

Radium isotopes in Cayuga Lake, New York: Indicators of inflow and mixing processes

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Abstract

Naturally occurring radium isotopes (^{223}Ra , ^{224}Ra , ^{226}Ra , and ^{228}Ra) were measured in lake and tributary water of Cayuga Lake, New York, during the course of a vernal inflow event in the spring of 2001. A large influx of groundwater, probably from a carbonate aquifer, entered the lake at its extreme southern end early in the vernal inflow event and spread northward, covering an extensive part of the southern end of the lake. The low $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of this water mass, compared with bulk lake water, allowed its identification through time. Estimates of mixing with bulk lake water were calculated from changes in the ^{226}Ra content. Groundwater inflow to the lake around the delta of a major tributary was detected on the basis of ^{223}Ra and ^{224}Ra activity of lake and tributary water. Inflow of a water mass to the surface of the lake was also detected using ^{223}Ra and ^{224}Ra activity. The integrity of this water mass was monitored using short-lived radium isotopes. Suspended sediment in the lake water is a source of the short-lived radium isotopes ^{223}Ra ($\sim 2 \times 10^{-4}$ dpm L^{-1}) and ^{224}Ra ($\sim 3 \times 10^{-3}$ dpm L^{-1}), but bottom sediments are a more significant source of ^{228}Ra . Radium isotopes can be valuable new tools in limnological investigations, allowing detection and monitoring of events and processes such as water inflow and mixing, determining sources of inflowing water, and monitoring introduced water masses as they move within the lake.

Physical processes occurring in lakes such as inflow, mixing, water-mass movement, and groundwater seepage are important aspects in understanding the dynamics of a limnological system, and they can be important aspects in the management of freshwater resources. The naturally occurring radium isotopic suite (^{226}Ra , ^{228}Ra , ^{223}Ra , and ^{224}Ra) has great potential for evaluating and quantifying many of these limnological processes. These isotopes have previously been used in salt marsh (Rama and Moore 1996; Krest et al. 2000; Kelly and Moran 2002), estuarine (Hussain et al. 1999; Hancock et al. 2000; Charette et al. 2001), and coastal marine (Moore 1999) regions to examine groundwater inflow to, and determination of residence time of water in, these environments. Although these isotopes behave identically chemically, their range of half-lives ($^{226}\text{Ra} = 1,601$ yr; $^{228}\text{Ra} = 5.8$ yr, $^{223}\text{Ra} = 11.4$ d, and $^{224}\text{Ra} = 3.7$ d) allows the examination of time-dependent processes that occur during mixing and transport. Radium isotopes are found to some extent in all natural waters, and recent instrumental developments (Moore and Arnold 1996) have made quantitative measurement of all naturally occurring isotopes practical for environmental water samples of all types.

Radium isotopes have been studied to a lesser degree in freshwater systems than they have been in estuarine and marine environments, possibly because of the assumption that radium isotopes are present in lower concentrations in freshwater than in more saline water, because of greater adsorption onto particulate matter. Several investigations, however, have shown that radium isotopes are measurable and useful in freshwater systems such as rivers (Kraemer and Curwick 1991; Eikenberg et al. 2001), small water bodies (Sidle et al. 2001), and the Everglades (Krest and Harvey 2003).

Radium isotopes form as daughter products during the decay of the naturally occurring elements uranium and thorium. They accumulate in water via interactions of the water with solid-phase material that contains the initial source of the isotope (uranium for ^{226}Ra and ^{223}Ra and thorium for ^{228}Ra and ^{224}Ra). The isotopes accumulate through the processes of dissolution, ion-exchange, and α -particle recoil (Kraemer and Genereux 1998). In a closed system, the daughter products reach a level of radioactivity equivalent to that of the long-lived head of the decay chain (e.g., ^{238}U for ^{226}Ra , ^{232}Th for ^{224}Ra and ^{228}Ra , and ^{235}U for ^{223}Ra), a condition that is known as "secular equilibrium. This results in the radium isotopic activities of the material equaling the U and Th activities. Where solid material is abundant compared with water (e.g., groundwater), radium isotopes will tend to accumulate to higher activities than where the solid material is relatively less abundant (e.g., rivers and lakes). As water transits from a groundwater environment to a low suspended-solid surface-water environment, the accumulation of radium isotopes largely ceases, but the water may retain the isotopes already accumulated. In this way, the water carries a marker of its original environment. The long-lived radium isotopes ^{226}Ra and ^{228}Ra can reveal information about the lithology through which the water traveled, such as whether the rock was higher in U than Th, as in marine carbonate rocks, or whether the Th concentration equals or exceeds that of U, as in clastic rocks and sediments. The

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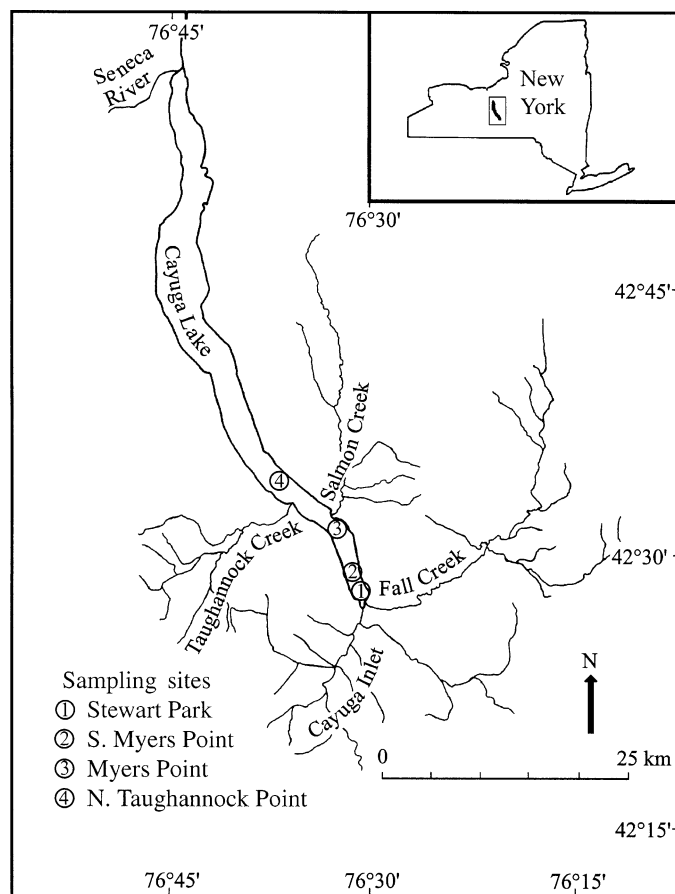


Fig. 1. Map showing location of Cayuga Lake in New York state with major tributaries. Smaller streams are not shown. Lake sampling locations are also indicated.

short-lived radium isotopes may reveal information about the timing of water emergence from the ground. These isotopes can, therefore, provide significant information about surface-water/groundwater interactions in the study of surface-water bodies such as lakes.

Materials and methods

Study area—Cayuga Lake (Fig. 1) is 1 of 11 Finger Lakes located in central New York. These lakes were formed by glacial scouring of preexisting drainage valleys during the Pleistocene, resulting in linear, narrow basins oriented with their long axes in an approximately north-south direction (von Englen 1961).

The area experiences warm summers and long, cold winters. Precipitation at Ithaca, New York, at the southern end of the lake, averages 33.7 inches annually. Nineteen percent of the precipitation falls during the winter months, December–February (Henson et al. 1961), which often results in large snow-mass accumulation in the Finger Lakes basins. As a result of rising temperature and additional precipitation in the spring, large water additions to the lake can occur during relatively short periods of time.

