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Biological treatment of oil and gas produced water: a review and meta-analysis

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Abstract

Biological treatment is effective but infrequently used for oil and gas produced water. To date, physical-chemical treatment methods have been favored due to the smaller space requirements and operational simplicity. Changing regulatory requirements and increased interest in recycling and beneficial reuse have led to increased interest in biological treatment. To elucidate its potential role, we reviewed and summarized 59 studies on the biological treatment of produced water. Oilfield produced water was predominantly studied (> 50%). More studies using real produced water were from China than from any other country (37%). Real produced water was used in most studies (73%). Studies were predominantly bench-scale experiments (69%). Fixed-film reactors were most prevalent (27%). Water quality of produced waters treated was variable; median total dissolved solids (TDS) was 28,000 mg L⁻¹ and median chemical oxygen demand (COD) was 1125 mg L⁻¹. Inhibition by salinity was variable according to the treatment system and study design, but efficacy generally decreased when TDS was above 50,000 mg L⁻¹. For studies treating real samples, average COD removal was 73% when TDS was less than 50,000 mg L⁻¹, and 54% when TDS was greater than 50,000 mg L⁻¹. Key issues were microbial acclimation, toxicity, biological fouling, and mineral scaling. Finding an inoculum was not problematic as microorganisms capable of degrading hydrocarbons were isolated from various environments. Treatment performance was better where synthetic produced water was used in lieu of real samples. Biological treatment is promising for producing effluents suitable for reuse, particularly where it functions as part of a larger treatment train.

Introduction

Oil and gas production, particularly in arid regions, introduces a challenge for water management (Clark and Veil 2009; Guerra et al. 2011). Water must be sourced for production. Produced water generated during production must be handled, treated, and disposed. The large volumes of produced water generated make this a significant environmental problem. In the USA alone, the total volume of produced water in 2012 was estimated as 3.4 billion m³ (Veil 2015). To date, injection of produced water has been the primary disposal method, often done to increase oil production (Veil 2015). Decreased availability of injection sites, changing regulations, and challenges associated with locating fresh water, have led the oil and gas industry to consider new strategies to treat and reuse produced water (Dahm 2014; Guerra et al. 2011; Heberger and Donnelly 2015). Recycling produced water for subsequent oil and gas production solves both sourcing

and disposal problems simultaneously. Beneficial reuse outside of oil and gas production solves the disposal dilemma and creates a needed water resource. Possible beneficial uses include irrigation, livestock watering, aquifer storage, streamflow augmentation, and municipal/industrials uses (Dahm 2014; Guerra et al. 2011; Heberger and Donnelly 2015; Interstate Oil and Gas Compact Commission and ALL Consulting 2006; Veil et al. 2004).

The desire to recycle and reuse produced water has led to increased interest in its treatment. Previous publications have reviewed existing produced water treatment technologies (Arthur et al. 2005; Dahm and Chapman 2014; Fakhru'l-Razi et al. 2009; Guerra et al. 2011; Hansen and Davies 1994; Igunnu and Chen 2012; Jimenez et al. 2018; Robinson 2013a, b, c). Most reviews have focused on physical-chemical treatment processes that dominate existing treatment trains. Physical-chemical treatment has been historically preferred because of the small footprint of facilities and the straightforward process control strategy; however, disadvantages include high capital and operating costs, production of hazardous sludges and brine solutions, and difficulties in removing trace contaminants (Fakhru'l-Razi et al. 2009). The high oxygen demand of produced water poses challenges for implementation of membrane technologies—that are advantageous for removing a range of contaminants—due to the potential for biological fouling and mineral scaling (Mondal et al. 2008; Xu et al. 2008).

Biological treatment has a long history of use in industrial waste treatment because of its ability in reducing oxygen demand, nutrients, metals, and trace organic contaminants (Tchobanoglous et al. 2014). Biological treatment can be used to remove specific contaminants of concern, such as arsenic that is elevated in some groundwaters (Katsoyiannis and Zouboulis 2004). Biological treatment can also function under extreme conditions of temperature, pH, and salinity (e.g., Margesin and Schinner 2001). Prior reviews have examined the biological treatment of oily wastewaters, including those from refineries and the shipping industry (Jamaly et al. 2015; Yu et al. 2017), and the treatment of salty wastewaters, including those from the agro-food industry, textiles dyeing, and tanneries (Castillo-Carvajal et al. 2014; Lefebvre and Moletta 2006; Xiao and Roberts 2010). In the previous reviews, treatment of produced water has been aggregated with other types of oil and salty wastewaters. To our knowledge, there are no review papers that solely evaluate the biological treatment of produced water generated from oil and gas production.

In this paper, we summarize peer-reviewed publications on the biological treatment of produced water and provide a meta-analysis of the data published. The summary includes information for fixed-film treatment, membrane bioreactors, wetlands and ponds, activated sludge treatment, anaerobic treatment, and bio-electrochemical treatment. We address questions regarding optimal reactor configuration, selection of process control parameters, impact of salinity, pretreatments, the need for specialized microbial consortia, and potential technological issues. Our goal

is to provide information useful for advancing in-industry recycling and beneficial reuse of treated effluents by including biological treatment into treatment trains.

Literature review methodology

Studies were located using the Web of Science database to search for peer-reviewed articles with various combinations of the following search words: produced water, oil and gas, biological treatment, and wetlands. Forward and backward searches of the selected articles were also done. Only studies of the biological treatment of produced water and flowback were included. Studies that generally addressed biological treatment of oily and salty water were not included, nor were studies of the treatment of refinery wastewaters. Studies were included if the water studied mimicked produced water. Only studies of biological treatment systems were included—biodegradation studies (bottle tests) were not included in the quantitative analyses although these studies are discussed as they support advancement of biological treatment.

Literature review summary

Fifty-nine studies of the biological treatment of produced water were reviewed (Table S1). The studies were published from 1979 to 2018. Various project motivations were identified by the study authors, although environmental concern was the most prevalent (Table 1). In most studies (51%), authors cited more than one motivation for completing their work. It is clear from the studies that researchers are interested in beneficial reuse and recycling of produced water effluent and that environmental regulations are influential in developing new treatment strategies. In 32% of studies, the benefits of biological treatment are cited as project motivations due to the relatively low cost and low impact compared with other technologies.

Table 1

Motivations for the studies reviewed, as described by the authors

Number of studies ^a	Stated project motivations
30 (51%)	Concern over environmental impacts
19 (32%)	Expanding opportunities for beneficial reuse and/or recycling
19 (32%)	Interest in biological treatment as a low-cost and low-impact approach
19 (32%)	Trying to meet environmental regulations
7 (12%)	Limited availability of injection wells and/or cost of injection
10 (17%)	No statements regarding study motivation

^aIn some studies multiple project motivations were described, so percentages do not add to 100

Real produced water was used in 43 of the studies (73%), with both real and synthetic produced water being used in four of these 43 studies. The real produced water originated from ten countries (Fig. S1). Of the 43 studies that used real produced water, 16 of these used produced waters from China (37%) while 12 originated from the USA (28%). Synthetic produced waters were composed of minerals, oil, and sometimes production chemicals such as surfactants. In some cases, synthetic produced water was formulated to mimic produced water from a specific region, such as Middle East oilfields (Shpiner et al. 2009a, b) or produced water from Africa where salinity is low and has total dissolved solids (TDS) of 704–1370 mg L⁻¹ (Horner et al. 2012). In one study, removal of the surfactant dodecyl benzene sulfonic acid (DBSA), an oilfield production chemical, was observed in the produced water from an oilfield where polymer flooding was practiced (Zhang et al. 2016). In another, the surfactant nonyl phenol glycol ether was added to synthetic produced water to observe its removal (Scholzy and Fuchs 2000). Most produced water samples originated from oilfields although a few were from gas production wells; produced water samples originated from both onshore and offshore oilfields (Table S1).

Most of the studies reviewed used a bench-scale apparatus (69%), meaning that small reactors were used (no larger than several liters capacity) and these reactors were operated in the laboratory. Pilot-scale studies were considered to be those that were larger in scale (e.g., over 100 L capacity reactors) and/or those done at a field site or outdoors (27%). Wetland studies were done in large totes placed outdoors so these were considered pilot studies (based on size) although synthetic produced waters were used in these studies. Observations were made on full-scale operational facilities (Lu et al. 2006; Wang et al. 2007), and in one study, both bench-scale and pilot-scale systems were observed (Kwon et al. 2011).

