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Groundwater transport and radium variability in coastal porewaters

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Radium isotopes (^{223}Ra, $t_{1/2} = 11.4$ d; ^{224}Ra, $t_{1/2} = 3.66$ d; ^{226}Ra, $t_{1/2} = 1600$ y; and ^{228}Ra, $t_{1/2} = 5.75$ y) are considered excellent tracers of groundwater movement and discharge in coastal systems. However, spatial and temporal variability in porewater radium activity have raised questions about the accuracy of these tracers. To better understand the factors affecting radium variability in coastal systems, measurements of porewater and surface water radium activity were made at an island in North Inlet Salt Marsh in Georgetown, South Carolina, from November 2009 to February 2011. Water salinity, temperature, pH, and redox potential were also recorded, and sediment samples were collected for analysis of bulk ^{228}Ra and ^{226}Ra activity. Hydraulic head observations during 2007–2008 from piezometers on the island were used to generate independent estimates of groundwater fluxes.

Porewater radium activities decreased with depth below the marsh surface and increased with distance from the creek banks. Salinity measurements were lower and redox potential higher near the marsh creeks. The stratigraphy of the island is typical of intertidal wetlands in the southeastern U.S., with a mud layer overlying a confined sandy aquifer; the observed patterns in porewater radium, salinity, and redox potential were consistent with (1) shorter porewater residence times in the permeable sand aquifer than in the low-permeability mud, (2) differences in grain size between the mud and sand, and (3) greater tidal exchange near the creeks. Temporal variations in porewater radium activity were not associated with salinity, pH, and redox potential although temperature provided significant control ($P < 0.05$, $r^2 < 0.47$) over variations in ^{228}Ra and ^{226}Ra activity. Lower mean sea water levels resulted in greater calculated groundwater discharge and were also associated with lower average porewater ^{224}Ra and ^{223}Ra activity, in that groundwater discharge variations strongly affected short-lived radium activity at this site. The $^{228}Ra/^{226}Ra$ activity ratios in the surface water and porewater signified that the confined aquifer, rather than the surficial mud, was the primary source of radium to the surface water. Our results highlight the importance of understanding the hydrology of any coastal system when interpreting radium results. It is also essential to identify and measure the correct porewater end-member(s) (i.e. source aquifers) when calculating radium budgets.

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1. Introduction

Groundwater discharge is an increasingly acknowledged and important source of solutes to coastal surface waters (King et al., 1982; Valiela et al., 1990; Rutkowski et al., 1999; Burnett et al., 2001; Kelly and Moran, 2002; Jahnke et al., 2003; Slomp and Cappellen, 2004; Moore, 2006; Kwon et al., 2014). However, because groundwater discharge is diffuse and spatially variable, it is not easy to directly measure. Radium (Ra) has been used with increasing frequency as a geochemical tracer useful for calculating spatially-integrated estimates of groundwater discharge to coastal waters. The power of this tracer has resulted in a large number of studies using radium-based estimates of groundwater discharge to estimate groundwater-borne solute and nutrient fluxes (Bollinger and Moore, 1984; Rama and Moore, 1996; Hancock et al., 2000; Krest et al., 2000; Charette et al., 2001; Kelly and Moran, 2002; Abraham et al., 2003; Charette et al., 2003; Krest and Harvey, 2003; Charette and Buesseler, 2004; Kim et al., 2005; Charette, 2007; Knee et al., 2008; Gonneea et al., 2013; Kwon et al., 2014).

Radium is useful as a naturally-occurring tracer in coastal systems for several reasons (Moore, 1987; Webster et al., 1994; Charette et al., 2001; Moore, 2003; Gonneea et al., 2008). Ra isotopes are generated from their respective parent isotopes either

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within or adsorbed to aquifer solids. The distribution coefficient (K_D) of Ra, the ratio of radium adsorbed to sediment surfaces to radium in solution, decreases with increases in salinity. This leads to higher radium activities in saline coastal porewater. A range of coastal sediment flushing and circulation processes are encompassed by the half-lives of ^{224}Ra and ^{223}Ra (3.66 and 11.4 days), and the half-lives of ^{228}Ra and ^{226}Ra (5.8 and 1600 years) allow them to be considered quasi-conservative tracers. This also means that the activities of ^{228}Ra and ^{226}Ra, once separated from the source aquifer, are affected primarily by dilution in the surface water. Therefore, the ratio of ^{228}Ra/^{226}Ra is largely unaffected because the average residence time of coastal surface waters is much shorter than the half-lives of these isotopes. This allows the source aquifers of water and solutes to coastal surface water to be traced by comparing ^{228}Ra/^{226}Ra activity ratios in groundwater and surface water (Moore, 2003).

Radium-based groundwater discharge studies in coastal systems over the past 30 years have revealed that the final discharge estimate is highly sensitive to variability in measurements of groundwater radium activity. Activity ($N\lambda$) is defined as the number of radium atoms (N) that decay within a specific amount of time (λ t^{-1}). Using radium as a tracer, estimates of groundwater discharge are made by measuring radium activity in the surface water of an embayment of interest and in the nearby ocean and groundwater as end-members. Excess radium in surface water (the contribution exclusively from groundwater inputs) is determined by eliminating radium inputs from other potential sources. These additional sources include generation of radium from parent isotopes within or adsorbed to surface sediment; molecular and turbulent diffusion of radium from the interstitial water in shallow surface sediment; release of radium via ionic exchange from new sediment deposits to coastal, salt water systems; river discharge; and inputs from near shore surface water at each incoming tide. These measurements are then combined with a simple mixing model to obtain an estimate of groundwater discharge. That is, at what rate will groundwater need to discharge to support the excess radium measured in the surface water?

