Research Article

# Study of Hybrid Performance of the Methods Applied for Recycling Aq Qala-Industrial Park Effluent

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## Abstract

**Aim:** Currently, recycling of secondary effluent can be used sustainably as a new water source to minimize the water consumption in areas which are exposed to water crisis. Hence, it is necessary to appraise hybrid advanced treatment technologies performance and to determine the best alternative for reusing secondary effluent of industrial park in full-scale. **Materials and Methods:** The secondary effluent (biological-treated) of the centralized industrial park wastewater treatment plant (Aq Qala, Golestan province, Iran) is introduced into combined systems such as (1) sand filter (SF) and membrane bioreactor (MBR), (2) SF, MBR and granular activated carbon (GAC) (3) SF and GAC (4) SF, MBR, GAC, and reverse osmosis (RO), as the hybrid advanced scenarios. **Results:** The effluent of SF/MBR/GAC/RO showed the highest quality (>99% removal efficiency). In this scenario, pH, silica, manganese, iron, total suspended solids, turbidity, total coliform, and chemical oxygen demand (COD), alkalinity, hardness, total dissolved solids (TDS), chloride, and sulfate were determined 6.93 ± 0.19, 1.4 ± 0.6 mg/L, not detectable (ND), ND, <2 ± 0.2 mg/L, <1 Nephelometric Turbidity unit (NTU), ND and <2 ± 0.2 mg/L, 54.8 ± 1 2.5 mg/L, 50 ± 17 mg/L, 100 ± 14.89 mg/L, 68 ± 10.9 mg/L, and 44 ± 3.67 mg/L were observed in the range of product water standard for sensitive industries. Also, the maximum of efficiency of SF/MBR, SF/MBR/GAC, and SF/GAC systems was obtained 97.75% (as total coliforms), 62.65% (as COD), and 55.8% (as COD), respectively. Other parameters removed slight about 2% to 40%. However, hardness, alkalinity, and manganese concentrations not reduced after these systems (0% efficiency). **Conclusions:** The hybrid system of SF/MBR/GAC/RO was produced a clean and suitable water supply for the sensitive industries (e.g., intermediate-pressure boilers, cooling water, textile, etc.) of Aq Qala industrial park according to the environmental protection agency standards.

**Keywords:** Advanced treatment plants, Aq Qala, hybrid systems, industrial park, recycle, reuse

## INTRODUCTION

Today, water resources are a critical issue around the world, particularly in the agricultural and industrial regions. The abatement of water resources quantity and quality has been provided the economic, social, and environmental concerns. However, the sustainable water resources management has been recognized as the major problem in the world. The organizations should employ the efficient politics and measures for providing required water.[1,2] One of these policies, exploring the new water supply source, which meets some of the water demands. Wastewater can be considered as a water resource to compensate for the water shortages, but it includes the high contaminants.[3] Due to the inefficiency of wastewater treatment plants (WWTPs), large quantities of effluent are discharged into the natural water resources with poor quality.[4,5] The most important effects of wastewater include significant river contamination, the destruction of living organisms,[6] the production of unpleasant odors and scenery, and the accumulation of hazardous material in the food chain. Therefore, human and environment health endanger seriously and is as an important concern worldwide.[4,5,7] United Nations Environment Program (UNEP) reported 90% of developing

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countries wastewaters introduced to the water sources without treatment.[8]

Among wastewaters, industrial wastewater is considered the most dangerous wastewater.[9] Industrial wastewater includes a wide range of contaminations from hazardous materials to heavy metals. If they are not treated properly, it can have severe effects on water, plants, and animals.[8] Currently, industrial wastewater is often used for reuse and reclamation applications as one of the most effective options to minimize the water consumption in areas which are exposed to water crisis.[9-12]

Moreover, recycling of secondary effluent can be used sustainably as a new water source for various industries (e.g., washing, painting, cooling water, or boilers-feed)[12] if the advanced wastewater treatment process is selected appropriately. Many factors are considered in the selection of the appropriate process such as the quantity, quality, and the application of effluent, which have to be affected the durability, efficiency, and cost-effectiveness of the process.[13]

The advanced treatment processes include advanced oxidation processes (AOPs), adsorption, membrane systems such as membrane bioreactor (MBR), and reverse osmosis (RO) to be used for effluent reuse.[14] The high oxidation of AOP causes by OH reactivity but it is changed by scavenger factors. These factors decline the oxidation force of OH with organic contaminants.[15] On the other hand, the adsorption process can apply for organic and inorganic contaminants. However, one of the disadvantages is its low removal efficiency in the reuse application.[16] Furthermore, the biological method (e.g., wetland) did not obtain the removal efficiencies of total suspended solids (TSS) and TDS according to the reuse water standard in the industry.[17]

Recently, literature review has been reported the performance of hybrid advanced treatment technologies for reuse applications.[9] Du *et al*. examined the ability of powdered activated carbon (PAC)-MBR system for groundwater treatment. These plants had the significant performance at high inorganic pollutants because of the microorganism's growth.[18] Yang *et al*. reviewed the role of membrane processes for reusing of municipal wastewater. They found these processes contribute reuse standards through reducing organic and inorganic contaminants, energy consumption, and increasing recovery rate.[19] Furthermore, Yang *et al*. described the combination of moving bed biofilm reactor with MBR is decreased retention time and is raised the removal efficiency of chemical oxygen demand (COD), TSS and color. Moreover, these processes effluent can be consumed in the textile industry and are decrease environmental and economic concerns.[20] Gunes and Gonder used electro-coagulation (EC) process as pretreatment stage before nanofiltration (NF) and RO plants for the treatment of textile effluent. They found EC process can be decreased cake resistance and TDS for NF plant.[21] The hybrid systems, especially membrane processes can be achieved high-quality water for industry. In addition, the combination of granular activated carbon (GAC) process with advanced oxidation processes (AOPs) (e.g., ozone) are provide disinfection process regardless of high removal of organic contaminants[22] or GAC plant is considered as a pretreatment before membrane filtration (MF) for alleviating membrane fouling.[23]

However, there are few reports on the successful application of the hybrid system for reusing of industrial park effluent in full-scale. Furthermore, we studied the ability of the hybrid treatment processes in water source production of the sensitive industries (intermediate-pressure boilers, cooling water, textile, pulp, and paper industries) in this research. We evaluated the removal efficiency of major and important physicochemical and microbial parameters of effluent during each hybrid processes. These parameters include pH, COD, TSS, TDS, chloride, sulfate, turbidity, iron, manganese, total coliform, alkalinity, and hardness (total).

