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Using BWRO for removal of Arsenic III from anaerobic groundwater

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ABSTRACT

Arsenic in groundwater poses significant health risks, especially in regions dependent on groundwater for drinking water. This study compares the performance of two brackish water reverse osmosis membranes for arsenic (III) rejection. The study explores membrane performance under conditions not extensively addressed before, such as using natural anaerobic groundwater with arsenic concentrations of 70–80 μg/L and examining the effect of sulfide complexation on arsenic rejection. The impact of sodium sulfide and antiscalant on arsenic rejection was investigated, with Na_2S tested as a complexing agent. The aim was to determine whether antiscalant addition affects sulfide stability or arsenic rejection, and whether it could lead to decreased membrane efficiency in the short term. Results show that membrane type contributes to a 5–8 % difference in arsenic rejection. Arsenic rejection mirrored boron's trend but was consistently higher. However, the addition of sodium sulfide, with or without the antiscalant, did not enhance arsenic rejection, indicating that membrane type remains the primary factor. These findings highlight the need for tailored membrane selection to optimize arsenic rejection under anaerobic conditions. Chemical treatment strategies should align with specific contaminant profiles and feedwater conditions, with further pilot-scale validation for real-world applications.

1. Introduction and background information

Freshwater aquifers worldwide are under increasing stress due to over-extraction and contamination, driving the adoption of alternative water sources in water-scarce regions [1–3]. In the Netherlands, excessive groundwater extraction has led to brackish groundwater intrusion, resulting in the closure of over 100 pumping stations, with the prediction that over 20 % of remaining wells will face salinization, a projection now confirmed by field observation [4]. Many groundwater-based drinking water companies are considering brackish water reverse osmosis (BWRO) as a key treatment strategy [5]. Advances in membrane system design, such as optimized spacer configuration [6,7] and orientation [8], have been shown to improve hydrodynamic conditions and mass transfer in laboratory settings, enhancing desalination efficiency. This progress expands the feasibility of using brackish groundwater and seawater in coastal and inland areas.

Arsenic contamination in groundwater primarily stems from natural geological processes [9], such as the weathering of arsenic-rich minerals, although human activities like mining can further elevate concentrations. Long-term exposure to arsenic in drinking water is associated with increased risks of skin, lung, and bladder cancers, as well as cardiovascular diseases [10–12]. Advanced treatment methods, including coagulation [13], adsorption [14], and membrane filtration [15], are being implemented to ensure safe water supplies. The World Health Organization (WHO) sets a drinking water limit of ≤ 10 μg/L for arsenic in drinking water [11], based on the health risks associated with long-term exposure. Some Dutch drinking water companies adopt an even stricter operational target of ≤ 1 μg/L to minimize risk [16,17]. To meet these rigorous standards, membrane-based treatment processes, including reverse osmosis, have gained attention as effective solutions for arsenic removal [18–20].

Given the significant health risks of arsenic contamination, effective removal methods are essential for safe drinking water. Reverse osmosis (RO) removes arsenic and purifies water [15,18]. Reverse osmosis (RO) effectively removes biological, chemical, and radioactive impurities from seawater and brackish water [21,22]. The process relies on applied pressure across a semi-permeable membrane. The ion rejection in these

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Nomenclature

Symbol/Abbreviation	Definition, Unit
°C	Degrees Celsius, °C
µg/L	Micrograms per liter, µg/L
µS/cm	Microsiemens per centimeter (electrical conductivity), µS/cm
AS	Antiscalant (used only in figure legends), –
As(III)	Arsenite (trivalent arsenic species), µg/L
As(V)	Arsenate (pentavalent arsenic species), µg/L
BD30	Biodegradable antiscalant (4AQUA OSM BD30), –
BWRO	Brackish Water Reverse Osmosis, –
Cf	Concentration in feed, mg/L
Cl^-	Chloride ion, mg/L
Cp	Concentration in permeate, mg/L
DO	Dissolved Oxygen, mg/L
γ	Salt (ion) rejection efficiency, –
GW	Groundwater, –
Jw	Water flux, $m^3 /m^2 \cdot s$
LMH	Liters per square meter per hour (flux), $L/m^2 \cdot h$
MTC	Membrane Transport Coefficient, $m^3 /m^2 \cdot s \cdot Pa$
Na^+	Sodium ion, mg/L
Na_2S	Sodium sulfide, –
NDP	Net Driving Pressure, Pa or bar
NF	Nanofiltration, –
RO	Reverse Osmosis, –
S/As	Sulfide-to-arsenic molar ratio, –
SEPA	Cross-flow membrane test cell (Sterlitech configuration), –
SWRO	Seawater Reverse Osmosis, –
TCF	Temperature Correction Factor, –
Virgin GW	Untreated groundwater (no chemical additions), –
Symbol/Abbreviation	Definition, Unit
RO	Reverse Osmosis, –
BWRO	Brackish Water Reverse Osmosis, –
SWRO	Seawater Reverse Osmosis, –
MTC	Membrane Transport Coefficient, $m^3 /m^2 \cdot s \cdot Pa$
γ	Ion (salt) rejection efficiency, –
NDP	Net Driving Pressure, bar or Pa
TCF	Temperature Correction Factor, –
Na_2S	Sodium sulfide, –
BD30	Biodegradable antiscalant (4AQUA OSM BD30), –
LMH	Liters per square meter per hour (flux), $L/m^2 \cdot h$

membranes is governed by size exclusion and electrostatic interactions between the ions and the membrane surface, allowing selective retention based on size, charge, and affinity [23,24]. Recent advances in membrane technology, such as surface-modified RO membranes [25] and novel hollow fiber configurations for arsenic removal [26], offer improved selectivity and efficiency. Integrated NF/RO systems also present opportunities for resource recovery and optimized performance in brackish water treatment [27], particularly when designed for energy and resource efficiency [28]. Pretreatment prevents fouling and ensures membrane longevity; post-treatment ensures the permeate quality meets drinking water standards [21]. A comprehensive review highlights the rapid evolution of arsenic removal technologies, supporting the need for real-world validation of new approaches [29]. Most studies remain limited to controlled laboratory conditions, and few evaluate commercial BWRO membranes under real-world anaerobic groundwater conditions, particularly concerning arsenic-sulfide complexation and antiscalant interactions.

The quality of produced water depends on the membrane type, feed water quality, and operational conditions [21,22]. While SWRO elements generally demonstrate higher ion rejection efficiency than BWRO elements, particularly for chloride ions (Cl^-) (Table 3), the performance of BWRO systems can vary significantly with salinity, applied pressure, and membrane type [15]. Brackish water RO elements also differ in performance, with denser, more selective membrane structures typically achieving higher rejection, as influenced by polyamide layer characteristics and surface properties [23]. This variability underscores the importance of evaluating commercial BWRO membranes under real-world conditions to identify optimal performance for challenging contaminants like arsenic.

