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Enabling Water Reuse by Treatment of Reverse Osmosis Concentrate: The Promise of Constructed Wetlands

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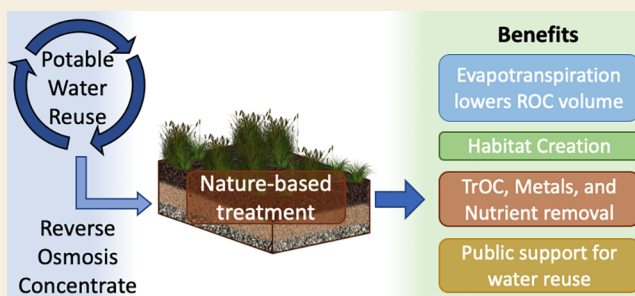


Article Recommendations



Supporting Information

ABSTRACT: As more cities experience water stress, the use of reverse osmosis (RO) membranes for wastewater treatment and reuse will expand. The concentrated waste stream resulting from RO treatment can pose chronic ecotoxicity risks if discharged to surface waters or shallow coastal ecosystems. Most existing RO concentrate treatment technologies are cost prohibitive, but constructed wetlands hold promise as a viable multibenefit solution because they have the potential to provide simultaneous treatment of nutrients, metals, and trace organic contaminants at a relatively low cost. They also are popular with the public. A handful of water-stressed cities have already begun experimenting with constructed wetlands for RO concentrate treatment. However,



further research is needed to reduce the land area needed for treatment and increase the reliability of constructed wetland systems.

KEYWORDS: reverse osmosis, wastewater treatment, constructed wetlands

INTRODUCTION

The United Nations forecasts that by 2050 urban dwellers will comprise about 70% of humanity, representing 2.5 billion more people than live in cities today.¹ In conjunction with this urban migration, water demand is expected to increase by around 80%.² Due to multiple demands on water resources and the effects of climate change, cities are likely to turn to alternative water sources, such as seawater desalination, rainwater capture, and potable water reuse.

Among these options, water reuse is attractive in part because it can be the least expensive option and simultaneously reduces pollution from municipal wastewater discharges. Although nonpotable water reuse—the practice of applying treated wastewater to crops or landscaping or using treated wastewater for industrial applications or in cooling towers—has been popular in many parts of the world for over 50 years, the reuse of wastewater to augment drinking water supplies (i.e., potable water reuse) is becoming a major focus of water planners in the United States,^{3,4} Singapore,⁵ Great Britain,^{6,7} South Africa,⁸ and Australia.^{9,10} Within the European Union, regulatory authorities, utilities, and researchers are beginning to reconsider potable water reuse after decades of skepticism about the safety of the practice.^{11,12}

As a result of the success of the first generation of potable water reuse systems in Southern California and Singapore, reverse osmosis (RO) has become the design standard for many advanced water treatment projects.^{13,14} Initially installed as part of an effort to lower the concentration of dissolved salts in recycled water, RO has become popular because it has

proven to be capable of removing waterborne pathogens and most chemicals.¹⁵ The few contaminants that are not well removed during RO (e.g., NDMA, 1,4-dioxane) are usually much easier to treat after the process when fewer organic compounds compete for oxidants or ultraviolet light.¹⁶ Advanced treatment trains incorporating RO are now gaining acceptance as safe processes for potable reuse, but the use of the technology results in a concentrated waste stream, RO concentrate, that requires treatment or disposal.

THE GROWING CHALLENGE OF MANAGING RO CONCENTRATE

Management of RO concentrate often poses a challenge for communities implementing water reuse projects. The simplest approach to disposing of RO concentrate is to blend it with municipal wastewater effluent, resulting in a smaller discharge volume containing nearly the same mass of contaminants. The resulting higher concentration that is discharged into the receiving water typically does not result in the exceedance of water quality criteria when a small fraction of a community's wastewater is reused. However, as a larger fraction of a city's wastewater is reused, concentrations of wastewater-derived

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contaminants in the effluent will increase, potentially leading to circumstances in which treatment will be necessary.

Considering that existing RO systems typically recover approximately 85% of the wastewater undergoing recycling, we predict that concentrations of contaminants in the blended outfall (i.e., when RO concentrate is mixed with the remaining fraction of the municipal wastewater effluent that is not being recycled) will double when approximately 60% of the wastewater entering the treatment plant is recycled (Figure 1). Concentrations will double again when approximately 85%

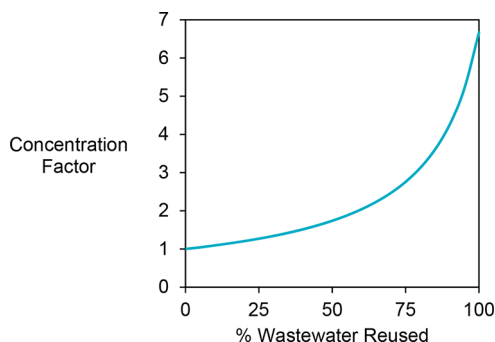


Figure 1. Factor by which the concentration of contaminants in discharged effluent increases relative to municipal wastewater when RO-based water reuse is implemented.

of the wastewater is recycled, and will be 6 to 7 times higher than concentrations in wastewater if all of the wastewater is recycled.

