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Transition towards Sustainable Carwash Wastewater Management: Trends and Enabling Technologies at Global Scale

Derrick Dadebo [1], Mona G. Ibrahim [1,2], Manabu Fujii [3] and Mahmoud Nasr [1,4,*]

[1] Environmental Engineering Department, Egypt-Japan University of Science and Technology (E-JUST), Alexandria 21934, Egypt; derrick.dadebo@ejust.edu.eg (D.D.); mona.gamal@ejust.edu.eg (M.G.I.)
[2] Environmental Health Department, High Institute of Public Health, Alexandria University, Alexandria 21544, Egypt
[3] Department of Civil and Environmental Engineering, Tokyo Institute of Technology, Tokyo 152-8552, Japan; fujii.m.ah@m.titech.ac.jp
[4] Sanitary Engineering Department, Faculty of Engineering, Alexandria University, Alexandria 21544, Egypt
* Correspondence: mahmoud.nasr@ejust.edu.eg

Abstract: Carwash wastewater (CWW) contains grease, oil, hydrocarbon residues, heavy metals, and surfactants, posing severe impacts to the environment and human health. Accordingly, various physical, chemical, and biological processes for CWW treatment have been demonstrated in recent research. In this study, a bibliometric approach was performed to comprehensively illustrate the recent progress, current direction, and future perspectives of CWW-related research. A keyword co-occurrence network was used to represent the results of the bibliometric analysis and to show the major pollutants in CWW effluents and the common systems for treating CWW via coagulation/flocculation, electrochemical, oxidation, membrane, adsorption, biological, and hybrid methods. An integrated anaerobic digestion/oxidation process has been reported to degrade CWW-associated pollutants and help develop an energy-efficient approach for waste management. The results demonstrated that the treatment of CWW has several benefits relevant to sustainable development, viz., good health and well-being, protection of life below water, bioenergy generation, and community awareness and acceptance towards wastewater reuse. Hence, these benefits could assist in meeting the environmental, economic, and social sustainable development goals (SDGs). These study outputs can encourage policymakers and stakeholders in implementing sensible regulations that control water usage and treatment in car sharing and personal vehicle services to either directly or indirectly adopt the agenda 2030 with its seventeen SDGs.

Keywords: bibliometric analysis; carwash wastewater; characteristics and treatment; three pillars of sustainability

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1. Introduction

Carwash wastewater (CWW) refers to the effluents of stations, garages, and bays that provide washing and cleaning services to vehicle users [1]. Washing practices utilize large amounts of detergents, soaps, and chemical products to remove particulate matter adhering to the automobile surface [2]. This matter can include dust, grime, sand, dirt, and mud, increasing the solid and colloidal contaminants in carwash effluent [3]. Automotive wash effluents can carry grease, oil, petroleum hydrocarbon residues, heavy metals, and surfactants, which are mainly released from the tyres, engine parts, and connections [4]. The chemical compounds in CWW are responsible for elevating organic and inorganic pollution in aquatic ecosystems, necessitating comprehensive study to develop appropriate CWW management strategies [5].

Several environmental issues, including degradation of marine habitat, loss of biodiversity, aquatic eutrophication, accumulation of metals in the food chain, and alterations in ecological balance have been associated with the unmanaged discharge of CWW [6]. Due to these negative impacts on aquatic and terrestrial environments, international regulations and guidelines have been established in order to maintain the pollutant concentrations in CWW at the allowable limits [7]. For example, the concentrations of chemical oxygen demand (COD), oil and grease, and total suspended solids (TSS) in CWW effluent should be reduced to 60, 2.5, and 25 mg/L before reaching the water bodies, respectively [8]. Consequently, various physicochemical and biological methods have been investigated for treating CWW before discharge into either sewer systems or water bodies.

The most common CWW treatment technologies include chemical coagulation/flocculation [9], adsorption [10], electro-Fenton [3], photo-assisted electrochemical oxidation [2], UV/H_2O_2 oxidation [5], photo-Fenton [11], electrocoagulation [12], electroflotation [13], biological-based systems [1,14], membrane-based systems [6,15,16], and hybrid processes [17,18]. The principles, applications, advantages, and limitations of each treatment method must be defined in order to provide accurate information about CWW management systems. Moreover, biological-based facilities have been applied to generate biogas/biofuel from oily wastewater via the process of anaerobic digestion [19]. In addition, advanced treatment methods are essential in obtaining a final effluent that can be reused in the carwash sector, allowing freshwater to be saved for human consumption [17].

The treatment of CWW is demonstrated by the tangible benefits expected from pollution reduction, human health protection, water security support, bioenergy production, and ought to be promoted by increasing public awareness, acceptance, and attitudes towards wastewater reuse [20]. These advantages are directly connected to the three pillars of sustainability (environmental, economic, and social aspects), meaning that further research is required in order to achieve the 2030 agenda's sustainable development goals ([21]. This agenda is composed of seventeen global goals with 169 targets introduced by the United Nations (UN), and seeks to balance the three dimensions of sustainability, especially for low-income countries [22]. In this context, understanding the available treatment technologies for CWW is essential in order for researchers and policymakers to either directly or indirectly adopt the 2030 agenda with its goals and targets.

A wide range of material, regarding CWW characteristics and the associated treatment technologies, pollution removal, and sustainable development is available in the literature. This information can be visualized graphically in the form of a simple and reliable network. which could be helpful to decisionmakers dealing with CWW management. Here, this network is provided via a bibliometric investigation that evaluates the current situation and growth patterns of this particular research field [23]. Bibliometric analysis can establish the ongoing research contributions among scholars, institutions, and countries.

There is a lack of research elucidating the interactions between CWW treatment and SDGs regarding ecosystem preservation, water-borne disease prevention, water and soil pollution reduction, and improving the proportion of treated wastewater and water-use efficiency. Hence, this study demonstrates the connection between the current CWW treatment applications and the three pillars of sustainable development using bibliometric analysis. As such, the study objectives are threefold: (i) to employ a bibliometric method to provide a comprehensive understanding of the recent progress, current direction, and future perspectives of CWW-related research; (ii) to describe the composition of CWW and the associated treatment technologies; and (iii) to determine the interactions between CWW management and achievable SDGs.

2. Materials and Methods

2.1. Search Strategy

Figure 1 shows the methodological scheme used to develop the bibliometric analysis. It is focused on CWW composition, treatment, reuse, and relevant international regulations, following the Durán-Sánchez et al. [23] hypothesis (2020). The search methodology was based on collecting data relevant to CWW from various online databases, including Scopus (https://www.scopus.com/search/, accessed on 1 January 2022), Google

Scholar (https://scholar.google.com/, accessed on 1 January 2022), Wiley Online Library (https://onlinelibrary.wiley.com/, accessed on 1 January 2022), and Web of Science (https://clarivate.com/web-of-science/, accessed on 1 January 2022). Detailed information on the available publications was collected using the search strings "Carwash" AND "Wastewater" AND "Treatment". Other keywords associated with "Carwash", such as "Vehicle", "Automobile", "Washing Services", and "Cleaning Bay/Garage" were used. Also, keywords such as "Remediation", "Carwash Effluents", "Sewage", "Treatment Technologies/Methods", and "Carwash Characteristics" were used for the bibliometric survey. The search protocol was performed using the criteria of (i) a 2010–2020 period; (ii) omitting duplicate publications; and (iii) original research studies written in the English language.

