Chapter 6

Marmara Seawater Desalination by Membrane Distillation: Direct Consumption Assessment of Produced Drinking Water

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Abstract

Drinking water was produced from Marmara seawater by membrane distillation (MD). The best operating conditions were determined by batch experiments as: 0.45 μm PTFE, 30°C distillate temperature and temperature difference, and 270–360 L/h cross-flow rates in feed-distillate. Seawater desalination was carried out with 99.93% solute rejection and 17.2 L/m^{2}h permeate flux in 66% concentration ratio by lab-scale pilot system. Since the desalinated water contained no organic carbon, turbidity, and nitrate, it seemed to be very suitable for immediate service with quality of 7.3 pH, clear, odor-free, 76.0 μS/cm, 47.1 mg TDS/L, <0.001 color, and 0.01 mg boron/L. The product water lacked of vital cations, especially Na^+, K^+, Ca^{2+}, Mg^{2+} that are essentials for promoting osmotic balanced body liquid and healthy development. A holistic management approach towards satisfying specific water quality requirements in direct service of MD effluents to human consumption was proposed that jointly included in injecting into urban potable water, adding appropriate chemicals into the effluent, and mixing effluents with raw or concentrated seawater (1:250/1:1000 for Marmara seawater) or brackish natural waters under hygienic precautions.

Keywords: seawater desalination, drinking water production, membrane distillation, product water quality, human consumption suitability

1. Introduction

The data of United Nations indicate that the rate of increase in water usage has been higher than twice the rate of increase in population over the past century. It is estimated that there

will be 1.8 billion people living in areas with scarce water resources by 2025, while two-thirds of the global population reside in regions of water stress due to use, growth, and climate change [1]. Potable water production has also become a worldwide concern, for many communities, increasing of industrial processes, population growth, climate change, over exploitation of ground water and nearby river systems as well as demand exceed of conventional available water resources [2–7]. For firmly preservation and sustaining stable development of life on earth at present situation, there needs to use plentiful salty water to produce freshwater supplies capable of meeting the increasing demand [7, 8]. Therefore, countries are progressively turning to desalination technologies as a solution to obtain direct potable water from seawater [4, 6, 9].

Commercial desalination technologies are mainly categorized under two main categories as pressure-driven membrane separation by reverse osmosis (RO) and thermal distillation by multi-stage flash (MSF) and multi-effect distillation (MED) [4, 10]. In spite of their growing popularity and improved technological applications in seawater desalination, available plants have potential negative impacts on coastal environments and marine ecosystems, which can be characterized with concentrate and chemical discharges to the sea, limiting effects on land use, salting of groundwater, noise pollution, air pollutants discharges to the atmosphere and high energy consumptions [11–17]. Total capacity of seawater desalination plants in the world about 50% is operated by RO-based membrane technology [4]. The decline in RO performance resulting from unavoidable membrane fouling, which needs expensive pretreatment, higher operation pressures, and frequent cleaning with chemicals which give harm to membranes, impairs the quality of permeate and accelerate membrane replacement, which pushes up the cost of water treatment and energy consumption [6, 18].

Membrane distillation (MD) could be an alternative to RO desalination process to overcome its negative impacts. MD is not highly affected by salt concentration in saline feed solutions, and hence, this technology can achieve good quality distillate with minimal brine discharge [19, 20]. MD, a thermal integrated membrane process, incorporates transporting vapor through microporous hydrophobic membranes, and its operation is based on the principle of vapor-liquid equilibrium as a basis for molecular separation. This process uses a gradient of temperature between the two sides of a porous membrane in order to establish a difference in vapor pressure that actually drives process [5, 19, 21–25]. Its four different configurations that consist of the type of the condensing design have been proposed, but direct contact membrane distillation (DCMD) is the most widely preferred technology because the step of condensation is performed within the membrane module, which brings about a simple mode of operation with no need of external condensers [19, 22, 24]. Additionally, more convenient membranes in MD process became available. Several materials such as polytetrafluoroethylene (PTFE), polypropylene (PP), polyethylene (PE), and polyvinylidene fluoride (PVDF) are used for producing hydrophobic MD membrane for desalination uses [24, 25]. The different commercial membranes have been used in several recent pilot plant studies, but there are still some problems that have not yet been clearly solved to use MD process under real scale [19].

In practice, the use of MD for desalination is still largely limited to pilot-scale studies. The commercial launch of MD desalination technology will first require certain technical issues to be resolved, such as its high energy consumption and the wetting of membrane pores [26]. Feng

et al. [27] used different nanofiber microfiltration membranes produced from PVDF material to obtain drinking water through the air gap MD process. This was the first time that an electrospun nanofiber membrane was used in MD. At temperature differences ranging between 25 and 83°C, they obtained flux values between 5 and 28 kg/m^2 h. They also reported that this new approach could potentially compete with other conventional desalination systems. Fard et al. [28] investigated bench-scale performance of DCMD process using flat sheet PTFE membrane under various conditions of inlet flow rates, temperatures, and salinity composition. A permeate flux of 35.6 L/m^2 h could be generated at a different temperature of 50°C between hot and cold stream sides. The rate of salt rejection during the conducted tests was really high with 99.9% and virtually independent of any operational parameters studied. It has been seen that DCMD is a viable and effective technology, which can produce high quality distillate in a consistent way from a very high salinity feed even with dramatic differences in quality compared to other methods of desalination like RO and MSF. Bouguecha et al. [29] attempted to run a DCMD pilot plant powered by solar energy using collectors plus PV panels under actual weather conditions in Jeddah, KSA, throughout two selected sunny days. It had been aimed to assess how the operating parameters affected the process performance in which the transmembrane temperature difference (ΔT) and the fluid mass flow rates (hot and cold) constituted the most remarkable operating parameters. A maximum permeate flux of 8.87 L/m^2 h was achieved at a ΔT of 60.5°C. According to their findings, it seems that the DCMD is a promising solution for the desalination of brackish water and also for seawater, particularly in distant places and/or whenever affordable low temperature sources are accessible.

In this paper, six different commercially available membranes were used to investigate the effects of different operating conditions including cross-flow rate, membrane type and pore size, solution temperatures, and membrane trans-temperature differences on dissolved ions rejections and permeate flux of DCMD process. Some characteristics of membranes such as roughness and wettability were additionally tested to more comprehensively understand the performances. At suitable operation conditions determined based on batch experimental runs, the process was operated along a 30-h period of time in which raw seawater was concentrated at approximately 70% to examine the direct usability of the MD output water as drinking water. The effluent quality established for direct supply to human consumptions was evaluated in terms of appropriateness to maximum allowable concentrations in the national standard and international guidelines, and a general framework for pragmatic solutions toward practical water quality management applications was proposed for facilitation to serve the MD desalination effluents to the human drinking directly.

2. Materials and methods

2.1. Seawater characterization

Raw seawater used as feed stream in all membrane distillation experiments was collected from Muallimköy Coast of Marmara Sea beside the coastal city of İzmit/Kocaeli in Turkey. It was taken from under one meter of seawater level and then pre-filtered through roughing filtration to remove large particles. The detailed characteristics of the seawater are given in **Table 1**.