For trace contaminants that pose risks to aquatic ecosystems at or near the concentrations in wastewater effluent (e.g., the neonicotinoid imidacloprid,¹⁷ the insecticide fipronil,¹⁸ and the antibiotic sulfamethoxazole¹⁹), a doubling of the concentration being discharged to the receiving water might result in chronic aquatic toxicity. One way to assess the impacts of trace organic contaminants is through a risk quotient analysis, which compares contaminant concentrations in effluent to concentrations that can cause aquatic toxicity. In the South San Francisco Bay, an area where wastewater effluent is not substantially diluted by water from other sources during a dry season that lasts about 6 months per year, a risk quotient

analysis for the discharge of RO concentrate generated from a water reuse facility that is expected to share a discharge pipe with a conventional wastewater treatment plant indicated that urban-use pesticides could pose a moderate chronic toxicity risk (i.e., risk quotient between 6 and 10 for imidacloprid) at concentrations present in wastewater, and that concentrations present in RO concentrate are up to approximately 50 times greater than chronic toxicity benchmark values.²⁰ Among the trace metals present in wastewater, copper is a concern due to its effects on aquatic organisms at low concentrations. In RO concentrate samples from five water reuse facilities, copper concentrations ranged from 7.4 to 38 $\mu\text{g/L}$,²¹ exceeding the EPA saltwater chronic toxicity concentration (3.1 $\mu\text{g/L}$)²² and potentially exceeding recommended water quality criteria for freshwater (which range from approximately 4 to 50 $\mu\text{g/L}$ depending on organic carbon concentrations and hardness).²³ The potential for chronic toxicity from contaminants such as these may motivate designers of water reuse projects to investigate methods to remove trace contaminants from RO concentrate.

For contaminants that pose potential risks to downstream drinking water sources, like nitrate and poly- and perfluoroalkyl substances (PFAS), increases in the concentrations of contaminants due to water reuse would increase human health risks, especially in effluent-dominated water sources typical of water-stressed regions.^{24,25} Further examples of contaminants of concern for ecotoxicological and human health and concentrations measured in reverse osmosis concentrate from water reuse facilities are provided in Table S1. Irrespective of the exact conditions and contaminants of concern, it is reasonable to assume that as water reuse becomes more popular, there will be a greater need for technologies capable of removing contaminants that pose ecological or human health concerns from RO concentrate.

■ CONCENTRATE MANAGEMENT LIMITATIONS NEAR WATER-STRESSED CITIES

To gain insight into the locations where treatment of RO concentrate may be needed in the future, we considered large cities in water-stressed regions. We found that among the 100 most populous cities in the world, 40 are located in regions

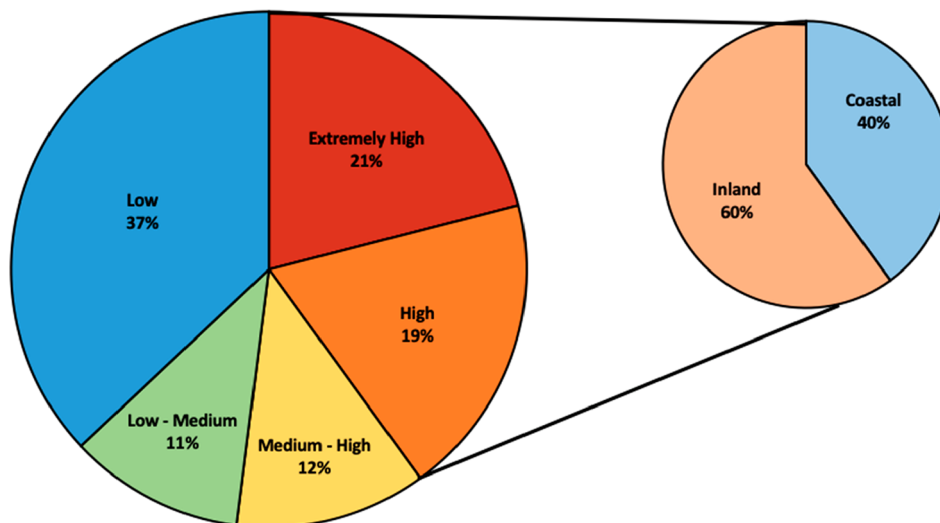


Figure 2. Water stress in the 100 most populous cities and proximity to the ocean for cities experiencing high or extremely high water stress.