Cayuga Lake is ~61 km long \times 5.6 km wide at its max-

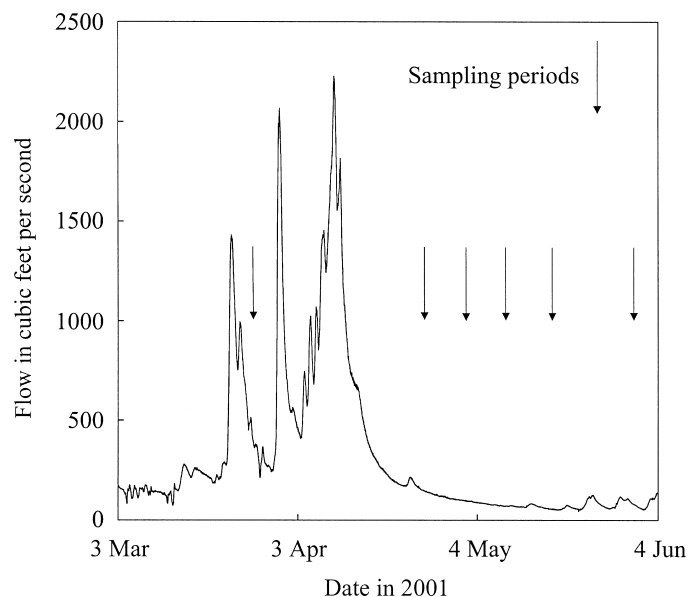


Fig. 2. Hydrograph of Fall Creek, which drains into Cayuga Lake, during the 2001 sampling. Sampling periods are marked by vertical arrows pointing down.

imum width, with a maximum depth of 133 m, a volume of 9.38×10^9 m³, and, typical for the finger lakes, a small drainage area of only 2,106 km² (Birge and Juday 1914; Henson et al. 1961). Four major tributaries (Fall Creek, Taughannock Creek, Salmon Creek, and Cayuga Inlet) deliver water from more than half of the total drainage basin area to the southern quarter of the lake (Wright 1969), and much of that water is delivered during March and April (Henson et al. 1961). The remainder of the inflow is from small, seasonal streams that occur rather uniformly throughout the basin and empty directly into the lake. Flow out of the lake is via the northern end. Seneca Lake, the largest of the Finger Lakes by volume (Birge and Juday 1914; Shaffner and Oglesby 1978), drains into the northernmost part of Cayuga Lake by way of the Seneca River but has a minimal effect on Cayuga Lake because of the inlet's close proximity to Cayuga's outlet (Effler et al. 1989). As a result of the almost direct outflow of Seneca Lake water, little interaction with the main body of Cayuga Lake takes place under most conditions. The mean residence time of water in Cayuga Lake has been estimated to be 9–21 yr (Wright 1969; Oglesby 1978; Michel and Kraemer 1995).

This combination of physical features causes a large part of the runoff generated during vernal thaw events to enter at the southern end of the lake and flow toward the northern end. The narrowness of the lake relative to its length enables convenient synoptic sampling of water and tracing of water movement as a result of the confining, narrow boundaries.

Sampling plan—During 2001, sampling was conducted as a coordinated effort to coincide with the vernal inflow, the largest that had occurred in the lake over the preceding several years. The hydrograph of Fall Creek is shown in Fig. 2. Inflow to the lake consisted of three major pulses peaking on 22 and 30 March and 9 April 2001, followed by a long

period of minimal input. The total volume of flow into the lake during this vernal event was $\sim 6 \times 10^8 \text{ m}^3$, or $\sim 6\%$ of the total lake volume, based on data from the Fall Creek hydrograph and estimates of relative annual percentages of stream flow contributions from the various tributaries presented by Wright (1969). Using Wright's estimate that half the drainage enters in the lowermost quarter of the lake, the surface inflow during the vernal inflow event into the southern quarter could be 12% of the lower part of the lake's volume.

The first sampling trip began on 26 March, on the falling limb of the first of the pulses. The other sampling trips began on 25 April and 2, 9, 17, and 31 May 2001, on the falling limb of the last major pulse of the vernal inflow. The lack of significant inflow after ~ 15 April 2001 allowed for the potential tracing of water movement and mixing that was uncomplicated by further major additions.

Radium sampling and isotopic analysis—The basic sampling procedure is to pass water through a manganese-impregnated acrylic fiber cartridge (referred to in this article as an Mn fiber), which extracts radium and other cations from solution (Moore 1976). The extraction apparatus consists of a hose, one end of which is placed in water at the appropriate depth and the other connected to a submersible pump just below the water surface. The pump feeds water first through a preweighed, 25-cm-long, 0.2- μm , polypropylene-medium filter cartridge with a filtration area of 3,300 cm^2 , then through a 25-cm-long Mn-fiber cartridge. A digital electronic flow meter (turbine type) is attached to the outlet of the Mn-fiber cartridge housing to measure the total sample volume and flow rate. The flow meter is precalibrated in the laboratory and is accurate to $\pm 2\%$ over a range of pumping rates. A sample volume of up to 500 liters is processed through the apparatus at a rate of 1–4 L min^{-1} for each sample, and the effluent is discharged away from the intake. For streams, the effluent was simply returned downstream from the intake; however, for lake stations, careful consideration of wind and current direction is essential to keep from resampling discharged water during shallow sampling.

Previous tests of this configuration have shown that radium is removed quantitatively from freshwater when this apparatus is used within the reported operating parameters. In these tests, two Mn-fiber cartridges were placed in series so that the second Mn-fiber cartridge processed the output of the first cartridge. In no case was activity on the second cartridge greater than a few percent of the first cartridge's activity. This is in agreement with previous work with these fibers (e.g., Moore and Reid 1973; Moore and Cook 1975; Moore 1976). The high degree of extraction efficiency in the present system is attributed to the large size and high capacity of the 25-cm-long Mn-fiber cartridge used, which allowed extensive contact of water with the fibers even at a high flow rate and to the low total dissolved solids of the samples, which reduced the saturation of the exchange sites on the Mn-fiber cartridge.

After the radium sample was collected, the volume was recorded, along with date, time sampling ended, and any other pertinent information. The Mn-fiber and sediment filter cartridge were removed from the housing, sealed in individ-

ually labeled polyethylene bags, and stored for transport back to the laboratory, where the sediment cartridges were air dried to constant weight and the data recorded for suspended sediment calculation. The Mn-fiber cartridges were placed in a photomultiplier tube-based counting system for the determination of ^{223}Ra and ^{224}Ra by delayed coincidence counting (Moore and Arnold 1996). An initial counting period measured the ^{219}Rn and ^{220}Rn produced by the decay of ^{223}Ra and ^{224}Ra , respectively, which were adsorbed onto the Mn-fibers. A second counting period was done ~ 1 month later, to measure any ^{220}Rn produced from ^{228}Th that may have been on the fiber and influenced the ^{224}Ra determination. The ^{223}Ra and ^{224}Ra activities in the water were calculated on the basis of the results of these two counting periods.

After the second counting period, the Mn-fibers were leached in 0.25 mol L^{-1} hydroxylamine hydrochloride and HCl to remove the manganese dioxide and adsorbed radium. The solution was filtered through a 0.45- μm membrane filter, and 6 ml of saturated $\text{Ba}(\text{NO}_3)_2$ solution was added to the filtrate and stirred. Sulfuric acid (17%) was then added to the solution to precipitate $\text{Ba}(\text{Ra})\text{SO}_4$, which was then collected, rinsed, dried, and stored for several days to allow daughter in-growth. It was then placed in the well of a high-purity germanium detector for the quantitative analysis of ^{228}Ra and ^{226}Ra by γ -ray spectrometry using a technique modified from Moore (1984). ^{228}Ra was quantified by measuring the γ -ray intensities of ^{228}Ac (338.4, 911, and 964.6–968.5 Kev), and ^{226}Ra was quantified by measuring the intensity of γ -rays of ^{214}Pb (295.2 and 351.9 Kev) and ^{214}Bi (609.3 Kev). γ -ray and photomultiplier detector systems were calibrated using standardized radium isotopic solutions adsorbed onto or precipitated with media identical to those used for sample analysis.