## MATERIALS AND METHODS

### Description of the hybrid treatment plants

The experiment was done at the industrial park WWTP, Aq Qala county, Golestan province, Iran. The capacity of WWTP is considered about 690 $m^3$/d. Industrial park includes 254 factories (e.g., boiler, food, chemical, cellulose, textile, metal, pharmaceutical solutions, electrical, etc.) introduce into centralized WWTP. Wastewater is treated through the combined extended aeration and sequence batch reactor (SBR) in the secondary treatment plants following conventional primary treatment plants. The geographical location of the industrial park WWTP is brought in Figure 1.

The secondary effluent (biological-treated) from the industrial park WWTP was induced into hybrid advanced treatment scenarios such as (1) sand filter (SF) and membrane-biological reactor (MBR), (2) SF, MBR and GAC, (3) SF and GAC, and (4) SF, MBR, GAC, and RO. The physical, chemical, and microbial characteristics of the secondary effluent and the qualities of produced water were compared to sensitive industries appropriate criteria[24,25] represented in Table 1. These parameters were analyzed according to available and existing standards of various industries in Aq Qala industrial park.

Four scenarios used the different tertiary plants to determine the performance of most suitable and available system in the sustainable development plans. The following treatment processes were performed by wastewater reclamation necessity for the intermediate-sensitivity industries in the industrial park. Initially, the effluent of the secondary treatment plant added to the SF to remove suspended solids. A pressure SF (with 20 $m^3$/h capacity) filled with two layers of anthracite (40 cm height) and four layers of silica. The anthracite layer placed at the bottom and top of the silica layers. The characteristics of silica layers are shown in Table 2. Then, the effluent transferred to MBR (SF/MBR) with 10000–15000 mg/L MLSS. The MBR system comprised two units with UF filters in size of 0.04µm (polyethersulfone (PES), German), and 15–30 $m^3$/h

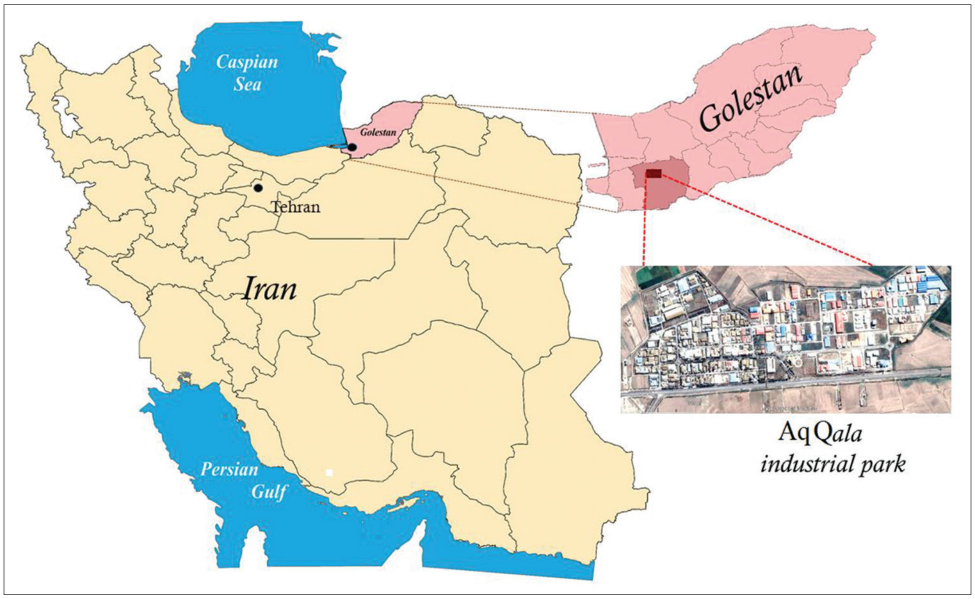

**Figure 1:** The geographical location of the industrial park wastewater treatment plant of Aq Qala

**Table 1: Physical, chemical and microbial characteristics of the secondary effluent and the water quality criteria for sensitive industries[24,25]**

| Parameter | Secondary effluent (mg/L) | Water quality |
|---|---|---|
| Iron (mg/L) | 0.07±0.04 | 0-0.1 |
| Manganese (mg/L) | 0.014±0.01 | 0-0.05 |
| pH | 7.53±0.26 | 6-10 |
| Hardness (total) (mg/L) | 501.4±164 | 0-100 |
| Alkalinity (mg/L) | 462.9±133.5 | 0-75 |
| Sulfate (mg/L) | 444.86±23.2 | 0-100 |
| TSS (mg/L) | 53±32.32 | 0-5 |
| TDS (mg/L) | 1631.7±150 | 0-100 |
| Silica (mg/L) | 42.2±7.3 | 0-10 |
| Chloride (mg/L) | 838.6±63.8 | 0-100 |
| COD (mg/L) | 55.43±22.45 | 0-10 |
| Total coliform (mg/L) | 293.6±42.9 | - |
| Turbidity (NTU) | 9.3±5.8 | - |

TSS: Total suspended solids, TDS: Total dissolved solids, COD: Chemical oxygen demand, NTU: Nephelometric turbidity units

**Table 2: The characteristics of silica layers used in sand filter**

| Silica layer | Particle size (mm) | Uniformity coefficient | Degree of purity (%) |
|---|---|---|---|
| 1 | 0.5-1.5 | 1.35 | >95 |
| 2 | 2-3.5 | 1.35 | >95 |
| 3 | 4-8 | 1.35 | >95 |
| 4 | 8-12 | 1.35 | >95 |

capacity. During the experiments of scenarios of SF/MBR/GAC and SF/GAC, MBR effluents were treated by GAC to remove odor and color. GAC designed a specific surface area of 1000 $m^2/g$ (Jacobi company) and 15 $m^3/h$ capacity before subjected to RO filtration (scenario of SF/MBR/GAC/RO). RO considered for the further reduction of COD and some anion & cation concentrations. RO had a circular structure and 15 $m^3/h$ capacity. Properties of RO are represented in Table 3. The operating conditions of hybrid scenarios must achieve the required standard for the water production of the intermediate-sensitivity industries.

A schematic of the hybrid system scenarios of the industrial park represents in Figure 2. Samples were collected twice monthly for four months and maintained at a temperature of 4°C and dark. Then, the considered parameters measured in each of the treatment stages.

