

1 ***Moringa oleifera* coagulation as pretreatment prior to microfiltration for**  
2 **membrane fouling mitigation**

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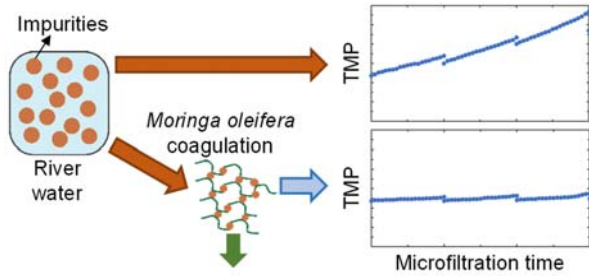
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15 **TOC contents**



16

17 Coagulation pre-treatment using *Moringa oleifera* in microfiltration of a river water achieved

18 membrane fouling mitigation and filtered water quality improvement.

19 **Abstract**

20 Coagulation using *Moringa oleifera* (MO), a natural coagulant, is an attractive approach in  
21 drinking water treatment to break away from conventional chemical coagulation using  
22 aluminium or iron salts. This study aimed to evaluate the effect of MO coagulation pretreatment  
23 on the fouling mitigation in microfiltration (MF) process. MF treatment of a river water without  
24 pretreatment promoted a considerable reduction in membrane permeability (i.e. membrane  
25 fouling), which was not sufficiently recovered by conventional backwashing. In contrast, MF  
26 treatment after MO coagulation substantially reduced membrane fouling. Over six filtration  
27 cycles (or 6 h filtration period), the fouling mitigation level with MO coagulation was  
28 comparable to that with aluminium sulphate (alum) coagulation. In addition, major water quality  
29 (turbidity and colour) after MF treatment was equivalent between MO and alum coagulation.  
30 Pretreatment by MO coagulation has an advantage of maintaining solution pH after MF  
31 treatment. The results obtained here suggest that MO coagulation can be employed as an  
32 effective and low-cost fouling mitigation technique for MF process in drinking water treatment.

## 33 **1. Introduction**

34 There is a growing interest in *Moringa oleifera* (MO) seed as an alternative to chemical  
35 coagulants such as aluminium sulphate (alum) and ferric chloride in recent years.<sup>1, 2</sup> MO seeds  
36 contain dimeric cationic proteins,<sup>3</sup> which can be extracted from MO seeds using water or salt  
37 solution and used for coagulation applications.<sup>4</sup> MO coagulation is based on the net-like structure  
38 coagulation mechanism, that is effective for turbidity removal.<sup>4</sup> For example, previous studies<sup>5-7</sup>  
39 demonstrated that MO coagulation achieved from 60% to almost complete removal of turbidity  
40 for high turbid surface waters with 50–450 NTU. MO is not toxic; thus, even when coagulation  
41 is not correctly performed causing overdosing, no adverse effects on public health are expected  
42 in case of overdosing. In addition, given its organic nature, coagulant sludge generated through  
43 MO coagulation can be processed for use as animal feed or plant fertilizer.<sup>8</sup> MO is a fast growing  
44 tree, commonly found in semiarid, tropical and sub-tropical areas including India, South and  
45 Central America, Africa and South East Asia.<sup>9</sup> Thus, MO coagulation is a pragmatic option to  
46 provide access to safe drinking water to rural communities in developing countries around the  
47 world as stated in the Millennium Development Goals by the United Nation.

48 Gravity driven membrane filtration is another technology that has significantly improved access  
49 to safe drinking water.<sup>10</sup> Notable examples include the Skyjuice<sup>11</sup> and Aqua CUBE systems, in  
50 which water is driven through a microfiltration (MF) by gravity. MF has been widely used for  
51 drinking water treatment to reliably remove suspended solids and pathogens such as protozoa  
52 and pathogenic bacteria.<sup>12, 13</sup> A major challenge in using MF for low cost drinking decentralised  
53 water treatment is membrane fouling, which is caused by the deposition of colloidal and organic  
54 substances on the membrane surface or in the membrane pores and can reduce membrane

55 permeability and separation performance.<sup>14-16</sup> Membrane fouling in MF can be controlled by  
56 frequent backwashing or chemical cleaning. However, these methods are not compatible with  
57 decentralised water treatment. A more suitable approach is to apply pre-coagulation prior to MF  
58 treatment.<sup>17</sup>

