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A revised operational limit for the water ²²⁶Ra activity concentration downstream of Ranger mine

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Peter Medley

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Executive summary

The current trigger value for the above background water radium-226 (²²⁶Ra) activity concentration at the Magela Creek compliance site is 10 mBq·L⁻¹ (in the total fraction). This has been derived from human radiation protection considerations, based on the potential for bioaccumulation of ²²⁶Ra in mussels downstream of the Ranger mine and subsequent ingestion by humans. The potential for radiological impact on the environment has not been considered in previous derivations of trigger values for ²²⁶Ra. The aim of this report is to provide a review of the trigger value for ²²⁶Ra, including assessment of the potential impacts on human health and also taking into consideration the potential radiological impacts on the environment.

The current ²²⁶Ra trigger value for Magela Creek is based on data collected and assessed in the 1980s and, as the major source of mine related doses to people from the ingestion of traditional food items, with new data available it was considered timely to conduct this review into the appropriateness of the current value. Data from more than ten years (2001-2013) of bioaccumulation monitoring in Mudginberri Billabong mussels are statistically evaluated and concentration ratios for Mudginberri Billabong mussels are calculated and used in subsequent assessments of radiation doses to human and the environment.

Our assessment is based on the above background total water ²²⁶Ra activity concentration in Magela Creek. It assumes that a 10 year old child consumes 2 kg (wet weight) of large mussels per year. The resulting ingestion dose to humans, as well as internal and external doses to mussels from ²²⁶Ra are assessed. It is shown that human radiological protection is more limiting on total water ²²⁶Ra activity concentration in Magela Creek than environmental radiation protection considerations.

A mine origin increase in total water ²²⁶Ra activity concentration of less than 3 mBq·L⁻¹ will lead to a mine origin ingestion dose to humans of less than 0.2 mSv per year. The same increase in total water ²²⁶Ra activity concentration of 3 mBq·L⁻¹, will lead to a maximum additional dose rate to a small number of mussels (1%) of 50 μ Gy·h⁻¹. This is much lower than the 400 μ Gy·h⁻¹ benchmark dose rate, which, according to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), would not have any detrimental effect at the population level in an aquatic environment.

Thus, the operational limit for the above background water total ²²⁶Ra activity concentration downstream of Ranger mine should be revised and set at 3 mBq·L⁻¹.

1 Introduction

The Ranger uranium mine (RUM) is located in the Alligator Rivers Region (ARR) in the wet-dry tropics of Australia's Northern Territory (Figure 1). It is surrounded by, though technically separate from, the World Heritage listed Kakadu National Park (KNP).

The mine is located along Magela Creek, which is part of the East Alligator River system. The headwaters of Magela Creek are located on the Arnhem Land Plateau. It flows through Bowerbird Billabong, a channel rock pool billabong near the headwaters of Magela Creek (Walker and Tyler, 1982) upstream of the mine. The creek continues its path through the lowlands within the Ranger mineral lease and then on to Mudginberri Billabong, a permanent channel billabong 12 km downstream from RUM and within KNP (Bollhöfer 2012) (Figure 1). Finally, the creek flows through the Magela Corridor and drains into the South Alligator floodplain which flows into Van Diemen Gulf.



Figure 1 Location of Ranger mine and Supervising Scientist Branch sampling sites along the Magela Creek channel. BBB, Bowerbird Billabong; MCUS, Magela Creek upstream; GTC, Georgetown Creek; MCDS, Magela Creek downstream; MBB, Mudginberri Billabong. GTB and CJB are Georgetown and Coonjimba Billabongs (from Supervising Scientist, 2008).

The climate of the ARR is characterised by distinct wet (November to April) and dry (May to October) seasons. The average annual rainfall over the past 100 years has been 1422 mm at Jabiru East (BoM 2015). More than 95% of rainfall occurs in the wet season and approximately two thirds occurs between January and March with individual rainfall events as high as 784 mm over 72 hours (Suradi et al 2014). It is these distinct seasons

and high rainfall intensities during the first three months of the year that lead to the requirement of intense water quality monitoring downstream of RUM.

Monitoring is focussed on Magela and Gulungul Creeks as both creeks receive inputs from areas within the RUM mineral lease, including:

- Runoff from the walls of the tailings storage facility (TSF)
- Runoff from land application areas (LAAs)
- Waste waters from retention ponds (in particular Retention Pond 1)
- Through the groundwater pathway (in particular Mn, see Iles 2004).

Water quality objectives have been set for key variables (Klessa 2001a & b) and were reviewed by Iles (2004). Whereas the water quality objectives for pH and turbidity have been derived from the reference site distribution of these parameters upstream of the mine site, for uranium, magnesium and manganese the recommended trigger values are based on ecotoxicity testing (Iles 2004; Hogan et al 2003; Sinclair & Tayler 2012, Harford et al 2014).

For radium-226 (²²⁶Ra), the trigger value is based on human radiation protection considerations (Klessa 2001b), based on the potential for bioaccumulation of ²²⁶Ra in mussels of Mudginberri Billabong downstream of RUM and subsequent ingestion by humans. The potential for radiological impact on the environment has not been considered in previous derivations of trigger values for ²²⁶Ra in Magela creek waters. The aim of this report is to provide a review of the trigger value for ²²⁶Ra, including assessment of the potential impacts on human health and also taking into consideration the potential radiological impacts on the environment.