### Effluent quality analysis

The spectrometry (Rayleigh spectrometry ultraviolet [UV] 9200) was applied for the determination of COD of the secondary effluent according to the standard methods[26] using closed reflux and colorimetric method.[27] Furthermore, pH and TDS were measured by Hatch pH meter and online probe. Iron (II), sulfate, and manganese concentrations were measured using phenanthroline and turbidimeter. Turbidity was measured through nephelometric method. Furthermore, silica was defined by the analytical method as molybdate-reactive silica. Titration was alkalinity, hardness ($Ca^{2+}$ and $Mg^{2+}$ cations), and chloride (Mohr).[22] Finally, the most probable number (MPN) test used for measuring total coliforms.[26]

### Statistical analysis

A Q-TEST analysis was used to eliminate statistical outliers in data. Then, the average and standard deviation of the result achieved for each parameter.

## RESULTS

The average of concentration of physicochemical and microbial parameters is summarized in Table 4. Different

**Figure 2:** Scenarios of hybrid advanced system in the industrial park (a) scenario 1, (b) scenario 2, (c) scenario 3, (d) scenario 4

**Table 3: The characteristics of reverse osmosis membrane**

| Parameter | Properties |
|---|---|
| Membrane material | PA semi-permeable |
| Membrane type | Spiral wound (Uniha, Austria) |
| Operating pressure (bar) | 15 |
| Membrane area (ft$^2$) | 80 |
| pH range | 4-9 |
| Temperature range (°C) | 2-45 |

PA: Polyamide

combinations of the process were evaluated as a scenario. Table 4 shows the parameters concentration studied in the proposed scenarios.

## pH

According to Table 4, pH reached 8.33 ± 0.2, 8.3 ± 0.3, 7.99 ± 0.21 and 6.93 ± 0.19 in SF/MBR, SF/MBR/AC, SF/AC and SF/MBR/AC/RO scenarios. The pressure SF process had a pH of 7.66. The standard pH range for sensitive industries are 6–10 (e.g., Textile, leather manufacturing, petrochemical, chemical, cement, paper and cardboard, food, and boiler industries). As shown in Table 1, pH of all scenarios was in a standard pH range and their pH is acceptable for the sensitive industries. SF/MBR/AC/RO scenario (pH = 6.93 ± 0.19) is not needed to add the chemical material for adjusting pH value. Furthermore, other physicochemical and microbial parameters are described as follows.

## Removal of total suspended solids and turbidity

As observed, TSS removal efficiency achieved 25%, 32%, 43%, and >99% from 53 mg/L of influent [Table 4] during SF/MBR, SF/MBR/GAC, SF/GAC, and SF/MBR/GAC/RO scenarios. As depicted in Table 4, the removal efficiency observed more in SF/GAC than SF/MBR, SF/MBR/GAC. Although TSS in SF/GAC effluent was decreased (30.20 ± 7.93 mg/L in effluent), it is not in the acceptable range of Table 1. The highest removal occurred in the scenario of SF/MBR/GAC/RO and then SF/AC. According to Table 1, only scenario of SF/MBR/GAC/RO (<2 mg/L) is provided in the standard range.

Figure 3a represents the average of effluent TSS values each treatment process of SF/MBR/GAC/RO during March, April, May, and June months. The effluent of filtration, MBR, activated carbon, and RO was 49.31, 40.49, 36.09, and <2 mg/L, respectively. Averagely, TSS removal efficiency was 6.96%, 23.60%, 32%, and >99%.

As shown in Figure 3a, TSS did not reach the standards of the sensitivity industries in the filtration, MBR, and GAC effluents. However, the GAC process has reduced TSS more completely during the sampling months. SF process in the studied scenarios did not remove TSS value in April. Furthermore, SF provided the lowest performance during sampling months [Figure 3a] and it increases loading TSS with MBR. The variations of removal efficiencies in the processes of scenarios were similar during sampling months. Moreover, secondary effluent turbidity (9.3 ± 5.8 NTU) acquired 2 ± 0.5, 2 ± 0.5 and 4.1 ± 1 NTU during SF/MBR, SF/MBR/GAC, and SF/GAC according to Table 4. Conversely, turbidity removal efficiency of SF/GAC determined 23% lower than SF/MBR and SF/MBR/GAC. The highest turbidity removal efficiency obtained in the scenario of SF/MBR/GAC/RO >SF/MBR = SF/MBR/GAC >SF/GAC [Table 4].

Figure 3b exhibits the average of turbidity in the effluent of filtration, MBR, activated carbon, and RO during March, April, May, and June months. The average removal efficiency of processes was 3.22%, 78%, 78%, and >99% during the sampling months. MBR and GAC had similar turbidity removal efficiency, but RO had an identical performance during the sampling months. In April, the performance of processes was determined best in comparison with other months. However, turbidity value received the required

**Table 4: The average of concentration of physicochemical and microbial parameters in different scenarios effluent**

| Parameter | Scenario | | | |
|---|---|---|---|---|
| | 1. SF + MBR | 2. SF + MBR + GAC | 3. SF + GAC | 4. SF + MBR + GAC + RO |
| pH | 8.3±0.2 | 8.3±0.3 | 7.99±0.21 | 6.93±0.19 |
| TSS (mg/L) | 40.49±10.4 | 36.09±5.3 | 30.20±7.93 | <2±0 |
| Turbidity (NTU) | 2±0.5 | 2±0.5 | 4.1±1 | <1±0 |
| TDS (mg/L) | 1656±146.6 | 1615.5±94.9 | 1509±55.63 | 161±14.89 |
| Silica (mg/L) | 49.9±8.7 | 33.5±5.7 | 45.1±5 | 1.4±0.6 |
| Iron (mg/L) | 0.029±0.01 | 0.047±0.01 | 0.04±0.01 | ND±0 |
| Manganese (mg/L) | 0.026±0.01 | 0.028±0.01 | 0.026±0.012 | ND±0 |
| Sulfate (mg/L) | 428.9±37 | 378.6±21 | 430.6±47 | 44.1±3.67 |
| Chloride (mg/L) | 989.3±80.3 | 1017.9±72 | 814.3±56.4 | 68.1±10.9 |
| Hardness (mg/L) | 532.9±36.4 | 527.1±45 | 512.7±24.7 | 50.8±17 |
| Alkalinity (mg/L) | 512.9±70.9 | 502.9±52 | 513.7±97.5 | 54.8±12.5 |
| COD (mg/L) | 26.86±5 | 20.7±5.9 | 24.5±7 | <2±0 |
| Total coliforms (CFU/100) | 6.6±2 | 124.9±20.3 | 420.4±30 | ND±0 |

TSS: Total suspended solids, TDS: Total dissolved solids, COD: Chemical oxygen demand, NTU: Nephelometric turbidity units, SF: Sand filter, MBR: Membrane-biological reactor, GAC: Granular activated carbon, RO: Reverse osmosis, ND: Not detectable

