TRACING THE MIXING AND MOVEMENT OF GROUND WATER INTO FLORIDA BAY WITH FOUR NATURALLY OCCURRING RADIUM ISOTOPES

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Abstract. Four naturally occurring isotopes of radium (^{223,224,226,228}Ra) have a range in half-life that extends from a few days to over 1,600 years. Unique geochemical attributes make these radium isotopes ideal to examine sediment/water interface exchange processes in coastal waters. Here we present initial radium isotopic data of Florida Bay, a heavily impacted coastal system in south Florida. Florida Bay is a shallow, brackish, semi-enclosed water body that receives most of its limited freshwater supply from the Everglades, principally by surficial water runoff through Trout Creek/Taylor River. Because the entire region is underlain by highly porous Key Largo limestone and due to other hydrologic constraints, there is the possibility that ground water exchange may be significant in Florida Bay. To evaluate the extent of such a subsurface contribution, radium isotopes are being determined in shallow wells, seepage meter sites, and a series of water column samples across the Everglades National Park-Florida Bay boundary.

All four radium isotopes were at least an order of magnitude greater in the two shallow well samples than in the water column samples. For example, ^{226}Ra ranged from about 0.50 dpm L^{-1} at a salinity of 5 to over 13 dpm L^{-1} in Well B (salinity = 47.2). Isotopic radium ratios reveal that the well waters (i.e., marine ground water) are geochemically distinct from surficial waters and are regenerated on a time-scale of several days (i.e., ^{224}Ra/^{223}Ra). Results indicate that this radium quartet can be used effectively in Florida Bay to examine the exchange of surficial water and ground water.

INTRODUCTION

Florida Bay, situated in southernmost Florida between the Everglades and the Florida Keys, has received increased attention during the last decades in response to dramatic environmental change, brought about by rapid and widespread development and extensive agriculture (Boesch et al., 1993; McPherson and Halley, 1996). As a consequence, the environmental wellbeing of Florida Bay and the surrounding regions has become threatened. This study is aimed at investigating one consequence of such a modified hydrologic regime. Using unique geochemical attributes of four naturally occurring radium isotopes, we are examining the cycling of nutrient-laden ground-water discharge within Florida Bay in the context of a potential additional source for coastal eutrophication (Johannes, 1980; Synder and Davidson, 1994).

BACKGROUND

In the U-Th decay series there are four radium isotopes ^{223}Ra ($t_{½}$ = 11.4 d), ^{224}Ra ($t_{½}$ = 3.6 d), ^{228}Ra ($t_{½}$ = 5.7 yr) and ^{226}Ra ($t_{½}$ = 1600 yr) with half-lives that coincide well with the time scales of many coastal processes (Swarzenski, 1998). Two important geochemical characteristics make radium ideal as a coastal freshwater-saltwater interface tracer: 1) having highly reactive thorium as its direct radiogenic parent and 2) exhibiting vastly different environmental behavior in freshwater and saltwater systems. Both of these criteria control the production and input of radium in shallow estuaries. In coastal waters, thorium is efficiently scavenged by particles/colloids and rapidly removed to the seabed. In freshwater, radium is bound strongly onto particle surfaces; however, as the ionic strength of a water mass increases during dilution into seawater, desorption reactions and the general absence of particles in the open ocean maintain Ra entirely in a dissolved (< 0.4 μm) phase (Cochran, 1982; Webster et al., 1995). Estuarine sediments thus provide a continuous source for radium isotopes to coastal waters, and the production rate is defined directly by their individual isotopic decay constants (Rama

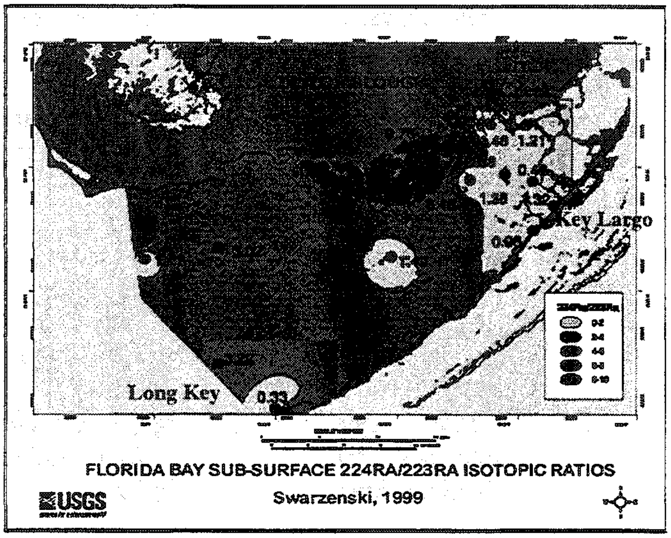

Figure 1. Shallow well $^{224}Ra/^{223}Ra$ activity ratios in Florida Bay.

and Moore, 1996). Source functions for radium in an estuary may thus include the following components: a) riverine, b) oceanic, c) estuarine sediments and d) ground water (Key et al., 1985, Bollinger and Moore, 1993; Rama and Moore, 1996). The relative significance of each of these source terms is defined by the particular environmental constraints of an estuary.