Results and discussion

Tributaries—Results of radium activity in the lake and water of the tributaries to Cayuga Lake are presented in Table 1. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of water from Taughanock Creek, Fall Creek, and Salmon Creek was 0.74–0.86, which indicates that tributary water has passed through material of a largely clastic nature (high Th/U ratio), resulting in a correspondingly high $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio. This is in agreement with the reported clastic nature of the glacial till covering the area (von Engel 1961). The absolute values of ^{228}Ra and ^{226}Ra activities vary significantly among tributaries, indicating differences of availability of these nuclides in the drainage basin material surrounding the lake.

The ^{223}Ra and ^{224}Ra activities of the tributary water were usually higher than that of the lake water, except where and when the lake has been influenced by surface or groundwater inflow (some lake samples actually exceed the tributary values; e.g., Stewart Park on 27 March), inasmuch as tributary water has only recently emerged at the surface after moving as groundwater in contact with solid-phase material, which supplies the short-lived isotopes.

The $^{223}\text{Ra}/^{226}\text{Ra}$ activity ratios of the main tributary waters were 0.035–0.041, and the $^{224}\text{Ra}/^{228}\text{Ra}$ activity ratios were

Table 1. Radium isotopic data for Cayuga Lake, New York.

Sample	Depth (m)	^{226}Ra (dpm [100L] $^{-1}$)	^{228}Ra (dpm [100L] $^{-1}$)	^{223}Ra (dpm [100L] $^{-1}$)	^{224}Ra (dpm [100L] $^{-1}$)	$^{223}\text{Ra}/^{226}\text{Ra}$ (activity ratio)	$^{224}\text{Ra}/^{223}\text{Ra}$ (activity ratio)	$^{228}\text{Ra}/^{226}\text{Ra}$ (activity ratio)
26–28 Mar 2001								
N. Taughanock Pt.	16.5	4.4 ± 0.2	3.6 ± 0.2	0.039 ± 0.006	1.0 ± 0.1	0.0089 ± 0.0014	0.28 ± 0.02	0.82 ± 0.05
Myers Pt.	0.3	6.7 ± 0.2	4.7 ± 0.4	0.019 ± 0.008	0.50 ± 0.01	0.0028 ± 0.0012	0.11 ± 0.01	0.69 ± 0.07
Myers Pt.	16.5	3.9 ± 0.1	2.9 ± 0.1	0.015 ± 0.003	0.27 ± 0.01	0.0038 ± 0.0008	0.09 ± 0.01	0.75 ± 0.03
S. Myers Pt.	0.3	4.9 ± 0.2	3.9 ± 0.4	0.16 ± 0.05	3.4 ± 0.1	0.032 ± 0.010	0.89 ± 0.10	0.79 ± 0.09
S. Myers Pt.	16.5	4.7 ± 0.1	3.4 ± 0.2	0.058 ± 0.008	1.1 ± 0.1	0.012 ± 0.002	0.30 ± 0.02	0.73 ± 0.04
Myers Pt. Plume	0.3	8.3 ± 0.3	6.5 ± 0.6	0.53 ± 0.05	9.2 ± 0.3	0.063 ± 0.007	1.4 ± 0.1	0.78 ± 0.07
Stewart Park	0.3	11.2 ± 0.2	4.4 ± 0.3	0.61 ± 0.05	3.7 ± 0.2	0.059 ± 0.005	0.83 ± 0.06	0.40 ± 0.03
Fall Creek	—	5.4 ± 0.3	4.0 ± 0.4	0.20 ± 0.02	4.6 ± 0.2	0.036 ± 0.005	1.1 ± 0.1	0.74 ± 0.08
Salmon Creek	—	8.4 ± 0.1	6.3 ± 0.2	0.29 ± 0.02	5.5 ± 0.1	0.035 ± 0.002	0.87 ± 0.03	0.75 ± 0.02
Taughanock Cr.	—	3.9 ± 0.2	3.4 ± 0.3	0.16 ± 0.03	2.4 ± 0.1	0.041 ± 0.007	0.70 ± 0.07	0.86 ± 0.09
24–25 Apr 2001								
N. Taughanock Pt.	16.5	5.7 ± 0.2	2.8 ± 0.3	0.037 ± 0.005	0.18 ± 0.01	0.007 ± 0.001	0.064 ± 0.010	0.49 ± 0.05
Myers Pt.	0.3	3.2 ± 0.1	2.2 ± 0.2	0.016 ± 0.003	0.24 ± 0.02	0.005 ± 0.001	0.11 ± 0.01	0.68 ± 0.07
Myers Pt.	16.5	8.4 ± 0.1	3.5 ± 0.2	0.29 ± 0.01	0.66 ± 0.03	0.034 ± 0.002	0.19 ± 0.02	0.41 ± 0.03
S. Myers Pt.	0.3	4.8 ± 0.1	3.8 ± 0.2	0.049 ± 0.005	1.2 ± 0.1	0.010 ± 0.001	0.31 ± 0.02	0.78 ± 0.04
S. Myers Pt.	16.5	6.5 ± 0.2	2.6 ± 0.3	0.17 ± 0.01	0.76 ± 0.03	0.026 ± 0.002	0.29 ± 0.04	0.40 ± 0.04
02 May 2001								
N. Taughanock Pt.	0.3	4.5 ± 0.3	3.4 ± 0.2	0.009 ± 0.003	0.27 ± 0.02	0.0020 ± 0.0006	0.079 ± 0.007	0.76 ± 0.07
N. Taughanock Pt.	16.5	3.8 ± 0.3	3.4 ± 0.2	0.007 ± 0.002	0.27 ± 0.02	0.0018 ± 0.0005	0.079 ± 0.008	0.89 ± 0.08
Myers Pt.	0.3	4.1 ± 0.1	3.2 ± 0.2	0.011 ± 0.004	0.42 ± 0.03	0.0027 ± 0.0010	0.13 ± 0.01	0.79 ± 0.05
Myers Pt.	16.5	4.8 ± 0.2	3.5 ± 0.2	0.032 ± 0.003	0.24 ± 0.01	0.0067 ± 0.0007	0.069 ± 0.004	0.73 ± 0.07
S. Myers Pt.	0.3	4.6 ± 0.1	3.8 ± 0.1	0.036 ± 0.003	1.1 ± 0.1	0.0078 ± 0.0007	0.28 ± 0.01	0.82 ± 0.03
S. Myers Pt.	16.5	4.1 ± 0.2	3.3 ± 0.2	0.013 ± 0.002	0.32 ± 0.01	0.0032 ± 0.0005	0.10 ± 0.01	0.79 ± 0.06
09 May 2001								
N. Taughanock Pt.	0.3	4.8 ± 0.1	3.8 ± 0.2	0.017 ± 0.004	0.25 ± 0.02	0.0036 ± 0.0009	0.066 ± 0.007	0.80 ± 0.06
N. Taughanock Pt.	16.5	4.7 ± 0.2	3.7 ± 0.2	0.011 ± 0.002	0.28 ± 0.01	0.0023 ± 0.0004	0.075 ± 0.005	0.79 ± 0.05
Myers Pt.	0.3	4.7 ± 0.1	3.8 ± 0.2	0.020 ± 0.004	0.63 ± 0.03	0.0043 ± 0.0009	0.17 ± 0.01	0.81 ± 0.05
Myers Pt.	16.5	4.0 ± 0.2	3.6 ± 0.2	0.022 ± 0.003	0.28 ± 0.01	0.0055 ± 0.0008	0.077 ± 0.005	0.91 ± 0.07
S. Myers Pt.	0.3	4.2 ± 0.1	3.4 ± 0.1	0.028 ± 0.003	0.92 ± 0.02	0.0067 ± 0.0007	0.27 ± 0.01	0.79 ± 0.03
S. Myers Pt.	16.5	4.6 ± 0.2	3.2 ± 0.2	0.028 ± 0.002	0.28 ± 0.01	0.0017 ± 0.0004	0.086 ± 0.007	0.70 ± 0.06
16–17 May 2001								
S. Myers Pt.	0.3	4.3 ± 0.2	3.6 ± 0.2	0.035 ± 0.004	1.1 ± 0.1	0.0082 ± 0.0010	0.30 ± 0.02	0.83 ± 0.06
S. Myers Pt.	16.5	4.0 ± 0.1	2.9 ± 0.2	0.016 ± 0.003	0.27 ± 0.02	0.0040 ± 0.0008	0.09 ± 0.01	0.72 ± 0.06
Myers Pt. Plume	0.3	5.1 ± 0.2	3.8 ± 0.3	0.033 ± 0.005	0.90 ± 0.03	0.0065 ± 0.0010	0.24 ± 0.03	0.74 ± 0.06
Myers Pt. Plume rep	0.3	4.2 ± 0.1	3.4 ± 0.1	0.041 ± 0.004	0.89 ± 0.03	0.0099 ± 0.0010	0.26 ± 0.01	0.81 ± 0.04
Stewart Park	0.3	4.5 ± 0.2	3.5 ± 0.3	0.070 ± 0.004	1.4 ± 0.1	0.016 ± 0.001	0.39 ± 0.03	0.78 ± 0.07
Stewart Park rep	0.3	4.6 ± 0.2	3.7 ± 0.2	0.030 ± 0.006	0.78 ± 0.03	0.0065 ± 0.0013	0.21 ± 0.01	0.81 ± 0.06
31 May 2001								
Myers Pt.	0.3	4.7 ± 0.1	3.1 ± 0.2	0.018 ± 0.003	0.17 ± 0.02	0.0039 ± 0.0007	0.054 ± 0.007	0.67 ± 0.05
Myers Pt.	16.5	4.4 ± 0.1	3.1 ± 0.1	0.010 ± 0.003	0.17 ± 0.02	0.0023 ± 0.0007	0.054 ± 0.007	0.71 ± 0.02
S. Myers Pt.	0.3	4.2 ± 0.2	3.2 ± 0.2	0.005 ± 0.002	0.27 ± 0.02	0.0012 ± 0.0005	0.084 ± 0.008	0.77 ± 0.06
S. Myers Pt.	16.5	4.6 ± 0.1	3.2 ± 0.2	0.017 ± 0.003	0.16 ± 0.01	0.0037 ± 0.0007	0.050 ± 0.004	0.69 ± 0.045
Stewart Park	0.3	5.6 ± 0.1	4.5 ± 0.1	0.091 ± 0.009	3.3 ± 0.1	0.016 ± 0.002	0.74 ± 0.02	0.81 ± 0.03
Stewart Park rep	0.3	5.6 ± 0.3	4.3 ± 0.2	0.093 ± 0.007	3.2 ± 0.1	0.017 ± 0.002	0.74 ± 0.04	0.76 ± 0.05

