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Saline Groundwater from Coastal Aquifers As a Source for Desalination

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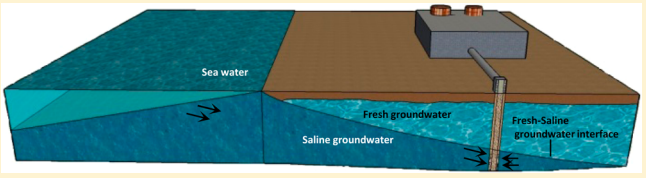
Supporting Information

ABSTRACT: Reverse osmosis (RO) seawater desalination is currently a widespread means of closing the gap between supply and demand for potable water in arid regions. Currently, one of the main setbacks of RO operation is fouling, which hinders membrane performance and induces pressure loss, thereby reducing system efficiency. An alternative water source is saline groundwater with salinity close to seawater, pumped from beach wells in coastal aquifers which penetrate beneath the freshwater-seawater interface. In this research, we studied the potential use of saline groundwater of the coastal aquifer as feedwater for desalination in comparison to seawater using fieldwork and laboratory approaches. The chemistry, microbiology and physical properties of saline groundwater were characterized and compared with seawater. Additionally, reverse osmosis desalination experiments in a cross-flow system were performed, evaluating the permeate flux, salt rejection and fouling propensities of the different water types. Our results indicated that saline groundwater was significantly favored over seawater as a feed source in terms of chemical composition, microorganism content, silt density, and fouling potential, and exhibited better desalination performance with less flux decline. Saline groundwater may be a better water source for desalination by RO due to lower fouling potential, and reduced pretreatment costs.



INTRODUCTION

The global shortage of fresh water in the world is only expected to increase in the coming decades;[1] arid and semiarid countries are under constant stress in freshwater supply due to a growing population and corresponding needs while experiencing gradual decrease in water resources availability due to contamination of groundwater.[2] Seawater desalination, particularly reverse osmosis, has a pivotal role in meeting the challenge of water scarcity and has a high potential to help cope with the water shortage. The global water intelligence (GWI) reports that source water for desalination is split with about 60% from seawater, 20% from brackish groundwater, and the remaining is surface water and saline wastewater.[3] In Israel, for example, over 600 million cubic meters per year (MCMY) of freshwater (~60% of the country's freshwater demand) is produced by seawater RO desalination.[4] The RO desalination process requires high energy and large plant area by the shore, thus increasing the product water cost.[3] The area required for seawater RO desalination plant is about 25 acres for a plant that produces 100 MCMY.[5] Desalination of brackish groundwater is a large water source, mainly due to the advantage of its lower salinity in comparison to seawater, hence it requires less energy. However, inland brine disposal poses a severe problem in brackish water desalination, as most brackish water desalination facilities are remote from the sea.[6]

In this study we examined the use of an alternative water source for desalination, saline groundwater (SGW) of coastal aquifers, as feedwater for RO desalination. This type of water results from seawater intrusion into coastal aquifers shifting the fresh-saline water interface (FSI) upward and landward, leading to replacement of fresh groundwater by saline groundwater.[7] This intrusion is mainly caused by over pumping of groundwater in coastal aquifers,[8,9] but also occurs naturally as a result of global sea level rising, seasonal change of groundwater table levels,[10] and groundwater tides.[11] This process is thus responsible for salinization and degradation of the groundwater quality in coastal aquifers.

Using coastal aquifer SGW, which is pumped significantly below the FSI, and has salinity close to seawater values for desalination, has been used and compared to seawater desalination.[12] However, a comprehensive research that tests

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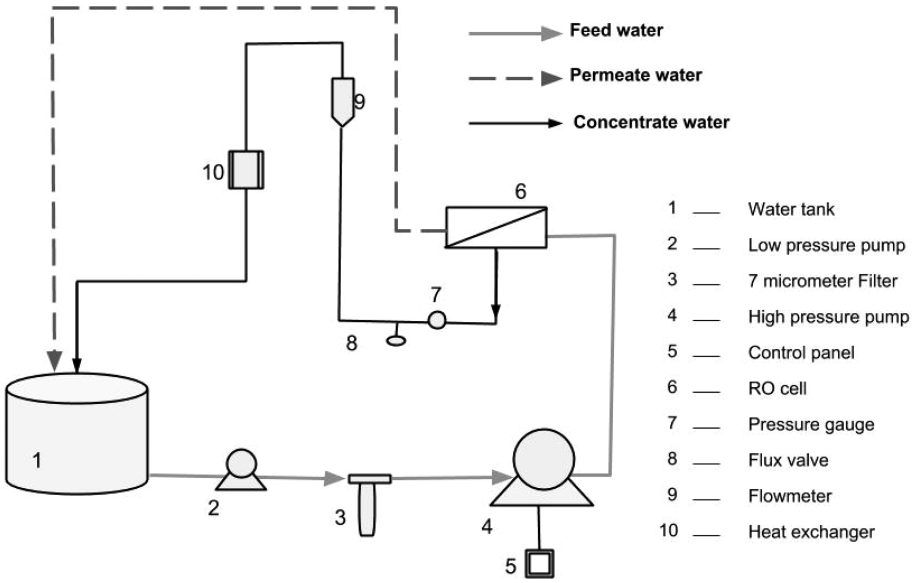

