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Radium-226 accumulation in Florida freshwater mussels

Mark Brenner[1]
Department of Geological Sciences and Land Use and Environmental Change Institute, University of Florida, Gainesville, Florida 32611

Joseph M. Smoak
Environmental Science, University of South Florida, St. Petersburg, Florida 33701

Douglas A. Leeper
Southwest Florida Water Management District, 2379 Broad Street, Brooksville, Florida 34604

Matt Streubert[2]
Environmental Science, University of South Florida, St. Petersburg, Florida 33701

Shirley M. Baker
Department of Fisheries and Aquatic Sciences, University of Florida, 7922 NW 71st Street, Gainesville, Florida 32653

Abstract

Selected lakes in Hillsborough County, Florida have been hydrologically augmented with groundwater to offset stage declines caused by excessive pumping of the Floridan Aquifer. Augmentation water can be relatively rich in ^{226}Ra (>5 decays per minute [dpm] L^{-1}). We measured ^{226}Ra activities in shells and soft tissues of adult bivalve molluscs (*Elliptio* cf. *buckleyi*) from groundwater-augmented and nonaugmented lakes to assess bioaccumulation of ^{226}Ra by mussels. Mussels from augmented lakes displayed higher ^{226}Ra in both shells and tissues than did mussels from nonaugmented lakes. Within a sample, ^{226}Ra activity in *Elliptio* tissues was higher than the value measured in shells. Highest activities were found in a composite mussel sample ($n = 6$) from an augmented lake; soft tissue activity was 619 ± 33 dpm g^{-1} dry weight and shell activity was 147 ± 7 dpm g^{-1} g dry weight. Large mussels displayed greater activities in soft tissues and shells than did small mussels. We transplanted animals from a nonaugmented lake into a groundwater-augmented water body. ^{226}Ra activity in dry tissue rose from 32 ± 1 to 196 ± 2 dpm g^{-1} within 2 months. When ^{226}Ra-rich mussels (232 ± 2 dpm g^{-1}) from the augmented lake were transferred to the nonaugmented lake, they showed no significant ^{226}Ra loss over the 69-d experiment. Large *Elliptio* mussels concentrated ^{226}Ra in their soft tissues to levels about 1,000 to 25,000 times concentrations in lake water. Pumping of groundwater in Florida for residential, agricultural, and industrial use contributes dissolved ^{226}Ra to some surface water bodies, where it can be bioaccumulated by bivalve molluscs.

The deep Floridan Aquifer is the principal source of domestic, irrigation, and industrial-use water for much of Florida. Throughout much of the state, the Floridan is separated from the overlying surficial aquifer system by a clay-rich, intermediate confining layer. In Hillsborough County (Fig. 1), long-term pumping of deep Floridan groundwater promoted downward seepage of both shallow groundwater and surface water. Rapid downward leakage from some local lakes led to stage declines and by the mid-20th century, littoral zones of some water bodies had been exposed. Drying was further exacerbated by droughts, stormwater diversion, roads, and housing construction (Stewart and Hughes 1974). As water levels fell, local residents became concerned that they would lose recreational uses of their lakes and that lakeside property values would decline. Beginning in the 1960s, some communities took action to reverse lake desiccation. In selected areas, wells were drilled to the Floridan Aquifer and deep groundwater was pumped directly into water bodies. Such hydrologic "augmentation" began at some Hillsborough County lakes in the early 1960s.

Studies were initiated in the 1970s to evaluate hydrological, chemical, and biological changes brought about by groundwater augmentation of water bodies (Stewart and Hughes 1974; Martin et al. 1976*a*; Dooris and Martin

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[1] Corresponding author (brenner@ufl.edu).

[2] Present address: USGS, 600 Fourth Street South, St. Petersburg, Florida 33701.

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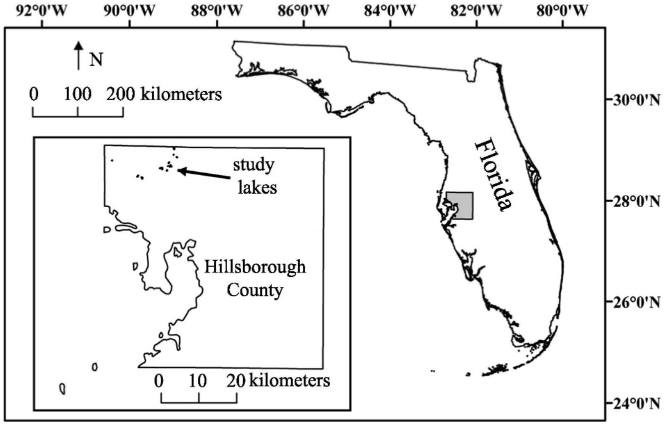

Fig. 1. Map of Florida showing the location of Hillsborough County. Inset map of Hillsborough County shows the location of the study lakes.

1979). Historically, the naturally soft-water lakes were dominated by sodium (Na^+), sulfate (SO_4^{2-}), and chloride (Cl^-) ions. Following groundwater augmentation, calcium (Ca^{2+}) and bicarbonate (HCO_3^-) became the principal dissolved ions, and relative proportions of lake-water ions were similar to those in deep groundwater. Augmented lakes were characterized by high hardness, bicarbonate concentration, and pH (Martin et al. 1976*a*). Phytoplankton diversity was greater in augmented lakes compared with lakes without augmentation (Dooris et al. 1982), and was correlated positively with water column inorganic carbon concentration. There was also speculation that augmentation might promote growth of the exotic macrophyte *Hydrilla* (Martin et al. 1976*b*).