1.1 Radium

Radium belongs to the alkaline earth metals (Group IIA) in the Periodic Table. The four radium isotopes occurring naturally in the environment are radium-226 (²²⁶Ra; t_{1/2}=1600 yr) of the uranium (²³⁸U) series (Figure 2), radium-228 (²²⁸Ra; t_{1/2}=5.75 yr) and radium 224 (²²⁴Ra; t_{1/2}=3.66 d) of the thorium (²³²Th) series and radium-223 (²²³Ra; t_{1/2}=11.8 d) of the actinium (²³⁵U) series. Radium and its radioactive properties were discovered in 1898 by Pierre and Marie Curie together with the discovery of another radioactive element, polonium (Curie et al 1898). Radium was hailed as the wonder drug for 'medical conditions with no known cure', and it took more than a decade to realise its potentially harmful properties (IAEA 1990). In the 1950s, it was Tsivoglou (1958) who first identified radium as a significant pollutant from uranium mining and milling activities because the longest lived natural radium isotope, ²²⁶Ra, is a member of the uranium decay series (Figure 2) and uranium mining residues generally still contain considerable activities of ²³⁰Th and its radioactive decay products, including ²²⁶Ra. A significant proportion of the radiation dose derived from ²²⁶Ra is also related to its short-lived decay products.

Radium-226 and these shorter-lived decay products contribute to radiation dose to humans and the environment via three principal pathways. Firstly, the short lived ²²⁶Ra decay products bismuth-214 (²¹⁴Bi) and lead-214 (²¹⁴Pb) emit γ -radiation, which can lead to external radiation exposure. Although ²³⁸U, ²²⁶Ra and their decay products emit characteristic gamma radiation, Monte Carlo modelling by Saito and Jacobs (1995) identified that more than 98% of the gamma dose field over an area containing ²³⁸U in

equilibrium with its progeny is due to the γ -emissions from short-lived ²¹⁴Bi and ²¹⁴Pb, whereas the contribution of ²³⁸U and ²²⁶Ra to the external γ -exposure is negligible.

Radium-226 is also the radioactive predecessor of the radioactive noble gas radon-222 (222 Rn; t_{1/2}=3.8 days; Figure 2), which can emanate from soil grains upon disintegration of 226 Ra, diffuse through the soil profile, exhale at the soil-atmosphere interface and mix in the atmosphere. Radon-222 then further decays to polonium-218 (218 Po), 214 Bi, polonium-214 (214 Po) and 214 Pb. The inhalation of these radon decay products can deliver a significant dose to the soft tissue of the lungs (Kendall-Smith 2002).



Figure 2 The uranium-238 (²³⁸U) decay series (from Martin 2000)

Finally, ²²⁶Ra and the other natural radium isotopes can be ingested by humans and wildlife with food and water. Radium is chemically similar to Ca, Mg, Ba and Sr and it is taken up as an analogue element to the essential elements Ca and Mg in plants (Medley et al 2013; Medley & Bollhöfer 2015) and animals, such as freshwater mussels (Johnston et al 1987; Bollhöfer et al, 2011). Plants and animals (including humans) transport the alkaline earth metals through similar biochemical channels and physiologically, they do not discriminate between them (Ivanovich and Harmon, 1982). The concentration of Ca and Mg in soil, sediment or water however can influence the magnitude of Ra uptake in plants (Medley & Bollhöfer 2015; Medley et al 2013; Bollhöfer et al 2011). In vascular plants for example, uptake of group II metals is exclusively via the roots (Strebl et al., 2007) and although the group II metals follow the same uptake pathway, Ca (and Mg) are preferentially taken up. Uptake in plants is discriminated against group II metals relative to their ionic radius with uptake of Ca(Mg)>Sr>Ba>>²²⁶Ra (Tagami et al., 2012; Medley & Bollhöfer 2015).

Uptake of ²²⁶Ra in animals also follows the transport pathways of Ca and Mg. In aquatic organisms, the lower trophic organisms (e.g. phytoplankton) display stronger accumulation trends than the higher trophic organisms (Vandenhove et al 2010). Certain hard tissues such as bones and shells accumulate relatively more ²²⁶Ra due to their higher Ca concentration compared to other tissues. This preferential uptake can create strong bioaccumulation of ²²⁶Ra in specific tissues and can lead to large differences in ²²⁶Ra activity concentrations in different parts of an organism (Ellis & Jeffree 1982). Mussels

for example bioaccumulate ²²⁶Ra in calcium phosphate granules in their soft tissue and ²²⁶Ra activity concentrations in these calcium phosphate granules is extremely high (Ellis & Jeffree 1982). The influence of group II metals on ²²⁶Ra uptake in mussels is such that if Ca and Mg concentrations in the (aquatic) environment are low, ²²⁶Ra uptake generally increases. For example, Brenner et al (2007) have reported much lower uptake of ²²⁶Ra for freshwater mussels in a groundwater augmented lake with high Ca concentrations, in contrast, uptake is particularly high from the low EC waters of Magela Creek (Bollhöfer et al 2011).

In mammals (and humans), most ingested ²²⁶Ra is promptly excreted again, however, some enters the bloodstream and is incorporated into bones. Radium-226 follows biochemical pathways as an analogue to Ca and Mg and can isomorphously substitute for Ca in the apatite mineral structure of bone tissue (Vandenhove et al 2010). Because of its preference for bone, Ra is commonly referred to as a bone seeker (National Research Council 1988). Once deposited in bones, ²²⁶Ra decays and delivers, either directly or via the short-lived daughter isotopes, a radiation dose to bone tissue. The ingestion of mussels with high concentrations of ²²⁶Ra can thus lead to significant radiation doses to humans from ²²⁶Ra.