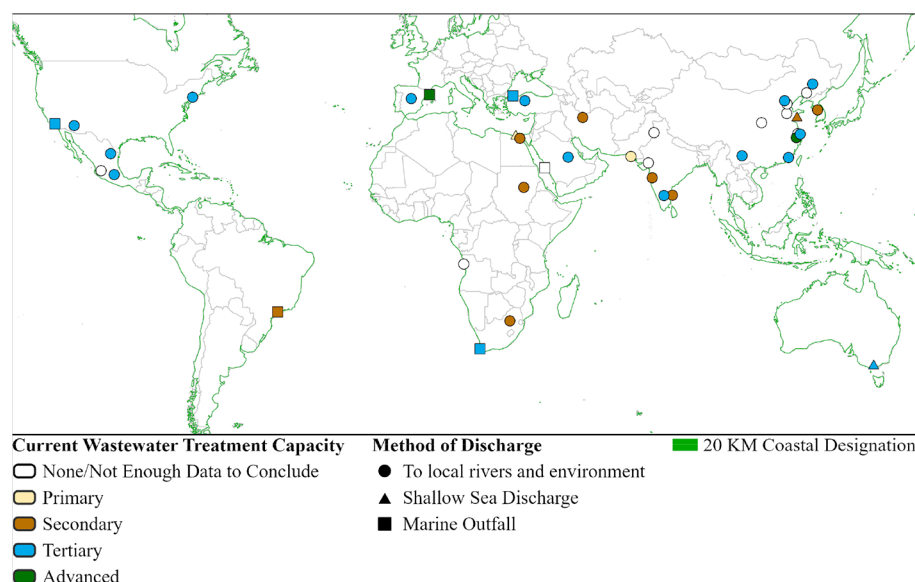


Figure 3. Wastewater treatment and discharge methods in the world's 40 most populous water-stressed cities. Shape corresponds to discharge method (square = marine outfall, triangle = shallow sea discharge, circle = to local surface waters). Color corresponds to level of wastewater treatment (green = advanced, blue = tertiary, brown = secondary, yellow = primary, white = none or inadequate data to determine).

that already experience high or extremely high physical water stress according to the Water Resource Institute's Water Risk Atlas (Figure 2; analysis details are in the Supporting Information). Among these cities, we expect that those located in wealthy countries will be the early adopters of potable water reuse due to the capital-intensive and complex nature of potable water reuse projects. In addition, smaller cities in arid regions within affluent countries (e.g., Perth, Denver, Athens) as well as large cities that face water stress due to population pressure and limited opportunities for expanding water storage or imported water supplies (e.g., London, Atlanta, Seoul) also may be part of the next wave of potable water reuse adopters.

To better understand the need for RO concentrate treatment in the world's largest cities that may pursue potable water reuse, we considered the discharge locations of wastewater treatment plants because existing discharge infrastructure is likely to be used as the ultimate means of disposal of RO concentrate. RO concentrate management approaches may differ between coastal and inland locations, so we considered the discharge strategies and implications of RO concentrate discharge in both settings.

For inland water-stressed cities, it is sometimes assumed that salts will be the primary concern for RO concentrate management, but our analysis suggests that nutrients and trace contaminants often should be a greater concern, particularly in arid regions where surface waters already have relatively high salt concentrations. For example, RO concentrate from a water reuse facility in Big Spring, Texas, is discharged to Beals Creek, which is allowed to receive RO concentrate discharge because it already contains an elevated concentration of total dissolved solids (TDS). Beals Creek is a tributary to the Colorado River just upstream of Lake Spence, which typically contains between approximately 1500 and 3500 mg/L TDS.^{26,27} Similarly, the Salt River in the southwestern United States has a TDS concentration of approximately 580 mg/L,²⁸ such that salts present in RO concentrate from a reuse project would not significantly impact downstream use. The city of Phoenix already treats its wastewater effluent at the Tres Rios wetland before it is

returned to the Salt River. In the future, if RO-based potable reuse were adopted in the city of Phoenix, RO concentrate discharges to the Salt River might be permitted in a similar manner, with the Tres Rios wetland receiving a mixture of RO concentrate and municipal wastewater effluent. In these cases, contaminants that pose risks to aquatic ecosystems, including trace organic contaminants, nutrients, and metals, may drive the need for RO concentrate treatment rather than concerns about the effects of salt on ecosystems or a downstream water supply. Similar drivers may be in place in other inland cities. For instance, nutrients would likely be the dominant concern for discharge of RO concentrate from Madrid, where wastewater is discharged to the Manzanares River, which can be up to 90% wastewater effluent, and where stricter limits are being considered to avoid downstream impacts of nutrients and biological oxygen demand (BOD).²⁹ Although TDS concentrations may be a consideration when RO concentrate is discharged to many freshwater systems, nutrients and trace contaminants may drive decisions related to water quality protection, especially in arid regions.

Coastal cities may avoid the need for RO concentrate treatment if appropriate discharge infrastructure exists (i.e., deep water outfalls) because of the substantial dilution in marine systems. However, not every coastal city will be able to discharge RO concentrate without consideration of its potential ecological impacts. Sixteen of the 40 water-stressed large cities in our analysis are located within 20 km of the ocean, but only six currently dispose of their wastewater primarily through deep ocean outfalls (Figure 3). Three large coastal cities from our analysis lack deep ocean outfalls and discharge directly to shallow coastal waters (i.e., Qingdao, Alexandria, and Melbourne). The remaining coastal cities discharge wastewater to rivers or shallow estuaries. In cases where cities discharge to shallow coastal waters or rivers, elevated contaminant concentrations in RO concentrate may pose a threat to local ecosystems, resulting in similar concerns as those faced by inland communities. Even in locations where deep ocean outfalls are used, ecological impacts might still be a concern near outfalls. These effects could be exacerbated by

the discharge of a more concentrated waste stream. Thus, coastal cities that do not already have access to infrastructure that will protect local ecosystems from contaminants in RO concentrate may need to weigh the costs of constructing deep ocean outfalls against those of treating RO concentrate.