Parameters[a]	Raw seawater		
	I[b]	II[b]	Average
T	24.1	24.2	24.15 ± 0.05
pH	8.69	8.62	8.66 ± 0.04
Ec	40,400	41,500	40,950 ± 550
TDS	25, 250	25, 772	25,511 ± 261
TOC	4.3	5.1	4.7 ± 0.4
DOC	4.1	4.9	4.5 ± 0.4
UVA_{254}	0.050	0.040	0.045 ± 0.005
SUVA	1.220	0.816	1.018 ± 0.202
Na^+	8880	8940	8910 ± 30
K^+	443	422	433 ± 11
NH_4^+	<0.1	<0.1	<0.1
Ba^{2+}	0.030	0.029	0.0295 ± 0.0005
Ca^{2+}	395	404	400 ± 5
Mg^{2+}	696	692	694 ± 2
Mn^{2+}	0.018	0.019	0.0185 ± 0.0005
Sr^{2+}	5.50	5.70	5.60 ± 0.10
B	2.95	3.05	3.00 ± 0.05
Si	0.89	0.91	0.90 ± 0.01
Fe	0.007	0.007	0.007 ± 0.000
Cl^-	13,996	14,481	14,239 ± 243
HCO_3^-	300	303	302 ± 2
NO_3^-	0.5	0.5	0.5 ± 0.0
CO_3^{2-}	0.0	30	15.0 ± 15.0
SO_4^{2-}	2314	2440	2377 ± 63
TKN	0.6	0.7	0.65 ± 0.05
Total nitrogen	1.1	1.2	1.15 ± 0.05
Alkalinity	300	333	317 ± 17
Total hardness	4986	5236	5111 ± 125
Color			
436 nm	0.0300	0.0300	0.0300 ± 0.0000
525 nm	0.0020	0.0100	0.0060 ± 0.0040
620 nm	0.0030	0.0020	0.0025 ± 0.0005

[a]Units of all parameters are mg/L except for temperature (°C), electrical conductivity (μS/cm), UVA_{254} (1/cm), SUVA (L/mg.m) and color (1/cm).

[b]Seawaters I and II are raw seawater samples that were used in batch and continuous MD experiments, respectively.

Table 1. Characteristics of Marmara seawater.

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2.2. MD membranes

Six different types of flat sheet hydrophobic microfiltration membrane with pore sizes of 0.2, 0.45, and 1.0 μm that were made of PTFE and PVDF materials (Membrane Solutions Inc., China) were used in the experiments. The characteristics and intrinsic performances of the membranes are presented in **Table 2**. The membrane thicknesses were provided by the manufacturer. The results of contact angle measurement below 90° indicate that the PVDF membranes demonstrated lower hydrophobicity than the PTFE membranes. Liquid entry pressure (LEP) parameter also shows that the PTFE membranes are more suitable for the MD processing of seawater due to its lower wettability and higher entry pressure values.

2.3. Experimental setup

The experimental setup of cross-flow lab-scale pilot DCMD system is schematically shown in **Figure 1**. The setup was composed of two thermostatic cycles, that is, feed and permeate, which were connected to a membrane module made by kestamid having an effective membrane area of 140 cm^2. The compartment cells of the module consisted of two machined parts compressing the rectangular MD membrane. Connected to a heating resistant, the feed flow side is kept under high temperature; on the other hand, the permeate flow side, connected to a cooling system, is maintained at a low temperature.

During the experimental investigations, rough-filtrated raw seawater from the Marmara Sea was preheated to intended temperatures and circulated through one side of the membrane (feed side), while de-ionized water was circulated through the other side of the membrane (distillate or permeate side) in a simultaneous way in a counter-current flow mode. The condensation of permeate vapor that was diffused across the membrane occurred in the cold distillate side. The temperatures of feed and permeate streams were controlled at desirable levels using a heater and chiller, respectively. The spacer placed in the feed flow side only to promote turbulence

Membranes	Material	Membrane thickness (μm)	Nominal pore size (μm)	Contact angle (±10°)	Liquid entry pressure (±2.5 kPa)	Water flux[a] (m^3/m^2h)
PTFE	Polytetrafluoro ethylene	160 ± 40	0.22	121	121.3	0.250
			0.45	126	81.1	0.333
			1.0	123	131.4	0.417
PVDF	Polyvinylidene fluoride	100 ± 10	0.22	68	58	6.0–9.0
			0.45	81	47.5	22.2–36.0
			1.0	84	22.5	–[b]

[a]The values given by manufacturer that measured at trans-membrane pressures of 0.2 and 1.0 bar for PTFE and PVDF membranes, respectively.

[b]Not available.

Table 2. Various properties of MD membranes used.

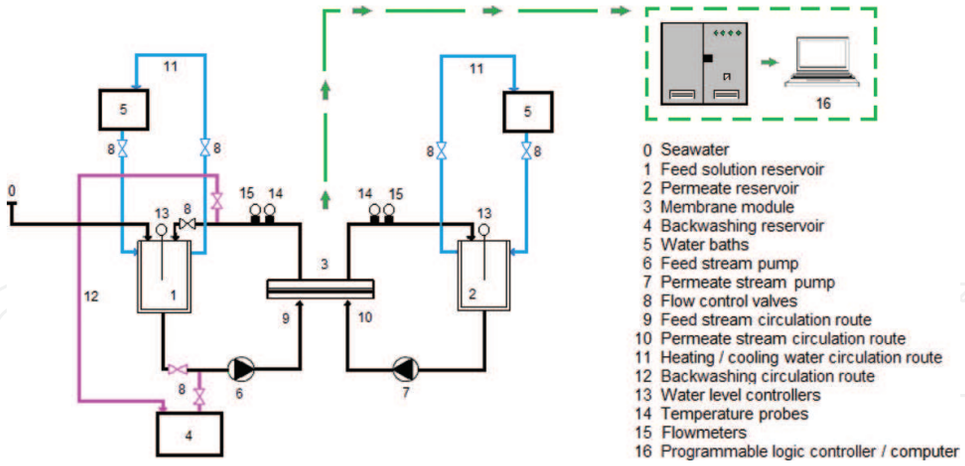

Figure 1. Schematic flow diagram of experimental MD setup.

and to support the membrane was obtained from Sterlitech Inc., USA. In the batch experiments, the storage tanks for the feed and distillate streams in the system were kept at equal volumes of five L. In the continuous experiments operated at concentration mode of the feed seawater of 12 L, the system was worked for 6 hours per day in a period of 5 consecutive days. During these experimental runs, the membrane active layers were cleaned in-place inside the module at the end of each operation day by means of using 1 L solutions in each one of the applications, which were operated by a flushing order of first applying "*1% HCl + distilled water*", and then followed by "*1% NaOH + distilled water*" except for the first operation day.

2.4. Surface morphology of membranes

The surface morphologies of clean and fouled membranes were marked by means of atomic force microscopy (AFM) to assess the vertically distribution of the fouling on the top layer or membrane surface roughness. After each membrane sample was let dry in air, the visual observations were provided using NanoScope IV AFM system (Digital Instruments, USA) operated in contact mode. The mean roughness (R_a), the root mean square (R_{rms}) of the average height of membrane surface peaks, and the mean difference in height among the five highest peaks and the five lowest valleys (R_z) were identified in order to compare the roughness of clean and fouled membranes. Due to its more representative findings for the surface foulings, the analyses were evaluated only via the variations of the measured R_z values of the specimens. In the AFM analyses, the value of mean roughness (R_a) stands for the mean value of surface in relation to centre plane and was calculated using the following equation [30]:

$$R_a = \frac{1}{P}\sum_{i=0}^{p}\left|z_{cu} - z_{av}\right| \quad (1)$$

where z_{av} is the average of the z values in the particular area, z_{cu} is the present value of z, and p refers to the number of points in a certain area. The root mean square of z values (R_{rms}) was calculated using the equation given below [30]:

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$$R_{rms} = \sqrt{\frac{\sum \left(z_{cu} - z_{av}\right)^2}{p}} \quad (2)$$

The average difference in height among the five highest peaks and the five lowest valleys, R_Z, was calculated in relation to the mean plane, on which the image data have a minimum variance [30, 31]. The cross-sectional morphologies of fouled membranes were monitored by scanning electron microscopy (SEM) (Philips XL30 SFEG) to visualize the fouling formations on the membrane surface. All the SEM observations were performed at 5 kV using Au-coated membrane specimens. Energy dispersive X-ray spectrometry (EDX) analysis of the components found on the membrane surface after the fouling was also done with EDX detector of the same device. The hydrophilicity or wettability of the membrane surfaces was analyzed with contact angle (°) measurements according to sessile-drop technique using a goniometry instrument (Attension Theta Lite Optical Tensiometer) [31]. In the analyses, 2 μL of pure water in tight syringe was manually dropped on the membrane surface. By means of the software processing the measurement data obtained from the camera of the device, the results were determined as the averages of contact angles at both sides of drops fall on five arbitrary places of the membrane surfaces.

