



OPEN An inclusive solution based on grey water footprint for performance evaluation of industrial wastewater treatment systems

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This study developed a new framework based on grey water footprint (GWF) to inclusively evaluate and compare the industrial wastewater treatment plants (WWTPs). The conventional approach typically reports the efficiency of treatment systems on abating the concentration of each pollutant, individually. As an alternative, GWF can simultaneously include multiple pollutants, pollution loads, and regional water quality standards in calculations. These advantages are critical for assessing an industrial WWTP treating complex wastewater, with hazardous pollutants and variable inflow. Moreover, we introduced four innovative criteria based on GWF and improved it as a multi-functional index. To verify the applicability of the proposed method, we chose two operating units in parallel, the activated sludge (AS) and membrane bioreactor (MBR), as case studies treating real industrial wastewater. Samples were obtained from both treated and untreated wastewater. 36 pollutants were examined and used for GWF accounting in different scenarios. These scenarios were based on different maximum allowable concentrations (C_{\max}). Multi-pollutant GWF reduction (%) was the first index evaluating the overall removal efficiency. The AS with 93.1% average GWF removal could outperform MBR with 87.1% removal. Operational reliability was the second index, showed that AS could reduce GWF variations from inlet to the outlet with 83.7% efficiency, while it was 77.5% for MBR. The third index was GWF per carbon footprint (GWCF). It quantified the equivalent abated stress from water bodies per increased pressure exerted by emitting greenhouse gas (GHG) during wastewater treatment. The GWCF of AS was $347.8 \text{ m}^3/\text{kg-CO}_2$ indicating a superior efficiency over MBR with $84.9 \text{ m}^3/\text{kg-CO}_2$. It means that MBR relatively emitted more GHGs for reducing less GWF. Heavy metal pollution reduction (HPI) was the fourth index quantified based on GWF. It evaluated the particular performance of treatment systems for abating hazardous pollutants. The AS with an average HPI of 56.7% outperformed MBR with 50.4% efficiency. Therefore, this study showed that GWF is a versatile and applicable index and can provide a more holistic framework for evaluating and comparing wastewater treatment units.

Keywords Activated sludge, Carbon footprint, Membrane bioreactor, Wastewater treatment, Water footprint

Industrial wastewater poses significant risks to human health and ecosystems¹. This wastewater contains various pollutants, e.g. chemical oxidation demand (COD), biochemical oxidation demand (BOD), and heavy metals^{2,3}. This complexity calls for well-performed wastewater treatment plants (WWTPs) with efficient units for pollution removal. For example, activated sludge (AS) and membrane bioreactor (MBR) are two commonly used wastewater treatment units. Recent studies reported the performance of the AS for COD and BOD removal were 82–90% and 88–98%, respectively^{4,5}. These efficiencies for MBR were 60–97% and 86–89%, respectively^{6–9}. However, these conventional metrics that account for certain and individual pollutant removal efficiency are insufficient for comprehensively evaluating or comparing treatment units. It is firstly due to the complexity of industrial wastewater that has multiple pollutants¹⁰. Moreover, the conventional approach ignores pollution loads and the regional constraints of the receiving environment for different pollution loads¹¹. As an alternative, grey water footprint (GWF) is an index with the advantage of including multiple pollutants, pollution load, and regional water quality standards in evaluations. It basically presents a different view of the performance evaluation of WWTPs and has the potential to develop a more holistic quantitative method for this purpose.

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The GWF is one key component of the water footprint (WF) that measures the equivalent pressure on water bodies due to pollution emissions¹². The GWF is the embedded volume of freshwater required to assimilate the discharged pollution loads regarding the regional capacities of the receiving environment¹³. It means that GWF evaluates water pollution loads of emission sources, e.g. industrial wastewater, and concurrently considers whether the receiving water body can handle the discharged pollution loads based on quality standards¹¹. Recent studies have revealed that WWTPs can significantly reduce the GWF. For example, twelve municipal WWTPs, using AS and MBR, could reduce the GWF of pharmaceuticals and conventional pollutants by about 26% and 90%, respectively¹⁴. In a large-scale study, the total annual industrial GWF of petroleum pollutants in China was reduced by 12–19%, underscoring the significant positive impacts of WWTP¹⁵. Connecting enterprises in an industrial park to a centralized WWTP could reduce the annual GWF up to 90%¹⁶. Municipal WWTPs could achieve an average 91% GWF reduction in 15 years, primarily based on ammonium (NH_4) and total phosphorus as key pollutants. Upgrading these systems with MBR could significantly reduce the GWF based on NH_4 and total suspended solids (TSS) up to 12.9 and 6.2 million cubic meters per month (MCM/month), respectively^{17,18}. More recently, a WWTP could reduce the GWF by about 4–7 MCM/month for BOD¹⁹. This literature shows that GWF application is increasing for reporting the performance of WWTPs. However, it obviously lacks addressing other required objectives for evaluating treatment systems. The sustainable performance of a wastewater treatment unit is not only reliant on pollution removal efficiency. It also depends on energy consumption, reliable operation, and effluent risk mitigation related to the toxic materials. These objectives have not been assessed yet in previous literature in the context of GWF.

The GWF of WWTPs indicates how much stress would be reduced from water bodies. However, it is not an only index measuring the equivalent stress on the environment. WWTPs consume energy and indirectly emit greenhouse gas (GHG), e.g. carbon dioxide (CO_2), during purification²⁰. Carbon footprint (CF) is an environmental index that reflects the equivalent GHG emissions from production and energy consumption²¹. The heavy metal pollution index (HPI) also determines the hazardous level of remaining pollutants in wastewater. The HPI considers multiple heavy metals and measures their combined adverse impacts on water quality²². A recent report showed that the HPI of a river subjected to industrial contamination ranged from 33 to 150 due to heavy metals, e.g. nickel (Ni), chromium (Cr), lead (Pb), cobalt (Co), cadmium (Cd), copper (Cu), and iron (Fe)²³. Moreover, literature demonstrated that the CF of AS was 68.9 ton- CO_2 /month, which can be reduced to 37.4 ton- CO_2 /month by using renewable energy sources²⁴. Likewise, MBR had the CF of 2.8–11.9 ton- CO_2 /month²⁵. Although CF and HPI are potential indices for performance evaluation of WWTPs, none of the above literature has evaluated the energy consumption, CF, or heavy metal toxicity in integration with or in the context of GWF. The questions remain are whether these indices can combine with GWF or used in association with GWF for comprehensive evaluation? Can we develop these indices based on GWF for performance evaluation of WWTPs?

Answering the questions above, we have developed a new framework based on multi-pollutant GWF to inclusively evaluate and compare the performances of industrial wastewater treatment systems. By this framework, we introduce GWF as a multi-functional index, applicable for performance evaluation in combination with CF and HPI. Here, four innovative GWF-based criteria were defined to include the overall pollution removal efficiency, operational reliability, CF, and HPI in this framework. In order to verify the proposed method, the study delved deep into the details of industrial GWF accounting. At first, we chose two fully operational treatment systems (AS and MBR) in parallel treating the complex real industrial wastewater. Sampling was conducted monthly from both treated and raw wastewater. 36 pollutants were examined for multi-pollutant GWF assessment. Four types of water quality standards and constraints were considered to account the GWF results in different scenarios. Finally, the results were fully discussed with other studies. It is noteworthy that the numbers were originally derived from the studied area. However, it obviously does not mean that the research objectives and main idea are case specific. On the contrary, these results verify the applicability of the proposed method worldwide with its new holistic approach toward the performance evaluation of WWTPs using GWF as the multi-functional index.