As an alternative to managing RO concentrate, some inland and coastal communities appear to be choosing to avoid RO in reuse projects altogether. For example, the SWIFT water reuse facility in Hampton Roads, Virginia, is planning to pursue potable water reuse without RO, in part to avoid the need to discharge RO concentrate to a nutrient-impacted estuary (i.e., the Chesapeake Bay). Elsewhere in the U.S., potable reuse facilities have also been built without RO treatment, relying instead on combinations of advanced oxidation and activated carbon filtration.³⁰ George, South Africa provides another example of reuse without RO. There, the wastewater treatment plant was upgraded to include ultrafiltration, after which the treated wastewater is discharged to a surface water reservoir.⁸ The reuse scheme also includes the addition of activated carbon to an existing drinking water treatment system.⁸ Although treatment trains without RO may gain more popularity in the future, RO is still considered among the most reliable treatment processes for removing wastewater-derived contaminants. Cost-effective RO concentrate treatment technologies could enable wider adoption of potable water reuse with RO and its commensurate water quality benefits.

■ THE PROMISE OF NATURE-BASED RO CONCENTRATE TREATMENT

Existing treatment options for RO concentrate are limited and often require multistep treatment trains to remove nutrients, trace organics, and metals. Ozone paired with biological activated carbon (O₃/BAC) may be technologically feasible for some aspects of treatment because it has proven to be effective at full scale for removal of trace organic contaminants from municipal wastewater effluent.^{31–33} Laboratory-scale studies indicate that it is also likely to be feasible for RO concentrate.^{34,35} However, when applied to RO concentrate, O₃ can form toxic byproducts, such as bromate. Furthermore, O₃/BAC cannot remove other important contaminants present in RO concentrate, such as metals and nutrients, although some nitrate removal may occur if a sufficiently large mass of carbon is added.³⁶ Other unit processes employed for treatment of residual wastewater effluent might also be used to treat contaminants in RO concentrate, but higher salt and contaminant concentrations will likely increase costs and decrease reliability.^{37,38} Irrespective of performance, the added cost of treating contaminants in RO concentrate is likely to slow the adoption of RO treatment for water reuse under conditions in which concentrate treatment is necessary.

In recognition of the need to develop more cost-effective and reliable treatment technologies for RO concentrate, researchers have begun to investigate new treatment methods. Proposed solutions for removal of trace organic contaminants from RO concentrate include advanced oxidation,³⁹ electrochemical treatment,^{40,41} activated carbon adsorption,⁴² photocatalysis, and sonolysis.⁴³ However, substantial barriers exist prior to implementation of these technologies, including cost, energy intensity, the need for highly trained operators, and the potential formation of toxic byproducts.⁴⁴

Nature-based treatment systems may be an appropriate option for RO concentrate treatment because they are cost-

effective, and well-designed nature-based systems have the potential to address multiple contaminant classes simultaneously.⁴⁵ Furthermore, wetland plants and microbes are well-adapted to the elevated salinity and solute concentrations encountered in RO concentrate. Nature-based systems also can create public support for water recycling projects because they provide ancillary benefits (e.g., aesthetic features, habitat) that are valued by community members.⁴⁶

Although constructed wetlands have not yet been tested at full scale for RO concentrate treatment, they have been widely used to treat wastewater that contains concentrations of salts and organic compounds that are in the range encountered in concentrate produced in potable water reuse projects (Figure 4).⁴⁷ For example, constructed wetlands have been used to

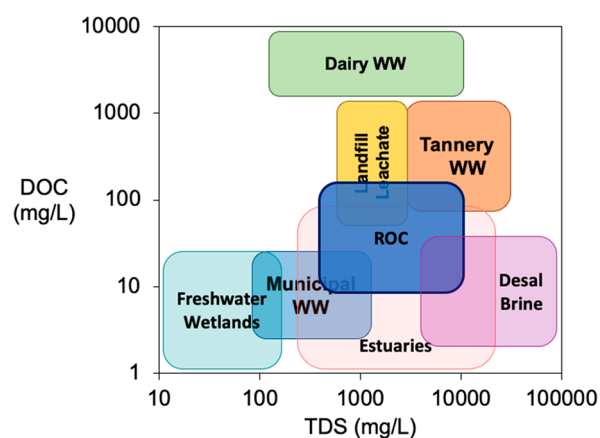


Figure 4. Total dissolved solids (TDS) and dissolved organic carbon (DOC) content of RO concentrate (ROC) compared to those in natural waters and wastewaters that have been treated with constructed wetlands.

remove labile organic carbon (i.e., the carbohydrates and organic compounds responsible for biological oxygen demand, BOD), suspended solids, and, in some cases, inorganic nitrogen from saline wastewater produced by tanneries and dairies.^{47,48} In terms of salinity and concentrations of organic matter, the conditions encountered in RO concentrate are well within the range of those encountered in wetlands, estuaries, and lagoons. Therefore, it is likely that the plants and microbes that are responsible for much of the treatment in these systems should be capable of adapting to the conditions encountered in RO concentrate from municipal water reuse projects.