Several factors influence RO performance, with temperature and pH playing significant roles. Higher temperatures increase water and salt permeability due to enhanced diffusion and reduced viscosity, as demonstrated in large-scale RO systems [21,22]. pH affects arsenic speciation and membrane surface charge, complicating its removal. At higher pH (pKa $\geq$9.2) [16,30], uncharged As(III) (H_3AsO_3) deprotonates to form the negatively charged $H_2AsO_3^-$, improving rejection via Donnan exclusion. The membrane's surface charge also becomes more negative, enhancing electrostatic interactions and affecting ion selectivity [31].

BWRO systems in the Netherlands commonly treat anaerobic groundwater, where dissolved oxygen is too low to support aerobic respiration. Dissolved oxygen concentrations above 1 mg/L typically indicate aerobic conditions, while concentrations below 1 mg/L signify anaerobic conditions [32]. These systems are often designed with minimum or no pretreatment, as anaerobic groundwater typically contains low levels of organic matter and aerobic microorganisms, making it less prone to biofouling [33]. This favorable feed water quality reduces the need for extensive pretreatment, simplifying system operation and maintenance. BWRO shows low rejection of heavy metals such as arsenic and boron under anaerobic conditions [34]. These metals exist in reduced, uncharged forms at natural pH, limiting their electrostatic repulsion and size-based exclusion by RO membranes [34,35]. SWRO systems typically use a two-pass RO process for high boron removal, where the pH of the first-pass permeate is raised above pH 10 to convert neutral $B(OH)_3$ into the negatively charged $B(OH)_4^-$, thereby enhancing boron rejection in the second pass [36,37].

Commercial SWRO membranes achieve 88–91 % boron rejection under laboratory standard conditions [22,36], but their performance in full-scale desalination plants is typically 10–15 % lower due to fouling, scaling, and suboptimal hydrodynamics [21]. Single-pass configurations are only suitable when boron rejection requirements are below 80 % [36]. RO design programs often fail to simulate boron and arsenic rejection under anaerobic conditions, as they are calibrated for aerobic, high-salinity feed waters. Membrane manufacturers typically provide rejection data based on test conditions that do not fully reflect the operating conditions of large-scale desalination plants. Assessing boron and arsenic rejection performance of commercial elements under real-world conditions, particularly in anaerobic brackish groundwater, is essential to bridge the gap between laboratory predictions and field performance.

Previous studies report 40–80 % arsenic (III) rejection by BWRO, depending on factors such as pH, ionic strength, and co-existing ions in the feed water [34,38]. Ahoulé et al. (2016, PhD research) compared As (III) removal using NF (NF270) and RO (TW30, similar to BW30) membranes in water containing 100 µg/L As(III). They found that NF barely rejected As(III) (2–5 %) at high recoveries (90 %), while RO achieved moderate rejection (40–60 %). For As(V), both NF and RO exhibited higher rejection, with NF rejecting over 70 % and RO over 93 %, at the same pressure and recovery. These findings highlight the challenge of As(III) removal in single-pass BWRO systems, particularly under neutral pH and anaerobic conditions, where As(III) remains uncharged and poorly rejected.

Chang et al. [31] investigated As(III) retention using NF (Desal HL) and low-pressure RO (Desal AK) membranes as a function of pH. They found that at pH ±9.5, the uncharged H_3AsO_3 transforms into the highly charged $HAsO_3^-$, significantly increasing arsenic (III) rejection by RO because of Donnan exclusion alongside size-based exclusion. For RO membranes, rejection increased from ±70 % ($pH < 8.5$) to 90 % ($pH > 9.5$), while for NF membranes, rejection rose from ±10 % ($pH < 8.5$) to 40 % ($pH > 9.5$). Their study concluded that while the direct application of NF on low-salinity brackish groundwater is economically attractive, it is insufficient to meet WHO guidelines when arsenic concentrations exceed 11 µg/L. This raises the question of whether significant differences exist between various BWRO membranes in terms of As(III) rejection under realistic operating conditions.

In conventional treatment, As(III) is oxidized to As(V), which is more effectively removed by adsorption and membrane processes [39]. This conversion uses aeration or chemical oxidants such as NaOCl and $KMnO_4$, which are more effective than NH_2Cl and ClO_2 [39]. Iron sulfide is another potential additive that interacts with arsenic, forming negatively charged thio-arsenic (S-As) complexes at sulfide concentrations above 10^{-4} M (0.1 mM), depending on the sulfide-to-arsenic ratio [35]. The presence of iron also influences this process by stabilizing sulfide species and promoting surface complexation [40]. These charged complexes offer potential for improved separation via membrane processes; however, little is known about their behavior during membrane filtration, particularly in real-world brackish groundwater systems.

Membrane type affects arsenic rejection. Akin et al. [15] compared arsenic removal between SWRO (SWHR) and BWRO (BW-30) membranes under controlled laboratory conditions using synthetic feed water. They found 85–95 % As(III) rejection for BWRO membranes and 95–99 % for SWRO membranes. The experiments were conducted using flat sheet flow cells at a recovery of 1 %, minimizing concentration polarization and fouling. While these results highlight the superior rejection capacity of SWRO membranes, they were obtained under idealized conditions that do not reflect the complexity of real-world brackish groundwater systems, particularly anaerobic, naturally contaminated waters.

This study compares the arsenic removal by two commercially available flat-sheet BWRO membranes (Hydranautics ESPA2-LD and CPA7-MAX) in treating natural anaerobic brackish groundwater with arsenic concentrations of 70–80 µg/L. Sodium sulfide (Na_2S) dosing promotes the formation of negatively charged thio-arsenic complexes, which can enhance arsenic rejection through mechanisms such as Donnan exclusion despite arsenic remaining in the + 3 oxidation state. Unlike As(III), these negatively charged complexes are more effectively repelled by the negatively charged polyamide membrane surface. The membranes' performance was evaluated under real-world conditions, including natural water composition and anaerobic chemistry, rather than controlled laboratory environments. This approach addresses key knowledge gaps regarding the behavior of arsenic-sulfide species during BWRO treatment and the performance of commercial membranes in full-scale-relevant scenarios.

This study also evaluates the impact of antiscalant on membrane fouling and arsenic retention, with a focus on sulfide stability under realistic operating conditions. Antiscalants prevent fouling and scale formation in real-world systems. A biodegradable antiscalant was added to assess interference with the sulfide dosing or complex formation that could reduce membrane performance. The objective was to determine if the antiscalant reduced sulfide availability, arsenic rejection, or membrane efficiency. This study addresses rapid sulfide depletion, a critical practical limitation overlooked in previous studies. The findings support optimizing arsenic removal in real-world settings.

2. Material and methods

2.1. Experimental setup

Fig. 1 presents a schematic of the experimental setup. Groundwater was extracted from an observation well at a depth of 220 m (Fig. 2) using a peristaltic pump (Eijkelkamp slangenpomp - Gemini BV) and pumped into a 60-liter feed tank. The average quality of the groundwater is mentioned in Table 1. The tank featured an overflow system to maintain a constant volume and ensure anaerobic conditions, preventing iron (hydr)oxide formation. From the feed tank, water was pumped through a SEPA cell (Sterlitech Corporation, USA) equipped with a 440 µm prefilter using a plunger pump (MarelliMotori 60034–1 motor with Hydra-Cell P200MSGSSA07S plunger system).

