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Perspective

Oil and Gas Produced Water Reuse: Opportunities, Treatment Needs, and Challenges

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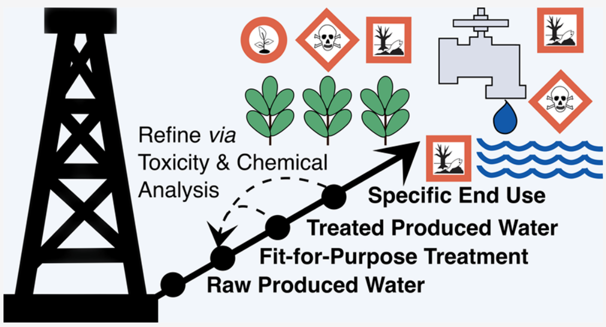

ABSTRACT: Advances in water treatment technologies paired with potential restrictions on oil and gas (O&G) produced water disposal could incentivize the beneficial reuse of treated produced water in the O&G industry. However, the remote nature of O&G operations limits the applicability of many of these solutions, which may be spatially inefficient, require operator supervision, or are ill-suited for the complex nature of produced water. Furthermore, the responsible, sustainable reuse of produced water as an alternative water source requires standardized analytical techniques for characterizing and determining the toxicity of treated produced water and improving our understanding of the fate and transport of various constituents. In the past decade, we made little progress in economically treating produced water for beneficial reuse outside of oilfield operations; the sole major breakthrough has been in the development of salt-tolerant fracturing chemicals that allow for reuse of produced water for fracking operations. Guided research should assist in the development of fit-for-purpose solutions to maximize the reuse of treated produced water. This is exemplified by the case studies presented here that detail currently operating treatment facilities for reclamation and reuse of produced water.

KEYWORDS: *Water Reuse, Hydraulic Fracturing, Toxicity, Technoeconomic Analysis, Wastewater Treatment*

1. INTRODUCTION

Use, disposal, and reuse of water associated with oil and gas (O&G) production has been a topic of interest to O&G operators, regulators, water users, and researchers for decades, but over the past decade, this interest has peaked due to the increase in hydraulic fracturing operations, water scarcity, and environmental and toxicological concerns. Recent analyses have highlighted the extensive volumes of produced water associated with different O&G production basins, as well as the potential management options for produced water across the basins.[1−3] These studies suggest that the recycling and beneficial reuse of produced water (e.g., reuse for irrigation, livestock watering, streamflow augmentation, municipal water supplementation, cooling water, dust suppression, ice control on roadways) must be viewed in terms of both regional availability and end user demand. Drivers for recycling and beneficial reuse of produced water often include reducing the freshwater intensity of O&G production,[4] minimizing seismicity associated with deep well disposal,[5] providing source water for other sectors,[6,7] and enhancing potential resource recovery from these waters.[8−10] Yet, despite a desire to increase produced water reuse, techno-economic, regulatory, and social challenges complicate beneficial reuse of produced water.[11] The confluence of these factors has resulted in the reinjection of ~92% of the 24.4 billion barrels (1.025 trillion gallons/508 million m^3) of produced water generated in the United States (US) in 2017 into the subsurface and minimal beneficial reuse.[1]

This dearth of reuse has not been the result of an absence of support for fundamental and applied research among the various stakeholders. However, due to widespread regional variability in produced water quality, generation, and management regulations, produced water research over the past several decades has involved predominantly a single or limited number of investigators or business-driven development of

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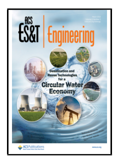

intellectual property, research papers, and field trials of technologies. In most cases, these studies addressed the characterization and treatment of specific waters and did not propose universally applicable produced water characterization or treatment technologies. This has led to a myriad of papers and literatures that often stymies stakeholder's search for relevant information, as highlighted in Tables S1 and S2 in the Supporting Information (SI), which categorize the range and breadth of topics covered. These tables identify select literatures that address water quantity, water quality, water treatment technologies, and water management issues.

Widespread regional variability in water availability and produced water quantity have been highlighted in several recent studies,[1−3] including Scanlon et al., who not only identified the regionality of water use but also connected it to potential reuse options.[3] The literature identified in Table S1 also highlights that produced water generation also differs both by basin and by well within the same basin due to the type of well (conventional or unconventional), drilling method, completion type, and age of well, resulting in water-to-oil ratios that range nationally from 3:1 to more than 10:1.[2,3,12,13] For example, in 2017, the Permian Basin, a relatively wet tight oil basin, produced upward of 1.66 billion barrels (69.7 billion gallons/264 million m^3) of produced water, while the Marcellus Basin, a relatively dry shale gas basin, only produced 0.033 billion barrels (1.34 billion gallons/5.25 million m^3) of produced water.[2] Conventional O&G wells are drilled into geological formations where oil and natural gas readily flow to the wellbore, while unconventional O&G wells tap into previously unconventional geological sources such as shale gas, coalbed methane (CBM), shale oil, tight oil, and oil sands. While conventional produced water is often reinjected into medium-to-high permeability reservoirs for pressure maintenance or enhanced oil recovery (EOR), unconventional produced water often cannot be reinjected into the low-permeability reservoirs associated with unconventional production.[14] For context, between 2009 and 2016, O&G operations in the Permian used 27 billion barrels (1.134 trillion gallons/4.29 billion m^3) of conventional produced water for EOR and disposed of 6.6 billion barrels (277 billion gallons/1.05 billion m^3) of conventional and 5.5 billion barrels (231 billion gallons/0.87 billion m^3) of unconventional produced water via saltwater disposal wells.[14] Recycling or beneficial reuse of unconventional produced water is further impeded by the temporally mismatched supply and demand for water within the hydraulic fracturing processes that can inhibit recycling within O&G without the presence of extensive storage, water handling, and conveyance infrastructures.[11]

Management of these spatially and temporally variable quantities of produced water is further confounded by the inconsistency of produced water quality and the resulting treatment challenges. Produced water contains variable concentrations of inorganics, organics, microorganisms, suspended solids, radioisotopes, and dissolved gases which vary by factors including the well's natural geologic formation, type of well, type of hydrocarbon being produced, and well production time.[1,12,15−22] Literatures revealing the variability in water quality are also presented in Table S1. In particular, total dissolved solids (TDS) concentrations, ranging from <1000 to >250,000 mg/L, can present substantial challenges for both treatment and residual management.[12,15,23] While organic matter in raw or pretreated produced water (total organic carbon (TOC) < 1500 mg/L) is generally not a limiting factor in disposal or recycle, sustainable beneficial reuse may be limited by concerns surrounding characterization, fate, and toxicity of constituents identified in produced water.[1,12,15−22,24,25]

A Google Scholar search of publications and patents illustrates the growing interest in produced water treatment (from 25 in the 1960s to 5780 in the 2010s) using commercially available technologies like reverse osmosis (RO) desalination, nanofiltration (NF), and membrane bioreactor (MBR) and such novel technologies as forward osmosis, osmotically assisted RO, membrane distillation, and eutectic freezing for removal of bulk TDS and/or organics, along with a range of technologies for precision separation of trace constituents. In addition to the numerous papers focused on individual technologies, since the 1970s more than 80 review papers, including several recent reviews, have discussed the generation, characterization, and treatment of produced water (Table S2).[26−32] For example, Conrad et al. specifically addresses the need for fit-for-purpose treatment to meet the varying end use water quality requirements.[13] Nevertheless, there are only a few examples in which novel technologies or treatment trains have been deployed for beneficial reuse of produced water.[33−35]

Moreover, no technological solution is complete without considering the disposal of residuals, regulatory constraints associated with disposal and reuse, and the environmental and health impacts of disposal and reuse. Regional, federal, and state regulations further influence the feasibility of various produced water management options. The U.S. Environmental Protection Agency (EPA) enacts national environmental regulations and grants primacy to state agencies to enact state-specific regulations that meet or exceed the stringency of the national regulations.[36] Title 40 of the Code of Federal Regulations Part 435 (40 CFR 435) Subchapter C does not allow onshore upstream O&G facilities east of the 98th meridian to discharge pollutants from produced water or other O&G fluids to surface waters.[37] In contrast, for onshore O&G facilities west of the 98th meridian, 40 CFR 435 Subchapter E allows for the discharge of produced water to surface water if the facility has a state and/or federally issued National Pollutant Discharge Elimination System (NPDES) permit, assuming that the effluent limitations of 40 CFR 437 are met. State-level produced water regulations depend on factors including the state's political and environmental climate, the state's water rights laws, the type of O&G resources, and the associated produced water quantity and quality. For more information on the produced water quantity, quality, and regulations surrounding produced water management, please refer to Table S1 in the Supporting Information.

While the treatment costs, residual management, and regulatory constraints remain significant barriers for produced water recycling, a secondary concern is the potential for toxicity associated with the beneficial reuse of produced water. As such, the design and operation of these treatment trains must mitigate risk associated with ecotoxicity, human toxicity, and soil and crop health relevant to specific end uses. Past work has highlighted common treatment technologies utilized by O&G companies that emphasizes the varying levels of treatment required for different end uses (Figure 1), but typically these technologies are selected based on the removal of target species rather than toxicity-relevant metrics for a specific beneficial use.

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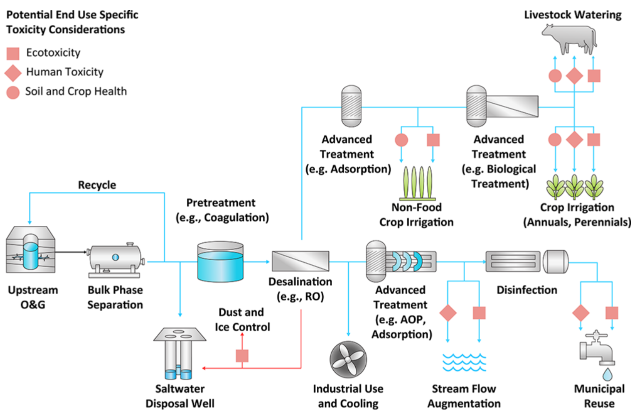

Figure 1. Common proposed produced water management options and corresponding potential toxicity concerns associated with specific end uses. Appropriate water treatment trains depend on both initial water quality and desired end use. Depending on these factors, water treatment could include the separation of oil, grease, and suspended solids (e.g., API gravity separator, dissolved air floatation, coagulation/flocculation, sedimentation), removal of bulk organics and target constituents (e.g., biological treatment, adsorption), desalination (e.g., electrodialysis, reverse osmosis), and post-treatment (e.g., advanced oxidation processes, ion exchange, disinfection). Red arrows indicate concentrate/brine streams.

To this end, there is a need to understand and address the current limitations associated with quantifying toxicity and environmental impacts to develop end use specific water quality criteria that can guide fit-for-purpose treatment strategies. This perspective utilizes several case studies to review current management practices for produced water and examines potential scenarios for expanding recycle and beneficial reuse; however, scientific advancements are needed for the better characterization and assessment of produced water being considered for beneficial reuse. Thus, a second integral component of this perspective is a discussion of both the challenges of characterizing the chemical composition and toxicity of produced water for specific end uses as well as a proposed strategy for assessing produced water toxicity to enable sustainable, beneficial reuse of produced water for specific end uses.

2. METHODS

Recycling or beneficial reuse of produced water has been implemented successfully in several cases. Analyses of these cases can be used to set the stage for identifying potential additional management options, as well as the current limitations and concerns surrounding expanded beneficial reuse. From disposal to recycling, treatment needs increase minimally in complexity, but as we expand from recycling to beneficial reuse applications, variable regulatory policies, treatment complexity, and public perception play more significant roles in realizing produced water reuse potential.

In this perspective, we utilize a literature-based analysis of current and potential management scenarios to assess the current potential for expanding both recycling and beneficial reuse. As highlighted earlier, most produced water is reinjected into the subsurface for either disposal or EOR. We chose six different case studies that highlight current common practices to represent possible produced water management approaches. The cases selected are (1) treatment and disposal of produced water via saltwater waste disposal (SWD) wells in the Permian Basin in Texas, (2) collection, treatment, and recycling of produced water in the Anadarko Basin in Oklahoma, (3) treatment, recycling, and beneficial reuse of produced water in the Marcellus Basin in Pennsylvania, (4) treatment and beneficial reuse of produced water for streamflow augmentation in the Denver—Julesburg (DJ) Basin in Colorado and in the Powder River Basin in Wyoming, (5) treatment and beneficial reuse of produced water for agricultural irrigation in the San Joaquin Basin in California, and (6) proposed treatment and beneficial reuse of produced water for municipal reuse in the Powder River Basin in Wyoming. These case studies were chosen to represent a wide geographic area, various commonly implemented or proposed end uses for produced water, and where data were readily available to model the treatment train.

For case studies where enough data were available, we modeled the treatment train using the Water Techno-Economic Assessment Pipe-Parity Platform (WaterTAP3). Specifically, three fit-for-purpose treatment approaches were evaluated based on previously reported literatures and assumptions in WaterTAP3 data to assess the levelized cost of water (LCOW, $\$/m^3$), energy intensity ($kWh/m^3$), and water recovery (%) using WaterTAP3. WaterTAP3 simulates a steady-state water treatment train and unit performance as well as capital and operating costs based on source water conditions, unit-level configurations for water treatment technologies, and system-level techno-economic assumptions. All unit processes achieve both flow and constituent mass balances.

WaterTAP3 estimates total installed costs for unit processes within the treatment train (e.g., capital cost of equipment and installation), fixed operating costs (e.g., employee salaries, plant maintenance), and variable operating costs (e.g., chemical addition, electricity) at the unit level. LCOW is then calculated from the total installed costs, operating costs, and a financial capital recovery factor with respect to the total volume of treated water stream. The following analysis presents an adjusted LCOW ($\$/m^3$) with respect to the total volume of influent produced water. WaterTAP3 calculates the system-level electricity intensity based on the electricity consumption of each unit within the treatment train and the volume of treated water by the system. The cost of energy ($/kWh) is based on the state-level cost of electricity for industrial purposes as reported by the U.S. Energy Information Administration. Facility locations are noted in the SI.

Analysis in WaterTAP3 is analogous to a Class 4 Feasibility Study as defined by the Association for the Advancement of Cost Engineering International Recommended Practice No. 18R-97.[38] As such, the uncertainty is −30% to +50% and is based on the availability of data, technology readiness level of the modeled technology, and the analysis approach. Additional details regarding the WaterTAP3 are available in Miara et al. and in the SI.[39]

Cases were evaluated for current economic and technological feasibility, and multiple scenarios with differing water qualities were generated and analyzed based on previously reported literature data. Alternate treatment scenarios endeavored to assess the applicability and potential of produced water management approaches in the context of increasing both recycling and beneficial reuse of produced water. Techno-economic modeling was performed to determine what research and development advances are needed to address cost trade-offs of produced water treatment trains.

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Also, while we compare the costs of water treatment to current water prices and disposal options, changing water availability trends and increasing prices for water may mean that some options for treating produced water for different end uses will become more economically competitive over time.

3. PRODUCED WATER TRENDS AND CASE STUDIES ANALYSES

3.1. Recycling. The upstream O&G industry uses billions of barrels of water per year to extract resources from underground geologic formations.[40,41] While water withdrawals for O&G constitute less than 1% of the total withdrawals in the US, O&G operations use of nearby surface and groundwater may exacerbate problems associated with water scarcity in semiarid and arid regions.[2,3,41−43] Furthermore, water use for unconventional O&G production nearly doubled between 2011 and 2018, and is anticipated to continue to increase.[4] Consequently, there is both industry and public interest in reducing the freshwater footprint of the upstream O&G industry.

Recycling of produced water within the upstream O&G sector is preferable when economically viable as it minimizes the use of external water sources, reduces produced water management liability concerns, and limits the management of produced water (e.g., treatment, conveyance, and disposal).[11,35] Before recycling, insoluble oil, microorganisms, iron, and boron are traditionally removed via fit-for-purpose treatment trains with oil−water separations, solids separation, disinfection, and iron removal.[11,44] Desalination is often not necessary for recycling as recent advances in hydraulic fracturing chemicals have enabled the recycling of produced water with TDS of nearly 300,000 mg/L.[45,46] However, this level of treatment may not be adequate to reduce public concerns surrounding spills and the potential resulting contamination of surface waters, groundwater, and soil.[47−49] In particular, in basins with relatively high salinity produced water, like the Permian and Bakken (often greater than 200,000 mg/L TDS), there are additional concerns related to scaling and potential for spills that may increase the costs of both treatment and conveyance.[11]

Expansion of recycling may alleviate the potential for the competition for water by O&G exploration and production with other end uses (e.g., agricultural, municipal);[42] nonetheless, recycling is frequently limited by logistics and economics instead of treatment capabilities. Within relatively dry basins (e.g., Marcellus, Eagle Ford, Niobrara, and Haynesville), recycling varies from more than 90% within the Pennsylvania region of the Marcellus to approximately 1% within the Haynesville and Eagle Ford Basins, respectively.[2,11,50] The wide variability in recycling has been attributed to factors such as regulatory limitations on SWD in the Marcellus leading to high produced water management costs for conveyance, the logistics of handling a relatively limited quantity of water within a large basin in the Eagle Ford, spatial and temporal changes of water demand and production over the life of the field in the Permian, and unfavorable economics for recycling within the Haynesville due to poor initial water quality and quantity.[11] While midstream water infrastructure (e.g., storage, conveyance, and treatment) may help to mitigate some of these challenges, widespread development of midstream water infrastructure within a basin often requires both substantial capital investment and time.[11] Thus, recycling is highly dependent on regional and local conditions. As such, widespread changes in industrial practices will likely require prioritized research in characterization, adaptable treatment trains, logistics, and regulations rather than broad improvements in treatment technologies. For example, if technology goals were guided by innovative, cost-effective toxicity removal approaches that are end use specific, this would be a disruptive change that fundamentally alters existing practice and revolutionizes treatment process engineering for complex waters. The following two case studies highlight limitations and state-of-the-art management practices to expand water recycling within the Permian and Anadarko Basins.

3.1.1. Disposal and Recycling: Permian Basin, Texas. Texas has cultivated a logistical and regulatory environment where recycling of produced water may be technically but not economically feasible.[51] Texas has a plethora of SWD (~8000 wells) with an estimated average pressure utilization of ~65% within the Permian Basin.[52−54] Consequently, approximately 54% of the 9.9 billion barrels of produced water generated in 2017 were disposed of via SWD wells.[1] In contrast, Texas has fewer facilities for the treatment and recycling of produced water, which can exacerbate economic and logistic challenges and lead to potential competition in water scarce years.[55] As such, the recycling of produced water (excluding EOR) within the Permian Basin in Texas is estimated at 10%−15%, albeit exact numbers are unknown.[11,51,56] Meanwhile, water use for O&G within the Permian Basin is currently estimated to be near 1.2 billion barrels (50 billion gallon/0.2 billion m^3) per year, with a potential to increase to an estimated 8.8 billion barrels (370 billion gallons/1.4 billion m^3) by 2030.[51] Thus, increasing the recycling of produced water within the Permian could provide an important avenue for meeting future water demand within the industry. However, over the life of the field, produced water volumes may overtake hydraulic fracturing water demand and limit the overall effectiveness of recycling operations, leading to planning and logistics concerns for operators.

The minimal treatment requirements, availability of SWD facilities, and associated reduced liability frequently enable SWD wells to be the most economic management option, in part due to the limited consideration of externalities. Traditional integrated treatment processes for injection include the separation of both suspended solids and oil and grease (e.g., gun barrel tanks, filtration) followed by chemical addition (e.g., antiscalants, corrosion inhibitors, biocides) to protect the well, formation, and related equipment.[2,12,57] A generalized flow diagram of this process is shown in Figure 2A.

WaterTAP3 analysis of SWD facilities in the Permian indicated an adjusted LCOW of \$1.11/m^3 (\$0.18/bbl) with an energy intensity of 0.41 kWh/m^3 (0.07 kWh/bbl), as shown in Figure 2B and C, respectively. These results are near those of actual SWD cost previously reported in the literature in the Permian that range from approximately \$2.50 to 12.50/m^3 (\$0.40−1.99/bbl).[43,58] Yet, conveyance, particularly trucking distances, is often a substantial portion of the cost for both SWD wells and recycling. Including conveyance, disposal fees are reported to range from approximately \$12.60 to 25.20/m^3 (\$2.00−4.01/bbl) within the Permian Basin.[43,58,59] Analysis in WaterTAP3 supports the idea that conveyance costs may be a significant driver for increasing recycling of produced water. For example, WaterTAP3 estimates a 9-fold increase in the adjusted LCOW if produced water is conveyed 50 miles via truck. While this effect may be mitigated in part through conveyance via pipeline, WaterTAP3 still estimates conveyance

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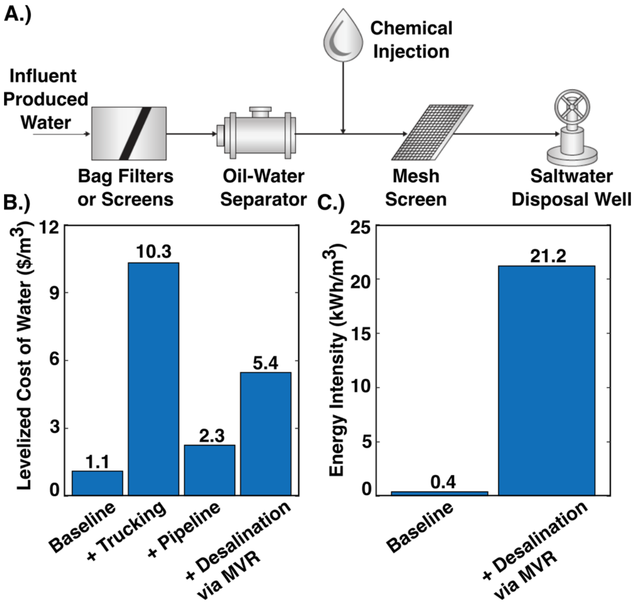

Figure 2. (A) Generalized process flow diagram of baseline treatment train for the disposal of produced water at an SWD facility. Variation in the (B) adjusted LCOW and (C) adjusted energy intensity of the baseline, baseline with 50 miles of conveyance via trucking, baseline with 50 miles of conveyance via pipeline, and baseline with intensified brine management. Additional details on the case study and WaterTAP3 inputs are available in the SI. Abbreviations are as follows: mechanical vapor recompression (MVR), levelized cost of water (LCOW), and energy intensity (EI).

via 50 miles of pipeline more than doubles the adjusted LCOW. Thus, there is interest in both shortening required conveyance distances and in developing and expanding oilfield water midstream operations (e.g., hydrovascular networks, storage, and centralized treatment facilities) within the Permian and other Texas basins.[60,61]

Recent changes (Texas House Bill 2771) have reduced permitting requirements for the discharge of produced water to only an NPDES permit from the Texas Commission on Environmental Quality (TCEQ).[62,63] The TCEQ permits still follow the federal effluent limitations of 40 CFR 435 as well as the state discharge effluent limitations laid out in 30 TAC 307. By easing this permitting process, the Texas legislature hopes to incentivize discharge of produced water for beneficial environmental purposes in the drought-stricken western portion of the state and potentially begin the process of assessing produced water for aquifer recharge.

Thus, intensification of treatment at either SWD facilities or centralized treatment facilities could enable water recovery from produced waters destined for disposal. In particular, pretreatment and desalination utilizing MVR to recover water has been evaluated and field tested in the Barnett Shale Play in Texas.[64,65] In this approach, produced water-precipitated iron, organics, and suspended solids are removed via coagulation and sedimentation. A set of modular MVR units then desalinates the produced water to generate a stream that can be recycled.[65] Yet, WaterTAP3 modeling indicates a nearly 5-fold increase in the adjusted LCOW of the process, with the MVR accounting for 83% of the adjusted LCOW and ~90% of the electricity intensity. The contributions of each unit process to the adjusted LCOW and electricity intensity are shown in Figures S5 and S6 in the Supporting Information. Thus, while recovering water and decreasing disposal volumes may be useful in semiarid and arid regions, the high cost may inhibit economic feasibility. Furthermore, there may still be toxicity concerns and additional treatment requirements for sustainable, beneficial reuse of the treated water recovered from produced water.

3.1.2. Recycling: Anadarko Basin, Oklahoma. Oklahoma, like Texas, does not explicitly quantify the recycled volume of produced water. Of the approximately 2.8 billion barrels (118 billion gallons/0.445 billion m^3) of water produced in 2017, the Oklahoma Corporation Commission (OCC) reported that approximately 45% and 55% were reinjected for EOR and SWD, respectively.[1] An increase in seismic events linked to hydraulic fracturing operations and SWD injection volumes, specifically within the Oklahoma Area of Interest (AOI), heightened interest in limiting disposal and promoting other produced water management techniques. Induced seismicity, particularly in the context of injection and seismicity in Oklahoma, has been a focus of past studies and is outside the scope of this perspective.[5,66−68] However, future regulations that limit SWD to mitigate induced seismicity concerns may lead to recycling of produced water or other management techniques.

The OCC's O&G Conservation Division has endeavored to reduce disposal volumes via SWD wells within the Oklahoma AOI to 40% below 2014 injection volumes to mitigate the risk of induced earthquakes.[69] To accomplish this objective, Oklahoma has begun to incentivize the development of centralized produced water treatment facilities and conveyance systems through regulatory measures (OK SB 1875), like those clarifying issues of ownership and liability of produced water while being transported to centralized treatment facilities and back to wells for recycling.[70] Clearly defining the ownership of and responsibility for produced water during conveyance allows O&G companies to better assess and manage the risks of conveying and treating produced water at centralized facilities that could be owned by a third-party company. However, logistics and costs for the conveyance of produced water could hinder recycling and beneficial reuse of Oklahoma produced water. Thus, the Oklahoma Produced Water Working Group (PWWG) anticipates that cooperative expansion of water distribution systems would likely facilitate increased recycling of produced water within (and potentially between) Oklahoma's STACK and Mississippi Lime plays.[70]

Newfield Exploration Company, an operator in the STACK play, installed an extensive network of storage, conveyance, disposal, and treatment systems to enable recycling of produced water.[11] This network includes approximately 150 miles of HDPE pipe, 10 million barrels (420 million gallons/1.6 million m^3) of freshwater storage, and 5 million barrels (210 million gallons/0.8 million m^3) of treated produced water storage. Newfield's extensive pipeline infrastructure has reduced truck traffic by approximately 60,000 round trips per year.[11] Along with reduced truck traffic, pipeline conveyance may reduce the chance of spills and labor costs.[49,71]

Newfield's Barton Water Recycle Facility provides sufficient treatment of produced water for recycling within the upstream O&G industry. The facility is capable of processing approximately 30,000 barrels per day (1.26 million gpd/4800 m^3/day) from approximately 40 well sites.[11,72] A simplified process flow diagram is shown in Figure 3. Influent wastewater is pumped into a series of tanks, where both insoluble organics and solids are removed. Effluent water from the pretreatment

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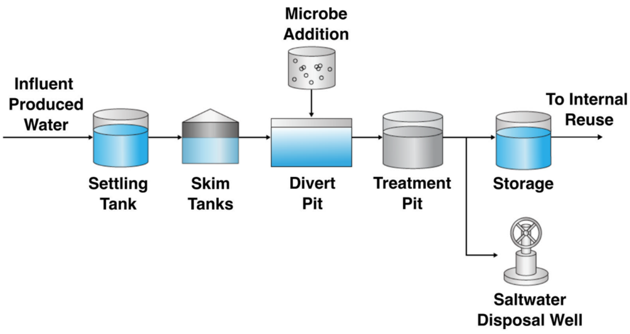

Figure 3. Simplified process flow diagram of Newfield Exploration Company's Barton Water Recycle Facility.

process is then transferred to aerated biological treatment holding ponds to oxidize contaminants; the hydraulic residence time in these ponds is approximately 21 days. Following biological treatment, the water reaches a quality suitable for recycling.[72] Water that is not recycled is disposed of via SWD wells.[73] The capital cost of the entire piping and treatment network was approximately \$90 million, and Newfield incurs roughly \$1.26/m^3 (\$0.20/bbl) to treat the water. Chemical costs are constant across the process.[11]

The OCC's continued effort to divert produced water management from SWD wells to centralized treatment facilities may help to increase the recycling of produced water. While the expansion of both recycling and beneficial reuse could aid in the economics of centralized treatment facilities, the Oklahoma PWWG identified that beneficial reuse is limited in part by a lack of toxicological understanding of risk to both the environment and public health for many commonly proposed beneficial reuse options.[70] These environmental and human health concerns must be addressed to expand the beneficial reuse of produced water responsibly and sustainably.

3.2. External Beneficial Reuse. The responsible beneficial reuse of produced water often necessitates extensive fit-for-purpose treatment and management practices. Beneficial reuse of produced water often raises concerns associated with the contamination of surface and groundwater,[49] plant health,[74,75] soil contamination,[76−78] and human toxicity that limit industry and public acceptance.[79−82] Frameworks and regulations are needed to address toxicology risks and public acceptance issues for beneficial reuse (discussed in Section 4).

Consequently, beneficial reuse of produced water generally requires the removal of constituents such as oil and grease, total suspended solids (TSS), TDS, biological oxygen demand, chemical oxygen demand, and pathogenic bacteria via conventional and advanced water and wastewater treatment processes.[3,12] More highly regulated beneficial reuse of produced water may also require further treatment for removal of constituents such as selenium, boron, radionuclides, and low molecular weight organics via desalination, advanced oxidation, and adsorption processes. For beneficial reuse, treatment trains might require a process dedicated to reducing the toxicity of the treated produced water to ensure public and environmental safety. Recently, promising studies have shown that photocatalysis-driven advanced oxidation processes (ozonation, rare metal catalyst, Fenton processes) can greatly reduce produced water toxicity.[83−88] Other technologies and processes have shown a potential for reducing toxicity, such as adsorption (granulated activated carbon, powder activated carbon, zeolite),[89,90] ion exchange, and biological treatments (activated sludge, MBR)[91−94] but require more research to determine their applicability for produced water treatment.[27] Furthermore, if primary, secondary, and tertiary treatments are needed to meet effluent requirements, ensure sustainability, and address public concerns, the resulting treatment system may not be economically viable. Some states, such as New Mexico, have passed regulations that prioritize beneficial reuse and promote research to balance the protection of human health and the environment with increased beneficial reuse of produced water to limit reliance on freshwater resources.[95] New Mexico also established a research center to identify needed technology and regulation research to address public health concerns and overcome the economic and safety challenges limiting produced water recycling.[96]

In particular, beneficial reuse of high salinity produced water remains a major challenge due to the current limited options for cost-effective desalination treatments that generate water of adequate quality for potential beneficial reuse, paired with the large volume of residuals that must be managed. Beneficial reuse of produced water with TDS as low as 2000 mg/L may require desalination via membrane-based (NF and RO) or thermal-based (eutectic freezing and MVR) processes to meet effluent requirements. If treatment of high salinity produced waters (TDS > 75,000 mg/L) were extended to ZLD, the volume of solid salts generated would be overwhelming. For example, produced water in the Permian Basin routinely exceeds TDS concentrations of 95 g/L that are mostly NaCl, and the produced water in the basin is expected to exceed 13,000 billion gallons (49 billion m^3) over the lifetime of the play (the next 50−100 years).[2] Assuming an average TDS of 100 g/L and a 100% water recovery, one O&G basin will generate nearly 4.92 billion metric tons of salt over the lifetime of the play, highlighting the potential salt management issues if ZLD was employed industry wide.[2,97,98] The salt, naturally occurring radioactive material (NORM), and brine residuals management associated with desalination and advanced treatment is nontrivial, and the economics of managing these residuals, whether through ultimate disposal or solidification, need to be considered in assessing the overall economic feasibility of specific treatment operations.

Enhanced resource recovery of either water or valuable constituents (e.g., insoluble hydrocarbons, lithium, iodine) may help offset the cost of produced water treatment.[8,9,11] For example, prior to disposal, recycling, or beneficial reuse of produced water, insoluble hydrocarbons may be recovered using conventional oil−water separation techniques[99−101] or novel higher-efficiency, modular methods.[102,103] In contrast, recovering inorganic compounds from the concentrated brines generated by treatment processes is often more technically and economically viable. Yet, advances in solute-tailored functionalization of membranes for enhanced selectivity could enable resource recovery earlier in the treatment process.[104−107] Further research must identify regions where saleable constituents are present in sufficient concentrations to have economic potential, as well as the fit-for-purpose solutions to best extract and refine these constituents.

While treating and adopting produced water for beneficial reuse poses greater challenges for achieving the desired water quality and pipe parity, some existing state-of-the-art management practices have already enabled the beneficial reuse of produced water. The following case studies discuss beneficial reuse scenarios with more stringent effluent standards and limitations: (1) recycling environmental surface discharge and

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resource recovery in the Marcellus Basin, (2) streamflow augmentation in the DJ and Powder River Basins, (3) agricultural irrigation in the San Joaquin Basin, and (4) municipal reuse in the Powder River Basin.

3.2.1. External Reuse, Recycling, and Surface Water Discharge: Marcellus Basin, Pennsylvania. Treatment, recycling, and beneficial reuse of produced water within the Pennsylvania portion of the Marcellus Basin is incentivized due to water scarcity and regulatory limitations for underground injection. The combination of Pennsylvania's geology and state regulations (25 Pa Code § 78.18) has resulted in fewer than 15 permitted SWD facilities in the state—culminating in the recycle of approximately 90% of produced water generated in Pennsylvania.[50,108] Pennsylvania has passed legislation that encourages the development of centralized treatment facilities and conveyance systems by streamlining permitting and reducing regulatory requirements for storage and conveyance (25 Pa Code § 299). Residual brine streams and remaining produced water are often transported to Ohio, where SWD regulations are less stringent, for injection at a total cost of up to \$94−126/m^3 (\$14.94−20.03/bbl).[11] Therefore, the O&G industry is financially motivated to pursue innovative options that enable both recycling and beneficial reuse of produced water within the Marcellus.[109]

Eureka Resources operates multiple facilities that receive produced water from more than 200 wells and multiple operators in the Marcellus Basin.[110] These centralized water treatment facilities include combinations of the treatment processes described in the following section to generate multiple qualities of effluent water, while, when possible, valorizing residuals.[73] The specific processes and extent of processing vary by facility based on multiple factors, including customer needs and influent water quality. A simplified, generic process flow diagram of the Eureka Resources process is shown in Figure 4.

In general, the treatment facilities have reported capacities of 4000−10,000 barrels (168,000-420,000 gallons/636−1590 m^3) of produced water per day.[109] Influent produced water is screened for various water quality parameters (e.g., screened by pH and concentrations of TDS, methanol, and TSS) before entering the treatment train. While influent produced water

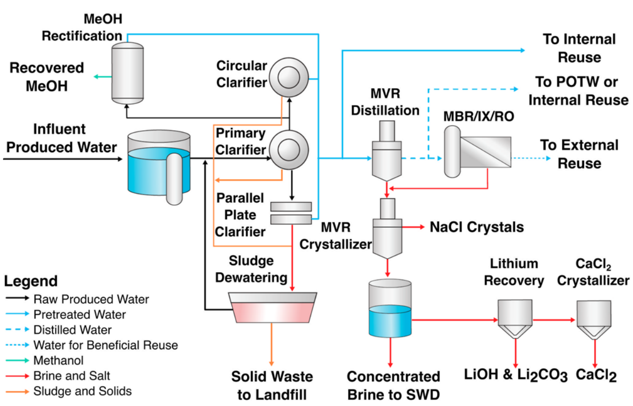

Figure 4. Simplified process flow diagram of various processes employed at Eureka Resources' centralized produced water treatment facilities.[110,111,114] Abbreviations are as follows: methanol (MeOH), mechanical vapor recompression (MVR), ion exchange (IX), membrane bioreactor (MBR), reverse osmosis (RO), publicly owned treatment works (POTW), and SWD well.

quality can vary substantially, the TDS are typically 100,000 mg/L.[111,112] Treatment costs may increase due to factors including increased chemical usage for produced waters with TDS in excess of 150,000 mg/L.[111,112] Oil and solids are first separated in a primary clarifier. Recovered oil is reclaimed for reuse, while the solid stream is dewatered prior to landfill disposal.[110,113] Eureka Resources estimates that 10−30 tons of solid waste per day is generated for produced water treatment plant with a capacity of 4000 barrels (168,000 gallons/636 m^3) per day.[111] At some facilities, water with elevated methanol concentrations (>500 mg/L) is pretreated using a rectification column to recover methanol for reuse within the O&G industry prior to further treatment.[109,114] The resulting methanol purity is typically 97%, with trace contaminants (e.g., BTEX, acetone, zinc, calcium, aluminum, water).[114] Deoiled produced water flows through the primary clarifier and then one of two parallel clarification treatment trains. While one treatment train is designed for higher TSS streams (e.g., drilling fluids), each train includes pH adjustment and chemical addition, a flash mixer, a clarifier, and an equalization tank.[73,109] Effluent water is classified as pretreated wastewater and may be used for internal purposes within the O&G industry.