Figure 1. Cross-flow RO system design: water from a 500 L water tank (1) is pumped by a secondary pump (2) through a filter (3). The intensity of the main pump (4) is controlled by the control panel (5). The water is pumped to the cross-flow membrane cell (6). The flow of the concentrated water out of the cell is controlled by a valve (8). The valve and the main pump control the pressure in the cell, which is monitored by a pressure gauge (7). The flowmeter (9) helps to maintain the same flux in all the experiments. The concentrated water flows back to the water tank through a heat exchange unit (10). The permeate flows to the water tank (dash line).

its advantages for desalination from various aspects is needed for determining the applicability of this water type. Desalination of SGW is expected to present several advantages over using seawater that include (1) Natural filtration of the groundwater through the porous sediment while pumped, thus reducing the need for feedwater pretreatment and saving plant area and costs;[13,14] (2) Temperature variations of groundwater are narrow (~24 °C), residing in the range of the desalination temperature;[15] (3) Pumping of saline groundwater for cooling systems (air conditioning systems) and seawater swimming pools in hotels is a common practice; water from existing wells can be used as feedwater for desalination; (4) Pumping saline groundwater below the FSI pushes the FSI toward the sea and therefore could result in the additional advantage of restraining seawater intrusion into coastal aquifers;[16,17] and (5) Fouling on RO membranes is expected to be lower with SGW than with coastal seawater used as feed due to lower values of several parameters pertinent to RO desalination, namely total organic carbon (TOC), dissolved oxygen (DO), turbidity and silt density index (SDI).[15] Fouling and biofouling are major obstacles in RO desalination. Thus, the maintenance and overall energy cost derived from periodical cleaning is expected to be lower when using SGW.[18] A recent study found that the membrane cleaning frequency in RO facilities that use SGW as feed was lowered by 25−75% as compared with facilities that use seawater as feed.[19] Furthermore, SGW has lower primary and bacterial production rates than those of coastal seawater and hence it is expected to have less biofouling.[15,20] Most of these advantages have only been suggested and studied preliminary, but further assessment and validation is still needed.

To date, desalination plants around the world use saline groundwater as their feedwater. One is located in Malta, where an RO desalination plant is operated by use of saline groundwater pumped from beach wells and supplies 50% of the fresh water production.[14] Another desalination plant that uses saline groundwater pumped below the FSI is located in Almeria (southeast Spain);[21] Almeria is semiarid area, thus the main water resource is the coastal aquifer, whose water quality has deteriorated in recent years.[22] Nineteen wells located 30−150 m away from the shoreline, pump saline groundwater and produce about 16 MCMY.[21] In other arid regions where there is no fresh groundwater source, SGW is used as feedwater for desalination and in some cases, the wells are located seawards.[19] Most facilities worldwide are small to medium scale, however in some places large scale RO facilities have been built using SGW as feed.[13] One example is the facility in Sur, Oman, which is the largest desalination facility that uses SGW as feed with permeate productivity of 60 MCMY.[23] By pumping SGW, FSI is pushed deeper and toward the sea.[24,25] In addition, water analyses showed that the groundwater is filtered while flowing through the sediment, which reduces the amount of particles (SDI) by more than 90%.[15,26]

The purpose of this study was to examine the suitability of SGW as feedwater for RO desalination by characterizing the chemical, physical and biological properties of the saline groundwater and of seawater, and by evaluating the performance, efficiency and fouling propensity of RO desalination of the two water types. The results indicated that using SGW as source for RO desalination in coastal aquifers may be highly beneficial in comparison to desalination of seawater.

MATERIALS AND METHODS

Water Sampling. FSI groundwater and the SGW were collected from an observation well, located 80 m away from the shoreline in the Nitzanim Nature Reserve (Latitude: 31°47′31″ N, Longitude: 34°38′58″ E, herein referred to as Nitzanim). The water was sampled by pumping, using a submersible pump (Grundfos, Bjerringbro, Denmark). FSI water was sampled from depth of ~30 m, and SGW from 45 m. The seawater for the desalination experiment was taken directly from the sea in the beach area adjacent to the study area in Nitzanim.

Saline groundwater (500 L) was also sampled in the Winter and in Summer from an active pumping well located 100 m

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from the shoreline, which pumps from 50 m below the surface in a rate of 1.7 MCMY at the Hilton hotel in Tel-Aviv (Latitude: 32°04′51″ N, Longitude: 34°46′50″ E). Silt particle concentration in the vicinity of a pumping well which pumps throughout the year is lower than the surrounding silt concentrations, which improves the water quality for desalination.[27] Water from a pumping well will ultimately provide feed for a desalination plant, thus better simulating the water quality of the SGW. Seawater for the fouling experiment was taken from the seawater inlet to the Ashkelon desalination plant (Latitude: 31° 40′8″, Longitude: 34°34′27″) in order to better simulate the source water that goes to the desalination industrial facility. In each sampling session, pH, DO, EC and temperature were measured on site.

