#### REPORT



# Present and paleo-hydrogeological reconstruction of a complex deep groundwater system in a tectonically active region (Golan Heights, Middle East)

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#### Abstract

The geochemical and isotopic composition of deep groundwater in sedimentary aquitards reveals a complex paleo-hydrological system affected by intensive tectonic activity. Water samples collected from deep research boreholes in the Golan Heights (Middle East) exhibit a unique combination of high salinity (>2,000 mg/L Cl) with low Na/Cl (<0.7) and Mg/Ca (<0.3) equivalent ratios, calcium chloride water type [Ca > (HCO<sub>3</sub> + SO<sub>4</sub>)], relatively low  $\delta^{18}O_{VSMOW}$  and  $\delta^{2}H_{VSMOW}$  values (-7 and -42%, respectively), and enriched <sup>87</sup>Sr/<sup>86</sup>Sr ratios compared to the host rocks. The salinity source is related to ancient lagoonary hypersaline brines (10–5 Ma) that existed along the Dead Sea Rift (DSR). These brines intruded into the rocks surrounding the DSR and, based on the current study, also extended away from the rift. Following their subsurface intrusion, the brines have been gradually diluted by <sup>18</sup>O- and <sup>2</sup>H-depleted freshwater recharged at high elevations, nowadays leaving only traces of the brines that originally intruded. It is also shown that variable hydraulic conductivities in different formations control the dilution rates and subsequently the preservation of the entrapped brines. A paleo-hydrological reconstruction is provided to demonstrate intrusion and backflow dynamics and also the relationship to freshwater dilution, which was triggered by a tectonically active basin of the nearby continental DSR. Brines that initially migrated from the rift have since been gradually flushed back to the rift through the current natural outlets. As the system discharges, it mixes and converges with a separate hydrogeological system, while still preserving some of the geochemical signals of the ancient brines.

Keywords Paleohydrology · Saltwater-freshwater interactions · Deep groundwater · Ca-chloride brines · Golan Heights

#### Introduction

Sedimentary basins often contain saline fluids in their deepest parts (Davisson and Criss 1996; Dickey 1969; Kreitler 1989). In some cases, saline fluids can persist in

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deep hydrogeological systems over millions of years, even in the presence of active regional groundwater flow (Bethke and Marshak 1990; Ferguson et al. 2018). Saline fluids exhibit different compositions depending on their origin and water-rock interaction processes. Some are remnants of water that was trapped in the pores during deposition (connate water), while others intruded later into the pore space. Saline intrusion can originate from various surface saltwater bodies (seawater, lagoons, salt lakes, etc.), or subsurface saline water bodies, which could be sourced from deeper strata and/or dissolution of highly soluble minerals (Bozau et al. 2015; Gaupp et al. 2008; Mirzavand et al. 2020). Thus, understanding the genesis of the saline fluids in the deeper parts of sedimentary basins can provide insights into the hydrological and chemical processes that have taken place in the subsurface, as well as the geological settings that enabled their intrusion and preservation in the system (Hanor 1994; Kharaka et al. 2007; Land 1995).

Deep formation waters in sedimentary basins are commonly saline and characterized by a Ca-chloride composition where Ca<sup>2+</sup> concentrations are higher than the sum of (HCO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) concentrations (meq/L), while Na/ Cl ratios are lower than the marine value (0.86) (Carpenter 1978; Hanor 1994; Starinsky 1974). Therefore, the chemical composition of Ca-chloride water is distinguishably different from plain seawater, or evaporated seawater. Saline Cachloride waters are found around the world in sedimentary basins (Bozau et al. 2015; Maghrebi et al. 2021), rift zones (Hardie 1983; Klein-BenDavid et al. 2004; Starinsky and Katz 2014), oilfields (Carpenter 1978; Egeberg and Aagaard 1989; Hanor and McIntosh 2007; Land 1995; Stueber and Walter 1991) and hydrothermal systems (Aulstead and Spencer 1985).

Previous studies (Carpenter 1978; Starinsky 1974) proposed that saline Ca-chloride brines in sedimentary basins originate from seawater that underwent evaporation (i.e., evaporation by a factor of  $\geq 10$ ) and/or mineral precipitation (aragonite, gypsum, halite), followed by water-rock interactions such as epigenetic dolomitization and gypsum formation (Ilani et al. 1988; Shelton et al. 2009; Stein et al. 2002). The evaporation produces highly dense hypersaline brines that intrude by density-driven migration into the subsurface (Babel and Schreiber 2014; Katz and Starinsky 2009; Stanislavsky and Gvirtzman 1999). Once the brines have intruded the subsurface, they can remain stagnant in the deep parts of the sedimentary basins for very long periods, up to tens of millions of years (Hanor 1994; Lowenstein et al. 2003). Alternatively, if conditions allow, they can be diluted or flushed by a different water source (Aulstead and Spencer 1985; Ferguson et al. 2018; Kim et al. 2022; Spencer 1987). The timescale of flushing large sedimentary basins can be in the order of millions of years (Darrah et al. 2015; Deming and Nunn 1991).

Hydrogeological and geochemical data collected from 4 new research boreholes (Ness wells, Fig. 1) provided information regarding the composition and sources of previously unexplored deep-seated aquitard sedimentary units. The water in those units contained traces of Ca-chloride brines, which were diluted by freshwater. Based on the data, the goal of the study was to reconstruct the paleo-hydrogeological history and determine the present conditions of these deep groundwater systems as affected by a tectonically active continental rift (Dead Sea Transform).

#### Study area

#### Geological background

The Golan Heights (GH), located in the Middle East (Fig. 1a), is a sedimentary syncline that was covered

during the Pliocene–Pleistocene by basaltic flows (Fig. 1b; Mor 1993; Sneh and Weinberger 2003). The sedimentary sequence underlying the basalts is exposed only at the northern and southern margins of the syncline, at the slopes of the Hermon and Ajloun anticlines, respectively (Fig. 1b). The western margin of the syncline is the active tectonic plate boundary of the Dead Sea Rift (DSR), which separates between the Arabian (east) plate and the African/Sinai (west) plate/subplate (Freund et al. 1968; Salamon et al. 2003; Shulman et al. 2004). The activity of the Dead Sea Rift transform resulted in a total left lateral displacement of 105 km between the plates (Quennell 1956). Activity over the past 25 Ma resulted in a 35-40-km shift (Freund et al. 1968; Walley 1998). The transform also branched eastwards towards the Golan area into several divergent faults, such as the Shamir and Sheikh Ali faults (Fig. 1b), which led to the discontinuation of the syncline structure and to the exhumation of the uplifted structure of the Hermon Anticline (Garfunkel 1981; Meiler et al. 2011; Ron et al. 1984).

The sedimentary section in the study area (Figs. 1, 2 and 3) consists of a thick Jurassic section (Arad Group), composed mostly of limestones, which constitutes most of the Hermon exposures. Units from the Cretaceous are composed of sandstones (Kurnub Group, Lower Cretaceous), dolomites and limestones (Judea Group, Upper Cretaceous; Michelson 1979; Sneh et al. 1998). Overlying the Judea Group is the Senonian Mt. Scopus Group. The base of the Mt. Scopus Group (Menuha Formation) is composed of massive white chalk (Mimran et al. 1985). The overlying Mishash Formation consists of chalk-chert alterations at its lower part (Margane et al. 2002; Mimran et al. 1985). The Upper Mishash and Ghareb formations are mostly inseparable and are composed of chalk. The Senonian rocks are considered to be extremely organic rich Type II-S source rocks that have partially matured from a paleo-thermal event (Reznik and Bartov 2021) and currently contain viscous immovable bituminous material (Rosenberg et al. 2017; Rosenberg and Reznik 2021). The uppermost Tagiye Formation of the Mt. Scopus Group is distinguishable from the underlying sequence, since it is consistently composed of marly chalk and lacks organic matter (Flexer and Honigstein 1984; Margane et al. 2002; Mimran et al. 1985).