An unanticipated consequence of groundwater augmentation was discovered in Round Lake, a small (0.05 km^2), shallow (mean depth = 2.4 m) water body that has received about half of its annual hydrologic budget from pumped groundwater since 1966. A paleolimnological study in Round Lake attempted to use ^{210}Pb dating to establish age : depth relations for recent sediments. Gamma spectroscopy revealed that surface deposits (0–4 cm) from two Round Lake sediment cores had ^{226}Ra activities of 26.9 ± 1.0 and 26.8 ± 0.3 decays per minute (dpm) g^{-1} dry (Brenner et al. 2000), the highest ^{226}Ra activities that had been measured in Florida lake sediments at that time (Brenner et al. 1994, 1997). Radium-226 activity in topmost Round Lake samples exceeded total ^{210}Pb activity, indicating disequilibrium between ^{226}Ra and "supported" ^{210}Pb (Brenner et al. 2000), a phenomenon that had been reported for only one other Florida water body (Brenner et al. 1994). Even higher ^{226}Ra activities and corresponding isotopic disequilibrium were subsequently identified in another groundwater-augmented system, Lake Charles (Brenner et al. 2004; DeArmond et al. 2006). Recently, high ^{226}Ra activities were documented in near-surface deposits of other groundwater-augmented lakes (Brenner et al. 2006).

High ^{226}Ra activities in the sediments of Round Lake were traced to pumped groundwater (Brenner et al. 2000), which had a ^{226}Ra activity of ~6.2 dpm L^{-1}. Lake-water ^{226}Ra activity was ~3.4 dpm L^{-1}, reflecting dilution of groundwater-derived ^{226}Ra by inputs of rainwater, surface runoff, and shallow subsurface seepage. Dissolved ^{226}Ra in the water column ultimately adsorbed to surface sediments. A study of groundwater-augmented Saddleback Lake also indicated that pumped groundwater was the source of elevated dissolved radium isotopes in that Hillsborough County water body (Smoak and Krest 2006).

High ^{226}Ra activities in groundwater are not surprising. The upper Floridan Aquifer can have high ^{226}Ra activities, especially where groundwater passes through ^{238}U-rich carbonate–fluorapatite deposits in the bedrock (Kaufmann and Bliss 1977; Upchurch and Randazzo 1997). Some coastal surface waters off west central Florida receive groundwater inputs and display high ^{226}Ra activities (Fanning et al. 1982; Miller et al. 1990). High activities of other radioisotopes in the ^{238}U series have also been detected in Florida groundwaters. Harada et al. (1989) measured high ^{210}Po activities in wells, with some of the highest values measured in Hillsborough County (Upchurch and Randazzo 1997).

Divalent ^{226}Ra behaves like Ca^{2+} biogeochemically and can substitute for calcium in plant tissues and the shells, bones, and flesh of animals. Therefore, the accumulation of radium in Round Lake flora and fauna was explored (Brenner et al. 2000). Radium-226 activity in the macroalga *Nitella prolonga* was 12.0 ± 0.4 dpm g^{-1} dry (Brenner et al. 2000). Radium-226 was undetectable in flesh from five of six fish species, though the lake chubsucker (*Erimyzon sucetta*) had measurable activity (0.6 ± 0.4 dpm g^{-1} dry). Fish bone displayed some ^{226}Ra activity, reaching 27 ± 1 dpm g^{-1} ash in chubsuckers. Molluscs had variable ^{226}Ra activities in shell and soft tissues. Planorbid snails had 3.9 ± 0.2 dpm g^{-1} dry in shells and 1.8 ± 0.3 dpm g^{-1} dry in soft tissues. *Pomacea* snail shells had 10.2 ± 0.4 dpm g^{-1} dry. Mussels (*Elliptio* cf. *buckleyi*) displayed relatively higher activities. Their shells ranged from 32 ± 1 to 44 ± 1 dpm g^{-1} dry, but activities in soft tissues were an order of magnitude higher, from 220 ± 2 to 455 ± 37 dpm g^{-1} dry (Brenner et al. 2000). Assuming mussel tissue was about 10% dry weight, bivalves in Round Lake had apparently accumulated ^{226}Ra in their tissues to levels >10,000 times the concentration in lake water.

Freshwater mussels are sometimes used to monitor environmental contaminants because of their ability to accumulate toxic compounds (Muncaster et al. 1990) and heavy metals (Duxbury et al. 2005) at concentrations much greater than those in water or sediments. Molluscs can also concentrate radionuclides in their shells and tissues (e.g., Van der Borght 1963; Jeffree 1985). Therefore, these bivalves may serve as sensitive bioindicators of elevated ^{226}Ra in Florida surface waters. Biological accumulation of ^{226}Ra by mussels in lakes may be an important pathway for transfer of radium to higher trophic levels of aquatic and terrestrial food webs. Furthermore, some of the radioactive daughters of ^{226}Ra, such as ^{222}Rn and ^{210}Po, are alpha emitters that have been linked to human health concerns.

Given the high ^{226}Ra activities measured in Round Lake mussels, we decided to further explore the relation between dissolved radium in lake waters and accumulation of ^{226}Ra

Table 1. Groundwater augmentation status, lake area, lake mean depth, and ^{226}Ra activity (dpm L^{-1}) in augmentation well water and lake water from water bodies in Hillsborough County, Florida. Analytical error was ~0.2 dpm L^{-1} for all samples.

Lake	Augmentation status	Area (km^2)	Mean depth (m)	Well	Lake
Brant	Intermittently since 1995	0.24	2.4	0.7	1.5
				0.2	1.1
				0.2	0.9
Charles	Regularly since 1968	0.06	1.8	2.9	1.8
				3.5	1.8
				2.9	1.3
Crystal	Intermittently since 1973	0.07	3.0	1.3	0.7
				1.1	0.9
				1.8	0.7
Jackson	Regularly since 1977	0.03	2.0	2.2	0.4
				3.3	0.4
				3.1	0.2
Little Hobbs	Intermittently since early 1970s	0.03	2.1	0.7	0.4
				1.1	0.7
				0.7	0.7
Round	Regularly since 1966	0.05	2.4	5.9	2.0
				5.9	1.3
				5.7	2.9
Saddleback	Regularly since 1968	0.15	1.7	3.3	1.8
				3.3	1.3
				3.1	0.4
Armistead	Not augmented	0.14	2.4		0.9
					0.4
					0.4
Commiston	Not augmented	0.06	3.0		0.4
					0.4
					0.9
Deer	Not augmented	0.14	3.6		0.2
					0.4
					0.4
Halfmoon	Not augmented	0.14	3.0		0.4
					0.7
					0.7

in Florida freshwater mussels. In this study, we (1) evaluated whether mussels from groundwater-augmented lakes display higher ^{226}Ra activities than mussels from nonaugmented lakes, (2) examined how radium-226 activity varies as a function of animal size, (3) determined where ^{226}Ra accumulates in the soft tissues of mussels, (4) assessed the rate at which animals acquire radioactivity when placed in a groundwater-augmented lake, and (5) examined whether animals that show relatively high ^{226}Ra lose their radioactivity when placed in a nonaugmented lake environment.

