

1 **Removal of chemical and microbial contaminants from greywater using a**
2 **novel constructed wetland: GROW**

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10 **Abstract:**

11 The availability of freshwater resources is becoming universally depleted, leading to the
12 requirement for a focused management strategy for treating and reusing wastewater. In particular
13 for urban and developing areas, small scale decentralized treatment systems are becoming
14 popular. The GROW (Green Roof-top Water Recycling System) constructed wetland is one such
15 option that provides a solution without a permanent land requirement and offering medium to
16 high treatment efficiency. The performance of the GROW system was monitored from
17 November 2013 to April 2015 in treating greywater from the Krishna Student Hostel in IIT
18 Madras. The performance of the GROW wetland cells were examined over four monitoring
19 periods in Phase 1 namely: 1) start-up stage, 2) seasonal variation 3) change of flow rate and 4)
20 change in organic fraction (26.8, 25.9 and 25.5 g COD/ cubic. meter/ day respectively). In Phase
21 2, the plants and the filling materials were changed and the performance of GROW wetland cells
22 were evaluated. The system was fed with greywater at a flow rate of 62, 70, 82, 100 and 120 L/
23 day respectively with hydraulic retention time of 0.7 – 1.3 days. The samples taken from the inlet

24 and the outlets of the GROW system were taken weekly and analyzed for the following
25 parameters; pH, COD, BOD, TSS, TN, NO₃ – N, TP, FC, SDS, PG and TMA. In the study, the
26 overall removal efficiency was greater than 82% for all the parameters. The GROW wetlands
27 reduced all the above mentioned parameters to within or closely to the USEPA standard limits
28 for reuse. The reusable effluent water is named ‘Green Water’.

29

30 ***Keywords:***

31 *Constructed wetland, Greywater, Recycle, Surfactants and Personal care products, Nutrients,*
32 *Organics*

33

34 **1. Introduction:**

35 Increasing stress on the availability of freshwater sources worldwide has forced water providers
36 to develop wastewater management strategies giving emphasis for recycling and reuse of treated
37 wastewater. Wastewaters from households are classified into two types, i.e., i) greywater and ii)
38 black water. Greywater includes wastes generated from bathroom sinks, baths or showers,
39 washing clothes and possibly dishwasher except the wastewater from toilet whereas black water
40 is the wastewater generated from toilets. Wastewater from dishwashers is usually excluded from
41 greywater, due to high loading of fats/oils/ greases (FOGs), organic content and bacterial
42 contamination, which makes the wastewater difficult to degrade and handle (Jefferson et al.,
43 2000; Avery et al., 2007). Greywater treatment and reuse is one of the efficient solutions which
44 offer the largest potential of water savings, accounting for 50-80% of freshwater water
45 consumption (Eriksson et al., 2002; Gross et al., 2007) in domestic purposes. Moreover,
46 greywater is lightly polluted and requires less expensive treatment prior to non-potable reuse

47 (Jefferson et al., 2000; Avery et al., 2007). There are various technologies available for treatment
48 of greywater such as activated sludge process (ASP), membrane bioreactors (MBR), sequential
49 batch reactor (SBR), rotating biological contractor (RBC), photocatalysis and electro coagulation
50 (Merz et al., 2007; Masi et al., 2010). However, capital/infrastructure cost, social acceptance and
51 power requirement may limit their application in rural and peri-urban areas in developing
52 countries.

53
54 Treatment and reuse of greywater (as 'Green Water') for non-potable/secondary applications
55 using various low cost less land intensive, sustainable and efficient technologies have been
56 carried out in the past. The greywater was treated using a novel organic cation octadecyl
57 trimethyl ammonium (ODTMA) with montmorillonite as a filtration unit along with a moving
58 bed biological reactor for decomposition of part of the organic matter in the GW. The ODTMA
59 complex was efficient in purifying GW due to its large surface area, positive charge and
60 existence of hydrophobic domains (Rakovitsky et al., 2016). Another study uses an anaerobic
61 filter followed by ultraviolet disinfection system for the treatment and reuse of greywater from
62 an airport in Brazil (Do Couto et al., 2015). In-order to improve the green area of the city and to
63 treat domestic greywater through a shallow horizontal subsurface constructed wetland that can be
64 located in a household roof. A Wetland roof (WR) system was developed by Thanh et al. (2014),
65 the system achieved an average COD removal efficiency of 77–78% or 20–28 kg COD/ha d for
66 both sunny and rainy days. The system was able to remove nutrients also effectively with a TN
67 removal efficiency of 88–91% or 17–20 kg TN/ha d, and a TP removal efficiency of 72–78% or
68 1.6 kg TP/ha d for different HLRs. A pilot installation of a green wall treating greywater from an
69 office building in Pune, Maharashtra State, India. Green walls were filled with LECA®

70 (lightweight expanded clay aggregate) and coconut fibers. COD removal efficiency of this
71 system was in the order of 14–86% (Masi et al., 2016).

