

## Quantifying saline groundwater flow into a freshwater lake using the Ra isotope quartet: A case study from the Sea of Galilee (Lake Kinneret), Israel

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### *Abstract*

We investigated the Ra isotope quartet in order to quantify the discharge of saline groundwater into a freshwater lake. The activities of <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>223</sup>Ra, and <sup>224</sup>Ra were measured in the Sea of Galilee, Israel, surrounding saline springs, and the Jordan River, coupled with U, Th, and <sup>226</sup>Ra measurements in sediment cores and laboratory Ra adsorption experiments under different salinity conditions. The <sup>226</sup>Ra activity (0.007–0.008 Bq L<sup>-1</sup>) and <sup>226</sup>Ra : Cl ratio in the lake were significantly lower than those of the inflowing saline springs, indicating that 75–86% of the incoming <sup>226</sup>Ra is removed from the lake water. This “missing” Ra is likely adsorbed onto suspended particles. Given the observed differential depletion of Ra isotopes, we present a model consisting of adsorption-desorption, recoil, and decay of the short-lived <sup>224</sup>Ra and <sup>223</sup>Ra isotopes. We predict a removal time of ~2 weeks for Ra, inferring the apparent residence time of suspended matter in the lake. A mass-balance calculation of the different <sup>226</sup>Ra inventories reveals a saline groundwater flux of 44–61 × 10<sup>9</sup> L yr<sup>-1</sup> for “Fuliya-type” water. The residence time of dissolved <sup>226</sup>Ra in the lake is estimated as 3–4 yr. The <sup>228</sup>Ra : <sup>226</sup>Ra ratio of the lake water and a mass-balance calculation show that <sup>228</sup>Ra arrives from regeneration through bottom sediments, in addition to groundwater flux. Mass-balance calculations of the expected saline fluxes before the diversion of saline inflows to the lake (early 1960s) provide <sup>226</sup>Ra activity estimates in the lake (0.018–0.020 Bq L<sup>-1</sup>) that are similar to the value (0.018 ± 0.0001 Bq L<sup>-1</sup>) measured in the lake in 1962.

Over the last three decades, radium isotopes have been used to trace and quantify groundwater discharges into oceans (Knauss et al. 1978; Burnett et al. 2006; Paytan et al. 2006) and estuarine environments (Elsinger and Moore 1983; Charette et al. 2003; Burnett et al. 2008). All of these studies focused on the interface between fresh groundwater and saline surface or subsurface water. Ra, a decay product within the U-Th decay chains, is an alkaline earth element with four naturally occurring isotopes, the half-lives of

which range from 3.6 d to 1600 yr (<sup>224</sup>Ra = 3.66 d; <sup>223</sup>Ra = 11.43 d; <sup>228</sup>Ra = 5.76 yr; <sup>226</sup>Ra = 1600 yr). Given the geochemical similarity to barium, its predecessor in the alkaline earth family of elements, Ra tends to co-precipitate with barium minerals, most notably with barium sulfate (Molinari and Snodgrass 1990; Paytan et al. 1996). Since the affinity of Ra for adsorption decreases with increasing salinity (Krishnaswami et al. 1991; Webster et al. 1995; Sturchio et al. 2001), it has been perceived as a conservative tracer (after accounting for radioactive decay) in saline environments such as estuaries and coastal zones. In freshwater lakes, however, Ra isotopes have not been applied except for a few studies. Kraemer (2005) used the Ra isotope quartet to quantify fresh groundwater fluxes into the freshwater of Cayuga Lake, New York. In that study, the <sup>224</sup>Ra : <sup>228</sup>Ra and <sup>223</sup>Ra : <sup>226</sup>Ra ratios were used to trace the mixing proportions of groundwater in the lake. By comparing ratios of short- to long-lived Ra isotopes, Kraemer (2005) was able to delineate the Ra sources that entered the lake and the flow rate of water masses in the lake. Moser et al. (1998) measured <sup>226</sup>Ra activities in the water column, shallow sediments, and pore water of Zürich Lake,

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Switzerland. They showed that Ra activity in the water column changes seasonally with dissolved oxygen; lower concentrations were observed during the winter when the dissolved oxygen levels were high. This was related to the interaction of Ra with redox-sensitive minerals in the sediment.

In this paper, we explore the potential of using naturally occurring Ra isotopes as a proxy for understanding water budgets and Ra behavior in freshwater lakes. In contrast to estuarine and coastal zones, where fresh groundwater discharges into saline environments, here we focus on an opposite situation, where saline groundwater flows into a freshwater lake. This study aims to explore the validity of Ra isotope geochemistry as a tool for quantifying groundwater discharge into freshwater lakes. We show that Ra in the lake water, which is primarily derived from saline groundwater discharge, is scavenged from the lake water due to the relatively low salinity of the lake ( $236 \text{ mg Cl L}^{-1}$ ). We used  $^{226}\text{Ra}$  variations to quantify the residence time of Ra in the water and to estimate the saline fluxes entering the lake.

## Methods

**Study area**—The Sea of Galilee (Lake Kinneret, Lake of Tiberias) is a freshwater lake ( $236 \text{ mg Cl L}^{-1}$ ) located in the northern part of the Jordan Rift Valley (Fig. 1). The average volume of the lake is  $4.3 \times 10^{12}$  liters over an area of  $166 \text{ km}^2$ , of which  $90 \text{ km}^2$  are pelagic (Nishri et al. 1999). The salinity of the lake is controlled by a balance between: (1) freshwater recharging the lake, mainly through the Jordan River and other surface tributaries; (2) saline springs, emerging mostly around the perimeter of the lake; (3) outflows, by pumping and through its natural outlet; and (4) evaporation.

Since the construction of the National Water Carrier (NWC) in 1964, the lake water has been extensively used to supply Israel's water demand. Water pumping from the lake through the NWC has been on order of  $420 \times 10^9 \text{ L yr}^{-1}$ , approximately one fourth of Israel's annual national consumption. During the 1930s, the Degania Dam was constructed on the southern outlet of the lake. The dam is kept closed, preventing flow to the Southern Jordan River during most years, except in unusually rainy years, when it is opened to prevent flooding of adjacent communities and fields. During the 1950s, when water-quality monitoring of the lake began, a constant increase in the  $\text{Cl}^-$  concentration of the lake was observed, peaking at  $\sim 400 \text{ mg Cl L}^{-1}$  during 1965. In an effort to reduce the salinity of the lake, a salinity diversion channel (SDC) was constructed to divert the flow of some of the saline springs that discharge into the lake. The SDC diverts three major groups of springs (Fig. 1): the Tiberias Hot Springs (THS,  $10^9 \text{ L yr}^{-1}$ ,  $\sim 18,400 \text{ mg Cl L}^{-1}$ ); the Tabgha Springs ( $15 \times 10^9 \text{ L yr}^{-1}$ ,  $\sim 2200 \text{ mg Cl L}^{-1}$ ); and the Fuliya Springs ( $0.75 \times 10^9 \text{ L yr}^{-1}$ ,  $1200\text{--}17,400 \text{ mg Cl L}^{-1}$ ). Since the construction and operation of the SDC and removal of the major saline inflows to the lake, the salinity of the lake has significantly decreased. Since the 1970s,  $\text{Cl}^-$  content has varied between 220 and  $270 \text{ mg L}^{-1}$ , which reflects variations in the annual precipitation and freshwater fluxes to the lake (Nishri et al. 1999; Gvirtzman 2002).

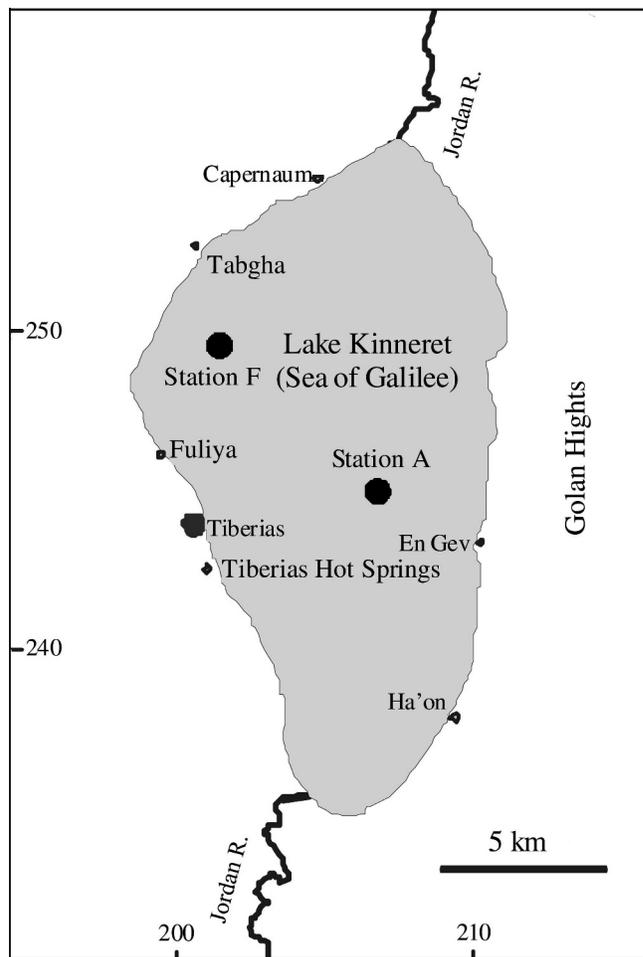


Fig. 1. A map of the Sea of Galilee, the major groups of springs surrounding the lake (Tiberias Hot Springs, Tabgha Springs, and Fuliya Springs) and the Jordan River. A and F mark sampling locations in the lake.

During the past decade, several attempts have been made to quantify the nature and extent of the unmonitored submarine saline discharge still entering the lake (Dror et al. 1999; Kolodny et al. 1999; Nishri et al. 1999). These studies have used the ratio of  $\text{Br}:\text{Cl}$  in the lake and the saline springs to distinguish between the different saline waters that flow to the lake, and they have suggested that "Fuliya-type" water containing  $\sim 1200 \text{ mg Cl L}^{-1}$  is the predominant salinity source that is still entering the lake, in spite of the SDC diversion. They estimated the flux of this unmonitored saline source to be  $61\text{--}68 \times 10^9 \text{ L yr}^{-1}$ .

