1 **Management of Concentrate and Waste Streams for Membrane-based**

2 **Algal Separation in Water Treatment: A Review**

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20 Water Research

21 (Re-submitted May 2020)

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Abstract

Frequent occurrence of harmful algal blooms (HABs) and red tides in freshwater and seawater poses serious threats to water treatment and drives the application of membrane-based technologies in algal separation. Despite the high removal efficiency of algal cells and their metabolites (*e.g.* organic matter and toxins) by membranes, the generation of concentrate and waste streams presents a major challenge. In this paper, we review the scenarios under which membrane-based processes are integrated with algal separation, with particular attention given to (i) drinking water production and desalination at low algal concentrations and (ii) cyanobacteria-laden water treatment/desalination. The concentrate and waste streams from backwashing and membrane cleaning in each scenario are characterised with this information facilitating a better understanding of the transport of algal cells and metabolites in membrane processes. Current strategies and gaps in managing concentrate and waste streams are identified with guidance and perspectives for future studies discussed in an Eisenhower framework.

List of abbreviations and acronyms

Abbreviations and acronyms	Definition
2-MIB	2-methylisoborneol
AOM	Algal organic matter
BR	Backwashing to feed solution volume ratio
BW_d	Backwashing duration
BW_f	Backwashing frequency
CF	Concentration factor, defined as the concentration ratio of concentrate over the feed solution
CIP	Cleaning in place
CR	Concentrate to feed solution volume ratio
DOC	Dissolved organic carbon
EOM	Extracellular organic matter
ERDs	Energy recovery devices
HABs	Harmful algal blooms
Hc-Htox	Scenario of cyanobacteria-laden water treatment/desalination
HPI	Hydrophilic fraction of AOM
HPO	Hydrophobic fraction of AOM
IOM	Intracellular organic matter
Lc	Scenario of drinking water production and desalination at low algal/toxin concentrations
MC, MC-LR	Microcystin, Microcystin -LR
MF	Microfiltration
MWCO	Molecular weight cut-off
NF	Nanofiltration
PAC	Powdered activated carbon
PRO	Pressure-retarded osmosis
RO	Reverse osmosis
SWRO	Seawater reverse osmosis
TEPs	Transparent exopolymer particles
TMP	Transmembrane pressure
UF	Ultrafiltration
WR	Water recovery, defined as the permeate to feed solution volume ratio

45

1. Introduction

Microalgae are an extremely large and diverse group of organisms and include both prokaryotes and eukaryotes capable of photosynthesis. They are critical to the aquatic ecosystem (and the biosphere) for oxygen production and carbon fixation (Rai et al. 2000). However, their excessive growth in some natural environments, such as commonly occurring in eutrophic or polluted water bodies, can result in severe oxygen depletion and damage to the aquatic habitats (Loza et al. 2014). Certain microalgae (*e.g. Microcystis aeruginosa*) can produce toxins, which pose a threat to public health through either drinking contaminated water or consumption of food that has been processed with contaminated water (Caron et al. 2010, Song et al. 2018b, Westrick 2008). For example, microcystin-LR (MC-LR), with amino acids leucine (L) and arginine (R) at positions 2 and 4 respectively, has been listed as one of the most potent water toxins in the United States (Greer et al. 2018). Due to their frequent occurrence (often at elevated concentrations), algal removal from affected source waters at high efficiency and low cost is of great practical importance.

Conventional technologies applied for algal treatment include coagulation, centrifugation, dissolved air-flotation, chemical oxidation (*i.e.* disinfection and pre-oxidation) and filtration (Christenson and Sims 2011, Henderson et al. 2008a, Naceradska et al. 2017). Despite substantial progress achieved with these methods, critical challenges remain. For example, coagulation, the most common water treatment method, can partially remove algal cells but has low efficiency against soluble algal toxins (Ghernaout et al. 2010). Centrifugation is considered to be one of the most effective separation methods, but the high shear force produced in this energy-intensive process can easily damage the cell membrane, resulting in the release of algal toxins (Chen et al. 2011, Christenson and Sims 2011). While dissolved air flotation of algal cells is attractive in view of its low operating cost, the presence of chemicals to aid this process raises other problems such as disposal of the chemical sludge (Granados et

4

71 al. 2012). Additionally, chemical oxidation may result in the generation of secondary

72 pollutants. For example, it has been reported that chloramination of waters containing

73 organics at elevated concentrations likely leads to the formation of harmful by-products such

74 as trihalomethanes and *N*-nitrosodimethylamine (Furst et al. 2018).

75   In recent years, membrane-based processes have gained increasing popularity as a means

76 of algal separation as a result of their high separation efficiency and low footprint (Liu et al.

77 2017). Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO)

78 have particular benefits for algal separation because they can achieve complete retention of

79 algal cells and, at least for the smaller pore sized membranes, remarkable removal of solutes

80 (*e.g.* organic matter and toxins) (Bilad et al. 2014, Teixeira and Rosa 2005). Following the

81 early trial by Koseoglu and Batchelor (1993) of use of membrane processes for algae

82 retention and the first systematic study of algae removal from source waters using

83 membranes by Chow et al. (1997), the continual improvement in membrane materials and

84 optimization of operating parameters has made this method competitive (in terms of both cost

85 and effectiveness) compared to centrifugation (Bilad et al. 2012). Nevertheless,

86 membrane-based algal separation faces two major drawbacks: (i) membrane fouling, which

87 leads to a notable decrease in membrane permeability (Heng et al. 2008, Hung and Liu 2006,

88 Zhang et al. 2013a) and consequently the need for frequent cleaning and maintenance (Judd

89 2016, Wang et al. 2014)), and (ii) the need for management of concentrate and waste streams

90 that may contain cells and toxins at much high levels.

91   Investigation of the algal-induced membrane fouling mechanism and associated control

92 strategies has become an important research topic, which has been addressed by a number of

93 research groups (Liu et al. 2017, Qu et al. 2012a, Villacorte et al. 2015, Zhang et al. 2012).

94 There are also comprehensive review papers on recent achievements and opportunities in

95 membrane materials and processes for algal separation (Liao et al. 2018, Luo et al. 2017,

96   Zhang and Fu 2018). Nevertheless, a systematic overview of the management and treatment

97   methods for the concentrate and waste streams is not yet available, which prompts us to

98   provide a comprehensive review on this important topic. Essentially, the volume ratio and

99   composition of the concentrate and waste streams is largely determined by the choice of

100   membrane processes in water treatment (Khan et al. 2009, Van der Bruggen et al. 2003), as

101   well as the algal (and toxin) concentrations and the salinity of the feed solution (Liu et al.

102   2017, Schurer et al. 2013). As such, we have based this review paper around two scenarios in

103   which membrane-based processes are integrated with algal separation: (i) drinking water

104   production and desalination at low algal concentrations, and (ii) cyanobacteria-laden water

105   treatment, especially under seasonal cyanobacterial blooms. The gaps in knowledge with

106   regard to managing concentrate and waste streams (generated from backwashing and

107   membrane cleaning) are identified, and guidance and perspectives for future studies in

108   membrane-based algal separation for water treatment are provided.

109

110   2. Classification of membranes and scenarios in algal separation

111   In this section, we will introduce the membranes used for algal separation and the streams (*i.e.*

112   the permeate, concentrate and waste streams) generated during the filtration. The application

113   scenarios are classified into two categories (*i.e.* (i) drinking water production and desalination

114   at low algal concentrations, and (ii) cyanobacteria-laden water treatment) with their

115   characteristics discussed individually.

116   2.1. Membranes

117   Table 1 summarises commonly-used membranes for algal separation. Specifically, MF and

118   UF are low pressure membranes. With their pore sizes smaller than microalgae cells, these

119   porous pressure-driven membranes can be used to address HABs in surface waters (Huang et

120   al. 2009a) or to pretreat the feed stream to RO in seawater desalination (Villacorte et al.

121 2015). Compared to their low pressure counterparts, RO and NF have smaller molecular

122 weight cut-off (MWCO) (Table 1) but require substantially higher pressure. High pressure

123 membranes are not expected to be directly used for algal separation (*e.g.* red tides during

124 RO-based seawater desalination) because of the severe membrane fouling and high operating

125 cost that would result. Nevertheless, issues related to the retention of dissolved organic matter

126 and toxins, if any, in the feed to RO and NF, must still be addressed (Seubert et al. 2012).

127 Considering that algal sizes are at the micrometer scale (*e.g. M. aeruginosa* 3.0−7.0 μm,

128 *Chlorella* sp. 3.0−8.0 μm and *Alexandrium tamrensis* 20−42 μm) (Granéli and Turner 2006),

129 all aforementioned membranes (Table 1) are capable of rejecting algal cells by size exclusion.

130 In comparison, MWCO governs the retention of dissolved organic matter, toxins and algae

131 derived odor compounds (*e.g.* 2-methylisoborneol (2-MIB) and geosmin) with much smaller

132 size.

133

134 Table 1

135

136 For a typical membrane process, the feed solution is separated into two streams as a result

137 of treatment (Fig. 1): the product water permeating through the membrane (*i.e.* the permeate)

138 and the rejected water containing concentrated contaminants (*i.e.* the concentrate). To

139 mitigate membrane fouling, backwashing (for the case of porous membranes) and chemical

140 cleaning are often utilised (Wang et al. 2014) with these processes generating additional

141 waste streams. In order to elucidate the challenges for management of the concentrate and

142 waste streams in a more systematic manner, we consider two major application scenarios: (i)

143 drinking water production and desalination at low algal/toxin concentrations (Low

144 concentration, L*c*) and (ii) cyanobacteria-laden water treatment/desalination (High

145 concentration and high toxicity, H*c*-H*tox*) (Fig. 1). It is worthwhile noting that the

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146　composition of the feed solution to the membrane process may differ substantially from that

147　of the raw water due to the presence of pre-treatment processes such as coagulation and

148　sedimentation. Furthermore, the composition of the concentrate and waste streams is affected

149　by the type of membrane operation and chemicals used, if any, which could differ

150　substantially for low (MF and UF) and high pressure (NF and RO) membrane processes.

151　Therefore, under each scenario, low and high pressure membrane processes are discussed

152　separately.