72

73 Constructed wetlands (CW) are also one of such systems considered as sustainable, cost effective
74 and a viable treatment option for treating greywater for small communities. Over the past few
75 years, CW has gained popularity due to its effectiveness, low capital investment and low cost of
76 operation with less maintenance over the conventional systems for treating various types of
77 wastewaters such as municipal wastewater, textile effluent and landfill leachate (Masi et al.,
78 2010). The earlier researches are mainly focused only on the treatment of real-time greywater
79 using CWs for the overall removal of organics, nutrients and pathogens (Avery et al., 2007;
80 Gross et al., 2007; Frazer Williams et al., 2008; Winward et al., 2008). But the present study
81 focuses on the performance of novel constructed wetland (GROW) under various operating
82 conditions (start-up, seasonal, hydraulic loading rate and organic loading rate) in treatment of
83 real-time greywater. Additionally, the current study also focuses on the removal of surfactants
84 and personal care products (SDS, PG and TMA) from real time greywater using GROW system.

85

86 There are various types of constructed wetland classified based on their flow pattern;

87 i) Horizontal subsurface flow constructed wetland,

88 ii) Vertical subsurface flow constructed wetland and

89 iii) Hybrid subsurface flow constructed wetland. The most commonly used hybrid flow CW is
90 that in which the wastewater flows first into a horizontal flow CW (HFCW) and then to a vertical
91 flow CW (VFCW) or vice versa, whereas in a few other studies hybrid systems are differentiated
92 from other systems by introducing the baffles in the bed to make horizontal and vertical flow

93 pattern in a single basin (Tee et al., 2012; Cui et al., 2015; Ramprasad and Philip, 2015). The
94 advantage of the hybrid system is that the nitrogen can be nitrified completely in vertical flow
95 CW and denitrified in horizontal flow CW (Sayadi et al., 2012). However, the disadvantage is
96 that it requires large areas of land and complex construction and operation. To overcome the
97 problem, a novel GROW constructed wetland (Green Roof-top Water Recycling System) was
98 developed which is suitable for use in urban areas where ground space is limited

99

100 The performance and working of the GROW system was originally monitored and subsequently
101 studied at Cranfield University, UK by Avery et al., (2007), Memon et al., (2007) and Winward
102 et al., (2008). A novel GROW system for treating 480 L/day of the hostel greywater with a
103 hydraulic retention time of 18.6 hours. The system consisted of a sequence of trough and weirs
104 that were placed above the wooden frame on a pitched roof. The troughs were filled with
105 expanded clay (size 0.1m) and gravel chippings (size 0.2m) and were planted with 8 varieties of
106 native aquatic species. They found that the GROW system was most effective in the removal of
107 suspended solids and turbidity (mean removal rates 91.2% and 98.2%, respectively). They also
108 reported a 4.2 log reduction of total coliforms in the system. With a COD and BOD removal
109 efficiencies of 59-80% and 84-92%, respectively, the treated water from the system was able to
110 meet the stringent United States Environmental Protection Agency (USEPA) standard for water
111 reuse (BOD <10 mg/L). They also claimed that the GROW system performed better than
112 horizontal and vertical flow constructed wetlands (Avery et al., 2007). A comparative studies on
113 the life cycle impact assessment of GROW system with other three biological treatment systems
114 like membrane bioreactors (MBR), membrane chemical reactors (MCR) and reed beds were
115 done by Memon et al. (2007). They concluded that the GROW system performed best in most of

116 the impact assessment categories and MCR appeared to be less environmentally friendly
117 (Memon et al. (2007)). Similar way, another study evaluated the presence of common pathogens
118 (total coliforms, *E. coli*, *Enterococci*, *Clostridia* and Heterotrophs) in greywater and compared
119 the performance of GROW, VFCW, HFCW, MBR and MCR in the removal of pathogens. These
120 systems were operated continuously with a flow rate of 480 L/day with an HRT of 2.1 days. It
121 was found that MBR system provided better quality treated effluent by meeting the stringent
122 USEPA standard limits for reuse followed by VFCW, GROW, HFCW and MCR (Winward et
123 al., 2008).

