

Efficient integrated module of gravity driven membrane filtration, solar aeration and GAC adsorption for pretreatment of shale gas wastewater

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1 Gravity driven membrane filtration combined with
2 solar aeration and GAC adsorption provides
3 excellent productivity and effluent quality as shale
4 gas wastewater pretreatment

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16 **ABSTRACT**

17 The rapid growth of shale gas extraction is associated to the increasing production of shale
18 gas flowback and produced water: efficient treatment processes are urgently needed to allow
19 better management of this wastewater. We propose a simple integrated pretreatment process for
20 on-site treatment, whereby gravity driven membrane filtration is combined with granular
21 activated carbon (GAC) adsorption and solar aeration. GAC and aeration significantly
22 increased the stable flux and improved the final effluent quality of the membrane process.
23 Specifically, the dissolved organic carbon removal rate of the integrated system was 44.9%,
24 and the stable permeate flux was 1.7 times higher than that of simple gravity-driven filtration,
25 which also showed negligible removal of organic. The high stable flux is attributed to a
26 reduction of extracellular polymeric substances accumulated on the membrane, as well as to
27 the more porous and heterogeneous biofilm formed thanks to the abundance and diversity of
28 eukaryotes with active predation behavior. The prevailing strains, *Gammaproteobacteria*
29 (35.5%) and *Alphaproteobacteria* (56.5%), played an important active role in organic carbon
30 removal. The integrated system has great potential as pretreatment for shale gas flowback and
31 produced water desalination due to its low energy consumption, low operational costs, high
32 productivity, and effluent quality.

33

34 **KEYWORDS:** Shale gas flowback and produced water (SGFPW); Gravity driven membrane
35 (GDM); Granular activated carbon (GAC); Aeration; Microbial community

36 **Introduction**

37 Shale gas may satisfy the current world's energy demand for over 60 years, and it is
38 considered as a better resource to replace traditional fossil fuels and to help reducing the
39 global carbon emissions (Chang et al., 2019a; Shaffer et al., 2013). However, the extraction of
40 shale gas is presently associated with severe environmental problems, especially related to the
41 great amount of freshwater that is consumed in the extraction activity and to the large flow of
42 refractory shale gas flowback and produced water (SGFPW), with $\sim 5200-25,870 \text{ m}^3$
43 generated per well (Chang et al., 2019b; Zou et al., 2018). SGFPW contains high
44 concentrations of salinity, radionuclides, heavy metals, and refractory organics, seriously
45 endangering human health, wildlife, and water ecosystems if not appropriately managed
46 before discharge (Abass and Zhang, 2020; Butkovskiy et al., 2017). Furthermore, its quality
47 and quantity change over time. For example, its salinity gradually increases, while the total
48 organic carbon concentration reduces gradually with the life of the well (Barbot et al., 2013;
49 Cluff et al., 2014). Shale gas extraction wells are often located in remote areas with scarce
50 transportation and power facilities, making SGFPW treatment even more challenging.

51 Membrane technologies are considered the most appropriate and effective way to reuse
52 SGFPW, achieving a sustainable cycle of water in the shale gas industry (Chang et al., 2019a;
53 Tong et al., 2019). Desalination may be achieved by nanofiltration, reverse osmosis, forward
54 osmosis, or membrane distillation. However, effective pretreatment is a significant factor in
55 the sustainable operation of desalination (Chang et al., 2019c). Luckily, pretreatment can be
56 effectively performed by low-pressure membrane processes, such as microfiltration (MF) and

57 ultrafiltration (UF) (Guo et al., 2018; Islam et al., 2019; Kim et al., 2018; Miller et al., 2013).
58 Nevertheless, the appeal of such technologies is limited by the operational problems
59 associated with membrane fouling (Chang et al., 2019c; Lee et al., 2019b). The recently
60 developed gravity driven membrane filtration (GDM) is more favorable than conventional
61 MF and UF in pretreating SGFPW (Chang et al., 2019c; Pronk et al., 2019). GDM has the
62 advantages of simple operation, low maintenance, low energy consumption, and lower costs
63 in general, mainly because its stable flux is realized by gravity and because the membrane
64 does not need cleaning (Pronk et al., 2019). This technology has specific potential for the
65 treatment of SGFPW generated from decentralized extraction wells.

66 In recent years, GDM technology has been successfully applied in many fields, such as
67 in the treatment of surface water (Boulestreau et al., 2012; Chawla et al., 2017;
68 Peter-Varbanets et al., 2010; Shao et al., 2019; Shi et al., 2020; Song et al., 2020a; Tang et al.,
69 2018b; 2020b; Truttmann et al., 2020), rainwater (Ding et al., 2017b; Du et al., 2019; Wu et
70 al., 2019), greywater (Ding et al., 2016; 2017a), sewage (Liu et al., 2020; Wang et al., 2017),
71 and seawater (Akhondi et al., 2015; Wu et al., 2016; Wu et al., 2017). In our previous research
72 (Chang et al., 2019c; Li et al., 2020), the suitability of the GDM process as a pretreatment
73 option for SGFPW desalination was discussed, also assessing the long-term effects of
74 operational parameters and analyzing the microbial community of the biofouling layer. While
75 the performance was better than that of traditional UF, the GDM process still needs significant
76 improvement. Chiefly, both the stable flux and the contaminant removal should be maximized
77 to alleviate the fouling potential of the stream entering the desalination process.

78 In recent articles, the combination of GDM with other separation processes was
79 discussed and broader conditions were evaluated, including the use of a biofilm reactor,
80 adsorption, coagulation, and aeration (Ding et al., 2018b; Lee et al., 2019a; Lee et al., 2019b;
81 Shao et al., 2017; Tang et al., 2018a; Tang et al., 2018b; Tang et al., 2018c). Specifically,
82 granular activated carbon (GAC) has been reported to remarkably improve the permeate
83 quality of GDM systems (Ding et al., 2018b; Lee et al., 2019b; Tang et al., 2018c). Regarding
84 stable flux, Ding et al. (2018b) indicated that this was reduced because the GAC layer
85 increased the filtration resistance. Lee et al. (2019b) attributed this effect to the lower
86 presence, predation, and mobility of eukaryotes. In contrast, Tang et al. (2018c) found that
87 GAC improved the diversity of eukaryotes with stronger predation ability in the biofouling
88 layer, thus producing a more permeable biofouling layer. Additionally, Ding et al. (2016)
89 analyzed the effect of aeration shear stress on a GDM system for greywater treatment. When
90 the aeration was positioned below the membrane module, shear stress resulted in a thinner,
91 denser, and less permeable biofouling layer with high EPS content, thus aggravating
92 membrane fouling. Meanwhile, Peter-Varbanets et al. (2011) and Ding et al. (2017a) reported
93 that high dissolved oxygen (7.9 mg/L, 6.0-6.5 mg/L) can promote high stable fluxes due to
94 the enhanced biological activity, larger surface roughness, and lower EPS content in the
95 biofouling layer. The effect of aeration on the GDM system is thus complex. On the one hand,
96 aeration increases the dissolved oxygen level and the permeability of the biofouling layer. On
97 the other hand, aeration shear stress might aggravate membrane fouling.

