תכנית-מחקר המוגשת לאישור כתכנית לעבודת-דוקטור

מרץ, 2009

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רדיום ורדון במערכת ההידרולוגית של אגם היפרסליני (ים המלח)

Radium and Radon in the Hydrological System of a Hypersaline Lake (The Dead Sea)

הריני מאשר את הנושא ואת התוכנית, ומסכים להדריך את המועמדת בביצוע עבודה זו:

אברהם סטרינסקי

יוסף יחיאלי

ישי וינשטיין

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Appendix A ^{222}Rn and ^{226}Ra activities in the Dead Sea and its region.

Site	Site type	Sampling date	^{222}Rn (dpm/L)	^{226}Ra (dpm/L)	^{222}Rn excess (dpm/L)	density (kg/L)	shore (km)	distance from Depth (m)	DS conditions
Arugot-Keshet	borehole	21/01/2008	1181.75	5.54	1176.21	1			
EG11 – 15.85m	borehole	21/01/2008	1288.89	5.96	1282.93	1.056			
EG16 – 11.5m	borehole	22/01/2008	262.52	58.88	203.63	1.235			
EG22 – 26.5m	borehole	21/01/2008	697.80	39.06	658.74	1.228			
EG16 – 7.05m	borehole	21/01/2008	2090.34	22.24	2068.10	1.149			
EG-16 – 6.05m	borehole	21/01/2008	1430.21	15.98	1414.23	1.073			
EG-16 – 7.05m	borehole	22/01/2008	1967.73	35.16	1932.57	1.148			
DS Wadi Arugot	lake	13/02/2008	303.98	156.43	147.54	1.24	0	0	40 cm waves
DS Air near EG16	Air	13/02/2008	0.53						
DS eg320	lake	27/02/2008	476.41	162.71	313.70	1.24	8	0	No waves
DS eg320	lake	27/02/2008	320.98	162.71	158.26	1.235	8	0	No waves
DS eg320	lake	27/02/2008	374.63	216.30	158.32	1.235	8	40	No waves
DS eg320	lake	27/02/2008	303.09	164.91	138.17	1.235	8	150	No waves
DS eg320	lake	27/02/2008	253.39	166.09	87.30	1.235	8	300	No waves
DS	lake	27/02/2008	440.68	178.22	262.46	1.235	0.5	0	No waves
DS	lake	27/02/2008	505.16	202.04	303.12	1.235	0.1	0	No waves
DS Wadi Arugot	lake	27/07/2008	284.56	210.95	73.62	1.2394	0	0.50	few cm waves
DS Wadi Arugot	lake	27/07/2008	230.64	152.87	77.78	1.2395	0	0.50	few cm waves
spring, Wadi Arugot	spring	27/07/2008	3007.03	18.33	2988.70	1.0833			
DS Arugot fan	lake	27/07/2008	112.35	156.94	-44.59		0	0.50	few cm waves
DS Arugot fan	lake	27/07/2008	242.14	142.42	99.72	1.24	0	0.50	few cm waves
DS Arugot fan	lake	27/07/2008	183.10	172.76	10.34	1.2372	0	0.5	few cm waves
DS Arugot fan	lake	27/07/2008	164.45	171.00	-6.55	1.2381	0	0.5	few cm waves
DS Arugot fan	lake	27/07/2008	151.47	166.64	-15.17	1.238	0	0.3	few cm waves
DS Arugot fan	lake	27/07/2008	199.42	185.21	14.21	1.2386	0	0.2	few cm waves
DS Arugot fan	lake	27/07/2008	176.12	172.73	3.40	1.2391	0	0.2	few cm waves
En-Qedem	spring	27/07/2008	4977.86	199.00	4778.86	1.1295			
En-Qedem	spring	27/07/2008	626.93			1.1302			
DS near En-Qedem	lake	27/07/2008	305.57			1.2295	0	0.1	few cm waves
DS near En-Qedem	lake	27/07/2008	446.44	159.72	286.72	1.2368	0	0.2	few cm waves
DS near En-Qedem	lake	27/07/2008	132.36	143.06	-10.70	1.2337	0	0.1	few cm waves
DS Wadi Arugot	lake	03/11/2008	180.57			1.24			10 cm waves
DS near En-Qedem	lake	03/11/2008	273.57						30 cm waves
DS near En-Qedem	lake	03/11/2008	264.68						30 cm waves
En-Qedem	spring	03/11/2008	1629.37						
En-Qedem	spring	03/11/2008	1031.70						
En-Qedem	spring	03/11/2008	1428.40						
DS eg320	lake	11/11/2008	189.80			1.24	8	260	10 cm waves
DS eg320	lake	11/11/2008	178.86			1.24	8	250	10 cm waves
DS eg320	lake	11/11/2008	198.31			1.24	8	210	10 cm waves
DS eg320	lake	11/11/2008	116.41			1.24	8	50	10 cm waves
DS eg320	lake	11/11/2008	185.15			1.24	8	60	10 cm waves
DS eg320	lake	11/11/2008	182.61			1.24	8	150	10 cm waves
DS eg320	lake	11/11/2008	193.60			1.24	8	75	10 cm waves
DS Wadi Arugot	lake	11/11/2008	243.97			1.24	0	0	10 cm waves
DS eg320	lake	11/11/2008	157.01			1.24	8	260	10 cm waves
DS eg320	lake	11/11/2008	0.00			1.24	8	150	10 cm waves

procedure (dependent on the actual concentration).

5.3 Simulations

All simulations will be done with the USGS SUTRA code (Voss, 1984). SUTRA is a saturated-unsaturated variable density groundwater flow model with solute or energy transport. The model is based on a numerical solution by finite element of the mass conservation equations of the groundwater and the solute (Bear, 1972).

SUTRA was found suitable for simulating the hydrological system around the Dead Sea with its high density (Kiro, 2006) and will be combined with a geochemical model.

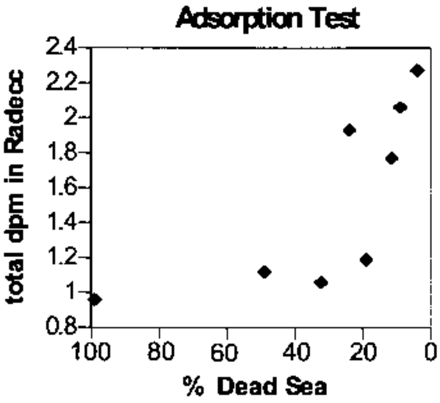


Figure 16: Results of an adsorption test of radium from the Dead Sea water. The efficiency increases with dilution.

of radium on the fibers was low (30–50%), probably due to its high ionic strength. Dilution of the Dead Sea water seems to significantly improve the adsorption. A preliminary adsorption test showed that the Dead Sea water should be diluted to at least 1:20 (Fig. 16). I hope to determine the optimal dilution proportions within a short while. However, since large volumes of Dead Sea water are needed in order to get good statistics of ^{223}Ra and ^{224}Ra, I would rather determine the efficiency of each sample by comparing the ^{226}Ra activity on the fibers with direct measurements on water samples.

The chance-coincidence of ^{224}Ra and ^{223}Ra according to this method must be studied thoroughly because of the high activity of ^{226}Ra in the Dead Sea. Giffin's (1963) correction is applicable only for small activities of ^{226}Ra and thus can not be applied to Dead Sea samples. An experiment is planned in which different activities of ^{226}Ra will be added to a ^{224}Ra standard, reaching the high activities of the Dead Sea. The correction methods that will be tested are Giffin's (1963) method and a correction based on Poisson distribution, which describes the probability of disintegration in radioactive decay.