Materials and methods

Case study

In order to verify the applicability of the developed method based on GWF, we have chosen two secondary treatment modules (AS and MBR) as the study area. These two biological units are operating in parallel at Jey Industrial Park WWTP. They receive the same raw complex wastewater from various industrial sectors, including metal plating, textile dyeing, food production, etc. Their pretreatment units are screening, grit chamber, and an equalization tank. The Jey Industrial WWTP covers approximately 27,800 m² and is located near the Industrial Park in Isfahan province, central Iran. Its coordinates are 32° 40' 2.74" N and 51° 50' 52.68" E. The average temperature and annual precipitation (1951–2017) are 16.3 ± 0.3 °C and 125 ± 21.2 mm, respectively²⁶.

There are some backgrounds and reasons for choosing the study area. First, Isfahan is an industrial province at the centre of Iran with more than 20 industrial parks. Here, Jey industrial WWTP is the largest wastewater treatment facility with a total daily capacity of 4000 m³. Second, studied area is a centre with complex industrial enterprises and consequently has complex industrial wastewater. Third, Jey industrial WWTP has two different biological treatment modules (AS and MBR) fully operational since 2021 in parallel feeding from the same pre-treated wastewater. This is an exclusive opportunity to our cause to control the applicability of the developed method for comparing the results (see Fig. 1). The treated wastewater of the study area is currently recycled for industrial use and irrigating unfruitful plantations. For more information, a block flow diagram (BFD) is depicted in Fig. 1B, and a process flow diagram (PFD) for WWTP is presented in Fig. S1 (as Supplementary file).

According to Table S1, the AS and MBR modules received about 73% and 27% of the total inflow of WWTP during the study period (October 2022–March 2023), respectively. On average, the AS treated 1079 ± 61 m³/day,

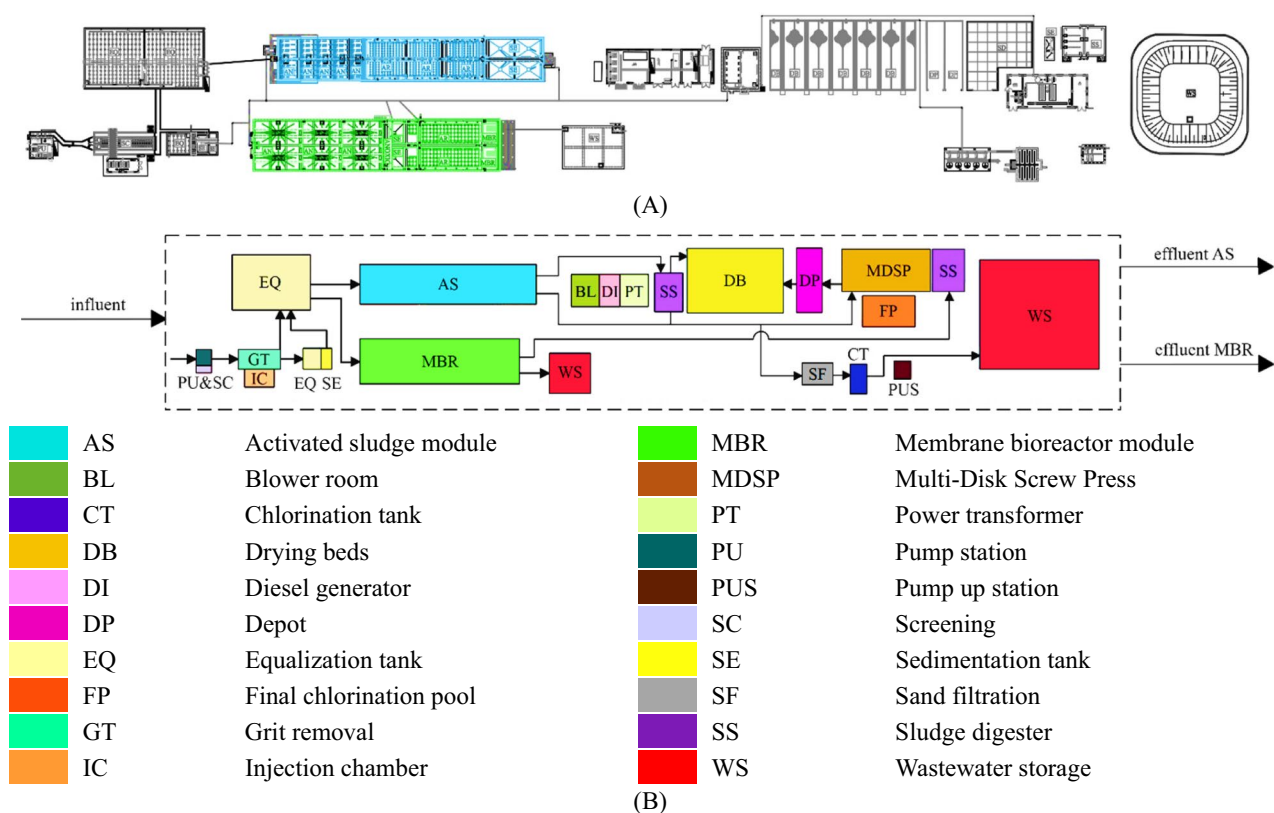


Fig. 1. **A** Studied industrial WWTP: AS (blue), MBR (green), **B** its flow diagram showing other units.

while it was $354 \pm 20 \text{ m}^3/\text{day}$ for MBR. Although the capacities of the two modules were different, this variation does not have an adverse influence on performance assessment. On the contrary, it was aligned with our cause. Since we were looking for a study area without limited characteristics, comparing two modules receiving different flow rate, but with the same concentrations, can better reflect the applicability of the developed method in GWF context. We should remind that accounting GWF is based on pollution loads, not pollutant concentration only. Accordingly, these boundaries are clearly defined to separately analyze both systems under their specific operational frameworks, ensuring robust and comparable insights into their performance.

Sampling and test

Over six months, from October 2022 to March 2023, samples were taken weekly and analyzed from both the inlet and outlet of the two modules. During this time, the study was carried out simultaneously for both modules, enabling a consistent evaluation of their efficiencies. A total of 72 samples were collected in triplicate to analyze 36 water quality parameters. The triplicate sampling ensured reliable experimental results, which were further validated for accuracy through cross-verification with the on-site data of the Jey industrial park laboratory during this period. The selected parameters (36 pollutants) also ensured a thorough evaluation of the GWF linked to the studied industrial wastewater. They reflect the wide range of pollutants found in industrial parks due to diverse sector activities. Other possible pollutants, e.g. cyanide, poly aromatic hydrocarbons (PAHs), polychlorinated biphenyl (PCBs), were not detected in samples as were insignificant regarding previous data of Jey laboratory. It is noteworthy that examining these 36 pollutants with 72 samples is fairly a rare study with the highest variety of pollutants for accurate multi-pollutant GWF assessment¹².

Samples were stored in sterile polyethylene terephthalate (PET) vessels (1.5 L capacity) and kept below 4 °C during transport to a certified laboratory 30 km away. Analytical procedures were followed regarding american public health association standard methods for water and wastewater examination²⁷. For tracing concentrations of pollutants, such as heavy metals, ICP-MS was employed according to american society for testing and materials (ASTM) standard methods²⁸, ensuring accuracy in the parts per billion (ppb) range. Detailed descriptions of the analyzed parameters can be found in Table S2 (see Supplementary Material).

Energy consumption and CF calculation method

Table S3 summarizes the average energy consumption of each treatment module in the examined WWTP, showing an average energy input of $112.3 \pm 5.3 \text{ kWh}$. The AS module used an average of 51.7 kWh, while the MBR module required 60.6 kWh. For CF accounting, it's crucial to identify electricity sources and quantify their GHG emissions. In Iran's industrial sector, electricity generation sources include natural gas, oil, coal, nuclear power, and various renewable energies like hydropower, wind, solar, biomass, geothermal, and tidal²⁹. CO₂ emissions

from electricity consumption depend on the average GHG emissions of each source in kg-CO₂ per megawatt-hour: natural gas (499 kg-CO₂/MWh), oil (733 kg-CO₂/MWh), coal (888 kg-CO₂/MWh), nuclear (66 kg-CO₂/MWh), and renewables (25 kg-CO₂/MWh)³⁰. From 2010 to 2023, natural gas was the main electricity source at 75%, followed by oil and coal at 18%, and low-carbon resources (nuclear and renewable energy sources) at 7%³¹. The average electricity generation was about 296.5 TWh, with 226.5 TWh from gas, 49.5 TWh from oil and coal, and 20.5 TWh from low-carbon resources as illustrated in Fig. S2²⁹. In 2023, low-carbon resources contributed only 6.2% of Iran's total electricity generation³¹.