98 In this study, the effect of GAC and aeration on GDM performance is explored and

99 analyzed in the pretreatment of SGFPW . The specific objectives are to assess the effect of
100 GAC and aeration on (1) stable flux, membrane fouling resistance, and permeate quality; (2)
101 the morphology and accumulated biofoulants on the membrane; and (3) the diversity of
102 bacterial and eukaryotic community present in the membrane biofilm. Therefore, an
103 integrated system is proposed to improve the efficiency of SGFPW pretreatment for
104 subsequent desalination.

105

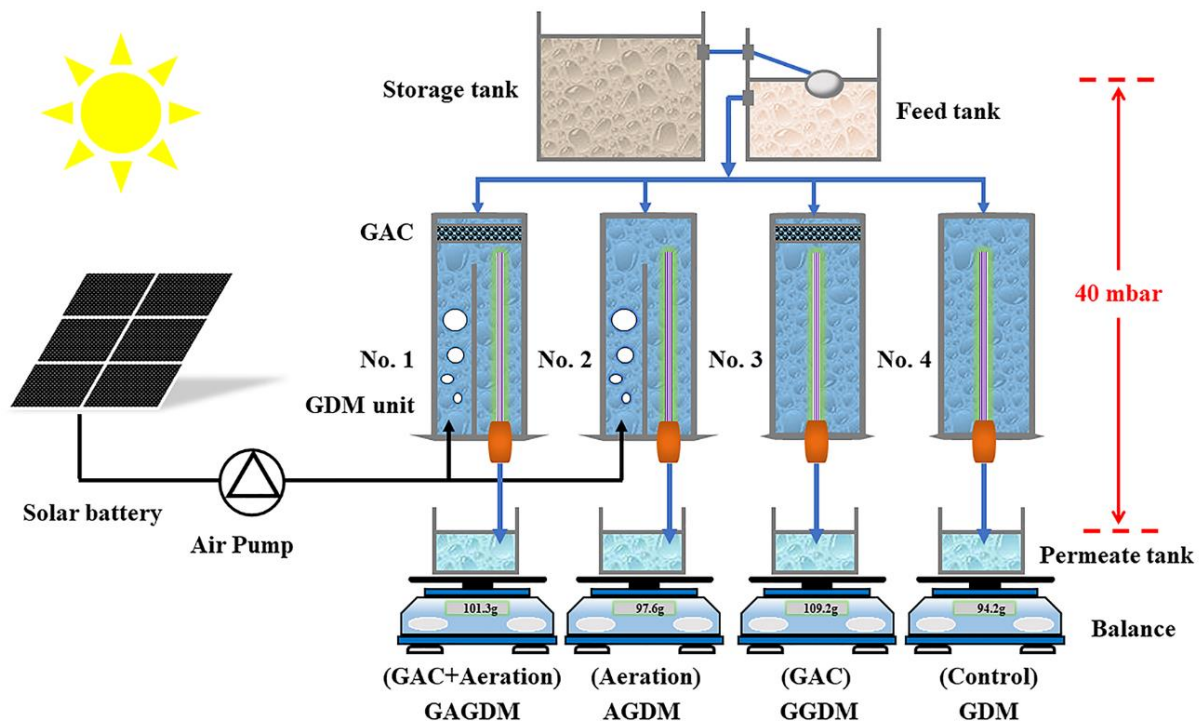
106 **Materials and methods**

107 *2.1 Gravity driven membrane filtration systems*

108 A schematic diagram of the four GDM systems utilized in this work is shown in **Fig. 1**.
109 The systems were operated in parallel at room temperature (~ 15°C) with hydrostatic pressure
110 of 0.04 bar. The characteristics of the poly(vinylidene fluoride) hollow fiber UF membranes
111 (Litree Purifying Technology Co., Ltd., China, with an effective membrane area of 15 cm²)
112 employed in this study can be found in previous reports (Li et al., 2020; Tang et al., 2020a).
113 After 30 days of operation, the microbial environment tended to become stable. Then, No. 1 and
114 No. 2 GDM units were aerated, and GAC was added to GDM units No. 1 and No. 3, to explore
115 the effects of aeration and adsorption on the systems in the following 30 days of operation.

116 Specifically, a single crystal silicon solar air pump (Koge, Xiamen, China) continuously
117 aerated No. 1 and No. 2 GDM units at a flow rate of 60 mL/min. To avoid direct erosion of
118 membrane by aeration, thus aggravating membrane fouling effects (Ding et al., 2016), the
119 aerators and the membrane modules were located on opposite sides of the reactors. The

120 concentration of dissolved oxygen measured by HQ30D dissolved oxygen analyzer (Hach
 121 Company, USA) was above 8 mg/L in aerated systems. In GDM units No. 1 and No. 3, 10 g
 122 GAC (CPG LF 12, Calgon Carbon Co., Ltd., USA) were added. GAC was cleaned with
 123 deionized water and dried before dosing. It was wrapped in gauze to prevent leakage, which
 124 might cause membrane fouling (Ding et al., 2018a; Ding et al., 2018b).



125
 126 **Fig. 1.** Schematic diagram of the GDM systems. Four systems (GAGDM: GDM with
 127 GAC+aeration; AGDM: GDM with aeration only; GGDM: GDM with GAC only; GDM:
 128 control GDM) operated for a total of 60 days at room temperature (~ 15°C) with hydrostatic
 129 pressure of 0.04 bar.

130
 131 *2.2 Wastewater samples and water quality analysis*

132 The shale gas flowback and produced water sample used in this study was collected from a
 133 drilling platform of the Weiyuan shale gas field in the Sichuan Basin, China. Compared to water

134 samples used in our previous research (Chang et al., 2019c; Li et al., 2020), the water samples
 135 utilized in this experiment were pale yellow and had a lower amount of suspended matters. The
 136 SGFPW samples were kept in sealed containers and in the dark to avoid changes in water
 137 quality. The pH value of the wastewater was measured using a pH meter (PB-10, Sartorius
 138 Scientific Instruments Co., Ltd., Beijing, China). The turbidity was determined by a HACH
 139 TL2310 turbidity meter (Hach, Loveland, CO, USA). The dissolved organic carbon (DOC) was
 140 determined with a TOC analyzer (TOC-L CPH, Shimadzu, Kyoto, Japan). The UV_{254}
 141 absorbance value was measured with a UV-vis spectrophotometer (Orion AquaMate 8000,
 142 Thermo Fisher Scientific Inc., MA, USA) at 254 nm wavelength. The concentration of total
 143 dissolved solids (TDS) and the electrical conductivity (EC) were determined using an
 144 Ultrameter II 6PFC portable multi-function apparatus (Myron L, Carlsbad, California, USA).
 145 The water quality characteristics of SGFPW samples are summarized in **Table 1**.