2.5. Analytical procedure

Water quality analyses in both MD streams were carried out based upon the parameters that included in temperature, pH, conductivity, turbidity, total dissolved solids TDS, total organic carbon (TOC), dissolved organic carbon (DOC), ultraviolet absorbance at 254 nm (UVA_{254}), specific ultraviolet absorbance (SUVA), total Kjeldahl nitrogen (TKN), NH_4^+, alkalinity, total hardness, Na^+, K^+, NH_4^+, Ba^{2+}, Ca^{2+}, Mg^{2+}, Mn^{2+}, Sr^{2+}, B, Si, Fe, Cl^-, HCO_3^-, NO_3^-, CO_3^{2-}, SO_4^{2-} and color at 436, 525, and 620 nm wavelengths that were measured according to "*Standard Methods for the Examination of Water and Wastewater*" [32].

Temperature, conductivity, TDS and pH were analyzed with the desktop multi-parameter with Hach HQ440 d (Hach-Lange GmBH). TOC was measured at 750°C by carbon analyzer equipped with a high pressure NDIR detector (Hach Lange IL550 TOC-TN) in which *5310 B-a high temperature catalytic oxidation method* was applied. DOC analyses were conducted on the samples filtered by Whatman syringe filter 0.45 μm using the TOC analyzer apparatus. UVA_{254} was measured by *5910 B-UV-absorbing organic constituents method,* and SUVA was calculated with values of UVA_{254} and DOC. TKN was analyzed by *4500-N_{org}B-macro-Kjeldahl method*, and ammonia was determined using the measurement probe of Hach HQ440 d by *4500-NH_3E-ammonia-selective electrode method*. Alkalinity and total hardness were determined in accordance with *2320B-titration method* and *2340C-EDTA titrimetric method*.

Na^+, K^+, Ba^{2+}, Ca^{2+}, Mg^{2+}, Mn^{2+}, B, and Fe concentrations were measured by Perkin-Elmer ELAN Optima 7000 DV with *3120-B inductively coupled plasma mass spectrometer* (ICP-MS) (Perkin-Elmer SCIEX Instruments, Canada). Si and Sr^{2+} were measured using flame atomic absorption spectrometer (Perkin Elmer 1100). Prior to the analyses, samples were filtered through a

0.45 μm pore-size membrane filter, and pH values of the samples were adjusted to <4.0 using HNO_3. $HCO_3^{2-}$and CO_3^{2-} were analyzed by *2320-B titrimetric method*, and SO_4^{2-} and Cl-were measured by *4500-SO_4^{2-}-E turbidity and 4500-Cl^--D potentiometric methods*, respectively. NO_3^{2-} was determined by *4500-NO_3^{2-}-C* spectrophotometric method, while color measurements at 436, 525, and 620 nm wavelengths were conducted by *2120C* spectrophotometric method for making possible the time-dependent calculations of MD water fluxes, the measurements of osmolalities of the feed and permeate solutions were executed at definite time intervals in duplicate for each data point using the Advanced Osmometer instrument (Model 3250—Advanced Instruments Inc., USA) by the method of freezing point depression.

2.6. MD performance analysis

The technical performance of MD was analyzed using not only the permeate flux values but the rejection values of the parameters as well, which are given in national and international guidelines for water reuse in drinking water quality. The flux was calculated from the time-dependent variations between the pretest and posttest values of TDS and osmolality as an explicit indicator for solute ions transports from seawater to distillate stream. So, after TDS and osmolality at both sides were individually measured for definite time intervals, V was calculated from the differences of sequential time points according to TDS/osmolarity-based mass balances calculations. From the average values of V volume calculated based on TDS and osmolality data measured at each time point, permeate water fluxes were totally obtained for the whole time scale of relevant experiment using Eq. (3).

$$J = \frac{1}{A}\frac{dV}{dt} \tag{3}$$

where J is the permeate flux; A is the effective membrane filtration area; V is the total permeate volume; and t is the filtration time.

The rejection performances in MD process for each water quality parameter were calculated from the water quality analyses results using Eq. (4):

$$R(\%) = \left(1 - \frac{C_p}{C_f}\right) \times 100 \tag{4}$$

where R, C_p, and C_f are the rejection, concentration in the permeate, and concentration in the feed stream, respectively [33].

3. Results and discussion

3.1. The effects of operating conditions on MD performance

3.1.1. Membrane type and size

The first part of the study was performed under batch experimental runs by using different membranes to examine the effect of different pore sizes and materials on the MD performance.

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The experimental performance levels were determined based on the passing water volume, the seawater concentration, and the water flux parameters. Initial experiments were performed with a 30°C difference between the solution temperatures, which are the generally recommended level of difference in the literature [21], by maintaining seawater and permeate water at temperatures of 55 and 25°C, respectively, and by using seawater and permeate water flow rates of 270 and 360 L/h, respectively. In general, available polymeric materials for manufacturing hydrophobic membranes suitable for MD are, typically, PP, PVDF, and PTFE [19, 21].

With PTFE membranes, water flux values for 0.22, 0.45, and 1.0 μm sized pores were determined as 27.7, 40.7, and 28.5 L/m^2 h, respectively. With PVDF membranes, these values were determined as 11.3, 19.4, and 29.6 L/m^{2}h, respectively. TDS values in output water for the PTFE membranes with 0.22, 0.45, and 1.0 μm pore sizes were determined as 4.3, 7.9, and 9.8 mg/L, respectively. **Figure 2** shows permeate flux and solute concentration for the MD membranes with different materials (PTFE and PVDF) and pore sizes (0.22, 0.45, and 1 μm) under batch operating conditions.

An evaluation of the permeate solute ion concentrations with the three different pore sizes revealed that increasing pore size led to higher concentrations in the permeate. Based on the observed water flux performances, it was determined that the PTFE membrane with 0.45 μm pore was the most suitable type of membrane for obtaining potable water from seawater. The PTFE (0.45 μm) membrane was hence chosen for the next stage.

Distillate contamination resulting from wetting of membrane pore is among the chief factors, which impede the broader application of the MD technology. In order to avoid pore wetting, it is necessary that the membrane material be hydrophobic with as high a contact angle as possible. Moreover, it is recommended that the membrane has a comparatively small maximum pore size [34]. The hydrophobic membrane can obstruct the penetration of liquid through surface tension force, but not that of vapor. Consequently, water vapor will be capable of passing from the hot solution side of higher vapor pressure to the cold distillate side of lower vapor pressure [35]. **Figure 3** shows measurement results of contact angle (θ°) and surface roughness (R_z) for surface characterizations of clean (θ_c and $R_{z,c}$) and fouled (θ_f and $R_{z,f}$) membranes.

Results of AFM and SEM evaluations performed before and after filtration on the active layers of the MD membranes are shown in **Figure 4**. The high pore sizes used in the experiments led to greater accumulation of inorganic pollutants inside the pores than on the surface. In addition, the surface fouling layer was observed to be soluble. Consequently, no significant differences were observed on the SEM images. An evaluation of the EDX results revealed that, depending on the structure of polymers used for producing the membranes, and the added chemicals, the membrane surfaces contained the elements boron (B), fluorine (F), carbon (C), nitrogen (N), and oxygen (O). It was observed that the fouling of the surface was largely local and lacked a large variety of ions, and when a prevalent distribution of ions was actually present, this was found to be due to the crystallized precipitates ($CaCO_3$, $CaSO_4$) formed under the effect of temperature [36]. Inorganic foulant materials were not observed on the membrane surfaces. It is hence possible to say that no different elements were observed on

