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### Authors

Keller, Arturo A  
Su, Yiming  
Jassby, David

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# Direct Potable Reuse: Are We Ready? A Review of Technological, Economic, and Environmental Considerations

Arturo A. Keller,\* Yiming Su, and David Jassby

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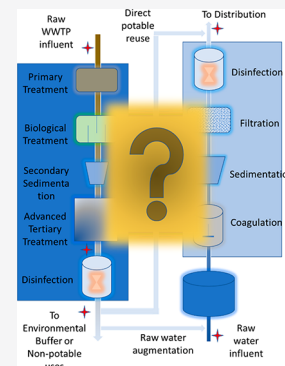
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**ABSTRACT:** Meeting future water demand will require serious consideration of direct potable reuse (DPR) for many water agencies. There has been tremendous progress in the technologies needed to address the concerns that conventional and novel water contaminants pose. Yet, to date, only a few relatively small DPR operations have been installed. As we get closer to the point where regulations are finalized and serious investments are planned, there is a need to ask: Are we ready? In this Review, we explore the technological, economic, and environmental considerations associated with DPR. In particular, we focus on the status of technologies for addressing the most challenging water pollutants, the cost estimates for DPR, and the energy demand and associated implications of DPR. We find that, although the technologies are nearly ready for DPR, the most critical issue will be real-time monitoring of a number of molecules that pose distinct challenges to advanced treatment trains. In addition, there is a need to consider emergency planning, both in terms of emergency buffer reservoir(s) and planning for situations in which the treated water does not meet specifications. Since any advanced treatment train will result in a significant increase in embedded energy, it will be particularly important to plan for renewable energy to minimize environmental impacts.

**KEYWORDS:** Emerging pollutants, water quality, reuse, monitoring, sensors



## INTRODUCTION

Rising temperatures, increasing variability in precipitation patterns, more extended droughts, growing populations, and limited alternatives for new traditional water sources in semiarid regions with frequent water scarcity episodes, such as the southwestern US and around the world, are major drivers for a serious consideration of direct potable reuse (DPR).<sup>1</sup> The concept of “closing the loop”<sup>2</sup> in terms of the urban water cycle, as part of the “Fourth Water Revolution”, considers potable water reuse as one of its key pillars.<sup>3</sup> The employment of high-quality treated wastewater for planned indirect potable reuse (IPR), by employing an environmental buffer as in the case of groundwater recharge or reservoir water augmentation, is already a current practice in many regions (e.g., NEWater Singapore,<sup>4,5</sup> Orange County Water District Groundwater Replenishment System (California),<sup>6</sup> Upper Occoquan Service Authority (Virginia),<sup>7</sup> Montebello Forebay Groundwater Recharge Project (Los Angeles, California), Western Corridor Recycled Water Scheme (South East Queensland, Australia)<sup>8</sup>). Summaries of several pilot or operational DPR and IPR systems are provided by Guo et al.<sup>9</sup> However, the direct introduction of high-quality treated effluent to a public water system or for raw water augmentation immediately upstream of a water treatment plant, as planned in DPR, requires additional considerations.<sup>10–12</sup>

Given the high cost of treating water for potable reuse, the first step must be to implement a community-wide water conservation plan. There are many success stories, throughout

California, the US, and around the world,<sup>13–20</sup> that implement appliances and fixtures with higher water (and energy) use efficiency, convert landscapes to vegetation that requires almost no watering or to permeable hardscapes, eliminate leaks throughout the entire water system, and provide incentives to consumers.<sup>21,22</sup>

The employment of an environmental buffer, essentially storing the treated water in a large compartment, either an aquifer or a reservoir, provides a dilution of constituents that may be present in the treated water as well as time for natural attenuation and detection of any unexpected changes in water quality.<sup>23–26</sup> In California, Title 22 requires that water for IPR be treated with reverse osmosis (RO) and an advanced oxidation process (AOP) plus a minimum of two months of subsurface travel time or reservoir retention time with specific considerations for dilution ratios.<sup>27</sup> In fact, a longer residence time, greater than 6 months, is desirable; less than two months requires additional considerations. The goal is to ensure that the concentrations of inorganic and organic chemicals are below their respective maximum concentration levels (MCLs), water quality objectives (WQOs), or notification levels (NLs), that

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levels of enteric viruses have a 12-log reduction, and that levels of *Giardia* cysts and *Cryptosporidium* oocysts have a 10-log reduction.<sup>27</sup> However, many communities do not have a nearby reservoir or aquifer capacity for the 2+ months of residence time or dilution ratios, or the hydrogeochemistry may be unsuitable for IPR. In addition, the elimination of the environmental buffer may be more cost-efficient than IPR.<sup>28</sup>

Many communities that extract their raw water from rivers, such as the Ohio River, the Colorado River, and many others around the world, already conduct unplanned IPR.<sup>29,30</sup> Upstream communities discharge their treated wastewater into the river, in many cases with only secondary treatment, which is diluted and then extracted downstream by other communities, with much less than the recommended two months of residence time. Technologies, regulations, and management approaches that are relevant for DPR are thus valid for many other locations.

The elimination of the environmental buffer raises concerns with regards to illegal or accidental discharges of chemicals into the wastewater sewer collection stream or a stormwater conveyance that connects to the wastewater system that may not be regularly monitored.<sup>31,32</sup> Concerns about water security also increase when there is a short time to react to such events. The current COVID-19 pandemic has been a wake-up call to also address unexpected, novel pathogens that may be present in wastewater.<sup>33–42</sup> Thus, even for DPR, a small emergency buffer should be built into the design to handle any flows that are suspected of not meeting drinking water quality goals and a means to divert flow temporarily to a receiving water body until goals are met.<sup>24,43,44</sup>

Closing the loop raises concerns with regards to the accumulation of common constituents (e.g., chloride, nitrate, borate) throughout the urban water system, even in the case of IPR, where the constituents can increase in the aquifer or reservoir. Current treatment levels for most wastewater treatment plants (WWTPs) do not entirely remove chemicals of emerging concern (CECs) such as pharmaceuticals and personal care products (PPCPs), per- and polyfluoroalkyl substances (PFASs), disinfection byproducts (DBPs), and nanomaterials and their residual ions as well as many industrial chemicals that are not regulated or monitored (e.g., acetone).<sup>6,45–50</sup> Conventional WWTPs are not required to remove these contaminants, and although many can be partially removed from the effluent, residuals can remain. The potential effects of chronic exposure to low-level residuals of these contaminants have not been fully characterized.<sup>51–56</sup> Thus, there is a need for advanced treatment and real-time monitoring of low (ng/L) levels of these currently unregulated contaminants. There is particular concern with low molecular mass (<200 Da) neutral chemicals that may not be removed entirely by RO and/or AOPs,<sup>6</sup> such as those presented in Table 1.

