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A modeling framework for technical, economic, energetic, and environmental assessment of produced water pretreatment from oil and gas industry

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ABSTRACT

Treatment and reuse of produced water (PW) generated from oil and gas industry can reduce PW disposal volume and costs, improve environmental sustainability, and offset freshwater uses. Due to the complexity and high scaling and fouling propensity of PW, PW pretreatment is essential to ensure the long-term operation of downstream desalination processes. This study developed a process modeling approach to evaluate pretreatment technologies through technical, economic, energetic, and environmental assessments to identify the impacts of each technology, such as costs, energy consumption, and carbon dioxide emissions, to make informed decisions for integrated treatment train development and applications. The evaluated individual and combined PW pretreatment technologies included chemical softening (CS), chemical coagulation (CC), electrocoagulation (EC), and granular activated carbon (GAC) for removing key fouling and scaling constituents, such as hardness, silica, and organics. The main evaluation parameters include levelized cost of water ($/m^3), cumulative energy demand (kWh/m^3), specific energy consumption (kWh/m^3), and carbon dioxide emissions (kg $CO_{2\text{-eq}}$/m^3). The case study evaluated the unconventional PW in the Permian Basin with total dissolved solids concentration of 130,000 mg/L. For pretreatment combinations, the implementation of EC+GAC was selected as the optimal choice due to its effectiveness and limiting the amount of waste for disposal. This study provided a modeling framework for optimization and integration of different pretreatment units accounting for three evaluation metrics (costs, energy, and CO_2 emissions) to effectively evaluate their viability in PW applications centered on minimal- or zero-liquid discharge.

1. Introduction

Produced water (PW) is wastewater generated during the production of oil and gas from both onshore and offshore wells. It is one of the largest and fastest-growing wastewater streams in the United States (U.S.), with approximately 25.8 billion barrels of onshore PW (4 billion m^3) being generated in 2021 [1]. Approximately 96 % of PW is disposed of by deep-well injection, which may lead to increased risks of induced seismicity, surface water contamination due to spills, and subsurface freshwater aquifer contamination due to downhole inter-zone communication within injection wells. Over the past decade, an increase in unconventional oil and natural gas development in the U.S. and an associated increase in the use of a large volume of water for hydraulic fracturing has led to a dramatic increase in both the demand for freshwater and the volumes of PW requiring treatment and/or disposal [2]. There is an increasing interest in treating PW for reuse inside and outside the oil and gas sectors to reduce PW volume for disposal and potentially provide a new water supply for fit-for-purpose applications [3,4]. Yet one of the major challenge for PW reuse is the formation of precipitates that can scale equipment for water treatment (especially desalination units) and potentially plugging brine disposal facilities such as saltwater

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Nomenclature

Al_2SO_4	Alum coagulant
$Ca(OH)_2$	Hydrated lime
CAPEX	Capital expenditure
CC	Chemical coagulation
CED	Cumulative energy demand
CO_2	Carbon dioxide emissions
CS	Chemical softening
DAF	Dissolved air flotation
EC	Electrocoagulation
$FeCl_3$	Ferric chloride coagulant
GAC	Granular activated carbon
LCOW	Levelized cost of water
MLD	Minimal liquid discharge
Na_2CO_3	Soda ash
NaOH	Sodium hydroxide
OPEX	Operational expenditure
PW	Produced water
SEC	Specific energy consumption
TDS	Total dissolved solids
TOC	Total organic carbon
TSS	Total suspended solids
ZLD	Zero liquid discharge

disposal wells [5,6].

PW has a complex composition consisting of suspended solids, oils, petroleum hydrocarbons, inorganic salts/minerals, heavy metals, naturally occurring radioactive materials, and chemical additives from fracturing [7–9]. Its high total dissolved solids (TDS) and variety of complex and toxic substances can be hazardous to the environment if disposed of or reused without appropriate treatment [10,11]. There have been a variety of studies on PW treatment using physical, chemical, biological, and membrane technologies, or a combination of them [6, 12–18]. Due to its complex water matrix, PW treatment typically consists of a variety of unit processes in series, i.e., an integrated treatment train, to tackle the contaminants of concern that may not be removed in a single process [9]. Although evaluations in the literature reported economic analysis and the life cycle impacts for PW treatment [19–22], these studies did not account for the evaluation of the integrated units needed for an effective beneficial reuse application and often just focused on the desalination aspects of treatment. Fig. 1 shows the required treatment stages typical for PW treatment and management. Both minimal liquid discharge (MLD) (up to 95 % water recovery) and zero liquid discharge (ZLD) (~100 % water recovery) applications offer a promising strategy for PW treatment and management since these involve the use of multiple treatment units to recover as much water as possible. These types of applications have been noted to be highly energy intensive, especially for PW due to its high TDS range, typically from 1,000 to 400,000 milligrams per liter (mg/L) [9,23–26].

The incorporation of pretreatment is imperative for successful PW treatment and MLD/ZLD applications to reduce potential scaling and fouling substances, but it is often overlooked due to most of the assessments being directed to the other downstream treatment stages focused on desalination, evaporation, and crystallization. There are limited studies that have quantified the impact that different pretreatment applications have on costs, energy, and the environment for a specific case study; this is especially true when considering an integrated PW treatment train. In addition, most of the focus on PW treatment has been centered on the desalination aspects of the treatment train [27]. It is often assumed the presence of solely the salinity/TDS (assumed to consist mainly of sodium and chloride ions) going to the desalination treatment stage and ignores the presence of other constituents such as inorganic minerals, heavy metals, and petroleum hydrocarbons, because these are assumed to have been removed before desalination.

Considering that pretreatment is essential to treating waters with high scaling and fouling potential, a thorough economic, energetic, and environmental impact assessment is valuable for the evaluation and selection of appropriate individual or combination of pretreatment technologies to minimize downstream scaling and fouling on integrated PW treatment trains. This study is focused on developing a modeling framework using a publicly available program - WaterTAP to evaluate the pretreatment units in terms of the costs, energy, and carbon dioxide (CO_2) emissions in a case study treating the unconventional PW in the Permian Basin, United States. WaterTAP is an open-source program developed by the National Alliance for Water Innovation (NAWI), which provides simulations of water treatment systems under steady-state conditions to effectively estimate performance, energy, and costs using a series of unit processes subjected to systematic constraints [28,29]. In addition to WaterTAP [28], additional software such as PHREEQC [30] and Avista's AdvisorCi [31] was incorporated into this assessment to evaluate water scaling potential. Evaluating pretreatment options in PW is crucial because inadequate pretreatment can lead to severe fouling, scaling, and membrane degradation in desalination units, significantly increasing operational costs and reducing system efficiency. While most research focuses on desalination performance in PW applications, the long-term viability and cost-effectiveness of these systems heavily depend on proper pretreatment, which ensures stable operation, extends equipment lifespan, and minimizes energy-intensive cleaning or