Though the Mt. Scopus Group is barely exposed at the surface in the GH north of the Lower Yarmouk Gorge (LYG), it is widely exposed to the south of the Yarmouk, at the Ajloun anticline (Fig. 1b). Overlying the Mt. Scopus Group are the Eocene chalks (Avedat Group) and the Neogene conglomerates and clay (Hordos Formation), representing a shift from a marine to a terrestrial environment (Sneh 1996). The top of the sedimentary section of the GH is marked by thick basaltic flows from the Pliocene–Pleistocene (Bashan Group). Most of the surface of the southern GH is covered by the basaltic flows from the Pliocene (5–3



**Fig. 1** a General location of the study area, modified after UN (2023), and **b** locations of water sources on top of a geological map (New Israel Grid Coordinates). Colors correspond to the stratigraphic table (**c**). The main outcrop of the Mt. Scopus and Judea groups at the foothills of Mt. Hermon are highlighted in yellow (Cretaceous outcrops).

Dashed lines (A–A' and B-B') mark the location of the cross sections in Figs. 2 and 3, respectively. DSR F - Dead Sea Rift Fault; HG -Hammat Gader; LYG - Lower Yarmouk Gorge. **b** is modified after Sneh et al. (1998) and **c** (stratigraphic table) is modified after Fleischer (2002). The water source locations are listed in Appendix 1 and 2



**Fig. 2** Schematic cross section through the Golan Heights (GH) from Mt. Hermon in the north to the Ajloun anticline in the south—A-A' in Fig. 1, after Burg and Gev (2019). Depths and locations of Ness and Meizar boreholes are indicated by light blue lines



**Fig. 3** Stratigraphic cross section through the studied boreholes in the GH including the Ajloun, northern Jordan (Fig. 1, B–B'). Depths of the drill stem tests (DSTs) are highlighted in orange. Open sections are highlighted in light blue for the Mt. Scopus Group and light green for the Judea Group. The same colors are used for water levels (triangles) and apparent hydraulic gradients (dashed line). Empty triangles mark the current water level in Meizar, which declined dramatically

Ma), known as the Cover Basalt (Heimann et al. 1996). In the northern GH and at the Yarmouk Gorge, younger basaltic flows that originated from local eruptions during the Pleistocene overlay the Cover Basalt (Mor 1993).

due to production at the LYG since its initial operation. According to the hydrogeological classification in Jordan, parts of the Mt. Scopus and the Judea groups are considered as one aquifer (A7/B2). The Ajloun stratigraphic section and water level (blue-green triangle) for the A7/B2 aquifer were compiled from Margane et al. (2002). Arrow thicknesses correspond with the relative contribution to Meizar wells, estimated by geochemical interpretation

#### Hydrogeological background

The study area is located at the margins of two hydrogeological systems with the following main flow directions: (1) The northern Golan Heights system that flows from the flanks of Mt. Hermon towards the Yarmouk (Arad and Bein 1986; Roded et al. 2013; Starinsky et al. 1979) and (2) The southern Ajloun system that flows from Ajloun to the Yarmouk (Bajjali et al. 1997; Margane et al. 2002).

The northern hydrological system of the GH includes two known and relatively well characterized regional aquifers: the Jurassic aquifer (Burg and Gev 2019) and the Basaltic aquifer (Dafny et al. 2003), which are recharged by direct precipitation through their respective outcrops (Fig. 1b). Other hydrogeological units (Kurnub, Judea, Mt. Scopus and Avedat groups) were not thoroughly studied. Further, these units were assumed to be hydrologically inactive mainly due to their limited outcrop area and/or low hydraulic conductivities (Roded et al. 2013; Tahal 1989).

The main southern hydrogeological systems are situated in the geological units that correlate to the Mt. Scopus and upper Judea groups—labeled B2 and A7, respectively, according to the Jordanian stratigraphic division (Margane et al. 2002; Siebert et al. 2014). These units are hydrogeologically active since they are widely exposed at the Ajloun anticline (Fig. 1b) and are readily available for direct recharge by precipitation (Margane et al. 2002), in contrast with the northern GH systems where the outcrops of these units are limited (Fig. 1).

In the LYG (Fig. 1b) the GH and Ajloun hydrogeological systems discharge through the thermal springs of Hammat Gader and Mukheiba (45.5 MCM year<sup>-1</sup>, <50 °C; Levitte et al. 1978). In this area, a series of Jordanian and Israeli production wells (e.g., Meizar, Mukheiba and JRV) have produced water since the 1980s from these systems (Bajjali et al. 1997; Margane et al. 1999; Rosenthal et al. 2020). Previous geochemical and numerical modelling (Arad and Bein 1986; Starinsky et al. 1979) suggested the water in the LYG represents a mixture of the two hydrological systems (GH and Ajloun). However, the water composition flowing to the LYG from the GH and the contributing units remained unknown due to a lack of deep boreholes.

#### **Materials and methods**

#### Sample collection

Water samples from the Ness boreholes were collected using three principal methods with the first being high-pressure injection of air (airlift) from open-hole sections during drilling intermissions. The airlift samples were collected during the drilling operation of Ness 02 and 12 (Fig. 3). As these wells were drilled underbalanced using foam/air, drilling induced little contamination. Regardless, to avoid contamination, the accumulated water column in the wells was completely removed at least once prior to sampling. The second method was bottom hole samples (BHS) collected from specific depths, and the third comprised drill stem tests (DSTs) that targeted eight different 15-m intervals within the studied formations (Fig. 3).

Production of fluids during the DSTs was performed either by mechanical swabbing or by N<sub>2</sub> lift. The samples that were collected during the DSTs were closely monitored to ensure minimum contamination of drilling and completion fluids as much as possible. This was done by continuously monitoring the produced water using an electrical conductivity (EC) probe (WTW, 1970i) until a stable water composition (<5% change) was attained. From each tested zone, at least three representative samples were selected, each collected at slightly different times during the production. The representative samples selected from each zone were analyzed for detailed chemical analysis and their results were averaged. To avoid any remaining contamination by completion fluids that were used in the DSTs (predominantly KCl), in the relevant cases, the equivalent concentrations of K<sup>+</sup> were subtracted from the Cl<sup>-</sup> concentrations: [Cl<sup>-</sup>]<sub>corrected</sub>  $(meq) = [Cl^{-}] (meq) - [K^{+}] (meq)$  (all values are presented in Table 1).

#### Water levels and hydraulic conductivities

Water levels of open sections were measured during intermissions in the drilling operations of Ness 02 and Ness 12 wells (Fig. 3). The levels were measured following the complete removal of the fluids present in the well and a buildup period of 48 h to ensure the water level represents hydrostatic conditions. The apparent hydraulic conductivity of each DST interval was calculated using pressure transient analysis (PTA) of buildup periods (Satter et al. 2008), based on bottom hole pressure readings (Quartz, Spartek Systems).

#### **Chemical and isotopic analyses**

Major cations and sulfate (Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Sr<sup>2+</sup>,  $SO_4^{2-}$ ) were measured by inductively coupled plasma optical emission spectrometry (ICP-OES, Optima 3300, Perkin Elmer,  $\pm 0.5\%$ ). The sum of Cl<sup>-</sup> and Br<sup>-</sup> was measured using automated potentiometric titration with AgNO<sub>3</sub> (702 SM Titrino, Methorn,  $\pm 0.5\%$ ). The concentration of Br<sup>-</sup> was measured using ion chromatography (IC, ICS-200, Dionex,  $\pm 2\%$ ). Br<sup>-</sup> concentration was subtracted from the sum to determine the Cl<sup>-</sup> concentration  $([Cl<sup>-</sup>] = [Cl<sup>-</sup> + Br<sup>-</sup>] - [Br<sup>-</sup>] \pm 3\%)$ . Alkalinity was measured either by automatic titration (785 DMP Titrino, Methorn) or manually using a pH meter with a 0.05 N HCl acid for titration down to a pH of ~4.5. Stable oxygen (<sup>18</sup>O) and hydrogen (<sup>2</sup>H) isotopes were analyzed using an isotope ratio mass spectrometer-IRMS, Delta plus XP, Thermo Finnigan;  $\delta^{18}O(\pm 0.1\%)$  and  $\delta^{2}H(\pm 1.0\%)$ —and were corrected using a Vienna Standard Mean Ocean Water (VSMOW) sample. Isotopic composition of Sr<sup>2+</sup> was measured using a multicollector inductively coupled plasma mass spectrometer (MC-ICPMS, Nu Plasma-I, Wrexham, UK). The calibration standard that was used for correcting <sup>87</sup>Sr/<sup>86</sup>Sr values was SRM-987 (55 ppb),  $2\sigma \le 1 \times 10^{-5}$ . All analyses were performed at the Geological Survey of Israel (GSI) laboratories.

