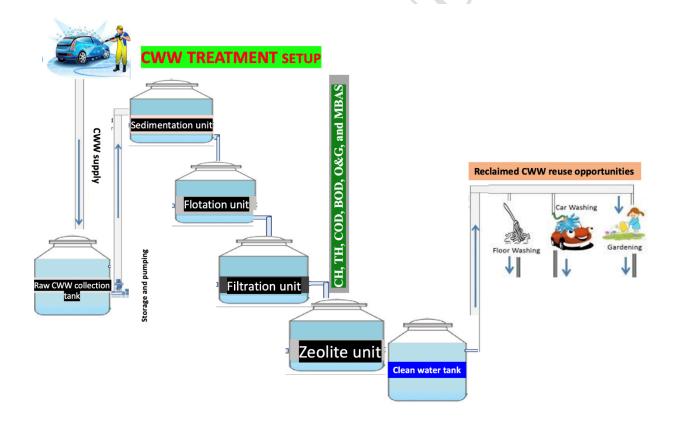
Sustainable surfactant removal and wastewater reuse in carwash systems using natural zeolite

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Graphical Abstract



Abstract

Jordan is the world's second water-poorest country. Treating and reusing of wastewater are significantly important at the national level. This research aims to investigate the use of natural and modified Zeolite for the removal of Methylene Blue Active Substances (MBAS) and lead (Pb), major constituents of carwash wastewater (CWW) that can pose great health and environmental risks. Five samples were collected from each station. Then, physical, chemical, and biological tests were conducted for each sample including Alkalinity, Calcium Hardness (CH), Total Hardness (TH), Total Dissolved Solids (TDS), Total Suspended Solids (TSS), Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), Oil and Grease, and MBAS. Raw wastewater was pre-treated starting with a settling step to flotation which precedes sand filtration and finally zeolite batch adsorption process. Results showed that the natural zeolite is effective in removing MBAS by 55.28% and 93.42% overall removal efficiency of the system. The best conditions of mixing time 30-minute, PH=6.8, and water temperature = 20°C, as this time the initial concentration of Sodium Dodecyl Benzene Sulfonate (SDBS), which is the main pollutant component of the CWW was decreased from 2.5 mg/L to 1.25 mg/L at the final step of treatment. Other contaminants like Pb were reduced during the treatment processes, with 46% removal by sedimentation unit. Reductions observed were alkalinity by 35%, CH by 75%, TH by 86%, COD by 67%, oil and grease by 99.6%, TS 62%, TSS by 76%, TDS by 18%, and BOD by 22%. Sensitivity analysis of the CWW treatment steps shows that the zeolite adsorption unit is the most effective, particularly for reducing parameters such as CH, TH, COD, BOD, O&G, and MBAS showing removal efficiencies up to 99%. Lifecycle Cost Analysis (LCCA) analysis indicates that the developed system is highly profitable and cost effective with a quick payback period, a high rate of return, and substantial net benefits over the lifecycle duration. Furthermore, this sustainable and

eco-friendly technique, which utilizes a natural material, is considered one of the most effective methods for enhancing water resources.

Keywords: Adsorption, Carwash Wastewater, Hardness, Methylene Blue Active Substances, Surfactant Removal, Sustainability.

1. Introduction

CWW is a source of environmental and health pollution as it contains suspended solids, detergents, organic compounds, and heavy metals such as lead and nickel (Kazembeigi et al., 2023). The impact of CWW on the quality of surface water and aquatic life were investigated by Susaj et al. (2023) in Tirana, Albania from November 2018–June 2021. They found that car wash wastewater significantly alter the surface water quality indicators such as water temperature, dissolved oxygen (DO), pH, biological oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids, turbidity, oil and grease the (O&G), and sulphate content. The estimated pollution loads of the CWW in the Kumasi Metropolis examined by Monney et al. (2020), ranged between 2-6 tons/year, which is considered significantly high and causes respiratory problems due to odor. Different studies investigated carwash treatment techniques and reuse. For instance, Gheethi et al. (2016) developed an integrated treatment system for CWW based on coagulation and flocculation as well as a natural filtration system in Parit Raja, Johor, Malaysia. This system efficiently treated raw carwash wastewater, maintaining the concentration of pH, DO, COD, and turbidity within the standard EQA 1974 regulation. Uçar (2017) investigated alternative treatments of car wash effluents in Atasehir/Istanbul, finding that settling, filtration, and membrane filtration processes decreased COD and conductivity by 10% and 4%, respectively. Baddor et al. (2015) analyzed the CWW produced from Aleppo, Syria, determining the optimal conditions for removing surfaceactive substances, the total dissolved solids, and residual oils and grease from car wash water using natural local materials (bentonite). They found that the best conditions for the highest removal efficiency of contaminants were pH=4, temperature= 20° C, and a mixing time =30 minutes. Fayed et al. (2023) assessed CWW treatment using an upgraded physical technique in Alexandria, Egypt, finding that physical treatment effectively lowered the O&G and COD by $79 \pm 15\%$ and $97 \pm 1.6\%$.