Activities of ^{228}Ra will be measured either by gamma spectrometry in the Physics laboratories of the Hebrew University or by MC-ICP-MS in the Geological Survey. Both methodologies involve the striping of the radium from the Mn fibers and precipitation with Ba-sulfate or Mn-sulfate. In the latter (MC-ICP-MS), the sample will probably also have to go through an ion exchange preconcentration

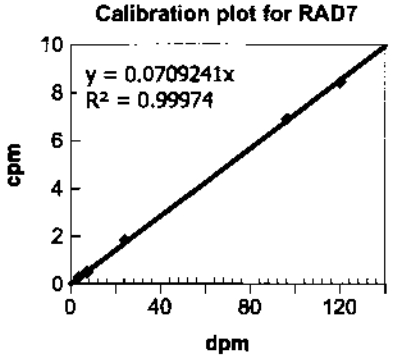


Figure 14: Calibration plot of ^{226}Ra standards for using the RAD7.

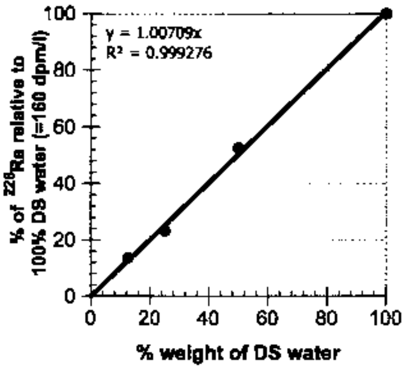


Figure 15: Results of bubbling test in the RAD7.

the Dead Sea water, the ordinary 250cc sampling bottles can not be used because of the penetration of sample water into the tubing. Thus, I changed to vacuum Erlenmeyer flasks with head space. The efficiency of this procedure was defined by ^{226}Ra standards as d.p.m=0.0709(c.p.m) (Fig. 14).

Another test was performed to determine the yield of radon from the Dead Sea water. This was done by diluting Dead Sea water to various degrees and measuring the ^{226}Ra activity with the RAD7. The results of this experiment show that the yield does not change with salinity and thus, that the RAD-H2O procedure is effectively bubbling out all the radon from the Dead Sea water samples (Fig. 15).

5.2 ^{223}Ra ^{224}Ra and ^{228}Ra measurements

Radium will be leached out of the water via adsorption on MnO_2-coated fibers. ^{223}Ra and ^{224}Ra will be measured by a Delayed Coincidence Counter (RaDeCC). In this methodology, ^{219}Rn and ^{220}Rn which are produced by the disintegration of the two radium isotopes are swept into a scintillation cell where alpha decay of the radon nuclides and their polonium daughters (^{219}Po and ^{220}Po, respectively) occurs. The scintillations are translated into electronic signals, and these are sent to a delayed coincidence circuit, which discriminates electronically decays of 219 and 215 (thus 223) from decays of 220 and 216 (thus 224) or others (Giffin et al., 1963; Moore and Arnold, 1996).

During the preliminary work, I found out that with the Dead Sea water the yield

The effect of salinity on radium activities in the Dead Sea brine type will also be examined.

A combination between hydrological and geochemical models will be developed during a 3–4 months visit to the US Geological Survey under the supervision of Dr. Cliff Voss in order to estimate the four radium isotope distribution in the aquifer and the Dead Sea and to define processes that control this distribution. These combined simulations will be performed for parameter estimation and a comprehensive understanding of this issue. This has not been done before and the simulations should give insight into the different processes depending on the hydrological and geochemical parameters under the dynamic system of the lake level changes. The objective is both to understand the processes that can occur in general and to determine the specific mechanism in the Dead Sea region.

4.4 Simulations of the hydrological system of the Dead Sea

In addition to the field work, simulations with the USGS SUTRA code (Voss, 1984) will be done as a part of all stages mentioned above. The effect of seasonal level changes on the groundwater system will be studied and fresh and saline groundwater discharge will be calculated.

5 Methods

5.1 ^{222}Rn and ^{226}Ra measurements

Analyses of ^{222}Rn and ^{226}Ra will be done by the radon-in-air RAD7 detector (DURRIDGE Co.). The RAD7 measures ^{222}Rn by detecting the α decay of its daughter ^{218}Po. The RAD7 is used with the RAD-H2O application, where the ^{222}Rn is bubbled out of the water sample and delivered with air to the RAD7, where it reaches secular equilibrium with the ^{218}Po. The samples are re-measured for ^{226}Ra after three weeks, the time needed for attaining a secular equilibrium with the ^{222}Rn.

During my preliminary work, I found out that because of the high viscosity of

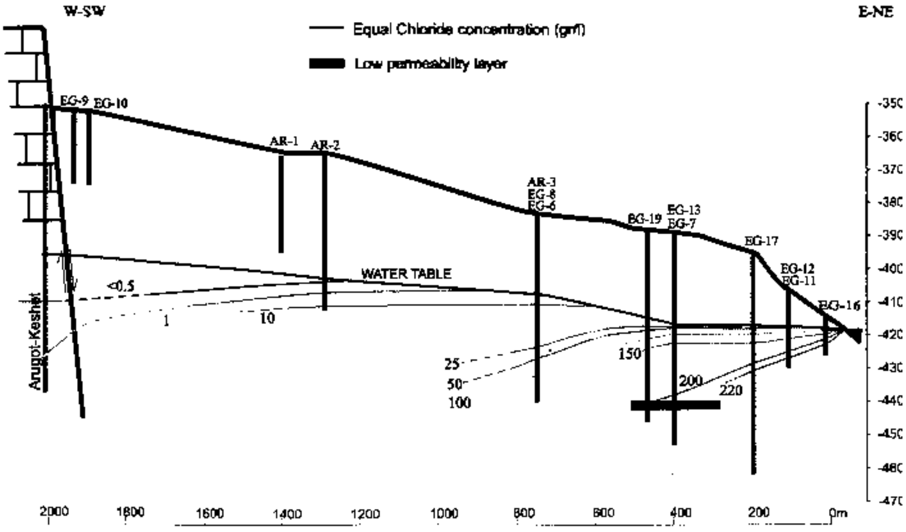


Figure 13: Cross section of the alluvial fan of Wadi Arugot. The solid lines are constant chloride concentrations.

The two short-lived radium isotopes and the long-lived ^{226}Ra will be studied along on-shore transects and at different depths in the aquifer. The isotope activities and ratios will be used to study the role of adsorption, precipitation, recoil and dilution in the aquifer. This includes analyses of all four radium isotopes in groundwater from boreholes at the alluvial fan of Wadi Arugot (Fig. 12). The samples will be taken at different depths and salinities on several occasions during the research period. The high activity of ^{226}Ra and the low activity of ^{224}Ra in the Dead Sea water allow distinguishing between the removal of radium by adsorption or precipitation, which is represented by the decrease of ^{226}Ra in the aquifer Dead Sea water, and recoil, which is represented by the enrichment of ^{224}Ra in the aquifer Dead Sea water, assuming that there is no isotope fractionation during adsorption or precipitation processes. Thus, all ^{224}Ra activity in the Dead Sea aquifer water is due to recoil. An additional piezometer will be installed immediately next to the Dead Sea shore in order to sample the Dead Sea waters close to their encounter with the aquifer.

In addition, lab incubation experiments will be performed on typical Dead Sea alluvium aquifer material in order to study the effects of the adsorption, recoil and dilution processes on the activities of radium isotopes in the aquifer water.