RO Cross-Flow Filtration Experiments. Flat-sheet membranes chosen for the RO filtration experiments were SW30-HR (Dow-FILMTEC), TM820−400 (Toray) and SWC-1 (Hydranautics). The experiments were conducted using a cross-flow system that contained a pressure cell (Sterlitech) with dimensions of 9.5 cm × 14.6 cm of active filtration area and applied pressure of 50 bar (Figure 1). The water was pumped from a 10 L water tank using a secondary pump in order to give the water initial pressure before using the main pump, which enabled high pressure. A flow rate of 150 L.hour^{-1} was kept constant during the experiments. The temperature of the system was kept at 24 °C by using a heat-exchange unit of the residual water and was monitored during the experiment.

The membranes used in the experiments were first immersed in isopropanol solution and then washed by distilled water. Prior to each experiment, the membrane performance was determined by measuring the permeate flux using distilled water as feed and the applied pressure was varied between 30, 40, and 50 bar. The linearity between the pressure and the flux was examined. $R^2 > 0.99$ suggested that the membrane was intact. Then, desalination of NaCl solution (0.55 M) was performed to determine the salt rejection of the membrane. Salt rejection (S.R) was determined by measuring the electrical conductivity (EC) of the feed (C_f) and the permeate (C_p), and expressed according to eq 1:

$$\text{S.R} = \left(1 - \left(\frac{C_p}{C_f}\right)\right) \times 100 \tag{1}$$

Membrane samples achieving NaCl rejection higher than 97% were used; the system was kept at 50 bar of pressure for 12−20 h, allowing compaction of the membrane and reaching permeate flux stabilization.

Comparative experiments investigating three different membrane types were performed with 4 L of SGW. A pressure cell of 8 cm × 2.8 cm active filtration was used. Permeate and concentrate samples were taken for chemical analysis (see below).

The membrane permeability (L_p) of the different water types (FSI, SGW, and seawater) was calculated using eq 2:

$$L_p = \frac{J_v}{\Delta p - \Delta \pi} \tag{2}$$

where J_v is the permeate flux, Δp is the applied pressure and $\Delta \pi$ is the osmotic pressure difference. L_p represents the flux through the membrane while considering the pressure on the membrane caused by concentration polarization phenomena. The experiments were conducted under a pressure of 50 bar for 6 h, using 8 L of each water type. Salt rejection was calculated using eq 3[28]

$$\frac{1 - R_0}{R_0} = \frac{1 - R}{R} \times \exp\left(\frac{J_v}{k}\right) \tag{3}$$

where R_0 is the apparent rejection that is calculated with eq 1, R is the true rejection, J_v is the flux and k is the mass transfer coefficient, defined by eq 4 according to Sutzkover et al. (2000):[29]

$$k = \frac{(J_{v_{salt}})}{\ln\left\{\frac{\Delta p}{\pi_b - \pi_p} * \left[1 - \left(\frac{J_{v,salt}}{J_{v,H_2O}}\right)\right]\right\}} \tag{4}$$

Where $J_{v,salt}$ is the flux measured with the salt solution and J_{v,H_2O} is the flux measured with distilled water; π_b and π_p are the osmotic pressure of the bulk water and the permeate, respectively.

Fouling Experiments. Fouling experiments were performed with SGW and seawater. Seawater was sampled from the intake pipe of Ashkelon seawater desalination plant (Israel) which takes the water 1 km away from the shore; the SGW was taken from the groundwater pump of Hilton hotel in Tel-Aviv. The experiments were 120 h long and a large feedwater reservoir of 500 L was used. A pressure cell of 8 cm × 2.8 cm active filtration area was used; these method modifications were made in order to exhaust the amount of foulants in the water.

Water Chemistry Analysis. Na^+, Ca^{2+}, Mg^{2+}, K^+, Sr^{2+}, and Boron were analyzed using inductively coupled plasma-optical emission spectroscopy (ICP-OES, Optima 5300 V) with 2% precision. Cl^-, Br^-, and SO_4^{2-} were analyzed by ion chromatography (Dionex 4000i) with a precision of 2%. HCO_3^- was analyzed by titration device (Titrino 785 Metrohm) using 0.02 N HCl solution. TOC was measured with multi N/C 2100s with 0.1% precision. Electrical conductivity (EC), pH, and DO were measured with EC, pH, and DO meter (WTW-KS multi 3xxi).

Silt density index (SDI) measurements were conducted using 20 L of source water in a PVC water container connected to a condensed air system and a pressure gauge applying 30 psig inside the container. Water was filtered by 0.45 μm filter. The water flow was measured at t_0 (flux at $t = 0$) and at t_{15} (flux at $t = 15$ min) and SDI was calculated using eq 5:[30]

$$\text{SDI} = \frac{1 - \left(\frac{t_0}{t_{15}}\right)}{15} \times 100 \tag{5}$$

SDI represents the percentage drop of the flux in 1 min.

