



# Mapping radioactivity in groundwater to identify elevated exposure in remote and rural communities

Ross Kleinschmidt<sup>a,b,\*</sup>, Jeffrey Black<sup>b</sup>, Riaz Akber<sup>a</sup>

<sup>a</sup> Queensland University of Technology, Faculty of Science and Technology, Discipline of Physics, 2 George Street, Brisbane, Queensland 4000, Australia

<sup>b</sup> Health Physics Unit, Queensland Health Forensic and Scientific Services, 39 Kessels Road, Coopers Plains, Queensland 4108, Australia

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## ABSTRACT

A survey of radioactivity in groundwater (110 sites) was conducted as a precursor to providing a baseline of radiation exposure in rural and remote communities in Queensland, Australia, that may be impacted upon by exposure pathways associated with the supply, treatment, use and wastewater treatment of the resource. Radionuclides in groundwater, including  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$ ,  $^{228}\text{Ra}$ ,  $^{224}\text{Ra}$  and  $^{40}\text{K}$  were measured and found to contain activity concentration levels of up to  $0.71\text{ BqL}^{-1}$ ,  $0.96\text{ BqL}^{-1}$ ,  $108\text{ BqL}^{-1}$ ,  $2.8\text{ BqL}^{-1}$ ,  $0.11\text{ BqL}^{-1}$  and  $0.19\text{ BqL}^{-1}$  respectively. Activity concentration results were classified by aquifer lithology, showing correlation between increased radium isotope concentration and basic volcanic host rock. The groundwater survey and mapping results were further assessed using an investigation assessment tool to identify seven remote or rural communities that may require additional radiation dose assessment beyond that attributed to ingestion of potable water.

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## 1. Introduction

Radiation exposure derived from Australian groundwater supplies is generally attributed only to ingestion of potable water. Radiological water quality guidelines have been developed regionally (ANZECC/ARMCANZ, 2000; NHMRC, 2004) and globally (WHO, 2008; USEPA, 2000; EU, 1998; Kocher, 2001) for potable, livestock, watering irrigation and recreational uses. These documents provide guidance in management and optimisation of water supply with a view to ensure that the total committed ingestion dose is maintained at less than 1 mSv in a year. While ingestion of water may contribute significantly to the dose of a critical group member, other less obvious exposure pathways need to be considered in assessing the full impact of radioactive constituents in groundwater supplies. Generation of Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) from water resource exploitation is a topic both locally (Cooper, 2003; Kleinschmidt and Akber, 2008; RHSAC, 2004) and internationally (IAEA, 2003). The USEPA have produced a guideline document detailing management practices for radioactive residuals derived from drinking water technologies

(USEPA, 2005), however, these and other assessments (Kleinschmidt and Akber, 2008) tend to focus on sludges produced from conventional water treatment plants typical of large urban systems, large scale water conditioning, reverse osmosis, and private point-of-entry treatment systems.

It has been recognised that the water supply systems of small, remote communities may differ from those of urban centres, quite often based on economical and environmental factors (DNRM, 2005). In many cases these communities rely on groundwater for their water supply. Often a sole, local resident is responsible for maintenance of water supply and sewerage infrastructure including headworks, reticulation and wastewater treatment. If the person resides and works in a community relying on groundwater containing elevated levels of NORM, then exposure pathways other than ingestion of water may need to be considered. Comprehensive information is not available on dose estimation for situations such as that described. Reference to studies on individual sources of exposure however, exists. For example those associated with radium scale in water supply distribution systems (Valentine and Stearns, 1994), hot water tanks (DeVol and Woodruff, 2004), exposure from radium and radon in water supplies and spring waters (Alabdulhman and Maghrawy, 2010; Koulouris et al., 1996), and water treatment plants (Toussaint and Burkett, 1996). Kleinschmidt (2007) identified a number of potential exposure pathways for individuals working and residing in small communities, recommending the need for a detailed exposure

\* Corresponding author. Health Physics Unit, Queensland Health Forensic and Scientific Services, 39 Kessels Road, Coopers Plains, Queensland 4108, Australia. Tel.: +61 7 3274 9124; fax: +61 7 3274 9123.

