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3 **Development of alum sludge-based constructed wetland: An**
4 **innovative and cost-effective system for wastewater treatment**

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17

18 **Abstract**

19 This paper describes (in a summarised manner) a research attempt to integrate the dewatered alum
20 sludge, a residual by-product of drinking water treatment process, into a constructed wetland (CW)
21 system for the purpose of enhancing the wastewater treatment performance, thus developing a so
22 called alum sludge-based constructed wetland system. A multi-dimensional research project
23 including the batch tests of phosphorus (P) adsorption onto alum sludge followed by the model
24 CWs trials of single and multi-stage CWs, has been conducted since 2004. It has been successfully
25 demonstrated that the alum sludge-based CW is capable of enhanced and simultaneous removal of P
26 and organic matter (in terms of BOD₅ and COD), particularly from medium and high strength
27 wastewater. The sludge cakes act as the carrier for developing biofilm for organics removal and also
28 serve as adsorbent to enhance P immobilization. Batch P-adsorption tests revealed that the alum
29 sludge tested possesses excellent P-adsorption ability of 14.3 mg-P/g.sludge (in dry solids) at pH 7.0
30 with the adsorption favored at lower pH. The results obtained in a 4-stage treatment wetland system
31 suggest that high removal efficiencies of 90.4% for COD, 88.0% for BOD₅, 90.6% for SS, 76.5%

32 for TN and 91.9% for $\text{PO}_4^{3-}\text{-P}$ under hydraulic loading of $0.36\text{m}^3/\text{m}^2 \text{ d}$ can be achieved. The field
33 demonstration study of this pioneering development is now underway.

34

35 **Keywords:** Alum sludge; constructed wetland; reuse; substrate; wastewater treatment

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37

38 **Introduction**

39 In line with sustainable wastewater treatment technologies which are environmentally friendly, easy
40 to operate, less energy-intensive and cost-effective, constructed wetland (CW) has been recognised
41 as a “green technology” that falls into the category of sustainable wastewater treatment
42 technologies. Although CWs have been shown to be very efficient in removing suspended solids
43 and organics (in terms of BOD_5 and COD), their performance has been inconsistent and often low
44 as regards nutrient removal. There is even more concern when they are used to treat medium to high
45 strength wastewaters. ^[1] Consequently, prominent research goals of CWs are to continually seek
46 specialized substrates with conducive physico-chemical properties to improve nutrient removal
47 (especially P) and to improve its design and operation, so as to facilitate enhanced oxygen
48 availability in order to achieve a higher degree of organics removal and nitrification. ^[2]

49

50 In the past five years, series of projects of different scales have been conducted at the University
51 College Dublin, Ireland, towards developing the novel alum sludge-based CW system for
52 wastewater treatment. Alum sludge refers to a resultant residual sludge, which is derived from
53 potable water treatment process that employs aluminium sulphate as coagulant to reduce the levels
54 of suspended particles, colour and organic matters in source water. Although the most prevalent
55 final disposal of the dewatered alum sludge is as a “waste” to landfill worldwide, the reuse of the
56 alum sludge is now attracting international attention. ^[3-5] The idea of reuse dewatered alum sludge as

57 main substrate in CW lies in its specific characteristics of abundant Al content and easy, local and
58 huge availability.

59

60 The main steps of the development towards the alum sludge-based CW system for P-rich
61 wastewater treatment are shown in Fig. 1. In the first step, emphasis was placed on assessing the
62 feasibility of the use of the dewatered alum sludge as an adsorbent and its capacity for P adsorption.

63 [6,7] Accordingly, the alum sludge was extensively characterized and the P adsorption behaviour
64 coupled with the P adsorption capacity of the dewatered alum sludge was examined. [8,9] Thereafter,
65 the optimal configuration of the proposed alum sludge based system, which will give the best
66 operational, hydraulic and treatment performance was determined using four different model reed
67 beds systems, configured with different proportions of the alum sludge and fed with a high P-rich
68 agricultural wastewater. Emphasis during this phase of the study was placed on isolating the
69 magnitude of the overall pollutant removal in the different model systems. The potential for bed
70 clogging and possible release of substances/elements from the alum sludge into the treated
71 wastewater upon passage through the system was also examined.