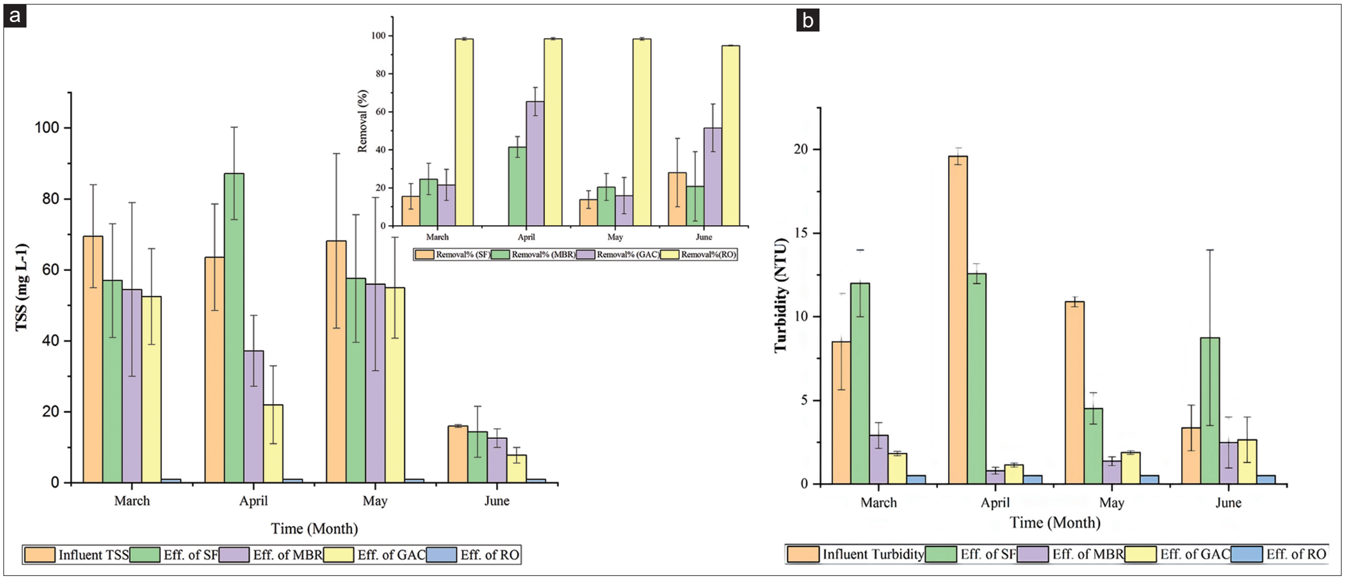

**Figure 3:** Variations of (a) TSS and (b) turbidity of plants effluent of hybrid treatment scenario 4 during the sampling time. TSS: Total suspended solids

standard of the industry in MBR and GAC effluent before the RO process.

## Removal of total coliforms

As observed in Table 4, SF/MBR after SF/MBR/GAC/RO presented the highest removal of total coliforms which their residue concentration was determined 6.6 ± 2 CFU/100 mL and not detectable respectively. Hence, the performance of SF/MBR was evaluated almost similar with SF/MBR/GAC/RO in the removal of total coliforms. Then, the residue concentration in SF/MBR/GAC and SF/GAC has designated 124.9 ± 20.3 and 420.4 ± 30 CFU/100 mL in effluent.

Moreover, the removal efficiency and effluent concentration of hybrid plants are depicted in Figure 4. The average density of total coliforms was also decreased from 293.5 MPN (in the secondary effluent) to 431.7, 6.6, 124.9 CFU/100 mL, and not detectable after the filtration, MBR, activated carbon, and RO [Figure 4]. Hence, the removal efficiency 0%, 97%, 57%, and >99% in the sand filtration (SF), MBR, GAC, and RO. The lowest of MPN number obtained during RO and MBR processes in the scenarios and its highest during filtration, respectively.

## Removal of iron, manganese, silicate, and sulfate

Iron, manganese, sulfate, and silicate concentrations measured after each scenario [Table 4] and treatment process [Figure 5].

Sulfate removal efficiencies were shown 3.2% and 3.6% in scenarios of SF/MBR and SF/GAC [Table 4]. The impressiveness of these treatment processes was insignificant due to the used operating conditions of this study, so that residue concentration of sulfate was found 428.9 ± 37, 378.6 ± 21 and 430.6 ± 47 in SF/MBR, SF/MBR/GAC and SF/GAC effluents. On the other hand, around 90% of sulfate removal took place in SF/MBR/GAC/RO [Table 4] especially RO process according to Figure 5a. The sulfate concentration in the scenario of SF/MBR/GAC/RO is reached the required standards at sensitivity industrials. However, sulfate did not reach the required water quality for sensitive industries in scenarios SF/MBR, SF/MBR/GAC, and SF/GAC [Figure 5a and Table 1].

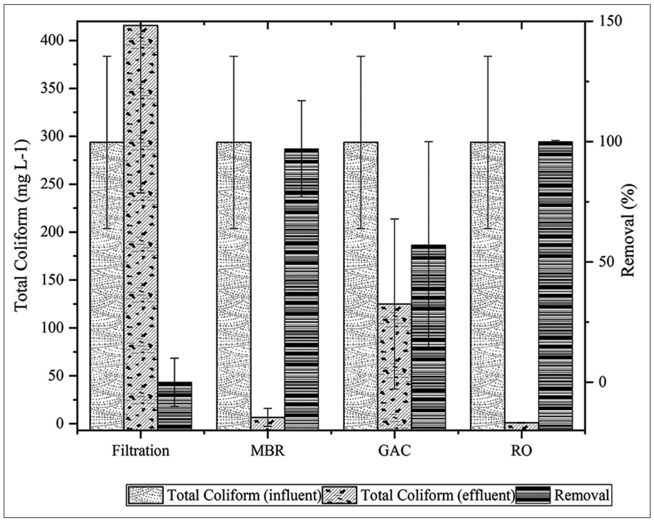

**Figure 4:** Total Coliform removal by the SF, MBR, GAC, and RO plants in the hybrid scenario 4. SF: Sand filtration, MBR: Membrane bioreactor, GAC: Granular activated carbon, RO: Reverse osmosis

All three scenarios were contributed to the removal of iron, which were 59%, 33%, 45%, and >99% during SF/MBR, SF/MBR/GAC, SF/GAC, and SF/MBR/GAC/RO according to Table 4. The highest and lowest residue iron were identified in scenarios SF/MBR/GAC and SF/MBR/GAC/RO. However, the iron of secondary effluent was lower than the water standard range in Table 1. In scenario of SF/MBR/GAC/RO, most of the iron was removed by the RO (>99%), which shown to be one of the most appropriate methods for its removal.

