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Using radon-222 and radium-226 isotopes to deduce the functioning of a coastal aquifer adjacent to a hypersaline lake in NW Iran

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ABSTRACT

This study aims to assess the hydrogeochemistry of coastal groundwater, the occurrence of ^{222}Rn and ^{226}Ra, and their isotopic response to salinity and associated chemical compositions of groundwater in the coastal Urmia Aquifer (UA) at the western side of Urmia Lake (UL). The results of the PCA show that 87.3% of groundwater chemistry changes are controlled by six principal components. The interaction between groundwater and coastal igneous and metamorphic rocks in eastern areas (next to the UL) results in complex hydrogeochemical conditions than western areas. Based on correlation of U and salinity, some coastal samples display conservative and the others non-conservative behaviors. Differed from most previous studies, ^{226}Ra and ^{222}Rn concentrations in coastal groundwater samples of UA do not show a good correlation with salinity. Given 10% of groundwater ^{222}Rn is originated from host rocks, the radon concentrations recorded in the coastal groundwater samples are relatively in range that can effectively be supplied by the local rocks (5–49 Bq/l). Results of different chemical and isotopic parameters in this area indicate that there is no direct connection between fresh groundwater and UL saltwater. This is because that the hard and thick salty layer in the lakebed acts as an impermeable barrier to prevent the underground hydraulic connection. Results show that removing the salty layer of UL as an option to progress in rehabilitation program of this lake may result in more hydraulic connection between the lake and groundwater resources in some areas.

1. Introduction

The groundwater salinization is a globally significant phenomenon; in many areas salinization occurs due to high groundwater demand where saline or salt groundwater is presented at depth or where salinity intrudes from adjacent surface water resources. Overexploitation of coastal groundwater may result in mix of sea/salt water, fossil brines, agricultural effluents, and sewage. In addition to the increase of total dissolved solids (TDS), which can lead to the abandonment of wells, salinization can intensify water-rock interaction and lead to high values of naturally occurring trace elements and radionuclides such as radium (Ra) and radon (Rn), and further damage water quality (Vinson et al., 2013).

These radionuclides may get into groundwater due to water-rock interaction process. However, it is expected that the ground/surface waters in some aquifers have higher values of one or more of the natural radionuclides, depending on the water chemistry, the nature of the aquifer host rocks such as the natural radioactivity content of the involved rock formation (Shabana and Kinsara, 2014).

Radium-226 (^{226}Ra) and radon-222 (^{222}Rn) are two of the most common naturally occurring radionuclides in groundwater. The longest-lived isotope of radium is ^{226}Ra with a half-life of 1600 years, which is produced by the alpha (α-) decay of ^{230}Th ($T_{1/2}$ = 75,200 yrs) in the decay chain of ^{238}U. Radium has 33 known isotopes from ^{202}Ra to ^{234}Ra. The most stable isotope of radon is ^{222}Rn (with a half-life of 3.8235 days), which is a decay product of ^{226}Ra. There are 39 known isotopes of radon from ^{193}Rn to ^{231}Rn, all of which are radioactive (Roba et al., 2012). Radon-222 is one of the densest gas nuclides under normal condition. The main sinks for ^{222}Rn are radioactive decay and loss to the pore space above groundwater level due to degassing (Kiro et al., 2015). ^{222}Rn is soluble in water and can be incorporate to groundwater flows. The activity of ^{222}Rn in groundwater depends on various factors such as aquifer characteristics, residence time of water in aquifer media, ^{226}Ra content of rocks, etc. (Moreno et al., 2014). High activities of ^{222}Rn are common in groundwater and are usually about 5–10% of the total radon produce by the decay of ^{238}U and its immediate parent ^{226}Ra in aquifer grains (Kiro et al., 2015).