72

73 Upon the determination of the optimal configuration of the reed bed system, the model CWs (in
74 single column form) with dewatered alum sludge as main substrate were operated for short term [10]
75 and long term trial over two years. [11] Thereafter a multi-stage CW system was designed and
76 continuously operated to treat an agricultural wastewater with strong organic and nutrient pollutants
77 under high organic loading. The trial of the multi-stage system will continue to focus on addressing
78 the lifespan of the system as well as the final reuse of the alum sludge as resourceful material when
79 it becomes fully saturated in the reed bed treatment system.

80

81

[Fig. 1 here]

82 The main objective of this paper is to present the overall technical progression towards the
83 development of the alum sludge-based CW system. Therefore, the overall view of such the
84 development, rather than the detailed results and analysis, is given in this paper.

85

86 **Experimental approaches**

87

88 *Dewatered alum sludge and wastewater*

89 Dewatered alum sludge cake (with moisture content of 72-75%) was obtained from a local Water
90 Treatment Plant outside Dublin, Ireland where aluminium sulphate is used for flocculating reservoir
91 water. After collection, the sludge was air-dried, ground and sieved to prepare the sludge for batch
92 P-adsorption test and model CW trials. The aluminium content (expressed as Al_2O_3) in the sludge
93 tested by ICP-AES (inductively coupled plasma-atomic emission spectrometry) was 24.7-46.3%,
94 depending on the seasonal dosage of aluminium sulphate. Artificial P solutions were used in batch
95 tests while raw wastewater collected from an animal farm with about 2000 livestock units including
96 sheep, pigs, cattle and horses was used for the model alum sludge-based CW trial. The typical
97 concentration of such animal farm wastewater (after settlement) was 322-510 mg/L (SS), 720-1523
98 mg/L (COD), 540-850 mg/L (BOD_5), 210-350 mg/L (TN), 147-275 mg- PO_4^{3-} /L (P) and 6.7-7.6
99 (pH). During the experiment, the wastewater was diluted with tap water if necessary and used as
100 influent to the CW system.

101

102 *Batch P-adsorption tests*

103 P-adsorption was studied by a series of batch adsorption tests using a standard Stuart Orbital Shaker
104 (SSL 1, Bibby Sterilin Ltd.). Different weight of prepared dewatered alum sludge was poured into
105 plastic bottles which were filled with artificial P solution using 3 kinds of P-containing chemicals
106 including orthophosphate (KH_2PO_4), polyphosphate ($\text{BDH}(\text{NaPO}_3)_6$) and organic phosphate

107 (C₁₀H₁₄N₅O₇P · H₂O). Adsorption tests were conducted under varying pH of the P solution, initial P
108 concentration and agitating time. The maximum adsorption capacity (mg/g) was obtained by fitting
109 of experimental data with Langmuir isotherm. [7]

110

111 ***Optimal configuration trial***

112 Four identical Pyrex columns of 1000 mm in height and 95 mm in diameter were designed using
113 different configurations (not shown in schematic diagram). The columns were filled with prepared
114 alum sludge and overlain with pea gravel in different proportions ranging from 0 to 60%. In other
115 words, the proportion of the alum sludge in the four columns was 100%, 80%, 60% and 40%. A real
116 agricultural farm wastewater contained in a single feed tank was equally pumped to each of the
117 columns and the treatment efficiency for the pollutants including SS, P and COD was monitored for
118 25 weeks under hydraulic loading rate ranged between 1.23-1.86 m³/m².d.

119

120 ***The model CWs***

121 The schematic diagrams of a single model CW and a multi-stage CW system used in the study are
122 jointly shown in Fig. 2. The model CWs were made of Perspex columns. The single model CW is
123 1000 mm in height and 145 mm in diameter. The model CW was filled with dewatered alum sludge
124 up to 350 mm in depth with a bottom layer of gravel (100mm depth) which serves as support
125 material at the bottom of the model CW. The multi-stage CW system consists of four identical
126 single CW. Each stage was 900 mm in height and 95 mm in diameter. The prepared alum sludge
127 was filled into the CWs up to a depth of 550 mm and with a support base of 100mm-depth of
128 gravel. Common reeds, *Phragmites australis*, were planted on each of the CWs. Both the single and
129 4-stage model CWs were operated using a novel operational strategy referred to as ‘tidal flow’ in
130 which the ‘tide’, i.e. the rhythmical filling and draining of the bed medium, was generated by
131 peristaltic pump controlled by timer. [2,12] The hydraulic loadings of the single model CW and the 4-

132 stage CW system were $0.50\text{m}^3/\text{m}^2$ d and $0.36\text{m}^3/\text{m}^2$ d, respectively.