Study sites

Here we report results from 11 lakes in Hillsborough County, Florida (Fig. 1) that were sampled over an 18-month period beginning in January 2000. Seven of the lakes had a history of groundwater augmentation (Brant, Charles, Crystal [aka South Crystal], Jackson, Little Hobbs [aka Lutz], Round, and Saddleback) and four lakes had not been deliberately augmented before this study (Armistead, Commiston [aka Commston], Deer, and Halfmoon) (Table 1). In the 2 yr preceding the study, Halfmoon Lake received some groundwater input during the preparation and testing of an augmentation well. The amount of groundwater delivered to the water body at that time was believed to be small relative to the lake volume. Furthermore, the water residence time of Halfmoon Lake is probably relatively short given its shallow mean depth (3 m). We therefore considered Halfmoon Lake nonaugmented for this study. Brant Lake was the largest water body (0.24 km^2) in the group. The other study lakes ranged in size from 0.03 to 0.15 km^2 (Table 1). Mean depths for the lakes ranged from 1.7 to 3.6 m (Table 1). We also report findings from a reciprocal transplant experiment in which *E.* cf. *buckleyi* were transferred between groundwater-augmented Round Lake and nonaugmented Commiston Lake, and ^{226}Ra levels in the mussels were monitored over a period of 69 d.

Field methods

We collected three water samples for ^{226}Ra analysis from each study lake at a water-column depth of ~0.5 m. Triplicate water samples were also collected from augmentation wells at Lakes Brant, Charles, Crystal, Jackson, Little Hobbs, Round, and Saddleback. Well casings were purged and well-water samples were collected at wellheads or from pipes that discharged groundwater into the lakes. At each study site, triplicate lake water or well-water samples were collected on the same day. Water samples for ^{226}Ra analysis were collected in plastic bottles and delivered to the Florida Department of Health (FDH) Safe Drinking Water Laboratory in Orlando, Florida. Triplicate lake water samples were also taken at the same time for calcium analysis. Water samples for Ca analysis were collected in plastic bottles and delivered to the Southwest Florida Water Management District Chemistry Laboratory in Brooksville, Florida.

Mussels representing several taxa were collected by hand or with a dip net in the shallow littoral zone of the study lakes. In deeper water, mussels were collected using a dip net or Ekman dredge. Live mussels were generally collected at several sites within each lake. Multiple individuals from each site were placed in a labeled plastic bag and transported on ice to the University of Florida for ^{226}Ra analysis.

In the reciprocal transplant experiment, large *E.* cf. *buckleyi* were collected from nonaugmented Lake Commiston and placed in groundwater-augmented Round Lake. Likewise, animals were collected from Round Lake and placed in Lake Commiston. The mussels were collected and transplanted in small cages on 17 July 2002, i.e., day zero of the experiment. Initial ^{226}Ra activity in tissues was measured on a composite sample of three mussels from each lake, and thereafter, three transplanted bivalves were collected from each lake for ^{226}Ra measurement at intervals of 3–8 d until 24 September 2002. Mussels harvested during the experiment were transported to the University of South Florida for radium analysis.

Laboratory methods

Radiochemical analysis of water samples was conducted at the FDH Safe Drinking Water Laboratory using U. S. Environmental Protection Agency methods (USEPA 1980, 1984). Individual mussels from a site within a lake were grouped to constitute a single sample for ^{226}Ra analysis. If large numbers of mussels were collected from a single location, a subsample consisting of multiple individuals was processed. Samples from Lakes Charles, Round, Crystal, Saddleback, and Armistead were separated into two nonoverlapping size classes on the basis of shell length to examine the relation between organism size and ^{226}Ra activity in tissues and shells. Each mussel sample, consisting of multiple individuals, was weighed (total wet mass) on an electronic balance. Next, whole individuals were freeze-dried. Dry tissues and shells were separated and weighed. We assumed that all weight loss on drying represented water loss from soft tissues. Wet mass of tissue was calculated as total wet mass minus shell mass. The percentage of wet tissue mass represented by dry tissue mass was calculated as soft tissue dry mass divided by soft tissue wet mass × 100. Samples from the transplant experiment were treated in a similar manner, with three individuals combined for each sampling date.

Shell and soft tissue samples were prepared separately for analysis of ^{226}Ra activity. Soft tissues from composite samples were homogenized in a small food processor. The right shell (valve) of each individual in a composite sample was ground to a powder with a mortar and pestle. The left valve of each mussel was measured for length and height using a ruler or calipers and archived. Each composited shell and soft tissue sample was stored individually in a labeled plastic scintillation vial before analysis for ^{226}Ra activity.

Nine *E.* cf. *buckleyi* mussels from three sites in Lake Charles were dissected to determine in which soft tissues ^{226}Ra accumulates. Soft tissues were separated into five classes: (1) mantle, (2) muscle (anterior and posterior adductors), (3) foot, (4) gill, and (5) body (i.e., remaining soft tissues, including organs and gut contents). Material from each tissue type from the nine individuals was combined to form a sample, then freeze-dried and ground in preparation for ^{226}Ra analysis.