Microbiology Analyses. Microbiology analyses were conducted for the SGW from Hilton hotel in Tel Aviv and in seawater from the intake pipe of Ashkelon desalination plant. To measure chlorophyll *a*, triplicate samples (300 mL) were filtered onto glass fiber filters (nominal pore size 0.7 μm, Whatman). The filters were stored at −20 °C in a dark box for up to 1 week until analysis. Samples were extracted in 5 mL 90% acetone overnight, at 4 °C in dark. Chlorophyll *a* concentrations were determined using a luminescence Trilogy fluorometer (7200−000) with a 436 nm excitation filter and a 680 nm emission filter.[31]

Picophytoplankton, heterotrophic and autotrophic bacterial abundance were enumerated by flow cytometry. Samples of 1.8

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Table 1. Average Physicochemical Parameters of Eastern Mediterranean Seawater and Saline Groundwater

	SGW			seawater			
parameters	mean	range	stddev	mean	range	stddev	SGW/seawater ratio
TDS, meq L^{-1}	1150	1052−1384, n = 46	85.6	1344	1267−1390, n = 10	41.6	0.86
Cl^-	526	457−632, n = 46	40.7	624	597−637, n = 10	12	0.84
Na^+	432	388−528, n = 46	36.1	506	459−534, n = 10	26.5	0.85
Mg^{2+}	100	88−115, n = 46	8.59	117	103−122, n = 10	5.8	0.85
SO_4^{2-}	53	46.1−66.1, n = 46	4	61	57.3−65.7, n = 10	3.8	0.86
K^+	8	6.6−11.5, n = 46	1.3	11.5	10.3−13.9, n = 10	0.92	0.70
Ca^{2+}	30	19.5−38.9, n = 46	3.9	23	19.8−24.4, n = 10	1.35	1.33
Br^-	0.73	0.64−0.81, n = 37	0.15	0.83	0.74−0.83, n = 10	0.07	0.88
Sr^{2+}	0.2	0.13−0.22, n = 46	0.02	0.18	0.12−0.2, n = 10	0.02	1.09
B	0.28	0.23−0.3, n = 3	0.03	0.4	0.37−0.41, n = 2	0.02	0.70
dissolved oxygen, mg L^{-1}	3.2	0.7−6.7, n = 26	1.8	8.5	7.2−9.36, n = 5	1.0	0.38
total organic carbon, mg L^{-1}	0.64	0−1.7, n = 3	0.75	1.63	1.48−1.78, n = 2	0.15	0.39
temperature-summer, °C	26.0	23.3−29.6, n = 18	1.77	28.6	27.5−29.6, n = 4	0.8	
temperature-winter, °C	22.4	20.4−26.7, n = 16	2.1	18.6	17.7−19.5, n = 3	0.74	
pH	7.1	6.58−7.42, n = 16	0.2	8.2	8−8.27, n = 7	0.09	
silt density index, % min^{-1}	3.3	2−4.4, n = 13	0.9	4.6	4.2−5.4, n = 6	0.4	0.72
osmotic pressure, bar	26.2	23.8−31.6, n = 46	2	30.8	29−31.9, n = 10	0.9	0.85

mL were immediately fixed after sampling with 6 μL of 50% glutaraldehyde (Sigma G-7651), left at room temperature for 10 min, frozen in liquid nitrogen, and subsequently kept at −80 °C until analysis. Prior to the analysis, fixed samples were fast thawed at 37 °C. Analysis was performed using an Attune cytometer (Applied Biosystems, MA), fitted with an argon lasers (405 and 488 nm). One μm beads (Applied Biosystems, Massachusetts, USA) served as standards.[32] The taxonomic discrimination was based on cell side-scatter as a proxy of cell volume, forward scatter as a proxy of cell size, and orange and red fluorescence of phycoerythrin and Chlorophyll *a* (585 and 630 nm, respectively). Heterotrophic bacteria were stained (200 μL of the initial sample) with SYTO 9 Green Fluorescent Nucleic Acid Stain[33] and enumerated by discrimination based on green fluorescence (530 nm) and side scatter.

Bacterial productivity was estimated using the [4,5-^{3}H]-leucine incorporation method (Amersham; specific activity: 160 Ci $mmol^{-1}$).[34] Briefly, three aliquots (1.7 mL each) from each sample were incubated with 100 nmol L^{-1} of [4,5-^{3}H]-leucine (PerkinElmer, specific activity 160 Ci $mmol^{-1}$) for 4 h at an ambient temperature in the dark. Preliminary experiments indicated that this was a saturating level of ^{3}H-leucine, and that incorporation was linear during this time. A triplicate addition of trichloroacetic acid (TCA) served as a control. The incubations were terminated with 100 μL of cold (4 °C) TCA (100%), followed by the microcentrifugation protocol.[35] After adding 1 mL of scintillation cocktail (Ultima-Gold) to each vial, the samples were counted using a TRI-CARB 2100 TR (Packard) liquid scintillation counter. Production rates were calculated using a conversion factor of 1.5 kg C mol^{-1} with an isotope dilution factor of 2.0.[36]

Photosynthetic carbon fixation rates were measured by the ^{14}C incorporation method.[37] All samples were analyzed in quadruplicates with dark and zero time controls. To determine the quantity of added radioactivity, 50 μL of each sample were immediately mixed with 50 μL of ethanolamine and stored for analysis. The incubations were terminated by filtering the spiked seawater onto GF/F filters (Whatman). The filters were incubated overnight in 5 mL scintillation vials containing 50 μL of 32% HCl in order to remove excess ^{14}C-bicarbonate. After adding 3 mL of scintillation cocktail (Ultima-Gold) to each vial, the radioactivity was measured using a TRI-CARB 2100 TR (Packard) liquid scintillation counter.