The high radiotoxicity of radium to humans upon ingestion is evidenced by the high dose conversion coefficient (DCC) for ²²⁶Ra of 0.28 μ Sv·Bq⁻¹ for adults and 0.8 μ Sv·Bq⁻¹ for a 10 year old child. Only ²¹⁰Po and ²¹⁰Pb have higher DCCs than ²²⁶Ra within the elements of the uranium series decay chain (1.2 μ Sv·Bq⁻¹ for an adult and 2.6 μ Sv·Bq⁻¹ for a child for ²¹⁰Po; 0.69 μ Sv·Bq⁻¹ for an adult and 1.9 μ Sv·Bq⁻¹ for a child for ²¹⁰Pb).

Both ²¹⁰Po and ²¹⁰Pb also have the potential to bioaccumulate in animal tissues, however, in the freshwater environment they are less mobile than ²²⁶Ra and have a stronger affinity for suspended particles (Alam & Mohamed 2010); both isotopes are quickly adsorbed on suspended sediment and removed from the water column into bottom sediments. Martin et al (1998) for example report ²¹⁰Po and ²¹⁰Pb activity concentrations in Retention Pond 2 (RP2) water more than 100 times lower than the activity concentration of ²²⁶Ra. This is similar to the ratios reported in Johnston et al (2005) where ²¹⁰Po and ²¹⁰Pb activity concentrations in RP2 water are assumed to be approximately 70 times lower than ²²⁶Ra. The ²¹⁰Po and ²¹⁰Pb activity concentration in Mudginberri Billabong measured during that time period (Martin et al 1998) confirming fast removal of these two radioisotopes from the water column into the sediment of RP1. Total (*filtered*) activity concentrations for ²²⁶Ra, ²¹⁰Po and ²¹⁰Pb in Mudginberri Billabong in the 1980s have been reported by Martin et al (1998) as 3.4 (0.7) mBq·L⁻¹, 4.5 (1.6) mBq·L⁻¹ and 6.3 (1.3) mBq·L⁻¹ respectively.

The activity concentrations of ²¹⁰Po and ²¹⁰Pb in mine waters is much lower than that of ²²⁶Ra and the contribution (if any) of ²²⁶Ra to the downstream environment from RUM is very low (Supervising Scientist 2015). Thus, although not directly measured, it can be assumed that mine origin ²¹⁰Po and ²¹⁰Pb do not contribute to the presence of these isotopes in water downstream of RUM and consequently do not contribute to the activity concentrations of these two radionuclides in mussels. This assumption is confirmed by the results of measurements of stable lead (²⁰⁶Pb, ²⁰⁷Pb, ²⁰⁸Pb) isotope ratios in mussel flesh, which confirm that the contribution of mine origin stable Pb to Pb in mussel flesh is consistently low at approximately 2 per cent (Bollhöfer 2012).

In a model to estimate mine related doses to people from the ingestion of traditional food items Martin (2000) used a concentration ratio for mussels for ²²⁶Ra from the water column (CR_{Ra-226,mussel}) of 19,000 L·kg⁻¹, which was determined by Johnston et al (1984, 1987). For the other radionuclide-tissue combinations in the model, CRs are published in Martin et al (1998). For a hypothetical release of RP2 water from RUM Martin (2000) has shown that ²²⁶Ra is the main contributor to a mine related ingestion dose (>92.2%) downstream of RUM and the main food item responsible for this dose is the consumption of freshwater mussels¹, *Velesunio angasi*, (85.1%).

The ²²⁶Ra trigger value for Magela creek is based on data collected and assessed in the 1980s and, as the major source of mine related doses to people from the ingestion of traditional food items, with new data available it is timely to conduct this review into the appropriateness of the current value used.

1.2 Radionuclide concentration ratios

Concentration ratios (CRs) are commonly used to quantify radionuclide transfer to biota from the surrounding environmental media that generally function as the reservoirs for nutrients and contaminants (Doering 2013). The CR method is a simplistic approach and does not take into account the chemical form of the radionuclide, competing ions, pH, and other chemical parameters that can affect the transfer of radionuclides. Nevertheless, it is consistent with the approach used in many human and environmental assessment models for quantifying radionuclide transfer (Brown et al 2008; USDOE 2004; Yu et al 2002). CRs for biota in aquatic systems are typically expressed as:

$$CR = \frac{Activity \ concentration \ in \ biota \ tissue \ (Bq \ kg^{-1} \ fresh \ weight)}{Activity \ concentration \ in \ water \ (Bq \ l^{-1})} \tag{1}$$

The CR method assumes equilibrium conditions between the receptor and donor compartments. Equilibrium conditions can reasonably be expected to exist in natural and undisturbed environments or where the environment is receiving continuous (steady) inputs of radionuclides from a site. In the uranium mining context, the CR method is likely to be valid for conditions of normal operation and also in the post-rehabilitation phase (Doering 2013).

The mussel CR for ²²⁶Ra has previously been determined as the average CR of four billabongs in the Alligator Rivers Region (Johnston et al 1984). These billabongs were Georgetown, Mudginberri, Leichhardt and Corndorl billabongs. Table 1 reproduces the CR values for ²²⁶Ra in freshwater mussel tissue from Johnston et al (1984). More recently, Bollhöfer et al (2011) have reported CRs for ²²⁶Ra that range from 28 000 to 33 000 with an average of 30 500 for sites in Magela Creek, downstream of Bowerbird Billabong.