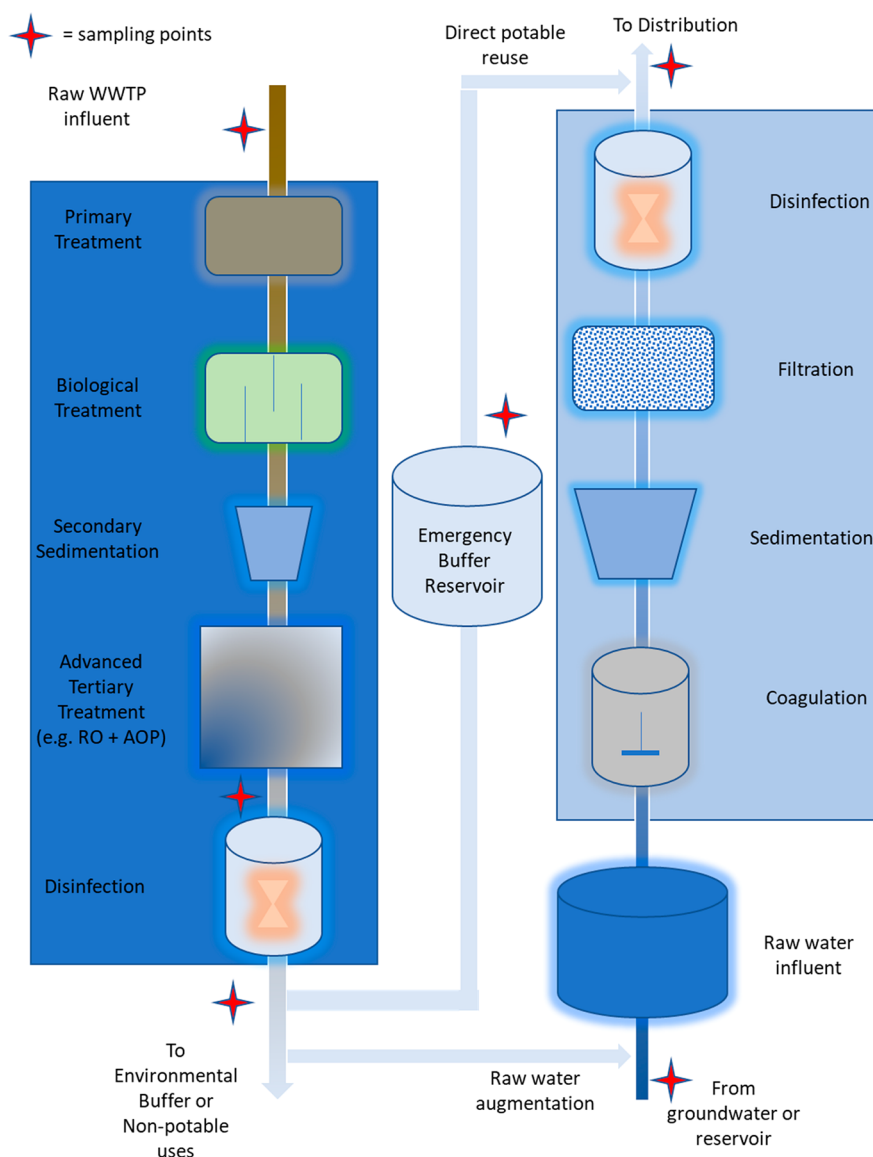
To address these concerns, advanced online monitoring of chemicals and rapid off-line analytical capabilities will be necessary.<sup>57–63</sup> Online monitoring of unregulated CECs cannot rely on currently available total organic carbon (TOC) sensors, even if they can detect organic molecules at 0.5 mg/L, or even 0.1 mg/L, since that is still orders of magnitude greater than the ng/L levels at which the CECs may be present. Although nontargeted and semitargeted analysis can be employed, these methods are capital intensive and have high labor costs,<sup>64</sup> requiring novel tools for real-time monitoring. In the long term, online monitoring will have to be reliable, convenient, and affordable for WWTPs. Several sampling points will be needed along the treatment trains and distribution systems (Figure 1).

**Table 1. Partial List of Chemicals of Potential Concern after Advanced Treatment**

chemical	potential effects	reference
Inorganics		
arsenic (arsenite)	cancer and other diseases	65
boron	developmental effects, toxic for plants	66
bromate	carcinogen, developmental neurotoxicity, negative effects on crop plants	67, 68
chlorate	carcinogen	69–71
Organics		
1,4-dioxane	liver and kidney damage	72
2,4,6-trichlorophenol	carcinogen	71
2,4,6-trichloroanisole	organoleptic threshold	73, 74
2,4-dichloroanisole	organoleptic threshold	73, 74
2-methyl-isobomeol	organoleptic threshold	73, 74
acetaldehyde	carcinogen	71
acetone and other VOCs	taste and odor	75
atenolol	developmental toxicity	76, 77
benzoquinones	DNA and protein damage	78
bisphenol A	endocrine disruption	79
bromoacetonitrile	carcinogen	71
bromoform	carcinogens	80, 81
carbamazepine	toxic to pregnant women and fetuses	82–84
chloroacetonitrile	carcinogen	71, 80, 81
chloroform	carcinogens	80, 81
dichloromethane	carcinogen	71
enedials	damage to hepatic proteins	85, 86
estrogen	endocrine disruption	87
fipronil	liver toxicity	88, 89
formaldehyde	carcinogen	71
geosmin	organoleptic threshold	73, 74
glyoxal	oxidative stress	90, 91
halogenated disinfection byproducts	carcinogens, mutagens	92
imidacloprid	reproductive toxicity	88, 93
nitrosamines	carcinogens	71, 92, 94, 95
N-nitrosodimethylamine (NDMA)	hepatotoxic and carcinogen	96
oxoenals	damage to hepatic proteins	85, 86
perfluoroalkyl substances (PFASs)	thyroid disorders, cancer	97, 98
tri(2-chloroethyl) phosphate (TCEP)	carcinogen	76, 99–101
tris(1-chloro-2-propyl) phosphate (TCPP)	DNA-damage potential	102
triclosan	endocrine disruption	103

In addition, raw wastewater contains a high level of pathogens, including enteric bacteria, viruses, and protozoa. Raw wastewater may contain virus concentrations of 107 to 109 gene copies per liter for qPCR-based analyses, virus concentrations of 3 to 1300 per liter for culture-based analyses, and protozoa concentrations of 6 to 17 000 per liter.<sup>104</sup> Their mass loading significantly decreases through the conventional WWTP process, and most can be removed by advanced technologies such as RO and AOPs; however, there is a need to be vigilant and implement online monitoring to avoid the risk of exposing the population to these pathogens.<sup>105,106</sup>

Sewersheds that include a significant number of industrial and commercial activity will need additional administrative measures, such as identification of all possible sources, inventories of chemicals in use, regular updating of inventories, enforcement of



**Figure 1.** Process diagram for direct and indirect potable reuse with potential sampling locations.

pretreatment (i.e., treatment at the industrial or commercial source), and education of all employees as to the potential effects of any discharge into the sewer that could result in unexpected chemicals passing through the treatment systems and into the water distribution system.

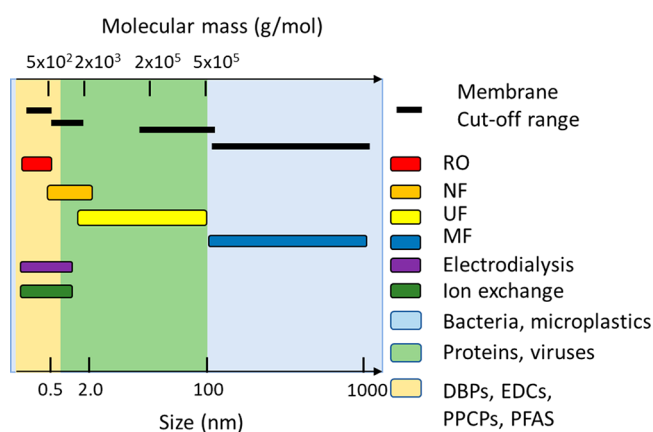
Other important considerations for DPR include (1) very low total dissolved solids (TDS) after RO, which requires proper dosing of dissolved solids to avoid corrosion problems that can result in distribution system failures (e.g., leaking old distribution pipes) and possible leaching of Pb and Cu;<sup>107–109</sup> (2) disposal of the RO brine, which can be a challenge for communities far from the ocean and with sensitive nearby habitats;<sup>110,111</sup> (3) higher embedded energy by incorporating RO, AOP, chlorination, and any additional processes after the conventional WWTP and the corresponding impacts on greenhouse gases and other issues;<sup>11</sup> (4) additional pumping costs to transport water upgradient to the high points of the watersheds;<sup>112</sup> (5) increased capital and operating costs; (6) additional complexity and training within the treatment plants; (7) redundant capital investment in key processes (e.g., RO, NF, activated carbon) to ensure these treatment steps are 100%

operational; (8) the need for close cooperation and coordination between the WWTP(s) and the agencies in charge of raw water treatment and distribution to maintain uniform flows, manage emergency reservoir(s), and implement action plans in case the treated effluent does not meet water quality objectives.

In this Review, we consider the: (1) technologies available for advanced tertiary treatment applicable for DPR and their ability to address the contaminants indicated in Table 1; (2) tools for a cost-effectiveness comparison; (3) energy and other considerations for advanced tertiary treatment trains; (4) the critical role for sensors that are being proposed to meet the challenges of real-time monitoring at multiple locations for chemicals of potential concern as well as for pathogens; (5) final recommendations on our readiness to deploy DPR.

### ■ TREATMENT TECHNOLOGIES: EFFECTIVENESS AND CHALLENGES

Low-pressure microfiltration (MF) and ultrafiltration (UF) membranes remove a significant fraction of particulates and large (>200 Da) organic molecules remaining after conventional (primary and secondary) wastewater treatment (Figure 2).



**Figure 2.** Size and molecular mass of pollutants and pathogens removed by different techniques. DBPs = disinfection byproducts, EDCs = endocrine disrupting chemicals, PPCPs = pharmaceuticals and personal care products, and PFASs = per- and polyfluoroalkyl substances. Membrane cutoff ranges from ref 145.