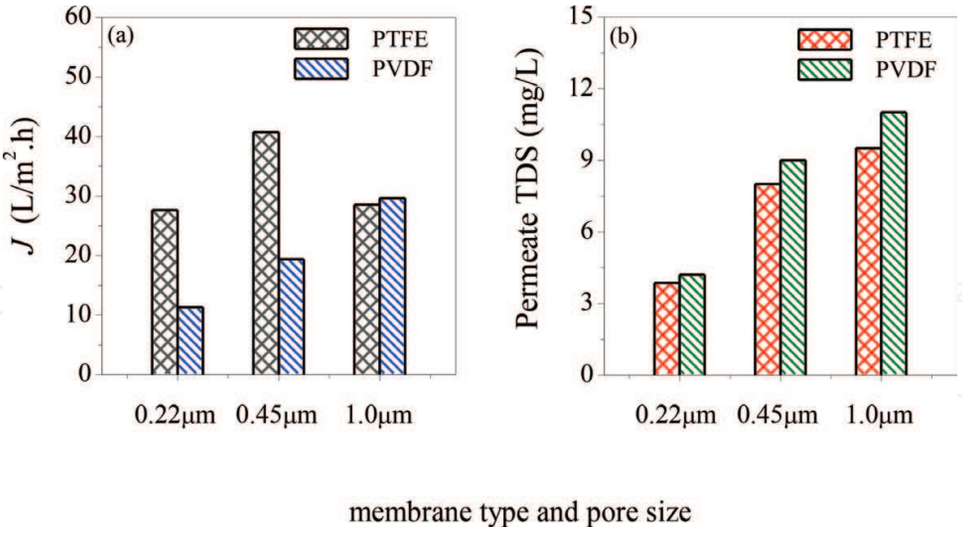

Figure 2. Permeate flux (a) and solute concentration (b) for MD membranes with different materials (PTFE and PVDF) and pore sizes (0.22, 0.45 and 1.0 μm) under batch operating conditions (seawater/permeate: 55/25°C (ΔT: 30°C) and 270/360 L/h cross-flow rates at seawater/permeate streams).

the membranes at the end of the process. The presence of such low and dilute levels of fouling during the experiments clearly indicated that under effective pass vapor pressure and low levels of pollution, the MD membrane is fairly efficient in providing drinking water from seawater.

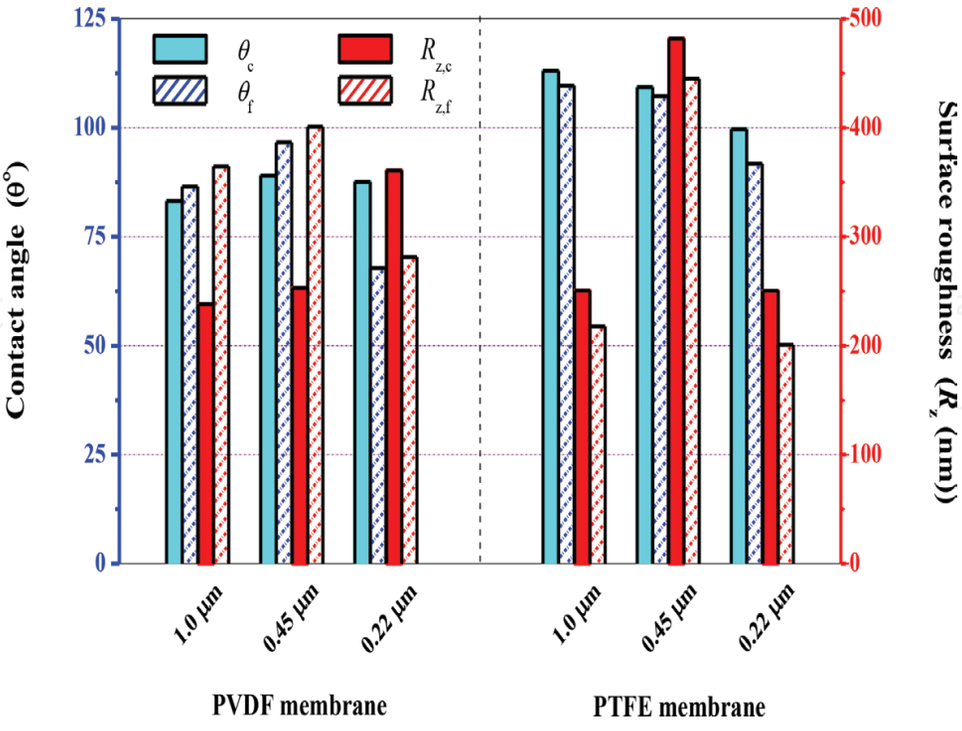

Figure 3. Measurement results of contact angle (θ°) and surface roughness (R_z) for surface characterizations of clean (θ_c and $R_{z,c}$) and fouled (θ_f and $R_{z,f}$) membranes.

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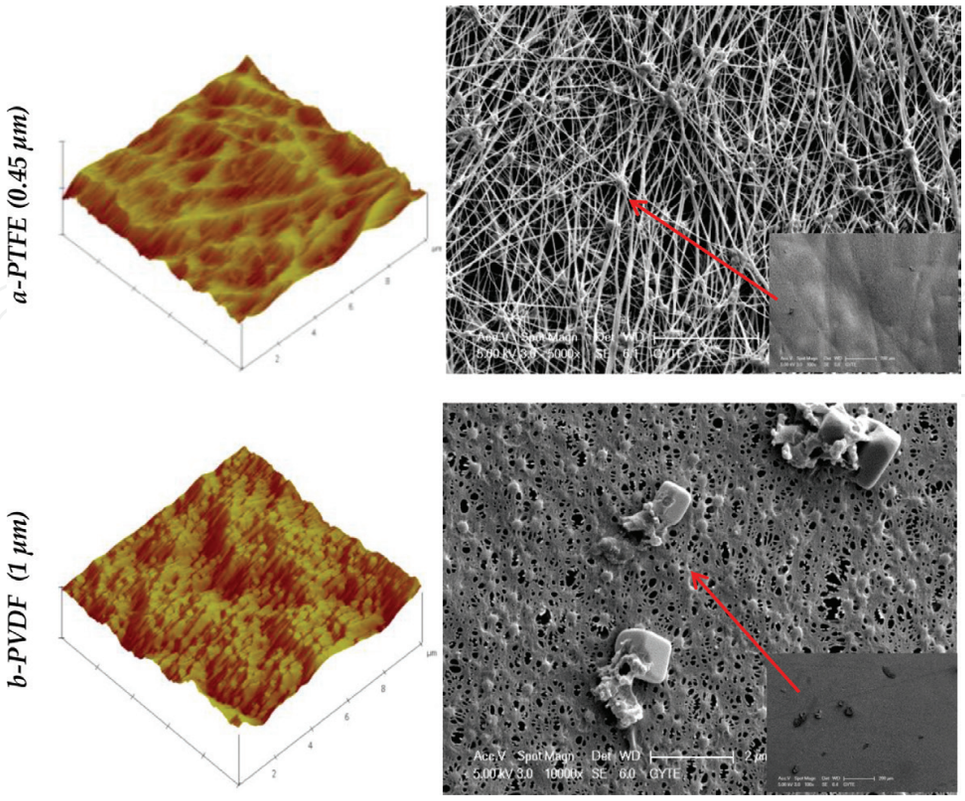

Figure 4. AFM (10 μm × 10 μm) images and SEM (1.4 mm × 0.9 mm) microphotographs of fouled membranes (PTFE-0.45 μm and PVDF-1 μm).

3.1.2. Cross-flow rate

The effect of the flow rates in membrane channel during the MD process was evaluated by applying flow rates with different Reynolds values (90/120, 180/240, 270/360, and 360/480 L/h seawater/permeate water flow velocity values) through a PTFE membrane at a fixed membrane trans-temperature difference of 30±0.5°C. It was observed that increasing flow rate led to a higher volume of water passing from the seawater to the permeate flow, which resulted in an increase in permeate flux performances. With increasing flow rates, water flux levels consecutively reached 23.4, 29.9, 40.7, and 43.4 L/m^2 h. Output water TDS values were determined as 5.3, 6.0, 7.9, and 9.75 mg/L, respectively, and output water with low solute ions was obtained in all flow velocity experiments. **Figure 5** shows permeate flux and salt concentration for different cross-flow rates (L/h) at seawater/permeate water under batch operating conditions.

Under the turbulence regime, similar performance values were observed in the 270/360 L/h and 360/480 L/h seawater/distilled water flow rate pairs. Taking into account that increasing flow rates might increase costs in practice, suitable flow rates were determined as 270 L/h (Re_{av}: 4320) for seawater and 360 L/h (Re_{av}: 4222) for permeate water.