Concentration ratios for ²²⁶Ra uptake in freshwater mussels are calculated relative to total water ²²⁶Ra activity concentrations. This is because mussels are filter feeders, meaning they take up food (plankton, algae and other nutrients and particles) suspended in the

¹ This is assuming that 2 kg of wet mussel tissue is consumed by a 10 year old child per year.

water column through trapping in the mucus of their gills (Humphrey & Simpson, 1985; Riisgård et al 2011). Plankton and algae are known to accumulate natural radionuclides to high concentrations (Fisher et al 1987; Vandenhove et al 2010) and it is thus important to include the activity concentration associated with suspended particles in the calculation of CRs.

Isotope	Georgetown BB	Mudginberri BB	Leichhardt BB	Corndorl BB	Average
²³⁸ U	50	>225	>615	>490	
²²⁶ Ra	6700	27,000	5200	38,000	19,000
²¹⁰ Pb	890	8400	3300	7800	5100

Table 1 Concentration ratios in $L \cdot kg^{-1}$ (relative to total water ²²⁶Ra activity concentrations) for freshwater mussel flesh (from Johnston et al 1984)

1.3 Radiation protection of humans

1.3.1 Ingestion pathway approach

To determine effective doses from the ingestion of radionuclides downstream of an operating mine, the total effective dose H to local people harvesting aquatic bushfoods and ingesting certain biota tissues is calculated using the following equation:

$$H = \sum_{i} \sum_{j} A_{i} \cdot CR_{i,j} \cdot m_{j} \cdot DCC_{i}$$
⁽²⁾

With:

A_i: activity concentration (Bq·L⁻¹) of radionuclide i in water

 $CR_{i,i}$: concentration ratio (L·kg⁻¹ wet tissue) of radionuclide *i* in food item tissue *j*

m_i: weight of tissue type *j* consumed (kg)

DCC: dose conversion coefficient of radionuclide *i* (Sv·Bq⁻¹)

1.3.2 Existing operational radium-226 water quality limit based on human radiological protection criteria

Water quality upstream and downstream of Ranger has been measured routinely in Magela Creek by *eriss* since 2000. Klessa (2001a,b) has summarised previous data to derive water quality objectives for mine origin inputs downstream of RUM. While many of the water quality objectives have been derived from statistical analysis of upstream data, the current limit for ²²⁶Ra (10 mBq·L⁻¹) is based on human radiological protection considerations (Klessa 2001b).

The limit for ²²⁶Ra in Magela creek applies to the increase above natural background in total ²²⁶Ra activity concentration in surface waters downstream of RUM. It was derived based on the following assumptions: (i) a dose constraint of 0.3 mSv per year above natural background from the ingestion of ²²⁶Ra in mussels, (ii) a 10 year old child consuming 2 kg (wet weight) of mussels annually, and (iii) a CR for mussels of 19,000 L·kg⁻¹ for ²²⁶Ra from the water column (Johnston et al 1984; 1987). With these

assumptions and considering the ²²⁶Ra activity concentration in mussels only, equation (2) simplifies and reduces to:

$$H_{Ra-226} = m_{mussel} \cdot CR_{Ra-226,mussel} \cdot A_{Ra-226} \cdot DCC_{Ra-226}$$
(3)

With:

A _{Ra-226} :	total activity concentration (Bq·L-1) of ²²⁶ Ra in water above background
CR _{Ra-226,mussel} :	concentration ratio for mussels for $^{226}\mathrm{Ra}$ from the water column (L·kg ⁻¹)
m _{mussel} :	weight of mussel tissue consumed (kg)
DCC _i :	dose conversion coefficient for 226 Ra (Sv·Bq ⁻¹).

With the 3 assumptions above and using equation (3), a 226 Ra activity concentration limit of 10 mBq·L⁻¹ above natural background in Magela creek water will lead to an annual dose of 0.3 mSv for a 10 yr old child from the ingestion of 2 kg wet weight of mussels.

Iles (2004) revised the water quality objectives for Magela Creek. For the total water ²²⁶Ra activity concentration the objective is: "The median total ²²⁶Ra activity concentration for the wet season at the downstream site will not be more than 10 mBq/L greater than that at the upstream site". A measure of success for meeting this objective is that "the downstream median total ²²⁶Ra activity concentration for the wet season minus the upstream median total ²²⁶Ra activity concentration for the wet season minus the upstream median total ²²⁶Ra activity concentration for the wet season is not greater than 10 mBq/L".

1.4 Radiation protection of the environment

1.4.1 Approach

Revisions of the Australian Radiation Protection and Nuclear Safety Agency's (ARPANSA) National Directory for Radiation Protection (republished in February 2014) and Safety Fundamentals in Radiation Protection Series F-1 (ARPANSA 2014b) were undertaken to take account of more recent recommendations by the ICRP (ICRP 2007; 2008) and the revised IAEA Basic Safety Standards (IAEA 2014) to explicitly include protection of the environment from the harmful effects of ionising radiation in national guidance documents. The Safety Fundamentals in Radiation Protection Series F-1 (which is the top tier document in the Australian national framework to manage risks from ionising radiation) explicitly includes environmental exposures of wildlife in the natural environment and state that "*Regulatory consideration of scenarios that may put the environment at risk (either individuals or species that may be protected for conservation purposes, or populations or ecosystems) protects against effects of ionising radiation of environmental concern"*. Consequently, radiation doses to wildlife from radionuclides released into the environment affects exists (Supervising Scientist 2014).