Moise et al. (2000) surveyed the  $^{224}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{226}\text{Ra}$  activities in the saline springs surrounding the lake. They showed that the ratio of  $^{228}\text{Ra}:$  $^{226}\text{Ra}$  decreases with increasing  $\text{Cl}^-$  content, and that high correlation exists between  $\text{Cl}^-$  concentration and  $^{226}\text{Ra}$  activity within each group of saline springs. Nishri and Stiller (1997) calculated the residence time of radium in the lake and showed significant  $^{222}\text{Rn}$  contribution to the upper water layer due to recoil from suspended matter in the thermocline zone;

this characteristic is more strongly expressed during the summer months when the lake is stratified.

*Water sampling and analysis*—Large-volume (50–300 liters) water samples from the main water body of the Sea of Galilee and saline springs entering the lake were collected during the course of three sampling campaigns between March 2005 and June 2006. A 250-liter sample of the Upper Jordan River was collected during February 2008. Water temperature, conductivity, dissolved oxygen, pH, and redox potential were measured in the field. The large-volume samples were run through a plastic column containing Mn oxide-covered acrylic fibers (Moore 1976), which efficiently adsorb the radium isotopes. The fibers from samples SG1–18 were sent to Stanford University for immediate measurement of their short-lived  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  isotopes by delayed coincidence alpha counting (Moore and Arnold 1996). Samples SG21–29 were measured by the same method at the Laboratory of Environmental RadioNuclides (LEARN) at Duke University. The fibers were then incubated in a sealed glass cylinder for 3 weeks and measured for  $^{226}\text{Ra}$  using a Radon-in-Air monitor (RAD7, DurrIDGE Inc.), following the method of Kim et al. (2001). Samples SG1–18 were counted at Ben-Gurion University (BGU) in Israel, whereas samples SG21–29 were measured at Duke University. All the fibers were then smashed and sealed in 90-mL tin cans, and their  $^{228}\text{Ra}$  was measured using a Canberra DSA2000 BEGe gamma detector at LEARN at Duke University.

Small-volume subsamples were collected for general chemistry and trace metals analysis. Trace metals of samples SG21–29 were measured on a VG PlasmaQuad-3 inductively coupled-plasma mass spectrometer (ICP-MS) at Duke University. Trace metals and cations of samples SG1–18 were measured on a Thermo Jarrell-Ash 61E ICP-ES in the Water Commission Laboratory at the Volcani Institute at Bet-Dagan, Israel. Cations of samples SG21–29 were measured on an ARL-Fisons Spectraspan 7 Direct Current Plasma (DCP) spectrometer at Duke University. Anions of samples SG21–29 were measured on a Dionex Ion Chromatograph (IC) DX-500 at the Wetland Centre of The Nicholas School of the Environment and Earth Sciences at Duke University. Anions of samples SG1–18 were measured on a Dionex IC model 600, with a CD-20 detector, a GP-40 pump, and an AS-14 column at Ben-Gurion University. Cl and  $\text{HCO}_3^-$  concentrations of samples SG1–18 were measured on a Radiometer TIM800 Titration Manager combined with an ABU 901 Autoburette. Cl titration with  $0.015 \text{ mol L}^{-1} \text{ AgNO}_3$  was monitored by a Radiometer M25Ag silver electrode. Alkalinity titration with  $0.02 \text{ mol L}^{-1} \text{ HCl}$  was monitored by a Radiometer Phc2401-8 pH electrode. Alkalinity of samples SG21–29 was measured by titration with  $0.02 \text{ mol L}^{-1} \text{ HCl}$  using a YSI pH100 meter. The reaction error (i.e., the balance between anions and cations) was checked for all samples and found to be below 4%.

*Sediment sampling and analysis*—During the June 2006 sampling campaign, three gravimetric sediment cores were collected at Sta. A (Fig. 1). The cores (26–28 cm in length)

were then sliced into 3-cm intervals and sealed in plastic cups pending analysis. Each sediment sample was placed in a piston, and a lever was used to force the pore water out. The pore-water samples were collected in plastic vials and stored for chemical analysis. The partially dried sediments were emptied into preweighed 90-mL tin cans and placed in an oven at  $50^\circ\text{C}$  for 24 h. The dried sediments were then crushed to a diameter  $<2 \text{ mm}$  using a mortar and pestle, returned to the tin can, weighed, covered, and sealed with electric tape to prevent gas escape. The sealed cans were incubated for at least 3 weeks (to allow  $^{226}\text{Ra}$  to reach secular equilibrium with its  $^{214}\text{Bi}$  granddaughter), and each sample was counted in a Canberra DSA2000 BEGe gamma detector at LEARN in Duke University for 2–3 d to measure nuclides from the U-Th series. The  $^{226}\text{Ra}$  activities were obtained through the 609-keV energy line of its radioactive granddaughter,  $^{214}\text{Bi}$ , assuming secular equilibrium. The activities of all these nuclides were calibrated using CCRMP U-Th ore standard DL-1a and Canberra Multi-Gamma Ray standard MGS-5C, measured under physical conditions identical to the samples (e.g., can size, material type). Extracted pore waters were analyzed for anion concentrations using a Dionex IC DX-500 with CD-20 detector, a GP-40 pump, and an AS-14 column at Duke University.

*Adsorption experiment*—Dredge samples of sediments collected from the bottom of the lake at Stas. A and F (Fig. 1) were used as the solid material for Ra adsorption experiments. Pore waters from sediments of both samples were extracted using a centrifuge at 3800 rpm for 20 min. After separating the water from the solid, five samples of 2 g each were weighed from each site and inserted into a centrifuge tube. An artificial solution with chemical composition and ionic ratios similar to those of the Sea of Galilee was prepared with Cl content of  $1000 \text{ mg L}^{-1}$ . Subsamples of the initial solution were diluted to contain 750, 500, and 250 (i.e., identical to the Sea of Galilee)  $\text{mg Cl L}^{-1}$ . Thirty milliliters of each solution were added to one of the 2-g samples from each site, and 30 mL of deionized water were added to a fifth sample from each site. The initial  $^{226}\text{Ra}$  activity in the dissolved phase of each subsample ( $\sim 5.3 \text{ Bq}$ ) was obtained by adding 0.02 g of a standard (NIST 4966) containing  $263 \text{ Bq g}^{-1} \text{ }^{226}\text{Ra}$ . Additionally, two sediment-free samples were prepared containing the same amount of Ra as the other 10 subsamples, one using 30 mL deionized water and the other with 30 mL of the  $1000 \text{ mg Cl L}^{-1}$  solution, to determine if Ra was lost through adsorption to the surface of the plastic centrifuge tube. The 12 tubes were then shaken for a week in a temperature-controlled room ( $23\text{--}25^\circ\text{C}$ ). The liquid and solid phases were then separated using a centrifuge for 20 min at 3800 rpm. The pH of the water was measured, after which the samples were diluted using deionized water to a volume of  $\sim 150 \text{ mL}$ . The diluted solutions were gravity fed to columns containing  $\text{MnO}_2$  fibers, and the process was repeated seven times for each sample to assure that all the Ra from the solution was adsorbed to the fibers. The fibers were then manually squeezed and individually incubated in glass tubes for at least 3 weeks, after which their  $^{226}\text{Ra}$  activity was counted

on a Radon-in-Air monitor at LEARN at Duke University, following the method described previously.

## Results

**Water samples**—The chloride contents and Ra isotopic activities in the saline groundwater, freshwater of the Jordan River, and fresh lake water of the Sea of Galilee are summarized in Table 1. The Ra isotopic results of the saline springs reported here are consistent with the measurements reported by Moise et al. (2000) for the same saline springs. The activities of the four Ra isotopes are linearly correlated with chloride concentrations, particularly in the Fuliya Springs (Fig. 2). All samples from the Sea of Galilee were collected during the summer months when the lake is stratified (i.e., thermocline depth is  $\sim 10.5$  m, see Table 1). The average  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{223}\text{Ra}$ , and  $^{224}\text{Ra}$  activities measured along the lakes' water column, which are used in our discussions, are 0.0073, 0.00058, 0.0000068, and  $0.00012 \text{ Bq L}^{-1}$ , respectively (Table 2). These activities are several orders of magnitude lower than those measured in the saline springs entering the lake (Table 1). The  $^{228}\text{Ra}:^{226}\text{Ra}$  ratio in the lake water ranged between 0.069 and 0.108, (average of 0.079) and is similar to the ratio at the Tiberias Hot Springs (THS; 0.061–0.105; average 0.087) and Fuliya Group Springs (0.041–0.092). The ratio in the lake, however, is lower than the ratio range in Tabgha Springs (0.179–0.280; an average of 0.24) (Fig. 3). The  $^{224}\text{Ra}:^{223}\text{Ra}$  ratio in lake water (12 to 28) overlaps with the ratio in Fuliya B spring (13–19), but it is significantly higher than in Fuliya A (6.5), Tabgha (7.8), and THS (7.3) Springs (Table 1). The  $^{224}\text{Ra}:^{228}\text{Ra}$  ratio varies from  $\sim 1.5$  in the Fuliya and Tabgha Springs, to  $\sim 1$  at THS spring and 0.25 in the lake water. The Ra : Cl ratios in the lake water ( $4.93 \times 10^{-7}$ ,  $2.89 \times 10^{-8}$ ,  $2.27 \times 10^{-6}$ , and  $3.11 \times 10^{-5}$  for  $^{224}\text{Ra}$ ,  $^{223}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{226}\text{Ra}$ , respectively; Table 2) are significantly lower than those in the Fuliya, Tabgha, and THS Springs.

The  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{223}\text{Ra}$ , and  $^{224}\text{Ra}$  activities of water collected from the Jordan River are 0.00153, 0.000665, 0.0000235, and  $0.000591 \text{ Bq L}^{-1}$ , respectively.

**Sediment samples**—The  $^{226}\text{Ra}$  activities of the sediments from the two cores systematically increase with depth (Fig. 4). In contrast, the U activity ( $^{238}\text{U}$ ; measured by ICP-MS) in the sediments decreases with depth and shows no apparent correlation to the changes in the activity of  $^{226}\text{Ra}$  (Fig. 4).