146 **Table 1.** Water quality characteristics of the SGFPW samples.

Constituents	SGFPW samples of Weiyuan shale gas field	
	This study	Previous literature (Chang et al., 2019c; Li et al., 2020; Shang et al., 2019; Tang et al., 2020a)
Turbidity (NTU)	35.9-42.7	32.5-215
pH	7.26-7.48	6.76-7.82
TDS (mg/L)	21,780-22,630	16,040-18,900
EC (mS/cm)	35.15-36.45	26.67-31.14
DOC (mg/L)	16.81-16.91	12.45-38.03
UV_{254} (cm^{-1})	0.162-0.173	0.057-0.165
DO (mg/L)	4.14-5.11	-

147 2.3 Membrane permeate flux and hydraulic resistance

148 The measurement and calculation methods of membrane permeate flux ($\text{L m}^{-2}\text{h}^{-1}$, LMH),
149 total fouling resistance (R_t) and its components, *i.e.*, membrane inherent resistance (R_m),
150 reversible resistance (R_{re}) and irreversible resistance (R_{ir}), were identical to our previous study
151 (Chang et al., 2019d; Li et al., 2020).

152 2.4 Analysis of the membrane fouling layers

153 The EPS measuring method and details about the measurement of contact angle can be
154 found in a recent study (Li et al., 2020). The surface of the fouled UF membrane samples were
155 observed and analyzed by scanning electron microscopy (SEM) (FE-SEM, Regulus-8230,
156 Hitachi, Japan) with energy dispersive spectroscopy (EDS) (X-MAX Extreme,
157 Oxford-Instruments, UK) at an acceleration voltage of 15 kV. Before microscopy, dried
158 membrane samples were sputter-coated with gold (MSP-2S, IXRF Systems Inc., USA).

159 2.5 Microbial diversity analysis

160 To explore the effects of aeration and GAC on microbial communities of GDM systems,
161 part of the hollow fiber membranes (about 8 cm^2) were collected after filtration and quickly
162 transferred to a sealed sterile tube. To prevent the decomposition of genetic material, the
163 membrane samples were frozen with liquid nitrogen and stored in a refrigerator
164 (906GP-ULTS, Thermo Scientific, USA) at -80°C . Details about DNA extraction, polymerase
165 chain reaction (PCR) amplification, and Illumina Miseq sequencing are presented in **Text S11**
166 of the Supporting Information and in our previous study (Chang et al., 2019c). Briefly, for
167 PCR amplification, the amplified primer sets of 18S rRNA genes for eukaryon and 16S rRNA

168 genes for bacteria were SSU0817/1196R and 338F/806R, respectively. UPARSE software
169 (version 7.1 <http://drive5.com/uparse/>) was utilized to analyze the cluster operational
170 taxonomic units (OTUs) with 97% similarity cutoff. The analysis of the alpha diversity, the beta
171 diversity, and microbial community composition were performed with the Majorbio I-Sanger
172 Cloud Platform (www.i-sanger.com).

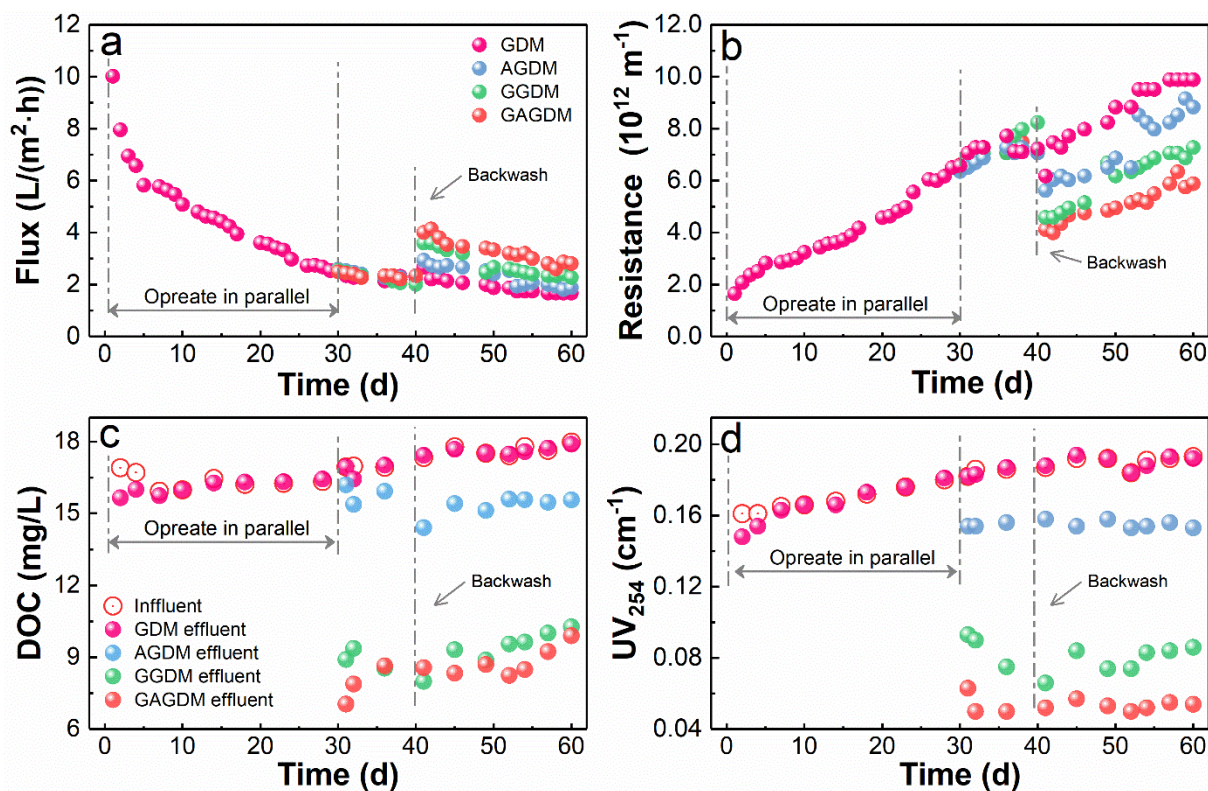
173

174 **Results and discussion**

175 *4.1 Permeate flux and fouling resistance*

176 The permeate flux profiles and the corresponding total fouling resistance of the four
177 GDM systems observed during the 60 days of filtration are presented in **Figs. 2a** and **2b**.
178 During the first 30 days, the systems were all equally run with no aeration and in the absence
179 of GAC. In this period, the permeate flux decreased from 10.0 LMH to 2.53 LMH. Between
180 the 30th to 40th day, the fluxes of all the systems decreased to values in the range 2-2.33
181 LMH at a slow rate, suggesting that GAC and aeration had negligible effect on flux decline.
182 The GDM systems were backwashed on the 40th day to also analyze the effect of
183 backwashing: to this purpose, some of the permeate was used as backwashing solution for ten
184 minutes with 5 LMH back flux. Backwashing allowed recovery of a portion of permeate flux
185 in all the systems. In particular, the flux of the control GDM system recovered only slightly
186 from 2.28 LMH to 2.67 LMH (17% increase). The fluxes of AGDM, GGDM, and GAGDM
187 units increased 26%, 80%, and 72%, respectively. This result suggests that the integrated
188 features, especially the presence of GAC, improved the reversibility of membrane fouling.