Environmental exposures are typically quantified as the above-background absorbed dose rate to wildlife from a radionuclide contaminated environment and are typically placed in a risk context by comparing to a benchmark dose rate (Doering 2013). The benchmark dose rate is an absorbed dose rate value that is considered to provide an acceptable level of protection to the environment; generally the prevention of deleterious

impacts to wildlife populations and ecosystem biodiversity (Doering & Bollhöfer 2015a; ICRP, 2007, 2008). The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) concluded from review of the scientific literature on radiation effects that "maximum dose rates of 400 uGy/h to a small proportion of the individuals in aquatic populations of organisms would not have any detrimental effect at the population level" (UNSCEAR, 1996, 2008).

Doering (2010) has reviewed several approaches that are used to assess radiation doses to wildlife, to provide some national guidance on available frameworks for radiation protection of wildlife, identified through the National Directory for Radiation Protection. He concludes that the ERICA Integrated Approach and ERICA Tool (Brown et al 2008) provide a practical framework for assessing absorbed dose rates to wildlife. Using the ERICA tool, absorbed dose rates to wildlife can be modelled and compared in a 3-Tier approach. Other tools are available, such as ResRad-Biota (Yu et al 2002) and K-Biota (Kum et al 2010). It is important to note that currently, radiation exposure of wildlife is not formally considered when deriving radiological water quality criteria for Magela Creek.

1.4.2 Aquatic organisms

Johnston et al (2005) calculated the weighted absorbed dose for nine species of biota resulting from a hypothetical release of four radionuclides (polonium, radium, thorium and uranium) into Magela Creek (Figure 3). They used and modified a spreadsheet based program developed by the UK Environment Protection Agency (Copplestone et al 2001), commonly referred to as R&D 128. Johnston et al (2005) found that the freshwater mussel (*Velesunio angasi*) showed absorbed doses rates at least one order of magnitude higher than any other studied organism, primarily (95%) resulting from the alpha decay of bioaccumulated ²²⁶Ra and its decay products (Sauerland et al 2005).



Figure 3 Predicted contributions to the weighted absorbed dose for organisms in Magela creek with an RP2 discharge rate of 2 $m^3 \cdot s^{-1}$ (from Johnston et al 2005).

For their assessment Johnston et al (2005) used a similar approach to Martin (2000) and Martin et al (1998) for human dose assessment, in so far as they assumed a scenario in which RP2 water is released from the mine site, with water ²²⁶Ra activity concentrations approximately 70 times higher than activity concentrations of ²¹⁰Po. Figure 3 is a reproduction of their results, showing that the magnitude of the absorbed effective dose to mussels will be from ²²⁶Ra. It also shows that mussels are the limiting organism, as far as doses to aquatic wildlife in the Alligator Rivers Region from an assumed release of mine waters are concerned.

In this report we use the ERICA tool (Brown et al 2008) to undertake a dose assessment for mussels in Magela Creek that receive internal exposures from bioaccumulated radionuclides and external exposures from radionuclides in water and sediment.

2 Methods

2.1 Data provenance

The data used in our study originates from annual collections of mussels and surface waters from Mudginberri Billabong (Figure 1). These collections were made by eriss between 2000 and 2013 (inclusive) as part of its routine bioaccumulation monitoring program. Summaries of each collection have been published in Supervising Scientist Annual Reports (Supervising Scientist, 2008, 2009 2014, 2015).

2.1.1 Mussels

As part of the bioaccumulation monitoring program of the Supervising Scientist, mussels are collected each year from Mudginberri Billabong at the end of the dry season in September or October (Ryan et al 2005). The 2007 collection was done in May at the end of the wet season as part of a larger study of radium bioaccumulation in mussels along Magela Creek (Bollhöfer et al. 2011). In 2008, mussels were collected at three different locations within Mudginberri Billabong, effectively giving three separate collections for that year (Supervising Scientist 2009). Mussels collected in each year other than 2009 and 2010 were aged and all mussels of the same age were combined to form a single sample for analysis. In 2009 and 2010, all mussels collected were combined into one bulk sample; results from these two years have not been used in this data analysis. Further details on the methods of mussel collection, processing, aging and analysis are provided in Ryan et al (2005) and Bollhöfer et al. (2011).

2.1.2 Water

Surface water samples from Mudginberri Billabong are collected at the same time that mussels are collected. The samples were filtered in all years other than 2013. Analysis of both the filtered water and particulate fractions was done in 2000, 2002, 2007, 2008 and 2012. Only the filtered water fraction was analysed in other years. Further details on the methods of water collection, processing, and analysis are provided in Ryan et al (2005) and Bollhöfer et al. (2011).

To calculate total water ²²⁶Ra activity concentrations, which have been used in this study to calculate CRs for mussels, the following data manipulations were applied:

- results of the filtered water and particulate fractions were summed for those years where measurements on both had been made;
- for those years where only the filtered water fraction was analysed, the analysis result was multiplied by the arithmetic mean of the ratio of total/filtered ²²⁶Ra activity concentration in water samples collected from Magela Creek and Mudginberri Billabong during the years 2000 to 2013 (inclusive);
- no manipulations were applied to the 2013 data as the water sample was not filtered prior to analysis and results were for the total water fraction.