Samples of ground mussel shell or soft tissue were placed in preweighed plastic Sarstedt tubes that were reweighed to determine sample mass. Next, samples were covered with epoxy glue and tubes were capped. Sealed samples were left for >3 weeks before counting to allow ingrowth of ^{226}Ra daughters ^{214}Pb and ^{214}Bi, which were measured as proxies for ^{226}Ra activity. Samples were measured by gamma counting (Appleby et al. 1986; Schelske et al. 1994) with well-type germanium detectors connected to a 4,096-channel multichannel analyzer. Radium-226 activity was estimated by averaging the activity of ^{214}Pb (295.1 keV), ^{214}Pb (351.9 keV), and ^{214}Bi (609.3 keV) (Moore 1984) and is expressed in units of decays per minute per gram of dry material (dpm g^{-1}). Errors associated with activities are reported as 1 SE of measurements on the basis of counting statistics for individual samples (Friedlander et al. 1981). Gamma counting of survey and dissected mussel samples was done at the University of Florida. Samples from the reciprocal transplant experiment were counted at the University of South Florida.

Because organisms utilize ^{226}Ra similarly to calcium, we also evaluated Ca concentrations in lake waters and tissues of mussels from augmented and nonaugmented lakes. Calcium in lake water samples was measured by the Southwest Florida Water Management District Chemistry Laboratory using the direct nitrous oxide–acetylene flame method (311D) (Am. Public Health Assoc. 1992). For mussels, we digested ground tissue samples in hot 1 N HCl (Andersen 1976) and brought the digestate to constant volume. Calcium concentrations in waters and digestates were measured using atomic absorption spectrophotometry. Calcium concentrations in mussels were figured per gram of dry tissue. Significant differences in ^{226}Ra activities or Ca concentrations between sample populations were tested using the nonparametric Mann–Whitney *U*-test.

Results

Radium-226 activity in the water column of augmented lakes ranged from a low of 0.2 dpm L^{-1} in one Lake Jackson sample to 2.9 dpm L^{-1} in a sample from Round

Table 2. Radium-226 activity (dpm g^{-1} dry) in mussel soft tissues and shells from 11 Florida lakes. Brant, Charles, Crystal, Jackson, Little Hobbs, Round, and Saddleback received groundwater supplements. Lakes Armistead, Commiston, Deer, and Halfmoon were nonaugmented. n = individual mussels in composite sample. Error (1σ) is 1 SE on the basis of counting statistics. Length = range of shell lengths for all individuals in a sample. Tissue/ind = mean dry tissue mass per individual in the sample (total dry mass ÷ n). %dry mass = dry tissue mass ÷ wet tissue mass × 100. Shell/ind = mean shell mass per individual in the sample (total shell mass ÷ n). * indicates only shells of dead animals were found. na = not available. ND = not detected.

Lake	Taxon	n	Tissue (Ra-226)	Tissue (1σ)	Shell (Ra-226)	Shell (1σ)	Length (mm)	Tissue/ind (g)	%Dry mass	Shell/ind (g)
Brant	*Utterbakia*	2	5	4	2	1	na	0.20	3.1	1.43
	**Utterbakia*	7	na	na	5	1	na	na	na	na
	**Utterbakia*	8	na	na	4	1	na	na	na	na
	*Unidentified Unionid	2	na	na	5	1	na	na	na	na
	Corbicula	11	4	1	13	1	na	0.13	3.6	5.22
	Corbicula	13	7	2	17	2	na	0.11	2.5	5.72
Charles	*Elliptio*	6	619	33	147	7	63–68	1.01	6.6	20.32
	Elliptio	10	434	23	120	6	48–65	0.67	7.4	8.62
	Elliptio	10	231	14	90	6	47–57	0.71	8.2	8.55
Crystal	*Elliptio*	7	104	6	42	2	51–58	0.78	10.1	7.32
	Elliptio	6	164	8	51	2	36–55	0.57	7.9	5.69
	Elliptio	9	43	4	24	2	33–49	0.36	8.9	2.55
	Utterbakia	1	17	4	5	1	61	0.24	2.9	1.82
Jackson	*Utterbakia*	15	19	1	35	2	71–78	1.01	7.2	3.31
Little Hobbs	*Elliptio*	10	98	8	53	2	47–56	0.87	10.0	7.44
	Elliptio	10	65	2	60	6	47–59	0.83	9.7	6.95
	Elliptio	10	140	9	38	2	45–57	0.77	10.0	6.93
Round	*Elliptio*	16	426	20	54	5	53–65	0.88	8.4	11.77
	Elliptio	15	315	17	38	3	50–59	0.79	9.2	8.74
	Elliptio	15	221	14	34	3	42–49	0.46	8.4	4.98
	Elliptio	24	156	19	32	2	29–45	0.34	9.2	3.20
Saddleback	*Elliptio*	14	79	6	44	1	50–60	1.06	10.1	10.52
	Elliptio	26	54	2	24	1	24–42	0.35	9.5	2.51
Armistead	*Elliptio*	16	38	2	4	1	46–57	0.61	9.1	9.28
	Elliptio	30	29	1	3	<1	28–41	0.27	8.6	3.58
	Corbicula	25	ND	ND	2	<1	30–35	0.34	8.5	7.86
Commiston	*Elliptio*	8	36	4	6	1	54–67	0.92	8.6	9.74
	Elliptio	8	9	1	5	1	51–65	0.78	7.5	11.61
	Elliptio	8	37	2	5	2	56–60	0.71	7.6	8.54
Deer	*Elliptio*	8	30	1	7	<1	45–52	0.42	6.2	6.19
	Elliptio	8	49	4	8	1	44–50	0.36	6.6	5.11
	Elliptio	8	25	2	6	<1	41–49	0.34	6.4	3.99
Halfmoon	*Elliptio*	12	86	5	16	1	48–54	0.61	8.9	6.05