In Jordan, carwash systems consume about 20 litters per wash, higher than in other countries (Hussein and Hussam, 2019). Based on the number of registered vehicles in Jordan in year 2021 which was 1,794,073, the total water consumption in carwash stations is approximately 35.88 million litters per day) (Ministry of Water and Irrigation Report, 2016). However, Jordan lacks studies or attempts at treating and reusing wastewater generated in the carwash stations. Therefore, this study suggests that treating and reusing the wastewater produced in the carwash stations in Jordan for rewashing cars or irrigation purposes is feasible. This is achieved through four main physical and chemical treatment steps for carwash water produced from five stations in Amman, the capital of Jordan. This study also indicates that zeolite, a natural material, has high efficiency in removing detergents and heavy metal from CWW with different mixing time. This sustainable approach not only mitigates environmental pollution but also conserves water resources, highlighting the critical importance of this research.

2. Materials and methods

Site visits were conducted in December 2023 to collect the raw water samples from the sewage in five carwash systems in Amman, in addition the general information's such as: the source of the supply water, paths of wastewater, management of wastewater, sampling techniques, preservation, transport, detergents used, the reuse of carwash wastewater, and water consumption as seen in

Table 1 below. These site visits were critical for understanding the operational practices and challenges faced by carwash systems in managing wastewater produced. The following sections will discuss the adopted methodology, which includes sampling methods, laboratory experimental procedures, and methods of treatment used. Table 1 showed data collected from five carwash station s in Jordan noticed that there weren't treatment units in all stations and all samples stored at 20°C.

Table 1. Description of the five CW stations surveyed in this study.

Name of carwash station	Transport	Detergents	Water consumpti on
Alozi station	Jubeha -By car	Carwash shampoo	18 L each car
Total	Swelih -By car	Carwash shampoo, and glass cleaner	16 L each car
Almanaseer	Airport road-By car	Carwash shampoo, liquid wax, and glass cleaner	18 L each car
Alwataneh	Abu nisar-By car	Carwash shampoo	20 L each car
Alhajawi	Almadenh almonawarah street- By car	Carwash shampoo, liquid wax, and glass cleaner	18L each car

2.1. Sampling

During the site visits, systematic sampling methods were employed to ensure that representative samples of wastewater were collected from different stages of the carwash process. Samples were taken from the supply water, post-wash runoffs, and the intermediate storage. The sampling was

conducted following standard protocols to avoid contamination and ensure the reliability of the data collected (Rowe and Abdel-Magid 2020). This study investigated the wastewater generated in five carwash stations in Amman - Jordan. Five samples were collected from each station at December 2023. Then, tests were conducted for each sample and the average value was taken at a different time of sampling. Figure 1 shows the water consumption in litters per car per wash in different water wash systems in Amman city.

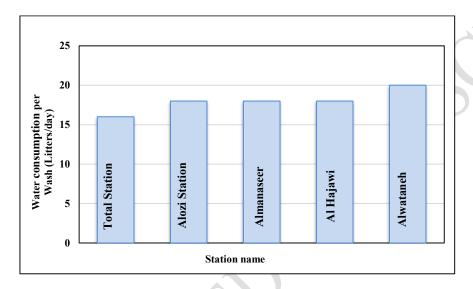


Figure 1. Carwash water consumption for five stations in (litters per car wash) in Amman city.