The SEPA cell was configured with a 138 cm^2 flat sheet membrane (either ESPA2-LD or CPA7-MAX), a 28-mil feed spacer, and a 9-mil permeate spacer. The system operated at a flow rate of 69 L/h, corresponding to a cross-flow velocity of approximately 0.2 m/s, within the typical range (0.1–0.35 m/s) for spiral wound reverse osmosis modules [41]. Table 1 summarizes the membrane properties under the manufacturer's test conditions.

Feed pressure, electrical conductivity, and temperature were continuously monitored at the permeate outlet. Permeate and concentrate were collected separately and discharged. Since the groundwater temperature remained stable, no additional temperature control was required. Water quality analysis was conducted at Aqualab Zuid (the Netherlands), with samples stored at 4°C prior to analysis. Field measurements of pH and conductivity were performed using a Hach Sension pH meter and an MM150 conductivity meter.

The permeate flow rate was measured using a Jadever JWE-6K scale (Taiwan) with 0.2 g precision, recording the permeate mass every minute. Instead of maintaining a constant flux, the feed flow was kept constant throughout the experiments. Achieving a stable flux was

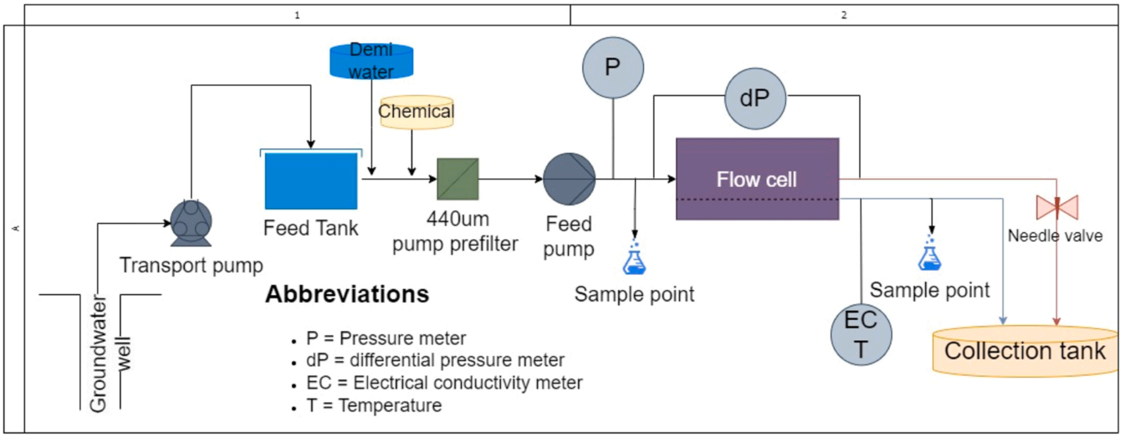

Fig. 1. A schematic view of the experimental setup.

Fig. 2. The observation well that is used for the extraction of water for this experiment.

Table 1
Average groundwater quality at the study location.

Parameter	Value	Unit
pH	7.7	-
Conductivity	1560	µS/cm
Temperature	14.5	oC
Boron	387	µg/l B
Iron	780	µg/L Fe
Manganese	943	µg/L Mn
Sodium	193	mg/L Na
Sulfide	0.1	mg/L S
Arsenic	74	µg/L As

challenging due to the membrane's low active surface area, which resulted in low permeate production. The combination of limited permeate yield, scale precision, and measurement frequency made exact flux determination difficult. However, for short-duration experiments like this, maintaining a constant feed flow provided a practical alternative to achieve near-constant flux.

CPA7-MAX required higher feed pressure than ESPA2-LD to achieve comparable fluxes. The pressure was adjusted using the concentrate valve, ranging from 10.4–11.0 bar for ESPA2-LD and 11.9–12.5 bar for CPA7-MAX. Identical fluxes could not be achieved. The resulting fluxes were 35 LMH for ESPA2-LD and 39 LMH for CPA7-MAX.

2.2. Methodology

The experiments aimed to assess the arsenic rejection performance of ESPA2-LD and CPA7-MAX membranes under varying feed conditions using brackish groundwater.

Before each experiment, membranes were flushed with demineralized water for at least two hours to remove residual salts or fouling from manufacturer testing, storage solutions, or membrane preparation (e.g., cutting). Clean water flux was measured to determine the baseline membrane transport coefficient (MTC), referred to as "MTC Before GW" (Table 2). This measurement served as a benchmark for subsequent comparisons. Each experiment lasted 2–3 h, including flushing, chemical conditioning, and permeate collection.

All experiments were conducted in triplicate (3x) to ensure result reliability. The cut membrane coupons were stored in preservative solution at 4 °C until the experiment and flushed with demineralized water prior to each use. Chemical clean-in-place (CIP) procedures were not applied, as the short test duration and low recovery (<1 %) minimized the risk of irreversible fouling.

Table 2
Characteristics of the membrane elements at test conditions applied by the manufacturer.

Membrane	Manufacturer	Salt Rejection (1500 mg/L NaCl solution)
ESPA2-LD	Hydranautics	99.6 %
CPA7-MAX	Hydranautics	99.8 %

Membrane transport involves water and salt transport. Water transport is measured by the membrane transport coefficient (MTC), and salt by salt rejection efficiency (γ). Checking these parameters provided insight about arsenic rejection. The MTC, commonly used in diffusion models, describes water flux through the membrane [41] and is expressed in $m^3/m^2 \cdot s \cdot Pa$. It is a function of the net driving pressure (NDP, in Pa), water flux (Jw, in $m^3/s.m^2$), and the temperature correction factor (TCFw, dimensionless). The calculation methodology is detailed elsewhere [41]. MTC was determined from experimental data to quantitatively assess membrane performance under varying conditions and evaluate the impact of chemical additions on water transport.

$$MTC = \frac{J_W \bullet NDP}{TCF_w} \tag{1}$$

Ion (salt) rejection measures the membrane's efficiency in removing dissolved ions from the feed water. It is calculated using Eq. 2 based on ion concentrations in the feed and permeate samples.

$$\gamma = 1 - \frac{C_p}{C_f} \tag{2}$$

Following the baseline MTC measurements, three experimental series were conducted to evaluate arsenic rejection using groundwater as feed water under varying chemical conditions. These experiments assessed the effects of sodium sulfide (Na_2S) and a biodegradable antiscalant (BD30) on arsenic removal and membrane performance. The tested conditions included:

- Virgin Groundwater (Virgin GW): Groundwater without chemical additions.
- Groundwater with Na_2S and Antiscalant (GW + Na_2S + BD30): Groundwater dosed with 1 mg/L Na_2S and 1 mg/L BD30.
- Groundwater with Na_2S (GW + Na_2S): Groundwater with 1 mg/L Na_2S only.

Sodium sulfide (Na_2S) dosing was calculated based on previous studies to ensure effective arsenic-sulfide (As-S) complexation [35,40]. These studies indicate that thioarsenite formation requires a minimum sulfide-to-arsenic (S/As) ratio of 3:1 and a sulfide concentration of at least 10 µM. The brackish groundwater used in the experiments naturally contained approximately 1 µM sulfide (±0.3 mg/L), contributing to the required ratio.