**Adsorption experiment**—The fraction of Ra that was adsorbed during the adsorption experiment,  $\text{Ra}_{\text{ads}}$ , was calculated by subtracting the dissolved activity measured at the end of the experiment from the initial Ra activity, corrected to the weight of sediments used for the experiment to yield activities in units of  $\text{Bq g}^{-1}$ . The dissolved activities measured at the end of the experiment,  $\text{Ra}_{\text{dis}}$ , were divided by the volume of the solution to yield activities in units of  $\text{Bq mL}^{-1}$ . The experiments show that the ratio of  $\text{Ra}_{\text{ads}}:\text{Ra}_{\text{dis}}$  decreases with chloride concentration (Fig. 5), clearly showing that the affinity of Ra to adsorption decreases with increasing salinity.

## Discussion

**Geochemical indicators for radium sources and sinks in the Sea of Galilee**—The Ra isotopic composition of the Sea of Galilee is controlled by (1) the Ra isotopic ratios of the saline inflows, which are considered the major sources for dissolved constituents in the lake (Kolodny et al. 1999; Nishri et al. 1999); (2) dissolved and particulate Ra entering through the Jordan River; (3) possible recoil from particulate matter in the water column (Kraemer 2005); (4) removal through adsorption and/or co-precipitation into barite minerals; (5) diffusion from bottom sediments (Key et al. 1985; Krest et al. 1999; Kraemer 2005); and (6) radioactive decay. Given the residence time of water in the Sea of Galilee ( $\sim 6$  yr), the radioactive decay applies only to the relatively short-lived nuclides of  $^{228}\text{Ra}$ ,  $^{224}\text{Ra}$ , and  $^{223}\text{Ra}$ , but not to  $^{226}\text{Ra}$ .

The Ra activities and their ratios in the different saline springs are presented in Fig. 2 and in Table 1. The data show that  $^{226}\text{Ra}$  in the lake water is depleted relative to  $^{226}\text{Ra}$  activities in the saline springs when extrapolated to the Cl content of the lake water (Fig. 2). We therefore suggest that in addition to dilution, which would equally affect Ra and Cl, Ra is being substantially removed from the liquid phase. Given that (1) the Sea of Galilee is undersaturated with respect to the mineral barite (Saturation Index [SI] =  $-0.2$ , calculated by PHREEQC software); (2) there is no evidence for the occurrence of barite in the upper sediment layer (Halitz and Nishri unpubl.); and (3) the residence time of water in the lake is  $\sim 6$  yr, we suggest that  $^{226}\text{Ra}$  is removed from the lake water through adsorption onto suspended particulates and sedimentation. Our adsorption experiment (Fig. 5) confirms that under the salinity conditions of the Sea of Galilee (Cl  $\sim 250 \text{ mg L}^{-1}$ ), a large fraction of Ra is removed by adsorption.

Nishri et al. (1999) and Kolodny et al. (1999) suggested that “Fuliya-type” water with an estimated salinity of  $\sim 1200 \text{ mg Cl L}^{-1}$  is the predominant unmonitored saline submarine input that has continued to flow into the lake after the SDC diversion. In spite of that, in the following discussion, we consider all known major saline end members (Fuliya, Tabgha, and THS), as well as the Jordan River, as potential salinity (and Ra) contributors to the lake. For each of the saline end members, the expected  $^{226}\text{Ra}_{\text{initial}}$  value in the specific source (if dilution was the only process that controls Ra activity in the lake) was determined by multiplying the ratio of each radium isotope to  $\text{Cl}^{-}$  by the salinity of the lake (e.g.,  $0.0292 \text{ Bq L}^{-1}$  for Fuliya; Fig. 2; Table 2). The difference between the measured  $^{226}\text{Ra}$  in the lake ( $0.00735 \text{ Bq L}^{-1}$ ) and the expected  $\text{Ra}_{\text{initial}}$  suggests that the lake water is depleted by 75% to 86% in  $^{226}\text{Ra}$  compared to that expected from simple dilution of Fuliya and THS water, respectively (Table 2; Fig. 2). Likewise, we show even larger depletions in  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$  activities in the lake water relative to the expected Ra for diluted saline groundwater (95–99% for Fuliya and Tabgha water, respectively; Fig. 2C,D). The depletion in  $^{228}\text{Ra}$  activity in the lake (Fig. 2B) is between 57% (Fuliya-type water) and 94% (Tabgha Springs). These depletions indicate a significant sink for Ra in the lake,

Table 1. Radium and chloride in the saline springs, the Jordan River, and the Sea of Galilee. Data are from this study combined with results previously reported by Moise et al. (2000).

	This study	Moise et al. (2000)	Collection	$^{226}\text{Ra}$ (Bq L <sup>-1</sup> )	$^{228}\text{Ra}$ (Bq L <sup>-1</sup> )	$^{223}\text{Ra}$ (Bq L <sup>-1</sup> )	$^{224}\text{Ra}$ (Bq L <sup>-1</sup> )	Cl (mg L <sup>-1</sup> )	$^{224}\text{Ra}$ : $^{223}\text{Ra}$	$^{228}\text{Ra}$ : $^{226}\text{Ra}$	$^{224}\text{Ra}$ : $^{228}\text{Ra}$	$^{223}\text{Ra}$ : $^{226}\text{Ra}$
Tiberias Hot Springs												
Hamei Tveria Main Spring	SG 2	TR 105	13 Mar 05	4.514	0.333	0.040	0.291	18,526.7	7.213	0.065		0.0089
Hamei Tveria Main Spring		TR 140	16 Nov 94	5.095	0.233			18,625.0		0.068		
Hamei Tveria Main Spring		TR 169	02 Feb 95	3.636	0.265		0.351	18,373.0		0.073	1.327	
Hamei Tveria Main Spring		TR 190	22 Feb 95	3.961	0.241			18,231.0		0.061		
Hamei Tveria Roman Spring	SG 3	TR 104	13 Mar 05	3.735	0.266	0.033	0.246	18,475.0	7.429	0.067		0.0089
Hamei Tveria Roman Spring		TR 141	16 Nov 94	3.996	0.583			18,822.0		0.105		
Hamei Tveria Roman Spring		TR 202	30 Nov 94	5.561	0.303		0.315	18,444.0		0.071	1.038	
Hamei Tveria Roman Spring		TR 255	16 Mar 95	4.297	0.443		0.403	18,475.0		0.097	0.910	
Hamei Tveria Roman Spring		TR 275	18 Jun 95	4.545	0.366		0.340	18,634.0		0.088	0.927	
Hamei Tveria Roman Spring		TR 286	11 Mar 96	4.164	0.291		0.273			0.083	0.937	
Hamei Tveria Roman Spring		TR 293	10 Jun 96	3.506	0.368		0.298			0.087	0.810	
Hamei Tveria Roman Spring		TR 293	11 Jul 96	4.229	0.368		0.298			0.087	0.810	
Tabgha												
Sartan-Eiver	SG 5	TR 99	13 Mar 05	0.507	0.072	0.011	0.091	2383.5	8.233	0.179		0.0217
Sartan-Eiver		TR 134	16 Nov 94	0.400	0.138			2889.0		0.218		
Sartan-Eiver		TR 259	30 Nov 94	0.633	0.128		0.173	2765.0		0.280	1.351	
Sartan sweet	SG 6	TR 259	18 Jun 95	0.458	0.073	0.006	0.045	3030.0	7.305	0.280		0.0163
Sartan sweet		TR 235	13 Mar 05	0.380	0.073		0.117	1528.5		0.256	1.591	
Sartan sweet		TR 235	30 Apr 95	0.286	0.073		0.117	1773.0				
Fuliya												
Fuliya B Spring	SG 7	TR 207	20 Apr 05	0.166	0.008	0.001	0.014	1432.0	13.525	0.076		0.0061
Fuliya B Spring		TR 102	16 Nov 94	0.837	0.115		0.010	8622.0		0.137		0.0046
Fuliya B Spring		TR 137	23 Jun 05	0.157	0.006	0.001	0.010	1230.0	13.509	0.161		0.0045
Fuliya B Spring		TR 181	06 Jun 06	0.154	0.008	0.001	0.013	8724.0	18.985	0.117	2.078	0.0103
Fuliya A Spring		TR 201	23 Jun 05	0.195	0.014	0.002	0.013	1532.0	6.479	0.089		0.0183
Fuliya A Spring		TR 220	06 Jun 06	0.275	0.025	0.005	0.032	2174.6	6.442	0.076	1.320	
Fuliya A Spring		TR 207	16 Mar 95	0.118	0.009		0.017	1215.0		0.137	1.852	
Kinneret 5 (pump)		TR 102	16 Nov 94	0.837	0.115		0.010	8622.0		0.137		
Kinneret 5 (pump)		TR 137	30 Nov 94	0.954	0.153		0.010	8724.0		0.161		
Kinneret 5 (artesian)		TR 181	22 Feb 95	0.500	0.058		0.010	4700.0		0.117		
Kinneret 5 (artesian)		TR 201	16 Mar 95	0.418	0.055		0.098	4616.0		0.131	1.788	
Kinneret 5 (artesian)		TR 220	09 Apr 95	0.466	0.057		0.092	4824.0		0.121	1.618	
Kinneret 5 (artesian)		TR 261	06 Jul 95	1.382	0.196		0.306	13,494.0		0.142	1.559	
Kinneret 10 (110–453 m)		TR 267	31 Aug 95	1.748	0.193		0.261	17,131.0		0.110	1.353	
Kinneret 10 (462–639 m)		TR 271	18 Sep 95	1.815	0.461		0.658	17,412.0		0.254	1.426	
Kinneret 10 (600–882 m)		TR 328	23 Dec 96	1.832	0.203		0.220			0.111	1.082	
Kinneret 10 (600–882 m)		TR 328	23 Dec 96	1.832	0.203		0.220			0.111	1.082	
Lake water												
Sta. A 5.5 m	SG 13	TR 328	21 Jun 05	0.00613	0.000463	$6.27 \times 10^{-6}$	0.000125	231.6	19.888	0.043	0.289	0.0010
Sta. A 5.5 m	SG 25	TR 328	07 Jun 06	0.00829	0.000572	$8.26 \times 10^{-6}$	0.000102	237.5	12.317	0.043	0.289	0.0010
Sta. A 10.5 m	SG 28	TR 328	08 Jun 06	0.00737	0.000502	$9.71 \times 10^{-6}$	0.000128	237.5	13.180	0.077	0.226	0.0013
Sta. A 30.5 m	SG 14	TR 328	21 Jun 05	0.00602	0.000652	$4.29 \times 10^{-6}$	0.000072	233.9	16.660	0.077	0.226	0.0007
Sta. A 35.5 m	SG 26	TR 328	07 Jun 06	0.00889	0.000698	$5.51 \times 10^{-6}$	0.000156	239.7	28.236	0.079	0.223	0.0006
Jordan River												
Jordan River (Arik Bridge)	SG 29	TR 328	17 Feb 08	0.00153	0.000665	$5.35 \times 10^{-5}$	0.000591	52	25.163	0.434	0.889	0.0153