#### Results

#### Water levels and apparent hydraulic conductivities

Figure 3 presents borehole sections and water levels in the Mt. Scopus and Judea groups derived from the Ness wells in the GH, along with compiled literature data from Ajloun, Jordan. Open section water level values in Ness 12 and Ness 02 (Mt. Scopus Group) were 19 and -22 m above mean sea level (AMSL), respectively (Fig. 3). Water level of the Upper Judea Group in Ness 12 was slightly higher than the Mt. Scopus Group at the same well (24 m AMSL). Shortly after the Meizar 3 well (Mt. Scopus Group) was drilled (1984), the water level was 74.8 m above ground level (-30.2 m AMSL). The water level in the same well during sample collection (2016) was lower by more than 60 m. at -93.4 m AMSL. In Meizar 2 (Judea Group) the 1984 and 2016 artesian groundwater levels were -42.3 and -83.1 m AMSL, respectively (Simon et al. (1997) and Israel Hydrological Service). In Ajloun, both the Upper Judea (A7) and the Lower Mt. Scopus Groups (B2) are considered as one hydrogeological unit, which serves as the main water source in northern Jordan (Margane et al. 1999). The water levels in the A7/B2 aquifer, 4 km southward of the LYG (Fig. 1b) are ~25 m AMSL (Fig. 3; Margane et al. 2002).

Apparent hydraulic conductivities calculated from the DSTs were the same  $(2 \times 10^{-7} \text{ m/s})$  for all intervals that included the Ghareb-Upper Mishash formations (GUMF). However, the overlying Taqiye Formation exhibited a hydraulic conductivity that was lower by two orders of magnitude  $(2 \times 10^{-9} \text{ m/s})$ . Values and depths are detailed in Appendix 1.

#### **Geochemical composition**

The salinities of the analyzed samples varied spatially, spanning over three orders of magnitude between different locations and depths in the Mt. Scopus Group (Table 1). Water collected from the Mt. Scopus Group in Ness 12 was fresh (<420 mg/L total dissolved solids, TDS). Similarly, the water collected from the Upper Judea Group in the same well was fresh (<700 mg/L TDS). The water collected during the DSTs in the Upper-Mishash and the Ghareb formations (GUMF) in Ness 02 and Ness 03 was moderately saline (5,000–6,300 mg/L TDS) and the water from the Taqiye Formation in Ness 02 was saline (~17,000 mg/L TDS). Water from the open section in Ness 02 (Fig. 3), which includes all the tested formations during the DST and additional contributing layers, was considerably less saline (2,800 mg/L TDS) than the DST samples in the same well (5,000–6,300 mg/L TDS).

Despite the range in salinities, the brackish to moderately saline water from the tested DST intervals had similar equivalent ratios (Table 2). Na/Cl ratios were lower than the marine value (<0.86). Mg/Ca ratios were low and with limited variation (0.2–0.3), and all samples exhibited a Cachloride composition. In contrast, the less saline water collected from the open sections in both Ness 12 (fresh) and Ness 02 (brackish) had higher Na/Cl (0.9–1.5) and Mg/Ca (0.7–0.9) ratios and was not Ca-chloride (Table 2). The electrical charge balance in all the samples that were analyzed did not exceed  $\pm 3\%$ .

#### **Isotopic composition**

The  $\delta^{18}$ O and  $\delta^2$ H (VSMOW) of the studied water are presented in Table 2. Freshwater samples from both the Mt. Scopus and Upper Judea groups in Ness 12 showed depleted  $\delta^{18}$ O and  $\delta^2$ H values ( $\leq -7.3$  and  $< -42\%_o$ , respectively). Brackish-moderately saline water from the Mt. Scopus Group in Ness 02 and Ness 03 had slightly less depleted  $\delta^{18}$ O and  $\delta^2$ H values ( $\leq -7$  and  $< -40\%_o$ , respectively). Saline water from the Taqiye Formation in Ness 02 had significantly higher  $\delta^{18}$ O and  $\delta^2$ H values of -6 and  $-36\%_o$ , respectively.

 ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  values of the studied water from the southern wells (Ness 02 and 03) were mostly similar (Table 2).  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  values of the water from the Mt. Scopus Group were 0.70766–0.70782. The freshwater from the northern well (Ness 12) had significantly lower  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  values (0.70746 in the Mt. Scopus Group and 0.70737 in the Judea Group).

#### Discussion

#### **Regional hydraulic gradients**

Calculation of preproduction hydraulic gradients based on the water level measurements highlight the differences between the southward flowing GH and northern flowing Ajloun systems: firstly, in the GH, the gradients from Ness 12 to Meizar in the Mt. Scopus and Judea groups are moderate (1.7 and 2.3‰, respectively). Despite the similar gradients, the water levels in the Mt. Scopus and Judea groups in both the Ness 12 and Meizar wells are different, indicating that the water systems are isolated. Secondly, in the

			5		, ,		+21 +	Ca <sup>24</sup>	- Ma <sup>2+</sup>	Sr <sup>2+</sup> CJ	ر ریستان 			
						Na	4		SIVI			- Br <sup>-</sup> S	$D_4^{2-}$ HCO	- TDS
Vess 02	Ghareb-Upper Mishash boundary	DST- avg. of thre	æ samples	26	6/04/2016	6.7 824	4 422	: 757	87.0	18.2 2,	921 2,538	10.5 3	1.9 479	5,391
Vess 02	Ghareb	DST- avg. of fou	r samples	10	)/05/2016	6.3 90	3 55	3 433	68.6	12.9 2,	451 1,950	23.7 5	1.9 607	5,090
Vess 03	Ghareb	DST- avg. of thr	e samples	18	3/03/2016	6.7 1,1	89 34	784	85.0	22.4 3,-	433 3,123	18.6 28	3.6 552	6,270
Vess 02	Taqiye	DST- avg. of three	æ samples	15	0/05/2016	7.3 4,4	89 16	127	1 169	43.4 9,	- 196	55.7 4(	).2 548	16,740
Vess 02	Lower Tagiye-Lower Mishash	Airlift		02	2/07/2016	8.3 717	7 46.	0 101	53.0	8.80 1,	240 -	8.10 7.	00 565	2,746
Vess 12	Lower Tagiye-Lower Mishash	Airlift		21	/12/2015	8.1 76.	0 8.0	0 27.0	14.0	1.28 90	- 0.	0.50 13	3.0 187	417
Ness 12	Upper Judea Gr (Bina-Dir Hana)	Airlift and BHS-	avg. of five s	amples M	arch-May 202	0 8.0 10	9 13.	0 59.5	24.5	2.27 11	- 0	, ,	79 190	687
<b>3orehole</b>	Formation	Equivale	int ratios					δ <sup>18</sup> C	(	S <sup>2</sup>	H	d-exce	(200) SSS	<sup>87</sup> Sr/ <sup>86</sup> S
		Na/CI	Mg/Ca	Mg/Cl	Ca/Cl	Ca/(HCO	$O_3 + SO_4$	) (%	VOMOV	() ()	se VSMOW)			
Vess 02	Ghareb-Upper Mishash bound	ary 0.50	0.19	0.10	0.53	4.70		-7.4	5	1	:0.7	18.9		0.7077
Vess 02	Ghareb	0.71	0.26	0.10	0.40	2.24		-7.2	4	4	1.0	17.0		0.70773
Vess 03	Ghareb	0.59	0.18	0.08	0.44	4.04		-6.9	6	4	2.1	13.8		0.70766
Vess 02	Taqiye	0.69	0.22	0.05	0.23	6.48		-6.0	1	Ч,	6.3	11.8		0.70782
Vess 02	Lower Taqiye-Lower Mishash	0.89	0.87	0.12	0.14	0.54		-7.2	0	4	2.4	15.2		0.70784
Vess 12	Lower Taqiye-Lower Mishash	1.30	0.86	0.45	0.53	0.40		-7.2	9	4	2.0	16.0		0.70746
Vess 12	Upper Judea Group	1.53	0.68	,	0.96	0.43		-7.4	Ľ	4	4.40	15.4		0.70737

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Ajloun system, where the Mt. Scopus and Judea groups are considered hydraulically connected (Margane et al. 2002) significantly steeper gradients were calculated towards the Meizar wells (14‰ in Mt. Scopus Group towards Meizar 3 and 17‰ in the Judea Group towards Meizar 2).