153

154　Fig. 1

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156　2.2. Lc scenario: Drinking water production and desalination at low algal concentrations

157　This is the most common scenario for drinking water production from surface water

158　treatment (with typical feed salinity < 500 mg/L) and seawater desalination (salinity typically

159　~35 g/L) using membrane technologies. In this scenario, the algal (and toxin) concentration

160　in source waters is relatively low; for example, the green algae alert by WaterNSW, Australia,

161　according to the National Health and Medical Research Council, is 500 cells/mL of *M.*

162　*aeruginosa* for fresh water or 10 cells/mL of *Karenia brevis* for coastal water (NHMRC

163　2008), and the World Health Organization guideline for MC-LR in fresh water is < 1 μg/L

164　(Sikka and Hellstrom 2017). Implementation of conventional processes such as coagulation

165　and activated carbon adsorption generally meets the standards for turbidity and heavy metals

166　(Fig. 2a). Membrane-based technologies are capable of "polishing" the effluent by removal of

167　residual colloidal matter and/or dissolved solutes (Table 1). In this case, algal cells may be

168　occasionally involved in membrane fouling, and algal organic matter (AOM) typically

169　contributes only a small portion of the membrane foulant. Despite the low concentrations of

170　algal toxins in the Lc scenario, the concentrate and backwashing streams are expected to

171   contain much more organic matter compared to the feed solution. As such, the "10%

172   recycling rule" (*i.e.* the recycle flow <10% of the raw water flow) has been implemented by

173   the US Environmental Protection Agency (EPA 2002). In general, there are few challenges

174   with regard to disinfection and management of the outfalls for the discharge of MF/UF

175   concentrate and backwashing streams into the environment. However, the discharge of RO

176   concentrate back to seawater can result in critical issues such as the generation of a dense

177   plume that sinks to the seafloor and disrupts marine communities (Pérez-González et al. 2012)

178   though it is now recognized that the implementation of an appropriately designed diffusion

179   device would largely avoid the adverse impacts (Missimer and Maliva 2018).

180

181   2.3. H*c*-H*tox* scenario: Cyanobacteria-laden water treatment/desalination

182   This scenario is applicable to the treatment of source waters during seasonal cyanobacterial

183   blooms. Table 2 summarises the characteristics of algae and toxins in cyanobacteria-laden

184   waters. Numerous studies have reported that paralytic shellfish poisoning and cyanobacterial

185   toxin poisoning can pose great threats to human health. For example, the toxicology of

186   paralytic shellfish poisoning (by *Alexandrium* sp.) in red tide blooms is similar to that of

187   tetrodotoxin (Etheridge 2010). Uptake of more than 80 μg of saxitoxin to 100-g of tissue may

188   lead to death in severe cases (Table 2) (Etheridge 2010). Cyanobacterial toxin poisoning

189   mainly occurs in freshwater areas where ingestion of toxin-contaminated water is more likely

190   (Westrick 2008). Among the various HABs in the H*c*-H*tox* scenario, *M. aeruginosa* and

191   *Anabaena flos-aquae* are particularly widely distributed hazardous species. *M. aeruginosa*

192   can produce the most widely distributed hepatotoxin, microcystin (MC) (Granéli and Turner

193   2006). MCs have a strong resistance to proteolytic enzymes with over 70 variants found (*e.g.*

194   MC-LR, MC-AR and MC-YR) (Carmichael 1992). According to National Health and

195   Medical Research Council guidelines (NHMRC 2008), the alert for a Hc-Htox scenario is

196 raised when >50,000 cells/mL of *M. aeruginosa* are present or a biovolume of all toxin

197 producing cyanobacteria exceeds 4 mm^3/L (or > 10 mm^3/L for all algae) in fresh water

198 while >10 cells/mL of *Karenia brevis* is applied for coastal water. The high abundance of

199 cyanobacteria at these alert levels would lead to a concentration of total microcystins >10

200 μg/L (NHMRC 2008). In spring 2007, a cyanobacteria bloom covered almost one-third of

201 Taihu Lake, which directly led to restriction of water supplies for around 5 million residents

202 of Wuxi city in China (Li et al. 2011). Moreover, although neither 2-MIB nor geosmin (the

203 typical algal odor compounds) has any known adverse biological or pathological effects

204 (Giglio et al. 2011), a level of ≥10 ng/L in water at HABs is considered to be above the taste

205 and smell threshold (Reynolds 2010) (Table 2). Likewise, non-toxic algal blooms will not

206 necessarily lead to serious health issues, but may result in deterioration of aquatic habitats

207 and water quality via generation of mucilage and/or transparent exopolymer particles (TEPs)

208 (Bar-Zeev et al. 2015, Gotsis-Skretas 1995, Hamilton et al. 2005).

209 Due to the relatively high algal concentration and the possibility that the source waters

210 exhibit high toxicity and unpleasant odor, the efficiency of pretreatment processes such as

211 coagulation is typically compromised. More importantly, the removal of dissolved toxins and

212 odor compounds likely requires additional chemical oxidation steps (Fig. 2a). Although

213 membrane-based separation can be integrated to improve process performance (Table 1), it

214 results in the accumulation of algal cells and toxins in the concentrate. The backwashing

215 waste in the H*c*-H*tox* scenario becomes more difficult to deal with (Fig. 1) because of the

216 release of algal cells (and organic matter) and toxins from the cake layer (Li et al. 2014). In

217 practical processes, the backwashing water is alternatively from the disinfection tank, and the

218 secondary byproducts (*e.g.* aldehydes, ketones and halogenated byproducts) from the reaction

219 of AOM and disinfectants would be contained in backwashing waste. In comparison, the

220 composition and toxicity of the waste stream from chemical cleaning are strongly influenced

221 by the chemical reagents and the dose used (with the possibility of generation of other

222 products from reactions with the cleaning reagents (Wang et al. 2014)). While the use of

223 oxidants can degrade a portion of the dissolved toxins, this practice can damage cell integrity

224 and result in the massive release of intracellular toxins (Hall et al. 2000, Zamyadi et al. 2012)

225 into the chemical waste.

226

227   Table 2

228

229   Fig. 2

230

231 **3. Concentrate and waste streams in L*c* and H*c*-H*tox* scenarios**

232 This section is focused on the review of concentrate and waste streams in the established L*c*

233 and H*c*-H*tox* scenarios. Standardisation of the basic characteristics (*e.g.* volume ratio and

234 concentration factor) is provided and detailed assessment of these characteristics is provided

235 for MF/UF, NF and RO in L*c* and H*c*-H*tox* scenarios. Particular attention is paid to the

236 transformations of AOM, toxins and odor compounds during filtration and the impacts of

237 these transformations on the composition of concentrate and waste streams.

238 3.1. Basic characteristics of the concentrate and waste streams

239 **Concentrate.** In this review paper, the concentrate represents either (i) the retentate stream in

240 cross-flow or semi-dead-end operation of MF/UF (Zeman and Zydney 2017) or (ii) the

241 retentate stream and the intermediate forward flushing stream in cross-flow operation of

242 NF/RO (Pérez-González et al. 2012) (Fig. 1). As forward flushing is usually conducted

243 during desalination (*i.e.* during the generation of the retentate), the volume of the NF/RO

244 concentrate is made up of the sum of the retentate and forward flushing streams. The

245 concentrate to feed solution volume ratio (*CR*) is related to the apparent water recovery

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246   (WR_{app}, $i.e.$ the permeate to feed solution volume ratio) by eq.1 in Table 3 (Van der Bruggen

247   et al. 2003). Typically, the CR is 1−10% for MF (EPA 2002) and UF, 15−30% for NF, and

248   15−60% for RO (Khan et al. 2009). Assuming that the mass loss due to fouling is minimal

249   compared to the mass in the feed, concentrate and permeate, the concentration factor (CF,

250   defined as the concentration ratio of concentrate over the feed solution $c_{c,i}/c_{f,i}$) can be

251   obtained by an overall mass balance (eq. 2 in Table 3) (Greenlee et al. 2009). The rejection of

252   all membranes for algal cells is close to 1, $i.e.$, $CF \approx 1/CR$. Determination of the rejection

253   and CF for AOM, toxins and odor compounds is more complicated and will be discussed in

254   Section 3.2.

255

256   Table 3

257

258   **Backwashing waste.** In membrane-based algal separation, hydraulic cleaning such as

259   backwashing is always used for porous membranes ($e.g.$ MF/UF) to remove the "physically

260   reversible fouling" and to restore the permeability (Ferrer et al. 2016). This cleaning process

261   generates a backwashing waste stream (Fig. 1). The backwashing to feed solution volume

262   ratio (BR) is determined by the backwashing TMP ($\Delta P_{BW,t}$), backwashing duration (BW_d, min)

263   and backwashing frequency (BW_f, time per min) (eqs. 3 and 4 in Table 3) (Ferrer et al. 2016,

264   Wang et al. 2014). An empirical flow ratio of 0.8−1.2 can be applied to q_{BW}/q_f, with BR

265   values of 5−15% documented in some published papers (Chew et al. 2016, Oh et al. 2006).

266   As such, the WR_{app} (eq. 1) should be adjusted to $WR_{adj} = 1 - CR - BR$. In comparison,

267   estimation of the concentration factor for the backwashing waste is challenging because it

268   depends on various operating parameters such as the total mass of the cake layer, the portion

269   released and the source of the backwashing water. It is easier to directly measure the

270   concentration of constituents ($e.g.$ algal cells, AOM and toxins) in the backwashing waste.

271   **Chemical cleaning waste.** Chemical cleaning (typically known as "cleaning in place"

272   (CIP) (Shi et al. 2014, Wang et al. 2014)) removes scale, organic fouling and biofouling and

273   results in a chemical waste stream. The volume (and volume ratio) of this stream is small (< 1%

274   especially in the Lc scenario) compared to CR and BR though the composition is more

275   complex and depends on the chemicals added (such as citric acid and EDTA for scale

276   removal, and NaOCl for organic fouling and biofouling removal (Villacorte et al. 2015, Wang

277   et al. 2010)). Moreover, consideration should be given to the presence of high concentrations

278   of coexisting ions (*e.g.* Na$^+$ and Cl$^-$ in seawater) in addition to the residual chemicals.

279

280   3.2. MF/UF concentrate and waste streams in Lc and Hc-Htox scenarios

281   MF/UF is increasingly used in disinfectant-free production and distribution of drinking

282   water (Schurer et al. 2019), particularly given the good performance of UF in rejecting high

283   MW organic carbon that plays an active role in the regrowth of *Aeromonads* (Hijnen et al.

284   2018). The MF/UF concentrate is typically termed a recirculation loop/stream because this is

285   the general practice for concentrate processing in an MF/UF system. As such, CR is rarely

286   discussed in the Lc scenario though the resultant organic concentrations in the recirculation

287   loop can be as high as >100 mg-carbon/L (but still lower than the recommended criteria of

288   ~600 mg-carbon/L) (Oh et al. 2006). In pilot tests, backwashing is conducted every 20−30

289   min (*i.e.* BW_f = 1/20−1/30 per min) with a BW_d of ~0.5 min and $\Delta P_{BW, t}$ = 0.2 MPa. As

290   summarised in Table 4, the BR value for the Lc scenario is generally <5%. It has been

291   suggested that a combined concentrate and backwashing stream should follow the "10%

292   recycling rule" (EPA 2002), *i.e.* $CR + BR$ <10%. Nevertheless, an industrial scale study by

293   Chew et al. (2016) showed that the percentage of water losses by combined pretreatment and

294   backwashing in a UF system (~11%) could be roughly double that of a conventional media

295   filtration system. Compared with the relatively dilute backwashing waste, the concentration

13

296   of the chemical cleaning waste (based on total organic carbon) can be 1−2 orders of

297   magnitudes higher (Oh et al. 2006, Zhang et al. 2011). It is worthwhile noting that current

298   studies of the small-volume chemical waste (volume ratio < 1%) (Table 4) is largely related

299   to improving our understanding of the membrane foulant components that are resistant to

300   hydraulic cleaning (Wang et al. 2010), rather than investigation of suitable methods for

301   disposal of the chemical waste.