189 After backwashing, normal operation was resumed for 20 more days and, at the end of the
 190 experiment, the fluxes of control GDM, AGDM, GGDM, and GAGDM systems were stable at
 191 values of 1.67, 1.87, 2.27, and 2.80 LMH, respectively. In this study, the stable fluxes were
 192 higher than those reported in our previous articles (Chang et al., 2019c; Li et al., 2020).



193
 194 **Fig. 2** The variation of (a) permeate flux, (b) total fouling resistance, (c) DOC of influent and
 195 effluent streams, and (d) UV₂₅₄ of influent and effluent stream during the 60 days of filtration
 196 of the four GDM systems.

197
 198 According to the flux results, the total fouling resistance of the filtration systems
 199 increased from about $1.64 \times 10^{12} \text{ m}^{-1}$ to about $6.51 \times 10^{12} \text{ m}^{-1}$ during the first 30 days of
 200 operation. Addition of GAC and of aeration diversified the evolution trends of total fouling
 201 resistance for the varioussystem. At the end of the experiment, the resistances in GDM,

202 AGDM, GGDM, and GAGDM units were 9.89×10^{12} , 8.83×10^{12} , 7.27×10^{12} , and 5.89×10^{12}
203 m^{-1} , respectively. Overall, the performance was enhanced by aeration and was significantly
204 improved by addition of GAC, for reasons that are discussed in the following sections.

205 *4.2 Organic matter removal performance*

206 **Figs. 2c** and **2d** present the removal efficiency of the filtration systems for DOC and
207 UV_{254} during the 60 days of testing. At the beginning of the experiment, the DOC of the raw
208 water was 16.9 mg/L, and the DOC in the effluent of control GDM system was 15.7 mg/L.
209 Thus, the DOC removal rate was 7.5%. As the unit consists of a dead-end filtration reactor,
210 the DOC of the feed solution was constantly increasing (Li et al., 2020; Wu et al., 2019). In
211 the course of the filtration test, the DOC removal rate of the control GDM system was always
212 negligible or even negative, because of the poor rejection combined with the effect of the
213 biological layer on the membrane: macromolecular organic matter was likely degraded into
214 smaller molecules by the biofilm, and passed more easily through the membrane pores
215 compared to the starting material in the influent water (Akhondi et al., 2015; Derlon et al.,
216 2016; Derlon et al., 2014; Peter-Varbanets et al., 2011; Tang et al., 2018c; Wu et al., 2019). A
217 biofilm already formed on the membrane after short-term operation.

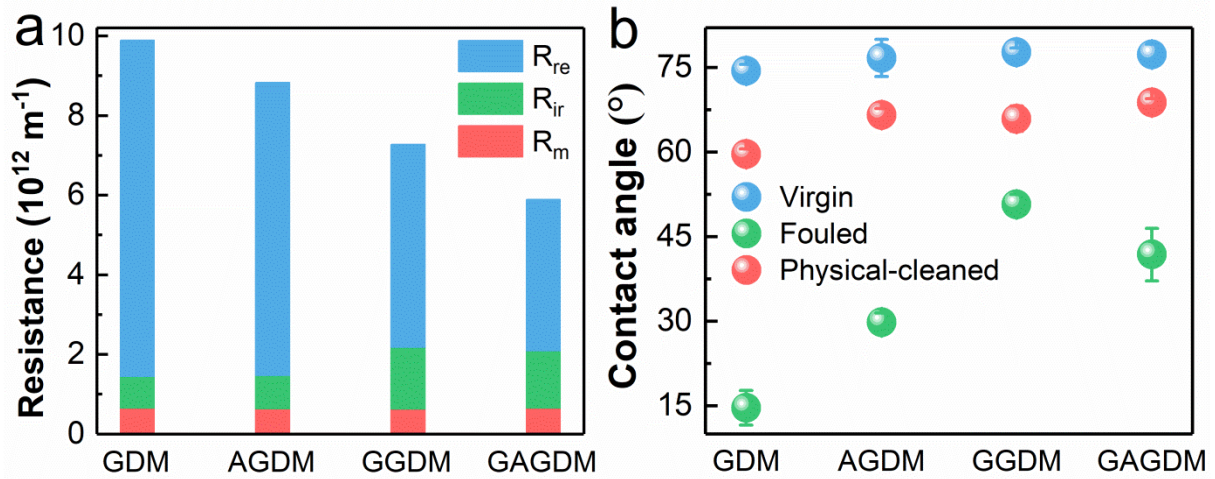
218 Upon addition of GAC and aeration, the DOC removal rate in AGDM, GGDM, and
219 GAGDM units was 4.4%, 47.4%, and 58.4%, respectively, and stable during filtration. This
220 sequence results in a DOC of the effluent equal to 17.9, 15.6, 10.3, and 9.90 mg/L, at the end
221 of the tests. These observations indicate that aeration promoted DOC removal, probably
222 because of the enhancement of microbial activity and the increase of the biomass

223 concentration in the two aerated reactors (Ding et al., 2017a; Lee et al., 2019a). Also, aeration
224 seemed to improve the ability of GAC to adsorb organic matter, as already observed by
225 previous studies (Karanfil et al., 1996; Lee et al., 2019a). For the two GDM systems provided
226 with GAC, adsorption on activated carbon was the main reason for the high DOC removal
227 rate in the initial period upon GAC addition; subsequently, the mechanism of DOC removal
228 changed to slower bioadsorption and biodegradation (Lee et al., 2019b), as suggested also by
229 previous investigations (Riley et al., 2016; Xing et al., 2008).

230 The UV_{254} removal efficiency was analogous to that of DOC. At the end of the filtration
231 period, the UV_{254} of the effluents from control GDM, AGDM, GGDM, and GAGDM units
232 was 0.192, 0.153, 0.086, and 0.054 cm^{-1} , respectively. Notably, aeration and especially GAC
233 addition significantly improved the effluent water quality with great potential benefits for the
234 subsequent desalination processes (Lee et al., 2019b).

235 *4.3 Fouling reversibility and surface characteristics of membrane fouling layers*

236 In order to investigate the effects of aeration and GAC on the recoverability of
237 membrane fouling, we measured the pure water flux and pure water contact angle relative to
238 the virgin membrane, the fouled membrane, and the fouled membrane after physical cleaning
239 (Li et al., 2020); see the results summarized in **Fig. 3**.