The rate of CO₂ emitted based on electricity consumption is calculated as the carbon footprint (CF) of electricity, which is the mass of CO₂ emissions per unit of time (ton/yr), as defined by Eq. 1^{21,32}:

$$CF = EN \times CEI \quad (1)$$

Where:

EN = The total energy input by the purification modules of the WWTP (energy/time).

CEI = The CO₂ emission intensity (CEI) caused by electricity consumption (mass-CO₂/energy).

GWF calculation

The GWF of multiple pollutants for both modules can be calculated by Eq. 2¹²:

$$GWF_i = \left(\frac{[Q_{eff} \times C_{eff}] - [Q_{abstr} \times C_{act}]}{[C_{max} - C_{nat}]} \right)_i \quad (2)$$

Where:

Q_{eff} is the volumetric flow of the inlet or outlet (volume/time).

Q_{abstr} is the abstracted water volume (volume/time).

C_{eff} is the pollutant concentrations in an inlet or outlet (mass/volume).

C_{act} is the pollutant's actual concentration in consumed water (mass/volume).

C_{nat} is the pollutant natural concentration in the receiving water body (mass/volume).

C_{max} is the pollutant's maximum allowed concentration in the receiving water body (mass/volume).

Since in the study area, the treated wastewater does not return to the original water body from which it was drawn, the calculation excludes the terms of Q_{abstr} and C_{act}. Therefore, Eq. 3 has been used for GWF instead¹².

$$GWF_i = \left(\frac{Q_{eff} \times C_{eff}}{C_{max} - C_{nat}} \right)_i \quad (3)$$

The GWF for multiple pollutants is determined by identifying the maximum value among the individual GWF_i for each pollutant, as outlined in Eq. 4¹². The dominant GWF is dictated by the pollutant requiring the largest volume of embedded water to reduce concentrations below allowed limits¹¹. When determining the GWF over a time interval, calculate the average GWF for each contaminant, then identify the maximum among these averages. If the GWF_i is less than zero (<0), it is considered null (GWF_i=0)¹².

$$GWF = \max \{GWF_i\}. \quad (4)$$

In this study, the GWF index is analyzed under various scenarios characterized by the strictness of the maximum permissible concentrations of pollutants (C_{max}). These scenarios, detailed in Table S4, cover 36 pollutants. Four scenarios for C_{max} range from strict (S1) to lenient (S4), tailored to the end-use of treated wastewater, including discharge into aquatic systems, freshwater, recreational water, and reclaimed water for irrigation. The C_{nat}, or natural concentration of pollutants, is based on historical data and regional analyses from a period with no significant human influence¹¹.

Combined grey water-carbon footprint

Assessment of GWF reduction per energy consumption (GWEN) necessitates calculating the net grey water footprint (GWF_n) based on the inlet and outlet of the treatment plant using Eq. 5. After that, Eq. 6 can assess the energy efficacy needed for reducing the GWF³³.

$$GWF_n = GWF_{inlet} - GWF_{outlet}. \quad (5)$$

$$GWEN = \frac{GWF_n}{EN}. \quad (6)$$

It calculates the GWF_n as the difference between the inlet and outlet GWFs, per consumed energy (EN). It measures how much GWF would be decreased by one unit of consumed energy for each treatment module.

This study proposed the composite index of GWF and CF (GWCF) instead of GWEN (Eq. 7). It calculates the ratio of GWF_n per CF to assess treatment performance. GWCF evaluates GWF reduction with respect to the emitted CF. This index considers the potential increases of other environmental pressures (GHGs). It allows for analyzing treatment efficiency in reducing GWF based on CF generation (m³/kg-CO₂).

$$GWCF = \frac{GWF_n}{CF}. \quad (7)$$

This innovative index quantifies the net reduced pressure from aquatic system (GW_n) and links it with the corresponding CO_2 emissions (kg-CO_2) through wastewater treatment processes (Fig. S3). $GWCF$, for the first time, can integrate the GW and CF for WWTPs, as two distinct concerns related to water and air environment, respectively. By linking water quality (GW) and energy consumption (CF), $GWCF$ introduces a robust framework for sustainable water management practices. Recent literature calculated equivalent water required (WF) for generating electricity demand³⁴ or GW reduction per unit of energy input during wastewater treatment³³. The $GWCF$ accounts for this relationship, indicating which category, GW_n or CF , is comparatively more critical for WWTP operations. If $GWCF > 1$, GW_n is dominant, signifying the priority of major water pollution removal than GHG reduction. If $GWCF < 0.1$, more energy usage is not justifiable, highlighting the need for energy-efficient equipment or low-energy treatment units. These thresholds are the primary recommendations of this study, and can be further updated in future studies. Yet, these assumptions would not impose biases for the developed method.

Heavy metal pollution index reduction

HPI evaluates the status of heavy metal pollution in water and wastewater²². It assesses multiple heavy metals and their combined impacts on water quality. HPI calculation involves assigning weights to each pollutant based on regulatory guidelines, reflecting each metal's contribution to overall pollution and potential damage to health and the environment. An $HPI < 100$ indicates a non-contaminated outlet suitable for human use and irrigation³⁵, while an $HPI > 100$ suggests significant contamination and hazardous levels. It should be noted that these thresholds, weights and even pollutants can be updated in future studies. By any updates, HPI can still be applicable and updated in GW context as the following equations.

$$HPI = \frac{\sum_{i=1}^n \left(\frac{1}{S_i} \times \left[\frac{C_{eff} - C_{id}}{S_i - C_{id}} \right] \right)_i}{\sum_{i=1}^n \left(\frac{1}{S_i} \right)_i} \quad (8)$$

In Eq. 8 we can assume that the ideal concentration (C_{id}) of heavy metals in water bodies matches their C_{nat} in the receiving environment, effectively considered zero. Moreover, the standard value (S_i) for each parameter is equal to their C_{max} . Based on these assumptions, Eq. 8 can be reformulated as Eq. 9. This new equation establishes a relationship between the HPI and GW . The updated equation offers a novel approach, integrating HPI with GW to provide a comprehensive method for evaluating the environmental impacts of heavy metal pollution.

$$HPI = \frac{\sum_{i=1}^n \left(\frac{1}{C_{max}} \times \left[\frac{GW}{Q} \right] \right)_i}{\sum_{i=1}^n \left(\frac{1}{C_{max}} \right)_i} \quad (9)$$

Finally, HPI reduction efficiency is determined as Eq. 10 based on inlet and outlet concentrations. It can evaluate the treatment system's effectiveness in abating heavy metals and related risks from industrial wastewater.

$$HPI \text{ reduction} = 100 \times \left(\frac{HPI_{inlet} - HPI_{outlet}}{HPI_{inlet}} \right) \quad (10)$$

Results

Basic results and estimations

Prior to GW assessment, pollution removal efficiencies of the AS and MBR modules were evaluated using the conventional approach to benchmark their treatment capabilities. Inlet flow rates for both modules were meticulously recorded to account for GW . Additionally, the energy intensity of AS and MBR systems was calculated, reflecting energy consumption (kWh) per unit volume (m^3) of treated wastewater.

