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High Naturally Occurring Radioactivity in Fossil Groundwater from the Middle East

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High levels of naturally occurring and carcinogenic radium isotopes have been measured in low-saline and oxic groundwater from the Rum Group of the Disi sandstone aquifer in Jordan. The combined ^{228}Ra and ^{226}Ra activities are up to 2000% higher than international drinking water standards. Analyses of the host sandstone aquifer rocks show ^{228}Ra and ^{226}Ra activities and ratios that are consistent with previous reports of sandstone rocks from different parts of the world. A compilation of previous data in groundwater from worldwide sandstone aquifers shows large variations in Ra activities regardless of the groundwater salinity. On the basis of the distribution of the four Ra isotopes and the ratios of the short- to long-lived Ra isotopes, we postulate that Ra activity in groundwater is controlled by the balance of radioactive decay of parent Th isotopes on aquifer solids, decay of the dissolved radium isotopes, and adsorption of dissolved Ra on solid surfaces. The availability of surface adsorption sites, which depends on the clay content in the aquifer rocks, is therefore an important constraint for Ra activity in sandstone aquifers. These findings raise concerns about the safety of this and similar nonrenewable groundwater reservoirs, exacerbating the already severe water crisis in the Middle East.

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Introduction

The rise in population and associated increased water demands in the Middle East have placed tremendous pressure on available water resources, which in turn has accelerated the rates of their depletion and contamination (*1*). Jordan is considered one of the 10 most water-deprived nations in the world, with approximately 160 m^3 of available water per capita per year, and water demand (1500 million m^3/year) exceeds supply (900 million m^3/year) (*2, 3*). On the basis of a multimodel compilation, the Intergovernmental Panel on Climate Change Report (*4*) predicts a significant reduction in precipitation in the Middle East during the next few decades, which will likely exacerbate the water crisis in the region. In an effort to accommodate increasing water demands, water authorities in the region are seeking alternative water sources, including exploitation of nonrenewable ("fossil") groundwater. Massive amounts of nonrenewable groundwater are already extracted from the vast Nubian sandstone basins in the Arabian Peninsula ("Saq aquifer"), Sinai Peninsula and Negev, and northeastern Africa (*5–9*). In Jordan, fossil groundwater from the Disi aquifer is utilized by the domestic sector in Karak and Aqaba and has long been considered the future drinking water source of the kingdom, with a potential to provide 125 million m^3/ year of high-quality water for the next 50 years (*2, 3*). Recently, Jordan has launched a large-scale project that aims to transfer the Disi groundwater to the capital Amman.

The quality of groundwater from the Nubian sandstone basins in the Middle East is mostly high (e.g., low salinity with total dissolved solids below 300 mg/L). Thus, a challenge facing water authorities in the region is the absence of modern replenishment of these aquifers, i.e., the lack of sustainability of "mining" fossil water. In this paper we show that a high concentration (henceforth, activity) of the radionuclide radium can be another important limiting factor for utilization of this type of groundwater. Radium has four isotopes, ^{226}Ra (half-life of 1600 years) derived from the ^{228}U decay chain, ^{228}Ra (5.6 years) and ^{224}Ra (3.6 days) that are part of the ^{232}Th decay chain, and ^{223}Ra (11.4 days) from the ^{235}U decay chain. The high level of naturally occurring radium in drinking water has severe health implications (*10–15*). As a result, radium is defined as a group A carcinogen (*10*). The maximum contaminant levels (MCLs) for combined Ra activities set by the U.S. EPA (*10*), EU (*16*), and WHO (*17*) are listed in Table 1.

In this paper we present, for the first time, the radium isotope data of groundwater from the Disi aquifer system in southern Jordan. The objectives of this study are (1) to evaluate the sources of radium in the Disi aquifer and the possible mechanisms of radium mobilization from the host aquifer rocks and (2) to evaluate the impact of this phenomenon of future water utilization from similar aquifer basins in the Middle East.

Analytical Methods

Thirty-seven groundwater samples were collected from pumping wells in the Cambro-Ordovician sandstone Disi (Rum Group) aquifer (*5*) and the overlying Khreim Group in the Disi-Mudawwara (*18*) and Dead Sea areas in southern and central Jordan (Figure 1). Groundwater samples were measured for major and trace elements and for the four Ra isotopes. In addition, sandstone rocks from the Cambro-Ordovician sandstone in Disi, Jordan, and Lower Cretaceous Nubian sandstone rocks in the Negev, Israel, were measured for their ^{228}Ra and ^{226}Ra contents. Dissolved oxygen, pH, and