Following the intensive water production that took place in the Meizar wells in the past 40 years, the hydraulic gradients towards the Meizar wells have steepened to ~10 and 30% for the GH and Ajloun systems, respectively. Assuming that similar hydraulic conductivities are present north and south of the Yarmuk, the significant gradient differences (either pre- or post-production) suggest faster natural flow rates from the Ajloun system compared to the GH system.

#### Chemical composition and sources of salinity

The Cl<sup>-</sup> concentrations in the water from the DSTs performed in Ness 02 and Ness 03 wells (Fig. 1b) are between 2,400 and 10,000 mg/L. The highest Cl<sup>-</sup> concentration was found in the water from the Taqiye Formation (~10,000 mg/L), which is also higher than any concentration previously recorded in GH groundwater. Along with high Cl<sup>-</sup> concentrations, brackish-saline water in the Mt. Scopus Group exhibits Na/Cl and Mg/Ca equivalent ionic ratios that are lower than the marine values with a Ca-chloride composition (Table 2). While remnants of seawater can explain such salinities, the ionic ratios of the samples are not similar to those of seawater (Fig. 5). Further, water-rock interactions between meteoric water and host rock cannot explain the high Na<sup>+</sup> and Cl<sup>-</sup> concentrations. Halite dissolution which could increase the Na<sup>+</sup> and Cl<sup>-</sup> concentrations is also overruled as this would have resulted in Na/Cl values of 1 (Carpenter 1978; Kreitler 1989). Therefore, the composition of the studied water cannot be explained without an additional salinity source that formed separately from the host rock in which it is found today.

Waters with Ca-chloride compositions are abundant along the Dead Sea Rift (DSR) Valley, including the modern Dead Sea as well as in ancient brines that intruded groundwater systems around the rift. This includes the subsurface surrounding Lake Kinneret (Klein-BenDavid et al. 2004; Mazor and Mero 1969; Starinsky and Katz 2014) where Ca-chloride saline waters (termed as the Kinneret brines) are found also with Cl<sup>-</sup> concentrations in the range of 1,000–25,000 mg/L and low Na/Cl ratios. Furthermore, a few kilometers north of Lake Kinneret, a hypersaline water body was found in the Rosh Pina-1 well (Fig. 1b) with a Cl<sup>-</sup> concentration of more than 100,000 mg/L. The Kinneret brines and Rosh Pina Brine (RPB) were previously defined as remnants of the DSR brines (Starinsky 1974; Starinsky and Katz 2014). The DSR brines evolved from seawater that transgressed during the Neogene through Yizra'el Valley (Fig. 4a) into the tectonic depression that formed along the DSR to form a lagoon (Lagoonary stage; Neev 1967; Zak 1997; Zak and Bentor 1972). The seawater underwent evaporation which led to an increase in salinity and to the precipitation of aragonite, gypsum and halite (decrease in Na/Cl and increase in Mg/Ca). The increase in salinity was accompanied by an increase in density which resulted in a density-driven migration into the surrounding rocks (Stanislavsky and Gvirtzman 1999).

Following the intrusion, at the subsurface, the brines interacted with carbonates to form dolomite. The dolomitization process increased Ca<sup>2+</sup> concentrations, which led to gypsum precipitation and a subsequent decrease in  $SO_4^{2-}$  concentrations. The combination between the dolomitization and mineral precipitation resulted in a decrease in Mg/Ca and increase in  $Ca^{2+}/(HCO_3^{-}+SO_4^{2-})$ , which led to the Ca-chloride composition. Lagoon level drops led to the backflow of previously intruded brines while gradually decreasing the Mg/Ca ratio. Following the terminal disconnection of the lagoon from the sea (lacustrine stage; Neev 1967; Stein 2014), inflow of bicarbonate-rich freshwater led to aragonite precipitation (increase in Mg/Ca). The geochemical evolution of the DSR brines enabled division of the saline water found in the DSR into two groups (Katz and Starinsky 2009): (1) brines from the lagoonary stage, with Mg/Ca ratios lower than 1, and (2) brines from the lacustrine stage, with Mg/Ca higher than 1 (Fig. 4b).

In the south of Israel, brines from the earlier lagoonary stage were found at a distance of up to 90 km from the rift, while brines from the later lacustrine stage were only found within the DSR (Fig. 4a; Katz and Starinsky 2009; Stein 2001). In northern Israel, lagoonary brines are found in springs west of Lake Kinneret, while lacustrine brines are found east of Lake Kinneret in the shallow alluvial basin fill units (Fig. 4a, empty purple triangle). The RPB hypersaline brine was found in the research well Rosh Pina-1, located in the Hula Valley, north of Kinneret Lake. The RBP was found at a depth of 3,800 m in Jurassic limestones. The limestones in which the RPB was found are a part of a block within the DSR, which is separated from its surroundings by deep faults (Heimann and Ron 1993).

To examine the relationship between the studied water from the southern GH and surrounding saline water sources, cation concentrations were plotted vs. Cl<sup>-</sup> concentration (Fig. 5a–e). Detailed chemical compositions are presented in Appendix 2. Also plotted, are RPB-freshwater and seawater–freshwater mixing lines. The freshwater endmember composition selected is the water found upstream in Ness 12. Note that due to extreme salinity differences between hypersaline brines and freshwater, even significant



Fig. 4 a Distribution of marine transgression according to evaporitic deposits (gray polygon) and intruding DSR brines found at the subsurface surrounding the rift (lagoonary and lacustrine brines in red and purple, respectively; modified after Zak and Bentor 1972); **b** Na/Cl vs. Mg/Ca of the DSR brines and the studied water (modified

brine dilution (>1% brine fraction) has little effect on the ionic ratios of the brines, thereby preserving the original ionic ratios of the ancient brines. Figure 5 indicates that the water found in the GUMF formation in the GH shares a common salinity source as the RPB. This means that a DSR brine with similar composition to the RPB intruded the GH subsurface.

Assuming that the RPB represents the closest brine composition to the brine that intruded the GH, the residual fraction of the hypersaline endmember can be calculated. Calculation of the RPB fraction based on Na<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup> and Cl<sup>-</sup> suggest that the water from the GUMF retains 2–3% of the saline fraction, while the remaining 97% are freshwater that diluted and flushed the original brine. The Na/Cl ratios in the water from the Taqiye Formation are slightly different from those in the GUMF, suggesting that the composition of the hypersaline brine that intruded it was slightly different. Thus, when taking the RPB as the hypersaline end member, the calculated volume fractions in the Taqiye have a larger

after Starinsky and Katz 2014). GH water chemical compositions are detailed in section 'Geochemical composition' (Tables 1 and 2). DSR brine compositions were reported in Bergelson et al. (1999) and Starinsky and Katz (2014). The compositions are detailed in Appendix 2

scatter: based on Na<sup>+</sup> and Cl<sup>-</sup>, the Taqiye contains 10–13% brine (with a composition of the RPB) and only 5–6% based on the Mg<sup>2+</sup> and Ca<sup>2+</sup> concentrations. The higher dilution degrees in the GUMF in comparison to the Taqiye Formation demonstrates the separation between the two units as well as their different hydraulic characteristics—i.e., less dilution is exhibited in the less-permeable Taqiye compared to the GUMF. Nevertheless, it should be noted that, despite the two orders of magnitude difference in the formation hydraulic conductivities, the saline endmember in the Taqiye has also been significantly diluted by freshwater (more than 87%).

#### Source of freshwater

In contrast with the dissolved ions that are mostly affected by the hypersaline brine endmember, the isotopic signal of the studied water should be governed by the freshwater component, as the samples are significantly diluted.



