

Recent advances in industrial water reuse: techno-economic insights into selective reuse and brine segregation in the pulp and paper sector

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ABSTRACT

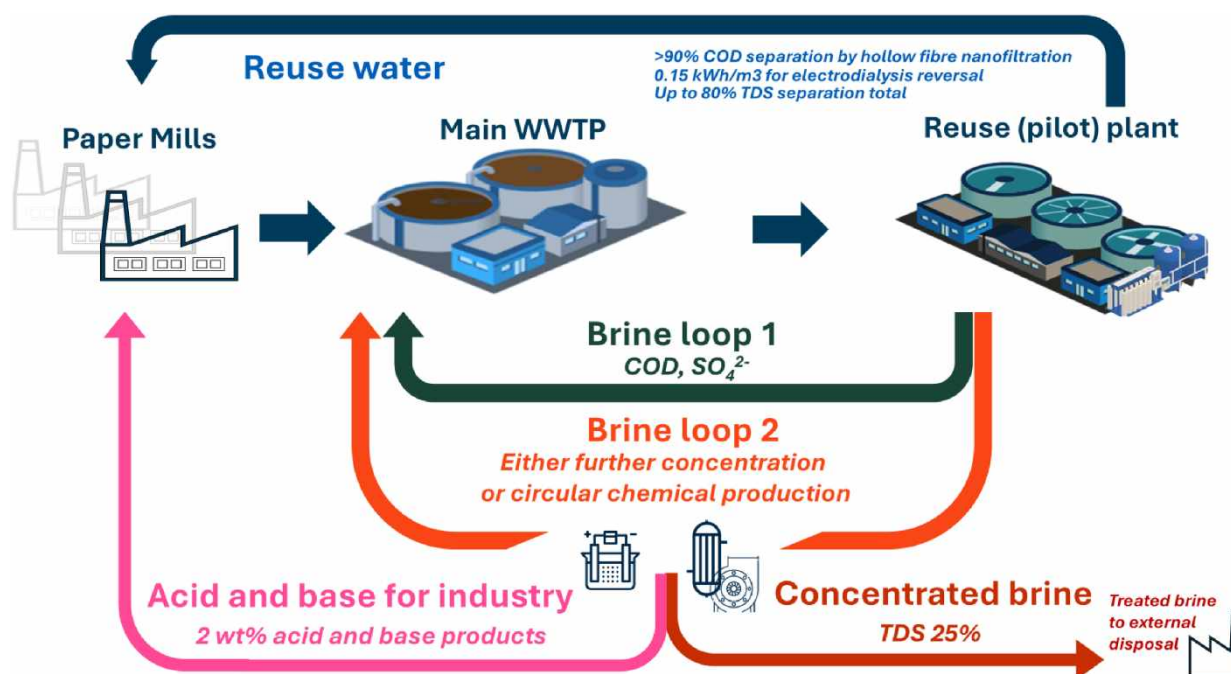
Growing pressure on freshwater resources in the Netherlands has increased the need for industrial water reuse, particularly in the water-intensive pulp and paper sector. Conventional end-of-pipe systems based on ultrafiltration (UF) and reverse osmosis (RO) are reliable but remain sensitive to fouling, scaling and variable intake quality. This study evaluates the techno-economic feasibility of a more selective and flexible treatment train. A pilot system combining hollow fibre nanofiltration (HFNF), granular activated carbon (GAC) polishing and electrodialysis reversal (EDR) was operated for 180 days on wastewater from the Industriewater Eerbeek Industrial Wastewater Treatment Plant (IWE WWTP). High removal of colour, chemical oxygen demand (COD) and sulphate was observed at the HFNF system, whilst the EDR showed great flexibility in desalination. Reuse water met all paper mill specifications at the 99th percentile. Selective treatment enabled segregation of organic-rich HFNF concentrate and salt-rich EDR brine. The HFNF concentrate was treated by coagulation and flocculation and recirculated over the WWTP, while the EDR brine was further concentrated and allowed for more research on brine management. Techno-economic comparison showed a total expenditure (TOTEX) of 1.85 EUR/m³ for the innovative system versus 2.14 EUR/m³ for the conventional system. These findings demonstrate a cost-effective pathway toward effective water reuse in end-of-pipe configurations.

Key words: brine management, electrodialysis reversal (EDR), hollow fibre nanofiltration (HFNF), pulp and paper industry, water reuse

HIGHLIGHTS

- First large-scale pilot in the Netherlands proves the reuse of treated paper mill effluent to cut groundwater use.
- Combines hollow fibre nanofiltration, granular activated carbon and electrodialysis reversal to meet strict water quality standards.
- Maintains 99th percentile compliance under variable conditions.
- Segregates brines for tailored treatment enabling minimal liquid discharge.
- Explores multiple brine management options for side streams of water reuse and identifies challenges and the most promising routes.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Potable freshwater availability for drinking water production is increasingly under pressure in the Netherlands due to a growing economy, a rising population and the impacts of climate change. The Dutch National Institute for Public Health and Environment (Rijksinstituut voor Volksgezondheid en Milieu, RIVM) indicates that there is no certainty that the supply of both groundwater and surface water will suffice for the supply of drinking water to the Dutch population in 2030 (van Leerdam *et al.* 2023).

With this increasing pressure on the availability of (fresh)water resources in the Netherlands, provinces such as the Province of Gelderland, are actively assessing options to mitigate this pressure. The paper and pulp industry is marked as a major extractor of fresh- and groundwater resources, consuming between 369 and 501 m³/tonne of paper produced (varying from newsprint, to printing and writing paper as well as paper board) (van Oel *et al.* 2009), due to the water-heavy processes from raw material to paper (Bajpai 2015). To alleviate pressure on the Dutch water footprint, water reuse incentives are of interest to the Province of Gelderland (the Netherlands).

In the context of this study, paper mills in the Eerbeek-Loenen (Gelderland, the Netherlands) surroundings are taking in about 3.6 million m³ of groundwater every year (Pilot Water Roundabout Eerbeek: reducing groundwater 2023). After in-factory utilisation and internal recirculation of this water, the process water is sent to a dedicated wastewater treatment plant (WWTP), which was erected in 1960 to treat the wastewater coming from all the paper mills in this area. In order to alleviate the local water footprint of the paper and pulp industry in this area, an incentive was created by the paper mills and Industriewater Eerbeek to set up a pilot test for efficient end-of-pipe WWTP effluent water reuse.

This paper focuses on the techno-economic evaluation of an end-of-pipe wastewater reuse strategy for the pulp and paper sector, emphasising both treatment performance and brine management. As freshwater scarcity increases in the Netherlands, cost-effective and robust reuse of WWTP effluent has become essential. Conventional ultrafiltration (UF), followed by reverse osmosis (RO), is widely applied for industrial reuse, but these systems operate in dead-end mode, which makes them sensitive to fouling, scaling and fluctuations in intake quality. Their limited operational flexibility and reliance on intensive chemical cleaning can restrict long-term performance under variable loading conditions, which are frequently shifting in variability of paper mill production schedules.

To address these limitations, the participating industries and the IWE WWTP sought a reuse configuration capable of handling dynamic organic and saline loads. An alternative treatment train was therefore designed, replacing UF with hollow fibre nanofiltration (HFNF) and RO with electrodialysis reversal (EDR).

The resulting innovative pilot system integrates selective HFNF pretreatment with flexible EDR desalination and targeted brine management. This combination aims to maximise recovery, stabilise performance under fluctuating influent conditions and create brine streams suitable for downstream treatment or valorisation. The pilot was operated with real industrial wastewater and ran for a prolonged period of 180 days. The work presented here evaluates the technological feasibility and economic implications of this integrated concept, representing, to our knowledge, one of the first demonstrations of such a configuration at this scale in the Netherlands and internationally. The paper aims to illustrate the use of brine loop segregation and the benefits of the innovative system setup.

2. MATERIALS AND METHODS

2.1. Existing upstream treatment system

The total wastewater feed flow to the IWE WWTP ranged from 4,070,000 m³/year in 2020 to 4,120,000 m³/year in 2021. The average daily flow between 2020 and 2021 was 11,205 m³/d (24 hours per day).

Wastewater coming from the paper mills was generally high in turbidity, organic matter, colour, calcium and sulphate, but loading varies periodically based on production schemes of the various paper mills (Pokhrel & Viraraghavan 2004; Patel *et al.* 2021a, b; Wang *et al.* 2021). The IWE WWTP is equipped to treat the wastewater to discharge into the IJssel river. The existing WWTP incorporates primary settling, anaerobic treatment, biogas desulphurisation and aerobic treatment with secondary clarification, followed by softening and 20-µm filtration (Janssen *et al.* 2009).

2.2. Conventional reuse treatment system

The UF-RO system is portrayed in the introduction as the conventional system choice for end-of-pipe water reuse. The UF unit removes suspended solids, colloids and pathogens from biologically treated effluent and operates through filtration cycles with hydraulic and chemically enhanced backwashing to control fouling. Coagulant dosing is often applied to stabilise flux when influent quality varies. The permeate from UF is directed to the RO system, while the UF concentrate containing retained solids and colloidal material is discharged without further recovery in the conventional setup.