Pollution removal

Table 1 summarizes the monthly average removal (%) of pollutants concentrations with their standard errors (SE) and significance levels (p-values) from the inlet to the outlet of the AS and MBR. The highest reduction was attributed to TSS with 97.6% for AS and 99.3% for MBR. Conversely, the lowest reduction was attributed to nitrite (NO_2) and nitrate (NO_3) (< 1%) due to nitrification, indicating the need for tertiary treatment units. The non-parametric Mann-Whitney test was used to evaluate the significance level with a 95% confidence interval. It assessed whether pollutant concentrations in the module outlets were significantly decreased compared to their inlet concentrations. For example, the p-values of TKN and BOD in the AS and MBR modules were < 0.05, indicating a significant reduction of these pollutant concentrations for each module. However, the p-values of most pollutants, especially heavy metals, were > 0.05. The statistical distribution of all pollutant concentrations are depicted in Fig. S4 (supplementary file). This figure can illustrate the background statistical analysis leading to Table 1.

The results (Table 1 and Figure S4) verify that the conventional evaluation method has limitations for comparing the performances of treatment units for multiple pollutant assessments. For example, MBR has comparatively higher tin (Sn) removal (59.1%) than the AS (31.6%), while MBR has less cadmium (Cd) removal (70.2%) than AS (91.3%). In addition, P-values of these pollutants does not approve that the concentration removals are significant from the inlet to outlet. It underscores lacking an inclusive evaluation system that determines the superiority of any units.

However, the experimental results imply that both systems were effective on improving water quality. The AS and MBR systems could reduce pollutants concentrations from influent to their effluents. For example

Pollutants	Removal \pm SE (%)		p-value		Pollutants	Removal \pm SE (%)		p-value	
	AS	MBR	AS	MBR		AS	MBR	AS	MBR
TKN	96.2 \pm 2.70	99.1 \pm 0.30	0.005	0.005	Cr	46.1 \pm 18.7	36.4 \pm 8.30	0.230	0.378
BOD	97.2 \pm 1.20	98.0 \pm 0.80	0.005	0.005	Cs	68.4 \pm 5.10	62.2 \pm 10.0	0.471	0.471
TSS	97.6 \pm 0.90	99.3 \pm 0.20	0.005	0.005	Cu	33.8 \pm 13.3	57.9 \pm 7.10	0.575	0.230
P	57.1 \pm 15.2	72.5 \pm 4.30	0.128	0.093	Fe	61.0 \pm 12.2	63.7 \pm 12.2	0.173	0.066
PO ₄	86.0 \pm 2.20	86.6 \pm 1.90	0.005	0.005	Ga	44.9 \pm 15.6	33.4 \pm 10.4	0.230	0.575
COD	94.9 \pm 0.60	96.3 \pm 0.60	0.005	0.005	Hg	66.2 \pm 9.90	69.2 \pm 12.6	0.471	0.471
FOG	92.4 \pm 1.60	90.6 \pm 3.70	0.005	0.005	Li	36.9 \pm 12.9	31.2 \pm 9.20	0.337	0.423
Cl	18.4 \pm 8.10	19.0 \pm 8.10	0.378	0.378	Mn	56.2 \pm 12.7	78.2 \pm 10.1	0.230	0.045
NO ₂	0.00	0.00	–	–	Mo	49.3 \pm 12.7	42.0 \pm 10.5	0.378	0.471
NO ₃	0.00	0.00	–	–	Ni	68.0 \pm 13.6	69.1 \pm 7.50	0.230	0.230
ABS	84.4 \pm 2.40	86.9 \pm 2.50	0.081	0.081	Pb	74.6 \pm 15.6	76.1 \pm 9.70	0.230	0.230
Ag	24.3 \pm 10.2	51.3 \pm 7.20	0.471	0.230	Sb	66.0 \pm 9.30	69.4 \pm 3.30	0.936	0.936
Al	84.0 \pm 5.90	87.7 \pm 4.20	0.060	0.037	Se	59.0 \pm 9.10	69.2 \pm 10.4	0.378	0.378
As	42.3 \pm 15.3	41.4 \pm 4.60	0.471	0.378	Si	48.0 \pm 16.2	41.6 \pm 15.4	0.093	0.128
Ba	53.4 \pm 13.4	66.2 \pm 5.30	0.631	0.337	Sn	31.6 \pm 14.1	59.1 \pm 11.3	0.471	0.298
Be	28.4 \pm 7.10	22.3 \pm 13.3	0.471	0.575	Sr	50.8 \pm 13.5	56.4 \pm 12.7	0.173	0.173
Cd	91.3 \pm 3.20	70.2 \pm 12.7	0.230	0.230	V	68.8 \pm 4.90	67.1 \pm 10.7	0.471	0.471
Co	60.8 \pm 11.0	59.3 \pm 11.6	0.230	0.230	Zn	58.7 \pm 5.20	68.2 \pm 11.9	0.230	0.230

Table 1. Pollutant concentrations removal (%) with the standard errors (SE) of the AS and MBR modules, in addition to the statistical significance levels (P-value) of pollutants concentration from the Inlet to outlet in each module.

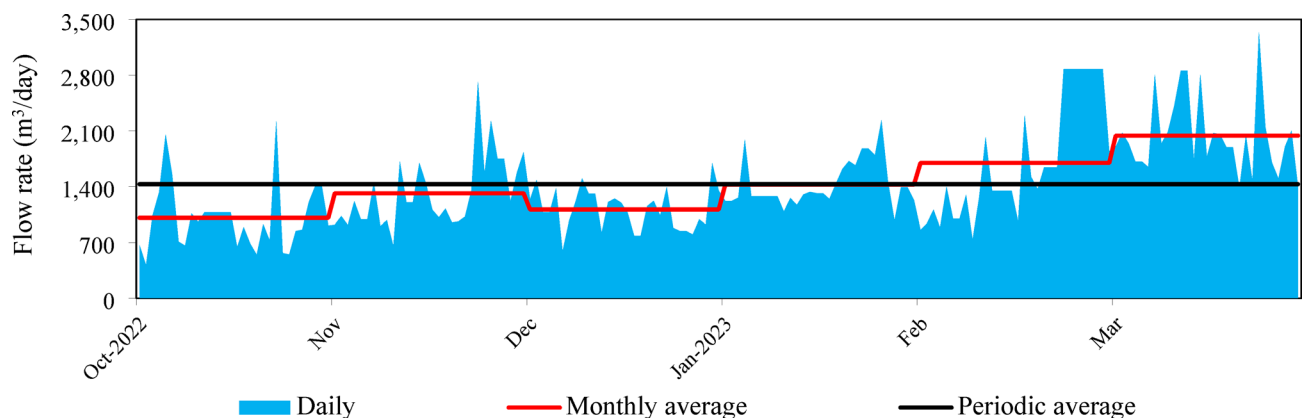


Fig. 2. Daily, monthly, and periodic average flow rate (m³/day) in the inlet of the WWTP.

TKN dropped from 193.7 ± 7.7 mg/L in inlet to 8.09 ± 6.0 mg/L (AS) and 1.8 ± 0.7 mg/L (MBR) in outlets. Likewise, BOD reduced from 1054.7 ± 300.2 mg/L to 21.3 ± 7.1 mg/L (AS) and 13.5 ± 2.2 mg/L (MBR). COD decreased from 1658.5 ± 258.7 to 80.0 ± 8.4 (AS) and 54.8 ± 5.2 mg/L (MBR). PO₄ removed from 133.3 ± 14.3 to 18.1 ± 2.6 mg/L (AS) and 17.2 ± 2.2 mg/L (MBR). The concentrations of heavy metals were also declined. For instance, Aluminum decreased from 456.4 ± 154.5 μ g/L to 57.1 ± 22.1 μ g/L (AS) and 52.1 ± 27 (MBR). Manganese abated from 474.2 ± 116.4 μ g/L in inlet to 232.3 ± 75.0 μ g/L and 140.1 ± 71.6 μ g/L in the outlets of AS and MBR, respectively. More details about pollutants concentration were illustrated in Fig. S4 (supplementary file).