Lake (Table 1). Nonaugmented lakes displayed a smaller range, from 0.2 dpm L^{-1} in one sample from Deer Lake to 0.9 dpm L^{-1} in one sample each from Lakes Armistead and Commiston (Table 1). Radium-226 activity was significantly higher in augmented lakes than in nonaugmented lakes ($p < 0.01$). Among the seven hydrologically augmented lakes, ^{226}Ra activity in well water used for augmentation ranged from a low of 0.2 dpm L^{-1} in the Lake Brant well to a high of 5.9 dpm L^{-1} measured in the Round Lake well (Table 1). Lake Brant was the only augmented lake that displayed ^{226}Ra activities in lake water that were greater than those measured in the augmentation well. Calcium concentrations ranged between 6.7 and 57.0 mg L^{-1} in waters of augmented lakes ($n = 21$) and from 7.0 to 13.3 mg L^{-1} in nonaugmented lakes ($n = 12$), and were significantly greater in augmented lakes ($p < 0.0005$).

We found three unionid mussel taxa in the study lakes, including *E.* cf. *buckleyi* (Florida shiny spike), *Utterbakia imbecillis* (paper pondshell), and an unidentified unionid (Table 2). We also collected the corbiculid, *Corbicula fluminea* (Asian clam). Live *Elliptio* were collected in all lakes except Brant and Jackson. Live *Utterbakia* were found in substantial numbers in Lake Jackson, but only one live specimen was collected in Crystal Lake, and just two live animals were taken from Brant Lake, though numerous shells of deceased animals were found. *Corbicula* were taken in Lakes Brant and Armistead. Among *Elliptio* mussels, the percentage dry mass of tissues varied from 6.2% to 10.1% (Table 2).

Radium-226 activity in mussel soft tissues ranged from undetectable in *Corbicula* from Lake Armistead to 619 ± 33 dpm g^{-1} in *Elliptio* from Lake Charles (Table 2). Shell values ranged from 1.8 ± 0.3 dpm g^{-1} in the *Corbicula*

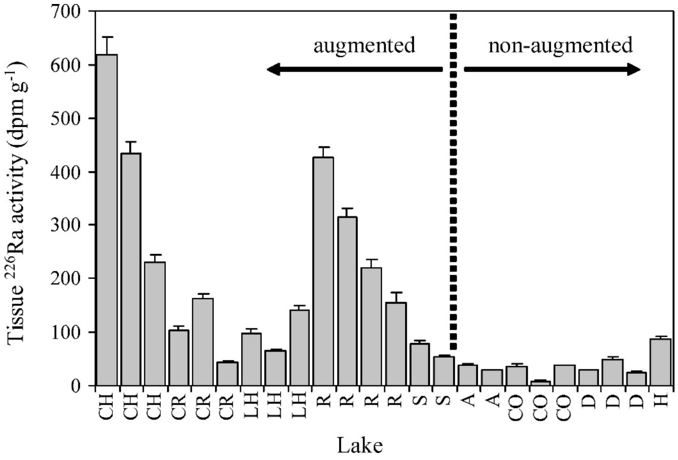

Fig. 2. ^{226}Ra activity (dpm g^{-1}) in dry tissues of *Elliptio* cf. *buckleyi* mussels from groundwater-augmented Lakes Charles (CH), Crystal (CR), Little Hobbs (LH), Round (R), and Saddleback (S), and from nonaugmented Lakes Armistead (A), Commiston (CO), Deer (D), and Halfmoon (H).

sample from Lake Armistead to 147 ± 7 dpm g^{-1} in an *Elliptio* sample from Lake Charles (Table 2). *Elliptio* tissues showed higher activities than *Elliptio* shells (Table 2). Activities in *Elliptio* tissues were also greater than activities in tissues of *Utterbakia* ($p < 0.01$) and *Corbicula* ($p < 0.01$). Given the propensity for *Elliptio* to accumulate ^{226}Ra, and the fact that *Elliptio* were found in nearly all the lakes, we selected this taxon for further analyses.

Radium-226 activity in tissues of *Elliptio* from groundwater-augmented lakes was significantly greater than activity in tissues of animals from nonaugmented lakes ($p < 0.001$) (Fig. 2). Likewise, shells of animals from the augmented lakes displayed higher ^{226}Ra than did shells of animals from nonaugmented lakes ($p < 0.001$) (Fig. 3). Calcium content in tissues of mussels from augmented lakes ranged from 8 to 40 mg g^{-1}. In nonaugmented lakes, Ca content in tissues displayed a similar range, from 12 to 35 mg g^{-1}. There was no significant difference in Ca content in tissues between populations from augmented and nonaugmented lakes ($p > 0.05$). Larger mussels demonstrated higher activity than smaller mussels in both their tissues and shells (Figs. 4 and 5). Dissected mussels from Lake Charles showed that ^{226}Ra activities in isolated tissues ranged from a high of 2,927 ± 123 dpm g^{-1} in the body (organs and gut contents) to a low of 10 ± 4 dpm g^{-1} in the foot (Fig. 6).

In the reciprocal transplant experiment, mussels taken from nonaugmented Lake Commiston for transplant into

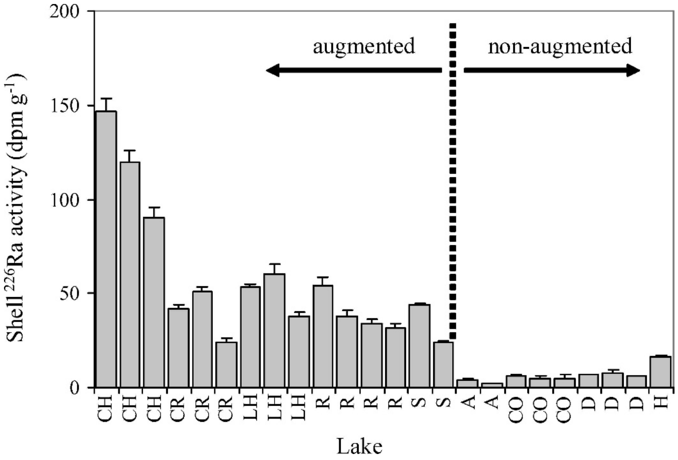

Fig. 3. ^{226}Ra activity (dpm g^{-1}) in shells of *Elliptio* cf. *buckleyi* mussels from groundwater-augmented Lakes Charles (CH), Crystal (CR), Little Hobbs (LH), Round (R), and Saddleback (S), and from nonaugmented Lakes Armistead (A), Commiston (CO), Deer (D), and Halfmoon (H).