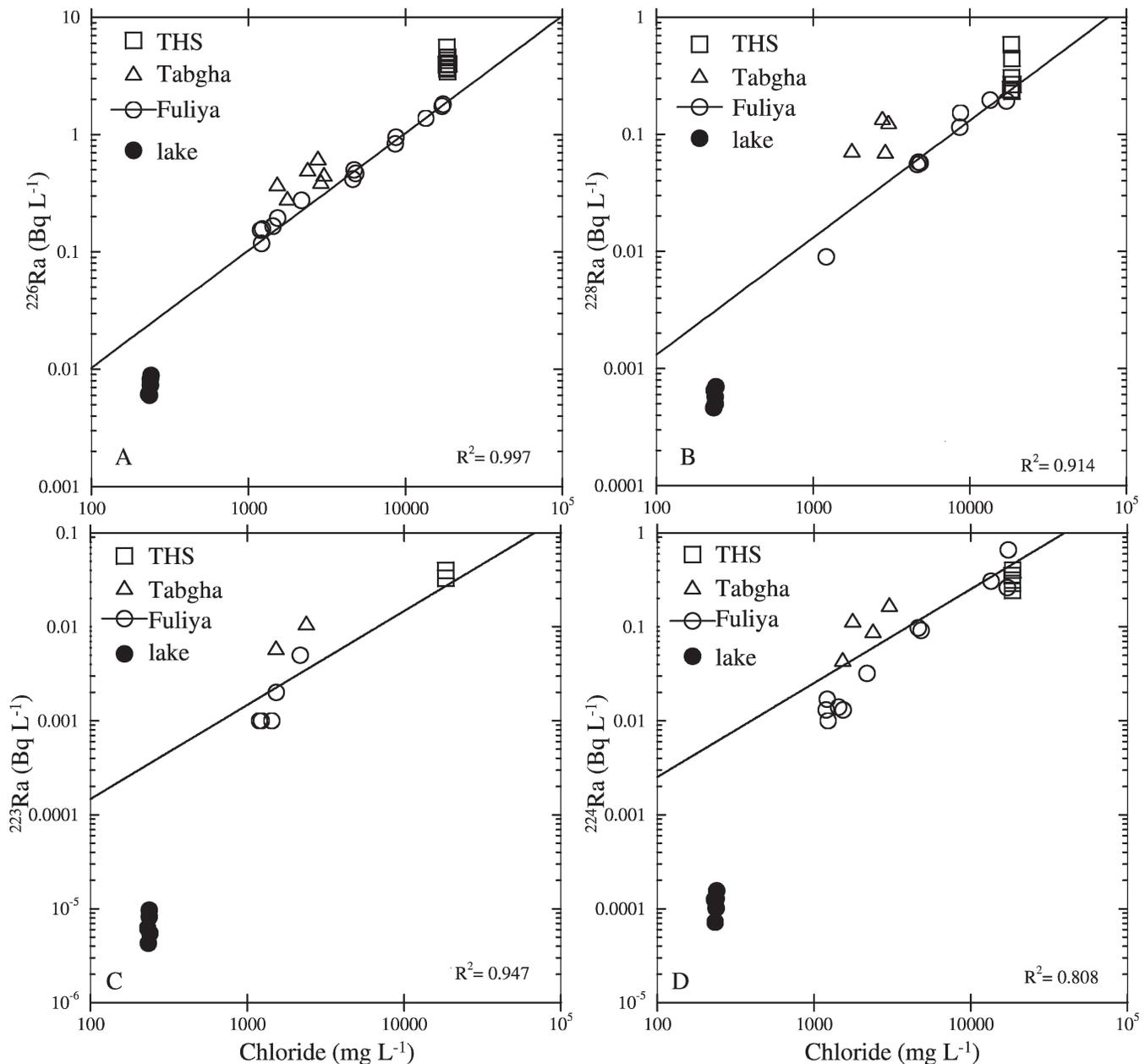


Fig. 2. Activities of the four Ra isotopes versus chloride concentrations (logarithmic scale) in the saline springs and Sea of Galilee. Radium activities in the lake are significantly lower than the Ra activities in the saline springs extrapolated for the lake salinity.

which changes the isotope ratios from those seen in the saline springs, and makes the activity of Ra in the lake significantly lower than expected from dilution of the saline springs. While several processes might contribute to the scavenging of Ra through the sediments, in the following discussion, we assume the major scavenging mechanism is via adsorption.

A possible additional Ra source to the lake (in addition to saline springs) is recoil from suspended particles in the water column (Kraemer 2005). Given the short residence time of particulate matter in the lake water column (a few weeks; Stiller and Imboden 1984), we suggest that recoil could contribute only short-lived Ra nuclides with similar

half-lives ( $^{224}\text{Ra}$ ,  $^{223}\text{Ra}$ ), while  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$  are derived predominantly from the saline springs that flow to the lake. Assuming that (1) the upper 3 cm of the sediment cores represent the particulate matter in the lake, with U and Th activities of  $0.046$  and  $0.013 \text{ Bq g}^{-1}$ , respectively (Fig. 4); (2) the small size of particulate matter results in 100% recoil efficiency to the lake water; (3) nuclides in the U and Th decay series are in secular equilibrium within the particulate matter (i.e.,  $^{224}\text{Ra} = ^{228}\text{Ra} = ^{232}\text{Th}$  and  $^{238}\text{U} = ^{226}\text{Ra}$ ); and (4) the concentration of particulate matter in the water column of the lake is  $2.1 \text{ mg L}^{-1}$  (Stiller and Imboden 1984), we can calculate the expected  $^{222}\text{Rn}$  ( $9.45 \times 10^{-5} \text{ Bq L}^{-1}$ ),  $^{224}\text{Ra}$  ( $2.73 \times 10^{-5} \text{ Bq L}^{-1}$ ), and  $^{223}\text{Ra}$  ( $7.0 \times$

Table 2. Average measured activities of Ra isotopes in different saline springs, Jordan River, and the Sea of Galilee. Expected activities refer to dilution factor, using the Ra : Cl ratios.

Water type	$^{224}\text{Ra}$ (Bq L <sup>-1</sup> )	$^{223}\text{Ra}$ (Bq L <sup>-1</sup> )	$^{228}\text{Ra}$ (Bq L <sup>-1</sup> )	$^{226}\text{Ra}$ (Bq L <sup>-1</sup> )	Cl (mg L <sup>-1</sup> )	$^{224}\text{Ra}$ : Cl × 10 <sup>-5</sup>	$^{223}\text{Ra}$ : Cl × 10 <sup>-6</sup>	$^{228}\text{Ra}$ : Cl × 10 <sup>-5</sup>	$^{226}\text{Ra}$ : Cl × 10 <sup>-4</sup>
<b>Tiberias Hot Springs</b>									
measured activity*	0.315	0.0368	0.336	4.211		1.738	1.987	1.803	2.265
expected activity†	0.00410	0.000469	0.00426	0.0535					
% depletion‡	97%	99%	86%	86%					
<b>Tabgha</b>									
measured activity*	0.107	0.00862	0.103	0.444		4.767	4.340	3.963	1.903
expected activity†	0.0112	0.00102	0.00935	0.0449					
% depletion‡	99%	99%	94%	84%					
<b>Fuliya</b>									
measured activity*	0.145	0.00190	0.128	0.735		0.950	0.630	0.566	1.238
expected activity†	0.00224	0.000149	0.00134	0.0292					
% depletion‡	95%	95%	57%	75%					
<b>Jordan River</b>									
measured activity*	0.00059	0.0000235	0.000665	0.00153		1.137	0.452	1.280	0.295
expected activity†	0.00268	0.000107	0.00302	0.00695					
% depletion‡	96%	94%	81%						
<b>Lake</b>									
measured activity	0.00012	0.00000682	0.000578	0.00735	236.04	0.049	0.029	0.228	0.312
$T_{1/2}$ (d)	3.66	11.43	2102.40	584,000					
$\lambda$ § (d <sup>-1</sup> )	0.189	0.061	0.00033	0.0000012					

Spring values were compiled from data obtained during this study and by Moise et al. (2000).

Lake data were compiled from samples collected at Sta. A during the course of this study.

\* Average Ra : Cl ratio is based on the data displayed in Table 1.

† Expected activity = (Ra : Cl)<sub>water type</sub> × [Cl]<sub>lakes</sub>, for each isotope and each water type (THS, Tabgha, Fuliya, Jordan River).

‡ % depletion = 100 × (1 - measured activity/expected activity), for each isotope and each water type (THS, Tabgha, Fuliya, Jordan River).

§  $\lambda = \ln(2)/T_{1/2}$ , for each isotope.

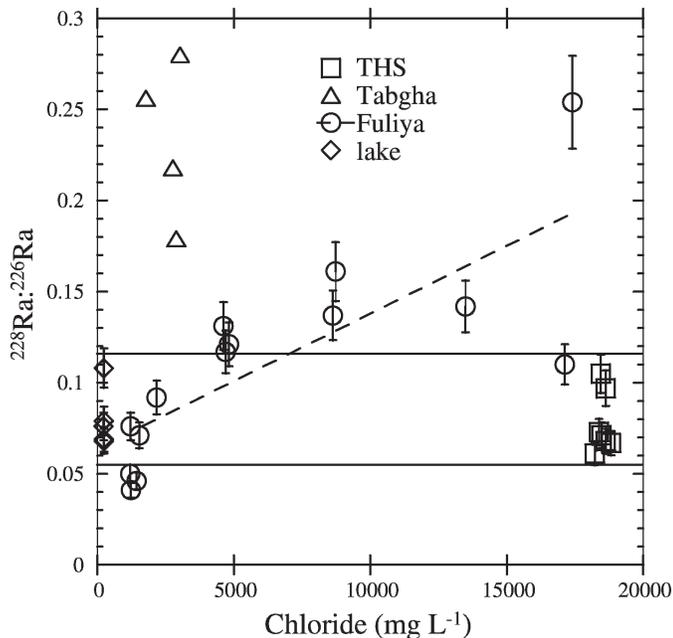


Fig. 3.  $^{228}\text{Ra}:^{226}\text{Ra}$  ratios versus chloride concentrations in the saline springs and Sea of Galilee. The  $^{228}\text{Ra}:^{226}\text{Ra}$  ratio in the lake is higher than that of “Fuliya-type” water, suggesting the latter cannot be the only source of Ra entering the lake.