The most commonly used treatment configuration was fixed-film treatment (32%) (Fig. 1). The fixed-film reactors included tanks filled with high-surface area media, granular activated carbon filters, rotating biological contactors, and aerobic filters. The second most commonly used biological treatment method was the MBR (20%). Constructed wetlands and ponds, including free surface and subsurface flow wetlands, were used in 17% of studies. Each of the studies reviewed was characterized by the predominant treatment type under study (Fig. 1), although there was some ambiguity in characterization. One of the anaerobic systems could have also been classified as a sequencing batch reactor (Li et al. 2010), while three other studies of anaerobic treatment used fixed-film media (Ghorbanian et al. 2014; Khong et al. 2012; Liu et al. 2013).

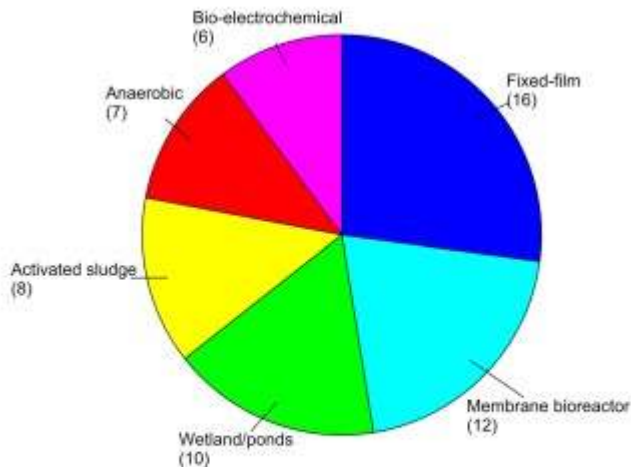


Fig. 1 Types of biological treatment used in the studies reviewed; three of the anaerobic treatment studies also used fixed-film media in their reactors

In the studies reviewed, pretreatments were commonly used. At a minimum, oil-water separation was often done and sometimes pretreatment included air flotation or stripping or other technologies. Removal of floating oil is recommended because it can interfere with aeration and biological processes (Robinson 2013b). Many researchers obtained their samples after oil-water separation at a full-scale facility (Table S1). Anaerobic pretreatment, with fixed-film media to retain biomass, was used in some studies to hydrolyze organic matter (termed hydrolysis acidification) and make it more amenable to treatment (Guo et al. 2014; Li et al. 2010; Lu et al. 2009; Su et al. 2007, 2009; Wang et al. 2007; Zhang et al. 2010, 2016). Increased biodegradability and removal of recalcitrant organic compounds was observed in the anaerobic reactors (Li et al. 2010; Liu et al. 2013; Su et al. 2009). In one study, adsorption by surfactant modified zeolite was used as a pretreatment to reduce benzene, toluene, ethylbenzene, and xylene (BTEX) prior to biological treatment (Kwon et al. 2011). Nutrient limitations were addressed in many of the studies with nutrients added to support microbial growth in biological reactors (Ji et al. 2009; Liu et al. 2013; Lu et al. 2009; Piubeli et al. 2012; Tellez et al. 2005; Tong et al. 2013). Lu et al. (2009) used commercial maize powder to supplement carbon for enhanced reduction of ammonia and other contaminants. In a bottle test study of produced water, COD removal increased from 20% without nutrients to 65–80% with addition of phosphorus and carbon substrates, demonstrating the importance of nutrient addition (Piubeli et al. 2012).

In some studies, temperature stabilization was necessary prior to biological treatment. In two related studies, produced water temperature (60 °C) was reduced in stabilization ponds prior to wetland treatment (Ji et al. 2002, 2007). Lu et al. (2009) studied a treatment facility in China (Shengli Oilfield) where the produced water temperature was 55 °C and the ambient temperature varied during the study from – 15 to – 5 °C. Lu et al. (2006)

used influent produced water in a heat exchange system to maintain the reactor temperature.

Total dissolved solids (TDS) and COD of the real and synthetic produced water samples varied among the studies (Fig. 2). The mean \pm standard deviation of TDS of samples treated—in the 50 studies where TDS was reported—was $64,118 \pm 76,024 \text{ mg L}^{-1}$ (median = $28,000 \text{ mg L}^{-1}$). The mean \pm standard deviation of COD of samples treated—in the 46 studies where COD was reported—was $1397 \pm 1270 \text{ mg L}^{-1}$ (median = 1125 mg L^{-1}). Note that in some studies more than one sample was treated and these samples are handled separately within the meta-analysis performed herein. Where conductance was reported in lieu of TDS, the relationship $\text{SpC} = 1.6 \cdot \text{TDS}$ was used to calculate TDS. In some studies, produced water TDS was increased by adding sodium chloride in order to observe its impact (Fakhru'l-Razi et al. 2010; Pendashteh et al. 2010, 2012; Sharghi et al. 2014). If only real produced water samples are considered—and samples where sodium chloride was added to increase salinity are excluded—the TDS of samples was $28,702 \pm 40,383 \text{ mg L}^{-1}$ (median = $16,135 \text{ mg L}^{-1}$) and the COD was $1154 \pm 1254 \text{ mg L}^{-1}$ (median = 727 mg L^{-1}). Seventeen of the total samples treated had $\text{TDS} \leq 10,000 \text{ mg L}^{-1}$ and 49 had $\text{TDS} \leq 40,000 \text{ mg L}^{-1}$ suggesting that a large portion of the produced water tested could be treated and potentially reused (Guerra et al. 2011).

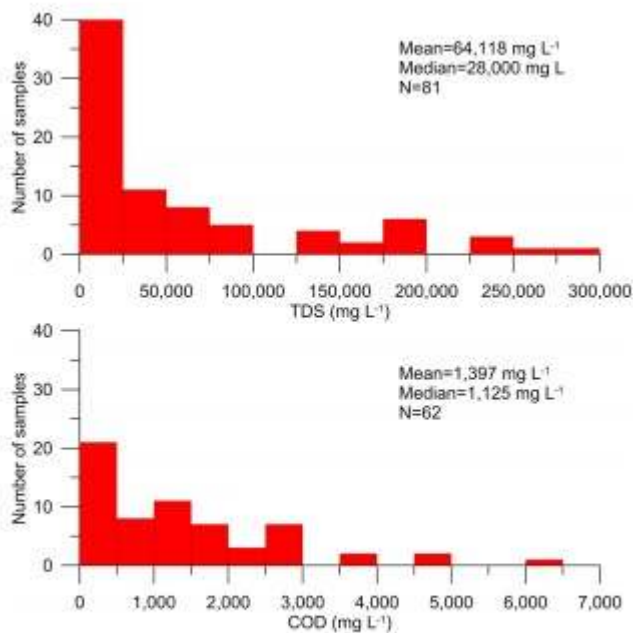


Fig. 2 Histogram of total dissolved solids (TDS) and chemical oxygen demand (COD) of the produced water treated

Treatment efficacy using different reactor configurations
Fixed-film treatment

Fixed-film treatment was the most commonly used treatment method, used aerobically in 16 studies (Table 2). Fixed-film reactors are favored in many industrial treatment processes because biofilm-bound microorganisms are retained and resistant to extreme conditions and shock loadings (e.g., extreme pH, high salinity, toxicity) (Gavrilescu and Macoveanu 2000).