2.2. Laboratory experimental procedures.

The collected samples were then transport to the laboratory under controlled conditions to preserve their integrity. In the laboratory, various experimental procedures were carried out to analyse the physical and chemical properties of the wastewater. Many parameters were tested and carried out for water samples collected from each carwash station before and after each treatment stage. This includes measuring parameters such as pH, Alkalinity, CH, TH, TS, TDS, TSS, COD, BOD, O&G, and MBAS. Table 2 shows the selected tests methods used according to standard methods of water and wastewater examination with corresponding (Rowe and Abdel-Magid, (2020; Rice et al.,

2017) code:

Table 2. The standard method for physical, chemical, and biological characteristics examination tests

No.	Test	ASTM
-,00		(Method)
1	Chemical Oxygen Demand(COD)	D1252 - 06
2	PH Measurement and Water Conductivity	D5464 - 11
3	Acidity or Alkalinity of Water	D1067 - 11
4	Total Dissolved Solids and Total Suspended Solids in	D5907-10
	Water	
5	Hardness in Water	D1126-12
6	Determining heavy metal Concentration in Water Samples	D3977 - 97
7	Methylene Blue Active Substances(MBAS)	D2330-02

2.3 Description of CWW system.

This study explored multiple treatment methods to evaluate their effectiveness in removing contaminates from carwash wastewater. These methods include sedimentation, flotation, filtration, and the use of zeolite for adsorption. The efficiency of these treatment methods was assessed based on the reduction in contaminant levels and compliance with local water quality standards J893 (JSMO, 2021; Bdour et al., 2022). Raw wastewater was pre-treated starting with a settling step which precedes filtration; it aims to enhance the filtration process by removing particulate matter; the water was left to flow through a rectangular basin at a slow enough velocity and left for 24 hours to permit the particulate matter to settle to the bottom of the basin before the water exits the basin. This water was then sent to a flotation and aeration tanks to remove the oil from water. Next, the water continued to enter a sand filtration unit filled with (Swieleh Sand) sand used as a filtration media. This adsorbent media has specifications describe as follows: the gravel particle diameter is 1.5 mm, the sand particle diameter is 0.05 mm, specific gravity is 2.65 and bed porosity is 0.82. The schematic diagram in Figure 2 below illustrates the proposed wastewater treatment unit for carwash station's water produced in various parts of Amman city, the raw samples was pre-treated

starting with a settling step with glass box dimensions (60x20x20) cm³; the water with flow equal to 7600 L was left to flow through a rectangular basin at a slow enough velocity and left for 24 hours to permit the particulate matter to settle to the bottom of the basin then the water was sent to a flotation and aeration tanks to remove the oil from water. Next, the water with flow rate = 7010L and a hydraulic velocity of 10 m³/m²/hour continued to enter a sand filtration unit for also 24 hours with same dimensions of settling glass box filled with (Swieleh Sand) sand used as a filtration media with sand particle diameter =0.05 mm, specific gravity=2.65, bed porosity =0.82, sand depth of 35 cm. This adsorbent media has specifications describe as follows: the gravel particle diameter is 1.5 mm, the sand particle diameter is 0.05 mm, specific gravity is 2.65 and bed porosity is 0.82, the water passed through the sand were collected to adsorption bat system to study the adsorption isotherms lines of MBAS on the synthesized zeolite used as an adsorbent. Different concentrations of zeolite were added (0.1, 0.5 and 1.0 grams) at different temperatures (25, 35 and 45° C) and pH = 6.8 which is the suitable value were most of the research papers (Tran et al., 2023) and (Rashid et al., 2021) adopted. The mixture was shaken at different times (5, 30, 60, 120) minutes, after which it was filtered and the residual concentration of SDBS-MBAS at the treated samples were measured by the same standard method mentioned above.

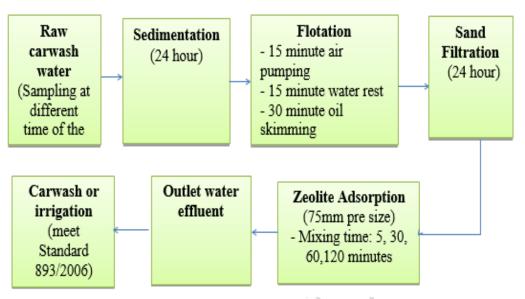


Figure 2. Suggested wastewater treatment processes including sedimentation, floatation, sand filtration and zeolite adsorption units.