240

241 **Fig. 3** (a) Composition of membrane fouling resistance and (b) variation of water contact

242 angle on membranes from GDM systems with different operation conditions.

243

244 As shown in **Fig. 3a**, the total fouling resistance (R_t), reversible resistance (R_{re}) and

245 irreversible resistance (R_{ir}) of the control GDM system was $9.89 \times 10^{12} \text{ m}^{-1}$, $8.45 \times 10^{12} \text{ m}^{-1}$,

246 and $0.79 \times 10^{12} \text{ m}^{-1}$, respectively. R_{re} and R_{ir} accounted for 85.5% and 7.9% of R_t , in that order.

247 All the systems were characterized by a high reversibility resistance ratio (Chang et al., 2019c;

248 Ding et al., 2018b; Lee et al., 2019a). Compared with the R_t of the control system, the R_t of

249 the other filtration units was lower, with a reduction of 10.7%, 26.5% and 40.4%, respectively,

250 in AGDM, GGDM, and GAGDM systems. The trend of R_{re} was similar to that of R_t , whereas

251 R_{ir} increased for the systems in the presence of GAC, almost doubling in the GGDM unit.

252 This phenomenon suggests that GAC significantly reduced the R_{re} , which accounted for a

253 large proportion of the R_t , while increasing the R_{ir} , with aeration able to partly thwart this

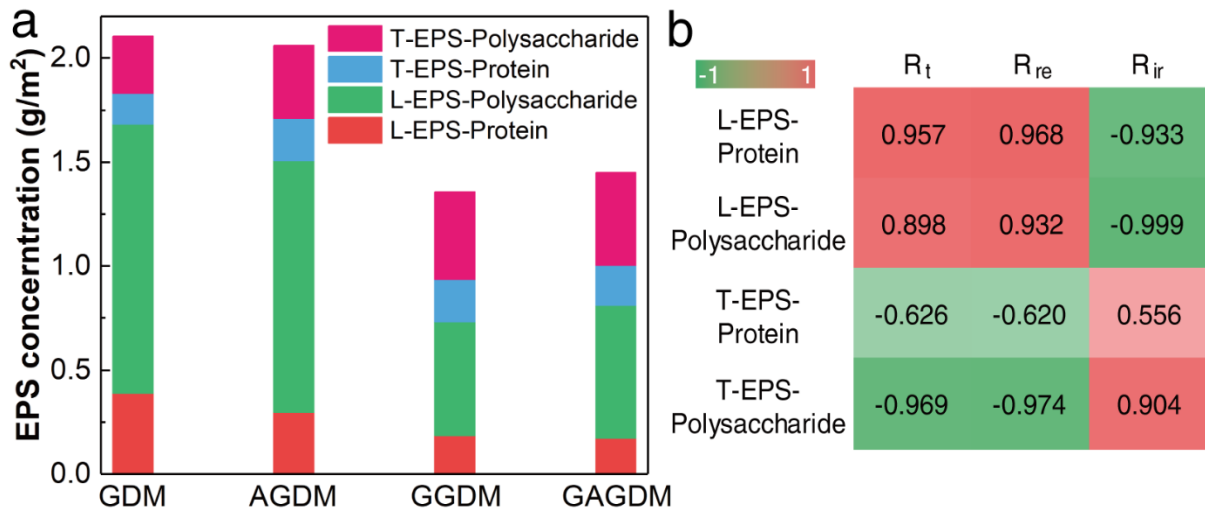
254 effect. This phenomenon was not observed in previous research (Lee et al., 2019a; Tang et al.,

255 2018a; Tang et al., 2018c).

256 As shown in **Fig. 3b**, the water contact angles for all fouled membranes were markedly
257 reduced, indicating the presence of hydrophilic foulants (Chang et al., 2019c). After physical
258 cleaning, the contact angles recovered to values equivalent to roughly 80% of those measured
259 with virgin membranes, which were in the range 75-77°. Similar to conclusions suggested by
260 previous studies (Li et al., 2020; Wang et al., 2017), the pure water contact angle of the fouled
261 membranes could be restored by physical cleaning to a level slightly lower than the original
262 membrane, showing that the membrane fouling of GDM systems had high recoverability. The
263 surface topographies of membrane fouling layers were observed with SEM and presented in
264 **Fig. S1**. Compared to the sample employed in the control GDM unit, relatively loose and
265 heterogeneous biofilm structures were found in the other three systems, in addition to many
266 pores and cracks of different size.

267 *4.4 Analysis of EPS on the fouling layer*

268 The concentration of EPS on the membrane surface was measured at the end of the
269 filtration tests. EPS was divided into loosely bound EPS (L-EPS) and tightly bound EPS
270 (T-EPS), according to the different extraction methods (Li et al., 2020). The concentration of
271 polysaccharide and protein was also measured, and all the results are presented in **Fig. 4**. The
272 concentration of L-EPS and T-EPS in the membrane fouling layer of control GDM system
273 was 1.68 and 0.42 g/m², respectively. The L-EPS consisted mostly of polysaccharides, while
274 the fraction of protein represented about one third of the T-EPS.



275

276 **Fig. 4 (a)** The component of EPS accumulated on the membrane per unit area and **(b)** clustering

277 correlation analysis between EPS and membrane fouling resistance.

278

279 The effect of aeration on the concentration of L-EPS was not significant. However, it

280 seemed to lead to a certain increase in T-EPS concentration in the fouling layer, which might

281 be due to the shear force produced by aeration increasing the density of the layer (Pronk et al.,

282 2019). The concentrations of polysaccharide and protein in L-EPS is positively correlated

283 with R_t and R_{re} , as shown in **Fig. 4b**. GAC significantly reduced L-EPS concentration in the

284 membrane fouling layer, which might be the reason for the decrease of total fouling resistance

285 and of the reversible resistance. Similarly, some studies proposed that the reduction of EPS in

286 the biofouling layer is one of the main reasons for the increased stable flux in GDM (Tang et

287 al., 2018b; Tang et al., 2018c). In addition, the concentrations of polysaccharide in T-EPS is

288 positively correlated with R_{ir} . In fact, GAC increased the concentration of polysaccharide in