Although the manganese had the optimum range of water industry standard, it removed completely at SF/MBR/GAC/RO especially RO process (>99%). However, manganese residue concentration was much in the effluent of SF/MBR, SF/MBR/GAC and SF/GAC scenarios [Table 4] and processes, e.g., activated carbon, filtration, and MBR [Figure 5c].

Silica was persistent to remove appropriately by filtration as well as MBR or by the application of GAC during the scenarios of SF/MBR (49.9 mg/L ± 8.7) and SF/GAC (45.1 mg/L ± 5) [Table 4 and Figure 5d]. The efficiency of 20% of silica removal occurred

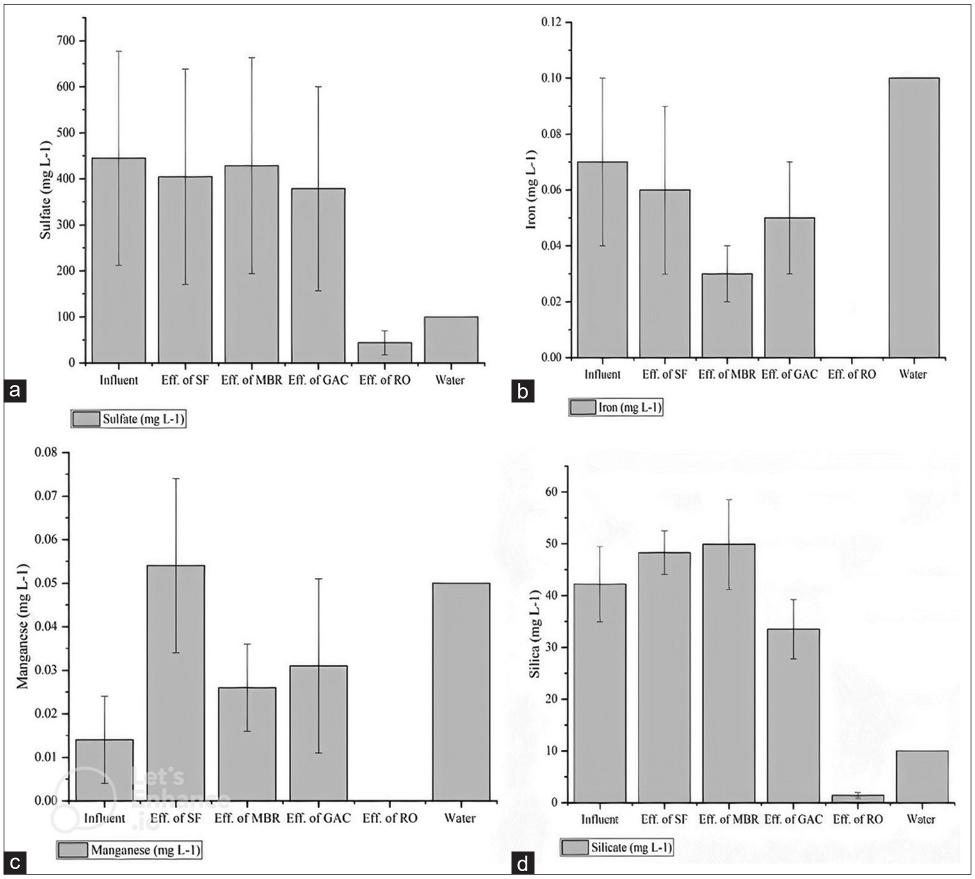

**Figure 5:** The concentration of (a) sulfate, (b) iron, (c) manganese, and (d) silicate in treatment processes effluent of scenario 4

during the scenario SF/MBR/GAC, due to the bind silica in the high pH of the activated carbon process.

### Removal of chemical oxygen demand

In the current study, the concentration of COD detected in the range of 20-26 mg/L in effluent SF/MBR, SF/MBR/GAC, and SF/GAC scenarios, and <2 mg/L in the effluent of SF/MBR/GAC/RO shows in Table 4, while influent COD determined 55.43 ± 22.45 mg/L. Scenario of SF/MBR/GAC/RO is provided suitable water quality for high sensitive industries without obstruction problems and a decrease in the production quality [Table 1]. Other scenarios (SF/MBR, SF/MBR/GAC, and SF/GAC) are appropriate for lower sensitivity industries. Averagely 51%, 62%, 60%, and >99% of COD removal obtained in effluent of scenarios SF/MBR, SF/MBR/GAC, SF/GAC, and SF/MBR/GAC/RO.

### Removal of chloride, hardness, alkalinity, and total dissolved solids

In the present work, none of SF/MBR, SF/MBR/GAC, and SF/GAC could reduce TDS, hardness, alkalinity, and chloride to standard of product water. In hybrid scenarios, SF plant effluent decreased only TDS as a level of 3.37%. According to Table 4, TDS removal detected 0%, 0.99%, and 7.5% by SF/MBR, SF/MBR/GAC, and SF/GAC.

The high removal of TDS (RE = 91.1%), hardness (RE = 89.9%), alkalinity (RE = 88.2%), and chloride (RE = 91.9%) is obtained during SF/MBR/GAC/RO.

### Cost-estimation of hybrid treatment processes

On the other hand, economic issues should be considered to predict the cost-effective application of hybrid systems for industrial park effluent reuse. The costs estimated for the studied treatment plants and scenarios annually. According to Table 5, the highest and lowest costs are related to RO and GAC plant.[9,25] On the other hand, scenarios of SF/MBR/GAC/RO and SF/GAC were estimated the highest and lowest treatment total costs, respectively, 81300$ and 20832$. Also, SF/MBR and SF/MBR/GAC were calculated 60973$ and 71000$ as total costs. The annual variable cost includes the costs of maintenance and operation such as chemical material, electrical current and repairs.

### A qualitative classification of product water

The quality and quantity of industrial water depended on processes, which select in an industry.

The industrial processes recognize the appropriate range of water used in the industry by qualitative classification. The qualitative classification of recycled effluent and product water for industrial applications is explained in Table 6.

Accordingly, the qualitative classification of the studied hybrid treatment scenarios is given in Table 7.

According to Table 7, product water of SF/MBR/GAC/RO can use for sensitive industries except for the food and hygiene industry. The product water of scenario of SF/MBR/GAC/RO can be considered in the intermediate and low-pressure boilers consumptions as one of the sensitive industries. High-pressure boilers cannot use product water of scenario SF/MBR/GAC/RO due to high hardness, alkalinity, and TDS of water. Although the water of scenario SF/MBR/GAC is appropriate for metal and chemical industries the consumption is limited due to high TDS value.