$10^{-7} \text{ Bq L}^{-1}$ ) activities and  $^{224}\text{Ra}:^{223}\text{Ra}$  ratio (39) in the lake water derived entirely from recoil processes from the nuclides in suspended matter. Given that the adsorption rate of Ra is extremely fast (i.e., minutes; Krishnaswami et al. 1982), one would expect that Ra adsorption onto particulate matter would change the Ra activities in the suspended matter and thus their recoil products. For example, Nishri and Stiller (1997) reported significantly

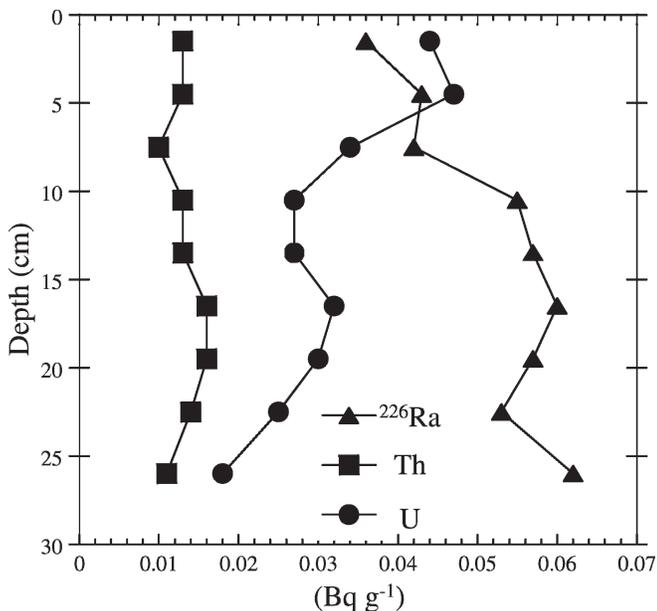


Fig. 4.  $^{226}\text{Ra}$ ,  $^{238}\text{U}$ , and  $^{232}\text{Th}$  activity variations in sediments along core 2 collected from Sta. A. The data show no apparent correlation between the activities of  $^{226}\text{Ra}$  and  $^{238}\text{U}$ .

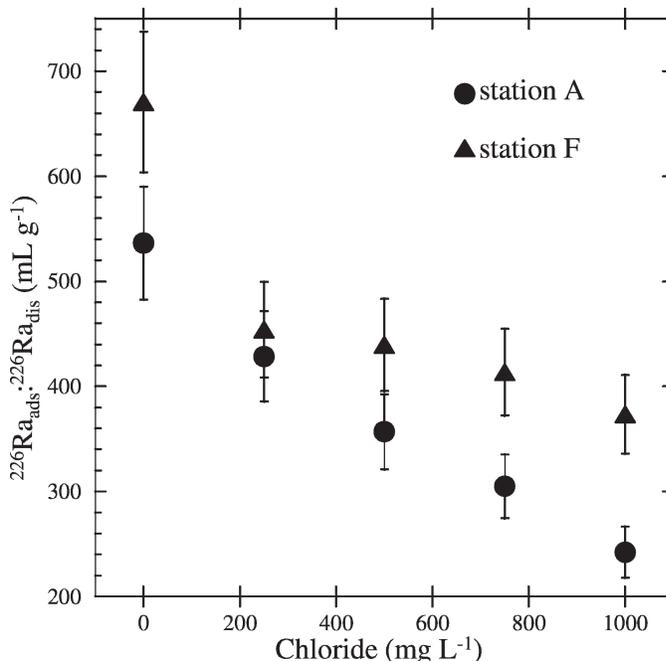


Fig. 5. Variations of the ratio  $\text{Ra}_{\text{ads}}:\text{Ra}_{\text{dis}}$  with chloride concentrations as measured in adsorption experiments using samples collected at Stas. A and F (Fig. 1). The data suggest that Ra adsorption decreases with salinity.

higher  $^{222}\text{Rn}$  levels in the water column of the Sea of Galilee (an average of  $0.0343 \pm 0.0208 \text{ Bq L}^{-1}$ ), which could reflect higher activities of  $^{226}\text{Ra}$  in suspended matter derived from adsorption from the lake water. Likewise, the measured  $^{224}\text{Ra}$  (an average of  $1.2 \times 10^{-4} \text{ Bq L}^{-1}$ ) and  $^{223}\text{Ra}$  ( $6.8 \times 10^{-6} \text{ Bq L}^{-1}$ ) activities in the lake water are higher (4.3- and 9.7-fold, respectively) than the expected activities derived from recoil. Moreover, the rapid Ra adsorption relative to the decay rate of even the short-lived Ra isotopes would result in high ratios of the short-lived to long-lived isotopes (e.g.,  $^{224}\text{Ra}:^{228}\text{Ra} > 1$ ,  $^{223}\text{Ra}:^{226}\text{Ra} > 0.046$ ) upon recoil combined with adsorption (Krishnaswami et al. 1982), when, in fact, the measured  $^{224}\text{Ra}:^{228}\text{Ra}$  and  $^{223}\text{Ra}:^{226}\text{Ra}$  ratios in the lake water are much lower, 0.25 and 0.0009, respectively. In a later section, we will quantify the different Ra sources and sinks that affect the Ra isotopic ratios in the Sea of Galilee.

The ratio of  $^{228}\text{Ra}:^{226}\text{Ra}$  in the lake water (an average ratio 0.079) is slightly higher than the ratio of Fuliya-type water containing  $1200 \text{ mg Cl L}^{-1}$  (0.065). Given the residence time of water in the lake ( $\sim 6 \text{ yr}$ ) and the expected decay of  $^{228}\text{Ra}$ , one would expect the ratio of  $^{228}\text{Ra}:^{226}\text{Ra}$  in the lake to be lower than the ratio measured in the saline groundwater source. The relatively higher  $^{228}\text{Ra}:^{226}\text{Ra}$  ratio of the lake water is therefore an indication of an additional source with a higher  $^{228}\text{Ra}:^{226}\text{Ra}$  ratio. Aside from Fuliya-type water, other potential water sources to the lake are the Tabgha Springs ( $^{228}\text{Ra}:^{226}\text{Ra} = 0.237$ ), THS (0.087), and the Jordan River (0.434; Table 1). Contribution from any of these sources might increase the  $^{228}\text{Ra}:^{226}\text{Ra}$  ratio in the lake above the ratio anticipated from discharge of Fuliya-type water alone.

Alternatively, the elevated  $^{228}\text{Ra} : ^{226}\text{Ra}$  ratio might be the result of regeneration of  $^{228}\text{Ra}$  from the bottom sediments (Key et al. 1985; Krest et al. 1999; Kraemer 2005), where the  $^{228}\text{Ra} : ^{226}\text{Ra}$  ratio (0.61 at the upper 3 cm) is significantly higher than the ratio measured in both the lake and in Fuliya-type water.

*Modeling adsorption and decay of radium isotopes*—The long-lived  $^{226}\text{Ra}$  isotope abundance in the lake is controlled predominantly by adsorption processes. In contrast, the relative depletion of the other Ra isotopes is controlled by recoil, radioactive decay, and adsorption. In order to quantify these effects, we consider four possible scenarios to explain the isotopic variations of the four Ra isotopes. In the different models described here, we use the average  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{223}\text{Ra}$ , and  $^{224}\text{Ra}$  activities in the lake water and assume that Fuliya springs are the predominant Ra source entering the lake (Kolodny et al. 1999; Nishri et al. 1999). First, we consider that all Ra isotopes are removed solely by adsorption (model A). This approach results in 75–86% depletion of  $^{226}\text{Ra}$ , 57–94% depletion of  $^{228}\text{Ra}$  (measuring error up to 25%), and 95–99% for both  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  (measuring error is less than 10%) relative to the expected Fuliya input. In our second approach, we consider that all short-lived  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$  isotopes are removed only through radioactive decay (model B). We use the basic radioactive decay equations

$$D = D_0 e^{-\lambda t} \quad (1)$$

and

$$t = -\frac{1}{\lambda} \ln \frac{D}{D_0} \quad (2)$$

where  $t$  is the decay time,  $\lambda$  is the decay constant for the specific Ra isotope,  $D_0$  is the initial (calculated) activity at time  $t_0$ , and  $D$  is the measured activity after time  $t$ . We define  $D_0$  values as the initial activities in the lake had dilution been the only process affecting the activity of radium in the lake (normalized to Cl content in the lake; see Fig. 2B–D; “expected activity” in Table 2) and  $D$  as the measured activities. The calculated decay times using this model are 15.6–24.1 and 50.8–82.7 d for  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$ , respectively. The third approach (model C) combines adsorption and radioactive decay. We consider a model where all isotopes are adsorbed to the same extent, while the remaining Ra differences are the result of removal through radioactive decay. In this model,  $D_0$  becomes the percentage of each radium isotope that remains in solution after subtracting the adsorbed portion, whereas  $D$  remains the activity measured in the lake water. Assuming that  $^{226}\text{Ra}$  is removed only by adsorption, model C uses 75–86% as the fraction of each Ra isotope that is removed by adsorption, converting Eq. 2 to the following form,

$$\begin{aligned} t &= -\frac{1}{\lambda} \ln \frac{D}{D_0} = -\frac{1}{\lambda} \ln \frac{\text{Ra}_{\text{measured}}}{\text{Ra}_{\text{expected}} - \text{Ra}_{\text{adsorbed}}} \\ &= -\frac{1}{\lambda} \ln \frac{\text{Ra}_{\text{meas}}}{0.25\text{Ra}_{\text{exp}}} \text{ or } -\frac{1}{\lambda} \ln \frac{\text{Ra}_{\text{meas}}}{0.14\text{Ra}_{\text{exp}}} \end{aligned} \quad (3)$$

This model results in decay times of 8.3–13.8 and 28.0–50.2 d for  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$ , respectively.