The methodology of Hou et al. [42] was applied to achieve a stable arsenic-sulfide complex, where a 25 µM Na_2S dosage was applied, attaining a 10:1 S/As ratio. A 10 µM Na_2S dosage ensured complex stability under test conditions. Na_2S was dissolved in demineralized water and buffered with $NaHCO_3$ to stabilize pH and reduce H_2S volatilization that could compromise dosing efficiency. However, as shown in the results, sulfide concentrations declined rapidly, indicating that H_2S loss could not be fully prevented under the applied conditions. Sodium sulfide and BD30 were precisely dosed using a membrane dosage pump (DDA type, Grundfos, Denmark) to achieve the target concentrations.

The biodegradable antiscalant 4AQUA OSM BD30 (Aquacare BV, Netherlands) was used to evaluate the impact of Na_2S on membrane performance. BD30 was chosen based on industry trends favoring biodegradable solutions for reverse osmosis (RO) systems. Its addition allowed assessment of its effect on membrane fouling, particularly in combination with Na_2S. BD30 prevents scaling in RO systems, but the low recovery rate (~1 %) in these short-term experiments minimized scaling risks.

Demineralized water was used after the groundwater experiments to measure the "MTC After GW" values, assessing potential membrane fouling during the short experimental period. Significant fouling within this timeframe would indicate that the Na_2S and BD30 combination is not suitable for long-term operation in full-scale systems. Arsenic rejection remained stable throughout individual experiments, indicating that membrane performance was not notably affected by short-term fouling.

3. Results and discussions

3.1. Membrane permeability

Fig. 3 presents the membrane transport coefficient (MTC) values for ESPA2-LD and CPA7-MAX under various feed water conditions. Each bar of the same color corresponds to a specific condition, as indicated in the legend. Overall, CPA7-MAX exhibits a lower MTC than ESPA2-LD, and a lower permeability, suggesting a tighter/thicker active layer, which may influence arsenic (III) removal efficiency under the tested conditions.

Fig. 4 compares the MTC values of both membranes using demineralized water before and after groundwater experiments. “MTC After GW” is slightly lower than MTC Before GW, suggesting minor fouling. The differences (black error bars) are negligible (<10 %), suggesting no measurable fouling. Short experiment duration likely prevented significant fouling.

Fig. 5 compares the mass transfer coefficient (MTC) of membranes using groundwater as feed water, with demineralized water as the baseline. The data indicate that groundwater generally results in a lower MTC compared to demineralized water. This reduction is attributed to the higher ion Concentration and osmotic pressure in groundwater, which lowers the net driving pressure (NDP) across the membrane.

Table 3
Summary of Experiments Performed.

Experiment Name in Graphics	Membrane Type	Feed Water	Chemical Dosage
MTC Before GW	ESPA2-LD	Demineralized water	-
Virgin GW	ESPA2-LD	Anaerobic groundwater	-
GW+Na_2S+BD30	ESPA2-LD	Anaerobic groundwater	1 mg/L Na_2S + 1 mg/L BD30
GW+Na_2S	ESPA2-LD	Anaerobic groundwater	1 mg/L Na_2S
MTC After GW	ESPA2-LD	Demineralized water	-
MTC Before GW	CPA7-MAX	Demineralized water	-
Virgin GW	CPA7-MAX	Anaerobic groundwater	-
GW+Na_2S+BD30	CPA7-MAX	Anaerobic groundwater	1 mg/L Na_2S + 1 mg/L BD30
GW+Na_2S	CPA7-MAX	Anaerobic groundwater	1 mg/L Na_2S
MTC After GW	CPA7-MAX	Demineralized water	-

3.1.1. Effect of adding sulfide and AS on MTC

The CPA7-MAX membrane exhibits a reduction in the mass transfer coefficient (MTC) when chemicals, either Na_2S with antiscalant or just Na_2S, are added to the feed water. This suggests that the increased osmotic pressure from these chemical additives contributes to the observed MTC reduction.

In contrast, the ESPA2-LD membrane exhibits a lower MTC without chemical additions but shows an increase when Na_2S or Na_2S with antiscalant is introduced. This contradicts the hypothesis that chemical additives would reduce MTC due to increased osmotic pressure. ESPA2-LD may respond differently due to variations in surface composition or membrane structure. Short experiment duration may have prevented the membranes from reaching equilibrium conditions.

The data do not support definitive conclusions on the contrasting behavior of these membranes in response to chemical additives. Further investigation is needed to clarify the underlying mechanisms. Long-term studies on membrane performance and stability under varying chemical conditions are valuable. This will be explored in a forthcoming 16 m^3 /h pilot system.

3.2. Membranes’ rejection

Simulations using the Integrated Membrane Solutions Design Software (Hydranautics) confirm that CPA7-MAX has superior rejection of sodium and chloride compared to ESPA2-LD. The results are presented in Table 4.

The projection results (Table 4) show about 1 % lower rejection compared to factory test conditions (Table 2), likely due to real-world factors such as feed water composition and system pressure. However, the overall trend confirms that CPA7-MAX provides better ion rejection than ESPA2-LD.

Table 4 shows that the projection program assigns identical rejection values for Na^+ and Cl^-. In practice, rejection can differ due to ion properties and membrane interactions. This reflects a limitation of membrane projection software, which typically applies average salt rejection values based on empirical test conditions, without simulating ion-specific transport mechanisms.

Fig. 6 compares the ion rejection by CPA7-MAX and ESPA2-LD. In all groundwater-based conditions, with or without Na_2S and antiscalant, both membranes achieved over 95 % rejection of sodium, iron, and manganese. The rejection differences between the membranes were ±1 %, making them statistically insignificant and inconclusive.

Dissolved sulfide rejection was significantly lower than sodium, iron, and manganese. At the same time, both membranes followed a similar removal trend, likely because sulfide exists in a different form compared to these ions. The rejection of sulfide in membrane filtration can be influenced by its ionization state, which changes depending on the pH of the feed water. In its uncharged molecular form (e.g., H_2S), sulfide can pass through the membrane more easily due to its neutral charge. On the other hand, when in its ionic form, such as sulfide ion (S^{2-}) or hydrosulfide ion (HS^-), it may experience electrostatic repulsion, depending on the membrane surface charge and pore size. Smaller, highly charged ions like sodium (Na^+) or iron (Fe^{2+}) are typically more effectively rejected due to their size and stronger electrostatic interactions with the membrane surface. This is supported by Shen et al. [23], who found that smaller ions are rejected more efficiently due to their higher charge density and smaller hydrated size [23]. Larger, neutral molecules, in contrast, interact more weakly with the membrane surface, which can reduce their rejection. Liu et al. [24] further supported this concept, showing that membranes with a denser active layer, which increase