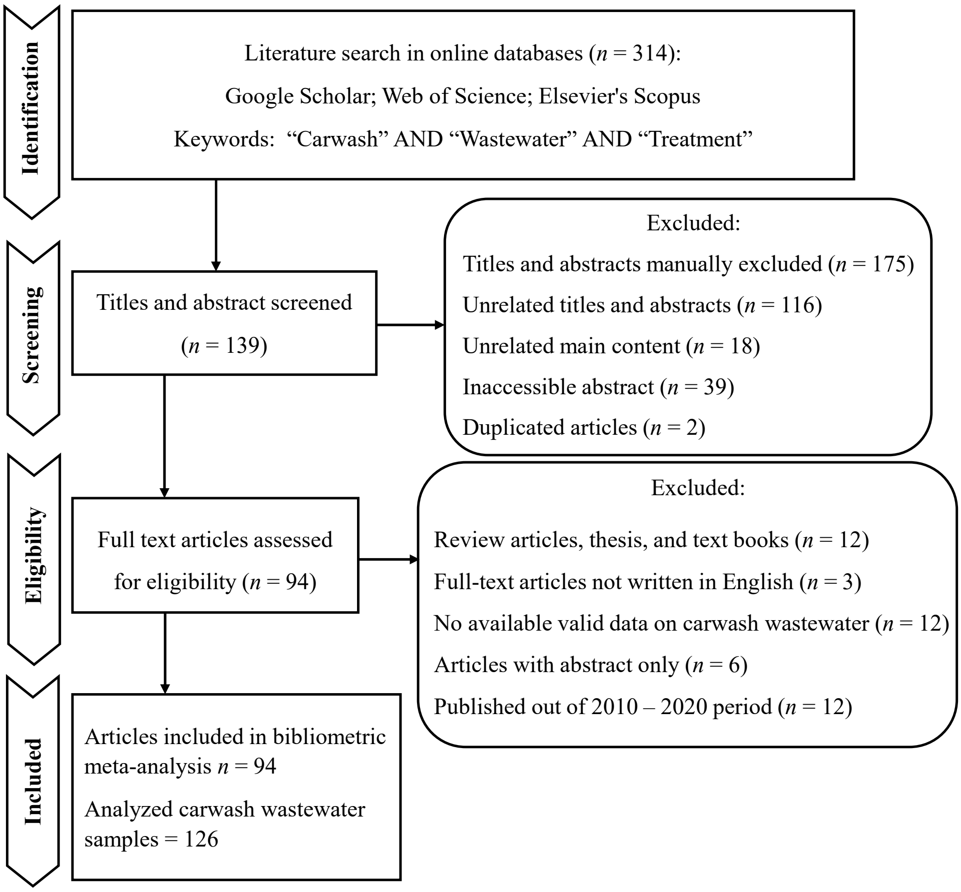

Figure 1. Framework of bibliometric methodology using search strings relevant to "Carwash" AND "Wastewater" AND "Treatment" in various online databases including Scopus, Google Scholar, Wiley Online Library, and Web of Science.

2.2. Data Classification

Types of data used to provide a systematic description of the bibliometric patterns and meta-approaches relevant to CWW Treatment included:

(i) carwash water consumption (in litres per vehicle), depending on the washing method (manual or automatic) and type and size of a vehicle;

(ii) carwash wastewater characteristics, including COD, oil and grease, surfactants, total dissolved solids (TDS), total suspended solids (TSS), and turbidity [24]);

(iii) treatment methods, covering the most common physical/chemical, electro-based, and biological technologies and/or their combinations;

(iv) contaminant removal efficiency, as estimated by Equation (1)

$$R = \left(1 - \frac{C}{C_o}\right) 100 \quad (1)$$

where R is pollutant removal efficiency (%) and C_o and C are the initial and final pollutant concentrations (mg/L), respectively.

2.3. Data Analysis

A bibliometric meta-analysis was performed with 126 carwash samples to represent the physicochemical characteristics of CWW and the performance of the associated treatment methods. The most attractive research fields and future research directions relevant to CWW treatment were represented using Visualization of Similarities (VOS) viewer (https://www.vosviewer.com/, accessed on 1 January 2022) version 1.6.16 software. VOSviewer was employed to map bibliometric information about CWW research based on the co-occurrence network of the keywords. Other procedures involving bibliometric, text mining, and content analyses were conducted as previously reported [25]. Post hoc comparisons of means among groups were analyzed using Tukey's honestly significant difference (HSD) test at $\alpha = 0.05$.

3. Results and Discussion

3.1. Carwash Wastewater Quantity and Composition at the Global Scale

Table 1 indicates that water consumption per carwash varies considerably among continents. This variation depends on the type of washing, which can be self-serve (manual), automated, or semi-automated. For instance, to clean/wash a single car automated processes consume about 200 L of water; this increases to about 45 kL under the self-serve scenario [26]. Moreover, this amount depends on the vehicle type and size, viz., 97 L for motorbike, 158 L for salon car, 197 L for SUV/pick up, 370 L for bus/van, 1139 L for truck, and 1405 L for grader/loader [27]. The socio-economic life and growing population in a country control the amount of water per washed car. The COD concentration in CWW varies from 459 to over 10000 mg/L due to the presence of large amounts of carbonaceous organic pollutants such as surfactants, oil, grease, paint residues, and volatile organic compounds (VOCs). The oil and grease level changes significantly, from 10 to 1395 mg/L, exceeding the WHO threshold value of 2.5 mg/L [8]. Oil is a mixture of hydrocarbons, whereas greases contain a thickener and a lubricating fluid; generally, both of them are hydrophobic [28]. Surfactants are detected in CWW due to the application of detergents, soaps, rinses, and waxes during washing procedures in automobile service stations, workshops, or garages. These anionic surfactants can transfer with wastewater to water bodies, causing toxicity to aquatic organisms (e.g., destruction of chlorophyll–protein complex in plants) and a shift in ecological balance [29]. CWW composition includes metal ions (Cr^{2+}, Fe^{3+}, Zn^{2+}, and Cu^{2+}) released from car wheels, bumpers, engines, surface paints, brake drums, and batteries. In addition to cations (e.g., Ca^{+} and Mg^{2+}) and anions (e.g., carbonates, chlorides, and PO_4^{3-}), these metal ions tend to increase the TDS concentration. Hence, the average values of TDS in CWW across continents are above the WHO effluent standards of 300 mg/L [8]. Moreover, CWW carries organic and inorganic solids, including tire dust, debris, rust stain, engine abrasion, sand, and films from brakes, removed during the vehicle cleaning process. The conventional asphalt pavements in certain countries can facilitate the re-suspension of particulate matter in the atmosphere under certain dynamic conditions, leading to the accumulation of fine dust on vehicle surfaces [30]. During rainy seasons, large amounts of grime, mud, and dirt can adhere to a vehicle's tires and body, resulting in elevated levels of TSS and TDS (Table 1).

Table 1. Variation of CWW quantity and composition among the continents.

	Asia	Africa	Europe	North America	South America	Oceania
Volume (L/vehicle)	462–758 (*n* = 9)	150–347 (*n* = 3)	275–425 (*n* = 7)	97–171 (*n* = 2)	310–450 (*n* = 4)	145–200 (*n* = 3)
pH	6.4–13.8 (*n* = 39)	6.9–8.6 (*n* = 26)	5.5–11.5 (*n* = 17)	6.3–7.8 (*n* = 3)	4.4–7.7 (*n* = 11)	6.1–8.5 (*n* = 6)
COD (mg/L)	1418–2032 (*n* = 51)	936–1413 (*n* = 11)	9716–14010 (*n* = 16)	682–1024 (*n* = 3)	459–683 (*n* = 13)	580–944 (*n* = 6)
Oil and grease (mg/L)	860–1395 (*n* = 34)	10–50 (*n* = 18)	76–125 (*n* = 4)	300–448 (*n* = 5)	71–112 (*n* = 8)	25–83 (*n* = 3)
Surfactants (mg/L)	145–189 (*n* = 12)	2–9 (*n* = 7)	214–290 (*n* = 10)	3–9 (*n* = 4)	77–119 (*n* = 8)	82–106 (*n* = 2)
Turbidity (NTU)	786–1194 (*n* = 32)	2810–4000 (*n* = 24)	212–366 (*n* = 8)	547–925 (*n* = 5)	242–426 (*n* = 11)	689–1000 (*n* = 5)
TSS (mg/L)	940–1750 (*n* = 26)	2055–3417 (*n* = 5)	1450–2300 (*n* = 9)	319–538 (*n* = 7)	68–260 (*n* = 7)	807–1275 (*n* = 5)
TDS (mg/L)	5521–7920 (*n* = 26)	448–686 (*n* = 20)	674–1054 (*n* = 2)	11876–17268 (*n* = 4)	520–803 (*n* = 9)	527–818.4 (*n* = 4)