dpm [100 L] $^{-1}$ disintegration per minute per 100 L, errors reported as SE based on counting statistics.

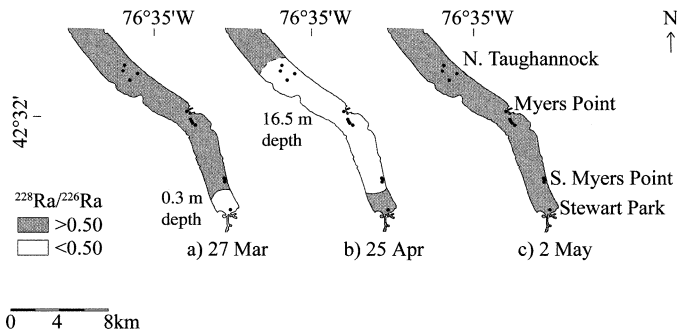


Fig. 3. Location map showing position of low $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio water in Cayuga Lake on (a) 27 March, (b) 25 April, and (c) 2 May 2001, apparently caused by groundwater inflow from carbonate aquifer to the lake as a result of elevated water table during the vernal thaw. Dots represent locations of samples during all sampling times.

0.70–1.1. The absolute activities of ^{223}Ra and ^{224}Ra in the tributaries were higher than lake values, except, again, where and when the lake receives flow from the tributaries. The spread of values of the activity ratios for the tributaries were near or even higher than the closed system values of 0.0466 for $^{223}\text{Ra}/^{226}\text{Ra}$ and 1.00 for $^{224}\text{Ra}/^{228}\text{Ra}$ that would be expected on the basis of secular equilibrium and natural abundances, attesting to recent contact with aquifer material. Under conditions of the rapid transport of water through porous clastic material, these ratios can become higher than the closed-system values described above. This is the result of the α -particle recoil process, whereby short-lived isotopes accumulate to their equilibrium values faster than longer-lived isotopes.

Stewart Park water mass—The first sampling trip (26–28 March 2001) occurred early during the vernal inflow event, just after the first major pulse of water. The results from the Stewart Park sampling site on 27 March 2001 were quite different from all of the other samples taken at this time (including tributary samples), in that they had the highest ^{226}Ra and ^{223}Ra activities (11.2 and 0.61 disintegrations per minute per 100 L [$\text{dpm } 100 \text{ L}^{-1}$], respectively) and lowest $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio (0.40) of any of the other samples obtained during the period when the lake was receiving high inflow (Fig. 3a). This indicates that a source of water unlike any of the sampled surface inflows was contributing significantly to the lake at this shallow southern site. Because of the low $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio, it is inferred that the water in the southern part of the lake was supplied from a carbonate aquifer, which would be characterized by a low Th/U ratio typical of marine carbonate deposits. The Tully limestone crops out along the extreme southern end of the lake (Oglesby 1978) and is a possible source for this water. It is possible that a high water table occurs in the surrounding basin during spring inflow events. This temporary condition may provide enough head to force groundwater from the carbonate aquifer through the lake bottom and into the lake during these times.

Water with $^{228}\text{Ra}/^{226}\text{Ra}$ isotopic activity ratios similar to those found at Stewart Park in March 2001 was observed

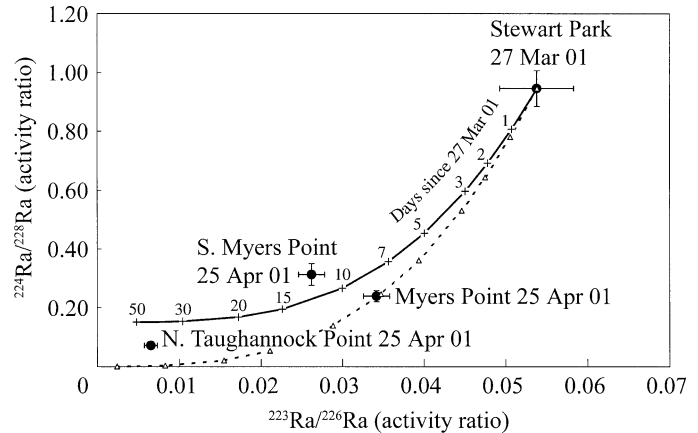


Fig. 4. $^{223}\text{Ra}/^{226}\text{Ra}$ and $^{224}\text{Ra}/^{228}\text{Ra}$ decay plot of Stewart Park water body with time. The heavy curved line represents the evolution of radium isotopes in the water body as they decay from the initial ratios of 0.0538 and 0.95, respectively, to a final value sustained by suspended sediment (represented by the 50-d point arbitrarily chosen to fit through the data points). The numerals represent the time, in days, since 27 March 2001. The data points represent the radium isotope activity ratios for the locations sampled on 25 April 2001.

on the subsequent sampling trip starting on 16 April 2001 at the 16.5-m sampling depths at the North Taughannock Point, Myers Point, and South Myers Point sampling sites (Fig. 3b). The sampling time was ~ 15 d after the last (and largest) peak inflow on 9–10 April. Water with a similarly low $^{228}\text{Ra}/^{226}\text{Ra}$ isotopic ratio was not observed in the shallow locations at these sites, only the higher $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio water more commonly found in the lake and surface streams during this (and previous) samplings. The water that was in the extreme southern end of the lake at Stewart Park in March had migrated northward by as much as 14 km, to at least as far as the North Taughannock Point sampling site, as an elongating pod encompassing the 16.5 m depth, perhaps confined vertically by density equilibrium after coming off the shallow southern shelf of the lake, before sinking below 16.5 m depth, mixing with old lake water, or being flushed beyond North Taughannock Point before the 2 May 2001 sampling (Fig. 3c).