133

134 [Fig. 2 here]

135

136 *Sampling and analysis*

137 Procedure of the batch tests and sampling were described elsewhere. ^[7,9] Samples of batch P-
138 adsorption tests were subjected for P analysis using a HACH DR/2400 Colorimeter (CAMLAB Ltd,
139 UK) according to its manual. During the optimal configuration trial and CWs experiments of single
140 model CW and multi-stage CW system, samples of influent and effluent from the CW (or each
141 stage of the CWs) were collected and analyzed for SS, COD, BOD₅, TN, P, and pH. A PHM62
142 standard pH meter was used for the pH analysis. BOD₅ was determined using a BODTrack
143 apparatus (CAMLAB Ltd., UK). The COD value was read directly on the HACH DR/2400
144 Colorimeter after the sample was digested at 150 °C for 2 hrs in a HACH COD digester according to
145 the dichromate method. The SS value was read directly with the HACH DR/2400 Colorimeter.
146 PO_4^{3-} value was also measured with HACH DR/2400 Colorimeter after reaction with
147 Molybdovanadate. TN was determined according to persulfate digestion method and the value was
148 read after 5 min of reaction with TN Reagent C in a 25 mL standard glass cell.

149

150

151 **Results**

152 The maximum P adsorption capacity (Q_0) determined via Langmuir isotherm from the batch tests
153 when the source P of KH_2PO_4 was used is listed in Table 1, which shows that the dewatered alum
154 sludge has a high P immobilization ability and can be used as an adsorbent for P removal. However,
155 the adsorption ability is significantly favoured at lower pH.

156

157 [Table 1 here]

158

159 The typical results of the optimal configuration trial are given in Table 2. It can be seen that the
160 alum sludge can be employed as substrate to serve as filter medium for SS removal and as biofilm
161 carrier for COD reduction although the COD removal efficiency is quite low due to the high
162 hydraulic loading of 1.2-1.9 m³/m².d. More significantly, the enhanced P immobilization proves the
163 advantage of such use in the column. The inclusion of pea gravel at the infiltrative surface of the
164 columns did not prove to have any significant beneficial advantage. In addition, all the four
165 columns exhibited clogging tendencies irrespective of the substrate configuration. Interestingly,
166 removal of SS in gravel-containing columns seemed lower than that of pure alum sludge column
167 (see Table 2). However, there is lack in further SS removal measurement regarding clogging
168 investigation in this study.

169 [Table 2 here]

170 The results of the single model CW (refer to Fig. 2) tests are summarised in Table 3. It can be seen
171 that while the removal of organics in the model CW increased gradually with time during the testing
172 period, the removal of P was very high in the initial stages, and in particular, it remained high
173 throughout the experimental period. It is interesting to note from Table 3 that as the experiment
174 progressed and the system further stabilized, the removal of organics was considerably significant
175 in the system, and even at higher loadings. It can be suggested that the initial removal of the
176 carbonaceous substrates is through filtration while improved removal was obtained gradually
177 through aerobic degradation as the system stabilized and matured.

178

179 [Table 3 here]

180

181 The results of the 4-stage CW system (refer to Fig. 2) operated for 110 days using an animal farm

182 wastewater are illustrated in Fig. 3. An average removal percentage of 90% for COD and 88% for
183 BOD₅ was achieved. These results are very similar with those obtained from a reed bed system
184 using soil and gravel as main substrate. [13] The performance of the system on SS, P and TN removal
185 showed that significant removal of P was achieved in the system and 92% of the influent PO₄³⁻-P
186 was consistently removed. This is believed to be attributable to the adsorption of P onto the alum
187 sludge due to its strong affinity. [6] The average removal of TN and SS is 76% and 91%
188 respectively, as shown in Fig. 3. A higher TN removal efficiency appears to be observed in the later
189 stage of the trial. If this is the case, the promoted removal may be due to the progressively matured
190 microbial activities and the reed uptake. High SS removal in the system is believed to be due to the
191 filtration and the physical trapping of the particles by the alum sludge, which serves as the filter
192 medium in the system.