Pretreated wastewater is desalinated via MVR to produce distilled water that may be recycled or discharged to publicly owned treatment works.[109] This distilled produced water may be further treated with an MBR followed by ion exchange and RO to generate water that meets Pennsylvania's dewasting effluent standards (Pennsylvania Department of Environmental Protections' WMGR123 Appendix A General Permit Requirements) for direct discharge to surface waters.[73,113] While high-purity MVR distillate traditionally does not require further polishing, the advanced treatments are necessary to meet the dewasting standards due to more stringent requirements for the WMGR123 permit. To further evaluate this extended treatment train performance, Eureka Resources worked with the Center for Sustainable Shale Development to conduct additional toxicity and characterization testing with the goal of mitigating toxicity.[109]

Eureka Resources endeavors to valorize residual streams from desalination and advanced treatment through resource recovery from the concentrated brine generated during desalination.[73,109] Concentrated brine (either NaCl or 20% $CaCl_2$) generated during this process can be reused as a drilling fluid additive, treated via an advanced brine management train, or transported to an SWD facility for disposal.[73,109,110] Eureka Resources' facilities may also produce NaCl crystals via crystallizer for reuse outside of O&G.[109,113] Finally, Eureka Resources is currently in the process of developing lithium and $CaCl_2$ precision separation technology to further valorize the brine generated during this process.

Ultimately, this case study highlights the relevance of flexible, fit-for-purpose produced water treatment that includes resource recovery valorization and reductions in produced water disposal.[73,109,114] Yet, the viability of this extensive treatment process is contingent on the elevated cost of produced water management in Pennsylvania.[11] Eureka Resources has reported treatment costs for reuse of flowback and produced water of \$40.63−62.50/m^3 (\$6.50−10.00/bbl).[111,112] For more extensive treatment, the reported crystallization costs are approximately \$68.75/m^3 (\$11.00/bbl), with the potential to reduce to approximately \$43.75/m^3 (\$7.00/bbl) with integrated, commercially viable resource recovery.[111,112] Thus, while this treatment and management approach is often viable within the Marcellus, the elevated cost of treatment and management far exceeds disposal or recycle costs within the Permian and Anadarko Basins. Further, relatively similar facilities, even within the Marcellus, have been idled, highlighting the often volatile economic viability of produced water reuse.[115] In particular, shifts in the price of oil affect exploration, production, and consequently the generation of produced water. This instability in the produced water supply may undermine the profitability of these large, centralized treatment facilities. However, management approaches like this may become more common if concerns related to the adequate treatment of produced waters and our understanding of the risks associated with beneficial reuse alter regulatory requirements.

3.2.2. Streamflow Augmentation: Denver−Julesburg Basin, Colorado, and Powder River Basin, Wyoming. To enable streamflow augmentation with produced water, operators need to meet and receive local and NPDES permits for specific contaminants and water qualities. An example is a permit that an operator has requested from the Colorado Department of Public Health and Environment for discharging treated/blended produced water from a tight shale formation into the South Platte River and St. Vrain Creek in the DJ Basin in northeastern Colorado (PEL230027). Water from a large network of pipelines in the basin that contains a mix of purchased surface and groundwater and treated produced water (MBR or RO desalination with conventional and ion exchange pretreatment) (first dilution step) is discharged to streams (second dilution step). The discharge limit is based on an elaborate mass balance of various constituents in the pipeline produced water and the water flow rate in the two streams. For the DJ produced water considered in this evaluation, the ratio between discharge flow and streamflow ranges from 1:43 (St. Vrain Creek) and 1:114 (South Platte River) for chronic low flow that represents the 30-day average low flow recurrence in a three-year interval. No data regarding the presence or quantification of organics were submitted to the division with the Preliminary Effluent Limits (PEL) application; therefore, testing for organics would be submitted with the permit application to determine which specific organic compounds are present and need to be removed.

Another example of surface discharge for streamflow augmentation was described by Plumlee et al. in a study that presented a decision support tool that was developed as part of a new framework for produced water treatment and beneficial reuse.[6] Average water quality was obtained from approximately 90 wells in the Powder River Basin in Wyoming, with an average TDS concentration of ∼900 mg/L; this TDS concentration is less than the limit for irrigation (2000 mg/L) and for livestock watering (5000 mg/L) in Wyoming. With respect to streamflow augmentation, the average conductivity values met standards for the Powder River, but the average sodium adsorption ratio (SAR) exceeded the standard. Thus, the produced water would require post-treatment prior to any stream discharge. The decision support tool developed by Plumlee et al. proposed a short treatment train consisting of chemical disinfection, media filter, and potentially NF desalination for sensitive receiving streams. That said, considering the high quality of the raw produced water, the decision support tool gave higher priority to beneficial reuses that demand higher water quality (e.g., aquifer recharge, fisheries, irrigation, livestock watering, and other industrial applications) than streamflow augmentation.

3.2.3. Agricultural Irigation: San Joaquin Basin, California. Treatment of produced water for irrigation could help alleviate the water demand of drought-stricken states with large agricultural practices, such as California and Texas. Currently, the beneficial reuse of conventional produced water for agricultural purposes is done on a small scale, and one operation within the San Joaquin Basin in Kern County, California, has been in operation for over 20 years. Federal regulations in 40 CFR 435 allow for the discharge of O&G produced water for environmental or agriculture use. In California, the Regional Water Quality Control Boards manage the NPDES program and require companies discharging produced water for agricultural uses to provide a list of chemicals added to the water before drilling, the volume of produced water being discharged, and evidence that the discharged produced water meets the effluent limits set by the board for irrigation use. However, the use of produced water for irrigation in Kern County should be viewed as an exception that may not be replicable on a national scale because the background TDS concentrations of produced water in parts of Kern County are very low (<1000 mg/L), and the produced water is co-located with elevated agricultural irrigation demand.[2,116] For wider use of produced water for agriculture in other oilfield play, cost-effective treatment technologies are needed to reduce high TDS values in other regions to a level similar to Kern County.

The Cawelo Water District ponds, located in Kern County, California, receive treated produced water from neighboring O&G sites, blend it with irrigation water (up to 50% produced water), and then distribute the water for agricultural irrigation (e.g., citrus fruits, nuts). The ponds have consistently provided reclaimed produced water for agricultural beneficial reuse (longer than any other facility) since the early 1990s and constitute one of the only such facilities in the US. Due to the age of this facility and the requirements of the water district, there is a large amount of historical data on the process and environmental impacts associated with this facility.[116−119]

One treatment facility, Station 36, upstream of the Cawelo ponds can treat 900,000 barrels (38 million gallons/143,000 m^3) per day of produced water from the Kern River oil field. The Cawelo ponds may also receive up to 175,000 barrels (7.4 million gallons/27,800 m^3) per day of water from the Valley Water Management Company from the Kern Front oil field. Treatment of high-quality produced water for agricultural beneficial reuse in Kern County focuses on oil−water separations before blending with surface water and pumped groundwater to lower the concentration of dissolved constituents such as As, Na, B, Cl, and Se. As shown in Figure 5A, produced water entering the treatment facility first undergoes mechanical/gravity separation, followed by sedimentation, air flotation, and finally filtration through walnut shell filters. Most of the pretreated water is pumped to a series of reservoirs for polishing (evaporation of VOCs), blending, and eventual transfer to the agricultural irrigation systems.[119]

Beneficial reuse of produced water in Kern County is enabled by the proximity to agricultural needs, low salinity, and low concentrations of constituents like boron. These factors allow for the treatment train to focus on low-cost oil removal, while subsequently utilizing blending with freshwater to lower the concentration of toxic constituents. WaterTAP3 analysis indicates that the adjusted LCOW for the baseline Kern

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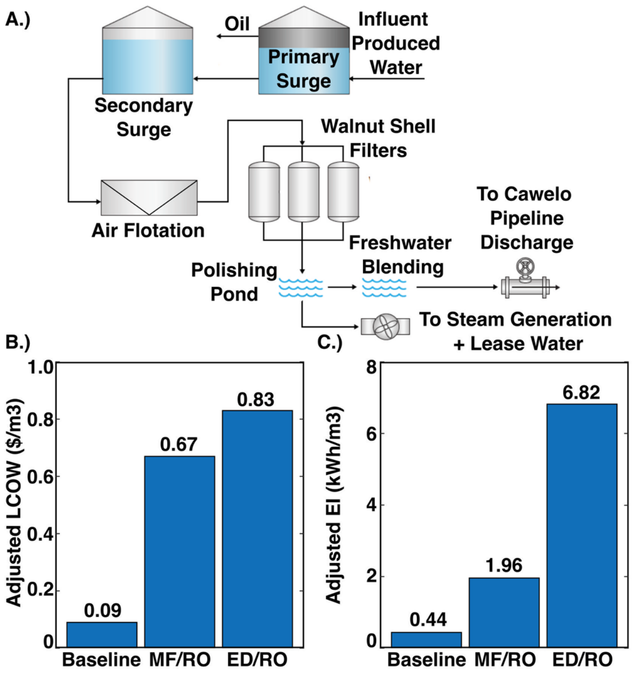

Figure 5. (A) Simplified process flow diagram of the produced water treatment train for agricultural irrigation in Kern County.[119] Variation in (B) adjusted LCOW and (C) adjusted energy intensity of the baseline, baseline with additional ED/RO treatment for elevated boron, and baseline with additional MF/RO treatment for elevated TDS. Additional details on the case study and WaterTAP3 inputs are available in the SI. Abbreviations are as follows: levelized cost of water (LCOW) and energy intensity (EI).

County scenario is \$0.09/m^3 (\$0.01/bbl) with a corresponding energy intensity of 0.44 kWh/m^3 (0.07 kWh/bbl) as shown in Figure 5B and C, respectively. The contributions of each unit process to the adjusted LCOW and electricity intensity for the Kern County baseline study can be seen in Figures S10 and S11 in the Supporting Information, respectively. As with the SWD case studies, increasing the required conveyance distances reduces the economic viability of the produced water management option. WaterTAP3 analysis indicates that increasing either the piping or trucking distance to 50 miles will increase the adjusted LCOW to \$0.04/m^3 (\$0.01/bbl) and \$0.07/m^3 (\$0.01/bbl), respectively.

Yet, blending may not be sufficient for sustainable soil and plant health when irrigating with lower quality produced water.[74,75,120] When the background TDS concentrations are higher (as is typical in other O&G plays), the water will require further pretreatment and desalination to remove salts before use for irrigation, which will result in an overall higher cost of treatment in order to meet SAR guidelines to prevent soil sodicity. Due to these sensitivities, agriculture irrigation requires a much higher water quality than does SWD or recycling. For a brackish produced water with TDS of 10,000 mg/L, WaterTAP3 indicates that incorporating desalination approaches like microfiltration (MF) with RO increases the adjusted LCOW of the blended water stream to \$0.67/m^3 (\$0.11/bbl) with a corresponding energy intensity of 1.96 kWh/m^3 (0.31 kWh/bbl) The contributions of each unit process to the adjusted LCOW and electricity usage are shown in Figures S10 and S11 in the Supporting Information, respectively. Yet, as most produced waters have much higher salinities, these values and this approach would still likely have limited applicability.

Furthermore, for produced waters with elevated boron concentrations (>0.5 mg/L), treatment processes may be necessary to meet recommendations for protecting plant and soil health.[74,121] Common methods for boron removal in produced waters include adsorption, ion exchange, and membranes (e.g., RO, electrodialysis (ED), electrodialysis reversal (EDR)).[122] Efficient boron removal in RO membranes often requires operation at high pH, as nonionized boric acid is the dominant species in relevant pH ranges (i.e., 6−8) and may diffuse through the membrane.[123−125] Yet, high pH may exacerbate the fouling of the membrane surface (e.g., calcite scaling).[126−129] One novel approach utilizes a hybrid ED/RO membrane system to reduce the concentration of anions and cations via ED, increase the pH to alter boron speciation, and remove boron via RO.[124,130] WaterTAP3 analysis of a hybrid ED/RO membrane system and baseline pretreatment for enhanced boron removal produces an adjusted LCOW of \$0.83/m^3 (\$0.13/bbl) with a corresponding energy intensity of 6.82 kWh/m^3 (1.08 kWh/bbl). The WaterTAP3 simulation models ED performance; however, replacing the ED unit with an EDR unit could potentially reduce the LCOW and energy intensity of these treatment trains. The contributions of each unit process to the adjusted LCOW and electricity usage are shown in Figures S10 and S11 in the Supporting Information. While these case studies consider the presence of both elevated salinity and boron, the presence of other recalcitrant constituents could require additional treatment processes, further increasing both the adjusted LCOW and energy intensity of the treatment approach. Furthermore, with agricultural water costs in California generally ranging from \$0.014 to 0.89/m^3 (\$0.002 to 0.14/bbl), these options may not be economically viable at the present time.[131]

While similar treatment processes for produced water beneficial reuse in agricultural irrigation may have the potential for adoption in other low salinity basins (e.g., San Juan Basin, Raton, Powder River Basin), expansion to basins with higher salinity produced water with trace contaminants and larger required conveyance distances are unlikely to be economically feasible. Prior assessments of minimally treated CBM from the Powder River Basin for use in agricultural irrigation has demonstrated that minimally treated CBM produced water allows for the short-term growth of crops, with negligible detrimental effects to the crops, but accelerates the long-term degradation of soil health.[132−134] Furthermore, there has been some social backlash from consumers to the beneficial reuse of produced water for agricultural irrigation.[135] To combat these public concerns, regulatory agencies have performed sampling tests for the health of crops irrigated with produced water (e.g., almonds, garlic, mandarins) and tests on the soil health irrigated with the produced water.[136−138] Additional research into human and ecological toxicity may be necessary to confirm the safety or dictate additional treatment needs for widespread use of treated produced water for agriculture irrigation.

3.2.4. Municipal Reuse: Powder River Basin, Wyoming. While beneficial reuse options like irrigation and streamflow augmentation are of interest in semiarid regions, water scarcity in severe drought-stricken regions may justify assessing the viability of municipal reuse or even direct potable reuse (DPR). In particular, many regions with significant CBM production experience water stress, which could be partially

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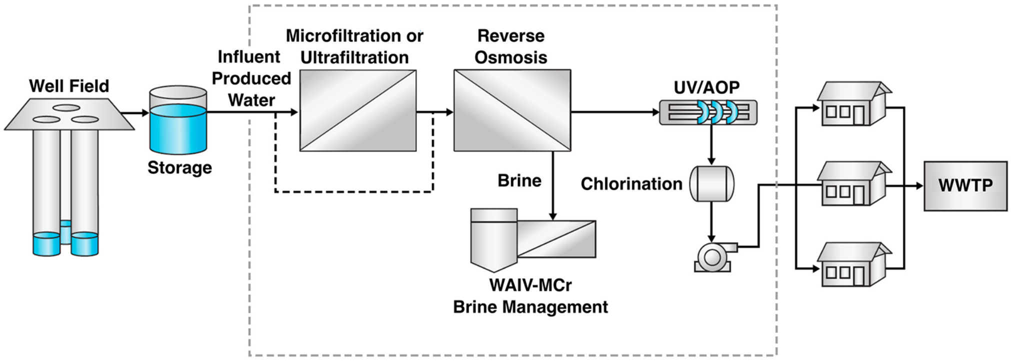

Figure 6. Process flow diagrams for a proposed centralized treatment systems for municipal reuse. Abbreviations are as follows: Wind-Aided Intensified eVaporation and Membrane Crystallization (WAIV-MCr), ultraviolet (UV), and advanced oxidation process (AOP). Additional details on the case study and WaterTAP3 inputs are available in the SI.

mitigated through the beneficial reuse of low salinity CBM produced water. For example, in the Damodar Valley Basin in eastern India, a techno-economic analysis indicated that RO-treated CBM produced water could provide high-quality water for an estimated 3.5 million people over 20 years.[139] Another techno-economic analysis by Meng et al. suggested that RO-treated unconventional and conventional produced waters in California could provide drinking water for one million residents per year.[131]

Singh and Colosi evaluated the feasibility of DPR of CBM produced water from the Damodar Valley Basin for both centralized and decentralized RO treatment systems.[139] In the centralized system, the RO-treated produced water is pumped through a pipe network into homes. In contrast, in the decentralized system, raw produced water is treated in homes for point-of-use RO treatment. For both cases, a non-commercial desalination technology, Wind-Aided Intensified eVaporation and Membrane Crystallization (WAIV-MCr), was modeled for brine management. WAIV-MCr is a process that concentrates the brine 10 times through evaporative processes, and a subsequent membrane process further intensifies the brine, resulting in solid salt byproducts, a clean water stream (50% recovery), and a membrane brine purge.[140] WAIV-MCr may have limited applicability due to its ambient condition requirements (e.g., wind) and its inability to recover freshwater.[141] While Singh and Colosi evaluated RO as the only treatment step in the centralized DPR treatment train, the WaterTAP3 results presented herein focus on the centralized treatment train and incorporate UV inactivation and addition of chlorine to comply with US drinking water regulations (e.g., disinfection credits, chlorine residual) (Figure 6). These additional treatment steps were added to inactivate pathogens and prevent microbial regrowth in the distribution system.

The potential for municipal reuse of produced water in the US was evaluated using CBM produced water from the Powder River Basin. WaterTAP3 analysis of the modified centralized treatment train indicates an adjusted LCOW of approximately \$2.20/m^3 (\$0.35/bbl) with an energy intensity of 1.40 kWh/m^3 (0.22 kWh/bbl) for a 4500 m^3/day treatment facility. Similar to the original analysis, greater than 50% of the adjusted LCOW can be attributed to the WAIV-MCr (Figure S13), highlighting the importance of cost-effective brine and residual management approaches in achieving pipe parity.[139] Thus, improvements in either the brine management technologies or co-location of the centralized treatment facilities with SWD could help to improve the economic viability of this produced water management approach. Yet, while the estimated LCOW is above the municipal water costs from freshwater sources (\$0.30−0.80/m^3 or \$0.05−0.13/bbl), they are relatively similar to those of brackish water or seawater desalination for municipal use (\$0.90−1.70/m^3 (\$0.14−0.27/bbl) and \$1.80−4.20/m^3 (\$0.29−0.67/bbl), respectively) and could provide a potential solution for rural homeowners rather than build out brand new municipal water treatment districts.[142] In response to growing water scarcity, some municipalities have started to rely on brackish and seawater desalination facilities to provide municipal water, and thus, CBM produced water for municipal reuse may be financially feasible.

However, while it may be economically viable for these treatment trains to achieve potable drinking water standards, the lack of toxicological information on unregulated constituents in this water limits the ability to safely adopt low salinity produced water for potable use. The general lack of toxicological studies on produced water poses both a scientific and a social hurdle in the adoption and general public's acceptance of these waters for more sensitive uses (e.g., food crop irrigation and municipal usage). Furthermore, many states lack regulations concerning municipal wastewater DPR and those that do have complex regulations and limitations concerning DPR.[143] Given the social and regulatory hurdles encountered during the implementation of DPR with municipal wastewater, it is difficult to imagine the drivers that would enable DPR with produced water in the US. Ultimately, responsible, sustainable beneficial reuse of produced water may require holistic chemical characterization and toxicological assessment to inform treatment train development to appropriately mitigate risk to the public and the environment (as depicted in Figure 1).

4. CHEMICAL CHARACTERIZATION AND TOXICOLOGICAL CHALLENGES

The complex chemistry of produced water creates characterization challenges that, when coupled with the understudied nature of drilling/fracturing fluids and the transformation products that form in the well, leads to difficulties in evaluating the success or failure of produced water treatment trains. Current research is attempting to overcome some of these challenges in characterizing produced water[31,144] and suggests that numerous unregulated or proprietary chemicals may be present in any particular untreated produced water stream[25,145,146] and therefore potentially exist in treated

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produced water. The difficulty in characterizing constituents in produced waters, in addition to the lack of data, has likely hampered policymakers and regulatory agencies in developing policies that enable advanced treatment of produced water to facilitate alternative water reuse. This, coupled with the lack of data on completion and production chemicals, and with clear and defined treatment goals, likely creates concerns that the water may not be adequately characterized or treated for the desired end use. Therefore, future research efforts must identify analytical and bioanalytical methods for indicator/priority compounds or transformation products in produced water. These indicators, which would serve as metrics for the evaluation of treatment technologies for beneficial use of produced water, must capture the breadth of water quality concerns for the targeted end use.

There is a critical need for developing rigorous protocols that ensure sustainable management and beneficial reuse of treated produced water. Previous studies have identified these needs and suggested that such protocols should include components to address monitoring, process control, treatment effectiveness, and potential environmental and health risks. In addition, phased approaches for evaluating produced water management options have been developed (GWPC) that incorporate both initial and beneficial reuse evaluations, legislative and regulatory assessment, logistics, economics, and benefits (Phase I).[11] Evaluation is followed by the identification of contaminants for treatment and risk analysis using pilot testing and effluent characterization (Phase II), traditional risk assessment methodologies (Phase III), and risk management (Phase IV).[11] While these recently developed approaches have significant potential for addressing produced water management at scale, they have failed to address the underlying gaps associated with assessing the suitability of produced water for particular end uses.

Untreated produced water matrix complexity (e.g., salts, organics, microorganisms) creates challenges for results in the application failure of traditional water and wastewater analytical methods, often due to interferences induced by specific constituents present at high concentrations in these waters and a dearth of analytical methods for analyzing unknown organic compounds present in these fluids and brines. Because an appropriate array of indicators and bioanalytical tools has yet to be identified or developed, these current methods fail to connect treatment process selection to chemical- or toxicity-based end points relevant for a specific use. Consequently, the systematic framework required to properly assess the chemical or toxicity end points of a waste product as complex as produced water has not yet been developed. Thus, while Figure 1 highlights the need to incorporate toxicity end points for particular end uses, an appropriate framework to link technology performance to end use specific toxicity measures is still lacking.

In terms of bioanalytical tools, several studies have allowed for a basic assessment of which particular fractions might contribute the most to certain toxicological end points, but they have rarely extended to whole organisms. This work has been performed on various model invertebrate and vertebrate systems and has been carried out almost exclusively with raw produced water. End points that have been studied include mortality (LC50),[147] developmental effects in early life stages,[148] endocrine disruption,[147,149] estrogenicity,[150] physical and behavioral impairment,[78,150,151] cardiovascular effects,[152−154] oxidative stress,[155,156] ionoregulatory stress,[151] metabolic stress,[157] and genotoxicity.[150] While these studies have contributed to our general knowledge of the toxicity associated with raw produced water, none have outlined approaches that could be followed to assess the toxicity of treated produced water.

Treated produced water matrices with, for example, lower salt and organics will be substantially different from the raw water matrix. A common practice of diluting water samples before analysis often results in the inability to detect compounds that were originally present at low concentrations. Not only does this practice create challenges in comparing pretreated and post-treated water, but it also may result in a gap in assessing synergistic chemical interactions during toxicity assessments. The analytical challenges are compounded by the multitude of unknown compounds present in produced water from either the subsurface,[158] proprietary fracturing fluid additives,[159] subsurface transformation products,[160] or the formation of chemicals during treatment.[161] Thus, significant efforts are needed to either identify or find indicators to assess the toxicity of treated produced water for use outside the O&G industry.

4.1. Toxicological Considerations. Toxicity analysis, both acute and chronic, is the most reliable approach for determining the long-term suitability and safety of produced water for beneficial reuse. The use of toxicity assays for assessing treated effluent quality for surface discharges is often required. Whole effluent toxicity (WET) is employed within the NPDES permits program to evaluate the toxicity of the entire waste stream and should be considered as part of pretreatment compliance inspections of municipal wastewater treated effluent.[162] While the selection of methodologies for toxicity testing of treated produced water will likely vary depending on the desired end use, both short-term and long-term impacts need to be considered.

Acute toxicity, generally defined as an adverse outcome after short-term exposure, can be evaluated through classic toxicology methods such as LC50 assays[163] or focused on specific end points (e.g., estrogenicity to genotoxicity) using in vitro bioassays. The evaluation of chronic toxicity (an adverse outcome after long-term exposure) is challenging because it must consider the life cycle of an organism with respect to exposure duration and adverse effects.[164] This can make causal relationships difficult to determine because additional confounding factors can play a role concurrently during the exposure period (i.e., synergistic or antagonistic effects). Synergistic effects are adverse outcomes amplified by the presence of other compounds;[165] this is especially concerning because produced waters are complex heterogeneous mixtures with many compounds that have the potential to interact with other natural organic and inorganic matter. This brief review of treated produced water toxicity focuses on two of the alternative beneficial reuse pathways discussed above: beneficial reuse for crop irrigation and streamflow augmentation. While different end uses of treated produced water may have differing acceptable levels or optimal assessment tools for toxicity (i.e., cooling tower reuse vs irrigation for crops), the two examples selected represent beneficial reuse options of near-term interest that may have direct impacts on ecological systems and indirect impacts on human health.

4.1.1. Irrigation. The agricultural sector has the greatest potential for beneficial reuse of treated produced water[2] but also has potential for toxic exposures to a variety of end points/biological systems. Beneficial reuse for irrigation involves

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indirect human exposure through consumption of food products irrigated with treated produced water but also includes environmental exposure to animals, plants, freshwater, and soil systems. The long-term impact on soil health in cropland is an essential component of evaluating this beneficial reuse pathway and the potential accumulation of inorganic chemicals, such as simple salts, boron, arsenic, NORMs, or metals, that could prove detrimental to agricultural soil health. There has been little research to date in this area, with the only long-term use of treated produced water for irrigation occurring in the San Joaquin Basin, which was detailed in Section 3.2.3. This paucity is compounded since the water in that basin is not representative of produced waters found elsewhere in the US (primarily its low TDS values).

Recent greenhouse studies have investigated the irrigation of crops with diluted, untreated produced water to address several questions regarding the role of simple dilution when irrigating crops.[74,75] These projects demonstrated that even when salinity concentrations meet local irrigation recommendations, diluted produced water (e.g., 5% produced water, 95% freshwater) can still adversely affect soil health by impacting the soil's physical properties, changing the soil microbiome, and overall decreasing crop yield.[120] Similar studies suggest that diluted produced water induces greater suppression of the plant's immune response[74] and promotes plant stressors (i.e., saline or oxidative stress) that lead to greater decreases in crop yield compared to controls containing the same concentration of salts but no other known contaminants.[75] Thus, current dilution targets that focus solely on salinity are not necessarily appropriate; targeted end points should address soil health and plant toxicity as depicted in Figure 1.

The studies described above complement the analysis of agricultural fields in Kern County, California, that demonstrates an accumulation of sodium and boron in fields irrigated with diluted produced water, even in cases when the water met local criteria for boron (less than 1 mg/L) or contained only 17% more sodium than local groundwater.[116] In general, the effects of these inorganic contaminants on soil health and plant physiology are well understood,[166,167] so treatment approaches can be designed to reduce their impacts using existing knowledge. For example, the scenario options identified for the Kern County case studies include desalination and boron removal using membrane technology. Evaluation of the impact of these improvements on salt accumulation and soil and plant health is needed. However, even in this case, more work is also needed to determine if there are relevant synergistic effects involving inorganic contaminants within this complex mixture, and future toxicity analyses need to focus on the impacts of treated produced water on the soil fauna necessary for a healthy soil (i.e., bacteria, fungi, nematodes, or earthworms).

There is little to no research on how treated produced water impacts the surrounding environment and its many receptors (e.g., ecotoxicological risks to plants or wildlife). The only research to date is on spills or releases of untreated produced water that correlate O&G activity to increases in endocrine disrupting compounds (EDCs) in nearby surface water,[168−170] compounds that can cause adverse reproductive effects in freshwater organisms.[147] Thus, thorough characterization of the TOC remaining in treated produced water will be necessary because excess irrigation water could drain from agricultural fields into nearby surface water systems, and the uptake and accumulation of organic compounds found in flowback and produced water have been demonstrated in wheat plants.[79] Uptake and accumulation of complex organic molecules such as PAHs,[171] EDCs, personal care products, and pharmaceuticals have also been shown to occur in corn,[172] leafy vegetables,[173] wheat,[174] and root vegetables[175] through irrigation with reclaimed wastewater. While flowback and produced water likely does not contain any pharmaceuticals or personal care products, the uptake of these compounds into plants shows the potential for crops to transport similar complex organic molecules into the plant biomass. Overall, our understanding of plant uptake, the formation of toxic metabolites through plant metabolism,[176] and the potential for synergistic effects of these organic compounds with pesticides or inorganic compounds[177] remains limited. In the absence of rigorous research and analytical methods for assessing end use specific toxicity of treated produced water, it will be difficult to develop metrics to guide engineers and regulators and reassure the public.

4.1.2. Streamflow Augmentation. Streamflow augmentation is another method that O&G producers use to discharge their produced water. This practice is allowed in arid states west of the 98th meridian and is regulated under the NPDES permit system (Section 3.2.2). Requirements for obtaining these permits and effluent limits of specific parameters may be site specific and vary from state to state or by EPA region. For example, at a discharge site in Wyoming, the NPDES permit has specific effluent limits for oil and grease, TDS, specific conductance, chloride, sulfate, Ra226, and pH, while acute toxicity is also analyzed every 6 months.[7,80] This is in contrast to the NPDES permitting in Colorado that also requires an assessment of chronic toxicity.[80] Produced water discharged for streamflow augmentation typically undergoes some type of treatment prior to release to meet NPDES regulations for oil and grease levels (<10 mg/L at this site). The treatment prior to discharge can be through separators (heat, gravity, or chemical), settling ponds, flotation, and/or skimming.[7]

Analysis of streamflow augmenting produced water effluent at one site in Wyoming showed that these treatment trains are effective at meeting NPDES requirements at the discharge point. However, downstream four of the six regulated parameters increased in concentration, and specific conductance increased above the permissible limit for NPDES regulated effluent (only regulated at the effluent discharge).[7,178] At the discharge point, the produced water effluent underwent comprehensive chemical characterization that showed the presence of over 20 unregulated volatile/semivolatile organic compounds and three different types of surfactants.[7,179] Concurrent mutagenicity assessments of effluent at the discharge point showed increased mutation rates with four different mutation types when compared to a negative control, but these mutation rates decreased as the discharge flowed down the augmented stream.[80] This produced water discharge was eventually consumed by cattle downstream in an ephemeral lake, but consumption also occurred as close as ∼100 m to the effluent discharge. Eventually, the augmented stream terminated upon entering a perennial river or at times even dried out before reaching the river.[7] This example highlights the importance of linking treatment trains to end use toxicity measures as suggested in Figure 1. The presence of both salinity and organic compounds downstream of the discharge point suggest that more advanced treatment is needed. Both advanced oxidation and adsorption processes are capable of reducing toxicity. As a result, treatment trains such as that provided within the Eureka

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Resources case study may be necessary. More importantly, this work brings to question the use of the current NPDES permitting approach and highlights the need for thorough chemical characterization and toxicity assessments to fully understand the risks of discharging treated produced water to the environment. Within the context of discharge to natural water bodies, the presence of bioaccumulating contaminants in complex produced water discharges can represent another threat that must be considered.[180] These issues have added importance in the arid west, where produced water effluent might be the major water source of an ephemeral water body that is consumed by various animals/livestock; alternatively, if the effluent flows into a perennial river, it would eventually be utilized downstream for either agriculture or as a drinking water source.[181] While the cost of treatment and toxicity testing may deter streamflow augmentation with produced water, treatment process advances and improved toxicity testing approaches may prove to be cost effective in the future. Indeed, future toxicity testing may be less expensive and more informative than existing analytical tools such as high resolution chromatography/mass spectrometry.

4.1.3. Bioanalytical Tools. Toxicological characterization of produced water to date has primarily focused on diluted raw produced water and on either organic or inorganic fractionated components of raw water. Chemical separation techniques (e.g., solid phase extraction) have been used to partition organic components from inorganics without dilution, allowing for bioassay testing of each fraction at a variety of dilutions. However, we currently lack the standard separation/dilution methods to facilitate comparisons among research studies and between raw and treated samples. Future methods must address this limitation to allow the development of generalizable insights and produced water toxicological databases.

4.2. Proposed Method for Assessing Toxicity in Treated Produced Water. The complexity of produced water makes it difficult to establish methods to determine biological toxicity. However, even more challenging is the fact that each produced water has a distinct composition, and when volatile organic compounds dissipate, the composition and toxicity of the water will change. This makes it exceedingly difficult to formulate thresholds for concentrations of constituents in either raw or treated produced water that might be expected to generate adverse environmental effects. One approach to solving this problem is the implementation of an adverse outcome pathway. The adverse outcome pathway concept uses existing toxicity testing methodologies at all levels of biological organization relevant to human and ecological risk assessment.[182] Adverse outcome pathways identify molecular and biochemical changes following exposure to a given chemical or effluent, and use of these as sublethal, early warning signs of eventual toxicity at the individual and/or population level provide a useful approach to follow in evaluating a treatment process that meets that water's specific end use (Figure 7).

Adverse outcome pathways could be integrated into an assessment of biological toxicity as follows. Preliminary "first tier" testing should initially establish links between mortality and exposure utilizing, for example, traditional toxicity tests such as LD/LC50 assays with early life stages, usually the most sensitive developmental stage. Ideally, such assessments would be performed chronically (the most realistic exposure scenario) and should involve a wide variety of different produced waters extracted from different geological formations that have gone

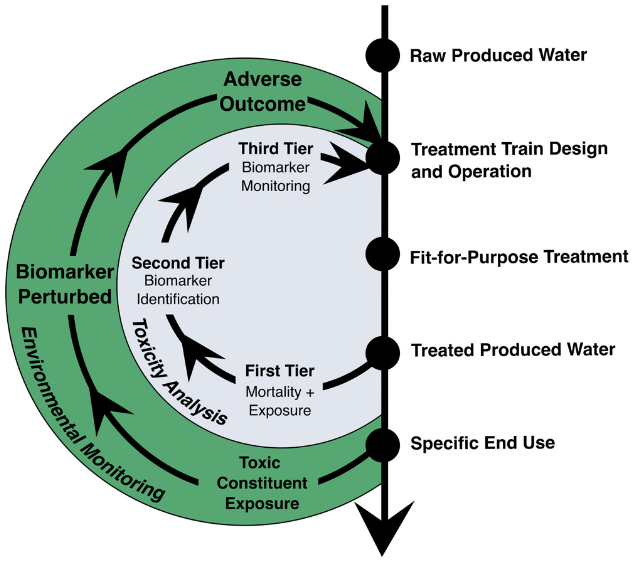

Figure 7. Proposed method for incorporating adverse outcome pathways and toxicity analysis into the design and operation of fit-for-purpose treatment trains for specific end uses of produced water.

through the same range of treatment steps. Chronic assessments would also allow samples to be taken over time for determination of sublethal changes, allowing linkage of these more subtle effects to mortality. The specific sublethal end points of interest should focus on pathways known to be impacted by salts (i.e., whole body ion homeostasis and associated enzymes, such as sodium/potassium ATPase), organic compounds (i.e., induction of biotransformation pathways, such as cytochrome P450 enzymes), and metals (e.g., metal handling pathways, such as metallothionein).