Nanofiltration (NF) membranes can remove many additional small molecules but do not remove low molecular mass monovalent ions and cannot alone meet TDS requirements for DPR. Reverse osmosis (RO) membranes remove most of the remaining organic molecules (down to 150–200 Da) and divalent ions as well as a fraction of the monovalent ions, meeting TDS requirements. More than 90% of semivolatiles and the majority of PPCPs and PFAS are well rejected by NF and RO.<sup>113–115</sup> However, a significant fraction of small molecules, such as chloroform, bromoform, bromodichloromethane, and dibromochloromethane, are poorly rejected.<sup>116</sup> Short-chain PFAS, TCEP, and other phosphoric acid esters can also pass to the permeate at detectable concentrations.<sup>76,117</sup> A major challenge for membrane technologies, in particular for RO and NF membranes, is the irreversible adsorption of natural organic matter and the formation of biofilms, which obstruct the passage of water, known as fouling.<sup>118–120</sup> Particulate deposition can also result in fouling.<sup>121</sup> Deposits of inorganic ions (i.e., scaling) can also affect membrane performance over time, increasing the pressure needed to maintain water flux.<sup>121</sup> Considerable effort has been spent on reducing fouling and scaling via membrane design, chemical additions, and pretreatment.<sup>122,123</sup> Biofilm formation (biofouling) can be limited using disinfectants (e.g., chloramine), scaling by lowering pH and with antiscalants, and NOM and particle deposition via UF and MF pretreatment.<sup>123,124</sup>

Significant advances have been made in membrane design and performance using simulation tools from the molecular level to system wide optimization. Molecular dynamics simulation of membranes can serve to evaluate different membrane materials in terms of treatment performance, energy requirements, and fouling. In these studies, there is generally a focus on inorganic ion (e.g., Na<sup>+</sup>, Cl<sup>-</sup>) transport through different membrane materials. Molecular dynamics can serve to guide membrane material development (e.g., use of functionalized carbon nanotubes,<sup>125</sup> graphene,<sup>126</sup> and boron nitride nanotubes<sup>127,128</sup> for higher ion rejection and water flow rates), selectivity based on membrane characteristics (e.g., pore size, membrane structure and thickness, surface modifications), water and ion dynamics, and fouling.<sup>120,129</sup> Membrane performance simulations at larger scales with specific input water conditions (e.g., salinity, operating pressure and temperature, flow rate) can be

used to select among the many types of membranes available for water reuse.<sup>130</sup> Simulations are also used to optimize system design, consider single- vs two-step configurations, and incorporate energy recovery devices and pretreatment as a means to reduce energy requirements as well as other costs.<sup>131</sup> Process optimization can also be achieved using adaptive control strategies for backwash frequency, disinfectant addition, RO flux, water recovery (i.e., fraction of feedwater present in treated water), pH, and antiscalants dose.<sup>132</sup> In addition to lowering energy and chemical use, optimization can also increase membrane lifetime and important operating costs.<sup>132</sup>

AOPs can generally be divided into O<sub>3</sub>-based, UV-based, plasma-based, and electrochemical approaches and can remove many of the remaining organic molecules that pass through the RO membranes, albeit with different effectiveness. For AOPs, there have been a number of studies implementing artificial neural networks (ANN) for determining removal efficiencies, operational control, and optimization<sup>133</sup> as well as for predicting performance.<sup>134</sup> In terms of removal efficiencies, the approach to date is to consider specific target chemicals (e.g., textile dyes,<sup>135–138</sup> pharmaceuticals,<sup>139</sup> pesticides, MTBE<sup>140</sup>) generally using a specific AOP,<sup>141</sup> but there is a need for a more comprehensive approach. Overall, there is a major research gap in the development of tools for predicting cost and performance of AOPs in part due to the wide range of chemicals considered, processes, and operational conditions.

A low-energy alternative to AOPs is granular activated carbon (GAC), which can also be biologically enhanced (BAC) to increase removal efficiency. For BAC, models have been developed to predict the adsorption and biodegradation performance as a function of operational conditions with good accuracy.<sup>142,143</sup> Modeling was more accurate for poorly to moderately adsorbing trace organics, indicating that the model is more accurate for biodegradation if the kinetics are known.<sup>143</sup> A few studies have modeled the combination of ozone and BAC to predict the removal efficiency of trace organics in water reuse.<sup>144</sup> However, there is also a need for performance and optimization models as well as to more accurately predict costs and energy requirements based on operating conditions.

The focus of this section is on those molecules that pose significant challenges for DPR, such as small monovalent ions and organics.

**Small Inorganic Molecules.** The majority of heavy metals (e.g., copper, chromium, nickel, etc.) are found in an ionic form at neutral pH, are well-rejected by RO membranes, and are not anticipated to be an issue in most streams.<sup>146</sup> However, As(III) and boron, which are found as uncharged oxides/hydroxides at neutral pH, are not well-rejected by RO membranes and could pose a danger (although boron is a problem primarily for plants and not mammals).<sup>147–150</sup>

Arsenic is naturally present in many minerals, and although typically dissolved concentrations are low, it can be found at up to 2000 μg/L in some groundwater sources. While many water supply systems in the US and around the world remove a significant fraction of the arsenic load from their raw water, it could be introduced into the system by domestic or industrial users using local wells; careful monitoring is needed to avoid buildup of As concentrations in a DPR system. Even at low levels, arsenic can lead to a number of cancers (skin, lungs, bladder, liver, kidney), and the effects may not be observed for years until they are irreversible.<sup>151</sup> Although a fraction of the particulate and dissolved arsenic can be removed by coagulation or adsorption, it may not be sufficient. MF/UF have pore sizes

that are too large to reject the ionic form.<sup>152</sup> RO can reject 60–98% As(V) and/or 65–80% As(III) in the influent; thus, in some cases, two or more steps are needed to reach WQOs.<sup>147</sup> If the source water is anoxic, preoxidation of arsenite to arsenate is recommended, since at typical pH arsenite is present as a neutral form, while arsenate is anionic, increasing rejection. However, if pH adjustment is needed to remove arsenate, it may be more cost-effective to have two-step RO filtration.<sup>147,148</sup>

Sea water (SWRO) and brackish water (BWRO) polyamide membranes reject boron by 80–93% and 30–80%, respectively. Consequently, a single-pass RO process is usually unable to remove boron down to WQOs. Boron removal can be improved by (a) pre- and post-treatment techniques;<sup>153</sup> (b) double-pass RO;<sup>153</sup> (c) membrane modification;<sup>149,150</sup> (d) electroactive membranes.<sup>154</sup> Removal of boron using a second pass or alternative method increases water cost by approximately 10–20%.<sup>153</sup>

While Br<sup>-</sup> by itself does not pose a concern, the potential formation of bromate (BrO<sub>3</sub><sup>-</sup>) and other brominated disinfection byproducts (Br-DBPs) during ozone-based disinfection processes can be a problem if local surface or groundwater contains high levels of Br<sup>-</sup> (e.g., Australia). While Br<sup>-</sup> removal efficiency of the RO process usually ranges from 93% to 99.3% when total dissolved salt (TDS) concentrations are low, it could decline to 74% when TDSs increased to 400 ppm.<sup>155</sup> In terms of Br-DBPs, their removal efficiencies in the RO process are relatively low, ranging from 0 (i.e., bromomethane) to 80% (i.e., 1,2-dibromoethane).<sup>156</sup> Moreover, while AOP processes can degrade Br-DBPs, the risk of forming bromate is quite high. To lower the levels of Br-DBPs, it is important to remove Br<sup>-</sup> before the AOP process. Double-pass RO is an option,<sup>157</sup> and new approaches such as Br<sup>-</sup> selective ion-exchange resin and membrane capacitive deionization are being explored.<sup>157,158</sup>

**Low Molecular Mass Volatile, Semivolatile, and Nonvolatile Organic Compounds.** The (secondary) biological treatment in a WWTP can efficiently remove many volatile organic compounds (VOCs) or semi-VOCs (e.g., 96.7 ± 1.6% of 2-butanone, 91.7 ± 5.9% of acetone), but the degradation of halogenated VOCs declines significantly and may be even nil (e.g., chloroform, *cis*-1,3-dichloropropene, and 1,2-dichloroethane).<sup>159</sup> Some may be lost to the atmosphere, and a reaeration step may help lower their concentrations. VOCs are poorly rejected by RO due to their small molecular mass and neutral charge.<sup>156</sup> For instance, the rejection ratios of acetonitrile, acrylonitrile, chloroethane, 1,1-dichloroethene, and trichloroethene are 23 ± 10.6%, 18 ± 8.1%, 15 ± 3.6%, 17 ± 9%, and 46 ± 2.9%, respectively.<sup>156</sup> Moreover, membrane fouling caused by many VOCs significantly decreases water flux. For instance, fouling caused by hydroquinone, 4-nitrophenol, and 4-chlorophenol reduced permeate water flux by 2.7%, 4.8%, and 8.6%, respectively,<sup>160</sup> indicating that high loads of these pollutants may deteriorate membrane performance.