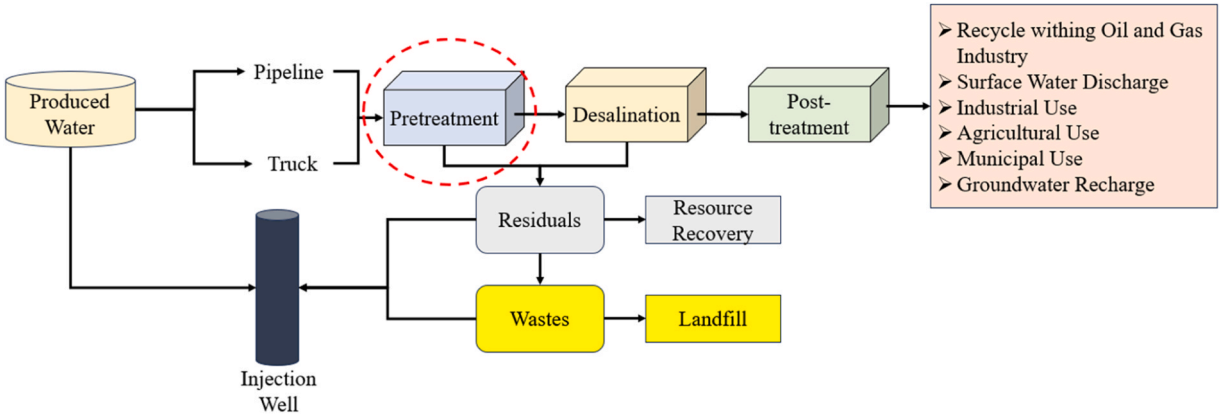

Fig. 1. Overview of pathways for produced water treatment and reuse.

replacement. It is important to note that our findings are intended as guiding estimates, offering a framework rather than rigid solutions. The choice of pretreatment methods should remain flexible, tailored to the unique properties of the PW and the specific economic context of the application site.

2. Pretreatment of PW

Pretreatment is critical to enable downstream desalination units in MLD/ZLD to have a higher water recovery and extend their operational lifetime [32]. An analysis of up to 50 studies reported that the most prominent foulants that contribute to the different fouling mechanisms in membrane-based treatment result from the organic constituents in PW, specifically oil and the total organic carbon (TOC) content [33]. In addition, the elevated levels of inorganic minerals and salts present in PW may result in significant scaling and colloidal accumulation. Some of the most common scales reported in oilfield PW are calcium carbonate ($CaCO_3$), calcium sulfate ($CaSO_4$), barium sulfate ($BaSO_4$), strontium sulfate ($SrSO_4$), ferrous carbonate ($FeCO_3$), ferrous sulfide (FeS), ferrous hydroxide ($Fe(OH)_2$), ferric hydroxide ($Fe(OH)_3$), calcium silicate ($CaSiO_3$), and magnesium silicate ($MgSiO_3$) [34–36].

The mechanisms involved in fouling and scaling of membrane-based desalination processes include particulate fouling, concentration polarization, pore blocking, cake formation, inorganic precipitation, organic adsorption, and biological fouling [37,38]. These fouling/scaling mechanisms, except pore blocking, are also applicable for thermal desalination. Because PW has a complex water matrix, MLD/ZLD applications need to have a greater focus on reducing fouling and scaling to enhance the performance of the downstream membrane or thermal desalination processes [39,40]. A thorough assessment of the PW chemistry, identification of the removal requirements, and cost/environmental limitations must be considered to effectively design appropriate pretreatment process combinations for MLD/ZLD applications [32,37].

Some common PW pretreatment technologies include chemical softening (CS), chemical coagulation (CC), and electrocoagulation (EC), of which all three were chosen in this study to be evaluated as effective pretreatment alternatives [41–43]. The descriptions and mechanisms of these pretreatment technologies are discussed in the reference [44,45]. Adsorption using granular activated carbon (GAC) is also considered in this work, as it is effective in removing a range of organics present in oil & gas wastewater [46–49]. The adsorption capacities of GAC depend on the pore size, the internal surface area of the pores, and its surface properties [50]. It is important to note that the ionic strength of PW can greatly influence the adsorption capacity of an adsorbent, either positively or negatively, through altering its surface potential [51–53]. There are very limited studies using GAC in highly saline water treatment; it is generally believed that the high salinity does not pose a significant impact on the adsorptive capacity of adsorbents and rather sometimes enhances the organic adsorption through the salting-out effect, which promotes the transfer of organic molecules from the aqueous phase to the adsorbent surface, enhancing the adsorption capacity [54–64]. For this assessment, it is assumed that the PW salinity does not affect the adsorptive capacity of organics in GAC. Applications of GAC in PW treatment have often been focused on the integration of a biologically active layer in the surface of the adsorbent, known as biologically active filtration. This application has been demonstrated to effectively remove $>$ 40 % of the incoming organics in PW [48,65,66]. Other pretreatment technologies such as membrane filtration (e.g., microfiltration or ultrafiltration), biological treatment, and advanced oxidation were excluded from this study due to their high scaling and fouling propensity, operational challenges and toxicity to the microorganisms, and high unit economic and energetic demand, respectively [32,37,67]. Ion exchange pretreatment was also excluded since the high TDS of the PW will exhaust the exchange capacity of the resins significantly [54,68]. Table 1 lists reported removal efficiencies in PW treatment for the different pretreatment units in this study.

3. Methods

3.1. Case study description

In this study, the Permian Basin is selected for the case study to treat the PW from unconventional oil and gas extraction. The Permian Basin in southeastern New Mexico and west Texas is the most productive oil province in the United States [10,76]. With high oil and gas production comes a large volume of PW that is traditionally managed by injection to deep wells for disposal. However, this has been linked to induced seismicity in the region, raising concerns about long-term sustainability. The increasing water demand for hydraulic fracturing in the Permian Basin and water shortage have drawn significant interest in PW treatment and reuse in water-stressed regions such as New Mexico and Texas. These issues may be alleviated by reusing PW for the hydraulic fracturing of new wells after partial treatment. It has been reported that the PW volumes exceed the hydraulic fracturing water demand in the Permian Basin [77], which poses an opportunity for PW treatment and fit-for-purpose reuse outside of oil and gas fields, such as rangeland restoration, agricultural irrigation, and other industrial applications [78,79]. Like many other unconventional PW, such as in Marcellus formation in Pennsylvania, the PW in the Permian Basin is hypersaline with average TDS of 130,000 mg/L, along with hydrocarbons, heavy metals, and naturally occurring radioactive materials (NORM) [80]. Table 2 presents the respective water quality characteristics for the Permian Basin PW case study. The composition of PW is highly variable

Table 1
Removal efficiencies for the different pretreatment options reported in the literature for organics, hardness, and silica in oil and gas PW applications.