302

303   Table 4

304

305   In Hc-Htox, the rejection of MF/UF membranes for algal cells is close to 1 with cell

306   deposition insignificantly affecting the mass balance. CF for algal cells in the concentrate is

307   therefore around 10−100. In comparison, the composition of AOM and toxins in the

308   concentrate and waste streams is related to the membrane rejection (and then backwashing

309   and cleaning) mechanism. Note that since odor compounds have comparable MW with small

310   toxins, their removal by MF/UF membranes should occur in a similar manner (and will not

311   be discussed separately). Considering that the MWCO of MF/UF (Table 1) is typically larger

312   than the MW of algal toxins (*e.g.* 165.2 Da of anatoxin-a in Table 2), four pathways can be

313   proposed for the mechanism of removal of these toxins (Fig. 3):

314   (I) Size exclusion of intracellular/cell-bound toxins. In the Hc-Htox scenario, the rejection

315   of toxins by MF/UF largely depends on the cell integrity because algal toxins are originally

316   intracellular. Minimal cell breakage (2−5%) has been found during UF filtration, resulting in

317   pathway (I) in Fig. 3 accounting for over 95% rejection of toxins (Gijsbertsen-Abrahamse et

318   al. 2006, Liu et al. 2017).

319   (II) Size exclusion and (III) adsorption of aqueous toxin assemblages (*i.e.* toxin-AOM$_{(aq)}$)

320   by membranes (Lee and Walker 2008). AOM can be classified into two groups − those

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321 remaining inside cells and referred to as intracellular organic matter (IOM), and those

322 secreted into the solution and referred to as extracellular organic matter (EOM) (Zhou et al.

323 2014). Since IOM is inside algal cells with minimal amount released into the bulk solution

324 due to the cell lysis (Fig. 3), EOM is expected to dominate the interaction between AOM and

325 the aqueous toxin. As such, the fate of EOM and its active role in toxin rejection (*i.e.*

326 rejection of toxin-EOM$_{(aq)}$) should be taken into account in pathways (II) and (III). Table 5

327 classifies the characteristics of EOM of representative algal species (*e.g. M. aeruginosa*

328 blooms in fresh water and *Chaetoceros muelleri* in coastal water). Most organics in EOM are

329 hydrophilic with the hydrophilic fraction (HPI) accounting for >60% (Table 5) while the

330 hydrophobic fraction (HPO) is present at lower abundance. The MW distribution of EOM is

331 quite wide. For instance, the MW of EOM of the representative algal species (except for

332 *Asterionella formosa* (Henderson et al. 2008b)) is divided into two size regions, that is,

333 30−60% > 20−100 kDa (an MWCO for biopolymers) and 40−60% < 1 kDa (an MWCO for

334 humics) (Table 5). These findings are generally in accord with the rejection (~50%) of EOM

335 by MF/UF membranes (Li et al. 2014). Although the hydrophilicity of toxins and the

336 relatively large MWCO of UF membranes are expected to lead to lower rejection,

337 unexpectedly high rejection rates have been reported in some studies (Campinas and Rosa

338 2010). More specifically, in addition to hydrogen bonding effects, the electrostatic interaction

339 between the negatively charged EOM and the functional groups of toxins (*e.g.* the amide

340 group in MCs) (Table 2) is expected to generate larger toxin-EOM$_{(aq)}$ assemblages which can

341 be rejected and/or adsorbed by MF/UF through pathways (II) and (III) in Fig. 3.

342 (IV) Rejection and adsorption of aqueous toxins and/or toxin assemblages by the cake

343 layer on membranes (Liu et al. 2017). It has been reported that MF/UF fouling during the

344 filtration of cyanobacteria-laden water in the H*c*-H*tox* scenario is initially governed by pore

345 blocking by algal cells and large MW EOM, followed by the buildup and compression of a

346   cake layer on the surface due to the continual deposition of cells and AOM (Fig. 3) (Liu et al.

347   2018). Pathway (IV) relates to the secondary effect of membrane fouling in this scenario. For

348   instance, Liu et al. (2017) indicated that there was a positive correlation between the

349   reversible resistance of UF membranes and MC-LR rejection values ($R^2 = 0.95$).

350

351   Fig. 3

352

353   Table 5

354

355   Overall, the contaminants in the MF/UF concentrate in the H*c*-H*tox* scenario include algal

356   cells (with IOM and intracellular toxins), HPI of EOM with large MW (*i.e.* >20−100 kDa)

357   and minimal free toxins (Fig. 3). While the rejection of UF membranes for AOM and

358   toxin-AOM$_{(aq)}$ ($r = 46−98\%$) has been documented (Table 2), the high *CF* (*i.e.* 5−25 at *CR* =

359   4−10%) and concentrations of toxins (for example, >100 μg-toxins/L) in the concentrate have

360   received limited attention, especially among the academic community. High algal mass in the

361   UF retentate has been observed in pilot/full-scale studies but discharge to sewers seems to be

362   a common practice (Dixon et al. 2011, Sorlini et al. 2013) in addition to the internal

363   recirculation.

364   Deposition of algal cells and large MW EOM largely accounts for the formation of the

365   cake layer (and reversible resistance) on MF/UF membranes (Babel and Takizawa 2010)

366   which results in the loss of membrane permeability but increases the membrane selectivity

367   via pathway (IV) in Fig. 3. With the development of the cake layer on MF/UF membranes,

368   backwashing is frequently applied (Fig. 1). During this process, deposited cells, AOM and

369   toxins are transported from the membrane surface to the waste stream. While there has been

370   limited investigation of the nature of toxins in the concentrate and backwashing waste, it is

expected that intracellular/bound toxins are dominant in the concentrate while toxin-AOM$_{(aq)}$ and free toxins are likely to be found in the backwashing waste. This is because the shear stress of cross-flow during filtration causes minimal cell lysis in the bulk solution (Gijsbertsen-Abrahamse et al. 2006) while the release of intracellular toxins is likely to occur during the compression of the cake layer (Liu et al. 2017) which will be flushed into the backwashing stream.

The characteristics of the backwashing waste have been analysed in recent studies (Table 6). While the concentration of organics in the backwashing waste is comparable with the feed, the composition is likely different. For example, the study by Li et al. (2014) showed that EOM with MW higher than ~10 kDa was substantially found in the backwashing waste (compared to EOM with MW in the ranges of 0.3–10 kDa and 10–300 kDa in the feed). A similar phenomenon was observed by Qu et al. (2012b). Although MF/UF is designed for disinfectant-free production, disinfectants can be found in the backwashing water because either the backwashing water is from the disinfection tank or chemically-enhanced backwashing is practical to improve the cleaning efficiency (Chang et al. 2017, Zheng et al. 2011). Ferrer et al. (2014) reported that the addition of 7 mg/L NaOCl in the backwashing water could significantly increase the removal of biopolymers from the fouled membrane. However, due to the reaction between oxidants and AOM, this method likely results in the presence of aldehydes, ketones and highly toxic halogenated byproducts in the backwashing waste. It has been reported that the specific disinfection by-products yields of *Anabaena* were in the range of 2–11 μmol/mmol C for total trihalomethane and 2–17 μmol/mmol C for total haloacetic acid (Huang et al. 2009b). In addition, there are continual requirements to include an engineer's perspective in the scientific research. For example, the aims of reported studies of backwashing waste have been mainly focused on developing an understanding of reversible fouling on membranes and, as such, unreasonably high *BR* values (30−90%) have

396   been applied (Li et al. 2014, Qu et al. 2012b).

397

398   Table 6

399

400   Chemical cleaning (*e.g.* CIP) can be periodically used to remove the residual and

401   irreversible fouling, which is related to, for example, adhesion of metal oxides and HPO of

402   AOM on/in membranes (Liao et al. 2018, Zhang and Fu 2018). The typical chemical cleaning

403   protocols used in the H*c*-H*tox* scenario and the composition of the waste are summarised in

404   Table 6. Acid cleaning (*e.g.* HCl and HNO$_3$) aims at eliminating inorganic/metal oxide

405   fouling with proton-assisted and double decomposition reactions considered to be the key

406   mechanisms (Fig. 4a) (Wang et al. 2014). It is worthwhile noting that citric acid forms strong

407   solution complexes with metals, and the adsorption of citric acid can further weaken the

408   bonds in the proximity of a surface metal ion (M^+) center followed by slow detachment of the

409   surface M^+ to solution (Zhang et al. 2018). Zhang et al. (2011) compared the efficacy of 500

410   mg/L HCl and NaOH reagents in chemical cleaning of algal-fouled membranes. The results

411   of these studies showed that the HCl solution exhibited higher removal of irreversible

412   resistance than NaOH with the concentrations of dissolved organic carbon (DOC), protein

413   and carbohydrate in the chemical waste being 2−10 times of those of the feed (Zhang et al.

414   2011). However, more studies indicate that NaOH cleaning is more efficient and neutral

415   fractions dominate in the organic matter in the chemical cleaning waste (Table 6). Under

416   basic conditions, large organic matter is expected to be hydrolyzed and solubilized into small

417   aqueous molecules (Fig. 4b) (Naafs and van Bergen 2002, Yu et al. 2013) with the

418   electrostatic repulsion between the negatively charged membrane surface and AOM

419   facilitating the transport of foulants to the bulk solution (Wang et al. 2014). To improve the

420   efficacy of chemical cleaning at high pH, oxidative reagents such as NaOCl are frequently

added. The major mechanisms relate to the oxidation of the functional groups of AOM to ketonic, aldehydic, and carboxylic groups which, consequently, increases the hydrophilicity and reduces the adhesion of AOM (Fig. 4c) (Wang et al. 2014). The residual fouling layer is expected to change into a looser and more open structure (Wang et al. 2018). The study by Zhang et al. (2011) indicated that ~90% recovery of the UF permeability during treatment of algal-rich water could be achieved with use of 100 mg/L NaOCl solution though the composition of the waste was not analysed. Compared with acid and base cleaning, oxidant cleaning provides the possibility of also degrading toxins. For example, the second order rate constant for reaction of chlorine with microcystins is $100-500$ $M^{-1}s^{-1}$ (Ho et al. 2006). Nevertheless, the oxidative reagent dose is related to the solution matrix (*i.e.* the mass of the residual foulants after hydraulic cleaning) and also influences the distribution of toxins. Hall et al. (2000) indicated that a higher oxidative dose was required for algal toxin (MC-LR) removal when the solution matrix became more complicated (*i.e.* higher organic matter concentration). Moreover, on exposure to oxidant, transformation of intra- to extracellular toxins initially occurs due to cell lysis (Figs. 4c). Hall et al. (2000) and Zamyadi et al. (2012) have observed a temporal increase in extracellular/dissolved toxins at lower oxidant dose and/or contact time. This highlights the concern that CIP at insufficient dose of oxidants may generate a more toxic waste stream in the H*c*-H*tox* scenario.