Understanding the treatment and reuse potential of CWW is crucial for Jordan for several reasons. Firstly, it helps in the national efforts in mitigating water shortage through assessing and introducing non-conventional water resources, especially considering the positive attitudes of Jordanians towards wastewater reuse (Tarawneh et al., 2024). Secondly, it addresses the environmental and health risks associated with discharge of raw CWW containing harmful substances. Finally, it provides a sustainable solution for water-conservation, especially in regions like Jordan where water scarcity is a significant issue. By demonstrating the effectiveness of natural zeolite in removing contaminants, this study offers a viable, eco-friendly treatment option that can be implemented widely.

3. Results and discussion

The accuracy and errors in measurements of different parameters are very important. Therefore,

to ensure higher accuracy, reliability, and reproductively of the estimated and calculated data for the five carwash stations at different sites in Amman city, all experimenters were carried at least three times. Subsequently, mean resultant values of the measured parameters were presented and adopted.

This study has classified and reported the results into three main categories: physical, chemical, and biological parameters. The following section will describe each of these parameters. The wastewater characteristics were investigated after each treatment steps and there were significant changes through treatment steps but there was a slight change in concentration of the heavy metals such as: Pb, Cd and Zn and all the values within the reclaimed wastewater for irrigation purposes JS893 (JSMO, 2021), as illustrated in Table 3.

Table 3. The average results of CWW effluents physical, chemical, and biological parameters for the five carwash stations studied, and the Jordanian standards for reclaimed wastewater for irrigation.

Test	Raw CWW	Sedimentation tank	Flotation Unit	Filtration Unit	Zeolite Adsorption Unit	JS 893/2021 Standard value (mg/L)
Alkalinity (mg/l as Calcium Carbonate CaCO ₃)	990	850	665	650	422	500
Removal efficiency %	-	14%	22%	2%	35%	
CH (mg/l)	580	220	220	210	53	400
Removal efficiency %		62%		4%	75%	
TH (mg/l)	860	410	410	400	56	500
Removal efficiency %		52%		2%	86%	
TS (mg/l)	1737	664.3	635.8	630.4	367.4	1560
Removal efficiency %		62%	4%	1%	42%	
TDS (mg/l)	407	334.5	305.3	300.4	213.2	1500
Removal efficiency %		18%	9%	2%	29%	
TSS (mg/l)	1330	320	233.4	230.5	126.4	150
Removal efficiency %		76%	27%	1%	45%	
COD (mg/l)	640	480	395	295	98.2	150
Removal efficiency %		25%	18%	25%	67%	
BOD (mg/l)	19.8	15.5	13.4 mg/L	13.4	7.37	60
Removal efficiency %		22%	14%		45%	
O&G (mg/l)	23365	20117	15087	15033	6	8

Removal efficiency %		14%	25%	36%	99.6 %	
Pb (ppm)	0.5	0.27	0.20	0.20	0.18	0.2
Removal efficiency %		46%	14%		4%	
Zn (ppm)	0.01	0.020	0.020	0.020	0.02	5.0
Removal efficiency %						
MBAS (%SDBS)	32.2	30.5	27.6	19.0	1.05	25
Removal efficiency %		5.28%	9.0%	26.7%	55.28%	

3.1. Physical water quality parameters (TS, TDS, and TSS).

The TS concentration is significantly reduced by up to 62% and TSS up to 76% removal efficiency after the sedimentation process. This is expected since all sediments and suspended matter either settle down or float up with the part oil and graces contaminates. Additionally, the TDS concentration also decreases slightly after sedimentation treatment step, achieving a removal efficiency of 18%.

This is illustrated in Figure 3. These results align with the research conducted by kumar et al. (2010), who assessed the removal efficiency of TSS and TDS after primary and secondary sedimentation process in two wastewater treatment plants in Nellakedaranahalli village, India. Their study found TSS, TDS removal efficiencies of 78 % and 20%.

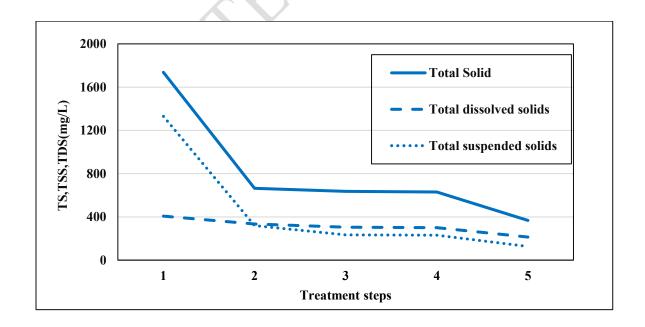


Figure 3. TSS and TDS concentrations (mg/L) after each treatment steps: (1. raw water, 2. sedimentation, 3. floatation, 4. sand filtration and 5. zeolite adsorption).