Constructed wetland treatment may reduce the number of steps needed in a treatment train because physical, chemical, and biological removal mechanisms happen simultaneously in wetlands, whereas other treatment systems make use of only one set of processes at a time (Figure 5). This feature means that wetlands may be able to remove nutrients, trace metals, and trace organic contaminants without the need for additional engineered treatment. Many existing constructed wetlands were built to remove BOD and nitrogen, but wetlands purposely designed to treat trace organic contaminants (e.g., pharmaceuticals and personal care products)⁴⁹ and trace metals⁵⁰ have become more common recently. In particular, wetlands are now being designed to treat (i.e., “polish”) municipal wastewater effluent by simultaneously removing nitrate and trace organic contaminants.^{51,52} For example, open-water unit process wetlands reduced concentrations of the β -adrenergic blockers atenolol, metoprolol, and propranolol in

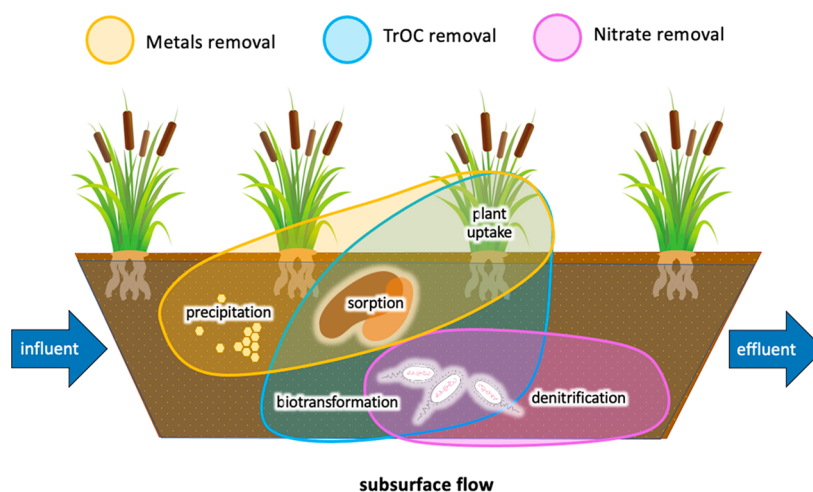


Figure 5. Main metals, trace organic contaminants, and nitrate removal mechanisms occurring simultaneously in subsurface-flow wetlands.

municipal wastewater effluent by approximately 85–95% via a combination of sunlight photolysis and microbial biotransformation and simultaneously removed more than 60% of the influent nitrate via denitrification.^{53,54} Open-water wetlands have also been demonstrated to reduce concentrations of nitrate by up to approximately 30–50% and trace organic contaminants by up to approximately 90% in RO concentrate at the pilot scale.^{20,55} In subsurface-flow wetlands treating municipal wastewater effluent, simultaneous biological removal of nitrate and trace organic contaminants to below detection limits (i.e., >90% removal) has also been observed.⁵² Although metals have not been the focus of research on constructed wetlands treating municipal wastewater effluent, copper and zinc concentrations have been reduced by up to 60–80% in wetlands treating industrial process water via plant uptake, sorption, and precipitation.^{50,56,57} Presumably, these removal mechanisms for metals could be achieved in wetlands that are also designed to remove nutrients and trace organics from RO concentrate.

Insight into the construction, operations, and maintenance costs of constructed wetlands for treating RO concentrate can be obtained by considering costs for systems employed in the treatment of municipal wastewater effluent. For example, the cost of wastewater effluent polishing at the 1600 ha surface-flow wetland system operated by the Tarrant Regional Water District and North Texas Municipal Water District on the Trinity River has been estimated to range from \$0.17 to \$0.30/m³.⁵⁸ The wetland area needed to treat each cubic meter of RO concentrate would likely be higher than that for wastewater because of the higher contaminant mass loads, but the overall area per mass of contaminant removed may be lower due to the lower volume of RO concentrate compared to wastewater effluent and because treatment may be more efficient with higher starting concentrations.⁵⁵ In contrast, ozone treatment combined with biological activated carbon for municipal wastewater costs approximately \$0.89/m³.⁵⁹ The annualized cost of using ozone to treat RO concentrate has been estimated at approximately 50% lower than the cost for wastewater when the volume reduction associated with the process is taken into account.³⁶

Land acquisition and site maintenance are important factors affecting the cost of wetland treatment because little else is needed for operation and maintenance.⁶⁰ Although no full-scale RO concentrate treatment systems have been built, they

already have been considered as options in several locations. Using design knowledge from projects in which constructed wetlands were used for effluent polishing, engineering consultants have concluded that surface-flow constructed wetlands would be less expensive than existing concentrate management technologies including evaporation ponds, injection wells, and zero liquid discharge systems.^{61,62} As more experience is gained through pilot projects, costs may decrease further as the footprint required for wetland treatment is reduced. Nonetheless, a lack of research and documentation on the performance of wetlands specifically designed to treat RO concentrate from potable water reuse projects is likely to slow their progress in the near future.

