



Contents lists available at ScienceDirect

Applied Geochemistry

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

Alpha-emitting isotopes and chromium in a coastal California aquifer

Jill N. Densmore^{a,*}, John A. Izbicki^b, Joseph M. Murtaugh^c, Peter W. Swarzenski^d, Thomas D. Bullen^e^a US Geological Survey, Placer Hall, 6000 J St., Sacramento, CA 95819, United States^b US Geological Survey, 4165 Spruance Rd., San Diego, CA 92101, United States^c CHMM, MCIWEST-MCB CPEN, Box 555008 (Bldg 22165), MCB Camp Pendleton, CA 92055-5008, United States^d US Geological Survey, 400 Natural Bridges Dr., Santa Cruz, CA 95060, United States^e US Geological Survey, 345 Middlefield Rd., Menlo Park, CA 94025, United States

ARTICLE INFO

Article history:

Available online 16 October 2014

Editorial handling by M. Kersten

ABSTRACT

The unadjusted 72-h gross alpha activities in water from two wells completed in marine and alluvial deposits in a coastal southern California aquifer 40 km north of San Diego were 15 and 25 picoCuries per liter (pCi/L). Although activities were below the Maximum Contaminant Level (MCL) of 15 pCi/L, when adjusted for uranium activity; there is concern that new wells in the area may exceed MCLs, or that future regulations may limit water use from the wells. Coupled well-bore flow and depth-dependent water-quality data collected from the wells in 2011 (with analyses for isotopes within the uranium, actinium, and thorium decay-chains) show gross alpha activity in marine deposits is associated with decay of naturally-occurring ²³⁸U and its daughter ²³⁴U. Radon activities in marine deposits were as high as 2230 pCi/L. In contrast, gross alpha activities in overlying alluvium within the Piedra de Lumbre watershed, eroded from the nearby San Onofre Hills, were associated with decay of ²³²Th, including its daughter ²²⁴Ra. Radon activities in alluvium from Piedra de Lumbre of 450 pCi/L were lower than in marine deposits. Chromium VI concentrations in marine deposits were less than the California MCL of 10 µg/L (effective July 1, 2014) but $\delta^{53}\text{Cr}$ compositions were near zero and within reported ranges for anthropogenic chromium. Alluvial deposits from the nearby Las Flores watershed, which drains a larger area having diverse geology, has low alpha activities and chromium as a result of geologic and geochemical conditions and may be more promising for future water-supply development.

Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/3.0/>).

1. Introduction

Selected radionuclide activities and chromium concentrations are elevated in some wells in marine and alluvial deposits in a coastal California aquifer. Radiation may be contributed to water from naturally-occurring isotopes within the uranium, actinium, and thorium decay chains (Fig. 1). Gross alpha and gross beta measurements, intended as screening tools for rapid quantification of radioactivity in water, are sensitive to the combined activity of a number of isotopes. However, these methods do not measure (1) volatile isotopes such as radon, and (2) gross beta measurements can be affected by ingrowth of radioactive progeny between sample collection and analyses (Thomas et al., 1993). Radon-222 (²²²Rn), an alpha-emitter within the uranium decay-chain, poses the largest risk to human health of naturally-occurring radionuclides (National Research Council, 1999), but other naturally-occurring radioactive isotopes such as uranium, and radium also have Maximum Contaminant Levels (MCL's) for drinking water

established by the U.S. Environmental Protection Agency (<http://water.epa.gov/drink/contaminants/#List>, accessed December 3, 2013). Understanding the chemistry and distribution of isotopes within the uranium, actinium, and thorium decay-chains allows increased understanding of the sources of radiation in water from wells, and facilitates planning future water supply development and management practices intended to limit these constituents in wells.