Fig. 4

3.3. NF concentrate and chemical waste streams in the H*c*-H*tox* scenario

NF membranes can be used to polish the MF/UF permeate whilst retaining essential minerals/salts in the final product (Dai et al. 2019). In the H*c*-H*tox* scenario, the rejection mechanism of NF membranes is straightforward in view of their small MWCO (Table 1)

446 (Teixeira and Rosa 2005); as such, the proposed pathways of (II) size exclusion of toxin$_{(aq)}$

447 and (III) adsorption of toxin$_{(aq)}$ are expected to dominate (Fig. 3). Gijsbertsen-Abrahamse et

448 al. (2006) investigated the efficacy of the Trisep membrane (MWCO = 200 Da) in rejecting

449 MC-RR, MC-LR, MC-YR, MC-LA and anatoxin-a (at concentrations of 1.2−9.4 μg/L) with

450 the results showing that the permeate concentrations were far below the WHO-guideline

451 value of 1 μg/L. Compared to MF/UF, NF is also more efficient in removing odor compounds.

452 Choi et al. (2010) investigated the removal of seasonal odor compounds, 2-MIB (long-term

453 occurrence in January–May and August–September) and geosmin (short-term occurrence in

454 July–September and March–April) from the Han River water by NF membranes. Results

455 showed that both loose and tight NF membranes exhibited high rejection (r) of ≥98%.

456 Moreover, because MF/UF are always implemented prior to NF in practical applications in

457 the Hc-Htox scenario, the low MW neutral of HPI should be the dominant component in the

458 NF concentrate. It has been shown that in a full-scale nanofiltration plant, WR_{app} will be >75%

459 (*i.e.* CR < 25%), resulting in higher concentrations of low MW neutral and toxins in the

460 concentrate (Gijsbertsen-Abrahamse et al. 2006). However, to the best of our knowledge,

461 there has been no investigation of the management of the concentrate and chemical waste in

462 NF treated cyanobacteria-laden waters up to date.

463

464 3.4. RO concentrate and chemical waste streams in the Lc and Hc-Htox scenarios

465 RO is the state-of-the-art technology for seawater desalination (*i.e.* SWRO) while recent

466 attention has been given to innovative solar-driven and electrical desalination alternatives

467 (Ma et al. 2016, Xia et al. 2019). In the Lc scenario (at least for SWRO plants located in

468 Australia and Caribbean coastal areas according to the RO plan distribution map developed

469 by Villacorte et al. (2015)), the CR and CF values are largely related to salt rejection. In the

470 past forty years, the footprint of SWRO plants has been significantly reduced because of the

471   continual increase in water recovery (from CR ~75% in 1980s to $CR < 40\%$ if a second stage

472   is applied now) (Ghaffour et al. 2013). This also creates the opportunity for energy recovery

473   from the concentrate. While it is open to question whether water recovery in a single stage

474   can be further increased, high water recovery at low pressure has been achieved in SWRO

475   plants utilizing multi-stage operation (Ahunbay 2019). For the concentrate (or brine), a

476   critical challenge of SWRO in the Lc scenario however relates to the use of antiscalants (*e.g.*

477   polyacrylic acid and polymaleic acid) to prevent the formation of scale on pumps and

478   membranes. Typically, a dose of $0.5-10$ ppm is added to the feed solution (depending on the

479   scale-forming potential of the feed water, WR, and manufacturer's recommendations (Singh

480   2006)), which is then concentrated (to $1-20$ ppm) in the brine (Khan et al. 2009). Moreover,

481   while there has been substantial progress in development of membrane cleaning protocols

482   and reagents (*e.g.* NaOH and more specific products such as SUEZ Kleen* MCT cleaners

483   and AMI acid and alkaline membrane cleaners) for RO membranes (Melián-Martel et al.

484   2013), little attention has been paid to either the analysis of the composition of the chemical

485   cleaning waste or the sustainable management of the waste (Missimer and Maliva 2018).

486   In regard to the use of conventional primary and secondary pre-treatments (such as

487   coagulation, dissolved air flotation and/or MF/UF membranes (Busch et al. 2010)) in SWRO

488   systems (Fig. 3), algal cells are unlikely to be detected in the feed of the RO unit even in

489   HABs (*i.e.* in the Hc-Htox scenario), with toxins such as saxitoxin and domoic acid present at

490   relatively low concentrations. RO membranes can reject toxins (via pathways of (II) size

491   exclusion of toxin$_{(aq)}$ and (III) adsorption of toxin$_{(aq)}$ shown in Fig. 3) even though they are

492   designed for salt removal. Seubert et al. (2012) monitored the temporal concentrations (from

493   2005 to 2009) of saxitoxin, domoic acid, brevetoxin and okadaic acid within the intake and in

494   desalinated water from a pilot RO desalination plant in El Segundo, CA. Results showed that,

495   while saxitoxin and domoic acid were occasionally found in the intake waters, all toxins were

not detected in the desalinated water (Seubert et al. 2012). Full removal (>99%) of saxitoxin, brevetoxin and domoic acid has been shown to be achieved, even at elevated source water concentrations, in lab-scale studies (Laycock et al. 2012, Seubert et al. 2012).

From published literature (Li et al. 2016, Schurer et al. 2012, Schurer et al. 2013, Villacorte et al. 2015), it is expected that the transport of algal cells, AOM and toxins to the concentrate and waste streams in the MF/UF pretreatment units should be similar to that in drinking water treatment (Fig. 3) though the species present will be significantly different (Table 2). Of particular note is that transparent exopolymer particles (TEPs) have been recognised as a fouling indicator in SWRO but not in drinking water treatment though they are ubiquitous in marine and freshwater environments (Bar-Zeev et al. 2015). TEPs can be further specified as particulate TEP (pTEP, particle size > 0.4 μm) and colloidal TEP (cTEP, 0.05 μm < particle size < 0.4 μm) in the membrane field (Villacorte et al. 2009). cTEP can pass through UF membranes and will accumulate in the RO concentrate. RO is capable of retaining, if any, toxins and cTEP present in the feed solution (*i.e.* UF permeate) which, however, results in their accumulation in the concentrate and chemical waste. This can be challenging for decontamination and discharge because the abundant chloride ions in SWRO concentrate are a well-known scavenger for various strong oxidants (*e.g.* hydroxyl radicals).

4. Opportunities and perspectives for management of concentrate and waste streams

Following review of the characteristics of concentrate and waste streams in L*c* and H*c*-H*tox* scenarios, we will identify the opportunities and perspectives for their management. In regard to the cost and feasibility of management in existing plants, current strategies typically employ coagulation, adsorption, chemical oxidation (or disinfection) and energy conversion processes (Fig. 2b) which aim at (i) increasing the overall water recovery, (ii) beneficial use of the concentrate and/or (iii) decontamination of the concentrate and waste streams.

521 Guidance and perspectives for future studies will be discussed in an Eisenhower framework.

522 4.1. Opportunities for management of concentrate and waste streams

523 4.1.1. Recycling of MF/UF concentrate to increase the water recovery in the Lc scenario

524 Instead of direct discharge of concentrate and waste streams into sewers, one may consider

525 either installing a secondary MF/UF unit to recover the waste or blend the waste with the raw

526 water to the main MF/UF process (Gora and Walsh 2011, Zeman and Zydney 2017).

527 Although the concentrate and backwashing waste are significantly different in their

528 characteristics (Section 3.2 and Table 4), these two streams are often combined in the Lc

529 scenario, with the combined stream accounting for approximately 5−10% of the feed volume

530 (CR_{TOT} = 5−10%). In view of the high CF values of the concentrate/waste with respect to

531 turbidity (>20 at c_f of 1−3 NTU) and total organic carbon (>10 at c_f of 2−4 mg/L) but low CF

532 values for DOC (1−2 at c_f of 2−4 mg/L), coagulation can be applied (Fig. 2). Gora and Walsh

533 (2011) reported that a dose of 5−10 mg/L of alum (aluminium sulphate) or aluminium

534 chlorohydrate was sufficient for removing DOC by 20−50% and UV_{254} by 65−80%. Blending

535 of the combined concentrate (CR_{TOT} = 10%) with the raw water can likely improve the

536 removal efficiency of DOC, UV_{254} and true color (Gora and Walsh 2011). Moreover,

537 coagulation may benefit the membrane operation in the subsequent filtration step. Strong

538 flocs with low distribution spreading index in the feed solution of MF/UF are expected to

539 lower the specific cake resistance for these membranes (Huang et al. 2011).

540 4.1.2. Beneficial use of RO concentrate in the Lc scenario

541 There has been some discussion on possible alternatives to the management of desalination

542 brine (*i.e.* the concentrate) from SWRO plants such as use for establishment of solar ponds

543 for thermal energy and electricity production and use of the salts through the chlor-alkali

544 process (Khan et al. 2009). The hydraulic energy associated with the concentrate (due to the

545 high pressure operation of the RO unit) is recoverable via either a direct route to pressurise

546   the feed to RO or an indirect route to drive a turbine (Gude 2011). Typically, energy recovery

547   devices (ERDs) include Pelton turbine (centrifugal type), turbocharger (centrifugal type) and

548   isobaric ERD (positive displacement type) (Fig. 2b) (Stover 2007). The isobaric ERDs

549   demonstrate higher energy recovery efficiency (95−97%) than the centrifugal types (*i.e.*

550   Pelton turbine: ~87% and turbocharger: ~85%) though the latter is now more widely used in

551   SWRO as a result of their mechanical simplicity and operational flexibility (Gude 2011).

552   Another interesting opportunity relates to the use of the chemical potential (known as salinity

553   power) of the high-salinity concentrate (Logan and Elimelech 2012, She et al. 2012). For

554   example, the energy present as a result of the salinity difference can be recovered as a

555   hydraulic pressure that can be used to spin a turbine in the pressure-retarded osmosis (PRO)

556   process (She et al. 2012). At the same time, the brine after PRO treatment is diluted to

557   prevent the formation of a density plume during its discharge (Prante et al. 2014).

558   Thermodynamic and thermo-economic analyses indicated that the integration of PRO with

559   SWRO can reduce the energy consumption of desalination by ~40% (Prante et al. 2014,

560   Sharqawy et al. 2011). Alternatively, there has been increasing interest in development of

561   electrochemical processes (*e.g.* reverse electrodialysis (Mei and Tang 2017, Mei and Tang

562   2018)) to recover the salinity power of the concentrate. Nevertheless, both PRO and

563   electrochemical processes are still too expensive for full scale implementation due to their

564   relatively low power density in addition to membrane fouling and stability issues.