The fourth model considers adsorption of Ra isotopes that is proportional to the recoil production: activity ratio of the different Ra nuclides, which depends on their decay constants. This model (model D) follows the approach of Krishnaswami et al. (1982), which showed that under exchange conditions (e.g., adsorption-desorption reactions) between dissolved Ra and Ra adsorbed onto surface sediments, the retardation factor ( $R_f$ ) is equal to the ratio of production (by recoil) to Ra activity. In their model, Krishnaswami et al. (1982) showed that shorter-lived isotopes experience less retardation than longer-lived ones. This is derived from the higher supply rate of the shorter-lived Ra isotopes through recoil. Kraemer (2005) suggested that recoil of the short-lived Ra nuclides can occur from suspended matter in the water column of the lake. The retardation of Ra isotopes ( $R_f$ ) can be described as

$$R_f = \frac{k_1 + k_2 + \lambda}{k_2 + \lambda} \quad (4)$$

where  $\lambda$  is the decay constant ( $\text{min}^{-1}$ ), and  $k_1$  and  $k_2$  are first-order adsorption and desorption rate constants ( $\text{min}^{-1}$ ), respectively. This model assumes exchange (adsorption-desorption) reactions between suspended matter in the lake water and sufficient time for recoil of the short-lived Ra nuclides to occur from the suspended matter back to the solution. Assuming desorption rate is significantly lower than adsorption in freshwater, Eq. 4 can be approximated as:

$$R_f \approx \frac{k_1 + \lambda}{k_2 + \lambda} \quad (5)$$

By extrapolating the results of Krishnaswami et al. (1982) to the salinity conditions of the Sea of Galilee, we find values of  $10^{-2}$  and  $10^{-6} \text{ min}^{-1}$  for the  $k_1$  and  $k_2$  parameters, respectively. Since the  $\lambda$  values for the short-lived Ra isotopes are significantly smaller than  $k_1$  (for  $^{224}\text{Ra}$ ,  $^{223}\text{Ra}$ , and  $^{228}\text{Ra}$ , the  $\lambda$  values are  $1.3 \times 10^{-4}$ ,  $4.2 \times 10^{-5}$ , and  $2.3 \times 10^{-7} \text{ min}^{-1}$ , respectively), Eq. 5 can be rewritten as

$$R_f \approx \frac{k_1}{k_2 + \lambda} \quad (6)$$

We have used these assumptions for setting the  $R_f$  ratios between the different Ra isotopes. Using the  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  differences to develop the model, the ratio of the retardation factors  $R_{f,223}$  to  $R_{f,224}$  (defined as  $\mathfrak{R}$ ) is

$$\mathfrak{R} = \frac{R_{f,223}}{R_{f,224}} = \frac{\frac{k_1}{k_2 + \lambda_{223}}}{\frac{k_1}{k_2 + \lambda_{224}}} = \frac{k_2 + \lambda_{224}}{k_2 + \lambda_{223}} \quad (7)$$

Next, we define  $X$  as the ratio between the adsorbed Ra and the expected Ra activity from the saline source that enters the lake (corrected for dilution):

$$\frac{^{224}\text{Ra}_{\text{ads}}}{^{224}\text{Ra}_{\text{exp}}} = X \quad (8)$$

Hence, the decay of Ra isotopes from the initial saline Ra, corrected for Ra adsorption is

$$\begin{aligned} {}^{224}\text{Ra}_{\text{meas}} &= [{}^{224}\text{Ra}_{\text{exp}} - {}^{224}\text{Ra}_{\text{ads}}] \cdot e^{-\lambda_{224}t} \\ &= (1 - X)^{224}\text{Ra}_{\text{exp}} \cdot e^{-\lambda_{224}t} \end{aligned} \quad (9)$$

$${}^{223}\text{Ra}_{\text{ads}} = \mathfrak{R} \cdot X \cdot {}^{223}\text{Ra}_{\text{exp}} \quad (10)$$

Combining Eqs. 8–10 for all Ra isotopes and solving for  $t$  yields

$$t = \frac{1}{\lambda_i} \left[ \ln(1 - \mathfrak{R}X) + \ln\left(\frac{i\text{Ra}_{\text{exp}}}{i\text{Ra}_{\text{meas}}}\right) \right] \quad (11)$$

where  $i$  signifies the specific Ra isotope ( ${}^{224}\text{Ra}$ ,  ${}^{223}\text{Ra}$ , or  ${}^{228}\text{Ra}$ ). Application of Eq. 11 to the  ${}^{224}\text{Ra}$ - ${}^{223}\text{Ra}$  and  ${}^{224}\text{Ra}$ - ${}^{228}\text{Ra}$  isotope pairs yields decay time of 10.7–13.8 d (12.3 d on average) when using Fuliya-type water as the saline end member, and 13.4–15.8 d (14.6 d on average) when using THS-type water as the saline end member. In spite of the fact this model is not based on actual measurement of adsorbed Ra: expected Ra, the internal consistency of the saline end members in model D indicates that this model, which includes Ra adsorption-desorption exchange and preferential recoil ingrowths of Ra isotopes based on their decay constant, can provide consistent decay times for all short-lived Ra isotopes.

Following Simpson et al. (1982), this calculated range of decay times is defined as the “permanent burial time” ( $\tau$ ) of Ra isotopes in the Sea of Galilee. This “permanent burial time” in fact refers to an average “residence time” of suspended matter in the lake water that consists of the major sink for Ra nuclides. This estimation of 12.3–14.6 days is similar to, though slightly lower than, the residence time of suspended matter in the lake calculated by Stiller and Imboden (1984) of 53 d.

*${}^{226}\text{Ra}$  mass balance in the Sea of Galilee*—Assuming that the principal Ra source that enters the lake is saline seepage through springs, we use the conservation of mass equation to provide information on the flux of saline water entering the lake,

$$\frac{d[{}^{226}\text{Ra}]_{\text{SG}}}{dt} = 0 = {}^{226}\text{Ra}_{\text{IN}} - {}^{226}\text{Ra}_{\text{OUT}} \quad (12)$$

where  $d[{}^{226}\text{Ra}]_{\text{SG}}$  is the net change in radium activity in the lake over time. Assuming the lake is well mixed and is in steady state, the incoming Ra flux is expected to be equal to the outgoing flux. The  ${}^{226}\text{Ra}$  sinks in the lake are described in Eq. 13 and consist of  ${}^{226}\text{Ra}$  exiting the lake through the NWC and through sedimentation of particles within the lake:

$${}^{226}\text{Ra}_{\text{OUT}} = {}^{226}\text{Ra}_{\text{NWC}} + {}^{226}\text{Ra}_{\text{SED}} \quad (13)$$

where  $[{}^{226}\text{Ra}]_{\text{SED}}$  is the bulk Ra activity measured in sediments and represents the sum of “supported” (i.e., Ra in secular equilibrium with U) and “adsorbed” (i.e., scavenged from the lake water) Ra found in suspended matter in the lake. Assuming that sedimentation occurs in the pelagic area where Ra is being permanently removed from the

lake water,

$$\begin{aligned} {}^{226}\text{Ra}_{\text{SED}} [\text{Bq/yr}] &= [{}^{226}\text{Ra}]_{\text{SED}} [\text{Bq/g}] \times (\text{rate}) [\text{g/m}^2 \cdot \text{yr}] \\ &\quad \times SA [\text{m}^2] \end{aligned} \quad (14)$$

Stiller and Imboden (1984) measured  $0.052 \text{ Bq g}^{-1}$  of  ${}^{226}\text{Ra}$  in suspended matter from the lake. We did not collect a sample of suspended matter from the lake in this study, but we measured an average of  $0.037 \text{ Bq g}^{-1}$  of  ${}^{226}\text{Ra}$  in the most shallow samples from cores 2 and 3 (the upper 3 cm of the cores; Fig. 4). We therefore use the range of  $0.033$  to  $0.050 \text{ Bq g}^{-1}$  of  ${}^{226}\text{Ra}$  to represent what we define as  $[{}^{226}\text{Ra}]_{\text{SED}}$ . The sedimentation rate (*rate*;  $1.4 \pm 0.4 \text{ kg m}^{-2} \text{ yr}^{-1}$ ) and surface area ( $SA$ ;  $90 \text{ km}^2$ ) were calculated by Serruya (1978) and used by Nishri et al. (1999) to describe sedimentation in the pelagic area.

Also,  ${}^{226}\text{Ra}_{\text{NWC}}$  is the amount of Ra lost per year through pumping ( $F_{\text{NWC}}$ ) the lake water to supply the National Water Carrier:

$${}^{226}\text{Ra}_{\text{NWC}} [\text{Bq/yr}] = [{}^{226}\text{Ra}]_{\text{SG}} [\text{Bq/L}] \times F_{\text{NWC}} [\text{L/yr}] \quad (15)$$

The  ${}^{226}\text{Ra}$  activity of the water pumped to the NWC is the same as that of the lake water. On average, a volume of  $420 \times 10^9$  liters is pumped through the NWC each year.