RESULTS AND DISCUSSION

Source Water and Characteristics. Initially, chemical and biological parameters of SGW and seawater were analyzed and compared. The results of water parameters that were measured in this study are depicted in Table S1 (Supporting Information (SI)), specifying values for each location separately. In order to represent typical parameters for SGW and seawater from Eastern Mediterranean area, average values of these parameters for SGW and seawater were calculated, including data from a previous study.[38]

Our calculated average chemical and physical parameters for SGW and seawater are given in Table 1; as can be seen in Table 1, the salinity of the SGW is lower than that of the Mediterranean seawater. The Cl^- concentration in the SGW (526 mequiv L^{-1}) is 84% of that of the Mediterranean seawater, indicating 16% dilution with fresh groundwater. Salinity of groundwater, which is lower than the adjacent seawater, was found in other coastal aquifers around the world[39,40] and was previously studied in arid environments.[38,41,42] However, in many areas, for example around the Red Sea, the groundwater is highly saline, in some cases even higher than the ocean salinity.[43] The ion concentration of the SGW deviates from simple mixing of seawater with fresh groundwater (84% of seawater values). The most obvious example is the Ca^{2+} enrichment in the groundwater which increases from 23 mequiv L^{-1} in seawater to 30 mequiv L^{-1} in SGW. Sr^{2+} follows the Ca^{2+} trend with enrichment of typical values of 0.18 mequiv L^{-1} for seawater to 0.2 mequiv L^{-1} for SGW. Typical values of K^+ were 11.5 mequiv L^{-1} in seawater at the Israeli coastline and 8 mequiv L^{-1} in SGW, which shows a 30% depletion. The typical concentration of boron in seawater is 0.4 mequiv L^{-1} and in the SGW is 0.28 mequiv L^{-1}, which shows a 30% decrease and depletion from pure mixing of seawater and fresh groundwater. The substantially lower boron concentration of SGW is highly beneficial for desalination, as the boron permeability of RO membranes is high, and special removal of boron is needed during seawater RO desalination (mostly post-treatment);[44,45] hence, lower boron values may eliminate

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Table 2. Temporal Variations of the Initial Biological Characteristics of the Water Used in the Different Experiments

	SGW		seawater		SGW/seawater ratio	
parameter	winter	summer	winter	summer	winter	summer
chlorophyll *a*, μg L^{-1}	0.14 ± 0.03	0.32 ± 0.04	0.30 ± 0.01	0.85 ± 0.09	0.46	0.38
cyanobacteria, cells $L^{-1} \times 10^4$	2.28 ± 0.10	3.33 ± 0.82	5.61 ± 0.07	4.30 ± 0.39	0.39	0.77
picoeukaryotes, cells $L^{-1} \times 10^4$	0.14 ± 0.03	0.12 ± 0.03	0.14 ± 0.02	0.32 ± 0.06	1	0.38
heterotrophic bacteria, Cells $L^{-1} \times 10^4$	50.08 ± 4.12	51.20 ± 6.35	79.35 ± 11.82	76.25 ± 8.08	0.63	0.67
primary production, μg C L^{-1} h^{-1}	0.83 ± 0.14	1.37 ± 0.21	2.16 ± 0.66	0.85 ± 0.07	0.38	1.61
bacterial production, μg C L^{-1} h^{-1}	0.75 ± 0.19	0.68 ± 0.14	0.88 ± 0.08	0.60 ± 0.09	0.85	1.13

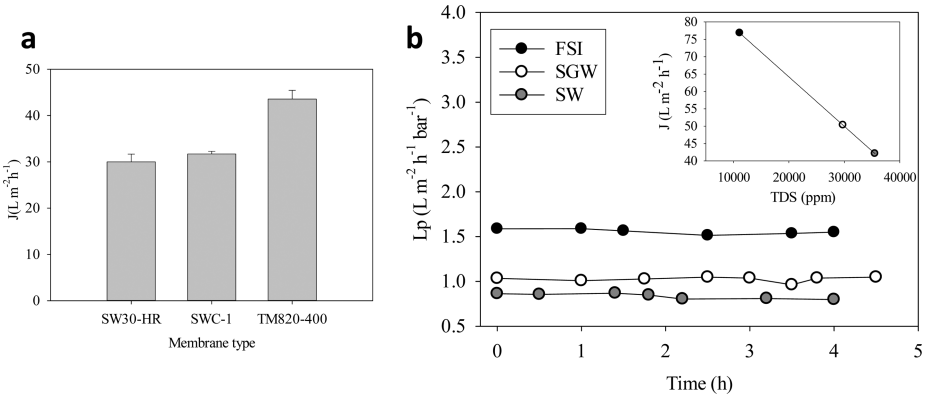

Figure 2. (a) The permeate flux (J) of three membrane types SW30-HR, TM820-400 and SWC-1 in RO filtration of SGW sampled from the Hilton hotel in Tel Aviv, under pressure of 50 bar. (b) Permeability (Lp) of three different water types in RO filtration experiments by a cross-flow system using TM820-400 membrane. SGW and FSI were sampled from Nitzanim observation well during winter; seawater was taken from the shoreline of Nitzanim, 80 m away from the well. The inset shows a correlation of the initial flux with the TDS concentration of each water type.

the need for the boron post-treatment and reduce operational costs. These processes of enrichment and depletion are mainly due to cation exchange, but other geochemical processes such as precipitation and dissolution of minerals (e.g., $CaCO_3$) occur during seawater intrusion into the aquifer and are partially responsible for these values.[10,39−41,46−49,43] It should be noted that higher pumping rates may reduce the residence time of seawater in the aquifer and thus reduce the inorganic compositional differences between SGW and seawater; however, this needs to be further explored.