The RO system consists of a three-stage spiral wound installation operated at recoveries of about 70–85%, which is typical for secondary effluent. Operating pressure is adjusted to the feed salinity, and periodic chemical cleaning is required to manage scaling and organic fouling. The process yields a single mixed brine stream with inorganic salts and residual organics and offers limited selectivity for downstream valorisation. Permeate quality is maintained through continuous monitoring of flux, pressure, conductivity and turbidity.

2.3. Reuse pilot plant setup

To separate the organic fraction from the wastewater, the pilot incorporated a HFNF step directly after softening and mesh filtration at the WWTP. HFNF was selected because its membrane geometry and chemistry allow more intensive hydraulic and chemical cleaning than spiral-wound nanofiltration, reducing fouling and supporting stable operation (Jonkers *et al.* 2023). The system operates in a feed and bleed crossflow mode in which recirculated concentrate controls recovery through a concentrate valve that adjusts pressure and transmembrane flux. This flexible regime limits fouling and allows for lower cleaning frequency and enables high-velocity chemical cleaning. HFNF therefore removes the organic fraction effectively while also reducing part of the hardness without requiring additional pretreatment.

A granular activated carbon (GAC) filter downstream of HFNF provided polishing to remove residual dissolved organics, particularly in the case of fibre damage. As the GAC effluent contains minimal organics and mostly inorganic salts, additional desalination is required to reach paper-grade reuse quality or meet MLD and ZLD objectives. Literature identifies electrodialysis (ED) and multi-stage RO as suitable technologies for high salinity papermaking effluents (Qiu *et al.* 2021).

To maximise the reuse of water production while meeting quality requirements, the pilot integrated EDR. HFNF already acts as an effective microbial barrier, eliminating the need for additional disinfection. The EDR unit also operates in feed and bleed mode, with a concentrate valve regulating hydraulic recovery and effluent conductivity determining the desalination setpoint. Current density automatically increases or decreases according to the incoming salt load, allowing substantial energy savings during low salinity periods.

ED has been shown to be more energy efficient than multi-stage RO for salinities up to about 3 g/L TDS and recoveries up to 73% (Patel *et al.* 2024). The brines treated in this study fall well below this threshold, making EDR favourable from an energy perspective. Although EDR typically has 20–30% higher capital expenditure than RO, its operational flexibility and ability to adjust power demand to changing influent conditions support its selection for the pilot.

Two recirculation loops were considered. In recirculation loop 1, HFNF concentrate underwent coagulation, flocculation and decanting before being recycled to the WWTP, which in full-scale configuration would eliminate HFNF concentrate discharge and increase total system recovery. In recirculation loop 2, EDR concentrate was softened using weak acid ion exchange and then concentrated in a single-stage RO system. The resulting RO permeate could be blended with EDR diluate, while the concentrated brine was evaluated for advanced brine management suitable for minimum liquid discharge (MLD) and zero liquid discharge (ZLD). Both EDR brine and RO brine were tested to compare physico-chemical softening, bipolar ED and thermal evaporation as further brine reduction and valorisation strategies.

The pilot plant system is shown in Figure 1. It shows the first and second stage HFNF system, the transfer tank, the coagulation flocculation and solids separation system, the GAC filter, the EDR system, the IEX system and the RO system.

All data used in this paper were collected between June 2023 and December 2023. Data as displayed in graphs is also from this period unless otherwise specified. Technological results were obtained by a mix of online monitoring and laboratory measurements. These results generate the foundation of the technological results and give input to the financial model, which is set up to assess costs for a full-scale system in the techno-economic comparison. All relevant analytical methods and equations are described in Appendix A of the Supplementary material.

3. RESULTS AND DISCUSSION

Pilot research was carried out from June to December 2023 at the IWE industrial WWTP. This section presents the key technological performance results of the innovative treatment train and compares them with the conventional UF RO

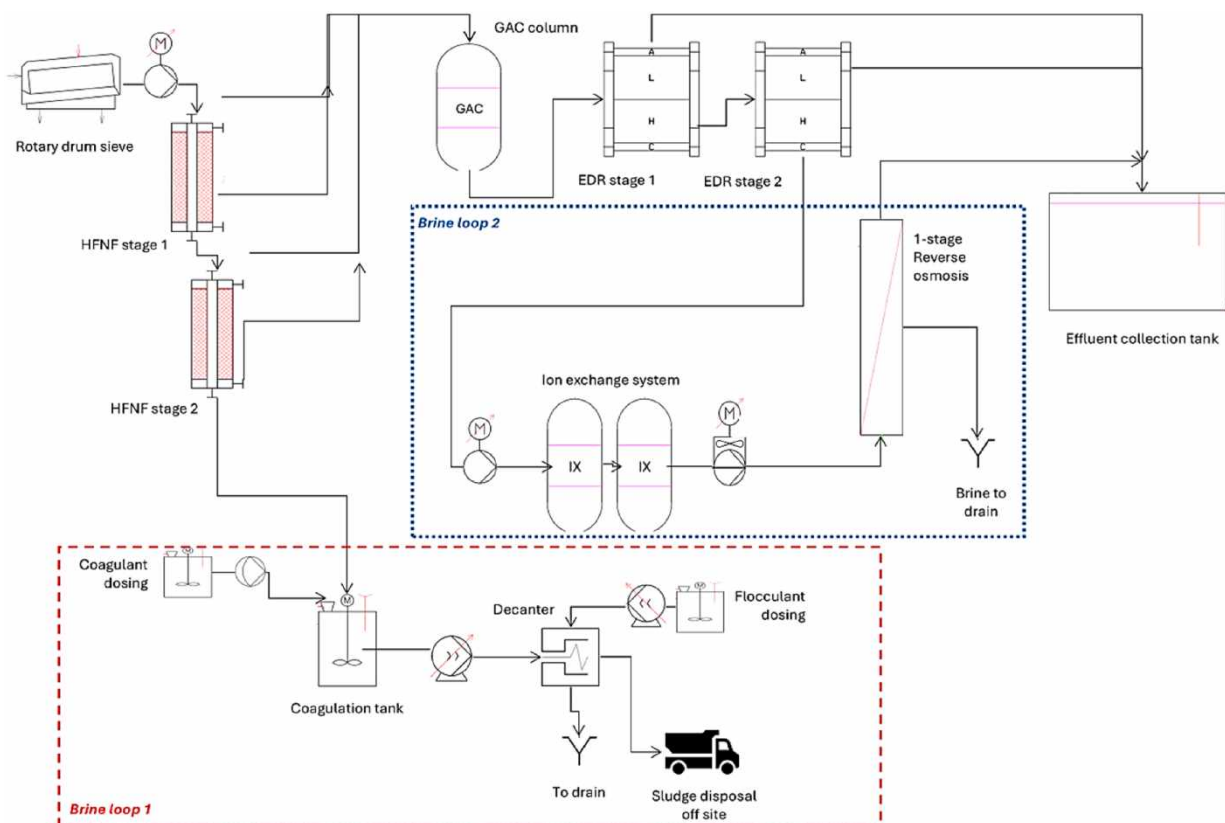


Figure 1 | Schematic setup of the IWE pilot installation. Brine loop 1 referring to the organic matter brine coming from the HFNF system and brine loop 2 is referring to the dissolved solids brine coming from the EDR system.

configuration. It then evaluates the produced water and brine quality, highlighting differences relevant to downstream brine valorisation. Finally, the techno-economic comparison of both treatment trains and the implications for brine management are discussed.

3.1. Treatment system choices and consequences compared with conventional treatment

3.1.1. Membrane pretreatment robustness and effectiveness

Conventional water reuse systems often gear towards the usage of metal-based coagulants dosed inline of the feed flow to allow for more stable operation and extended operational time between cleaning procedures (Stachurski *et al.* 2024). The HFNF consisted of two stages to further push the recovery of the total system whilst reducing fouling in the first stage. The HFNF system with these two stages operates at a high overall recovery. This is a consequence of filtration time, backwash interval, backwash duration and feed flux. With the choice of an HFNF system, no chemical dosing is required in the feed flow; simultaneously, the system is less prone to fouling due to sudden changes in water quality (Duin *et al.* 2000).

HFNF system retention was measured to be 99 ± 1 , 92 ± 4 , 62 ± 14 and $29 \pm 38\%$ for colour, COD, sulphate and calcium, respectively. The second stage shows comparable system retentions with 97 ± 1 , 95 ± 2 , 64 ± 14 and $52 \pm 40\%$, respectively, indicating near similar results except for calcium system retention in the second stage, with the crucial note that the standard deviation spread does not allow for quantifiable comparability of calcium retention between the first and second stages. Comparing this with removal rates of dead-end UF systems, research by Lidén & Persson (2015) indicated the removal rates for dead-end hollow fibre UF (with coagulant dosing) to be 55 ± 9.5 , 2.7 ± 2.3 , and 6.0 ± 5.3 for TOC, hardness and calcium, respectively, as listed in Table 1. This indicates that the choice for the HFNF system allows for significantly more COD and calcium removal, allowing more beneficial inorganic-focused brine treatment of the desalination system further downstream in the reuse plant.