This "first tier" approach would be conducted in combination with "second tier" methods, based on the end use of the water and its potential to impact broader receptors in the environment. These may include omics techniques (e.g., transcriptomics, proteomics, epigenomics, and metabolomics), which are wide-scale screening techniques that can be used to identify other end points that are consistently changed upon produced water exposure.[183] First and second tier approaches would be of greatest efficacy if performed in standard model organisms across multiple phyla. Standard model organisms offer a greater availability of genetic information along with the ease of culturing such animals in the laboratory, and the past use of standard organisms will facilitate knowledge of exposure history. These studies will be critical to establish robust adverse outcome pathways and to establish the strongest linkages between sublethal change and eventual mortality. Once established, biomarkers that have been identified as early warning signals of future adverse effects will have to be verified in region-specific monitoring for the "third tier". Ideally, field-collected biota (of greatest relevance to the specific region and end user) can be sampled and assessed for the sublethal changes. This may ultimately require lab-based verification that the relationship between exposure and effect holds for the species of interest. Following this multilevel adverse outcome pathways approach, treatment technologies could be evaluated for their ability to not just reduce toxicity but ideally eliminate it from these complex industrial waters.

Overall, the adverse outcome pathway method can be used to continuously monitor toxicity regardless of the effluent's end

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use. Adverse outcome pathway analysis would be able to verify the effectiveness of any proposed treatment for produced water. If toxicity is appearing in vitro, then the specific biomarkers affected can help identify which class of chemicals is causing toxicity, and the treatment system can be modified accordingly. Once a certain treatment system has been established, the adverse outcome pathway method allows for a framework to establish credible regulations/policy because the targeted biomarkers are reliable indicators of toxicity that can be used to monitor the effectiveness of the treatment and beneficial reuse systems *in situ*. Therefore, if O&G producers want to beneficially reuse produced water at a specific location for a specific outcome, they could concentrate on the biomarkers that would be the focus of any regulatory requirements, then independently develop a treatment system and analyze the potential toxicity of the treated produced water in key model organisms (i.e., organisms defined by the end use and environmental restrictions). Lastly, long-term monitoring of the biological systems exposed to this treated produced water would be essential and should be conducted by testing different phyla of animals in the field for these established bioindicators; this monitoring would act as an alarm system, indicating adverse outcomes that might be manifesting in the environment.

5. CONCLUSIONS AND RESEARCH NEEDS

This perspective presents a current baseline of treatment methods of produced water in the O&G industry. We first provided a baseline of current water treatment practices for different end uses of produced water and then provided context and analysis for six case studies. These six case studies were chosen to represent different water treatment and disposal methods, different geographic regions, and different end uses of treated produced water. Where data were available, we modeled these case studies in the WaterTAP3 model to assess the LCOW and energy use for different treatment trains and source waters. On the basis of these modeled case studies, we also examined sensitivities and scenarios to determine the implications of utilizing treatment in other regions and water qualities. Even though the LCOW of current treatment practices may not be competitive with current water and treatment costs, technology innovations, changing water availability, mutable regulations, and rising water costs over time are likely to cause these treatment trains to become competitive with other water sources, especially when examining opportunities in water scarce regions. In cases such as CBM for municipal reuse, the economics may appear to be competitive with similar nontraditional water sources such as brackish water, but water treatment economics may not be the sole driver for future increased beneficial reuse of produced water. Regulatory frameworks are needed to ensure that environmental and health risks of beneficial use are communicated and managed. Frameworks should address monitoring, process control, treatment effectiveness, and potential environmental and health risks and can build off past developed frameworks (GWPC).

This perspective attempts to provide a framework that (a) links such tools to treatment train selection and optimization to reduce the potential toxicity associated with the beneficial reuse of produced water and (b) can be applied to case studies of treated produced waters of varying composition and end use targets, with the goal of identifying appropriate chemical indicators, biomarkers, and bioanalytical assays that correctly assess the risk associated with each particular end use. Thus, research is needed that employs a comprehensive adverse outcome pathway methodology for a range of waters and uses. The resulting data can be used to develop monitoring tools or optimize treatment trains that are relevant to produced water beneficial reuse for a specific end point. This analysis should begin with a baseline case in which produced water beneficial reuse has been employed for a long period of time, such as Kern County agricultural beneficial reuse. Comprehensive adverse outcome pathway testing on treated water, plant species, and soil samples would provide a baseline for the study of more complex waters in Colorado, Wyoming, Texas, and New Mexico. While this approach is necessary for reducing the risks associated with the beneficial reuse of produced water, it can also be applied to other industrial reclaimed water reuse scenarios, as well as to other unconventional water sources.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestengg.1c00248.

Process flow diagrams of the simulated WaterTAP3 baselines and what if scenarios, tabulated techno-economic outputs of the WaterTAP3 baselines and what if scenarios, and plots of adjusted LCOW and energy intensity per unit for baselines and what if scenarios (PDF)

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Notes

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Review

Produced water characteristics, treatment and reuse: A review

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ABSTRACT

In oil and gas industry, produced water is considered as the largest waste stream, which contains relatively higher concentration of hydrocarbons, heavy metals and other pollutants. Due to the increase in industrial activities, the generation of produced water has increased all over the world and its treatment for reuse is now important from environmental perspective. Treatment of produced water can be done through various methods including physical (membrane filtration, adsorption etc.), chemical (precipitation, oxidation), and biological (activated sludge, biological aerated filters and others) methods. This paper aims to highlight characteristics of produced water in detail and physical, chemical, and biological techniques used for its treatment. In addition, reuse of produced water for different purposes has been discussed. At the end, few case studies from different countries, related to the treatment and reuse of their produced waters have been included.

1. Introduction

In oil and gas industry, the produced water is a term used for the water associated with oil during the extraction process. It is one of the largest streams of wastewater generated in these industries, estimated to be > 70 billion barrels per annum in the world in 2009, out of which, 21 billion barrels is produced by US alone [1]. Produced water results from two processes in the oil and gas industry. First, during extraction, this gives a mixture of water and oil; the source of which is usually seawater surrounding the oil well. Second, the water injected into the oilfield to bring the deep oil to the surface also ultimately becomes part of produced water or wastewater. Based on the origin, produced water can be classified as produced water from natural gas, oilfield or coal bed methane [2].

Naturally, occurring rocks in subsurface formations are usually permeated with water, oil, gas or a combination of these fluids. It is believed that, prior to the petroleum invasion and trapping, rocks were completely saturated with water in most oil-bearing formations. This water could be either flowing from above, below or within the hydrocarbon zone or could flow from the injected fluids and additives coming from the production activities. Before the production process of the reservoir starts and the fluids brought to the surface, the produced water is known as formation water or connate water. Thus, any water present in the hydrocarbon reservoir and produced with crude oil or natural gas and brought to the surface is known as produced water. Furthermore, produced water removed through the production of coal bed methane (CBM) have almost the same properties as the produced water from crude oil or conventional gas production with some differences in its composition [3]. Moreover, extraction of oil and gas from offshore and onshore wells result in produced water generation, regardless if the fuel is extracted from conventional or unconventional sources including CBM, tight sands and gas shale [4].

Most of the volume of waste stream in oil and gas production operations on offshore platforms is produced water, and it represents 80% of the residuals and wastes produced through the production of natural gas. Furthermore, as the age of the well increases and the decline of oil and gas production results, the amount of produced water generation increases [5]. Volume of produced water could reach 98% in nearly depleted fields with only 2% of fossil fuel production [6]. During extraction of oil, the water to oil ratio is around 3:1 [7]. Even though the ratio has increased, the production of produced water at global level is still as much as 39.5 $Mm^3\,day^{-1}$. Due to the ageing of wells, it is also expected that the water to oil ratio will be averaging 12 (v/v) for crude oil resources by 2025 [1,6,7]. Thus, the market growth for the management and reuse of produced water is expected to grow further.

In this review paper, the produced water volumes across different countries, its characteristics in general, and physical, chemical, and biological techniques used for its treatment are discussed in detail. In addition, reuse of produced water after treatment for different purposes is highlighted. At the end, different case studies related to produced water treatment and reuse has been included. The aim behind this review paper is to stress and promote the treatment and reuse of produced water in order to reduce the reliance on limited freshwater resources.

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Table 1
Volumes of produced water associated with gas production in different countries.

Country	Produced water (bbl/year)	References
USA	21,000,000,000	[8,12,13]
Australia	207,570,000	[14–16]
China	45,917,000	[17]
Colorado	92,274,300	[18,19]
Iraq	105,853,190	[20,21]
Oman	1.84×10^9	[22–24]
Qatar	50,508,816.54	[25]

2. Produced water volumes

Produced water from oil or natural gas production does not have specific and constant volume (Table 1) as it depends on location and the technology used for extraction [8]. In 2003, it was reported that about 667 million metric tons of produced water were discharged to the offshore in the World. Out of which, 21.1 million tons were discharged to US Gulf of Mexico offshore water and between 358 and 419 million tons were discharged to the North Sea of the Europe offshore waters [9,10]. Additionally, in 2007, 256,000 m^3/day of produced water were produced during the production process of oil and natural gas in US Federal offshore waters in which 234,000 m^3/day was discharged to the ocean after treatment and the rest was reinjected [8]. Moreover, according to the International Association of Oil and gas Producers (IOGP), in 2014, 0.6 tons of produced water was discharged, and 1.2 tons of hydrocarbon produced were reinjected and 92% of the produced water was generated from offshores operations. The volume of produced water from a specific reservoir does not remain constant. Initially, the water production is very less but it increases with the age of the reservoir [11]. Since, the volume of produced water being produced is very high in most of the countries (Table 1), therefore, the management of this kind of wastewater is now a need.

2.1. Characteristics of produced water

Produced water is not a single product, it has a simple to complex composition that is variable, and it is considered as a mixture of dissolved and particulate organic and inorganic chemicals. Chemical and physical properties of produced water vary considerably which depends on several factors including, geographic location of the field, age and depth of the geological formation, hydrocarbon-bearing formation geochemistry, extraction method, type of the produced hydrocarbon, as well as its chemical composition in the reservoir. The toxicity of produced water discharged from gas platforms is 10 times higher than the toxicity of the oil wells discharge. However, the volumes from oil production are much higher than gas production [26]. Specific studies for each region should be done as its characteristics varies from region to region and such studies will also help in investigating the environmental risks of its discharge.

The main components found in produced water are categorized and summarized in Table 2 along with their concentrations from the literature. Generally, the major constituents that are present in produced water include: salt content (measured as salinity), total dissolved solids (TDS) or electrical conductivity; oil and grease (O&G); polyaromatic hydrocarbons (PAHs), benzene, toluene, ethylbenzene, and xylenes (BTEX), phenols, organic acids, natural organic and inorganic compounds that cause hardness and scaling (e.g., calcium, magnesium, sulfates, and barium); and chemical additives such as biocides and corrosion inhibitors that are used during drilling, fracturing and operating process of the well [27].

2.1.1. Conductivity, salinity and total dissolved solids (TDS)

The conductivity of produced water can vary widely as it was found that the conductivity of produced water from natural gas ranged from 4200 to 180,000 μS/cm [47]. In another research, the conductivity was found to be in a range of 136,000–586,000 μS/cm [55]. The salinity of produced water ranges from few parts per thousand (‰) to ∼300‰ (saturated brine) which is much higher than the salt concentration of seawater which is in the range of 32–36‰ and that is why produced water is generally denser than seawater. Higher salinity results due to the presence of dissolved chloride and sodium mainly as the concentrations of calcium, magnesium and potassium are usually lower [29]. According to a study done by Guerra et al. [34], TDS was in the range of 370–1940 mg/l due to the increased concentrations of both sodium and bicarbonate.

Recently, TDS concentration over time for produced water was investigated [56]. Results showed that the quality of produced water changes over time, which affects the management and reuse of produced water. Variations in the concentration of TDS occurs due to several reasons including the location of the well in the well field, geological variations between basins and the resource of the produced water. Furthermore, the concentration of TDS varies between the conventional and unconventional wells since it was found that the concentration of TDS was < 50,000 mg/l in CBM wells while it was as high as 400,000 mg/l in the conventional wells [34] (Table 2).

2.1.2. Inorganic Ions

Chloride and sodium are considered as the most abundant salt ions found in produced water, while phosphate has the lowest concentration. In produced water from both conventional and unconventional wells; sodium is considered as the dominant cation with 81% in conventional wells and more that 90% in unconventional wells [34]. However, the anions makeup in conventional and unconventional wells is not the same. The conventional wells are almost completely chloride anions representing 97% of the total anions, while 66% and 32% of the unconventional wells contain bicarbonate and chloride anions respectively [34].

Furthermore, sodium, chloride, magnesium, sulfate, bromide, potassium, iodide and bicarbonate are found abundantly in produced water with high salinity (Table 2). The presence of sulfate and sulfide ions in produced water can leads to insoluble sulfate and sulfide at high concentrations in produced water. Moreover, the presence of bacteria in the anoxic produced water, cause the reduction of sulfate and in turn leads to the presence of sulfides (polysulfide and hydrogen sulfide) in the produced water [32]. However, the concentration of these anions and cations varies from location and their ranges are presented in Table 2.

2.1.3. Metals

Produced water may contain certain metals like Fe, Cr, Ba, Ni, Zn and others. However, differences in the type, concentration, and chemical content of the metals are influenced by the geological age and features, injected water volume and chemical composition [57]. Commonly, mercury, zinc, barium, manganese, and iron are found in produced water at higher concentration than the seawater concentration [58]. For instance, Hibernia produced water have high concentrations of barium, iron, and manganese as compared to seawater. In addition, it was also reported that the barium, sodium, iron, magnesium, potassium and strontium in produced water from natural gas production field are present at higher concentrations [49].

2.1.4. Total suspended solids (TSS), total organic carbon (TOC) and total nitrogen (TN)

Total suspended solids (TSS) in produced water may include the floating or drifting materials found in the water such as silt, sediment, sand, algae and plankton. It has been noted that TSS concentration in produced water is in the range of 14–800 mg/l [47] and 8–5484 mg/l [54]. Moreover, another study conducted by Tibbettes [28], for oilfield produced water found that the TSS concentration was in the range of

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Table 2
Main components and their concentration found in produced water.

Parameter	Concentration (mg/l)	References	Parameter	Concentration (mg/l)	References
Major parameters			Metals		
COD	1220–2600	[11,28–30]	Na	0–150,000	[28,33,40–46]
TSS	1.2–1000	[2,11,28,29,31]	Sr	0–6250	[28,40–47]
TOC	0–1500	[2,29,32]	Zn	0.01–35	[28,41–46]
TDS	100–400,000	[18,29,31,33–35]	Li	0.038–64	[28,40–46]
Total organic acids	0.001–10000	[2,32,36]	Al	0.4–410	[28,40–46]
Production treatment chemicals			As	0.002–11	[29,30,41–43]
Glycol	7.7–2000	[2,37]	Ba	0–850	[28,40–47]
Corrosion inhibitor	0.3–10	[2,37]	Cr	0.002–1.1	[28,41–43]
Scale inhibitor	0.2–30	[2,37]	Fe	0.1–1100	[28,41–46]
BTEX			Mn	0.004–175	[28,41–46]
Benzene	0.032–778.51	[9,32,39,40]	K	24–4300	[28,41–46]
Ethylbenzene	0.026–399.84	[9,32,39,40]	Pd	0.008–0.88	[28–30,48]
Toluene	0.058–5.86	[9,32,39,40]	Ti	0.01–0.7	[28,41–43]
Xylene	0.01–1.29	[9,32,39,40]	Other ions		
Total BTEX	0.73–24.1	[9,28,31,38]	B	5–95	[28,40–47]
Other pollutants			Ca^{2+}	0–74,000	[28,33,41–46]
Saturated hydrocarbons	17–30	[32,36]	SO_4^{2-}	0–15,000	[28,33,41–47,49–53]
Total oil and grease	2–560	[11,29,31,41–43]	Mg^{2+}	0.9–6000	[28,41–46]
Phenol	0.001–10,000	[2,11,28,31]	HCO_3^-	0.15,000	[11,28,33,41–43]
			Cl^-	0–270,000	[28,33,41–47,54]

1.2–1000 mg/l (Table 2).

Furthermore, Rosenblum et al. [56], investigated the time variation of levels of TSS in produced water and found that there was almost 59% reduction in the concentration of TSS within first 4 days followed by no more significant variation noted for next few days. However, further 40% decrease was observed in the concentration of TSS in the period of 55th day of monitoring till 80th day.

According to a study conducted by OGP [59], the range of TOC found in produced water is from 0 to 1500 (mg/l) (Table 2). Various naturally occurring water have TOC concentration between less than 0.1 mg/l and to greater than 11,000 mg/l [32]. According to Ayers and Parker [51], the mean concentration value of TOC found in produced water from Hibernia platforms is 300 mg/l, while it was in the range of 67 to 620 mg/l in produced water from Louisiana rigs [60]. Similarly, TOC in the range of 0–1500 mg/l has been reported for produced water samples collected from various sources [29].

Kim et al. [33], conducted a study on produced water samples which were collected over a 200-day time period from two wells and used TOC concentration as a macro-indicator for the quality of the produced water and found that the concentration of TOC from both wells before 30 days was fluctuating significantly, but after this period of time it was stabilized at 2000 mg/l.

Total nitrogen is the cumulative sum of all the nitrogen compounds in the water, including ammonia-nitrogen (NH_3-N), nitrate-nitrogen (NO_3-N), nitrite-nitrogen (NO_2-N) and organically bonded nitrogen [34]. According to UNITAR [34], total nitrogen by Total Kjeldahl Nitrogen (TKN) is the total organic nitrogen compounds and ammonia with excluding nitrate-nitrogen and nitrite-nitrogen. According to Veil et al. [60], the presence of nitrogen and other nutrients lead to the formation of hypoxic zones.

According to Metcalf et al. [35], it is more difficult to remove the non-biodegradable part of the organic nitrogen than the biodegradable part which is easier to treat and less harmful for the environment. Furthermore, separation of particulate is easier than the soluble part. Therefore, the removal of TKN before the injection of the water back to the environment is crucial. According to Veil et al. [60] and Bierman et al. [61], who investigated the presence of NO_3^-, NO_2^-, NH_3, NH_4 in produced water from fifty platforms of either gas, oil or mixed production wells, found that the highest mean concentration of NO_3^- (2.71 mg/l) was found in produced water from mostly gas wells, while the highest concentration of NH_3 and NH_4 was found in produced water from mostly oil wells (92 mg/l). On the other hand, same concentration of 0.05 mg/l was measured for NO_2^- in produced water from all tested wells

2.1.5. Chemical oxygen demand (COD) and biochemical oxygen demand (BOD5)

According to studies done by Fillo et al. [47], and Johnson et al. [49], the estimated COD concentration in produced water was between 2600 mg/l and 120,000 mg/l. A research conducted in East China, where onshore produced water samples were collected from treatment plant at various sampling points, showed that the concentration of COD (mg/l) at each sampling point was: 285.5 ± 76.1 for the influent, 108.9 ± 29.2 for effluent, 195.2 ± 32.9 for EOS (effluent of oil separation tank), 109.5 ± 58.4 for EBO (effluent of bio-contact oxidation tank), and 190.7 ± 53.8 for EF (effluent of flotation tank). Therefore, it was found that these concentrations are higher than the acceptable limit set by the Environmental protection agency of China, i.e. < 150 mg/l [62]. Moreover, according to a research conducted by Zhao et al. [63], the concentration of COD was 280 mg/l in produced water from oil fields in Canada. Similarly, Shakrollahzadeh et al. [64], found that COD level of produced water extracted from gas stream in an Iranian gas refinery was 270 mg/l. On the contrary, Gomes et al. [65], obtained high range of 27,000 to 35,000 mg/l for produced water samples collected from oil fields in USA. Another research in which the physico-chemical characteristics of produced water collected from two oil facilities in Nigeria were tested, noted that the COD does not vary much among the two locations as it was found to be 3.91 ± 1.32 mg/l for both locations [66], which was less than the permissible limit of 125.0 mg/l [33].

The BOD of 75–2870 mg/l has been reported for produced water from natural gas field [47]. Reduced inorganic elements such as Fe and Mn, used fluids for well drilling, and additive chemicals can results in higher BOD concentrations in produced water obtained directly from the well. According to Adewumi et al. [67], high volumes of organic materials in drilling fluids can lead to the high BOD values in produced water. Furthermore, dissolved oxygen can severely deplete in water bodies receiving produced water with high BOD content, thus, substantial oxidation of this water should be ensured to prevent the discharge of waste water with high BOD into natural waters.

2.1.6. Oil and grease (O&G)

According to a study conducted by Fillo et al. [47], the concentration of O&G in natural gas, produced water was in the range of

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6–60 mg/l. Similar range has also been reported by Johnson et al. [49] and USEPA [54]. They found the concentration of 2.3–60 and 2.3–38.8 mg/l of O&G in produced water. Moreover, another study was conducted on western United States' produced water and the concentration of O&G was found to be 40 mg/l to as high as 2000 mg/l [68].

2.1.7. Organic acids

Main organic acids that are found in produced water are monocarboxylic acids and dicarboxylic acids (COOH) of both aliphatic and aromatic hydrocarbons having low molecular weight, such as formic acid, hexanoic acid, butanoic acid, acetic acid, propanoic acid, and pentanoic acid [69–71]. However, the most abundant organic acids in produced water are formatic acid and acetic acid [72]. Previously, it has been reported that the concentration of formic acid was from not detectable levels to 68 mg/l, acetic acid from 8 to as high as 5735 mg/l and propionic acid up to 4400 mg/l in produced water samples collected from Mexico gulf off the Texas and Louisiana coast and in the Santa Maria Basin off the California coast [72].

2.1.8. Benzene, toluene, ethyl benzene, and xylene (BTEX)

BTEX are volatile aromatic compounds that are naturally present in oil and gas products including natural gas, gasoline, and diesel fuel, thus they easily escape to the atmosphere during the water treatment process [48]. Benzene is abundantly found in produced water (Table 2), however, increasing the alkylation lead to the decrease in the concentration of benzene [39]. Furthermore, according to a study carried out by Dorea et al. [39], the concentration of BTEX present in produced water collected from oil field in Gulf of Mexico, the concentration of benzene was found to be highest i.e. 0.44–2.80 mg/l, followed by toluene, xylene, and ethyl benzene. These results are in consistent with the results of Neff [32], in which benzene was present at highest concentration i.e. 0.084–2.30 mg/l in produced water, followed by toluene, ethyl benzene and xylene. Similarly, Dorea et al. [39] investigated the characteristics of Permian basin produced water and they found that highest concentration was for benzene with 1.5–778.51 mg/l, followed by ethyl benzene, xylenes, and toluene.

2.1.9. Phenols

Phenols or phenolics are part of aromatic organic compounds that include one or more hydroxyl group attached to an aromatic hydrocarbon group. Various levels of phenols are present in produced water from oil and gas-operating wells, however, gas condensate production was found to have the highest concentration of phenols [73]. The comparison of the concentration of phenol in produced water from oil and gas field revealed that gas field-produced water has higher concentrations of phenol than oil field-produced water. Moreover, Neff [32] and Johnsen et al. [37] carried out a study to investigate the concentration of phenol in produced water collected from the Louisiana Gulf Coast and Norwegian Region of the North Sea found that the concentration range of phenols in produced water was in the range of 2.1–4.5 mg/l and 0.36–16.8 mg/l, respectively.

2.1.10. Production chemicals (treating chemicals)

Production chemicals are usually added to the oil or gas field for the management of the operational problems such as to facilitate oil, gas, and water separation process, prevention of pipeline corrosion and methane hydrate formation in the gas production system. The required chemicals for the production process are unique and vary along with the various production systems, and they can be categorized into 3 broad groups i.e. gas processing chemicals, simulation and work over chemicals and production treating chemicals such as scale, corrosion, hydration inhibitors, biocides, water treating chemicals like flocculants and anti-foams, emulsion breakers, reverse emulsion breakers, and coagulants, which are used in hydrocarbons' recovery and pumping. These production chemicals are soluble in oil, eliminating the need for the mechanism of disposal. The chemicals such as corrosion inhibitor and biocides are negatively affecting the environment, their use has been reduced as they were found at very low concentration in produced water [37,74].

3. Produced water treatment

Since, produced water contains several different contaminants with varying concentrations, therefore, numerous treatment technologies have been proposed for produced water treatment. The wide variety of produced water treatment methods have been reported previously [29,75–78]. The treatment system usually requires a series of individual unit processes for contaminants removal that might not be removed through a single process. Treatment of produced water can help in facilitating additional options for water management including its reuse for agricultural and industrial purposes. According to Arthur [27], the treatment of produced water should be able to remove dispersed and free oil and grease, soluble organics, suspended solids and dissolved gases, naturally occurring radioactive materials, salts and microorganisms. As a result, it is challenging to choose the type of treatment system suitable to remove most of the contaminants from produced water. Generally, the cheapest method is the most preferable one and the cost of the produced water treatment mainly depends on influent quality, electricity price, plant's capacity, as well as the intended quality of the effluent [79]. Furthermore, treatment of offshore produced water is more challenging due to the absence of adequate space or weight capacity for the equipment used for the treatment process as they should be designed for operation in remote and harsh environments.

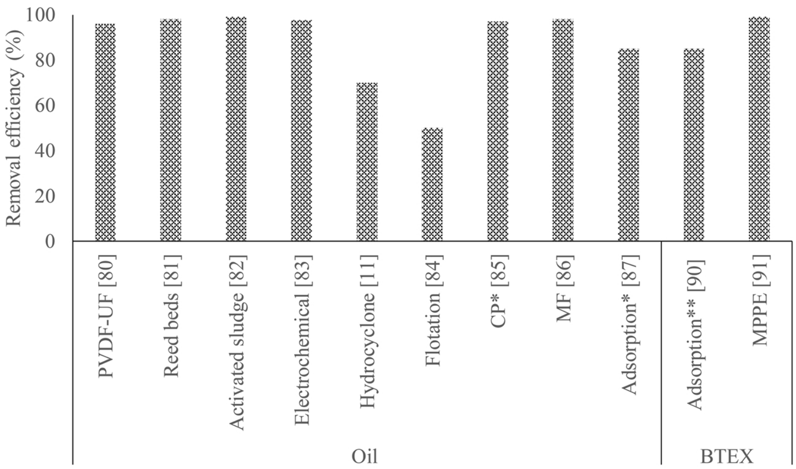
In general, produced water treatment process has three main stages i.e. pre-treatment, main treatment step, and final polishing treatment step. The pre-treatment step is done to remove large oil droplets, coarse particles and gas bubbles to reduce dispersed contaminants. The main treatment step involves primary treatment in which small oil droplets and particles removal will be achieved and will be done by using skim tanks, plate pack interceptors and API separator. The secondary treatment will involve removal of much smaller oil droplets and particles using gas flotation, hydro-cyclones and centrifuges. The polishing step is usually employed to remove ultra-small droplets and particles, dispersed hydrocarbons (< 10 mg/l) using techniques like dual media filters, cartridge filters and membranes. The optional step (tertiary treatment) is sometimes used to remove dissolved matter, gases and dispersed hydrocarbons (< 5 mg/l).

According to Fakhru'l-Razi et al. [29], combination of physical, chemical and biological treatment processes should be used for the achievement of the different treatment goals. Fig. 1 shows the removal efficiency of several treatment techniques for different pollutants. It is evident from Fig. 1 that most of the techniques such as adsorption, membrane filtration and chemical precipitation, widely studied in the literature, possesses higher removal efficiencies of above 90 for various produced water constituents and therefore, the cost of treatment and the intended purpose of treatment (reuse or discharge) with associated standards can affect the choice of suitable technique. In the following subsections, physical, chemical and biological techniques for produced water treatment are individually discussed in detail.

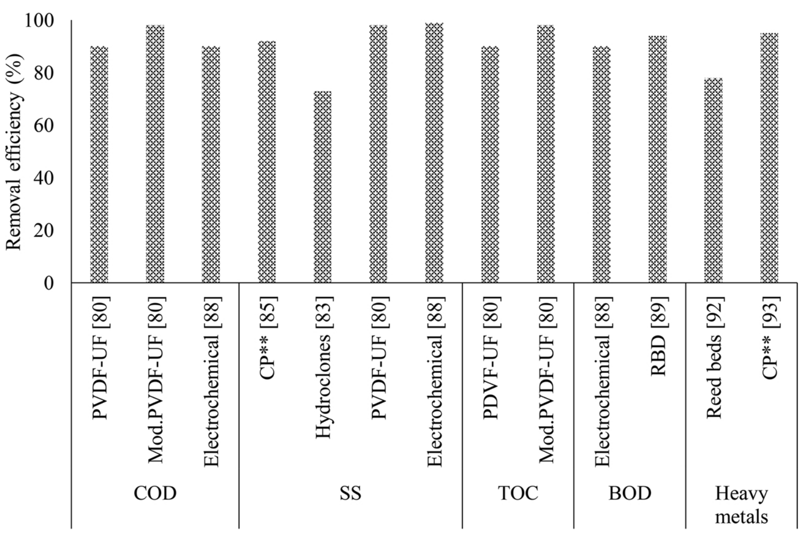
3.1. Physical treatment process

3.1.1. Filtration

Filtration is relatively simple technique used in water and wastewater treatment process, which is based on the use of porous filter media to allow only the water but not the impurities to pass through it. There are various porous materials that can be used as filter media, such as sand, crushed stone, and activated carbon. However, the widely used material is sand due to its availability, low cost and efficiency [94]. As proposed by Adewumi et al. [67], removing metals by sand filtration process should be done after the pretreatment stage which consists of



(A)



(B)

Fig. 1. Treatment technologies for removal of different contaminants (A) Oil and BTEX; (B) COD, Suspended solids (SS), TOC, BOD and Heavy metals from produced water. (CP*: Chemical precipitation by using FMA which is inorganic mixed metals (Fe, Mg, and Al) polynuclear polymer; CP**: chemical precipitation by lime; PVDF-UF: tubular polyvinylidene fluoride ultrafiltration 35 kDa; Mod. PDVF-UF: modified polyvinylidene fluoride ultrafiltration membrane with alumina nanoparticles; MF: tubular ceramic microfiltration (α-Al_2O_3) with pore size of 0.2 µm; Adsorption*: copolymers beads based on methyl methacrylate (MMA) and divinylbenzene (DVB) were prepared by suspension polymerization technique; Adsorption**: by zeolites; RBD: rotating biological disks; MPPE: macro-porous polymer extraction [80–93].

three steps: (1) adjustment of the pH for oxidation reduction initiation; (2) increasing the oxygen concentration for the reaction through the aeration unit and (3) adequate retention time for the settlement of precipitated solids in the solid separation unit. After these steps, the fine solids that were not removed during pre-treatment stages will be removed by sand filtration.

In the slow sand filtration treatment system, the pretreated water is passed in a downward direction through a filter which is made up of a layer of sand and specific features to control the water flow and the water filtration rate ranges between 0.1 and 0.4 $m^3/m^2/h$ [94]. Filter beds are mainly composed of fine grains with 0.15–0.35 mm diameter range and 1 m depth before starting the filtration process. As the filtration proceeds, the higher part of the treatment system will contain the colloidal and suspended particles coming from the untreated water, and as these particles build up they will clog the system and reduce its efficiency. Therefore, it is necessary to scrap off the top layer of the sand which is full of impurities to remove the clogging materials. Moreover, the filtration process is a combination of various processes including mechanical straining, chemical and biological activity, adsorption, and sedimentation for overall removal of impurities [94,95].

The removal of various ions from water through sand filtration was investigated by Wathugala et al. [96], and it was noted that sand filtration results in higher percentage removal of COD and nitrogen and the filtered water was free from ammonia and phosphorus. A new engineered sand filtration technique was developed by Cha et al. [97], in which it was combined with ozonation technique to treat the produced water. The technique helped to reduce the COD from 320 mg/l to 102 mg/l and oil content to 20 mg/l. Similarly, sand filtration was used to remove oil and grease from produced water and 95.8% removal was obtained [98].

Furthermore, membrane filtration technique can also be used for the treatment of produced water. Table 3 shows the comparison between various type of membranes used for the treatment of produced water [2].

Recently, it was demonstrated that the ceramic microfiltration membranes were able to sustain permeability of 1400 $l/m^2/h/bar$ when real produced water from Arabian oilfield was used as feed water, through the application of back-flushing, during cleaning in place (CIP) between cycles of filtration [99]. The research done by Chen et al. [100], on the treatment of produced water obtained from oil field using ceramic crossflow microfiltration membrane showed that the TSS level was < 1 mg/l and O&G value was < 5 mg/l. Another research was

Table 3
Comparison of the membrane technologies used for produced water treatment [2].

Technology	Feasibility	Used chemicals	Pre/post-treatment	Life cycle	Advantages	Disadvantages
Ceramic MF/UF membrane	Can be used for the treatment of all produced water types and especially oilfield produced water, but it is not very applicable for produced water with high concentrations of TDS and salts.	Coagulant agents for pre-coagulation: ferric/polyaluminum chloride and aluminum sulfate. Cleaning agents: surfactants, acids, and bases.	Pre-treatment: coagulation and cartridge filtration. Post-treatment: depends on the produced water, could require polishing.	> 10 years	- Totally free product water of SS. - Operates in both filtration modes: dead-end or cross flow - Has 90%-100% recovery of product water - Longer life cycle than other membranes	- Periodic cleaning of the membrane - Disposal, recycling, or further treatment of the generated waste during the cleaning and backwash process is required - Significant amount of iron can concentrate in the water feed when irreversible membrane fouling occurs.
Polymeric MF/UF membrane	Can be used for the treatment of produced water with high salinity and TDS concentration.	Coagulant agents for pre-coagulation: ferric/polyaluminum chloride and aluminum sulfate. Cleaning agents: surfactants, acids, and bases.	Pre-treatment: coagulation and cartridge filtration. Post-treatment: depends on the produced water, could require polishing.	≥7 years	- Totally free product water of SS. - Has 85–100% recovery of product water	- Periodic cleaning of the membrane. - Disposal, recycling, or further treatment of the generated waste during the cleaning and backwash process is required.
NF	It is inappropriate method to be used alone for produced water treatment and can be used for the treatment of produced water with 500–25,000 mg/l of TDS.	Fouling preventing: by scale and caustic inhibitors. Cleaning agents: NaOH, HCl, Na_2SO_4, H_2O_2, Na_4EDTA	Pre-treatment is extensively needed for fouling inhibiting.	3–7 years	- Tolerate high pH - Automatic operation system - Implementing subsystems of energy recovery can decrease the energy costs - Solid waste disposal is not required Has 75–90% recovery of water.	- Has high sensitivity toward organics and inorganics present in the feed water - Multiple cycles of backwashing are required. - 45 °C is the maximum temperature that the membrane can withstand.
RO	To be used in produced water treatment, it is important to extensively pre-treat the produced water feed in which many studies failed to use it due to the poor pre-treatment process.	Fouling preventing: by scale and caustic inhibitors. Cleaning agents: NaOH, HCl, Na_2SO_4, H_2O_2, Na_4EDTA, H_3PO_4.	Pre-treatment is extensively needed for fouling inhibiting.	3–7 years	- Tolerate high pH - Automatic operation system - Implementing subsystems of energy recovery can decrease the energy costs - Excellent performance for treatment of pre-treated produced water	- Has high sensitivity toward organics and inorganics present in the feed water - 45 °C is the maximum temperature that the membrane can withstand.

Table 4
% Removal efficiency of the treatment of produced water by nanofiltration (NF) and reverse osmosis (RO) [103,104].