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TABLE 1. Salinity (Total Dissolved Solids (TDS), mg/L), Trace Element Concentrations (μg/L), Radium Activities (Bq/L), and Ra Activity Ratios Measured in Groundwater from the Disi Aquifer System in Southern and Central Jordan[a]

area and well name	well ID	date	TDS	[Ba]	[Mn]	[U]	^{226}Ra activity	^{228}Ra activity	^{224}Ra activity	^{223}Ra activity	^{228}Ra/^{226}Ra activity ratio	^{224}Ra/^{223}Ra activity ratio	^{224}Ra/^{228}Ra activity ratio	^{223}Ra/^{226}Ra activity ratio	%EU[b]	%EPA[c]	%WHO[d]
Unconfined Rum Group																	
Sahl El Suwan SS-6	ED1506[e]	10/5/07	246	24.3	1.3	4.1	0.34	0.37	0.62	0.011	1.08	55.43	1.67	0.035	252	384	465
Sahl El Suwan SS-5A	ED1505[e]	10/5/07	245	23.9	1.2	4.5	0.41	0.51	0.85	0.027	1.25	31.09	1.69	0.068	335	494	635
M 14 (Rum Co.)	ED1612	10/5/07	238	20.5	1.1	2.2	0.25	0.47	0.66	0.020	1.91	33.24	1.40	0.092	286	390	566
Sahl El Suwan SS-4	ED1504[e]	10/5/07	231	26.2	1.2	1.1	1.13	1.25	2.10	0.097	1.11	21.53	1.68	0.081	852	1287	1583
M 4 (Rum Co.)	ED1623	10/5/07	337	31.3	11.2	5.5	0.10	0.25	0.47	0.012	2.47	40.44	1.84	0.120	148	193	313
SS20 (Sahl El Suwwan)	ED1614[e]	10/5/07	410	33.5	0.6	6.7	0.88	1.43	2.95	0.121	1.62	24.33	2.06	0.143	893	1253	1828
M 5 (Rum Co.)	ED1624	10/5/07	392	34.6	6.2	5.5	0.12	0.24	0.63	0.012	2.06	53.50	2.60	0.127	145	195	319
Mneisheer M6	ED1540	10/5/07	235	21.2	3.3	3.4	0.34	0.96	1.35	0.042	2.82	32.22	1.40	0.120	548	702	1132
SS24 (Sahl El Suwwan)	ED1608	10/5/07	249	24.1	0.9	5.4	1.11	2.11	4.99	0.159	1.90	31.30	2.37	0.142	1275	1738	2732
Mneisheer W-2 /M 8	ED1402[e]	10/5/07	438	35.1	1.4	6.4	0.69	2.14	3.45	0.073	3.12	47.17	1.61	0.118	1207	1527	2560
Q'a Abu Suwana M2	ED1509[e]	12/9/04	257	23.2			0.10	0.20			2.05				118	158	207
Qa Disi well no. 3	QD3[e]	12/9/04	246	34.3			1.27	1.44			1.13				976	1468	1570
Quweirah well no. 3	S5[e]	13/9/04	631	37.8	4.9		0.21	0.47			2.24				277	367	491
Confined Rum Group																	
Gramco G 6	K1034	9/5/07	229	14.4	0.3	1.2	0.43	1.16	1.67	0.036	2.70	46.05	1.44	0.153	664	857	1371
Gramco G 3	K1031	9/5/07	237	15.9	0.8	1.3	0.62	1.72	3.19	0.068	2.78	46.72	1.86	0.111	984	1264	2108