Moreover, the product water of scenarios of SF/MBR, SF/MBR/GAC, and SF/GAC cannot apply to the sensitive industries due to the TDS, alkalinity, hardness, TSS, COD, chloride, sulfate, and silica values are higher than the standard range is brought in Table 1.

## DISCUSSION

pH parameter is important in industry processes because of the corrosion or scale formation problems on the equipment. This issue is critical in the reuse and reclamation of effluent.

As shown in Table 4, the pH of SF/MBR, SF/MBR/GAC, and SF/GAC became more than influent (pH = 7.53) introduced to hybrid systems. This can be due to the activity of denitrifying microorganisms in MBR that it is observed in the literature.[28] Denitrification reactions follow as:

$$NO_3^- + 2H^+ + 2e^- \rightarrow NO_2^- + H_2$$

$$NO_2^- + 2H^+ + e^- \rightarrow NO + H_2O$$

$$2NO + 2H^+ + 2e^- \rightarrow N_2O + H_2O$$

$$N_2O + 2H^+ + 2e^- \rightarrow N_2 + H_2O$$

While Azis *et al*. reported an ion exchange process between wastewater and activated carbon can be promoted the pH value after activated carbon, which is in combination with SF. The initial pH was increased from 6.75 ± 0.01 to 7.22 ± 0.04 in study of Azis *et al*. This result is similar to pH of SF/GAC effluent (pH = 7.99 ± 0.21) but it is lower than the effluent pH of SF/MBR/GAC (pH = 8.33 ± 0.3) in the current study.[29]

### Removal of total suspended solids and turbidity

The high TSS and turbidity imply higher COD.[30] The oxygen content drops and it provides the anaerobic conditions in the

**Table 5: Annual fixed and variable costs per plant and scenario**

| Cost | Fixed cost ($) | Annual variable cost ($) | Annual total cost ($) |
|---|---|---|---|
| Sand filtration | 10,000 | 675 | 10,675 |
| MBR | 40,000 | 894 | 40,894 |
| Activated carbon | 9000 | 589 | 9589 |
| RO | 20,000 | 960 | 20,960 |
| SF + MBR | 60,000 | 973 | 60,973 |
| SF + MBR + GAC | 70,000 | 1000 | 71,000 |
| SF + GAC | 20,000 | 832 | 20,832 |
| SF + MBR + GAC + RO | 80,000 | 1300 | 81,300 |

SF: Sand filter, MBR: Membrane-biological reactor, GAC: Granular activated carbon, RO: Reverse osmosis

**Table 6: Proposed classification of product water[24]**

| Parameter (mg/L) | Class A* | Class B* | Class C* | Parameter (mg/L) | Class A | Class B | Class C |
|---|---|---|---|---|---|---|---|
| Iron | <0.3 | <1 | >1 | Sulfate | <250 | <500 | <500 |
| Manganese | <0.3 | <1 | >1 | Silica | <20 | <50 | <50 |
| pH | 6-9 | 6-9 | 6-9 | TSS | <50 | <100 | <100 |
| COD | <20 | <75 | <75 | TDS | <500 | <1000 | <1000 |
| Total hardness | <250 | <500 | <500 | Chloride | <200 | <500 | <500 |
| Alkalinity | <150 | <500 | <500 | | | | |

*Class A: This class includes very high-quality waters, which require in sensitivity processes of industry. Water does not need the treatment process or it needs minimum treatment, Class B: The class includes intermediate-quality waters, which require in low- sensitivity processes of industry, Class C: This class includes low- quality waters, which need to be treated for industrial applications. COD: Chemical oxygen demand, TSS: Total suspended solids, TDS: Total dissolved solids

**Table 7: A qualitative classification of product water from the studied scenarios**

| Parameter | Scenario 1 | Scenario 2 | Scenario 3 | Scenario 4 |
|---|---|---|---|---|
| Iron | Class A | Class A | Class A | Class A |
| Manganese | Class A | Class A | Class A | Class A |
| pH | Class A | Class A | Class A | Class A |
| COD | Class B | Class A | Class B | Class A |
| Total hardness | Class C | Class C | Class C | Class A |
| Alkalinity | Class C | Class C | Class C | Class A |
| Sulfate | Class B | Class B | Class B | Class A |
| Silica | Class B | Class B | Class B | Class A |
| TSS | Class A | Class A | Class A | Class A |
| TDS | Class C | Class C | Class C | Class A |
| Chloride | Class C | Class C | Class C | Class A |

COD: Chemical oxygen demand, TSS: Total suspended solids, TDS: Total dissolved solids

treatment processes. Furthermore, the consumption of reused water with high COD (due to high TSS) in industries cause to interfere in the bleaching process of paper and textile industry.[31]

In the present study, the maximum removal of TSS and turbidity occurred during SF/MBR/GAC/RO scenario. Although the high removal determined at the RO process of SF/MBR/GAC/RO scenario (>99%), there must be pretreatment processes to prevent fouling of RO membranes.[32] In fact, applied pretreatments are essential for post-treatment. Hence, the hybrid system of SF/MBR/GAC/RO provided the appropriate TSS value for high-sensitivity industries (TSS <2 mg/L).[9,33]

As showed in previous works,[34] the effect of the use of GAC pretreatment on RO solids control was evaluated across different media. They found that the removal of turbidity by GAC was dependent on adsorption rate, bulk flow, and adsorbent particle size. Hatt *et al*. reported at least 80 percent turbidity removal efficiency using GAC. The removal in study of Hatt *et al*. was observed agreement with result of this study [removal efficiency of turbidity was 78% in Figure 3b].[34] The characteristics such as the size of GAC, flow rate, EBCT in GAC can be identical in both study. The value of turbidity in MBR effluent [Figure 2b] was more than turbidity value in the study of Naghizadeh *et al*. who used hollow fiber microfiltration in a bioreactor and reported that turbidity and TSS removal were high.[35] However, the ability of MBR process in this study was low (RE = 32%). Afterward, the scenario needs RO process to meet the required standards for industrial.

MBR and GAC had the same performance in the removal of turbidity [Figure 3b]. Due to bio-flocculation and UF filters in MBR, particulates decrease in MBR.[36] As shown in Figure 3a, the percentages of TSS removal in GAC were more than MBR during sampling months. Hence, more TSS value remove in SF/GAC>SF/MBR/GAC>SF/MBR. This could due to the long solids retention time (20d) in MBR.[33] Thus, SF/MBR decreased TSS removal efficiency to 25% according to Table 4.

Further removal has occurred by the hybrid application of RO in the scenario of SF/MBR/GAC/RO, which is related to the advanced removal of inorganic (ionic matter) and organic matter.