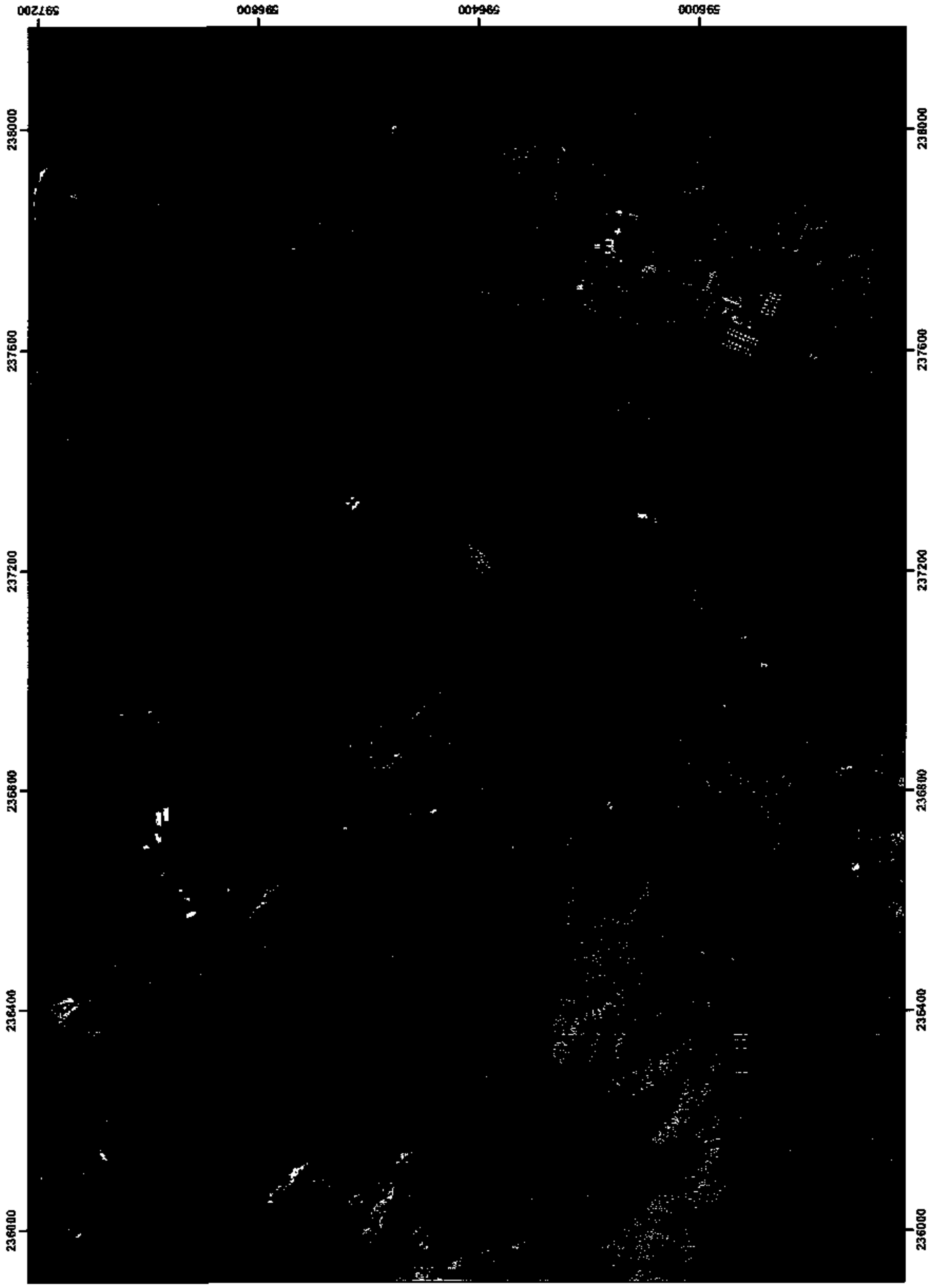


Figure 12: Location map of borholes in the alluvial fan of Wadi Arugot.

of radium and thorium on suspended particulates and bottom sediments.

5. Examination of the Ba-Sr-Ca sulfates system vs. radium.

4.2 Groundwater discharge to the Dead Sea.

The higher activities of radon and short-lived radium isotopes in the aquifer allow the assessment of fresh and brine groundwater discharge to the Dead Sea by mass balances of the excess radon. This will be done by analysis of Dead Sea water from two sites. The first is En-Gedi (Arugot), where radon is enriched in the fresh water and brines, albeit not in the circulated Dead Sea water in the aquifer, and radium is enriched in all saline water. The use of both radon and radium isotopes will allow the distinction between the three sources. The second site is En-Qedem, which represents brine discharge to the lake. In both sites, sampling will be conducted in several locations along the coastline in order to identify discharge spatial variability.

The boreholes around En-Gedi (Fig. 12) will be sampled for radium and radon isotopes and for chemical composition in order to define the end members of the groundwater discharging to the Dead Sea. Sampling will be performed at various depths and distances from the lake, as to get samples of different salinities (different proportions of Dead Sea/aquifer mixing) and of the aquifer brine (Fig. 13).

The seasonal effect and the impact of continuous lake level drop on the groundwater discharge will be studied by repeated sampling of radon and radium isotopes (4–6 times a year for two years) at one coastal site. A high resolution time series measurements of ^{222}Rn, ^{226}Ra, ^{224}Ra and ^{223}Ra will also be conducted at this site in order to identify and quantify short period variability in the discharge. If variability is identified, it will be repeated twice a year for two years.

4.3 Circulation of saline water in the aquifer

The study of saline water circulation includes the understanding of the behavior of the radium isotopes in the aquifer through three different approaches: collection of field data, lab experiments and simulations.

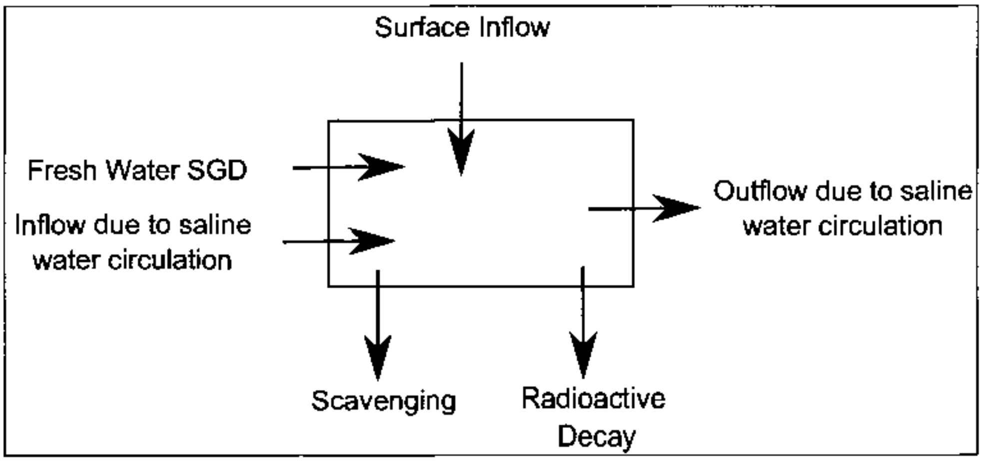


Figure 11: Box plot of sources and sinks of radium into the Dead Sea.

4 Research Plan

4.1 Radium mass balances in the Dead Sea

This will include mass halances of ^{228}Ra and ^{226}Ra. The different half lives of these two isotopes can reveal processes in different time scales concerning the radium mass balance and residence time in the Dead Sea. For this purpose, all sources and sinks of radium (Fig. 11) in the Dead Sea system should be estimated. This study includes:

1. Transects and depth profiles of ^{228}Ra and ^{226}Ra in the Dead Sea.

2. Re-analysis of ^{226}Ra in historic (1970's) Dead Sea water samples, to be compared with present radium data in order to establish the resemblance or differences in the radium and radon inventory between the meromictic stage and the current monomictic stage. The water samples are stored in the Dead Sea water bank in the Israel Geological Survey under the supervision of I. Gavrieli and M. Stiller.