Gramco G 4	K1039	9/5/07	254	17.9	1.0	1.3	0.50	1.41	2.24	0.037	2.80	59.84	1.60	0.216	804	1032	1685
Wafa 3	K1043	9/5/07	245	13.1	3.0	0.9	1.00	2.64	3.01		2.64		1.14	0.099	1518	1965	3038
Wafa 2	K1028	9/5/07	250	17.4	1.6	1.1	0.79	2.30	3.03	0.055	2.90	55.09	1.32	0.087	1307	1670	2685
Wafa 1	K1027	9/5/07	235	12.7	1.0	1.0	0.53	1.49	2.37	0.056	2.82	42.42	1.59	0.060	849	1089	1782
Arab Agriculture Co. 1	K1016	9/5/07	247	14.2	1.3	1.1	0.65	1.98	3.34	0.062	3.06	53.94	1.69	0.127	1118	1418	2382
Arab Agriculture Co. 3	K1020	9/5/07	231	14.2	0.7	1.0	0.56	1.71	3.38	0.051	3.03	65.72	1.98	0.104	966	1227	2106
Arab Agriculture Co. 6	K1026	9/5/07	235	14.6	0.8	1.2	0.70	2.47	3.45		3.54		1.40	0.156	1376	1715	2888
Suleiman Mar'l El A'taneh	K3023	9/5/07	253	15.3	1.9	1.3	0.85	3.11	3.81		3.64		1.22	0.189	1727	2144	3578
Al-Arabiya well no. 9	K1041	12/9/04	226	14.5			0.62	2.20			3.53				1223	1525	2260
Suleiman Abu Juweied		12/9/04	235	14.7			0.71	1.89			2.66				1088	1406	1962
Halet A'mmar 1 (HA1)	K3021	12/9/04	249	12.7	4.4		0.85	1.93			2.27				1135	1503	2015
Khreim Group																	
Al Hodood well	11S1	12/9/04	531	54.4	14.9		0.09	0.14	0.12	0.004	1.56	29.72	0.87	0.047	89	126	164
Fawwaz Jeryes El Halaseh (BH9)	ED1602	13/9/04	733	32.9	13.0		0.05	0.09	0.50	0.020	2.05	24.68	5.29	0.439	56	76	150
Hasan Salameh El Hawashleh 1	ED3009	13/9/04	651	35.0	42.8		0.08	0.10	0.27	0.011	1.28	25.14	2.68	0.137	66	96	136
Mohammad Odeh El Njadat	ED3008	13/9/04	1006	34.6	14.2		0.04	0.08	0.16	0.008	1.87	19.67	2.09	0.200	47	64	98
Halet A'mmar 2 (HA2)/W16	K3000	9/5/07	464	51.4	12.4		0.11	0.15	0.16	0.005	1.43	33.42	1.02	0.032	98	140	179
Halet A'mmar 2 (HA2)/W16	K3000	12/9/04	460	65.5	17.2		0.08	0.12	0.12	0.003	1.50	34.90	0.94	0.040	78	111	143
Central Jordan																	
Lajjun deep well	Lajjun[e]	12/6/04	640	72.6	582.6		0.31	0.86			2.75				495	637	895
Potash well 2-TA2	DA1039	12/6/04	650	94.5	228.2		0.30	0.34			1.12				229	345	367
Potash well 1-TA1	DA3023	12/6/04	2149	84.5	47.3		0.72	0.84			1.18				563	841	912
Potash well 1-TA1	DA3023	6/5/07	687	91.8	205.4	<0.5	0.30	0.48	0.52	0.015	1.62	35.79	1.09	0.049	300	420	510
Potash well 2-TA2	DA1039	6/5/07	2532	201.2	209.0	<0.5	0.78	1.10	0.86	0.028	1.40	31.45	0.79	0.035	705	1015	1175