While HFNF is less sensitive to sudden influent fluctuations due to its crossflow operation than UF, its higher selectivity for organics and divalent ions means that more material is intentionally retained at the membrane surface. This strengthens pretreatment performance but also leads to a different fouling profile than UF, requiring more structured cleaning to maintain permeability. The fouling layers caused the transmembrane pressure to increase with fouling building up over time, whilst the permeability decreased over time. Every filtration cycle would end with a hydraulic backwash, removing any built-up fouling, which could be removed hydraulically. A qualitative comparison of HFNF and dead-end UF is illustrated in Table 1. It is argued that fouling control and robustness during upsets are better in the HFNF system over the dead-end UF system. This is further quantified by data shown in Figure 2.

Figure 2 shows the effect of the sequential chemically enhanced backwash steps. The initial clean with 400 ppm citric acid at pH 6 restored 29.6% permeability. A subsequent 300 ppm NaOCl plus 1,100 ppm NaOH clean added 8.3%, while the following acid clean (1,700 ppm HCl with 180 ppm citric acid) produced no measurable improvement. A final alkaline-oxidative step restored an additional 25%. In total, the cycle yielded a 56.7% permeability recovery, with citric acid contributing most of the initial removal and the final caustic step removing remaining organic layers. Although such intensive cleaning is not standard practice, the sequence illustrates how multiple foulant layers can be effectively removed. The crossflow configuration of HFNF enhances this cleaning efficiency by providing higher shear forces than conventional dead-end UF.

Table 1 | Performance comparison of HFNF and dead-end UF for pulp and paper end-of-pipe water reuse

Parameter	HFNF	Dead-end UF ^a
Flux	25 LMH	60 LMH
Operation	Crossflow 0.3–0.4 m/s	Dead-end
COD removal	$92 \pm 4\%$	$55 \pm 9.5\%^b$
Calcium removal	$29 \pm 38\%$	$6.0 \pm 5.3\%^b$
Chemical dosing required	No	Yes (6 mg Fe3 + /L)
Robustness during upstream WWTP upsets	++	+ / –
Fouling control	+	+ / –

^aDead-end UF values based on third-party projections.

^bCOD and Ca²⁺ removal based on literature data Lidén & Persson (2015) with COD removal benchmark aligned with Sousa *et al.* (2018).

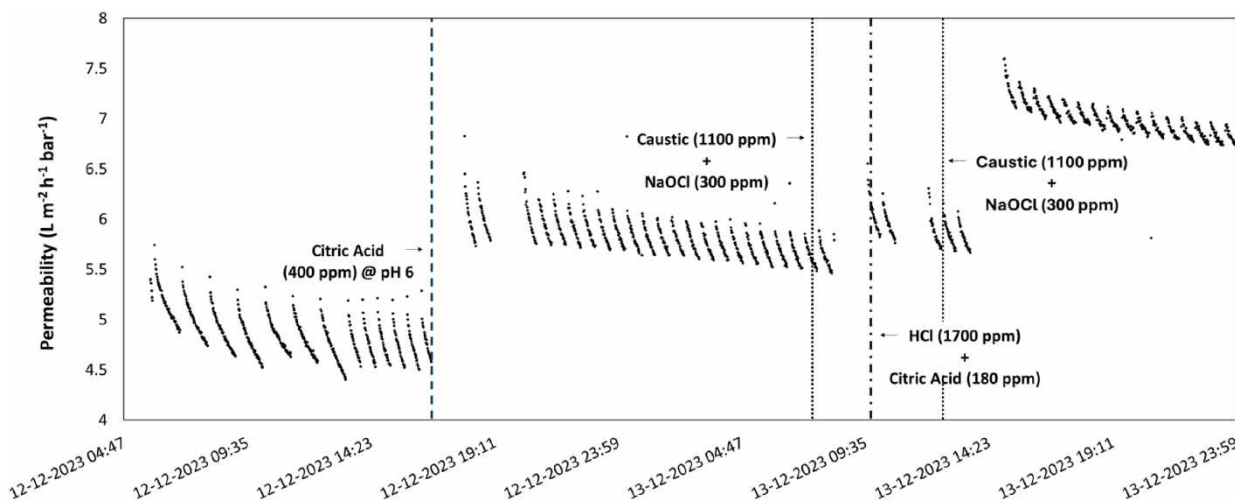


Figure 2 | Permeability of produced permeate over the first stage HFNF system and the effect of various cleaning regimes.

3.1.2. GAC filtration polishing filter

The GAC filter, positioned downstream of the HFNF system, was designed at an empty bed contact time between 7 and 9 minutes, which is shorter than the normally designed 10–20 minutes for drinking water and groundwater remediation (Jin *et al.* 2003). The filters' purpose in this study was to work as a polishing filter for residual organics and other <800 Da compounds in the HFNF permeate. The objective of this filter was to mitigate EDR fouling if HFNF fibre breakage would occur. The total bed volumes treated were calculated to be 6,478 throughout the pilot period with simultaneously no breakthrough measured based on outlet COD values, ensuring no fiber breakage in the HFNF system, which was confirmed during autopsy.

3.1.3. Desalination energy efficiency and selectivity

The EDR system, receiving the HFNF and GAC-treated water, is comprised of two stacks operated in series. This configuration allows for dividing the power consumption and energy distribution over two systems. In conventional RO, which relies on hydraulic pressure to drive ion separation, the energy demand can only be reduced to the point where the applied pressure approaches the osmotic pressure of the feedwater. Below this limit, further specific energy consumption (SEC) reduction is not possible (Wang *et al.* 2020). EDR, driven by convection, diffusion and ion migration in an electric field, allows the energy demand to adjust much more dynamically. As a result, the power input of the system can increase or decrease sharply depending on the required desalination setpoint. Patel *et al.* (2021a, b) described the SEC of EDR to increase in an exponential trend, with the gradient depending on the inlet TDS concentration and the total TDS retention (%). If only one stack were employed, this SEC would increase significantly compared with two stacks running at individually lower salt removal rates. The total energy consumption over the complete period ranged between 0.0 and 0.27 kWh/m³ permeate (between 0 and 1,400 W for stack 1 and between 0 and 638 W for stack 2) with a total average of 0.10 ± 0.06 kWh/m³ permeate. The salt removal averaged at 39 ± 13.7 and 42 ± 11.6% for stack 1 and 2, respectively, and averaged at 61.2 ± 14% for the duration of the pilot with a variety of settings applied.

Figure 3 shows the salt flux in a near-linear relation to the current density of the stacks. The salt flux, i.e., is the flux of ions transferred from the feed channels to the concentrate channels. This flux of cations through the cation exchange membranes and anions through the anion exchange membranes is possible by a sufficient electromotive force, which is created by the cathode and anode, which are energised by the stacks' power supply.

The stacks were operated using a permeate conductivity setpoint, meaning the salt flux depended on both the incoming TDS and the required outlet concentration. Ion transport behaviour was evaluated through relative transport numbers, which compare the movement of each ion to that of sodium or chloride. Increasing current density provided a stronger driving force for ion migration, producing the diluate in the feed channel and the concentrate in the adjacent channels. Because the wastewater contains substantial calcium, electroneutrality requires corresponding transport of bicarbonate and other

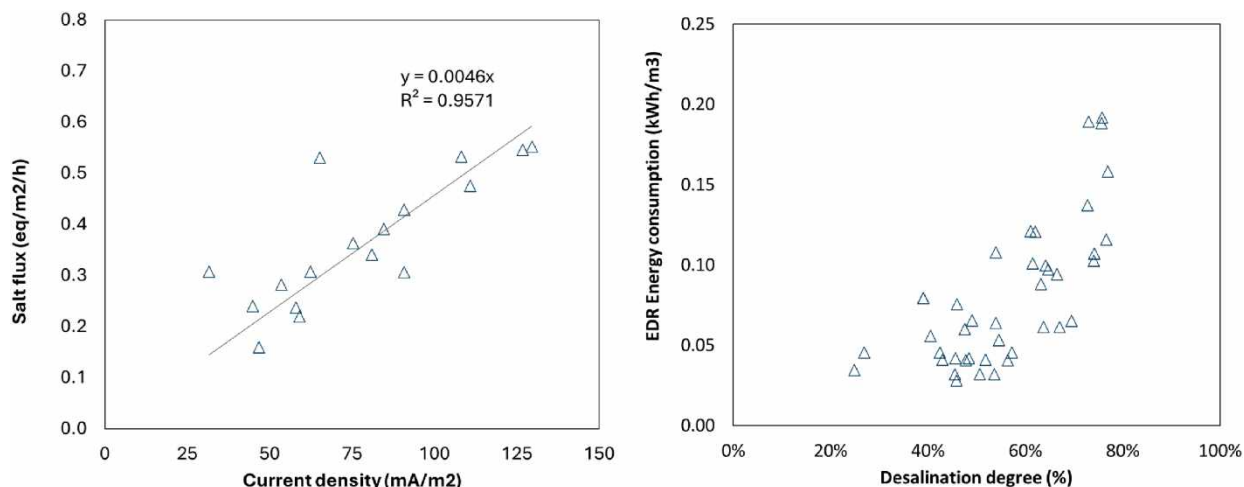


Figure 3 | Salt flux over stack 1 and 2 plotted against the utilised current density (left graph). Energy consumption in kWh/m³ diluate produced plotted against the desalination degree (right graph).

anions across the membranes. As a result, the system shows inherent selectivity between monovalent and multivalent ions based on their relative abundance and charge.