1.1. Uranium, other radionuclides, and chromium occurrence in groundwater

Uranium is soluble in oxic groundwater and insoluble in reduced groundwater (Rai and Zachara, 1984; Szabo and Zapeczka, 1991). In the absence of complexing ions, uranium in oxic groundwater is present in the +6 valence as the uranyl (UO_2^{2+}) ion. In this form, the solubility of uranium is limited by sorption to iron coatings on mineral grains, clay mineral-edge structures, and organic material (Langmuir, 1978). Uranyl complexes with fluoride, phosphate, or most commonly with inorganic carbon (carbonate) enhance uranium solubility (Langmuir, 1978; Hsi and Langmuir,

* Corresponding author. Tel.: +1 916 278 3163; fax: +1 916 278 3070.

E-mail address: jidensmo@usgs.gov (J.N. Densmore).

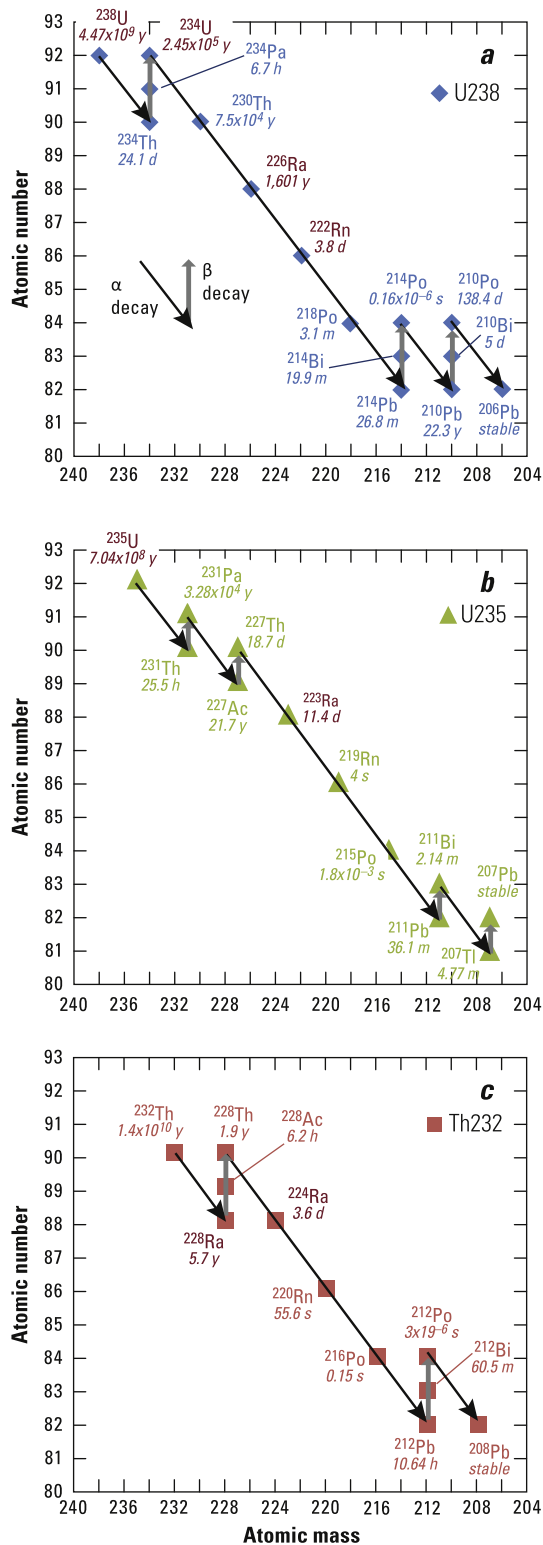


Fig. 1. Decay series for (a) Radon (Uranium-238), (b) actinium (Uranium-235) and (c) thorium (Note: isotopes measured as part of this study are shown in red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

1985; and Jurgens et al., 2009) and the presence of calcium can enhance desorption of uranyl-carbonate complexes from aquifer material (Fox et al., 2006).