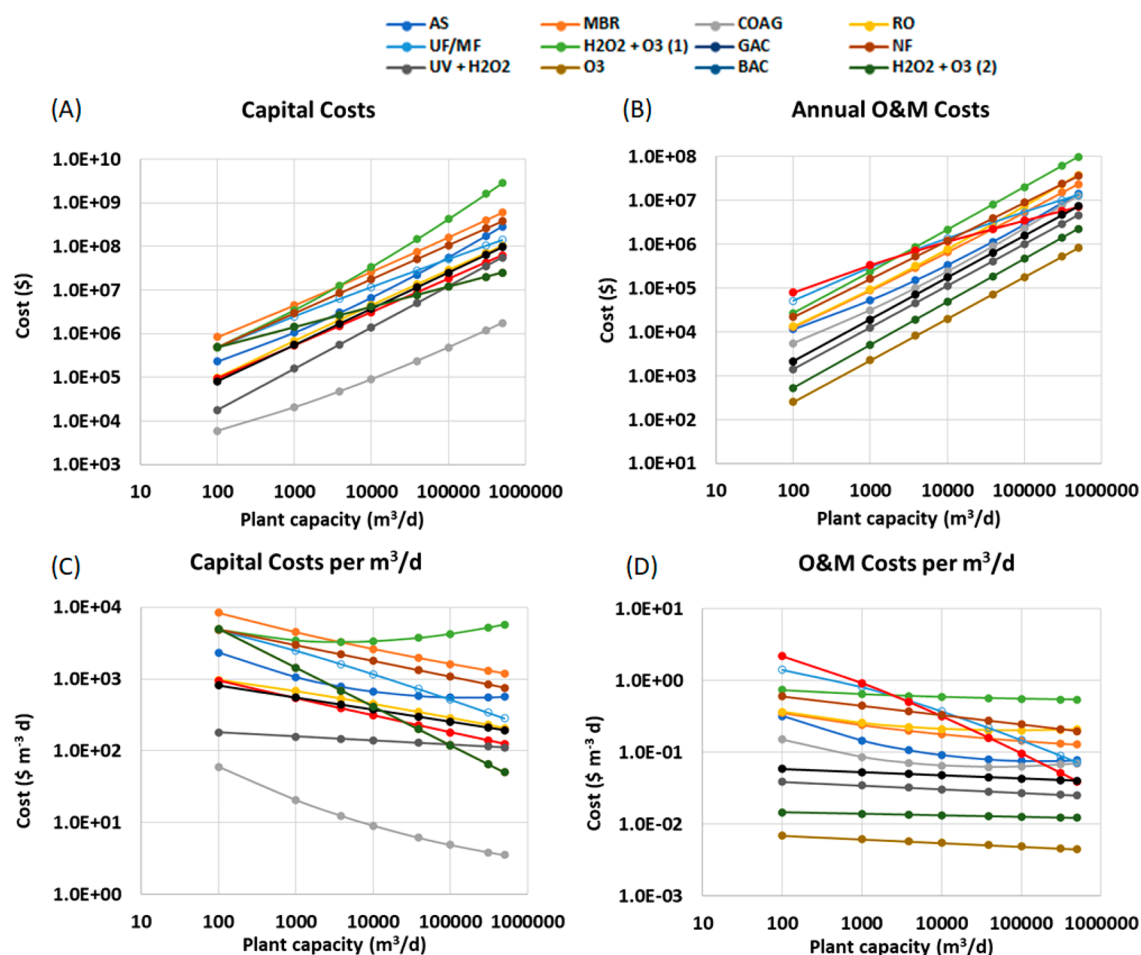
GAC and other porous materials (e.g., zeolites) can remove most organic compounds to a large extent, particularly if it is biologically active.<sup>161–163</sup> VOCs can be removed by interception, hydrophobic interactions, electrostatic interaction, multiple hydrogen bonding, and various types of  $\pi\cdots\text{Cl}$  interactions.<sup>161,162</sup> The massive number of adsorption sites on the huge surface areas of these porous materials are responsible for the large removal capacity and nanoadsorbents are in development for specific classes of pollutants.<sup>164,165</sup> Biofilms can form on the GAC or other adsorbents to further increase the

removal capacity of many halogenated VOCs (e.g., methylene chloride, chlorobenzene, carbon tetrachloride, tetrachloroethylene, 95% *o*-chlorophenol, trichloroethene, *cis*-1,2-dichloroethylene, *trans*-1,2-DCE, and vinyl chloride).<sup>166,167</sup> Removal and destruction of volatile and semivolatile organic compounds can be enhanced by combining it with ozone, since activated carbon can serve to produce hydroxyl radicals from ozone.<sup>168</sup> This can also remove many of the precursors to disinfection byproducts. To increase the effectiveness, combinations of AOPs with GAC, such as O<sub>3</sub> + GAC + UV/H<sub>2</sub>O<sub>2</sub>, can be used to enhance the destruction of the organics.<sup>169</sup> The most cost-effective configuration for many organics was O<sub>3</sub> + GAC + O<sub>3</sub>.<sup>169</sup> Photoreactors, in some cases using nano-TiO<sub>2</sub> and UV, are being considered as alternatives for the degradation of challenging organics.<sup>170</sup> Many PPCPs can be removed by these processes, either GAC alone or in combination with AOPs.<sup>171–174</sup> PFASs can also be removed to a certain extent with GAC, although short-chain PFASs exhibited desorption, and branched PFASs and those with carboxylic acids exhibited lower adsorption than PFASs that are linear or contain sulfonic acids.<sup>175</sup>

Electrochemical systems can also function as advanced oxidation/reduction processes for VOC degradation. Over 80% of chloroform, benzene, toluene, and trichloroethene in solution can be oxidized within 2 h on Ir/Pd doped titanium electrodes.<sup>176</sup> In addition, halogenated VOCs can be efficiently electrochemically reduced to halogen-free products on different types of cathodes at relatively low potentials from -0.3 to -1.4 V (versus standard hydrogen electrode).<sup>177,178</sup> Thus, heterogeneous AOP processes, including the electrochemical system, are increasingly widely adopted for organic pollutant removal.<sup>179,180</sup> In these processes, the reduction/oxidation rate is positively correlated to the surface areas of the catalysts; thus, the application of nanoscale materials becomes attractive.<sup>180</sup> The introduction of nanomaterials not only concentrates the trace pollutants on electrodes but also delivers electrons onto adsorbed pollutants, which can significantly increase pollutant removal performance and energy efficacy as well.<sup>181,182</sup> In the future, the heterogeneous AOP process for VOC degradation may play an important role in DPR.

Two molecules pose particular challenges, namely, *N*-nitrosodimethylamine (NDMA) and 1,4-dioxane. NDMA (74.08 Da), both a former industrial chemical and a chloramination byproduct, is detected in drinking water and wastewater treatment plants, typically at levels below 100 ng/L,<sup>183–185</sup> but in industrial areas, concentrations up to 8230 ng/L have been observed.<sup>186</sup> The USEPA screening level is 0.42 ng/L. The removal efficiency of NDMA and other nitrosamines in conventional WWTP biological processes ranges from 0% to 96% from plant to plant and, even the same plant, can experience a wide range of removal,<sup>183,184</sup> making it particularly challenging for DPR. The minimization of nitrite yield in the biological process and/or the addition of sand filtration after secondary sedimentation can reduce NDMA levels in secondary treatment effluent,<sup>183,187</sup> but it still remains as a challenge to fully eliminate it through biological treatment. Due to the existence of NDMA precursors in secondary effluent, it is very likely that NDMA and other nitrosamines would be generated again during disinfection if they are not fully removed before this step.