Transport numbers (P_C^{Na}) for cations and (P_A^{Cl}) for the specific ions corresponded to 9.0 for calcium, 8.2 for magnesium, 3.2 for potassium, 1.0 for sodium, 1.2 for sulphate, 0.8 for bicarbonate and 1.0 for chloride. This indicates the relative ion transport of calcium to be nine times higher than for sodium and the relative transport of magnesium to be 8.2 times higher than that of sodium. Moreover, this also means the transport of sulphate is only 1.2 times higher than chloride, and bicarbonate is 0.8 times lower than chloride. This implies a strong selectivity for multivalent cations when treating pulp and paper effluent with an EDR system, whilst less transport of monovalent cations and anions in general is observed. These transport numbers are calculated based on relative ion abundance in meq/L, which is graphically shown in Figure 4.

Figure 4 shows the relative ion compositions in the EDR feed, diluate and brine. The system displays clear ion selectivity, with monovalent anions such as chloride and bicarbonate transported more readily to the brine than sulphate, leading to sulphate enrichment in the diluate. Divalent cations are preferentially removed, resulting in a sodium-dominated diluate and a hardness-rich brine. This behaviour aligns with observations by Mubita *et al.* (2022), who reported that non-selective cation exchange membranes favour transport of Ca^{2+} and Mg^{2+} over Na^+ due to higher membrane affinity and stronger electromigration forces. The resulting brine composition indicates a high calcium carbonate scaling potential, requiring appropriate downstream treatment.

A similar selectivity trend is reported for RO by Biesheuvel *et al.* (2020), who showed that ion passage depends strongly on the Na^+ to Ca^{2+} ratio of the feedwater. Because the pilot wastewater contains relatively more calcium, an RO system in this configuration would be expected to pass more Ca^{2+} , Mg^{2+} and sulphate, potentially requiring additional polishing. Percentile analyses shown in Figure 5 confirm that the EDR brine periodically reaches elevated Ca^{2+} and Mg^{2+} levels consistent with oversaturation and high $CaCO_3$ scaling risk.

A caveat in interpreting selectivity data is that EDR behaviour depends directly on current density. Kim *et al.* (2012) showed that lower current densities favour retention of multivalent cations, whereas higher current densities promote monovalent ion transport due to increased concentration polarisation. Current density effects were tested by operating only one stack to reach the required diluate conductivity setpoint. Between day 110 and 123, the single-stack current density averaged 39.2 ± 6.7 , compared with 36.2 ± 12.3 mA/m² during two-stack operation. Despite this difference, no measurable increase in monovalent ion rejection was observed when operating with one stack at higher current density.

Thus, even though slightly higher current densities were applied with one stack only in operation, no shift in more monovalent ion transport could be observed (Table 2). Nevertheless, the flexibility of the EDR system, allowing ramping up and down in energy consumption, still agrees with the research of Patel *et al.* (2024) as earlier described. The sole benefit of the EDR thus remains flexibility and selectivity, with the latter being harder to control as it is a function of multiple variables.

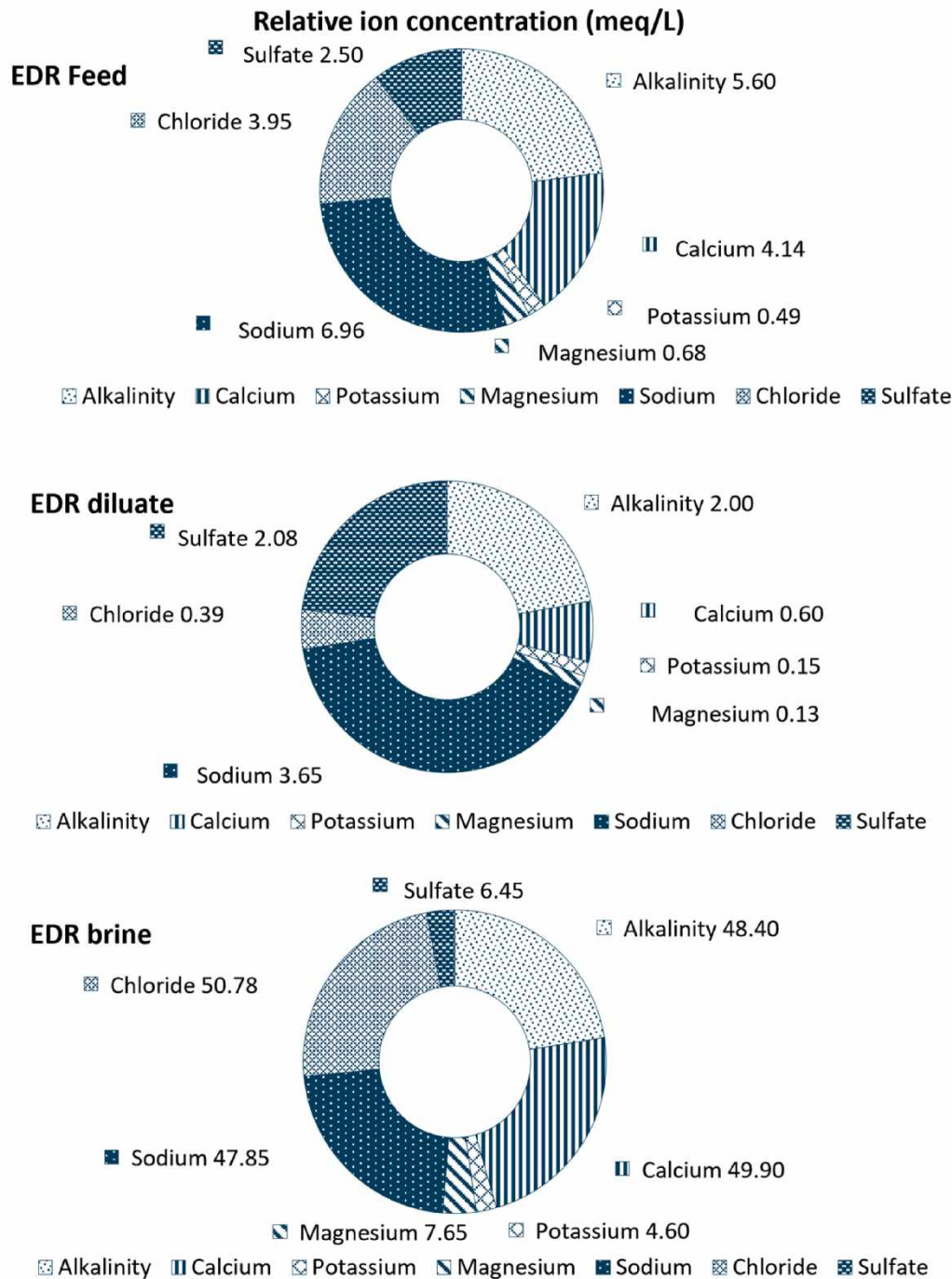


Figure 4 | Relative ion composition of EDR feed, diluate concentrate (brine).

In a conventional system setup, the dead-end UF system would not have retained as much COD, making the desalination system more prone to fouling. At a relative energy consumption of 0.3 kWh/m^3 permeate (*sub-supplier IP*), this HFNF system would be more efficient in removing this bit of additional scaling, which would otherwise cost 0.54 kWh/m^3 with a three-stage RO system running at similar recovery (Membrane Technology 2011). The EDR, on the other hand, was projected to require only 0.13 kWh/m^3 of diluate produced on full-scale (*sub-supplier IP*), not taking pilot non-continuity into account.