3.2. Chemical water quality parameters (Alkalinity, CH, TH, O&G, MBAS, and COD).

Figure 4 shows that alkalinity is reduced up to 35 % when the treated water is discharged from the treatment unit (i.e. after Zeolite adsorption treatment step). This indicates that these consecutive treatment steps are effective in the reducing solution alkalinity, which is expected in primary physical primary treatment with a highly basic media effluent. The concentrations of calcium hardness, total hardness, and chemical oxygen demand concentrations are reduced slightly by approximately 62%, 52%, and 25%, respectively, after sedimentation process. This reduction is anticipated as all sediments and inorganic matter settle down sharply. Before the zeolite treatment steps, the reduction percentages of CH, TH, and COD are 62.3%, 57.1%, and 20%. After the zeolite treatment, the reductions are 75%, 86%, and 67%. These results are consistent with the findings of Aragaw and Ayalew (2019) on the removal of water hardness using zeolite synthesized from Ethiopian kaolin by hydrothermal method. Their study showed similar reductions in salts ions, inorganic matter, and chemicals in water by zeolite. Additionally, these results align with the findings of Mkilima et al. (2024) in Nairobi using microbial fuel cells consisting of Zeolitic Imidazolate Framework-8 (ZIF-8). They estimated the removal efficiency of CH, TH, and COD concentrations from CWW to be 65%,75, and 70%.

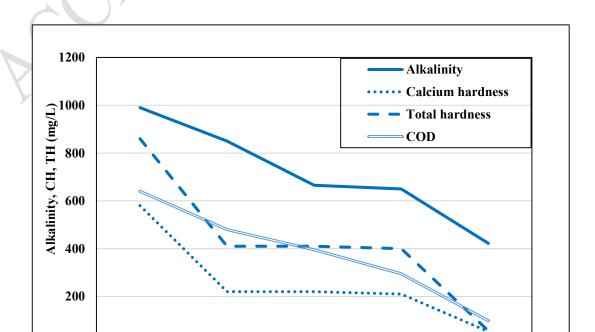


Figure 4. Chemical analysis test results after five treatment steps (1. raw water, 2. sedimentation, 3. floatation, 4. sand filtration and 5. zeolite adsorption).

Figure 5 shows a reduction of O&G pollutants concentration by approximately 25% after the floatation process. This is expected, as oils and fats are skimmed and removed by a mechanical skimmer during flotation. Moreover, the O&G concentration is reduced by 99.6% of its initial concentration after zeolite adsorption, highlighting the effectiveness of zeolite treatment in removing the O&G from the wastewater. These results are consistent with the findings of Sanghamitra et al. (2021) in India, who used biological treatment (BT) technique and modified zeolite adsorption to remove O&G, achieving a removal efficiency of 99%.

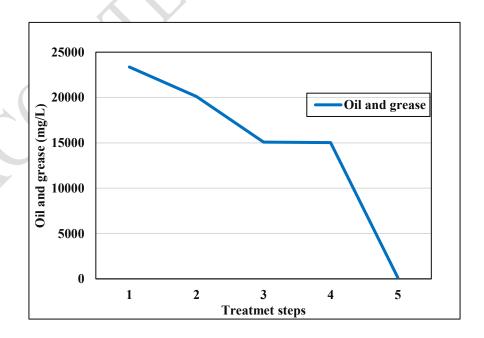


Figure 5. Oil and grease test (mg/L) after five treatment steps (1. raw water, 2. sedimentation, 3. floatation, 4. sand filtration and 5. zeolite adsorption).

Figure 6 shows the concentration values MBAS after each step in CWW treatment system, with the concentration being sharply reduced to a removal efficiency of 93.42 % of its initial value. The highest removal efficiency occurs after the zeolite adsorption step, which is highly effective in adsorbing all kind of surfactants. These results align with the findings of Li (2007), who evaluated the efficiency of clinoptilolite zeolite as a sorbent material for removing cationic surfactant from water in Kenosha, USA. His study showed that an MBAS removal efficiency of about 94%.