3.2. Carwash Discharge into the Environment

Pollutants associated with CWW (e.g., sewage rich in surfactants and detergents) transfer from carwash services to treatment facilities and/or water bodies. The direct discharge of CWW into surface waters or without proper treatment triggers the degradation of aquatic habitats, water quality, and biodiversity (Figure 2). For example, an elevated COD level in CWW is responsible for depleting the DO of water bodies, adversely impacting aquatic life, water potability, and agricultural practices. Oil and grease in CWW are responsible for clogging sewerage systems, forming a thick and dense layer above water bodies, and harming aquatic life, with long-lasting impacts. Because surfactants are amphiphilic, they can form foams on the surface of rivers and curtail oxygen diffusion in the water. These pollutants can damage fish eggs, livers, and muscles and hinder animal and plant growth in the receiving water bodies [31]. Oil, fats, grease, and inorganics in wastewater can cause clogging of sewer systems, increasing the occurrence of overflow on streets and forming stagnant pools. Hence, wastewater treatment units tend to remove these contaminants via multiple physicochemical, biological, and/or hybrid processes for meeting the discharge standards. The treated effluent can either be reused for car cleaning or disposed of to water bodies. CWW reuse is an essential strategy in conserving limited water resources, and therefore in paving the path for sustainability.

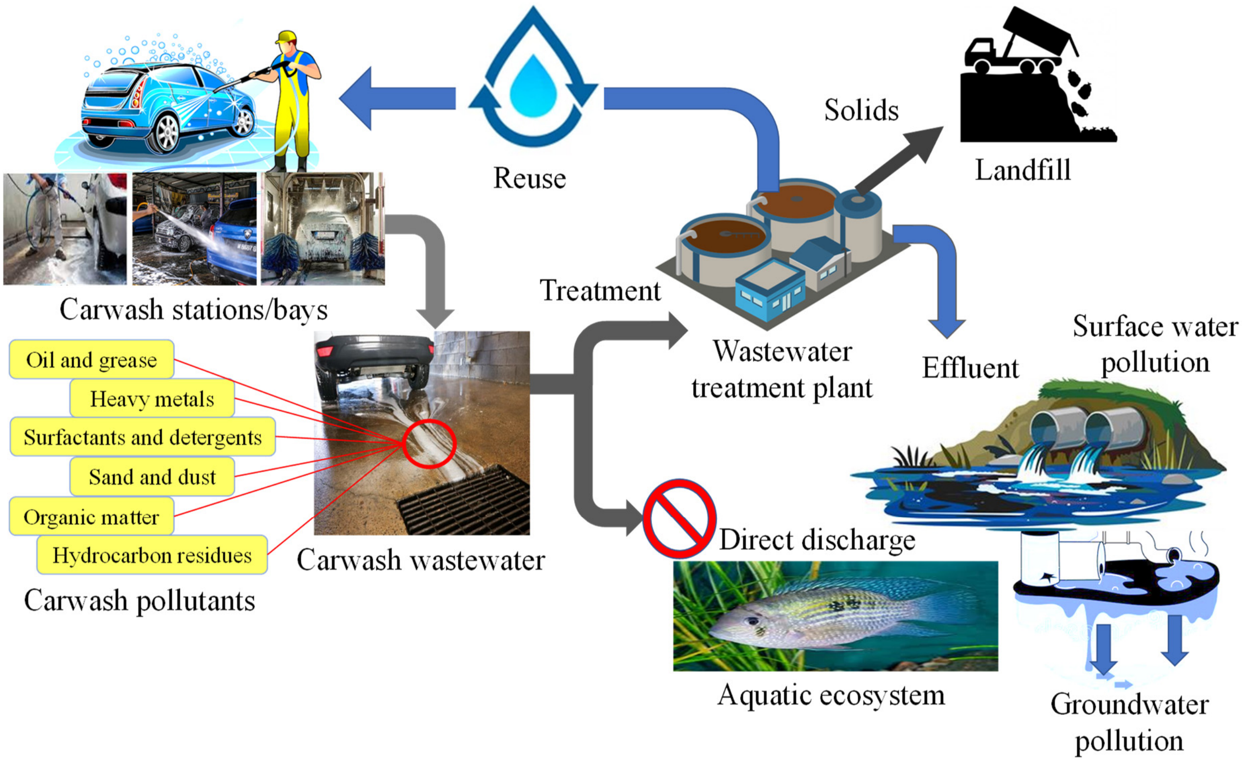

Figure 2. Transfer of carwash wastewater to the environmental matrix.

3.3. Carwash Wastewater Treatment Technologies

3.3.1. Adsorption

Adsorption has shown a wide range of applications for removing particulates from CWW owing to its simplicity, high performance, and short treatment time (Figure 3a). Adsorption is used to eliminate pollutants from CWW through multiple mechanisms, including hydrophobic interaction, chelation, and electrostatic and van der Waals interactions [30]. The size of particulate pollutants (e.g., road dust, tire/brake wear, mud, and dirt) in CWW are considered large enough to facilitate the solid/liquid separation process; hence, TSS removal by adsorption is sufficient. As such, the particle size of raw CWW has a normal distribution of about 11.4 µm (50% of particles) and can reach up to 144 µm [16]. It has been suggested that the neutral-to-alkaline condition encourages the physical adsorption of oil particles onto coagulants via a sweep flocculation mechanism [7]. An adsorbent material having sufficient amounts of negative charge on its surface would form a strong electrostatic attraction to positively-charged contaminants, depending on solution pH and ionic strength [32]. Chemical modifications could be performed for the cationization of adsorbents, after which they become able to form a strong electrostatic attraction with anionic contaminants. Moreover, abundant surface functional groups, including CO, NH_2, and aliphatic and aromatic COOH, are essential for binding to various ions in CWW, leading to greater adsorption ability [33]. For instance, activated carbon prepared from bituminous minerals has been used to remove anionic surfactants from CWW, achieving an adsorption capacity of 5.65 mg/g after 2 h [34]), and a macrocomposite adsorbent containing activated carbon, aggregates, sand, zeolite, and cement has been used to remove COD (1.3–1.8 mg/g) and suspended solids (1.5–2.1 mg/g) from CWW in a fixed-bed adsorption column [33].

