.

Ryan, B., Medley, P., Bollhöfer, A., 2008a. Bioaccumulation of radionuclides in terrestrial plants on rehabilitated landforms. In: Jones, D.R., Humphrey, C., van Dam, R., Webb, A. (Eds.), Eriss Research Summary 2006–2007. Supervising Scientist, Darwin NT, pp. 99–103. Supervising Scientist Report 196.

Ryan, B., Bollhöfer, A., Martin, P., 2008b. Radionuclides and metals in freshwater mussels of the upper South Alligator River, Australia. J. Environ. Radioact. 99, 509–526.

Van Dam, R.A., Humphrey, C.L., Martin, P., 2002. Mining in the Alligator Rivers region, northern Australia: assessing potential and actual effects on ecosystem and human health. Toxicity 181–182, 505–515.