2.1.3 Concentration ratios

A CR dataset for mussels was generated by dividing the fresh weight ²²⁶Ra activity concentration in each age-composited mussel sample from each year by the total ²²⁶Ra activity concentration in water. The arithmetic mean ²²⁶Ra activity concentration in total water over <u>all</u> years was used to calculate CRs as it was considered to better represent the

long-term ²²⁶Ra exposure conditions of mussels in Mudginberri Billabong compared to the activity concentration in water on the day of mussel collection.

2.2 Human dose assessment

2.2.1 Exposure scenario

The exposure scenario for humans was based on a 10 year old child consuming 2 kg of mussels (fresh weight) from Mudginberri Billabong per year, with the further assumption that larger sized mussels were consumed in preference to smaller sized mussels since they offer greater nourishment for the effort expended in collecting them.

Mussels from the Magela Creek system reach approximately 90% of their lifetime size by the age of 3 years (Bollhöfer et al., 2011) and it was assumed that only mussels of this age and older were collected and consumed.

2.2.2 Concentration ratio

The geometric mean CR of mussels aged 3 years and older was calculated from the CR dataset taking into account the number of individual mussels in each age composited sample from each year. This value was used in the assessment as it represents the most likely CR of any mussel aged 3 years or older randomly collected from the billabong, as would be the case for an Aboriginal person collecting mussels opportunistically for sustenance.

2.2.3 Benchmark dose rate

The member of the public dose constraint for Ranger mine is 0.3 mSv (ERA 2014). This value denotes an upper bound on the annual doses that members of the public should receive from all above-background radiation exposures traceable to the mine such that radiation protection could be considered optimised – it is not a dose limit (ICRP, 2007). Doses from the ingestion of ²²⁶Ra in mussels represents radiation exposure to the public from a single radionuclide via a single pathway only. Although this is expected to be the dominant radionuclide-pathway combination contributing to public doses downstream of the mine following a release of retention pond water (Martin et al. 1998), other radionuclides and bush foods will also contribute to dose, and so too will the inhalation of radon progeny in air. To account for dose contributions coming from radionuclides and pathways other than ²²⁶Ra in mussels, the guideline value was not back-calculated from the dose constraint of 0.3 mSv, but from a lower annual benchmark dose of 0.2 mSv.

2.2.4 Assessment approach

Calculation of the guideline value was done by dividing the benchmark dose by the product of the geometric mean CR of mussels aged 3 years and older, the ²²⁶Ra ingestion dose coefficient for a 10 year old child (8.0x10⁻⁷ mSv mBq⁻¹, ICRP (1996)) and the weight of mussels consumed per year.

2.3 Environmental dose assessment

2.3.1 Exposure scenario

The exposure scenario for the environment was based on mussels in Magela Creek receiving internal exposures from bioaccumulated radionuclides and external exposures

from radionuclides in the water and sediment. A previous assessment by Johnston et al. (2005) found that mussels were the limiting organism for the creek and, in the case of a release of mine waters, would receive their dose predominantly from bioaccumulated 226 Ra.

2.3.2 Benchmark dose rate

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) is the international authority on the effects of ionising radiation and has concluded from a review of the scientific literature on radiation effects that "maximum dose rates of 400 μ Gy/h to a small proportion of the individuals in aquatic populations of organisms would not have any detrimental effect at the population level" (UNSCEAR, 1996, 2008).

2.3.3 Assessment approach

The tier 3 assessment module in the ERICA Tool 1.2 (Brown et al., 2008) was used to determine probabilistically the absorbed dose rate to the organism (ie – a mussel) per unit of ²²⁶Ra activity concentration in water. This absorbed dose rate was then compared to the benchmark dose rate of 400 μ G·h⁻¹ to derive a total ²²⁶Ra activity concentration that would result in a dose rate of less than the benchmark to 99% of the population.

3 Results

3.1 Mudginberri mussel ²²⁶Ra activity concentrations

Figures 4a and 4b show a summary of all ²²⁶Ra activity concentrations measured in the flesh of age classed samples of freshwater mussels (including 0, 1 and 2 year old mussels), *Velesunio angasi*, collected between 2001 and 2013.

Radium-226 activity concentrations (Bq·kg⁻¹ dry weight) are lognormally distributed (p = 0.25; Anderson-Darling statistic: 0.466), with a geometric mean of 780 Bq·kg⁻¹ and an arithmetic mean of 940 Bq·kg⁻¹. The average (both arithmetic and geometric) dry to wet weight ratio in mussels collected between 2001 and 2013 is 0.10.



Figure 4 (a) Histogram and (b) cumulative probability plot of all ²²⁶Ra activity concentrations (in Bq·kg⁻¹ dry weight) measured in flesh of age classed freshwater mussels, *Velesunio angasi*. The blue lines are lognormal fits to the distribution and associated 95% confidence intervals.

3.2 Water ²²⁶Ra activity concentrations

3.2.1 Mudginberri Billabong

Figures 5a and 5b show the time series of all water ²²⁶Ra activity concentrations measured by *eriss* in Mudginberri Billabong since the early 1980s, total and filtered respectively. Data have been extracted from the BRUCE tool described in Doering (2013).



Figure 5 Time series of (a) total and (b) filtered ²²⁶Ra activity concentrations in Mudginberri Billabong from the 1980s to 2013.

Figures 6a, b, c and d show a statistical summary of all measured ²²⁶Ra activity concentrations in Mudginberri Billabong water collected between the early 1980s and 2013. Radium-226 activity concentrations in Mudginberi Billabong water follow a log-normal distribution with low Anderson-Darling statistics and p-values above 0.35. Table 2 shows that total ²²⁶Ra activity concentrations appear to be higher in the 1980s (which could be an effect of sample collection), but filtered ²²⁶Ra activity concentrations have not changed over the sampling periods.