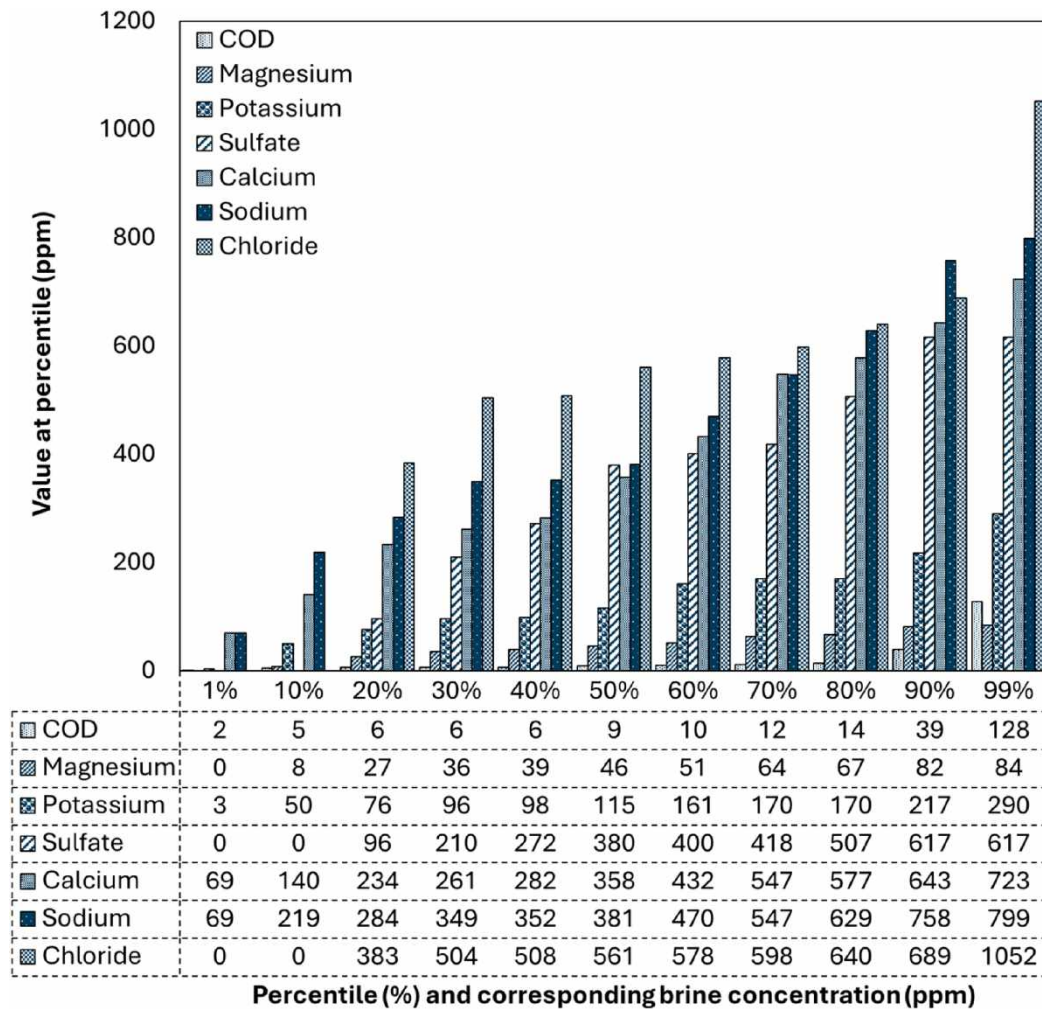


Figure 5 | Brine composition after HFNF, GAC and EDR treatment.

Table 2 | Comparative table of total salt removal, specific energy consumption (SEC) and corrected SEC per kg TDS separated whilst showing the corresponding applied current density for operation with one stack vs. two stacks in series

Parameter	Units	Stack 1 only	Stack 1 & 2 in series
Total salt removal	%	48.1 ± 11.6	63.4 ± 11.6
SEC	kWh/m ³ permeate	0.03 ± 0.01	0.10 ± 0.06
SEC/kg TDS separated	kWh/m ³ permeate/kg TDS	0.005 ± 0.003	0.008 ± 0.009
Applied current density ^a	mA/m ²	39.2 ± 6.7	36.2 ^a ± 12.3

^aFor the average of stack 1 and 2.

The technological advantages of the piloted end-of-pipe water reuse configuration for pulp-and-paper effluent can be summarised as follows: it provides (i) enhanced organic removal compared with UF due to the HFNF barrier, (ii) significant hardness reduction during HFNF that lowers downstream desalination energy demand, (iii) more selective ion removal when applying EDR instead of RO while maintaining a lower overall energy consumption, (iv) flexible desalination control that allows production of tailor-made permeate qualities, and (v) the formation of two distinct concentrate loops, the first one predominantly of organic nature with limited hardness and the other one of inorganic nature and highly enriched in hardness. This fifth aspect enables fit-for-purpose concentrate and brine management, supporting targeted recirculation

or discharge pathways and ultimately improving overall hydraulic recovery when integrated within the existing full-scale WWTP

3.2. Reuse water quality and brine composition comparison

The final produced water from the pilot was verified against the quality specifications as defined by the paper mill technologists. The main reuse water specifications were set to limits of <550 $\mu\text{S}/\text{cm}$ conductivity, <75 ppm chloride, <100 ppm sulphate, <10 ppm COD, <55 ppm calcium and <7.5 ppm magnesium. All limits were met at a 99th percentile as indicated in Figure 6.

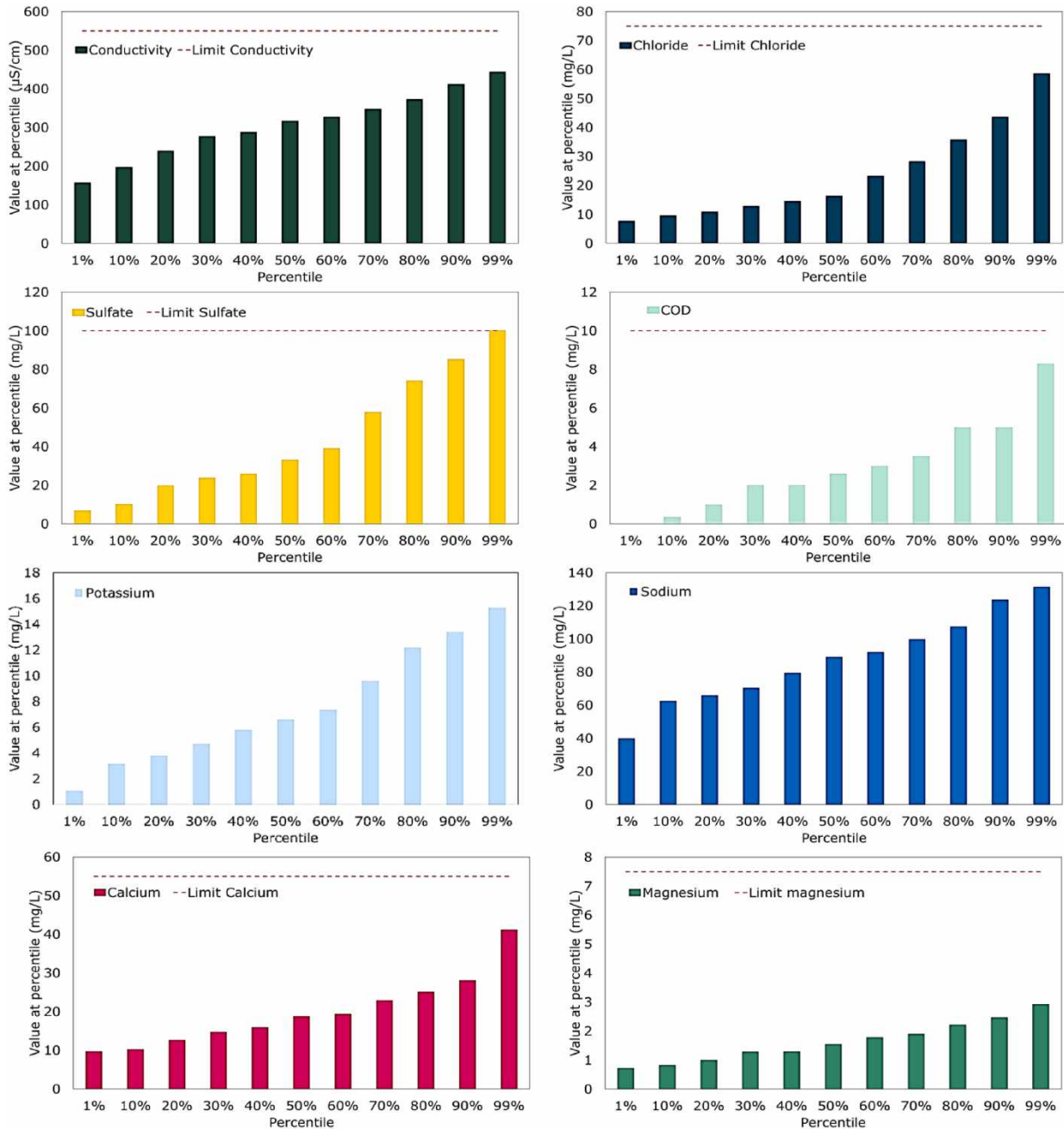


Figure 6 | Reuse pilot effluent quality displayed in 99th percentile graphs. *Limit values which are not displayed are limits that are not specified by the paper mill technologists.

Meeting the calcium limit was possible due to upstream physico-chemical softening at the IWE plant, which reduced the calcium content entering the EDR. The sulphate limit was easier to achieve due to the high selectivity of the HFNF system. However, in order to maintain these specifications at full-scale and with brine loop recirculation, the concentrations in this recirculation must be controlled by further treatment steps or existing treatment capacity in the upstream WWTP, as will be elaborated on in the next paragraphs.