Radium activity in the porewater of a single aquifer was initially assumed to be constant in space and time, and any changes in excess surface water Ra activity were attributed to changes in groundwater discharge rates. Thus, porewater radium activity was measured only once, or multiple measurements were averaged to a single value for use in the mixing model along with a series of surface water samples collected over an extended period of time (Bollinger and Moore, 1993; Scott and Moran, 2001; Yang et al., 2002; Charette et al., 2003). It is now understood that radium activity in porewater can vary greatly in natural settings, which can produce high variance in radium-based discharge estimates (Gonneea et al., 2008; Hougham et al., 2008; Gonneea et al., 2013).

Radium activity in porewater is affected by five factors: (1) radioactive production in the sediment as a function of thorium (Th) content; (2) sediment grain size and surface area (Beck and Cochran, 2013), including a strong inverse relationship between sediment grain size and thorium content (Bollinger and Moore, 1993); (3) radioactive decay in the water; (4) groundwater transport (advection and dispersion) and discharge; and (5) sorption as controlled by porewater and sediment chemistry. Porewater radium activity controlled by generation and decay is isotope-dependent and based upon the half-lives of the Th parent isotopes as well as the individual Ra isotopes, which vary by several orders of magnitude. The porewater radium controls of groundwater transport and discharge, sorption, and differences in sediment grain size are universal across the four Ra isotopes. Measurements of radium activity in sediments include what can be considered immobile as well as mobile fractions. The immobile fraction of radium is generated from thorium held within the sediment grain matrix. This fraction has a greater probability of release into the porewater via alpha recoil if the thorium atom is close to the grain surface (Sun and Semkow, 1998). The mobile fraction may be defined in two parts. Radium may be generated from thorium adsorbed to the sediment grain surfaces or iron- and manganese-oxide grain coatings, and radium may itself be adsorbed to the grain surface or grain coatings.

Changes in porewater and sediment chemistry result in sorption or desorption of both thorium and radium from the sediment surface (mobile sediment radium fraction). Radium sorption to sediment and to Fe and Mn oxide/hydroxide sediment coatings is affected by variations in salinity, temperature, pH, and redox potential (Elsinger and Moore, 1980; King et al., 1982; Webster et al., 1995; Rama and Moore, 1996; Hancock et al., 2000; Gonneea et al., 2008; Beck and Cochran, 2013). In coastal aquifers, changes in salinity across the fresh water/salt water interface provide the greatest control on distributions of porewater radium (Krest et al., 2000; Gonneea et al., 2008) with increases in radium activity of up to two orders of magnitude as salinity increases from 0 to 25 (Abraham et al., 2003; Gonneea et al., 2008). Thus variations in radium in the surface water resulting from groundwater discharge can be very difficult to interpret in coastal systems with a mobile fresh water/salt water interface.

The distribution of Ra has also been shown to be variable in coastal aquifers without fresh water/salt water transition areas. Radium activity in porewater still varied by one order of magnitude in space and time in an intertidal salt marsh (North Inlet, South Carolina) where porewater salinity exceeded 17 and porewater was reduced (Krest et al., 2000). In that study, spatial variations in porewater radium were controlled by radium generation, decay, and sediment grain size. Temporal variations in porewater radium were interpreted to be changes in sorption controls (other than salinity) and variable groundwater transport.

Although elevated porewater salinity and reduced conditions are typical of tidal salt marshes along the southeast Atlantic coast (Wiegert and Freeman, 1990), seasonal redox changes in the shallow (0–25 cm) porewater and sediment at this site were reflected in porewater iron speciation changes observed to a depth of 100 cm (Hughes et al., 2012). Pyrite oxidation within the shallow marsh sediment during periods of extended marsh surface exposure (winter and early spring) were believed to be the cause of the iron speciation changes and be accompanied by lower porewater pH. However, reduced porewater conditions prevailed for most of the year.

We hypothesize that groundwater transport provides a primary control on porewater Ra distribution in saline systems with generally reduced porewater conditions. To test this hypothesis, we conducted a field and modeling study on a marsh island at North Inlet, South Carolina. High porewater salinities and typically reduced porewater conditions at this site mean that salinity and redox have the potential to be eliminated as factors affecting porewater radium variability. The overall goal of this study was to investigate the role of groundwater flow on the spatial and temporal distribution of porewater radium activity. In order to determine the controls on spatial and temporal variability, we measured bulk sediment radium activity within the marsh mud and underlying confined sand aquifer. Simultaneous measurements of radium, temperature, salinity, redox potential, and pH were made in the groundwater and surface water. Finally, to investigate the role of groundwater flow and residence time on porewater radium activity, we used previously measured hydraulic head at this site to estimate groundwater discharge rates.

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