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Geological and hydrogeochemical controls on radium isotopes in groundwater of the Sinai Peninsula, Egypt

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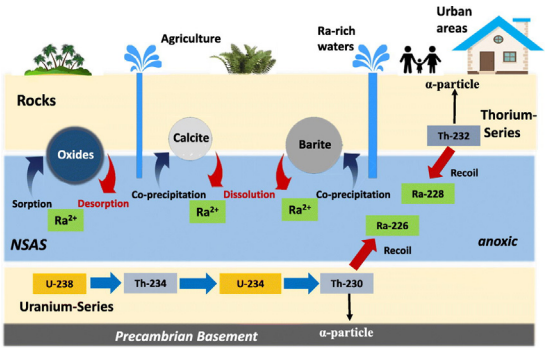
HIGHLIGHTS

- Radium isotopes were investigated in groundwater of the Sinai Peninsula, Egypt.
- Radium levels are > the US EPA, EU, and WHO maximum contaminant levels.
- Mechanisms controlling mobility of radium isotopes in the aquifer were considered.
- Groundwater solute composition and aquifer lithology are important.
- Potential health implications of groundwater utilization are discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Radium isotopes (^{226}Ra and ^{228}Ra) were analyzed in 18 groundwater samples from the Nubian Sandstone Aquifer System (NSAS) and the shallow alluvial aquifers overlying the basement complex of the Sinai Peninsula, Egypt. Groundwater samples from deep Nubian aquifer wells (total depths 747 to 1250 m) have ^{226}Ra and ^{228}Ra activities ranging from 0.168 to 0.802 and 0.056 to 1.032 Bq/L, respectively. The shallower Nubian aquifer wells (63 to 366 m) have ^{226}Ra and ^{228}Ra activities ranging from 0.033 to 0.191 and 0.029 to 0.312 Bq/L, respectively. The basement shallow alluvial aquifers have ^{226}Ra and ^{228}Ra activities ranging from 0.014 to 0.038 and 0.007 to 0.051 Bq/L, respectively. Combined Ra activities in most wells were generally in excess of the US Environmental Protection Agency (EPA), the European Union (EU), and the World Health Organization (WHO) maximum contaminant levels (MCL) for drinking water. Radium in groundwater is produced mainly by decay of parent nuclides in the aquifer solids, and observed activities of dissolved Ra isotopes result from a combination of alpha-recoil, adsorption/desorption, co-precipitation/dissolution processes. The observed correlation between Ra activities and salinity indicates that adsorption/desorption processes may be the dominant factor controlling Ra mobility in Sinai groundwater. Radium activities in central and northern Sinai are generally higher than those in southern Sinai, consistent with a gradual increase in salinity and water-rock interaction with increasing groundwater age. Barite is approximately saturated in the groundwater and may limit maximum dissolved Ra

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concentration. The results of this study indicate that Sinai groundwater should be used with caution, possibly requiring Ra removal from water produced for domestic and agricultural consumption.

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1. Introduction

Radionuclides occur naturally in drinking water supplies and their activities vary according to the source of the water (Milvy and Cothern, 1990). The common radionuclides in groundwater are from the uranium and thorium decay series. Radium has been identified as the most potentially hazardous naturally occurring radionuclide likely to occur in drinking water (Gilkeson et al., 1983; Cothern and Lappenbusch, 1984; Kay, 1999). Four Ra isotopes are commonly found in the environment: ^{226}Ra (half-life = 1600 years), ^{228}Ra (half-life = 5.8 years), ^{223}Ra (half-life = 11.4 days), and ^{224}Ra (half-life = 3.6 days). The two Ra isotopes of most concern in drinking water are ^{226}Ra and ^{228}Ra owing to their relatively long mean lifetimes compared to the others. ^{226}Ra is the daughter of ^{230}Th (half-life = 7.52×10^4 a) in the ^{238}U series and decays by alpha emission. ^{228}Ra is the daughter of ^{232}Th (half-life = 1.39×10^{10} a) series and decays by beta emission.

Natural radioactivity is increasingly being recognized as a limiting factor in the potability of groundwater reserves in the Middle East. An investigation of fossil groundwater from the Lower Cretaceous Nubian sandstone (Kurnob Group) and the overlying Upper Cretaceous carbonate (Judea Group) aquifer in the Negev and Arava Valley, Israel, revealed Ra activities above the MCL for Ra (Vengosh et al., 2007). Moreover, high levels of natural radioactivity in excess of the MCL, primarily from Ra isotopes and Rn, were reported in groundwater from the Nubian sandstone (Rum Group) of the Disi aquifer in Jordan (Vengosh et al., 2009).

Distribution of Ra isotopes between groundwater and aquifer solids is generally related to groundwater chemistry and to the concentration and distribution of the U and Th parent isotopes within the solids (Ku et al., 1992; Sturchio et al., 2001). Uranium forms soluble U(VI) carbonate complexes under oxic conditions, and thereby it can be transported by groundwater over long distances. Under anoxic conditions, U(IV) precipitates from groundwater and can form secondary U deposits. Consequently, ^{226}Ra is widely distributed and tends to be highly concentrated at sites where U is enriched or in carbonate rocks. In contrast, Th is insoluble and generally immobile in groundwater. As a result, ^{228}Ra is directly controlled by the local distribution of Th in the aquifer materials (Michel, 1990).