As a result of mixing with the fresh groundwater, the osmotic pressure of the SGW was 15% lower than that of the seawater (Table 1), which is crucial for the efficiency of the desalination process. Lower osmotic pressure in SGW is highly beneficial due to reduced energy demand. The temperature of the groundwater (both FSI and SGW) was in the range of 22.4° to 26 °C throughout the study, while the seawater sampled varied from 18.6° to 28.6 °C; this variation in seawater temperature is in accordance with the literature, which indicated that Eastern Mediterranean water temperature experiences wide seasonal variability (16−30 °C).[50] Hence, SGW may need less temperature adjustment than seawater for desalination.[51] The pH of seawater was alkaline (8.2), whereas the SGW pH was around 7. The decrease in the pH is associated with the fact that the aquifer contain carbonate cementation (partly built of calcareous sandstone) while in siliciclastic aquifers it may not always occur. In large scale RO plants, the feedwater is usually acidified to pH 5−7 in order to prevent calcium carbonate scaling.[51] In that respect, the SGW would require lower addition of chemicals for pH adjustments. Moreover, the desalination efficiency increases at lower pH values,[52] giving SGW another advantage. Dissolved oxygen (DO) in seawater was at saturation levels (8.5 mg L^{-1}; Table 1), while the groundwater DO was reduced to about 65% from saturation (3.2 ± 1 mg L^{-1}). Typical DO values in saline groundwater are even lower, mostly less than 1 mg L^{-1}.[38,53] The DO is essential for aerobic bacteria, and thus high DO values in the feedwater might accelerate biofilm formation on the membrane.[54,55] Hence, SGW has an advantage because it is expected to present lower biofouling of the membranes in comparison to seawater.

The total organic carbon (TOC) concentration measured for SGW was 60% lower than that of seawater (0.63 mg L^{-1} and 1.64 mg L^{-1}, respectively). The mean TOC concentration from the Spanish Mediterranean coast measured by Sola et al. showed similar trend: The TOC in seawater was 0.76 mg·L^{-1}, while it was only 0.19 mg·L^{-1} in the nearby SGW.[15] TOC is used by heterotrophic bacteria as a substrate for biofilm growth.[56] Thus, the seawater possesses a greater potential for biofouling on the membrane with subsequent deterioration of the membrane performance.

Evaluation of SDI showed 30% lower index for SGW in comparison with seawater (4.6%.min^{-1} and 3.3%.min^{-1} for seawater and SGW, respectively; Table 1). To avoid membrane clogging, the commercial membrane manufacturers recommend an SDI value between 2 and 4 which gives clear advantage to SGW over seawater. The differences of DO, TOC, pH, temperature, SDI, boron concentrations and salinity between SGW and seawater render SGW more appropriate and advantageous for desalination, as less pretreatment and smaller energy demand are expected.

Microbial abundance and activity was determined in the SGW and in seawater. Overall, the microbial abundance was higher in the seawater than in the SGW (Table 2). Chlorophyll *a* level was two- to 3-fold higher in the seawater than SGW, and picophytoplankton (cyanobacteria and small-size eukaryotes) abundances followed the same trend. Heterotrophic bacterial

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abundance in the seawater was moderately higher than in the SGW (1.5 fold; see Table 2). In contrast to the higher microbial abundances found in the seawater compared with the SGW, bacterial productivity remained unchanged in all samplings. Primary productivity rates of seawater were 3-fold higher than SGW during the summer and ∼50% lower during winter samplings. Concurrent with the similar bacterial productivity rates and abundance, bacterial diversity based on 16S analyses showed similar composition in both water types with dominance of proteobacterial phylotypes (>80% of all OTUs; Table S2, SI). The lower microbial abundance and activity in the SGW than the seawater may pose further advantages for SGW desalination, especially due to the lower autotrophic biomass and activity (Table 2) which may restrain membrane biofouling in RO operations.

RO Desalination of SGW. Desalination experiments in RO cross-flow system were performed for FSI water, SGW and seawater to evaluate the desalination process parameters such as salt rejection and permeate flux of the three types of water. Prior to studying the different water types, a comparison of three types of commercial seawater-RO membranes was conducted with SGW as a feed, to choose the most appropriate membrane for subsequent studies. The permeate flux in the RO filtration experiments with SW30-HR and SWC-1 membranes was lower than that of the Toray TM820-400 membrane (30 ± 1.6 $L{\cdot}m^{-2}\ h^{-1}$, 31.7 ± 0.6 $L{\cdot}m^{-2}\ h^{-1}$, and 43.5 ± 1.8 $L{\cdot}m^{-2}\ h^{-1}$, respectively; Figure 2a). The permeate flux of TM820−400 was about 30% higher than that of the other two membranes, giving it an advantage. The membrane with the best salt rejection property was SW30-HR membrane, with 99% rejection. The salt rejection of TM820-400 was 98% and of SWC-1 was 95.2%. Taking the results of permeate-flux and salt rejection into consideration, the TM820-400 membrane was chosen for further experiments with SGW.