Beyond their attraction as cost-effective treatment systems, constructed wetlands are also appealing to decision-makers because members of the community appreciate the additional benefits that wetlands can provide. For example, constructed wetlands can be designed to create wildlife habitat,^{63,64} improve biodiversity,⁶⁵ and provide recreation and educational opportunities.⁶⁰ Results of a mixed-methods stakeholder analysis that assessed the importance of these ancillary factors in the selection of nutrient reduction strategies indicated that ancillary benefits (e.g., sea level rise protection, habitat creation) combined with the water quality benefits of constructed wetlands made them more favorable than other approaches like installation of advanced treatment technologies at existing wastewater treatment plants.⁴⁶ RO concentrate treatment wetlands that provide a public interaction component may be able to capitalize on the positive association that decision-makers have with wetlands, especially because they could enhance overall public support for the entire potable water reuse project.

■ EARLY-STAGE EXPERIMENTS ON THE USE OF CONSTRUCTED WETLANDS FOR RO CONCENTRATE TREATMENT

Although the ability of constructed wetlands to remove contaminants from wastewater effluent or effluent-impacted surface water in a cost-effective manner is well-established, the use of nature-based systems for treatment of RO concentrate is still in the early stages. However, several cities are experimenting with nature-based RO concentrate treatment

systems. These projects are at different stages of development and are motivated by diverse project drivers (Table 1).

Installation of a subsurface wetland system for treating RO concentrate from the Torrele Water Reuse Plant in Belgium was motivated by a desire to reduce permit fees associated with the discharge of nutrients and metals to the North Sea. In a pilot project that started in February 2011, RO concentrate was passed through a 28.5 m² wetland cell containing various species of willow trees (*Salix* spp.). During these experiments, a 30–35% reduction in the mass of phosphorus and nitrogen discharged by the system was observed. This load reduction would allow the Intermunicipal Water Company of Veurne-Ambacht (IWVA) to save a minimum of 20,000 €/yr in discharge fees if the wetland were to be scaled up to treat all of the RO concentrate from the reuse facility. The IWVA has designed a full-scale system and has applied for permits to construct a full-scale subsurface RO concentrate treatment system.

In the San Francisco Bay, nature-based RO concentrate treatment is also under consideration as the region's water utilities develop plans to build potable water reuse systems. RO concentrate discharge in combination with a lack of mixing could exacerbate water quality issues in the lower South San Francisco Bay. In an effort to reduce contaminant concentrations while expanding water reuse in the region, a local utility is investigating the use of three different types of nature-based RO concentrate treatment systems.⁷¹ A pilot-scale study of RO concentrate treatment using open-water unit process wetlands to remove nitrate and trace organic contaminants operated between 2017 and 2019.^{20,55} Removal of nutrients, trace metals, and trace organics is now being studied in subsurface-flow and floating wetland systems, with the aim of identifying an efficient and cost-effective RO concentrate treatment method.

Another motivating factor for cities considering constructed wetland treatment of RO concentrate is habitat creation. The City of Oxnard, California, has identified habitat creation as a driving factor for managing the city's RO concentrate. Although this coastal city has access to a deep ocean outfall for wastewater effluent discharge, they use their RO concentrate to create a brackish wetland habitat.⁷² The wetland currently treats a small fraction of the RO concentrate generated at the advanced water purification center, but additional RO concentrate from this facility may be used for wetland habitat creation and restoration in the future. The use of RO concentrate for habitat creation has also been employed by the city of Goodyear, Arizona. In this case, the city tested the use of RO concentrate from brackish groundwater desalination for the restoration of riparian habitat along the Gila river.⁷⁰ These projects highlight that habitat creation is not just a cobenefit but may be a driver when selecting RO concentrate management options.

These initial studies suggest that nature-based systems for RO concentrate management are attractive for multiple reasons: water quality benefits, cost savings, sea level rise adaptation, and habitat creation. However, the experiments conducted thus far do not provide a thorough understanding of the potential for removal of the full suite of contaminants of concern. For example, the fate of trace metals and/or trace organic contaminants have only been considered in a few studies despite the fact that these contaminants may cause adverse impacts in receiving waters. The data from this small set of pilot-scale projects are encouraging, but constructed