Characteristics	% Removal efficiency	
	NF	RO
TOC	31–85	66.9–97
TSS	90–99.9	90–99.9
O&G	90–97.2	90–99.9
Chloride	41.2–99.2	97.9–98.6
Sulfate	99.9	99.9
Calcium	77.2–98	99.3–99.9
Magnesium	70.5–99.9	99.5–99.9

conducted by Mueller et al. [101], on the removal of oil from synthetic produced water by ceramic membranes and results showed that 99.3–99.9% removal of oil could be achieved using this technique. Moreover, according to a case study conducted by the New Logic Research in USA, the treatment of produced water using combination of NF and RO membrane resulted in removal of most of the pollutants from produced water as shown in Table 4. Direct contact Membrane distillation (DCMD) technique has also been used recently for the treatment of produced water. It was noted that nanoporous hydrophobic hollow fiber membranes fabricated during the research was able to remove greater than 99.5% of TDS from feed water even after continual operation for 100 hours. The treated water was found to be suitable for industrial reuse. The combination of microfiltration and membrane distillation has also been evaluated for treatment of produced water and minerals recovery and it was found that the combined technique can recover various salts such as sodium, calcium, magnesium and barium from produced water [102] Membrane filtration technique has also been used in conjunction with adsorption to treat produced water. The double staged treatment system comprising carbon-bentonite adsorbent and nano-hybrid membrane helped to reduce TDS (72%), turbidity (6%) and salinity (90%). The use of adsorbent at a pretreatment stage helped to stabilize membrane flux, reduce membrane fouling and prolong the membrane lifetime [5]. The selection of suitable pre-treatment technique can also help to improve the performance of NF and RO as previously demonstrated by Ozgun et al. [31]. It was noted that the application of membrane bioreactors (MBRs) as a pre-treatment helped to reduce 83% COD from produced water and therefore, it was optimum treatment technique in combination with RO in terms of COD and conductivity reduction. Whereas, microfiltration and ultrafiltration membranes were able to remove less than 25% of the COD and therefore, were not as effective as MBR in terms of pre-treatment techniques [31].

3.1.2. Electrodialysis

Electrodialysis (ED) and electrodialysis reversal (EDR) are both separation processes that are driven by electrochemical charge and are used for the treatment of brackish water, seawater desalination and wastewater reclamation, as well as being tested for the treatment of produced water at laboratory-scale [105]. Salts in produced water could be removed by electrodialysis in which it utilizes a stack of alternating anion and cation selective membranes that are separated by spacer sheets [106]. Electrical current is applied to the cell after passing the water through membranes stack, which lead to the migration of anions and cations into opposite directions. Alternating cells of diluted and concentrated solutions are produced between the membranes as the migrating ions intersects the selectively permeable membrane. This process requires less energy than reverse osmosis as it operates under lower pressure and it can reduce the concentration of salt to < 200 mg/l. This treatment method is usually used for produced water with low TDS concentrations [107]. On the other hand, the drawbacks of this technology include its limited ability to remove non-ionic constituents such as organic molecules, occurrence of membrane fouling, relatively high cost, periodic disposal of concentrate and highly skilled labor requirement [105]. Moreover, this process could remove salts from produced water but other constituents such as oil, heavy metals, and other pollutants present in produced water may not be removed.

The main advantages of this technology include: withstanding harsh conditions, has membrane lifetime that is estimated to be in the range of 4–5 years, no need for special infrastructure and considered as an excellent treatment process to be applied for produced water [2].

3.1.3. Flotation

In this method, fine gas bubbles are utilized for the separation of suspended particles that cannot be removed by sedimentation. This process is done by injecting gas into the water to be treated, then both suspended particulates and oil droplets will attach to the air bubbles, and then they will rise to the surface leading to the formation of foam that is removed by skimming. This process can be done using air, nitrogen or other types of inert gases. The technique can be used for the removal of volatile organics, oil and grease from produced water [108]. Dissolved gas flotation (DGF) and induced gas flotation (IGF) are two subdivisions of the gas flotation technology and the difference between them is in the method used for the generation of the gas bubbles and the resultant bubble size. The efficiency of the process mainly depends on the contaminants to be removed, density differences of liquids, temperature, and the size of the oil droplet. Flotation process does not work ideally with high temperature feed streams, however, it works well under cold temperature and can be used for the treatment of produced water with high and low TOC concentrations, as well as, water with oil, grease and particulates with less than 7% solids [34]. Particles with 25 μm size can be removed by dissolved air flotation process, and when using coagulation as pretreatment step, contaminants with size of 3–5 μm can be removed as well. As reported by Fakhru'l-Razi et al. [29], there are multiple advantages and disadvantages of gas flotation process. The advantages includes: (i) coalescence increase the process efficiency; (ii) ease of operation; (iii) has no moving parts; (iv) it is durable and robust. While the disadvantages are: (i) large amount of airis generated; (ii) skim volume; (iii) 4–5 min retention time. The percentage oil removal of up to 93% has been obtained using flotation process [109]. Moreover, Beyer et al. [89], used induced-air flotation technique as a pre-treatment for oilfield produced water having TDS of 20,000 mg/l and it was found that the COD and TOC level decreased to 595 mg/l and 115 mg/l, respectively.

Thus, using these techniques, oil content, VOCs, suspended solids etc. can be removed from produced water without using any additional chemicals. In addition, coagulants can be used sometimes to increase the efficiency of treatment. Nevertheless, the main drawback remains the disposal of sludge at the end of treatment which increases the cost of treatment [110].

3.1.4. Adsorption

Adsorption is considered as one of the best treatment techniques for achieving better water quality as it can reduce the concentration of the contaminant to very low levels [111]. According to Spellman [112], nearly 100% produced water recovery and 85% removal of heavy metals can be accomplished through adsorption process. However, the main disadvantage of using the adsorption method is the cost of the installation and maintenance of the system, but this drawback can be solved by using more economical adsorption media such as activated carbon that can make the adsorption process more competitive. Another disadvantage of adsorption process is the requirement of waste disposal for the produced waste and spent media through the regeneration process of media. Furthermore, different organic and inorganic compounds can be retained by various adsorbent materials. Several adsorbents have been proven to remove manganese, iron, TOC, BTEX, oil and heavy metals (> 80%) from produced water.

Activated carbon have been used widely for a long time as an

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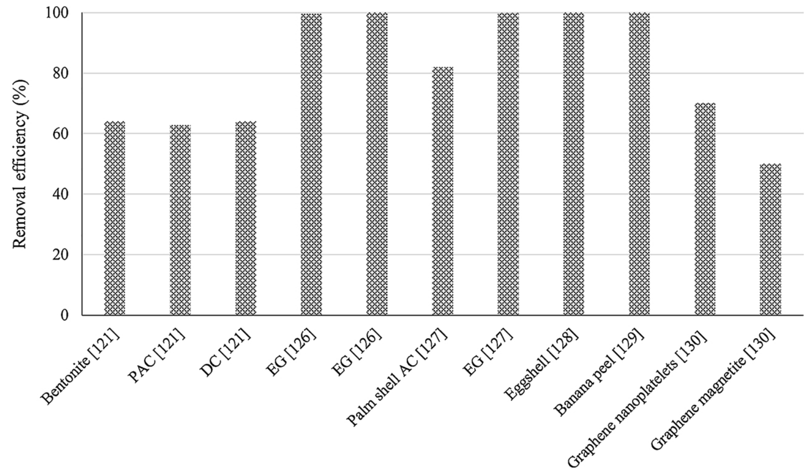

Fig. 2. Removal of oil and organic pollutants from produced water by various adsorbents. PAC: powdered activated carbon; DC: deposited carbon; EG: exfoliated graphite; AC: activated carbon [126–130].

effective adsorbent to remove various pollutants from contaminated water. Using AC as an adsorbent is preferable for water decontamination process rather than other techniques because it is less expensive, relatively simple, and efficient. Producing reasonably cheap and excellent adsorbent from AC is done through different treatment methods that helps to develop internal pore structure and large surface area [113]. Activated carbon has various unique characteristics including high grade of surface reactivity, high adsorption ability, extended surface area, and microporous structure [114]. In addition, activated carbon has several functional groups that are accountable for the diversity of physiochemical and catalytic characteristic [115]. Using AC, contaminants that can be removed include: cadmium, mercury, natural organic matter, BTEX compounds and synthetic organic chemicals such as benzo(a)pyrene, dioxin, radionuclides, di(2-ethylhexyl)phthalate, and hexachlorobenzene [34]. There are various materials that can be used for preparing AC, such as fossil fuel, wood, or some agricultural wastes using different preparation processes of physical or chemical nature [116]. Physical activation method is not widely used as it has been reported to give specific surface area less than 1500 m^2/g only [117]. On the other hand, in the chemical activation method, chemical activation agents such as KOH, NaOH and H_2SO_4 are used for the impregnation of the used precursors for AC preparation, followed by carbonization process. KOH is the most preferred chemical activation agent as it results in formation of K_2CO_3 after the interaction between char and KOH, leading to prevention of excessive burning of the sample giving higher yield and well developed internal porosity. Another advantage of using KOH is that it is considered as more environment friendly chemical than other activators [118]. Several different parameters influence the quality of the prepared AC including the impregnation ratio (IR), activation time and activation temperature [117]. AC has around 650 m^2/g to 1000 m^2/g adsorption surface area which is considered as an extremely large amount of adsorption surface area.

Activated carbon removal efficiency can range from 70 to 85%, but the presence of suspended particles within the produced water can decrease the removal efficiency [29]. Regeneration of activated carbon is necessary after few batches of treatment to regain the pollutant removal efficiency as it dramatically decreases with time [119]. Various chemicals such as acids, bases, redox agents, and organic solvents are used during the regeneration process of activated carbon, leading to the increase in cost of the treatment process [120]. Furthermore, regeneration can be done onsite in case the AC plant is large enough, however, it is usually done off site because onsite regeneration is only effective in the case of having 910 kg/day carbon exhaustion rate. Moreover, there are certain factors that determine the reactivation frequency including concentration, contaminant type, water usage rate and carbon type used [109].

Okiel et al. [121] investigated the efficiency of using bentonite, deposited carbon (DC), and powdered activated carbon (PAC) for oil removal from produced water, the results showed that increasing the weight and contact time of the adsorbent lead to an increase in the percentage removal of oil which highlighted the importance of contact time in adsorption technique.

According to Al-Ghouti et al. [122], who used activated carbon (AC) on reducing organosulfur compounds (ORS) from diesel-non-aqueous medium; found that there are excellent adsorption capabilities of granular bead form of AC (NORIT PK 1-3, Holland). The research illustrates that AC particle size affects the elimination efficiency of ORS in which the adsorption mainly occurs on the external surface area. Moreover, it was proved that AC has the ability to remove BTEX and free hydrocarbons participating in the total petroleum hydrocarbons (TPH). There are various parameters that influence the absorber effectiveness which are pH, temperature, salinity, and low concentration of heavy metals and dissolved organic chemicals. In addition, Doyle et al. [123], developed a new system composed of a modified polymer, bentonite or organoclay and bed column packed with AC to eliminate hydrocarbons resulting in the reduction of TPH and BTEX to non-detectable limits, but the need to regenerate the absorbent material repeatedly is the main disadvantage of this system.

Another research was carried out by Luukkonen et al. [124], in which 4 commercial ACs were utilized to investigate the TOC elimination from makeup water of power plant where continuous flow bench scale AC filters at steam boiler desalination plant were used to carry out the experiment. Results showed that all the tested ACs had similar removal efficiency of 42–45% of TOC and 58–68% of dissolved organic carbon after operation for 30 days. Removal efficiencies of TOC were steady throughout the experiment ranging between 41.6% and 44.8% for all the used ACs, and the residues of TOC after AC filtration were between 126 ppb and 260 ppb. Since deionized water nutrient content are too low for allowing any biological activity, it is assumed that the removal of TOC is primarily due to adsorption. Furthermore, Halim et al. [125], did a comparison study demonstrating the AC adsorption ability of COD and found that AC has higher removal capacity than other adsorbents such as composite and zeolites and the adsorption capacities of COD were 37.88 mg/g, 22.99 mg/g, and 2.35 mg/g, respectively. Fig. 2 summarizes the efficiency of various adsorbents for oil and organic pollutants removal from produced water. It is evident from the Fig. 2 that the adsorbents like exfoliated graphite and the ones obtained from organic source (eggshell and banana peel) achieved

100% removal of oil and organic pollutants.

Modification of the adsorbent's surface enhances their elimination efficiency and adsorption selectivity of specific toxic materials. Modification of activated carbon can be done with hydrophilic groups that include cations, anions, and zwitterions leading to structured molecular assemblies that depends on the group nature. The research group of Nadeem et al. [131], used sodium dodecyl sulfate (SDS) as a negative charged surfactant and cetyltrimethyl ammonium bromide (CTAB) as a positively charged surfactant for cadmium removal from aqueous solution. It was found that SDS-AC had the highest percentage removal of Cd (98%) among all other adsorbents, as well as having better surface area and greater porosity. Therefore, such modified adsorbents can be used for target removal of certain pollutants from produced water.

AC has also been used as a pre-treatment technique for produced water treatment. It was noted that the use of AC (PAC and granular activated carbon, GAC) with microfiltration as pre-treatment technique increased the % reduction in COD from 10 to 30 and 48%, respectively. Furthermore, it also helped to improve the performance of final treatment step with NF and RO techniques. In conclusion, the GAC with microfiltration followed by RO was suggested as a best technique for optimum removal of heavy metals and reduction in conductivity [132]. Nevertheless, the overall cost of the treatment technique including different pre-treatment methods needs to be taken into consideration to determine best technique for treatment of produced water.

3.2. Chemical treatment process

3.2.1. Precipitation

Precipitation is considered as one of the conventional chemical treatment processes of produced water [133]. Through this process, up to 97% removal of suspended and colloidal particles can be accomplished [134]. Flocculants and coagulants which are mainly comprised of inorganic metals such as iron, magnesium and aluminum polymers are usually used in the chemical treatment process and they were found to be effective in removing contaminants [85]. Other studies removed particulate metals, phosphorous and carbonaceous compounds by applying flocculants like ferric chloride ($FeCl_3$) and anionic polymer in ballasted flocculation unit, however, these flocculants were found to be less efficient for the removal of hydrophilic compounds and nitrogen [135]. Moreover, Zhou et al. [85], reported that the addition of coagulation chemicals can remove almost 97% of suspended solids and oil from produced water.

3.2.2. Chemical oxidation

This technology is usually used for the removal of COD, BOD, odor, color, organics, and some inorganics from produced water. According to Igunnu and Chen [2], it is not possible for free electrons to be present in solution, therefore, this treatment process depends on the reactions of oxidation and reduction as they occur together in the produced water. According to Huang [136], strong oxidants and catalysts can be used for decomposing the organic impurities present in the produced water. Generally, multiple pollutants can be broken down by using several oxidants like chlorine, ozone, peroxide and oxygen. Furthermore, there are various parameters that affect the oxidation rate of this technology including: dose of the chemical, used oxidant's type, quality of the raw water, and contact time between water and used oxidant [2]. The main advantages of this treatment process are; minimal requirement of equipment, does not generate any waste, does not need any pretreatment process and can achieve almost 100% water recovery rate. However, the main drawbacks are; it has high chemical cost, maintenance and calibration of the chemical pump is required regularly and production of byproducts through the process that are not easily removed [2]. Moreover, Igunnu and Chen [2], mentioned that a final treatment process is required for particulate matter removal after the oxidation process. Advanced oxidation processes (AOP) is considered as a recent development in water treatment field which is an effective solution for quickly oxidizing the organic pollutants through the addition of oxidants or mixture of oxidants [136]. This process utilizes ozone, iron, and hydrogen peroxide as chemical oxidizers. Furthermore, hydroxyl radicals like zinc oxide, titanium dioxide, and iron oxide are also introduced in this treatment process [137].

3.2.3. Electrochemical technologies

Although this process has been used widely in the treatment of various wastewater types, it is rarely used for the treatment of produced water. However, this technique is being suggested as the future produced water treatment technology. The advantages of these technologies over other treatment technologies is that it is low-cost green technology and it does not utilize any additional chemicals nor generate secondary waste. Furthermore, it can remove organic materials efficiently, can produce and save energy and help to recover valuable materials from produced water without negatively affecting the environment. This can be achieved through harmonizing various electrochemical techniques such as water electrolysis, fuel cell, electrodeposition, and photo-electrochemistry, that includes photo-electrolysis, photo-catalysis, and photo-electrocatalysis into one electrochemical process [2].

Photo-electrolysis is a chemical process that uses light for breaking down large molecules into smaller ones. As reported by Fujishima and Honda [138], removal of organics from produced water could be achieved through the use of TiO_2 electrodes for the photocatalytic decomposition of water and Adams et al. [139], reported that semiconductor photocatalysis can decrease the content of hydrocarbons present in produced water effectively in 10 min by 90%. Furthermore, Li et al. [140], conducted a study on synthetic produced water that mimics the original produced water with its constituents to investigate the removal efficiency of COD by photo-electrocatalysis and found that it has higher removal efficiencies than electrochemical oxidation and photocatalysis. Another study carried by Ma and Wang [88], on the removal of organics from produced water obtained from oilfield by setting up a catalytic electrochemical pilot-scale plant, in which they used iron as a cathode material, double anodes with graphite and active metal, in addition to noble metal having large surface as a catalyst. Results showed a reduction on the level of COD and BOD by more than 90% in 6 min. Furthermore, in 3 min, they observed that the suspended solids reduced by 99%, content of Ca^{2+} by 22%, 98% reduction in the rate of corrosion, and 99% decrease in the presence of sulfate reducing bacteria and iron bacteria.

Moreover, fuel cell is one of the most important electrochemical technologies that converts energy into electricity and produces heat and water as by-products. This technology plays crucial role in the future of the treating produced water. However, successful research on reducing the cost of this technology, as well as improving the efficiency and increasing its life span are important factors for the application of the fuel cell technology on the treatment of produced water in the future [141]. In addition, there are various established treatment technologies of produced water that can remove heavy metals, but they cannot recover the metals removed from produced water. However, electrodeposition is a technology that is usually used for the recovery of metals. It has been utilized for the recovery of Cu from synthetic produced water and results showed the excellent ability of electrodeposition to recover metals that would be lost during the removal process from produced water [142].

3.3. Biological treatment process

Biological treatment process is considered as one of the least expensive processes for removal of pollutants, in which either aerobic or anaerobic conditions are maintained [143]. Furthermore, algae, fungi and bacteria with 0.2–10 μm size are generally present in produced water, and they can be utilized for produced water treatment as these microorganisms will use the pollutants as nutrient source for growth purposes [144]. Several different processes and technologies like sequencing batch reactors and biological aerated filters can be used for biological treatment of produced water [29]. This process is mostly effective in feed water with COD < 400 mg/l, BOD < 50 mg/l, oil level < 60 mg/l [34], and concentration of chloride < 6600 mg/l [145]. According to research carried out by Li et al. [146], on the removal of COD with concentration of 2600 mg/l from produced water and removal efficiency of 90% can be achieved by utilizing the immobilized *Basillus* sp. under aerobic conditions. Furthermore, microbial community of *Methanosarcina*, *Rhodopseudomonas*, and *Clostridia* were used for removal of COD from produced water under anaerobic conditions and achieved removal efficiency of 65%. Kose et al. [147], tested the operational stability of membrane bioreactors (MBRs) for the treatment of real produced water. Despite of variation in the influent quality and solids retention times (SRT), it was found that MBR provides stable quality of treated water with 80–85% removal of COD and 99% removal of hydrocarbons. Moreover, spiral microbial electrochemical cell (SMXC) as fuel cell was developed to improve the treatment of produced water by Naraghi et al. [148], in which consortia of halophile and halotolerant anaerobic microbial community were used to remove organic compounds from feed water with > 200,000 mg/l salinity, and they achieved 90% removal of organics. In addition, Stoll et al. [149], biodegrade organics present in produced water by using microbial capacitive desalination cell (MCDC), and they successfully removed 6.4 mgTOC/h in biological reactor as well as biodegradation of 36 mgTDS/g carbon of electrode/h. However, one of the major disadvantages of biological treatment is the generation of huge quantity of biological sludge, which requires further treatment and relatively lower efficiency and more contact time. Another drawback is the stationary infrastructure of the common biological processes which needs long assembly and operation time [150].

3.3.1. Activated sludge

Activated sludge is one of the commonly used aerobic treatment process of wastewaters, in which it can adsorb and occlude soluble and insoluble materials [29]. Furthermore, it has been reported that using the solids retention time (SRT) of about 20 days; the removal of total petroleum hydrocarbons (TPH) of 98–99% can be achieved through activated sludge treatment [151]. Similarly, another research group used the mixture (45% and 35% (v/v)) of produced water and sewage and treated it using SBR (sequence batch reactor) and acclimated sewage sludge. The results showed that the %COD removal was in a range between 30 and 50% [152]. In addition, they also noted that the salinity does not seem to affect the treatment efficiency. However, due to the recalcitrant feature of produced water; the efficiency of biological treatment is generally low [152]. According to Fakhru'l-Razi et al. [29], this treatment process can remove trace and suspended solids, in addition to the removal of metals. Moreover, activated sludge is considered as cheap, clean, and simple treatment technology, but it requires oxygen, large filter dimensions, and it produces sludge as waste after the treatment process is over. It normally requires post-treatment for the separation of precipitated solids, biomass, and dissolved gases.

3.3.2. Biological aerated filters (BAF)

BAF system is consisting of media that is permeable and have diameter of 4 in. that prohibit pore spaces clog when sloughing happens. This class of biological technologies works under aerobic conditions to remove organics from polluted water and assist the biochemical oxidation process. Furthermore, for the complete usage of the filter bed, upstream and downstream sedimentation are required [2]. BAF treatment process was used for the removal of various pollutants from produced water and is considered most effective with produced water containing < 6600 mg/l chloride [153]. It was found that BAF can achieve removal efficiencies of 70%, 80%, 60%, 95%, and 85% for nitrogen, oil, COD, BOD, and SS, respectively [153]. Moreover, since the generated waste is removed in the form of solid, BAF has nearly 100% water recovery. Therefore, there is a requirement for solid disposal of the sludge accumulated in the sedimentation basins that accounts for almost 40% of the total cost of this treatment process, which is considered as the main drawback of this technology. On the other hand, there are various advantages such as it does not require post-treatment; can easily be adapted for wide range of water quantity and quality; requires little maintenance; does not require use of any chemicals; and it is expected to have long life cycle.

Furthermore, according to Mohan et al. [154], salinity and C/N ratio could hugely affect wastewater treatment process as denitrification and accumulation of nitrate will increase with the increase in the C/N ratio but decrease with increase in salinity. Thus, maintaining specific salinity levels is very important, which can be done by proper wastewater homogenization with the batch reactor at the inlet. Similarly, nitrogen removal from the municipal wastewater in the biological aerated filtration (BAF) system is highly affected by the C/N ratio [155,156]. Nitrogen removal efficiency could be affected by the hydraulic retention time, in which the removal efficiency increased to 95–96% when different hydraulic retention time was used [156]. The BAF system was proven the best technique for nitrogen removal from wastewater in which the denitrification performance under total COD and TKN ratio of 3:6 gave the best results [155,156]. In addition, removal efficiency of COD, ammonium, and TN reached 83.7%, 93.1%, and 84.6%, respectively, when the COD/N ratio was 5 and a dramatic reduction in the system's performance occurred when the COD/N ratio decreased [155]. On the other hand, raceway pond and photobioreactors are usually used in processes that are based on the use of microalgae [157]. Operational systems and microorganisms' inoculation affect the removal efficiency of pollutants from produced water [158].

3.3.3. Innovative microbial capacitive desalination cell (MCDC) treatment process

Microbial desalination cell (MDC) is a recent and new technology that was developed from the traditional technology of microbial fuel cell (MFC) process that generates an electrical current; ensure desalination of water and treats wastewater. The development method of MDC from MFC was achieved through the construction of a reactor by installing a desalination chamber in-between the cathode and the anode chambers [159]. Furthermore, MCDC is derived from the MDC design with the integration of capacitive deionization (CDI) into the design of the MDC in which it prevents the transport of salts into the anode and the cathode because it uses porous electrodes that sorb ions electrically [160]. The advantage of MCDC over the traditional MDC is that it can overcome the pH changes and imbalance by utilizing two cation exchange membranes (CEM) instead of one as used in MDC, allowing the protons to transfer freely across the system which in turn prevents the significant changes in pH. Stoll et al. [149], conducted a study on the use of MCDC for the removal of salts, organics from shale gas produced water, and the efficiency of the desalination of MCDC was estimated based on the removal of ions in the cathode, anode, and desalination chambers. This study was conducted to proof the demonstrated concept that the biodegradable organic matter constituents of the shale gas produced water are sufficient for running the MCDC, generating an electrical potential for desalination of 0.25–0.28 V. The dissolved organic carbon (DOC) and aromatic compounds were removed from the anode chamber through biodegradation and sorption, in addition, a consistent potential was successfully generated by the microbial community throughout the experiment. Moreover, salt was removed effectively from the desalination chamber through electro-sorption with removal percentage ranging between 64% and 70%, with sorption capacity 5–18 times greater than the conventional CDI, which utilize activated carbon as electrodes [161]. Furthermore, MCDC have salt removal rate that is 1.1–12 times higher than the traditional MDC system due to the additional sorption capacities with the advantage of

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keeping salts out of the cathode and anode chambers by preventing its transportation and accumulation [162]. The sequential operation of MCDC is required for the removal of salts and organics, so it can meet the quality standards for the reuse of produced water [149]. In addition, MCDC can also be used as pretreatment for the partial degradation of organics and to desalinate water in order to reduce membrane fouling and scaling. However, this treatment technology requires more research for better understanding of the long-term negative effects of highly saline produced water and its contaminants on membranes, assemblies of the electrodes, microbial communities, and the overall performance, which in turn will assist in optimizing the reactor for the desalination and treatment of produced water, as well as in power generation. It is also crucial to determine the factors affecting the regeneration process of MCDC, in which optimizing the regeneration before implementing MCDC on a large scale is a must [149]. Shrestha et al. [163], compared the MCDC performance with microbial fuel cell (MFC) and found that MFC achieved higher % reduction of COD (88%) than MCDC (76%). However, MCDC was able to remove two times more dissolved solids than MFCs. It was also noted that both techniques suffered from impedance due to fouling during the later operational stages.

3.3.4. Microalgae based treatment process

Currently, Eco-technology is a new approach that has been introduced for the treatment processes of produced water in which higher removal rate of pollutants from produced water can be reached [164]. Therefore, the use of microalgae-based treatment as a sustainable solution for the treatment process is defined by these Eco-technology approaches. Generally, bio-remediation of produced water effluents can be done through using the microalgae due to their ability in utilizing certain pollutants as nutrient source [165]. According to a study done by Takacova et al. [166], that BTEX can be utilized as a sole carbon source by specific microalgal species such as *Parachlorella kessler*. In another study, water soluble fraction (WSF) gasoline was used to investigate its toxicity and it gives an important foundation for the effect of BTEX on the growth of microalgae [167,168]. However, 50% growth inhibition on cultures of microalgae is caused by increasing the concentration of BTEX with more contact time [169]. Heavier hydrocarbons have higher toxicity on microalgae growth [170]. Furthermore, produced water generally contains sufficient concentration of nitrogen and phosphorus which sometimes act as growth limiting factors for microalgae [29]. Furthermore, there are various trace elements other than nitrogen and phosphorus that are important for microalgal growth and they are also present in produced water. Thus, growing microalgae in produced water has the potential to be used as efficient treatment process in which the microalgae biomass production is increased during treatment process. Furthermore, cultivated microalgae biomass can be used as alternative feedstock for generation of energy [171]. The microalgae strains used in different water treatments for pollutants bioremediation includes *Monoraphidium* sp., *Chlorella* sp., and *Scenedesmus* sp. [88,165,172].

4. Reuse of produced water

Since the demand and production of oil and gas is continuing to increase globally, the environmental footprints associated with this production are increasing, such as produced water. Furthermore, as the scarcity of freshwater supply is increasing, produced water can be a crucial source of water after suitable treatment. There has been an increased attention on reclaiming, reusing, and recycling of water that is usually wasted to meet the communities' needs of freshwater source [173]. Different standards for reuse of treated water have been provided based on intended purpose. The US-EPA provides standards for reuse of treated water as drinking water [54]. In addition, standards for reuse in irrigation and for livestock has been provided by US Department of Agriculture Natural Resources Conservation Service [174].

Table 5
Standards for water reuse for drinking, irrigation and livestock purposes [174,175].

Component	Drinking (g/ m^3)	Irrigation (g/ m^3)	Livestock (g/ m^3)
Li^+	–	2500	–
K^+	–	–	–
Na^+	200	Based on SAR	2000
NH_3	1.5	–	–
$Ca2^+$	–	Based on SAR	–
Mg^{2+}	–	Based on SAR	2000
Br^-	–	–	–
Cl^-	250	–	1500
HCO_3^-	–	–	–
SO_4^{2-}	250	–	1500
TDS	500	2000	5000
Conductivity (dS/m)	–	2.5	1.5–5
Sodium adsorption ratio (SAR)	–	0–6	–

Table 5 shows standards for water reuse based on different purposes. As expected, the standards for drinking water are more stringent and therefore, more extensive treatment of produced water is needed.

There are several alternatives for utilization of produced water such as drinking water, irrigation, livestock watering, habitat and wildlife watering, fire control, and industrial uses such as dust control, oil field uses, and power generation. Based on the characteristics of produced water discussed previously, treatment of the produced water is required to meet the quality standards before re-using it. The treatment level or degree required depends on the application that it will be reused in, for example, minimal treatment regime is required for using produced water in oil and gas and industries, as well as dust control. On the other hand, higher treatment level is required for uses such as drinking water and agriculture [176]. Furthermore, cost efficiency is an important factor that determines the treatment regime and reuse option. For example: it was calculated that the produced water treatment with traditional techniques such as hydrocylcone, gravity separation and media filters will yield treated water suitable for re-injection at the cost of $\$0.509/m^3$ of water. However, for recycling of produced water, advanced desalination technique like Mechanical vapor compression (MVC) with other suitable technique will yield recyclable water at the cost of $\$3.808/m^3$ of water [177].

4.1. Livestock watering

Water quality consumed by livestock usually have lower standards than the quality of water for human consumption as the contaminant tolerance of livestock is better than humans. However, contaminants present in the water used by animals should be under certain limit to avoid negatively affecting their health [3]. For instance, water with < 1000 mg/l of TDS can be used as a water source for livestock. However, it can affect the health of livestock by causing diarrhea if the value exceeds 7000 mg/l [109]. This idea was applied in some projects of CBM in which they established watering stations for livestock to utilize produce water as drinking water [3].

4.2. Habitat and wildlife watering

Produced water can be used after semi-intensive treatment and ensuring its harmless nature to create artificial reservoir for providing drinking water source for wildlife as well as offering habitat for waterfowl and fishes. These impoundments can collect and retain produced water in large volumes as they have large area of several acres [178].

4.3. Irrigation

After the treatment of produced water and removal of all pollutants, if the quality of treated water is meeting with certain standards and having low enough TDS, we can consider produced water as a valuable resource for irrigation of crops [178]. Reuse of produced water for irrigation especially in dry lands has been recommended and reviewed [179]. Due to high content of salts (TDS = 35–472,000 mg/l, Na = 3–435,000 mg/l); its reuse for irrigation remains a challenge [179]. The main challenges include sodicity, salinity, specific ion toxicity, and alkalinity which are magnified because of the lower produced water quality, therefore, it is very crucial to consider the crop type when using produced water for irrigation [78].

A research was done by Sirivedhin et al. [178], to treat the synthetic solutions with variable TDS concentrations to mimic produced water using electrodialysis technique. Results showed that all 3 samples with low TDS concentration meet the standards of both livestock watering and drinking water. However, they had higher SAR values than the standard values for irrigation use, but this problem can be solved by the addition of Ca^{2+} and/or Mg^{2+} to these waters in small amounts in order to decrease their SAR value and make them usable for irrigation. On the other hand, water samples with high TDS concentrations did not meet the standards for reuse highlighted in Table 5. Therefore, it can be concluded that electrodialysis under the experiment operating conditions is an inadequate process for reclaiming produced water to be used in these applications.

4.4. Algae production

There are limited number of studies that reported the growth rate of algae, productivity or biomass yield, and lipid production by algal strains grown in produced water as nutrient medium. However, to date, Godfrey [180], conducted the most comprehensive study for using produced water as a medium to grow microalgae for the aim of producing lipid for biofuel conversion. Total of 8 microalgae strains were grown successfully in produced water and all produced neutral lipids. *Amphora coffeiformis* optimized its growth and lipid production with 150 mg/l sodium nitrate and without the addition of phosphate, while 300 mg/l of sodium nitrate were added to optimize the growth and high lipid productivities of *Chaetoceros gracilis*, and *Chlorella* sp. Moreover, pretreatment of produced water for the removal of hydrocarbon through filtration by activated carbon, centrifugation, or settling, can increase the growth and lipid productivity. Results also showed that there is a remarkably higher biodiesel productivity of strains grown on produced water as the lipid content reached 63% and 63.8 mg biodiesel/l/day. Thus, it was concluded that replacement of growth media with produced water supplemented only with essential nitrogen and phosphorus nutrients is more environmentally and economically logical because similar lipid and biomass productivities can be achieved with lower cost without the requirement of freshwater.

5. Case studies (produced water treatment and reuse)

5.1. Reuse of produced water for microalgae production (Qatar)

The produced water from one of the natural gas field in Qatar was collected by the research team of Al-Ghouti et al. (unpublished data) and was used to investigate the removal of heavy metals using microalgae. The produced water samples collected were first filtered using 0.45 μm Millipore filter to remove most of the suspended solids and other major pollutants (Table 6). The filtered water was then used to grow various species of microalgae to investigate their capabilities to remove heavy metals. Table 6 shows the ability of species to remove various metals from produced water.

As shown in Table 7, 100% removal efficiency of Al, Zn, and Fe from produced water was achieved through microalgae, while K had the lowest removal efficiency of 11.27% [181]. According to Cai et al. [182], toxicity of the produced water could increase with the increase in the concentration of these elements. According to the results obtained in this research, *Dictyosphaerium* sp. can recover more elements since it had better growth than other species. Our findings agreed with previous researches [183,184] which shows that *Dictyosphaerium* sp. has the ability to grow within metal rich water.

In this research, among 14 metals were found to be present in the collected produced water, almost half of them are considered as micronutrients, such as K that plays an important role in various enzymatic reactions [185]. Along with K, Fe and Cu have crucial role in the photosynthetic electron transport system [186], while Zn is utilized by the microalgae through the transcription process of DNA and uptake of phosphorus [187]. On the other hand, Monteiro et al. [188], mentioned that certain metals such as Cd and Cr could negatively affect the cell division and decrease the photosynthetic ability if present at high concentration. According to Millach et al. [189], intensity of Chlorophyll a in *Scenedesmus sp.* could significantly decrease if Cr is available in concentration higher than 0.75 ppb. Unlike *Scenedesmus* sp., some species were found to tolerate higher concentrations of Cr, such as *Dictyosphaerium* sp. that can tolerate up to 13–17 mg/l, and *Chlorella pyrenoidosa* that can tolerate up to 2 mg/l [190,191]. Furthermore, high biomass yield of *Scenedesmus* sp., *Dictyosphaerium* sp., and *Chlorella* sp., was observed in this study, which can be attributed to the low Cr concentration present in the tested produced water. This case study shows that the produced water after minimal treatment, can be used for microalgae production. Although, microalgae can help to remove certain metals from the produced water. Nevertheless, the effect of various constituents of produced water on the growth of microalgae needs to be investigated.

Table 6
Characteristics of produced water collected from Natural Gas field in Qatar.

Parameters	Characteristics of produced water	
	Raw produced water	Filtered water
Total organic carbon (mg/l)	389.1	317
Total nitrogen (mg/l)	35.77	27.6
Total phosphorus (μg/l)	277.78	180
Benzene (mg/l)	21	16.1
Toluene (mg/l)	3.8	3.21
Ethylbenzene (mg/l)	1.22	1.05
Xylene (mg/l)	3.43	3.11

Table 7
Removal of trace metals from produced water using Microalgae.