124

125 In general constructed wetlands performances were affected by various factors such as climatic
126 conditions, greywater characteristics, native plant species and substrate materials. The literature
127 on GROW system were found to be mostly concentrated in the temperate maritime climate. The
128 substrate (filling) material and plant species used in the earlier studies were mostly indigenous to
129 the UK. Hence, it is necessary to evaluate the performance of the GROW system in different
130 climatic conditions, vegetation patterns and greywater characteristics to determine the suitability
131 of the system in other regions. Moreover, previous studies on GROW systems were conducted
132 mostly at one particular flow rate, at constant HRT and at single organic loading rate (OLR).
133 Information regarding the fate of surfactants and personal care products in GROW systems, is
134 also lacking. Therefore, the present study focused on the evaluation of the performance of the
135 GROW system in Indian tropical conditions and with native filling materials (sand, brick bat and
136 gravel (1:1:1)) and 8 different plant species commonly available in India (*Canna indica*, *Canna*
137 *flaccida*, *Canna lily – hybrid*, *Cardamina pratensis*, *Plectranthus amboinicus*, *Crossandrain*
138 *fundibuliformis*, *Phragmites australis*, *Solanum trilobatum*), at different flow rates (62, 70, 82,

139 100 and 120 L/day), and organic loading rates (26.8, 25.9 and 25.5 g COD/ cubic. meter/ day).
140 The study also evaluated the effect of seasonal variations, change of plant species and substrate
141 materials on the performance of GROW system. The fate of surfactants in GROW system was
142 also evaluated.

143

144 **2. Materials and Methods:**

145 **2.1 GROW constructed wetlands:**

146 A novel constructed wetland system, Green Roof-Top Water Recycling System (GROW), was
147 developed by Water Works UK Ltd., London, UK and was fabricated and installed in Krishna
148 Hostel, IIT Madras, Chennai, India (GPS coordinates 12° 59' 1.266" N; 80° 13' 57.3852" E).
149 Chennai lies on the thermal equator and features a tropical wet and dry climate with the
150 temperature ranging from 18°C - 42 °C and average annual rainfall of 1400 mm. The pilot scale
151 experimental system for the treatment of greywater from the hostel was in operation from
152 November, 2013 to December, 2016. The GROW system consisted of four rows of troughs
153 connected laterally and placed on a mild steel scaffolding frame. Each row consisted of two
154 troughs mounted in series and butted up to each other. The scaffolding frame was placed on the
155 leveled ground surface, and the top row of the troughs 'A' was placed 0.8 m above the ground
156 surface and the lowest one (trough 'D') was positioned at 0.4 m above the ground surface (Fig.
157 1). The troughs of the GROW system were made of high density polyethylene sheet of 6 mm
158 thickness 4 m length and 2 m wide. The trough had a depth of 25 cm with a water holding
159 capacity 125 L per trough. The troughs were fitted with intermediate 'baffles' and 'weirs'
160 arranged in such a way that the wastewater was forced to have contact with the whole depth of
161 media/ substrate and thereby reducing any short-circuiting. In first phase of the study, troughs

162 were filled with a support medium which consisted of a mixture of sand, brick bats and gravel in
163 equal proportion (1:1:1) to approximately 15 cm depth. The total volume of the GROW system
164 was 1.84 cubic. meter and each substrate material occupied a volume of 0.4 cubic meters.

165
166 The troughs were planted with 8 varieties of native plant species of *Canna indica*, *Canna*
167 *flaccida*, *Canna lily – hybrid*, *Cardamine pratensis*, *Plectranthus amboinicus*, *Crossandrain*
168 *fundibuliformis*, *Phragmites australis*, *Solanum trilobatum*. The planting plan employed for the
169 study is shown in Fig 2. Trough 1 was used only with substrate without any plants to act as an
170 additional settling unit; trough 2 was planted with 4 plants of *Canna indica*, trough 3 was
171 planted with 4 *Canna flaccid*, trough 4 was planted with *Canna lily – hybrid* (3 numbers), trough
172 5 was planted with 3 plants of *Cardamine pratensis and*, trough 6 was planted with 3 plants of
173 *Plectranthus amboinicus*, trough 7 was planted with 1 plant of *Canna indica and* 2 plants of
174 *Solanum trilobatum*, trough 8 was planted with 1 plant of *Phragmites australis* and 1 plant of
175 *Crossandrain fundibuliformis*, trough 9 was planted with 4 numbers of *Crossandrain*
176 *fundibuliformis*, trough 10 was planted with 2 numbers of *Canna lily – hybrid*, and 3 varieties of
177 *Canna flaccid*, trough 11 and 12 were planted with 5 numbers of *Canna indica*. In Phase 1,
178 above mentioned plant species were planted on the trough having a surface area of 8 square.
179 meter with a plant density of 4 plants per square. meter. In phase 2, the substrate material filled
180 was removed and replaced with gravel of size < 5 cm and the plants were replaced with *Canna*
181 *Sp* with a planting density of 4 plants per square. meter.