The total incoming  ${}^{226}\text{Ra}$  flux ( ${}^{226}\text{Ra}_{\text{IN}}$ ) is composed of dissolved Ra delivered through the Jordan River ( ${}^{226}\text{Ra}_{\text{JR,DISS}}$ ), particulate Ra entering the lake through the Jordan River ( ${}^{226}\text{Ra}_{\text{JR,SED}}$ ), and the unmonitored saline groundwater flux ( ${}^{226}\text{Ra}_{\text{SAL}}$ ), assumed to be predominantly of Fuliya type:

$${}^{226}\text{Ra}_{\text{IN}} = {}^{226}\text{Ra}_{\text{JR,DISS}} + {}^{226}\text{Ra}_{\text{JR,SED}} + {}^{226}\text{Ra}_{\text{SAL}} \quad (16)$$

$${}^{226}\text{Ra}_{\text{JR,DISS}} [\text{Bq/yr}] = [{}^{226}\text{Ra}]_{\text{JR,DISS}} [\text{Bq/L}] \times F_{\text{JR,W}} [\text{L/yr}] \quad (17)$$

We estimate that the annual water flux from the Jordan River,  $F_{\text{JR,W}}$ , is  $400 \times 10^9 \text{ L yr}^{-1}$  with a  ${}^{226}\text{Ra}$  activity of  $0.0015 \text{ Bq L}^{-1}$  (Table 1). Based on measurements of suspended matter arriving from the Jordan River ( $0.033 \text{ Bq g}^{-1}$ ) conducted by Stiller and Imboden (1984), we estimate the Jordan River  ${}^{226}\text{Ra}$  sedimentary flux,  ${}^{226}\text{Ra}_{\text{JR,SED}}$ , as

$${}^{226}\text{Ra}_{\text{JR,SED}} [\text{Bq/yr}] = [{}^{226}\text{Ra}]_{\text{JR,SED}} [\text{Bq/g}] \times F_{\text{JR,SED}} [\text{g/yr}] \quad (18)$$

where  $F_{\text{JR,SED}}$  is the annual sediment flux from the Jordan River and is calculated to be  $40 \times 10^6 \text{ kg yr}^{-1}$ . This flux is the main source of allochthonous sediments arriving to the lake (Stiller and Imboden 1984). Using Eqs. 12–18, the amount of  ${}^{226}\text{Ra}$  entering through unmonitored saline groundwater discharge can be calculated as

$${}^{226}\text{Ra}_{\text{SAL}} [\text{Bq/yr}] = {}^{226}\text{Ra}_{\text{OUT}} - {}^{226}\text{Ra}_{\text{JR,DISS}} - {}^{226}\text{Ra}_{\text{JR,SED}} \quad (19)$$

Assuming that the saline groundwater source to the lake is Fuliya-type water containing  $1200 \text{ mg Cl L}^{-1}$  and  $0.12 \text{ Bq } {}^{226}\text{Ra L}^{-1}$ , the annual Fuliya-type flux entering the lake is calculated to be between  $44 \times 10^9 \text{ L yr}^{-1}$  and  $61 \times 10^9 \text{ L yr}^{-1}$ , corresponding to  $[{}^{226}\text{Ra}]_{\text{SED}}$  values of  $0.033$  and  $0.050 \text{ Bq g}^{-1}$ , respectively. This calculated Fuliya-type saline flux would insert  $52.7 \times 10^6$  to  $73.4 \times 10^6 \text{ kg Cl yr}^{-1}$  to the lake. This is similar, yet slightly lower, than the  $\sim 75 \times 10^6 \text{ kg}$

Table 3. Flux values calculated for different contribution proportions of Fuliya and THS groundwater discharge to the Sea of Galilee.

Assumption	Fuliya-type flux* ( $\times 10^9$ L yr $^{-1}$ )	THS-type flux† ( $\times 10^9$ L yr $^{-1}$ )	Cl $^{-}$ flux ( $\times 10^6$ kg yr $^{-1}$ )	$^{228}\text{Ra}_{\text{REG}}$ ( $\times 10^7$ Bq yr $^{-1}$ )
100% Fuliya type‡	43.90	N/A	52.68	5.15
99% Fuliya type, 1% THS type‡	32.92	0.32	45.21	6.07
98% Fuliya type, 2% THS type‡	26.34	0.51	40.74	6.62
100% Fuliya type§	61.16	N/A	73.39	2.48
99% Fuliya type, 1% THS type§	45.87	0.44	62.99	3.12
98% Fuliya type, 2% THS type§	36.69	0.71	56.76	3.50

\* Fuliya-type water is taken as having 1180 mg Cl L $^{-1}$ , 0.122 Bq  $^{226}\text{Ra}$  L $^{-1}$ .

† THS type water is taken as having 4.21 Bq  $^{226}\text{Ra}$  L $^{-1}$ .

‡ [ $^{226}\text{Ra}$ ] in lake suspended matter is 0.033 Bq g $^{-1}$ .

§ [ $^{226}\text{Ra}$ ] in lake suspended matter is 0.050 Bq g $^{-1}$ .

yr $^{-1}$  estimated by Nishri et al. (1999) using a chloride mass-balance alone.

Dror et al. (1999) used the ratio of Br:Cl in the lake and its sources to show that Fuliya-type water alone cannot explain the Br:Cl ratio observed in the lake and postulated an additional contribution from THS-type water with a high Br:Cl ratio. Contribution of THS-type water with a higher  $^{228}\text{Ra}$ : $^{226}\text{Ra}$  ratio could also explain why the lake water has a higher  $^{228}\text{Ra}$ : $^{226}\text{Ra}$  ratio than that of Fuliya-type water. We show, for example, that for a scenario in which the saline groundwater flow is composed of 98% Fuliya-type water and 2% of THS water mixture (Table 3), the saline flux of Fuliya-type water would contribute only 26–37  $\times 10^9$  L yr $^{-1}$  and 40.7  $\times 10^6$ –56.8  $\times 10^6$  kg Cl yr $^{-1}$  using [ $^{226}\text{Ra}$ ] $_{\text{SED}}$  values of 0.033 and 0.050 Bq g $^{-1}$ , respectively.

Since the  $^{226}\text{Ra}$  activity measured in the bottom sediments in the lake represents both “supported Ra,” arriving as the sedimentary flux of the Jordan River, and “adsorbed Ra,” which is derived from scavenging of saline groundwater discharge, we calculate the relative proportion of dissolved Ra being lost through scavenging by:

$$^{226}\text{Ra}_{\text{SCAV}}[\%] = \frac{^{226}\text{Ra}_{\text{SED}}[\text{Bq/yr}] - ^{226}\text{Ra}_{\text{JR,SED}}[\text{Bq/yr}]}{^{226}\text{Ra}_{\text{SAL}}[\text{Bq/yr}]} \quad (20)$$

$\times 100$

Our calculations suggest that 54–67% of the dissolved  $^{226}\text{Ra}$  entering the lake is lost through adsorption. This estimate is similar to our independent observation that uses the difference between the  $^{226}\text{Ra}$ :Cl ratio in the lake and saline springs to show that 75–86% of the  $^{226}\text{Ra}$  entering the lake through the unmonitored saline groundwater discharge is being scavenged to the sediments.

Our flux calculations can now be used to evaluate the residence time of  $^{226}\text{Ra}$  in the Sea of Galilee by dividing the existing  $^{226}\text{Ra}$  reservoir in the lake (standing stock) by the yearly entering or exiting flux (Eq. 21):

$$T_{\text{Ra}}[\text{yr}] = \frac{\text{existing}}{\text{entering/year}} \quad (21)$$

$$= \frac{[^{226}\text{Ra}]_{\text{SG}}[\text{Bq/L}] \times \text{Vol}[\text{L}]}{(^{226}\text{Ra}_{\text{JR,DISS}} + ^{226}\text{Ra}_{\text{JR,SED}} + ^{226}\text{Ra}_{\text{SAL}})[\text{Bq/yr}]}$$

where [ $^{226}\text{Ra}$ ] $_{\text{SG}}$  is the average  $^{226}\text{Ra}$  activity in the lake (0.00735 Bq L $^{-1}$ ), Vol is its average volume of the lake (4.3  $\times 10^{12}$  liters), and the incoming fluxes are calculated from Eqs. 18–20. Using Eq. 21, the calculated residence time of Ra in the lake is 4.3 and 3.4 yr for [ $^{226}\text{Ra}$ ] $_{\text{SED}}$  values of 0.033 and 0.050 Bq g $^{-1}$ , respectively. The similarity of this calculation to previous estimations made by Nishri and Stiller (1997) (residence time in the order of 3.8 years) strengthens the validity of our estimation.

*Reconstructing the pre-SDC  $^{226}\text{Ra}$  activity in the Sea of Galilee*—Assuming the pre-SDC lake (before 1964) was in steady state, we can now use mass-balance equations to calculate the expected  $^{226}\text{Ra}$  activity in the lake during the early 1960s, several years before the diversion of the saline inflows to the lake took place. Ra sources to the pre-1964 lake, as described in Eq. 22, were through the Jordan River ( $^{226}\text{Ra}_{\text{JR,DISS}}$ ), the now-diverted saline springs of Fuliya, THS, and Tabgha ( $^{226}\text{Ra}_{\text{FUL,pre64}}$ ,  $^{226}\text{Ra}_{\text{THS,pre64}}$ , and  $^{226}\text{Ra}_{\text{TAB,pre64}}$ , respectively), and the unmonitored saline flux still entering the lake and calculated using Eq. 19 ( $^{226}\text{Ra}_{\text{SAL,now}}$ ):

$$^{226}\text{Ra}_{\text{IN,pre64}} = ^{226}\text{Ra}_{\text{SAL,now}} + ^{226}\text{Ra}_{\text{FUL,pre64}} + ^{226}\text{Ra}_{\text{THS,pre64}} + ^{226}\text{Ra}_{\text{TAB,pre64}} + ^{226}\text{Ra}_{\text{JR,DISS}} \quad (22)$$

We assume the saline flux that entered the pre-1964 lake to equal the average saline flux diverted away from it today; that is, 0.75  $\times 10^9$ , 1  $\times 10^9$ , and 15  $\times 10^9$  L yr $^{-1}$  for the Fuliya, THS, and Tabgha Springs, respectively. We assume that all of the diverted Fuliya Springs are of “Fuliya type,” bearing 0.122 Bq  $^{226}\text{Ra}$  L $^{-1}$ . THS and Tabgha activities are taken as the averaged values measured in these springs (4.211 and 0.444 Bq  $^{226}\text{Ra}$  L $^{-1}$ , respectively; Tables 1, 2). The flux of dissolved Ra entering through the Jordan River is assumed to be the same as today (Eq. 17). To account for the flux of Ra currently entering the lake through unmonitored flow from the saline springs, we use the water fluxes calculated after Eq. 19 (Table 3). As discussed earlier, we consider a scenario where all the unmonitored saline flux is of Fuliya-type water, as well as other scenarios where up to 2% of the water is derived from THS-type water.