Trace metals	Feed water (ppb)	Filtered water (ppb)	Microalgae species	% Removal
K	736.18×10^2	677.40×10^2	*Scenedesmus* sp.	11.27
Mg	417.15×10^2	392.57×10^2	*Dictyosphaerium* sp.	13.9
Sr	111.98×10^2	105.73×10^2	*Dictyosphaerium* sp.	21.23
B	425.9×10^2	374.7×10^2	*Dictyosphaerium* sp.	20.23
Mn	318.56	318.56	*Neochloris* sp.	87.80
Cu	224.97	180.78	*Dictyosphaerium* sp.	91.65
Fe	287.94	100.19	*Neochloris* sp.; *Chlorella* sp.	100
Ba	55.69	43.35	*Monoraphidium* sp.	13.06
Cr	24.09	17.20	*Dictyosphaerium* sp.	19.36
Al	114.41	13.68	*Neochloris* sp.	100
–	–	–	–	–
Ni	7.83	3.71	*Dictyosphaerium*	92.29
V	1.87	1.46	*Scenedesmus*	36.26
Cd	0.09	0.06	*Chlorella*	97.37

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5.2. Reuse of produced water for internal reuse in gas industry (China)

China possess the largest shale gas resources in the world with Sichuan Basin being the largest shale gas play [56]. The volume of produced water generated from each well is approximately 20,000 m^3 [49], which is even higher than the volume generated in the US [192]. However, the TDS values in this produced water [193,194] has been found to be lower than the shale plays in the US [76,195,196]. Due to the better quality of produced water and its higher volumes, the feasibility to reuse this water within the industry was studied [197]. The reuse of produced water within the industry such as for fracturing operations has been gaining more attention [198] because it can reduce both the volume of wastewater as well as consumption of freshwater resources. Hence, the integrated treatment technique comprising coagulation followed by UF and NF was investigated for its suitability to meet the standards of reuse.

In this case study [197], two different types of coagulants (i.e. aluminum sulfate octadecahydrate and ferric chloride hexahydrate) were used. The coagulated water was then pumped to the UF system. The UF membrane used was of molecular weight cut-off (MWCO) 100 kDa and was operated at a constant flux of 50 $l/m^2/h$ with backwashing for 5 min after every one hour. The UF filtered water was then further treated with NF membrane at different pressures of 100, 200, 300, and 400 psi. The NF system was operated at the water recovery of 50, 70, and 85%.

Thus, the integrated coagulation-UF-NF treatment system reduced the turbidity by 99.9%, COD by 94.2% and alkalinity by 94.1%. The cations such as calcium, magnesium, barium and strontium were removed by 72–83% as shown in Fig. 3. When compared with the standards for reuse (Table 8), it was found that all the parameters of treated water obtained through integrated treatment system are much lower and therefore, it can be reused within the industry.

Table 8
Standards for reuse within the industry considered in this case study [199].

#	Parameter	Recommended values (maximum) (mg/l)
1	TSS	50
2	Total hardness	2500
3	Total alkalinity	300
4	TDS	50,000–65,000
5	TOC	< 25
6	pH	6–8
7	Chloride	45,000
8	Sulfates	50
9	Calcium	8000
10	Sodium	36,000
11	Potassium	1000
12	Others (iron, barium, strontium)	10

5.3. Reuse of produced water for irrigation of biofuel crops (USA)

The volume of produced water is among the highest in USA in the world. It has been estimated that around 90% of the produced water is being injected back to the wells [200,201]. However, this practice of produced water re-injection is being linked with the seismic activities and frequent earthquakes in Texas as the area contains relatively larger number of injection wells [202,203]. The growth rate of biofuel production is projected to increase [204], which is also water-intensive process. Therefore, the use of produced water for irrigation in the region can help to reduce the consumption of freshwater resources. However, the presence of hydrocarbons, organic matters and high salt concentrations are the major parameters that need to be considered for reuse of produced water in irrigation.

Thus, in this case study, the effect of produced water on biofuel crops was studied [205]. The switchgrass and rapeseed were chosen as they can tolerate broad range of salinity, pH and temperature conditions [206,207].

The produced water from central processing facility in the Denver-Julesburg Basin was used that serves more than 500 wells and contains higher TOC and TDS concentrations both higher than 20,000 mg/l. The effect of salinity and organic matter concentrations were studied at highest and lowest concentrations to cover both worst- and best-case scenarios.

The produced water was treated with different techniques to achieve different quality of water. As a control, good quality reservoir water (TDS–310 mg/l, TOC–3.3 mg/l) was used for irrigation (designated as irrigation water A). The best quality of treated water was obtained through treatment with coagulation and ultrafiltration followed by adsorption through granular activated carbon. The effluent water was then diluted using tap water to obtain the TDS of 400 mg/l and TOC of 3 mg/l (irrigation water B). Similarly, the dilution was done to obtain another two qualities of water with both having TDS of 3500 mg/l, but with different TOC concentrations i.e. 65–100 mg/l (low TOC) and 215–235 mg/l (high) designated as irrigation water C and D, respectively. Furthermore, the produced water was pre-treated using hydrogen peroxide oxidation followed by coagulation and ultrafiltration to obtain treated water with both relatively higher TDS (15,000–20,000) and TOC (1500 mg/l) concentrations (irrigation water E). The aim of choosing different water qualities was to determine water with minimal quality and treatment required to obtain suitable

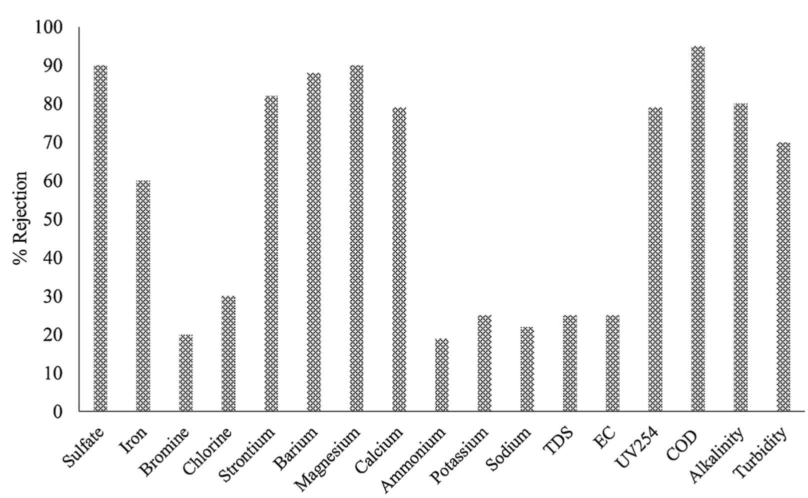

Fig. 3. Removal efficiency of integrated coagulation-UF-NF treatment system [198].

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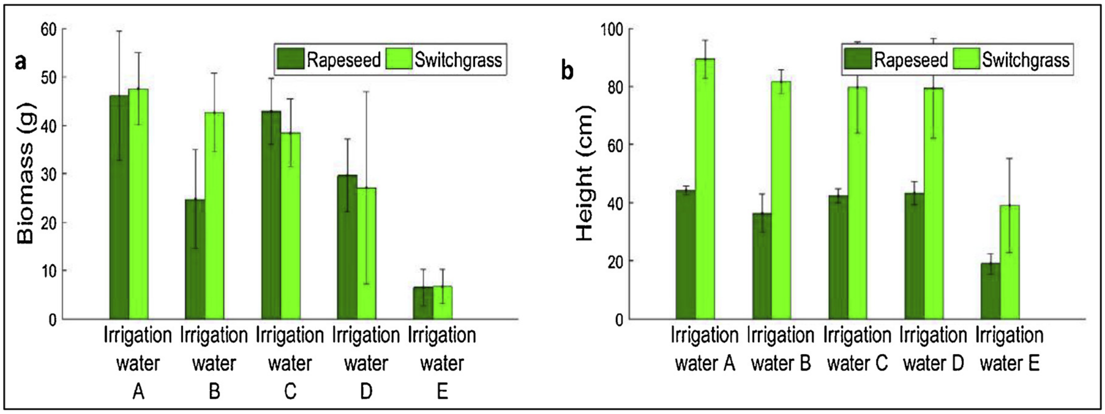

Fig. 4. Effect of water quality on mean plant biomass yield (a), root weight (b) and height [205]. (Irrigation water (A) water used as control with TDS – 310 mg/l and TOC – 3.3 mg/l, (B) high quality treated water with TDS – 400 mg/l, TOC – < 5 mg/l, (C) medium quality treated water with TDS – 3500 mg/l, low TOC – 65–100 mg/l. (D) Medium quality treated water with TDS–3500 mg/l, high TOC–215–235 mg/l. (E) Pre-treated water with high TDS – 15,000–20,000 mg/l, high TOC–1500 mg/l.).

biomass growth using produced water.

As expected, the plant growth and subsequently biomass yield decreased for both biofuel crops (switchgrass and rapeseed) with decrease in water quality due to higher salinity and organic matter concentrations (Fig. 4). The difference between the plant growth and biomass yield obtained with irrigation water having highest TOC and TDS was statistically significant as compared to the irrigation water with lower concentrations. The quality of water was found to be significantly correlated with the accumulation of salts in soil samples. The salinity of the water was found to be very important characteristic for the growth of perennial energy crops. The physiological characteristics of crops i.e. biomass production on ground and underground, were significantly affected by the irrigation water of TOC of nearly 232 mg/l when compared with the one having low TOC of around 38 mg/l. Thus, higher concentrations of organic matter were shown to affect the EL (electrolyte leakage) value of the leaf which confirmed that the crops are sensitivity to TOC levels in produced water. Hence, both the salinity and the organic matter are important characteristics of produced water which needs to be considered for their reuse in irrigation of biofuel crops.

It was concluded from this case study that the produced water should be treated enough to reduce the TOC to 3500 mg/l and to lower content of organic matter in order to consider its reuse for irrigation. It was also concluded that the extent of treatment to reduce TOC and the organic matter content is optimum from feasibility, scalability and cost point of view. This holds true for the treatment of produced water as one-step or two-step treatment methods are not considered enough to meet the reuse standards [208,209].

Since, the production of produced water in oil and gas industry is an inherent problem, more research is needed to develop techniques to reduce the volume of its production. Jafari et al. [210], studied the ways to reduce the water production in oil and gas wells. It was concluded that using newly designed equipment, adopting production rates lesser than the critical rates, and plugging wells can help to control the volumes and impacts of produced water.

6. Conclusion

Considering the shortage of fresh water resources and environmental problems caused by Oil and Gas industry, produced water treatment and reuse are generating immediate attention. The detailed literature review showed that the characteristics of produced water widely differs from location to location. Therefore, the treatment technique needs to be evaluated for a particular source of produced water. The cost of treatment and aim of reuse can also affect the choice of treatment technology. In literature, numerous techniques and combination of different techniques were found to be used for produced water treatment. However, practical demonstration of reuse of produced water for purposes such as for irrigation or microalgae growth and others is seldom reported. In order to meet the rising water demands and to reduce the environmental impacts of oil and gas industry, it is dire requirement now to promote the reuse of produced water. Its reuse will not only reduce the environmental impacts of the industry but also reduce the stress on fresh water resources. This is particularly important for water stress countries, where, population and economic growth is continuing to increase stress on limited water resources of the region.

Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Produced Water Beneficial Use Case Studies

Case Stusies Document (posted 6/14/2011).

Produced water can have a variety of beneficial uses, facilitating augmentation of fresh water resources, providing water for livestock and wildlife, for irrigation in arid regions, and for industrial applications. The following is a summary of recent and current beneficial use practices in the U.S.

Groundwater Augmentation

In Colorado, produced water from oil wells near Wellington has been treated as a raw water resource to augment shallow water aquifers[1]. The Wellington Oil Company, operating in Larimer County, Colorado, reported that 98.5% of its fluid production is produced water. The company utilized a deep injection well that re-injected the produced water into the underground formation from which it was pumped at a cost of approximately $1 per barrel[2]. The company needed to find an efficient and cost-effective way to manage produced water because their ability to dispose of the water has a direct impact on how many pumps can be online and thus how much oil they can recover. Therefore, the company embarked on a groundwater augmentation project to increase oil production. The steps to beneficial use of the water included[3]:

- Pilot testing of water treatment processes to demonstrate water quality target and treatment process efficiency
- Determination of non-tributary status of the groundwater by State Engineer
- Water Quality Control Division permit assessment
- Issuance of permit by Colorado Oil and Gas Conservation Commission
- Construction of the water treatment plant
- Completion of an RO drinking water treatment plant

The produced water at Wellington is treated through dissolved air floatation, pre-filtration, ceramic microfiltration, and activated carbon adsorption. The treated water is piped 4,000 feet to the groundwater recharge site, which is a rapid-infiltration pit that allows the treated produced water to percolate into a tributary aquifer. The shallow aquifer supplies water to a reverse osmosis plant that provides drinking water to the Town of Wellington and northern Colorado water users. The Wellington project provides environmental benefits, reduced waste disposal costs, and increased water supply; however, it required a significant financial investment as described in[1].

Under Colorado water law, one of the legal hurdles for the Wellington reuse project was to identify which agency should issue the discharge permit. In this case, the Attorney General's (AG) office decided that the Colorado Oil and Gas Conservation Commission (COGCC) should issue the permit instead of the Colorado Water Quality Control Division (CWQCD). The COGCC was considering whether to promulgate new rules to accommodate projects like Wellington in the future.

IOGCC and ALL described an example in which CBM produced water has been injected into the aquifer of a city's well field[4]. In the example, the City of Gillette, WY, had depleted its local wells by unbalanced pumping for many years[5]. The well field was completed in Lower Fort Union sands at a depth of approximately 1,500 feet.

The city coordinated with a CBM production operator to install Class V aquifer recharge wells with the capacity equals to the produced water flowrate from a small CBM producing project.

Irrigation

Irrigation with produced water has become an attractive alternative for producers to manage and use water that otherwise will require an NPDES discharge permit. Thousands of acres in the Powder River Basin have been transformed to productive agricultural land using CBM produced water[6]. Most CBM product water is characterized as sodic water and is high in the sodium (Na^+) concentration relative to concentrations of calcium (Ca^{2+}) and magnesium (Mg^{2+}). Excess sodium can lead to soil dispersion and loss of soil infiltration capability[7]. Some trace elements in produced water (e.g., boron) can harm plants when present in elevated concentrations.

ALL Consulting reported two examples from Wyoming[8]. The first project was conducted by Fidelity Exploration and Production. They irrigated livestock forage using only CBM produced water on some plots, and a blend of surface water and CBM produced water on other plots. Both tests resulted in adequate crop production; yet, the CBM produced water had to be applied at a higher rate because the plants did not utilize it as efficiently as the surface water blend. The second project was conducted by Williams, a CBM producer. Large areas were irrigated that previously had supported only the local drought-tolerant vegetation. Following irrigation with CBM produced water the land was able to support healthy grass crops that served as feed for livestock. Between watering intervals, Williams applied gypsum and other soil supplements to counteract the high SAR in the produced water.

DeJoia described a successful managed irrigation project[9]. After two years of applying soil amendments and CBM produced water, the test sites were converted from overgrazed range land to highly productive grasslands yielding livestock and wildlife benefits.

Paetz and Maloney[17] reported on a project where 12,500 bbl/d of CBM produced water was used to irrigate 100 acres of arid land in the Powder River basin to produce a forage crop. The carefully managed approach resulted in the successful production, harvesting, and sale of the forage crop.

Operators intending to use CBM produced water without causing long-term harm to crop and soil employ managed irrigation. This technique involves careful monitoring of soil chemistry. Different soil supplements are added to provide the necessary chemical and mineral balance[10]. CBM produced water can be neutralized with an acid like sulfur. Calcium amendments such as gypsum can be added to offset the sodium-rich water. Adding sulfur and gypsum is equivalent to adding standard fertilizers because these standard fertilizers often contain the same elements.

More than 30 options exist under the category of managed irrigation. Several automatic and manual irrigation systems are available, including: center pivot sprinklers, side-roll/wheel line sprinklers, hand-moved or fixed solid set sprinklers, big gun sprinklers, surface drip, subsurface drip, gated pipe flood and ditch flood[6]. Ranchers are commonly using center pivot and side-roll sprinklers.

More recently, the use of subsurface drip irrigation method has grown rapidly. BeneTerra, LLC, a water management company, has developed subsurface drip irrigation technology to disperse CBM produced water[11]. The system works by evenly applying small amounts of water over a large surface. Produced water is filtered, treated, and pumped through a labyrinth of polyethylene tubing which spreads it uniformly across the land. The tubing has emitters attached to the inside, which regulate flow from openings on the tubing. It is placed in the soil with a chisel plow to depths ranging from 18 to 48 inches. Plants derive moisture from roots in the subsoil while the topsoil remains relatively dry. Haying operations can continue while the field is being irrigated. Heavy equipment cannot compact dry soil and weeds cannot germinate. BeneTerra irrigation systems are designed to utilize the native calcium and magnesium already present in the soil to offset the effects of sodium. Then the salts percolate to a lower depth in the soil. Because the CBM water is naturally warmer, freezing is not an issue. This system provides year-round water dispersal for energy operators while producing bountiful crops.

Livestock Watering

Some CBM projects on ranch land have created impoundments or watering stations to provide CBM produced water as a drinking water source for livestock[7]. ALL Consulting reported the results from tests at the 7 Ranch near Gillette, Wyoming, where livestock are watered from small reservoirs containing CBM produced water[8].

Animals can tolerate a wide variety of water quality. However, highly saline water or water containing toxic elements may be hazardous to animals and may even render the milk or meat unfit for consumption. In evaluating the usability of any particular water, a number of factors should be considered, including local conditions, availability of alternate supplies, source water quality, seasonal changes, age, species, and health conditions of the animals, and composition rate[12].

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Wildlife Watering and Habitat

Some CBM projects collect and retain large volumes of produced water in impoundments that may have surface areas of several acres. The impoundments provide a source of drinking water for wildlife and offer habitat for fish and waterfowl in an otherwise arid environment. It is important to ensure that the quality of the impounded water will not create health problems for the wildlife[8]. ALL Consulting[1] also presents information on siting and construction of wildlife watering impoundments.

Fisheries

ALL Consulting reported that untreated CBM produced water is currently being used to sustain privately owned fishponds in some states, including Wyoming[8]. Meanwhile the State of Wyoming discontinued fish stocking programs in certain ponds due to a general lack of available water volume needed to sustain the systems. CBM produced waters are now being beneficially used to supplement the ponds, allowing for continuation of the State's stocking program. The application and success of this use would depend on applicable state guidelines, public demand, water quality, drainage, and geographic region. Water rights rules and regulations vary by state; it is important to determine local water rights as they apply to this management option.

Constructed Wetlands

Constructed wetlands are an alternative treatment and use option for oil and gas produced water[13]. Constructed wetlands could increase wildlife distributions, reduce displacement, and enhance diversity by improving quality habitat[2]. Research sponsored by Marathon Oil Company in 2000 involved construction of an artificial sedge wetland system to treat CBM produced water. The purpose of the project was to determine if constituents found in CBM produced water, mainly iron, barium, and unbalanced SAR, could be treated cost-effectively. A report by Montana State University further supported the results, concluding that "clean water is needed to supplement sodicity and saline treatment by vegetation and soil[14]."

Wetlands may have significant ecological and environmental impact. They provide areas that can be utilized by wetland birds and animals and aquatic life. Wetlands can also be utilized for livestock and wildlife watering purposes[15]. On the other hand, the contaminants in CBM produced water may affect fish and wildlife. For example, the research conducted by the USGS has demonstrated acute and chronic sodium bicarbonate toxicity to aquatic species. CBM produced water discharges containing selenium in concentrations above 2 μg/L may cause bioaccumulation in sensitive species[16]. In addition, if the wetlands are constructed as part of direct discharge, they will change habitat from increased flows and increased erosion. Impacts to downstream users due to direct discharges would be comparatively higher with increased flows during traditional low flow periods and increased sedimentation from erosion.

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water

Review

Advances in Produced Water Treatment Technologies: An In-Depth Exploration with an Emphasis on Membrane-Based Systems and Future Perspectives

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Abstract: This comprehensive review focuses on treatment technologies for produced water, with a particular emphasis on membrane-based systems. These systems offer significant advantages, including high contaminant removal efficiencies, compact design, and the potential for resource recovery. The review emphasizes the application of these technologies, their performance in meeting regulatory standards, and the challenges they face, such as operational efficiency and fouling. It highlights the need for further research and for the optimization of processes to enhance their efficiency. The integration of conventional methods with advanced treatment processes is also explored, with a vision toward developing hybrid systems for improved treatment efficiency. Overall, membrane-based systems show great promise for the treatment of produced water, but further advancements, sustainability considerations, and integration with other technologies are essential for their successful implementation in large-scale applications.

Keywords: produced water; membrane bioreactors; fouling; desalination; oil-water separation; electrodialysis

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1. Introduction

The petroleum industry generates approximately 250 million barrels per day of produced water, making it a substantial byproduct and the primary waste stream in terms of volume [1–3]. It is also referred to as formation water, which coexists with petroleum in the reservoirs of the Earth's crust. This water accumulates alongside hydrocarbons, as illustrated in Figure 1. Petroleum reservoirs can be categorized into conventional and unconventional types. Conventional reservoirs involve the entrapment of naturally occurring hydrocarbons such as natural gas and crude oil, by impermeable rock formations situated above them. Conversely, unconventional reservoirs are characterized by rocks possessing a low permeability and high porosity, which effectively confines the hydrocarbons in place and eliminates the need for a cap rock. The composition of produced water is indeed complex, consisting of various important components. These include oil in both dissolved and dispersed forms, organic and inorganic compounds, hydrocarbons, carbon dioxide,

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hydrogen sulfide, total dissolved solids (TDS), total suspended solids (TSS), inorganic salts, and heavy metals, among others [4]. Table 1 describes the quality of produced water based on reported studies. Hence, discharging untreated produced water into the environment can result in significant environmental impacts, affecting both human health and aquatic life.

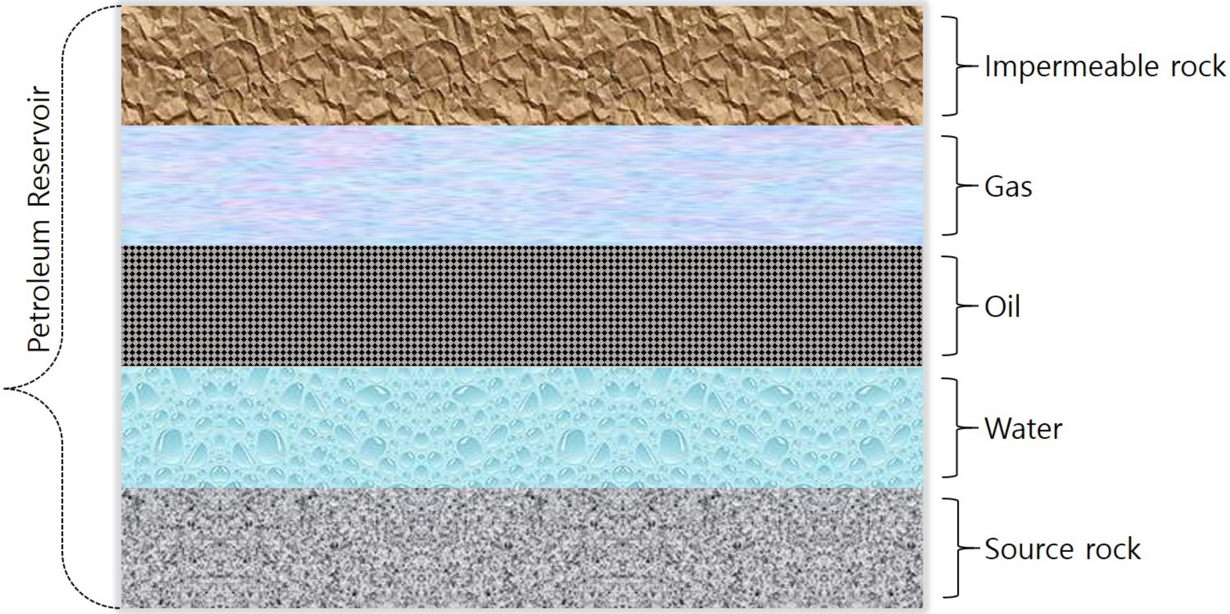

Figure 1. Schematic illustration of a representative reservoir.

The organic compounds present in produced water can cause harmful consequences, including increased resistance to biodegradation, carcinogenic properties, and high toxicity to marine life [5]. Exposure to these compounds can lead to various health issues such as tumors and cancer. Additionally, produced water can have endocrine-disrupting effects, affect non-endocrine systems, and impact the reproductive system [6,7]. It is crucial to properly treat and manage produced water to mitigate these environmental and health risks. Therefore, strict regulatory standards have been established to govern the discharge of produced water into the environment. Various technologies are employed to treat and purify produced water [8]. These technologies can be categorized into biological, chemical, and physical methods, including adsorption, microbial degradation, filtration, ion exchange, chemical oxidation, electrochemical oxidation, gas flotation, coagulation, photocatalysis, and membrane separation, among others [9–14].

The literature indicates that coalescence and destabilization mechanisms play a crucial role in achieving efficient oil-water separation, particularly for fine-sized oil droplets. Additionally, chemical demulsification has been recognized as a suitable and highly efficient method for oil-water separation operations [15]. Hydrocyclones have established their effectiveness in reducing the oil content present in produced water. However, they are often plagued by issues such as clogging and fouling [1]. Pressure-driven membrane-based processes have emerged as promising techniques for the treatment of produced water. However, membrane fouling represents a significant drawback associated with membrane-based processes, as it can progressively reduce their efficiency over time [16,17]. Additionally, the high energy costs associated with membrane operations are another limiting factor that needs to be addressed [2].

Table 1. Quality of produced water. The table is prepared based on data reported in earlier studies [18–20].

Parameters	Range	Unit
Conductivity	4200–58,600	(µS/cm)
pH	4.3–10	-
Density	1014–1140	(kg/m^3)
Turbidity	182	(NTU)
Surface Tension	43–78	(dyne/cm)
COD	1220	(mg/L)
TOC	0–1500	(mg/L)
TSS	1.2–1000	(mg/L)
Total oil	2–565	(mg/L)
Volatiles	0.39–35	(BTEX; mg/L)
Petroleum hydrocarbon (total)	>20	(TPH)
Non-volatile oil and grease (total)	275	(µg/L)
Bicarbonate	77–3990	(mg/L)
Chloride	80–200,000	(mg/L)
Sulfate	<2–1650	(mg/L)
Volatile fatty acids (VFA's)	2–4900	(mg/L)
Sodium	132–97,000	(mg/L)
Calcium	13–25,800	(mg/L)
Potassium	24–4300	(mg/L)
Lithium	3–50	(mg/L)
Iron	<0.1–100	(mg/L)

Membrane bioreactors (MBRs) have demonstrated an excellent treatment performance for removing both inorganic and organic components in comparison with physio-chemical processes and trickling filters. MBRs combine the biological treatment of wastewater with membrane filtration, resulting in enhanced removal efficiencies. The membrane barrier effectively separates suspended solids, microorganisms, and contaminants, resulting in higher treatment efficiency. This makes MBRs an attractive option for producing high-quality treated water, surpassing the performance of traditional physio-chemical processes and trickling filters [21–23]. However, it is worth noting that the majority of published reviews have focused on the generalized performance of MBRs for normal wastewater. In contrast, there has been relatively less attention given to the application of MBRs for the treatment of produced water [24].

The objective of this review is to provide an updated overview of the treatment systems employed for produced water, highlighting their advantages, disadvantages, limitations, and efficiency. Specific attention will be given to the application of these systems to different types of produced water. The review will also identify research gaps in the development of technologies for purifying oilfield-produced water. Furthermore, this article aims to assess the effectiveness and performance of membrane-based technologies, particularly membrane bioreactors and electrodialysis, for the treatment of produced water. By evaluating these membrane-based processes, their suitability and efficiency for produced water treatment will be estimated. Overall, this review seeks to offer valuable insights into the current state of produced water treatment systems, address research gaps, and evaluate the potential of membrane-based technologies for the purification of produced water.

2. Treatment Technologies for Produced Water

The primary objective of treating produced water is to address various aspects such as the removal of oil content (de-oiling), desalination, elimination of suspended particles and sand, removal of organic and inorganic components, elimination of dissolved gases, and extraction of heavy metals [25]. To achieve this objective, a range of treatment processes, including physical, chemical, biological, integrated, and membrane-based methods, are employed. Table 2 provides an overview of the different operations and technologies used in physical, chemical, biological, and membrane-based treatment processes. The table also highlights the effectiveness of these methods at removing specific constituents present in produced water.

2.1. Conventional Treatment Approaches

2.1.1. Adsorption

Adsorption is the most common water treatment technology [26]. The influential factors of adsorption are attached to the surface tension of solutions, temperature, nature, and quantity of the adsorbed substances [27]. Normally, no chemicals are required for adsorption [28]. Adsorption exhibits a remarkable efficiency of over 80% when employed to eliminate oil, total organic carbon, and heavy metals that are found within produced water [29]. Various materials, such as organoclays [30], zeolites [31], chitosan, and activated carbon [32], are used for adsorption. Numerous studies have provided evidence of the successful utilization of magnetic-based materials in the treatment of wastewater, showcasing their effectiveness at removing various contaminants such as heavy metals, organic pollutants, dyes, and pharmaceutical compounds [33,34].

Natural superwetting materials offer a promising solution for addressing the global issue of oil contamination due to their affordability, environmentally friendly properties, and widespread accessibility. Utilizing natural materials for oil-water separation holds great potential in tackling this challenge that has been widely recognized worldwide [35]. Coconut pith (CP), olive leaves powder (OLP), and eggplant peel powder (EPP) are utilized as adsorbents for the effective removal of oil and metals [36,37]. A study employed δ-Bi_2O_3 for the removal of bromide from aqueous solutions that contain low concentrations of chloride [38].

Akhlamadi et al. presented a sustainable solution for oil-water separation by introducing a superhydrophobic cellulose nanocrystal-based aerogel derived from waste tissue paper [39]. The aerogel exhibited a remarkable sorption capacity ranging from 69 g/g to 168 g/g for six different oils and eight organic solvents tested. In addition, the reusability experiments demonstrated that the aerogel maintained more than 92% of its sorption capacity even after undergoing 20 cycles of sorption squeezing. This indicates the excellent reusability and durability of the aerogel for oil-water separation applications. Azad et al. developed a hydrophobic and superoleophilic adsorbent by applying a coating of candle soot onto the surface of a recycled egg carton material [40]. The resultant carbon-coated adsorbent demonstrated an exceptional oil absorption capacity across a wide range of densities, eliminating the necessity for pre-treatments or surface modifications. The developed adsorbent exhibited successful absorption of various oils including diesel, engine oil, petrol, coconut oil, mustard oil, and refined oil. The adsorbent displayed a remarkable maximum absorption capacity of 3 g/g, indicating its high effectiveness in oil absorption.

Yu et al. utilized waste plastic to produce alveolate polystyrene (PS) foam [41]. The synthesis of the PS foam was achieved through a one-step process employing a high internal phase Pickering emulsion technique. The emulsion was effectively stabilized by a co-Pickering system comprising SiO_2 particles and Span 80 surfactant. The resulting SiO_2@PS foam displayed a unique multi-order pore structure and possessed superhydrophobic and superoleophilic properties. This made it highly effective at selectively removing various oily contaminants from water. The SiO_2@PS foam demonstrated an excellent adsorption capacity ranging from 20.4 g/g to 58.1 g/g (Figure 2), and it achieved rapid adsorption rates. One of the notable advantages of the SiO_2@PS foam was its reusability. After oil adsorption, the material could be easily reused through a simple centrifugation process. Even after 10 cycles, the decline in oil adsorption capacity was less than 1%, indicating its robust and durable performance. Overall, the SiO_2@PS foam developed by Yu et al. exhibited great potential for the treatment of oily water. Its superhydrophobic and superoleophilic properties, high adsorption capacity, and reusability make it a promising material for applications in oily water treatment and remediation.

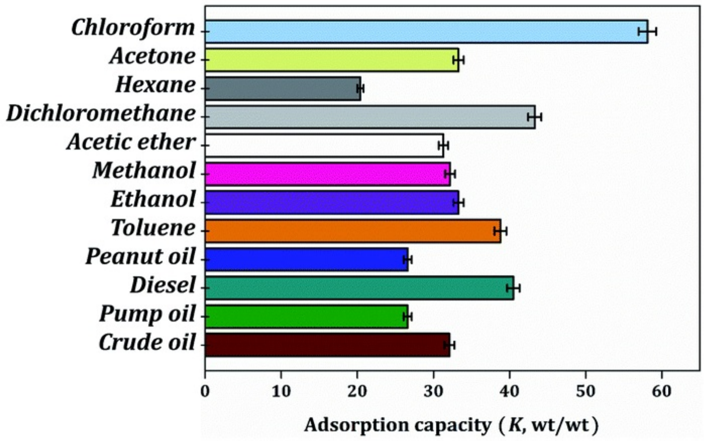

Figure 2. Different oil adsorption capacities of SiO_2@PS foam [41]. Image reused under the Creative Commons attribution license.

The performance of the adsorbent is influenced by temperature, pH, suspended solids, salts, and oils. The presence of suspended oil particles in produced water can lead to plugging of the medium and minimize the efficiency [42]. The usage rate of the adsorption medium directly impacts the operational cost of the adsorption treatment. During the replacement process of adsorption agents, disposal of solid waste becomes necessary [43,44]. Typically, this process is used in combination with other units, rather than alone. The combination of organoclays and activated carbon is efficient for the elimination of total petroleum hydrocarbons [45]. EARTH (Canada) Corporation has successfully developed a multistage process dedicated to the recovery of dispersed oil droplets found in produced water, specifically targeting those with a size greater than 2 microns [46]. The efficiency of adsorbents can vary under different conditions [27].

2.1.2. Cyclonic Separation

The cyclonic separation processes are also well-known in oil-water separation. A hydrocyclone is a device used to separate solid particles and/or immiscible liquids from a liquid, typically water. It is named hydrocyclone because the liquid (water) is considered the primary phase [47]. The principle of density difference between two liquids and/or a liquid and solid is employed in hydrocyclones. It is a physical technique where the strong rotational motion creates a radial acceleration, serving as the separating force. Hydrocyclones are constructed using metals, plastics, and other materials. They typically have a conical base and a cylindrical top, and their performance is intricately tied to the angle of the conical section [48]. The liquid stream is tangentially fed from the top, resulting in two discharge streams: one located at the top, known as the overflow or product stream, and another at the bottom referred to as the underflow or reject stream. The top stream is utilized for discharging the lighter phase of the input stream, while the bottom stream is used to discharge the heavier phase [49].

Produced water usually consists of oil droplets, surfactants, and suspended solids. Hydrocyclones, depending on the specific model used, are capable of removing particles within the size range of 5 to 15 μm. However, it should be noted that hydrocyclones are ineffective at removing soluble constituents present in produced water. Many companies utilize hydrocyclones for the treatment of produced water, with a capacity of treating approximately 8 million barrels per day [48]. Hydrocyclones operate without the need for any chemicals, making them highly efficient and cost effective. Additionally, they eliminate the necessity for any pre- or post-treatment steps, making the hydrocyclone the sole essential equipment for the treatment process. They are commonly employed as a pre-treatment step in conjunction with other technologies. Hydrocyclones have a long lifetime, low space requirements, small footprint, and do not need any additives [47,48]. However, they can experience significant pressure drops. The waste stream from the hydrocyclone (bottom output) consists of a concentrated slurry of solids that requires appropriate disposal measures [49]. This substantial production of concentrated solid slurry is a major drawback of this technique [48].

The efficiency of oil-water separation in hydrocyclones can be influenced by various factors, including processing capacity, the density difference between the two phases, geometry, temperature, pressure drop, and oil droplet size. Researchers have shown through their studies that separation efficiency ranges from 90.3 to 99.12% in the underflow, and the oil concentration in the overflow ranges from 77.8 to 98.82%, indicating a decrease in solid particle separation efficiency by 8.86% and an increase in oil droplet separation efficiency by 11.91% [50].