Pretreatment Unit[a]	Feed TDS, mg/L	Organics	Hardness	Silica	References[b]
Chemical Softening	11000	NR	96 %	NR	[43]
	237,680	NR	99.5 %	NR	[69]
	41,420	39 %	50 %	32 %	[70]
Chemical Coagulation	1050	42 %	6.4 %	NR	[71]
	200,000	35 %	NR	NR	[41]
Electrocoagulation	47,500	91 %	NR	NR	[72]
	50,650	NR	70 %	99 %	[42]
	1500	67 %	86 %	NR	[73]
Granular Activated Carbon	129,300	50 %	-	-	[74]
	10,631	78.2 %			[75]
	16,716	92 %			[48]
	65,000	40 %			[47]

[3]NR = Not Reported

[a] The reported removal percentages are dependent on the type of water, and its respective water quality of the study, as well as operational goals, inputs, materials, and conditions established by the authors.

[b] Salinity and removal efficiencies reported in order of citation.

Table 2
Water quality characteristics of the Permian Basin PW.

Parameter	Permian basin PW			
	Minimum	Mean	Maximum	Standard Deviation
pH	3.9	6.6	8.1	±1.4
Temperature, °C	30	35	40	-
TDS, mg/L	100,830	128,645	201,475	±21,581.6
Conductivity, μS/cm[a]	201,660	257,290	402,950	-
Total Hardness, mg/L[b]	3417	12,621	28,198	-
Total Alkalinity, mg/L[b]	100	267	870	±187.4
Calcium, mg/L	880	3821	8186	±2246.1
Magnesium, mg/L	295	745	1877	±436.7
Sodium, mg/L	25,080	40,896	68,985	±7013.5
Chloride, mg/L	57,543	78,648	120,200	±13,578.6
Silica, mg/L	4	108	195	±47.3
Manganese, mg/L	0.01	1.24	1.49	±373.9
Sulfate, mg/L	151	496	965	±236.2
Turbidity, NTU	23	116	200	±82.6
TOC, mg/L	2.5	105	248	±73.6
Oil and Grease, mg/L[c]	47	104	273	±80.1

[d]Data collected from [10] based on 45 samples.
[a] Based on TDS measurements as sodium chloride (NaCl)
[b] Hardness was calculated based on calcium and magnesium ions present – units of measurement are in mg/L as $CaCO_3$
[c] Oil and grease from PW taken as the sum of present petroleum hydrocarbons (diesel, gasoline, motor oil)

in the Permian Basin [10] as it covers more than 75,000 square miles. The Permian Basin is comprised of several sub-basins and platforms with the three main subdivisions including the Delaware Basin, Central Basin Platform, and the Midland Basin [78,81]. Despite the variability of PW quality in the entire basin, this study used the average water quality for modeling simulation, assuming the PW treatment plants would be built in saltwater disposal facilities that receive PW with relatively consistent water quality.

The average Langelier Saturation Index of the Permian Basin PW was estimated to be 0.71 using AdvisorCi Online [31]. The potential precipitates (saturation indices > 0) that may affect the desalination processes for the Permian Basin PW were calculated with the PHREEQC software [30], as shown in Table 3. The scaling potential of the different minerals was also evaluated for an additional 45 % water recovery scenario of PW with a seawater RO unit modeled in the WAVE 1.83 Water Treatment Design Software (DuPont Water Solutions), at which the different minerals are further concentrated in the output brine of the system, hence making the PW continuously harder to desalinate for other treatments aiming to recover additional water/minerals. In addition, having poor disposal water quality can compromise the effective injectivity of the PW even in carbonate or sandstone formations, which can result in economic failures and costly workovers [82]. Scales and possible precipitates are of concern in the deep-well disposal of PW since they can clog the injection well, but other considerations in the disposal of the PW, such as the ionic composition, suspended solids, oil, and grease, hydrocarbons, incondensable gas, and bacterial content can also have a significant influence in effective disposal.

Table 3
Oversaturated chemical species present in the Permian Basin PW and their change when considering an additional 45 % water recovery.

Minerals – PW	Saturation Index	Saturation Index (Additional 45 % recovery)
Albite ($NaAlSi_3O_8$)	6.01	8.92
Alunite ($KAl_3(SO4)_2(OH)_6$)	4.97	3.94
Anorthite ($CaAl_2Si_2O_8$)	4.14	6.94
Aragonite ($CaCO_3$)	0.52	0.84
Barite ($BaSO_4$)	0.59	1.07
Ca-Montmorillonite ($Ca_{0.3}Al_2Si_4O_{10}(OH)_2$)	10.92	13.62
Calcite ($CaCO_3$)	0.68	1.01
Chalcedony (SiO_2)	1.09	1.74
Chlorite−14A ($Mg_5Al_2Si_3O_{10}(OH)_8$)	5.86	15.24
Dolomite ($CaMg(CO_3)_2$)	1.17	1.91
Ferric hydroxide ($Fe(OH)_3$)	1.68	2.91
Gibbsite ($AlOH_3$)	2.89	2.87
Goethite (FeOOH)	7.95	9.23
Hematite (Fe_2O_3)	17.99	20.59
Hydroxyapatite ($Ca_5(PO_4)_3OH$)	7.69	13.82
Illite ($K_{0.6}Mg_{0.25}Al_{2.3}Si_{3.5}O_{10}(OH)_2$)	10.75	13.88
Jarosite-K ($KFe_3(SO_4)_2(OH)_6$)	0.65	3.39
K-feldspar ($KAlSi_3O_8$)	6.39	9.07
K-mica ($KAl_3Si_3O_{10}(OH)_2$)	17.92	20.65
Kaolinite ($Al_2Si_2O_5(OH)_4$)	9.65	10.97
Silica quartz (SiO_2)	1.48	2.14
Talc ($Mg_3Si_4O_{10}(OH)_2$)	4.34	11.41
Vivianite ($Fe_3(PO_4)_2{\cdot}8\ H_2O$)	0.94	2.86