The average ratio of ²²⁶Ra activity concentrations in the total water relative to the filtered water in samples taken from 2000 onwards is 1.6 which is the same as the ratio in Magela Creek samples (see below). This ratio was used to determine a total ²²⁶Ra activity concentration in water samples from Mudginberri Billabong where only the filtered water fraction was analysed. Arithmetic and geometric means of 0.0020 and 0.0018 Bq·L⁻¹ were calculated for the total ²²⁶Ra activity concentration in Mudginberri Billabong between 2001 and 2013.



Figure 6 (a, c) Histograms and (b, d) cumulative probability plots of all total and filtered water ²²⁶Ra activity concentrations (in Bq.L⁻¹) measured in Mudginberri Billabong. The blue lines are a lognormal fit to the distribution and associated 95% confidence intervals.

3.1.2 Magela Creek

Figures 7a and 7b show the time series of all water ²²⁶Ra activity concentrations measured by *eriss* in Magela Creek (upstream and downstream sites) since the commencement of ²²⁶Ra monitoring in 2000. Data shown are for both total and filtered water activity concentrations. Data have been extracted from the BRUCE tool, described in Doering (2013) and sites include all sites along the Magela Creek channel from which water samples have been collected.

Total water ²²⁶Ra activity concentrations in Magela Creek follow a log-normal distribution with an Anderson-Darling statistics value of 0.482 and p = 0.227 (Figure 8), a statistical summary of the data is provided in Table 2.



Figure 7 Time series of (a) total and (b) filtered ²²⁶Ra activity concentrations in Magela Creek, from 2000 to 2013.



Figure 8 (a, c) Histograms and (b, d) cumulative probability plots of all total and filtered water ²²⁶Ra activity concentrations (Bq·L⁻¹) measured in Magela Creek from 2000 onwards. The blue lines are lognormal fits to the distribution and associated 95% confidence intervals.

 Table 2 Summary of total and filtered water ²²⁶Ra activity concentrations (Bq·L-1). Arithmetic means and standard deviations are shown. Geometric means are in brackets.

	Ra-226 total	Ra-226 filtered	total/filtered
Mudginberri Billabong			
All data	0.0026± 0.0019	0.0012 ±0.0009	2.1
	(0.0021)	(0.010)	(2.1)
1980s	0.0031 ± 0.0023	0.0012 ± 0.0011	2.5
	(0.025)	(0.009)	(2.8)
Post 2000	0.0020 ± 0.0010	0.00012 ± 0.0007	1.6
	(0.018)	(0.010)	(1.6)
Magela Creek			
Post 2000	0.0021 ± 0.0008	0.0013 ± 0.0005	1.6
	(0.0020)	(0.0013)	(1.6)

3.3 Radium-226 concentration ratios

Figure 9 shows a histogram of all ²²⁶Ra CRs (rather than concentrations) measured in flesh of age classed freshwater mussels (including 0, 1 and 2 year old mussels), *Velesunio angasi*, collected between 2001 and 2013 from Mudginberri Billabong. This dataset was generated by dividing the fresh weight ²²⁶Ra activity concentration in each age-composited mussel sample from each year by the arithmetic mean ²²⁶Ra activity concentration in total water of 0.0020 Bq·L⁻¹ measured in Mudginberri Billabong post 2000. The average was used to calculate CRs as it was considered to better represent the long-term ²²⁶Ra exposure conditions of mussels in Mudginberri Billabong compared to the activity concentration in water on the day of mussel collection. In addition, the CRs were weighted by the number of mussels in each composite age class: when an aged composite sample from a particular collection consisted of *i* mussels, the CR value is represented *i*-times in Figure 9. Table 3 shows the summary statistics for this dataset.



Figure 9 (a) Histogram and (b) cumulative probabilities of CRs [L·kg⁻¹_{wet}] measured in all mussels collected since 2001 in Mudginberri Billabong. The blue line represents a lognormal fit to the data.

Table 3	Statistical summary for t	the whole CR data-set.	AM: arithmetic mea	an; AMSD: ar	ithmetic mean
	standard deviation; GM:	geometric mean; GMS	D: geometric mean	standard dev	iation.

	Ra-226 CR [L·kg⁻¹ _{wet}]
N: number of data	1106
AM ± AMSD	36,100 ± 23,900
GM ± GMSD	30,000 ± 1.9
Median	31,200
(25-75 percentile)	(19,700-44,000)

Figure 10 shows a histogram of all ²²⁶Ra CRs measured in flesh of age classed freshwater mussels (excluding those mussels that are 0, 1 and 2 year old), collected between 2001 and 2013 from Mudginberri Billabong and Table 4 shows the summary statistics for this data set.



Figure 10 Concentration ratios ([L/kg_{wet}] measured in mussels older than 2 years, collected since 2001 in Mudginberri Billabong. The blue line represents a lognormal fit to the distribution.

Table 4 Statistical summary of the CR data-set excluding 0, 1 and 2 year old mussels. AM: arithmetic mean; AMSD: arithmetic mean standard deviation; GM: geometric mean; GMSD: geometric mean standard deviation.

