



Land application of mine water causes minimal uranium loss offsite in the wet-dry tropics: Ranger Uranium Mine, Northern Territory, Australia



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ABSTRACT

Ranger Uranium Mine (RUM) is situated in the wet-dry tropics of Northern Australia. Land application (irrigation) of stockpile (ore and waste) runoff water to natural woodland on the mine lease is a key part of water management at the mine. Consequently, the soil in these Land Application Areas (LAAs) presents a range of uranium (U) and other metals concentrations. Knowledge of seasonal and temporal changes in soil U and physicochemical parameters at RUM LAAs is important to develop suitable management and rehabilitation strategies. Therefore, soil samples were collected from low, medium, high and very high U sites at RUM LAAs for two consecutive years and the effect of time and season on soil physicochemical parameters particularly U and other major solutes applied in irrigation water was measured. Concentrations of some of the solutes applied in the irrigation water such as sulphur (S), iron (Fe) and calcium (Ca) showed significant seasonal and temporal changes. Soil S, Fe and Ca concentration decreased from year 1 to year 2 and from dry to wet seasons during both years. Soil U followed the same pattern except that we recorded an increase in soil U concentrations at most of the RUM LAAs after year 2 wet season compared to year 2 dry season. Thus, these sites did not show a considerable decrease in soil U concentration from year 1 to year 2. Sites which contained elevated U after wet season 2 also had higher moisture content which suggests that pooling of U containing rainwater at these sites may be responsible for elevated U. Thus, U may be redistributed within RUM LAAs due to surface water movement. The study also suggested that a decrease in U concentrations in LAA soils at very high U (>900 mg kg⁻¹) sites is most likely due to transport of particulate matter bound U by surface runoff and U may not be lost from the surface soil due to vertical movement through the soil profile. Uranium attached to particulate matter may reduce its potential for environmental impact. These findings suggest that U is effectively adsorbed by the soils and thus land application may serve as a useful tool for U management in the wet-dry tropics of northern Australia.

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1. Introduction

Ranger Uranium Mine (RUM) receives monsoonal rainfall, with 700–2200 mm (average 1540 mm) falling in the wet season (November–April) which makes water management a major issue at the mine. In addition, the mine is surrounded by Kakadu National Park, which has world heritage listing. Water from the site is stored in retention ponds, RP1 and RP2. RP1 is a part of the sediment control system on the mine-site and water is discharged into Magela Creek during most wet seasons (Akber et al., 2011a). RP2

stores runoff water from the low-grade ore and waste stockpiles and other areas on the minesite. Uranium, probably in the form of UO_2^{2+} , sulphur in the form of SO_4^{2-} , copper (Cu^{2+}), iron (Fe^{2+}) and manganese (Mn^{2+}) are the major ionic species present in RP2 water (Zimmermann and Lu, 2011). Lead (Pb^{2+}) and calcium (Ca^{2+}) ions are also present in small amounts. Uranium in the RP2 water is under oxidising to mildly reducing conditions. Under such conditions, uranium (U) in solution predominantly exists as uranyl ion (UO_2^{2+}) or as soluble carbonate complexes ($(\text{UO}_2)_2\text{CO}_3(\text{OH})^{3-}$, UO_2CO_3 , $\text{UO}_2(\text{CO}_3)_2^{2-}$, $\text{UO}_2(\text{CO}_3)_3^{4-}$ and possibly $(\text{UO}_2)_3(\text{CO}_3)_6^{6-}$ in carbonate containing waters at near neutral or basic pH (Ciavatta et al., 1981; Duff and Amrhein, 1996; Grenthe et al., 1992). The uranyl species may also form complexes with sulphate, fluoride and possibly chloride where concentrations of these anions are high (Grenthe et al., 1992). At a sulphate level of 100 mg kg^{-1} , UO_2SO_4 may be a significant species up to pH 7 (Langmuir, 1978). Uranyl hydroxy complexes such as $\text{UO}_2(\text{OH})^+$ and $(\text{UO}_2)_3(\text{OH})^{5+}$ are also formed in carbonate-depleted alkaline water. In RP2 water, the sulphate concentration is $700\text{--}850 \text{ mg kg}^{-1}$ but the pH ranges from 7 to 9, which may not favour sulphate complexes. This suggests that the U may predominantly be present as the uranyl hydroxy complexes such as $\text{UO}_2(\text{OH})^+$ and $(\text{UO}_2)_3(\text{OH})^{5+}$ in RP2 water because these species predominate under U concentration greater than $1000 \text{ }\mu\text{g/L}$ (concentration present in RP2 water) and pH greater than 5 (Krupka and Serne, 2002; Markich, 2002).