Table 2 Summary of fixed-film treatment studies for produced water

Reference	Sample type	TDS (mg/L)	COD (mg/L)	Pretreatments	Biological reactor			Key results
					Process	Size (L)	Parameters	
Freedman et al. (2017)	Real	10,460, 18,170, 14,230 (Piceance PW, Denver-Julesburg PW, FFB)	770, 1080, 6360 (Piceance PW, D-J PW, D-J FFB)	Coagulation-flocculation, sedimentation	Biological GAC columns	1.27 cm dia., 30 cm GAC (bench-scale), 5.1 cm dia., 76 cm GAC (lab-scale)	EBCT = 3 min (bench-scale), 5–60 min (lab-scale)	78% COD removal (Piceance PW), 82% COD removal (D-J PW)
Riley et al. (2016)	Real	12,615, 14,569, 31,193 (Piceance PW, D-J PW, D-J FFB)	85, 1157, 2872 (Piceance PW, D-J PW, D-J FFB)		Biological GAC columns, UF, NF	5 cm dia., 76–304 cm GAC	HRT = 3.7 h (Piceance PW), 7.4 h (DJ PW and FFB), HLR = 2.44 m h ⁻¹	72% COD removal (Piceance PW), 89% COD removal (D-J PW), 90% COD removal (D-J FFB)
Zhang et al. (2016)	Real	3075	283	Flocculation, air flotation	HA, BAF	4200	HRT = 33.6 h, OLR = 0.249 kg COD m ⁻³ d ⁻¹	75% COD removal
Guo et al. (2014)	Real	6920	303		HA, sedimentation, BAF, biological GAC columns	4200 (BAF), 76 (GAC)	HRT = 36 h (BAF), 1.5–3.6 h (GAC), OLR = 0.14 kg COD m ⁻³ d ⁻¹ (HA), 0.06 kg COD m ⁻³ d ⁻¹ (BAF), 0.56 kg COD m ⁻³ d ⁻¹ (GAC)	60% COD removal after BAF, 80% COD removal after GAC
Tong et al. (2013)	Real	2696	209		CAS, BAF	10,000 (aeration), 27,000 (BAF)		64% COD removal (HRT = 18 h)
Dong et al. (2011)	Real	5090	354	OWS	Moving bed biofilm reactor w/suspended media	5	HRT = 10–36 h, OLR = 1.17–4.21 kg COD m ⁻³ d ⁻¹	79% COD removal (HRT = 18 h)
Lu et al. (2009)	Real	46,530	345	Sedimentation	HA, sedimentation, BAF	4200		64% COD removal (HRT = 32 h, 0.28 kg COD m ⁻³ d ⁻¹)
Su et al. (2009)	Real	25,000	330	OWS	Anaerobic baffled reactor, BAF	2900		58–75% COD removal
Chavan and Mukherji (2008)	Synthetic	–	4513 (0.6% diesel)		RBC	4	HRT = 21 h, OLR = 27.3 g TPH m ⁻² d ⁻¹	97.8% COD removal, 99.4% TPH removal

Table 2 (continued)

Reference	Sample type	TDS (mg/L)	COD (mg/L)	Pretreatments	Biological reactor			Key results
					Process	Size (L)	Parameters	
Su et al. (2007)	Real	25,000	340	OWS	Anaerobic baffled reactor, BAF, sedimentation	2900	HRT = 11.4, 16, and 26.7 h	75% COD removal (HRT = 26.7 h), 58% COD removal (HRT = 11.4 h)
Wang et al. (2007)	Real	5560	470		Anaerobic/aerobic reactors with fixed media	2,970,000	HRT = 12 h (each), OLR = 0.72–0.75 kg m ⁻³ d ⁻¹	86% COD removal, 97% BTEX removal, 90% TPH removal
Lu et al. (2006)	Real	–	286	OWS, flotation	BAF	–		95% O&G COD removal, 62% TSS COD removal
Zhao et al. (2006)	Real	Conductance = 8700 uS cm ⁻¹	124		BAF	1.57	HRT = 4–20 h, OLR = 1.07 kg COD m ⁻³ d ⁻¹	64–78% TOC removal, 84–90% PAH removal
Campos et al. (2002)	Real	80,470	1622	Coarse filtration, MF	BAF	1	HRT = 12, 24, and 48 h	65% COD removal (HRT = 12 h)
Woolard and Irvine (1994)	Synthetic	152,225	–		Biofilm SBR	1	HRT = 48 h, cycle time = 24 h	> 99% phenol removal
Palmer et al. (1981)	Real	20,000, 28,000 (Carpinteria, Huntington Beach)	–	OWS, air flotation	RBC	6057	HRT = 2 d, HLR = 0.041 m d ⁻¹ (Carpinteria), 0.014 m d ⁻¹ (Huntington Beach)	> 94% BOD5 removal (Carpinteria); 72% BOD5 removal, 24% COD removal (Huntington Beach)

BAF biological aerated filter, BOD5 biochemical oxygen demand, 5 days, BTEX benzene, toluene, ethylbenzene, and xylene, CAS conventional activated sludge, COD chemical oxygen demand, EBCT empty bed contact time, FFB fracturing flowback, GAC granular activated carbon, HA hydrolysis acidification, HLR hydraulic loading rate, HRT hydraulic retention time, MF microfiltration, NF nanofiltration, O&G oil and grease, OLR organic loading rate, OWS oil–water separator, PAC powdered activated carbon, PAH polycyclic aromatic hydrocarbons, PW produced water, RBC rotating biological contactor, SBR sequencing batch reactor, TDS total dissolved solids, TOC total organic carbon, TPH total petroleum hydrocarbons, TSS total suspended solids, UF ultrafiltration

BAF biological aerated filter, *BOD5* biochemical oxygen demand, 5 days, *BTEX* benzene, toluene, ethylbenzene, and xylene, *CAS* conventional activated sludge, *COD* chemical oxygen demand, *EBCT* empty bed contact time, *FFB* fracturing flowback, *GAC* granular activated carbon, *HA* hydrolysis acidification, *HLR* hydraulic loading rate, *HRT* hydraulic retention time, *MF* microfiltration, *NF* nanofiltration, *O&G* oil and grease, *OLR* organic loading rate, *OWS* oil-water separator, *PAC* powdered activated carbon, *PAH* polycyclic aromatic hydrocarbons, *PW* produced water, *RBC* rotating biological contactor, *SBR* sequencing batch reactor, *TDS* total dissolved solids, *TOC* total organic carbon, *TPH* total petroleum hydrocarbons, *TSS* total suspended solids, *UF* ultrafiltration

Most fixed-film treatment systems used packed media within an aerated reactor (Campos et al. 2002; Dong et al. 2011; Guo et al. 2014; Liu et al. 2013; Lu et al. 2006, 2009; Su et al. 2007, 2009; Tong et al. 2013; Wang et al. 2007; Woolard and Irvine 1994; Zhang et al. 2016; Zhao et al. 2006). In some studies, an airlift aeration configuration was used where compressed air was introduced at the bottom of the tank in tubes and was directed into a circulating vertical pattern (Campos et al. 2002; Guo et al. 2014; Zhang et al. 2016). Other fixed-film treatment systems consisted of biologically active carbon filters (Freedman et al. 2017; Guo et al. 2014; Riley et al. 2016), rotating biological contactors (RBC) (Chavan and Mukherji 2008; Palmer et al. 1981), a biofilm sequencing batch reactor (SBR) (Woolard and Irvine 1994), and an anaerobic reactor filled with media (Ghorbanian et al. 2014; Khong et al. 2012). Ghorbanian et al. (2014) compared the performance of an anaerobic upflow fixed-bed reactor with that of an anaerobic SBR, and found that the upflow reactor with fixed-film media was more efficient in removing aromatic compounds.

The process control parameters used in the studies indicate some variability (Table 2). The hydraulic retention time (HRT) in fixed-film reactors varied from four to 48 h except in biologically active granular activated carbon (GAC) column where the HRT was lower (Freedman et al. 2017; Riley et al. 2016). Organic loading rates (OLR) varied: Guo et al. (2014) reported 0.14 kg COD m⁻³ d⁻¹ while Dong et al. (2011) used an OLR as high as 4.21 kg COD m⁻³ d⁻¹. In most studies, researchers used COD as one of the performance metrics, which made it possible to compare the studies. In some studies total petroleum hydrocarbons (TPH), dissolved organic carbon (DOC), total organic carbon (TOC), oil and grease (O&G), or 5-day biochemical oxygen demand (BOD5) were used in addition to, or in lieu of, COD. In some studies, multiple metrics of treatment efficacy were used. As an example, Palmer et al. (1981) reported 72% BOD5 removal and 24% COD removal in an RBC treatment system (Huntington Beach location), indicating that much of the COD was recalcitrant. Su et al. (2007) showed similar results where removal of COD was 75% while BOD5 removal was 93% (HRT = 26.7 h). The study results indicate good COD removal—typically around 60–80%—provided that the operating parameters (e.g., HRT, OLR) are

adequate. Researchers also reported removal of ammonia, sulfides, and specific constituents in TPH (Guo et al. 2014; Riley et al. 2016; Tong et al. 2013; Zhang et al. 2016).