	Ra-226 CR [L·kg⁻¹ _{wet}]
N: number of data	774
AM ± AMSD	44,100 ± 900
GM ± GMSD	39,500 ± 1.6
Median	38,500
(25-75 percentile)	(29,200-54,500)

3.4 Human dose assessment

The exposure scenario for humans is based on a 10 year old child consuming 2 kg of mussels (fresh weight) per year from Mudginberri Billabong. A recent review conducted by Energy Resources of Australia Ltd (Garde 2015) has shown that the diet used in Ryan et al (2008) is most likely "*still accurate in 2014*". It has thus been assumed that adults consume 4 kg of mussels (wet weight) per year while a 10 year old child's consumption is 50% of that. A further assumption is that larger sized mussels (3+ years) were consumed in preference.

Using equation 3, with a typical CR of 39,500 ($L\cdot kg_{wet}^{-1}$) for ²²⁶Ra in mussels (Table 4) and the ²²⁶Ra ingestion dose coefficient for a 10 year old child ($8.0\cdot10^{-7}$ mSv·mBq⁻¹ from ICRP 1996), the average above background total water ²²⁶Ra activity concentration in Magela Creek should be less than 3.2 mBq·L⁻¹ to stay below a dose constraint of 0.2 mSv per year. Choosing a limit for the difference between the downstream and upstream water ²²⁶Ra activity concentration of 3 mBq·L⁻¹ will ensure that doses will remain below the 0.2 mSv per year benchmark for radiation exposure of humans from the consumption of freshwater mussels downstream of RUM.

3.5 Environmental dose assessment

Table 5 gives the input parameter values used for the ERICA assessment.

Figure 11 shows the distributions of internal and external dose rates for an ERICA Tier 3 assessment for the freshwater mussel, *Velesunio angasi*, from an assumed water 226 Ra activity concentration of 1 Bq·L⁻¹ and using parameters in Table 5. Only around 1% of the dose rate to the mussels is from external gamma radiation.

The 99th percentile of the output probability distribution of the total dose rate was chosen to represent the maximum dose rate to 'a small proportion of individuals' in the mussel population and was subsequently used to back-calculate the guideline value from the 400 μ Gy·h⁻¹ benchmark dose rate adopted from UNSCEAR (1996, 2008). The total water activity concentration of ²²⁶Ra leading to an exposure of 400 μ Gy·h⁻¹ to 1 per cent of the mussel population was calculated to be 24 mBq·L⁻¹.

Parameter	Value
Radionuclides	²²⁶ Ra
Organisms	Mollusc - bivalve
CR	The arithmetic mean (36,100) and standard deviation (23,900) of the total mussel population were entered as a lognormal distribution with a lower bound of zero and an upper bound of infinity.
Kd	The arithmetic mean (227,500) and standard deviation (47,170) of values presented in Humphrey and Simpson (1985) for ²²⁶ Ra in Magela Creek were entered as a lognormal distribution with a lower bound of zero and an upper bound of infinity.
Occupancy factor	100% at sediment-surface
Radiation weighting factors ^a	10 for alpha, 1 for gamma/beta and 3 for low energy beta
Water activity concentration	1 Bq·L ⁻¹
Simulations	10000
Seed	0

Table 5 Input parameter values used in the tier 3 assessment module of the ERICA Tool 1.2 to calculate the absorbed dose rate to mussels per unit activity concentration of 226Ra in water.

^aThe UNSCEAR dose rate of 400 μGy h⁻¹ refers to the effects of exposure to low-LET radiation. Where a significant part of the incremental radiation exposure comes from high-LET radiation (alpha particles), it is necessary to take account of the different relative biological effectiveness of the radiation.





Figure 11 Distributions of internal and external an/d dose rates modelled for freshwater mussels, *Velesunio angasi*, from an assumed water ²²⁶Ra activity concentration of 1 Bq·L⁻¹

4 Summary and conclusions

Doses to humans and the environment arising from ²²⁶Ra in Magela Creek water have been assessed. As it has previously been shown (Martin et al 1998; Martin 2000; Johnston et al 2005) that for a potential release of mine water from Ranger mine, ²²⁶Ra in mussels will be the most important contributor to radiation doses to both humans and the environment, the assessment presented here focussed on ²²⁶Ra in water only.

Our assessment of ingestion doses to humans and internal and external doses to mussels from ²²⁶Ra has shown that human radiological protection is more limiting than considerations for radiation protection of the environment. The assessment is based on the above background total water ²²⁶Ra activity concentration in Magela Creek downstream of RUM and by assuming that a 10 year old child consumes 2 kg (wet weight) of large mussels per year. Using a typical CR for ²²⁶Ra in 3+ year old mussels from Mudginberi Billabong collected between 2001 and 2013 we have shown that a mine derived increase in total water ²²⁶Ra activity concentration of less than 3 mBq·L⁻¹, will lead to a mine origin ingestion dose to humans of less than 0.2 mSv per year. The same increase in total water ²²⁶Ra activity concentration of 3 mBq·L⁻¹, will lead to a maximum additional dose rate to a small number of mussels (1%) of 50 µGy·h⁻¹.

Total water ²²⁶Ra activity concentrations in Magela Creek are log-normally distributed, thus the revised operational water quality objective for ²²⁶Ra in Magela Creek should be: "*The geometric mean of the total ²²⁶Ra activity concentration for the wet season at the downstream site will not be more than 3 mBq/L greater than that at the upstream site*".

This will ensure that doses to humans from the ingestion of mussels will be below 0.2 mSv, and that the radiological impact from ²²⁶Ra in the water column to the aquatic environment in Magela Creek will be negligible.

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