[a] Columns %EC, %EPA, and %WHO represent percent values over the maximum contaminant levels of the European Union, the U.S. EPA, and the World Health Organization drinking water standards, respectively. [b] %EU = [(^{226}Ra activity/0.5) + (^{228}Ra activity/0.2)] × 100 (percent over EU MCL values). [c] %EPA= [(^{226}Ra activity + ^{228}Ra activity)/0.185] × 100 (percent over EPA MCL values). [d] %WHO = [^{226}Ra activity + (^{228}Ra activity/0.1) + ^{224}Ra activity + ^{223}Ra activity] × 100 (percent over WHO guideline). [e] The well is used for public water supply.

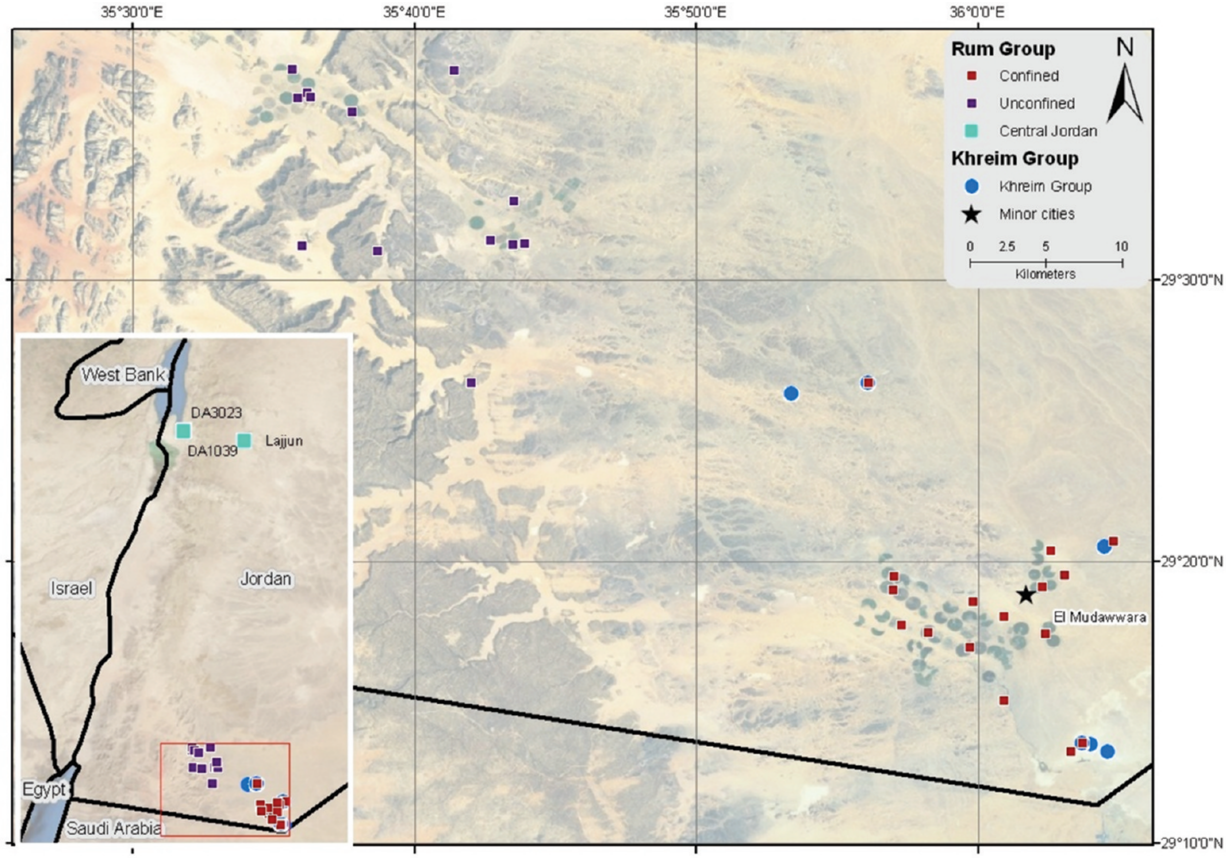

FIGURE 1. Location map of investigated wells from the unconfined (purple) and confined (red) areas of the Rum Group and wells from the Khreim Group (blue) in the Disi-Mudawwara area in southern Jordan. Also marked are deep wells (green) from the Disi aquifer in central Jordan.

temperature were measured at the field. Water samples were measured for major and trace elements using inductively coupled plasma atomic emission spectroscopy (ICP-AES) and ion chromatography (IC). Radium was extracted by Mn fiber (*19, 20*) without preflirtation. ^{226}Ra, ^{228}Ra, and ^{224}Ra−^{223}Ra isotopes were determined by a radon counter (*21*), γ spectrometer, and delayed coincidence α counter (*22*), respectively (see the Supporting Information, Supplementary Text S1).

Results and Discussion

Radium Occurrence in Groundwater from the Disi Aquifer. Results from the Disi aquifer show four groundwater clusters (Figures 1 and 2, Tables 1 and S1 and S2, Supporting Information): (1) the unconfined zone of the Rum Group with a salinity range of 230−630 mg/L, pH of 7.0−7.9, temperature of 27−31 °C, and dissolved oxygen (DO) content of 6−8 mg/L; (2) the confined Rum Group with low salinity (240 mg/L), pH of 6.5−7.7, temperature of 32−35.4 °C, and DO content of 6−8 mg/L; (3) Khreim Group that is overlying the confined Rum Group with a salinity range of 460−1000 mg/L, pH of 7.0−7.8, and temperature of 28−30 °C; (4) confined groundwater from the Disi aquifer in central Jordan with a wide salinity range of 640−2530 mg/L, pH of 6.8−8.0, temperature of 36.4−44.6 °C, and DO content of of 3 mg/L. The data show that groundwater from the Rum Group has high Ra activities, exceeding by several orders of magnitude the threshold level acceptable by international drinking water standards (Table 1, Figure 2). While groundwater from the unconfined zone has a wide range of Ra activities, in the confined zone the Ra activity is high throughout, within a smaller range. High Ra activities have also been found in pumping wells in central Jordan. In contrast, groundwater with higher salinity from the Khreim Group has significantly lower radium activities. The groundwater clusters also differ in their ^{228}Ra/^{226}Ra ratios (Figure 3). In addition, the groundwater clusters differ in their redox state; groundwater from the Rum Group has a high level of DO and low contents of Mn and U, while groundwater from the Khrein Group and from deep wells in central Jordan are reduced, as indicated by the low DO content, high Mn content, and below-detection-limit concentration of apparently insoluble U (Tables 1 and S1). The inverse correlation between dissolved oxygen and Mn^{2+} is a typical indicator of the redox state of the water (*23*).