Treatment options, before feeding the secondary treatment effluent into an RO system, include filtration (i.e., sand filtration, GAC adsorption, and nanofiltration) and coagulation/flocculation, which all exhibit limited to moderate removal capacity for



**Figure 3.** Estimated (A) total capital costs; (B) annual operating expenses; (C) capital costs per 1000 m<sup>3</sup> of capacity; (D) operating annual expenses per 1000 m<sup>3</sup>. From Guo et al.: RO = reverse osmosis, UF/MF = ultrafiltration/microfiltration, MBR = membrane bioreactor, AS = activated sludge, COAG = coagulation, H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub> (1) = peroxone, and GAC = granular activated carbon.<sup>9</sup> From Plumlee et al.: NF = nanofiltration, UV + H<sub>2</sub>O<sub>2</sub> = ultraviolet + hydrogen peroxide, O<sub>3</sub> = ozone, BAC = biologically activated carbon, and H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub> (2) = peroxone.<sup>218</sup> All costs adjusted for inflation to 2021.

NDMA (16–44% with an NDMA concentration ranging between 14 and 370 ng/L) and NDMA precursors.<sup>188</sup> Removal efficiency of these processes varied widely depending on raw water qualities and the specific biological treatment process. In fact, studies report that coagulant dosing and biofiltration can contribute to the formation of NDMA precursors.<sup>188</sup> Nanofiltration (NF) can remove up to 90% of NDMA precursors,<sup>189</sup> but there could be significant leaching of NDMA precursors (~180–450 ng/L in permeate) after membrane fouling.<sup>189</sup>

Due to its small molecular radius (0.248 nm), high hydrophilicity, and neutral charge at pH 6–8, NDMA is not effectively removed by RO membranes.<sup>190,191</sup> To enhance NDMA removal, strategies such as (a) heat treatment of three types of RO membranes (HYDRA, ESPAB, and ESPA2) improved NDMA rejection from 74%, 62%, and 53% to 88%, 79%, and 62%, respectively;<sup>192</sup> (b) plugging the RO membrane with dodecylamine increased rejection from 42% to 81%;<sup>193</sup> (c) modification of RO with graphene oxide nanosheets enhanced NDMA rejection from 76.5% to 82.7%.<sup>194</sup> However, while these strategies increased NDMA rejection, the trade-off was that the membranes exhibited lower permeability.<sup>194</sup> These strategies are still in the research stage, and with current RO membranes, there is no guarantee that NDMA would be completely removed; for instance, it was reported that the concentration

of NDMA in RO permeate ranged from 8.8 to 31 ng/L.<sup>195</sup> Moreover, the rejection of NDMA precursors declined with increasing membrane age and after membrane cleaning; the NDMA rejection ratio decreased 6–9% during the first 4 h before increasing back to the precleaning rejection ratio.<sup>196,197</sup>

In terms of AOPs, H<sub>2</sub>O<sub>2</sub>, ozone, and ClO<sub>2</sub> alone decrease the concentrations of NDMA and NDMA precursors to some degree, but the potential formation of NDMA in subsequent chloramination and its partial rejection of RO highlight the need for a combined AOP.<sup>198,199</sup> Since the removal efficiencies of NDMA using UV + H<sub>2</sub>O<sub>2</sub>, UV + O<sub>3</sub>, and UV + monochloramine are all above 95% and that using UV/free chlorine is 81–95%, UV-based technologies are favored.<sup>195</sup> However, excessive UV radiation and oxidant dosage can increase NDMA formation potential.<sup>96</sup>

1,4-Dioxane (88.11 Da) is a stabilizer added to chlorinated organic solvents, which is found as a contaminant in some urban groundwater basins.<sup>72</sup> It is also poorly rejected by NF and RO due to its high polarity and small size.<sup>200,201</sup> Traditional AOPs such as O<sub>3</sub> or UV + H<sub>2</sub>O<sub>2</sub> are not sufficiently effective to remove it from the RO permeate.<sup>202,203</sup> Thus, research is ongoing to achieve higher removal efficiencies for 1,4-dioxane, such as reductive electrochemical activation of hydrogen peroxide.<sup>203</sup>

BAC and O<sub>3</sub>-BAC may also be cost-effective approaches for 1,4-dioxane.<sup>204,205</sup>

**Pathogens.** Pathogens, including viruses, have a particle size large than 10 nm,<sup>206</sup> and thus, RO should be able to achieve 100% removal based on size exclusion theory.<sup>207–209</sup> Forward osmosis followed by RO treatment was reported to achieve 6.7-log removal of spiked MS2 bacteriophage in graywater and sewage, 5.4-log removal of native *Escherichia coli* (*E. coli*) in graywater, and 7.9-log removal of native *E. coli* in sewage.<sup>210</sup> However, regrowth of the native bacteria in RO permeate was still observed, indicating that additional disinfection is needed.<sup>210</sup> In addition, monitoring virus removal after RO still presents challenges.<sup>211,212</sup> Thus, UV, O<sub>3</sub>, and/or chlorination treatment is highly recommended after RO. For instance, dosing sodium hypochlorite can completely eliminate SARS-CoV-2 in septic tank wastewater.<sup>213</sup>

## UNIT PROCESSES AND TREATMENT TRAIN COST ESTIMATES

There are many combinations of advanced tertiary treatment trains that can be considered for a given location for DPR,<sup>29,214</sup> on the basis of local water sources and conditions, existing WWTP infrastructure and space, availability of brine disposal to the ocean vs inland, energy costs, funding mechanisms, etc. These factors as well as the selected treatment train, scale (i.e., flow rate), and local regulatory requirements can substantially affect the final installed capital and operating costs. Nevertheless, it is useful to consider the cost of the various treatment units to plan for DPR to make an informed decision about the various water supply options. A particular challenge in obtaining cost information is that most publications are based on small-scale lab studies or a relatively limited number of large-scale operations with very different conditions.<sup>214–216</sup> Guo et al. conducted a very detailed study of the capital (CAPEX) and operating (OPEX) expenses of many of the unit operations (e.g., RO, UF + MF, membrane bioreactor (MBR), activated sludge (AS), coagulation (COAG), peroxone (H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub>), and granular activated carbon (GAC)) and generated scaling equations.<sup>9,217</sup> Figure 3 presents the inflation-adjusted CAPEX (Figure 3A) and OPEX (Figure 3B) costs (from 2012 to 2021) based on equations for plant capacities ranging from 100 to 500 000 m<sup>3</sup>/d. Chlorination is expected to be a very small fraction of the total cost of the DPR train.<sup>217</sup> To complement the information, the cost equations developed by Plumlee et al. were also included in Figure 3, which were also adjusted for inflation (from 2011 to 2021).<sup>218</sup> These authors consider that the CAPEX and OPEX for RO are exactly the same as for NF (thus, they are not presented in Figure 3). In terms of overall capital costs, the capital investment per unit operation for a 100 000 m<sup>3</sup>/d (~26.4 million gallons per day) plant ranges from around \$10M to \$162M, excluding coagulation and peroxone treatment,<sup>111</sup> as estimated using the information from Guo et al.<sup>9</sup> Most IPR and DPR plants built to date have a capacity of around 100 000 m<sup>3</sup>/d.<sup>219</sup> While in general the cost functions from Guo et al.<sup>9</sup> and Plumlee et al.<sup>218</sup> are comparable in their estimates for a given technology, there is a major discrepancy in the estimates for peroxone (H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub>), CAPEX, and OPEX. Both equations used to estimate this are presented in Figure 3. The data used for Figure 3 is provided in Tables S1 and S2.