Different types of fixed-film media used. Campos et al. (2002) used 2 mm diameter polystyrene media in an airlift reactor configuration. Dong et al. (2011) added porous ceramic fixed-film carriers to an activated sludge system and found that inclusion of the carriers increased COD removal from 62 to 77%, while the modification of the carriers further increased COD removal to 79%. Ghorbanian et al. (2014) used 1 cm³ polyurethane foam cubes. Several researchers used plastic rings with fibers attached (Lu et al. 2009; Wang et al. 2007; Zhang et al. 2016).

Given the high mineral content of produced waters and potentially elevated carbonate concentrations, the potential for scaling of fixed-film media and its effect on biofilm formation should be considered. Zhang et al. (2016) reported mineral scaling within biofilms of a pilot-scale aerated lift system; however, the scaling did not inhibit biological activity as was the case in the commercial full-scale system treating the same produced water. Palmer et al. (1981) also reported mineral scaling on the disks of RBCs although scaling did not inhibit biological growth over the six month study period. Most studies focused on short-term feasible tests. Longer-term tests are needed to assess scaling potential or, alternatively, geochemical modeling can be used to assess scaling potential (Lester et al. 2015).

Membrane bioreactors

After fixed-film treatment systems, membrane bioreactors were the most commonly used treatment method (Table 3). Membrane bioreactors (MBRs) are good candidates for produced water treatment since good settling sludge is not necessary because membranes are used for solids separation (Tchobanoglous et al. 2014). Since external clarifiers are not used, MBRs typically have a small footprint compared with other types of biological treatment systems.

Table 3 Summary of membrane bioreactor studies for produced water

Reference	Sample type	TDS (mg/L)	COD (mg/L)	Pretreatments	Biological reactor			Key results
					Process	Size (L)	Parameters	
Janson et al. (2015)	Real	5189	1222	OWS, air stripping	MBR	1	HRT = 16–32 h, SRT = 60–120 d, temp = 22–38 °C, flux = 3–15 L m ⁻² h ⁻¹	60% COD removal, 91% TPH removal
Sharghi et al. (2014)	Synthetic	144,000, 184,000, 255,000, 299,000 (4 phases)	2600	–	MBR	5	HRT = 24 h, SRT = 80 d, OLR = 2.6 kg COD m ⁻³ d ⁻¹ , flux = 1.89 L m ⁻² h ⁻¹	82–95% COD removal
Ozgun et al. (2013)	Real	8100	2165	–	MBR, NF, RO	5.1	HRT = 15 h, SRT = 30 d, flux = 2.2 L m ⁻² h ⁻¹	83% COD removal, > 99% TPH removal
Sharghi and Bonakdar-pour (2013)	Synthetic	64,400	1800, 2600, 2600 (Phases 1, 2, and 3)	–	MBR	5	HRT = 24 and 36 h, SRT = 80 d, OLR = 1.2–2.6 kg COD m ⁻³ d ⁻¹ , flux = 1.26–1.89 L m ⁻² h ⁻¹	86–87% COD removal
Sharghi et al. (2013)	Synthetic	64,400	600, 1200, 1800 (Phases 1, 2, and 3)	–	MBR	5	HRT = 48 h, SRT = 80 d, OLR = 0.3, 0.6, 0.9 kg COD m ⁻³ d ⁻¹	83% COD removal
Kose et al. (2012)	Real	8367	2371	–	MBR	5.1	SRT = 30 d, flux = 10 L m ⁻² h ⁻¹	80–85% COD removal, 99% TPH removal
Pendashteh et al. (2012)	Both	35,000, 50,000, 100,000, 150,000, 200,000, 250,000 (synthetic), 16,400 (real)	1240 (real)	–	MSBR, RO	5	HRT = 24–96 h, cycle time = 12–48 h, OLR = 0.281–3.372 kg COD m ⁻³ d ⁻¹ , temp = 30 °C	98% COD removal (synthetic PW) and 86% COD removal (real PW) (OLR = 1.124 kg COD m ⁻³ d ⁻¹ , HRT = 48 h)
Kwon et al. (2011)	Both	10,000 (synthetic), 10,717 (field)	–	OWS, sand filtration, MF, SMZ	MBR with PAC added in some experiments	8	HRT = 9.6 h, SRT = 100 d	74–92% TOC removal (SMZ and MBR), 95% BTEX removal in MBR (field)
Pendashteh et al.	Synthetic	35,000	2250	–	MBR	5	HRT = 48 h, HLR = 80 L m ⁻² h ⁻¹ , cycle time = 22.5–23.5 h, temp = 30 °C	Mg, Al, Ca, Na, K, Fe, rod-shaped bacteria in foulants, contributed
Fakhru'l-Razi et al. (2010)	Real	35,000, 100,000, 150,000, 200,000, 250,000 (NaCl added), 16,400 (real)	1240	–	MSBR, RO	5	HRT = 8–44 h, cycle time = 12–48 h, OLR = 0.62 kg COD m ⁻³ d ⁻¹	91–92% COD removal

Table 3 (continued)

Reference	Sample type	TDS (mg/L)	COD (mg/L)	Pretreatments	Biological reactor			Key results
					Process	Size (L)	Parameters	
Zhang et al. (2010)	Real	9242	626		Anaerobic tank, MBR, coagulation-flocculation	13.5	HRT = 6–16 h, temp = 45 °C	24% COD removal in MBR
Scholzy and Fuchs, (2000)	Synthetic	–	0–7877 (8 trials)		MBR	9	HRT = 6.7 and 13.3 h, OLR = 0–9.82 kg oil m ⁻³ d ⁻¹ , temp = 35 °C	77–97% COD removal, 93–98% COD removal (3–5 kg oil m ⁻³ d ⁻¹)

COD chemical oxygen demand, *HLR* hydraulic loading rate, *HRT* hydraulic retention time, *MBR* membrane bioreactor, *MF* microfiltration, *MSBR* membrane sequencing batch reactor, *NF* nanofiltration, *OLR* organic loading rate, *OWS* oil–water separator, *PAC* powdered activated carbon, *PW* produced water, *RO* reverse osmosis, *SMZ* surfactant modified zeolite (adsorption), *SRT* solids retention time, *TDS* total dissolved solids, *TPH* total petroleum hydrocarbons

In the studies reviewed, COD removal was typically higher than 80%, indicating good treatment and there was variability in process control parameters (Table 3). The HRT used varied from 6 to 96 h. Cycle times in membrane sequencing batch reactors (MSBR) were 12–48 h (Fakhru'l-Razi et al. 2010; Pendashteh et al. 2012). Solids retention times (SRT) in the MBRs were long: 30–100 days. The flux reported was $2.2 \text{ L m}^{-2} \text{ h}^{-1}$ and lower in several studies (Ozgun et al. 2013; Sharghi and Bonakdarpour 2013; Sharghi et al. 2014), although $10 \text{ L m}^{-2} \text{ h}^{-1}$ and higher was reported (Kose et al. 2012). Similar to the results for fixed-film treatment studies, it appears that MBRs can achieve high COD removal provided that the operating parameters are controlled. The COD removal remained high even in the presence of surfactants (Scholzy and Fuchs 2000). Kwon et al. (2011) mitigated BTEX compounds using pretreatment by modified zeolite adsorption and mitigated organic loading by the addition of PAC in the MBR. In the MBR studies, fewer pretreatments appeared to be used although Zhang et al. (2010) used anaerobic pretreatment.