Measurements of Cambro-Ordovician sandstone rocks from Disi (Rum Group) in Jordan and Lower Cretaceous Nubian sandstone rocks in the Negev, Israel, show ^{228}Ra and ^{226}Ra activities and ^{228}Ra/^{226}Ra activity ratios (~1.6) that are consistent with previous reports of sandstone rocks from different parts of the world (Figure 4) (*24*). While aeolian sand dunes have typically low Ra activities and a ^{228}Ra/^{226}Ra ratio of ~1.1, beach sands with higher contents of heavy minerals (e.g., zircon, monazite) have significantly higher Ra concentrations and higher ^{228}Ra/^{226}Ra ratios (~1.6) (*24*). Consequently, the radium content in the host Nubian sandstone aquifer is not different from that of other worldwide sandstone basins.

Radium in groundwater can be derived from multiple sources including (1) Ra in-growth via decay of the dissolved U or Th parents in the solution, (2) dissolution from the aquifer minerals, (3) α-recoil from the parent nucleus in the aquifer rocks and on the surface coating located on clay minerals and oxides, (4) adsorption/desorption exchange with Ra adsorbed on the surface coating, clays, and oxides, and (5) coprecipitation with and/or dissolution of secondary

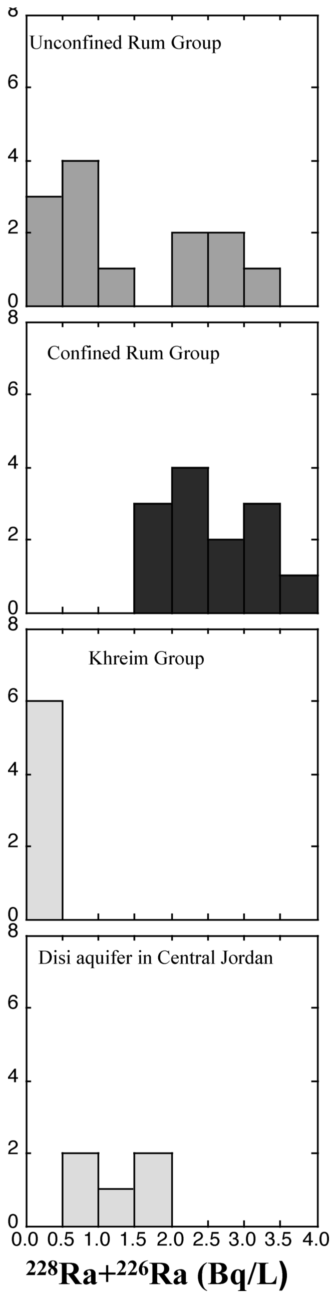

FIGURE 2. Histograms of combined ^{228}Ra and ^{226}Ra activities (1 Bq = 1 (disintegration/s)/L) in groundwater from the unconfined Rum Group, confined Rum Group, Khreim Group, and deep wells from the Disi aquifer in central Jordan. Most wells show high Ra activities exceeding the drinking water MCL values of the U.S. EPA (combined ^{226}Ra and ^{228}Ra activities 0.185 Bq/L), EU (^{226}Ra activity 0.5 Bq/L, ^{228}Ra activity 0.2 Bq/L), and WHO (^{226}Ra activity 1 Bq/L, ^{228}Ra activity 0.1 Bq/L).

minerals (e.g., barite) (*25*–*28*). Dissolution of Ra-containing minerals would result in low ratios of the short-lived to long-lived Ra isotopes (e.g., ^{228}Ra/^{226}Ra) relative to the host aquifer rocks (*25*–*28*), given the slow dissolution rate and relatively faster decay of the short-lived Ra isotopes. In contrast, combination of the recoil process and decay of the dissolved radium isotopes and their rapid adsorption would increase the relative abundances of the short-lived Ra isotopes (*25*–*28*). Previous studies have emphasized that the recoil process balanced by adsorption on clay minerals and oxides is the predominant process that controls Ra activity in groundwater (*25*–*28*). The results from the Disi aquifer are consistent with this assumption, as the ^{224}Ra/^{228}Ra and ^{223}Ra/