59 The effectiveness of coagulation pretreatment with alum or ferric chloride to mitigate MF  
60 membrane fouling has been demonstrated in the literature.<sup>18, 19</sup> The reduced membrane fouling  
61 can be attributed to coagulation process capable of removing suspended solids and dissolved  
62 organic matter.<sup>20</sup> Although alum and ferric coagulation have been successfully used as a  
63 membrane fouling mitigation technique in drinking water treatment, the use of these heavy metal  
64 coagulants may be inappropriate in some developing countries due to the unaffordable high costs  
65 of imported chemicals<sup>5</sup> and MO coagulation can be an alternative pretreatment. In fact, several  
66 studies have successfully applied MO coagulation to pretreat dairy wastewater<sup>21, 22</sup> and  
67 secondary oxidation pond effluent<sup>23</sup> prior to MF operation. MO coagulation pretreatment can  
68 possibly substitute backwashing to reduce the complexity of MF operation in drinking water  
69 applications, making it more amendable to small rural communities.

70 This study aimed to evaluate the effects of MO coagulation on the fouling mitigation of MF  
71 treatment in drinking water applications. MO dose in a surface water with low turbidity was  
72 optimised for turbidity removal and fouling mitigation. Thereafter, MO coagulation on the  
73 fouling mitigation of MF treatment was evaluated over six filtration cycles, and the results were  
74 compared with that of alum coagulation. This study also examined water quality after  
75 coagulation followed by MF treatment.

## 76 **2. Materials and methods**

### 77 **2.1 Chemicals**

78 Analytical grade alum ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ ) and sodium chloride (NaCl) were purchased from  
79 Wako Pure Chemical Industries (Tokyo, Japan). A stock solution of alum coagulant was  
80 prepared in Milli-Q water to obtain 1000 mg-Al/L. A NaCl solution of 1 M was also prepared in  
81 Milli-Q water and used for MO coagulant extraction. Tap water was treated using a reverse  
82 osmosis system and used for backwashing water. River from Nagasaki, Japan was used to  
83 present surface water. The river water collected in December was stored at 4 °C and was used  
84 within one week.

### 85 **2.2 MO coagulant**

86 MO seeds were collected from Tamil Nadu, India. The MO seeds were first ground into powder.  
87 Then 1 g of MO powder was dissolved with 100 mL Milli-Q water at 30 °C. Thereafter, the  
88 obtained MO solution was stirred for 5 min and allowed to settle for 10 min. The supernatant of  
89 MO solution was filtered by a 0.45  $\mu\text{m}$  filter (Thermo Scientific, Waltham, NA, USA) and the  
90 filtrate was discarded. Solid residue on the filter was washed by 100 mL of 1 M NaCl. The  
91 obtained solution was filtered again via the same protocol described above (i.e. stirred for 5 min,  
92 settled for 10 min, and filtered to obtain 100 mL of clear filtrate). The final filtrate was used as  
93 MO coagulant for subsequent experiments.

### 94 **2.3 Membrane module and filtration system**

95 A hollow fibre polyvinylidene fluoride (PVDF) MF membrane with nominal pore size of 0.1  $\mu\text{m}$   
96 (USP-043, Asahi Kasei, Tokyo, Japan) was used in this study. The MF membrane was supplied

97 in a module with length of 132 mm, external diameter of 1.4 mm, internal diameter of 0.7 mm  
98 and effective membrane area of 120 cm<sup>2</sup>. The membrane module is designed to operate in the  
99 inside-out filtration orientation. The module is integrated into a laboratory-scale dead-end  
100 filtration system comprising 2 L glass feed reservoir, flow meter, a peristaltic pump for filtration  
101 (Kros Flo<sup>®</sup> Research pump, Spectrum Laboratories, CA, USA), a peristaltic pump for  
102 backwashing (Q-100, Tacmina, Osaka, Japan), a temperature control unit (ACE-2000, Tokyo  
103 Rikakikai, Tokyo, Japan), valves and pressure gauges (**Fig. S1**).

## 104 **2.4 Experimental protocols**

### 105 **2.4.1 Coagulation experiment**

106 Coagulation experiments were performed using a jar test system (JMD-3E, Miyamoto Riken  
107 Industry, Osaka, Japan). This jar test system was equipped with a series of 1 L beakers and  
108 stirring shafts. The stirring speed of these shafts can be changed automatically. Each beaker was  
109 filled with 1 L of river water. Immediately after coagulant addition, the beakers were rapidly  
110 mixed for 2 min at 150 rpm. The coagulant dose was from 0 to 8 mL-MO/L and 0 to 10 mg-Al/L  
111 for MO and alum, respectively. After rapid mixing, the solution was flocculated for 30 min at a  
112 mixing speed of 30 rpm. Then, the beakers were left undisturbed for sedimentation for 1 h. The  
113 supernatant was then collected for subsequent filtration experiments and water quality analysis.