Membrane fouling and scaling were observed in several of the MBR studies. Treating real produced water, Janson et al. (2015) observed reduced membrane permeability and allowed the pH to remain low (4.9–6.0) to minimize precipitation; the effect on the microbial population was not studied. In another study, membrane fouling was only observed when the organic loading rate was high ($2.6 \text{ kg COD m}^{-3} \text{ d}^{-1}$) (Sharghi and Bonakdarpour 2013). Membrane fouling occurred as the result of excess extracellular polymeric substances (EPS) production and viscous sludge bulking (Sharghi and Bonakdarpour 2013). Based on microscopy by Fakhru'l-Razi et al. (2010), membrane fouling was the result of a dense cake layer containing bacteria (primarily rod-shaped) and minerals (e.g., containing magnesium, aluminum, silica, calcium, and iron). Reduced transmembrane pressure, indicative of fouling, was noted by Kwon et al. (2011). Pendashteh et al. (2011) reported membrane fouling in a MBR treating synthetic produced water, concluding that both biological and inorganic scaling were responsible. Fouling was not improved by flocculation pretreatment (Pendashteh et al. 2011). Sharghi et al. (2014) observed good treatment and no membrane fouling for treatment of a synthetic produced water with high salinity (TDS of 100,000–250,000 mg L^{-1}) although most of the salinity was from sodium chloride. In another study based on treatment of synthetic produced water, fouling was similarly not observed (Sharghi et al. 2013).

Wetlands and pond treatment

Wetlands and treatment ponds were investigated in 10 studies (Table 4). Constructed wetland treatment systems have advantages over mechanical treatment such as lower cost and maintenance as well as providing ecosystem services (e.g., wildlife habitat). The use of treatment wetlands by the petroleum industry has been reviewed by Knight et al. (1999) although most of the data available at the time originated from studies done on refinery wastewater.

Table 4 Summary of wetland and pond treatment system studies for produced water

Reference	Sample type	TDS (mg/L)	COD (mg/L)	Pretreatments	Biological reactor			Key results
					Process	Size (L)	Parameters	
Beebe et al. (2015)	Synthetic	Conductance = 337 μ S/cm (average)	–	–	FWS wetlands (series with 4 cells)	1060	HRT = 4–8 d	Complete ammonia oxidation in 3–4 months
Pardue et al. (2014)	Synthetic	–	–	OWS	FWS and SSF wetlands (series with 4 cells)	1512	HRT = 2 and 4 d	> 90% O&G removal when O&G loading < 10 mg/min
Alley et al. (2013)	Synthetic	Conductance = 9109 μ S/cm	–	OWS	FWS wetlands (series with 4 cells)	600	HRT = 6 d	94–95% removal of oil compounds (depth = 15, 23 cm)
Horner et al. (2012)	Synthetic	Conductance = 210 μ S/cm	213	–	FWS and SSF wetlands (series with 4 cells)	1512	HRT = 4 d	> 98% O&G removal
Shpiner et al. (2009a)	Synthetic	–	1200	–	Photobioreactor	10	HRT = 8 d, depth = 15 cm, 15 h light d^{-1}	> 85% COD removal
Shpiner et al. (2009b)	Synthetic	–	–	–	Photobioreactor, intermittent slow sand filter	10	Depth = 15 cm, 15 h light d^{-1}	95% Ni, Cd, Cr removal
Ji et al. (2007)	Real	–	390	Pond for temperature stabilization	SSF wetlands	140,000	HRT = 7.5 and 15 d	71–80% COD removal, 77–88% BOD5 removal
Murray-Gulde et al. (2003)	Real	6554	–	Filtration, ion exchange, filtration, RO	SSF wetlands (series with 4 cells)	260	HRT = 5 d total	80% TOC removal
Ji et al. (2002)	Real	–	479	Pond for temperature stabilization	SSF wetlands	540,000	HRT = 3 d, HLR = 0.52 $m d^{-1}$	81% COD removal, 89% BOD5 removal
Beyer et al. (1979)	Real	20,000	–	Flotation	Aerated lagoons	60,000	HRT <= 30 d	41% COD removal, 83% BOD5 removal, 63% TOC removal

BOD5 biochemical oxygen demand, 5 days, *COD* chemical oxygen demand, *FWS* free water surface, *HLR* hydraulic loading rate, *HRT* hydraulic retention time, *O&G* oil and gas, *OWS* oil-water separation, *RO* reverse osmosis, *SBR* sequencing batch reactor, *SSF* subsurface flow, *TDS* total dissolved solids, *TOC* total organic carbon

Most of the wetland studies reviewed used large containers to construct and simulate wetlands. Beebe et al. (2015) used a series of plastic containers (265 L) planted with cattails. Pardue et al. (2014) also used a series of plastic containers (378 L each) planted with reeds to simulate subsurface wetlands; free surface wetlands were simulated by planting the containers with bulrush and cattails. Alley et al. (2013) used four plastic containers (150 L each) in series, planted with bulrush and cattails, where the water depth was controlled and altered during the study. Shpiner et al. (2009a) studied pond treatment of a synthetic produced water in a laboratory setting with fluorescent lights. Murray-Gulde et al. (2003) simulated subsurface wetland treatment using a series of four 132 L barrels, with the barrels turned on the side and filled halfway. Horner et al. (2012) tested three wetland systems: one series of four plastic containers (379 L each) mimicking free surface wetlands and two series of four containers mimicking subsurface wetlands. Shallow depth was observed to be important for contaminant removal by Shpiner et al. (2009a), as was reactor baffling and recycle flows. Ji et al. (2007) used surface flow wetlands planted with reeds in a three-year experiment. In Ji et al. (2002), subsurface reed beds with variable hydraulic loading rates were observed. Aerated lagoon treatment was investigated by Beyer et al. (1979) in plastic lined steel tanks.

In the studies reviewed, wetland treatment was often used for nitrogen and metals removal in addition to reduction of oxygen demand. Beebe et al. (2015) observed both nitrification and denitrification when supplemental aeration and carbon were added in pilot-scale surface and subsurface wetlands treating a simulated produced water ($\text{TDS} < 1000 \text{ mg L}^{-1}$). Pardue et al. (2014) observed removal of O&G, iron, and manganese under oxidizing conditions as well as removal of nickel and zinc under reducing conditions; iron removal was variable. Redox conditions were controlled by altering the O&G loading rate (Pardue et al. 2014). In the laboratory-based photobioreactor, Shpiner et al. (2009b) reported precipitation by sulfide salts and other removal mechanisms (e.g., bio-adsorption) to remove metals (cadmium, chromium, nickel) from synthetic produced water in a laboratory photobioreactor. Alley et al. (2013) found that increased water depth increased removal of metals (cadmium, copper, nickel, and zinc) but that removal of trace contaminants (1,2-benzofluorenone and 1-methylcyclopentanol) was better in shallow wetlands. Alley et al. (2013) measured 1,2-benzofluorenone to indicate the hexane fraction of hydrocarbons and 1-methylcyclopentanol to indicate the chloroform fraction of alcohols. Ji et al. (2007) found that wetlands effectively reduced total Kjeldahl nitrogen in addition to COD. Horner et al. (2012) produced effluents suitable for livestock watering although the nickel was too high for irrigation. These studies show that wetland and pond treatment can be effective for COD removal. With the exception of one study, all studies reported COD removal rates $> 70\%$ (Table 4).

Pretreatment for wetland treatment systems is important for reducing influent loadings and stabilizing produced water temperatures. Pardue et al. (2014) found that use of an oil-water separator was important for increasing dissolved oxygen and soil redox potential within the wetland, and for maintaining oxidizing conditions. Murray-Gulde et al. (2003) used water softening and reverse osmosis upstream of a constructed wetland in a pilot-scale demonstration, observing a reduction of aquatic toxicity and 94% removal of TDS in the pretreatment processes. Temperature control was important in for wetland treatment in some studies (Ji et al. 2002, 2007).

Activated sludge

Activated sludge treatment, including conventional activated sludge and sequencing batch reactors, was investigated in eight studies (Table 5). Conventional activated sludge (CAS) treatment is a mature technology providing effective removal of oxygen demand and other contaminants; it is considered to have a low-cost and lower environmental impact compared with other treatment methods (Tchobanoglous et al. 2014). Sequencing batch reactors (SBRs) are similar to conventional activated sludge treatment but have the advantage of a smaller footprint because a separate clarifier is not required.