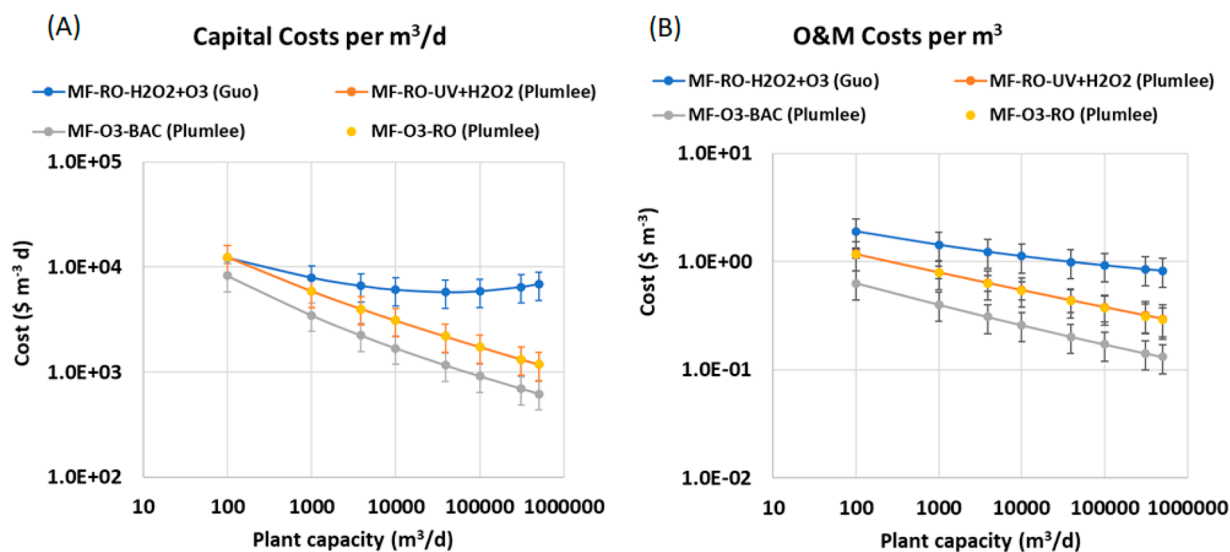
On the basis of these equations, at 100 000 m<sup>3</sup>/d, CAPEX is ranked as follows: COAG ≪ O<sub>3</sub> = H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub> (Plumlee et al.<sup>218</sup>) = UV + H<sub>2</sub>O<sub>2</sub> < GAC < BAC < RO < UF/MF < AS < NF < MBR < H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub> (Guo et al.<sup>9</sup>); OPEX is ranked as O<sub>3</sub> < H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub>

(Plumlee et al.<sup>218</sup>) < UV + H<sub>2</sub>O<sub>2</sub> < BAC < AS < COAG < GAC < UF/MF = MBR < RO < NF < H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub> (Guo et al.<sup>9</sup>). These cost estimates have an uncertainty of -30/+50%, and local conditions may result in significant differences. Thus, there could be substantial overlap in the range of estimates for the various technologies, which affect the ranking. Naturally, the assumptions embedded in the cost estimate equations are important and may differ for each WWTP. For example, BAC as estimated here assumes a 10 min<sup>218</sup> empty bed contact time (EBCT), but other EBCTs may increase or decrease both CAPEX and OPEX.<sup>218</sup> O<sub>3</sub> is considered pre-RO, while H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub> is intended to be post-RO. O<sub>3</sub> dosage is considered at 6 mg/L in the influent, but since some is consumed by the organic matter, it is estimated at an effective dose of 3 mg/L with a 5 min hydraulic contact time.<sup>218</sup> The increase of the effective dose of ozone from 1.5 to 9 mg/L can result in an increase of 8% in CAPEX for an ~100 000 m<sup>3</sup>/d plant and 22% increase in OPEX.<sup>218</sup> Low pressure (e.g., MF/UF) and high pressure (e.g., RO, NF) membranes and labor costs can be combined, reducing overall OPEX.<sup>218</sup> The cost of electricity can be quite significant as a fraction of OPEX and will differ substantially for each region.

The effect of scale is clearer when CAPEX and OPEX are normalized by plant capacity (m<sup>3</sup>/d) and annual flow (m<sup>3</sup>), respectively (Figure 3C,D). CAPEX decreases substantially with increasing scale for most technologies, except for UV + H<sub>2</sub>O<sub>2</sub>, which remains near \$115 per m<sup>3</sup>/d, and for peroxone as calculated from the equation in Guo et al.,<sup>9</sup> in which CAPEX first decreases slowly until the plant capacity increases above ~10 000 m<sup>3</sup>/d, and then unit costs begin to increase again with increasing plant capacity. This behavior differs considerably from the prediction using the equation in Plumlee et al.<sup>218</sup> for peroxone, which has a continuous decrease in CAPEX with scale. In terms of OPEX, most technologies exhibit a gradual but consistent decrease with scale, except GAC and UF/MF, for which OPEX decreases very rapidly with increasing plant capacity. However, BAC, a very similar technology to GAC, appears to reach a flat cost of around \$0.045 per m<sup>3</sup> above a plant capacity of 10 000 m<sup>3</sup>/d.

The CAPEX and OPEX of NF are expected to be similar to those of RO.<sup>218,220</sup> NF can operate at slightly lower pressure than RO, which reduces OPEX.<sup>76</sup> The retentate from NF and RO contains a significant fraction of the CECs that were not fully removed in the conventional wastewater treatment process and may require additional treatment before being disposed of, for example, using AOPs, such as ozone.<sup>220</sup> Since NF and RO have a recovery ratio of 50–85%,<sup>220</sup> the waste stream to treat and dispose can be significant. If ozone is used for this treatment, with or without UV, the transfer efficiency of ozone from the gas phase where it is generated to the liquid phase needs to be high, around 70–90%.<sup>221,222</sup> Although some NF can perform very well compared to RO in the removal of most CECs, they may not meet the California requirement of TOC < 0.5 mg/L and also have a poor rejection of nitrate and other monovalent ions.<sup>76</sup> For example, a study to remove a combination of PFAS using NF from concentrate estimated OPEX of around \$0.25–0.50/m<sup>3</sup> at a flow rate of 11.5 m<sup>3</sup>/d, depending on the treatment goal, which is similar to the estimate using the equation in Plumlee et al.<sup>218</sup> (Figure 3D).<sup>115</sup> Some NF can lower most CEC concentrations below 100 ng/L and, if bromide is not a concern, NF can be more economical due to the lower pressure requirements and fouling potential, which can result in 50% fewer cleanings and higher overall utilization compared to RO.<sup>76</sup> The cost of fouling is significant and can represent up to 24 ± 3%





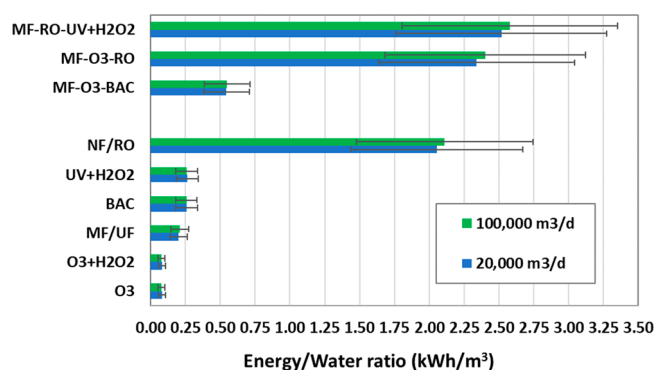
**Figure 4.** Integrated advanced tertiary treatment train (A) capital costs; (B) annual operating expenses. The treatment train costs are calculated with the equations from Guo et al.<sup>9</sup> and Plumlee et al.<sup>218</sup> All costs adjusted for inflation to 2021.

of OPEX for RO and  $11 \pm 1\%$  of OPEX for NF.<sup>223</sup> Fouling results in additional energy, earlier membrane replacement, and minor additional cleaning costs.<sup>223</sup> Fouling also results in additional downtime, which can be an important loss of use if cleaning-in-place events are frequent.<sup>223</sup>

Four advanced tertiary treatment trains were considered (Figure 4), although there are many other possible combinations. The least expensive in terms of CAPEX and OPEX is MF–O<sub>3</sub>–BAC as proposed by Snyder et al.,<sup>224</sup> which can produce high effluent quality largely eliminating most CECs, although it may not achieve TDS treatment goals and thus may be more suitable for IPR.<sup>224</sup> CAPEX and OPEX are essentially the same for MF–RO–UV + H<sub>2</sub>O<sub>2</sub> and MF–O<sub>3</sub>–RO,<sup>218</sup> and these two treatment trains have the potential to achieve very high effluent quality and meet the strict California requirements. The most expensive treatment train would be MF–RO–H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub>,<sup>218</sup> which also has the lowest economies of scale. The CAPEX estimated here corresponds well (within  $-30/+50\%$ ) to those in operational or pilot-scale plants in the US.<sup>219</sup> In all cases, the added cost of these advanced tertiary treatment trains will have an important impact on the overall cost of water for ratepayers. However, CAPEX for RO and NF continues to decrease with increasing installed capacity,<sup>225,226</sup> which has not been taken into consideration in these estimates. On the other hand, we are approaching the thermodynamic limit of separation<sup>227,228</sup> with costs for membranes reaching an asymptote.<sup>229</sup>