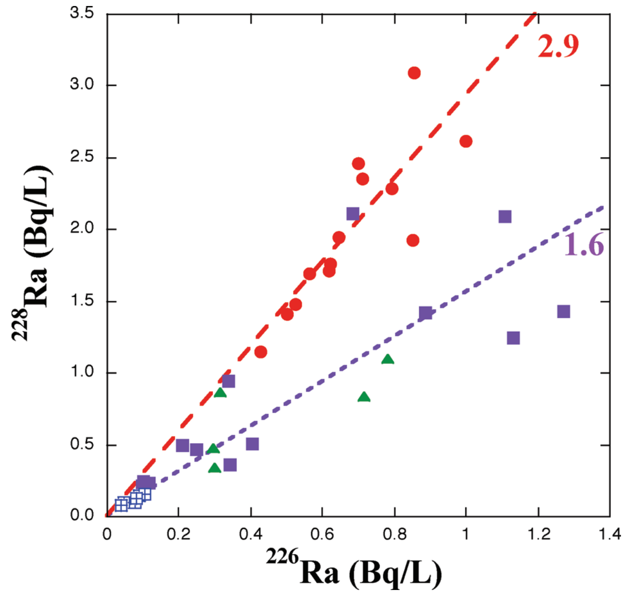

FIGURE 3. ^{228}Ra vs ^{226}Ra activities in groundwater from the unconfined zone (purple squares) and confined zone (red circles) of the Rum Group, the Khreim Group (checkered squares), and deep wells from the Disi aquifer in central Jordan (green triangles). Groundwater from the confined Rum group is distinguished by high ^{228}Ra/^{226}Ra ratios (slope 2.9) relative to groundwater from the unconfined zone, Khreim Group, and central Jordan (all 1.6) as well as the ^{228}Ra/^{226}Ra activity ratios in sandstone rocks (~1.6).

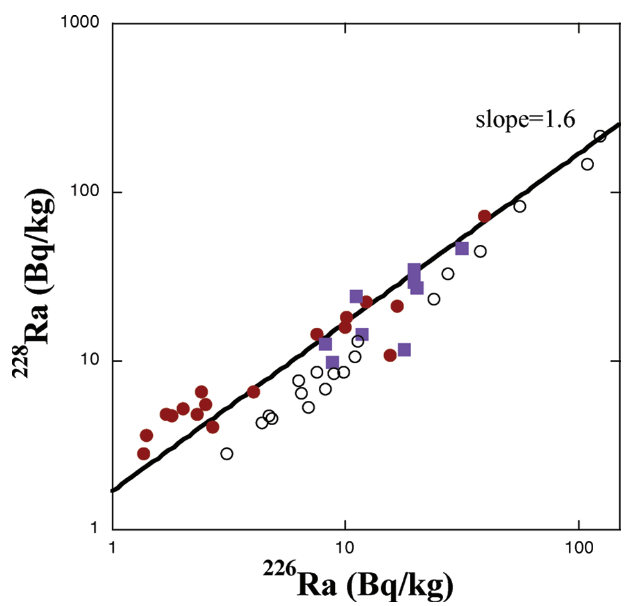

FIGURE 4. ^{228}Ra vs ^{226}Ra activities (in logarithmic scale) of Lower Cretaceous Nubian sandstone rocks in the Negev, Israel (brown circles) and Paleozoic Disi sandstone in southern Jordan (purple squares) and compiled data of worldwide sandstone (open circles, data from ref *24*). Each of the worldwide sand data points represents a mean value of sandstone rocks from different basins. The radium activities and the ^{228}Ra/^{226}Ra ratios measured in this study are consistent with the worldwide sandstone composition and indicate a uniform ^{228}Ra/^{226}Ra slope of ~1.6 in sandstone rocks.

^{226}Ra ratios in the groundwater (Table 1) are higher than the expected ratios in the aquifer rocks (1 and 0.046, respectively).

The rapid adsorption of Ra onto clay minerals and oxides (minutes to days) (*25*) infers higher abundances of the short-lived isotopes (^{223}Ra and ^{224}Ra) relative to the long-lived isotopes (^{228}Ra and ^{226}Ra), but the ^{228}Ra/^{226}Ra ratio is expected

TABLE 2. Compiled Data of Published Mean TDS (mg/L) and Radon and Radium Activity Values (mBq/L) of Groundwater from Sandstone Aquifers[a]