Table 5 Summary of activated sludge treatment studies for produced water

Reference	Sample type	TDS (mg/L)	COD (mg/L)	Pretreatments	Biological reactor			Key results
					Process	Size (L)	Parameters	
Kardena et al. (2017)	Synthetic	11,000	1120	–	CAS	7	HRT = 8–20 h, SRT = 10–25 d	82% COD removal (SRT = 20–25 d, HRT = 20 h)
Lester et al. (2015)	Real	22,500	1218	MF	SBR	0.1	HRT = 6 h, cycle time = 8 h	50% DOC removal
Pendashteh et al. (2010)	Both	35,000, 100,000, 150,000, 200,000, 250,000 (synthetic), 16,400 (real)	1240	–	SBR	5	Cycle time = 24 h, OLR = 0.9–3.6 kg COD m ⁻³ d ⁻¹ , temp = 30 °C	> 90% COD removal (synthetic PW, TDS = 35,000 mg L ⁻¹ , OLR = 1.8 kg COD m ⁻³ d ⁻¹), 74% COD removal (synthetic PW, TDS = 250,000 mg L ⁻¹), 81% COD removal (real PW)
Tellez et al.	Real	35,023	431	OWS, aeration	CAS	945	MLSS = 300– 700 mg L ⁻¹	97% COD removal, 99% TPH removal (SRT = 20 d)
Tellez et al.	Real	35,023	431	OWS, aeration	CAS, filtration	945	SRT = 0.5–40 d	97% COD removal, 99% TPH removal (after filtration)
Freire et al. (2001)	Real	52,100	2000	Coarse filtration, car- tridge filtration	SBR	1	Cycle time = 24 h, dilution of PW with domestic wastewater (10–45% v/v)	30–50% COD removal, 95% ammonium removal, 65% phenol removal
Dalmacija et al. (1996)	Real	32,300	400	Coagulation-floccu- lation	CAS with PAC added	5	HLR = 0.5–6.8 d ⁻¹ , PW diluted with river water	k = 0.012 d ⁻¹ , PAC = 400 mg L ⁻¹ , dilution = 3:1 river water to PW
Woolard and Irvine (1995)	Synthetic	152,000	–	–	SBR	1	Cycle time = 12 h	> 99.5% phenol removal

CAS conventional activated sludge, COD chemical oxygen demand, DOC dissolved organic carbon, HLR hydraulic loading rate, HRT hydraulic retention time, MF membrane filtration, MLSS mixed-liquor suspended solids, OLR organic loading rate, OWS oil–water separator, PAC powdered activated carbon, PW produced water, SBR sequencing batch reactor, SRT solids retention time, TDS total dissolved solids, TPH total petroleum hydrocarbons

Only four studies were completed using SBRs (Table 5). Lester et al. (2015) treated hydraulic fracturing flowback prior to reverse osmosis, a treatment combination that was able to produce effluent meeting irrigation standards. Pendashteh et al. (2010) used a SBR to investigate treatment of synthetic and real produced water with varying TDS concentrations (sodium chloride was added to increase TDS). The COD removal was lower in real produced water (81%) than in synthetic produced water (typically > 90% at moderate TDS) (Pendashteh et al. 2010). Freire et al. (2001) managed toxicity and added nutrients in treating a real produced water sample by diluting the influent with domestic wastewater. Biological inhibition was apparent because COD removal rates increased when the produced water was further diluted (Freire et al. 2001). Woolard and Irvine (1995) observed phenol removal in an SBR, treating a synthetic produced water.

Only four studies were completed on CAS, and two of them were done at the same facility (Table 5). In Kardena et al. (2017), 82% COD removal was achieved using process control parameters typically used at domestic wastewater facilities (Tchobanoglous et al. 2014). Tellez et al. (2002; 2005) demonstrated effective activated sludge treatment (> 95% COD removal) for treatment of produced water using a typical solids retention time (SRT) of 20 days. In the study by Dalmacija et al. (1996), microbial inhibition was remedied by diluting the produced water with river water. Powered activated carbon (200 mg L⁻¹) was added to the aeration basin, which likely provided a substrate for biofilm formation and mitigated toxicity (Dalmacija et al. 1996). In the CAS studies, COD removal was 82% and higher, where reported.

Anaerobic treatment

Anaerobic treatment was used in seven studies and in three of these studies, fixed-film media were also used (Table 6). Anaerobic treatment is beneficial because of the additional benefit of energy production. Although anaerobic treatment can be used for produced water, the salinity can be inhibitory and most of the hydrocarbons found in produced water are degraded aerobically, excluding some recalcitrant halogenated aromatics (Xiao and Roberts 2010).

Table 6 Summary of anaerobic biological treatment studies for produced water

Reference	Sample type	TDS ^a (mg/L)	COD (mg/L)	Pretreatment	Biological reactor			Key results
					Process	Size (L)	Parameters	
Ghorbanian et al. (2014)	Synthetic	20,000	–	–	(1) UASB with fixed-film media, and (2) anaerobic SBR	5	HRT = 24 h, OLR = 0.95, 1.45, 2.50 kg TPH m ⁻³ d ⁻¹	99.6% TPH removal (UASB), 98.5% TPH removal (SBR) (infl. TPH = 1450 mg/L)
Liu et al. (2013)	Real	1506	684	OWS, flotation	UASB, BAF	25,000 (UASB), 12,500 (BAF)	HRT = 12 h	74% COD removal (total)
Khong et al. (2012)	Real	19,070	1597	OWS	UASB with fixed-film media	5	HRT = 5 d, temp = 35 °C	61% COD removal, 76% COD removal (PW:tap water = 1:4)
Li et al. (2010)	Real	–	274	Micro-electrolysis (80 g/L iron, 40 g/L carbon)	Anaerobic reactor	30	15 d tests	53% COD removal (total)
Ji et al. (2009)	Real	13,050	550	–	Anaerobic reactor	75	HRT = 2.5 d, OLR = 0.20 kg COD m ⁻³ d ⁻¹	65% COD removal, 88% heavy oil removal
Vieira et al. (2005)	Real	75,890	4730	–	Anaerobic reactor	1.5	HRT = 6 and 15 d, pH control, temp = 35 °C	57% COD removal, 58–78% phenol removal (15 d)
Rincon et al. (2003)	Real	–	1150, 890, 275 (light, medium, heavy crude oil PW)	OWS	UASB	4	HRT = 18 (light) and 24 h (medium and heavy), OLR = < 1.80, 0.53–1.09, 0.18–0.33 kg COD m ⁻³ d ⁻¹ (light, medium, heavy), temp = 37 °C	87%, 20%, 37% COD removal (light, medium, heavy oil PW)

BAF biological aerated filter, COD chemical oxygen demand, HRT hydraulic retention time, OLR organic loading rate, OWS oil–water separation, PW produced water, SBR sequencing batch reactor, TDS total dissolved solids, TPH total petroleum hydrocarbons, UASB upflow anaerobic sludge blanket

Different configurations of anaerobic treatment were used. In four studies, upflow anaerobic sludge blanket (UASB) reactors were used, and in two studies fixed-film media were incorporated into the UASB (Ghorbanian et al. 2014; Khong et al. 2012; Liu et al. 2013; Rincon et al. 2003). Liu et al. (2013) used a UASB followed by an aerated biological filter. Ji et al. (2009), used a baffled reactor for anaerobic treatment.

The removal rates in anaerobic treatment studies varied depending on the study conditions, and COD removal ranged from 37 to 87% (Table 6). While COD was not reported, Ghorbanian et al. (2014) reported almost complete removal (> 98%) of TPH in both an anaerobic UASB with fixed-film media and in an anaerobic SBR. Li et al. (2010) increased COD removal to 53% by including a micro-electrolysis pretreatment step, which partially degraded hydrocarbons and increased their biodegradability. Khong et al. (2012) increased COD removal by diluting the produced water treated. In one study, 74% COD removal was the result of both anaerobic and aerobic treatment, and 94% removal of ammonia was observed (Liu et al. 2013). The HRT in the anaerobic reactors varied from 12 h to 15 days, which likely explains the variability in COD removal observed.

The differing study conditions allowed for observation of different issues that can be helpful in future studies. Mineral crystallization (Fe_2O_3 , FeS, and CaCO_3) was observed in the anaerobic sludge in one study, suggesting that mineral precipitation could be a concern in anaerobic reactors treating produced water (Ji et al. 2009). Control of hydrogen sulfide—done by altering the pH and periodically purging with nitrogen gas—was an issue in another study (Vieira et al. 2005). Rincon et al. (2003) studied produced waters originating from the separation of light, medium, and heavy crude oil, which yielded different COD removal rates (87, 20 and 37%), demonstrating treatment variability depending on the produced water characteristics.