3.1.3. Solutions' temperatures

In the next part of the study, experiments were conducted with 0.45 μm PTFE membranes, a fixed membrane trans-temperature difference value (30°C), a seawater and permeate water

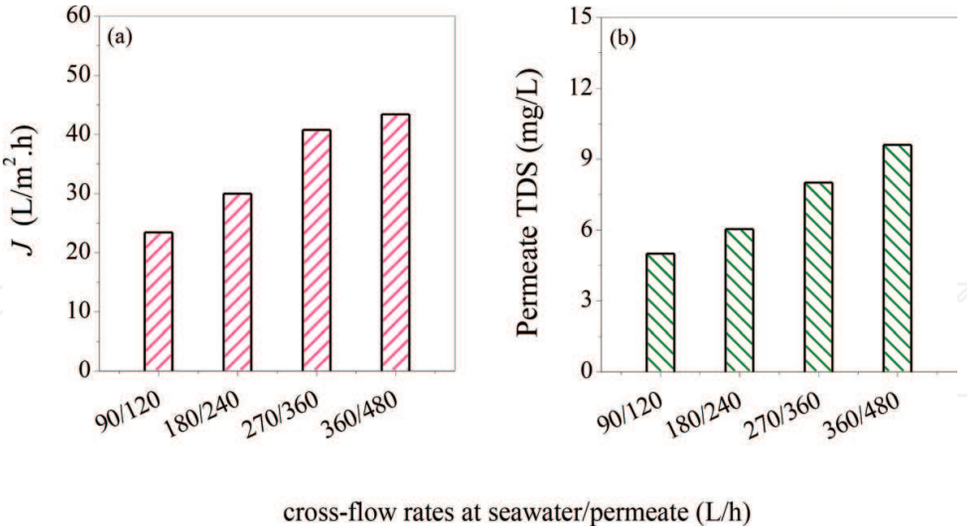

Figure 5. Permeate flux (a) and solute concentration (b) for different cross-flow rates (L/h) at seawater/permeate water under batch operating conditions (PTFE—0.45 μm and seawater/permeate: 55/25°C (ΔT: 30°C)).

flow rates of 270 and 360 L/h, respectively, and distilled water temperatures of 15, 20, 25, and 30°C in order to determine the effect of solution temperature on process performance. The performance results of the experiments were evaluated at seawater-permeate water operation temperatures of 45–15, 50–20, 55–25, and 60–30°C based on the water flux and output water TDS parameters.

It was determined that the water flux values of the process gradually increased in parallel with increasing solution temperature, as 40.3, 32.7, 36.8, 40.7, and 48.0 L/m² h, respectively. Total solute concentrations in MD output water were determined as 7.2, 7.5, 8.0, and 8.7 mg/L, respectively, and it was clearly observed that the output water had low dissolved solids. **Figure 6** shows permeate flux and salt concentration for different temperatures at seawater/permeate solutions under batch operating conditions.

The $CaCO_3$ and $CaSO_4$ compounds, which have lower solubility at high temperatures, can cause a serious reduction in water permeation performances of MD membranes by fouling occurring on the membrane surface during the processing. A previous study reported that increasing the input feed flow temperature from 80 to 90°C led to a fourfold decrease in flow level [36]. At membrane, considering the possibility of Ca^{2+} precipitation that might result continuous operation and constant membrane trans-temperature difference, the most suitable was to obtain high flux outputs to continually operate the membranes at fixed permeate water and seawater flow temperatures of 30 and 60°C, respectively.

3.1.4. Membrane trans-temperature difference

It was observed that increasing membrane trans-temperature difference (20, 30, and 40°C) led to an increase in the volume of water passing from the seawater to the permeate water. At a permeate flow temperature of 15°C, the water flux values for membrane trans-temperature differences of 20, 30, and 40°C were determined as 17.0, 32.7, and 53.3 L/m² h, respectively. At a

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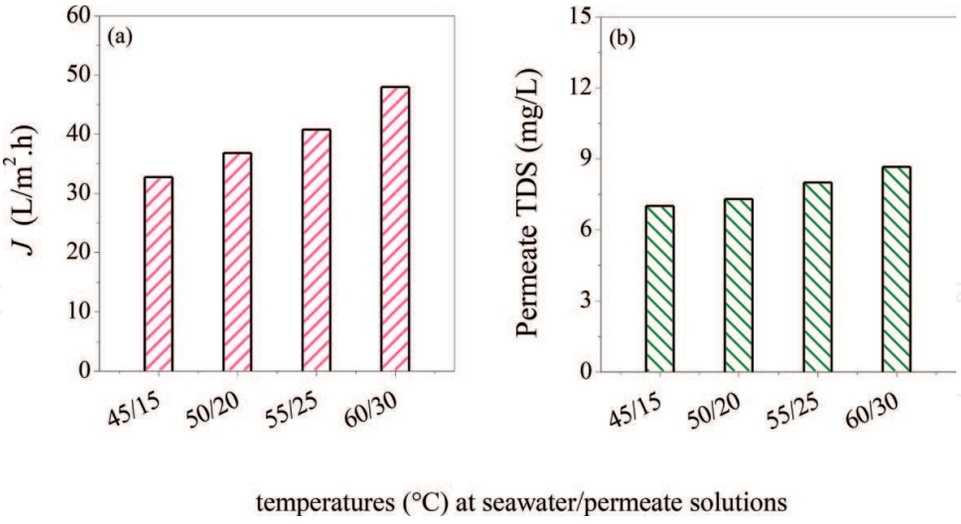

Figure 6. Permeate flux (a) and solute concentration (b) for different temperatures (°C) at seawater/permeate solutions under batch operating conditions (PTFE—0.45 μm, 270/360 L/h cross-flow rates at seawater/permeate, ΔT: 30°C).

permeate flow temperature of 30°C, these values were increased to 31.3, 48.0, and 63.0 L/m^2 h, respectively. At a permeate flow temperature of 15°C, the output water TDS values for membrane trans-temperature differences of 20, 30, and 40°C were determined as 6.5, 6.8, and 7.4 mg/L, respectively. On the other hand, at a permeate flow temperature of 30°C, the output water TDS values for membrane trans-temperature differences of 20, 30, and 40°C were determined as 6.75, 8.3, and 10.3 mg/L, respectively. In all temperature difference experiments, the output water contained low levels of dissolved solids. **Figure 7** shows permeate flux and effective permeate flux per unit of membrane trans-temperature difference and total solutes concentration for varying trans-temperature differences.

As described in the literature, continuous operation at higher seawater temperature may result in $CaCO_3$ and $CaSO_4$ precipitates that foul/clog the pore entrances on the membrane active layer, as well as the pore spaces within the membrane [36]. Based on this consideration, 30°C can be evaluated as the suitable trans-difference among the solutions for the continuous

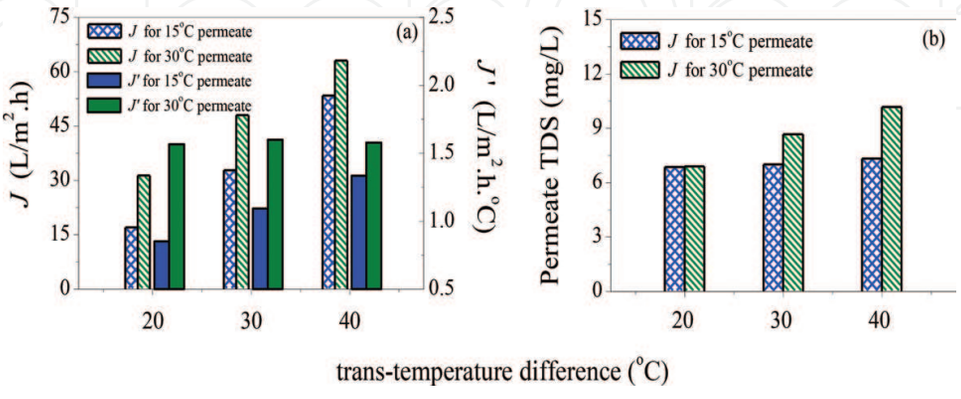

Figure 7. Permeate flux and effective permeate flux per unit of trans-temperature difference (a) and solute concentration (b) for varying trans-temperature differences (PTFE—0.45 μm, 270/360 L/h cross-flow rates at seawater/permeate, 15 and 30°C temperatures at permeate).

operation in which seawater or feed stream would be heated up to max 60°C. Taking into account the fact that energy costs represent the most important obstacle standing in the way of the rising uses of MD process compared to RO seawater desalination plants, it was comprehended that 30°C and some below or above differences would render further possible to arrive the optimal cost solutions in on-land MD plants due to lower heat energy consumption per unit of potable water to be produced.