E-mail address: [ross\\_kleinschmidt@health.qld.gov.au](mailto:ross_kleinschmidt@health.qld.gov.au) (R. Kleinschmidt).

assessment to include not only potable water dose contributions, but also those associated with recreational, workplace and waste disposal activities. The purpose of this study is to provide a means of identifying those communities that may be at increased risk of exposure to radioactivity associated with groundwater.

Screening surveys have previously been used, particularly for  $^{222}\text{Rn}$  maps, to facilitate the radiological characterisation of an area of interest. Mapping radioactivity levels in air, water and the terrestrial environment may be used as a precursor to carrying out more detailed surveys that serve to validate, or extend, existing data and to identify areas of potential public harm, or to provide baseline data prior to the commencement of a new radiation practice (Synnott and Fenton, 2005; WHO, 2009). Knowing the groundwater radioactivity characteristics for the reference site, in conjunction with the identified exposure pathways, allows for implementation of a simple risk based assessment of the potential impact of elevated groundwater radioactivity levels established during the mapping process.

## 2. Method

### 2.1. Survey and sampling design

A groundwater screening program was developed to provide initial data on the extent and magnitude of radiological properties of groundwater supply and use in the state of Queensland, Australia. The sampling was designed to include as many aquifer systems as possible, particularly those serving a community for potable, recreational or livestock water use. Sampling regions were chosen to cover the range of aquifer lithology descriptors as provided by the Queensland Water Resources Commission (QWRC, 1987). As a large physical land area was to be covered (approximately 1.7 million square kilometres serving a population of 4 million), a 'mail-out' water sampling kit was developed (Fig. 1). Sampling kits comprising of prepared polyethylene sample bottles, detailed sampling instructions (see Supplementary Material) based on standard water sample collection methods (AS/NZS, 1998) and a questionnaire were assembled with packaging and return freight instructions. These kits were then forwarded to a number of regional shire councils and selected sampling agents. The questionnaire included pre-assigned sampling location descriptions and laboratory codes, fields for entering data on the physical location, latitude and longitude, water treatment processes, physical characteristics of the bore including its depth and yield, and the population served.

### 2.2. Radioanalytical methods

Radon ( $^{222}\text{Rn}$ ) analysis via liquid scintillation spectrometry, and radium ( $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ) & uranium ( $^{235}\text{U}$  and  $^{238}\text{U}$ ) isotope analysis by high resolution gamma spectrometry were considered suitable for the screening program. Potassium 40 in water was determined from total potassium (natural abundance of 0.0117%; IUPAC, 1998) analysis using atomic absorption spectrometry, or

inductively coupled mass spectrometry as dictated by laboratory instrumentation availability.

The concentration of  $^{222}\text{Rn}$  in water was determined using the direct counting method described by Kleinschmidt and Akber (2008). The sampling instructions included detailed information on minimisation of delays in submitting samples for analysis, and additionally the radioanalytical laboratory assessed all samples for time limitation compliance. To further monitor sampling effectiveness and reproducibility, duplicate samples were collected at each sampling location. Low diffusion Teflon<sup>®</sup> coated liquid scintillation vials (supplied by Perkin Elmer) were used. Sample aliquots of 10 mL were prepared in similar Teflon coated vials, by introducing the sample water under 5 mL of Mineral Oil<sup>®</sup> (Perkin Elmer) scintillation cocktail. Samples were shaken to mix, and then held for at least 4 h before counting so that equilibrium between  $^{222}\text{Rn}$  and its decay progeny was attained. Analysis was performed using TriCarb 3170TR/SL, TriCarb 3180 TR/SL and QUANTULUS 1220 (Perkin Elmer Pty Ltd) alpha/beta discriminating liquid scintillation analysers depending on instrument availability. All  $^{222}\text{Rn}$  concentration results were corrected for decay back to the date and time of sampling. A minimum detection level of 20 mBqL<sup>-1</sup> was achieved for the method using a count time of 120 min, this value considered as being adequate for a screening program.