**Fig. 5** Major cations vs. Cl<sup>-</sup> equivalent concentrations for both the studied water and nearby saline water bodies in the GH region. Subplots in the right panel are enlargements of the subplots in the left panel. Theoretical mixing lines between freshwater and Rosh Pina Brine (RPB) were added. Additional mixing lines with seawater were added to the subplots in the right panel for comparison. The brine fraction (upper horizontal axis) was calculated based on the Cl<sup>-</sup> con-

The  $\delta^{18}$ O of the freshest and most upstream water sample is -7.3% (Ness 12, Fig. 1b). This value is similar to the  $\delta^{18}$ O values found in the deep aquifers that are recharged in Mt. Hermon and its elevated vicinity (Dafny et al. 2006; Kattan 1997). In contrast, the  $\delta^{18}$ O value of RPB is +3% (Gat et al. 1969), i.e. particularly enriched and indicative of water that underwent significant evaporation. Figure 6a

centration. The lagoonary brines west of the Kinneret show slight compositional differences compared to the RPB mixing line, while the lacustrine brines east of the Kinneret exhibit a completely different end-member composition, as also shown in Fig. 4. DSR brine compositions were reported in Bergelson et al. (1999) and Starinsky and Katz (2014). The compositions are detailed in Appendix 2

presents the  $\delta^{18}$ O values vs. Cl<sup>-</sup> concentrations of the studied water with a mixing line between the RPB and the freshwater found in Ness 12. The results show that the  $\delta^{18}$ O values of the water samples fall on the suggested mixing line, exhibiting increasing  $\delta^{18}$ O values with increasing salinity. Most notably, the least diluted water from the Taqiye Formation exhibit the highest  $\delta^{18}$ O values (-6.0%),



**Fig. 6** a  $\delta^{18}$ O and b  $\delta^{2}$ H values vs. Cl<sup>-</sup> concentrations in the studied water and nearby water bodies. c  $\delta^{2}$ H vs.  $\delta^{18}$ O values with global and local meteoric water lines (GMWL and LMWL, respectively). LMWL is based on Mt. Hermon and Syria precipitation values after Al Charideh and Abou Zakhem (2010). Also plotted are the mixing lines (black) between the Mt. Scopus Group freshwater endmember

and RPB, where dashed lines are mixing lines with estimated  $\delta^2 H$  value of the saline endmember. The brine fraction (upper horizontal axis) was calculated based on the Cl<sup>-</sup> concentrations. Cl<sup>-</sup> concentrations,  $\delta^2 H$  and  $\delta^{18}O$  of the northern DSR brines (triangles) were taken from Gat et al. (1969) and are fully detailed in Appendix 3

as it is slightly more affected by the heavier isotopic composition of the saline endmember.

Similarly, Fig. 6b presents  $\delta^2$ H values vs. Cl<sup>-</sup> concentrations of the studied water with a mixing line between the hypersaline and the freshwater endmembers (Ness 12, -42‰). Since the  $\delta^2$ H value of the RPB was not measured, the value was assumed to be +4.2‰, in accordance with the linear relationship between  $\delta^{18}$ O and  $\delta^2$ H found in the Dead Sea brine (Horita and Gat 1989). The  $\delta^2$ H values of the studied water also increase with salinity and conform with the mixing line, which reinforces the conclusion drawn based on  $\delta^{18}$ O.

The calculated RPB volume fraction, based on the mixing lines of both  $\delta^{18}$ O and  $\delta^{2}$ H, yielded a value of 12% in the water from the Taqiye Formation, which falls within the range defined by the dissolved ions (Na<sup>+</sup> and Cl<sup>-</sup>, 10–13%). Likewise, the brine volume fraction in the water from the GUMF was less than 3% (based on both  $\delta^{18}$ O and  $\delta^{2}$ H), which matched the mixing ratios derived based on the dissolved ions (2–3%). Thus, the isotopic compositions of the water further support the two end-member mixing model for the studied water in the Ness wells. It should further be noted that in contrast with the studied water in the GH, the Kinneret brines exhibit more enriched  $\delta^{18}$ O and  $\delta^{2}$ H compositions, indicating they were diluted by different freshwater sources (Fig. 6).

The isotopic composition of the freshwater endmember is very light (-7.3 and -42% for  $\delta^{18}$ O and  $\delta^{2}$ H, respectively). The closest such depleted isotopic compositions are found in rainwater that precipitate over elevations higher than 1,000 m AMSL, such as Mt. Hermon and its foothills (Fig. 1b), or the distant Palmyride Mountains (Dafny et al. 2006; Kattan 1997). The depleted  $\delta^2$ H values of the studied water  $(\sim -42\%)$ , resemble the values on the southern flanks of the Hermon Mt. (< -40%) and suggests the source of the freshwater is probably not from the distant slopes of the Palmyride Mountains, where  $\delta^2 H$  values are more enriched (> -35%) (Kattan 2021). The northern water source also conforms with the north-south modern regional hydraulic gradient found in the present study for the Mt. Scopus and Judea groups in the southern GH (Fig. 3). This also suggests that the dilution and flushing process is ongoing and therefore the light isotopic composition and brine remnants should be detected downstream.

# Lower Yarmouk Gorge outlets: water and salinity sources

The gradient and chemical and isotopic composition suggest that the Mt. Scopus and Judea groups are not inactive, as previously suggested, and may potentially serve as one of the contributing units to the LYG outlets. The salinities of the LYG springs are relatively low, yet their chemical composition suggests that their salinity source relates to the lagoonary stage of the DSR brines (Starinsky et al. 1979). To investigate the relationship between the northern GH systems, the southern Ajloun system and wells and springs located downstream, such as the Meizar wells and LYG spring outlets, the compositions of different water sources in the area were compiled and analyzed (Fig. 7).

Figure 7a shows high Na/Cl values in the freshwater samples that are not affected by the brine composition. With increasing salinity, however, the Na/Cl ratios decrease below the marine value due to a higher volume fraction of DSR brines remnants. The decrease in Na/Cl values with increasing salinity indicates that the source of salts in both the GH and the downstream LYG has a lower Na/Cl value (Fig. 7a). The isotopic composition shows two distinctly different freshwater sources: the relatively saline and isotopically light GH component and the fresh and isotopically heavier Ajloun component (-5 to -6.5%; Bajjali et al. 1997). The data of the LYG fall within a range defined by the Ajloun component and the GH component, which by itself is a mixing product of hypersaline brine and isotopically light freshwater. This mixing explains the increase in salinity with decreasing  $\delta^{18}$ O values observed in the LYG water (Fig. 7b, orange lines).

Based on this mixing pattern, the water in the Mt. Scopus Group in Meitzar 3 is mainly derived from the south (relatively enriched  $\delta^{18}$ O and low salinity; Fig. 7b, dark blue square), which is typical for rainwater that precipitated on outcrops of the Mt. Scopus Group at the Ajloun slopes. In contrast, the isotopic composition in the underlying Judea Group in Meitzar 2 indicate that the water is mainly derived from the GH system (depleted  $\delta^{18}$ O and relatively higher salinity; Fig. 7b, green squares).

 $^{87}$ Sr/ $^{86}$ Sr values add additional information regarding the source of the salt and the host rock the water interacted with. The isotopic  $^{87}$ Sr/ $^{86}$ Sr signature of the freshwater is largely acquired by water–rock interactions (Kendall and Doctor 2003) with the latter having the seawater composition at the time that they precipitated (Burke et al.

1982). However, when the isotopic signal of the water and the host rock differ significantly-the difference can be explained by an additional source of Sr, such as remnants of brine (Chaudhuri and Clauer 1993). In the case of the studied water from the GH, only the freshwater samples are congruent with the host rocks (Fig. 7c, Ness 12 and Meitzar 03). The rest of the brackish-saline samples (Ness 03, 02, Meitzar 02, LYG springs) show higher  $^{87}$ Sr/ $^{86}$ Sr isotopic values (0.7077–0.7079) with respect to the values of the Late Cretaceous rocks (0.7073–0.7077) (Burke et al. 1982; McArthur et al. 1994), excluding the data point of the Ghareb Formation in Ness 03 that falls slightly out of range. This is evident in both the Mt. Scopus Group and Judea Group waters that show a prominent increase in <sup>87</sup>Sr/<sup>86</sup>Sr along the flow path from Ness 12 (Fig. 7c). The increase in <sup>87</sup>Sr/<sup>86</sup>Sr is explained by the presence of lagoonary brines, with values as high as 0.708 (Starinsky and Katz 2014). Similarly to the  $\delta^{18}$ O mixing lines (Fig. 7c), the <sup>87</sup>Sr/<sup>86</sup>Sr values of the water samples as a function of salinity can be explained by mixing an enriched lagoonary brine with freshwater that inherited the <sup>87</sup>Sr/<sup>86</sup>Sr values of the Mt. Scopus and Judea rocks (Fig. 7c).