Flow-rates

Figure 2 shows the measured daily flow rates of the raw wastewater entering the whole WWTP from October 2022 to March 2023. These flow rates were measured using flow meters installed separately for each module, with exact daily recordings maintained. The flow increased sharply in March (2040 ± 89 m³/day) when the maximum daily volume of wastewater was 3337 m³ (recorded on the 23rd of March). The monthly average flow rates were recorded, ranging from 1008 m³/day in October to 2040 m³/day in March. The average flow rate of overall raw wastewater during the studying period was about 1430 ± 43 m³/day. The wastewater flow rate is an essential factor in calculating the GWF and serves as a crucial element in thorough evaluations of wastewater treatment performance.

Carbon emission intensity (CEI) and footprint (CF)

The average electricity intensity for each module was also determined. Average energy consumption was 51.7 kWh for AS and 60.6 kWh for MBR, with daily flow rates of 1079 ± 61 m³/day for AS and 354 ± 20 m³/day for MBR. The AS module had an average electricity intensity of 1.15 kWh/m³ of treated wastewater, significantly lower than the MBR's 4.11 kWh/m³, within previously discussed ranges^{36,37}. From 2010 to 2023, an evaluation of the CEI in Iran revealed a steady reduction in CO₂ emissions per unit of electricity produced, declining from 529.3 kg-CO₂/MWh to 486.4 kg-CO₂/MWh. The average CEI over this period was calculated at 508.6 ± 8.1 kg-CO₂/MWh (Fig. S5). In comparison, the mean CEIs for China, the USA, Germany, and South Africa were 1170, 720, 680, and 990 kg-CO₂/MWh, respectively³⁸. Energy consumption for the AS and MBR modules resulted in CF estimates of 18.9 and 22.2 ton-CO₂/month, respectively.

GWF assessment

While pollutant removal rates (%) are valuable, they are insufficient to fully assess the performance of WWTP. Thus, accounting GWF is introduced instead for more inclusive evaluation of WWTPs, considering multiple pollutants, pollution loads, and receiving environmental standards. Based on the experimental results (Sect. **Pollution removal** and **Flow-rates**), the monthly average GWFs of multiple pollutants were quantified in four scenarios (S1–S4), as illustrated in Fig. 3. In S1 (Fig. 3A), due to the strictest C_{\max} , the GWFs of raw and treated wastewater in the AS and MBR modules were relatively higher than S2–S4. Relaxing the standard limits by increasing C_{\max} (S2–S4) results in decreasing GWFs. Since stricter standards (e.g. S1 and S2) are required for water bodies with high pollution vulnerability, reflecting this regional limit on pollution assimilation in calculations is an advantage for GWF. By considering C_{\max} and C_{nat} , GWF can include this potential in the performance evaluation of WWTPs. Higher pollution assimilation potential (larger $C_{\max} - C_{\text{nat}}$) can reduce pressure on treatment systems, decreasing the GWF index. For example, the monthly average GWF of nitrate (NO₃) ranged between 0.07 (S4) and 1.2 (S1) MCM/month for the AS outlet, and 0.04 (S4) to 0.7 (S1) MCM/month for the MBR outlet.

In certain scenarios, the C_{nat} of specific pollutants has surpassed the C_{\max} levels. For instance in S1, some pollutants (e.g. TSS and COD) have C_{\max} less than C_{nat} (GWF < 0), and their GWF registered zero for the inlets and outlets of both modules. It indicates that these parameters could also be critical pollutants in other scenarios and require targeted control measures¹⁶. The ability to modify the GWF based on the C_{\max} , more or less than C_{nat} , can be seen as a challenging aspect of GWF. Therefore, it is crucial to accurately determine the C_{\max} for pollutants, taking into account the specific discharge locations.

The total GWF of a treatment system was determined as the maximum of calculated GWFs for multiple pollutants (Eq. 4), providing a single index for more inclusive performance assessments of industrial WWTPs. Figure 4 shows the multi-pollutant GWF with different C_{\max} . TKN (S1), BOD (S2), TSS (S3), and TKN (S4) were the leading pollutants for the GWF in the AS and MBR inlets, having the maximum monthly average GWF in each scenario. However, the leading pollutants in the outlets were different from those in the inlets. This unique specification of GWF enabled independent multi-pollutant assessments of treated wastewater (outlet) from its background (inlet) in WWTPs. The total calculated GWFs in four scenarios for the two modules were as follows: In S1, the GWF of the AS inlet was 14.436 ± 2.786 MCM/month (by TKN), reducing to 1.222 ± 0.588 MCM/month (by NO₃) in the outlet, indicating 91.5% performance. For MBR, the inlet GWF was 4.689 ± 0.489 MCM/month (by TKN), decreasing to 0.699 ± 0.172 MCM/month (by NO₃) in the outlet with 85.1% reduction efficiency. In S2, the AS module reduced the GWF from 7.041 ± 3.284 MCM/month (by BOD) in the inlet to 0.451 ± 0.217 MCM/month (by NO₃) in the outlet, showing 93.6% efficiency. The MBR module reduced the GWF from 1.871 ± 0.374 MCM/month (by BOD) in the inlet to 0.258 ± 0.063 MCM/month (by NO₃) in the outlet with 86.2% efficiency. In S3, the GWF of the AS inlet was reduced from 5.679 ± 1.667 MCM/month (by TSS) to 0.387 ± 0.186 MCM/month (by NO₃) in the outlet, indicating 93.2% efficiency. For MBR, the inlet and outlet GWFs were 1.784 ± 0.439 MCM/month (by TSS) and 0.221 ± 0.054 MCM/month (by NO₃), respectively, showing 87.6% efficiency. In S4, the AS and MBR inlet GWFs were 1.274 ± 0.246 MCM/month and 0.414 ± 0.043 MCM/month (both by TKN), respectively. The outlet GWFs were 0.077 ± 0.065 MCM/month (by TKN) and 0.043 ± 0.011 MCM/month (by NO₃), with 94% and 89.6% reduction efficiencies for AS and MBR, respectively. Using GWF_n (Eq. 5) was more valid than the GWF based on effluent quality alone, particularly for industrial wastewater with multiple pollutants. Monthly average GWF_n for AS were 13.21 (S1), 6.59 (S2), 5.29 (S3), and 1.2 (S4) MCM/month; for MBR, they were 3.99 (S1), 1.61 (S2), 1.56 (S3), and 0.37 (S4) MCM/month. Both modules effectively improved the pollutant profile, transitioning from higher-risk pollutants (e.g., TKN) at the inlet to lower-risk pollutants (e.g., NO₃) at the outlet. Heavy metals did not predominate as leading pollutants in any scenario, suggesting their impacts should be checked with a more innovative approach in the GWF framework. HPI will be discussed further in this research.

The GHG emissions of each wastewater treatment system were typically reported per inflow volume. Previous studies indicated that the AS emitted 0.94 kg-CO₂/m³ and MBR emitted 1.56 kg-CO₂/m³³⁹. However, this study introduced GWCF (Eq. 7) to evaluate WWTP performance by indicating the pressure alleviated on water bodies per unit of CO₂ emission. The monthly average GWCFs of AS were 699.2 (S1), 348.7 (S2), 280.1 (S3), and 63.3 (S4) m³/kg-CO₂ as illustrated in Fig. 4. In comparison, MBR's GWCFs were 179.7 (S1), 72.7 (S2), 70.4 (S3), and 16.7 (S4) m³/kg-CO₂. AS was found to be more efficient than MBR with GWCF perspective, reducing equivalent pollution from water bodies by 63–699 m³ (S4–S1) per unit CO₂ emission, while MBR reduced 17–180 m³ (S4–S1). This discrepancy was primarily due to MBR's higher energy consumption. Additionally, the higher GWCF of AS was attributed to its larger treatment volume. Optimizing energy usage, transitioning to sustainable energy sources, and enhancing pollution removal efficiency were deemed crucial for improving environmental performance based on GWCF.