### 114 **2.4.2 MF treatment**

115 Prior to each MF filtration experiment, integrity of the membrane module was confirmed by  
116 measuring the clean water permeability at 20 °C. Thereafter, the feed reservoir was filled with a  
117 river water (a) without pre-treatment, (b) after MO coagulation or (c) after alum coagulation.

118 Each filtration cycle comprised of two steps: filtration and backwashing. Filtration was  
119 conducted at the permeate flux of 140 L/m<sup>2</sup>h (at 20 °C) for 1 h. Membrane fouling was  
120 monitored by the transmembrane pressure (TMP). At the end of the filtration step, backwashing  
121 was applied using clean water at the flux of 280 L/m<sup>2</sup>h (at 20 °C) for 1 min. The next filtration  
122 cycle was then repeated as described above. After each experiment, chemical cleaning was used  
123 to restore the membrane permeability to within 1% the initial value. Chemical cleaning was  
124 conducted by soaking the membrane overnight in 1% sodium hydroxide and 400 ppm of sodium  
125 hypochlorite at room temperature. The membrane was then rinsed with a copious amount of  
126 clean water to remove any residual chemicals.

## 127 **2.5 Analytical techniques**

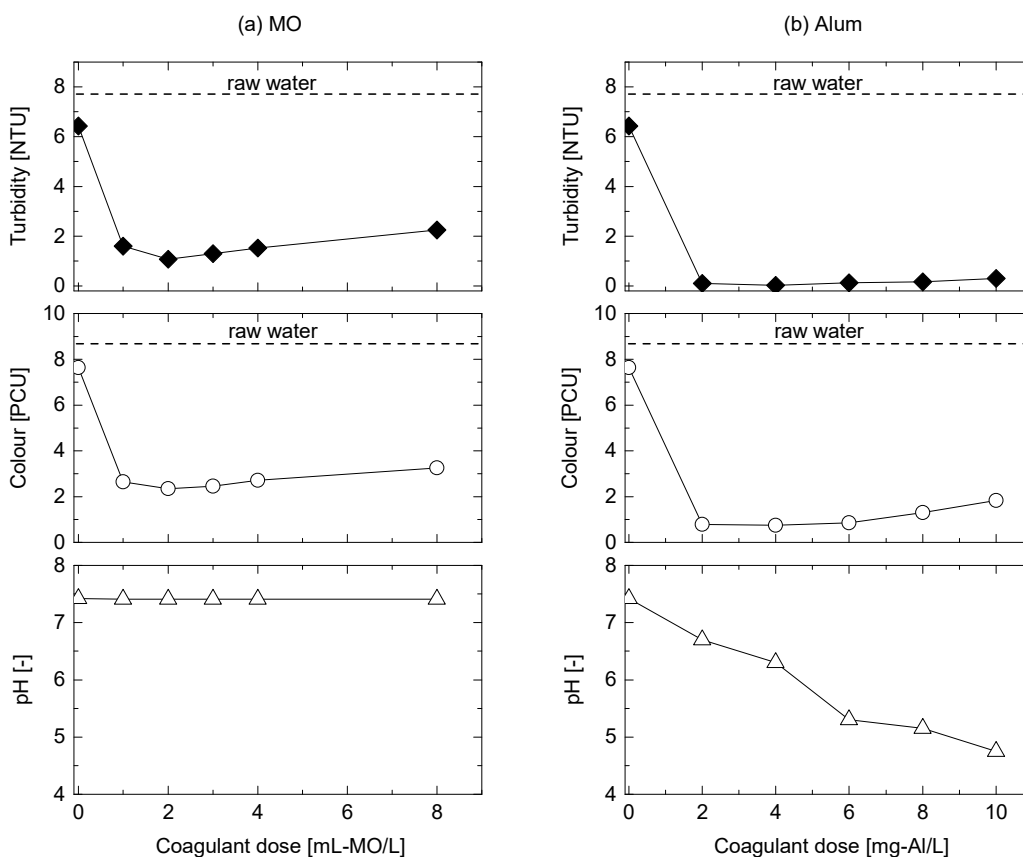
128 The water quality parameters analysed here include turbidity, colour, pH, electrical conductivity  
129 and total organic carbon (TOC). Turbidity and colour measurements were conducted using water  
130 analyser turbidity meter (WA1, Nippon Denshoku, Tokyo, Japan). The pH was measured using a  
131 pH meter (SK-620 PH II, Sato, Tokyo, Japan). Electrical conductivity was measured using a  
132 conductivity meter (AS 710, As One Corporation, Shanghai, China). TOC was measured using a  
133 total organic analyser (TOC-VCSD, Shimadzu, Kyoto, Japan).