3.2. WaterTAP & pretreatment unit design

Pretreatment units were simulated through the open-source Python based WaterTAP program [28]. The unit models for CC and adsorption through GAC are readily available in the WaterTAP open-source library along with the EC and CS with hydrated lime ($Ca(OH)_2$) and soda ash (Na_2CO_3) models, both developed by the research team at New Mexico State University (NMSU) in collaboration with WaterTAP developers. The CS, CC, and EC models are deemed as assumed performance models where the removal efficiency of target pollutants is based on reported efficiencies in the literature [42,69,71]. Sludge management costs were included in this study, accounting for processes such as thickening and dewatering and disposed waste was assumed to be hazardous due to the presence of radionuclides in the PW. The cost for hazardous waste disposal is state- and site-specific, and at the federal level, the oil field wastes are covered by the exemption from the hazardous waste provisions of the Resource Conservation and Recovery Act (RCRA). This simplifies the disposal of oil field waste and allows for reduced disposal costs [83]. Typical hazardous waste disposal costs can range from $10 to $500 per metric ton of waste, in the current study, a total disposal cost of $100 per metric ton of waste ($0.11/kg) was assumed for off-site commercial disposal of naturally occurring radioactive material inclusive of handling and off-site transportation fees [83–86]. The use of a dissolved air flotation (DAF) for the removal of oil, grease, and total suspended solids (TSS) in the input water was accounted for in the costs of all the units as a preliminary treatment before the pretreatment units. The key inputs of the CC, EC, and GAC models are presented in Table 4.

Effluent water quality for the CS model chemical dosing assumes that

Table 4
WaterTAP model inputs.

Design Input	Value
Coagulant dose $Al_2(SO_4)_3$[a]	600 mg/L [71]
Coagulant dose $FeCl_3$[a]	900 mg/L [71]
Polymer dose[a]	5 mg/L [41]
Current density[b]	350 Amperes per m[b] [87]
Electrode gap[b]	0.02 m [74]
Current efficiency[b]	100 %
Retention time – Flocculation basin[b]	15 minutes [74]
Fe^{2+} electrode dose[b]	350 mg/L [88]
Al^{3+} electrode dose[b]	135 mg/L [89]
Freundlich isotherm k parameter[c]	0.269 [90]
Freundlich isotherm 1/n parameter[c]	0.5134[90]
Empty bed contact time (EBCT)[c]	600 s [91]
Superficial velocity[c]	13 m/hr [91]
GAC particle porosity[c]	0.5 [91]
GAC bulk density[c]	400 kg/m[c] [91]
GAC bed void[c]	0.5 [91]
GAC particle diameter[c]	0.001 m [91]

[a] Chemical coagulation
[b] Electrocoagulation
[c] Granular activated carbon

both Ca^{2+} and Mg^{2+} are lowered to their practical solubility limit levels of 30 mg/L as $CaCO_3$ and 20 mg/L as $CaCO_3$ [91]. The CS chemical costs adapted in this study were \$0.24/kg of 95 % $Ca(OH)_2$, \$0.28/kg of 98 % Na_2CO_3, and \$0.59/kg of 50 % NaOH. For CC the costs of the coagulants were \$0.88/kg $FeCl_3$ and \$0.69/kg $Al_2(SO_4)_3$ [88]. The electrode costs for the EC unit were adapted from the literature as \$2.23/kg Al and \$3.41/kg Fe [92]. For GAC, this study includes the cost of regeneration (\$4.28/kg of regenerated GAC) and replacement/makeup of the media (\$4.58/kg of new GAC). The GAC WaterTAP model assumes a 70 % regeneration rate for spent media, with 30 % designated for disposal, a fixed parameter from the model that was retained in this study due to limited data on GAC regeneration/replacement rates in PW treatment. Descriptions of costs considered in all pretreatment units are further discussed in Supporting Information (SI).

3.3. Economic, energetic, and environmental assessment

Providing an economic, environmental, and energy evaluation can aid decision-making when considering the system design and optimization of large-scale applications. It is essential to identify key factors of the different pretreatment technologies and their operation. This modeling study aims to evaluate pretreatment technologies for the effective reduction of the fouling and scaling potential in PW applications with a focus on (i) levelized cost of water (LCOW), (ii) specific energy consumption (SEC), (iii) cumulative energy demand (CED), and (iv) carbon emissions as carbon dioxide (CO_2), for treatment of one m^3 of PW. The LCOW consists of units of 2024 U.S. dollars (\$) per m^3 (\$/m^3) to account for recent inflation trends, considering both capital and operational expenses of the technologies, a lifetime of 30 years, and a 5 % interest rate. In the case of energy consumption, the specific energy consumption (SEC) is used as the energy indicator, in kilowatt-hours (kWh), for the treatment of one m^3 of PW (kWh/m^3) of a specific treatment unit, while the cumulative energy demand (CED) focuses on a systems total energy demand accounting for the manufacturing of replaced chemicals and materials [93]. CO_2 emissions throughout the operation and manufacturing/production of the different chemicals and materials used in the pretreatment alternatives were also accounted for in this assessment due to the increasing interest in the decarbonization of the water sector [94–98]. The unit for evaluation for CO_2 emissions is kilograms of $CO_{2\text{-eq}}$ for the treatment of one m^3 of PW (kg $CO_{2\text{-eq}}$/m^3). Chemical/waste transportation energy and emissions were excluded from this evaluation since there is limited information for accurate estimates. The impact of the construction phase of the different units was also excluded since the operational phase impact is larger than that of the construction phase [99,100]. The SI provides a summary of the data and assumptions relating to costs, energy, and emission factors accounted for in this study; their adaptation was based on an extensive search for reported literature values.

4. Results and discussion

WaterTAP was used to model the different pretreatment units of interest in this analysis. Design aspects of the different units are shown in SI, with most design values adapted from literature data and experimental work accounting for a similar PW quality. The design flowrate to evaluate all the pretreatment units was selected as 11,356 m^3/d (71,430 barrels/day, or 3 million gallons per day (MGD)). The design flowrate was selected considering that the production rate of PW at the Permian basin can range from 11 million barrels/day (242 MGD) to 15 million barrels/day (330 MGD) [101,102]. The selection of the 3 MGD flow represents ~1 % of the total daily flow generated in the Permian basin for PW. CC will be focused on the use of both ferric chloride ($FeCl_3$) and alum ($Al_2(SO_4)_3{\cdot}14\ H_2O$) in combination with a polymer-based coagulant aid [41], while CS will consider the use of both $Ca(OH)_2$ and Na_2CO_3, as well as a separate CS application focused on caustic soda (NaOH).

4.1. Evaluation of individual pretreatments

Figs. 2 and 3 show the LCOW and CO_2 emissions, and SEC of the different individual pretreatment units considered in this study, respectively. Table 5 summarizes the CAPEX and OPEX values of individual pretreatment technologies in PW. The cost breakdown of the individual components contributing to the CAPEX and OPEX of the pretreatment units is shown in the SI – Figures S1 to S18.