3. Analysis of the radium isotope quartet in water sources of the Dead Sea, which includes fresh and saline groundwater from boreholes, fresh and saline springs and rivers from various sites around the Dead Sea.

4. Assessment of radium scavenging, as well as of supported radium, by analysis

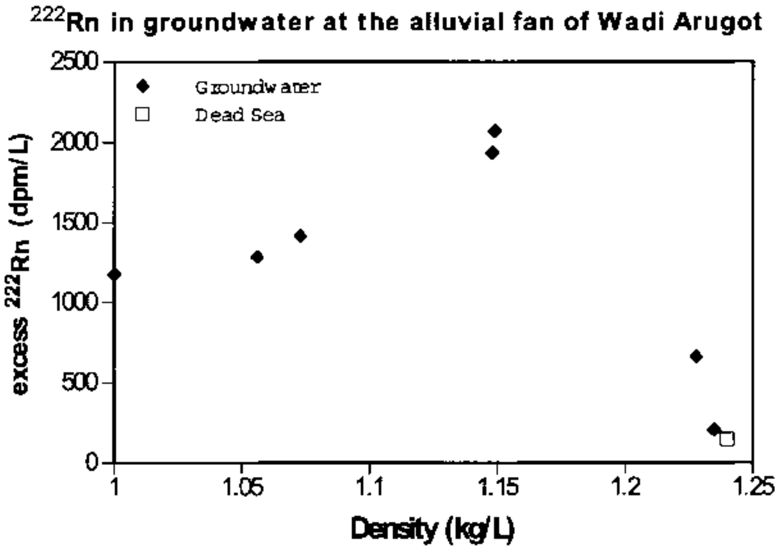
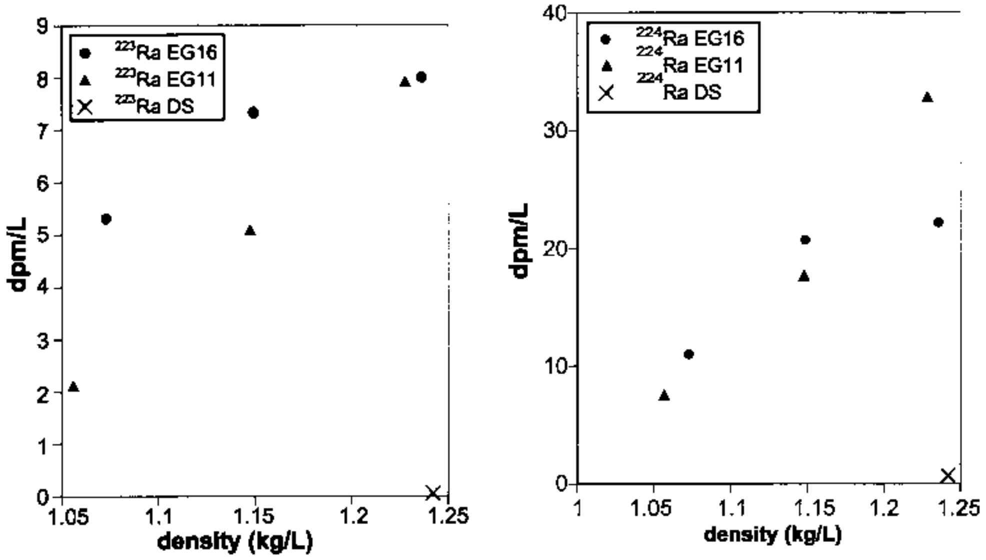


Figure 10: ^{222}Rn in the groundwater at the alluvial fan of Wadi Arugot. The highest values are in groundwater which contains a brine component.

3 Research Goals

The main goal of my thesis is to study the effect of the Dead Sea level changes on the groundwater system and lake water circulation in the aquifer. This includes the study of:

1. The source of radon and radium in the Dead Sea water.
2. The role of saline water circulation in the alluvial aquifer and its implication on the radium and radon mass balance of the Dead Sea.
3. Study of the radium and the water mass balance in the Dead Sea system.
4. The effect of the Dead Sea level changes (seasonal changes and the lake level drop) on the hydrogeological system, with emphasis on discharge of fresh and saline groundwater to the lake.



(a) (b)

Figure 9: ^{224}Ra and ^{223}Ra in the aquifer. The activities are significantly greater than in the Dead Sea and increase with salinity.

fresh groundwater at Arugot-Keshet borehole is 1182 dpm/L and in En-Shulamit (both in the Cretaceous aquifer) it is around 3000 dpm/L (Appendix A). However, the relation between the density and the activities of ^{222}Rn in the groundwater is not linear (Fig. 10), suggesting that there is some contribution from another high-Rn source, probably a brine. The preliminary results suggest that the sources of the excess ^{222}Rn in the upper water of the Dead Sea and near the coast are fresh groundwater, saline springs such as En-Qedem, and freshwater springs such as Enot-Zukim. The higher activity of the short-lived radium isotopes near the shore is probably due to the discharge of circulated Dead Sea water. This will be further studied in a quantitatively manner.

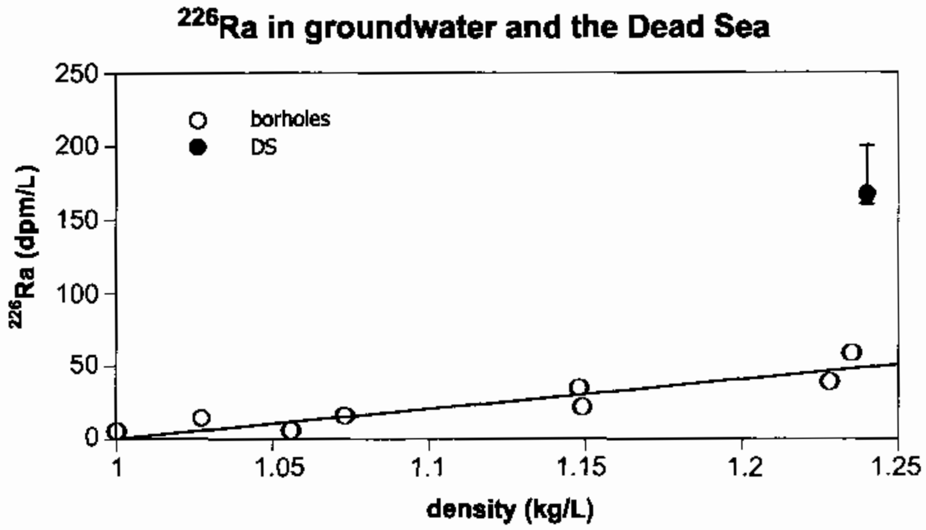


Figure 7: ^{226}**Ra in the Dead Sea and in groundwater in the alluvial fan of Wadi Arugot. The** ^{226}**Ra decreases in the aquifer Dead Sea water due to adsorption or precipitation and then decreases due to dilution.**

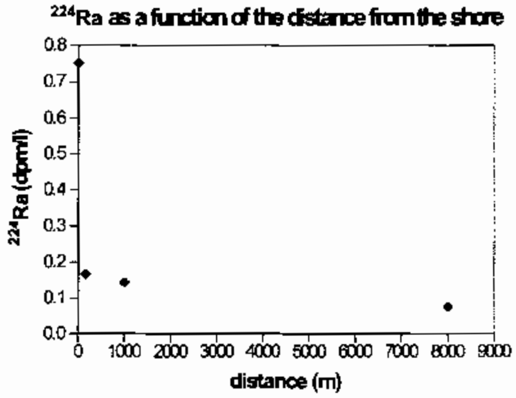


Figure 8: ^{224}**Ra activities in the Dead Sea. The** ^{224}**Ra activity increases toward the shore due to saline water inflow to the lake.**

ity decreases with the salinity due to dilution by radium-poor fresh groundwater (Fig. 7). On the other hand, activities of ^{224}Ra are higher in groundwater by several orders of magnitude than in the Dead Sea. The ^{224}Ra activity in the Dead Sea ranges from 0.75 dpm/L near the shore to 0.07 dpm/L at 8 km away from the shore (Fig. 8) while in the Dead Sea water inside the aquifer the activity reaches 22 dpm/L (Fig. 9). The activity is lower in the fresher groundwater due to dilution. Similar pattern was observed in the activities of ^{223}Ra.