### 134 3. Results and discussion

#### 135 3.1 Coagulation

136 Coagulant doses of MO and alum coagulants were optimised based on the removal of turbidity  
137 and colour. The optimum coagulant doses of MO and alum were at 2 mL-MO/L and 4 mg-Al/L,  
138 respectively (**Fig. 1**). MO coagulation reduced turbidity and colour from 7.8 to 1.1 NTU and  
139 from 8.7 to 2.4 PCU, respectively. Alum coagulation resulted in better turbidity and colour  
140 removal, achieving 0.1 NTU and 0.8 PCU after coagulation, respectively. The optimum turbidity  
141 removal by alum was 99.7%, which was considerably better than that by MO (86%). Similarly,  
142 alum coagulation resulted in 91% colour removal, which was higher than MO coagulation (72%)  
143 (**Fig. 1**). Lower turbidity and colour removal efficiencies by MO coagulant can be explained by  
144 their lighter and smaller flocs compared to those from alum. MO flocs had poor settleability as  
145 previously reported by Okuofu.<sup>24</sup> For both MO and alum, increasing the coagulant dose beyond  
146 the optimum value resulted in increased residual turbidity and colour. Over-dose of MO can  
147 cause the saturation of the polymer bridge sites and increased charge repulsion due to MO's  
148 cationic poly-electrolyte.<sup>7</sup> These phenomena can destabilize colloids and particles, inhibiting the  
149 removal of turbidity. Results in **Fig. 1** indicate that MO coagulant dose needs to be precisely  
150 regulated to optimise turbidity removal.



151  
 152 **Fig. 1** – Removal of turbidity and color, and change in pH in the surface water at various  
 153 coagulant doses of (a) MO and (b) alum.

154 In terms of other major water quality parameters, solution pH was not affected by MO  
 155 coagulation at 0–8 mL-MO/L dose (**Fig. 1**). In contrast, alum coagulation resulted in a  
 156 considerably drop in the solution pH from 7.4 (initial surface water) to 6.3 when the alum dose  
 157 increased to 4 mg-Al/L. On the other hand, a small but discernible increase in conductivity was  
 158 observed from 270 to 286  $\mu\text{S}/\text{cm}$  at the optimum MO dose (**Table 1**). The increase in  
 159 conductivity can be ascribed to the addition of NaCl used for extracting MO coagulation as  
 160 described in section 2.2. In addition to conductivity, MO coagulation resulted in a slight increase  
 161 in TOC concentration, from 15.3 to 15.7 mg/L at the optimum dose. An increase in the

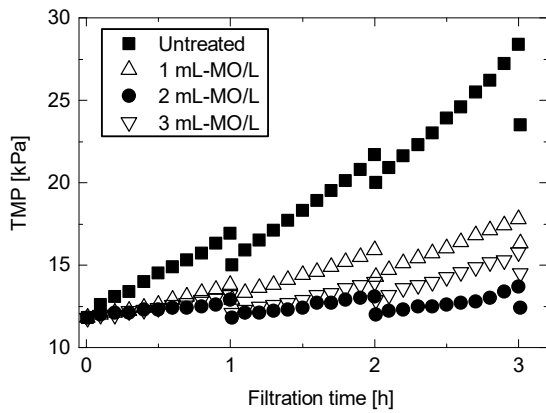
162 concentration of organic matter in treated water is one of the major drawbacks of MO.<sup>7, 8, 25</sup> In  
 163 contrast, alum coagulation could achieve about 8% TOC removal (**Table 1**). The reduction in  
 164 TOC concentration in alum coagulation is mainly due to adsorption and complexation with  
 165 positively charged aluminium ions under acidic conditions, forming insoluble particulate  
 166 aggregates.<sup>26</sup>

167 **Table 1** – Water quality before and after coagulation at their optimum doses (2 mL-MO/L and 4  
 168 mg-Al/L).