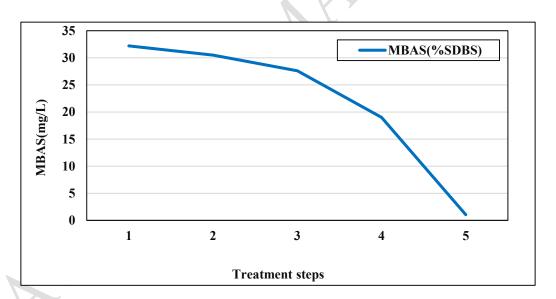


Figure 6. MBAS concentration (mg/L) after five treatment steps (1. raw water, 2. sedimentation, 3. floatation, 4. sand filtration and 5. zeolite adsorption).

3.3. Biological water quality parameter (BOD).

The BOD concentration is reduced slightly about 22% after each treatment steps, as shown in Figure 7. This is expected because the treatment used here is a physical treatment, not biological

one, and the test was conducted to investigate the ability of zeolite to adsorb chemicals and other pollutants. The results indicate that zeolite is highly effective in removing chemicals and solids but slightly effective in removing pathogens. Ultimately, BOD removal is reaches about 45% after adsorption, as shown in Figure 7. These results are consistence with those achieved in a study by Makisha (2021), who improved the secondary treatment of wastewater in an aerobic reactor in Moscow, Russia, increasing the BOD removal efficiency to 55%.

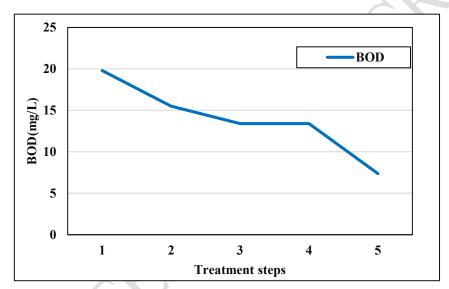


Figure 7. Biological Oxygen Demand (BOD) concentration (mg/L) versus treatment steps: (1. raw water, 2. sedimentation, 3. floatation, 4. sand filtration and 5. zeolite adsorption).

3.4. Sensitivity analysis

Sensitivity analysis was conducted to evaluate the effectiveness of each treatment step. This involved identifying which treatment step contributed most significantly to the reduction of each parameter, including the sedimentation unit, flotation unit, filtration unit, and zeolite adsorption unit. The analysis involved calculating and validating removal efficiencies, comparing treated effluent values with regulatory standards, determining which treatment steps were most effective for each parameter, and identifying areas where the treatment process failed to meet standards, indicating the need for further optimization or additional treatment steps.

The sensitivity analysis of the CWW treatment steps reveals that the zeolite adsorption unit is the most effective, particularly for reducing parameters such as CH, TH, COD, BOD, O&G, and MBAS (%SDBS) showing removal efficiencies up to 99%. The sedimentation tank also plays a crucial role in significant initial reductions, especially in TS, TSS, and Pb with 62%, 76%, and 46% removal efficiencies. The treated CWW in compliance with the Jordanian standards (JS 893/2021) for reclaimed wastewater for irrigation. Overall, the analysis highlights the importance of the Zeolite Adsorption Unit and Sedimentation Tank in the treatment process.

3.5. CWW system lifecycle cost analysis (LCCA)

The LCCA of the CWW system includes four major items as illustrated in Table 6 Initial investment 1) which include storage tanks, pumps, drainpipes, pipe fittings, filter media, and electrical control device; 2) Operating and maintenance costs (OMC) which includes costs of filer medium replacement every 8 months, and fittings and parts replacements; 3) Assuming the revenues and savings (RS) of using CW system will approximately save 60% of the water utility bill which is 165 US Dollars (USD) per month (1,980 USD) yearly; 4) Resale or Salvage value; assuming that CWW system needs replacements approximately every 10 years and costs 150 USD. To perform financial calculations and evaluate the economic viability of the proposed system over its entire lifespan, Table 4 shows the values used in the economic feasibility calculations.