A recent study examined the effect of flow structures on the efficiency of hydrocyclones designed for oil-water separation, specifically focusing on single and dual inlets [51]. The study found that the use of a single inlet resulted in unsteady wavering flow, primarily caused by an imbalance in the flow immediately upon entering the cyclone. This unsteady flow adversely affected the separation efficiency, as it could transport water droplets located near the reverse flow core boundary into the overflow stream. Moreover, the presence of frequent recirculation zones led to the incomplete separation of fluid droplets. In contrast, the dual inlet hydrocyclone exhibited a uniform and steady fluid flow structure. This stable flow pattern facilitated the separation of oil and water into their respective core regions, with some inner cores rich in oil and some outer cores rich in water. The dual-inlet hydrocyclone demonstrated superior separation efficiency compared with the single-inlet hydrocyclone. At a 0.5 m^3/h flowrate, the dual inlet hydrocyclone achieved an efficiency of 82.3%, whereas the single inlet hydrocyclone achieved 73.7%. Figure 3 illustrates the oil superficial velocity vectors for both the single and dual inlet hydrocyclones. It focuses on the region starting from the inlet area and extending down to the tapering section, which is where the bulk of the separation process takes place [51]. The oil superficial velocity vectors provide a visual representation of the oil flow patterns within the hydrocyclones, offering insights into the fluid dynamics and separation efficiency of the two configurations. For higher flowrate (1.0 m^3/h), the dual inlet hydrocyclone demonstrated an excellent separation performance with an efficiency of 93.6%, while the single inlet hydrocyclone achieved a lower separation performance of 88.5% under the same feed conditions [51].

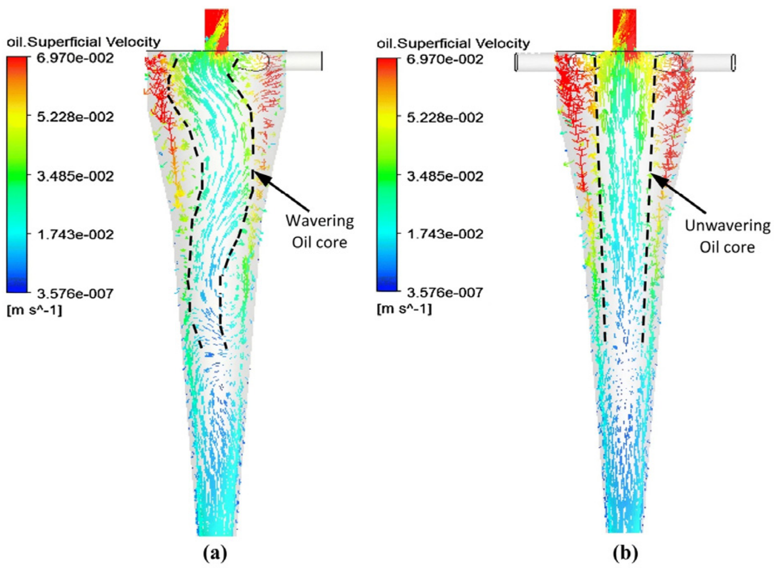

Figure 3. The oil superficial velocity vectors at a flowrate of 0.5 m^3/h flowrate were analyzed for a mixture consisting of 10% oil and 90% water in both the single (**a**) and dual (**b**) inlet hydrocyclone configurations [51]. Image reused under the Creative Commons attribution license.

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This technology offers significant advantages and plays a crucial role in downhole oil-water separation. It reduces the cost of lifting produced water and helps control the moisture content of water. It is also employed to extend the exploitation of aging oil wells [48]. In the downhole oil-water separation process, the traditional liquid-liquid separation hydrocyclone (LLSH) is utilized in conjunction with screw pumps, causing the hydrocyclone to rotate around its axis. This rotational motion creates vortex systems within the fluid, where the outer vortex moves in the direction of underflow, whereas the inner vortex (reversed) moves in the direction of overflow. In the LLHC, the less-dense oil is directed towards the center of the device, while the denser water is forced to move towards the wall. As a result, water, with a higher water-to-oil ratio, exits the LLHC through the underflow outlet, while oil, with a higher oil-to-water ratio, exits through the overflow [52]. It is worth mentioning that the utilization of compact hydrocyclones with smaller and lighter specifications offers significant advantages in offshore environments, where space is limited. In offshore gas and petroleum industry applications, the capacity of the equipment can only be adjusted by blocking specific cyclone tubes within the equipment [53].

2.1.3. Sand Filtration

The removal and reduction of turbidity from water through filtration is a cost-effective and efficient process. Filtration involves the physical separation of dispersed compounds using a porous medium [54]. Sand filters are commonly recommended for water treatment while retaining organic matter [55–57]. These filters collect contaminants throughout the sand bed and effectively retain organic matter, making them superior to other filters [58]. As a result, sand filters have been extensively used in the treatment of residual water [59,60]. Cha et al. achieved reasonable outcomes in oil removal from produced water by employing a combined process of ozonation and filtration [61].

To effectively remove metals from produced water, several pre-treatment steps can be employed prior to sand filtration. These steps include adjusting the pH to initiate oxidation reactions, using an aerator to enhance O_2 concentration, employing a solid separation unit to allow sufficient retention period for the settling of precipitated solids, and employing sand filtration to remove fine solid particles that cannot settle effectively. Numerous systems have demonstrated over 90% removal efficiency for iron using this process [19]. While sand filters are commonly employed for metal removal from produced water [62], they are not typically utilized as a filtering medium for oil removal [61].

2.1.4. Dissolved Air Precipitation (DAP)

A study introduced DAP method to generate bubbles for solvent sublation bubble columns [63]. The solvent sublation process, introduced by Sebba in 1996, is a non-foaming bubble separation technique used for removing organic components from water [64]. This process involves applying high pressure, up to 820 kPa, to saturate the air in a packed column saturator. Afterward, the valve releases the pressure into the water column, causing the air to condense and create bubbles with a diameter ranging from 60 to 100 µm. This initiates flotation, leading to the separation of aromatic hydrocarbons and aliphatic compounds. At the pilot scale, the removal efficiencies for dissolved ethylbenzene, micro-dispersed decane, and dissolved octane were 40%, 75%, and 95%, respectively [63].

The influence of pressure on bubble size was significant at low salt concentrations but became insignificant at high salt concentrations. The removal of alkanes was faster compared with the aromatic compounds. The DAP/SS (dissolved air precipitation/solvent sublation) system was efficient at removing the total oil and grease (TOG) contents from produced water, achieving up to 70% removal efficiency [65].

Table 2. The known technologies and their applicability for produced water treatment [48,49,66].

Technology	Desalting	De-Oiling	Softening (Mg and Ca Removal)	Removal of Suspending Particles	Iron Removal	Trace/Soluble Organics Removal
Reverse osmosis (RO)	√	√	√	√	√	√
Nanofiltration (NF)	√	√	√	√	√	√
Ultrafiltration (UF)		√		√		√
Electrodialysis (ED)	√		√		√	
Thermal desalination	√		√	√		
Chemical treatment processes		√				√
Biological treatment processes						√
Activated carbon (AC)		√	√		√	√
Ion exchange process (IOP)			√		√	
Precipitation				√	√	
Aeration and sedimentation				√		
Deep bed filter		√		√		
API separator		√		√		
Hydrocyclone		√		√		

2.1.5. Gravity Separator/Coalescing Filter

Produced water, when treated using conventional gravity separation methods such as API separators, is often unsuitable for injection disposal or release into the environment. Gravity separation may not be efficient for heavy oil due to its similar density to water, requiring long detention times for effective treatment [67]. Therefore, alternative treatment methods, such as coalescence/filtration, are necessary to effectively treat produced water contaminated with oil.

In coalescence/filtration, which is a flexible method for accelerating the merging of small oil droplets into larger ones, water containing dispersed oil droplets in a hyper-saline continuous phase can be treated [68]. The coalescence process involves three steps: the droplets striking and adhering together, the coalescence of captured drops on the medium, and the growth of larger droplets that separate and settle. The key component of the coalescence/filtration process is the coalescing medium. Shirazi et al. conducted a study using a pilot plant (refer to Figure 3), to investigate the coalescence/filtration of wastewater contaminated with oil [69]. They used an electrospun nanofibrous filter made of polystyrene as the coalescing filtration medium and examined the influence of thermal treatment on the properties and efficiency of the filters. The thermally treated filters exhibited better efficiency for the separation of oil droplets, demonstrating the effectiveness of this process for separating oil from produced water streams.

Coalescers offer several advantages over other settling methods, such as gravity settlement. One notable advantage is their compact size, making them suitable for installations where space is limited. Additionally, coalescers demonstrate high separation accuracy, allowing them to effectively separate emulsified oil droplets that are smaller than 10 μm. This is a significant improvement over hydrocyclones, which struggle to separate such fine droplets [70]. The compact structure of coalescers contributes to their efficiency and effectiveness. They are designed for convenient operation and have a long service life for coalescence materials. Unlike other separation methods, coalescers do not require the use of additional reagents, making them cost-effective and environmentally friendly. As a result of these advantages, mechanical coalescers have gained widespread use in liquid-liquid separation processes. They have found applications in various industries where efficient separation of immiscible liquids is required, such as oil and gas, chemical, and wastewater treatment sectors.

Chen et al. analyzed the coalescing separator mechanism based on the principles of gravity separation, shallow pool theory, and equal flow theory [71]. They innovatively designed horizontal and vertical separators with different coalescing components, which were linked to make a multistage multiphase separation system (MMSS). Fluent computational fluid dynamics software was employed to study the flow field within the vertical separator. In their study, laboratory experiments were conducted to examine the influence of the flowrate, coalescing components, and parallel vertical separator on the separation efficiency of MMSS. The results demonstrated a significant improvement in separation efficiency compared with a one-stage horizontal separator. Four types of coalescing components, namely spiral tracks, semicircular baffles, 4-hole plates, and 7-hole plates, were tested to assess their impact on the separation of oil−water emulsions (Figure 4). It has been observed that the spiral track component is particularly suitable for handling small flow separation, whereas the orifice plate component demonstrates excellent performance in scenarios involving large flow separation.

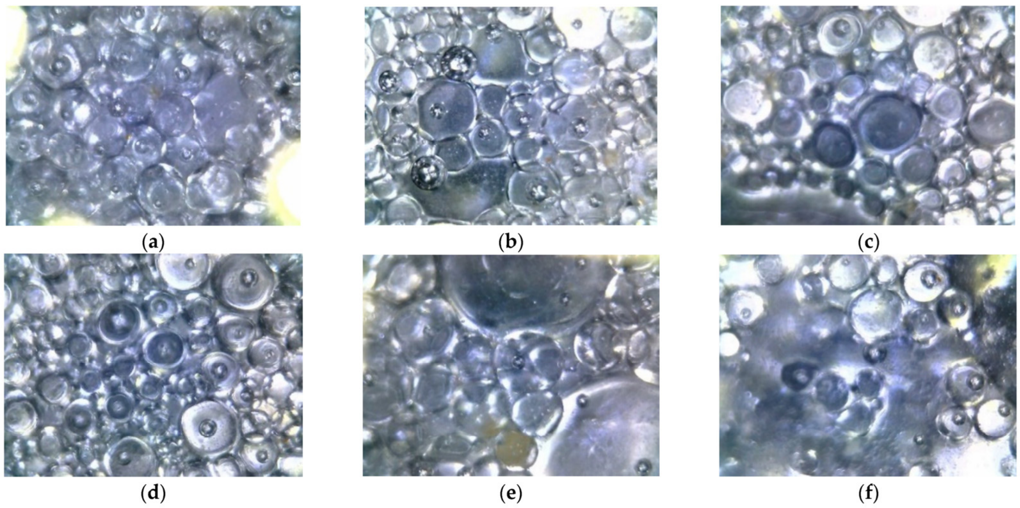

Figure 4. Morphological examination of the oil droplets at outlet of the vertical separator under various working conditions. The sub figures (**a**–**f**) signify a blank tube, semicircular baffle, spiral track, 4-hole plate, 7-hole plate, and all five risers are open, respectively [71]. Image reused under the Creative Commons attribution license.

2.2. Thermal Treatment Processes

2.2.1. Evaporation

The treatment of produced water using direct evaporation systems offers several advantages by eliminating the need for multiple chemical and physical treatment processes [25]. Various effective methods, such as vertical tubes, vapor compression evaporation, and falling film, have been employed for this purpose. The working principle of these techniques involves providing latent heat to the inlet water, causing it to evaporate and form vapor. The vapor is then condensed back into liquid water through cooling. This process allows for up to 98% recovery of produced water (high-quality distillate), with low levels of non-volatile inorganic TDS (<10 mg/L). The resulting distillate can be used as feedwater for Once Through Steam Generators (OTSG) or conventional boilers, improving their reliability and overall performance [72].

One of the significant advantages of direct evaporation systems is the elimination of physical separation and chemical treatment processes. This leads to reduced maintenance requirements, lower material and labor costs, and a decrease in de-oiling equipment for produced water. Additionally, the feed water quality for OTSG is improved, resulting in more efficient operation. However, direct evaporation systems have limitations related to the presence of an excessive concentration of solid salts in the produced water, which makes the reuse of concentrated salts unfeasible [73]. The energy requirements for these systems are also relatively high, leading to increased operating costs [74].

To optimize energy efficiency and minimize fouling, falling film vertical tube evaporators have been used. These evaporators have a high heat transfer coefficient and effectively remove oil contents from produced water. Prior to entering the evaporator, the produced water is preheated and deaerated to effectively remove non-condensed gases. The deaerated brine is then introduced into the evaporator, where it flows down the heat transfer tubes. As the process continues, a fraction of the brine evaporates, while the remaining liquid descends back into the sump for recirculation. Vapor condensation is achieved using a compressor [25].

Overall, direct evaporation systems offer a viable solution for produced water treatment, providing efficient separation and a high-quality distillate output. However, consideration should be given to the disposal of concentrated salts and the associated energy requirements.

Evaporation ponds, also known as solar evaporation ponds, are artificial ponds that are specifically designed to facilitate the efficient evaporation of water using solar energy [75]. These ponds serve various purposes, such as preventing the subsurface infiltration of water or controlling the downward movement of produced water. Evaporation ponds are often considered an economical option and have been widely utilized to treat produced water, both offsite and onsite [29]. They provide a natural and passive method for water treatment, relying on solar radiation and evaporation to remove water from the system while leaving behind concentrated contaminants.

In the operation of evaporation ponds, the produced water is directed into the pond, where it is allowed to disperse and cover a significant surface area. As the water is exposed to sunlight, and solar energy causes evaporation to occur. Over time, the water evaporates, leaving behind the concentrated contaminants, which can then be further managed or disposed of accordingly. Evaporation ponds offer several advantages, including simple process, low energy requirements, and cost effectiveness. They can be implemented in various locations and are particularly suitable for areas with abundant sunlight and available land.

However, it is important to consider the potential environmental impacts and ensure that the design and management of evaporation ponds comply with regulatory requirements to prevent any adverse effects on surrounding ecosystems or groundwater resources. Overall, evaporation ponds provide a practical and viable solution for the treatment of produced water, offering an efficient and environmentally friendly method for water management and disposal.

2.2.2. C-Tour

The C-Tour process is a method that employs solvent extraction principles to effectively remove residual hydrocarbons from produced water. The process involves injecting condensate into the produced water stream and allowing inline mixing to take place. By means of this mixing, the hydrocarbon impurities present in the produced water are extracted and combined with the injected condensate [76]. During the extraction process, the impurities and condensate coalesce, forming lighter and larger oil droplets. These oil droplets can be subsequently separated from the produced water stream using downstream treatment apparatus. The separation can be achieved through hydraulic or mechanical means, depending on the specific setup and system requirements. Once separated, the oil droplets are directed to the appropriate oil process streams for further treatment or utilization. This allows for the recovery and proper management of the hydrocarbons present in the produced water, reducing waste and potentially providing additional value.

The C-Tour process provides an efficient and effective solution for treating produced water while simultaneously recovering valuable hydrocarbons [76,77]. By employing solvent extraction principles and optimizing the mixing and separation steps, the C-Tour process facilitates the efficient extraction of hydrocarbon contaminants. This results in cleaner produced water and enhances water management in oil and gas operations as a whole.

The C-Tour technique has demonstrated its ability to effectively remove dispersed oil by 50 to 70% from produced water. Additionally, it has the capability to disperse dissolved organic material. Compared with other cleaning processes such as Epcon, C-Tour is more efficient at extracting PAHs (polycyclic aromatic hydrocarbons) and BTEX (benzene, toluene, ethylbenzene, and xylene), which are dissolved in water [78,79]. It has been found to increase the discharge of BTEX by 17% compared with other methods.

The performance of the C-Tour process can be influenced by the initial oil content present in the water and the percentage of NGL (natural gas liquids) injected [77]. A higher oil content has a greater effect on compounds within the C4–C5 range and phenols, while lower reference values also show an effect on PAHs (2–3 ring) and PAHs (4–6 ring). The impact of oil content increases with higher NGL injection rates. A pilot-scale study demonstrated promising results using real oil and real condensate in the C-Tour process. It achieved a remarkable 90% reduction in dispersed oil contents, indicating its effectiveness at removing oil from the produced water [76]. Furthermore, the C-Tour process has shown 90% removal efficiency for PHA compounds, highlighting the capability of C-Tour to target and extract specific contaminants from the water. These results from the pilot-scale plant validate the effectiveness of the C-Tour process at reducing dispersed oil and removing PHA compounds, indicating its potential for larger-scale implementation in oil and water treatment applications.

2.2.3. Freeze-Thaw Evaporation (FTE)

The FTE process is designed for managing produced water by utilizing natural temperature fluctuations. It involves freezing, thawing, and evaporation to generate a comparatively large volume of pure water suitable for various applications. In the FTE process, the produced water is initially preserved in a holding pond until the air temperature drops below 0 °C. Pumps are used to transfer the water from the pond, which is then sprayed onto a freezing pod. The freezing pod contains an elevated pipe grid with strategically placed sprinklers. The water undergoes freezing and thawing cycles within the pod. To remove the concentrated brine, the water is transferred to separate storage ponds [80,81].

During the winter season, the FTE process allows for the recovery of approximately 50% of the water, taking advantage of freezing and thawing mechanisms. However, in other seasons when freezing conditions are absent, the FTE operates similar to a traditional evaporation pond, and no water recovery occurs. The FTE process is highly efficient, achieving over 90% removal of total suspended solids, heavy metals, and hydrocarbons present in the produced water [81,82].

The FTE system has a lifespan of over 20 years and is designed for easy operation and monitoring [82]. It offers a chemical-free treatment approach, although its effectiveness relies on ample land space and specific climatic conditions. Proper waste disposal is an important aspect to consider when implementing FTE technology, as it produces a significant volume of concentrated brine and oil that require appropriate management strategies.

A study investigated the oil recovery from high-moisture oily sludge employing freeze−thaw and the solvent extraction process. Three solvents, namely CHX, MEK, and EA, were evaluated to determine the better solvent for oil recovery. The researchers found that by conducting a 30 min extraction at a solvent to sludge ratio of 4:1, approximately 40% of the oil could be recovered. Furthermore, more than 80% of the solvent used in the extraction process was successfully recycled. However, the recovered oil exhibited a relatively low total petroleum hydrocarbon (TPH) content, approximately 30% when using CHX and 40% when using MEK or EA. This lower TPH content was ascribed to the emulsified water present in the extracted oil. To address this issue, the researchers employed the freeze−thaw method, which improved the contents of TPH in the restored oil obtained from EA or MEK extraction. Specifically, the TPH content increased from

40 to 60% due to the dewatering effect achieved during the freeze−thaw process. Overall, the combination of freeze−thaw and solvent extraction techniques exhibited promising results in the recovery of oil from high-moisture oily sludge. This integrated approach has the potential to improve the TPH content in the recovered oil by effectively addressing the issue of emulsified water [83].

3. Advanced Thermal Separation Processes

Prior to the advent of membrane technology, thermal separation technologies were commonly utilized in large-scale desalination plants [48,84]. These technologies were employed in regions where the cost of energy for water treatment was comparatively affordable [84]. The primary thermal desalination technologies include vapor compression distillation (VCD), multistage flash (MSF) distillation, and multi-effect distillation (MED). MED-VCD plants are integrated thermal desalination facilities that aim to achieve a maximum performance [85]. Although membrane processes are generally favored over thermal approaches, recent advancements in thermal processes have made them more attractive for treating highly contaminated water, such as produced water [49].

3.1. Multi-Stage Flash (MSF) Distillation

MSF technology can indeed be employed for the desalination of various wastewaters, including produced water. Figure 5 illustrates the schematic of an MSF unit. The working principle of MSF involves reducing the pressure of the evaporation water rather than raising its temperature. The preheated feed water is introduced into a low-pressure chamber [86]. MSF technology is approximately 20% effective in terms of water recovery, and post-treatment is also required [86]. The system cost depends on many factors such as the site location, materials used for construction, type of feed water, size, and desalination capacity [87]. The energy consumption for the MSF process typically ranges from 3.35 to 4.70 kWh per barrel [88]. Moreover, MSF units have a lifespan of over 20 years [49].

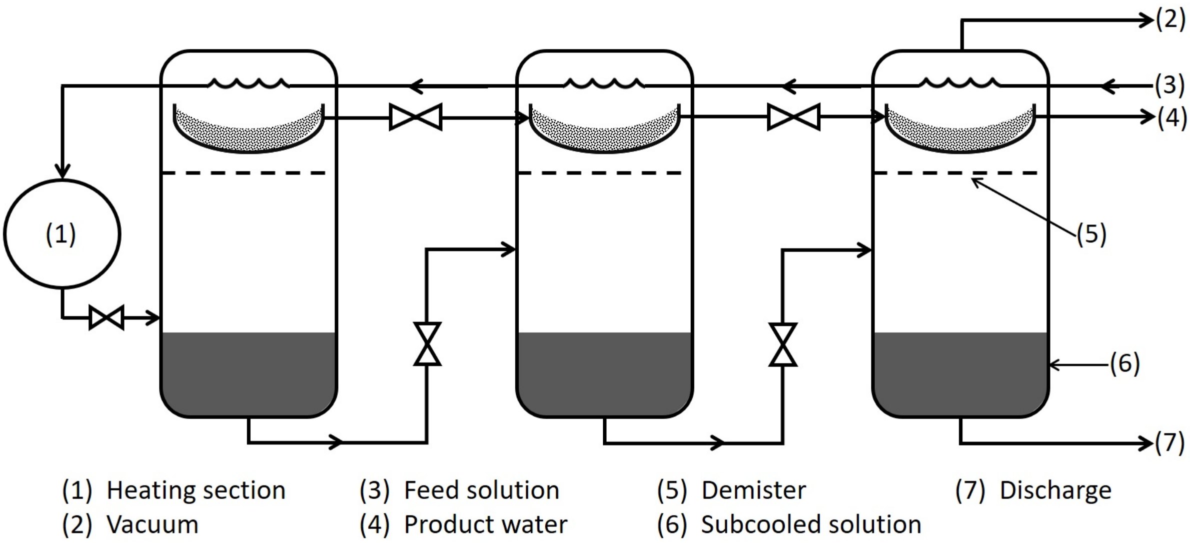

Figure 5. A schematic illustration of a three-stage MSF system to treat produced water.

A recent innovation introduced a system and method to process and recycle water utilized in an oil region steam operation [89]. A vaporizer-desalination unit was employed to separate a polluted water flow into two distinct streams: a flow designated for contaminated disposal and a separate flow consisting of clean water vapor. After the separation process, the polluted water flow is acquired by extracting it from the combined flow of oil and water originating from an oil well. Subsequently, the flow of clean water vapor is preferably routed via a steam generator to produce the steam required for the oil region steam operation. The generated steam is introduced into the oil region of a

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specified well, where it is utilized, and then retrieved as the combined flow of water and oil. Once adequately supplied by external water, the operational setup is structured to function continuously with minimal replenishment requirements, thanks to the effective water−vapor−steam cycle.

US11034605B2 introduced a new apparatus, system, and method for purifying produced water and removing valuable metals and minerals [90]. The apparatus comprises a device for flowing produced water from a wellbore to the produced water purification apparatus. It also includes at least one device for removing heavy metals from the produced water and at least one brine removal device for eliminating brine from the produced water. The method involves using the apparatus, and the system consists of a control panel that operates at least one device for removing heavy metals and at least one sensor in a coordinated manner. A series of connected sections can be created within the heat exchanger by engineering a combination of selected openings, baffles, perforated tubing, shunts, screens, and their combinations. This arrangement enables the operation of MSF systems. As the fluids pass through each section, gravity assists in the separation process, with the heavier liquids containing contaminants settling while the lighter vapor moves on to the subsequent section with reduced levels of contaminants. To enhance efficiency, the pressure can be lowered by incorporating a pump at the outlet located on the top of the heat exchanger. This adjustment facilitates the swift exit of purified vapor from the heat exchanger and lowers the boiling point of the fluid, further optimizing the overall performance [90].

3.2. Multi-Effect Distillation (MED)

MED technology is employed to improve efficiency and reduce energy consumption. This System involves the movement of feed water through a series of evaporators in which produced vapors evaporate water for the subsequent evaporator [91]. The produced water recovery rate can vary from 20 to 67% depending on the specific evaporator type employed. MED is particularly advantageous for treating produced water with a high TDS content [85]. These systems have a life cycle of 20 years and can be utilized for a wide range of influent qualities, such as MSF technology [88,91].

Li et al. evaluated the feasibility of MED process for treating high-salinity organic RO concentrates (ROCs) generated from wastewater treatment in the refining and chemical industries [92]. The experimental results revealed that approximately 6% of organic impurities volatilized (during processes of evaporation) and became part of the produced water, while around 8% entered the tail gas. Both the produced water and tail gas, which complied with relevant Chinese national standards, had the potential to be reused or directly released without requiring additional treatment. However, significant fouling issues occurred due to calcium sulfate when the water recovery reached approximately 30%, indicating the need to remove hardness from the ROCs prior to evaporation [92]. To further analyze the thermal and economic aspects of ROCs treatment, a forward flow MED model was developed using the Aspen Plus platform. A performance analysis was conducted, which involved studying the specific heat transfer area, fresh steam flow, and gained output ratio (GOR as functions of the heating steam temperature and effect number). The outcomes of study demonstrated that the efficiency of MED system was notably affected by the heating steam temperature and effect number. Enhancing the effect number in the MED system resulted in an improved thermodynamic performance. However, it should be noted that increasing the effect number also led to higher capital costs associated with the system [92].

Recent research demonstrated an impressive enhancement in produced water production through the utilization of MED modified with flash-box desalination. The study successfully generated 806 m^3/day of potable water, surpassing the output of conventional MED systems [93]. This achievement was made possible by reusing brines used as a secondary feed for the production of potable water.

3.3. Vapor Compression Distillation (VCD)

VCD is a unit equipped with a compressor that generates vapor extracted from the evaporator, and condenses it within a tube bundle. Various configurations of this unit have been developed to enhance heat exchange during the evaporation of saline water [86]. In recent years, mechanical vapor recompression (MVR) has been increasingly utilized for produced water treatment [94]. MVR technology offers several advantages over conventional VCD, including reduced system complexity and emissions from water streams. Moreover, the MVR process solely requires an electricity source to drive the system.

Figure 6 illustrates a flow diagram of the MVR process. To ensure optimal efficiency and prevent issues such as precipitation and corrosion on the plate heat exchangers, proper pre-treatment of the feed is necessary for the MVR system. The pre-treated produced water feed is subsequently pumped into the evaporator condenser, which is equipped with a vertical tube bundle. Within this configuration, the produced water undergoes evaporation, resulting in the production of steam.

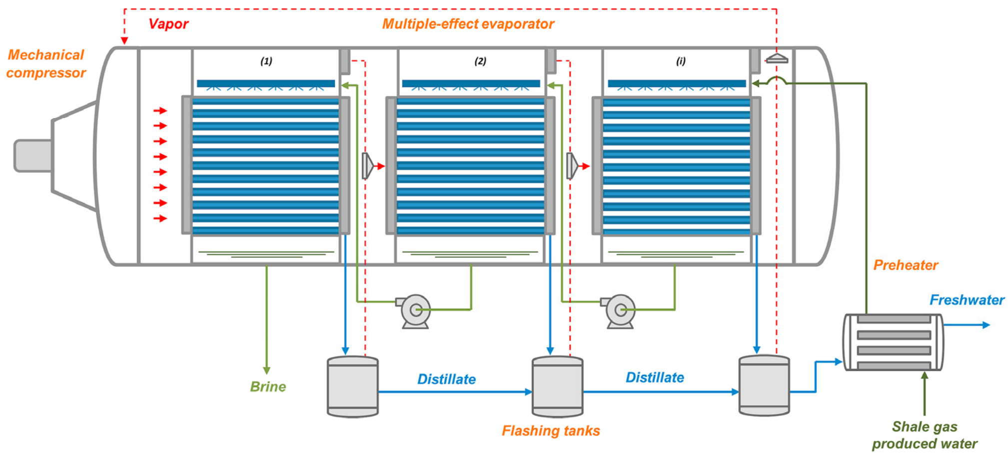

Figure 6. A schematic illustration of MVR system for the treatment of produced water [94]. Image reused under the Creative Commons attribution license.

3.4. MED–VCD Hybrid System

The hybrid MED−VCD process is a newly developed method specifically appropriate for produced water treatment [95]. It aims to enhance both energy efficiency and water recovery rates. By employing this process, significant improvements can be achieved by efficiently treating produced water from oil fields. This innovative technology is poised to replace the older MSF plants, as highlighted in the reported literature [95,96]. It offers distinct advantages over conventional produced water treatment methods. These benefits include reduced chemical dosage, lower overall costs, improved storage capabilities, mitigated fouling issues, easier handling, softer sludge generation, and more efficient management of waste streams [97]. Canada has already witnessed the installation of over 16 potable water evaporators, and the trend is expected to continue with the installation of more units in various regions across the globe [98].

ENTROPIE/SIDEM, a renowned company under Veolia (Aubervilliers, France), specializes in installing MED-VCD systems that guarantee high purity of treated potable water, with a claimed TDS level of <2 mg/L [95]. These systems allow for the direct utilization of the treated water in various industrial applications, with the option to use it either as is or after undergoing minor polishing if necessary. These applications cover a broad spectrum of industries, including the production of process water, boiler feed water, and water for closed-loop cooling systems.

The capital cost for the MED-VCD systems is estimated to be approximately $250 per barrel per day, as indicated in [48,87]. Conversely, the operational costs vary depending on the energy usage associated with the process.

4. Membrane-Based Treatment Systems for Produced Water

The membrane is a microscopic semipermeable material that separates substances of varying sizes by applying a driving force [99]. Membranes are typically composed of polymeric, ceramic, or metallic materials, which determine their properties, effectiveness, and performance in water purification processes [100]. Membranes can be categorized based on their pore size and particle rejection into four distinct types, including microfiltration (MF), UF, RO, and NF [101]. The pore size of MF membranes ranges from a few micrometers to 0.1 μm, UF membranes have pore sizes ranging from 0.1 to 0.01 μm, NF membranes have pore sizes ranging from 0.01 to 0.001 μm, and RO membranes have extremely small pore sizes ranging from 0.001 to 0.0001 μm [102]. Researchers have devoted significant efforts to improving the membrane performance through various approaches, such as material modifications and pore structure refinements [103,104]. Extensive work has already been completed, and ongoing research continues to explore new avenues for enhancing membrane efficiency [105].

Microfiltration and ultrafiltration are commonly used for removing oil from water. In the treatment of produced water, membrane technology can be employed for various purposes, ranging from minor treatment to the removal of suspended solids, oil, and desalination. Besides pressure-driven membrane systems (RO, NF, MF, and UF), the membrane bioreactor (MBR), which combines membrane filtration and biological processes, is another viable option for treating produced water. Membrane distillation, a membrane-based process, shows potential for the reuse of produced water [106]. Electrodialysis (ED) is another membrane-based desalination process [107]. The following membrane-based treatments are available for produced water, each with their own performance and efficiency characteristics.

4.1. Electrodialysis (ED)

The electrochemical separation process involves the ions being transported through an ion exchange membrane using a direct current voltage. This process is commonly employed for the desalination of wastewater. By applying a driving force, ionic substances from the source water move through the cathode and anode towards a concentrated waste water stream, resulting in the creation of a more dilute stream [108]. Dissolved solids in the produced water exist in the form of both anions and cations. These ions can be attracted towards electrodes that possess an opposing charge, as shown in Figure 7. Membranes are positioned between pairs of electrodes. Positively charged membranes selectively permit the passage of anions, while negatively charged membranes exclusively allow cations to pass through [29]. To facilitate the flow of feed water along the surface of each membrane, a spacer sheet is positioned between each pair of membranes [25].

According to Figure 7, positively charged ions such as sodium ions (Na^+) move towards the cathode, while negatively charged ions such as chloride ions (Cl^-) move towards the anode. Ions that have the same charge as the ion exchange membrane are rejected during this movement. Consequently, water undergoes concentration, resulting in desalted water being left behind in the neighboring compartment of the ED unit. In the subsequent section, both the desalted water and concentrate are continuously extracted from the unit. The principal unit of ED is a membrane stack consisting of several hundred cell pairs. These cell pairs are joined together with electrodes located on the outside. Prior to the ED process, pre-treatment of the water is essential to protect the membranes from potentially harmful substances, as the water passes through narrow passages. To transform alternating current (AC) into direct current (DC), a rectifier is employed.

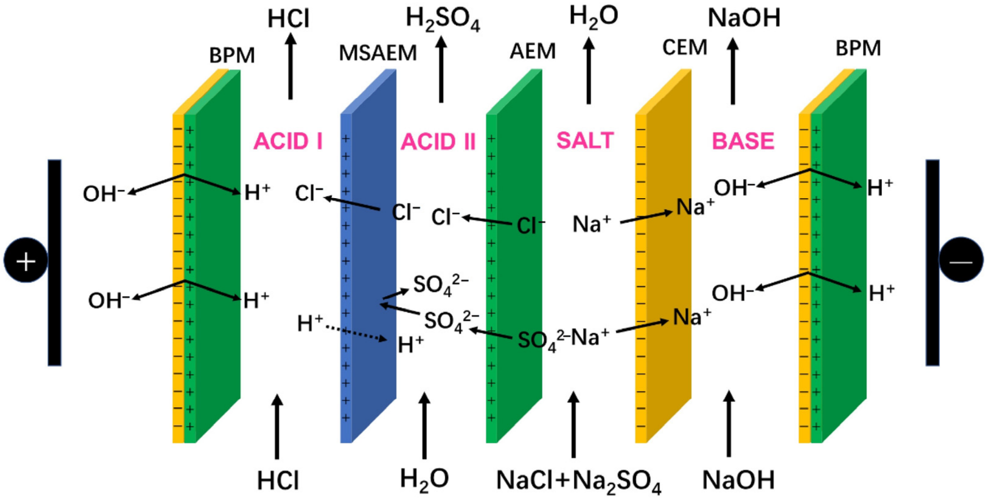

Figure 7. Schematic illustration of the innovative ED stack (SBMED) [109]. Image reused under the Creative Commons attribution license.

Post-treatment of the water involves stabilizing it and preparing it for distribution. This process may include the removal of gases such as hydrogen sulfide and adjusting the pH [25]. ED is a suitable method for treating low TDS concentrations, but it is typically not considered economical for treating concentrated produced waters due to various factors [110]. While ED is considered an outstanding technology for produced water treatment, particularly for relatively low saline-produced water, its application is currently limited to laboratory-scale experiments [111]. The life cycle of ED membranes is typically around four to five years. However, the main challenges associated with this technology include frequent membrane fouling and high treatment costs.

4.2. Membrane Bioreactors

The membrane bioreactor (MBR) is considered the best system for treating produced water due to its unique capabilities [24]. This process eliminates the need for a secondary clarifier, which is typically required in conventional activated sludge (CAS) treatment systems, as the membranes used in MBR serve as a means of solids separation [112,113]. Figure 8 visually compares MBR and CAS systems.