Parameter	Untreated water	MO-treated water	Alum-treated water
Turbidity (NTU)	7.8	1.1	0.1
Colour (PCU)	8.7	2.4	0.8
Conductivity ( $\mu\text{S}/\text{cm}$ )	270	286	351
TOC (mg/L)	15.3	15.7	14.0
pH	7.4	7.4	6.3

### 169 **3.2 Optimisation of coagulant dose for fouling mitigation**

170 Results from MO optimisation experiments were used to evaluate the performance of MO  
 171 coagulation pretreatment to mitigate MF membrane fouling. MF treatment of the river water  
 172 without pretreatment resulted in a rapid increase in TMP from 12 to 17 kPa within the first  
 173 filtration cycle (0–60 min) (**Fig. 2**). Moreover, backwashing with clean water was not capable of  
 174 restoring membrane permeability. As a result, after three filtration cycles TMP reached 24 kPa,  
 175 twice higher than the initial TMP.



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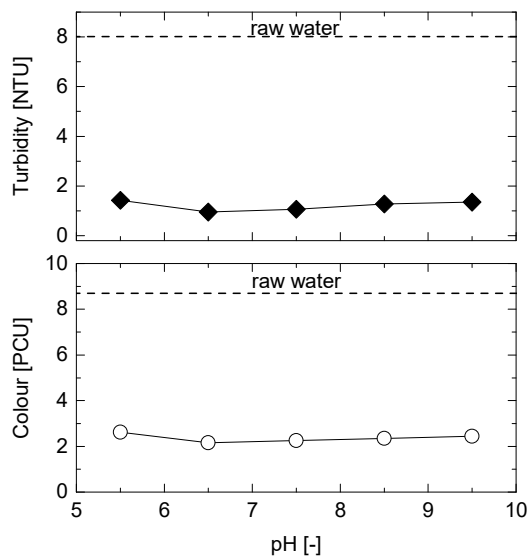
177 **Fig. 2** – TMP increase in MF treatment of waters treated with MO coagulant (permeate flux =  
 178 140 L/m<sup>2</sup>h, backwashing flux = 280 L/m<sup>2</sup>h and backwashing time = 1 min).

179 MO coagulation performed prior to MF effectively reduced membrane fouling at all MO doses  
 180 selected here (1, 2 and 3 mL-MO/L), which correspond to residual turbidity removals of 75%,  
 181 86% and 83%, respectively. The significant reduction in membrane fouling by MO coagulation  
 182 is due likely to the improved transformation and removal efficiency of colloids from water prior  
 183 to MF treatment. During the coagulation and sedimentation processes, aggregation of small  
 184 colloids and dissolved matter can lead to a larger and effective particle size, which can result in  
 185 less specific membrane resistance.<sup>27</sup> In addition, organic matter and colloidal particles  
 186 transformed into flocs can easily be removed by backwashing.<sup>28</sup> The results revealed that the  
 187 effect of fouling mitigation by MO coagulation can be maximized at 2 mL-MO/L dose, which  
 188 resulted in only an increased TMP from 12.0 to 12.4 kPa over three filtration cycles (**Fig. 2**). In  
 189 contrast, 3 mL-MO/L appeared to be an overdose, resulting in a discernible increase in TMP  
 190 from 12 to 15 kPa in three filtration cycles. Likewise, a lower dose at 1 mL-MO/L was not  
 191 sufficient to effectively mitigate membrane fouling. The results here indicate that the MO dose  
 192 optimised for turbidity removal can be an important control parameter for membrane fouling

193 mitigation in MF process. Therefore, monitoring turbidity in MF feed (i.e. coagulation effluent)  
194 can be an operating parameter to maintain the minimum level of membrane fouling.