Ground-water discharge, which can consist of recycled marine water *or* fresh water, may enhance the diffusive/advective flux of dissolved Ra from the seabed to a coastal water column wherever the hydraulic gradients and sediment transmissivities are favorable (Moore, 1996). In coastal systems where ground water is discharged to coastal waters either continuously or ephemerally due to tidal forcing, a localized disequilibrium between ^{228}Ra, ^{228}Th and ^{224}Ra can develop. This disequilibrium occurs because dissolved Ra will be rapidly advected into the water column while thorium remains attached to bottom sediments (Bollinger and Moore, 1993). In surficial sediments that are flushed by the upward movement of ground water, $^{224}Ra/^{228}Ra$ activity ratios can become quite large as a result of this process. Below this diagenetically active surface sediment layer one can expect secular equilibrium to develop from ^{232}Th down to ^{224}Ra. Such radioactive equilibrium in the ^{232}Th decay series requires approximately 20 years of sediment storage (Rama and Moore, 1996). Resuspension, bioturbation, chemical dissolution as well as ground water flow will allow these two sediment layers to interact (Aller and Cochran, 1976). Moore and colleagues have used such disequilibria to derive a ground-water flux rate in a South Carolina salt marsh (Rama and Moore, 1996). In their model, the flux of ^{228}Ra from the surface sediments will deplete the activities of its progeny, ^{228}Th and ^{224}Ra as a function of the bioturbation rate (derived from the ^{210}Pb profile), the tidal flushing rate and the ground water flux rate. One can solve a mass balance of these terms by successive iteration.

PRELIMINARY RESULTS AND DISCUSSION

To assess the role of ground-water exchange in a system as hydrogeologically complex as Florida Bay, the various water mass input and removal terms must be clearly identified. The vast drainage system of Taylor River/Trout Creek carries most surficial (sheet flow and channeled) water into upper Florida Bay through a series of cuts across Buttonwood Ridge. At present, the role of ground water exchange in this water supply has not been resolved but is likely to be important. Short-lived radium isotopes ($^{223,224}Ra$) can be used to identify what appear to be discreet water parcels within Trout Creek and lower Taylor River. Trout Creek has a surface water $^{224}Ra/^{223}Ra$ value of 1.46 while the ratio for Taylor River at upper Madeira Bay is 1.65. The oceanic $^{224}Ra/^{223}Ra$ value of 6.48 reflects background-levels of ^{223}Ra (0.006 dpm L^{-1}) and a very low ^{224}Ra activity (0.04 dpm L^{-1}) generated in the water column from the decay of ^{228}Th. These values are comparable to other oceanic radium values (Key et al., 1985; Moore and Todd, 1993).

In Florida Bay, all four radium isotopes are consistently an order of magnitude greater in ground water than in surficial waters. The ground water isotopic radium signature or fingerprint is thus easily discernable and can be used to evaluate the exchange across the sediment-water interface and to identify unique water parcels. A ground water sample collected bay-side at Long Key was highly enriched in excess ^{224}Ra (13.50 dpm L^{-1}) and ^{223}Ra (4.47 dpm L^{-1}). This suggests a longer ground water residence time which is consistent with other ground water tracer studies (Corbett et al., 1999). The largest surface water-ground water exchange appears to occur at a site bay-side off Key Largo, where the limestone is in direct contact with the water column.

$^{224}Ra/^{223}Ra$ activity ratios can be used as a proxy for apparent water mass ages; higher values generally imply younger water. The $^{224}Ra/^{222}Ra$ activity ratio is greatest (9.76) in all of the shallow well sites at Little Madeira Bay (Fig. 1), which is the receiving basin for the Taylor River distributary system. It is thought that this region is the 'burial ground' for water that has recently drained the Everglades (Nuttle, personal communication). Indeed, subsurface salinities are also generally highest in this region (Reich et al., 1998). These observations support the general hypothesis that evapotranspiration of brackish bay water occurs within the lower reaches of the Everglades and the shallow mudbanks of upper Florida Bay. This process is highly dependent on precipitation and is thought to be responsible for the hyper-saline subsurface waters present today below Florida Bay.

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