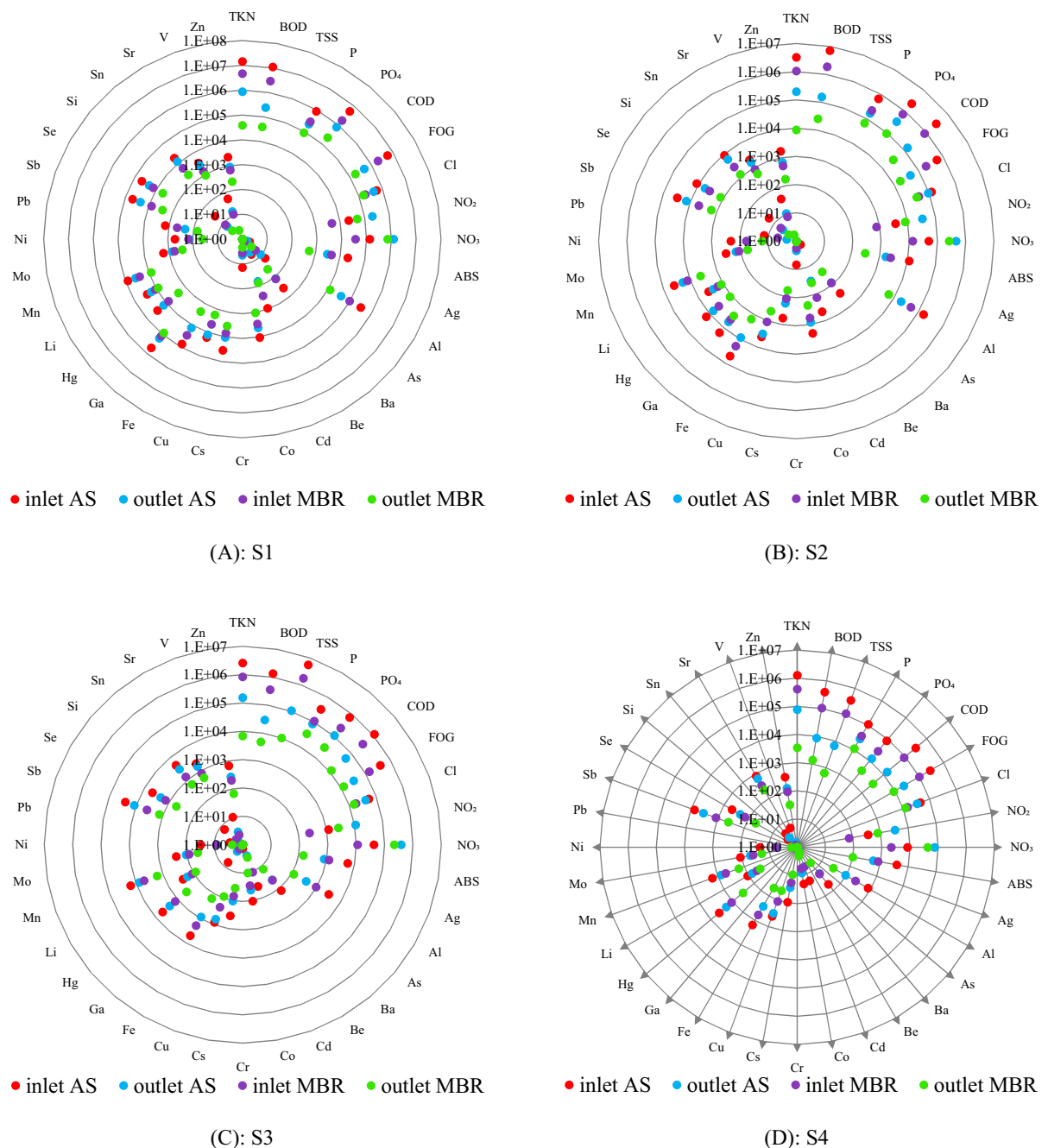


Fig. 3. The monthly average of GWF in logarithmic scale (m^3/month) based on the scenarios: S1–S4.

Figure 5 illustrates the statistical analysis of the GWF_n and $GWCF$ of both treatment units regarding their variations across all scenarios (S1–S4). Here, the monthly average GWF_n of AS and MBR are 6.57 and 1.88 MCM/month, respectively. Furthermore, the monthly average $GWCF$ s of these modules are $347.8 \text{ m}^3/\text{kg-CO}_2$ and $84.9 \text{ m}^3/\text{kg-CO}_2$, respectively.

The variations and trajectory of the GWF s for both modules in the inlet and outlet across different months (October to March) were shown in Fig. 6, along with their monthly GWF reduction efficiency (%). In S1, the highest GWF for the AS inlet was 36.292 MCM in March, while for MBR, it was 6.977 MCM in December. Similar complexity was observed in the outlets. For instance, in S1, the highest GWF for the AS outlet was 4.561 MCM in March and 1.318 MCM in October for MBR. Since GWF is monthly variable due to leading pollutants and pollution loads, it was recommended that GWF s be calculated monthly in both inlet and outlet to ensure accuracy. The maximum GWF reduction in S1 for AS and MBR were 95.7% (October) and 92.1% (January), respectively. The monthly average GWF reduction efficiency for AS was 91% (S1), 93% (S2), 93% (S3), and 94% (S4), while for MBR, it was 85% (S1), 86% (S2), 87% (S3), and 89% (S4).

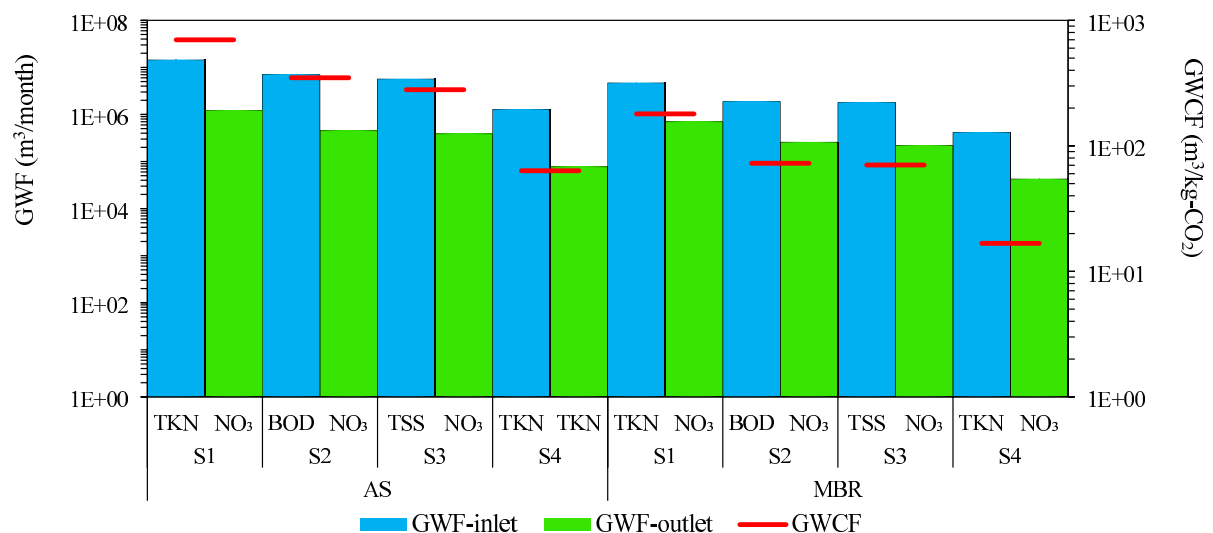


Fig. 4. The monthly average GWF and GWCF of the AS and MBR modules in different scenarios (S1–S4).