3.3. Mass balance implications and recirculation treatment requirements

At full scale, recirculating untreated HFNF concentrate would lead to progressive contaminant accumulation in the WWTP, as shown in Figure 7. Mass balance calculations indicate increases of 6% for COD, 49% for chloride, 71% for sulphate and 37% for TDS, which would raise osmotic pressure, energy demand and scaling risk in the HFNF and EDR systems. This shift

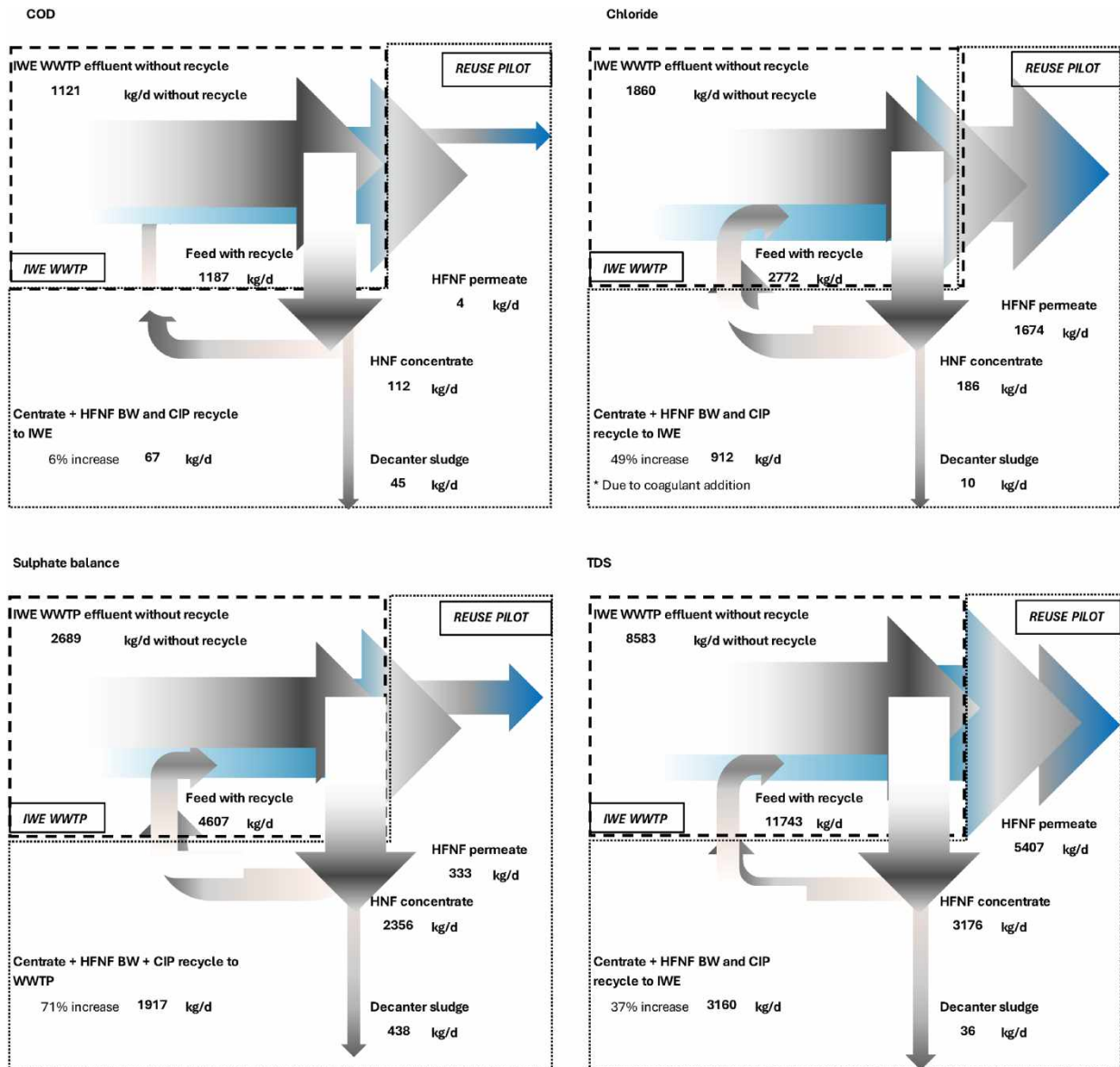


Figure 7 | COD, chloride, sulphate and TDS mass balance over IWE WWTP and reuse pilot if implemented at full-scale assuming no adequate treatment step to control the accumulation other than waste activated sludge or decanter sludge disposal from the pilot or salt separation by brine loop 2 in the reuse plant scheme. * Feed with recycle also accounts for higher steady state values due to iterative mass balancing.

would push the WWTP toward a new steady state, placing higher loading on the anaerobic and aerobic reactors, increasing sulphur input to the biological desulphurisation step and boosting sludge production. Although no major CAPEX upgrades are expected, the operational cost impact is significant. Chemical consumption in the desulphurisation process would rise proportionally to the 71% increase in sulphur load, with a cost reference of 13.65 EUR/kg S-H₂S removed (Cano *et al.* 2018). Sludge disposal costs would increase by roughly 6% at the current rate of 130 EUR per tonne dry solids. Costs associated with the disposal of calcite from physico-chemical softening remain variable at this site. Overall, the main OPEX increases stem from higher desulphurisation utility demand and increased sludge handling.

These implications are central when designing industrial water reuse schemes that incorporate recirculation loops over a factory or WWTP. Additional flow to the existing WWTP affects overflow rates in the primary and final clarifiers, increasing the potential for solids carryover. The recirculated COD has already undergone biological degradation and is further concentrated by the HFNF system, increasing the fraction of slowly biodegradable COD in the influent. This raises oxygen demand, sludge production and potentially effluent COD concentrations. The extra sulphur load also leads to increased elemental sulphur formation in the desulphurisation reactor, contributing to chemical consumption. Downstream, elevated calcium and magnesium concentrations promote precipitation in the physicochemical softening step, effectively turning it into a sink for these ions. Selecting a coagulant with lower chloride content can help reduce the recycled salt load to the EDR system while remaining within operational limits.

Overall, recirculation of the HFNF concentrate establishes new steady-state conditions in the upstream WWTP, but these effects are foreseeable and manageable with appropriate design and operational adjustments. A more challenging stream to manage is the high-TDS brine from the EDR (brine loop 2), which is not easily treated by the existing WWTP without causing significant TDS discharge or imposing a substantially higher TDS load on the downstream water-reuse system.

3.3.1. Recirculation loop 1 treatment strategy – organic dominant HFNF concentrate

The HFNF concentrate contains mainly non-biodegradable COD, which would increase aeration demand and OPEX if recycled directly to the WWTP. To limit this load, coagulation, flocculation and solid-liquid separation were applied to remove the hard-to-oxidise organic fraction before recirculation. Jar tests showed that FeCl₃ achieved 49 ± 11% COD removal, while tannin-based coagulants (Biofloc NG30 and NG57) achieved about 58%. These tannin-based coagulants (lower chloride concentrations) were therefore selected, together with A130 flocculants, as the most effective treatment chemicals. During pilot operation, the decanter achieved an average solids capture of 69 ± 22%, and overall COD removal from the combined coagulation, flocculation and dewatering step averaged 46 ± 18%. Although lower than the 95–98% capture achievable with larger decanter systems, this removal effectively reduces the organic load entering the WWTP and helps contain aeration and sludge-related OPEX while preventing elevated COD from impacting downstream reuse performance.

3.3.2. Recirculation loop 2 treatment strategy – inorganic EDR brine

Brine loop 2 presented a greater challenge than loop 1 because the existing WWTP has minimal capability to remove inorganic salts. The EDR brine exhibited high scaling potential with LSI values between 1.5 and 2.0, and its elevated concentrations of Cl[−] and SO₄^{2−} prevented discharge to the WWTP outfall. Recirculating this brine untreated would increase scaling risks in upstream equipment, increase loading on the biological desulphurisation and softening systems, and promote operational problems such as diffuser calcification and corrosion. Effective conditioning and concentration of this stream were therefore essential for both valorisation and cost-efficient disposal.

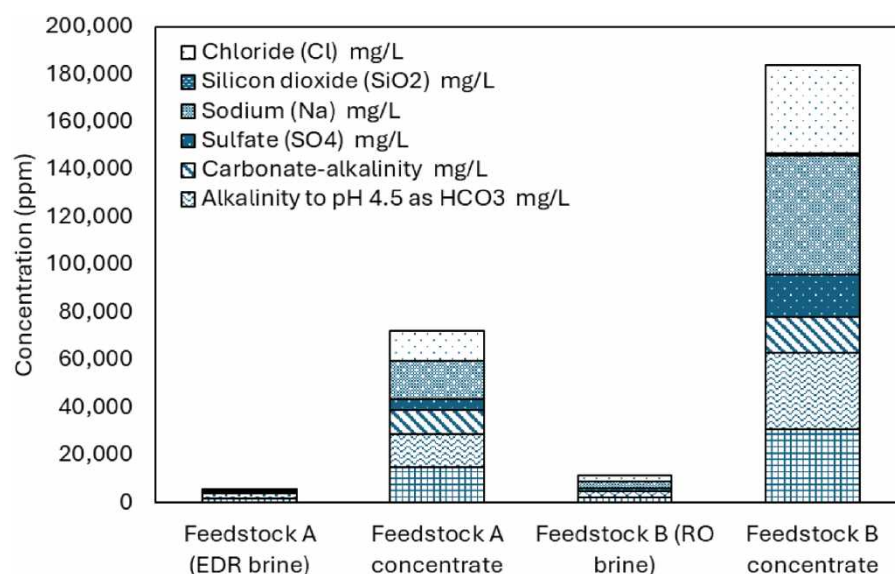
Two brine strategies were evaluated, namely evaporation and bipolar membrane ED for chemical production. Because Ca²⁺ and Mg²⁺ limit the achievable concentration of the brine, a weak acid cation exchange system was applied prior to further treatment, removing 92 ± 5% of Ca²⁺ and 84 ± 21% of Mg²⁺. A simpler lab-scale aeration-based softening test (a lower cost alternative) achieved 61% Ca²⁺ removal and 6% Mg²⁺ removal, while decreasing HCO₃[−] from 51.2 to 15.8 meq L^{−1} (70% reduction). Following softening, the brine was concentrated in a single-stage RO system, which operated stably at 60–90% recovery (average 71.2 ± 11.2%), with a feed flux of 19.2 ± 2.5 L m^{−2} h^{−1} and no significant scaling observed.