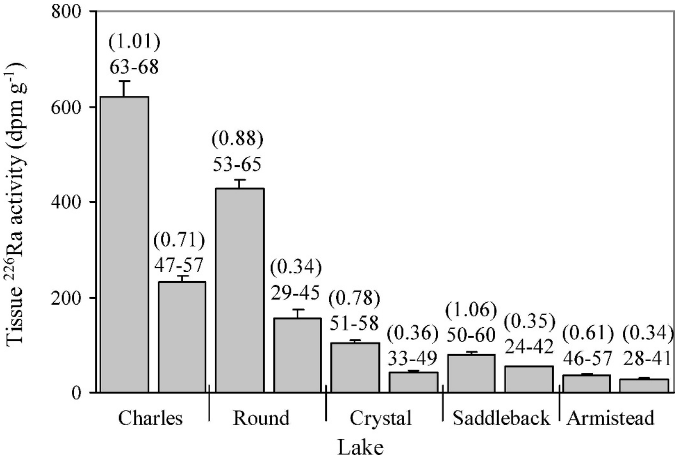

Fig. 4. ^{226}Ra activity (dpm g^{-1}) in dry tissues of *Elliptio* cf. *buckleyi* mussels from groundwater-augmented Lakes Charles, Round, Crystal, Saddleback, and nonaugmented Armistead. Within a lake, each sample is composed of multiple individuals representing a distinct shell length range, shown above each bar rounded to the nearest millimeter. Values in parentheses are the mean dry tissue mass per individual in the sample.

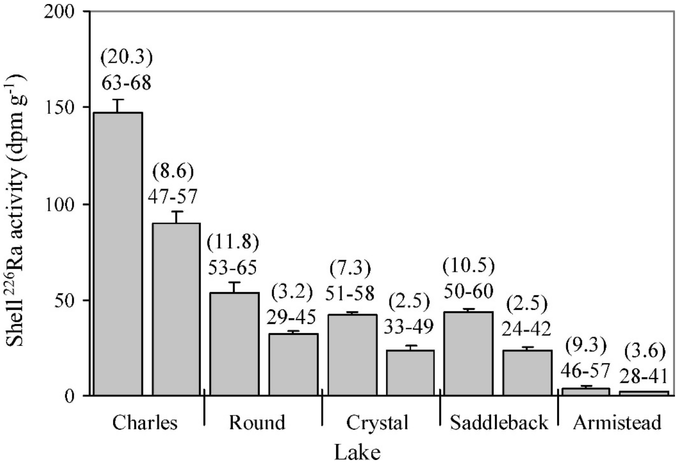

Fig. 5. ^{226}Ra activity (dpm g^{-1}) in shells of *Elliptio* cf. *buckleyi* mussels from groundwater-augmented Lakes Charles, Round, Crystal, Saddleback, and nonaugmented Armistead. Within a lake, each sample is composed of multiple individuals representing a distinct shell length range, shown above each bar rounded to the nearest millimeter. Values in parentheses are the mean dry mass of shell per individual in the sample.

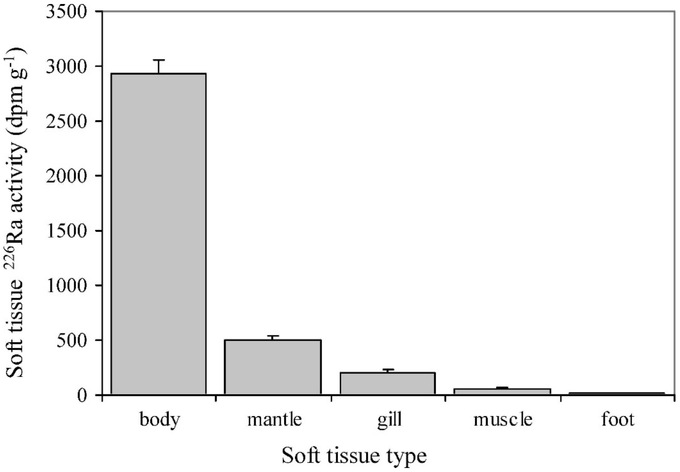

Fig. 6. ^{226}Ra in dissected dry tissues of *Elliptio* cf. *buckleyi* mussels from groundwater-augmented Lakes Charles. Dissected tissues were composited from nine large individuals. "Muscle" is composed of anterior and posterior adductors and "body" is remaining soft tissues, including organs and gut contents.

groundwater-augmented Round Lake had an initial tissue ^{226}Ra activity of 32 ± 1 dpm g^{-1}. Over the course of the experiment, mussels transplanted to Round Lake accumulated ^{226}Ra, and samples of animals harvested on the last two collection dates of the experiment, days 61 and 69, had activities of 196 ± 2 and 151 ± 6 dpm g^{-1}, respectively (Fig. 7). Animals taken from groundwater-augmented Round Lake for transplant to nonaugmented Lake Commiston had starting activities of 232 ± 2 dpm g^{-1}. Throughout the 69-d experiment, transplanted animals retrieved from Lake Commiston displayed variability in their activity, between 177 ± 3 and 279 ± 16 dpm g^{-1}, but there was no trend in activity with time, and animals maintained high activities throughout the duration of the experiment (Fig. 7).