In this paper, we present the data for the long-lived Ra isotopes in groundwater from shallow alluvial aquifers in granitic basement and from the Nubian sandstone aquifer throughout the Sinai Peninsula, Egypt. We examine the relations between Ra activities and aquifer characteristics to better understand the geological and geochemical controls on the distribution and behavior of Ra isotopes in groundwater of the Sinai Peninsula. In addition, we address the potential radiological hazard presented by using untreated groundwater in the Sinai Peninsula for drinking water or irrigation.

2. Regional geology and hydrogeology

The Sinai Peninsula is a 61,000 km^2 land area located in northeastern Egypt between latitudes 27°43′ and 31°19′ North and longitudes 32°19′ and 34°54′ East. It is bound by the Gulf of Aqaba to the east, the Gulf of Suez to the west, and the Mediterranean Sea to the north. The peninsula is separated from the African continent on the west by the Gulf of Suez extensional rift system, and from the Arabian Peninsula on the east by the sinistral Gulf of Aqaba fault system.

The Sinai can be divided into three physiographic districts: (1) exposed Precambrian basement forming an intricate complex of high and rugged igneous and metamorphic mountains at the southern tip of the peninsula; (2) a dissected plateau consisting of Mesozoic and Cenozoic limestones, dolostones, and sandstones in the central part of the peninsula; and (3) a sandy plain that parallels the coastline in the north (Jenkins, 1990; Kusky and El-Baz, 1999) (Figs. 1 and 2).

The Nubian Sandstone (NS) formation is the main source of water for the Sinai Peninsula. It is a part of the Nubian Sandstone Aquifer System (NSAS) of northeast Africa that constitutes the world's largest known fossil water system (Thorweihe, 1990). The NSAS in Sinai is mainly composed of ferruginous sandstone with shale and clay intercalations, having a thickness that ranges between 140–230 m. In Sinai, the NSAS extends laterally in the subsurface for approximately 10,000 km^2 from the northern margin of the Precambrian basement in central Sinai to the Gulf of Suez in the west and the Dead Sea in the northeast. The aquifer is recharged through sandstone outcrops at the foothills of the Precambrian basement complex in central Sinai and to a lesser extent through erosional windows in Sinai and Negev (Isaar et al., 1972; Sultan et al., 2011) (Figs. 1 and 2).

3. Material and methods

Fieldwork was conducted in January and June of 2010 to sample groundwater from drilled wells which tap the NSAS, and from drilled and open-pit wells tapping alluvial aquifers overlying fractured basement in southern Sinai. The sampled wells were distributed over the northern, central, and southern parts of the Sinai Peninsula and along the Gulf of Suez coastal zone (Fig. 1). The samples from the NSAS groundwater wells are divided into two groups on basis of total depth (TD), depth to static water level (DWL) and proximity to recharge areas. The deep NSAS group samples were collected from 11 wells with a wide range in total depth (TD) and depth to water level (DWL) (TD: 747–1250 m; DWL: 137–377 m), and from a spring (Ayun Musa), at various distances from the recharge area (Fig. 1). The shallow NSAS group samples were collected from four wells (TD: 63–366 m; DWL: 19–56 m) that are located close (<40 km) to recharge zones (Fig. 1). In addition, three samples were collected from shallow wells tapping alluvial aquifers in the basement complex (Fig. 1).

For analysis of ^{226}Ra and ^{228}Ra, 25 L of water was collected from each groundwater well and poured into a large open container prior to extraction of radium by adsorption on Mn-oxide coated acrylic fiber (Moore and Reid, 1973; Kim et al., 2001; Dulaiova and Burnett, 2004). Water was aerated during filling of the container to cause degassing and oxidation to help ensure quantitative extraction of Ra by the Mn-fiber as recommended by Dulaiova and Burnett (2004). Turbidity was negligible and water was not filtered. The water was passed slowly (<1 L per minute) by gravity feed through ¼-inch plastic tubing into the inlet of a 100 cm^3 flow-through cartridge containing 14 g of Mn-coated acrylic fiber (Scientific Computer Instruments, Columbia, SC). The Mn-fiber adsorbed Ra from the water and after draining and removal from the cartridge it was transferred to a labeled plastic zip-loc bag for transport to the laboratory. Extraction efficiency was not evaluated in this study, nor were duplicate samples collected because of logistical constraints in the field. The sampling procedure has been used extensively for seawater and groundwater and found to be quantitative when flow rate through the Mn-fiber column is maintained at <1–2 L per minute (Kim et al., 2001; Dulaiova and Burnett, 2004). In case of incomplete extraction, Ra activities could be somewhat higher than those reported, but ^{228}Ra/^{226}Ra activity ratios would not be affected.

The Mn-fiber samples were sealed in labeled aluminum containers and Ra isotope activities were measured by gamma spectrometry

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