Both the fresh and the brine groundwater are very rich in ^{222}Rn, which makes them traceable when they discharge to the Dead Sea. The ^{222}Rn activity in the

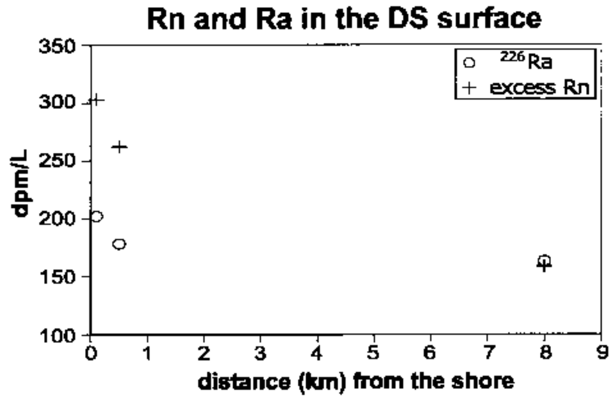


Figure 4: Excess ^{222}Rn along the Dead Sea. The ^{222}Rn increases towards the shore.

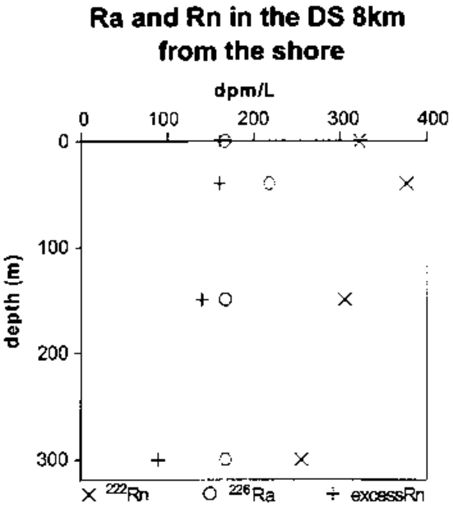


Figure 5: Radium and radon profiles in the Dead Sea. The ^{226}Ra is almost constant in the Dead Sea and the ^{222}Rn decreases with depth.

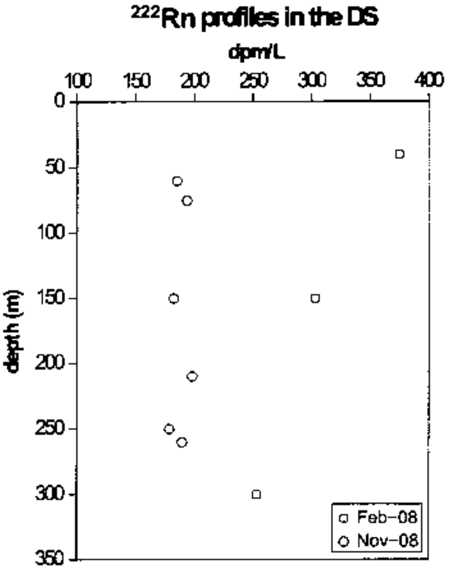


Figure 6: ^{222}Rn profiles at eg320 (8km from shore). The ^{222}Rn activities are higher during the winter.

for these samples).

Dead Sea activities of total ^{222}Rn in the vicinity of the hot springs of En-Qedem were higher than those near Wadi Arugot during the summer, ranging from 300 to 450 dpm/L during the summer (August-2008, Appendix A).

2.1.2 Groundwater activities of ^{226}Ra, ^{224}Ra, ^{223}Ra and ^{222}Rn

Groundwater with Dead Sea salinity (circulated lake water) has significantly lower ^{226}Ra activities than in the lake (~60 dpm/L; Fig. 7).This may be due to adsorption or precipitation of the ^{226}Ra onto the aquifer sediments. Radium activ-

Date	Dpeth (m)	^{226}Ra (dpm/L)	Source
Nov-57	0	138	(Mazor, 1962)
Mar-63	0	148	(Gilboa, 1963)
Apr-63	0–40	145	(Stiller and Chung, 1984)
Apr-63	80–	111	(Stiller and Chung, 1984)
Mar-77	35–87	114–121	(Stiller and Chung, 1984)
Mar-77	137–289	96–99	(Stiller and Chung, 1984)
Dec-77	0	104	(Stiller and Chung, 1984)
Dec-77	220	88	(Stiller and Chung, 1984)
Feb-78	0–150	142	(Stiller and Chung, 1984)
Feb-78	175–	120	(Stiller and Chung, 1984)

Table 2: Historic activities of ^{226}Ra in the Dead Sea

2 Preliminary Results

2.1 Field data

Groundwater samples were taken from boreholes in the Wadi Arugot alluvial fan. Dead Sea samples were taken along a cross-shore transect up to 8 km off-shore from the Arugot area. A depth profile down to a depth of 300 meters was taken at the 8 km station during the winter and autumn. Another transect was conducted (August 2008) along a 2 km shoreline of the Arugot delta in order to study the differences in the activity of ^{222}Rn and radium isotopes between the center and the margins of the alluvial fan.

2.1.1 Dead Sea activities of ^{226}Ra and ^{222}Rn

The ^{226}Ra activity along the Dead Sea transect was very high and close to uniform, ranging from 160 to 200 dpm/L (most of the samples were around 160 dpm/L), with no seasonal variability (Table 1). These values agree with previous measurements in the upper water body of the Dead Sea. However, the present values are slightly greater than 1960's and 1970's values (Table 2), which could be due to a real change of the ^{226}Ra activity in the Dead Sea or due to intercalibration problems. ^{222}Rn activity shows significant variability with the highest activity found next to shore (300 dpm/L excess ^{222}Rn; Fig 4). The ^{222}Rn activity decreases towards the centre of the Dead Sea and with depth (Fig. 4, 5). In the summer, the total ^{222}Rn activity was much lower, ranging from 143 to 210 dpm/L compared to 303–505 in the winter (Fig. 6; Appendix A; the excess ^{222}Rn is not available yet

Location	**Sampling date**	**^{226}Ra (dpm/L)**	**Distance from shore (km)**	**depth (m)**
DS eg320	27/2/2008	162.71	8	0
DS eg320	27/2/2008	216.30	8	40
DS eg320	27/2/2008	164.91	8	150
DS eg320	27/2/2008	166.09	8	300
DS – Wadi Arugot	27/2/2008	178.22	0.5	0
DS – Wadi Arugot	27/2/2008	202.04	0.1	0
DS – Wadi Arugot	27/7/2008	210.95	0	0.5
DS – Wadi Arugot	27/7/2008	152.87	0	0.5
DS Arugot fan	27/7/2008	156.94	0	0.5
DS Arugot fan	27/7/2008	172.76	0	0.5
DS Arugot fan	27/7/2008	171.00	0	0.5
DS Arugot fan	27/7/2008	166.64	0	0.3
DS Arugot fan	27/7/2008	185.21	0	0.2
DS Arugot fan	27/7/2008	172.73	0	0.2
DS En-Qedem	27/7/2008	159.72	0	0.2
DS – Wadi Arugot	13/2/2008	156.00	0	0
DS eg320	27/2/2008	163.00	8	0
DS Zeelim	27/7/2008	152.00	0	0
DS Zeelim	27/7/2008	163.00	0	0