Uranium, thorium and radium screening analysis was conducted using high resolution gamma spectrometry after sample preparation via barium and iron hydroxide co-precipitation based on the method described by Parsa et al. (2005). A 1000 mL sample of water was acidified to pH ~2 with 9 M H<sub>2</sub>SO<sub>4</sub>, and  $^{133}\text{Ba}$  tracer, of nominal activity 0.5 BqL<sup>-1</sup>, added to determine radium chemical recovery. The barium carrier solution was added and the sample heated to 50° C for 30 min while stirring to allow co-precipitation of radium isotopes with barium sulphate. The iron carrier is added and the sample neutralised by adding dilute NaOH until a brown precipitate forms. The precipitate was progressively separated by settling, decanting, centrifuging and rinsing into a 90 mm × 14 mm diameter polyethylene tube. The resulting precipitate 'plug' was dried in a block heater at 80° C and then sealed in the tube pending counting. Counting was performed using a low background, well type high resolution gamma-ray spectrometer (EG&G 150-15 well germanium detector and EG&G DSpec Plus<sup>®</sup> spectrometer). The gamma-ray spectrometer was calibrated using reference pitchblende material known to be in equilibrium ( $^{238}\text{U}$  of activity 101 Bqg<sup>-1</sup>, Sill and Hindman, 1974) in a geometry replicating that of the sample. Approximately 15% of the samples were counted within 48 h of preparation for  $^{224}\text{Ra}$  determination, with all prepared samples then stored for a minimum of 20 days to allow  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  decay progeny to attain secular equilibrium before counting. The mean chemical yield, as measured using the  $^{133}\text{Ba}$  tracer as an analogue for radium isotopes, was measured to be 80 ± 8%, however all result sets were corrected by the sample specific chemical yield factor. Uranium and Thorium chemical yield was considered to be 100% for the purposes of this screening method (Chou and Moffatt, 2000). For a counting time of 20 h and a 1000 mL sample volume, minimum detection levels of 50 mBqL<sup>-1</sup> for radionuclides of primary concern, i.e.  $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , can be achieved and is adequate for the purposes of the mapping program.  $^{238}\text{U}$  and  $^{232}\text{Th}$  were determined using respective, immediate decay progeny, and  $^{133}\text{Ba}$  via direct measurement of individual characteristic photopeaks.

### 2.3. Mapping

Groundwater activity concentration results for the radionuclides of interest were geographically mapped according to location and magnitude of activity. A map indicating the major groundwater aquifer types and associated lithology (Table 1) of Queensland is shown in Fig. 2. Small disjointed aquifers associated with both surface systems within the Great Artesian Basin and localised fractured rock systems are not well represented and the assignment of aquifer lithology was based on either interpretation of printed hydrology maps (QWRC, 1987), or the bore strata log where available. In all cases where a result for radioactivity concentration was below the calculated minimum detection level, a value of one half the MDL was used for plotting purposes.

**Table 1**  
Aquifer lithology key (QWRC, 1987).

| Code | Aquifer lithology                            | Examples                       |
|------|----------------------------------------------|--------------------------------|
| AI   | Acid to intermediate volcanics               | andesite, rhyolite, tuff       |
| BI   | Basic intrusives                             | gabbro, serpentine             |
| BV   | Basic volcanics                              | basalt                         |
| Ca   | Carbonates                                   | limestone, dolomite            |
| DL   | Complex alternation of different lithologies | —                              |
| MR   | Metamorphic rocks                            | schist, quartzite              |
| SS   | Sedimentary strata                           | sandstone, shale, conglomerate |
| US   | Unconsolidated sediments                     | sand, gravel                   |



**Fig. 1.** Groundwater sampling kit including 1 × 500 mL acid washed polyethylene bottle, 2 × 20 mL Teflon coated polyethylene liquid scintillation vials, sampling instructions and questionnaire, and reusable shipping container with prepaid consignment note.

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