The CS application has the highest LCOW due to the high chemical input required to mitigate the hardness and SiO_2 content through both $Ca(OH)_2$ and Na_2CO_3, and with NaOH. There have been applications and investigations of chemical softening for PW in the literature [13, 103,104], all focused on either warm (30°C – 80°C) or hot (81°C – 125°C) softening applications with successful removal of both hardness and SiO_2 due to the accelerated chemical reactions at high temperatures. In this case study, the reported mean temperature of the PW was 35°C (Table 2), hence it is assumed that the softening application involves warm softening. There have been limited cost assessments for CS applications in PW treatment. A previous study for a warm softening pretreatment of PW, with NaOH as a substitute for lime, and the addition of magnesium chloride, reported a cost range of \$0.40 – \$0.72/m^3 (\$0.064 – \$0.115/barrel) and was heavily dependent on the chemical dosages and design capacity [104]. Another study estimated a cost of \$158/m^3 (\$25.36/barrel) using both a surfactant and soda ash for enhanced oil recovery operations and minimization of hardness [105]. The calculated cost for CS with $Ca(OH)_2$ and Na_2CO_3 in this study (\$6.06/m^3) accounts for the addition of both $Ca(OH)_2$ and Na_2CO_3 and CO_2 for pH regulation. Na_2CO_3 requires the highest dosage of the chemicals to mitigate the non-carbonate hardness, accounting for 71 % of the total operational costs of the system, with sludge disposal (14.4 %) and the lime chemical purchase (10 %) the second and third highest, respectively. The high dosage requirements of $Ca(OH)_2$ and Na_2CO_3 chemicals led to a high CED of 42.9 kWh/m^3, based on the production of the high quantity of chemicals required. Subsequently, this high chemical demand also leads to CO_2 emissions up to 6.36 kg $CO_{2\text{-eq}}$ /m^3. The CS with NaOH was demonstrated to be the highest of the pretreatment alternatives (\$12.09/m^3) since it required a higher dosing requirement and had a higher cost per mass of softening chemical (\$0.59/kg of 50 % bulk NaOH), making it cost-prohibitive for PW applications. The highest cost driver in the LCOW for the CS with NaOH was the chemical purchasing cost, accounting for 57.3 % of the total operating cost. Since only NaOH is being dosed, the CED (26.7 kWh/m^3) and CO_2 emissions (8.12 kg $CO_{2\text{-eq}}$ /m^3) are lower due to less demand in the production of chemicals, in comparison to CS with both $Ca(OH)_2$ and Na_2CO_3.

Granular activated carbon (GAC) had one of the highest LCOWs (\$5.01/m^3), mainly driven by media regeneration (68.6 %) and replacement (31.4 %) costs. Frequent regeneration due to high organic content in PW led to high energy demand (21.5 kWh/m^3) and CO_2 emissions (9.24 kg CO_2-eq/m^3). Prior studies reported lower GAC treatment costs (\$3.0/m^3), likely due to differences in PW composition [106]. It is important to note that the WaterTAP GAC model only supports the adsorption of a single solute species while all others are considered inert, which is not normally the case in real applications of GAC in PW applications. The different volatile organics (e.g., benzene, toluene, ethylbenzene, and xylene) present in the PW may affect the replacement rate of the GAC media as well as the removal rate of the organics present. The solute of focus selected in the WaterTAP model was TOC, as it serves as an indicator of total organics in the PW (additional information on SI). The WaterTAP model predicted to remove 67 % of TOC present in the input water, yet the GAC unit design and operation may need to consider the multiple solutes present in PW and experimental data to effectively validate the WaterTAP model simulation. Additionally, TSS in PW may necessitate pre-filtration (e.g., DAF) to prevent media clogging. The use of EC as a standalone treatment approach is very promising, and this has also been demonstrated in PW

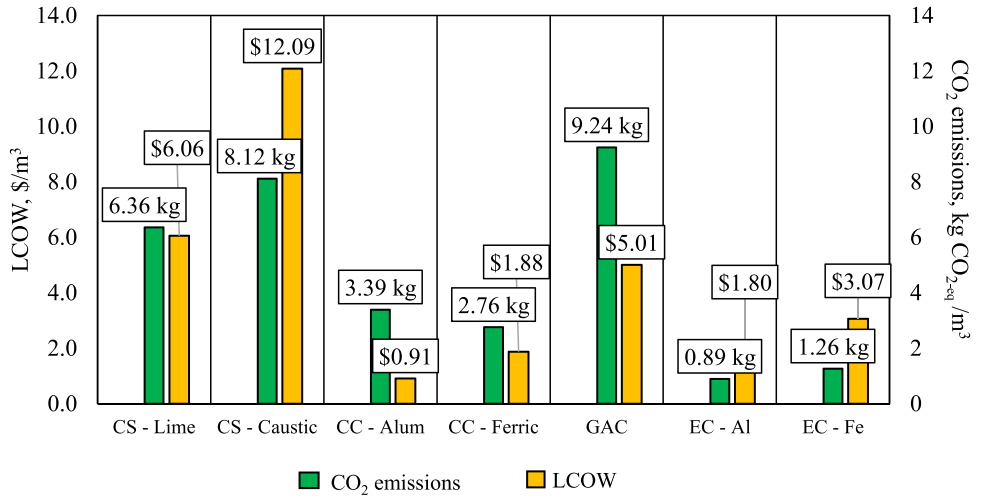

Fig. 2. LCOW and CO_2 emissions for the different individual pretreatment units for treating PW from the Permian Basin.

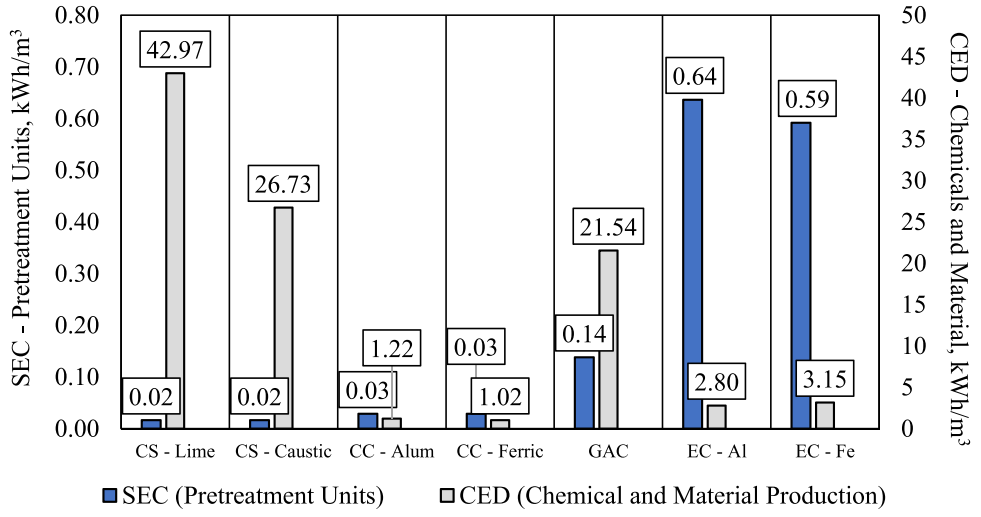

Fig. 3. The specific energy consumption of pretreatment units and the cumulative energy demand of the pretreatment units with chemical/material energy for treating PW from the Permian Basin.