Table 1: ^{226}Ra activities in the Dead Sea

Chung (1987) and Chung and Craig (2004) suggested scavenging on particles as an additional possible sink for the radium, but they also agree that the main sink of radium in the lake is by radioactive decay. Activities of ^{222}Rn are also very high in springs around the Dead Sea (Moise et al., 2000) reaching 19,000 dpm/L in En-Qedem and 59,000 dpm/L in Enot-Zukim which is not supported by ^{226}Ra. Radon was hardly studied in the Dead Sea itself. Chung and Craig (2004) report excess radon of about 18 dpm/L in the deep layer before overturn. The steady-state model of Stiller and Chung (1984) does not include submerged groundwater around the Dead Sea, circulated saline water and sources of the eastern Dead Sea, which may be significant. The high activities of ^{228}Ra also do not agree with the steady-state model in which the ^{228}Ra should have decayed in the lower water mass and the activities at the upper water mass should have been smaller. Thus, the ^{226}Ra inventory may not be in steady state or there are unknown sources and sinks of radium.

of the Dead Sea (in Jordan) but there is not enough data about the discharge and composition of this water.

1.5 Radium and radon isotopes in the Dead Sea region

The Dead Sea is extremely enriched in radium relative to most natural water. The ^{226}Ra activity in the Dead Sea is 1,800 times that of Pacific surface water ($\sim$0.08 dpm/L) and almost 400 times ($\sim$0.35 dpm/L) that of Pacific deep water (Chan and Chung, 1987; Chung and Craig, 2004; Mazor, 1962; Stiller and Chung, 1984) whereas the salinity of the Dead Sea is only 10 times that of ocean water. The ^{226}Ra activities before the overturn were 141 and 121 dpm/L in the upper and lower water masses, respectively, and after the overturn they were 137 dpm/L (Chan and Chung, 1987; Stiller and Chung, 1984). These values yield the same ^{226}Ra inventory (Chan and Chung, 1987). The activity of ^{228}Ra in the Dead Sea is also higher by 2–3 orders of magnitude compared to the ocean (Somayajulu and Rengarajan, 1987). The ^{228}Ra activity during the meromictic stage was 1.57 dpm/L in the upper water mass, 0.13–0.22 dpm/L at a depth of 175 m and 0.42 at depth of 275 m. The higher values at the bottom of the profile can be due to diffusion from bottom sediments or influx of submarine springs (Somayajulu and Rengarajan, 1987). The activities of ^{226}Ra in the onshore springs range from 20 to 750 dpm/L (Gilboa, 1963; Mazor, 1962; Moise et al., 2000), and the ^{228}Ra/^{226}Ra ratios range from 0.028 to 0.740. Thus, the activities of ^{228}Ra range from 2 to 472 dpm/L (Moise et al., 2000). It seems that there is no correlation between the salinity of the springs and the ^{226}Ra activity. For example, the springs near En-Gedi have low activities of ^{226}Ra (8 dpm/L), while at Enot-Zukim relatively fresh water activities reach 400 dpm/L. Stiller and Chung (1984) suggested that the inventory of ^{226}Ra in the lake was built up a long time ago and that it is now in steady state with a relatively low input from the Jordan (1–2 dpm/L; Stiller and Chung, 1984), from other runoffs and from onshore and submerged springs. They estimated a total inflow of dpm/yr, balanced by the slow decay rate of the ^{226}Ra in the lake and a minor outflux via deposition with aragonite. Chan and

(2007) suggest, based on numerical simulations, that under steady-state conditions lake water sinks through the faults and then ascends via other faults at the lake shores together with fresh groundwater. During a lake level decline, the circulated Dead Sea water flows backwards through the faults and discharges at the bottom of the lake.

1.4.1 Brines in the Dead Sea area

Saline springs are common along the Dead Sea coast. At the western part of the lake, the discharge is concentrated along a coastline of ~3km at the Qedem-Shalem area. The amount of discharge in this area was estimated at 10^7 m^3/yr in 2005 (B. Rophe, Hydrological Service, pers. Comm.). The salinity of these brines is about half that of the Dead Sea water and their composition is somewhat different and is characterized by higher SO_4 concentration, as well as higher Na/Cl and Ca/Mg ratios (Gavrieli et al., 2001). These brines are abundant in the alluvial aquifer (Kiro, 2006; Yechieli and Arad, 1997; Yechieli et al., 2007a). It has been suggested that these brines infiltrated the subsurface during a high stand of an older lake that occupied the Dead Sea basin, and therefore represent a previous stage in the evolution of the Dead Sea brines (Yechieli et al., 1996). The relatively high temperature of some of these brines (~45°C) suggests an ascent from a depth of ~1000 m via faults in the Dead Sea rift (Gavrieli et al., 2001; Shalev et al., 2007). The rate of brine discharge to the Dead Sea was estimated by hydrological simulations to be significantly larger in recent years due to the drop in the lake level (Shalev and Yechieli, 2007).

Gavrieli and Stein (2006) suggested that the discharge mechanism of the thermal springs is similar to that proposed for Lake Kinneret (Goldschmidt et al., 1967; Gvirtzman et al., 1997). However, the hydrogeological configuration of the two lakes is different, and it has recently been suggested that the main driving force is the circulation of lake water via faults. The rate of this circulation is not clear and could range between hundreds to thousands of years (Shalev et al., 2007).

There is a relatively large amount of hot springs discharging to the rivers east

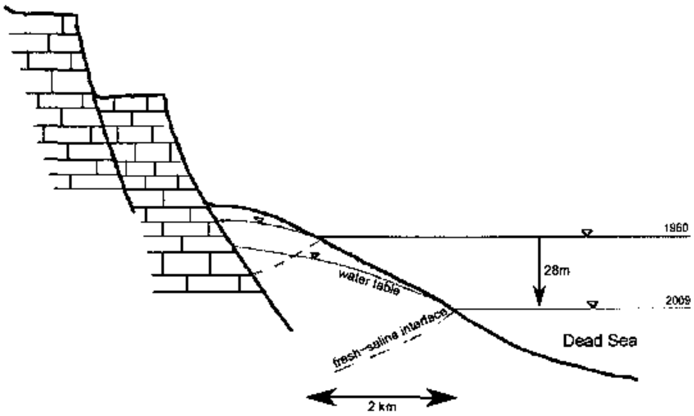


Figure 3: An illustration of the effect of the Dead Sea level drop on the groundwater system in the alluvial aquifer. The water table and the fresh-saline interface are dropping as a response to the lake level drop.

rate compared to the Dead Sea level drop (Yechieli et al., 2007b), while at Wadi Arugot the groundwater level decline rate is similar to that of the Dead Sea (Kiro et al., 2008). The Dead Sea level drop probably increases the fresh and saline groundwater inflow into the Dead Sea. The continuous lake level drop will cause an increase in the fresh groundwater discharge until a constant value is attained (Kiro et al., 2008). This issue will be tested in the field during my research.

The current Dead Sea salinity and density are 340 g/L and 1.24 kg/L, respectively. The extremely high density of the Dead Sea induces a very shallow transition zone between the fresh and saline water. According to the Ghyben-Herzberg approximation, the depth of the transition zone is 4.35 times the groundwater elevation, compared to 40 times in aquifers next to the ocean (Yechieli, 2000).

The saline groundwater also responds to the Dead Sea level drop. Field data and simulations show that the fresh-saline interface drops and widens over time. Furthermore, the drop also affects the saline water circulation in the aquifer. The lake level drop causes a saline water flow towards the lake due to a flushing process. However, simulations (Kiro et al., 2008) and geochemical data (Kiro, 2006) also show that there is still a flow of saline water from the Dead Sea into the aquifer.