The short-lived radium isotopes ^{223}Ra and ^{224}Ra provide additional information about the Stewart Park water pod. The $^{223}\text{Ra}/^{226}\text{Ra}$ versus $^{224}\text{Ra}/^{228}\text{Ra}$ data are shown in Fig. 4. The curved lines represent the theoretical decay of ^{223}Ra and ^{224}Ra isotopes in a water mass from an initial dissolved value (i.e., Stewart Park on 27 March) over 50 d within the Stewart Park pod. The decay curves in Fig. 4 are based on normalizing each short-lived isotope to a longer-lived isotope, to decrease any effect of chemical removal (e.g., adsorption or precipitation) while preserving the variability due to decay alone. The dotted curved line represents the decay path that Stewart Park water would take if the ^{223}Ra and ^{224}Ra decreased by radioactive decay alone, with no other source of loss or supply. The solid curved line represents this same decay process but with a constant resupply of a small amount of ^{223}Ra and ^{224}Ra from suspended sediment. The suspended sediment-generated values used in constructing

the solid curved line (0.0047 for $^{223}\text{Ra}/^{226}\text{Ra}$ and 0.152 for $^{224}\text{Ra}/^{228}\text{Ra}$) were selected to give the best fit through the 26 April data points for North Taughannock Point, Myers Point, and South Myers Point. A more detailed discussion of the sediment supply of these nuclides is discussed in a later section.

The data points for the North Taughannock Point, Myers Point and South Myers Point samples plot around the augmented theoretical decay line constructed from the Stewart Park water point, with the oldest water of the sample set corresponding with the northernmost location at North Taughannock Point. This apparent age relationship with distance north supports the concept of a body of water growing in size by continuous addition from the south, moving and spreading as a unit and mixing with old lake water, rather than water infiltrating into the lake along a great length of shoreline simultaneously, which is a possible alternative scenario. Considerable error may result in trying to assign an age to the water from such small ^{223}Ra and ^{224}Ra activities as are present in the North Taughannock Point sample. This is probably the reason that the calculated age of the North Taughannock Point water is greater than the time between the start of the vernal inflow and sampling (~ 39 d). In addition, the data set is not totally internally consistent, in that the water at the Myers Point site may be interpreted as slightly younger than the water at South Myers Point, 5 km further south. This plot serves to illustrate, however, that short-lived radium isotopes may be useful in monitoring the relative passage of time with respect to water bodies emplaced into a lake.

Myers Point delta groundwater inflow—A second inflow-lake interaction event detected using radium isotopes during the 2001 vernal inflow event occurred in the lake near Myers Point delta, where Salmon Creek enters the lake (Fig. 1). The Myers Point Plume sampling site is a few meters offshore from the mouth of the creek. Because of the relatively high volume of flow in the creek on the March 2001 trip and the nearness of the sampling site to the mouth of the creek (Fig. 5), it was hypothesized that the water at this site would consist of significant fractions of lake and creek water and that mixing relationships would dominate the data, both in the chemical and radium isotopic analyses. The results of the chemical analyses (Table 2) indicate that, for the most part, the water at the Myers Point Plume site is nearly indistinguishable from Salmon Creek water and is very different from the rest of the lake. For example, Salmon Creek and Myers Point Plume samples had significantly higher and nearly identical HCO_3^- , Mg^{+2} , Ca^{+2} , and NO_3^{-2} concentrations and lower Cl^- and Na^+ concentrations than the other lake samples. It is clear that the water at these two sites is related. However, there were significant differences in the $^{223}\text{Ra}/^{226}\text{Ra}$ and $^{224}\text{Ra}/^{228}\text{Ra}$ activity ratios (Table 1). These ratios were higher in the Myers Point Plume sample (0.063 and 1.4, respectively) than in the Salmon Creek sample (0.035 and 0.87, respectively), indicating that the plume sample is neither unaltered Salmon Creek water nor a mixture of Salmon Creek water and lake water. The isotopes ^{223}Ra and ^{224}Ra , being short-lived, would decay if they were not resupplied from some solid source containing uranium

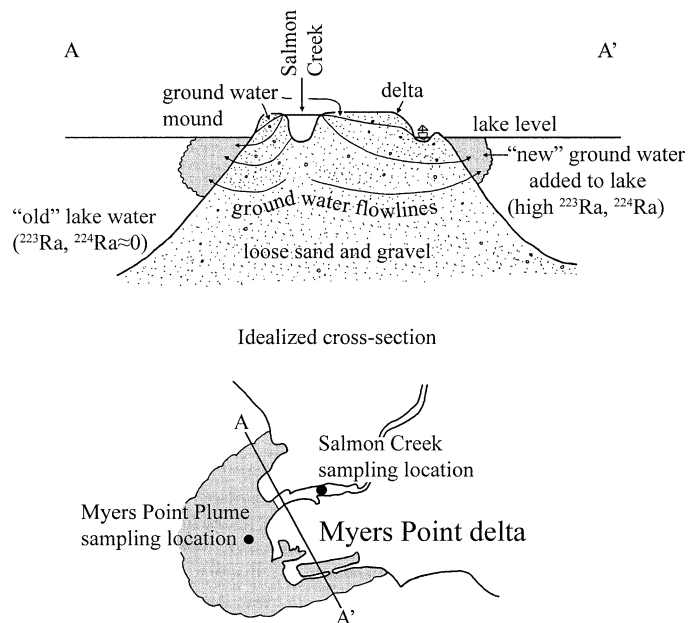


Fig. 5. Idealized cross section of the Myers Point delta showing the mounding of groundwater hypothesized to occur during the high waters of a vernal inflow. Groundwater flow and water from the channel of Salmon Creek infiltrates through the sand and gravel of the delta, gaining short-lived ^{223}Ra and ^{224}Ra by α -particle recoil from the solid phase, then flows into the lake below the surface as groundwater inflow.

and thorium, but, in this case, the water at the Myers Point Plume location has gained additional quantities of short-lived radium isotopes compared with Salmon Creek water.

The most reasonable way to account for the observations at the Salmon Creek–Myers Point Plume area is to hypothesize that the high head of the mounded water table in and landward of the delta caused by infiltration of water through the upland glacial till and stream-bed deposits forces the groundwater through the delta toward its lake-bound margins where it emerges below lake level as groundwater inflow into the lake (Fig. 5). In so doing, the major-element chemistry of the water is little changed from the sampled Salmon Creek composition except for the supply of ^{223}Ra and ^{224}Ra to the water from the clastic material (or reintroduction, in the case of surface water reentering the deltaic material through the stream bed). This results in the higher ratios found in the lake at the sampling site. The $^{223}\text{Ra}/^{226}\text{Ra}$ and $^{224}\text{Ra}/^{228}\text{Ra}$ activity ratios in the lake some distance from the delta (e.g., Myers Point location) were very low, representing only the small amount of short-lived radium isotopes supplied from uranium and thorium decay from particulate matter suspended in the water column.