193

194

[Fig. 3 here]

195

196 **Discussion**

197 Studies on several potential CWs substrates including minerals and rocks, soils, marine sediments,
198 industrial by-products and man-made products show their P-removal capacity to vary from 0.025 to
199 32mg-P/g. [14] In comparison, the dewatered alum sludge used herein can be seen to have a
200 significant and comparable P adsorption capacity (Table 1). A variation of the adsorption maxima
201 with pH could possibly be attributed to the change of surface potential of the adsorbent particles
202 (alum sludge) and the combined effects of phosphate speciation and pH both on the adsorbent
203 particles, and in solution.

204

205 After identifying the adsorption capacity of dewatered alum sludge, an attempt to develop the alum
206 sludge-based CW was made. It should be pointed out that such development is hinged on the basis

207 that: (1) alum sludge is a locally, easily and hugely available material; (2) reuse of such water
208 industrial by-product falls into the theme of environmental sustainability which encourages “reduce,
209 reuse and recycle”; (3) P removal is an universal environmental problem and the reuse of such
210 sludge will enhance the removal of P from P-rich wastewater. Research of different phases in the
211 development of such novel CW provided convincing data to demonstrate that the dewatered alum
212 sludge is a promising material, which can be reused as a substrate in CW for wastewater treatment.
213 Removal of COD and BOD₅ can be achieved at about the same level as in soil and gravel-based
214 CW system. ^[13] This is a good demonstration of the dewatered alum sludge being a carrier for
215 biofilm development. It provided a surface for biofilm attachment which served to enhance
216 microbial activity in the CW. More importantly, P removal can be significantly enhanced (Fig. 3),
217 and this provides evidence to show that the dewatered alum sludge as a novel adsorbent and
218 substrate in CW can considerably improve the P-immobilization capacity. Due to the fact that alum
219 sludge is currently treated as a waste for landfill, the reuse of such sludge in CW will be reasonably
220 recognised as a cost-effective solution in CW development.

221

222 It is interesting to note that the lifespan of the alum sludge in CW regarding its saturation for P
223 adsorption was estimated according to the P-adsorption capacity and the operation condition of the
224 CW system. ^[11] In case of domestic wastewater treatment using alum sludge-based CW, the lifespan
225 of the alum sludge can be 9-40 years ^[11] when the total P discharge per person per day of 2.3g of P
226 was estimated and CW area of 5 m² per person was proposed. ^[15] Even in the case of P-rich animal
227 farm wastewater treatment tested in this study, the lifespan of the dewatered alum sludge is
228 estimated as 2.5–3.7 years. ^[11] Moreover, in most cases of the practical use of CW system, a multi-
229 stage treatment system is usually applied. Therefore, the lifespan is reasonably expected to be
230 longer. This is also an ongoing investigation in the multi-stage model CW system. In addition,
231 studies on the P desorption and recovery from the saturated alum sludge after use was conducted.

232 Several kinds of acids (HCl, HNO₃, and H₂SO₄) and bases (NaOH and KOH) were tested to extract
233 P from saturated alum sludge, which was used as media in CW. The results, which are given in
234 Table 4, show that either acid or base is efficient for P extraction and the efficiency relies mainly on
235 the concentrations of H⁺/OH⁻, and not on the type of acid or base. Under 0.1M H⁺/OH⁻, around 90%
236 of P can be extracted from the used alum sludge by acids, but only 65-66% by bases within 1 hour.
237 By considering the convenience and safety of extraction operation, ^[16] H₂SO₄ seems to be the best
238 reagent for such the P extraction.

239

240 It should be pointed out that during the testing period, especially in the 4-stage model CW system
241 trial, no obvious and serious bed clogging occurred. However, regarding the practical operation in
242 real situation of long term point of view, possible clogging behaviour and measures to
243 reduce/mitigate clogging should be investigated, since clogging is the most serious problem in
244 practice. This reflects the need for large scale and long term demonstration study of such novel CW
245 treatment system development, which is currently underway.

