Journal of Environmental Radioactivity 154 (2016) 83–92

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

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

Dissolved radon and uranium in groundwater in a potential coal seam gas development region (Richmond River Catchment, Australia)

Marnie L. Atkins [a, b, *], Isaac R. Santos [a, b], Anita Perkins [a, b], Damien T. Maher [a]

[a] School of Environmental Science and Management, Southern Cross University, Lismore, NSW, 2480, Australia
[b] National Marine Science Centre, School of Environment, Science and Engineering, Southern Cross University, Lismore, NSW, 2480, Australia

ARTICLE INFO

Article history:
Received 8 October 2015
Received in revised form
20 January 2016
Accepted 24 January 2016
Available online 8 February 2016

Keywords:
Groundwater
Radon
Uranium
Coal bed methane
Catchment
Baseline research
Unconventional gas

ABSTRACT

The extraction of unconventional gas resources such as shale and coal seam gas (CSG) is rapidly expanding globally and often prevents the opportunity for comprehensive baseline groundwater investigations prior to drilling. Unconventional gas extraction often targets geological layers with high naturally occurring radioactive materials (NORM) and extraction practices may possibly mobilise radionuclides into regional and local drinking water resources. Here, we establish baseline groundwater radon and uranium levels in shallow aquifers overlying a potential CSG target formation in the Richmond River Catchment, Australia. A total of 91 groundwater samples from six different geological units showed highly variable radon activities (0.14–20.33 Bq/L) and uranium levels (0.001–2.77 μg/L) which were well below the Australian Drinking Water Guideline values (radon; 100 Bq/L and uranium; 17 μg/L). Therefore, from a radon and uranium perspective, the regional groundwater does not pose health risks to consumers. Uranium could not explain the distribution of radon in groundwater. Relatively high radon activities (7.88 ± 0.83 Bq/L) in the fractured Lismore Basalt aquifer coincided with very low uranium concentrations (0.04 ± 0.02 μg/L). In the Quaternary Sediments aquifers, a positive correlation between U and HCO_3^- ($r^2 = 0.49$, $p < 0.01$) implied the uranium was present as uranyl-carbonate complexes. Since NORM are often enriched in target geological formations containing unconventional gas, establishing radon and uranium concentrations in overlying aquifers comprises an important component of baseline groundwater investigations.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Natural gas extraction from unconventional resources such as shale and coal seam gas (CSG; also referred to as coal bed methane) is rapidly expanding globally with potential impacts on shallow groundwater systems reported through increased aquifer connectivity and faulty infrastructure (Iverach et al., 2015; Jackson et al., 2013; Osborn et al., 2011). Unconventional gas extraction often targets geological layers with high naturally occurring radioactive materials (NORM) (Dai et al., 2014; Fouad and El-Rakaiby, 2009). If aquifer connectivity is increased during CSG production processes, radionuclides may migrate from the coal seam aquifers to overlying shallow aquifers. Currently, the literature lacks studies focussing on groundwater chemistry in shallow aquifers overlying target gas formations. Establishing baseline groundwater conditions for shallow aquifers in CSG drilling zones can assist in detecting and monitoring possible enhanced mobilisation and distribution of NORM, including uranium and radon.

Radon (^{222}Rn), primarily present in groundwater, is an inert radioactive noble gas with a half-life of 3.82 days (Swarzenski et al., 2006). Radon is the fifth product in the uranium (^{238}U) decay series and is produced from the decay of radium (^{226}Ra) present in soils and rocks. Radon has many field applications such as identifying fault locations (Han et al., 2006; Papp et al., 2010), seismic precursor (İçhedef et al., 2014; Miklavčić et al., 2008; Ramola et al., 1990), uranium exploration (Card, 1982; Roy, 2000; Wattananikorn, 1995), as a hydrological tracer (Burnett and Dulaiova, 2003; Peterson et al., 2010; Santos et al., 2015; Schmidt et al., 2008) and as an atmospheric chemistry tracer (Schmidt et al., 1996; Tait et al., 2015). In a CSG field in Australia, elevated atmospheric radon concentrations were related to either point or diffuse soil sources (Tait et al., 2013). This may indicate geological structure alterations resulting from CSG activities, highlighting the