source[b]	location	TDS	^{222}Rn activity	^{226}Ra activity	^{228}Ra activity	^{228}Ra/^{226}Ra activity ratio
1	Long Island, NY	63 (9)	3183 (8)	0.57 (9)	1.15 (9)	1.83
2	Ojo Alamo aquifer, New Mexico	632 (11)	4283 (9)	3.55 (8)	11.5 (8)	2.96
3	Kombolge Sandstone, Australia			95.5 (16)	187.7 (16)	1.67
4	Cambrian-Ordovican aquifer, eastern Wisconsin	511 (7)	9700 (7)	75.8 (7)	45.5 (7)	0.73
5	unconfined Cambrian-Ordovican, southeastern Wisconsin	425 (7)		109.8 (7)	87.3 (7)	0.97
5	Confined Cambrian-Ordovican, southeastern Wisconsin	907 (7)		164.8 (6)	238.3 (6)	1.46
6	Mt. Simon, Minnesota	632 (10)	11066 (7)	200.0 (9)	268.3 (10)	1.60
7	Nubian sandstone aquifer, Negev, Israel	2564 (31)	1946 (22)	263.2 (35)	420.0 (25)	1.57
8	Bahariya Oasis, western Egypt	195 (4)		700 (3)		
9	unconfined Rum Group, Jordan	329 (13)	11415 (7)	534.4 (13)	906.0 (13)	1.90
9	confined Rum Group, Jordan	240 (13)	6272 (9)	678.0 (13)	1990.6 (13)	2.93

[a] Values in parentheses correspond to the number of samples in the different aquifers. [b] Data sources: (1) ref *29*, (2) ref *30*, (3) ref *48*, (4) ref *49*, (5) ref *50* (6) ref *51*, (7) ref , (8) ref *38*, (9) this study.

to mimic its parent ^{232}Th/^{230}Th ratio in the aquifer solids (*28−30*). Our data show (Figure 3) that the ^{228}Ra/^{226}Ra activity ratios in groundwater from the unconfined Rum Group, Khreim Group, and central Jordan are consistent with the ^{228}Ra/^{226}Ra ratio measured in the host aquifer rocks ($\sim$1.6), but the ^{228}Ra/^{226}Ra ratios in the confined Rum Group are higher (2.9). A higher ^{228}Ra/^{226}Ra ratio in groundwater from the confined Rum group could be derived from either local source rocks with a higher Th/U ratio (*28*) or differentiation in the rate of decay of the parent ^{232}Th and ^{230}Th nuclides on old surface coatings that would result in excess ^{232}Th over ^{230}Th in the solids and thus a higher ^{228}Ra/^{226}Ra ratio in groundwater (*29, 30*).

While Ra mobilization from the rocks seems to be controlled by recoil, which is a physical process, Ra adsorption depends on the water chemistry and decreases with salinity, acidity, temperature, and reducing conditions (*26−28*). Consequently, saline (*31−33*), reduced (*34*), acidic (*35*), and thermal (*36*) waters have typically high levels of Ra activities. The high Ra in the Disi aquifer in central Jordan can be related to the high salinity and reducing conditions (Table S1, Supporting Information), but the high Ra activity in the Rum Group, which is associated with low-saline, neutral-pH, and oxygenated groundwater (Tables 1 and S1) is not consistent with this conceptual model. Calculation of Ra species distribution in the groundwater indicates that most of the dissolved radium in the low-saline water is in the form of Ra^{2+} ($\sim$90%) and only a small fraction ($\sim$10%) is in the form of $RaSO_4^0$ species (Table S2, Supporting Information). Groundwater from the Rum Group is also largely unsaturated with respect to barite mineral (Table S2). Consequently, the high Ra activity in the Rum Group is not derived from reducing conditions, salinity effect, low pH, formation of $RaSO_4^0$ species that would not be adsorbed onto the aquifer rocks, or dissolution of Ra-rich barite mineral (which would reduce the short-to-long Ra isotope ratios). Moreover, the high Ra content in the Disi aquifer cannot be explained by anomalous Ra content in the host aquifer rocks, as we show that Ra activity and ^{228}Ra/^{226}Ra ratios in the Disi sandstone rocks are not different from those of other worldwide sandstone rocks (Figure 4).

General Radium Occurrence in Sandstone Aquifers. Compilation of reported isotopic data of groundwater in sandstone aquifers from different parts of the world and the results of the present study show that typically radon activities in groundwater have a narrow range, while the Ra activities are significantly lower and vary by several orders of magnitude in different aquifers (Table 2). Since radon in groundwater is derived primarily from recoil from its parent ^{226}Ra on the aquifer solids (*25−28*), the data shown in Table 2 indicate that radon emanation is relatively uniform in sandstone aquifers, regardless of the different mechanisms that generate radon in sandstone aquifers (*37*). This suggests uniform recoil of Ra isotopes, assuming that ^{226}Ra is in secular equilibrium with ^{230}Th in the aquifer solids. In contrast, the large variations in Ra activities, the range of ^{228}Ra/^{226}Ra activity ratios (0.7−3) in groundwater from the sandstone aquifer, and the high ^{224}Ra/^{228}Ra and ^{223}Ra/^{226}Ra ratios reported in this study suggest that the significant variation of Ra in the different aquifers is due to adsorption. Since the compiled data (Table 2) and our study show that radium variation in sandstone aquifers is not associated with water salinity, we propose that the availability of surface adsorption sites, which depends on the clay content and oxides in the aquifer rocks, is another factor that controls Ra activity in sandstone aquifers. In spite of the higher salinity and reducing conditions of groundwater in the Khreim Group, the Ra activity is lower, which suggests a higher clay content that would result in more effective Ra retardation. In contrast, the high permeability of the Rum Group infers a lower clay content (*18*) and less potential adsorption sites and, consequently, lower salinity but higher Ra activity. The results of the present study suggest that Ra may occur more frequently in fresh groundwater hosted by sandstone aquifers and that the limited available database (Table 2) may be biased.