289 T-EPS, which might be the reason for the increase in the irreversible resistance in systems

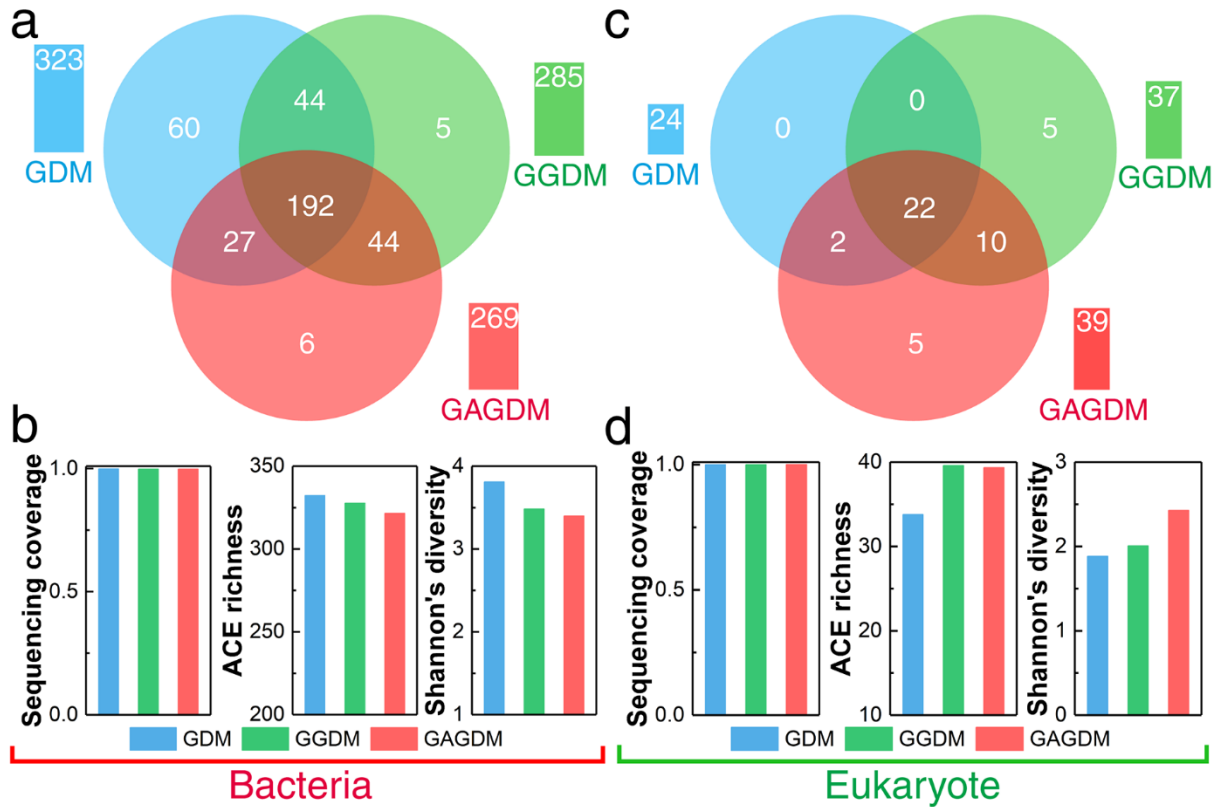
290 comprising activated carbon (Fig. 3a).

291 *4.5 Microbial diversity analysis*

292 The Venn diagram of bacterial and eukaryotic communities at OTU level and the alpha
293 diversity of bacterial and eukaryotic communities are shown in **Fig. 5**. Overall, the diversity
294 and richness of microorganisms in this work was higher than in samples analyzed in our
295 previous research (Chang et al., 2019c), and than in other samples from the Sichuan basin
296 (Zhang et al., 2017). However, it was far lower than that of waste sludge, soil, or wastewater
297 samples, due to the harsh water quality characteristics of SGFPW (Wang et al., 2019). The
298 coverage values of all samples were higher than 99.9% (**Figs. 5b** and **5d**), indicating that the
299 sequencing depth was sufficient to cover most of bacteria and eukaryotes. The rarefaction
300 curves (**Fig. S2**) also suggest that the sequencing depth was adequate.

301 In terms of bacterial communities, a total of 378 OTUs were found across all samples
302 with a range of 269-323 OTUs present in each sample. The highest OTU number was found
303 for the biolayer evolved in the control GDM system. The number of OTUs from GGDM and
304 GAGDM units was close and slightly lower than that of the control system, indicating that the
305 addition of GAC and aeration only affected a small portion of the bacteria in the fouling layer.
306 The decrease in ACE index and Shannon index also indicated that GAC and aeration slightly
307 reduced the richness and diversity of the bacterial community.

308



309

310 **Fig. 5** Venn diagram of bacterial (a) and eukaryotic (c) communities and the alpha diversity of
 311 bacterial (b) and eukaryotic (d) communities.

312

313 The richness and diversity of eukaryotic communities were much smaller than those
 314 relative to bacteria, but opposite trends were observed upon combination of the GDM system
 315 with GAC and aeration. A total of 44 OTUs were detected in the eukaryotic community of
 316 biolayer from three GDM systems. The control group did not have a unique OTU, while a large
 317 number of new OTUs were detected in the biofilm from the GGDM and GAGDM units. The
 318 variation of ACE index and Shannon index indicated that addition of GAC enriched the
 319 richness and diversity of the eukaryotic community in the membrane fouling layer, while
 320 aeration had only a small further enhancing effect.

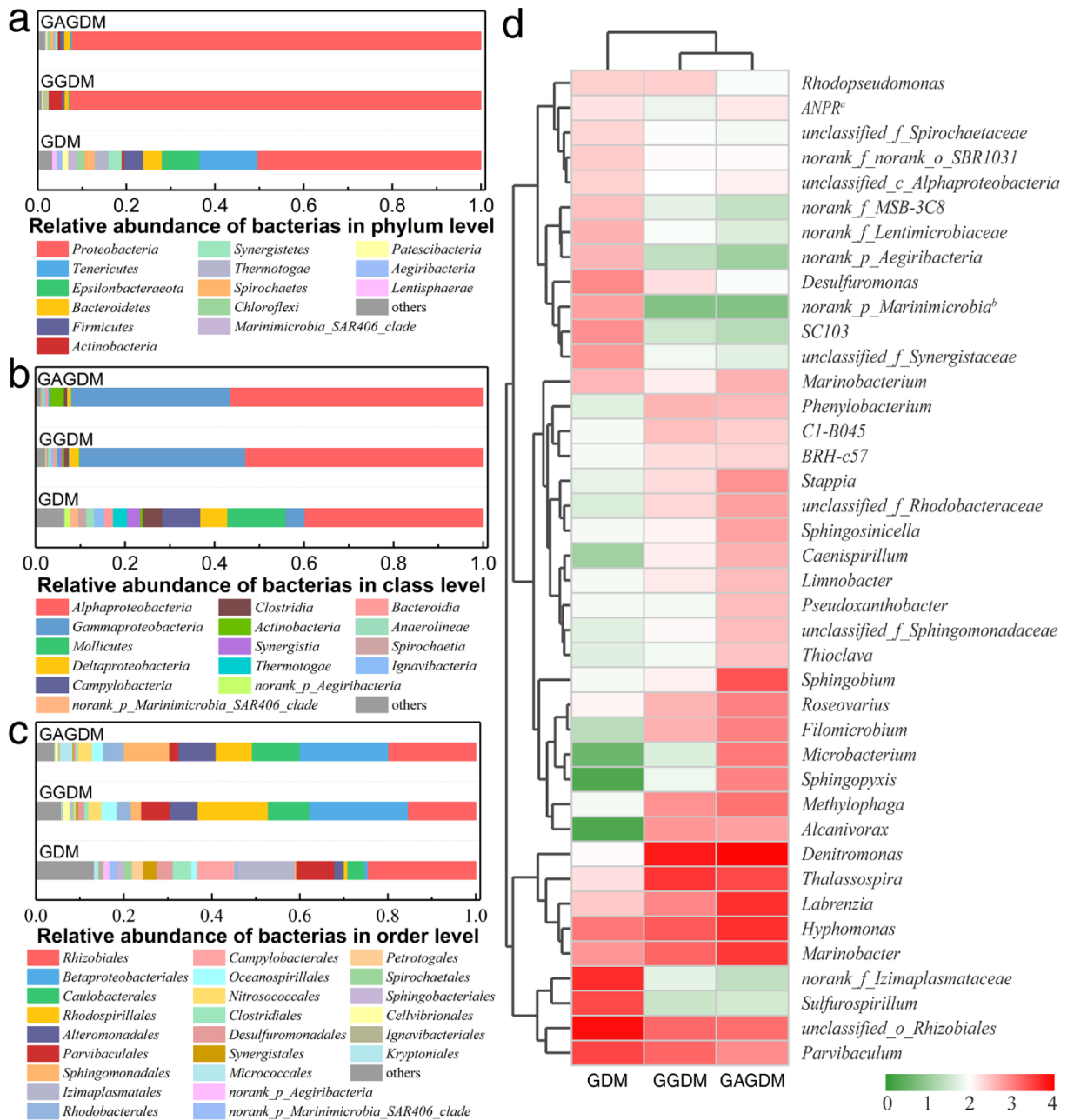
321 *4.5.1 Bacterial community of the biofouling layer in GDM systems*