565   The beneficial use of membrane chemical cleaning waste is generally difficult because of

566   (i) the relatively small volume generated (3−5 times per year) and (ii) the very complex

567   composition (*e.g.* citric acid and EDTA at higher concentrations compared to antiscalants in

568   the concentrate). While chemical cleaning waste in SWRO plants is usually disposed to

569   sanitary sewers (Khan et al. 2009), more work is needed to better characterise the nature of

570   the waste streams and, subsequently, their potential environmental impacts.

4.1.3. Decontamination of MF/UF concentrate and waste streams in the H*c*-H*tox* scenario

In the H*c*-H*tox* scenario, recycling of the MF/UF concentrate (*i.e.* a combined stream of the concentrate and the backwashing and forward flushing waste with a typical *CR* of 10%) to the pretreatment units has been considered (Fig. 2b). The concentrations of algal cells, AOMs and total toxins are similar or lower than those in the feed water (*e.g.* typical values of 10^6-10^7 cell/mL, ~5 mg-DOC/L and $1-20$ μg/L) considering the dilution by backwashing waste. Conventional flocculants, including aluminum salts, polyacrylamide or chitosan, can form larger aggregates that largely remove algal cells (and intracellular toxins) and toxin-AOM$_{(aq)}$ in the concentrate (Dixon et al. 2011). Coagulation removes 95−99% of algal cells (and intracellular toxins) and 20−50% of DOC with an alum dose of 5−20 mg/L (Gora and Walsh 2011). Prior to clarification, a powdered activated carbon (PAC) dose of typically 5−25 mg/L can be applied to remove free toxins (*e.g.* 50−60% removal of 22 μg-anatoxin-a/L at a 5 mg/L PAC dose and 90% removal of 10 μg-anatoxin-a/L at a 11 mg/L PAC dose) (Vlad et al. 2014). The combined use of PAC with coagulants can further improve the removal of cyanobacteria (and intracellular toxins) such as *Anabaena*, *Microcystis* and *Pseudanabaena* to 99−99.9% (Zamyadi et al. 2013), which avoids the accumulation of algal cells in the concentrate during internal recycling. However, this practice results in elevated algal cell numbers and toxin concentrations in the clarifier (Fig. 2b). If the chemical sludge is not properly managed, breakthrough of cells and toxins in the effluent may occur (Zamyadi et al. 2012).

In order to reduce the final discharge of algal cells and toxins into the environment in the H*c*-H*tox* scenario, there has been increasing interest to integrate oxidation with membrane-based algal separation, although most studies focus primarily on membrane fouling control rather than the management of the concentrate (Liu et al. 2017, Ma et al. 2018). Chlorine-based oxidants (chlorine, chloramine and chlorine dioxide) are commonly

25

596   used reagents to oxidize cyanobacteria and toxins. While chlorine has reasonable activities

597   for microcystins (*e.g.* k_{app} of 100−500 $M^{-1}s^{-1}$) (Ho et al. 2006) and cylindrospermopsin, very

598   low rate constants (< 1 M^{-1} s^{-1}) have been observed for anatoxin-a (Vlad et al. 2014). In

599   comparison, ozone, permanganate and hydroxyl radicals show higher activities for

600   microcystins, anatoxin-a and cylindrospermopsin (despite the low second-order rate constant

601   for cylindrospermopsin oxidation with permanganate < 1 M^{-1} s^{-1}). The efficacy of the

602   oxidants is significantly influenced by the water matrix. For example, higher AOM content in

603   the feed will result in strong scavenging effects. In addition, the use of ozone and

604   permanganate can induce cell lysis that causes the transformation of intracellular toxins to

605   extracellular (or free) toxins (Hall et al. 2000). To mitigate this problem, the oxidation unit

606   can be placed after the coagulation and clarification unit (Fig. 2b) so that the influent algal

607   cell and AOM concentrations are minimized. The dose of oxidant is usually determined by

608   the organic matter concentration (*e.g.* 0.1−0.5 mg O_3/$KMnO_4$ per mg DOC at a typical

609   contact time of 30 min) rather than by the toxin concentration (Naceradska et al. 2017, Vlad

610   et al. 2014) because of the competition of AOM over low-abundant toxins for oxidants (*e.g.*

611    $k_{HO•,DOC} = 3.6 \times 10^8$ M^{-1} s^{-1} and $k_{HO•,MC\text{-}LR} = 2.3 \times 10^{10}$ M^{-1} s^{-1} with two orders of magnitude

612   difference in first-order rate constants) (Song et al. 2009, Westerhoff et al. 1999). This

613   strategy would nevertheless increase the cost of chemicals.

614   Considering the much smaller volume of the combined concentrate (*i.e.* <10%) compared

615   to the feed solution, the oxidation unit may be alternatively included in the recycling loop

616   (Fig. 2b). Pre-oxidation prior to coagulation may offer some benefits. For example,

617   Naceradska et al. (2017) found that permanganate is capable of preventing the formation of

618   Fe-peptide/protein complexes that inhibit coagulation, with the adsorption of organic matter

619   onto the hydrous MnO_2 generated improving the efficiency of the following coagulation step.

620   Essentially, the challenge relating to the reuse of the concentrate in the H*c*-H*tox* scenario

621 compared to that in the L*c* scenario is contributed by the algal concentration in the raw water,

622 with source reduction of the intake gaining in popularity (Figure 2b). For instance, a patented

623 flowing g-C$_3$N$_4$ photocatalyst capable of inactivating *M. aeruginosa* under solar irradiation in

624 contained cyanobacteria-laden water has been developed by Song et al. (2018b). Results

625 showed that up to ~90% removal and degradation of *M. aeruginosa* at an initial concentration

626 of 2.7×10^6 cells/mL can be achieved under 6 h of irradiation at a catalyst dose of 2 g/L. Cell

627 lysis occurs mainly as a result of the attack of HO• and h$^+$ generated on the semiconductor

628 surface. The intracellular and extracellular MC-LR is further degraded by the oxidation of the

629 Adda chain (Song et al. 2018a, Song et al. 2018b).

630 4.1.4. Critical pretreatment for RO in the H*c*-H*tox* scenario

631 For SWRO plants in the H*c*-H*tox* scenario, the potentially beneficial use of the concentrate

632 largely relies on the MF/UF pretreatment unit (*i.e.* reduction from the feed) because (i) the

633 high rejection of RO membranes will result in the accumulation of toxins in the concentrate

634 and (ii) decontamination of the concentrate at large volume is not cost-effective in view of

635 the scavenging effect of the complicated matrix. While the efficacy of ozonation is less

636 affected by chloride ions, more toxic byproducts can be formed in the presence of bromide

637 (Vlad et al. 2014). At the same time, best practice with respect to solid reuse of the

638 concentrate is under open discussion (Fig. 2b). More studies are required to understand the

639 relationship between the presence of toxins (and at what level) in the concentrate and

640 contamination of the salts in the chlor-alkali industry.

641 4.2. Perspectives for management of concentrate and waste streams

642 Fig. 5 summarises the relative state of knowledge and importance of topics discussed in

643 this review paper. Overall, there have been numerous studies on the removal of algae, AOM

644 and toxins (and odor compounds) by membrane separation, adsorption and oxidation in pure

645 water (or simple) matrices, such that these topics deserve low priority in future research

646　(lower right quadrant of Fig. 5). In comparison, more studies are needed on the impacts of

647　membrane permeability and selectivity on the transport of AOM and toxins in

648　membrane-based algal separation, especially at pilot scale with realistic water matrices (see

649　issues of ongoing-concern, upper right quadrant of Fig. 5). For example, the incorporation of

650　hydrophilic metal organic frameworks into the membrane matrix has been investigated to

651　create selective nanochannels for enhancing the rejection of micropollutants (Dai et al. 2019).

652　Results of the relevant studies have facilitated the construction of nanochannels with specific

653　characteristics (*e.g.* charge density and terminal functional groups) capable of improving the

654　water flux/decreasing *CR* whilst achieving selective removal of low-abundant compounds

655　(*e.g.* toxins and odor compounds). Currently, there is limited investigation on the formation

656　and control of byproducts when the backwashing water contains disinfectants. As such, this

657　issue requires ongoing attention in future studies (upper right quadrant of Fig. 5).

658

659　　　　　　　　　　　　　　　　　　Fig. 5

660

661　　　As previously mentioned in Section 4.1, there is a lack of studies on the disposal of

662　chemical waste generated from membrane cleaning in the H*c*-H*tox* scenario. The widespread

663　practice of discharge to the sewers can cause critical problems due to the presence of toxins

664　in the chemical waste (Fig. 2). Also, the reaction of algal cells, AOM and toxins with chlorine

665　likely results in the formation of highly toxic byproducts (Huang et al. 2009b), which has

666　been rarely considered in past studies. As such, priority should be given to the better

667　understanding and monitoring (*e.g.* through on-line electrochemical sensors) (Zhang et al.

668　2019) of the secondary pollution that may be generated during the disposal of the waste

669　streams in the H*c*-H*tox* scenario. Similar importance is assigned to the management of the

670　chemical sludge from the pretreatment step such as coagulation of the feed solution and

28

671   recycling of the concentrate (upper left quadrant of Fig. 5).

672   While adsorption followed by oxidation is very efficient in regenerating the concentrate,

673   critical challenges remain in regard to the low selectivity of the target pollutants (*e.g.* toxins)

674   that are present at much lower concentrations than the bulk natural organic matter (*e.g.* at

675   µg/L level compared to DOC at mg/L level). These needs call for novel methods for

676   management of the membrane concentrate in drinking water treatment (upper left quadrant of

677   Fig. 5). In this regard, design of heterogeneous AOPs with the catalysts/electrodes capable of

678   selective uptake of target pollutants to the interface followed by oxidation (Chen et al. 2020)

679   offers more opportunity for the future; however, research in this field is still in its infancy.

680   In addition, areas requiring additional research include the need for development of

681   composite membranes and new desalination technologies for minimal generation of

682   concentrate (*e.g.* spatially isolating salt crystallisation from water evaporation as described by

683   Xia et al. (2019)) (lower left quadrant of Fig. 5). Magnéli phase titanium suboxide, Ti_4O_7, has

684   the ability to be used in the preparation of reactive electrochemical membranes that enables

685   fine filtration whilst oxidising the pollutants deposited on the membrane surface (Chaplin

686   2014). This strategy is conducive to decreasing the *BR* of the backwashing waste. Long-term

687   evaluation and economic and life-cycle analyses of these alternatives will be invaluable in

688   identifying viable, low-cost processes for better management of the concentrate and waste

689   streams for membrane-based algal separation in water treatment and desalination.