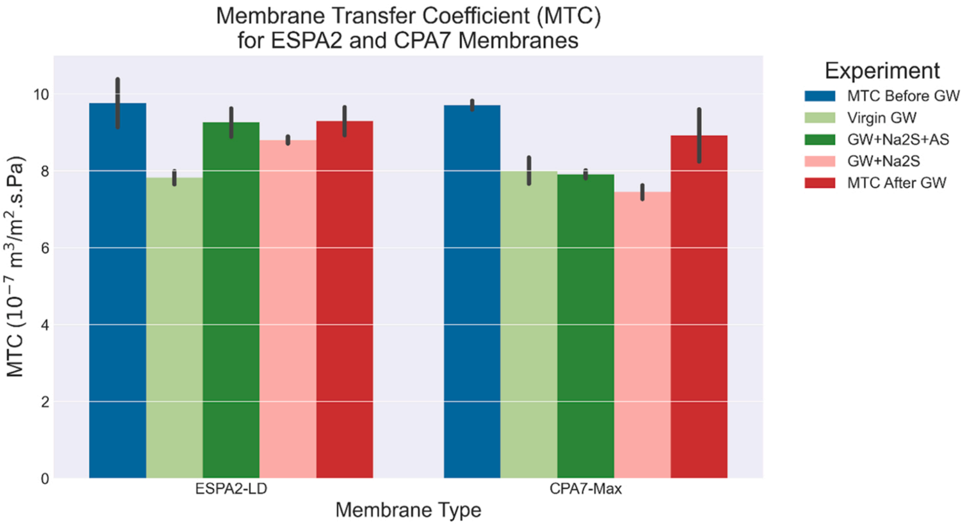

Fig. 3. Membrane transport coefficient (MTC) values for ESPA2-LD and CPA7-MAX under different feed water conditions. MTC Before GW represents measurements with demineralized water before groundwater introduction. Virgin GW corresponds to untreated groundwater without chemical additions. GW + Na_2S + AS includes sodium sulfide (Na_2S) and antiscalant. GW + Na_2S contains only Na_2S. MTC After GW refers to measurements with demineralized water after groundwater experiments. Error bars represent the standard deviation of triplicate tests (n = 3).

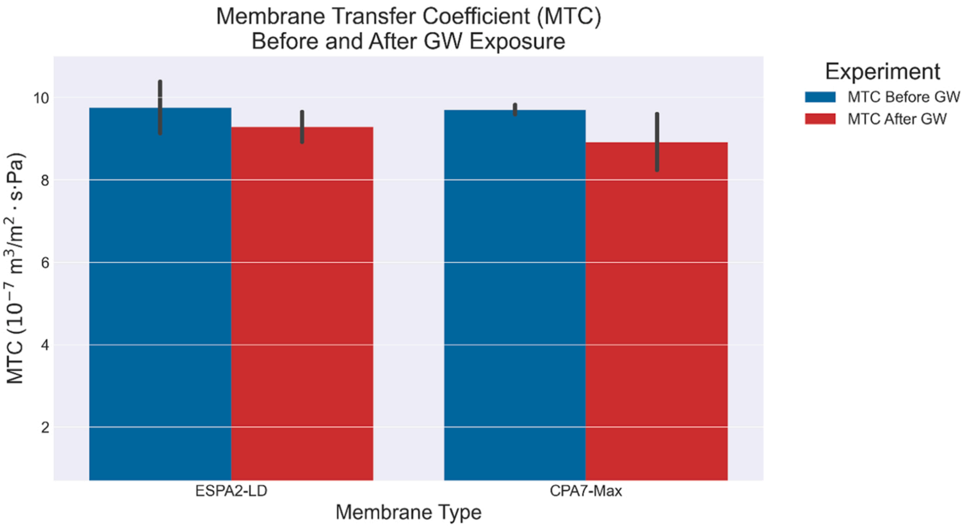

Fig. 4. Potential membrane fouling based on MTC values (MTC ≥ 7.0 × 10^{-7} $m^3/m^2\cdot s\cdot Pa$). The figure is zoomed in to emphasize higher MTC values. MTC Before GW represents measurements with demineralized water before groundwater experiments, while MTC After GW corresponds to measurements with demineralized water after completing all groundwater experiments. Error bars represent standard deviation of triplicate tests (n = 3).

charge interactions, reject ions more effectively compared to larger or neutral species, although their study mainly focused on ion rejection and Donnan exclusion [24]. The addition of Na_2S, with or without antiscalant, reduced sulfide removal efficiency. While CPA7-MAX showed slightly higher sulfide rejection, the difference was not significant when considering the error margin (represented by error bars in the graph).

Observed Na^+ rejection was slightly lower than the projection program results, particularly for the CPA7-MAX membrane. Fig. 7(y-axis zoomed to 95–100 %) shows consistently high Na^+ rejection (>96 %) with minor variations. The addition of Na_2S and/or antiscalant did not significantly affect Na^+ rejection. A full assessment of membrane selectivity would require Cl^- rejection analysis, which was not included in this study. The experimental setup and projection model use different assumptions and conditions, including flux, recovery, and feedwater composition. These differences likely explain the deviation between measured and simulated Na^+ rejection.

Fig. 8 compares the removal efficiency of Arsenic (III) and Boron using CPA7-MAX and ESPA2-LD membranes under three conditions: Virgin GW (groundwater only), GW + Na_2S + AS (with sodium sulfide and antiscalant), and GW + Na_2S (with sodium sulfide only). The data show that both membranes achieve over 60 % removal efficiency for arsenic and boron. Notably, arsenic removal is consistently higher than boron removal across all conditions, aligning with the findings of Teychene et al. [34].

This observed pattern is likely related to differences in molecular size, charge, and speciation behavior. At neutral pH and under anaerobic conditions, As(III) typically exists as the uncharged molecule H_3AsO_3, while boron primarily occurs as the uncharged species $B(OH)_3$. Both species are poorly rejected by RO membranes due to their small size and lack of charge, limiting electrostatic repulsion. However, As(III) has a larger molecular radius than boron, leading to slightly higher rejection through steric hindrance. Additionally, differences in membrane surface charge and density may influence rejection; tighter membranes with higher surface charge (like CPA7-MAX) enhance Donnan exclusion and

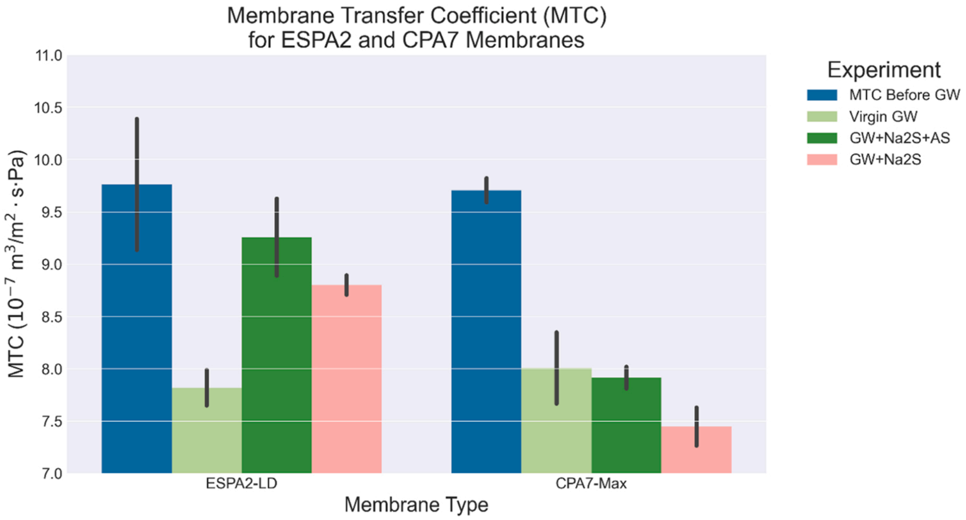

Fig. 5. Comparison of membrane mass transfer coefficients (MTC) under different groundwater (GW) conditions, using MTC Before GW (demineralized water) as the baseline. Conditions include Virgin GW (untreated groundwater), GW + Na_2S + AS (groundwater with sodium sulfide and antiscalant), and GW + Na_2S (groundwater with sodium sulfide only). Error bars represent the standard deviation of triplicate tests ($n = 3$).