Evaporation tests compared raw EDR brine with the pre-softened and RO-concentrated brine, and results are shown in Table 3 and Figure 8, showing a clear performance gap between the two feedstocks. The RO brine contains more than double the total solids of the EDR brine (1.13 wt% vs 0.46 wt%), a conductivity more than twice as high, and significantly elevated sodium, chloride, sulphate and alkalinity concentrations. As a result, MVR evaporation of RO brine achieves far

Table 3 | Evaporation test results for EDR brine and RO brine

Evaporation feed	Total solids (TS) (wt%)	Density (kg/L)	pH	Conductivity ($\mu\text{S}/\text{cm}$)	COD (mg/L O ₂)
RO brine	1.13	1.00	7.5	11 340	166
EDR brine	0.46	1.00	7.5	5 020	16
Evaporation condensate	Total solids (TS) (wt%)	Density (kg/L)	pH	Conductivity ($\mu\text{S}/\text{cm}$)	COD (mg/L O ₂)
RO brine	NM	1.00	5.4	79.9	< 5
EDR brine	NM	1.00	5.3	9.0	< 5
Evaporation concentrate	Total solids (TS) (wt%)	Density (kg/L)	Viscosity @ 85 °C	Boiling point elevation (°C)	
RO brine	35.3	1.42	5 cP	8.0	
EDR brine	22.9	1.24	5 cP	3.4	

NM, not measured.

**Figure 8** | Mechanical vapour recompression (MVR) evaporation results with EDR brine and RO brine feedstocks.

greater concentration: the final concentrate reaches 35.3 wt% TS compared with only 22.9 wt% for EDR brine, despite similar viscosity limits. This translates to a 16.1× concentration factor for RO brine versus 11.7× for EDR brine, meaning RO brine achieves 37.6% higher final TDS at equal recovery.

Figure 8 further stresses the brine compositions, confirming the thermodynamic advantage created by pre-softening and RO pre-concentration, allowing for higher concentrated MVR concentrates. Because RO brine enters MVR at a higher strength and lower volume, the evaporator utilises less kWh/t brine evaporated as well as lower volumetric required capacity, thereby saving on both CAPEX and OPEX. Additionally, the EDR brine contains more hardness and possesses a lower TDS concentration, yielding a lower evaporator concentrate concentration and an increased scaling potential. Together, Table 3 and Figure 8 show that RO-derived brine is markedly more suitable for MVR evaporation.

Brine strategy exploration: Bipolar membrane ED. Two bipolar ED configurations were evaluated on RO brine: a three-chamber BPED producing acid, base and a weakened brine, and a two-chamber BPED producing a carbonate-free brine and a base. In the three-chamber system, a 1.3 wt% brine was treated in sequential stages, each removing approximately 60% of the salinity and generating equal acid and base volumes. Feed conductivity decreased from 17 to 7–8 mS/cm per stage, while product conductivities increased. Operation at 200 A/m² continued until the feed reached roughly 40% of its initial

conductivity. After five stages, the acid and base streams reached 1.2 and 1.3 wt% respectively, with production rates of 0.11 kg/m²/h (acid) and 0.13 kg/m²/h (base) at SECs of 7 and 6 kWh/kg. A continuous counter current approach would reach about 0.2, 0.6 and 1.2 wt% at 100, 200 and 300 A/m².

In the two-chamber system, the brine was acidified to remove bicarbonate, converting HCO₃⁻ to CO₂ and lowering pH from 6 to 4. The process operated at 200 A/m² and 15.8V, consumed 0.018 kWh and produced a base at 0.12 kg/m²/h. The treated brine contained less than 6 mg/L bicarbonate, with stable Cl⁻ and SO₄²⁻ and the chemical product contained 0.32 wt% carbonate free base suitable as precursor for NaOCl generation with 99% purity at 5 kWh/kg.

The three-chamber system yielded both acid and base, while the two-chamber configuration produced only base but required fewer membranes and lower voltage. In both cases, final product concentrations remained below industrial norms, although higher salinity feeds or increased current densities could raise strengths. While the two-chamber process effectively removes bicarbonate, the resulting low-ionic-strength brine still requires evaporation for minimum liquid discharge (MLD) or ZLD applications, limiting its standalone viability.

Energy requirements were substantial for both mechanical vapour recompression and bipolar membrane ED. MVR required about 25 kWh/m³ to reach a 30 wt% brine at 90% recovery. These findings lie between the 10 and 14 kWh/m³ reported by Yang *et al.* (2016), at 42% recovery and the 40 kWh/m³ reported by Hayes *et al.* (2014) at 72.5% recovery. The three-chamber BPED configuration consumed about 72 kWh/m³ of brine fed at 200 A/m², while the two-chamber system required about 18 kWh/m³, consistent with values reported by Fernandez-Gonzalez *et al.* (2017) for comparable current densities. Economic viability depended strongly on brine disposal cost and possible chemical revenue. Sensitivity analysis indicated that increases in electricity price and CAPEX depreciation affected both MVR and BPED similarly, while disposal cost and achievable product value were the dominant parameters.

Two valorisation strategies were examined: volume reduction by evaporation and recovery of acid and base via BPED. RO pre-concentration proved essential for both routes. It produced brine that was 80.8% more concentrated than untreated EDR brine before evaporation and 148.3% more concentrated after evaporation, underlining the importance of early volume reduction. Hardness removal remained the primary limitation for achieving high RO recovery, and although ion exchange required frequent regeneration, physical chemical softening provided a more economical alternative.

For BPMED, product concentration depended on feed salinity, recirculation rate and current density, in line with the transport behaviour described by Wilhelm (2001). This stresses the need for preconcentration or an increasingly more CAPEX-oriented BPMED solution, which is capable of applying higher recirculation rates in order to generate higher product concentrations.

3.4. Techno-economic comparison

Financial starting points are summarised in Table 4. Together with Table 5, they form the basis for the output of the economic comparison displayed in Tables 6 and 7.

Tables 6 and 7 present the full TOTEX assessment for a 3,650,000 m³/year reuse plant, with all process blocks cost-calculated using internal databases and supplier quotations. The total TOTEX, including treatment of both brine loops, was 2.14 EUR/m³ for the conventional UF GAC RO configuration and 1.85 EUR/m³ for the innovative HFNF GAC EDR configuration.

Although both estimates carry an accuracy of approximately ±30%, the innovative setup consistently shows the lower overall cost. Model sensitivity is strongly linked to the piloted recoveries and salt removal efficiencies, which differ from design expectations typically applied in the design of UF and RO systems.

A key observation is the shift in cost distribution. The innovative system has a more CAPEX-oriented water line compared with the conventional system, but has significantly lower brine treatment costs. This reflects both the lower desalination degree required for EDR compared with RO and the higher concentration of HFNF concentrate relative to UF concentrate. As shown in Table 5, the cost burden in conventional systems moves rapidly toward the brine treatment stage, where costs scale sharply with brine volumes produced. In contrast, the tailored approach of the innovative configuration reduces OPEX at the expense of higher CAPEX. Major OPEX contributors for the innovative system include (i) HFNF module replacement, (ii) chemical consumption in recirculation loop 1 and (iii) electricity use. Electricity demand is substantially lower for EDR than for RO because only 65% desalination is required rather than 96%.

Table 4 | Economic model initial conditions

Item	Unit	Unit price ^a
Electrical power	EUR/kWh	€ 0.11
NaOH 20%	EUR/tonne	€ 220
Citric acid 99%	EUR/tonne	€ 1,750
NaOCl 12.5%	EUR/tonne	€ 400
HCl 30%	EUR/tonne	€ 200
FeCl ₃ 40%	EUR/tonne	€ 250
Antiscalant	EUR/tonne	€ 6,660
Biocide	EUR/tonne	€ 20,000
H ₂ SO ₄ 98%	EUR/tonne	€ 160
Polymer	EUR/tonne	€ 4,000
Nitric acid 53%	EUR/tonne	€ 350
UF module replacement	EUR/module	€ 3,900
RO module	EUR/module	€ 800
GAC regeneration/replacement	EUR/m ³	€ 1,200
EDR stack replacement	EUR/stack	€ 6,875
Sludge disposal costs	EUR/wet tonne	€ 130
Brine disposal costs	EUR/wet tonne	€ 100
CAPEX depreciation (linear)	CAPEX depreciation years	10

^aUnit prices corresponding to December 2023 at the time of pilot execution and are location-specific.