applications [73,107,108]. The EC dosage requirement was selected through the literature [74,107], while the reactor material was assumed polyvinyl chloride (PVC) to avoid issues with corrosion [109]. The EC pretreatment LCOW for both Al and Fe electrodes falls in the range of \$0.05 - \$6.3/m^3, the reported operational cost for wastewater, mining, and oil and gas applications [74,107,108,110–113]. Waste disposal (60 % for Al, 30 % for Fe) and electrode replacement (33 % for Al, 66 % for Fe) were the major OPEX drivers. The CO_2 emissions accounted for in both EC systems consist of electrical production and electrode material production emissions. CO_2 emissions for both applications differ by 30 %. For the CED considering the production of the electrode material of choice and the units energy requirement, the EC-Al pretreatment demonstrated to be the lowest in comparison with the EC-Fe pretreatment since the energy required to produce Fe is lower than Al. As for the SEC, both the Al (0.64 kWh/m^3) and the Fe (0.59 kWh/m^3) are the highest SEC of the pretreatment units. Both SEC values fall in the range of 0.1–25 kWh/m^3 reported in the literature for wastewater, mining, and oil and gas production [74,107,114–117]. Very few studies have accounted for the overall energy consumption considering the production of the electrode material of choice in an EC system [107]. Compared to CC, EC avoids high coagulant consumption but requires careful design considerations (e.g., current density, electrode gap, electrical conductivity) to optimize performance. The CC pretreatment was focused on the effective removal of both organics and SiO_2 in the PW. With the increase of organic content entering the system, a higher chemical dosage of both $Al_2(SO_4)_3$ and $FeCl_3$ is often required [71]. The LCOW for the CC with $Al_2(SO_4)_3$ (\$0.91/m^3) is lower than CC with $FeCl_3$ (\$1.88/m^3) due to the difference in dosages as well as the cost of the chemical (\$0.69/kg of $Al_2(SO_4)_3$ vs \$0.88/kg of $FeCl_3$). The two main drivers in OPEX of the CC pretreatment are the chemical purchasing costs, with 83 % of the total for $Al_2(SO_4)_3$ and 78 % for $FeCl_3$. The CED of both CC approaches differ slightly due to the different production energy demands that each chemical has 1.2 kWh/m^3 for $Al_2(SO_4)_3$ vs 1.0 kWh/m^3 for $FeCl_3$. CO_2 emissions show the same trend, as the production of the $FeCl_3$ coagulant has less emission per kg of chemical product than $Al_2(SO_4)_3$, this is attributed to selected emission factors (presented in SI) and differences in the production processes between the two coagulants.

4.2. *Evaluation of coupled/combined pretreatment processes*

In addition to individual pretreatment units, an evaluation of different pretreatment combinations was performed to reduce all three pollutants of focus in the PW, i.e., organics, hardness, and SiO_2, to

Table 5
CAPEX and OPEX values for 11,356 m^3/d individual pretreatments in PW in 2024 U.S. dollars.

Pretreatment Unit	CAPEX, $M	OPEX, $M/year	Highest contributor in the CAPEX	Highest contributor in the OPEX
CS – Lime	$3.65 M	$24.87 M/yr	Sedimentation basin (23 %)	Na_2CO_3 dosing (71 %)
CS – Caustic	$10.02 M	$49.43 M/yr	NaOH feed system (71 %)	NaOH dosing (57 %)
CC – Alum	$8.30 M	$3.22 M/yr	-	$Al_2(SO_4)_3$ dosing (83 %)
CC – Ferric	$8.93 M	$7.19 M/yr	-	$FeCl_3$ dosing (78 %)
GAC	$2.11 M	$20.61 M/yr	GAC Contactor (42 %)	Regeneration of spent GAC (68 %)
EC – Al	$3.95 M	$7.22 M/yr	Sludge management units (77 %)	Waste disposal (59 %)
EC – Fe	$4.35 M	$12.42 M/yr	Sludge management units (77 %)	Electrode replacement (66 %)

Note: CAPEX and OPEX account for the costs of sludge management units, gravity thickener, and filter press. CAPEX values account for the application of the default WaterTAP cost factors. The highest contributor of CAPEX for CC pretreatment is not specified in the zero-order WaterTAP model. The cost of the DAF is accounted for in the CAPEX.

effectively minimize fouling and scaling in downstream processes through interconnections between the units. Each of the proposed couplings was evaluated in terms of costs, energy, and emissions impact based on the previous individual unit evaluations. Developing pretreatment combinations is essential to expanding the scope of removal for the different fouling and scaling pollutants in the PW. The use of CC with $FeCl_3$ and EC with Al electrodes was considered in this section due to their good overall results in the individual pretreatment evaluation, as in the case of CS the consideration of $Ca(OH)_2$ and Na_2CO_3 is accounted for instead of NaOH due to its lower cost. The incorporation of GAC aims to minimize waste disposal, especially the use of CC, CS, and EC in PW applications may generate different amounts of solids waste that may be classified as hazardous, such as precipitation of radioactive materials. The combinations of pretreatment options considered for each case study along with the selected target removal rates for effective desalination applications are shown in Fig. 4.

The coupling of chemical softening with granular activated carbon (CS+GAC) has not been documented in PW treatment applications, but the goal of this combination is for the CS unit to remove upwards of 90 % and 75 % of the hardness and silica, respectively. The application of CS before the GAC aims at removing the incoming hardness and SiO_2 and avoiding any possible constituents clogging the media that may affect the adsorption capacity of the GAC media. The use of GAC aims at removing the incoming organics. Treatment combinations of electrocoagulation coupled with chemical coagulation (EC+CC) and with granular activated carbon (EC+GAC) for different types of waters have been reported in the literature [118–120]. The goal of the EC+CC and EC+GAC coupling aims at EC removing the incoming SiO_2 and hardness while CC and GAC tackle the organics. The benefit of the CS+GAC and EC+GAC pretreatment combinations is less PW waste disposal (which accounts for precipitates and removed target pollutants) in comparison to the EC+CC pretreatment application since it would generate less precipitates to dispose of. The scaling precursors of focus for the combinations will be organics, SiO_2, and hardness. Figs. 5 and 6 show the modeling results for different pretreatment combinations for the PW from the Permian Basin. Table 6 presents the CAPEX and OPEX values of the combined pretreatment technologies in PW.