Several faults were described at the bottom of the Dead Sea (Lazar and Ben-Avraham, 2002; Niemi and Ben-Avraham, 1997). These faults could serve as conduits for water migration between the aquifer and the lake. Shalev and Yechieli

1979). Since then, the lake changed to a monomictic regime (Anati et al., 1987; Lensky et al., 2005). Estimates of the duration of meromixis in the lake range between 170 to 320 years (Assaf and Nissenbaum, 1977; Chung and Craig, 2004; Stiller and Chung, 1984).

1.4 Hydrogeology of the Dead Sea basin

There are three main aquifers in the Dead Sea region: The Lower Cretaceous Kurnub aquifer, the Upper Cretaceous Judea aquifer and the Quaternary alluvial aquifer (Arad and Michaeli, 1967; Yechieli et al., 1995). The latter is adjacent to the Dead Sea, and it mainly consists of clastic sediments deposited in fan deltas (gravel, sand, and clay) and of lacustrine sediments (clay, aragouite, gypsum, and salt). Alternations between gravel and clay create several sub-aquifers that differ in their groundwater level and chemical composition. The alluvial aquifer is bounded on its west by normal faults, which set Cretaceous carbonate rocks of the Judea Group against Quaternary alluvial and lacustrine sediments. The freshwater recharge of the alluvial aquifer is through lateral flow from the Judea Group aquifer, which is replenished in the highlands 10–30 km to the west and by flash floods. Direct rain on this aquifer is negligible because of the arid climate in the Dead Sea region. Based on energy and mass balances, the annual influx to the Dead Sea is calculated today to be 265–335 million m^3, of which 60 million m^3 at most is subsurface inflow (Lensky et al., 2005). Other estimations of fluxes from specific sources into the Dead Sea include 60–150 million m^3/yr from the Jordan River (Holtzman et al., 2005) and 150–270 million m^3/yr from other inflows around the Dead Sea (Israel Hydrological Service, Salameh and Udluft, 1985).

There is a hydraulic relationship between the Dead Sea and the adjacent groundwater system in the alluvial aquifer (Fig. 3), expressed in a relatively rapid (a few days) water level response to level changes in the Dead Sea (Yechieli et al., 1995). Groundwater levels respond to the lake level drop at different rates according to the aquifer parameters and the slope of the lake boundary in different locations. For example in the Tureibe region, groundwater level declines at a lower

The lake level has been dropping from -390 m in the 1930's (Klein and Flohn, 1987) to -420 m in 2007, with a steeper decline since the 1960's reaching a rate of 1 m/yr during the past few years (Lensky et al., 2005). This level decline is the result of the increased utilization of water in the northern part of the basin, particularly the creation of the Israeli National Water Carrier in the 1960's and of the Jordanian Jordan Valley (King Abdullah) Canal in the 1970's. The Israel Dead Sea Works and the Jordanian Arab Potash Company significantly added to this negative balance by increasing the evaporation from the lake. Altogether, these changes in water management resulted in a decrease of the average annual inflow to the lake from 1600–2000 million m^3 in the early 1900's (Klein, 1998; Neumann, 1958; Salameh and El-Naser, 1999) to a current 265–335 million m^3 (Lensky et al., 2005). Assuming the present climate conditions, the evaporation is expected to decrease with time until it stops due to the decrease both in surface area and in the evaporation rate due to the increased salinity (Krumgalz et al., 2000; Yechieli et al., 1998). Calculations show that the Dead Sea is expected to reach a new steady state in about 300 years at a level of around -550 m (Yechieli et al., 1998).

The composition of the Dead Sea water is Ca-chloridic (Starinsky, 1974) with low Na/Cl ratio compared to that of normal ocean water (0.23 vs. 0.87, respectively). This composition is the result of the evaporation of seawater, which intruded the rift valley, followed by the dolomitization of carbonates in the adjacent aquifer. The Na/Cl ratio is still decreasing due to the present precipitation of halite in the lake (Gavrieli, 1997).

Until 1979, the Dead Sea was meromictic with an upper water mass that received inflows from sources such as rivers, springs and submerged groundwater and a lower water mass which remained isolated. In 1959 the pycnocline was at a depth of 40 m and there was a significant density difference between the lower and upper water masses (1.233 kg/L and 1.205 kg/L respectively). The declining lake level and the negative water balance caused an increase in the density of surface water which lead to a gradual increase in the depth of the pycnocline during the 1970's and eventually to a complete overturn in 1979 (Steinhorn, 1985; Steinhorn et al.,

^{228}Ac). In the aquifer and in surface water bodies, all the radium isotopes are produced by the decay of insoluble thorium parents, which are adsorbed on sediment grains or on suspended material. In freshwater, radium is mainly tightly bound to the surface of sediment grains (e.g., Lazar et al., 2008), while in saline water the radium is much more soluble and readily undergoes ion exchange with other adsorbed cations (e.g., Krishnaswami et al., 1991; Moore, 2000). In terms of hydrological and oceanographic applications, the short-lived isotopes ^{224}Ra and ^{223}Ra (half lives of 3.66 and 11.4 days, respectively) are used for studying short term processes such as coastal mass balances and mixing with the open sea (e.g., Moore, 1996, 2000; Shellenbarger et al., 2006), while ^{226}Ra (half life of 1,600 years) may be used for long-period studies such as residence times in the ocean and lakes (Broecker et al., 1967; Stiller and Chung, 1984). The combination of ^{226}Ra and the shorter-lived isotope ^{228}Ra (with half life of 5.7 years) may help in assessing processes and residence times in surface seawater (Moore et al., 2008).

Radon is a noble gas, which is produced by the decay of ^{226}Ra and disintegrates into ^{218}Po (half life 3.82 days). It has been shown to be a very powerful tool in tracking groundwater discharge to the sea (Burnett and Dulaiova, 2003; Weinstein et al., 2007). Usually, both radium isotopes and radon are enriched in groundwater compared to sea water and are used as a tracer for SGD (e.g., Cable et al., 1996; Corbett et al., 1999; Kelly and Moran, 2002; Moore, 1996). A combined study of radon and radium isotopes will allow the detection of the type of the discharging groundwater, as radium activity depends on water salinity (e.g. Krishnaswami et al., 1991; Weinstein et al., 2007), while radon is usually enriched in both fresh and the saline groundwater (e.g. Mulligan and Charette, 2006).

1.3 The Dead Sea

The Dead Sea is a terminal lake located in a deep pull-apart basin along the Dead Sea Transform which drains groundwater and surface water from a wide area. The deepest point in the lake is about -720 m. It is a terminal hypersaline lake with a current (2007) area of 620 km^2 and a drainage basin of 40,000 km^2.