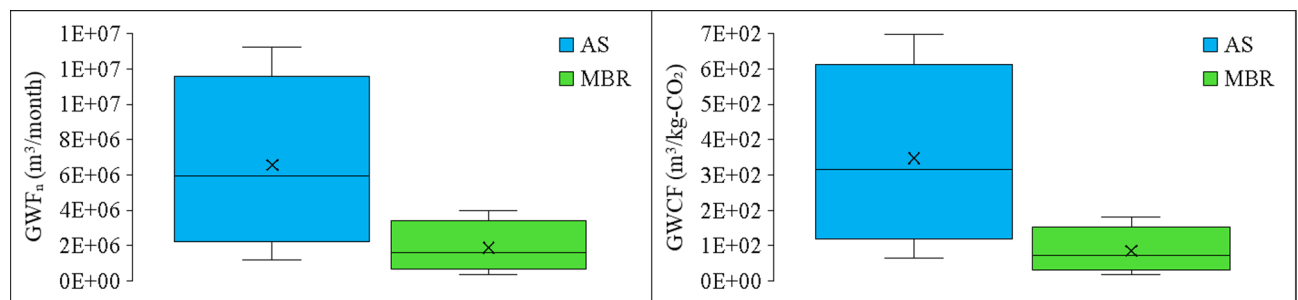


Fig. 5. GWF_n and GWCF box plots based on the monthly average GWF by scenarios (S1–S4).

Comparative study through GWF

The monthly multi-pollutant GWF calculation for the raw and treated wastewater of both modules was found to provide an inclusive quantification framework. Within this framework, multi-pollutant GWF_n , equivalent GWCF, and overall GWF removal efficiency (%) were calculated as tools for comparing the performances of the AS and MBR. This new framework demonstrated advantages over conventional evaluation methods. This framework for comparative studies was established by emphasizing four main criteria derived from industrial GWF evaluations.

- 1) **Removal efficiency:** The 1st criterion for evaluating wastewater treatment systems was the multi-pollutant GWF removal efficiency (%). The AS achieved 91–94% GWF removal efficiency, while the MBR achieved 85–89%. Consequently, the AS consistently outperformed the MBR with an average GWF reduction of 93.1%, compared to 87.1% for the MBR, indicating the superiority of AS in overall pollution reduction (Table 2).
- 2) **Operational efficiency:** The 2nd criterion was the operational reliability (OR_e) and efficiency of wastewater treatment units in controlling and reducing GWF variations from the inlet to the outlet of modules. This criterion is measured by the reduction (%) amount of standard errors for monthly GWFs ($OR_e = 100 \times [SE_{inlet} - SE_{outlet}] / SE_{inlet}$). Higher OR_e indicated reduced fluctuations and greater stabilization of GWF at the outlet. The AS demonstrated slightly higher OR_e than the MBR, especially under stricter standards (S1–S3). The average OR_e of the AS was 83.7%, compared to 77.5% for the MBR, indicating the AS's superiority in reducing pollution load fluctuations and providing a more consistent outlet (Table 2).
- 3) **Carbon efficiency:** The 3rd criterion was the GWCF attributed to energy consumption and carbon emission for GWF reduction. In all four scenarios, the AS exhibited higher GWCF, with an average of 347.8 $m^3/kg-CO_2$ compared to 84.9 $m^3/kg-CO_2$ for the MBR (Table 2). This indicated that the AS performed better in reducing GWF regarding CO_2 production during the study period.
- 4) **Heavy metals reduction efficiency:** The 4th criterion evaluated the efficiency of heavy metals pollution reduction by Eq. 10, assessing the AS and MBR modules' performance in mitigating heavy metals contamination. Although typical pollutants like TKN or COD were leading in GWF assessments, heavy metals required

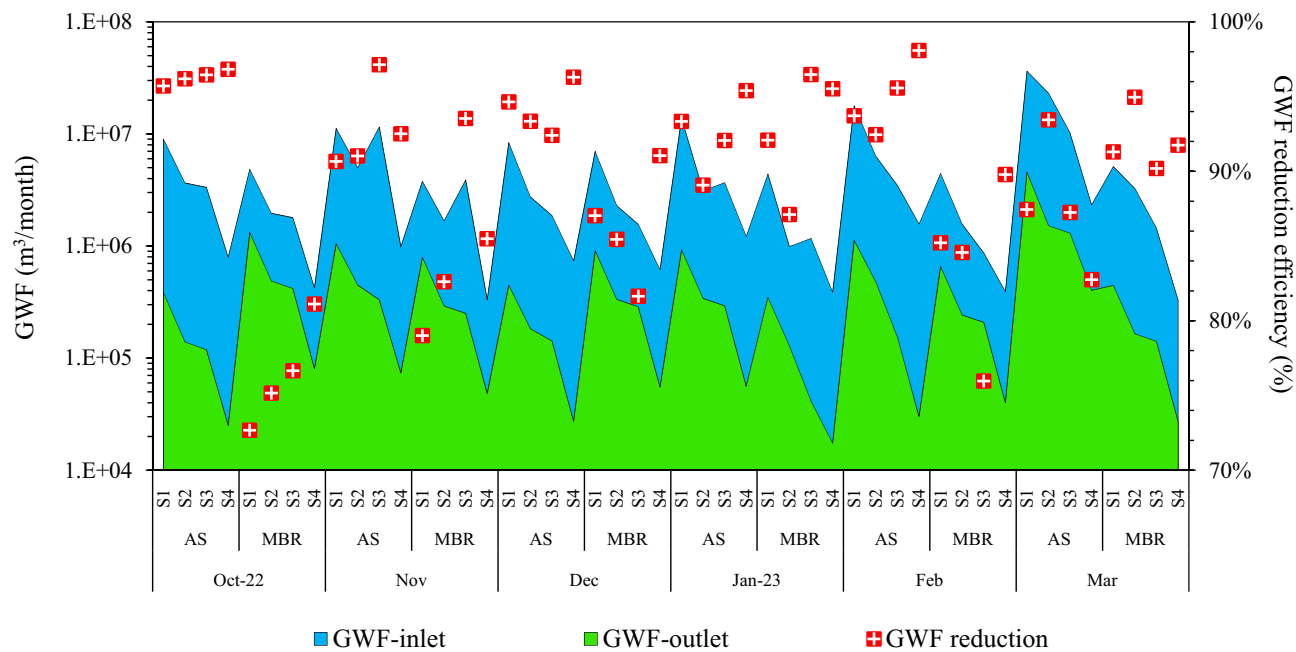


Fig. 6. Monthly GWF of inlets and outlets on a logarithmic scale (m³/month) with their reduction efficiency (%).

Scenario	GWF reduction (%)		OR _e (%)		GWCF (m ³ /kg-CO ₂)		HPI reduction (%)	
	AS	MBR	AS	MBR	AS	MBR	AS	MBR
S1	91.5	85.1	78.9	64.8	699.2	179.7	59.5	33.8
S2	93.6	86.2	93.4	82.9	348.7	72.7	57.7	41.7
S3	93.2	87.6	88.8	87.7	280.1	70.4	51.1	62.9
S4	94.0	89.6	73.6	74.4	63.3	16.7	58.4	63.1
Average	93.1	87.1	83.7	77.5	347.8	84.9	56.7	50.4

Table 2. GWF reduction, OR_e, GWCF, and HPI reduction criteria to compare the performance of the AS and MBR modules.

additional consideration due to their health and environmental risks. HPI reduction was introduced as the fourth index, calculable using the GWF. The AS reached an average HPI reduction of 56.7%, outperforming the MBR's 50.4% efficiency (Table 2), indicating better performance in heavy metals risk alleviation. The AS excelled in S1 and S2 scenarios, while MBR performed better in S3 and S4. In scenario S1, the HPI value for the inlet was 196, which was reduced to 80 by AS and 130 by MBR, with significant reductions in both the inlet and outlets in the more lenient scenarios (S2–S4). Therefore, evaluating treatment system performance must consider the standards and constraints of receiving environment.