* Corresponding author. School of Environmental Science and Management, Southern Cross University, Military Rd, Lismore, NSW, 2480, Australia.
E-mail address: marnieatkins1@gmail.com (M.L. Atkins).

http://dx.doi.org/10.1016/j.jenvrad.2016.01.014
0265-931X/© 2016 Elsevier Ltd. All rights reserved.

need for baseline investigations prior to CSG development.

Radon's short-lived alpha emitting decay products pose potential health hazards if inhaled or ingested (NRC, 1999). Risks of developing stomach and gastrointestinal cancers are significantly increased by ingesting drinking water with very high radon levels (Kendall and Smith, 2002; Zhuo et al., 2001). However, inhaled radon poses a greater risk than ingested radon (Folger et al., 1994; Khan et al., 2010) with 89% of radon related cancer deaths due to radon inhalation (lung cancers) and 11% of deaths due to radon ingested from drinking water (stomach cancers) (USEPA, 1999). Increased radon exposure in indoor environments occurs through radon gas diffusion from groundwater or underlying soils and rocks (Nazaroff, 1992). Identifying groundwater radon concentration in domestic environments is of vital importance to prevent excessive radiation exposure and predict potential health hazards.

Uranium is a ubiquitous naturally occurring trace element which is considered a toxic groundwater contaminant. Although the different uranium isotopes are naturally radioactive, uranium's chemical toxicity is 6 orders of magnitude more harmful than its radioactivity (Kurttio et al., 2002; Milvy and Cothern, 1990). Ingesting elevated uranium concentrations in drinking water can primarily lead to kidney disease (WHO, 2011; Zamora et al., 1998), although there is a lack of information relating uranium to other chronic adverse health conditions (EFSA, 2009; WHO, 2011). Although uranium content in groundwater is mainly derived from natural deposits, anthropogenic activities such as nuclear power emissions (EFSA, 2009), application of phosphate fertilisers (Birke et al., 2009; Schnug and Lottermoser, 2013), mining activities (Baborowski and Bozau, 2006; Carvalho et al., 2005) and coal and other fuel combustion (WHO, 2011) also contribute uranium to groundwater.

In natural waters, uranium minerals usually exist in two main oxidation states; the soluble hexavalent (U(VI)) or the insoluble tetravalent (U(IV)) form (Langmuir, 1978). Under oxidising conditions and at low pH (<5), the uranyl ion (UO_2^{2+}) dominates while at higher pH values, highly soluble carbonate complexes are formed with uranium (UO_2CO_3) which can significantly enrich dissolved uranium levels (Langmuir, 1978). Under reducing conditions, the sparingly soluble uraninite (UO_2) dominates and the aqueous uranium concentration can be quite low (Langmuir, 1978). Elevated uranium concentration in groundwater can be attributed to high uranium contents in coal bearing strata (Wu et al., 2014b). During unconventional gas development activities, produced water is removed from the target gas layer and may contain uranium concentrations much higher than overlying aquifers (Dahm et al., 2011; Khan and Kordek, 2013).

In this paper, we report groundwater radon and uranium levels in shallow aquifers overlying a target coal seam gas formation in order to detect possible migration of radon or uranium into overlying shallow aquifers. Our results establish baseline groundwater conditions prior to the development of gas fields and should allow for comparisons to be made if gas mining is established.