690

5. Conclusions

692   In this paper, recent research advances in relation to the management of concentrate and

693   waste streams from membrane-based algal separation processes have been reviewed. From

694   the viewpoint of the characteristics of membranes (*e.g.* MWCO), concentrate and waste

695   streams (*e.g.* concentrations of algal cells, AOM and toxins as well as *CR* and *CF*), two

696 scenarios, (i) drinking water production/desalination at low algal concentrations and (ii)

697 cyanobacteria-laden water treatment/desalination are comprehensively discussed. A summary

698 of the fate of algal cells and metabolites in membrane processes facilitates the understanding

699 of the impacts of these transformations on the composition of concentrate and waste streams.

700 Current strategies are updated in regard to (i) recycling of MF/UF concentrate and beneficial

701 use of RO concentrate in the Lc scenario and (ii) decontamination of MF/UF concentrate

702 (and wastes) and pretreatment for RO feed in the Hc-Htox scenario. Identification of the

703 knowledge gap thereby provides insights to future studies of, for example, increasing the

704 low-pressure membrane rejection of toxins and management of the generation of secondary

705 pollutants in backwashing and chemical cleaning wastes.

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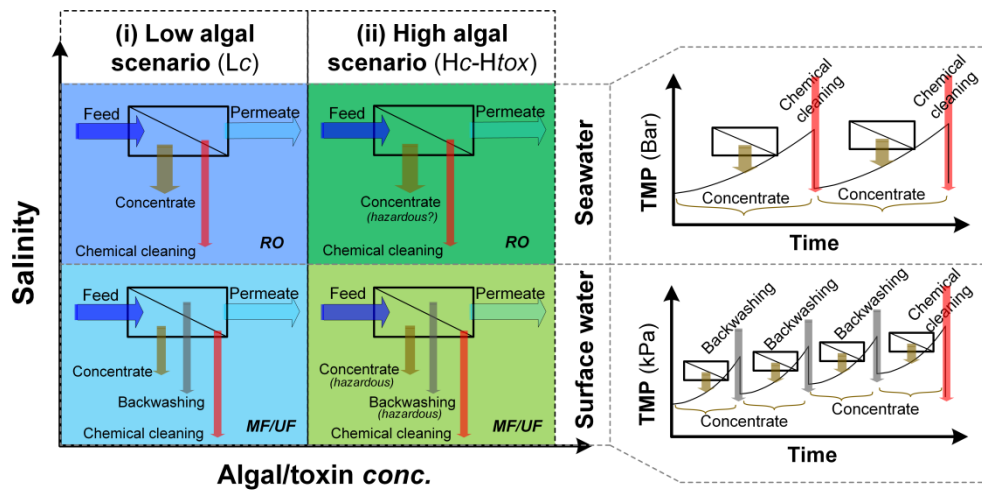
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Fig. 1. Two typical scenarios for membrane-based algal separation: (i) drinking water production and desalination at low algal/toxin concentrations (Low concentration, L*c*) and (ii) cyanobacteria-laden water treatment/desalination (High concentration and high toxicity, H*c*-H*tox*). The transmembrane pressure (TMP) *vs* time plots depict the temporal generation of the concentrate and waste streams (from backwashing and chemical cleaning).
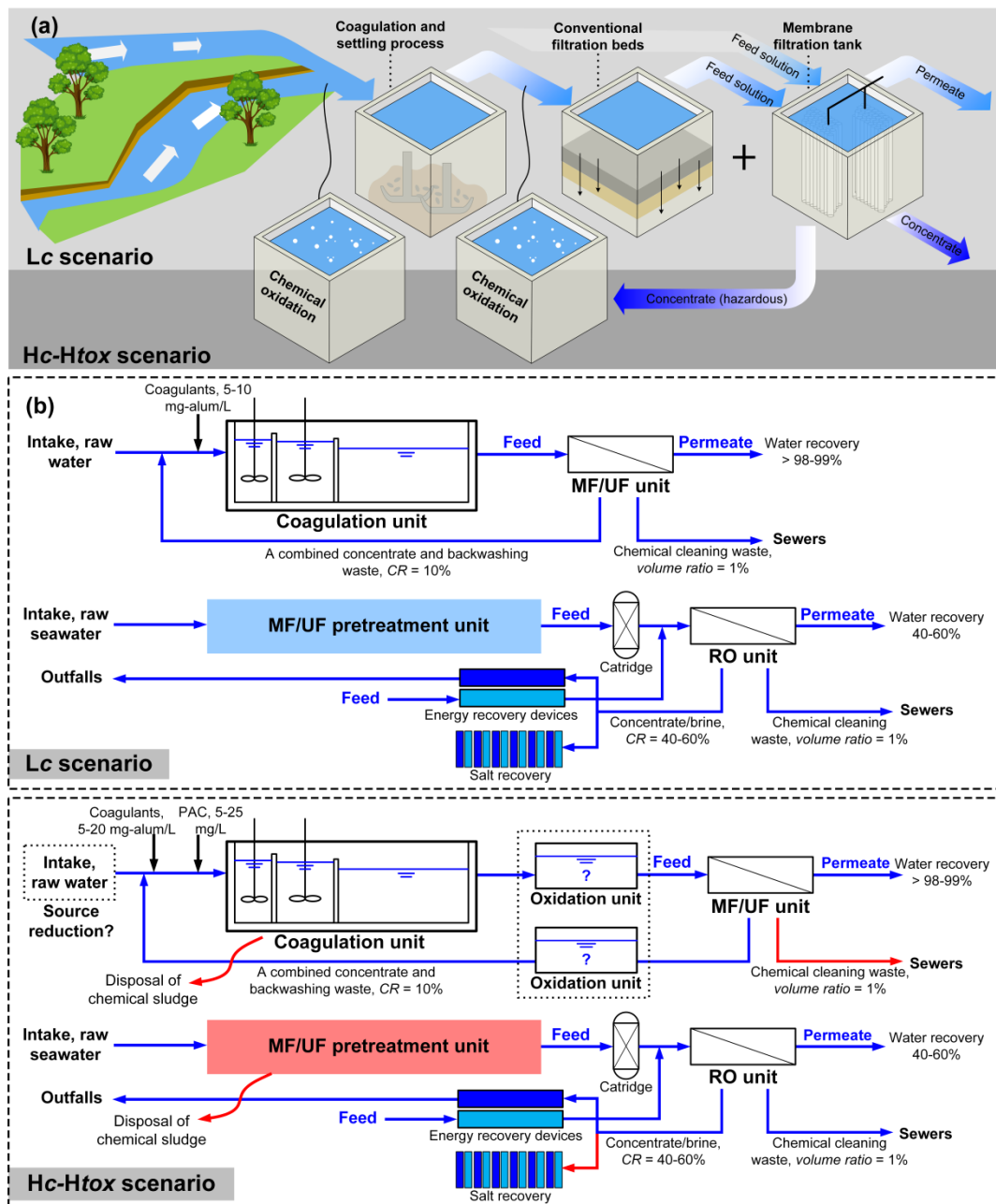
Fig. 2. (a) A flow diagram of treatment units likely included in L*c* and H*c*-H*tox* scenarios. The treatment processes are drawn based on Calgary's water treatment plants, Canada (https://www.calgary.ca/UEP/Pages/home.aspx). (b) Current methods and opportunities for management of the concentrate and waste streams in L*c* and H*c*-H*tox* scenarios.

Fig. 3. Composition of algal organic matter (AOM) and toxins, and their removal pathways by MF/UF, NF and RO membranes. The arrows of the flow diagrams indicate the transformation and fate of AOM and toxins in the concentrate and waste streams. The box chats describing the composition of AOM and toxins are for visualization purpose. Toxin-IOM$_{(aq)}$ and toxin-EOM$_{(aq)}$ respectively denote the aqueous toxin-IOM and toxin-EOM assemblages. From a general perspective, MF/UF is implemented as the pretreatment for NF and RO in the H*c*-H*tox* scenario.

Fig. 4. Comparison of the reaction mechanisms in (a) acid, (b) base and (c) oxidant cleaning, and effect of oxidant doses of (d) ozone and (e) chlorine on the distribution of intra- and extracellular (or total and dissolved) MC-LR. Data in Fig. 4d were retrieved from the study by Hall et al. (2000) with the figure created by the authors. Fig. 4e was adapted from Zamyadi et al. (2012). Copyright 2012 Elsevier.

Fig. 5. Eisenhower framework of research perspectives in management of the concentrate and waste streams for membrane-based algal separation.

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1085 **Table 2.** Typical characteristics of algae, toxins and odor compounds in harmful algal blooms

1086 (HABs) and red tides (Aune et al. 2007, Blanco et al. 2002, Brunson et al. 2018, Cusick and

1087 Sayler 2013, Furey et al. 2010, Granéli and Turner 2006, Jüttner and Watson 2007, Phukan et

1088 al. 2011, Villacorte et al. 2015, Watkins et al. 2008, Yoshida et al. 1997).

1089 **Table 3.** Summary of the basic indicators of the concentrate and waste streams (Chew et al.

1090 2016, Ferrer et al. 2016, Oh et al. 2006, Van der Bruggen et al. 2003, Wang et al. 2014)

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Figure 1

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Figure 2

Fig. 2. (a) A flow diagram of treatment units likely included in L*c* and H*c*-H*tox* scenarios. The treatment processes are drawn based on Calgary's water treatment plants, Canada (https://www.calgary.ca/UEP/Pages/home.aspx). (b) Current methods and opportunities for management of the concentrate and waste streams in L*c* and H*c*-H*tox* scenarios.

Figure 3

Fig. 3. Composition of algal organic matter (AOM) and toxins, and their removal pathways by MF/UF, NF and RO membranes. The arrows of the flow diagrams indicate the transformation and fate of AOM and toxins in the concentrate and waste streams. The box chats describing the composition of AOM and toxins are for visualization purpose. Toxin-IOM$_{(aq)}$ and toxin-EOM$_{(aq)}$ respectively denote the aqueous toxin-IOM and toxin-EOM assemblages. From a general perspective, MF/UF is implemented as the pretreatment for NF and RO in the H*c*-H*tox* scenario.