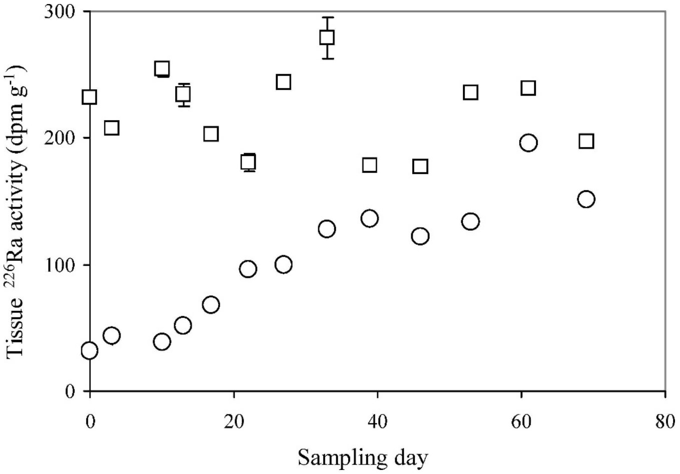

Fig. 7. ^{226}Ra activity (dpm g^{-1}) in dry tissues of *Elliptio* cf. *buckleyi* mussels from the reciprocal transplant experiment. Mussels were moved from nonaugmented Commiston Lake to augmented Round Lake, and vice versa. Three transplanted individuals were sampled from each lake at intervals over a period of 69 d. Circles are mussels moved to augmented Round Lake. Squares are mussels moved to nonaugmented Commiston Lake. Error terms were smaller than plot symbols for most samples.

Discussion

We examined ^{226}Ra activity in bivalve molluscs from groundwater-augmented and nonaugmented lakes in Hillsborough County, Florida. We found that (1) mussels in groundwater-augmented lakes had significantly higher ^{226}Ra activity than mussels from nonaugmented lakes, (2) larger mussels had higher activity than smaller mussels in both their shells and tissues, (3) ^{226}Ra accumulated primarily in the body of the mussels, as opposed to other tissues, and (4) animals rapidly gained activity when placed in a groundwater-augmented lake, but (5) did not lose radioactivity when transplanted to a nonaugmented lake. Our results indicate that groundwater augmentation contributes bioavailable ^{226}Ra to Florida lakes, where it is accumulated to high levels by freshwater mussels. This has implications for both monitoring of ^{226}Ra and the health of aquatic and terrestrial food webs.

Well water generally displayed higher activity than lake water among the groundwater-augmented systems. This confirms the finding of previous studies (e.g., Smoak and Krest 2006) that well water is a source of ^{226}Ra to lakes receiving hydrologic supplements and that ^{226}Ra is diluted by water that enters lakes in direct rainfall, runoff, and subsurface seepage. Brant Lake, the largest water body in the study, was an exception in possessing lake water with higher activity than well water. The well at Brant Lake had the lowest ^{226}Ra of the sampled wells, and we speculate that the augmentation well may not draw water from a depth with ^{226}Ra-rich deposits. The relatively higher values in the lake may reflect contributions of groundwater from residential wells in the basin that do in fact tap into rock with abundant ^{226}Ra.

Several factors may account for interlake differences in water column ^{226}Ra activity. Groundwater is a major contributor of ^{226}Ra to lakes and displays variable ^{226}Ra concentration from site to site. Furthermore, some augmented lakes are pumped on a regular basis, while others receive intermittent groundwater supplements. Lastly, groundwater probably represents a different proportion of the hydrologic budget for each lake, and this proportion may vary in each water body from year to year. Intralake variation in lakewater ^{226}Ra activity in augmented lakes may also reflect both analytical error and incomplete mixing of radium-rich well water throughout the lake. All ^{226}Ra activity values in well waters and lake waters were low relative to the state and federal combined ^{226}Ra and ^{228}Ra limit of 11.1 dpm L^{-1} for drinking water.

Mussels from augmented lakes showed higher ^{226}Ra activity in their tissues and shells compared with mussels from nonaugmented lakes. This also supports the claim that groundwater is a major source of ^{226}Ra in augmented lakes. The mussels are capable of concentrating ^{226}Ra in their tissues to levels far above concentrations in the lake water. Soft tissues have percentage dry weights of about 6–10%, so drying of tissues can account for only about a 10–

16-fold increase in activity as dissolved radium in and around the animal flesh adsorbs to desiccating tissues. For each *Elliptio* tissue sample, we calculated the ^{226}Ra "concentration factor" using the ^{226}Ra activity in the tissue sample, the percentage dry mass of the soft tissue sample (to remove the effect of concentration due simply to drying), and the mean ^{226}Ra activity g^{-1} of water in the lake. Concentration factors computed this way indicate that mussels concentrated ^{226}Ra to levels about 1,000 to 25,000 times those in the lake water.

When different size classes of mussels were compared with one another within five lakes, larger mussels had higher ^{226}Ra activity in their shells and soft tissues than did smaller mussels. This suggests that animals continue to accumulate ^{226}Ra as they grow, or that larger, and presumably older, animals were exposed to higher ^{226}Ra in the past. The reciprocal transplant experiment also suggests that animals may preserve a record of the high levels of ^{226}Ra to which they have been exposed in the past. The experimental organisms were shown to accumulate ^{226}Ra rapidly, and those that started with relatively high activity failed to lose radioactivity over a 2-month time span following placement in a nonaugmented lake. We suggest that soft tissues of larger (older), rather than smaller (younger), *Elliptio* individuals are preferable for use in monitoring ^{226}Ra accumulation.