The surface water bodies such as lakes and rivers are normally

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depleted in ^{222}Rn (Al-Masri and Blackburn, 1999; Cosma et al., 2008; Somlai et al., 2011). On the other hand, some studies (e.g., Cook et al., 2003; Dimova and Burnett, 2011; Luo et al., 2016) showed that very low concentrations of ^{222}Rn is detectable in most river and lake water bodies, and it can be used as an ideal tracers to quantify groundwater discharge in surface water bodies. ^{222}Rn is concentrated in groundwater, and uranium-rich crystalline rocks (Przylibski et al., 2004; Voronov, 2004; Skeppstrom and Olofsson, 2007; Freiler et al., 2016).

Concentrations of radium and radon in groundwater and their spatio-temporal variability have been investigated frequently in recent years (Ku et al., 1992; Luo et al., 2000, 2016; Tricca et al., 2000; Cook et al., 2003; Porcelli, 2008; Dimova and Burnett, 2011). The results of these studies are very important for understanding radon movement processes in the lithosphere and the role of groundwater as a carrier fluid (Przylibski et al., 2004). It is also very important to identify the role of geological structures and rock types on the occurrence of groundwater ^{222}Rn. The groundwater often moves through rock and soil containing radon and radon gets solubilized in the water (Duggal et al., 2014; Shivakumara et al., 2014).

The worldwide distribution of radium and radon in groundwater is linked with the specific geological characteristics of the formations. High radium and radon concentrations are mostly found in groundwater in the fractured system where groundwater is intensively interacted with host rock (Moreno et al., 2014).

It is documented that the concentrations of radium and radon in the groundwater are mostly controlled by uranium and thorium contents of the aquifer materials. In addition to the proved role of uranium and thorium contents of the aquifer in controlling the radium and radon activities in groundwater (Zukin et al., 1987; Isam et al., 2002), the groundwater chemistry may be impart as a secondary control on concentration of these radionuclides in the aquifers (Sturchio et al., 2001; Vinson et al., 2009; Vengosh et al., 2009).

Mobility and adsorption of radium can be influenced by various physico-chemical parameters such as increased TDS values, high salinity (Sturchio et al., 2001), oxidation/reduction potential (Beaucaire and Toulhoat, 1987; Szabo et al., 1992), or low pH (Riese, 1982; Portle, 1987; Szabo et al., 1992). Certain chemical processes-common ion effect (Miller and Sutcliffe, 1985; Sturchio et al., 2001), cation exchange process (Appello and Postma, 1993; Vengosh et al., 2009), oversaturation with respect to barite (Glundl and Cape, 2006), adsorption on iron (Fe) and manganese (Mn) oxides and hydroxides (Vinson et al., 2009) or on organic matter like humic substances (Buckau et al., 2005) may influence the mobility of radionuclides in the environment. However, the mechanisms controlling radionuclides mobility are still not fully understood (Roba et al., 2012).

Radium-226 and ^{222}Rn isotopes have been frequently used as tracers of groundwater discharge in several environments, including lakes (Kluge et al., 2007), streams (Burnett et al., 2010), coastal lagoons (Santos et al., 2008; Su et al., 2012) or wetlands (Charette et al., 2003; Cook et al., 2008; De Weys et al., 2011; Rodellas et al., 2012). However, only a few studies assessed the role of saline waters in radioactivity changes of coastal groundwater (e.g., Almeida et al., 2004; Condomines et al., 2012).

The actual mechanisms defining the radium and radon movement in salinized waters are not well-understood (Swarzenski et al., 2013). A more detailed understanding of the radium-radon-salinity relationship would consider factors such as other interacting ions introduced by water-rock interaction, redox-sensitive removal mechanisms, and estimation of salinity thresholds at which radium and radon behavior is not approximated by the simple dilution of saline waters (Vinson et al., 2013).