Table 4
Results of the projection program for comparing different membranes.

Description	Unit				
Membrane	-	ESPA2-LD	CPA7-MAX	CPA7-MAX	SWC 5 Max
Application	-	Brackish	Brackish	Brackish	Seawater
Feed flow	m3/h	10	10	10	10
Permeate flow	m3/h	6	6	6	6
pH	-	7	7	7	7
recovery	-	60 %	60 %	60 %	60 %
Temperature	oC	14	14	14	14
Feed pressure	bar	14.5	17.3	41.1	51.9
Concentration of Na in feed	mg/L	1200	1200	7000	7000
Concentration of Cl in feed	mg/L	1849.5	1849.5	10789	10789
Concentration of Na in permeate	mg/L	18.7	11.8	85	24.3
Concentration of Cl in permeate	mg/L	28.7	18.1	131	37.5
Rejection of Na	%	98.45 %	99.02 %	98.79 %	99.65 %
Rejection of Cl	%	98.45 %	99.02 %	98.79 %	99.65 %

size exclusion, improving arsenic retention more than boron. This is consistent with prior studies on polyamide membranes, which show that surface charge and pore size are key determinants of solute rejection [23,24]. The formation of negatively charged thio-arsenic complexes under sulfide dosing further enhances electrostatic repulsion, while neutral boron species remain poorly rejected [36].

pH was not adjusted and remained near neutral (pH ≈ 7.7) under all conditions. This study did not evaluate the pH effect on arsenic rejection. Future research should include controlled pH variation to understand its influence on arsenic speciation and rejection.

Boron and arsenic, both metalloids, exhibit intermediate properties between metals and non-metals, affecting their interaction with water and membrane rejection. Arsenic, with a larger ionic radius than boron, is more effectively rejected. This difference in removal efficiency is due to their distinct physicochemical properties.

Table 5 summarizes arsenic rejection efficiency, including mean and standard deviation from triplicate experiments. Membrane type caused a 7 % difference in arsenic rejection, with CPA7-MAX consistently outperforming ESPA2-LD under all tested conditions. This difference was statistically significant (two-tailed *t*-test; $p < 0.001$). The low p-value confirms the reliability of the observed effect.

This study reports arsenic (III) rejection of 83.5–89.1 % for commercial BWRO membranes under natural anaerobic groundwater conditions, which is notably higher than the 40–80 % range reported in previous studies [34,38]. This improvement is likely due to the tighter polyamide layer of BWRO membranes and the use of real groundwater with natural ionic strength, which may enhance size exclusion and reduce diffusive transport. Many prior studies used synthetic feed waters with simplified ionic compositions, which may not fully capture the concentration polarization and membrane-solute interactions present in real systems.

The consistent outperformance of CPA7-MAX over ESPA2-LD aligns with its higher salt rejection rating (99.8 % vs 99.6 %), suggesting that even small differences in membrane structure can significantly impact contaminant rejection under field-relevant conditions. This aligns with previous findings indicating denser, more negatively charged membranes show higher rejection of neutral and weakly charged species due to enhanced Donnan exclusion and steric hindrance [23,24]. CPA7-MAX's superior performance likely results from its tighter polyamide active layer, enhancing Donnan exclusion and steric hindrance.

Within each membrane type, the impact of sulfide and BD30 addition on arsenic rejection appeared to vary. For ESPA2-LD, Na_2S significantly reduced arsenic rejection compared to virgin groundwater ($p = 0.034$), whereas Na_2S + BD30 showed no significant effect ($p = 0.078$). Conversely, for CPA7-MAX, arsenic rejection was significantly reduced under Na_2S + BD30 ($p = 0.001$), but not under Na_2S alone ($p = 0.070$). These trends suggest a potential membrane-specific response to chemical conditions; however, given the limited replication ($n = 3$), these findings should be interpreted cautiously and validated in future studies.

For CPA7-MAX, the addition of Na_2S with antiscalant or Na_2S alone has a negligible effect on boron and arsenic removal efficiency, indicating stable performance regardless of feed water composition. In contrast, ESPA2-LD shows a decline in removal efficiency when Na_2S is introduced. Na_2S may interact with the membrane material, altering surface properties or pore structure, reducing its rejection efficacy for both ions.

As previously discussed, sulfide addition did not enhance arsenic and boron rejection, contrary to expectations and prior studies. This unexpected result led to additional experiments to verify the accuracy of sulfide dosing. To assess this, sulfide concentration was measured over time, with the results presented in Fig. 9.