South Myers Point area—The third major inflow-lake interaction phenomenon was observed at the South Myers Point sampling site and persisted from March to May 2001. Radium isotopic data indicated that a body of water was added to the surface of the lake during the vernal inflow event and retained its identity as a separate water body for several weeks after emplacement. Unlike the deeper Stewart

Table 2. Chemical analysis of Cayuga Lake, New York.

Sample	Date	Ca (mg L ⁻¹)	Mg (mg L ⁻¹)	Na (mg L ⁻¹)	K (mg L ⁻¹)	Sr (mg L ⁻¹)	Si (mg L ⁻¹)	HCO ₃ (mg L ⁻¹)	Cl (mg L ⁻¹)	Br (mg L ⁻¹)	NO ₃ (mg L ⁻¹)	SO ₄ (mg L ⁻¹)
Taughannock Cr.	26 Mar 2001	44	8.4	17	1.8	0.08	1.8	145	32	0.27	12	21
Fall Creek	27 Mar 2001	41	7.9	20	1.5	0.07	1.6	135	38	0.08	8.5	15
Salmon Creek	28 Mar 2001	70	18.7	13	2.4	0.11	1.7	230	22	0.05	35	24
Myers Pt. Plume surface	27 Mar 2001	68	18.0	13	2.5	0.11	1.8	215	22	0.03	35	23
N. Taughannock Pt. 16.5 m	26 Mar 2001	41	10.8	25	2.4	0.21	0.8	145	41	0.04	5.8	33
Myers Point 16.5 m	28 Mar 2001	42	10.7	24	2.3	0.20	0.9	140	41	0.04	6.0	32
S. Myers Pt. 16.5 m	28 Mar 2001	40	10.4	25	2.4	0.20	0.9	135	42	0.72	6.1	32
Salmon Creek	24 Apr 2001	59	19.0	13	2.7	0.12	0.5	205	24	0.36	22	25
Taughannock Cr.	24 Apr 2001	47	8.9	13	1.8	0.08	1.3	155	27	bdl	7.7	21
N. Taughannock Pt. 16.5 m	25 Apr 2001	41	10.8	24	2.4	0.20	0.9	135	42	0.02	6.1	33
Myers Pt. 16.5 m	25 Apr 2001	41	10.5	24	2.3	0.20	0.9	133	42	bdl	6.1	32
S. Myers Pt. 16.5 m	25 Apr 2001	41	10.4	24	2.4	0.19	0.9	140	42	0.03	6.4	32
N. Taughannock Pt. 16.5 m	2 May 2001	41	10.7	24	2.4	0.20	0.9	130	40	0.02	4.3	32
Myers Point 16.5 m	2 May 2001	42	10.6	23	2.4	0.20	0.9	125	40	0.02	4.4	32
S. Myers Pt. 16.5 m	2 May 2001	42	10.7	24	2.5	0.20	1.0	130	39	0.01	4.1	31
N. Taughannock Pt. 16.5 m	9 May 2001	40	10.3	23	2.4	0.20	0.6	130	43	0.05	5.5	23
Myers Point 16.5 m	9 May 2001	37	9.5	23	2.3	0.19	0.7	120	42	0.02	5.5	23
S. Myers Pt. 16.5 m	9 May 2001	36	9.2	22	2.1	0.18	0.2	125	42	0.02	5.4	23
Myers Pt. Plume	17 May 2001	36	9.5	22	2.2	0.19	0.35	120	43	0.02	5.2	23
S. Myers Pt. 16.5 m	16 May 2001	37	9.6	22	2.2	0.19	0.5	125	43	0.02	5.1	23
Stewart Park	16 May 2001	39	10.1	23	2.3	0.20	0.6	125	43	0.02	5.1	23
Myers Point 16.5 m	31 May 2001	35	9.1	22	2.0	0.18	bdl	120	43	0.02	5.1	23
S. Myers Point 16.5 m	31 May 2001	37	9.8	23	2.3	0.20	bdl	125	43	0.02	5.2	24
Stewart Park	31 May 2001	32	7.1	22	1.5	0.12	0.22	110	39	0.02	5.3	18

bdl = below detection limit (Br = 0.01 mg L⁻¹, Si = 0.05 mg L⁻¹).

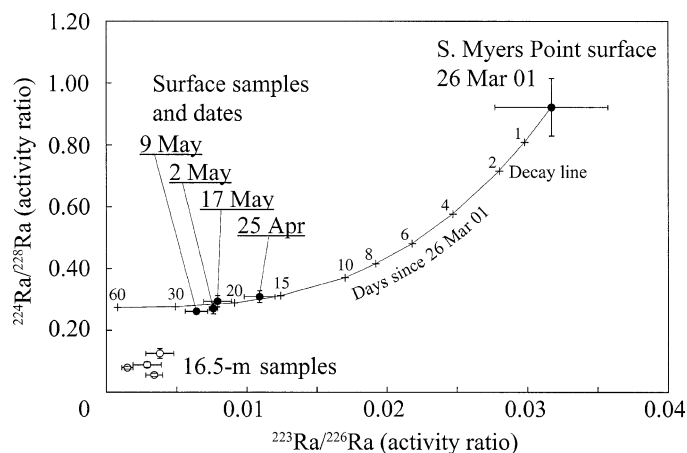


Fig. 6. $^{223}\text{Ra}/^{226}\text{Ra}$ vs. $^{224}\text{Ra}/^{228}\text{Ra}$ activity ratio decay plot of surface water in Cayuga Lake at the South Myers Point sampling site. The solid line represents the path that radium isotopes would follow as the water at South Myers Point sampled on 26 March 2001 aged as a distinct water body without mixing with the bulk lake water, (represented by the 16.5-m samples). The numerals are the time, in days, after 26 March 2001. The surface samples at South Myers Point are distinct from the 16.5-m depth samples and fall on or near the decay line, indicating that this water remained in place and distinct from the bulk lake water for a period of several weeks. The values for the suspended-sediment maintained ratios were arbitrarily chosen, as in Fig. 4, to fit through the surface data points.

Park pod, however, this water body did not move rapidly to the north but persisted in the same location for a considerable period of time.

The South Myers Point samples (at depths of 0.3 and 16.5 m) had high $^{223}\text{Ra}/^{226}\text{Ra}$ and $^{224}\text{Ra}/^{228}\text{Ra}$ activity ratios for March 2001, in particular, and the 0.3-m samples were also relatively high throughout most of the subsequent sampling periods. These values, as well as the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios, were similar to the Myers Point Plume site when it was first sampled in March, which may indicate a similar mode of shallow groundwater emplacement. Southward transport of water introduced at Myers Point could account for this occurrence, or there could be groundwater input into the lake all along the southeast section of the lake. It is not likely, however, that this water is from the same source that was active at Stewart Park in March, because the $^{228}\text{Ra}/^{226}\text{Ra}$ ratio was distinctly different (0.40 for Stewart Park and 0.73 for South Myers Point at a depth of 16.5 m on 26 March 2001).

The data from the South Myers Point site for sampling periods from March through May 2001 are shown in Fig. 6. The shallow samples plot as a distinct class above the deeper water (16.5 m) samples and include the sample taken on 26 March 2001, which has very high $^{223}\text{Ra}/^{226}\text{Ra}$ and $^{224}\text{Ra}/^{228}\text{Ra}$ activity ratios. If this 26 March surface sample represents water that was emplaced during the vernal surface-water inflow period, the radium isotopes would decay with time along the line shown in Fig. 6, under the assumption of a final equilibrium $^{223}\text{Ra}/^{226}\text{Ra}$ activity ratio of 0.001 and a final equilibrium $^{224}\text{Ra}/^{228}\text{Ra}$ activity ratio of 0.025 (selected, as in the Stewart Park discussion, so that the line would fit through the surface points and is governed primarily by equilibrium with the suspended material in the water). This

Table 3. Suspended sediment concentrations of Cayuga Lake water and tributaries.