In summary, based on Na/Cl and <sup>87</sup>Sr/<sup>86</sup>Sr, the salinity sources of the GH and Yarmouk are similar, while the  $\delta^{18}$ O distinguishes between the freshwater sources of the GH and Ajloun. The water found in both Meitzar 2 and LYG springs was shown to be geochemically related to the northern component that contains brine remnants. Therefore, it is concluded that flushing of the remaining DSR brine from the Mt. Scopus and Judea groups in the GH, and possibly from other units as well, still continues today through discharge in the LYG springs.

#### Paleo-hydrological reconstruction

During the past 9.5 Ma, significant tectonic and basin changes have occurred in the area due to the dextral displacement along the Dead Sea transform. The overall movement during the last 25 Ma is estimated at 35–40 km (Freund et al. 1968; Walley 1998), which would have placed the Ness boreholes several kilometers closer to the Kinnarot basin prior to the movement (Fig. 8a). This tectonic movement also resulted in the uplift of the Hermon Mountain ridge (Garfunkel 1981; Heimann et al. 1990) which initiated the freshwater hydrogeological system sourced in the north.

Given the foregoing, a conceptual model demonstrating the intrusion of the hypersaline brine to the subsurface and their subsequent dilution and flow reversal, due to the major tectonic shifts over time, is presented



**Fig. 7** a Na/Cl equivalent ratio, b  $\delta^{18}$ O and c  ${}^{87}$ Sr/ ${}^{86}$ Sr vs. Cl<sup>-</sup> concentrations of the studied water of Golan Heights (GH). Mixing lines were added to show the change in geochemical properties as a function of salinity.  ${}^{87}$ Sr/ ${}^{86}$ Sr values of the northern lagoonary brines were taken from Starinsky and Katz (2014) (0.7077–0.708).  ${}^{87}$ Sr/ ${}^{86}$ Sr values of the Mt. Scopus and Judea Groups rocks are from McArthur

et al. (1994). The chemical and isotopic compositions of the LYG water were taken from previous publications (Bajjali et al. 1997; Gavrieli and Burg 2002), as well as analysis performed by the Israel National Water Co. (Mekorot Co.). All compositions are detailed in Appendix 3 and 4



9 - 5 Ma – halite precipitation and lagoonary brines intrusions:

5-0 Ma - Lacustrine brines intrusions and lagoonary brines dilution:



**Fig. 8** A conceptual model demonstrating the intrusion and flushing of the northern DSR brines. The intrusion of the lagoonary brines to the subsurface occurred concurrently with the precipitation of halite in the Kinnarot basin (**a**–**b**, red). The brines' ongoing migration created spatial chemical composition differences. Subsequently, lacustrine brines that were formed after the disconnection from the

Mediterranean Sea intruded into shallow units inside and nearby the DSR (c, purple). Following the intrusion, freshwater activity led flow reversal, dilution and flushing of the lagoonary brines (increasingly lighter red) into the natural outlets along the LYG (d). Dashed lines mark the locations of the cross sections in Appendix 5

in Fig. 8. Initially, during the Neogene, seawater transgressed (Fig. 4a) into a tectonic depression that developed along the DSR and formed a semiclosed lagoon (lagoonary stage). The seawater underwent intensive evaporation which led to an increase in solution density and salinity, and to precipitation of evaporites (predominantly halite). Concurrently, several pull-apart basins developed along the Dead Sea Rift and provided sufficient accommodation space for the halite to accumulate through rapid subsidence. The subsidence enabled the deposition of massive evaporite bodies near Lake Kinneret within the Kinnarot basin, where 900 m of halite were deposited across a 2.5-km section of alternating magmatic and sedimentary beds (Marcus and Slager 1985; Rozenbaum et al. 2019).

For halite to precipitate from seawater, a degree of evaporation of tenfold and above is required. At such degrees of evaporation, the density of the evaporated seawater is higher by >20% relative to freshwater. The elevated density increases the hydraulic head within the basin compared to the surrounding hydrogeological systems, which can result in the necessary driving force for the intrusion of the brines to the surrounding rocks (density-driven migration) while displacing freshwater (Stanislavsky and Gvirtzman 1999). In the subsurface, the brines can migrate over long distances (tens of kilometers) assuming no hydraulic barriers (Stanislavsky and Gvirtzman 1999). The fact that no brines were found northwards of Ness 03 suggests that the intrusion may have been restricted by the rising water head which is due to the elevated geological structure towards the Hermon Mountain range (Fig. 2).

Ar–Ar ages of basaltic flows from the top and base of the halite deposits in the Kinnarot Basin provide a general time constraint of ~9.5–5 Ma for the deposition of the halite sequence (Stein 2014). Based on the dating results, a similar time interval was defined as the geological period of the lagoonary brines formation and intrusion (Fig. 8a–b, red–orange). During this time window, dilution/evaporation cycles occurred in the lagoon (Raab et al. 1997; Rozenbaum et al. 2019), which could have led to numerous intrusion events of brines with different densities and chemical compositions.

During the subsequent lacustrine stage that followed the lagoonary stage (5 Ma to present), the basin disconnected from the sea. During this period, hypersaline lake water was significantly diluted by freshwater and halite stopped to precipitate in the Kinnarot basin. These diluted lacustrine brines are found only in shallower units within the DSR, likely due to their lower density compared to the lagoonary brines (Fig. 8c, purple). Over time, active fresh groundwater flow led to gradual subsurface dilution of the brines. In order for the brines to be flushed out of a given hydrogeological unit, the freshwater system has to exhibit a driving force that can overcome the high density of the brine.

With the lack of a driving force, brines will remain stagnant over geological periods. Elevated recharge areas were shown to create a topography-driven force that can gradually flush brines from deep sedimentary units (Ferguson et al. 2018). The extent of dilution depends on the recharge rate of the freshwater component and the hydraulic conductivity of each unit.

The high altitude of the Hermon range in the north (Fig. 1b) provides the necessary driving force for the flow and flushing to take place and overcome the high density of the brines. The brines that were flushed by freshwater discharged through the natural outlets in the LYG, while gradually decreasing the salinity of the water (Fig. 8d).

#### Conclusions

The water compositions found in the deep hydrogeological systems in the southern Golan Heights (GH) allowed for reconstruction of the unique paleo-hydrogeological setting of the area. The Upper Cretaceous rocks from the Mt. Scopus and Judea groups were shown to contain remnants of ancient Ca-chloride lagoonary Dead Sea Rift (DSR) brines, estimated to have formed 10–5 Ma ago in a subsiding basin within the DSR, where a massive halite section was deposited.

The intrusion process was controlled by lake level and density on one hand, and the groundwater level elevations in the surrounding hydrogeological systems on the other. The combination of all these processes resulted in intrusion/backflow where some of the brines that were chemically modified by water-rock interactions back-flowed into the rift. These dynamics have led to compositional differences in area as well as in different geological units.

Following the intrusion to the subsurface, the brines were diluted by freshwater with depleted isotopic composition, which originated from the margins of Mt. Hermon in the north through the subsurface. The high-altitude source enabled the flushing and dilution of the brines, as it provided the necessary driving force to overcome the hydraulic head provided by the high density of the brines. Due to the relatively low hydraulic conductivities and limited outcrop area of the studied formations, freshwater dilution took place over geological periods and is still currently taking place. Independent calculation of dilution factors based on salt composition and water isotopes showed higher degrees of dilution in the more permeable units (GUMF and Upper Judea Group) compared to the less permeable Taqiye Formation. This implies that brine intrusion and dilution is likely to exhibit a hysteretic behavior, where intrusion is less sensitive to hydraulic conductivities compared to subsequent flushing by freshwater (Akbarabadi and Piri 2013).

The intrusion and dilution processes were tectonically controlled. On one hand, the movement along the DSR led to the development of the subsiding basin west of the GH syncline, which accommodated the formation of the Ca-chloride brine, while on the other, the tectonic activity also led to the uplift and the exhumation of the elevated recharge areas of Mt. Hermon, which provided the necessary driving force to displace and flush the intruding brines. Flow was also assisted by the continuation of the subsiding DSR and the receding water levels of the rift lakes that enabled the gradual flushing and drainage towards the rift and the LYG. Additionally, brine intrusion and/or subsequent dilution may have displaced or assisted in the biodegradation of the hydrocarbons that were formed during a Pliocene paleothermal event in the GH from the Mt. Scopus source rocks (Reznik and Bartov 2021). The fact that the Mt. Scopus Group is more hydrologically active than thought may provide an additional explanation to the low oil saturations found in the source rocks (Mishash and Ghareb Formation) and/or the low quality of the hydrocarbons found (mostly extremely viscous bitumen).