Table 4: CWW system LCCA parameters

Parameter	Value	
Initial Investment (II)	\$1200 USD	
Operating and Maintenance Costs (OMC) per year	\$180 USD	
Revenues and Savings (RS) per year	\$1,980 USD	
Lifecycle Duration (LD)	10 years	
Discount Rate (DR)	10%	
Resale Value (SV)	\$150 USD	

Herein, the net present value (NPV) has been calculated to investigate the LCCA with a minimum acceptable rate of return (MARR) of minimum 5% (Juan *et al.*, 2016). This analysis used three main investment using Eq. 1 (Abdallat *et al.*, 2024).

With Net Present Value (NPV)

$$NPV = \sum \left(\frac{RS - OMC}{(1 + DR)^2}\right) - II + \frac{SV}{(1 + DR)^2}\right)$$
 (1)

With Payback Period (PP), using Eq. 2 (Abdallat et al., 2024).

$$PP = \frac{II}{\Sigma(RS - OMC)} \tag{2}$$

With Benefit Cost Ratio (BCR), using Eq. 3 (Abdallat et al., 2024).

$$BCR = \frac{\sum RS}{II + \sum OMC} \tag{3}$$

Where, II: Initial investment of SFS system

OMC: Operating and Maintenance cost of SFS per year.

RS: Revenues and Savings per year which represents the difference between water bills with and without the SFS system.

LD: Lifecycle Duration which indicates the expected operational lifespan of the setup.

DR: Discount Rate which accounts for the time value of money.

SV: Resale Value or Salvage Value which represents the potential value that can be recovered if the system is sold.

OMCs were considered as part of the LCCA, as outlined in Table 4. The OMC includes the costs of filter medium replacement every 10 months, along with fittings and parts replacements. The frequency of maintenance is designed to ensure optimal and sustained system performance. The 8-month interval for filter medium replacement is based on empirical observations and aims to address any potential decrease in filtration efficiency over time.

Table 5 shows the economic feasibility analysis of the developed CWW system. These results demonstrate that the CWW system is economically viable and financially sound. The positive NPV, the high initial rate of return (IRR), the relatively short PP, and a high BCR greater than 5 all indicate that investment in the developed system is highly profitable and efficient with a quick payback period, a high rate of return, and substantial net benefits over the lifecycle duration.

Furthermore, this system will be attractive to owners of CW stations due to its profitability, buying this system will allow them to treat and reuse a good portion of the water used for showers and toilets. This treated CWW can be reused safely for car washing and irrigation purposes without threatening their health and the environment. Although the system's revenue is vital, applying this system will ultimately help in lessening the water scarcity in Jordan by introducing an alternative new non-conventional water resource, also, considering that the developed system entails no consumables (non-chemical, non-hazardous materials) to address the sustainability of CWW treatment.

Table 5: Results of SFS economic feasibility investigations

Parameter	Value
NPV	11,024 \$US
IRR (NPV=0)	149.98 %
PP	≈ 1 year
BCR	≈ 5.28

4. Conclusion

This study demonstrates the treatment and reuse of CWW in Amman City, Jordan. The findings indicate that natural zeolite is highly effective in removing MBAS, achieving an overall removal efficiency of 93.42% under optimal conditions of a 30-minute mixing time, pH of 7, and water temperature of 20°C. Zeolite also has a significant impact on other contaminants, reducing alkalinity by 35%, CH by 75%, TH by 86%, COD by 67%, O&G by 99.6%, TS 62%, TSS by 76%, TDS by 18% and BOD by 22%. Using zeolite in removing of with carwash stations contaminants was approved efficient removal by this study.

This study confirms that using zeolite for removing contaminants from CWW is efficient. Additionally, the technique is considered as one of the most effective methods for increasing water resources. The treatment technologies used in this research are viable alternatives, providing large amount of water that can be reused in carwash operations. The treatments applied in this study were found to be effective in removing various contaminates with different removal efficiencies. This study shows that natural and modified zeolite have a high potential for removing COD, TDS, TSS, TH, O&G, heavy metals, MBAS. The sensitivity analysis of the CWW treatment steps reveals that the zeolite adsorption unit is the most effective, particularly for reducing parameters such as CH, TH, COD, BOD, and O&G, showing removal efficiencies up to 99%. LCCA analysis indicate that the developed system is highly profitable and cost effective with a quick payback period, a high rate of return, and substantial net benefits over the lifecycle duration.

The methods used in the proposed treatment system are characterized by low operating and

maintenance costs. Therefore, the adopted processes considered environmental-friendly techniques that do not involve the use of any chemical operations.

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