From 1985 to 2008–09, water stored in RP2 during the wet seasons was disposed of onsite by irrigating Land Application Areas (LAAs) during subsequent dry seasons (Table 1). The concentration of metals and radionuclides varied between LAAs largely due to different durations of water application, variable water quality (wetland treated vs untreated water), different distance of these LAAs from the source of irrigation (e.g., distance from sprinkler head) and method of irrigation (spray vs flood irrigation) (Table 1). All sites at LAAs are well drained to excessively drained soils generally having high gravel content (20–50% of soil mass) composed of quartz and ferruginous material. These soils have low soluble salt contents, acidic pH and low concentration of secondary iron and manganese oxides (Chartres et al., 1991). The soils are also low in clay content, usually <20% and deficient in organic matter (<1%) (Chartres et al., 1991). Vegetation at RUM LAAs includes open eucalypt forest dominated by *Eriachne tetradonta*, *Eriachne miniata*, *Eriachne bleeseri* and *Eriachne porrecta*. The understory consists of *Acacia* sp., *Livistona humilis* and *Gardenia megasperma* with a variable grass cover of *Sorghum* sp., *Themeda triandra* and *Eriachne trisetata* (Chartres et al., 1991). The cation exchange capacities (CEC) of the soils are extremely low (<10 cmol [+]/kg) with no detectable anion exchange capacity (Willett et al., 1993). Although the soils have limited capacity to assimilate major cations, manganese (Mn^{2+}), uranyl (UO_2^{2+}) and radium (Ra^{2+}) ions present in RP2 water are likely to be taken up in the soil through high affinity and adsorption reactions possibly mediated by the ferruginous gravels (Chartres et al., 1991; Willett and Bond, 1995).

Soil U can be sorbed onto soil particles and in pore water it can occur in complexes, or in reduced and precipitated forms. These different forms affect the mobility and fate of U in the soil environment (Zhou and Gu, 2005). The forms of U in the RP2 water will influence its adsorption to the soil particles in the LAAs. Carbonate complexing results in reduced adsorption of U leading to its release from soils (Pabalan and Turne, 1997; Waite et al., 1994) while soils have greater affinity for positively charged uranyl hydroxy complexes (Echevarria et al., 2001) – the complexes which are expected in RP2 water. Chemical fractionations showed that U is not in exchangeable forms in the soils at RUM LAAs but reacted to produce less mobile forms (Willett and Bond, 1998). Consequently, U is retained in the surface 10 cm and is preferentially adsorbed to the fine earth and to the iron oxides in the soil thereby effectively immobilizing the U before it can reach the regional aquifers (Brown et al., 1998; Hollingsworth et al., 2005). Uranium could be mobile in the sandy soils present in the lower slope sites in the Magela LAA and Retention Pond 1 (RP1) LAA, when applied at high concentrations (Hollingsworth et al., 2005; Willett and Bond, 1995). The concentration of radionuclides adsorbed in the soil due to land application of RP2 water may require some RUM LAAs to be rehabilitated at mine closure (Akber et al., 2011a). Understanding the seasonal and temporal effect on soil U and physicochemical parameters at RUM LAAs is important for adopting sustainable management and rehabilitation practices. Therefore, the aim of this study was to measure the effect of time and season on LAA soil physicochemical parameters, particularly the concentration of U and other major solutes. Existing data indicate that major radionuclides and divalent cations present in irrigation water are efficiently retained by the soils at RUM LAAs and the loss of these radionuclides and divalent cations offsite after the wet season rainfall is minimal (Chartres et al., 1991; Hollingsworth et al., 2005; Willett and Bond, 1998). In light of this, we predicted that there would not be a significant change in soil physicochemical properties between years and between wet and dry seasons. Further, we also hypothesized that U concentrations would not differ significantly between years or seasons. To address these hypotheses, we measured soil physicochemical parameters and U concentrations at the end of wet and dry seasons for two consecutive years and analysed the temporal and seasonal differences in soil physicochemical properties.

2. Materials and methods

2.1. Sites and sampling

Soil samples were collected from 18 sites at the RUM LAAs (Fig. 1, Table 2). The selection was based on the distance of these sites from the source of irrigation, year commissioned, the quality of irrigated water and method of irrigation (Table 1) so as to determine their effect on the U and other solute concentrations in the soil. Historical soil chemistry data were used to categorise sites as low, medium,

Table 1
Ranger Uranium Mine Land Application Areas (adapted from Akber et al., 2011a).

Land Application Area and abbreviation	Water quality	Irrigation method	Total area (ha)	Year commissioned
Magela (MLAA)	Unpolished RP2 water	Spray	33	1985
Magela Extension (MLAA EXT)	Unpolished RP2 water	Spray	20	1994
Retention Pond 1 (RP1LAA)	Polished RP2 water	Flood	46	1995
Djalkmara (DLAA)	Polished RP2 water	Flood	18	1997
Djalkmara Extension (DLAA EXT)	Polished RP2 water	Flood	20	1999
Jabiru East (JELAA)	Unpolished RP2 water	Spray	52	2006
RP1 Extension (RP1 EXT)	Unpolished RP2 water	Spray	8	2006
Corridor Creek (CCLAA)	Unpolished RP2 water	Spray	141	2007

Unpolished = raw RP2 water, Polished = water filtered through wetlands.

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