There are two naturally-occurring primordial isotopes of uranium, uranium-238 (²³⁸U) and uranium-235 (²³⁵U), and a

naturally-occurring radiogenic isotope uranium-234 (²³⁴U), formed as part of the uranium decay series (Fig. 1). The half-lives of these isotopes are long: 4.47×10^9 , 7.04×10^8 , and 2.45×10^5 years, respectively (Osmond and Cowart, 2000). ²³⁸U, the most abundant isotope, composes about 99.275% of crustal uranium; ²³⁵U, the fissionable form used in reactors and nuclear weapons, composes about 0.72% of the crustal uranium; while ²³⁴U, a daughter of ²³⁸U decay, composes only about 0.005% of the crustal abundance. Uranium isotope abundance is measured as an activity in picoCuries per liter (pCi/L). Despite the difference in crustal abundance, the activity ratio (AR) of ²³⁸U and ²³⁴U in sediments is commonly near 1 as a result of secular equilibrium, a steady-state condition where the rate of production of a shorter-lived daughter isotope equals the rate of decay of the longer-lived parent isotope. Departures from this ratio result from a combination of alpha-recoil and specific geochemical conditions within aquifers. Activity ratios characteristic of groundwater from different settings can be used to evaluate geochemical reactions and the hydrologic history of groundwater (Osmond and Cowart, 1976 and Osmond and Cowart, 2000).

In contrast to uranium, radium in the +2 valence is strongly sorbed and insoluble in oxic groundwater (Szabo and Zapeczka, 1991). Radium is soluble in acidic, anoxic, chloride-rich groundwater with elevated total dissolved solids (Tanner, 1964; Dickson, 1985; Szabo and Zapeczka, 1991). There are four naturally-occurring radium isotopes. Radium-226 (²²⁶Ra), within the uranium decay-chain (Fig. 1), is the longest lived radium isotope having a half-life of about 1,600 years. Radium-223 (²²³Ra), within the actinium (²³⁵U) decay-chain (Fig. 1b), has a half-life of 11.4 days. Radium-228 and radium-224 (²²⁸Ra and ²²⁴Ra, respectively) within the thorium decay-chain have half-lives of 5.7 years and 3.6 days, respectively. ²²⁸Ra is a beta-emitter and does not contribute to gross alpha activity. However, the MCL for combined activity of ²²⁸Ra and ²²⁶Ra is 5 pCi/L. Consistent with the low abundance of its parent isotope ²³⁵U, ²²³Ra activity is usually much lower than the activity of the other radium isotopes.

Radon is a noble gas and, although soluble, radon is otherwise non-reactive in groundwater (Szabo and Zapeczka, 1991). As previously discussed, of the naturally-occurring radionuclides, radon poses the largest risk to human health (National Research Council, 1999). ²²²Rn is part of the uranium decay-chain that includes ²³⁸U, ²³⁴U and ²²⁶Ra (Fig. 1). ²²²Rn is produced directly by alpha decay of ²²⁶Ra and has a half-life of 3.8 days. Radium is insoluble and radon in solution is released from the solid phase as a result of direct recoil during alpha decay, and to a lesser extent by indirect recoil and subsequent diffusion (Cecil and Green, 2000). Because of its relatively short half-life, ²²²Rn is reflective of geologic conditions near the sample collection location, and ²²²Rn activities can differ greatly over short distances even within the same geologic unit (Cecil and Green, 2000). Radon-219 and radon-220, produced as part of the actinide and thorium decay chains, have half-lives of 4 and 56 s, respectively—too short for most hydrologic applications (Cecil and Green, 2000).

Chromium and uranium have similar aquatic chemistry, in that both elements are present in the +6 valence, soluble under alkaline, oxic conditions, and insoluble under reduced conditions (Rai and Zachara, 1984). In that in alkaline, oxic groundwater chromium is present as the oxyanion form, CrO_4^{2-} , while uranium is present as the uranyl ion, or as uranyl complexes with either fluoride, phosphate, or most commonly with carbonate ions (Langmuir, 1978; Hsi and Langmuir, 1985; and Jurgens et al., 2009). Chromium is higher in mafic rocks and uranium is higher in granitic rock (Reimann and de Caritat, 1998). Although these trace elements are not commonly associated with the same geologic materials or hydrologic processes, co-occurrence of chromium and uranium has been observed in unconsolidated sand and gravel

Download English Version:

<https://daneshyari.com/en/article/6335108>

Download Persian Version:

<https://daneshyari.com/article/6335108>

[Daneshyari.com](https://daneshyari.com)