Figure 4
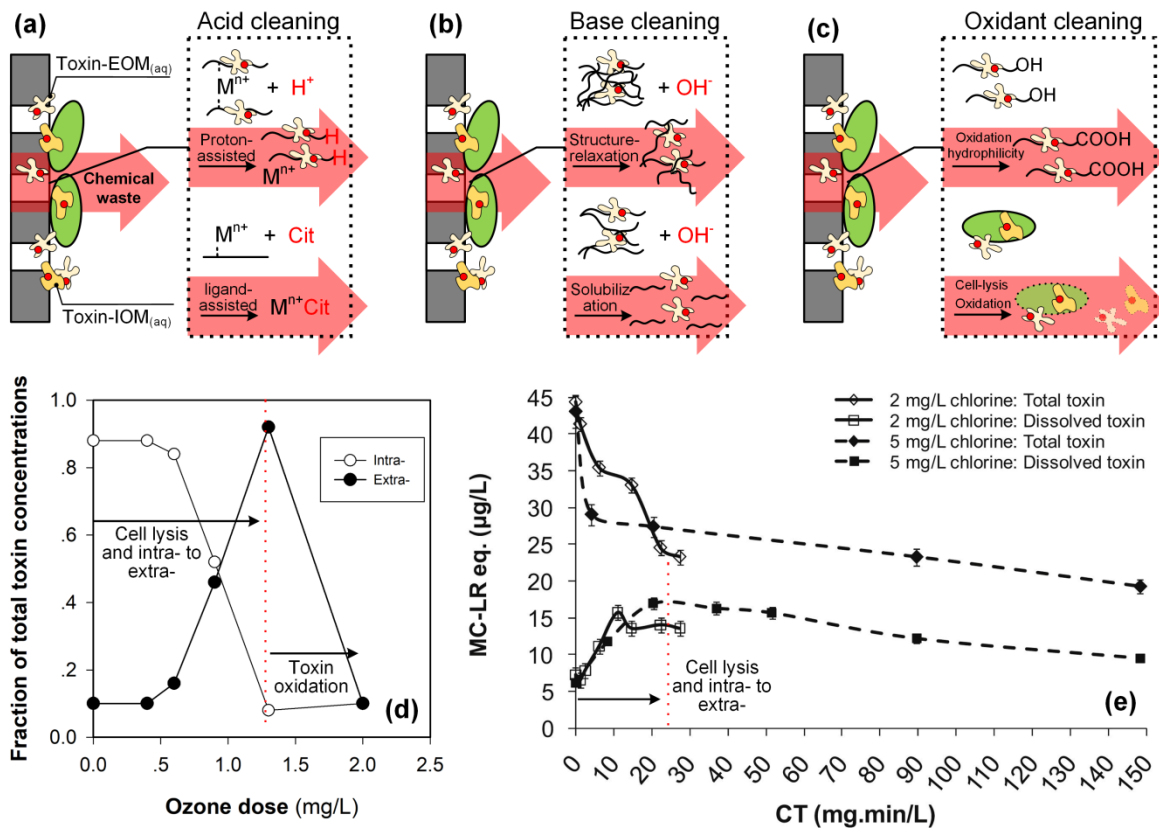
Fig. 4. Comparison of the reaction mechanisms in (a) acid, (b) base and (c) oxidant cleaning, and effect of oxidant doses of (d) ozone and (e) chlorine on the distribution of intra- and extracellular (or total and dissolved) MC-LR. Data in Fig. 4d were retrieved from the study by Hall et al. (2000) with the figure created by the authors. Fig. 4e was adapted from Zamyadi et al. (2012). Copyright 2012 Elsevier.

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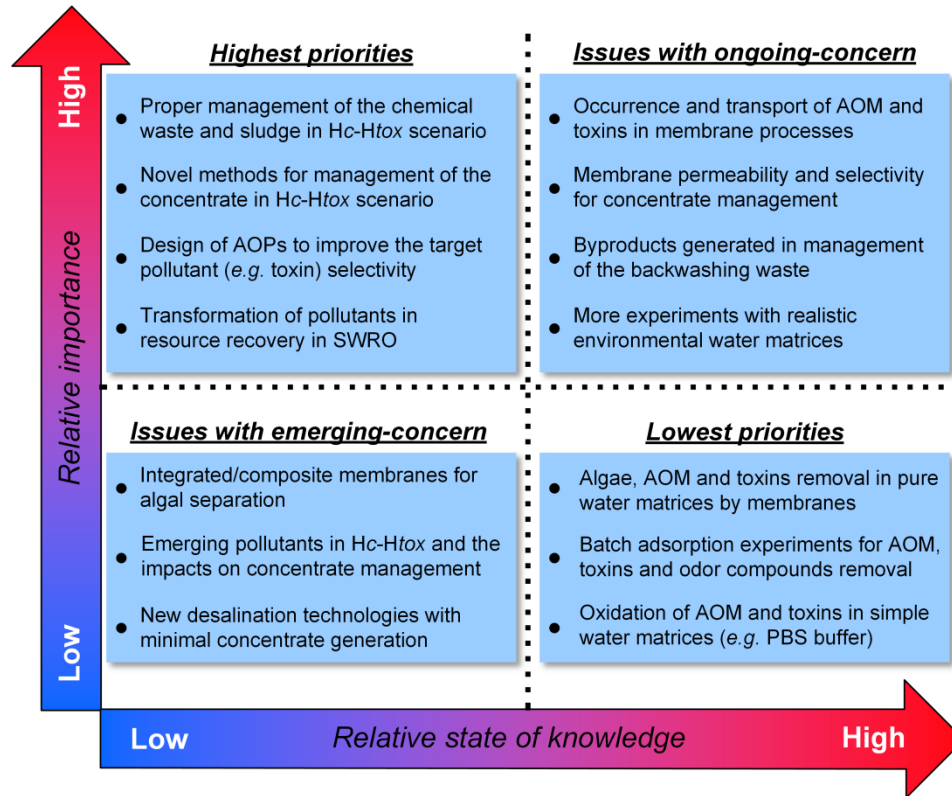
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Table 1

Table 1. Classification of membranes for algal separation in water treatment

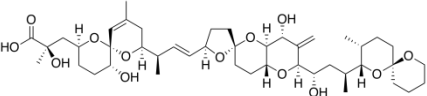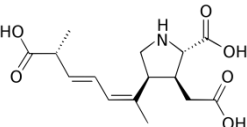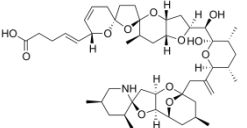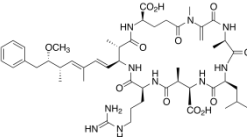| Type[*]	Pore Size	MWCO	Main applications[#]	Refs.
MF	0.1−1 μm	>1000 kDa	Desalination pretreatment in L*c* and H*c*-H*tox*	Villacorte et al. (2015)
UF	2−100 nm	10−100 kDa	Drinking water production in L*c*, colloidal organic matter and algal cell removal;	Davey and Schäfer (2009)
			Drinking water production in H*c*-H*tox*, algal cell, colloidal organic matter and toxin removal;	Gijsbertsen-Abrahamse et al. (2006), Liu et al. (2017)
			Desalination pretreatment in L*c* and H*c*-H*tox*	Villacorte et al. (2015)
NF	1−10 nm	100−1000 Da	Toxin and odor compounds[$] removal in H*c*-H*tox*;	Choi et al. (2010), Dixon et al. (2010)
RO	~1 nm	~100 Da	Desalination in L*c* and H*c*-H*tox*, toxin and odor compounds removal in H*c*-H*tox*	Villacorte et al. (2015) Schoonenberg Kegel (2010)
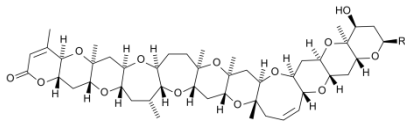
* MF: microfiltration; UF: ultrafiltration; NF: nanofiltration; RO: reverse osmosis; [#] L*c*: drinking water production and desalination at low algal concentrations; H*c*-H*tox*: cyanobacteria-laden water treatment/desalination at high concentration and toxicity.
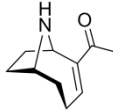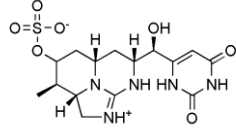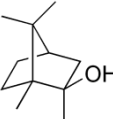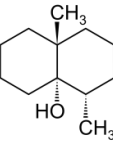
[$] The most common algae derived odor compounds are 2-MIB and geosmin.

Table 2

Table 2. Typical characteristics of algae, toxins and odor compounds in harmful algal blooms (HABs) and red tides (Aune et al. 2007, Blanco et al. 2002, Brunson et al. 2018, Cusick and Sayler 2013, Furey et al. 2010, Granéli and Turner 2006, Jüttner and Watson 2007, Phukan et al. 2011, Villacorte et al. 2015, Watkins et al. 2008, Yoshida et al. 1997).

Syndrome	Toxin/odorant	Molecular weight, Da	pK_a	Toxicity $(LD_{50})^*$	Solubility	Growth area	Representative species
Paralytic shellfish poisoning	Saxitoxin	299.3	$pK_{a1} = 8.22$; $pK_{a2} = 11.3$	3–10	Water	Marine	*Alexandrium catenella*, *Alexandrium tamrensis*, *Gymnodinium catenatum*, *Pyrodinium bahamense*, etc.
Neurotoxic shellfish poisoning	Brevetoxin			170–350	Lipid	Marine	Dinoflagellate: *Karenia brevis*, etc.
	BTX PbTx-1	867.1					
	BTX PbTx-2	895.1	$pK_a = 13.7$				

			(Predicted)				
Diarrhetic shellfish poisoning	Dinophysistoxin		$pK_a = 3.76$	200	Lipid	Marine	*Dinophysis* spp, *Prorocentrum donghaiense*
	Okadaic acid	805.0					
Amnesic shellfish poisoning (ASP)	Domoic acid	311.3	$pK_{a1} = 1.85$; $pK_{a2} = 4.47$; $pK_{a3} = 4.75$; $pK_{a4} = 10.6$	4000	Water	Marine	*Pseudo-nitzschia*
Azaspiracid shellfish poisoning	Azaspiracid	842.1	$pK_a = 3.95$; (Strongest Acidic)	200		Marine	*Azadinium spinosum*
Cyanobacterial toxin poisoning	MCs MC-LR	900~1100	$pK_a = ~3.5$	~60	Water (MC-LR > 1 g/L)	Fresh water	*M. aeruginosa*

	Compound	MW	pK_a	LD$_{50}$*	Solubility	Water type	Species
	Anatoxin-a	165.2	pK_a = 9.36	380	Water	Fresh water	*Anabaena flos-aquae*
	Cylindrospermopsin	415.4	pK_a = 8.8	>2000	Water	Fresh water	*Cylindrospermopsis raciborskii*
Algae driven odor	2-MIB	168.3	n.a. [$]	Low/non-toxicity	Water (0.45 g/L)	Fresh water	*Oscillatoria limosa, Planktothrix agardhii, Pseudanabaena catenata*
	Geosmin	182.3	n.a.	Low/non-toxicity	Water (0.05-0.15 g/L)	Fresh water	*Aphanizomenon flos-aquae Planktothrix agardhii, Pseudanabaena catenata*
Non-hazardous microalgae				Low-toxicity		Fresh water & marine	*Chlorococcales, Chlorella* sp, etc.