## ENERGY AND OTHER ENVIRONMENTAL CONSIDERATIONS

Energy considerations are paramount to potable reuse, given the significant increase in the energy required per m<sup>3</sup> of treated water. For the unit operations being considered for potable reuse, the largest requirement would be from the high-pressure membranes (Figure 5) followed by the energy requirements for UV + H<sub>2</sub>O<sub>2</sub>, which are very similar to those of BAC and MF/UF. As with the cost estimates, these values have an uncertainty of  $-30/+50\%$ , and local conditions may result in significant differences. The range of values also depends on the level of technology (e.g., older vs newer membranes, ozone generating system, UV lamps). Thus, treatment trains that consider RO (or



**Figure 5.** Energy/water ratio for different unit processes as well as three possible treatment trains for DPR or IPR. Energy data from Plumlee et al.<sup>218</sup>

NF) would have 80–90% higher energy requirements than alternatives such as MF–O<sub>3</sub>–BAC. These estimates based on modeling equations can be compared to a recent study that found the electricity intensity in 70 operating, planned, or pilot fully advanced treatment systems to range from 0.9 to 2.2 kWh/m<sup>3</sup> with operational systems reporting from 1.1 to 1.4 kWh/m<sup>3</sup>.<sup>219</sup>

To compare the energy required to degrade CECs, it is useful to consider the concept of electrical energy per order (EE/O, in kWh/m<sup>3</sup>), which is the electrical energy needed to degrade a particular chemical by 1 order of magnitude in 1 m<sup>3</sup> of water.<sup>230</sup> A recent review of the EEOs of 13 AOPs for a large number of molecules concluded that O<sub>3</sub> alone had in general the lowest median EEO followed by H<sub>2</sub>O<sub>2</sub> + O<sub>3</sub> < electron beam < UV + chlorine < UV + persulfate < UV + O<sub>3</sub> < UV + H<sub>2</sub>O<sub>2</sub> < photo-Fenton < plasma.<sup>231</sup> The median EEO values for the first seven EEOs in the ranking are  $<1$  kWh/m<sup>3</sup>. For photo-Fenton and plasma, the median EEO values are 3–5 kWh/m<sup>3</sup>. There is a significant difference in the EEO for the various chemicals, depending on the molecular structure and physicochemical properties, in some cases ranging over 5 orders of magnitude (e.g., from 10<sup>-3</sup> to 10<sup>1</sup> kWh/m<sup>3</sup> for O<sub>3</sub>).<sup>231</sup> EEO is also dependent on the dose of H<sub>2</sub>O<sub>2</sub>, UV lamp type and arrangement, and other operating conditions such as the efficiency with which

O<sub>3</sub> can be transferred from the gas phase to water.<sup>231–238</sup> EEOs are also reported only for the AOP process itself and does not take into account energy requirements for purchased chemicals or other life-cycle stages.<sup>231</sup> While it is useful to consider EEO, it is important to note that the removal efficiency generally decreases as follows: plasma < UV-based AOPs < O<sub>3</sub>-based AOPs.<sup>239</sup> To select the technology for a given DPR system, one will need to consider the most recalcitrant contaminants in wastewater that are not removed by the previous or subsequent processes in the treatment train.

The advanced tertiary treatment train not only will increase the energy footprint of the WWTP and the embedded energy but also, depending on the composition of the electrical grid, may result in increased emissions of greenhouse gases, particulate matter of all sizes, mercury, and other air pollutants as well as its on water demand.<sup>240,241</sup> On the other hand, as the electrical grid relies more and more on renewable energy, in general with lower environmental impacts, the increased energy/water ratio may not result in a much higher environmental footprint. It should also be noted that the transport of water and wastewater to the corresponding treatment plants requires a considerable amount of energy, in some cases more than the energy of conventional wastewater treatment.<sup>242</sup> Domestic water heating also represents a much higher energy intensity (35–70 kWh/m<sup>3</sup>, depending on inlet and outlet temperatures as well as fuel source)<sup>243</sup> than those in these treatment trains, highlighting the need to minimize unnecessary water heating; proper accounting of the water volumes should be taken into consideration in these assessments. Since domestic water heating is controlled by consumers, it is important to educate them on the potential energy savings that can be achieved at the consumer level. In addition, there is an opportunity to use DPR systems as a means to store renewable energy by using it only when it is cheap and abundant on the grid (i.e., during peak solar hours during the day in California or at peak wind hours at night in Texas) and then store the treated water in the emergency buffer reservoir.<sup>244</sup> Creative thinking can be used to solve these two challenges (i.e., storing renewable energy and implementing DPR).

### ■ CRITICAL ROLE OF SENSING IN DPR

IPR and conventional water treatment systems already monitor a suite of physical, chemical, and biological contaminants on both continuous and intermittent schedules.<sup>245</sup> However, since the DPR process provides little buffer capacity (if any), contaminants that do pass through the treatment train can rapidly spread throughout the distribution system, exposing consumers to enhanced risk.<sup>245,246</sup> Therefore, the development and use of real-time sensing capabilities are critical for the safe use of DPR, as these capabilities can be used to inform operators and consumers, in real time, of danger to their drinking water supply.<sup>247</sup> In addition, this information can be used as input to automated systems used to control the treatment and distribution of reclaimed water, which can alert operators, intensify treatment, shut the process down, and divert contaminated water from the distribution system. However, while there is a clear need for real-time sensing and monitoring capabilities, few real-time sensing platforms are currently used, particularly for the detection of pathogens, trace organic pollutants (e.g., PPCPs), and trace metals/metalloids, all of which are present in wastewater at elevated concentrations and present risks during DPR.<sup>248</sup>

The sensing of DPR treatment train performance can be separated into two categories: (1) sensing of the performance of individual steps in the treatment train (e.g., the sensing of RO rejection); (2) sensing of contaminants of concern in the final treated water before it is introduced to the distribution system. In many ways, the evaluation of individual treatment steps is easier, as there are easily measurable water quality properties that correlate with the overall performance of each step. When one considers a common treatment train used in potable reuse (e.g., MF/UF–RO–AOP), the performance of each one of these steps can be readily inferred by using off-the-shelf sensing platforms. For instance, optical turbidity meters and UV-transmittance measurements that deliver readings in real time are used to monitor the performance of MF and UF membranes;<sup>249,250</sup> an increase in permeate turbidity can indicate damage to the membrane, which would require the membrane to be pinned (if in a hollow-fiber module) or replaced.<sup>251</sup> For RO membranes, which are designed to reject all charged species (including monovalent ions), monitoring permeate conductivity is a simple way to evaluate RO performance, in real time, although the sensitivity of this method is rather low;<sup>252,253</sup> since solution conductivity is highly sensitive to ion concentrations, damage to the RO membrane will manifest in a measurable increase in conductivity, informing operators that the RO process needs attention.<sup>253</sup> Fluorescence excitation–emission measurements (which can measure the presence of small organic molecules) are another method to evaluate the integrity of an RO membrane in near real time.<sup>254</sup> In terms of the performance of AOPs, there are numerous examples of hydroxyl radical sensors as well as sensors that evaluate the concentrations of hydrogen peroxide (used as a hydroxyl radical source in the UV/H<sub>2</sub>O<sub>2</sub> and O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> systems). Direct measurement of hydroxyl radicals (the main reactive component in AOPs) and the measure of H<sub>2</sub>O<sub>2</sub> disappearance (H<sub>2</sub>O<sub>2</sub> is consumed during hydroxyl radical generation) give a measure of process performance.<sup>255–257</sup> However, in all the cases mentioned above, while these measurements can fairly accurately determine the overall integrity of the different treatment steps, these measurements say nothing about specific contaminants of concern. Also, these measurements are sensitive to feedwater quality. For example, increased turbidity and conductivity of the feed stream will result in elevated readings in the membrane permeate streams. Therefore, these measurements must be conducted in both the feed and permeate streams to properly evaluate process performance. Another example is the ionic composition of the feedwater; if there is a change in the composition (e.g., increased Na<sup>+</sup> or Cl<sup>−</sup> concentrations), this can lead to a drop in the observed RO rejection and could lead to an erroneous conclusion that the treatment step is failing.