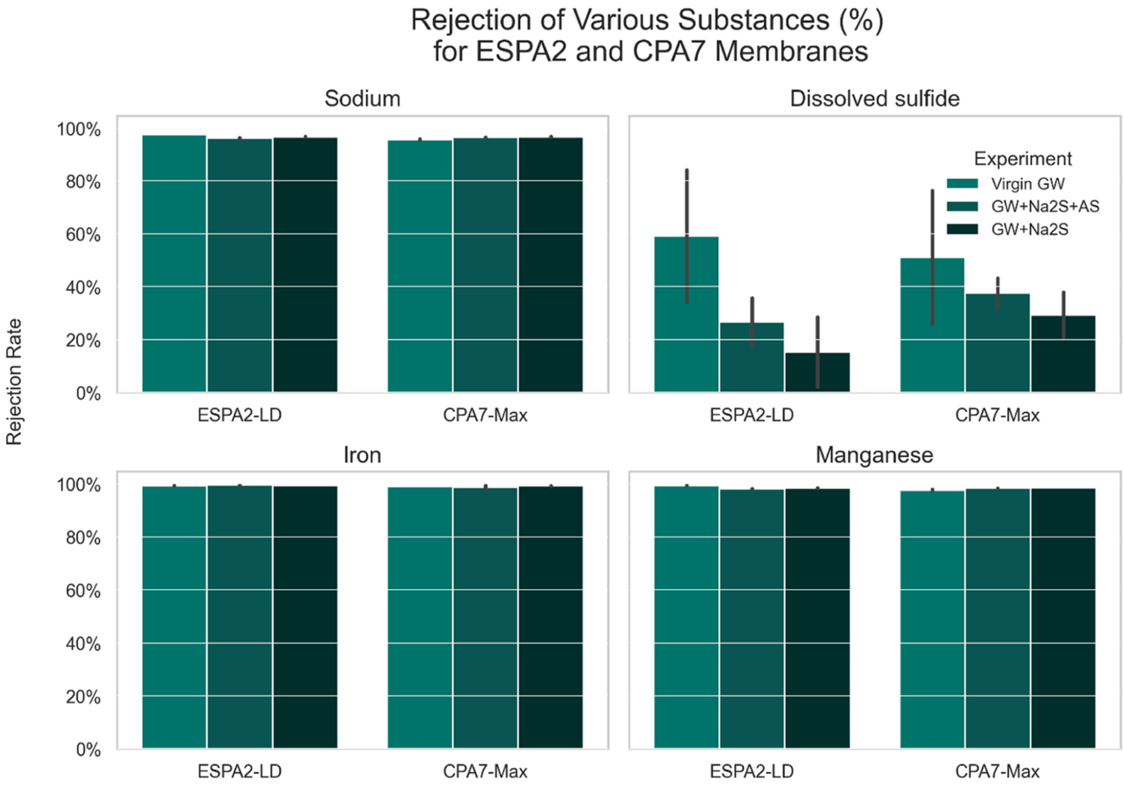

Fig. 6. Comparison of ion rejection in percentage (Sodium, Iron, Dissolved Sulfide, and Manganese) by ESPA2-LD and CPA7-MAX membranes. Error bars represent the standard deviation of triplicate tests (n = 3).

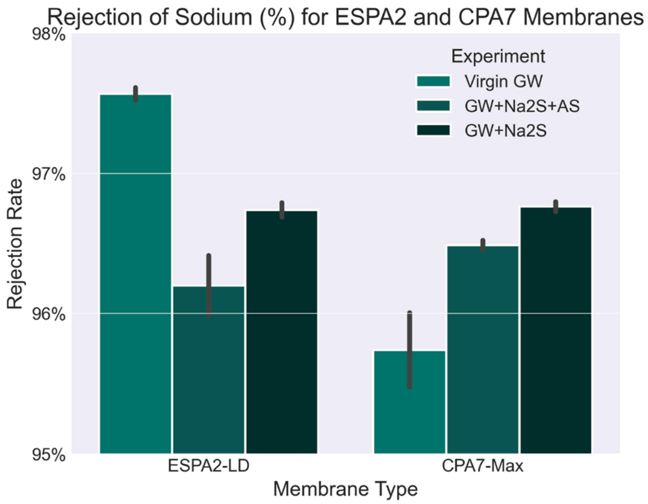

Fig. 7. Sodium rejection by ESPA2-LD and CPA7-MAX membranes in percentage (95–100 % zoomed range). Error bars represent the standard deviation of triplicate tests (n = 3).

The results show a rapid initial decline in sulfide concentration, dropping by ±72 % within 0.5 h (0.02 days). This suggests rapid oxidation or volatilization immediately after dosing, occurring independently of antiscalant presence.

A distinct difference in sulfide concentration emerges from Day 4 onward. Without antiscalant, sulfide levels stabilized at ±8.2 mg/L on Day 4, gradually decreasing to 6.3 mg/L by Day 6. In contrast, with antiscalant, sulfide concentrations were lower, ranging between 4.5 and 5.6 mg/L during this period. This suggests that the antiscalant accelerates sulfide depletion, likely through oxidation, precipitation, complexation, or other chemical interactions. The rapid decline in sulfide concentration indicates that pH buffering with $NaHCO_3$ did not prevent H_2S volatilization under the applied conditions.

Sulfide dosing solutions were prepared one or several days before use. By the time they were applied to the feed water, the actual sulfide concentration had already declined, resulting in significantly lower effective dosing than initially calculated. This rapid decline explains the lack of improvement in As(III) rejection, as previous studies [35] have indicated that a minimum concentration of approximately 1 mg/L (±30 μM) sulfide is required to promote effective sulfide-arsenic complexation. It is likely that sulfide concentrations in our study were insufficient to achieve this threshold under the tested conditions.

Sulfide complexation with arsenic depends on both the concentration and stability of sulfide in the feed water. Prior studies have shown that at sufficient concentrations and under reducing conditions, thioarsenite or thioarsenate species may form, particularly when sulfide-to-arsenic ratios exceed 3:1 [35]. These negatively charged complexes could, in principle, be more easily rejected by RO membranes via Donnan exclusion. However, in the present study, rapid sulfide depletion through oxidation or volatilization likely prevented meaningful complex formation. In the absence of sustained sulfide availability, arsenic speciation remains dominated by the neutral form As(III), resulting in no measurable improvement in rejection efficiency.

The results show that sulfide dosing does not reliably enhance arsenic rejection in BWRO due to rapid depletion during storage before dosing. Antiscalant further reduces sulfide stability, likely accelerating its loss. Further investigation is needed to identify the specific chemical mechanisms behind sulfide depletion, including oxidation, volatilization, and/or precipitation. Exploring alternative stabilization methods, such as pH adjustment or closed systems, could help maintain effective sulfide levels for a longer duration. Alternatively, preparing sulfide solutions shortly before use or employing in-line dosing systems could help maintain sufficient sulfide concentrations in the feed water. However, the experiments revealed that rapid sulfide depletion—likely due to oxidation or volatilization—prevented stable complex formation with

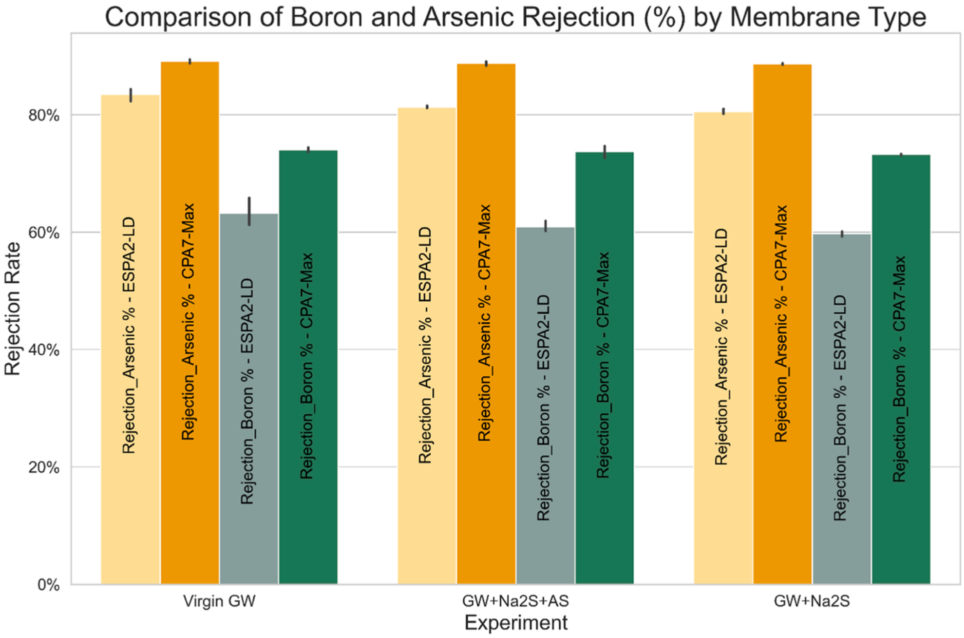

Fig. 8. Rejection of Arsenic (III) and Boron in percentage (%) by ESPA2-LD and CPA7-MAX membranes under three conditions: Virgin GW (groundwater only), GW + Na_2S + AS (groundwater with sodium sulfide and antiscalant), and GW + Na_2S (groundwater with sodium sulfide only). Error bars represent the standard deviation of triplicate tests (n = 3).