322 There were 30 kinds of bacterial phyla in the membrane biofouling layer of three GDM
323 systems; see **Fig. 6a**. *Proteobacteria* (50.3%), *Tenericutes* (13.1%), *Epsilonbacteraeota* (8.6%),
324 *Bacteroidetes* (4.2%), and *Firmicutes* (4.3%) were the major phyla and constituted 80.5% of the
325 bacteria in the control GDM system. Some halotolerant and halophilic bacteria existed in phyla
326 *Proteobacteria* and *Bacteroidetes* (Frank et al., 2017; Zhang et al., 2017). Consistent with what
327 discussed above, GAC and combined GAC with aeration significantly decreased the diversity
328 of the bacterial community and changed the community structure at the phylum level. In
329 particular, the vast majority of the phyla was represented by *Proteobacteria* in samples from
330 GGDM and GAGDM units (> 90%). The existence of *Proteobacteria* is common to wastewater
331 because of their ability to decompose carbohydrates (Frank et al., 2017; Song et al., 2020b).

332 *Alphaproteobacteria* (39.8%), *Mollicutes* (13.1%), *Campylobacteria* (8.5%),
333 *Deltaproteobacteria* (6.1%), *Clostridia* (4.3%) and *Gammaproteobacteria* (4.2%) were the
334 main classes found in the biofilm from the control GDM system. In GGDM and GAGDM
335 samples, *Alphaproteobacteria* and *Gammaproteobacteria* affiliated to *Proteobacteria*
336 increased, especially *Gammaproteobacteria*, as shown in **Fig. 6b**. According to reports,
337 halophilic bacteria of these two classes can effectively degrade polycyclic aromatic
338 hydrocarbons in polluted seawater (Arulazhagan and Vasudevan, 2009). The
339 *Gammaproteobacteria* and *Alphaproteobacteria* might also play an important role in high DOC
340 removal. In the biolayer evolved in the GAGDM system, the relative abundance of
341 *Deltaproteobacteria* was obviously reduced to 0.8%: this result is not surprising because the
342 dominant organisms of this class have anaerobic metabolism function, including

343 sulfate-reducing bacteria and geobacter sub-phylum (Freedman et al., 2017).

344 At the genus level, 231 bacteria genera were detected in the samples from the three systems.
345 The core genera from the control GDM unit were *unclassified_o_Rhizobiales* (21.7%),
346 *norank_f_Izimaplasmataceae* (12.9%), *Parvibaculum* (8.6%), *Sulfurospirillum* (7.8%),
347 *Hyphomonas* (3.7%), and *Desulfuromonas* (2.8%). GAC and combined GAC with aeration
348 significantly changed the core genera, which for GGDM and GAGDM samples were
349 *Denitromonas* (18.3-21.5%), *Thalassospira* (6.2-14.0%), *Labrenzia* (3.5-9.6%), *Marinobacter*
350 (6.1-8.1%), *Hyphomonas* (7.6-9.7%), *Parvibaculum* (2.0-6.2%), *unclassified_o_Rhizobiales*
351 (3.3-5.9%), and *Roseovarius* (1.6-2.3%), significantly different with those extracted from the
352 membrane used in the control GDM system.