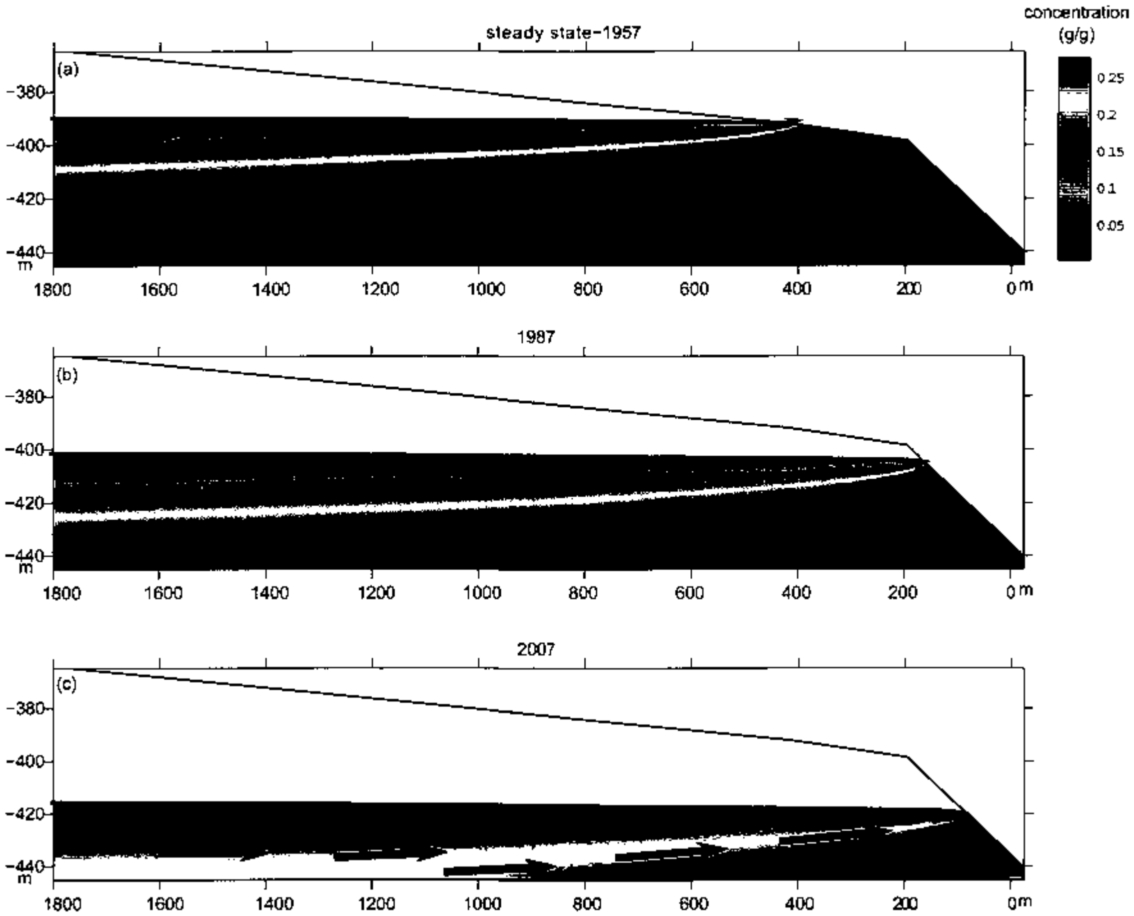


Figure 2: Saline water flow velocities during the Dead Sea level drop. The saline water flow direction can be towards the lake or from the lake into the aquifer.

SGD. The freshwater discharge increases with lake level decline until it is expected to attain a new constant value and the saline water is continuously flushing back into the lake. However, even in the case of a dropping lake level there still may be an inflow of saline water into the aquifer (Fig. 2; Kiro et al., 2008).

1.2 Radon and radium isotopes in groundwater and in the ocean

There are four naturally-occurring radium isotopes (the radium quartet), with half-lives ranging from 3.7 days to 1600 years. Two of them, the ^{226}Ra and ^{223}Ra, are the radioactive chain products of uranium isotopes (^{238}U and ^{235}U, respectively), while ^{228}Ra and ^{224}Ra are chain products of ^{232}Th. Three of the isotopes (^{226}Ra, ^{224}Ra and ^{223}Ra) disintegrate into radon daughters (^{222}Rn, ^{220}Rn and ^{219}Rn, respectively), while ^{228}Ra decays to ^{224}Ra via ^{228}Th (and the short-lived

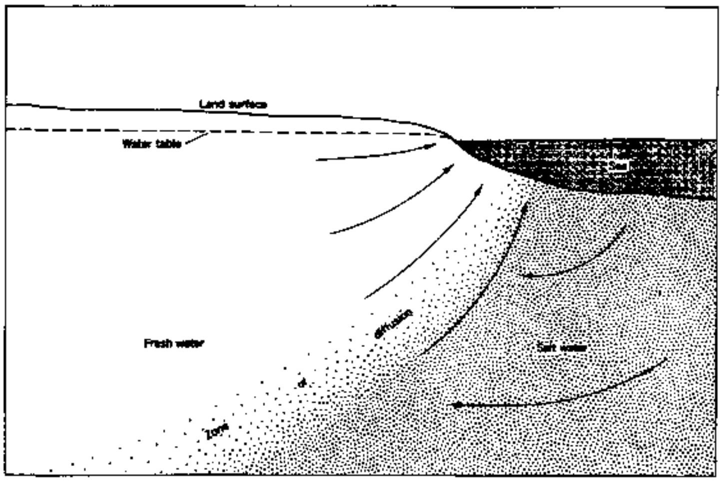


Figure 1: Saline water circulation in the aquifer according to the dispersive model (Cooper, 1959).

instein et al., 2007). In the last decade most balances were done with the short-lived radium isotopes. Moore (1996) concluded that direct groundwater discharge to the sea is a significant component of coastal geochemical mass balances and is about 40% of the river-water flux.

SGD can sometimes consist solely of fresh meteoric groundwater and may produce large fluxes, especially in karstic areas (e.g., in Northern Lebanon, where fresh SGD reaches values of hundreds of millions m^3/yr; Shaban et al., 2005). However, SGD is mostly a mixture of fresh groundwater and recirculated seawater (Michael et al., 2003; Moore, 1996; Moore and Church, 1996; Simmons, 1992) and the saline component is usually the dominant one. Fresh groundwater discharge is mainly driven by the hydraulic gradient between land and sea, while the driving force for the seawater circulation includes mechanisms at all scales such as waves, tides (Robinson et al., 1998), dispersive circulation (Fig. 1; Cooper, 1959) and seasonal changes (Michael et al., 2005).

SGD has also been measured in lakes (e.g., Boyle, 1994; Lee, 1977; Shaw and Prepas, 1990a). In some cases it can reach as much as 50% of the total inflow (Shaw and Prepas, 1990b) and it was found to be affected by lake level changes (Taniguchi and Fukuo, 1996).

A continuous base level drop affects both the fresh and saline components of

Abstract

The Dead Sea water system is unique in terms of its unusual geochemical composition, rapid lake level changes and the water compositions of the brines discharging along its shoreline. The groundwater interaction with the Dead Sea water and its effect on water mass balance of the Dead Sea has hardly been studied to date. The study of radium and radon isotopes in the Dead Sea can be used for understanding submarine groundwater discharge (SGD), processes of saline water circulation and water mass balances. For this purpose, this research will include an extensive study of radium in the Dead Sea.

The Dead Sea level has been dropping during the past 70 years due to human intervention in its water budget. This fast lake level drop makes the Dead Sea a suitable natural field lab for studying effects of base level changes on groundwater systems including SGD and saline water circulation. The field work will be accompanied by numerical simulations of groundwater flow and distribution of radium in order to have a complete understanding of all processes. The simulations will be done with the USGS SUTRA code.

Preliminary results show enrichment of ^{222}Rn in lake water next to the shore, especially during the winter, which implies seasonal changes in SGD. Furthermore, there is also an enrichment of the short-lived radium isotopes next to the shore, which implies saline water discharge. In this research I intend to define the role of SGD and saline water circulation in the lake water mass balance and to study how are these processes affected by the Dead Sea level drop using radium and radon isotopes.

1 Introduction

1.1 Submarine Groundwater Discharge

Submarine groundwater discharge (SGD) has been found to be a major process in coastal areas worldwide and is an important factor in coastal water mass balances (Moore, 1996; Simmons, 1992). SGD is assessed by various methods, including hydraulic gradient models or standard water balance considerations, as well as seepage meter measurements (Simmons, 1992) and radioisotope tracers such as radium isotopes (Moore, 1996) and ^{222}Rn (e.g., Burnett and Dulaiova, 2003; We-