RO desalination experiments of the three water types (seawater, FSI, and SGW) were performed using the TM820−400 membrane, and the obtained membrane permeabilities are shown in Figure 2b; as expected, the membrane permeabilities using FSI feedwater and SGW feedwater were higher than that of the seawater. Under pressure of 50 bar, the average permeate flux in desalination of groundwater from the FSI was 75.3 $L{\cdot}m^{-2}{\cdot}h^{-1}$, from the SGW the average permeate flux was 49.9 $L.m^{-2}{\cdot}h^{-1}$ and the seawater showed 40.7 $L{\cdot}m{\cdot}h^{-1}$. The average membrane permeabilities were 1.55, 1.02, and 0.83 $L{\cdot}m^{-2}{\cdot}h^{-1}bar^{-1}$, respectively. The salinity of the FSI groundwater is ∼30% of that of the seawater, and SGW salinity is about 85%. The permeate flux in the filtration experiments using FSI water was 82% higher than seawater, and SGW was 19% higher than seawater flux. A linear correlation between the initial permeate flux and the feedwater salinity was obtained (insert, Figure 2b).

Although the FSI water yielded the highest permeability among the water types investigated, FSI is not considered a viable source for desalination because there is a small volume of water in the mixing zone, and continuous pumping from the FSI is expected to shift the mixing zone to a different location in the aquifer. Moreover, constant pumping from the FSI zone may draw fresh groundwater from above and have a detrimental effect on the freshwater aquifer. Therefore, subsequent experiments were focused on desalination of SGW beneath the FSI zone.

Fouling Potential and Seasonal Variations of the Water Types. Fouling experiments were conducted in the RO cross-flow system with a Toray TM820−400 membrane to evaluate the fouling potential of SGW and seawater. The experiments were conducted with SGW sampled from the Hilton Hotel (Tel Aviv) pumping well, and with seawater that was sampled from the intake pipe of the desalination plant in Ashkelon (prior to treatment), both in summer and in winter.

The results from the winter sampling sessions showed that permeate flux fluctuated around its initial value without declining in both seawater and SGW (Figure 3a). On the other hand, the summer sampling session showed different behavior for SGW and seawater: SGW initial flux was 41.8 $L{\cdot}m^{-2}{\cdot}h^{-1}$ which, after about 10 h, declined to an average flux of 38.1 $L{\cdot}m^{-2}{\cdot}h^{-1}$, that is, a 9% decrease (Figure 3b). The seawater permeate flux decreased by about 15% during the first few hours from initial flux of 40.6 $L{\cdot}.m^{-2}{\cdot}h^{-1}$ and stabilized around the average value of 34.2 $L{\cdot}m^{-2}{\cdot}h^{-1}$. Hence, flux decline in seawater was more pronounced than in SGW, which may be explained by larger amount of foulants in the seawater. Permeate flux decline of seawater during the summer sampling are in accordance with higher SDI value for the seawater than SGW (Table 1). RO desalination of SGW showed less fouling than seawater desalination, therefore it is more efficient. The permeate flux measured for SGW in both winter and summer was higher than for seawater (as was also shown in Figure 2b).

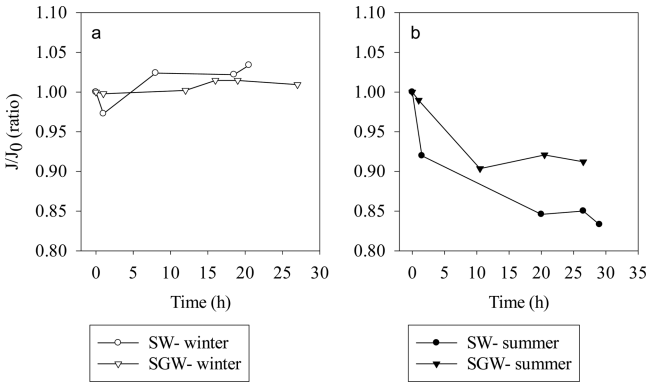

Figure 3. Comparison of the normalized flux of SGW and seawater sampled during winter (a) and summer (b) in RO filtration cross-flow system using TM820-400 membrane. Experiments were conducted under 50 bar, showing SGW initial permeate flux of 33.3 $L{\cdot}m^{-2}h^{-1}$ in winter, and 41.8 $L{\cdot}m^{-2}h^{-1}$ in summer; seawater initial permeate flux was 30 $L{\cdot}m^{-2}h^{-1}$ in winter and 40.6 $L{\cdot}m^{-2}h^{-1}$ in summer.

The differences in the flux behavior between the summer and the winter in both SGW and seawater may be associated with an increase in the microbial population during the summer. According to Yang et al. (2010)[57] the extracellular polymeric substances (EPS) secreted by microorganisms in the summer is much more detectable. The abundant EPS enhance fouling, mainly organic matter adsorption on the membrane surfaces, and thus there is higher fouling on the membrane in summer. In addition, high ionic strength might increase organic matter precipitation (accumulation) on surfaces;[58] the SGW ionic strength is 0.0125 mol/mol, lower than the seawater ionic strength (0.0136 mol/mol), showed 9% reduction while seawater flux reduced by 15%.