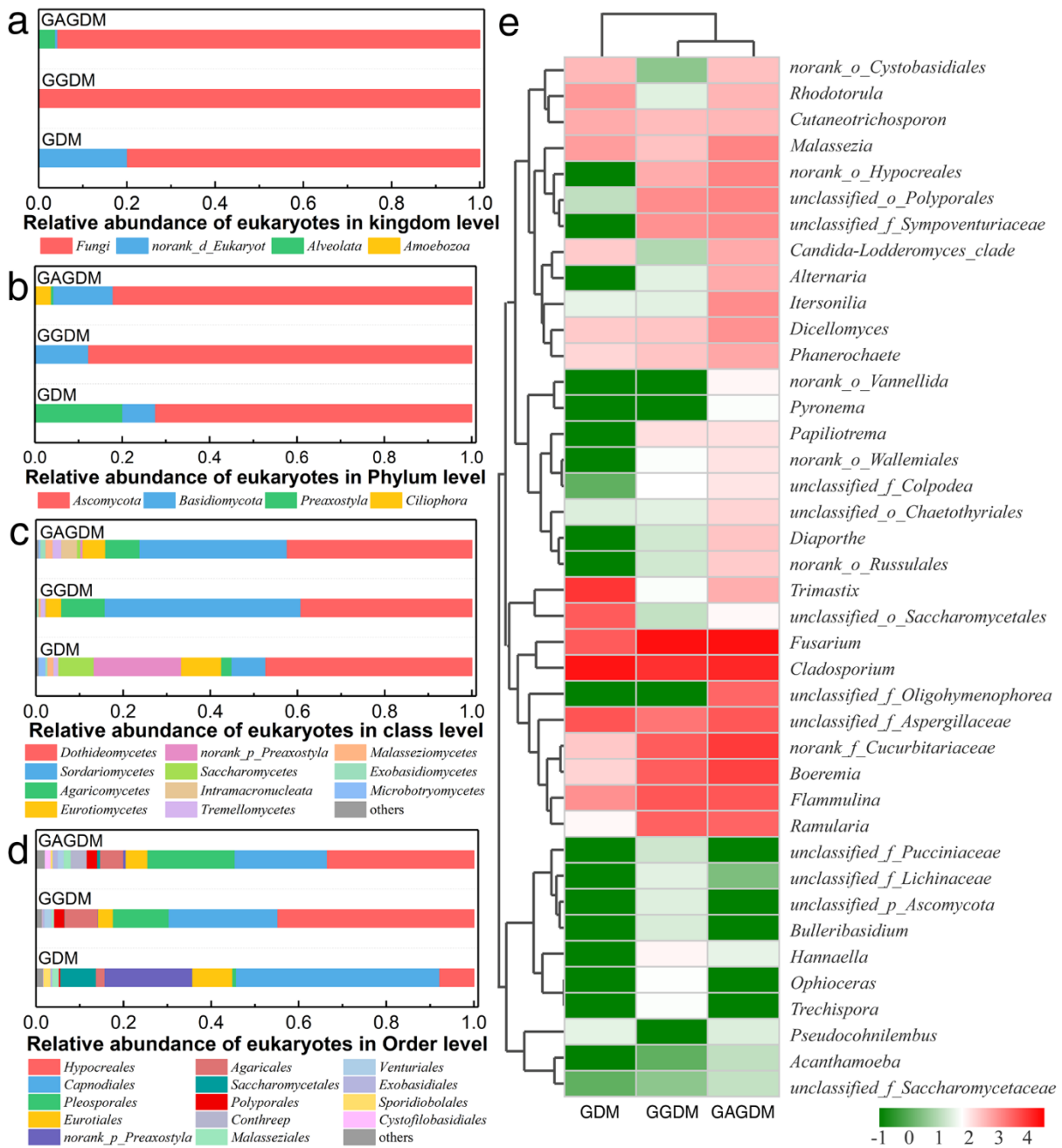


353
 354 **Fig. 6** Bacterial community compositions at (a) the phylum (>0.5%), (b) the class (>1%), (c)
 355 the order (>1%), and (d) the genus level (top 40). In (d), ANPR^a and norank_p_Marinimicrobia^b
 356 represent Allorhizobium-Neorhizobium-Pararhizobium-Rhizobium and
 357 norank_p_Marinimicrobia_SAR406_clade, respectively.

358
 359 4.5.2 Eukaryotic community of the biofouling layer in GDM systems

360 **Fig. 7a** shows that the eukaryotic kingdom of the biofilm from the control GDM system
361 consisted of *Fungi* (79.9%), *Excavata* (20.0%), and *Alveolata* (0.1%), which was a different
362 classification compared to our previous study (Chang et al., 2019c). The fraction of *Fungi*
363 increased significantly to 95.7-99.8% in GGDM and GAGDM samples, representing the
364 vastly predominant eukaryotic kingdom. At the phylum level, *Ascomycota* (72.4-87.7%) and
365 *Basidiomycota* (7.5-16.5%) were dominant in all systems, and a similar observation was
366 reported in other SGFPW samples from the Sichuan Basin (Zhang et al., 2017). The number
367 of classes observed in GDM, GGDM, and GAGDM samples was 7, 17, and 16, respectively.
368 Finally, as shown in **Fig. 7e**, the core genera of the biofilm evolved in the control GDM system
369 were *Cladosporium* (46.3%), *Trimastix* (20.0%), *unclassified_f_Aspergillaceae* (9.2%),
370 *Fusarium* (7.8%) and *unclassified_o_Saccharomycetales* (7.7%). The *Cladosporium* fungi can
371 produce extracellular hydrolytic enzymes, like monoacyl esterase, protease, and pectinolytic
372 enzymes (Barbosa et al., 2001). Addition of GAC and combined GAC with aeration changed
373 the environment significantly, thus modifying the core genera. Specifically, the fouling layer
374 environment of the latter unit was beneficial to the growth of *Fusarium* (31.8-44.0%),
375 *norank_f_Cucurbitariaceae* (6.5-10.4%), and *Boeremia* (6.2-8.9%). In contrast, it was
376 detrimental to the growth of *Cladosporium* (17.4-19.2%), *Trimastix* (0.1-0.5%),
377 *unclassified_f_Aspergillaceae* (3.4-5.0%) and *unclassified_o_Saccharomycetales* (0.02-0.1%).
378 Overall, the clear increase in abundance and diversity of eukaryotes by GAC and aeration
379 should be accompanied by more active predation behavior, resulting in a more porous and
380 heterogeneous membrane biofouling layer, thereby increasing the stable flux of the filtration

381 system (Chang et al., 2019c; Pronk et al., 2019; Tang et al., 2018a; Tang et al., 2018c).



382
 383 **Fig. 7** Eukaryotic community compositions at (a) the kingdom, (b) the phylum
 384 (>1%), (d) the order (>1%), and (e) the genus level.

385 **Conclusions**

386 To solve the issue about low stable flux and low organic removal of gravity driven

387 membrane filtration, solar aeration and in-situ GAC adsorption were combined with GDM to
388 pretreat SGFPW in Weiyuan. The performance of the integrated processes and the
389 characteristics of the membrane fouling layers obtained under different conditions were
390 evaluated and the following conclusions can be drawn:

391 (1) GAC and aeration, especially GAC, markedly enhanced the stable flux and reduced the
392 total fouling resistance of GDM systems. GAC significantly reduced R_{re} , which accounted for
393 a large proportion of R_t and it also slightly increased the R_{ir} of GDM systems. Aeration can
394 further reduce R_t of the GDM system when combined with GAC.

395 (2) Compared to traditional units, the system comprising both GAC adsorption and aeration
396 showed high DOC removal rate due to bioabsorption and biodegradation. GAC significantly
397 reduced the concentration of EPS in the membrane biofouling layer and this effect is one of
398 the main reasons for the increased stable flux of the integrated system.

399 (3) GAC and aeration observably changed the the microbial community structure. The
400 dominant *Gammaproteobacteria* (35.5%) and *Alphaproteobacteria* (56.5%) classes evolved in
401 the GAGDM integrated system played an important role in high DOC removal. In this unit, the
402 eukaryotic community richness and diversity significantly increased in the biofouling layer.
403 This is accompanied with more active predation behavior, resulting in a more porous and
404 heterogeneous membrane biofouling layer, thus translating into a higher system productivity.

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410 **Appendix A. Supplementary data**

411 The Supporting Information to this article is available at online. Detailed experimental
412 procedures and additional experimental data: DNA extraction, polymerase chain reaction
413 (PCR) amplification, and Illumina Miseq sequencing; SEM images and EDS analyses of
414 membrane fouling layers in different GDM systems; Rarefaction curves of OTUs for bacteria
415 and eukaryote in biofouling layers of three GDM systems.

416

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