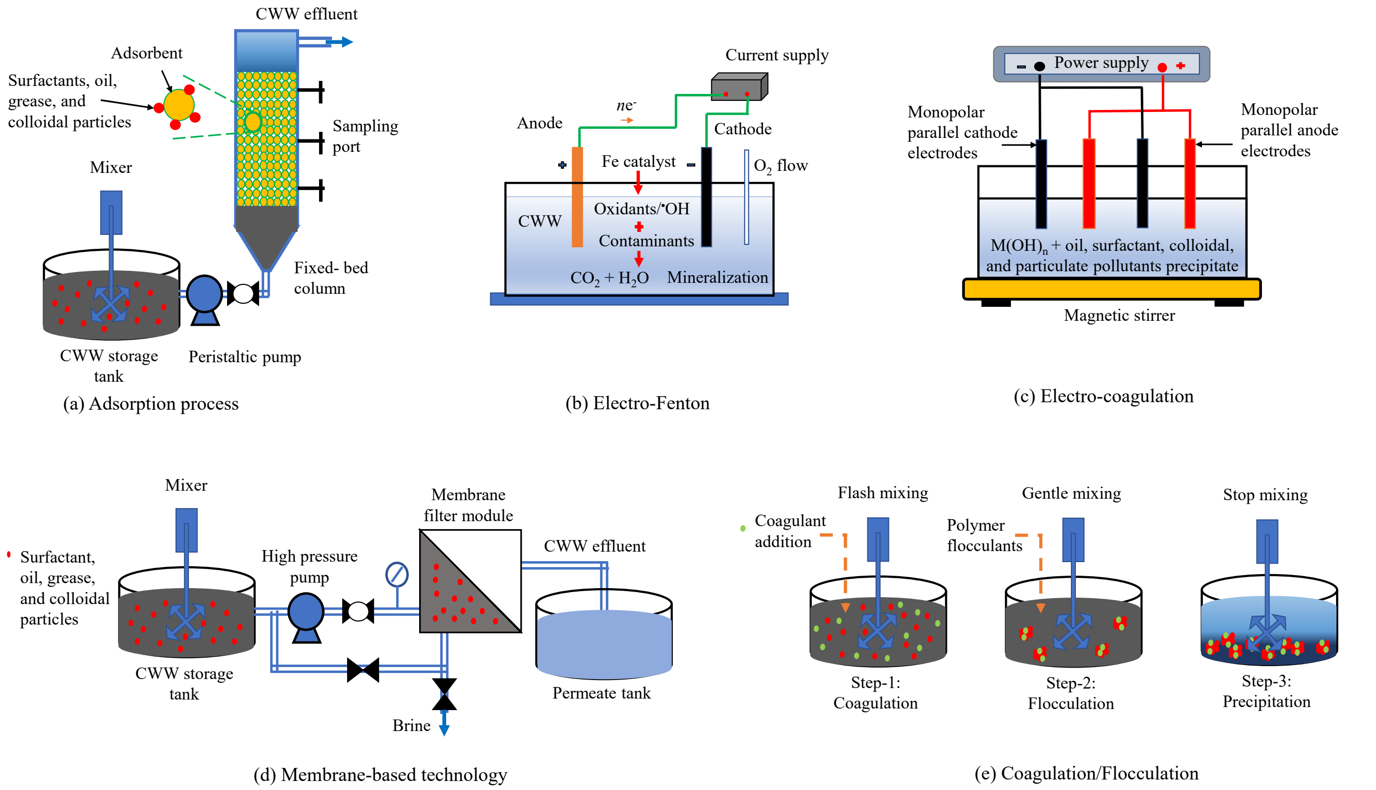

Figure 3. Common technologies for treatment of carwash wastewater (CWW).

3.3.2. Oxidation Processes

Ozonation, Fenton-related chemistry, and ultraviolet and ultrasound irradiation are common advanced oxidation processes used for treating wastewater containing oil residues [35]. For example, the degradation of organics by Fenton's method ensues by the oxidation of Fe^{2+} to Fe^{3+}, leading to the formation of hydroxyl radicals (OH) (Figure 3b). Advanced oxidation can be employed to destroy organic and inorganic contaminants in CWW as a pretreatment step prior to other biological-related processes. This pretreatment phase is essential because the toxic contents in CWW would inhibit the bacterial activity intended for removing organic compounds [36]. However, most oxidation mechanisms are subjected to pH adaptation (e.g., pH around 9 for ozonation and pH around 3 for Fenton) to maximize the COD reduction; hence, pH neutralization is required before biological treatment processes [18].

3.3.3. Electrocoagulation

During the electrocoagulation operation, metal ions (e.g., Fe^{2+}) are released from the anode of the reactive (iron) electrode, forming coagulants that destabilize the colloidal suspension (Figure 3c). The destabilized particles are aggregated to form flocs, which are further removed in a solid/liquid separation phase [13]. The electrocoagulation process has several advantages, such as simple operation, short processing time, and reduced sludge production. Several researchers have employed the electrocoagulation mechanism to cope with the chemical addition problem in the conventional coagulation/flocculation process. For example, Gönder et al. [37] demonstrated the application of electrocoagulation with Fe and Al electrodes to treat CWW. The optimum pH, current density, and operating time for the Fe electrode were 8, 3 mA/cm^2, and 30 min and for the Al electrode 6, 1 mA/cm^2, and 30 min [37]. Electrocoagulation operated with the Fe electrode achieved COD, oil–grease, and chloride removal efficiencies of 88%, 90%, and 50%, respectively. These removal rates were 88%, 68%, and 33% for the Al electrode, respectively [37]. It has been reported that the performance of electrocoagulation to treat wastewater containing micellar surfactant is influenced by several factors such as surfactant initial concentration, reactor polarity, current density, electrode material (e.g., Al or Fe), and processing time [38].

3.3.4. Membrane-Based

In membrane-related technologies (Figure 3d), high pressure drives CWW through fixed pore-sized membranes where contaminants are effectively retained, producing a high-quality effluent [28]. Based on pore size, the membrane modules used for CWW treatment include microfiltration, ultrafiltration, nanofiltration, and reverse osmosis. However, membranes suffer from low flux due to the deposition of oil droplets and particulates within the membrane pores. Therefore, enhancing the performance of membrane-based technologies to treat CWW is controlled by wettability (i.e., due to the hydrophobic properties of oil, grease, and surfactants) and the antifouling behavior of membranes [16]. The application of chemical and physical pretreatment steps before using membranes has been recognized as a viable option to mitigate flux-related and fouling-related problems. These pretreatment processes include oil/water separation (skimmers), induced air flotation, and dissolved air flotation.

3.3.5. Coagulation–Flocculation

The coagulation/flocculation process can form multiple mechanisms, such as charge neutralization, bridging, sweeping, and charge patching, to remove oil particles from wastewater (Figure 3e). The solids in CWW can be removed by chemical coagulants such as ferric chloride, alum, and ferric sulfate through electrostatic attraction and/or sweep flocculation mechanisms [9]. However, the addition of chemicals increases the dissolved ions, resulting in secondary pollution by TDS. Jiku et al. [39] reported that Poly Aluminum Ferric Chloride could be used as a suitable coagulant for CWW treatment and damaged the stability of oil colloidal contaminants. The pH of CWW can be adapted according to the point of zero charge (pH_{PZC}) to improve the electrostatic interaction between organic pollutants (as COD) and coagulant surfaces. However, coagulation/flocculation should be integrated with another treatment process such as membrane filtration, adsorption, or oxidation to improve pollutant removal effectiveness. For example, CWW with a high load of oil and grease can be treated using an integrated coagulation/flocculation and oxidation with hydrogen peroxide method [17]. The COD removal efficiency in the first stage was 92.4%, which was enhanced to 94.4% in the combined system [17].

3.3.6. Biological-Based Process

Biological processes have the advantage of converting hydrocarbons into less harmful end products rather than only separating oil and grease from wastewater, which involves disposal problems [19]. The degradation of oil and grease by bacteria, fungi, and algae (or the combination of particular enzymes in the same reactor) has been previously reported [14]. These microorganisms must be well acclimatized and adapted for an extended period (several months) using a sufficient amount of oxygen (for aerobic biological treatment) and nutrients. For instance, *Pseudomonas* has been employed as a lipase-producing strain for degrading oil and grease in industrial wastewater. Moreover, a two-stage anaerobic–aerobic treatment has been considered for promising biodegradation of hydrocarbons, requiring no additional pretreatment steps [1]. However, the success of biological processes to treat CWW is limited by the applied load of oil and grease, as well as the type and design of bioreactors. Moreover, biological-based methods can cause air pollution due to the volatilization of light hydrocarbons, requiring frequent monitoring, control, and maintenance to avoid microorganism deactivation.

3.4. Performance of Carwash Wastewater Treatment Methods

3.4.1. COD Removal Efficiency

Among the CWW treatment technologies, advanced oxidation is the best process for reducing COD contaminants, with a removal efficiency of about 94.0% (Figure 4a). The high reduction of COD by oxidation techniques can be assigned to the generation of $^{\bullet}OH$ or $SO_4^{\bullet -}$, which is responsible for degrading the recalcitrant organic pollutants. The highly reactive intermediates, irradiation, powerful oxidizing agents, and catalysts are suitable for attacking light and heavy hydrocarbons and other impurities in CWW. In wastewater laden with oil residues [11], a COD reduction of about 50% was maintained by photo-Fenton with pH= 3, H_2O_2 = 400 mg/L, and Fe^{2+} = 40 mg/L. Due to unacceptable final COD concentration levels, a physicochemical treatment process was employed as an initial stage, resulting in improved COD reduction to 75% [11]. The results of our bibliometric analysis showed that the high performance in COD reduction by advanced oxidation was, when followed by adsorption, equivalent to a removal efficiency of 88.7%. COD reduction by adsorption is limited by external (film) mass transfer resistance, which controls the diffusion of organic/inorganic pollutants from the CWW solution to the adsorbent surface [40]. The adsorption capacity of the solid phase depends on several parameters, such as CWW pH, initial COD concentration, and contact time. Coagulation/flocculation exhibited the lowest COD removal efficiency at 62.7%, having a significant difference (Tukey's HSD, $p < 0.05$) with other treatment methods (Figure 4a). This insufficient COD removal efficiency by coagulation/flocculation may be due to soluble organic pollutants in CWW, such as micelles of surfactants (COD caused by soluble materials) not being sufficiently trapped and coagulated into flocs.