Table 5
Arsenic (III) rejection efficiency (% mean ± standard deviation) of ESPA2-LD and CPA7-MAX membranes under three different feedwater conditions.

Membrane	Condition	Arsenic rejection (%) ± SD
ESPA2-LD	Virgin GW	83.50 ± 1.33
ESPA2-LD	GW + Na_2S	80.52 ± 0.43
ESPA2-LD	GW + Na_2S + BD30	81.33 ± 0.22
CPA7-MAX	Virgin GW	89.14 ± 0.36
CPA7-MAX	GW + Na_2S	88.68 ± 0.15
CPA7-MAX	GW + Na_2S + BD30	88.77 ± 0.36

arsenic. This limitation highlights that, under real-world conditions, Na_2S dosing may not consistently enhance arsenic rejection unless sulfide stability is ensured. These findings underscore the need to control sulfide reactivity and minimize storage time when designing arsenic removal strategies.

Although CPA7-MAX demonstrated superior ion rejection, sulfide dosing did not significantly enhance arsenic removal, contrary to expectations. Future studies should focus on optimizing sulfide stabilization to improve long-term rejection efficiency.

4. Conclusion

This study demonstrated the comparative efficiency of ESPA2-LD and CPA7-MAX membranes in arsenic removal from brackish

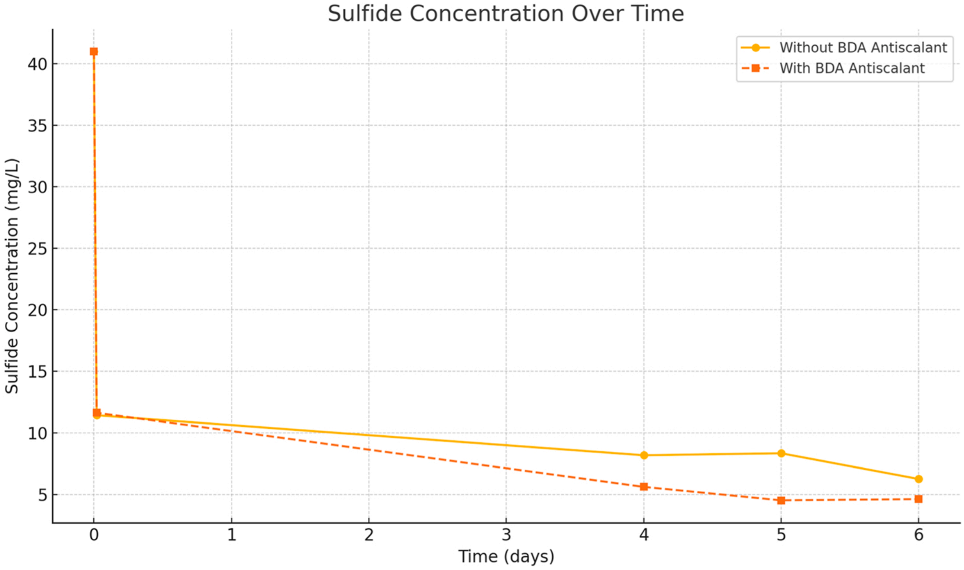

Fig. 9. Sulfide concentration over time in the presence and absence of antiscalant. A rapid initial decrease is observed within the first 0.5 h, followed by a slower decline. Error bars represent the standard deviation of triplicate tests (n = 3).

groundwater under anaerobic conditions, a configuration rarely evaluated in prior studies. The results showed that CPA7-MAX generally exhibited lower membrane transport coefficients (MTC) than ESPA2-LD, likely due to differences in surface structure or the polyamide separation layer. However, this study did not include measurement and analysis of these layers, as it was outside the scope of the practical research. Future research should focus on such membrane characterization for a more comprehensive understanding.

Both membranes experienced a reduction in MTC after exposure to brackish groundwater, indicating the influence of salts on membrane performance, likely due to the formation of a thicker concentration polarization layer.

Na_2S was added to enhance arsenic removal by facilitating the formation of chemical complexes with arsenic, and antiscalant, a standard procedure in membrane processes, to assess its potential impact on membrane performance as a consequence of the effects of sulfide complex formation under real-world conditions. The addition of Na_2S with antiscalant or Na_2S alone did not significantly affect membrane performance, suggesting that the surface composition or structure of the membranes primarily determines ion rejection. Both membranes consistently achieved high rejection rates (>95 %) for sodium, iron, and manganese, while sulfide rejection was notably lower, likely due to its molecular speciation.

However, experiments revealed that rapid sulfide depletion—likely due to oxidation or volatilization—prevented stable complex formation with arsenic. This limitation highlights that, under real-world conditions, Na_2S dosing may not consistently enhance arsenic rejection unless sulfide stability is ensured. These findings emphasize the need to control sulfide reactivity and minimize storage time when designing arsenic removal strategies.

Arsenic removal was consistently above 80 % for both membranes, with arsenic being more efficiently removed than boron, mainly due to arsenic's larger ionic size. The addition of Na_2S did not significantly affect CPA7-MAX but negatively impacted ESPA2-LD, likely due to interactions between Na_2S and the membrane material. Additionally, because the feedwater pH remained near neutral and was not adjusted during the experiments, this study does not assess the pH dependence of arsenic rejection.

These findings highlight that boron rejection can serve as an indicator for arsenic removal when both contaminants are present in the feedwater.

Future investigation is needed on the long-term performance and stability of these membranes in full-scale systems under varying chemical conditions. Specifically, the impact of sulfide dosing delay should be studied, and methods to stabilize sulfide for longer-lasting effects should be explored. Additionally, exploring surface and structure modifications to improve membrane resilience would be valuable.

This study adds new insight into the performance of BWRO membranes under anaerobic conditions using natural groundwater, including the role of membrane type, ion speciation, and operational dosing limitations. Overall, the findings underscore the importance of tailored membrane selection and chemical treatment strategies based on specific contaminant profiles and feedwater conditions.

CPA7-MAX's higher arsenic rejection may justify its use in areas with strict limits (≤1 μg/L), despite slightly higher energy needs. The lack of improvement with Na_2S dosing underscores the challenge of maintaining sulfide stability in real systems. Future pilot-scale studies will validate these findings for full-scale application, including economic tradeoffs in membrane and chemical selection.

CRediT authorship contribution statement

T. Rijnaarts: Writing – review & editing, Supervision, Project administration, Methodology, Formal analysis, Data curation. **E.R. Cornelissen:** Writing – review & editing, Supervision, Conceptualization. **T. van Dijk:** Writing – review & editing, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **W. G. Siegers:** Methodology, Investigation. **Haidari Amir Hoseen:** Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the author(s) used ChatGPT to improve the language and readability of the article. After using this tool, the author(s) reviewed and edited the content as needed and take full responsibility for the content of the publication.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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