Implications for Utilization of the Fossil Groundwater from the Nubian Sandstone Aquifer in the Middle East. Since most of the fossil groundwater in the Middle East is tapped from sandstone aquifers with hydrologeologic properties similar to those of the Disi aquifer (*5−7*), we hypothesize that some of the utilized groundwater has similar high Ra levels. Results from low-saline groundwater from a similar Nubian sandstone aquifer in Egypt (Bahariya Oasis) (*38*) and brackish groundwater from the Nubian sandstone aquifer in the Negev and Arava Valley in Israel (*39*) also show high Ra activities (Table 2) that exceed the international drinking water standards. Monitoring the Ra activity in groundwater extracted from the Nubian sandstone basins is therefore essential for evaluating the magnitude of the radioactivity impact on water quality in the region.

A study in New Jersey (*11*) has shown that the bone cancer incidence rate increased by 90% for individuals exposed to 0.185 Bq/L relative to background levels for combined ^{226}Ra and ^{228}Ra. Given that the average combined ^{226}Ra and ^{228}Ra activities in the unconfined and confined zones of the Rum Group are respectively 9 and 18 times higher, the cancer rate upon long-term consumption of this water is expected to be significantly higher. It is important to note that the preferential enrichment of ^{228}Ra over ^{226}Ra in the groundwater induces higher health risk, given the relatively short half-life of ^{228}Ra (*40*). In addition to the direct health implications for consuming drinking water with high Ra content, Ra in irrigation water could result in accumulation of Ra in the soil and some agricultural products (*41, 42*). Moreover, use of Ra-rich water for fish farming, which could be an ideal use of the brackish water from the Nubian sandstone (e.g., the Negev, Israel), would also be problematic given the high uptake of Ra by fish (*43, 44*). Future utilization of groundwater from the Disi aquifer and similar sandstone basins in the region, thus, requires a significant reduction of radionuclide levels. Compliance with the international drinking water standards can be achieved by sufficient blending with other Ra-poor water sources (e.g., the Khreim Group) and/or by treatment. The best available technologies for Ra removal are ion exchange, reverse osmosis desalination, and lime softening (*45, 46*). Each of these treatment technologies produces solid residuals (e.g., spent resins, membranes) and liquid residuals (e.g., brines, backwash water) that would be enriched in radionuclides and would require adequate disposal for suitable low-level radioactive waste (*47*).

This study has revealed high levels of Ra in groundwater from the Disi sandstone aquifer, which is considered the future drinking water resource in Jordan and other countries in the region. The high Ra content largely exceeds the international drinking water standards and poses a health risk upon long-term utilization. High Ra occurs in low-saline, neutral-pH, and oxygenated groundwater, which contradicts previous assumptions that high Ra would occur primarily in reduced, acidic, and/or saline groundwater. Compiled Ra data from other sandstone aquifers confirms the lack of correlation of Ra with salinity. On the basis of the relative distribution of the four Ra isotopes, this study provides a framework for interpreting the occurrence of Ra in sandstone aquifers. The study proposes that Ra in groundwater from sandstone aquifers is derived primarily from recoil from the parent nuclides in the aquifer solids (sandstone rocks, surface coating) and Ra adsorption on clay minerals and oxides. The relationship between uniform recoil of Ra isotopes and differential retardation is attributed to the geological properties of the aquifers; an aquifer with a higher content of clay minerals and oxides would provide more adsorption sites, which would enhance Ra retardation. In contrast, in highly conductive aquifers, such as the Nubian sandstone aquifers, the balance between the uniform recoil contribution of Ra from the aquifer solids and limited retardation due to the relatively low adsorption sites would generate Ra-rich groundwater. Future research should validate this hypothesis and expand the limited database on Ra isotopes in sandstone aquifers.

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Supporting Information Available

Description of the analytical techniques for the measurement of radium isotopes in water and rock samples. This material is available free of charge via the Internet at http://pubs.acs.org.

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