3.2. MD processing of Marmara seawater

Under controlled membrane cleaning conditions per day cycle where 1% HCl, 1% NaOH and distilled water were consecutively used for flushing the active layer of the membrane while inside the module, and the permeate flow was replaced with distilled water within a suitable time frame, the continuous processing experiments were performed to identify the design flow that would serve as a basis for the field application of the DCMD. In the study, the DCMD was operated in order to concentrate seawater flow to obtain daily concentration performances of 63–66%, and at the end of the 5-day operation period, a 60% volumetric concentration ratio was reached. To ensure that the continuous operation performance for the periods shown in **Figure 8** remained consistent, the initial volume of the feed flow was completed (i.e. brought to the desired level) every day by adding raw seawater.

At the end of days I, II, III, IV, and V, the permeate water fluxes were determined as 38.9, 31.1, 22.6, 16.7, and 15.5 L/m^2 h, respectively. It was found that the flux changes in the first 4-day period were generally characterized by a consistent decrease, while the flux value on day V was very close to the flux value of day IV (despite the fact that the permeate was changed with distilled water). Based on this observation, it is possible to say that the water flux performance almost reached a constant operation level by the end of the day V.

An MD optimization study using PP membrane operated under conditions of $\Delta P = 0.355 \times 10^{-5}$ Pa, $Q_f = 73.6$ L/h and $Q_p = 18.8$ L/h determined an MD process flux of 4.192 L/m^2 h [24]. A previous

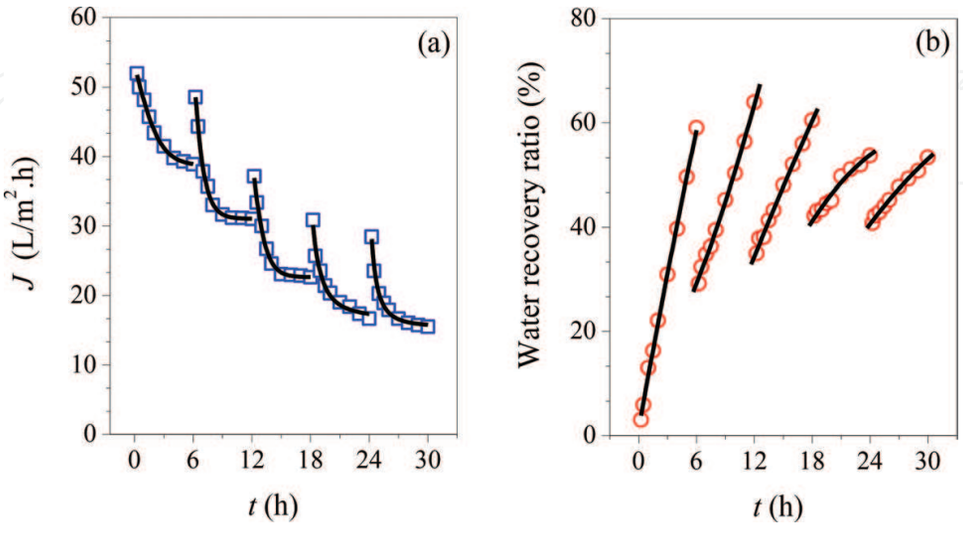

Figure 8. MD process performance under continuous operating condition ((a): permeate flux, (b): water recovery ratio).

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study using continuously operated MD process for producing drinking water from seawater made use of a PTFE membrane (0.22μm). In this study, where the feed temperature was 60°C and the permeate temperature was 20°C, the process flux level was reported to decrease from 23.76 to 14.36 L/m^2 h following a 1-month operation. Further, following membrane washing procedures, the same membrane was successfully reused in the DCMD system [22]. In another study using PTFE membranes, a flux level of 25 L/m^2 h was observed following the 70 h operation of the DCMD process with a temperature difference of 60°C [37].

Although the literature describes the use of various different acid and base derivatives for cleaning membranes, HCl use is common in MD [36, 38]. HCl is a quite effective chemical in cleaning of scaling and fouling from basic salts such as $CaCO_3$ [36]. In a study being used synthetic seawater, it was observed that citric acid and NaOH completely remedied for both the flux and the membrane's hydrophobicity [39]. In another study, washing MD membrane with HCl resolved the reduction in flux values caused by $CaCO_3$ fouling, bringing the flux values to their original level [38].

Within the context of the study experiments, the membrane cleaning at the end of day I was performed using only distilled water. On the following days, the cleaning was done using 1% HCl for 20 min, then 1% NaOH for 20 min, and lastly distilled water for 20 min. After flushing, the pH-conductivity values of the acid and base washing solutions changed from 2.05–51.1 and 12.7–52.9 to 2.04–44.9 and 12.71–46.6 mS/cm, respectively. Based on these results, it can be said that no significant changes were observed in the pH values. By the end of the day IV of operation, the changes in the conductivity of the acid and base washing solutions were 11.9 and 12.1%, respectively. For 1 month of a continuous cleaning at the same conditions per the applied operation cycle, it was calculated that the amounts of acid and base lost will reach to about 89.3 and 91.0%, respectively. With regard to the membrane flushing step that will be applied in a real-scale facility, it appears a necessary to renew the 1% acid and base solutions at certain time scales.

By maintaining seawater TDS value within an interval from 26,400 to a max of 78,900 mg/L throughout the processing (66,800 mg/L at the end of the operation), it was ensured that the DCMD system could be permanently operated with steady-state concentrating conditions at a constant temperature difference. In the operations, where distilled water is kept within the system for 24 h, conductivity values of the permeate flow at the end of days I, II, III, and IV were always below the threshold level of 500 μS/cm, being 14, 78, 117, and 375 μS/cm, respectively, while the TDS values for the days I, II, III, and IV permeates were 7, 49, 54, and 179 mg/L, respectively (*data were not shown*). Thanks to the distilled water replace at the beginning of day V, it was noted that these values decreased to 76.0 μS/cm and 47.1 mg TDS/L as adjacent values to the values at the end of the first 2 days. The qualities of concentrated seawater and potable water produced at the end of continuous operation are given in **Table 3**, together with MD rejection performances.

According to **Table 3**, the output water obtained was in very good quality in which the retentions of dissolved organics, boron, silicium, and iron as well as anions and cations were at above 99%. Too low color presence at 525 nm wavelength was observed resulting mostly probably from the trace level amounts of dissolved organics. Based on all results obtained

from the continuous operation, it can be suggested for successful field operations of DCMD seawater desalination plants that the distillate stream or product output water should not be remained for a longer operating time than about 10–12 h in the product water storage tank. Certainly, more accurate heuristic knowledge for safe plant operation would be ensured by means of taking into account the performance information to be retrieved from the data of the pilot and/or field scale installations.