This study has been carried out in Urmia Aquifer (UA), in the west of Urmia Lake (UL), Iran. This lake, as one of the top-ten largest saline lakes in the world, has faced extreme water loss in recent years due to overuse and mismanagement. It is, in fact, a hypersaline lake with a unique aquatic ecosystem for which it has been designated as a Biosphere Reserve by UNESCO and a National Park under the 1971 Ramsar Convention. Over the last thirty years, the population in the lake basin has been doubled and the agricultural area fed by water resources of the lake basin has tripled. In UL basin, precipitation decreases by 18% compared to its long-term record. Therefore, inflows to the lake have decreased drastically and the situation has been worsened by continuous droughts, resulting in reduction of renewable water resources and the lake's water levels at an alarming rate. The mean annual water level is currently more than four meters below the critical level (1274 m above sea level) needed to sustain ecosystems. In Oct 2015, the UL water level reached to the lowest level (1270 m) and the southern parts of it were entirely dried. The lake water level has plummeted about more than 7.84 m during the last 20 years due to a dramatic decline of its inflows which leads to a consequent increase in the salinity of the lake (from 166 g/l in 1995 to over 412 g/l in 2015) (Khatami, 2013; Amiri et al., 2016a). The recorded density of UL water in May 2015 was about 1.27 g/cm^3 (Amiri et al., 2016a,c).

Urmia aquifer (UA) is the largest groundwater resource connected to UL. Investigations of potential saltwater intrusion into UA were begun coinciding with the launch of Urmia Lake Restoration National Committee (ULRNC) mission to develop a roadmap to revive this lake. Amiri and his colleagues have conducted a series of studies in this area. Amiri et al. (2016b) investigated the salinization and freshening processes of groundwater through major ion and trace element indicators and reported that the percentage of saltwater in the groundwater samples of UA is very low, ranging between 0.001% and 0.79% in the wet season and 0.0004% and 0.81% in the dry season. Amiri et al. (2016a) by assessment of potential saltwater intrusion into UA revealed that most groundwater samples collected from the same wells during wet and dry seasons have the same chemical type, which indicates that the groundwater chemistry are not highly affected by seasonal changes. In Iran, research on using radium and radon as tracers for investigating the hydrogeology and water chemistry is very rare. This study aims to assess the hydrogeochemistry of coastal groundwater, identify the occurrence of ^{222}Rn and ^{226}Ra, and isotopic response to salinity and associated chemical compositions of groundwater in the UA. We wish to find the major mechanisms controlling the geochemistry of groundwater and assess the role of different factors on ^{222}Rn and ^{226}Ra distribution and existence in the coastal waters.

2. Materials and methods

2.1. Description of study area

This region is located between the eastern longitude of 44°, 20′ and 45°, 20′ and northern latitude of 37°, 05′ and 38°, 05′ (Fig. 1). The underground aquifer of this plain is a large natural collecting reservoir and the regulator of water inflowing from the large drainage area, which both retains and enables its useful utilization. The groundwater flow direction is from west to east (UL). Hydrological investigations have shown that this underground reservoir spreads under an area of approximate 868 km^2 and consists of confined and unconfined aquifers (Badv and Deriszadeh, 2005). Nazlu-Chai, Rowzeh-Chai, Shahr-Chai and Baranduz-Chai are the four main rivers, which are flowing in the plain. They originate from the western mountains and end in UL.

The oldest rock units of the Pre-Cambrian are formed by meta volcanic series, acidic tuff and diorite in the surrounding mountains of UA, as well as metamorphous amphibolites and gneiss. In this area, the Ophiolit units consist of basic and ultra-basic rocks with schist and radiolarite limestone. Tertiary rocks in this plain are represented by limestone, conglomerate, sandstone, and shale (Kamei et al., 1973).

This area, according to the Global Bioclimatic Classification System, is in a Mediterranean pluviseasonal-continental climate (Martinez et al., 1999). During recent 30-year period, mean annual temperature and precipitation in the area are 11.52 °C and 346.3 mm, respectively, while mean maximum and minimum temperature occur in July (31.2 °C) and