The highest LCOW ($11.09/$m^3$) in this study was observed for the CS + GAC combination, primarily due to high chemical dosages for CS (36 % of OPEX) and frequent GAC regeneration (27 % of OPEX). While CS effectively removes hardness and SiO_2 before GAC adsorption, its high energy demand (64.08 kWh/m^3) and CO_2 emissions (16.19 kg CO_2-eq/m^3) make it the least cost-effective option. The main contributors to these emissions are GAC regeneration (60 %) and soda ash production (54 %).

The coupling of EC+GAC has been documented extensively in the literature for different types of industrial wastewater [121–126] and also PW treatment [118,119]. Placing EC before GAC, preventing organic overloading and reducing adsorbent saturation [122,127]. The use of the EC, assuming a TOC removal efficiency of 40 % for EC with Al electrodes [42,74], can reduce the GAC replacement rate by approximately 30 %. The LCOW of the coupling of EC+GAC is shown to be the second highest of the pretreatment unit couplings in this case study at $4.42/$m^3$. It has the second highest overall total CED (22.97 kWh/m^3) and the second-highest CO_2 emissions (9.43 kg $CO_{2\text{-eq}}$ /m^3) compared to the other coupled units. Most of the costs emerging from this coupling are due to the OPEX contribution from the regeneration of the spent GAC (38 %) as well as the final disposal of the waste (34 %). In the case of the CAPEX, the major cost contributor is the waste management units (58 %).

In contrast, EC + CC, though less studied for PW, has shown high removal efficiency for hardness, silica, organics, and heavy metals [128–131].This combination had the lowest LCOW ($3.60/$m^3$), lowest CO_2 emissions (2.66 kg CO_2-eq/m^3), and lower energy demand (4.36 kWh/m^3), making it a cost-effective alternative. However, it generates more sludge (56 % of OPEX), increasing disposal costs. Ultimately, the selection of pretreatment depends on balancing cost, energy use, waste disposal, and target pollutant removal. The EC + CC coupling enhances hardness, SiO_2, and organics removal, optimizing coagulant use and reducing chemical costs. However, it generates higher sludge volumes, increasing disposal costs. EC + GAC, while reducing sludge, has higher energy and regeneration costs due to frequent adsorbent replacement. CS + GAC effectively removes hardness and organics, minimizing membrane fouling but incurs high chemical demand and regeneration expenses, making it the costliest option. Selection depends on balancing removal efficiency, waste management, and cost feasibility.

4.3. *Influence of water quality on selection of pretreatment*

The consideration of the constituents of the source water is a key

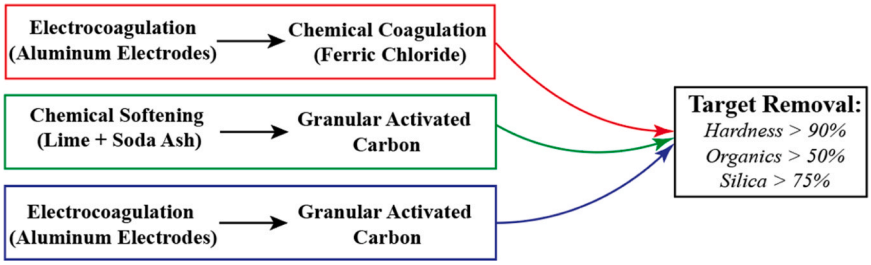

Fig. 4. Pretreatment combinations for PW considered in this section along with the maximum removal efficiency.

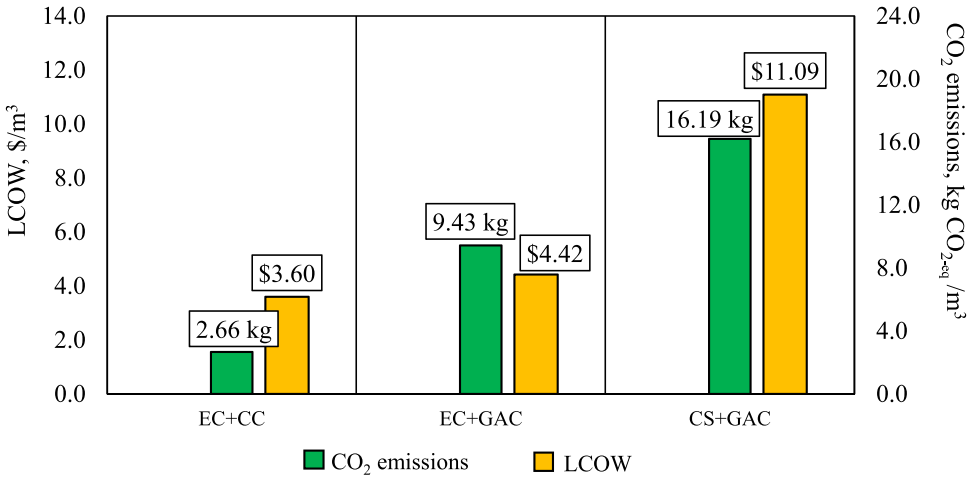

Fig. 5. LCOW and CO_2 emissions for coupled pretreatment units for the PW in the Permian Basin.

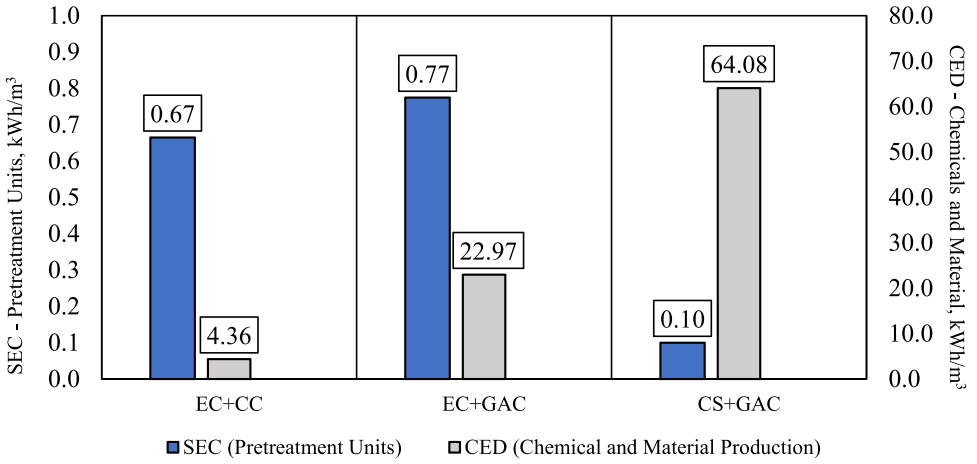

Fig. 6. The specific energy consumption of pretreatment units and the cumulative energy demand of the coupled pretreatment units with chemical/material energy for treating PW from the Permian Basin.