Location	Depth (m)	Date	Suspended solids (mg L^{-1})
N. Taughannock Pt.	0.3	2 May 2001	0.7
		9 May 2001	0.9
N. Taughannock Pt.	16.5	26 Mar 2001	1.0
		25 Apr 2001	1.4
		2 May 2001	1.0
		9 May 2001	0.8
Myers Point	0.3	26 Mar 2001	1.1
		25 Apr 2001	1.1
		2 May 2001	1.2
		9 May 2001	0.6
Myers Point	16.5	28 Mar 2001	1.4
		25 Apr 2001	1.7
		2 May 2001	1.0
		9 May 2001	0.9
Myers Point Plume	0.3	27 Mar 2001	12.6
		S. Myers Point	0.3
S. Myers Point	0.3	25 Apr 2001	3.0
		2 May 2001	2.9
		9 May 2001	1.1
		16 May 2001	1.9
		26 Mar 2001	1.4
		25 Apr 2001	1.7
S. Myers Point	16.5	2 May 2001	1.0
		9 May 2001	0.9
		27 Mar 2001	10.2
Stewart Park	0.3	27 Mar 2001	10.2
Taughannock Creek		26 Mar 2001	8.1
Salmon Creek		28 Mar 2001	7.9
Fall Creek		27 Mar 2001	7.1

provides strong evidence that the water added to the lake during the spring inflow remained at the surface of the lake at this site for an extended period of time and retained its physical characteristics during this period with minimal mixing with the bulk lake (lower short-lived radium isotope activity) water. The plotted location of the shallow samples above the deep samples on the graph represents the remaining short-lived isotopes initially present in the water plus the standing crop of the short-lived isotopes produced from suspended sediment in the introduced water body, which plots higher on the graph than in the deeper (older) lake water, in part because it has not had sufficient time to lose sediment by settling, compared with the older lake water. This is in agreement with the higher suspended-sediment concentration of the shallower waters, compared with most of the deep lake-water samples at this site on any sampling date (Table 3).

The measured values of the samples plotted very close to the decay curve of the 26 March sample in Fig. 6, and, for the most part, samples taken later plotted closer to the lower end of the curve than did earlier samples. The 17 May sample, however, was contrary to this trend, plotting out of sequence (younger) than the other samples. It is not prudent, therefore, to assign ages to the samples taken on various dates, especially because there was likely to have been continued inflow into the lake past the March sampling date.

Other lake water observations—On 31 May 2001, samples were taken after two relatively minor surface-water inflow events into the lake (Fig. 2). Definite effects were seen only at the Stewart Park site, where $^{224}\text{Ra}/^{228}\text{Ra}$ and $^{223}\text{Ra}/^{226}\text{Ra}$ activity ratios increased to 0.74 and 0.017, respectively (Table 1). This indicates that freshened water was added to the extreme south end of the lake. The relatively small addition caused by the minor inflow event was noticeable because of the shallow nature of this end of the lake. Significantly, the high $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of the water at this site does not imply a reintroduction of the same type or amount of water (low $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio) found in and near the southern end of the lake during the first two sampling trips. The event probably added surface inflow to the lake rather than groundwater inflow, with no resulting decrease in the $^{228}\text{Ra}/^{226}\text{Ra}$ isotopic activity ratio of the lake water and shows the sensitivity of these isotopes to changes in surface water systems.

Supply of radium isotopes from suspended sediment—Table 3 presents the results of the suspended sediment measurements for the sample locations and reveals low concentrations ($\sim 2 \text{ mg L}^{-1}$ or less) of suspended sediment in the lake, except where influenced by heavy vernal tributary inflow (such as at the Myers Point Plume and Stewart Park locations).

The average suspended sediment of lake water unaffected to any significant degree by the vernal inflow is 1.0 mg L^{-1} . This figure, along with the uranium content of suspended sediment, can be used to test the assumption that, in old lake water, the ^{223}Ra and ^{224}Ra activities in solution are due to supply from the suspended sediment. Suspended sediment from Cayuga Inlet and bottom sediment from the lake, taken before the present study and the only sources from which sufficient sample could be physically recovered for uranium analysis, is $\sim 3 \mu\text{g g}^{-1}$ of sediment, dry weight. This corresponds to $\sim 2.3 \text{ dpm g}^{-1}$ of ^{238}U in the sediment and $\sim 0.1 \text{ dpm g}^{-1}$ for ^{235}U , under the assumption of a $^{235}\text{U}/^{238}\text{U}$ activity ratio of 0.0466 for the uranium. Assuming 1.0 mg L^{-1} of suspended sediment and secular equilibrium between the ^{235}U and ^{223}Ra , the maximum amount of ^{223}Ra available from the suspended sediment to the water is $1 \times 10^{-4} \text{ dpm L}^{-1}$. This is similar to the average measured value of $2 \times 10^{-4} \text{ dpm L}^{-1}$ of ^{223}Ra in water that has not been influenced by addition of new water from tributary input. This implies that most of the ^{223}Ra in suspended sediment would enter the dissolved phase as a result of the dislocations caused by the α -decay process of parental nuclides in the decay chains.

A similar calculation for the ^{224}Ra would require a value for ^{232}Th for the sediment, which is unavailable. However, assuming a common 4:1 mass ratio for Th/U in the sediment, the ^{232}Th activity in the sediment would be $\sim 3 \text{ dpm g}^{-1}$, and the maximum dissolved value at 1 mg L^{-1} suspended sediment concentration would be $3 \times 10^{-3} \text{ dpm L}^{-1}$, very close to what was actually observed in lake water that has not received freshened inflow.

In an attempt to determine the distribution coefficient (K_d) of radium between suspended sediment and water in Cayuga Lake, the suspended-sediment filter cartridges were counted numerous times over a period of several months in an at-

tempt to detect unsupported ^{223}Ra and ^{224}Ra decay (Sun and Torgersen 1998). In theory, the quantity of radium isotopes that decay over periods longer than the nuclide half-lives would represent unsupported (adsorbed) radium. This quantity and the dissolved ^{224}Ra and ^{223}Ra activity could be used to calculate the radium distribution coefficient. Results, however, showed no clearly defined trends for ^{223}Ra or ^{224}Ra with time in these samples. The K_d values of radium in freshwater-sediment environments has been reported to be in the 10^2 – 10^5 range (Dickson 1990; Benes 1990). Excess ^{223}Ra and ^{224}Ra (unsupported by parents in the sediment particles) should be located on sediment surfaces and its presence easily detected (through its decay) by the means used in this study. This lack of excess surface activity on the suspended sediment, in conjunction with the fact that most of the ^{223}Ra in solution in the lake water has been calculated to come from decay of parent nuclide within, not necessarily only adsorbed onto the surface of, sediment suggests that the chemical equilibrium between sediment and water phases in this freshwater system is not achieved on the timescale of ^{223}Ra and ^{224}Ra half-lives. This, in turn, would imply that the short-lived radium isotopic observations described for the water masses in this article are largely unaffected by sediment-water equilibrium interactions; the low temperature of the water ($< 4^\circ\text{C}$) at that time of year may not promote attainment of chemical equilibrium and may be a significant factor in the observed non-equilibrium state.