Figure 4 shows the specific ion rejection of a few major ions. It can be seen that the ion rejection with the SGW was slightly higher than that of the seawater, yet these differences were not very significant. Higher salt concentration in the feedwater results in higher salt passage through the membrane.[59] In the

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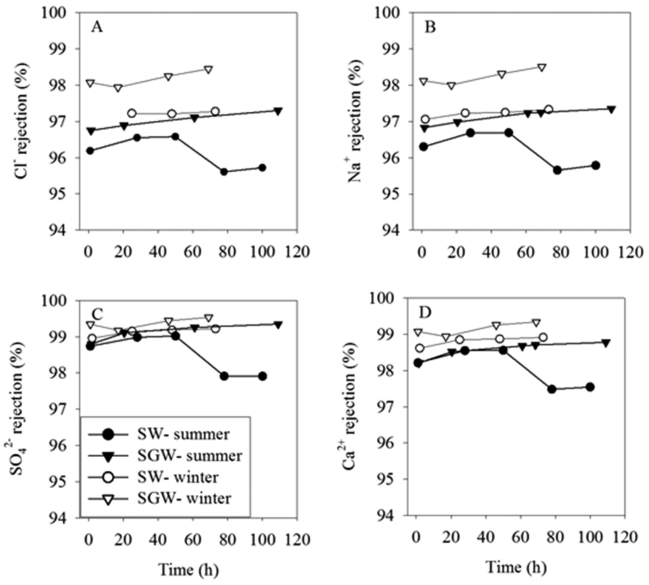

Figure 4. Major cation and anions rejection throughout the desalination process for SGW and seawater at winter and at summer.

summer experiments, the average Ca^{2+}, Na^{+}, SO_4^{2-}, and Cl^{-} rejection for SGW was 98.5%, 97.1%, 98.6%, and 96.5%, respectively. The rejection of seawater for the same ions for the first 50 h was 98.4%, 96.6%, 98.7%, and 96.5%, respectively. After 50 h, a decline in salt rejection was noted in the seawater experiments. The Ca^{2+}, Na^{+}, SO_4^{2-}, and Cl^{-} rejection declined to 97.5%, 95.9%, 97.6%, and 95.6%, respectively. This could be explained by enhanced trans-membrane osmotic pressure of the fouled membrane due to a biofilm layer growing on the membrane surface. The increase of the trans-membrane osmotic pressure is attributed to the (bio)fouling layer (deposited bacterial cells, EPS and organic matter) which increased the concentration polarization of salt near the membrane surface, and therefore increased the salt passage through the membrane.[60]

Boron rejection in desalination of seawater was 80%, almost similar to the rejection obtained in SGW desalination (79%). However, it is pertinent to note that the SGW possess 30% lower boron concentrations than seawater (Table 1), therefore SGW permeate is expected to possess reduced boron content, which is another advantage of using SGW as feedwater.

The winter experiments showed a different trend regarding salt rejection; the salt rejection of the SGW in the winter was still higher than that of seawater, but both were higher in comparison to the summer experiments. The ion rejection in seawater experiments for Ca^{2+}, Na^{+}, SO_4^{2-} and Cl^{-} was 98.8%, 97.2%, 99.1%, and 97.6%, respectively, and using SGW was 99.3%, 98.5%, 99.4%, and 98.4%. The distinct results of summer and winter could be explained by the temperature, which plays a role in salt passage: The salt rejection decreases with increase in temperature due to changes in membrane permeability and mass transfer.[61,62] Therefore, rejections in winter were generally higher than summer (Figure 4).

Implications. This work shows that using saline groundwater as source for desalination in coastal aquifers may be highly beneficial in comparison to desalination of seawater. The aquifer media filtration increases the feedwater quality and reduces the need for extensive pretreatment processes.[13] Normally, seawater RO desalination plants occupy large areas due to the need for large pretreatment facilities. Such vacant land is difficult to find in densely populated regions close to the shore. Desalination facilities for saline groundwater require small pretreatment facilities and thus occupy a smaller footprint. The availability of wells along the coastal areas allows for shorter transport distances between the source water and the desalination facility, as well as the end user of water.

There are seasonal changes of the seawater characteristics and quality for desalination. Seawater quality in summer deteriorates, while SGW quality remains the same. Hence, using SGW in summer is preferable.[63] The use of SGW has further advantages over using seawater as feedwater, such as a constant annual water temperature, lower SDI, lower dissolved oxygen, lower pH, and lower phytoplankton abundance and productivity. The lower salinity and lower fouling potential are important advantages, and are expected to yield higher permeate flux and more stable one. In conclusion, RO desalination process with SGW as feedwater is expected to be more efficient, with higher recoveries, lower use of chemicals, lower maintenance, and therefore lower overall operational costs.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b03634.

Location map of the study area, indicating the water sampling sites; general cross section of the coastal aquifer; chemical and physical parameters of the specific source water used for the fouling experiments; relative abundance of proteobacterial phylotypes in the source water and on the RO membrane; replicate of RO cross-flow fouling experiments of seawater, SGW and FSI water (PDF)

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Notes

The authors declare no competing financial interest.

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