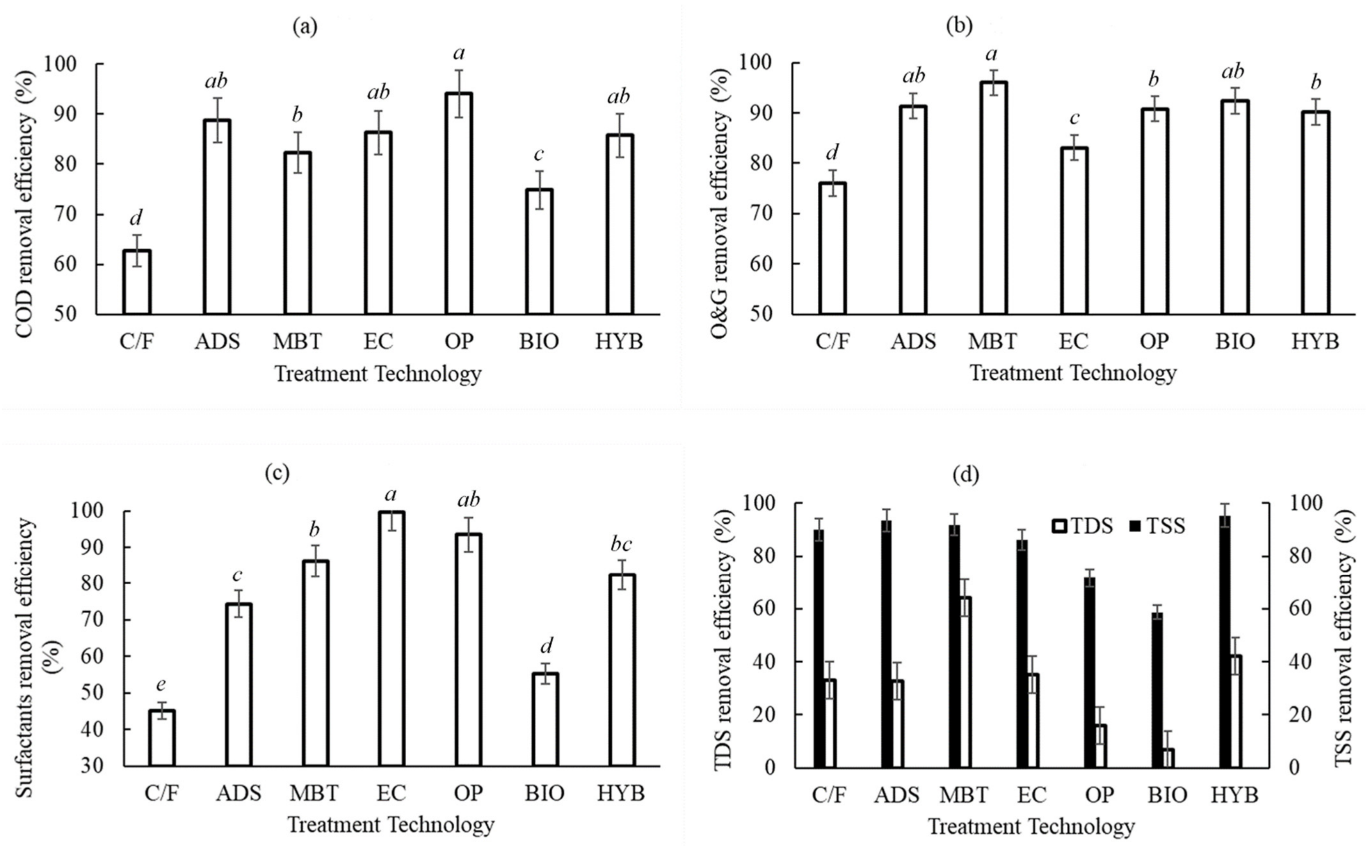

Figure 4. Removal efficiencies of (**a**) COD, (**b**) oil and grease, (**c**) surfactants, and (**d**) TDS and TSS from treated carwash wastewater (CWW) using various treatment technologies. Based on one-way ANOVA followed by Tukey post hoc test, removal efficiency values with same superscripts letters indicate an insignificant difference ($p > 0.05$).

3.4.2. Oil and Grease Removal

Membrane-based technologies showed the highest performance for oil and grease rejection from CWW (Figure 4b). Istirokhatun et al. [41] employed ultrafiltration membranes to treat CWW, achieving a complete rejection of oil and grease. The interaction between the oil droplet and the membrane surface depends on various parameters, such as feed oil concentration, trans-membrane pressure, operational temperature and pH, and cross-flow velocity. Despite high oil and grease removal by the membrane-based technologies, the accumulation of micrometer-sized oil droplets on the surface causes pore-clogging and critical fouling issues. The removal of oil and grease by coagulation/flocculation exhibited a low percentage, about 76.0% (Figure 4b). This unsatisfactory treatment performance can be assigned to the alkaline nature of CWW due to the presence of washing shampoos and detergents. Increasing the pH above the optimum value (mainly under alkaline conditions) might result in higher negative zeta potential values [15]. This condition causes a weak electrostatic attraction between coagulant surfaces and oil particles, reducing the aggregation ability of small (micro-sized) particles into larger flocs.

3.4.3. Surfactant Removal Efficiency

The data in Figure 4c imply that the removal of surfactants from CWW using electrocoagulation is a feasible scenario. During the electrocoagulation operational process with Al electrodes, aluminium–hydroxy complexes are generated to adsorb surfactants within a shorter period of time. For instance, Dimoglo et al. [13] found that electrocoagulation using Al electrodes had a surfactant removal efficiency in excess of 85%with a processing time of only 5 minutes. The presence of various functional groups such as sulfonate, carboxylate, and polyethylene glycol in surfactants facilitates binding performance. In a previous study, increasing the current density showed a positive impact on the removal of three different surfactants (i.e., cationic, anionic, and non-ionic surfactants) [13]. However, the anionic surfactants migrated and accumulated on the Al anode's surface, which further increased passivation behavior. Under this condition, the anode can suffer from a reduction of its effective surface area, resulting in a slower dissolution rate. For removing surfactants from wastewater by electrocoagulation, Önder et al. [12] found that the addition of H_2O_2 formed Fenton's reagent, improving the treatment performance of Fe electrodes.

Advanced oxidation processes have shown high performance in treating CWW thanks to their ability to generate highly reactive intermediates (hydroxyl radicals) that degrade surfactants. The common oxidation techniques used for surfactant degradation include H_2O_2/UV, Fenton (Fe^{2+}/H_2O_2), and photo-Fenton (Fe^{2+}/H_2O_2/UV), owing to their fast and efficient oxidation performance [18]. Moreover, the photochemical degradation of surfactants generates oxidation products such as aldehydes and carboxylic acids, which are further mineralized to CO_2 and H_2O with increasing the H_2O_2 concentration [35]. However, the concentration of H_2O_2 should be optimized in order to avoid the scavenging effects of $HO^\bullet$ by the water matrix components, which can reduce the surfactant degradation ability. Additionally, the oxidation processes should be operated at an optimum Fe^{2+} level in order to avoid further iron precipitation issues. In another study, the degradation of surfactants was effectively achieved by a UV/$K_2S_2O_8$ system thanks to the generation of $HO^\bullet$ and $SO_4{}^{\bullet-}$ radicals, which act as strong oxidants [5].