Bio-electrochemical systems

Although an evolving technology, six studies were located using bio-electrochemical systems (BES) for produced water (Table 7). In BES treatment, energy from microbially mediated redox reactions is harnessed via electron flow between electrodes (Jain et al. 2017). In the microbial aerobic conversion of organic compounds to carbon dioxide, electrons are released at the anode and water is produced at the cathode; organic carbon removal also occurs when compounds adhere to activated carbon surface used in BES (Forrestal et al. 2015; Stoll et al. 2015). An advantage of electrochemical treatment is that the electrical current disrupts the oil present in produced water, making it more amenable to treatment (Jamaly et al. 2015). Since BES technologies capture ions at cathodes and anodes, desalination occurs in addition to degradation of organic matter and oxidation of other contaminants.

Table 7 Summary of bio-electrochemical treatment studies for produced water

Reference	Sample type	TDS (mg/L)	COD (mg/L)	Pretreatments	Biological reactor			Key results
					Process	Size (L)	Parameters	
Shrestha et al. (2018)	Real	60,000	–	–	(1) MFC and (2) MCDC	–	Diluted PW with deionized water until infl. COD=10,000 mg L ⁻¹	85–96% COD removal (MFC), 71–85% COD removal (MCDC)
Sheikhyousefi et al. (2017)	Both	187,300 (real PW)	2000 (synthetic), 3685 (real)	–	MFC	0.125	HRT=20 d, 35 °C, 100 Ω resistor, diluted PW until infl. COD=2000 mg L ⁻¹	96–97% COD removal (synthetic), 94–96% COD removal (real)
Mousa (2016)	Real	66,985, 10,131, 17,665, 10,785 (Sites 1, 2, 3, 4)	–	–	EBC, biofilter	1.7	HRT=5.1 min, DC power w/10–30 V applied, energy consumption=32.6 mA cm ⁻²	75% TPH removal, 25% sulfate removal
Forrestal et al. (2015)	Real	15,870	950	Hydrocyclones, dissolved air flotation, air stripping	MCDC	0.062	HRT=4 h (desalination chamber)	85% COD removal, 75% recovered during regeneration
Naraghi et al. (2015)	Real	200,000	2750	–	EBC	0.09	Open circuit potential=330 mV, power density=0.65 mW m ⁻³ , diluted PW until infl. COD=700 mg L ⁻¹	70% COD removal (96 h), 89% COD removal (148 h), 400 mL H ₂ m ⁻³ d ⁻¹
Stoll et al. (2015)	Real	15,870	950	OWS, dissolved air flotation, air stripping	MCDC	0.062	–	74% COD removal (first run, lower in subsequent runs)

COD chemical oxygen demand, *EBC* electrobiochemical, *HRT* hydraulic retention time, *MFC* microbial fuel cell, *MCDC* microbial capacitive deionization cell, *OWS* oil–water separator, *PW* produced water, *TDS* total dissolved solids, *TPH* total petroleum hydrocarbons

The studies reviewed had different configurations (Table 7). In two studies microbial fuel cells (MFC) were studied that consist of anaerobic treatment with biofilm-coated anodes and cathodes that form a cell contained within the reactor (Sheikhyousefi et al. 2017; Shrestha et al. 2018). As described by Sheikhyousefi et al. (2017), the anodes and cathodes are constructed of treated carbon cloth. Microbial capacitive desalination cells (MCDC) were also studied where the anode and cathode are placed in separate compartments and a desalination compartment resides between the anode and cathode compartments (Forrestal et al. 2015; Shrestha et al. 2018; Stoll et al. 2015). As described by Forrestal et al. (2015), the desalination compartment consists of a series of nickel/copper electron collectors that have activated carbon cloth on either side and are separated from other collectors by plastic mesh. In one study, an electrochemical system was combined with a biological filter where a voltage was applied to titanium-mixed metal oxide electrodes prior to filtration (Mousa 2016). In another study, a spiral anode was used with a cathode in the center; the spiral anode was intended to grow the biofilm to facilitate the redox reactions (Naraghi et al. 2015).

The COD removal in the BES studies was consistently higher than 70%. In a study comparing MFC with MCDC, Shrestha et al. (2018) found that the MFC yielded higher COD removal rates (85–96%) than MCDC (71–85%). Conversely, TDS was better removed in MCDC (65–74%) than in MFC (20–40%) (Shrestha et al. 2018). Sheikhyousefi et al. (2017) observed almost identical COD removal in real produced water (96%) compared with synthetic produced water (96–97%). The COD removal rates reported are impressive, especially considering that the TDS must be approximately 100,000 mg L⁻¹ based on the water quality and dilution described (Sheikhyousefi et al. 2017). Although COD removal rates are high in EBS, recovery of ion adsorption capacity during regeneration can be limited as noted by Forrestal et al. (2015) where 75% recovery was observed. Also, COD removal rates can decrease as the ion adsorption capacity is reduced; Stoll et al. (2015) observed lower COD removal rates in subsequent runs of a MCDC. While still an evolving technology, EBS appear promising for high salinity wastes such as produced water and will likely improve with further testing (Jain et al. 2017).

Effect of salinity on biological treatment

Salinity can inhibit biological treatment (Castillo-Carvajal et al. 2014; Xiao and Roberts 2010). Here, treatment efficacy, as measured by a reduction in COD, appeared related to TDS concentration (Fig. 3). It also appears that TDS was more inhibitory in real produced water than in synthetic produced water. Not all of the studies reviewed reported TDS and COD, so not all studies are represented in Fig. 3. Where ranges of values were reported, mean values were used. Where the TDS of the produced water treated was altered, each test was plotted separately in Fig. 3. When only real produced water samples are considered, it appears that MBRs typically provided the

best treatment (Fig. S2). One activated sludge study with data available showed good removal (Tellez et al. 2002, 2005), but data from other activated sludge studies are not available to corroborate this result. The BES study using real produced water exhibited COD removal higher than other types of reactors with similar TDS (Sheikhyousefi et al. 2017).

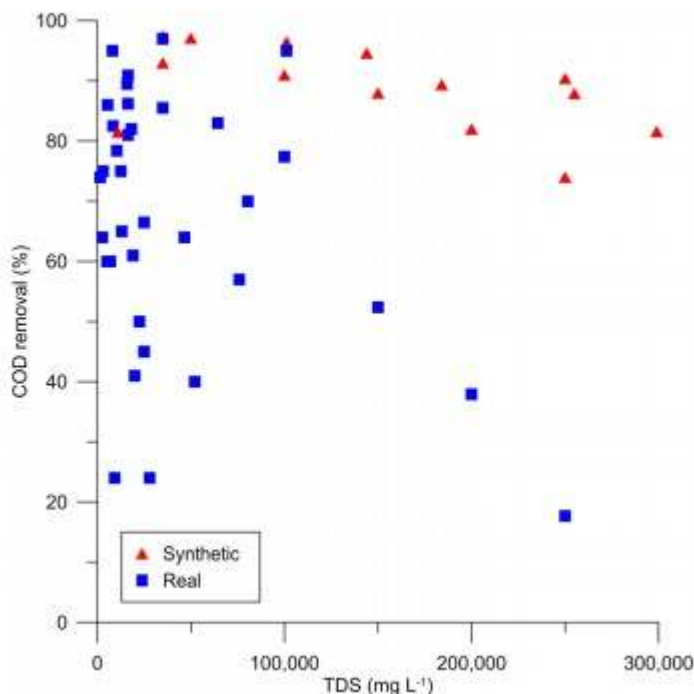


Fig. 3

Relationship between chemical oxygen demand (COD) removal and total dissolved solids (TDS) for biological treatment of produced water, with data separated by the type of sample

Controlled increases in salinity allowed for its impact on treatment efficacy to be studied in a controlled manner. For example, Sharghi et al. (2014) varied the TDS of synthetic produced water (144,000, 184,000, 255,000, and 299,000 mg L⁻¹), and observed an impact on COD removal (95, 89, 88, and 82%, respectively) and O&G removal (94, 92, 90, 85%, respectively). Pendashteh et al. (2012) observed similar results when the TDS of both synthetic and real produced water were varied (35,000, 50,000, 100,000, 150,000, 200,000, and 250,000 mg L⁻¹). Pendashteh et al. (2012) observed differences between real and synthetic produced waters that were apparent —when the TDS was 250,000 mg L⁻¹, COD removal was 90% in the synthetic produced water, but only 18% in the real produced water. In an earlier study, Pendashteh et al. (2010) observed greater than 90% removal of COD when the synthetic produced water TDS was 35,000 mg L⁻¹, but only 74% removal when the TDS was increased to 250,000 mg L⁻¹. In small-scale tests of hydraulic fracturing fluids and flowback, the effects of salinity on aerobic degradation have been studied—Kekacs et al. (2015) found that TDS > 40,000 mg L⁻¹ inhibited biological activity in bottle tests. In small-scale

treatability tests, Akyon et al. (2015) observed biodegradation of 1.45 mg COD g⁻¹ wet d⁻¹ when TDS = 91,351 mg/L.