*LD$_{50}$ is the single dose of a toxic substance at which 50% of the individual mice will be killed (µg/kg); $n.a.: not available

Table 3

Table 3. Summary of the basic indicators of the concentrate and waste streams (Chew et al. 2016, Ferrer et al. 2016, Oh et al. 2006, Van der Bruggen et al. 2003, Wang et al. 2014)

Indicator	Definition	General results
Concentrate		
CR	$$CR = \frac{q_c}{q_f} = 1 - WR_{app} = 1 - \frac{q_p}{q_f} \quad (1)$$ where q_f, q_c and q_p represent the volumetric flow rates of the feed solution, the concentrate and the permeate, respectively.	$CR_{MF/UF}$ = 1−10%, CR_{NF} = 15−30%, and CR_{RO} =15−60%
CF	$$CF = \frac{(q_c + q_p)c_{f,i} - q_p c_{p,i}}{q_c c_{f,i}} = 1 + \left(\frac{1}{CR} - 1\right)r \quad (2)$$ where $c_{f,i}$, $c_{c,i}$ and $c_{p,i}$ refer to the concentrations of component i in the feed solution, the concentrate and the permeate, respectively, and r is the rejection for component i.	For algal cells, the rejection of all membranes is close to 1, *i.e.*, *CF* $\approx$ 1/*CR*.
Backwashing waste*		
BR	$$BR = BW_d \times BW_f \times \frac{q_{BW}}{q_f} \quad (3)$$ $$\approx (0.8 - 1.2) \times BW_d \times BW_f$$ where q_{BW} is the volumetric flow rate of the backwashing stream. q_{BW} can be estimated according to Darcy's equation (eq. 4): $$q_{BW} = \int J_t A = \int \frac{\Delta P_{BW,t} A}{\mu(R_m + R_{rev,t} + R_{irrev})} \quad (4)$$ where R_m, $R_{rev,t}$ and R_{irrev} are the clean membrane resistance, and hydraulically reversible (at time t) and irreversible resistances respectively. J, A and μ are the flux, membrane area and solution viscosity.	q_{BW}/q_f = of 0.8−1.2 and *BR* = 5−15%

* The concentrate factors for waste streams are generally determined by direct measurement of the concentration of constituents (*e.g.* algal cells, AOM and toxins) in the waste.

Table 4

Table 4. Comparison of the basic characteristics of membrane concentrate and waste streams in L*c* and H*c*-H*tox* scenarios

Scenarios and membranes*	Feed solution	Concentrate	Backwashing	Chemical cleaning	Refs.
L*c*: Surface water					
SKMF01-106, SK Chemicals, Korea MWCO = 150 kDa	Raw water; Turbidity = 1.9–214 NTU (average 3.7 NTU).	Turbidity = 99 NTU; **CF = 27 (CR = ~4%)$**			Oh et al. (2006)
PAN UF (MWCO = 50 kDa)	Raw water; Turbidity = 23 NTU.		**BR = 2−5%**		Xia et al. (2004)
PVC UF (MWCO = 80 kDa)	Raw water; Turbidity = 6–20 NTU.		**BR = 1.7−3.3%**	Volume ratio: ~1%	Guo et al. (2009)
L*c*: Desalination					
RO membranes	Typical seawater; Salts = ~33−37 g/L.	**CR = 40−60%** **CF = 1.5−1.8**			Khan et al. (2009)
H*c*-H*tox*: Surface water					
PVDF MF (pore size = 0.1 μm)	Raw water; Cyanobacteria = 1.3−2.4 × 10^6 cells/L	Algal rejection, *r*: >98%	**BR = 0.8−1.6%**	Volume ratio: ~0.2−0.3%	Sorlini et al. (2013)

Hydrophobic PES membrane (Sartorius, Germany) MWCO = 100 kDa	Synthetic *M. aeruginosa* IOM^ and EOM solutions; DOC = ~8 mg/L; MCLR (in IOM) = 75.5 µg/L, MCLR (in EOM) = 7.85 µg/L.	***CR = 10%*** **Rejection, *r*:** IOM = 57%; EOM = 46%; MCLR-IOM$_{(aq)}$ = 78% MCLR-EOM$_{(aq)}$ = 98%		Li et al. (2014)
PVDF UF (pore size = 0.02 µm)	Raw water with dose of *A. circinalis* at 100,000 cells/mL; DOC = 4.0–6.2 mg/L, turbidity = 12–15 NTU; Saxitoxin = 2.2–2.7 µg/L STX-eq in the feed water of UF with extracellular of 0.7–0.8 µg/L STX-eq.	**Rejection, *r*:** Up to 78% for the total, and up to 47% for the extracellular STX-eq.	***BR = ~10%***	Dixon et al. (2011)
Trisep TS80 4040 NF (MWCO = 200 Da)	Tap water; MCs = 1.2−9.4 µg/L *or* Anatoxin-a = 4.6−4.8 µg/L	***CR = 90%*** **Rejection, *r* > 93−99%**		Gijsbertsen-Abra hamse et al. (2006)

H*c*-H*tox*: Desalination

RO membranes	Typical seawater;	Not detectable in the		Seubert et al.

Salts = ~33−37 g/L, total domoic   desalinated water   (2012)

acid = 0−14.1 μg/L

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*PAN: polyacrylonitrile; PVC: polyvinyl chloride; PVDF: polyvinylidene fluoride; PES: polyethersulfone; $Assume the rejection is ~100%; #TOC: total organic carbon; ^IOM and EOM: Intracellular organic matter and extracellular organic matter.

Table 5

Table 5. Characteristics of the EOM derived from representative algal species.

Algal species*	HPI/HPO, %		Molecular weight (MW) distribution
	HPI	HPO	
M. aeruginosa	69	28	Non-peptide: >30 kDa: 29−38%, <1 kDa: 44−48%
			Peptide/protein: >30 kDa: 5−46%, < 1 kDa: ~0%
Anabaena flos-aquae	85	17	>100 kDa: ~35%, <10 kDa: ~50%
Aphanizomenon flos-aquae	79	17	>100 kDa: ~46%, <10 kDa: ~50%
Chlorella vulgaris	60−71	11−22	> 30 kDa: 62%, < 1 kDa: 30%
Asterionella formosa	70−73	15−20	> 30 kDa: 9%, < 1 kDa: 81%
Melosira sp.	64	32	> 30 kDa: 30%, < 1 kDa: 53%
Chaetoceros affinis	n.a.		>20 kDa: 51%, < 1 kDa: 49%
Chaetoceros muelleri	n.a.		>20 kDa: 38.5%, < 1 kDa: 60.9%

*M. aeruginosa (Pivokonsky et al. 2014): EOM was extracted by filtering through a 0.22 μm membrane filter. The MW distribution was fractionated by centrifugation-driven filtration;

Anabaena flos-aquae and *Aphanizomenon flos-aquae* (Huang et al. 2014): AOM (mainly EOM) was extracted by centrifuging at 10,000 *g* for 15 min and then filtering through a 0.45 μm filter. HPI is a sum of negatively charged HPI and the neutral HPI. The MW distribution was determined by high performance size exclusion chromatography;

Chlorella vulgaris, *Asterionella formosa* and *Melosira sp.* (Henderson et al. 2008b): AOM (mainly EOM) was extracted by centrifuging at 10,000 *g* for 15 min and then filtering through a 0.7 μm Whatman GF/F glass micro-fibre. The MW distribution was fractionated by centrifugation-driven filtration;

Chaetoceros affinis (Tabatabai et al. 2014): AOM (mainly EOM) was recovered through a sedimentation method. The MW distribution was determined by liquid chromatography-organic carbon detection.

Chaetoceros muelleri (Chekli et al. 2017): AOM (mainly EOM) was extracted by filtering through a 0.45 μm nylon membrane filter. The MW distribution was determined by liquid chromatography-organic carbon detection.

$n.a.: not available.

Table 6

Table 6. Components or characteristics of backwashing and chemical cleaning wastes in L*c* and H*c*-H*tox* scenarios[*]

Backwashing

Feed	Backwashing water	Backwashing waste	Refs
Decanted water from the settling basin of a drinking water treatment plant (Barcelona, Spain). TOC = 4.1 ± 0.24 mg/L	UF permeate DOC = 3.6 mg/L	DOC = 4.7 mg/L	Ferrer et al. 2016
Surface water from Gueui Intake on the Han River (South Korea). DOC = 1.03–2.95 mg/L	Water Water + air	TOC = 1.5 mg[#] TOC = 2.1 mg	Oh et al. 2006
Schie Canal water (Delft, the Netherlands). DOC = 12.7 mg/L (Biopolymer (> 20 kDa) = 0.45 mg/L, humics (~1 kDa) = 8.39 mg/L)	DI water UF permeate (Biopolymer = 0.14 mg/L, humics = 7.92 mg/L)	Biopolymer = 1.81 mg/L, humics = 3.48 mg/L Biopolymer = 1.81 mg/L, humics = 8.49 mg/L	Li et al. 2012
Algal solution. DOC of EOM = 8.12 mg/L	Milli-Q water	EOM with MW higher than ~10 kDa	Li et al. 2014

Chemical cleaning

Feed	Chemical washing solution	Chemical cleaning waste	Refs
Surface water from Gueui Intake on the Han River (South Korea). DOC = 1.03–2.95 mg/L	0.1 N HCl 0.1 N NaOH	TOC = 9.4 mg TOC = 228.6 mg	Oh et al. 2006

Chitose river (Japan). DOC = 2.4 mg/L	0.01 M HCl	DOC = 2.4 mg/L (Protein: 2.2 mg/L)	Kimura et al. 2004
	0.01 M NaOH	DOC = 22.2 mg/L (Protein: 8.7 mg/L; carbohydrate: 5.2 mg/L and neutral fractions dominate)	
Lujing (Nantong) water supply plant (China) DOC = 1.55−2.24 mg/L	0.01 M HCl	DOC = 0.14−0.51 mg/L (Proteins, polysaccharides with high MW (>10 kDa))	Xiao et al. 2012
	0.01 M NaOH	DOC = 0.68−1.15 mg/L (Humic-like substances 1 kDa, and proteins, polysaccharides with high MW (>10 kDa))	
Algal-rich water (DOC = 1.69 mg/L, $M.\ aeruginosa = 1−1.5 \times 10^8$ cells/L)	500 mg/L HCl	DOC = 18.2 mg/L (Protein: 1.01 mg/L; carbohydrate: 3.05 mg/L)	Zhang et al. 2011
	500 mg/L NaOH	DOC = 13.9 mg/L (Protein: 0.73 mg/L; carbohydrate: 2.1 mg/L)	
	150 mg/L EDTA[$]	DOC = 15.3 mg/L (Protein: 0.75 mg/L; carbohydrate: 2.62 mg/L)	
Taihu Lake (Wuxi, China) (DOC = 3.3 mg/L, the main algae categories are *Microcystis* and *Chlorella*)	HCl[&]	TOC = 4.1 mg/L (Neutral HPI and HPO dominate)	Huang et al. 2015.
	NaOH+NaOCl	TOC = 19.3 mg/L (Neutral HPI and HPO dominate, with organics having three MW ranges (300	

kDa, 1400 Da, and 500 Da))