Various approaches have been explored for the monitoring of specific contaminants of concern in treated wastewater. These approaches can be further divided into two categories: (1) the sensing of specific contaminants; (2) the measurement of bulk water quality metrics (i.e., surrogate metrics) that are correlated to the presence of trace contaminants.<sup>29,258</sup> While the first approach offers a more accurate view of what is in the water, the large number of potential contaminants makes the utilization of this approach difficult, as it requires numerous sensors that need constant updating to keep up with the evolving field of potential contaminants.<sup>29</sup> Because the second approach relies on correlations between the presence of easily measurable species and trace contaminants, this approach is thought to be more cost-effective (and feasible), albeit while sacrificing specificity.<sup>29</sup>

However, these surrogate measurements are somewhat “water specific” and require “tuning” to a local water quality profile.<sup>259</sup>

A large amount of effort has gone into the development of sensors for various pathogens, including viruses, bacteria, and protozoa; a (non)comprehensive list of such sensors can be found in a recent review.<sup>260</sup> These sensors operate using different methods but can be grouped into (i) sensors that operate by monitoring changes to surfaces (e.g., electrical properties) when a target pathogen attaches to specific binding sites, which allows for detection in real time; (ii) sensors that detect specific pathogens by amplifying DNA; (iii) optical sensors (e.g., cytometry, fluorescence); (iv) colorimetric sensors.<sup>260</sup> While many of these sensors are highly accurate and sensitive, many require some time for the detection process to take place (e.g., to amplify DNA), making real-time detection impossible. For sensors that offer results in near real time, these sensors can become fouled by other aquatic species, such as dissolved organic matter that can form nonspecific (e.g., hydrophobic) interactions with the surface, which can block binding sites and reduce sensitivity.

Sensors for the detection of specific PPCPs (e.g., pharmaceuticals, endocrine disruptors, etc.) have also been explored. Approaches toward their detection include the fabrication of electrodes with specific binding sites to these compounds<sup>261,262</sup> as well as spectroscopic analysis (e.g., using fluorescence spectroscopy, Raman spectroscopy, and infrared spectroscopy) of water.<sup>263,264</sup> While these sensors often exhibit high sensitivity and specificity in pure samples, electrochemical sensors are prone to fouling, and spectroscopic signals can be complicated to deconvolute in a mixed sample environment. That being said, the highly treated water resulting from the multiple treatment step used for DPR has very low concentrations of other contaminants, making these detection methods highly attractive. In terms of metal/metalloid sensing, many reports have described online sensors for the detection of arsenic and other heavy metals.<sup>265–268</sup> Many arsenic electrochemical sensors have been demonstrated, using such methods as anodic stripping voltammetry and molecular imprinting of electrodes.<sup>267</sup> To the best of our knowledge, no online sensors have been developed for boron. There is likely an interesting opportunity to combine machine learning methods with monitoring tools for the rapid and flexible detection of a range of contaminants.<sup>269–272</sup> Specifically, the ability to train a computer to deconvolute and decipher the complex spectroscopic signals resulting from the analysis of aqueous streams may prove transformative, as the algorithm can be trained to identify target contaminants as they emerge. For instance, software using a convolutional neural network was able to recognize nitrates, some pharmaceuticals, microplastics, and their additives after being trained on a large set of spectra collected from a deep-UV Raman/fluorescence spectroscopy.<sup>271</sup> A multivariate model utilizing machine learning algorithms based on a back-propagation neural network was successfully developed and trained to accurately read various spectra collected from soil by laser-induced breakdown spectroscopy for the detection of trace element analysis.<sup>273</sup> UV–vis spectroscopic data was used to train a *fitness*-support vector machine classifier, which was then able to serve as an early warning system for water contamination events.<sup>274</sup> While they require significant amounts of high-quality data for training, machine learning approaches can offer high reliability for detecting and monitoring contaminants in different water matrices. For instance, after being trained with a data set of 12 560 UV spectra, a system using machine learning

algorithms successfully detected target contaminants in 107 times out of a total of 109 measurements and did not generate any false positive signals.<sup>274</sup>

The installation of sensor systems will likely increase the capital costs of water treatment systems. However, a system with a series of in-line sensors has the potential of not only minimizing operational failures but also informing data-driven system control algorithms (using artificial intelligence approaches) that optimize water treatment processes and reduce operational costs.<sup>275</sup> For example, it has been demonstrated that a data-driven approach could reduce pump energy consumption in a wastewater treatment plant by 18.5%;<sup>276</sup> a model based on artificial neural networks cut coagulant dosage by 10%,<sup>277</sup> and an in-line control system built on a genetic algorithm utilizing a fuzzy wavelet neural network algorithm provided robust and effective dissolved oxygen control and reduced the demand for aeration.<sup>278</sup> In addition, it is anticipated that the benefits of in-line sensor systems coupled to artificial-intelligence optimization algorithms will increase the adoption of such sensors, which will potentially reduce the costs of these sensors, bringing down the associated capital costs.

## RECOMMENDATIONS

A number of treatment trains (e.g., MF–RO–AOP, MF–O<sub>3</sub>–BAC) have been successfully tested (e.g., in pilot tests and IPR systems) and can produce the high-quality effluent needed for DPR. Costs and energy requirements continue to decrease for RO and NF, although thermodynamic limits are being reached, and a reduction in cost and energy is a technological challenge.<sup>145</sup> The most significant challenges are in online, real-time sensing of a number of small molecules that may be present in the influent; breakthroughs in this area will be needed for full deployment of DPR to ensure high reliability and consumer safety. Of particular concern would be high episodic loads from accidental or unreported discharges to the sewer system that could move quickly through the treatment system and pass to the distribution system at concentrations that could pose a concern as well as the rapidly evolving field of trace contaminants that find their way through the treatment system and can be harmful to consumers when chronically present in the water.

**Establish Online and Real-Time Water Quality Monitoring Systems.** There have been important advances in water quality monitoring, but there is a critical need to quantitatively monitor a number of molecules and conditions, which can serve as indicators of unit process or treatment train performance. Monitoring has to have a high degree of reliability and redundancy to avoid failures as well as provide warning in real time to operators and consumers. Since there is little buffering capacity, rapid measurements are critical to safety. There may be an attractive opportunity to utilize machine learning approaches to analyze spectroscopic data (which can be rapidly collected and analyzed) and inform operators of potential problems.

**Redundancy in Treatment Processes to Ensure a High Level of Removal.** If DPR is to get main-stream acceptance and become a regular water source, redundant treatment equipment will be needed to handle maintenance as well as load surges and other emergencies. This will increase the overall CAPEX. In any case, this will likely be the most expensive water source for most municipalities, even if CAPEX and OPEX costs, particularly those of RO/NF membranes, continue to decrease as more capacity is installed.

**Emergency Buffer and Plans for Dealing with Off-Spec Water.** If the monitoring system detects a deviation from specifications or there is a detected equipment failure or under-performance, there will be a need for an emergency buffer reservoir to store the off-spec water and a clear process for dealing with it for either reprocessing or discharge into an environmental receiving water body without causing undue environmental damage.

**Consideration of Renewable Energy.** Most DPR treatment trains will have a substantial impact on the embedded energy in water. This may become a barrier for adoption, unless there is a plan to employ renewable energy that will result in lower emissions of carbon and many pollutants associated with conventional fuels. Building renewable energy at the same pace as DPR is implemented must be part of the plan as well as developing creative ideas for storing renewable energy from the grid by filling the emergency reservoir with treated water from DPR. There is also a need to educate the consumer about opportunities for energy reduction in domestic water use, which can be as important as water treatment in the overall balance.

## ■ ASSOCIATED CONTENT

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsestengg.1c00258>.

Two tables with capital and operating expenses for different unit processes and treatment trains (PDF)

One spreadsheet with equations to calculate capital and operating expenses (XLSX)

## ■ AUTHOR INFORMATION

### Corresponding Author

Arturo A. Keller – Bren School of Environmental Science and Management, University of California Santa Barbara, Santa Barbara, California 93106, United States; [orcid.org/0000-0002-7638-662X](https://orcid.org/0000-0002-7638-662X); Email: [arturokeller@ucsb.edu](mailto:arturokeller@ucsb.edu)

### Authors

Yiming Su – Department of Civil and Environmental Engineering, University of California Los Angeles, Los Angeles, California 93106, United States; [orcid.org/0000-0001-6035-7384](https://orcid.org/0000-0001-6035-7384)

David Jassby – Department of Civil and Environmental Engineering, University of California Los Angeles, Los Angeles, California 93106, United States; [orcid.org/0000-0002-2133-2536](https://orcid.org/0000-0002-2133-2536)

Complete contact information is available at:

<https://pubs.acs.org/doi/10.1021/acsestengg.1c00258>

### Notes

The authors declare no competing financial interest.

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