246

247

248 **Conclusions**

249 It is expected that this pioneering investigation on the possible application of dewatered alum
250 sludge as main substrate in a constructed wetland system treating P-rich wastewater would serve as
251 a primer for eventual field application. The results have demonstrated the promise of such novel
252 application of the “waste” alum sludge. Batch P-adsorption tests revealed that the alum sludge
253 tested possesses excellent P-adsorption ability with the adsorption favored at lower pH. The results
254 obtained in the 4-stage treatment wetland system suggest that high removal efficiencies of 90.4%
255 for COD, 88.0% for BOD₅, 90.6% for SS, 76.5% for TN and 91.9% for PO₄³⁻-P under hydraulic
256 loading of 0.36m³/m² d can be achieved. However, long term operation should be conducted for

257 further investigation of such innovative approach, especially the engineering aspect of the large
258 scale application. Notwithstanding, the development so far supports the proposition that the
259 potential reuse of dewatered alum sludge as a substrate in constructed wetland system can be a
260 promising solution to transfer alum sludge as a “waste” to a useful raw material, in developing a
261 cost-effective treatment wetland system.

262

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334 **Tables**

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337 **Table 1 Maximum P-adsorption capacity (with sludge particle size <0.063mm, tested by KH₂PO₄**
338 **under initial P of 102 mg/L and equilibrium time of 48 hrs)**

pH	4.3	6.0	7.0	8.5	9.0
Q ₀ (mg-P/g.sludge)	22.4	18.3	14.3	1.1	0.9

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Table 2 Typical results of optimal configuration trial

	1# (100% sludge)	2# (80% sludge)	3# (60% sludge)	4# (40% sludge)
P (%)	89.3	88.9	70.2	60.6
SS (%)	64.6	63.0	45.5	49.4
COD (%)	30.5	36.2	27.7	30.9

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Table 3 Results (mean±SD) of the single model CW trial

Parameter		Period (weeks)				
		1-8	9-12	13-16	17-20	21-28
BOD ₅	Loading (g/m ² .d)	35.8±8.5	50.0±6.7	57.5±10	114.8±15	111.7±13
	Removal (%)	63.3±16	71.7±5.2	75.9±1.4	80.3±7.1	82.3±3.5
COD	Loading (g/m ² .d)	66.7±21	77.1±12	99.4±13	187.6±25	183.6±11
	Removal (%)	50.2±9.1	75.1±3.8	78.0±3.1	84.4±2.8	85.5±2.1
P	Loading (g/m ² .d)	3.3±1.3	4.7±0.9	6.0±1.3	8.4±1.3	10.3±1.3
	Removal (%)	88.9±2.9	90.2±0.5	90.3±1.3	89.0±1.0	88.1±0.5
SS	Loading (g/m ² .d)	23.9±2.9	36±1.3	38.1±1.7	75.6±5.5	84.6±3.3
	Removal (%)	76.4±20	79.8±14	83±9.2	86±12.7	91.7±3.2

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Table 4 P release efficiency by different acid or base with different H⁺/OH⁻ concentrations

	HCl	HNO ₃	H ₂ SO ₄	NaOH	KOH
0.01 M	4.2	4.2	4.8	16.3	17.8
0.03 M	41.2	42.0	37.5	46.0	52.5
0.05 M	75.0	72.2	72.3	60.8	61.2
0.075 M	85.8	89.8	88.5	65.5	63.3
0.1 M	90.7	91.0	88.5	66.1	65.3