Table 1. Early Constructed Wetland Projects for RO Concentrate

facility name and location	project driver	project description	area/flow rate	year	performance findings	development stage
Torrele Water Reuse Facility Koksijde, Flanders, Belgium	lower costs associated with nutrient discharges into the North Sea ⁶⁶	subsurface-flow sand bed planted with willow trees	28.5 m ² 250 L/h	2011	30% nitrogen removal no phosphorus removal minimal BOD/TOC removal ⁶⁶	moving to full-scale construction ⁶⁷
Silicon Valley Advanced Water Purification Center San Jose, California, USA	decrease the loading of nitrogen and other contaminants into the San Francisco Bay	shallow, open-water wetland	400 m ² 1560 L/h	2017	28–47% nitrate removal (varied seasonally and by year) ⁵⁵ 0–90% trace organic contaminant removal (varies by contaminant) ²⁰	under consideration ²⁰
		subsurface-flow wetland	583 m ² 204 L/h	2017	none yet	study ongoing ⁵²
		floating wetlands	48 m ²	2019	none yet	study ongoing
Groundwater Recovery Enhancement and Treatment (GREAT) program Oxnard, California, USA	restoration of coastal brackish wetland habitat through discharge of treated RO concentrate	horizontal subsurface-flow wetland, vertical subsurface upflow wetlands and surface-flow wetlands/ponds	3155 L/h	2016	96% nitrate removal	built at pilot scale ⁶⁹
Goodyear Bullard Water Campus ⁷ Goodyear, Arizona, USA	restoration of riparian habitat along the Salt River; preventing discharge of high salinity RO concentrate from brackish water desalination into municipal wastewater	vertical-flow wetlands followed by surface-flow marsh	125 m ²	2010	93% Se removal ⁶⁸ 60–90% removal of arsenic, selenium, and chromium >90% removal of nitrate ⁷⁰	moving to full-scale construction

^aThe pilot wetland in Goodyear, Arizona treated RO concentrate from a brackish groundwater desalination facility.

wetlands are not yet considered a proven treatment technology for RO concentrate treatment. For nature-based RO concentrate management to become more popular, additional research is needed to inform design, demonstrate performance, and decrease costs.

RESEARCH TO ENABLE WIDESPREAD USE OF NATURE-BASED TREATMENT OF RO CONCENTRATE

Although nature-based treatment systems offer attractive advantages for managing RO concentrate produced by potable water reuse projects, engineers may not include them in their designs due to perceived shortcomings in their performance, the absence of standardized designs, and a lack of data from pilot- and full-scale systems. To enable more rapid deployment of nature-based systems, further research is needed in several key areas.

Among the challenges associated with nature-based RO concentrate treatment systems, land acquisition is likely to be a major impediment, especially in urban areas where land is expensive. This issue also has proven to be an impediment to the use of constructed wetlands for effluent polishing. For example, wetlands built to remove nitrate from municipal wastewater effluent often achieve median areal load reduction rates of approximately $100 \text{ mg-N m}^{-2} \text{ day}^{-1}$,⁷³ which translates to a need for approximately 18 ha of wetland to remove 90% of the nitrate from 1 million liters per day (MLD) of effluent. During summer, when rates of denitrification are high, open-water wetlands treating municipal wastewater effluent required approximately 1 ha per MLD (Figure 6).⁷⁴ For comparison,

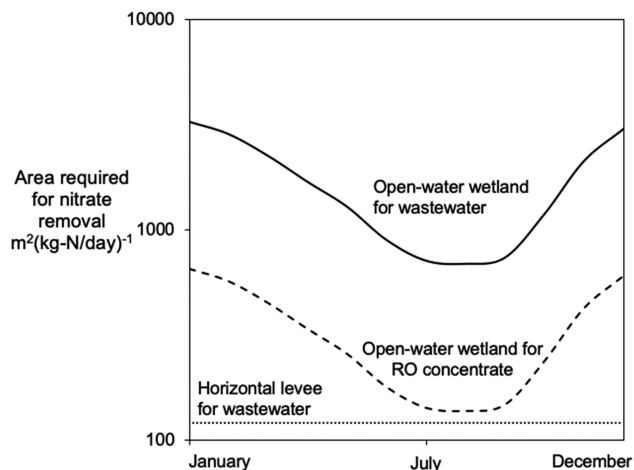


Figure 6. Effect of seasonality on footprint of constructed wetlands for removal of nitrate from municipal wastewater effluent and RO concentrate. Figure drawn using data from previous references.^{52,54,55}

large municipal wastewater treatment plants typically take up less than 0.5 ha for each MLD of design capacity (calculated based on wet-weather design capacity of 11 large wastewater treatment plants for which area estimates were available).

Treating RO concentrate instead of municipal wastewater effluent has the potential to reduce the footprint of constructed wetlands because the volume of water requiring treatment could be up to 85% lower, but the relationship between wastewater volume and required area is not one-to-one. In the case of nitrate, removal from RO concentrate by open-water wetlands was limited by the surface area available for

photosynthesis, which provided organic carbon to fuel denitrification, whereas carbon was not limiting in open-water wetlands treating municipal wastewater effluent with lower nitrate concentrations.⁵⁵ For trace organic contaminants, phototransformation may be slower in RO concentrate because the higher concentrations of organic matter increase light screening and scavenge reactive intermediates.²⁰ Other parameters may also influence RO concentrate treatment in a manner that is different from what is observed during the treatment of municipal wastewater. For instance, elevated sulfate concentrations could affect the microbial community in subsurface-flow wetlands and result in sulfide toxicity. Research is needed to determine controlling factors for RO concentrate treatment efficiency and to identify strategies for optimizing performance.