### 195 **3.3 Comparison with alum coagulation**

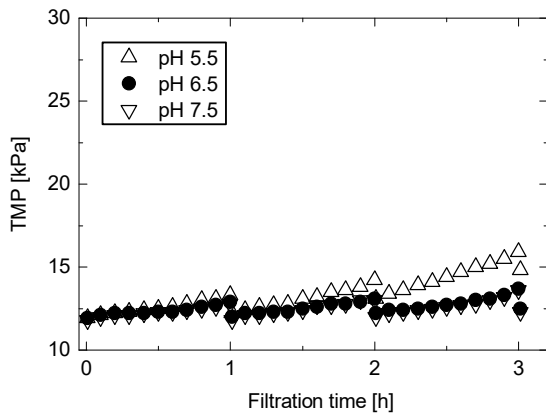
196 The impact of solution pH on the removal of turbidity and colour by MO coagulant was  
197 evaluated at the optimum MO coagulant dose (2 mL-MO/L). As reported in section 3.1, MO  
198 coagulant did not alter solution pH, thus solution pH was adjusted prior to coagulation. As a  
199 result, negligible impact was identified throughout the tested pH (5.5–9.5) (**Fig. 3**). A similar  
200 observation with negligible impact of solution pH on turbidity removal by MO coagulation has  
201 been reported in a previous literature.<sup>29</sup> MO coagulant in water at the tested pH is positively  
202 charged due to the isoelectric pH of the cationic proteins at 10.<sup>7</sup> Because colloidal particles in  
203 natural water are usually negatively charged, the dominant mechanisms of coagulant of charge  
204 neutralisation and adsorption effects<sup>9</sup> can effectively occur at the experiment pH value. This is a  
205 notable advantage over alum, which often requires pH adjustment for successful coagulation. A  
206 slightly high residual turbidity and colour was observed at the lowest pH (5.5). This may be due  
207 to the increased positive charge of the MO coagulant and reduced negative charge of particles in  
208 water.



209

210 **Fig. 3** – Removal of turbidity and colour by optimum MO coagulant dose (2 mL-MO/L) at  
 211 variable solution pH in the surface water. Optimisation of MO coagulant dose for the specific  
 212 river water is provided in **Fig. S2**.

213 The waters treated by MO coagulation at three pH (5.5, 6.5 and 7.5) were used to evaluate the  
 214 impact of MO coagulation pH on MF membrane fouling. Over the three filtration cycles, a  
 215 negligible difference was observed for pH 6.5 and 7.5 (**Fig. 4**). Coagulation using a surface water  
 216 is typically performed within the pH range. A slightly higher TMP was observed for pH 5.5,  
 217 which is in line with the results for turbidity removal. This indicates that turbidity removal by  
 218 MO coagulation is a key for achieving the optimum effect of membrane fouling mitigation.  
 219 Overall, the results indicate that pH adjustment prior to MO coagulant is not necessary to achieve  
 220 the optimum coagulation and fouling mitigation.



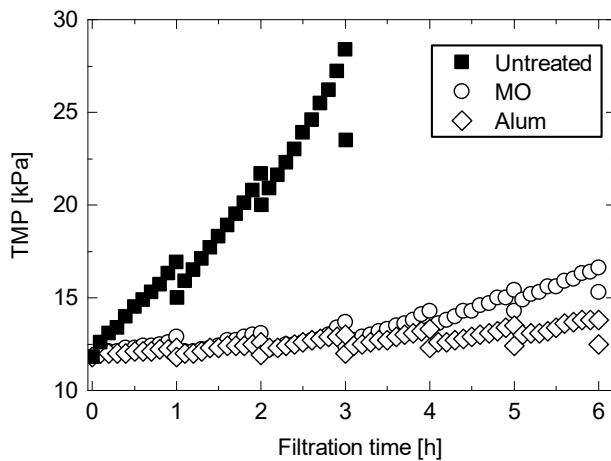
221

222 **Fig. 4** – Effect of solution pH in MF treatment of waters treated with the optimum MO coagulant  
 223 dose of 2 mL-MO/L (permeate flux = 140 L/m<sup>2</sup>h, backwashing flux = 280 L/m<sup>2</sup>h and  
 224 backwashing time = 1 min).

### 225 3.4 Comparison with alum coagulation

226 To demonstrate the potential of MO as an alternative coagulant of alum for MF treatment, the  
 227 effect of pretreatment on fouling mitigation between MO (2 mL-MO/L) and alum (4 mg-Al/L)  
 228 coagulation was compared. For both coagulants, minor fouling development was observed  
 229 during filtration (**Fig. 5**). MF treatment of the MO-treated river water revealed an increase in  
 230 TMP from 12 to 15 kPa over six filtration cycles. In contrast, alum coagulation prior to MF  
 231 treatment resulted in a less increase in TMP from 12 to 13 kPa. Despite of the relatively large  
 232 difference in residual turbidity between the two coagulants (MO = 1.1 NTU and alum = 0.1  
 233 NTU), they effectively mitigated membrane fouling and the subsequent increases in  
 234 transmembrane pressure in both cases did not differ significantly over six filtration cycles. This  
 235 indicates that sufficient level of membrane fouling mitigation in MF can be achieved through  
 236 MO coagulation. In addition, the results suggest that MO is a good alternative to alum and other  
 237 conventional chemical coagulants for coagulation pre-treatment prior to MF. Further