The data on TDS and COD removal indicate a complex and site-specific relationship between COD removal and TDS, but the data also indicate good treatment (Fig. 3). In treating real produced water samples, average COD removal was 73% when TDS was < 50,000 mg L⁻¹ and 54% when TDS was < 100,000 mg L⁻¹. While these COD removal rates suggest that sole use of biological treatment is insufficient for beneficial reuse and recycling, it does appear that biological treatment provides the benefit of reduced oxygen demand which may make effluents suitable for other treatment technologies to further reduce organic matter and other contaminants (Camarillo et al. 2016).

Biological treatment for membrane systems

Biological treatment can serve as a pretreatment for membrane technologies, reducing organic loads to reduce biofouling. In nine of the studies reviewed, biological treatment was investigated with the intent of biological treatment serving as a pretreatment for membranes (Campos et al. 2002; Fakhru'l-Razi et al. 2010; Freedman et al. 2017; Kwon et al. 2011; Lester et al. 2015; Ozgun et al. 2013; Pendashteh et al. 2012; Riley et al. 2016; Zhao et al. 2006). Riley et al. (2016) used a biologically active GAC filter followed by ultrafiltration and nanofiltration in series; the combined treatment reduced organic compounds by 99% and TDS by 94%, producing an effluent with TDS as low as 700 mg L⁻¹. Fakhru'l-Razi et al. (2010) used a MBR followed by reverse osmosis, reduced TDS to 450 mg L⁻¹ and removed most organics (effluent COD was 23 mg L⁻¹). Ozgun et al. (2013) compared an MBR with the combination of microfiltration (MF) and ultrafiltration (UF) as pretreatment steps for nanofiltration followed by reverse osmosis. The MBR provided better COD removal than MF/UF, achieving 83% COD removal, although O&G removal was better in the MF/UF system (Ozgun et al. 2013). Lester et al. (2015) found that biological treatment followed by RO was sufficient to produce effluents meeting standards for irrigation.

Chemical pretreatments for biological treatment

The efficacy of biological treatment could be improved by chemical pretreatments. Electrocoagulation appears technically promising as a pretreatment, as it can reduce many produced water constituents, including boron and those that contribute to hardness, COD, and total hydrocarbons (Esmailirad et al. 2015; Ezechi et al. 2014; Zhao et al. 2014). Lu and Wei (2011) investigated treatment with zerovalent iron (20 g/L) and EDTA (150 mg/L) prior to studying biotreatability of produced water from a polymer flooded oilfield—45% COD removal was observed despite the presence of recalcitrant polymer hydrolyzed polyacrylamide. Coagulation-flocculation was used as a pretreatment in an aerobic batch test study with other pretreatments (flotation, sedimentation, and filtration), resulting in 91% TPH removal (Steliga et al. 2015).

Although not specific to produced water, other studies outside of the 59 studies reviewed suggest pretreatments that would be effective for produced water. Fenton's reagent and ozonation were shown to increase biodegradability of process water from a sour gas sweetening plant in Mexico that contained diethanolamine (Duran-Moreno et al. 2011). Correa et al. (2010) used ozone-photocatalytic oxidation (O₃/UV/TiO₂) to treat petroleum refinery effluents prior to algae-based treatment, effectively reducing phenol, sulfide, COD, O&G, and ammonia. Nam et al. (2001) used hydrogen peroxide pretreatment to biodegrade soil contaminated with PAH.

Inoculum used in biological treatment

A variety of inoculum sources were used to introduce microorganisms acclimated to the high salinities and toxic conditions found in produced water. Researchers used indigenous microorganisms from the produced water to establish biofilms in reactors (Freedman et al. 2017; Riley et al. 2016). Microorganisms were isolated from oily sludge collected from an oil tank or settling tanks (Liu et al. 2013; Naraghi et al. 2015; Zhang et al. 2016), activated sludge was used that may or may not have been acclimated to the feedstock (Ghorbanian et al. 2014; Stoll et al. 2015), microbes were obtained from soil contaminated by oil spills (Lu and Wei 2011; Sharghi and Bonakdarpour 2013; Sharghi et al. 2013, 2014), commercial laboratory microorganisms were cultured and used (Shpiner et al. 2009a, b; Tong et al. 2013), some samples were scraped from lake rocks (Chavan and Mukherji 2008), and microorganisms were isolated from a salty lake (Woolard and Irvine 1994, 1995). In one study where the impact of using different commercially available cultured organisms were used as the inoculum, Zhao et al. (2006) observed 84% PAH removal using one strain and 90% removal using another. In another study, isolation and inclusion of heat-resistant bacteria in the inoculum was important (Guo et al. 2014). Zhang et al. (2016) used bioaugmentation and supported its efficacy by genetic analysis of the biofilms, which showed incorporation of the augmented bacteria into the biomass. In a study of biotreatability in bottle tests, immobilized cells were used to inoculate the solutions (Li et al. 2005).

Conclusions

This review successfully aggregates results from previous studies on the biological treatment of produced water. Information from the studies is summarized, including descriptions of the types and origins of produced water treated, influent water quality (COD and TDS), pretreatments, biological process under study, posttreatment, process control parameters, and major study results. We also reviewed the studies to determine the authors' motivations for performing their studies, adding insight into this global environmental issue. Inclusion of a meta-analysis allowed us to quantify COD and TDS of the produced water under study and to clarify the relationship between TDS and COD removal efficiency.

The results indicate that biological treatment of produced water is being studied globally. While most real produced water samples originated from China and the USA (37 and 28%, respectively), produced water from ten countries was used in the studies. While most researchers cited environmental concerns as the motivation for their study (51%), stringent regulations were also noted as a study motivator (32%), suggesting that increasing regulations are causing researchers to consider more treatment options. Most studies to date have been laboratory based, bench-scale studies (69%) and fewer larger-scale, field-based studies have been conducted. The most commonly studied biological processes were fixed-film technologies and membrane bioreactors (32 and 20%, respectively), although studies were located that used wetlands and treatment ponds, activated sludge, and anaerobic treatment. Several of the most recent studies have been conducted on innovative bio-electrochemical systems that integrate carbon oxidation and salt removal into one technology.

Overall, biological treatment of produced water appears a viable approach, particularly where salinity is not too high (e.g., below 50,000 mg L⁻¹). In the studies reviewed, COD removal was typically above 50% and much higher depending on study conditions. In real produced water samples where TDS was < 50,000 mg L⁻¹, average COD removal was 73%. Removal of COD appeared related to the produced water TDS although the relationship was not linear. Removal rates for COD in studies using synthetic solutions were higher than in studies using real produced water samples (given similar TDS).

Several conclusions can be drawn based on the results of the studies reviewed. Effective pretreatment and nutrient addition are important in the biological treatment of produced water. Chemical pretreatments that alter organic matter and make it more amenable to treatment are promising and should be further pursued. A specialized microbial consortium is likely unnecessary although microbial acclimation should be considered in start-up of reactors. The effects of salinity and other forms of toxicity can likely be mitigated in biological treatment systems using fixed-film media and other approaches. Special consideration should be given to mineral scaling in fixed-film and membrane treatment process because such scaling has been observed and can be detrimental to treatment outcomes. Based on the study results, it appears that biological treatment can effectively serve as pretreatment for membrane and desalination technologies, producing effluents suitable for reuse. Future studies are recommended where biological systems operate under realistic field conditions over a long period of time. Such studies would be useful for developing guidelines for recycling and beneficial reuse of produced water effluents.

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