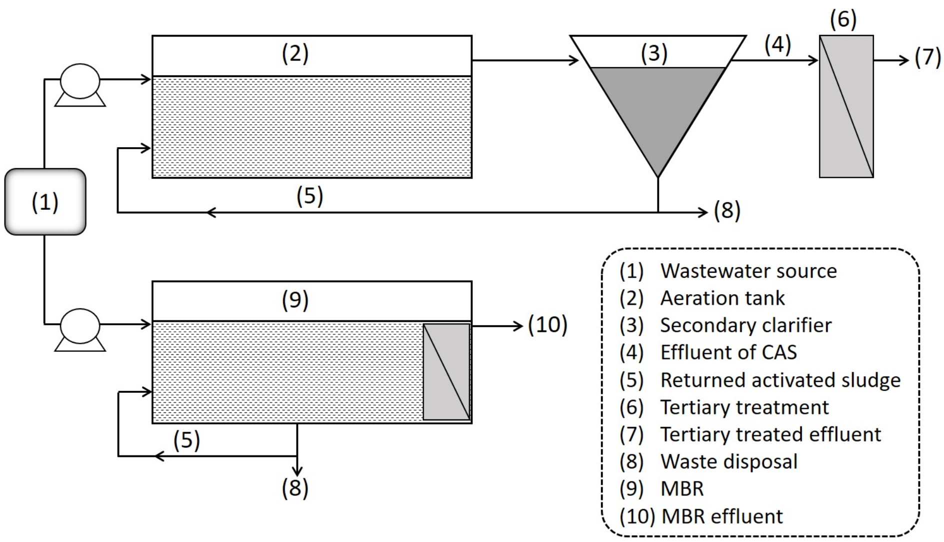

Figure 8. The schematic illustration of the conventional activated sludge (CAS) and MBR process.

MBR is a highly successful single system that integrates both physical and biological processes. In addition to integrating multiple processes, MBR technology offers several advantages, such as a simple control system, the ability to control the solid retention time (SRT), a reduced footprint, and ease of controlling the high retention time (HRT) [114]. The performance and purity of MBR systems depend on the pore size of the membranes. As the pore size decreases, the resistance to liquid flow increases, resulting in the need for higher operational pressures in the membrane system [102].

In a study conducted by Frank et al., a hybrid sequencing batch-reactor-MBR (SBR-MBR) process was employed for the treatment of both produced water and residential wastewater. The results demonstrated a remarkable reduction of over 90% in soluble chemical oxygen demand (COD) [115]. Two additional laboratory-scale studies conducted by Fakhru'l-Razi et al. [116] and Pendashteh et al. [117] showed impressive removal rates of 92% and 91% for TOC (total organic carbon) and TDS at concentration level of 16 g/L and 35 g/L, employing real and synthetic produced water, respectively.

In another study, a laboratory-scale MBR system was utilized to treat synthetic produced water with varying TDS levels. The results showed COD removal rates of 83% and 95% for synthetic produced water with TDS concentrations of 64.4 g/L and 144 g/L, respectively [118,119]. These studies highlight the effectiveness of MBR processes, both standalone and in hybrid systems, for the treatment of produced water, demonstrating significant reductions in various contaminants such as COD, TOC, and TDS.

A study was designed to evaluate the treatment of oil-field-produced water (OPW) using two submerged MBR setups, namely MBR-A and MBR-B [120]. Both systems operated using identical mixed liquor conditions, with pH 7 and 25 °C. The objective was to remove contaminants such as COD, oil and grease (O&G), and ammonia (NH_3) from the OPW. To maintain the desired dissolved oxygen (DO) content of approximately 3 mg/L, the air velocity in the systems was controlled using a rotameter set at 8 L/min. The results of the study indicate that both MBR-A and MBR-B achieved high removal efficiencies for COD, O&G, and NH_3, exceeding 90% under steady-state circumstances. Specifically, both MBR-A and MBR-B demonstrated an O&G removal efficiency of 96% [120].

While the MBR process has proven to be effective for treating high-strength wastewater [113], it is not without its challenges. Fouling and foaming are significant challenges associated with MBR systems [121]. The accumulation of solids and other substances on the surface of the membrane can lead to reduced filtration efficiency and hinder the overall performance of the MBR system [122]. This fouling phenomenon needs to be carefully managed to maintain optimal system operation and prevent any negative impacts on the quality of the effluent released into the environment [123].

The adverse impacts of fouling on both the efficiency of the MBR system and the effluent's quality, are significant concerns that should not be overlooked [124]. Membrane fouling can lead to decreased permeability, increased energy consumption, and reduced treatment efficiency [100]. Additionally, it can affect the removal of contaminants, including suspended solids, organic matter, and pathogens, potentially compromising the quality of the treated wastewater. It is important for operators and designers of MBR systems to implement strategies to mitigate fouling and foaming, such as proper membrane cleaning, optimized operating conditions, and the use of advanced monitoring and control techniques. Addressing these challenges is important to confirm the reliable and efficient operation of MBR systems and the production of high-quality effluent appropriate for environmental discharge or further reuse.

The cleaning of MBR systems is influenced by various factors, including operational conditions, the type and source of foulants, membrane material, and module configuration [100,125]. When it comes to cleaning methods, biochemical or biological cleaning is typically limited to in situ cleaning, meaning it takes place within the system itself. On the other hand, physical and chemical and cleaning approaches can be employed for both in-situ and ex situ cleaning, where the membranes are removed from the system for cleaning purposes. The selection criteria for the cleaning method highly depends on the specific circumstances and requirements of the MBR unit. Factors such as the composition and characteristics of the foulants, as well as the type of membrane used, play a crucial role in determining the most suitable cleaning approach. However, it is noteworthy that even with the application of physical, chemical, and biological cleaning methods, there may still be some instances where a portion of the fouling remains irrecoverable, especially in long-term operations. To optimize the cleaning effectiveness and minimize fouling-related issues in MBR systems, it is essential to implement a comprehensive cleaning strategy that combines appropriate cleaning methods, regular maintenance, and monitoring of system performance. This proactive approach can help maintain the long-term efficiency and reliability of the MBR system, while minimizing the impact of fouling on its operation.

5. Resource Recovery

Produced water is a complex mixture containing various valuable resources, including inorganic components, metals, crude oil, hydrocarbons, and water. The recovery of these resources, such as residual oil, hydrocarbons, and important inorganic components, can significantly offset the cost involved in produced water treatment and promote its reuse. Developing new techniques for resource recovery is essential in this regard [126].

One valuable metal present in produced water is lithium, which has significant applications in energy storage devices and electric vehicles to meet the growing demand [127,128]. The reclamation of produced water, particularly in enhancing oil production, involves a multistep process aimed at removing solids, oil, salts, and gases to ensure the water's quality for industrial, irrigation, livestock, and domestic use. The selection of suitable treatment technology for resource recovery depends on several factors such as treatment cost, the composition of produced water, environmental considerations, and water reuse standards. Figure 9 illustrates the treatment sequence for resource recovery and integrated treatment, which transforms produced water into a suitable resource for reuse by recovering oil and minerals, among other valuable components.

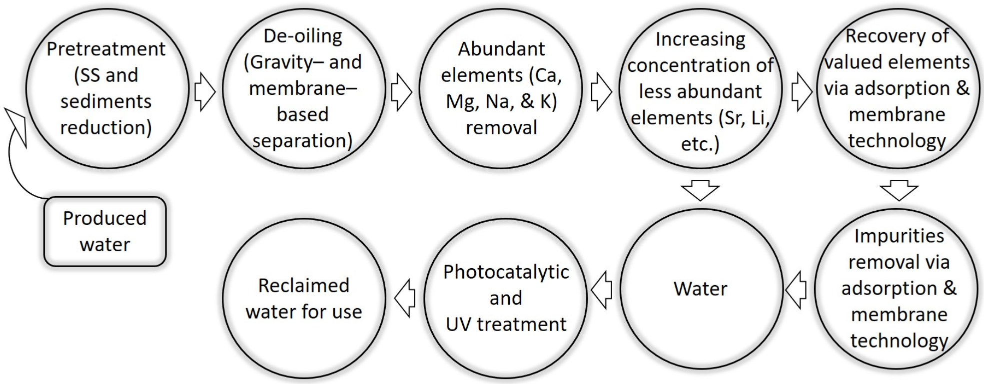

Figure 9. Flow diagram of the steps involved in resource recovery from produced water.

6. Future Perspectives

Produced water poses a significant challenge for countries involved in oil and gas production, considering that it constitutes the most substantial waste stream generated in the process. The production of each barrel of oil results in the generation of 3–10 barrels of produced water [19]. While approximately 95% of this water can be treated and reinjected to improve oil recovery, a substantial fraction remains. Unlike oil fields, water injection is not utilized in gas fields. As a result, oil and gas companies face the challenge of finding cost-effective and efficient treatment technologies to reduce contaminants in produced water for discharge or reuse purposes.

Because of the variations in the characteristics of produced water, such as differences between gas fields and oil fields, as well as variations among individual wells and based on good age, there is no universally recommended technique to meet entire environmental standards and recycling and reuse objectives [129]. The different methods discussed in this paper have their own advantages and disadvantages. The common shortcomings of physical methods include significant upfront investment and vulnerability to changes in water input. Chemical treatment, on the other hand, leads to the generation of hazardous sludge, which requires additional treatment and disposal efforts, along with major operational budgets and sensitivity to initial wastewater concentrations. Biological processes are sensitive to variations in organic substances and salt concentration in the incoming waste. These factors limit the feasibility of providing a single, overarching recommendation for the produced water treatment.

While there have been numerous studies examining the prospective reuse of treated produced water, government regulations pertaining to produced water reuse are currently limited [130]. Existing water regulations and guidelines primarily focus on water applications such as drinking water standards and irrigation guidelines set by organizations such as the Environmental Protection Agency (EPA), the United States, and the European Union water quality directive for groundwater (80/68/EEC) [129]. However, according to a recent research [131], more than 900 chemicals found in produced water do not have approved analytical procedures for the quantification or detection or in the regulatory framework established by the EPA. This implies that existing regulations may be insufficient to monitor the quality of treated produced water and ensure its safe reuse outside the oil and gas industry. To address this issue, it is advisable for regulatory bodies or organizations to establish comprehensive monitoring and assessment programs. These programs should thoroughly evaluate the potential effects and hazards associated with the reuse of reclaimed produced water, considering its impact on surface water, groundwater, ecological systems, and public health. It is important to conduct these evaluations prior to permitting the reuse of produced water outside the oil and gas industry. This would involve evaluating the effectiveness of treatment methods and implementing stringent monitoring protocols to ensure the safety of produced water reuse in other applications.

7. Conclusions

In conclusion, the produced water treatment is a critical challenge faced by the oil and gas industry. Various treatment technologies, including conventional approaches, thermal treatment processes, membrane-based systems, and resource recovery methods, have been explored. Each technology has its own advantages and limitations, making it crucial to select the most suitable approach based on the characteristics of the produced water and the desired treatment goals.

Among the technologies discussed, membrane-based systems, particularly electrodialysis and membrane bioreactors, demonstrate significant potential for efficient produced water treatment. These technologies offer high contaminant removal efficiencies, compact design, and the possibility of resource recovery, making them promising options for future implementation. However, further research is required to optimize the performance, cost effectiveness, and durability of membrane-based systems. Overcoming challenges related to fouling, membrane fouling, and operational costs will be critical to ensure the successful application of these technologies in large-scale produced water treatment.

Looking ahead, future efforts should focus on the development of hybrid treatment systems that combine multiple technologies, the integration of advanced treatment processes, and an increased emphasis on sustainability and resource management. By advancing treatment technologies and promoting responsible water management practices, the oil and gas industry can mitigate the environmental impact of produced water and contribute to a more sustainable energy sector.

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Managing Water in the West

Final Report No. ST-1601-2016-01

Summary of Current Research on Produced Water Treatment

Research and Development Office
Science and Technology Program

U.S. Department of the Interior
Bureau of Reclamation
Research and Development Office
Denver, Colorado

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Mission Statements

The U.S. Department of the Interior protects America's natural resources and heritage, honors our cultures and tribal communities, and supplies the energy to power our future.

The mission of the Bureau of Reclamation is to manage, develop, and protect water and related resources in an environmentally and economically sound manner in the interest of the American public.

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Research and Development Office
Advanced Water Treatment

by

Katie Guerra

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Acronyms and Abbreviations

bbl	barrel
CDT	capacitive deionization
DOE	Department of Energy
DWPR	Desalination and Water Purification Research and Development
EPA	United States Environmental Protection Agency
FT	freeze-thaw
FTE®	freeze-thaw process
gal/min	gallons per minute
GIS	Geographic Information System
IPSC	Integrated Precipitative Supercritical
Interior	Department of Interior
kWh	kilowatt-hour
LLC	limited liability company
MCL	maximum contaminant level
MF	microfiltration
mg/L	milligram/liter
NETL	National Energy Technology Laboratory
NF	nanofiltration
NORM	naturally occurring radioactive materials
NPDES	National Pollutant Discharge Elimination System
NSF	National Science Foundation
PSF	polysulfone
Reclamation	Bureau of Reclamation

RO	reverse osmosis
RPSEA	Research Partnership to Secure Energy for America
S&T	Science and Technology
SAR	sodium adsorption ratio
TDS	total dissolved solids
UF	ultrafiltration
ULPRO	ultra-low pressure reverse osmosis
UOG	Unconventional Oil and Gas
US	United States
USGS	United States Geological Survey
WWTP	Waste Water Treatment Plant

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Executive Summary

Water demand and water production in the oil and gas industry is significant and has the potential to impact water supplies (both positively and negatively) in the western United States (US). In recent years, the demand for fresh water for hydraulic fracturing has increased dramatically in areas where fresh water supplies are constrained. The water that is generated during oil and gas extraction, termed produced water, typically has elevated levels of salts, metals, and organic constituents and is seen as a waste by-product in the oil and gas industry.

When petroleum prices are low, as they currently are, the need for better water management practices to reduce cost is more important than ever. Identifying alternative water supplies for hydraulic fracturing and recycling the flowback water are critical to ensuring water supply sustainability and satisfying competing demands for fresh water. Furthermore, with suitable treatment, fracturing flowback water and produced water can be used to augment conventional water supplies for irrigation, livestock watering, and stream flow augmentation.

Many of the water treatment-related challenges currently addressed in the Bureau of Reclamation's (Reclamation) Science and Technology (S&T) Research Program and Desalination and Water Purification Research and Development (DWPR) Program are common to the oil and gas industry. Cost, energy consumption, concentrate management, chemical use, and operational complexity are recognized challenges in both the water treatment industry and the oil and gas industry. Advances in either of these industries have the potential to positively impact the other. For example, the oil and gas industry has more tolerance for risk and economic drivers that allow the use of newer, more cutting edge technologies. These same technologies may not be cost effective for municipal water treatment. Therefore, maintaining an expertise and awareness of water treatment issues in the oil and gas industry can help Reclamation identify new technologies and solutions to technical challenges that may benefit the municipal water treatment industry.

Two areas are identified, and discussed in detail in this report, for Reclamation involvement in water management related to oil and gas:

- Treatment of produced and flowback water for beneficial use, such as irrigation and livestock water

- Treatment and use of non-traditional water as an alternative to fresh water for hydraulic fracturing

This report summarizes (1) the potential impact of water produced and consumed in the oil and gas industry on Reclamation project areas, (2) past research by Reclamation and others in oil and gas water management, and (3) identifies current research thrust areas and potential for Reclamation involvement in future efforts.

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1. Introduction

Water is a critical part of oil and gas resource development and extraction; water is both consumed and produced at different stages of hydrocarbon production. Water is required for well completions, hydraulic fracturing, and other well stimulation techniques to increase oil and gas production. A water based solution (termed flowback water) flows back to the surface during and after the completion of hydraulic fracturing. Flowback water commonly contains clays, chemical additives, dissolved metal ions and total dissolved solids (TDS). The water usually has a murky appearance from high levels of suspended particles. Most of the flowback occurs in the first seven to ten days while the rest can occur over a three to four week time period (Schramm 2011).

Produced water is the water that exists naturally in oil and gas formations and is brought to the surface during petroleum extraction. Approximately 2 billion gallons of produced water are generated each day (Veil et al. 2004). Produced water is naturally occurring water found in shale formations that flows to the surface throughout the entire lifespan of the production well. This water has high levels of TDS and leaches out minerals from the shale including barium, calcium, iron and magnesium. It also contains dissolved hydrocarbons such as methane, ethane and propane along with naturally occurring radioactive materials (NORM) such as radium isotopes (Schramm 2011).

Many of steps in the well completion process that require water also require the transportation of water to and from the well field site, which has a significant energy requirement and environmental impact. Figure 1 shows the role of water in different phases of well completion and development.

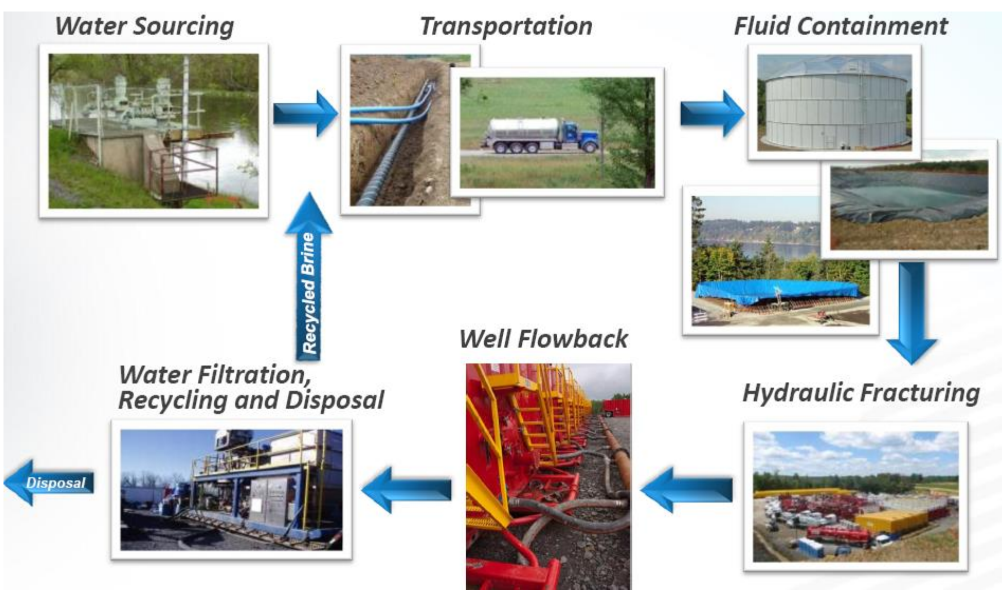

Figure 1.—Role of water in the lifecycle of well completion and development (Anon 2012).

For many years, managing, treating, and disposing of produced water in an environmentally-safe manner was the primary technical challenge for water managers in the oil and gas industry.

In the last decade, water management related to sourcing new supplies for hydraulic fracturing has become another significant challenge in the oil and gas industry. Large amounts of water are required for hydraulic fracturing, approximately 1 to 5 million gallons per well per fracturing event. Multiple wells exist on a development pad and each well can be fractured anywhere from 1 to 10 times. In many areas of oil and gas development, especially in the western United States (US), fresh water resources are already fully allocated, and sourcing fresh water for hydraulic fracturing is difficult. Treating hydraulic fracturing flowback or produced water for reuse on future fracturing jobs and for discharge back to the environment has become an important practice for water supply sustainability in the oil and gas industry.

Water treatment has the potential to both offset the water demand from the oil and gas industry and to make beneficial reuse of the water produced during oil and gas extraction. Identifying alternative sources of water for hydraulic fracturing, other than fresh water, may require treatment to make the water suitable for use. Furthermore, on-site water recycling can reduce the fresh water withdrawals needed for well completions and hydraulic fracturing. Finally, treatment of produced water can augment conventional water supplies to expand water supplies in the western US. Figure 2 shows the role water treatment can play in the oil and gas development process to both reduce water consumption within the industry and generate a useable water supply from produced water.

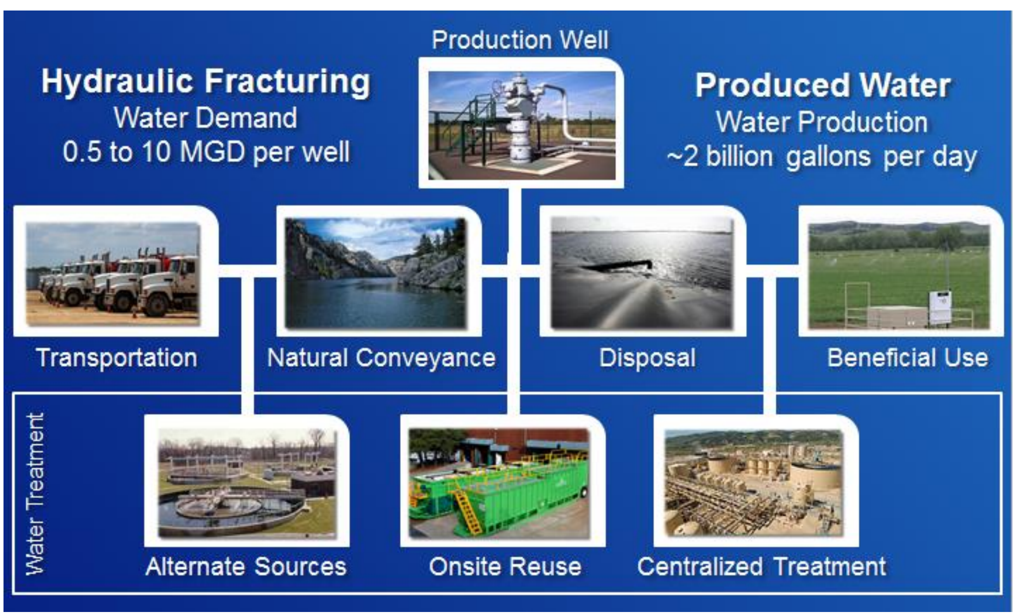

Figure 2.—Potential for water treatment to improve water use efficiency during oil and gas production (Dahm & Guerra 2013b).

Water management throughout the lifecycle of oil and gas production wells is important for water supply sustainability. Because of the large volumes of water produced and consumed by the oil and gas industry in arid regions of the western US, the Bureau of Reclamation (Reclamation) has maintained expertise and knowledge in water-related issues in the oil and gas industry. This report details past work conducted by Reclamation in produced water, current challenges in the industry, and provides recommendation for future Reclamation work in this area.

2. Oil and Gas Industry Water Challenges in Reclamation's Service Area

Approximately 80 percent of the oil and gas generated on-shore in the US occurs in the 17 western States (Veil et al. 2004). As oil and gas fields age, they tend to produce more water relative to oil and gas, therefore the amount of water produced will continue to increase as production wells age. Therein lies an opportunity for Reclamation to expand water supplies, as produced water represents an unused and non-traditional water source in the western US.

In addition to producing large amounts of brackish water, hydraulic fracturing and steam assisted gravity drainage require large amounts of water. This means that as oil and gas producers drill new wells and stimulate old wells, the demand for water in oil and gas producing areas will also increase. Many of these areas already have fully allocated fresh water resources, which means that the potential for conflict over water availability will increase. For example, according to a recent report, in Colorado and California, 97 percent and 96 percent of wells, respectively, were in regions with high or extremely high water stress. In New Mexico (NM), Utah, and Wyoming (WY) the majority of wells were in high or extremely high water stress regions. Texas, which has the highest concentration of hydraulic fracturing activity, had 52 percent of its wells in area with extremely high water stress (defined as over 80 percent of available surface and groundwater already allocated) (Freyman 2014).

The purple shaded areas in the map below (Figure 3) delineate oil and gas producing basins. The red, orange, and yellow shaded areas were identified in the Water2025 study as having a potential for conflict over water. Because of the geographical overlap between oil and gas production and areas that have limited fresh water supplies, there is a market driver for treatment and beneficial use of produced water to augment conventional supplies.

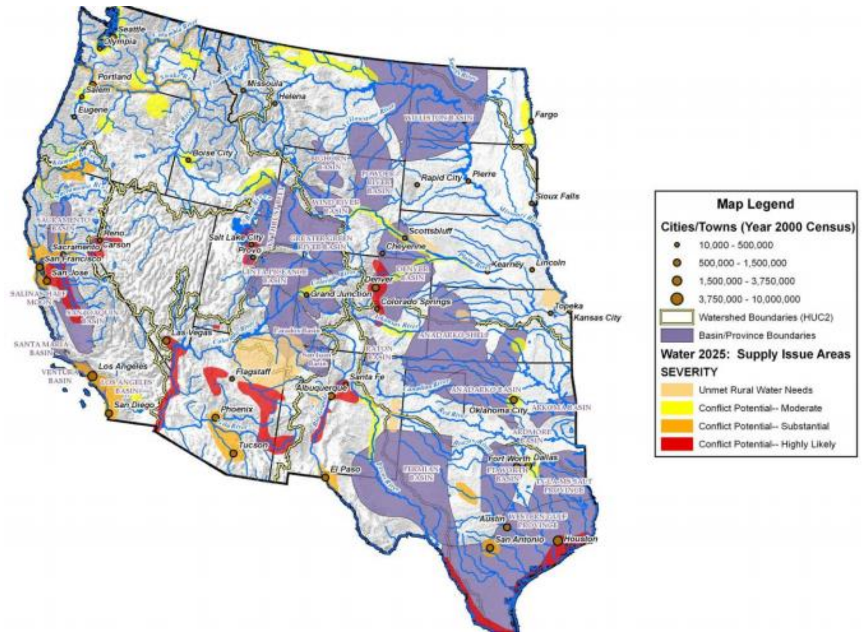

Figure 3.—Map showing oil and gas producing basins with Water2025 areas of potential water conflict (Guerra et al. 2011).

Additional drivers for collaboration between the petroleum industry and the water industry with respect to implementation of water treatment processes are:

- Reduce the cost of water treatment
- Minimize energy consumption
- Reduce chemical consumption
- Remove trace metals
- Minimize concentrate and process residuals
- Decrease system maintenance
- Reduce operational challenges associated with variable water quality and quantity
- Develop flexible, mobile, modular systems

Even though the price of oil is currently low and oil and gas production is down, there is still a need for efficient water management practices, including identifying alternative sources for hydraulic fracturing and treatment and beneficial use of produced water. Efficient water management practices enable companies to realize competitive advantages that can allow them to cost effectively produce wells that would otherwise be shut down due to water needs or excess water production.

Treatment of produced water for a valuable beneficial use can also offset water handling costs that would otherwise have been necessary. In many areas of the western US, water supplies are limited and the cost of new water rights is more expensive that treatment costs. Therefore, identifying alternative uses of produced water such as irrigation, livestock water, maintaining instream flows to allow for upstream water withdrawals can have very favorable financial implications. Therefore, even in what is considered a "low dollar environment" there is still a need for efficient water management and treatment practices in the oil and gas industry.

3. Reclamation Research

Reclamation has conducted a number of studies in produced water and water management for oil and gas. Reclamation's work in this area has had a significant impact on the industry because Reclamation has a perspective much different from the other government agencies and private sector companies working in this field. Using a mindset of increasing water supplies and balancing competing needs for water, Reclamation has provided a unique perspective on water management to the oil and gas industry. This section describes the research activities conducted by Reclamation in this area. Research is organized by funding source.

3.1 Science and Technology (S&T) Funded Research

3.1.1 Fouling Resistant Membrane for Produced Water Desalination (2006)

This project was done in partnership with Dr. Benny Freeman's research group at the University of Texas, Austin who received funding through the DWPR Program to develop novel, low fouling membranes for the treatment of produced water. Dr. Freeman's research group was investigating surface modifications for ultrafiltration membranes and were testing and evaluating the integrity of the coated membrane surfaces using synthetic water sources with vegetable oil in the laboratory.

Reclamation identified the need to conduct additional testing using a testing solution that would be representative of real produced water, as a next step in the evaluation of these membranes. In the effort to identify an example of produced water quality for further testing, Reclamation conducted a significant bench top study on the geographical occurrence and composition of produced water. While not the original intent of this study, the Research and Development Office agreed that a more detailed study on produced water generation and quality was needed.

Reclamation conducted an assessment of the United States Geological Survey (USGS) database on produced water. This database has since been updated to include newer well information. The current version of the database can be found on the USGS website (Blondes et al. 2014). The original database was used to obtain a general understanding of produced water variability. The TDS for each well in the database was used to generate Figure 4 (Benko & Drewes 2008). The vast majority of wells present in the database have a TDS that is significantly higher than the secondary maximum contaminant level (MCL) for TDS for drinking water. Many of the samples showed a TDS higher than seawater. Therefore, in order for produced water to be put to beneficial, desalination is necessary.

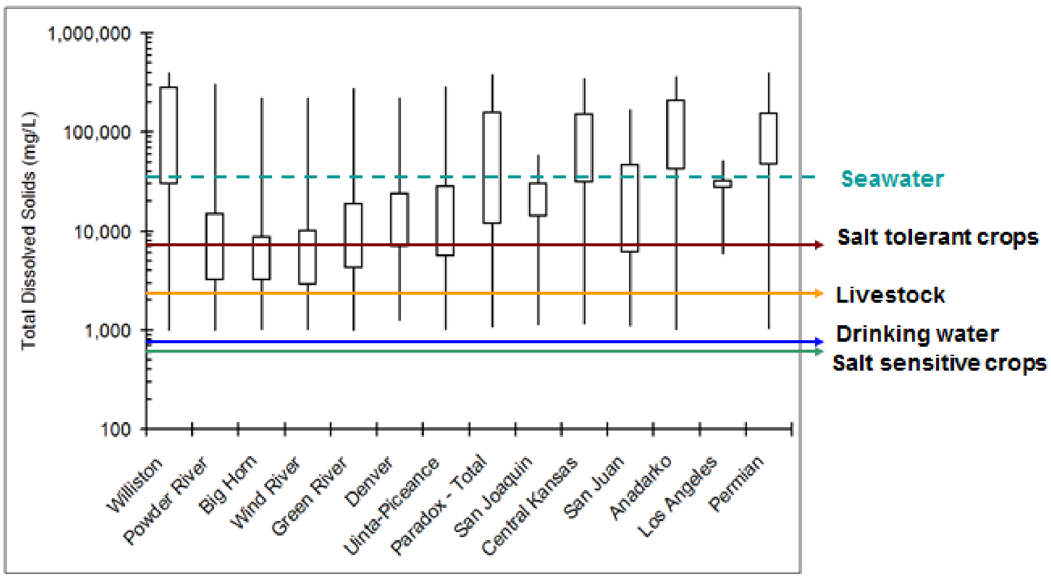

Figure 4.—TDS distribution of wells in the produced water database (Benko & Drewes 2008).

Data were available it the USGS database for the TDS concentrations and the following ions: calcium, magnesium, sulfate, and chloride. Sodium and chloride make up the majority of the ions in the produced water samples included in this database, as shown in Figure 5. The high sodium and chloride concentrations indicate that reverse osmosis would be a favorable technology for treating produced water. This work is provided in more detail in Benko and Drewes (2008).

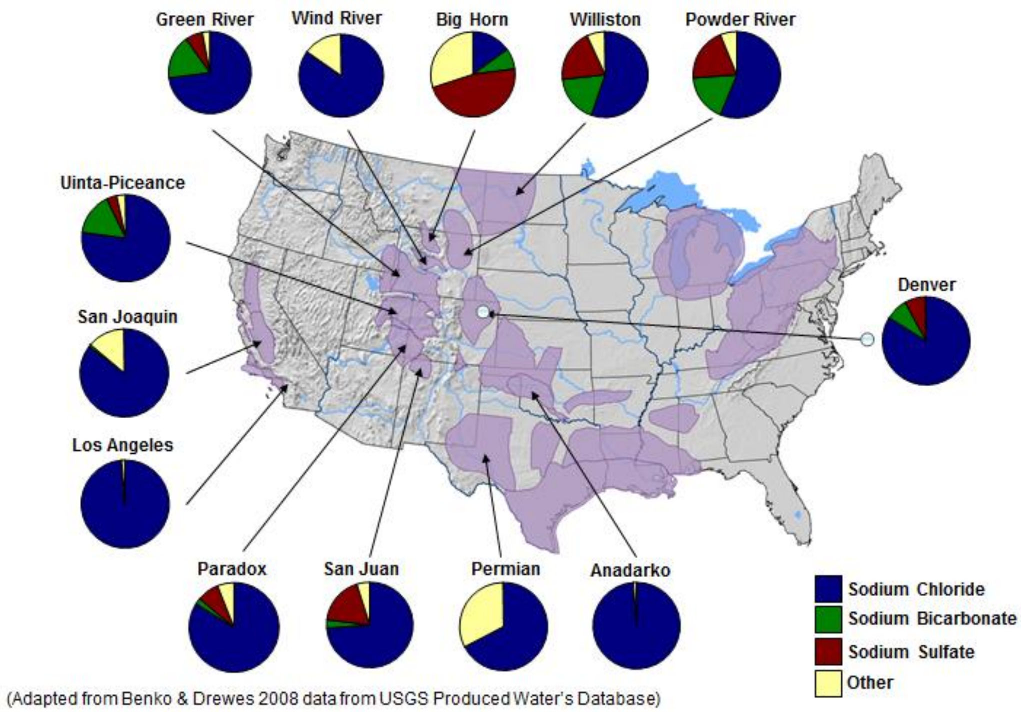

Figure 5.—Variability of produced water salt composition by basin.

3.1.2 Produced Water Workshop (2006)

The Produced Water Workshop was held in Fort Collins, Colorado, on April 4-5, 2006, to explore the potential opportunity for beneficial use of produced waters and the obstacles to making this a reality (Waskom 2006). The overriding goal of the workshop was to enhance our understanding of opportunities and challenges involved in converting produced waters to beneficial use. The workshop was attended by nearly 200 participants from government, energy companies, water users, water supply planners, government agency staff, researchers, industry representatives, and other interested parties.

The workshop did not focus on water treatment technologies because it was agreed upon that current water purification technology is generally adequate to treat produced waters where it is economically feasible. There is a portfolio of technologies available to apply depending on site-specific factors. However, final disposal of the concentrated waste from these processes is still an issue that requires further research and development.

The following conclusions were reached regarding the feasibility of treatment and beneficial use of produced water:

- The most promising opportunities to convert produced waters to beneficial use occur where produced water sources geographically align with markets for water.

- Water markets and the costs of disposal versus treatment will drive the value of produced waters and will be the fundamental factor in determining if produced waters are converted to beneficial use.

- The end users of the produced waters need to be willing to significantly offset the cost of treatment, storage, delivery, and management.

The following observations were made regarding the Federal and State role in produced water:

- States play the key role in water management and administration and must be in the lead on changing laws and policies to facilitate beneficial uses of produced waters.

- Federal agencies should provide leadership in helping to solve these problems as much of the production occurs on federal lands.

The business cultures in the energy and water industries are very different, and was a major source of discussion during the workshop. The following are key points from these discussions:

- Oil and gas producers react quickly to swings in the energy market while water suppliers are engaged in a more steady market without large swings in price, unless there is an extenuating circumstance such as drought.

- Energy companies work quickly in accessing their non-renewable supplies while raw water suppliers (generally government organizations) work over long time scales in planning new water projects.

- Energy companies often work with high risk, while water utilities/districts try to reduce risk to the lowest possible levels.

- The general approach to water management is in conflict with the longer time frame to plan and implement water projects. Planning should occur in advance of energy production on a watershed scale.

During the workshop the following research needs were identified:

- Social sciences to help remove institutional and social barriers to beneficial use of produced water.

- Understanding and managing the long-term adverse impacts to lands, ground waters, and ecosystems from produced water discharges and beneficial use.

- Pilot and demonstration projects to provide proof of concept from treatment to beneficial use of produced water in key basins.

One avenue for pursuing these research needs is for the Department of Energy's (DOE) National Energy Technology Laboratory (NETL) and Reclamation to explore joint projects. A formal interagency state and federal cross-cutting work group is needed to enhance communication among agencies and provide a point of contact for the industry. There were also suggestions of expanding the workgroup composition to include stakeholders in the oil industry and private sector.

3.1.3 Beneficial Use of Produced Water (2007 to 2010)

From 2007 to 2010, Reclamation funded work through the S&T Program to investigate the potential for treatment and beneficial use of produced water. The work was summarized in the Reclamation Desalination Series as report #157 (Guerra et al. 2011).

Reclamation gathered data from publically available sources to describe the water quality characteristics of produced water, performed an assessment of water quantity and quality in terms of geographic location (see Figure 6) and water quality criteria of potential beneficial uses, identified appropriate treatment technologies for produced water, and described practical beneficial uses of produced water.