238 improvement for membrane fouling mitigation by MO coagulation can be expected by further  
239 optimising coagulation or filtration conditions. Coagulation condition includes solution pH, and  
240 filtration conditions include permeate flux and frequency of backwashing. In addition,  
241 modification on the extraction method of MO coagulant from seeds may improve coagulation  
242 performance in terms of turbidity removal, which can enhance membrane fouling mitigation.  
243 However, these further optimisations are beyond the scope of this study and will be addressed in  
244 the future study.



245  
246 **Fig. 5** – Fouling development in MF treatment without pretreatment, with MO coagulation at 2  
247 mL-MO/L or with alum coagulation at 4 mg-Al/L (permeate flux = 140 L/m<sup>2</sup>h, backwashing flux  
248 = 280 L/m<sup>2</sup>h and backwashing time = 1 min).

249 Despite of the difference in coagulation-treated water quality between MO and alum coagulation,  
250 water quality after MF treatment was comparable. For example, turbidity and colour in MF  
251 permeate was not detectable or almost zero for both coagulants (**Table 2**), indicating that MF  
252 treatment plays a major role in the reduction of turbidity and colour. In contrast, conductivity and  
253 TOC in MF permeate varied notably due to the difference in coagulation performance (**Table 1**)



254 and the incapability of MF for their removal. MF membrane is not capable of removing  
 255 dissolved ions, resulting in the difference in conductivity in MF permeate (275 and 312  $\mu\text{S}/\text{cm}$   
 256 for MO and alum, respectively). Likewise, TOC in MF permeate of the MO treated river water  
 257 was as high as 15.3 mg/L, which was 2.9 mg/L higher than that of the alum-treated river water.  
 258 An advantage of MO over alum was solution pH. pH is an important parameter in product water  
 259 to avoid damaging infrastructure in a water distribution network. MO coagulation followed by  
 260 MF treatment maintained solution pH almost constant at 7.4, which is within the recommended  
 261 range of 6.5–7.5 by WHO guidelines.<sup>30</sup> By contrast, alum coagulation reduced the solution pH  
 262 from 7.4 to 6.3 (below the WHO guideline level), and it remained unchanged after MF treatment.  
 263 This necessitates pH adjustment by addition of chemical reagents, which is a cost factor.

264 **Table 2** – Water quality in MF permeate of raw river water, MO and alum treated-river water  
 265 with the optimum coagulant doses (2 mL-MO/L and 4 mg-Al/L) during the 1<sup>st</sup> filtration cycle.

Parameter	Untreated river water (control)	MO treated river water	Alum treated river water
Turbidity (NTU)	0.2	0.0	0.0
Colour (PCU)	1.9	0.3	0.0
Conductivity ( $\mu\text{S}/\text{cm}$ )	257	275	312
TOC (mg/L)	15.3	15.3	12.4
pH	7.4	7.4	6.3

#### 266 **4. Conclusions**

267 Pre-treatment of the surface river with MO and alum at their optimum doses led to a substantial  
 268 reduction in membrane fouling. Despite of the relatively large difference in residual turbidity  
 269 after MO (1.1 NTU) and alum (0.1 NTU) coagulation, their membrane fouling levels on MF  
 270 treatment did not differ significantly over six filtration cycles. This indicates that sufficient level

271 of fouling mitigation in MF can be readily achieved through MO coagulation. Major water  
272 quality (turbidity and colour) after MF treatment was comparable between MO and alum  
273 coagulation. While MO coagulation slightly increased the load of organics, alum notably  
274 increased conductivity. MO coagulation did not change solution pH, which is advantageous over  
275 alum coagulation, because post treatment for pH adjustment can be avoided. The results reported  
276 here suggest that MO is a good alternative coagulant to mitigate membrane fouling of MF  
277 treatment for decentralised drinking water applications.