In summary, the unique isotopically light saline Cachloride water found in the DSR tectonically active basin, was deconvoluted to explain the complex mixing dynamics that took place within the hydrogeological systems of the Mt. Scopus and Judea groups in the GH: (1) subsurface intrusion of DSR brines and mixing with geochemically distinct freshwater over prolonged geological periods and (2) flushing towards the LYG natural outlets where the GH and Ajloun hydrogeological systems converge.

# **Appendix 1**

**Table 3** Locations and elevations of the studied wells, as well as the depths of the open sections sampled. The apparent hydraulic conductivity was calculated based on the bottom hole pressure readings dur-

ing the drill stem tests. X-Y coordinates are presented in New Israel Grid units. *BGL* below ground level

Borehole	X coordinate	Y coordinate	Ground level eleva- tion (m AMSL)	Formation	Open section depths (m BGL)	Apparent hydraulic con- ductivity
Ness 02	268,472	742,853	370	Ghareb-Upper Mishash boundary	997-1012	$2 \times 10^{-7}$
				Ghareb	916–931	$2 \times 10^{-7}$
				Taqiye	816-831	$2 \times 10^{-9}$
				Lower Taqiye-Lower Mishash	850-1223	-
Ness 03	267,486	745,299	375	Ghareb	1,002-1,017	$2 \times 10^{-7}$
Ness 12	275,128	763,232	635	Lower Taqiye-Lower Mishash	1,150-1,530	-
				Upper Judea Gr (Bina-Dir Hana)	1,600–1,845	-

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Source	Well/spring	X Coordinate	Y Coordinate	Formation	Ref	Concentr	ation (n	ng/L)								Equivale	ent ratios	
						Na <sup>+</sup>	$\mathbf{K}^+$	Ca <sup>2+</sup>	$Mg^{2+}$	$\mathrm{Sr}^{2+}$	CI-	Br'	$\mathrm{SO_4}^{2-}$	HCO <sub>3</sub> <sup>-</sup>	TDS	Na/CI	Mg/Ca	Ca/ (HCO <sub>3</sub> + SO <sub>4</sub> )
East Kinneret	Bet Zayda	180,480	672,380	Quaternary fill	а	3,621	35.6	735	861	10.4	9,690	123	33.0	179	15,288	0.58	1.93	10.1
brines	1064	260,000	741,600	Quaternary fill	a	6,922	380	814	1,576		16,570	216	395	172	27,045	0.64	3.19	3.68
	1066	260,000	741,600	Quaternary fill	a	8,397	384	1,112	1,915		20,030	253	864	122	33,077	0.65	2.84	2.78
	1071	258,400	736,800	Quaternary fill	a	9,093	296	1,285	2,258		23,050		4.00	186	36,172	0.61	2.90	20.5
	1069	259,600	738,700	Quaternary fill	a	6,483	337	1,052	1,822		16,680	196	782	219	27,571	0.60	2.86	2.64
	1075	255,400	734,400	Quaternary fill	а	7,127	195	944	1,734		17,540	176	5.00	469	28,190	0.63	3.03	6.05
	Ha'on	258,620	736,960	Quaternary fill	a a	4,073	178	587	896	53.4	9,883	114	244		16,028	0.64	2.52	5.77
West Kinneret brines	Tiberias Sp. – main	254,207	763,057	Various units	æ	7,096	335	3,498	651	62.2	18,190		849	149	30,830	0.60	0.31	8.68
	Tiberias Sp. – roman	254,207	763,057	Various units	ci	7,154	356	3,592	692	69.3	18,630	245	809	ī	31,547	0.59	0.32	10.6
	Tiberias Sp. – roman	254,207	763,057	Various units	æ	7,029	348	3,524	675	67.7	18,480	247	833	ī	31,204	0.59	0.32	10.1
	Tabgha- Sar- tan Iver	255,756	756,105	Various units	c;	1,420	52.3	509	144	11.9	3,075	ı	260		5472	0.71	0.47	4.69
	Tabgha- Sar- tan Iver	255,756	756,105	Various units	c3	1,293	51.0	413	134	10.9	2,765	ı	233	342	5242	0.72	0.54	1.97
	Fuliya 6	248,546	748,421	Various units	a	7,718	305	1,912	844		16,260	115	2,140	224	29,518	0.73	0.73	1.98
	Fuliya new	248,550	748,420	Various units	a	672	22.6	228	93.0	2.90	1,323		172		2514	0.78	0.67	3.18
	Kinneret 10	249,750	745,720	Various units	a B	8,364	293	2,081	748	38.4	17,410	126	1,940	226	31,226	0.74	0.59	2.35
	Barbutim 1020/3	249,721	745,896	Various units	a	1,314	55.0	523	156		3,070	30.0	235	341	5724	0.66	0.49	2.49
	Barbutim 48"	249,721	745,896	Various units	q	1,438	54.3	535	162		3,335	32.0	28.0	295	5879	0.66	0.50	4.92
Rosh Pina brine	Rosh Pina-1	254,207	763,057	Jurassic lime- stones	c;	33,400	1160	27,000	2,920	ı	105,200	1740	006	171	172,491	0.49	0.18	62.5
Seawater	MOW				e	10,556	380	400	1,262	13.0	18,980	65.0	2,646	140	34,442	0.86	5.20	0.35
<sup>a</sup> Starinsky an <sup>b</sup> Bergelson et	d Katz (2014) al. (1999)																	

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Table 5 Chemical composition and ion	1 ratios of saline water	bodie	s near the	Dead S	ea southe	rn DSR b	rines								
Source	Well/spring	Ref	Concenti	ration (m	ıg/L)								Equival	ent ratio	
			Na <sup>+</sup>	$\mathbf{K}^+$	Ca <sup>2+</sup>	$Mg^{2+}$	$\mathrm{Sr}^{2+}$	CI-	Br <sup>-</sup>	$\mathrm{SO}_4^{2-}$	HCO <sub>3</sub> <sup>-</sup>	TDS	Na/CI	Mg/Ca	Ca/ (HCO <sub>3</sub> +SO <sub>4</sub> )
DSR southern lagoonary brines (northern Negev- R <sub>0</sub> )	Boqer-1	ci	31,080	314	15,870	1,660	427	81,060	ı	1,170		131,581	0.59	0.17	32.5
	Kurnub-1	5	31,950	565	13,950	1,062	331	77,660	ı	753	ı	126,271	0.63	0.13	44.4
	Makhtesh Qatan-2	a	25,950	901	14,800	2,550	ı	73,580	725	895	76	119,477	0.54	0.28	37.1
	Makhtesh Qatan-2	a	29,600	581	22,300	2,190	ī	89,190	924	531	39	145,355	0.51	0.16	95.2
	Qeren-1	5	41,260	1,010	15,407	1,585	284	95,760	733	497	158	156,694	0.66	0.17	59.4
	Qeren-1	es S	42,680	1,080	15,570	2,094	ı	99,630	816	505	146	162,521	0.66	0.22	60.2
	Sherif-1	es	35,000	300	17,900	2,128	684	94,150	782	644	110	151,698	0.57	0.20	58.7
	Zavoa-1	e	36,800	408	16,500	1,960	585	92,100	730	474	14	149,571	0.62	0.20	81.5
DSR southern lagoonary brines (R <sub>s1</sub> )	Arava-1	5	50,480	1,030	50,640	12,040	1205	202,200	3,670	122	61	321,448	0.38	0.39	714
	Heimar-1	a	26,920	1,250	39,780	4,107	ı	113,450	ı	1,230		186,737	0.37	0.17	77.5
	Heimar-1	e	24,000	1,600	43,010	3,630		137,800	2,750	1,280		214,070	0.27	0.14	80.5
	Massada-1	ei B	33,800	1,000	44,340	5,034		144,400	2,760	352	,	231,686	0.36	0.19	302
	Massada-1	es	35,000	1,100	46,700	4,888		153,800	2,780	247		244,515	0.35	0.17	453
	Zuk Tamrur-3	e	35,570	331	20,410	5,003	937	112,200	2,610	133	ı	177,194	0.49	0.40	368
DSR southern lacustrine brines $(R_{s2})$	Dead Sea	e	39,250	7,050	16,000	39,200	295	207,800		ı	261	309,856	0.29	4.04	187
	Dead Sea	a	37,800	7,530	17,500	43,800	309	221,000	5,100	440	265	333,744	0.26	4.13	64.7
	Dead Sea	e	39,500	7,590	16,500	24,600	241	218,500	4,900	510	265	312,606	0.28	2.46	55.0
	Dead Sea	e	38,890	7,500	16,250	43,690	338	223,500		ı		330,168	0.27	4.43	
	Dead Sea	es.	39,050	8,180	16,500	44,000	330	224,400		ı	ı	332,460	0.27	4.40	
	Dead Sea	a	29,910	6,220	15,020	37,040	285	200,600	ı	2,080	273	291,428	0.23	4.07	15.7
	Dead Sea	e	39,710	9,060	19,430	52,420	374	226,200	ı	1,320	264	348,778	0.27	4.45	30.5
	Dead Sea	e	36,100	8,400	19,400	50,200	360	235,000		ı	ı	349,460	0.24	4.27	
	Dead Sea	e	33,000	7,650	18,000	45,500	330	225,000	,500	2,000		336,980	0.23	4.17	21.6