## Removal of total coliforms

Many bacteria (e.g., coliforms) are in the water systems of industries. The total coliforms standard in 100 cc water of cooling towers is 93 MPN.[37]

As shown Figure 4, the number of bacteria reduced after MBR because of membrane rejection and bio-flocculation,[33] the MBR has played a major role in decreasing coliforms of SF/MBR, SF/MBR/GAC, and SF/MBR/GAC/RO. However, coliform density increased significantly because the biofilm growth and low contact time occurred during the activated carbon process of SF/GAC [Table 4].[32] Similarly, Purnell *et al*. (2015) reported MBR reduced fecal coliforms to 0.3 CFU/100 cc in the effluent.[38] The removal of bacteria in the study of Purnell *et al*. was more than our study that it can attributed to low influent MLSS level (MLSS = 7000 mg/L) and the more number of operation plants.[38] On the other hand, Baresel *et al*. showed MBR effluent was achieved removal efficiency more than 85% followed by GAC. However, this result indicates incompatibility with removal efficiency of 57.45% by SF/MBR/GAC.[39] All effluents of the studied scenarios can be consumed for the industrial application.

Although the density of coliforms is not considered as an indicator in industrial applications, it is necessary to provide the health standards.

## Removal of iron, manganese, silicate, and sulfate

In the treatment process, iron, silicate, and sulfate are very important in the reclamation of effluent for sensitive industries. The residues will cause to produce very hard and stable deposits in the equipment of industries. These deposits damage turbine nozzles and blades.[32] This gradually will cause pressure drops and affect the ability and productivity of the turbine.[32] Furthermore, the high concentrations of these parameters cause to change the color of leather in the tanning industry or paper industry.[40]

In Table 4 sulfate partly was removed during SF/MBR/GAC (RE = 15%), it may be through more biological reduction and oxidation processes by microorganism's growth such as sulfate-reducing bacteria (SRB) in SF/MBR/GAC than SF/MBR and SF/GAC. This result is acknowledged by study of Vallero *et al*.[41]

Gisi *et al*. reported sulfate removal to obtain 99.8% at 90 bar TMP and 74.7% recovery rate when the RO process combined with the activated sludge process as a pretreatment stage that confirm this study. Used transmembrane pressure (TMP) in the study of Gisi was more than the present study.[42] However, the recovery rate obtained 70% in this study at 15 bar TMP.

In stages of filtration, MBR, and GAC, the average removal efficiency decreased to 14.28%, 57%, and 28% [Figure 5b]. There is a significant difference in the removal efficiency using RO. This can indicate the ability of RO in removing iron of effluent. Several studies have found the efficient use of RO for iron removal.[30]

In the scenario of SF/MBR, the iron removal is determined by MBR more than SF plant, which can be exhibit higher biological oxidation because of the high SRT and the presence of iron-oxidizing bacteria.[18,43] Hence, SF/MBR showed further treatment after SF/MBR/GAC/RO. The GAC removes iron because of high surface and porous carbon as an enhancing bed of biofilm.[44]

Manganese removal in SF/MBR/GAC/RO was shown the highest among the studied scenarios because of the RO plant presence, which can be implied the growth of manganese oxidizing microorganisms. Furthermore, the presence of a very low concentration of manganese in RO feed causes to decrease in membrane fouling[45] thus it implies a good performance of secondary treatment processes in the Aq Qala industrial park. As cited before, SF/MBR, SF/GAC and SF/MBR/GAC scenarios were not changed manganese. Du *et al*. reported manganese further reduced by PAC-MBR (Mn <0.1 mg/L), however, this result is not agreement with Du *et al*. study.[18]

During the pH range 8–10, a stable ionic silica forms in that region. On the other hand, the effect of neutral and acidic pH results in decreasing silica removal.[46] Hence, silica removed slightly (RE = 20%) in this study because of pH value of 8.3 in the effluent of SF/MBR, SF/MBR/GAC, and SF/GAC. The silica removed by SF/MBR/GAC/RO (>96%) because of effluent natural pH, which has been explained as the appropriate treatment for meeting effluent standards of high-sensitivity industrial [Table 1]. However, other researchers such as Latour *et al*. found a low silica removal efficiency (RE = 10%), who was implemented softening processes using a polyaluminum coagulant.[47] As noted above, the processes (e.g., cartridge filter, UF, activated carbon…) before RO treatment are important.

## Removal of chemical oxygen demand

The presence of organic matter increases obstruction and corrosion in the heat exchangers and cooling systems. To monitor the industrial wastewater organic contamination level, we used chemical organic demand tests after each treatment stage. Furthermore, COD higher than standard limits in the sensitive industries cause to decrease the production quality.

Hence, RO effluent as the last plant in scenario of SF/MBR/GAC/RO revealed the lowest COD concentration in the effluent during sampling time [Figure 6 and Table 4]. However, SF showed the highest COD in the effluent during 70 days of sampling time [Figure 6]. This plant affected all scenarios. SF could be removed most suspended COD in hybrid scenarios. As illustrated in Figure 6, COD in GAC effluent was exhibited low concentrations in comparison with influent COD during sampling times of scenarios. Slight higher removal efficiency (75%) of GAC was reported by Zou (2015), who assessed the removal efficiency of color by an integrated system ozonation/activated carbon/biological filter and also determined removal efficiency by GAC had more removal than ozonation.[48] Furthermore, the integration of MBR as green technology with various wastewater treatment systems recommended in the research studies.[49] The various types of MBR may be produced the appropriate water for industrial applications. Kumari *et al*. showed complete removal of COD using MBR-PVDF (polyvinylidene fluoride) and MBR-ceramic and concluded these can be recycled water from a high organic load wastewater of dairy factory.[50] However, Figure 6 depicted COD concentration not removed completely using MBR, so that SF/MBR and SF/MBR/GAC scenarios demonstrated 51% and 62% removal [Table 4]. In this study, the type of MBR applied MBR-PES (Poly Ether Sulphone) that is affected at the residue COD.

To provide further treatment was used RO followed by GAC in the scenario of SF/MBR/GAC/RO. COD removed significantly by the application of RO. The results were corroborated by the literature review.[51,52] As shown in Table 4, COD value at RO effluent in SF/MBR/GAC/RO reached <2 mg/L that may be caused by the flow velocity, formed a secondary layer on the membrane, and charge of contaminants.[32,53] Similarly, it is seen in the research of Liu *et al*. (2011), who compared the RO treatment effect on the reusing textile effluent. Liu *et al*., reduced the COD concentration to <10 mg/L.[53] The lack of COD indicates the effluent of SF/MBR/GAC/RO have not any organic contaminations (e.g., emerging contaminants).