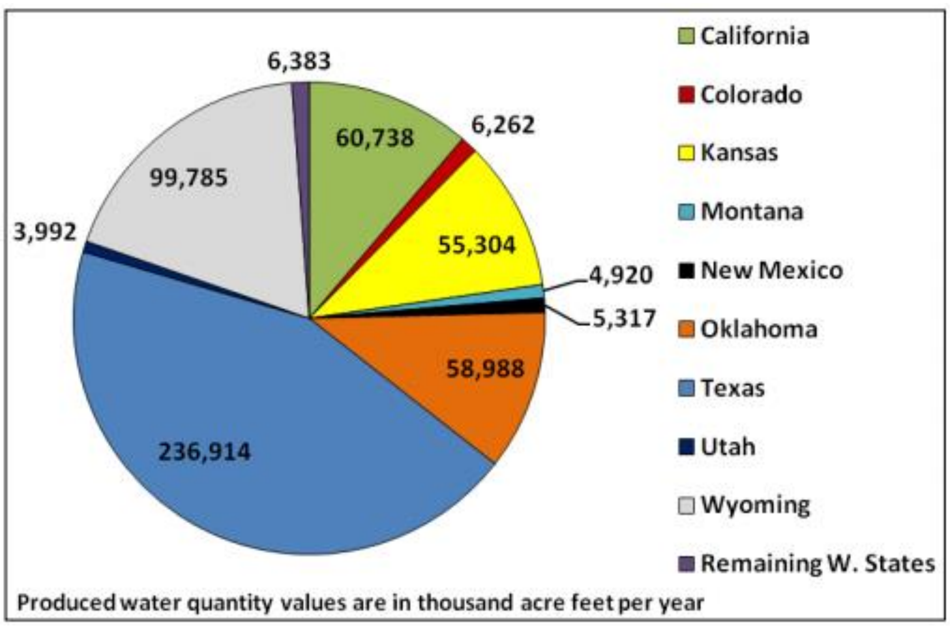

Figure 6.—Estimated volumes of produced water generated in the western U.S.

Produced water quality varies significantly based on geographical location, type of hydrocarbon produced, and the geochemistry of the producing formation. In general, the TDS concentration can range from 100 mg/L to over 400,000 mg/L. Silt and particulates, sodium, bicarbonate, and chloride are the most commonly occurring constituents in produced water. Benzene, toluene, ethylbenzene, and xylene compounds are the most commonly occurring organic contaminants in produced water.

The types of contaminants found in produced water and their concentrations have a large impact on the most appropriate type of beneficial use and the degree and cost of treatment required. Many different types of technologies can be used to treat produced water; however, the types of constituents removed by each technology and the degree of removal must be considered to identify potential treatment technologies for a given application. For some types of produced water, more than one type of treatment technology may be capable of meeting the contaminant removal target, and a set of selection criteria must be applied to narrow down multiple treatment options.

Beneficial uses of produced water include crop irrigation, livestock watering, stream flow augmentation, and municipal and industrial uses. Produced water also can be placed in aquifer storage for future use. The type of beneficial use most appropriate for a produced water application depends on the geographical location of the produced water generation, the location of the beneficial use, and the constituent concentrations in the produced water. A series of maps were produced to show the location of produced water generation in relation to areas with needs for additional water supplies. Figure 7 shows the geographic overlap between oil and gas production and areas of irrigated crops, as an example.

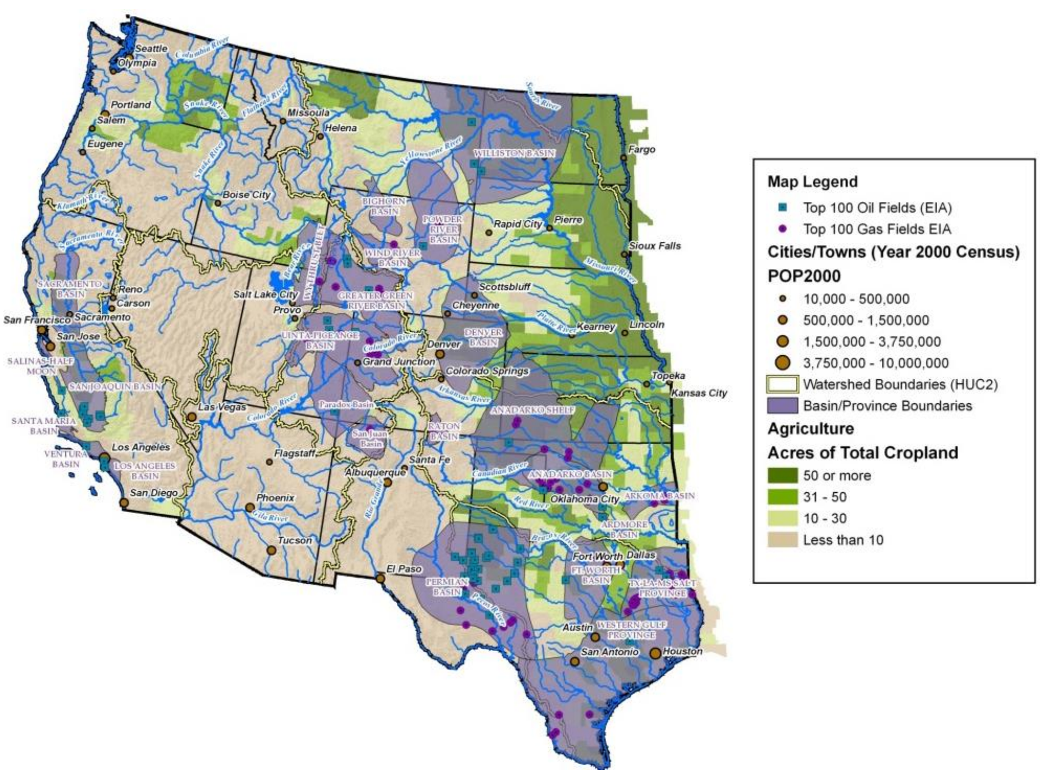

Figure 7.—Oil and gas production and acres of irrigated agriculture (Guerra et al. 2011).

Three case studies were also presented, which illustrated the large potential for beneficial use in the western US for different types of applications: agriculture, stream flow augmentation, and industrial use. Appropriate management techniques will allow produced water to be used as a resource rather than treated as a waste to meet the growing water demand in the western US.

3.1.4 Treatment and Beneficial Use of Produced Water (2011, 2012)

This project furthered the work conducted on during the 2007-2010 project. More research was conducted in collaboration with Colorado School of Mines on water quality characterization including the compilation of coalbed methane water quality data (Dahm et al. 2011). This study also proposed a method for identifying groundwater contamination from gas production using of fluorescence measurements and statistical analysis (Dahm et al. 2013). A conference presentation was given at the International Petroleum Environmental Conference summarizing the past Reclamation produced water research (Dahm & Guerra 2012).

3.1.5 Produced Water Treatment Primer for Oil and Gas Operations (2013, 2014)

This project produced a catalog of advanced water treatment technologies currently used in the oil and gas industry (Dahm & Chapman 2014). Each technology was identified as having positive and negative aspects with respect to chemical requirements, energy requirements, footprint, cost, and removal capability.

General information was included on a number of categories of applied technologies, including: a brief technology description, applicable contaminants removed, removal mechanisms, and qualitative notes on advantages and disadvantages. Links to case study examples are provided to industry information on technology applications, operational experience, and performance data.

3.2 Manuals and Standards

In 2014, Manuals and Standards funding was used to develop a guidance document for considering water use and production in the oil and gas industry for Reclamation water managers (Dahm & Guerra 2013a).

3.2.1 Guidance to Evaluate Water Use and Production in the Oil and Gas Industry (2014)

The guidance document developed in this study provides guidance to water managers on evaluating water use and production in the oil and gas sector of the energy industry (Dahm 2014). Water management strategies that highlight tradeoffs in water management options to reduce demand and increase water supply are discussed. Options highlighted include:

- Using alternative water sources to develop wells
- Providing on-site industrial water reuse or recycling
- Using produced water post-well completion in beneficial ways

This guidance includes formulas to calculate the amount of water used and produced in these various management strategies (Dahm & Guerra 2013a). This document also provides a standard assessment method for determining supply and demand with a focus on consumptive use calculations associated with energy production. A framework was developed for considering water use in the oil gas industry, shown in Figure 8.

A series of case studies were evaluated in which water treatment technologies were used to treat flowback and produced water. These case studies illustrated the technologies used and the benefits of treatment technology implementation (Dahm & Guerra 2013b).

3.3 Desalination and Water Purification Research (DWPR) Program

While the DWPR program has not had produced water treatment as a focus area, research funded through the program is applicable to the oil and gas industry. Three DWPR projects had a specific produced water focus and three others investigated technologies that have been or are currently used in use in the oil and gas industry for water treatment.

3.3.1 Multi-Beneficial Use of Produced Water Through High-Pressure Membrane Treatment and Capacitive Deionization Technology (2009)

In this study, researchers at the Colorado School of Mines investigated the use of ultra-low pressure reverse osmosis (ULPRO), nanofiltration (NF) membranes, and capacitive deionization (CDT) to treat produced water to nonpotable and potable water quality standards. Recovery of salable iodide was also investigated (Drewes et al. 2005).

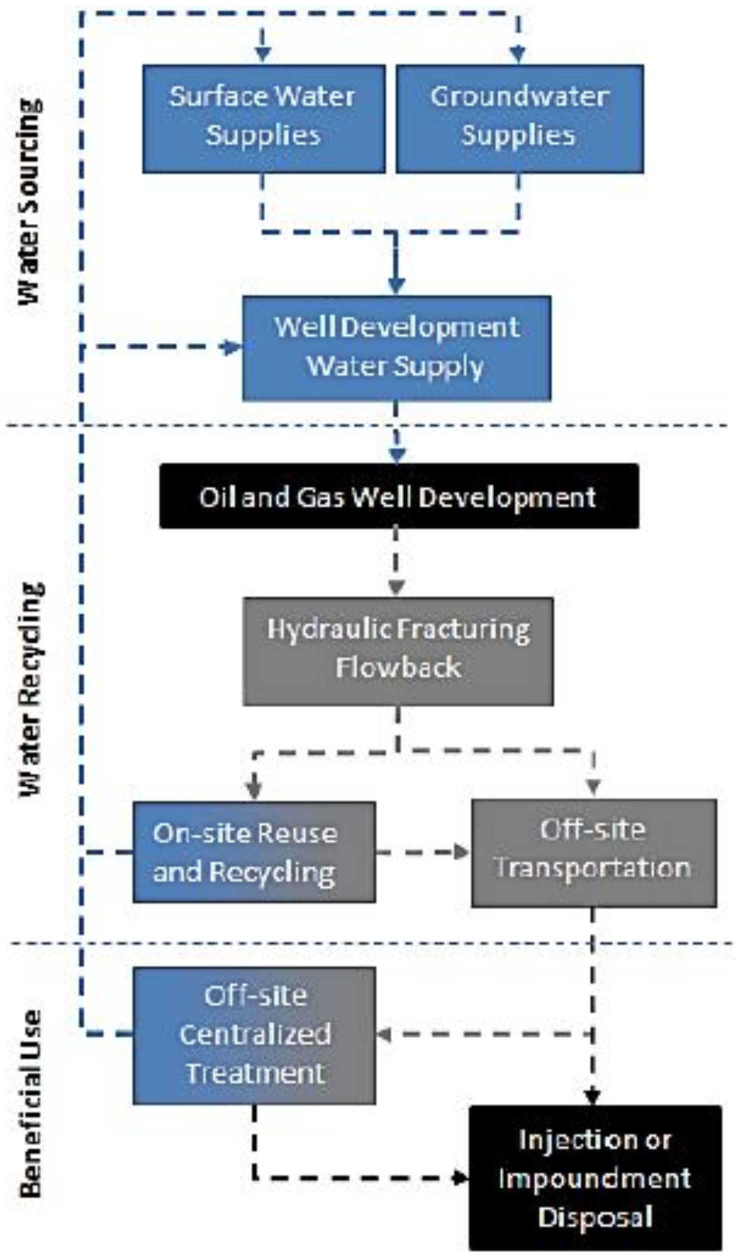

Figure 8.—Framework for alternative water use and treatment in the oil and gas industry (Dahm 2014).

Membrane fouling and scaling is the biggest challenge to employing membrane technology in produced water treatment. This study found that the fouling propensity of the membranes depended on membrane properties such as hydrophobicity and roughness. Smooth hydrophilic membranes exhibited less fouling than rough hydrophobic membranes. Chemical cleaning using caustic and anionic surfactant solutions was shown to restore permeate flux after fouling.

ULPRO membranes performed better than nanofiltration for producing high solute rejection and high recovery of iodide. ULPRO membranes were also more cost effective than nanofiltration membranes. NF membranes could be used to treat the produced water to primary and most secondary drinking water standards. Chloride and TDS exceeded the secondary standard in the NF permeate.

Compared to membrane technology, which often needs rigorous and complex pretreatment, CDT required minimum pretreatment (such as cartridge filters), and no chemicals for scaling control and chemical cleaning. However, because of slow mass transport rate of ions adsorbing onto and desorbing from carbon aerogels, water recovery for the CDT was low compared to the ULPRO process. A large amount of concentrate waste was produced during electrode regeneration and rinsing process. CDT may be a potential alternative to brackish water reverse osmosis (RO), however, the efficiency and system design need to be improved before the technology becomes economically feasible for commercialization.

Membrane technology was more cost-effective than CDT and provided a better overall performance in terms of product water quality, iodide recovery, and energy consumption. Both the product water from CDT and membrane technologies required post-treatment for stabilization, removal of boron, and adjustment of sodium adsorption ratio (SAR) for agricultural irrigation.

3.3.2 Novel Fouling Resistant Membranes for Water Purification (2008)

In this study, researchers at the University of Texas, Austin, developed an approach of applying a very thin coating of fouling-resistant polymer to membrane surfaces to increase the oil/water fouling resistance of commercially available ultrafiltration (UF), nanofiltration (NF), and RO membranes (Ju & Freeman 2006). Dynamic oil fouling filtration experiments in dead-end and crossflow modes were performed using well-characterized oil/water emulsions, and the coated polysulfone (PSF) membranes showed water flux values about five times higher than those of uncoated PSF membranes.

3.3.3 Membrane Technology for the Recovery of Produced Water (Not published)

This study, by Western Environmental, investigated microfiltration (MF), UF, NF, and RO membranes for the treatment of produced water. The study encountered challenges with testing resulting in membrane fouling and successful results were not obtained during the funding period.

3.3.4 DWPR Projects on Technologies Used in the Oil and Gas Industry

Before about 2005 the oil and gas industry was relying on the use of technologies originally developed for the municipal water treatment industry to solve its water treatment challenges. As a result, many of the technologies investigated in the DWPR program were also investigated for use in the oil and gas industry.

Typically, more novel and emerging technologies can be used economically in the oil and gas industry compared to technology use in the municipal water treatment industry. There are a number of factors that contribute to this, including:

- More favorable economic conditions
- Complex water chemistry
- Easier regulatory process for employing new technologies
- Less concern over public perception and less direct risk to public health

As a result, many emerging technologies may find commercial success in the oil and gas industry before being used for municipal purposes. In the future, technologies may be developed to the point in the oil and gas industry that they can be commercially viable for more traditional water treatment applications.

Research areas that are routinely funded by DWPR that are relevant to oil and gas water treatment include the following:

- Cost and energy reduction of treatment processes
- Concentrate management
- Zero liquid discharge
- Mineral recovery

Technologies that have been funded by DWPR that could also be applicable to the oil and gas industry include membrane distillation and forward osmosis.

The following sections describe technologies that were funded under DWPR for technology development; but weren't necessarily targeting technologies for oil and gas operations.

3.3.4.1 Dewvaporation

The DWPR program funded a pilot test of dewvaporation, a humidification-dehumidification cycle process, in 2003. In dewvaporation, feed water is evaporated by hot air on one side of a heat transfer wall and fresh water is condensed on the other side. The condensate or dew collects on the other side of the wall represents the purified water stream. Because the process uses low energy and low pressures compared to either conventional thermal and membrane processes, the operating and capital costs of the process are expected to be more favorable.

In the 2003 DWPR study (03-FC-81-0905), a 5,000-gallon-per-day dewvaporation pilot plant was designed, built, and operated at the 23rd Avenue Waste Water Treatment Plant (WWTP) in Phoenix, Arizona. The city of Phoenix Water Services Department, along with Reclamation Phoenix Area Office, cooperated to establish a pilot plant site. The pilot plant feed was the concentrate from a Tactical Water Purifier System RO unit with UF pretreatment. A 2,000 liter (mg/L) TDS waste water RO concentrate stream was treated by the pilot plant to more than 45,000 mg/L of TDS brine and 10 mg/L of TDS distillate. Recovery varied from 70 to 100 percent, with no decrease in distillate rate or increase in distillate contamination. The operating cost was estimated to be \$20.85 per 1,000 gallons. The use of waste heat or solar thermal would reduce the operating cost to the cost of water pumping and air blowing. Power needs of 0.5 kilowatt-hour (kWh) per 1,000 gallons at \$0.10 per kWh would amount to \$0.05 per 1,000 gallons (Beckman 2008).

Since the DWPR pilot project, Altela, Inc. has licensed the technology and now sells AltelaRain® treatment plants for uses such as brine minimization in the oil and gas industry. Treatment plants range in size from 2,100 to over 10,400 gallons per day (mobile system) and larger for fixed treatment plants.

An example project installation for the AltelaRain® technology for produced water treatment is Piceance Basin Waste-To-Asset Conversion project in western Colorado (Altela n.d.). Altela partnered with a natural gas exploration company it the Piceance Basin of western Colorado to use the AltelaRain® system to treat produced water and flowback water. Altela acquired precedent-setting regulatory approvals to discharge the clean, treated water to the Colorado River Basin. This was the first of its kind non-tributary water right approval for water discharged to the Colorado River Basin for beneficial reuse.

This project is a good example of the use of water treatment technology to create a usable, valuable water supply from produced water. Other AltelaRain® projects related to water management in the oil and gas industry are installed in the San Juan Basin, NM, Marcellus Shale Basin in Pennsylvania, and in northwestern Alberta, Canada.

Another project in New Mexico, within the Navajo Nation, uses the AltelaRain® system to treat produced water generated at natural gas wells located on Navajo Nation lands. These wells were generating produced water that was being transported approximately significant distances for underground injection and disposal. By deploying the AltelaRain® system, the per gallon disposal fee was reduced by approximately 30 percent. Following treatment, the purified water was made available, for free, to the Navajo Nation for valuable re-use.

The Altela system is now also being used for the treatment of RO concentrate in the South Platte River Basin (an alternative to injection wells). The clean, treated water can be surfaced discharged to the South Platte River for valuable return flow water right credits in accordance with western prior appropriations water law frameworks. By considering water value for beneficial use such as return flows, capital outlays for additional water rights can be reduced.

3.3.5 Natural Freeze-Thaw (FT) Process

In 1998, Reclamation funded the project "Demonstration of the Natural Freeze-Thaw Process for the Desalination of Water From the Devils Lake Chain to Provide Water for the City of Devils Lake." The project took saline feed water directly from Devils Lake and desalinated it using the natural FT process. Samples of feed, treated water, and concentrated brine were collected and analyzed during operations to determine the viability of a full-scale FT plant and to demonstrate the performance of the process (Boysen & Harju 1999).

The freeze-thaw process, called FTE® process, has been operated at a commercial-scale under Wyoming Department of Environmental Quality Permits and Regulation in the Great Divide Basin of WY since 1999. The initial plant (1999-2002) had a nominal 500 barrel (bbl)/day (21,000 gallons per day) capacity. The current plant (2003 to present) has a 1,000 bbl/day capacity. The FTE® process was also operated at a commercial-scale under Wyoming Oil and Gas Conservation Commission Permits and Regulation in the Jonah Field of WY from 1997 - 2004. Pilot-scale operation of the FTE® process was successfully operated in the San Juan Basin of NM (1996 and 1997), Devils Lake in North Dakota (1999), Farson, WY (1970's) and Evers Ranch in WY (1960's and 1970's (Boysen 2008).

4. Research Funded by Others

This section describes relevant current and past research and activities conducted by others in the area of water management for oil and gas.

This section contains a summary of research funded by DOE and National Science Foundation (NSF) on produced water. There is also a summary of research conducted by the national labs and non-profit organizations. Other agencies, such as the Environmental Protection Agency (EPA) have an interest in produced water, but do not appear to be actively conducting research in this area.

Numerous private sector companies have focused on oil field services, technology vendors, and sensor and instrumentation manufacturers are also conducting a significant amount of research and pilot testing in this industry. Those activities are too numerous to summarize here, however, those companies may make very suitable partners for future work.

4.1 Multi-Agency Collaboration on Unconventional Oil and Gas Research

DOE, EPA, and the Department of Interior (Interior) formed a multi-agency collaboration to investigate research needs in the oil and gas industry. The Interior agency participating in this effort is USGS. Results of this effort were recently published in a report to the Congress (Anon 2015).

The report focused on the following topics:

- Water quality
- Water availability
- Induced seismicity
- Resource assessments
- Ecological effects
- Human health

Within the areas of water quality and water availability as they related to treatment the three agencies described their role and activities:

DOE is developing technologies for water reuse and recycling to reduce the amount of water requiring disposal or treatment. DOE is investing in technologies that could reduce the utilization of valuable freshwater resources for hydraulic fracturing. DOE conducted pilot testing of pretreatment options to allow removal of naturally occurring radioactive material, salt crystal recovery, and reuse of produced water.

DOI is researching the potential impacts of Unconventional Oil and Gas (UOG) activities on surface water and groundwater quality. Research includes determining the baseline water quality conditions; assessing the potential for migration of methane gas and other hydrocarbons; investigating the environmental contaminants due to spills from UOG wastewater management activities, and developing geochemical methods and groundwater flow models to evaluate potential contamination of water supplies.

EPA is assessing the potential impacts of hydraulic fracturing activities on drinking water resources in the US to improve understanding of the factors and drivers that may affect the frequency and severity of these impacts. EPA is also investigating water withdrawal impacts on drinking water supplies. Water withdrawals for hydraulic fracturing have the greatest potential to impact drinking water availability in areas with, or in times of, low water availability, exacerbated by drought, and over allocation of water. EPA researchers published a study that examined the impacts of water withdrawals in the Upper Susquehanna and Upper

Colorado River Basins. EPA's analysis has been focused on water quality impacts to drinking water rather than water availability.

Goals of future research in this area include the following:

- Identify alternative sources of water for UOG development to replace the use of freshwater sources.

- Increase the number of wastewater treatment options.

Future research identified is the following:

- Determine the effect of water withdrawals for UOG production on headwater streams and drinking water aquifers.

- Develop technologies and management practices for reducing fresh water demand and increasing the recycling of produced water.

4.2 Department of Energy

DOE has the most comprehensive research program in the area of produced water of any of the government agencies. DOE works with states, other government agencies and non-governmental organizations to develop tools to aid operators in meeting the environmental and economic challenges in managing produced water. The overall goal DOE research in this area is to allow for the expansion of oil and gas production, while protecting the environment and increasing the supply of water for consumers.

Produced water research is funded by DOE through the NETL which administers the Research Partnership to Secure Energy for America (RPSEA). The latest request for proposals due dates listed on the RPSEA website are for 2013. Most projects have been completed and the on-going projects have completion dates in 2016.

4.2.1.1 RPSEA Unconventional Resources Program

Under RPSEA, the Unconventional Resources Program funds work on water management, including water use planning and treatment. The mission of the Unconventional Resources Program is to increase the supply of domestic natural gas and other petroleum resources by reducing the cost and increasing the efficiency of exploration for and production of such resources, while improving safety and minimizing environmental impact (http://www.rpsea.org/unconventional-resources-program/). Water is a critical component to meeting this program objective. A summary of relevant water treatment projects as part of the program is provided in Table 1.

4.2.1.2 RPSEA Small Producers Program Relevant Projects Summary

The Small Producer Program is established to benefit small producing companies in technology development for mature oil and gas fields, with the objective of extending the life and ultimate recovery of these fields. This is an important group to overall US production. The goal of this program is to carry out research, development and demonstration efforts that will assist small producers in reducing the cost and increasing the efficiency of exploration and production while operating safely and in a manner which does not harm the environment. Efficient, cost effective water management is a critical part of achieving that objective. A summary of relevant water treatment projects as part of the program is provided in Table 2.

4.3 National Science Foundation

The National Science Foundation (NSF) has funded research in the area of produced water treatment through the Small Business Innovation Research program. Through this program, NSF has funded small business to develop new technologies for treating produced water. Absorbent Materials Company, received a Phase I and Phase II award to investigate the use of swellable glass absorbents for the removal of organic compounds from produced water. Symbios Technologies also received an award to develop a technology to develop a plasma treatment system to oxidize organic compounds for treatment of waters such as produced water.

FloDesign Sonics developed a new, efficient separation technology to treat produced water. The process uses a method called acoustophoresis, in which droplets or particles within a liquid can be manipulated with a special acoustic wave pattern. Depending on their relative density compared to the liquid, these larger clusters either settle to the bottom or rise to the surface, where they can be separated easily. The technology can remove particles smaller than 20 microns without the addition of chemicals. The company has 7,000 -gallon per day prototype for pilot testing with plans to develop a 100,000 gallons per day system.

NSF is currently funded the Air Water Gas Network, Routes to Sustainability for Natural Gas Development, and Water and Air Resources in the Rocky Mountain Region, a Sustainability Research Network. Water treatment is a component of this larger, multi-disciplinary effort. The water treatment team members are working to develop sustainable techniques for on-site treatment of wastewater that could decrease the need for trucking and injecting wastewater into deep wells, and increase the feasibility of water re-use, including forward osmosis and membrane distillation.

Table 1.—Summary of Relevant Research from RPSEA Unconventional Resources Program.

Project Title	Conducting Organization	End Date	Brief Description
An Integrated Framework for the Treatment and Management of Produced Water	Colorado School of Mines	2011	This project developed a web-based treatment selection and screening tool that allow for the selection of the best fit for purpose technology for treating produced water from coalbed methane extraction.
Pretreatment and Water Management for Fracturing Water Reuse and Salt Production	General Electric Global Research	2011	This study investigated a lime-soda process for the removal of magnesium, calcium, and strontium for nonhazardous solid waste disposal followed by barium and radium precipitation as carbonates. The carbonates were re-dissolved and disposed of by well through the Underground Injection Control program. A second process was studied that used manganese dioxide as an adsorbent for barium and radium.
Novel Engineered Osmosis Technology: A Comprehensive Approach to the Treatment and Reuse of Produced Water and Drilling Wastewater	Colorado School of Mines	May 2016	The objective of this research effort is to investigate the osmotically driven membrane processes. The proposed research will advance the development and implementation of the forward osmosis, osmotic dilution, and a novel ultrafiltration processes for treatment and management of well drilling and stimulation wastewater and produced water in many unconventional and conventional gas and oil fields.
Advancing a Web-Based Tool for Unconventional Natural Gas Development with Focus on Flowback and Produced Water Characterization, Treatment and Beneficial Use	Colorado School of Mines	May 2016	The objective of this study is to provide a set of web-based tools that will enable producers and other users to characterize, treat, beneficially use, and manage produced water and fracturing flowback water from unconventional gas production. The goal is to sustain gas production while minimizing potential impacts on natural water resources, public health, and environment. Built upon the integrated decision making framework developed for CBM produced water management, the proposed study focuses on shale gas and tight sand production, the most difficult and least developed.
Development of Geographic Information System (GIS)-Based Tools for Optimized Fluid Management in Shale Gas Operations	Colorado State University	Sept 2016	The overall objective of the proposal is to develop GIS-based tools that can be used to optimize water management decisions during unconventional oil and gas development and production to minimize the environmental impact. The environmental impacts that will be directly assessed with the tool include the handling, treatment and disposal of produced water, air toxics and greenhouse gases associated with fluids handling, water footprint, and the optimal siting of wells and treatment facilities with respect to community impacts.
Advanced Treatment of Shale Gas Fracturing Water to Produce National Pollutant Discharge Elimination Program Quality Water	M2 Water Treatment	2015	The project will investigate an integrated approach using magnetic ballast clarification , vortex-generating,and NF membranes, and hydrogel media or precipitation/solidification/stabilization to treat produced water
Cost-Effective Treatment of Flowback and Produced Waters Via an Integrated Precipitative Supercritical (IPSC) Process	The University of Ohio	2015	A treatment process consisting of ultraviolet treatment, chemical precipitation/adsorption, and supercritical water was constructed and tested to validate technical feasibility and to acquire information necessary to design, construct, and operate a pilot-scale IPSC process unit. In the second phase of the project, performance of a pilot-scale IPSC process unit will be demonstrated in order to acquire engineering information necessary to develop detailed techno-economic evaluation and a commercial-scale engineering design package.

Table 2.—Summary of Relevant Research from RPSEA Small Producers Program

Project Title	Conducting Organization	End Date	Brief Description
Cost Effective Treatment of Produced Water Using Co-Produced Energy Sources for Small Producers	New Mexico Institute of Mining and Technology	2012	This project aimed to demonstrate a cost-effective process for produced water purification at the wellhead using a low-temperature distillation unit (humidification/dehumidification process). The researchers constructed a demonstration unit that can utilized solar energy and coproduced geothermal energy for wellhead produced water desalination (http://www.rpsea.org/projects/07123-05/).
Cost Effective Treatment of Produced Water Using Co-Produced Energy Sources, Phase II: Field Scale Demonstration and Commercialization	New Mexico Institute of Mining and Technology	2015	This project continued the development of the humidification/dehumidification process. The process was shown to use solar energy and other co-produced energy sources to reduce the electricity consumption of the process to 0.16 kWh/barrel resulting in a water production cost of approximately $0.18/barrel (http://www.rpsea.org/media/files/project/08495dd0/11123-03-TS-Cost-Effective_Treatment_Produced_Water_Co-Produced_Energy_Sources-12-14-12.pdf).
Treatment and Beneficial Reuse of Produced Waters Using a Novel Pervaporation-Based Irrigation Technology	University of Wyoming	2014	The project evaluated the application of a novel pervaporation-based irrigation technology to treat and reuse oil and natural gas produced water. The project found that pervaporation process showed promise to be used as a treatment and irrigation system. However, for treating the volumes of produced water generated using this technology for irrigation the membrane properties would need to be improved (http://www.rpsea.org/media/files/project/60b022ec/09123-11-FR-Treatment_Beneficial_Reuse_Produced_Waters_Novel_Pervaporation_Irrigation-03-20-14_P.pdf).
Basin-Scale Produced Water Management Tools and Options – GIS-Based Models and Statistical Analysis of Shale Gas/Tight Sand Reservoirs and Their Produced Water Streams	Utah Geological Survey	2015	This project had the following objectives: (1) create basin-wide, digital produced water management tools that integrate produced water character, water disposal/reuse, water transport, and groundwater sensitivity factors to allow for quicker and more efficient regulatory and management decisions related to unconventional gas developments in the Uinta Basin; (2) investigate the option of beneficial use of produced water treatment for geothermal heat recovery or power generation in the Uinta Basin; (3) promote maximized produced water reuse which will minimize use of freshwater in unconventional gas development and production; (4) compile Uinta Basin produced water management practices and recommend best practices; and (5) seek to increase protection of critical Uinta Basin alluvial aquifers.
A Portable, Two Stage, Antifouling Hollow Fiber Membrane Nanofiltration Process for the Cost Effective Treatment of Produced Water	New Mexico Institute of Mining and Technology	June 2016	The overall objective of this project is to develop and demonstrate the performance and cost-effectiveness of the portable Two-Stage, Antifouling Hollow Fiber Membrane NF process to convert produced water into a clean water product for a reused fluid or direct discharge.
Water Treatment System for Effective Acid Mine Drainage Water Using Hydraulic Fracturing	PPG Industries	July 2016	The project will develop a novel ion exchange membrane and water treatment process capable of reducing sulfates to no more than 500 ppm at bench-scale influent flow rates of 1-5 gal/min. Filter cartridges, skids, and system maintenance processes for the new system will be evaluated at intermediate scale influent flow rates of 10-50 gal/min. This design will be optimized so that one, non-specialized laborer can operate and maintain the system. This design will undergo field validation of the efficacy and ease of use of the new treatment process. The field tests will be conducted at drilling locations at targeted influent flow rates of 400-500 gal/min.

4.4 National Laboratories

National laboratories have conducted research in the area of produced water. The following table summarizes efforts by the labs, Table 3.

Table 3.—Government Labs Working In Oil and Gas Water Management

Agency	Summary of Research
Los Alamos National Laboratory	Developing novel technologies to treat produced water as new water source in NM, beneficial use of produced water for growing biofuel precursors
National Renewable Energy Laboratory	Investigating the use of water treatment technologies to increase energy and water supplies in a cost effective manner
National Energy Technology Laboratory	Administers DOE program on produced water, studied constructed wetland treatment systems for produced water
Argonne National Laboratory	Investigate water quality, quantity and identify treatment options for produced water. Produced White Water Paper: background and regulations on produced water as well as discussion on options for managing produced water (prepared for the DOE)
Sandia National Laboratory	Conduct applied research projects in desalination of brackish ground waters and produced waters Developed “Energy-Water Decision Support System” to enable planners to analyze the potential implications of water stress for transmission and resource planning

5. Recommendations for Future Work

Based on the past research by Reclamation and the research currently being conducted by others, recommendations are made for future work in this area. Future efforts by Reclamation should focus on partnering with others working in this area in order to make a meaningful contribution to solving Reclamation’s water supply challenges as well as improving water challenges in the oil and gas industry.

Two potential areas are identified for future work. These two potential projects are based on the premise of efficient water management and treatment of non-traditional water sources to increase water supplies for stakeholders with competing needs for water.

5.1 Increasing Agricultural Water Supplies Through Treatment and Beneficial Use of Flowback and Produced Water

A promising area for Reclamation involvement in oil and gas water management is the study of agricultural uses for fracturing flowback water and produced water. Due to public perception challenges associated with these water sources, the use of produced and flowback water for drinking is unlikely; however, there is a potential for produced water to be treated and used for irrigation and livestock watering. The use of flowback and produced water to offset conventional supplies currently used for irrigation would free up more water because standards for irrigation water are much lower than drinking water standards for other uses and may increase water usage efficiency.

Because the management of flowback and produced water typically requires disposal costs such as transportation and treatment, the treatment of these waters for agricultural purposes may be almost entirely offset by the disposal costs paid by the industry. Therefore, there is a huge potential for flowback and produced water to be treated for agricultural water affordably.

Based on the past research in produced water, it is clear that in order to make an impact, projects need to be multi-disciplinary and actively involve project partners and stakeholders. The beneficial use of produced water is multi-faceted and would require the successful, engagement of stakeholders and partners. Potential partners for a project in this area include the following:

- Irrigation district(s) in states with significant oil and gas development, such as Texas, California, Oklahoma, Colorado, or the Dakotas
- Relevant Reclamation area office
- Well field service companies and petroleum producers
- Treatment equipment suppliers

The project would include the selection of a relevant, representative study area. Selection of the study location should consider the type of petroleum produced, production water needs, and water production characteristics.

Water quality requirements for agricultural uses such as irrigation and livestock watering should be identified and characterized. The produced water and flowback water quality can be compared to the water quality requirements of the beneficial uses to determine treatment needs.

Institutional and regulatory issues will need to be considered to understand the implications of augmenting agricultural water supplies on water rights in the area.

This will help to determine how produced water can be used to offset current agricultural water uses in the area. A framework will be developed that can be used as a model for implementation in other areas.

Pilot testing of the treatment technology is necessary to understand the cost of the water produced as this will be a critical determining factor in the feasibility of the use of produced water.

5.2 Use of Alternative Water Supplies for Hydraulic Fracturing

The use of alternative water (such as brackish groundwater or treated wastewater) rather than conventional supplies for oilfield uses (such as hydraulic fracturing, drilling fluids, and other field water uses) is an area where Reclamation's water resources experiences, knowledge of local stakeholders needs and concerns, and water treatment experience could be leveraged to reduce the competing needs for fresh water resources in Reclamation project areas with oil and gas development. Potential partners for this project include:

- Well service companies and petroleum producers
- Local Reclamation area office

As in the previous project, the selection of the project focus area is critical and should be selected to include an area with a projected increase in the use of hydraulic fracturing. Areas to consider include those currently experiencing water stress due to increasing fresh water demand, changing water availability due to drought and climate change, and a include a diverse array of water users.

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