In dissected mussels from Lake Charles, highest ^{226}Ra activity was found in the mussel body (organs and gut contents), in which it approached 3,000 dpm g^{-1}. This portion of the animal constitutes a relatively small fraction of the total soft tissue dry mass, as ^{226}Ra activity for whole animal dry tissues in Lake Charles ranged from 231 ± 14 to 619 ± 33 dpm g^{-1}. The high activity detected in the body is offset by lower activities in the adductor muscles and foot. We initially considered the possibility that high activity recorded in the body might be largely attributable to ^{226}Ra-rich particles in the gut. Two lines of evidence argue against this. First, activity in the body is about two orders of magnitude higher than activities recorded in the surface sediments of Lake Charles (DeArmond et al. 2006) that probably contain large amounts of the same particle types found in the mussel diet. It might be argued that mussels selectively remove particles of a size class or geochemical composition that are particularly radioactive, and that such food particles differ from surface mud in terms of ^{226}Ra activity. If, however, the ^{226}Ra activity were largely associated with material passing through the mussel intestinal tract, then when highly radioactive animals purge their gut contents, there should be a decline in tissue ^{226}Ra activity. The reciprocal transplant experiment showed that ^{226}Ra-rich mussels from groundwater-augmented Round Lake did not lose radioactivity when placed in a nonaugmented lake, even after a period of more than 2 months. Although gut contents may contain radioactive particles, the high activities measured are evidently bound in the animal tissues. These findings are consistent with the results of experimental studies with freshwater mussels, which suggested that most of the ^{226}Ra incorporated into mussel tissue comes directly from the water rather than from the diet (Jeffree and Simpson 1986).

The reciprocal transplant experiment showed that *Elliptio* taken from nonaugmented Lake Commiston and moved to groundwater-augmented Round Lake accumulated ^{226}Ra rapidly during the 69-d duration of the study. Radium-226 activity in mussel tissues quickly approached levels found in the tissues of resident mussel populations in Round Lake. This indicates that it is possible to identify water bodies that have substantial dissolved ^{226}Ra by placing mussels with low ^{226}Ra activity in a lake for a relatively short period of time and monitoring their tissue ^{226}Ra activity. The use of mussels as "bioindicators" of dissolved ^{226}Ra in lakes enables rapid assessment without the need to filter large volumes of water through manganese fiber.

The transplant experiment demonstrated that mussels moved from groundwater-augmented Round Lake to nonaugmented Commiston Lake showed no significant loss of ^{226}Ra activity in their tissues over the 69-d study period. Long-term retention of ^{226}Ra has also been documented for the tropical freshwater mussel *Velesunio angasi*, which showed no significant loss of ^{226}Ra activity in ^{226}Ra-rich animals after 286 d of exposure to ^{226}Ra-free water (Jeffree and Simpson 1986). If ^{226}Ra accumulation in mussel tissues is irreversible, then measurements will reflect any period of relatively high ^{226}Ra activity in the lake over the lifetime of the mussel, even if recent lake water activity has been low. That is to say, mussels may preserve in their tissues a historical record of exposure to high ^{226}Ra concentrations in the water. We speculate that this could account for relatively elevated ^{226}Ra activities in the shells and tissues of animals from Halfmoon Lake. Although the lake was not receiving augmentation water at the time of the study, the earlier tests of the augmentation well may have introduced some dissolved ^{226}Ra into the lake.

Comparison of ^{226}Ra activity in *Elliptio* from groundwater-augmented and nonaugmented lakes strongly implicates groundwater as a primary source of ^{226}Ra in the tissues and shells of bivalves. A number of factors may influence differences seen both within and between lakes, with respect to mussel radioactivity. Within a lake, groups of mussels at sites throughout a basin may be exposed to different dissolved ^{226}Ra concentrations. In augmented lakes, this may be related to the proximity of animals to the augmentation well and the degree of well-water mixing throughout the basin. Some mussels may occur in areas where there is substantial subsurface inflow and are hence exposed to relatively lower concentrations of dissolved ^{226}Ra.

Experimental work with *V. angasi* showed that increased concentrations of calcium in waters reduced the rate of ^{226}Ra uptake by mussels (Jeffree and Simpson 1986). We examined the effect of calcium concentrations in augmented and nonaugmented lake waters on ^{226}Ra concentrations in *Elliptio* mussels. Despite higher calcium content in the water column of augmented lakes, there was no significant difference between Ca content in tissues of mussels from augmented and nonaugmented lakes ($p > 0.05$). Furthermore, in spite of the higher calcium levels in augmented lake waters, the mussels from augmented lakes display relatively higher ^{226}Ra activities than mussels from nonaugmented lakes.

Activities of ^{226}Ra in tissues and shells of *Elliptio* from nonaugmented lakes are relatively lower than values recorded in animals from augmented lakes, but nevertheless beg the question "what is the source of ^{226}Ra in mussels from nonaugmented lakes?" Brenner et al. (2004) reported up-core (i.e., recent) increases in ^{226}Ra activity in sediment profiles from several nonaugmented Florida lakes. They hypothesized that these water bodies, although not deliberately groundwater-augmented, receive ^{226}Ra-rich runoff from deep groundwater pumped for irrigation, residential use, industry, and mining. Any human activity that introduces ^{226}Ra-rich groundwater to lakes creates the potential for bivalves to bioaccumulate the radionuclide.

Our results indicate that the soft tissues and shells of *Elliptio* mussels can be used to identify surface waters that are or have been rich in dissolved ^{226}Ra. These long-lived organisms are capable of concentrating ^{226}Ra in soft tissues by 3–4 orders of magnitude above concentrations in the ambient water. The tissues of large individuals are recommended if animals are to be used as bioindicators. If individuals with low ^{226}Ra are introduced to a new water body, they will accumulate levels of ^{226}Ra comparable with resident populations within 2 to 3 months.

The ability of bivalves to bioaccumulate ^{226}Ra, especially in the soft tissues, raises further questions. Radium-226 and several of its daughter radionuclides (e.g., ^{222}Rn and ^{210}Po) are alpha emitters. Wildlife or humans that consume bivalves with high ^{226}Ra activity may be exposed to hazardous levels of radiation. It has been reported that regular consumption of *Elliptio* from Round Lake could pose an increased risk of cancer for humans (Hazardous Substance & Waste Management Research, Inc. 2000). Much remains to be learned about the impact of ^{226}Ra on mussels that accumulate it, as well as the fate of the ^{226}Ra as it moves from irradiated mussels into other organisms in the food web.

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