Parameters*	MD concentrated seawater	MD permeate (output water)	*R* (%)
T	24.2	24.3	–
pH	8.54	7.30	–
Ec	108, 200	76.0	99.93
TDS	66, 800	47.1	99.93
TOC	20.5	0.08	99.61
DOC	18.4	0.06	99.67
UVA_{254}	0.160	0.001	99.38
SUVA	0.870	1.667	–
Na^+	20, 445	0.114	99.99
K^+	1143	0.005	99.99
NH_4^+	<0.1	0.0	–
Ba^{2+}	0.058	<0.01	–
Ca^{2+}	1097	3.5	99.68
Mg^{2+}	1770	1.4	99.92
Mn^{2+}	0.044	0.0	100.00
Sr^{2+}	15.20	0.02	99.87
B	4.39	0.007	99.84
Si	1.22	0.008	99.34
Fe	0.017	0.0	100.00
Cl^-	46, 236	12.6	99.97
HCO_3^-	244	0.58	99.76
NO_3^-	1.3	0.0	100.00
CO_3^{2-}	112	0.0	100.00
SO_4^{2-}	7050	7.2	99.90
TKN	1.8	0.0	100.00
Total nitrogen	3.1	0.0	100.00
Alkalinity	356	0.58	99.84
Total hardness	16,035	27.1	99.83

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Parameters*	MD concentrated seawater	MD permeate (output water)	*R* (%)
Color			
436 nm	0.003	0.000	100.00
525 nm	0.004	0.001	75.00
620 nm	0.001	0.000	100.00

*Units of all parameters are mg/L except for temperature (°C), electrical conductivity (μS/cm), UVA_{254} (1/cm), SUVA (L/mg.m) and color (1/cm).

Table 3. Quality of potable water produced by continuous MD operation together with MD rejection performances.

3.3. Health impacts assessments of produced drinking water

Almost 75 million people across the world acquire drinking water from the sea by treating seawater in desalination plants, and it is expected that this number will go up as a result of the steadily growing demand for water [40].

Seawater abounds with certain ions like calcium, magnesium, sodium, chloride, and iodine but has a lower content of essential ions such as zinc, copper, chromium, and manganese. As a result of seawater desalination applications, the amount of almost all of these ions and minerals, which are essential to the human health and to the agricultural productivity, is dramatically reduced in drinking water [14, 40]. Some essential metals like Cu, Mn, and Zn are required for normal body growth and function [41]. For healthy body, calcium and magnesium are both essential elements, and they play protective roles in the human body development [14, 40, 42]. Calcium, which is a primary constituent of the skeletal structure, is also important for different key physical functions. Due to decreases in bone mass and mineral content, calcium deficiency results in an increased risk of fractures. Deficiency of magnesium, an essential element for physiological processes such as mineralization and skeletal development, cardiac excitability and vascular tone, contractility, reactivity, and growth, can give rise to the pathophysiology of hypertension [40, 43]. Depending on the beneficial health effects rendered by intake of Ca and Mg through drinking water, it has been recommended by World Health Organization (2007) that the minimum and optimum Ca and Mg levels in drinking water should be 20 and 40–80 mg/L and 10 and 20–30 mg/L, respectively [43].

Otherwise, it is also likely that high quantities of these metals will inflict harms for human health. High concentrations of Cd, Cr, Ni, and Pb are regarded to be extremely toxic for humans and aquatic organisms [41]. Furthermore, demineralized water is more aggressive to piping, and therefore, additional risks could be posed through exposure to extracted trace elements such as lead and cadmium. According to certain studies, the use of demineralized water for cooking escalates the loss of essential mineral content of foods, bringing about detrimental effects on health [14, 42]. When the quality parameter values were evaluated in the output water obtained during this study, produced water is low in ions such as Ca, Mg, Na, Cl and essential ions like Zn, Cu, Cr, and Mn. In addition to this, its low solute content of 47.1 mg TDS/L paves the way for corrosive effects to the metal distribution piping and thus the risks

by exposure to extracted trace elements like lead and cadmium into the pipe will increase. The allowable limit values for these ions in the relevant national and international drinking water standards were presented in **Table 4** together with the other drinking water quality parameters specific to the produced water.

As described in the literature, to fulfill the desirable values for these parameters under limit concentrations, one possible solution involves making certain additions to the distilled product water by taking into account hygienic requirements. This will make it possible to supply drinking water that does not pose any problems with regard to public health. It is possible to overcome mineral deficiencies directly through addition of minerals at the MD desalination plant or locally by means of chemical injection at specified locations into water distribution systems by also considering hygienic requirements. In an alternative way, quality standards can be reached by mixing high-quality and low-quality water [47], which lead to the sig-

Parameters*	Permeate	TSE 266 [44]	WHO [45]	EPA [46]
T	24.3	–	–	–
pH	7.30	6.5–9.5	6.5–8.5	6.5–8.5
Ec	76.0	2500 μS/cm	250 μS/cm	–
TDS	47.1	–	–	500 mg/l
TOC	0.08	–	–	–
DOC	0.06	–	–	–
UVA_{254}	0.001	–	–	–
SUVA	1.667	–	–	–
Na^+	0.114	200 mg/l	200 mg/l	20 mg/l
K^+	0.005	–	–	–
NH_4^+	0.0	–	–	–
Ba^{2+}	<0.01	–	0.7 mg/l	2 mg/l
Ca^{2+}	3.5	–	–	–
Mg^{2+}	1.4	–	–	–
Mn^{2+}	0.0	50 μg/l	0.4 mg/l	0.05 mg/l
Sr^{2+}	0.02	–	–	–
B	0.007	1 mg/l	0.5 mg/l	–
Si	0.008	–	–	–
Fe	0.0	200 μg/l	0.3 mg/l	0.3 mg/l
Cl^-	12.6	250 mg/l	250 mg/l	250 mg/l
HCO_3^-	0.58	–	–	–
NO_3^-	0.0	50 mg/l	50 mg/l	10 mg/l
CO_3^{2-}	0.0	–	–	–

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Parameters*	Permeate	TSE 266 [44]	WHO [45]	EPA [46]
SO_4^{2-}	7.2	250 mg/l	500 mg/l	250 mg/l
TKN	0.0	–	–	–
Total Nitrogen	0.0	–	–	–
Alkalinity	0.58	–	–	–
Total hardness	27.1	–	150–500 mg/L	–
Color				
436 nm	0.000			
525 nm	0.001	–	–	–
620 nm	0.000			

Table 4. Maximum allowable concentrations of specific water quality parameters in use of produced water as drinking water supply.

nificant savings since remineralization facilities and chemicals are no longer needed [40]. The most affordable option to increase the dissolved content of specific ions in desalinated water is blending remineralized desalinated water with treated brackish groundwater or treated seawater [48]. When the MD output water produced in this study is evaluated in terms of physicochemical water quality for directly human consumption, it was found to be much lower than necessary levels especially in terms of dissolved minerals concentrations, which also give corrosive properties to the demineralized water. Based also on the literature knowledge and land experiences, a general management approach for field-scale pragmatic solutions toward to directly use the MD desalination effluents as drinking water was holistically proposed in **Figure 9**; as being the first, adding into existing urban potable water at desirable ratios; the second, injecting appropriate chemicals into effluent by completely mixing; and the third, mixing effluents with raw/concentrated seawaters (1:250/1:1000 for Marmara seawater) or clean brackish natural waters under hygienic precautions.

As the third distinctive solution, the MD product water can be readily mixed with seawater concentrate at a ratio of 1/1000 or with raw seawater at a ratio of 1/250 as shown in **Table 5** after MD desalination of raw seawater. In the calculations, dissolved solutes concentration increased to 180–200 mg/L in the mix ratios of 1/500 and about 1/200 for concentrated and raw seawaters, respectively. But, Ca^{2+} and Mg^{2+} concentration levels increased by 1.7 and 3.5 times although they could not fulfill the desired levels. Low level of total hardness, which is reached to ~60 mg/L, needs to be still increased to the desired standard levels by applying carbonation to the produced water. Hence, the obtained mixtures would also still lack of the desired levels of minerals and ions to be targeted for healthy drinking water production. For this reason, it is deduced that stand-alone use of this solution for an absolute success would not be sufficient. In addition, the practical ways of the mixing applications can be evaluated as an insignificant improving factor for decreasing of the negative effects of the concentrated seawater discharges that importantly affect the sea life near discharge points.