Table 6
CAPEX and OPEX values for 11,356 m^3/d pretreatment couplings for PW and scaled to 2024 U.S. dollars.

Pretreatment Coupling	CAPEX, $M	OPEX, $M/year	Highest contributor in the CAPEX	Highest contributor in the OPEX
EC+CC	$9.90 M	$14.26 M/yr	Coagulation system (74 %)	Waste disposal (56 %)
EC+GAC	$4.07 M	$18.05 M/yr	Waste management units (58 %)	Regeneration of spent GAC (38 %)
CS+GAC	$7.21 M	$45.48 M/yr	Sedimentation basin (19 %)	Na_2CO_3 dosing (36 %)

Note: CAPEX and OPEX account for the costs of sludge management units, gravity thickener and filter press. CAPEX values account for the application of the default WaterTAP cost factors. The cost of the DAF is accounted for in the CAPEX.

design factor on all the pretreatment units in this work, and it has been demonstrated that incoming concentration of target pollutants have a significant influence on the evaluation metrics when considering a lower salinity stream such as reverse osmosis concentrate (ROC) in comparison to PW. For instance, pretreatment PW likely necessitates robust techniques to manage high salinity and potentially complex organic compounds, compared to ROC which might only require minimal intervention compared to PW. This leads to a careful selection of pretreatment technologies based on the water quality profile, since it can have a significant impact on rejection efficiency, operational cost, and environmental footprint. By tailoring pretreatment to the source water, treatment efficacy can be optimized while ensuring cost-effectiveness and minimizing environmental impact. PW presents a unique challenge due to its complex and variable composition. This complexity arises from a cocktail of contaminants, including hydrocarbons, dissolved salts, heavy metals, and NORM. These factors significantly increase the complexity of treating produced water compared to lower-salinity streams like ROC. As a result, pretreatment for PW often involves a multi-stage approach, will often require various techniques to address the diverse range of contaminants.

The evaluation of coupled pretreatment processes underscores the critical link between upstream removal targets and downstream treatment costs. As demonstrated by the varying performance and associated expenses of the EC+CC, EC+GAC, and CS+GAC combinations, the level of pollutant removal achieved in the pretreatment stage directly impacts the demands and consequently the costs of subsequent desalination processes. For instance, achieving higher removal rates of scaling precursors like hardness and silica through CS aims to minimize scaling in downstream reverse osmosis membranes, potentially reducing cleaning frequency and extending membrane lifespan. However, aggressively pursuing very high removal efficiencies (e.g., >90 % hardness and 75 % silica) can lead to substantial increases in operational expenditure due to

high chemical consumption and frequent regeneration cycles. Therefore, defining appropriate removal targets in the pretreatment phase requires a careful economic assessment that balances the costs of achieving higher removal with the potential savings in downstream operations.

Furthermore, the necessity of achieving higher removal rates, potentially exceeding 70 % for certain pollutants, is heavily contingent upon site-specific PW characteristics. In scenarios where the initial concentrations of the fouling and scaling agents are exceptionally high, or where downstream treatment technologies have stringent influent water quality requirements, achieving removal rates significantly above 70 % may become indispensable. Neglecting to do so could lead to rapid fouling, increased energy consumption in downstream processes, and ultimately, a higher overall cost of treatment. Consequently, a thorough characterization of the PW at a specific site is paramount to determine the necessary pretreatment removal targets and to guide the selection of the most economically and environmentally sound pretreatment technologies.

5. Conclusion

This study provides a comprehensive framework for techno-economic-environmental assessment of high-salinity PW pretreatment, evaluating costs (LCOW), energy (SEC|CED), and carbon emissions (CO_2) across treatment infrastructure, operation, and waste management. The findings highlight that pretreatment selection is a key determinant of desalination efficiency and cost-effectiveness, yet it remains an underexplored area in PW treatment. Electrocoagulation (EC) and chemical coagulation (CC) offer cost-effective and lower-energy alternatives, while granular activated carbon (GAC) minimizes waste disposal but requires frequent regeneration, increasing costs. For the Permian Basin PW case study, EC + GAC was found to be the most effective combination for removing hardness, silica, and organics, while also reducing sludge disposal challenges. However, this approach had the second-highest LCOW, SEC, and CO_2 emissions, indicating a trade-off between contaminant removal efficiency and economic feasibility. Despite these limitations, EC + GAC remains a promising pretreatment option, particularly for cases where waste minimization is a priority.

The results of this TEA underscore the importance of identifying key cost drivers and strategically targeting research and development efforts to enhance the economic viability of PW treatment. High-cost processes, such as energy-intensive treatment systems, present clear opportunities for innovation. To advance research in this area, future work should focus on optimizing pretreatment configurations to balance cost, energy consumption, and environmental impact. Future research that involves Monte Carlo simulations or scenario-based cost analyses could provide a more dynamic understanding of economic variability, accounting for fluctuations in water quality, electricity prices, and chemical demand. Additionally, pilot-scale testing and field studies should be prioritized to validate model predictions and improve the scalability of pretreatment strategies. Further exploration of hybrid pretreatment systems, alternative adsorbents, and renewable-powered treatment solutions will be essential to developing more sustainable and economically viable PW treatment technologies.

CRediT authorship contribution statement

Fthenakis Vasilis: Writing – review & editing, Investigation, Funding acquisition, Conceptualization. **Kuravi Sarada:** Writing – review & editing, Investigation, Funding acquisition, Conceptualization. **Lugo Abdiel:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Xu Pei:** Writing – review & editing, Writing – original draft, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Kurup Parthiv:** Writing – review & editing, Supervision, Investigation, Funding acquisition, Conceptualization. **Wang Huiyao:** Writing – review & editing, Validation, Project administration, Investigation, Funding acquisition, Conceptualization. **Kota Krishna:** Writing – review & editing, Investigation, Funding acquisition. **Sitterley Kurban:** Writing – review & editing, Validation, Software, Investigation, Formal analysis, Conceptualization. **Stoll Zachary:** Writing – review & editing, Validation, Investigation, Data curation. **Senanayake Punhasa S.:** Writing – review & editing, Validation, Investigation, Data curation. **Mejía-Saucedo Carolina:** Writing – review & editing, Validation, Investigation, Data curation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2025.117026.

Data availability

Data will be made available on request.

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