Ra outputs in the pre-SDC lake are through outflow to the southern part of the Jordan River and local use

( $^{226}\text{Ra}_{\text{SJR,DISS}}$ ) and scavenging to the sediments:

$$^{226}\text{Ra}_{\text{OUT,pre64}} = ^{226}\text{Ra}_{\text{SJR,DISS}} + ^{226}\text{Ra}_{\text{SCAV,pre64}} \quad (23)$$

Using Eq. 20, we calculate the current scavenging fraction (54–67%) for the lake. However, the salinity of the Sea of Galilee in the early 1960s was higher than today (more than 350 mg Cl L<sup>-1</sup>; Gvirtzman 2002), and, thus, following our adsorption experiment (Fig. 5), we expect lower adsorption fraction under the higher salinity of the Sea of Galilee during the early 1960s. Assuming Cl<sup>-</sup> content of 350 mg L<sup>-1</sup> during the early 1960s, the relative change in the  $^{226}\text{Ra}_{\text{ads}} : ^{226}\text{Ra}_{\text{dis}}$  ratio detected by our adsorption experiment (Fig. 5) indicates that the scavenging fraction during the early 1960s would have been 94% of the current scavenging rate (when Cl concentration is 250 mg L<sup>-1</sup>). Following this calculation, the early 1960s scavenging fraction is estimated at 51–63% ( $^{226}\text{Ra}_{\text{SCAV,pre64}}$  [%]). The amount of  $^{226}\text{Ra}$  lost through scavenging to the sediments in the pre-SDC lake would therefore be

$$^{226}\text{Ra}_{\text{SCAV,pre64}}[\text{Bq/yr}] = ^{226}\text{Ra}_{\text{SCAV,pre64}}[\%] \times (^{226}\text{Ra}_{\text{SAL,now}} + ^{226}\text{Ra}_{\text{FUL,pre64}} + ^{226}\text{Ra}_{\text{THS,pre64}} + ^{226}\text{Ra}_{\text{TAB,pre64}})[\text{Bq/yr}] \quad (24)$$

The remaining exiting Ra flux,  $^{226}\text{Ra}_{\text{SJR,DISS}}$ , is assumed to discharge the same amount of water as is currently pumped through the NWC ( $F_{\text{NWC}}[\text{L yr}^{-1}]$ ; Eq. 15), bearing the average activity of the pre-SDC lake ( $[^{226}\text{Ra}]_{\text{SG,pre64}} [\text{Bq L}^{-1}]$ ). Assuming steady state and combining Eqs. 22–24, we use Eq. 25 to calculate the  $^{226}\text{Ra}$  activity in the pre-SDC lake:

$$[^{226}\text{Ra}]_{\text{SG,pre64}}[\text{Bq/L}] = \frac{^{226}\text{Ra}_{\text{JR,DISS}}[\text{Bq/yr}]}{F_{\text{NWC}}[\text{L/yr}]} + (1 - ^{226}\text{Ra}_{\text{SCAV,pre64}}[\%]) \times \{ (^{226}\text{Ra}_{\text{SAL,now}} + ^{226}\text{Ra}_{\text{FUL,pre64}} + ^{226}\text{Ra}_{\text{THS,pre64}} + ^{226}\text{Ra}_{\text{TAB,pre64}})[\text{Bq/yr}] \div F_{\text{NWC}}[\text{L/yr}] \} \quad (25)$$

Our calculations suggest that the pre-SDC  $^{226}\text{Ra}$  activity in the lake was between 0.018 to 0.020 Bq L<sup>-1</sup>. This value is in agreement with the actual  $^{226}\text{Ra}$  activity ( $0.018 \pm 0.0012$  Bq L<sup>-1</sup>) measured in the Sea of Galilee during 1962 (Gilboa 1963). This agreement reinforces the validity of our estimation for the unmonitored saline flux to the Sea of Galilee.

*$^{228}\text{Ra}$  mass balance in the Sea of Galilee*—A similar approach to the  $^{226}\text{Ra}$  mass balance provides the base for conducting mass-balance calculations for  $^{228}\text{Ra}$  in the Sea of Galilee. Sinks for  $^{228}\text{Ra}$  in the lake (Eq. 26) consist of  $^{228}\text{Ra}$  exiting the lake through the NWC, scavenging to the sediments, and radioactive decay:

$$^{228}\text{Ra}_{\text{OUT}} = ^{228}\text{Ra}_{\text{NWC}} + ^{228}\text{Ra}_{\text{SCAV}} + ^{228}\text{Ra}_{\text{DEC}} \quad (26)$$

The value of  $^{228}\text{Ra}_{\text{NWC}}$  is defined by Eq. 15 as the annual amount of  $^{228}\text{Ra}$  exiting the lake due to pumping through the National Water Carrier. The activity of  $^{228}\text{Ra}$  in suspended matter derived from the Jordan River was not reported by Stiller and Imboden (1984), and thus we cannot use this direct measurement as we did for the  $^{226}\text{Ra}$  mass balance. We

evaluate the amount of  $^{228}\text{Ra}$  lost through sedimentation by assuming that the reactivity of all Ra isotopes with suspended matter is identical. We therefore assume  $^{228}\text{Ra}_{\text{SCAV}}$  (%) is equal to the  $^{226}\text{Ra}_{\text{SCAV}}$  (%) determined in Eq. 20:

$$^{228}\text{Ra}_{\text{SCAV}}[\text{Bq/yr}] = ^{228}\text{Ra}_{\text{SCAV}}[\%] \times ^{228}\text{Ra}_{\text{SAL}}[\text{Bq/yr}] = ^{226}\text{Ra}_{\text{SCAV}}[\%] \times ^{228}\text{Ra}_{\text{SAL}}[\text{Bq/yr}] \quad (27)$$

Since the half-life of  $^{228}\text{Ra}$  (5.7 yr) is in the same scale as the residence time of water (~6 yr) in the Sea of Galilee, we must account for radioactive decay as an additional sink for  $^{228}\text{Ra}$ . As described in Eq. 28,  $^{228}\text{Ra}_{\text{DEC}}$  accounts for the annual decay of all the sources and sinks of  $^{228}\text{Ra}$ :

$$^{228}\text{Ra}_{\text{DEC}}[\text{Bq/yr}] = (^{228}\text{Ra}_{\text{NWC}} + ^{228}\text{Ra}_{\text{SCAV}} + ^{228}\text{Ra}_{\text{JR,DISS}} + ^{228}\text{Ra}_{\text{SAL}}) \times (1 - e^{-\lambda_{228}t}) \quad (28)$$

The incoming flux of  $^{228}\text{Ra}$  ( $^{228}\text{Ra}_{\text{IN}}$ ) consists of dissolved  $^{228}\text{Ra}$  entering through the Jordan River ( $^{228}\text{Ra}_{\text{JR,DISS}}$ ) and dissolved  $^{228}\text{Ra}$  entering through the saline springs ( $^{228}\text{Ra}_{\text{SAL}}$ ). Any excess  $^{228}\text{Ra}$  found in the system is considered to be regenerated through the sediments ( $^{228}\text{Ra}_{\text{REG}}$ ), mainly by diffusion, as suggested earlier in our discussion, so

$$^{228}\text{Ra}_{\text{IN}} = ^{228}\text{Ra}_{\text{JR,DISS}} + ^{228}\text{Ra}_{\text{SAL}} + ^{228}\text{Ra}_{\text{REG}} \quad (29)$$

To account for the  $^{228}\text{Ra}$  arriving through the saline springs ( $^{228}\text{Ra}_{\text{SAL}}$ ), we considered the saline flux calculated using Eq. 19 to be all of “Fuliya type,” as well as scenarios where up to 2% of the Ra arrived from THS-type water (Table 3):

$$^{228}\text{Ra}_{\text{SAL}}[\text{Bq/yr}] = F_{^{226}\text{Ra,SAL}}[\text{L/yr}] \times [^{228}\text{Ra}]_{\text{SAL}}[\text{Bq/L}] = \frac{^{226}\text{Ra}_{\text{SAL}}[\text{Bq/yr}]}{[^{226}\text{Ra}]_{\text{SAL}}[\text{Bq/L}]} \times [^{228}\text{Ra}]_{\text{SAL}}[\text{Bq/L}] \quad (30)$$

Combining Eqs. 26–30m we can find the amount of regenerated  $^{228}\text{Ra}$  as

$$^{228}\text{Ra}_{\text{REG}}[\text{Bq/yr}] = ^{228}\text{Ra}_{\text{OUT}} - ^{228}\text{Ra}_{\text{JR,DISS}} - ^{228}\text{Ra}_{\text{SAL}} \quad (31)$$

Our calculations predict that of the  $^{228}\text{Ra}$  inventory in the Sea of Galilee, a range of 2.48 to  $6.62 \times 10^7$  Bq yr<sup>-1</sup> (Table 3) originates from regeneration and/or diffusion through bottom sediments. Consequently, we estimate that 4% to 10% of the dissolved  $^{228}\text{Ra}$  in the lake is derived via regeneration from the underlying sediments, 34% to 43% is from the Jordan River, and 48% to 61% comes through the unmonitored saline groundwater discharge.

This study investigates the Ra isotope geochemistry of the fresh lake water of the Sea of Galilee and the saline springs that constitute the major salt source to the lake. Integration of the data shows that a large fraction of the Ra entering the lake via the saline inflow is removed through scavenging to suspended matter and sedimentation. We propose that the removal is by adsorption onto suspended matter in the lake water, yet validation by direct measurements of Ra in suspended matter in the lake is

necessary. The relationships between Ra isotopes with different half-lives indicate that the reactions between lake water and suspended matter involve adsorption-desorption exchange, recoil, and decay of the short-lived  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  isotopes. Our data suggest an average residence time of suspended matter on the scale of 2 weeks. Mass balance was used to determine the residence time of the residual dissolved Ra (3 to 4 yr) in the lake system.

Our data suggest that the unmonitored saline groundwater flux varies between  $44 \times 10^9 \text{ L yr}^{-1}$  and  $61 \times 10^9 \text{ L yr}^{-1}$ , assuming the entering groundwater is of "Fuliya type." We also estimate that  $2.48\text{--}6.62 \times 10^7 \text{ Bq }^{228}\text{Ra yr}^{-1}$  enters the lake through regeneration from bottom sediments, which makes up 4% to 10% of the overall annual  $^{228}\text{Ra}$  flux entering the lake. We combined a mass-balance approach with the results of our adsorption experiment to calculate the expected  $^{226}\text{Ra}$  activity in the lake before the diversion of saline springs to the lake. Our calculated  $^{226}\text{Ra}$  activity,  $0.018\text{--}0.020 \text{ Bq L}^{-1}$ , is in agreement with the  $0.018 \pm 0.0012 \text{ Bq }^{226}\text{Ra L}^{-1}$  that was actually measured in the lake in 1962.

In spite of the nonconservative behavior of Ra in freshwater systems, this study demonstrates that variations of the Ra isotope quartet can provide essential information for quantifying the flow of saline groundwater into a freshwater lake and the residence time of dissolved Ra in the lake water.

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