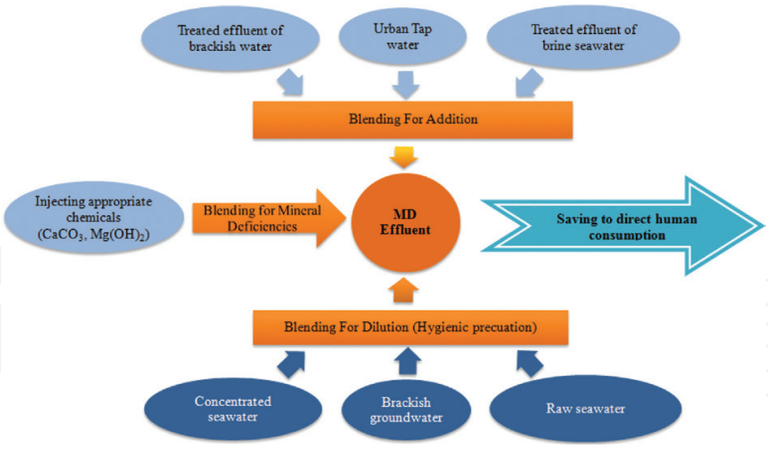

Figure 9. Holistic management approach for satisfying specific water quality requirements in long-term use of MD effluents demineralized from seawater as drinking water.

			Mixing ratio with							
Parameters	**Units**	**Permeate**	**Concentrated seawater**				**Raw seawater**			
			1/100	**1/250**	**1/500**	**1/1000**	**1/20**	**1/50**	**1/100**	**1/250**
T	°C	24.3	24.3	24.3	24.3	24.3	24.3	24.3	24.3	24.3
pH	–	7.30	7.31	7.30	7.30	7.30	7.3	7.3	7.3	7.3
Ec	µS/cm	76.0	1157	508	292	184	2092	882	479	222
TDS	mg/L	47.1	715	314	181	114	1254	530	289	134
TOC	mg/L	0.0	0.2	0.1	0.0	0.0	0.2	0.1	0.0	0.0
DOC	mg/L	0.0	0.2	0.1	0.0	0.0	0.2	0.1	0.0	0.0
UVA_{254}	1/cm	0.004	0.01	0.0	0.0	0.0	0.01	0.0	0.0	0.0
SUVA	L/mg.m	1.67	1.66	1.67	1.67	1.67	1.64	1.66	1.66	1.33
Na^+	mg/L	0.114	205	82	41	21	444	178	89	36
K^+	mg/L	0.01	4	2	1	0.0	22	9	4	2
Ca^{2+}	mg/L	3.5	14	8	6	5	23	11	7	4
Mg^{2+}	mg/L	1.4	19	8	5	3	36	15	8	4
Mn^{2+}	mg/L	0.001	0.001	0.001	0.001	0.001	0.002	0.001	0.001	0.001
Sr^{2+}	mg/L	0.02	0.17	0.08	0.05	0.04	0.29	0.13	0.07	0.04
B	mg/L	0.01	0.05	0.02	0.02	0.01	0.21	0.09	0.05	0.02
Si	mg/L	0.02	0.03	0.02	0.02	0.02	0.07	0.04	0.03	0.02
Fe	mg/L	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cl^-	mg/L	12.6	475	197	105	59	712	292	152	66
HCO_3^-	mg/L	0.58	3	2	1	1	16	7	4	2
NO_3^-	mg/L	0.0	0.01	0.01	0.0	0.0	0.0	0.0	0.0	0.0
CO_3^{2-}	mg/L	0.0	1	0.0	0.0	0.0	0.0	0.0	0.0	0.0

			Mixing ratio with							
Parameters	**Units**	**Permeate**	**Concentrated seawater**				**Raw seawater**			
			1/100	**1/250**	**1/500**	**1/1000**	**1/20**	**1/50**	**1/100**	**1/250**
SO_4^{2-}	mg/L	7.2	78	35	21	14	123	53	30	15
TKN	mg/L	0.0	0.02	0.01	0.0	0.0	0.0	0.0	0.0	0.0
Alkalinity	mg/L	0.58	4	2	1	1	16	7	4	2
Total hardness	mg/L	27.1	187	91	59	43	275	126	77	42
Color										
436 nm	1/cm	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
525 nm		0.001	0.001	0.001	0.001	0.001	0.0	0.0	0.0	0.001
620 nm		0.000	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001

Table 5. Changing values of output water quality parameters according to the ratios of mixings with concentrated or raw seawaters.

4. Conclusions

In this chapter, suitable operating conditions for continuous DCMD processing of seawater were first determined based on water flux and solutes rejection performances. Thereafter, by relative long-term MD desalination in seawater concentration mode at lab pilot-scale system, the water qualities of MD product water and seawater concentrate were investigated in light of physicochemical parameters specific to the seawater characteristics. In the final, it was elaborately examined whether the MD produced water is suitable for direct human use as drinking water.

Within the scope of the DCMD experiments under batch conditions, it was determined that the best operating conditions involved the use of hydrophobic PTFE membrane with 0.45 μm pore diameter; a flow velocity for seawater and permeate (distilled water) of 270 and 360 L/h, respectively (corresponding to mean Re numbers of 4320 and 4222); and flow temperatures for seawater and permeate streams of 60 and 30°C, respectively, which are associated with a membrane trans-temperature difference of 30°C.

By means of 30 h MD processing of rough-filtrated raw seawater from the Marmara Sea that was operated with steady-state permeate flux of 17.21 L/m^2 h and solutes retentions of >99% at seawater concentrating level reached to 66% in the constant temperature difference, the production of MD output water below threshold level of 500 μS/cm was continually carried out without replacing intrinsic distillated water of distillate stream. After its replacement was applied, ultimate product water was supplied with 76.0 μS/cm conductivity and 47.1 mg/L dissolved solids. A replacement time of about 10–12 h for initial clean distillate in the output water storage tank would be sufficient for field-scale operations of DCMD seawater desalination plants.

In case the MD product water is to be used as drinking water, it will be necessary to ensure that the dissolved minerals that are essential for a healthy life are found in the water in at least the minimum recommended levels, and to prevent the various trace elements (Cu, Fe, and Mn)

that can have toxic effects on living beings above certain levels or cause damage to the processing lines. Possible management options recommended in the literature to remedy these issues include the direct addition of minerals to final waters, the addition of chemicals to specific locations on process lines, and the blending of demineralized water with treated brackish groundwater/seawater. However, stand-alone generalization of each one of these practical solutions would not be made possible for all the challenges to be encountered in all application varieties of seawater desalination plants. In this frame, there need to develop unique approaches oriented on novel pragmatic solutions toward to the direct use of MD demineralized effluents as drinking water. In this respect, under an integrated conservative approach, a general management framework toward satisfying the specific water quality requirements in long-term use of MD effluents was proposed. With an aim of fulfilling the deficiencies of minerals and ions to be targeted for healthy drinking water supply, the developed holistic approach is jointly dependent on injections to urban water distribution systems at desirable ratios, mixings with raw/concentrated seawaters (1:250/1:1000 for Marmara seawater) or brackish natural waters under hygienic precautions, and additions in sufficient amounts of appropriate chemicals by completely mixing.

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Nomenclature

AFM	atomic force microscopy
C_f	concentration in the feed
C_p	concentration in the permeate
DCMD	direct contact membrane distillation
DOC	dissolved organic carbon
EDX	energy dispersive X-ray spectrometry
LEP	liquid entry pressure
MED	multi-effect distillation
MD	membrane distillation
MSF	multi-stage flash distillation
PE	polyethylene
PP	polypropylene

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PTFE	polytetrafluoroethylene
PV	photovoltaic
PVDF	polyvinylidene fluoride
Q_p	flow of permeate side (L/h)
Q_f	flow of feed side (L/h)
R	rejection (%)
Re	Reynolds number
RO	reverse osmosis
$R_{z,c}$	surface roughness of clean membrane
$R_{z,f}$	surface roughness of fouled membrane
SEM	scanning electron microscopy
SUVA	specific ultraviolet absorbance
TDS	total dissolved solids
TKN	total Kjeldahl nitrogen
TOC	total organic carbon
UVA_{254}	ultraviolet absorbance at 254 nm
WHO	world health organization
Greek symbols	
θ_c	contact angle of clean membrane (°)
θ_f	contact angle of fouled membrane (°)

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