## 278 **5. Conflicts of interest**

279 There are no conflicts to declare.

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***Moringa oleifera* coagulation as pretreatment prior to microfiltration for  
membrane fouling mitigation**

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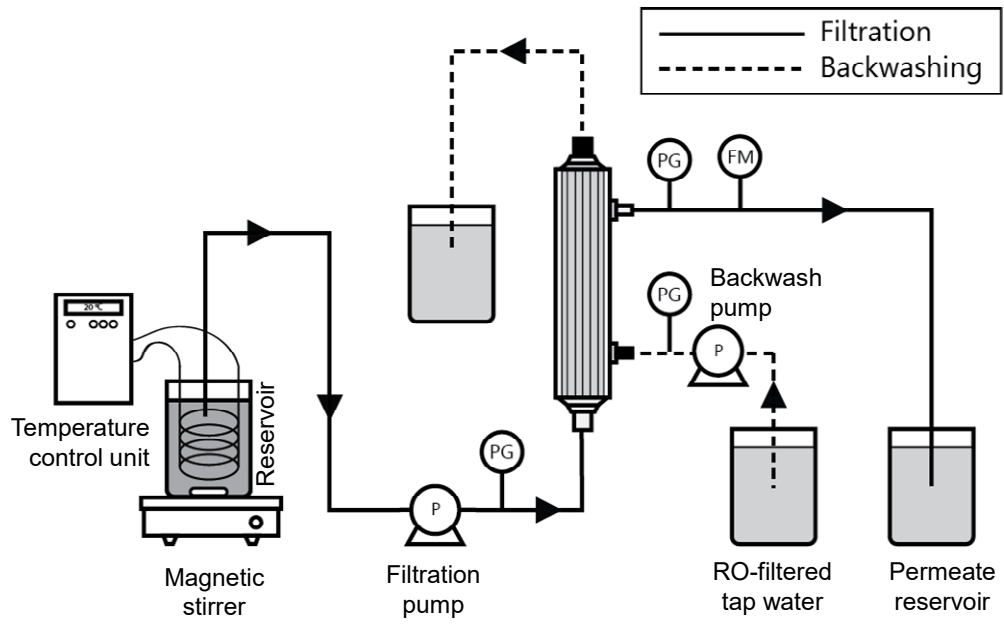
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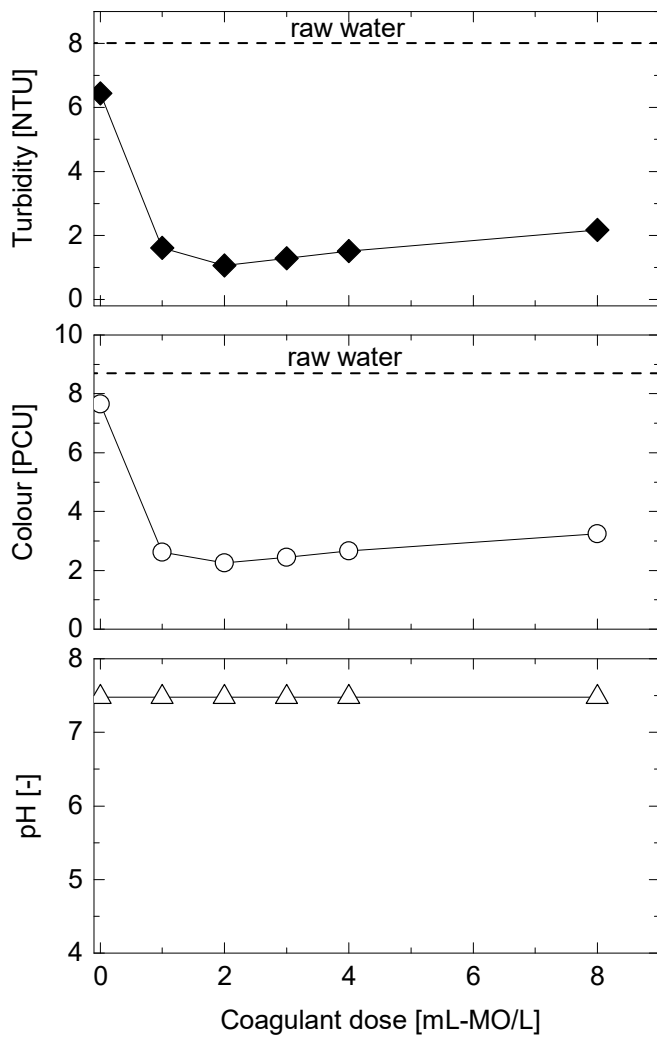
**Supporting Information**

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**Fig. 1** – Schematic diagram of the MF system.



**Fig. S2** – Removal of turbidity and color, and change in pH in the surface water at various MO coagulant doses (initial turbidity = 8.0 NTU and initial colour = 8.7 PCU).