The results of our bibliometric analysis show that conventional biological treatment processes are not always the best choice for treating surfactant-laden wastewater. For instance, surfactants such as nonylphenol ethoxylate can be biodegraded into more toxic metabolites. The fate and effects of these metabolites on aquatic and terrestrial environments are unknown, and require further comprehensive investigation. Moreover, these bio-processes produce a large amount of sludge containing surfactant compounds, adversely impacting the different ecological systems if disposed of improperly [14]. Biological processes for treating surfactants are limited by foam formation, biomass washout, and reduction of the functioning of bacterial communities. Accordingly, these bioremediation systems have been integrated with other treatment technologies to sufficient degrade various complex surfactants that require a longer retention time. Moreover, cationic surfactants such as quaternary ammonium compound can be degraded by aerobic biological processes, as they are utilized as a source of carbon and energy for certain microorganisms; however, the anaerobic biodegradability of cationic surfactants remains invalid [42]. Therefore, it can be assumed that cationic surfactants are better removed by adsorption onto a negatively-charged surface, electrochemical oxidation, or a combination of both. However, anionic and non-ionic surfactants have less toxicity on the environment than the cationic ones. Integrating a biodegradation process with advanced oxidation may enhance surfactant removal efficiency and reduce operating costs [39].

3.4.4. Removal of Total Dissolved Solids (TDS) and Total Suspended Solids (TSS)

The coagulation/flocculation process shows appropriate TSS removal efficiency from CWW at around 90.0%. The oil droplets and colloidal content in CWW are reduced by membrane technology, which has a TSS removal efficiency of about 92.0% (Figure 4d). The membrane-based approach shows better performance in eliminating TDS, with a removal efficiency of 64.3% (Figure 4d). Pressure-driven membrane processes such as nanofiltration (NF) and ultrafiltration (UF) have been used to remove TDS and other ionized constituent particles from CWW. The rejection performance depends on the passage of the dissolved fraction of contaminants through the membranes and their subsequent accumulation in the draw solution. The rejection of TDS by membranes is considered a function of time, feed TDS concentration, and transmembrane pressure. Interestingly, TDS removal efficiency improved to about 98% when the supernatant of coagulation/flocculation was subjected to filtration by sand layers and membranes (reverse osmosis). The TDS removal efficiency fell below 10% when CWW was treated with biological processes. The anaerobic biodegradation of the CWW organic fraction results in the generation of soluble metabolites and volatile fatty acids that increase the dissolved matter in the effluent. In addition, CWW contains dissolved organics that are considered complex and not completely bioconverted. For instance, Mallick and Chakraborty [1] found insufficient removal of TDS from CWW by bioremediation, owing to high salinity (1.23 $\pm$ 0.11 g/L) and metal composition, e.g., Na^+ (20.13 $\pm$ 3.12 mg/L), K^+ (37.44 $\pm$ 6.13 mg/L), Ca^+ (21.87 $\pm$ 6.33 mg/L), Mg^{2+} (1.09 $\pm$ 0.07), Zn^{2+} (0.85 $\pm$ 0.06), Pb^{2+} (1.02 $\pm$ 0.02 mg/L), and Mn^{2+} (1.22 $\pm$ 0.01 mg/L). Accordingly, most studies have employed a secondary stage of treatment after the biological process to maintain efficient TDS removal.

3.5. Optimization of Operational Factors for Enhancing Carwash Wastewater Treatment

The optimization of operational conditions is an essential step in enhancing the performance of CWW treatment. The findings in Table 2 indicate that hybrid wastewater treatment systems can be classified into physical–biological, chemical–biological, and physical–chemical systems. As such, a hybrid system of electrocoagulation and nanofiltration has been used to treat CWW, achieving COD and oil–grease removal efficiencies of 88% and 90% [37]. Their findings were reported under the optimum condition of 25 °C, stirring speed = 250 rpm, and monopolar electrodes in parallel connection. Collivignarelli et al. [43] demonstrated the application of an integrated biological process (aerobic membrane reactor), membrane (nanofiltration), and adsorption (activated carbon) to treat real wastewater containing non-ionic and anionic surfactants. Average surfactant removal reached up to 95.3 $\pm$ 0.8% using the combined system compared with 47.8 $\pm$ 5.1% for the standalone biological process. Their study demonstrated that the biological process effectively removed several anionic surfactants (such as linear alkylbenzene sulfonate), as their structure can be better attacked by microorganisms than by non-ionic surfactants [43]. It is recommended that further research be conducted to explore the techno-economic feasibility of CWW treatment via such physical–chemical–biological hybrid systems.

Table 2. Common CWW treatment technologies reported in the literature.

CWW Treatment Method	Operational Factors	Removal Efficiency (%)				Reference
		COD	Oil and Grease	Surfactants	TDS/TSS	
Coagulation/flocculation by Poly-Aluminium Chloride (PACl)	100 mg/L Na-bentonite, 20 mg/L Al^{3+}, and 0.5 mg/L anionic polyelectrolyte	59.0	85.0	–	–	[7]
Coagulation/flocculation by alum	12.5 mL 10% Alum, and 10 mL 5% PACl per 1 L CWW	67.4	–	–	97.9 (TSS)	[15]
Coagulation/flocculation by synthesized alum from bauxite waste	90 mg/L alum, 200 rpm@2 min, 25 rpm@5 min, and 34 min sedimentation	75.0	–	34.0	–	[44]
Oxidation by electro-Fenton (EF)	75.8 min, 58.8 mA/cm^2, pH 3.02, 1.62 mL/L H_2O_2/CWW, and 3.66 H_2O_2/Fe^{2+}	68.7	–	73.6	71.8 (TSS)	[3]
Oxidation by electrooxidation with H_2O_2 generation	0.5 mM Fe^{2+}, pH 3, and 500 mA	96.0	96.0	–	–	[31]
Oxidation by Fenton	pH 3.0, and 1 h	83.3–83.9	88.5–89.0	94.1–95.2	–	[45]
Oxidation by photo-Fenton	two 40 W UVA radiation, pH 3.0, and 1 h	92.3–93.9	98.9–99.6	100	–	[45]
Adsorption by macro-composite	10 mL/min flowrate, 38.2 min, and 0.1 cm/min surface loading rate	88.0	–	–	92.3 (TSS)	[33]
Membrane ultra-filtration	1 bar pressure, and 2.69 L/m^2/h	95.0	100	–	–	[41]
Membrane nano-filtration	3 bar pressure, and 58.5 L/m^2/h	70.9–91.5	–	–	60.0–61.5 (TDS)	[46]
Membrane ultra-filtration	3 bar pressure, and 58.5 L/m^2/h	54.9–83.9	–	–	17.6–31.5 (TDS)	[46]
Biological up-flow anaerobic sludge blanket (UASB) reactor	4 d hydraulic retention time	96.0	96.8	–	11.0 (TDS)	[47]
Biological anoxic - aerobic sequential reactor	24 h hydraulic retention time	94.0	–	–	–	[1]
Hybrid coagulation/flocculation + Adsorption	220 mg/L coagulant, and 2 h sorption	92.6	–	97.2	35.6 (TDS)	[34]
Hybrid bioreactor + UV Lamp + membrane filtration	6.6 J/cm^2 UV dosage, 10.13 L/m^2/h flux, 50.8 kPa, and 94 h	99.9	80.0	99.9	100 (TSS); 25.6 (TDS)	[48]
Hybrid flocculation-column flotation + sand filtration + chlorination	0.5 mg Cl_2/L	62.8	27.3	42.9	91.0 (TSS)	[4]
Hybrid aeration + coagulation/flocculation + oxidation	90 min aeration, 80 mg/L alum, and 2.5 mL/L waste H_2O_2	93.0	96.3	–	14.0 (TDS)	[17]

Table 2. *Cont.*

CWW Treatment Method	Operational Factors	Removal Efficiency (%)				Reference
		COD	Oil and Grease	Surfactants	TDS/TSS	
Hybrid coagulation/flocculation + sand filtration + ceramic ultrafiltration (UF) + reverse osmosis (RO)	45 mg/L $FeCl_3$ coagulant, 250 kPa UF, and1000 kPa RO	96.0	–	–	100 (TSS); 42.5 (TDS)	[16]
Hybrid electrocoagulation + electrooxidation	Fe electrodes, 25 mA/cm^2, pH 5, and 120 min	90.2	–	–	80.7 (TSS)	[29]
Hybrid electrocoagulation + adsorption	Al electrodes, and 22.5 g/L coagulant	99.1	100	–	95.9 (TSS)	[30]
Hybrid coagulation + flotation + ultrafiltration	150 mg/L coagulant, 0.2 m^3/h, 1:9 gas-liquid ratio, and 0.3–0.4 MPa	–	40.0	–	–	[39]
Hybrid electrocoagulation and nanofiltration	25 °C, 250 rpm, and parallel connection of monopolar electrodes	88.0	90.0	91.0	99.0 (TSS)	[49]
Hybrid coagulation/flocculation followed by sedimentation + sand filtration + ceramic membrane filtration	300 rpm @ 1 min (coagulation), 30 rpm @ 20 min, 30 min sedimentation, 3.5 m/h in filter, and 2 bar in membrane	78.3–79.8	–	–	14.5 (TDS); 100 (TSS)	[28]

3.6. Research Trends and Hotspots in Carwash Wastewater Management

Figure 5 (created using VOSviewer) shows a map of six clusters distinguished by colors, representing the keyword co-occurrence network of the bibliometric data. In this network, the size of bubbles increases with a higher occurrence of the number of keywords in research articles, whereas a line connecting the keywords increases in thickness for strong relationships.