It is apparent that suspended sediment is responsible for the ^{223}Ra and ^{224}Ra in lake water, but it is not likely that this source is responsible for what appears to be a large inventory of ^{228}Ra found in the lake water. The residence time of water in this lake is 9–21 yr, or 2–4 half-lives of ^{228}Ra . Therefore, water in the lake should have a considerably lower $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio than tributary water. It was observed however, that this ratio in lake water (0.67–0.91, excluding the Stewart Park pod samples) was not significantly lower than that of the output from the tributaries (0.74–0.86). The supply of ^{228}Ra from suspended sediment could not be larger than that calculated for ^{224}Ra ($0.3 \text{ dpm } 100 \text{ L}^{-1}$) which is considerably below that seen in the lake (2.2 – $4.7 \text{ } 100 \text{ L}^{-1}$). Therefore, a second source of ^{228}Ra must be present in the lake environment. It is unlikely that surface water or groundwater vastly different from those sampled is contributing to the bulk lake budget, but bottom sediment could be a source. Studies in the estuarine environment (Key et al. 1985; Krest et al. 1999) have shown that ^{228}Ra commonly is enriched in overlying water due to the regeneration of ^{228}Ra in the sediment by α -recoil from the particles. The results of this study provide evidence that the same process occurs in the sediments of freshwater environments and may be useful as a tool in studying sediment-water interaction in these environments.

Evidence of mixing in the Stewart Park water mass—An examination of the ^{226}Ra and ^{228}Ra activities of the Stewart Park water mass samples taken during the March and April sampling periods clearly show radium decrease with distance northward. This is most likely the result of mixing between the pod water and older lake water caused by turbulence as the pod moves through the lake. The high initial ^{228}Ra and

Table 4. Mixing ratios and age calculations in the Stewart Park pod water mass.

Sample	Date	^{223}Ra (dpm [100L] $^{-1}$)	^{224}Ra (dpm [100L] $^{-1}$)	^{226}Ra (dpm [100L] $^{-1}$)	^{228}Ra (dpm [100L] $^{-1}$)	% of old water
Mixing proportions of old lake water with Stewart Park pod water mass over time						
Stewart Park	27 Mar 2001	0.61	3.7	11.2	4.4	0
S. Myers Point	25 Apr 2001	0.17	0.76	6.5	2.6	69
Myers Point	25 Apr 2001	0.29	0.66	8.4	3.5	41
N. Taughannock Pt.	25 Apr 2001	0.037	0.18	5.7	2.8	81
Average of old lake water		0.016	0.28	4.4	3.3	100
				$^{234}\text{Ra}/^{223}\text{Ra}^*$ calculated	$^{224}\text{Ra}/^{223}\text{Ra}^\dagger$ corrected	Age ‡ (days) (equation 2)
Radium isotopes in Stewart Park pod water mass corrected for mixing						
Stewart Park	27 Mar 2001	0.61	3.7	6.06	5.70	0
S. Myers Point	25 Apr 2001	0.164	0.57	3.5	2.07	7.9 \pm 1.0
Myers Point	25 Apr 2001	0.28	0.56	4.1	1.13	12.6 \pm 1.5
N. Taughannock Pt.	25 Apr 2001	0.026	-0.05	—	—	—

† Corrected for mixing and ^{223}Ra and ^{224}Ra contributed by suspended sediment.

* Corrected for mixing only.

‡ Error based on propagation of counting statistical error only.

^{226}Ra activities in the pod allow monitoring of the mixing process and estimation of the degree of mixing. Table 4 shows the radium isotopic activities of the Stewart Park pod samples, plus the estimated activities of old Cayuga Lake water, defined as the average of all samples with a $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio >0.50 and a $^{223}\text{Ra}/^{226}\text{Ra}$ activity ratio <0.0067 (minus three samples excluded as outliers on the basis of being >2 SD from the mean for any isotope). A total of 16 samples was used to calculate the old lake water values.

Based on the ^{226}Ra data (the long-lived isotope that showed the widest spread of values), the samples taken on 25 April have apparently undergone mixing to varying degrees with the old lake water. Using Stewart Park and the old lake water as end members, the estimated mixing percentages are calculated using the equation

$$^{226}\text{Ra}_{\text{samp}} = X(^{226}\text{Ra}_{\text{old}}) + (1 - X)(^{226}\text{Ra}_{\text{SP}}) \quad (1)$$

where the subscripts samp, old, and SP refer to sample water (mixed water), old lake water, and Stewart Park water, respectively, and X is the fractional proportion of old Cayuga Lake water making up the sample. These calculated values are shown in Table 4. This information was then used to correct the ^{223}Ra and ^{224}Ra data to the unmixed values (Table 4). The North Taughannock Point location is calculated to have been $\sim 80\%$ old lake water, indicating extensive mixing of the original water body as it moved northward. Locations farther south (South Myers Point and Myers Point) showed less, but still considerable, mixing.

Mixing has a relatively minor effect on the ^{223}Ra and ^{224}Ra activities in these samples because of the low activities of these nuclides in the old lake water. This makes the $^{224}\text{Ra}/^{223}\text{Ra}$ activity ratio attractive as a potential age calculator for water masses of this type, an example of which is shown by the ages calculated for the unmixed (corrected) samples according to the equation

$$t = \ln([^{224}\text{Ra}/^{223}\text{Ra}]_t / [^{224}\text{Ra}/^{223}\text{Ra}]_o) / (\lambda_3 - \lambda_4) \quad (2)$$

where t is the time since a mass of water was emplaced, in days; λ_3 is the decay constant for ^{223}Ra (0.0606 d^{-1}); λ_4 is the decay constant for ^{224}Ra (0.1891 d^{-1}); samp and SP refer to the activity ratios of sample and the Stewart Park end member, respectively; and $[^{224}\text{Ra}/^{223}\text{Ra}]_o$ and $[^{224}\text{Ra}/^{223}\text{Ra}]_t$ represent the activity ratios of the Stewart Park sample and the ratio of the sample at time t , respectively.

To make an age calculation, the ^{223}Ra and ^{224}Ra activities must be corrected for the amount of these nuclides that would be supplied by suspended sediment in the water. Their values are estimated from the lowest activity bulk lake water as $0.014 \text{ dpm } 100^{-1} \text{ L}$ for ^{223}Ra and $0.26 \text{ dpm } 100^{-1} \text{ L}$ for ^{224}Ra . These ages are shown in Table 4. This form of the age equation assumes only simple exponential decay of the radium isotopes in a water body acting as a closed system, whereas water was likely being added to the pod over an extended period, violating the closed system assumption. The ages calculated by Eq. 2 therefore represent minimum ages only. The fact that the calculated ages increase to the north implies a continued input of Stewart-Park-type water from the south over time.

The South Myers Point water mass did not reveal the same mixing effects as the Stewart Park water mass in terms of ^{226}Ra activity variation, meaning that there was little mixing or the end-member waters are very close to each other with respect to their radium isotopic composition, or both. Therefore, a similar analysis cannot be carried out for this water mass.

Results of this investigation show that radium isotopes can be useful tools in limnology. The isotopes ^{228}Ra and ^{226}Ra were used to infer that water from a source different from any surface water in the area was entering the lake at the southern end near Stewart Park and formed an easily identifiable feature within the lake body itself. The source is most likely a carbonate aquifer, as based on the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio. The isotopic data also show that this emplaced water body, while mixing with bulk lake water, retained enough of

its identity to track over several weeks as it moved through the lake. A more stationary second water body was also observed using the short-lived ^{223}Ra and ^{224}Ra isotopes, and the loss of these isotopes followed a radioactive decay pattern. Additionally, strong evidence of groundwater inflow into the lake around a delta was observed from ^{223}Ra and ^{224}Ra data, as was apparently minor inflow of surface water at the Stewart Park site as a result of summer storms. The small amounts of ^{223}Ra and ^{224}Ra present in old bulk lake water is attributed to the presence of suspended sediment, which supplies the water with a small but constant amount though decay of parents contained within the grain. ^{228}Ra is also supplied to the lake to compensate for its decay in the water. The source of this ^{228}Ra is diffusion from bottom sediment where it is generated by α -particle recoil from the solid grains.

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