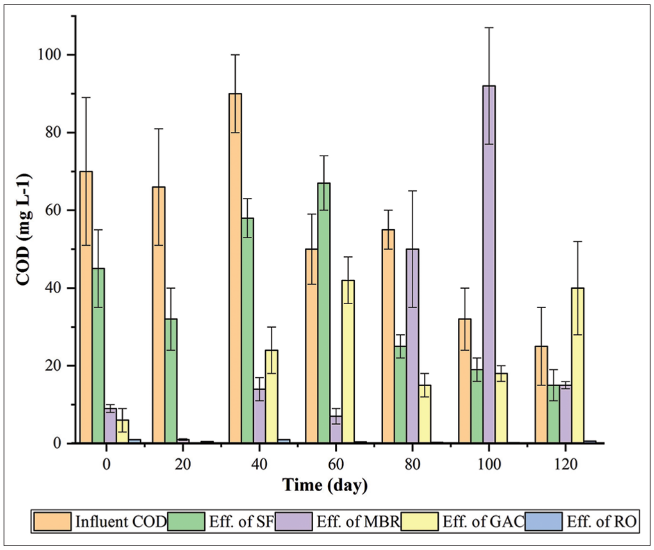

**Figure 6:** COD concentration in effluent of processes during operation time. COD: Chemical oxygen demand

## Removal of chloride, hardness, alkalinity, and total dissolved solids

Moreover, chloride, hardness, and alkalinity are the important factors, which can cause issues such as corrosion and deposit on the industrial equipment uses the reused water in the sections of industry. These parameters can interfere in the quality of final products. Therefore, the feed water for the industry must be treated by the processes.[31]

Table 8 presents removal of these parameters after the treatment processes. As shown in Table 4, hardness, alkalinity, and chloride values in scenario of SF/MBR/GAC/RO conform to the sensitive industry standards. This result could be due to the effect of RO process on the anions (e.g., chloride) and cations (e.g., calcium and magnesium) especially anions such as chloride. The produced water quality can be used for other industries such as intermediate-pressure boilers, cooling water, textile, and chemical industries. Amosa *et al*. (2016) reused only the palm oil industry effluent for low-pressure boilers.[9] The detected hardness, alkalinity, and TDS concentrations of the Amosa study (hybrid PAC-UF system) was more than the current study. Also, a study performed on ozonation and membrane processes, which was achieved lower removal of chloride and total hardness than the current study.[54] While, Yin *et al*. pointed out that the integrated system of SF/UF/RO could be reduced 82%, 87% and 92% for hardness, total alkalinity and chloride, however, this results is similar with SF/MBR/GAC/RO system.[55] As previously mentioned, none of SF/MBR, SF/MBR/GAC, SF/GAC scenarios were treated the chloride, alkalinity, hardness, and TDS.

**Table 8: The comparison of the average of total dissolved solids, hardness, alkalinity, and chloride in effluent with product water**

| Sample | TDS (mg/L) | Hardness (mg/L) | Alkalinity (mg/L) | Chloride (mg/L) |
|---|---|---|---|---|
| Influent | 1631±150 | 501.4±164 | 462.9±133.5 | 838.6±63.8 |
| Filtration effluent | 1576±126.3 | 568±100.9 | 553.3±91.5 | 843.6±235 |
| MBR effluent | 1656±139.6 | 532.9±89.2 | 512.9±99.6 | 989.3±279.3 |
| GAC effluent | 1616±129.4 | 527.1±154 | 502.9±123 | 1017.9±212.6 |
| RO effluent | 100±14.89 | 50.8±17 | 54.8±12.5 | 68.1±10.9 |
| Water | 100 | 100 | 75 | 100 |

TDS: Total dissolved solids, MBR: Membrane-biological reactor, GAC: Granular activated carbon, RO: Reverse osmosis

## Cost estimation of hybrid treatment processes

The cost was obtained ~1$ and <1$ per $m^3$ treated wastewater in SF/MBR/GAC/RO and SF/GAC and also other scenarios were obtained 0.5$. However, a study reported desalination of seawater (as an advanced technology) was provided the cost of more than 3$.[56] The costs of product water scenario SF/MBR/GAC/RO, which the best scenario in this study, are lower than desalination costs. The reuse of water can benefit from developing countries due to environmental issues and economical values, which are going to enhance in the future.[13] The proposed scenario (SF/MBR/GAC/RO) can increase the quality of the products of industry and can decrease the operation problems (e.g., deposits, corrosion, and obstruction of equipment), energy, and water consumption. As cited above, annual total cost in SF/MBR/GAC/RO was slightly higher than other studied scenarios. Afterwards, total cost of SF/MBR/GAC, SF/MBR and SF/GAC was estimated 10000$, 20000$ and 60000$ lower than SF/MBR/GAC/RO. The operation and maintenance costs of membrane filters such as MBR and RO are high and they include the annual variable cost. Annual variable cost of SF/MBR/GAC/RO shown 300$ more than SF/MBR/GAC. The membrane fouling at RO is more difficult than MBR and its cleaning stage is more expensive. Although RO is provided the highest cost so that the literature confirms the result[57] but the annual total cost of RO and MBR per plant was similar in the current study. Annual total cost of GAC plant in SF/GAC was affected total cost of scenario. The cost of electrical and chemical consumption of GAC was low in this study, however, Q. Adams and M. Clark indicated the total cost of GAC systems depends on the system dimensions.[58] The total cost of SF, MBR, AC, and RO was calculated based on the number of 2, 2, 2, and 1 plant, respectively.

## CONCLUSIONS

In this research, the efficiency of hybrid advanced technologies (SF/MBR, SF/MBR/GAC, SF/GAC and SF/MBR/GAC/RO) was investigated in reducing physicochemical and biological parameters to standards for reusing effluent.

According to the obtained results, the scenario SF/MBR/GAC/RO had the highest quality efficiency and cost-effective at reusing secondary treatment effluent for sensitive industries. Conversely, the product water of SF/MBR, SF/MBR/GAC, and SF/GAC cannot apply in the sensitive industries due to the high TDS, alkalinity, hardness, TSS, COD, chloride, sulfate, and silica values has been explained already.

Moreover, it suggests the manipulation of hybrid treatment processes for emerging pollutants. On the other hand, this study observed some limitations that should be considered future applications, such as providing feed water for the high-sensitivity industries.

## Ethics code

The authors approved this study in Ethical Number: ir.goums.rec. 1396.44.

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## Conflicts of interest

There are no conflicts of interest.

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