2. Methods

2.1. Study site

Groundwater samples were collected from the Richmond River Catchment (RRC) (Fig. 1), a sub-tropical coastal catchment located in northern New South Wales (NSW), Australia and spanning an area of almost 7000 km^2 (DPI, 2012). The upland ranges bordering the catchment remain mostly forested while agricultural activities dominate the lower coastal floodplains. The Richmond River extends for 170 km past several townships, before meeting the Pacific Ocean at Ballina. The region experiences high annual rainfall along the coastal fringe (1800 mm) and inland areas (1200 mm), with 65% of annual rainfall received during the wet summer and autumn months (December to April) (Atkins et al., 2013 and references therein). Intense rain periods during summer/autumn and prolonged dry periods during winter, contribute to contrasting flow characteristics within the catchment.

Complex geological sequences of Mesozoic consolidated sediments overlain by Cenozoic volcanics and Quaternary sediments occur throughout the catchment (Drury, 1982; McElroy, 1962) (Fig. 1). The East Richmond Fault is located in the western catchment. The Walloon Coal Measures (WCM), the CSG target layer, contains substantial CSG reserves; therefore, the region has potential for CSG development. Although almost 50 CSG exploratory wells (depths from ~620 m to 1520 m) have been drilled, operations ceased in March 2013. This work focuses mainly on the shallow aquifer system comprised of Quaternary sediments, Cenozoic basalts and Late Jurassic/Early Cretaceous sandstone bedrock in the northern RRC (Atkins et al., 2015 and references therein) (Fig. 1). Detailed groundwater information regarding hydrochemistry, other dissolved gasses and carbon stable isotopes, are reported in our companion paper Atkins et al. (2015).

2.2. Experimental approach

Between May and September 2013, a total of 91 groundwater samples were collected in the RRC from governmental monitoring bores (40) and private bores (51), allowing analysis of spatial variability within, and between, geological units. To ensure a representative groundwater sample, three bore volumes were purged prior to sampling, and stable temperature, conductivity and pH measurements were attained prior to sampling (Sundaram et al., 2009). The NSW Office of Water Bore Registry provided bore data logging records which were used to ascertain the aquifers from where groundwater was extracted. Regional geological maps (Brown et al., 2007; Henley et al., 2001) were used when the bore registry provided inadequate information (1 sample). Groundwater bore depths ranged between 5 m and 120 m and no groundwater samples were collected from the WCM (the CSG target layer).

Samples were collected for radon, uranium, dissolved inorganic carbon (DIC), pH, specific conductivity, temperature, dissolved oxygen (DO), total iron (Fe) and major ions ((calcium (Ca^{2+}), magnesium (Mg^{2+}), potassium (K^+), sodium (Na^+) and chloride (Cl^-)). To ensure sampling technique consistency, duplicate samples were collected every 20 sites. Uranium samples were collected in sample rinsed polypropylene syringe (60 mL) and filtered through a 0.45 μm syringe filter in to a polypropylene vial (10 mL). Uranium samples were analysed using a Perkin Elmer NexION 300D ICPMS using an iridium internal standard at a concentration of 20 ppb, resulting in an analytical uncertainty of approximately 6%. Discrete radon samples were collected in 6 L airtight HDPE plastic bottles (Stringer and Burnett, 2004) and measured using a RAD-7 (Durridge Co.) radon-in-air monitor (Burnett et al., 2001). The radon sample bottle, RAD-7 and Drierite column (to maintain humidity below 10%) were fitted in a closed air loop where radon was circulated via an internal RAD-7 pump. Radon equilibration (approximately 50 min for 6 L samples) between the air and water phase was achieved by degassing and recirculating radon through the closed air loop according to a method outlined by Lee and Kim (2006). Radon samples were counted for at least 2 h, with the second hour of values used to calculate radon concentrations, resulting in radon concentrations with counting uncertainties better than 5%. Samples for DIC, pH, HCO_3^-, specific conductivity, temperature, DO, Fe and major ions were collected and analysed as outlined in a previous

Download English Version:

https://daneshyari.com/en/article/1737750

Download Persian Version:

https://daneshyari.com/article/1737750

Daneshyari.com