Overall, the AS demonstrated superior performance and greater GWF_n , with lower overall CF than the MBR due to less energy consumption, indicating better environmental sustainability. Based on four GWF-based criteria and the developed method, now we can conclude that the AS excelled in reducing GWF (%), minimizing GWF variations (%), producing less CO₂ per GWF removal, and reducing HPI. Despite MBRs' effective performance in reducing certain pollutants by the conventional method (Sect. Pollution removal), the AS proved more suitable based on the multi-parameter GWF evaluation. However, in scenarios S3 and S4 with lenient limitations, the criteria became closer, showing the influence of ambient variables on this evaluation method.

Discussion
Pollution removal in WWTPs

Industrial WWTPs played a crucial role in mitigating pollution from industrial wastewater. Properly equipped and operated WWTPs achieved COD, total nitrogen, NH₄, and total phosphorus removal efficiencies up to 90%, 81%, 95%, and 80%, respectively, while saving 1.5 GWh of energy annually⁴⁰. In this study, MBR reduction efficiencies for Cd, Cu, and Fe were 70.2%, 57.9%, and 63.7% respectively. Notably, the MBR efficiencies for mentioned parameters in another study were equal to 85%, 24%, and 96%⁴¹. The AS reduced COD and

Reference	GWF reduction (%)	CF (ton-CO ₂ /month)
This study	87.1–93.1	18.9–22.2
²⁵	–	2.8–11.9
¹⁶	64.0–90.0	–
²⁴	–	37.4–68.9
⁴⁶	84.8–94.3	–

Table 3. Comparing GWF reductions and CFs in WWTPs.

P concentrations in this study by 95% and 57%, respectively, compared to 96% and 73% in the MBR. In the industrial sector, COD reduction for AS and MBR was determined 83% and 91% respectively⁴². The reduction efficiencies of heavy metals in the AS system were 74.6%, 33.8%, and 68% for Pb, Cu, and Ni respectively (as shown in Table 1). Another study reported that the removal efficiencies for Pb, Cu, and Ni ranged from 83.1 to 90%, 74.3–80.6%, and 0–6.2%, respectively⁴³. These findings indicated that operation methods and wastewater type significantly influenced reduction efficiencies. However, these reductions alone could not clarify advantages in comparative studies.

GWF reduction in WWTPs

The GWF was introduced for a comprehensive evaluation of WWTPs, considering multiple pollutants and receiving environmental standards. Hoekstra et al. (2011) believed WWTPs could reduce the GWF. Industrial WWTPs decreased GWF by up to 90%⁴⁴, similarly in another study, results showed significant GWF reductions, achieving 51.5% with secondary treatment units and 72.4% with tertiary units treating phosphorus⁴⁵. The characteristics of wastewater and specific pollutants also influenced GWF reduction. The AS and MBR systems reduced GWF for pharmaceuticals by 26% and for conventional pollutants by 90%¹⁴. Another study achieving up to 90% annual GWF reduction in an industrial complex, as reported in Table 3¹⁶. This study showed average GWF reductions of 93% and 87% for the AS and MBR, respectively, based on 36 pollutants. Upgrading a WWTP with MBR could achieve a GWF reduction of 12.9 MCM/month for NH₄ and 6.2 MCM/month for TSS¹⁷. Over 15 years, municipal WWTPs reduced GWF by over 91%, with NH₄ and TP as key factors¹⁸. In comparison, an even higher GWF reduction range of 84.8–94.3% in WWTPs observed as given in Table 3⁴⁶. Based on reviewed studies, the type of wastewater and industrial treatment processes significantly affected the pressure on the receiving environment.

Energy consumption and carbon footprint in WWTPs

Treatment units were evaluated not only on pollutant removal performance but also on energy consumption and CO₂ emissions, ensuring a balance between pollution reduction and GHG release. In the current study, AS utilized 1.15 kWh/m³ while this system in other studies consumed 0.109 to 1.63 kWh/m³³⁷, and in another study ranged from 0.128 to 2.28 kWh/m³⁴⁷. AS systems with chemical or physicochemical processes consumed 1.3 to 2.8 kWh/m³³²⁴. The MBR module had an average electricity intensity of 4.11 kWh/m³ in this study however, these technologies used from 0.73 to 0.88 kWh/m³^{348,49} to 1.17 kWh/m³³⁵⁰. Notably, side-stream MBR systems consumed 4 to 12 kWh/m³³³⁶. Accordingly, the GHG emissions varied, with the AS system emitting 0.94 kg-CO₂/m³ and the MBR system emitting 1.56 kg-CO₂/m³ of treated wastewater³⁹. The CFs of AS and MBR modules in this study are estimated about 18.9 and 22.2 ton-CO₂/month, respectively. However, these values can vary significantly, ranging from 2.8 to 68.9 ton-CO₂/month (Table 3), depending on factors such as their size, operational conditions, and the source of energy^{24,25}. A study showed that CF can be used instead of rough energy consumption in WWTPs to integrate GWF and CF within the life cycle assessment for comparing the overall performance of WWTPs⁵¹. By considering the critical interplay within the water-energy nexus, this innovative framework emphasizes the need to evaluate these interconnected sources collectively for sustainable applications. Despite its significance, the concept currently lacks a comprehensive quantification approach, presenting an opportunity for further refinement and adoption in environmental management practices. This study assessed WWTP efficiency by calculating GWF_n per CO₂ emitted (m³/kg-CO₂) as GWCF. The AS achieved an average GWCF of 347.8 m³/kg-CO₂, compared to 84.9 m³/kg-CO₂ for the MBR, suggesting the AS was more effective in mitigating GWF and potentially had a lower environmental impact on the receiving ecosystem.

Despite GWF and CF reductions, GWF has the potential to expand performance evaluation indices. Based on two additional criteria introduced in this study, the AS outperformed the MBR by showing less GWF fluctuations in the outlet and reducing HPI to stricter standards. These GWF-based criteria can expand the operational and environmental application of GWF assessment in WWTPs.

Conclusion

This study mainly introduced and developed an inclusive quantification method based on GWF to evaluate and compare the performance of wastewater treatment systems. The primary conclusion of this development was to introduce GWF as a pivotal multi-functional index. It has basic potentials in assessing multiple pollutants, including regional water quality limits and standards, and pollution loads instead of pollutants concentration in calculations. Now, it is verified that GWF can also be integrated with carbon footprint, heavy metal pollution index, and used for evaluating total pollution removal (%) and operational reliability of treatment units.

This study also carried out an accurate analysis for verifying the applicability and robustness of the developed method based on original data. Sampling real complex industrial wastewater, examining multiple pollutants simultaneously in the influent and effluents of two fully operational treatment units, controlling different water quality standards, and evaluating flow-rates and energy consumptions, were implemented. This methodological approach is highly recommended to follow for further studies in any cases. Some secondary conclusions can be as below:

- Multi-pollutant GWF accounting provided a framework for introducing four performance indices: (1) GWF reduction (%), (2) GWF variation reduction for operational efficiency (%), (3) GWCF for carbon efficiency per abated pollution, and (4) HPI reduction for reducing heavy metals impacts. These criteria were centered on GWF.
- The AS outperformed MBR in GWF removal (93.1% vs. 87.1%), operational reliability (83.7% vs. 77.5%), carbon footprint (345 vs. 85 m³/kg-CO₂), and heavy metals reduction (56.7% vs. 50.4%). By these data we can now conclude that the AS performed relatively more sustainable than MBR, particularly under stricter water quality standards, while MBR was comparable to the AS under lenient water quality standards.
- Heavy metals were not always dominant in GWF assessments compared to pollutants like TKN, suggesting their long-term impacts might have been underestimated. Therefore, HPI could add extra assessment toward the toxicity and long-term impacts of heavy metals in GWF context. Further research was proposed to address toxic elements more effectively without altering GWF's core concept.

Data availability

Data is provided within the manuscript or supplementary information files.

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Author contributions

K. A. worked on Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization, A.D. and S. J. were responsible for Conceptualization, Formal analysis, Validation, Supervision, Project administration, Writing – review and editing. All authors reviewed the manuscript.

Declarations

Competing interests

The authors declare no competing interests.

Additional information

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