Table 5 | Technological outline and comparison basis

Item	Units	HFNF	UF	EDR	RO
Treatment recovery	%	96	91	90	85
Treatment setup	–	Double-stage crossflow	Single-stage dead-end	Feed and bleed	Single pass 3 stage
Desalination degree	%	–	–	65 ^a	96 ^b

^aRequired TDS removal for EDR is displayed to produce sufficient quality reuse water, maximum desalination potential is higher.

^bRO desalination degree calculated based on Nitto Hydranautics RO projections (Membrane Technology 2011).

Table 6 | Innovative treatment train consisting of HFNF, GAC and EDR in the main water line

Item	Unit	Water line	Concentrate and brine treatment	Total
Total CAPEX	€/m ³ reuse water	€ 0.65	€ 0.17	€ 0.82
Electrical power	€/m ³ reuse water	€ 0.22	€ 0.11	€ 0.33
Chemicals	€/m ³ reuse water	€ 0.04	€ 0.19	€ 0.24
Consumables	€/m ³ reuse water	€ 0.41	€ 0.05	€ 0.45
Total OPEX	€/m ³ reuse water	€ 0.67	€ 0.36	€ 1.02
TOTEX	€/m ³ reuse water	€ 1.32	€ 0.53	€ 1.85

The effective cost of brine treatment depends strongly on (i) the volume and composition of brine produced, (ii) disposal costs, (iii) chemical and energy consumption and (iv) the potential for circular chemical valorisation. In this context, the performance advantages of the innovative system become clear. Successful end-of-pipe reuse relies on (i) selective removal of organics and hardness early in the process, where HFNF outperforms UF, (ii) partial desalination using EDR, which provides lower energy use under moderate salinity conditions, (iii) high recovery to reduce brine volume, (iv) effective hardness

Table 7 | Conventional treatment train consisting of UF, GAC and RO in the main water line

Item	Unit	Water line	Concentrate and brine treatment	Total
Total CAPEX	€/m ³ reuse water	€ 0.44	€ 0.20	€ 0.64
Electrical power	€/m ³ reuse water	€ 0.39	€ 0.20	€ 0.59
Chemicals	€/m ³ reuse water	€ 0.05	€ 0.44	€ 0.49
Consumables	€/m ³ reuse water	€ 0.17	€ 0.24	€ 0.42
Total OPEX	€/m ³ reuse water	€ 0.62	€ 0.88	€ 1.50
TOTEX	€/m ³ reuse water	€ 1.05	€ 1.08	€ 2.14

removal to prevent scaling and (v) flexibility to accommodate changing influent quality. Because local electricity and disposal prices vary significantly, they strongly influence the economic balance between the two configurations.

Finally, the valorisation potential of concentrated brines was explored by comparing evaporation with BPMED, summarised in Table 8. The results show that economic feasibility can shift depending on (i) energy price, (ii) achievable chemical revenue and (iii) disposal cost. Under favourable conditions, BPMED can offer additional circular value; however, evaporation remains more robust for this brine matrix at present. Overall, the HFNF GAC EDR configuration demonstrates a selective, energy-efficient and economically favourable route for reliable end-of-pipe water reuse in the pulp and paper industry.

The choice for an MVR evaporation system for the full-scale model of Tables 6 and 7 was not directly related to its economic benefits, as Table 8 shows the BPMED system to be more economically viable over an MVR evaporation system. Moreover, the BPMED chemical product also allows for a more circular benefit of producing chemicals for the pulp and paper industry. However, from a technical and supply chain point of view, the concentrations of the produced acid and base were not deemed of sufficient strength and purity for the paper mills to guarantee offtake for the calculated price per tonne. However, if an off-taker would be sufficiently interested in such a chemical of the quality and quantity specified in Section 3.3.2, a more positive business case is expected based on $\pm 30\%$ figures presented above.

Challenges remain in increasing the product concentration by applying an increased recirculation rate in the BPMED system to produce chemicals with a higher concentration. Moreover, softening is a key brine pretreatment step to avoid limiting operating conditions due to CaCO₃ scaling. Evaporation itself is also economically more viable after pre-concentration by RO compared to no pre-concentration. i.e. evaporating the EDR concentrate was less energetically favourable than evaporating the RO concentrate. The same holds for chemical production, which also benefits from elevated inlet concentrations as was found in the technological results from Section 3.3.2, highlighting the need for concentrated brine feedstock for these measures.

Table 8 | Evaporation vs BPMED for brine treatment and valorisation comparison, along with simplified sensitivity effects displayed at the end of the table

Criteria	Unit	MVR	BPMED
Depreciated CAPEX	€/m ³ reuse water	€ 0.05	€ 0.03
Electrical power costs	€/m ³ reuse water	€ 0.03	€ 0.05
Waste stream management	€/m ³ reuse water	€ 0.16	€ 0.06
Revenue (+)/costs (-) from fresh water	€/m ³ reuse water	€ -0.02	€ 0.01
Revenue/savings (+) from chemicals	€/m ³ reuse water	€ 0.00	€ -0.17
TOTEX (simplified)	€/m ³ reuse water	€ 0.22	€ -0.02
TOTEX modification:	Unit	MVR	BPMED
Electricity x3	€/m ³ reuse water	€ 0.27	€ 0.08
No chemical revenue BPMED	€/m ³ reuse water	€ 0.22	€ 0.15
No brine disposal costs	€/m ³ reuse water	€ 0.06	€ 0.09

4. CONCLUSIONS

This study provides important insights into the technological feasibility and practical implementation of water reuse systems for the paper and pulp industry, specifically in the Province of Gelderland, the Netherlands. The research explored whether a more selective and adaptive treatment approach could overcome the limitations of conventional UF- and RO-based end-of-pipe reuse systems. By comparing established water reuse technologies to an innovative treatment train centred on HFNF, GAC polishing and EDR, the work provides new evidence that a selectively staged configuration can improve process stability, energy efficiency and brine treatability under variable industrial conditions.

The pilot results demonstrate that HFNF provides a substantially stronger removal of organic matter and sulphate than UF, reducing fouling and improving downstream desalination performance. These outcomes confirm that the use of a selective barrier early in the process can meaningfully influence the overall behaviour of the treatment line, particularly when dealing with high variability in WWTP effluent quality. The integration of EDR further strengthens this effect, as its load-dependent energy adaptation and partial desalination capability offer operational flexibility that conventional RO cannot easily provide. While the Introduction outlined this potential, the pilot results show that the system achieves these benefits in practice, reaching high-quality reuse water under conditions where the conventional line would be more sensitive to fouling and scaling.

The separation of the concentrate into an organic-rich HFNF stream and a salt-rich EDR brine provides an additional advantage. It allows brine treatment and valorisation options to be tailored to each stream instead of relying on a single mixed RO brine. This separation was found to reduce the brine treatment burden, especially when combined with softening and pre-concentration of the EDR brine before evaporation or bipolar ED. Although both evaporation and BPED were technically feasible, the pilot clarified that the economic viability of BPED depends strongly on the achievable inlet concentration and the marketability of the produced chemicals. Nevertheless, the work illustrates that brine management, long viewed as a major barrier to large-scale reuse, can be approached more strategically through controlled selectivity and stream segregation.

From a techno-economic perspective, the innovative line achieved a somewhat lower total expenditure than the conventional UF RO line while producing reuse water of consistently high quality. The difference is not dramatic, but it is meaningful, especially considering that the innovative configuration performed better in terms of operational flexibility, energy responsiveness and robustness against variable inlet conditions. These outcomes provide practical evidence that next-generation reuse trains can shift from a purely membrane-driven strategy to a more selective, process-adaptive approach that reduces overall treatment stress while supporting minimum liquid discharge (MLD)-oriented brine handling.

In summary, this study confirms that end-of-pipe water reuse for the pulp and paper industry can be achieved reliably through a novel combination of selective HFNF filtration, adaptive EDR desalination and targeted brine treatment. While additional optimisation is required for full-scale implementation, especially regarding brine valorisation routes, the findings offer a realistic and actionable pathway that advances current practice. The work thereby closes a critical knowledge gap identified in the Introduction and provides the scientific and engineering community with new insights to guide future designs of flexible, resilient and economically feasible industrial water reuse systems.

ACKNOWLEDGEMENTS

This work was performed in the cooperation framework of the province of Gelderland, the Vallei en Veluwe Water Board, the Dutch Paper Mills Association and the municipality of Brummen. The authors would like to thank the team of Industrie-water Eerbeek for their cooperative and intensive efforts to ensure succeeding of the pilot. Furthermore, the authors would like to thank fellow water technologists from NX Filtration B.V., REDstack B.V., KWR, Mobile Water Solutions B.V., Salt-tech B.V. and Epcon B.V.

FUNDING

The pilot was funded under the subsidy of Provincie Gelderland and was initiated by the paper mills in the Eerbeek-Loenen periphery, as the subsidy was granted to Industrierwater Eerbeek B.V., which gave the project order to Nijhuis Saur Industries B.V.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The pilot study was initiated by paper mills located in the Eerbeek-Loenen periphery, and a direct relation was established for the purpose of collecting data on the required water quality of the produced reuse water. The authors declare that the paper mills had no influence on the design, execution, interpretation, or reporting of the work presented in this study.

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First received 17 December 2025; accepted in revised form 18 April 2026. Available online 9 June 2026