Further research is also needed to assess the fate of toxic trace metals in constructed wetland systems and to optimize their treatment. Removal of metals has been observed in wetland systems, but with varying results. For instance, metals may be sequestered in wetland sediments in the summer but remobilized during winter months if sulfide oxidation occurs.⁷⁵ Metal sulfides formed under anaerobic conditions may also undergo conversion to more stable sulfide minerals, such as pyrite, over time,⁷⁶ effectively immobilizing metals. Research is needed to understand mechanisms of metal removal and to design constructed wetlands for long-term metals treatment.

In addition to optimizing the efficiency of constructed wetlands, another strategy to reduce the footprint of RO concentrate treatment involves the use of hybrid systems. For instance, ozone pretreatment upstream of a pilot-scale open-water wetland resulted in trace organic contaminant removal in a smaller footprint than could be achieved by wetlands alone, while simultaneously removing contaminants that were less reactive with ozone.²⁰ Intensification of wetlands by incorporating aeration, electrolysis, or other modifications has also been proposed, although this approach requires balancing the modifications with maintaining the unique advantages of constructed wetlands.⁷⁷ An example of integrating electrochemical and wetland treatment approaches involves introduction of an electrically conductive material into the constructed wetland subsurface. In a biofilter system referred to as METlands (a hybrid microbial electrochemical technology/constructed wetland), the conductive material (e.g., electroconductive biochar or electroconductive coke) acted as an electron sink that increased biological removal rates for chemical oxygen demand (COD) and total nitrogen by connecting oxic and anoxic zones with a conductive material.^{78–80} The effects of wetland intensification strategies such as the METland on removal of metals and trace organic contaminants requires further research.

The decrease in performance of nature-based systems during winter is often seen as a severe limitation because the rates of microbial processes such as denitrification decrease with temperature. Although discharge permits often require uniform performance over the entire year, this concern may be less important in the context of water reuse because water demand tends to be highest in summer, when nature-based systems are most effective. Thus, advanced treatment facilities may curtail operations or be idled during winter when wetland treatment efficacy decreases. This may be especially relevant in cities considering direct potable reuse because extra water produced during winter cannot be stored. If the fraction of wastewater recycled decreases during winter, higher dilution would be

achieved at the discharge point and RO concentrate treatment may no longer be necessary. For example, the Livermore Water Reclamation Plant in Livermore, California, processes over 22 MLD of wastewater, almost all of which is reused in summer.⁸¹ During winter, a large fraction of the facility's wastewater effluent is discharged to San Francisco Bay because the recycling plant is expensive to operate and the additional water is not needed.

Where wintertime treatment is necessary, subsurface systems that are insulated from air temperatures may continue to provide treatment year-round, despite wastewater temperatures as low as 10 °C.^{82,83} For instance, a pilot-scale subsurface-flow constructed wetland treating municipal wastewater effluent maintained water temperatures above approximately 10 °C in the winter and exhibited little seasonal variation in nitrate removal, which occurred primarily via denitrification.^{52,84} Similarly, COD removal was maintained at temperatures as low as 4 °C in planted microcosms simulating subsurface wetlands.⁸⁵ In contrast, nitrification activity has been found to decrease in subsurface wetlands in the winter,⁸⁶ and in some cases, denitrification rates have slowed in subsurface wetlands at temperatures below 15 °C.⁸⁷ Surface-flow systems are subject to greater variations in treatment efficacy because they are more directly influenced by the ambient temperature and may rely on sunlight photolysis for trace organic contaminant removal. As a result, these systems may be most useful in locations that require minimal winter treatment or where temperatures are relatively warm year-round.

Future research may elucidate other opportunities for integrating nature-based systems into RO concentrate management. For instance, diatoms exhibited an ability to remove nutrients from RO concentrate while simultaneously reducing concentrations of silicate, the ion that often limits water recovery from concentrate due to its role in membrane fouling.⁸⁸ Although these treatment systems required long retention times (i.e., several days to weeks) and lacked the ancillary benefits of constructed wetlands, they suggest the potential for using nature-based systems to enable greater water recovery from water reuse. Wetlands could also be used as an alternative to evaporation ponds because high rates of evapotranspiration are achieved by salt-tolerant plants [e.g., saltcedar (*Tamarix ramosissima*) can tolerate TDS as high as 20,000 mg/L].⁸⁹ Thus, marine or halophytic plant species could become integral to near zero-liquid discharge systems, providing cost-effective volume reduction while simultaneously removing contaminants.

The potential of constructed wetlands to serve as RO concentrate treatment systems merits further attention. Although the complexities inherent in nature-based treatment systems have historically forced engineers to employ empirical (i.e., black-box) models to predict treatment capabilities, a new research focus on understanding underlying mechanisms and optimizing wetland design is needed to support the adoption of these efficient and desirable treatment technologies and to enable widespread potable water reuse.

■ ASSOCIATED CONTENT

■ Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsenvironau.1c00013>.

Representative contaminant concentrations in RO concentrate and details of analysis of water-stressed cities (PDF)

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Notes

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