Cluster 1 (red) represents the major pollutants present in CWW, such as anionic surfactants (representing the main organic constituent of CWW), oil, and grease. These elements can be introduced into CWW from waste lubrication oil from the engine, surfactant gasoline additives, and grease inside the oil distribution board [47]. Surfactants and oil receive special attention from researchers because they are resistant to most treatment technologies and toxic to the environment when discharged without proper treatment. For example, oil contaminants can reach the aquatic environment via CWW discharge, cover the gills of fish, and interfere with oxygenation levels [5]. In addition, the discharge of surfactants into water bodies can form a foamy layer on the surface that prevents oxygen transfer from the surrounding environment, leading to fish death due to septic conditions [44].

Cluster 2 (green) represents other components in CWW, such as COD and TSS. These parameters are enriched in the effluents of vehicle service stations, representing their soluble, particulate, and/or colloidal organic contents [33]. Colloidal particles can reach sewage drains through CWW along with cleaning street waste (e.g., sand, dust, and mud). Cluster 2 has a connection with the keyword "pollution", as these elements cause pollution to water resources, aquatic species, atmosphere, and crops.

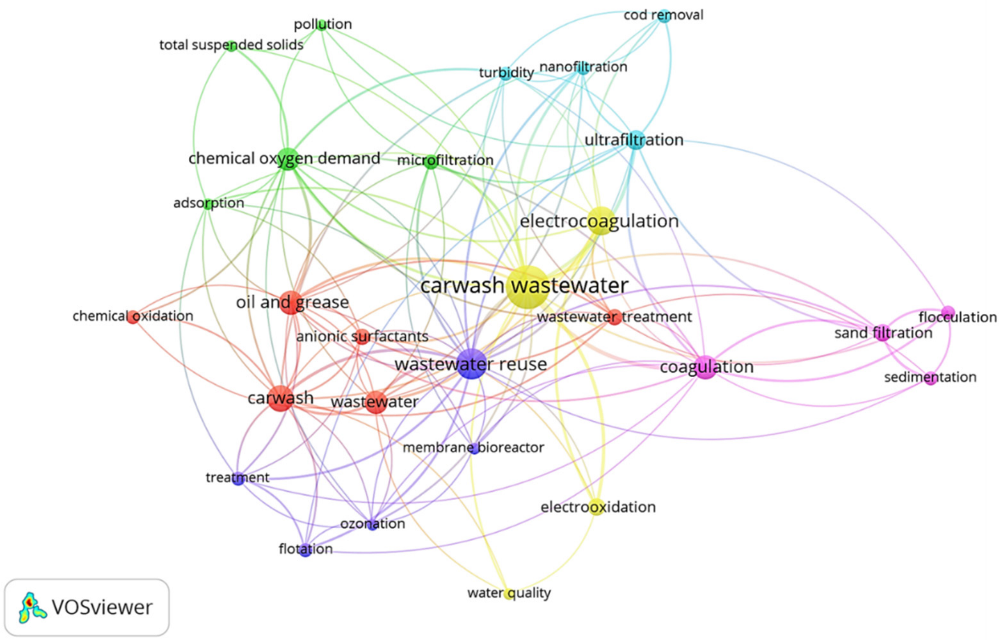

Figure 5. Co-occurrence network of top 28 keywords with at least three occurrences using search strings relevant to "Carwash" AND "Wastewater" AND "Treatment" in various online databases, including Scopus, Google Scholar, Wiley Online Library, and Web of Science during 2010–2020.

Cluster 3 (blue) shows that various researchers have endeavoured to explore the appropriate technologies for removing contaminants from CWW. These subjects include "flotation", "ozonation", and "membrane bioreactor", which comprise physical and biochemical mechanisms for CWW treatment [46]. The interaction between these keywords indicates that the conventional biological-based methods are not always preferable to treat wastewater containing oily materials. Several researchers have employed bioreactors coupled with membrane separation processes to reduce oil and grease concentrations in wastewater streams; however, fouling remains the major limiting factor in the membrane lifetime [14]. Although ozonation has been widely used to degrade toxic organic pollutants, additional investigations are required in order to eliminate the formation of undesirable byproducts associated with the oxidation reactions [17].

Cluster 4 (yellow) is relevant to the electrochemical techniques of CWW treatment, including electrooxidation and electrocoagulation processes. Electrooxidation technology has attracted the attention of researchers mostly due to its effectiveness in degrading different organic pollutants resulting from many industrial processes, such as olive oil, tannery, textile, and pulp and paper effluents. Moreover, various authors have paid increased attention to the application of electrocoagulation for treating heavily polluted effluents that contain oil, grease, and metals. The electrochemical treatment of surfactant-containing wastewater can be achieved via two steps: (i) conversion of non-biodegradable compounds into biodegradable matter by electrochemical conversion, and (ii) mineralization of organic contaminants by electrochemical combustion [43]. These mechanisms have shown better results than biological treatment processes, which require longer retention times and can generate huge quantities of byproducts from bacterial activities and metabolism.

Keywords of cluster 5 (purple) depict the common physicochemical and hybrid systems adopted by several authors to treat CWW [48]. These unit processes include coagulation, flocculation, sedimentation, and sand filtration, depending on the solid/liquid or liquid/oil/water separation performances. These hybrid systems for CWW treatment have found promising and feasible applications for oil, COD, and turbidity removal to meet strict effluent guidelines.

Cluster 6 (bright blue) indicates that nano- and ultra- filtrations are among the types of membrane-based technologies that have attracted much research attention for their possible applications in CWW treatment. These filtration methods have been employed as a secondary treatment stage to eliminate the remaining portions of COD and turbidity after the chemical and/or physicochemical processes [16]. Although various researchers have applied these filtration-related techniques, fouling is considered a major challenge during operation, causing flux reduction and low productivity. Hence, the number of studies relevant to membrane operation and frequent cleaning using low energy consumption is increasing worldwide.

3.7. Outlook for Achieving SDGs in Carwash Wastewater Management at Field Scale

Although CWW contains various organic and inorganic pollutants, its management has attracted several researchers worldwide seeking to achieve sustainable development [34]. The development objectives and targets associated with CWW treatment and reuse can be illustrated according to the three pillars of sustainability (Figure 6). This section represents the environmental, economic, and social SDGs retrieved from CWW management for providing the appropriate policy, investment, and management decisions.