<sup>a</sup>Starinsky and Katz (2014)

# Appendix 3

Source	Well/spring	Ref	δ <sup>18</sup> O (‰ VSMOW)	δ <sup>2</sup> H (‰ VSMOW)	d-excess	<sup>87</sup> Sr/ <sup>86</sup> Sr
West Kinneret brines	Ein Nur	с	-5.70	-28.0	17.60	-
	1047	с	-4.50	-20.0	16.00	-
	Nur 3	с	-5.90	-19.0	28.20	-
	Kinneret 8	a,c	-2.29	-9.06	9.26	0.70772
	1020/2	с	-5.50	-28.0	16.00	-
	1020/3	с	-4.60	-27.0	9.80	-
	1020/7	с	-5.90	-20.0	27.20	-
	Ein Mudwara	с	-5.20	-22.0	19.60	-
	1051	с	-4.90	-25.0	14.20	-
	Kinneret 10b	a,c	-1.40	-2.87	8.33	0.70787
	Kinneret 5	с	-4.30	-9.00	25.40	-
	Kinneret 6	с	-1.60	-9.00	3.80	-
	Kinneret 2	a,c	-2.71	-14.4	7.30	0.70777
	Tiberias Roman sp.	a,c	-3.30	-20.0	6.40	0.70777
	CH14	с	-3.90	-18.0	13.20	-
East Kinneret brines	D1069	с	-2.30	-9.00	9.40	-
	D1071	a,c	-1.70	-6.00	7.60	0.70574
	Ha'on 1	a,b	-1.50	-8.47	3.53	0.70682
	Ha'on 2	b	-1.83	-10.7	3.90	-
Rosh Pina-1	Rosh Pina 3150 m	с	3.00	-	-	-
Lower Yarmouk Gorge- Meizar wells	Meizar 3	d	-6.20	-31.6	18.0	0.70767
	Meizar 3	e	-5.90	-33.4	13.8	0.70765
	Meizar 2	d	-7.16	-34.5	22.8	0.70782
	Meizar 2	e	-7.05	-39.0	17.3	0.70769
Lower Yarmouk Gorge- Hammat Gader	Ein Makla	d	-5.90	-28.1	19.1	0.70782
thermal springs (Israel)	Ein Makla	e	-6.22	-34.6	15.1	-
	Ein Balsam	d	-5.90	-30.4	16.8	0.70773
	Ein Balsam	e	-6.03	-29.6	18.6	-
	Ein Reach	d	-5.90	-30.9	16.3	0.70779
	Ein Reach	e	-6.06	-30.4	18.1	-
	Ein Sehina	d	-5.75	-23.9	22.1	-
	Ein Sehina	d	-5.70	-24.0	21.6	0.70775
Lower Yarmouk Gorge- Mukheiba thermal	Balsam sp.	f	-6.00	-29.0	19.0	-
springs (Jordan)	Maqla sp.	f	-6.40	-31.0	20.2	-

Table 6 Isotopic composition of water bodies in the Golan Heights region

<sup>a</sup>Starinsky and Katz (2014)

<sup>b</sup>Bergelson et al. (1999)

<sup>c</sup>Gat et al. (1969)

<sup>d</sup>Gavrieli and Burg (2002)

<sup>e</sup>Israel National Water Co. (Mekorot Co.)

<sup>f</sup>Bajjali et al. (1997)

Appendix 4

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Table 7 Chemical composition and ion ratios of water sources at the Lower Yarmouk Gorge

Source	Well/spring	Formation	Ref (	Concent	ration (1	ng/L)							Equiva	lent ratio	
				Va <sup>+</sup> K	+ Ca	<sup>2+</sup> Mg <sup>2</sup>	+ Sr <sup>2+</sup>	Cl-	Br <sup>-</sup>	$\mathrm{SO}_4^{2-}$	HCO <sub>3</sub> <sup>-</sup>	TDS	Na/Cl	Mg/Ca	Ca/ (HCO <sub>3</sub> +SO <sub>4</sub> )
Lower Yarmouk Gorge- Meizar wells	Meizar 3	Ghareb-Upper Mishash Fm	a	8.0 4	.80 64.	5 20.5	1.10	57.0	0.20	3.60	342	541	1.30	0.52	0.57
	Meizar 3	Ghareb-Upper Mishash Fm	ч, ч,	2.0 4	20 79.	0 27.0	1.08	78.0	ı	20.0	352	614	1.03	0.56	0.64
	Meizar 2	Upper Judea Gr	a	88 2	1.2 169	36.8	5.00	340	4.30	305	273	1,343	0.85	0.36	0.78
	Meizar 2	Upper Judea Gr	сч 4	41 2	5.5 175	37.4	5.2(	393	5.90	315	267	1,465	0.95	0.35	0.80
Lower Yarmouk Gorge- Hammat	Ein Makla	Various units	e	11 5	20 162	2 43.3	5.2(	456	6.50	145	354	1,388	0.71	0.44	0.92
Gader thermal springs (Israel)	Ein Makla	Various units	сч 4	16 1	9.0 163	\$ 45.0	5.99	486	6.00	154	304	1,399	0.69	0.46	0.99
	Ein Balsam	Various units	a	34 1	1.5 130	36.6	3.3(	282	3.40	115	349	1,070	0.73	0.44	0.84
	Ein Balsam	Various units	q	37 1	2.0 133	38.0	3.58	285	4.00	122	333	1,068	0.74	0.47	0.83
	Ein Reach	Various units	e	43 1	1.5 139	38.7	3.2(	308	3.80	113	356	1,116	0.72	0.46	0.85
	Ein Reach	Various units	q	78 1	4.6 15]	37.5	4.01	362	4.70	120	355	1,227	0.76	0.41	0.91
	Ein Sehina	Various units	a	1.0 3	20 96.	5 29.6	0.50	61.5	0.30	47.6	378	658	1.03	0.51	0.67
	Ein Sehina	Various units	a	1.5 3	20 97.	0 30.0	0.52	60.7	0.27	46.3	378	658	1.05	0.51	0.68
Lower Yarmouk Gorge- Mukheiba	Balsam sp.	Various units	с. о	2.9 7	.04 99.	6 31.6	ī	109	ı	89.8	353	763	1.03	0.52	0.65
thermal springs (Jordan)	Maqla sp.	Various units	с э	22 1	2.5 118	33.4	ı	198	ī	158	326	968	0.95	0.47	0.68
<sup>a</sup> Gavrieli and Burg (2002)															

<sup>b</sup>Israel National Water Co. (Mekorot Co.) <sup>c</sup>Bajjali et al. (1997)

## **Appendix 5**

Fig. 9 Conceptual E–W cross sections demonstrating the intrusion of the DSR brines into the subsurface of GH. Based on the paleo reconstruction detailed in section 'Paleohydrological reconstruction'. Cross line locations are marked in Fig. 8



#### 9 - 5 Ma – halite precipitation and lagoonary brines intrusions:

5-0 Ma - Lacustrine brines intrusions and lagoonary brines dilution:



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#### Declarations

**Conflicts of interest** The authors declare that they have no conflict of interest in connection with the submitted material.

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