Unit: %

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Figure captions

Fig. 1 Road map of the development of the alum sludge-based CW

Fig. 2 The schematic of the experimental CWs systems

Fig. 3 Summarized performance of the 4-stage CW system

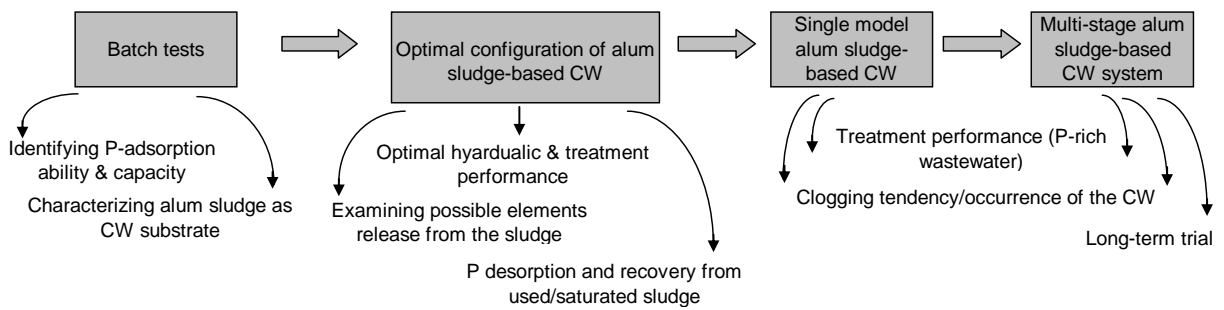
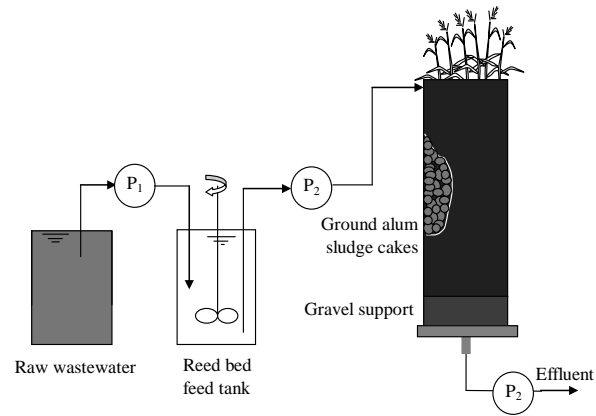
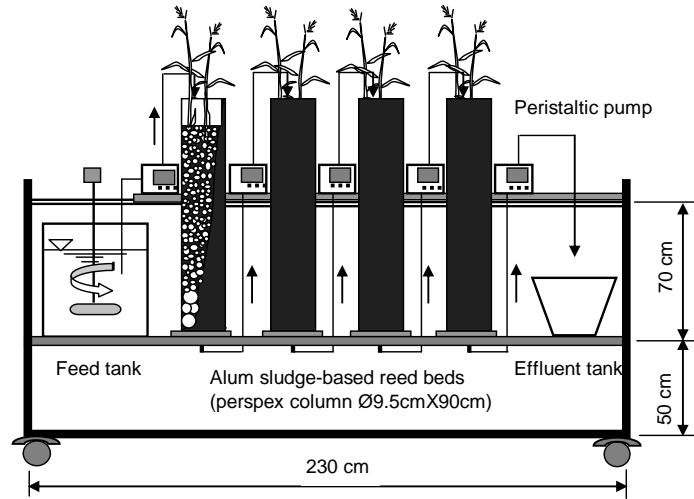


Fig. 1

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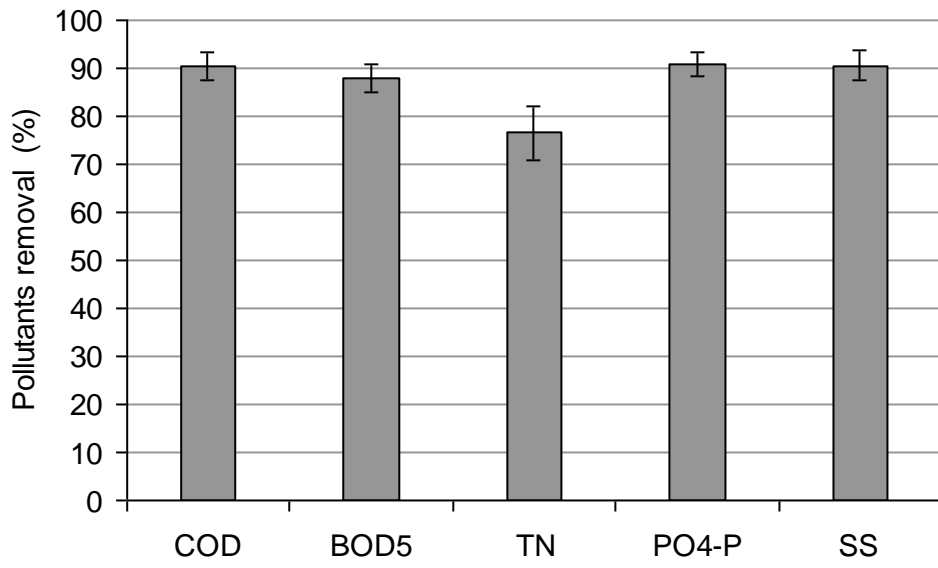


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Fig. 2



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Fig. 3