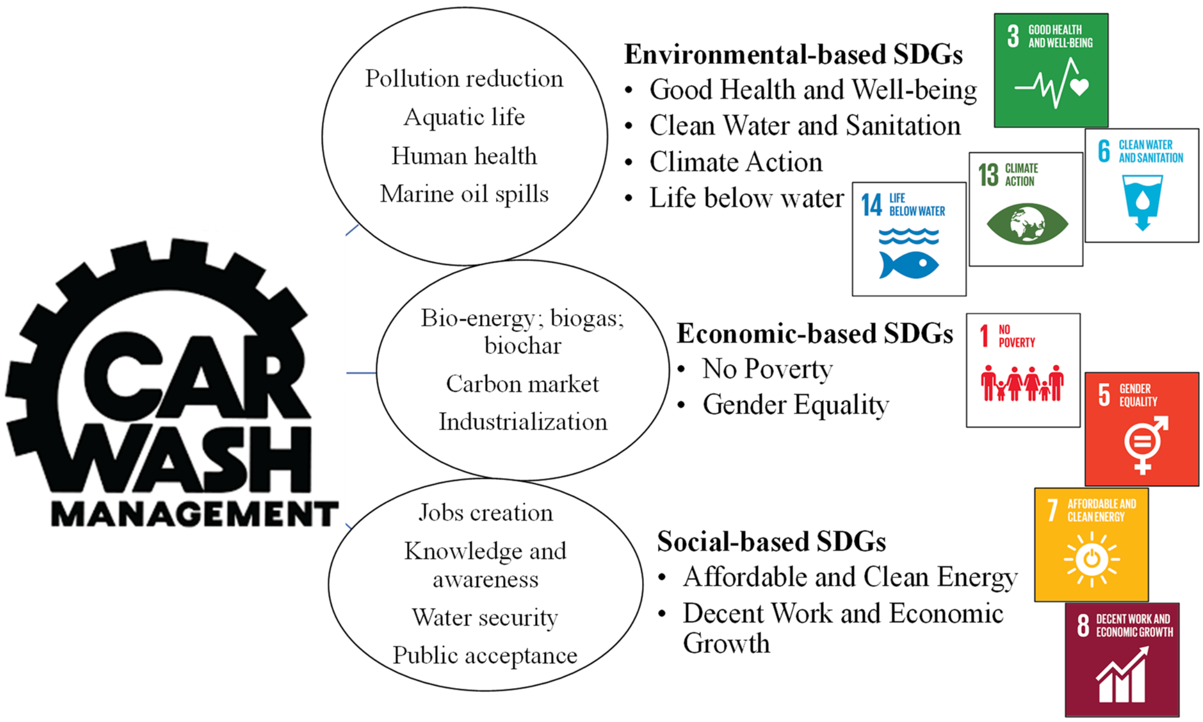

Figure 6. Environmental, economic, and social SDGs related to CWW management.

3.7.1. Environmental-Related SDGs

The current study illustrates the common treatment methods used to reduce the pollution load associated with CWW effluent before reaching the aquatic environment. The removal of oily compounds from wastewater is essential to protecting the environment and avoiding deterioration in the physicochemical properties of the receiving soil and water. This pattern has a direct correlation to SDG3, "Good Health and Well-being", as human exposure to untreated CWW can be detrimental to public health. As such, various diseases such as headaches, high blood pressure, anemia, gastric disturbances, nerve damage, cardiac arrhythmia, and cancer (stomach and lungs) accompanying the uncontrolled disposal of CWW would be minimized. Moreover, various researchers have employed advanced and hybrid systems for CWW treatment in order to produce a final effluent that complies with wastewater reuse and disposal standards. For instance, treated CWW can be reused to wash and clean cars, reducing the amount of fresh water utilized in automobile service centres. This pattern can partially meet Goal 6, "*Clean Water and Sanitation*", through (i) increasing recycling and reuse to support water security objectives, (ii) reducing pollution and minimizing dumping of hazardous compounds, and (iii) safeguarding aquatic life and protecting water-associated ecosystems. In parallel, the treatment of CWW minimizes accidental oil spills in the marine environment resulting from sewer effluent discharge into coastal ocean waters. This would ensure the conservation of watercourses, maintain sustainable fishing, and attain productive oceans, meeting SDG 14, "*Life below water*".

3.7.2. Economic-Related SDGs

The treatment of CWW by the biological processes can generate valuable products that have potential economic benefits. For instance, Li et al. [19] found that the anaerobic co-digestion of wastewater with fat, oil, and grease enhanced biogas production to 25.14 L/d with 70.2% methane content. Moreover, El Hanandeh et al. [10] prepared a biochar material via biomass pyrolysis containing oil products at 550 °C to achieve cleaner production and economic value. These byproducts (e.g., biogas, polyhydroxyalkanoates, and biochar) can be further used to upgrade energy services, meeting SDG7, "*Affordable and Clean Energy*". Moreover, previous studies have revealed the application of biochar as a charred carbon-enriched material to enhance the properties of agricultural soils [21]. Improving the physicochemical features of soil is essential to achieving a sustainable agriculture framework and attaining food security, complying with the targets of SDG2, "*Zero Hunger*". Hence, additional studies are essential to characterize biochar obtained from CWW treatment sludge for further application in the agricultural sector.

Governments can issue licences, permission, and approval to initiate carwash businesses based on information on the water sources used for washing and the presence of wastewater treatment facilities. Implementing suitable treatment technologies for CWW would facilitate the establishment of carwash investment projects, having a direct correlation to job creation and productive employment. Accordingly, governments and stakeholders should encourage the establishment of small-scale businesses relevant to automobile cleaning services with a focus on water usage quantities and characteristics. This pattern would elevate the number of employees working in the carwash sector, meeting SDG8, "*Decent Work and Economic Growth*".

3.7.3. Social-Related SDGs

Public opinion and knowledge about CWW management should be fostered to assist policymakers and stakeholders in implementing sensible regulations that control water usage and treatment in carsharing and personal vehicle services. Educational institutes should be encouraged to include training programs that enhance social awareness of the sustainability of CWW management systems regarding their environmental and economic benefits. This CWW management concept would have social acceptance through offering job opportunities, protecting environmental systems, and obtaining products with essential economic values. Hence, the quality of education should be improved to increase the public skills and knowledge needed to endorse sustainable development through CWW management, obeying SDG 4, "*Quality Education*".

3.8. Future Perspectives in Carwash Management

Based on our illustration of CWW composition and treatment technologies and performance along with the associated environmental and sustainability impacts, future studies related to CWW management should focus on:

- Integrating the "circular economy" concept into CWW management by boosting reuse and waste minimization scenarios.
- Providing appropriate analytical techniques and holistic tools to examine the cost of CWW treatment over various steps involving implementation, operation, and maintenance.
- Selecting suitable CWW treatment technologies that discharge no liquid effluent into surface waters, supporting efficient recycling and reuse.
- Raising environmental awareness in the population regarding the reuse of treated water in order to avoid water resource depletion and aquatic pollution.

- Applying internet of things, cyber-physical systems, and machine learning towards determining optimal CWW treatment techniques.
- Establishing dynamic models to optimize the performance of membrane systems and predict fouling issues due to the deposition of solids and oily particulates.
- Studying pilot and large-scale systems for CWW treatment representing real processes and environmental conditions.
- Encouraging the stakeholders, policymakers, and both the public and private sectors to invest in CWW treatment systems for pollution reduction and resource recovery and reuse.

4. Conclusions

Due to the severe environmental pollution associated with the direct disposal of CWW into the aquatic ecosystem, it is essential to determine appropriate treatment technologies that could maintain the major aspects of sustainable development. Here, we have elucidated the relevant SDGs using a keyword co-occurrence network of a bibliometric survey with the search strings “Carwash” AND “Wastewater” AND “Treatment”. This study has revealed for the first time the various physical, chemical, and biological processes used to treat CWW while meeting the three pillars of sustainability. A biological process combined with post-treatment (i.e., a hybrid system) showed an appropriate technique for CWW treatment with the dual benefits of pollution reduction and bioenergy generation. The treatment of CWW is interconnected to (i) environmental-related SDGs (protect soil and water, promote human health and well-being, safeguard terrestrial and aquatic systems, support water reuse schemes), (ii) economic-related SDGs (biogas/biochar production, establish carwash investment projects, create new jobs), and (iii) social-related SDGs (enhance the social awareness about wastewater reuse, improve public skills and knowledge about the sustainability of wastewater treatment). These study outputs should encourage policymakers and stakeholders to implement sensible regulations for managing CWW treatment and reuse in order to meet the SDGs of the 2030 Agenda. Further studies are essential in order to minimize the capital and operational costs of CWW treatment and to examine the implementation of risk assessment, especially in developing countries suffering from water stress.

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