182

183 2.2 Substrate Characteristics

184 In phase 1, the filter media used was a mixture of sand, brick bats and gravel of equal proportion
185 (1:1:1). Three different filter media were purchased commercially, sand with a particle size of
186 0.5 mm, gravel (10 mm) and brick bat of size < 5 cm were filled in the troughs for a depth of 15
187 cm. In phase 2, the old filter media were replaced completely with gravel of particle size <10
188 mm.

189

190 **2.3 Greywater Sources:**

191 The influent raw greywater was collected from the Krishna student hostels on IIT Madras
192 campus, Chennai, India. Wastewater from baths, showers, wash basins and washing machine
193 were collected separately and drained into a common settling tank from which 100 L was
194 pumped to an over head tank. The greywater from the overhead tank was allowed to flow by
195 gravity into the GROW system through a flow control valve. The water entered from one trough
196 to another (1 to 12) through the weirs and baffles continuously from the top trough to bottom
197 trough where it reached the outlet pipe. The greywater was supplied to the GROW system
198 continuously with a hydraulic loading rate of 53.1 – 58.9 L/ cubic. meter/ day with a hydraulic
199 retention time varied from 0.7 to 1.3 days. The operating history of GROW system is tabulated
200 in **Table 1**.

201

202 **2.4 Sampling and Analysis:**

203 The raw greywater and treated water samples were collected every week starting from
204 November, 2013 between 09:00 and 12.00 hours. In addition, samples were collected from the
205 end of each row of troughs 1, 2, 3 and 4 every month. The samples were carried to the laboratory
206 in air tight plastic bottles and were stored in refrigerator at 4°C. The water samples were further

207 examined for the physico-chemical and biological parameters as per standard methods for the
208 examination of water and wastewater (APHA, 2012). pH of the sample was analyzed using
209 Eutech cyberscan PCD 650 multi parameter kit (Thermo scientific, Singapore). Chemical oxygen
210 demand (COD) was measured using a closed reflux chromate titrimetric method, Biochemical
211 oxygen demand (BOD) was measured using the 5 day incubation method, Total organic carbon
212 (TOC) and total nitrogen (TN) were measured using total organic carbon analyzer V600 series
213 (Shimadzu, Japan). Nitrate nitrogen ($\text{NO}_3 - \text{N}$) and total phosphate (TP) was analyzed using UV
214 spectrometer (UV-VIS 8000, Shimadzu, Japan) (APHA, 2012). Fecal coliform (FC) was
215 measured by chromocult nutrient media plates supplied by Sartorius, Germany. The sodium do-
216 decyl sulphate (SDS) was measured calorimetrically at 467 nm using a UV 1800
217 spectrophotometer (Shimadzu, Japan). Propylene glycol and Trimethyl amine were measured
218 using gas chromatography fitted with flame ionization detector (PerkinElmer Clarus 500).

219

220 **2.5 Statistical Analysis**

221 The performance of the GROW constructed wetland system was statistically evaluated by
222 comparing the means of effluent concentrations of various parameters under different operating
223 conditions, using paired sample 't' test. The paired 't' test are commonly applied for comparing
224 the means of data's from two related samples or variables. The statistical analysis was performed
225 using IBM SPSS statistics 20 software at 95% confidence level ($p < 0.05$).

226

227 **3. Results and Discussion**

228 **3.1 Influent raw greywater quality**

229 The raw wastewater characteristics analyzed over a period of time is shown in **Table 2**. In
230 general the pollutant concentrations such as organics, solids and indicator organisms in
231 greywater are comparatively less than that in domestic wastewater. The greywater also has lesser
232 macronutrients (N and P) than the domestic wastewater. The organics concentration and fecal
233 coliforms of greywater used in this study were lower than the reported values. The mean value of
234 COD was 216–320 mg/L and BOD was 68-120 mg/L. The obtained values of COD and BOD
235 were lesser than earlier reported values by **Gilboa and Friedler, 2008**. The reason for lesser
236 concentration of these parameters is apparently due to the very high per capita water
237 consumption. The COD: BOD ratio was in the range of 2.7-3.0, which indicates that greywater
238 contains higher amount of recalcitrant organics than sewage (**Metcalf et al, 2010**). The reason
239 for higher COD: BOD ratio may be due to higher usage of surfactants and personal care products
240 during laundry services. As no urination bowls were connected with the separated greywater, the
241 concentrations of nitrogenous and phosphorus compounds in greywater were also lesser than the
242 reported values. The phosphorus present in the greywater mostly originated from the detergents
243 used in washing powders. The values of emerging contaminants i.e., surfactants namely sodium
244 do-decyl sulphate (SDS), propylene glycol (PG) and trimethyl amine (TMA) were present in the
245 concentration ranges of 14.9-35.9 mg/L, 11.6-46.6 mg/L and 8.7-15.5 mg/L, respectively. The
246 obtained values were similar to earlier reported values for SDS by **Gross et al., 2007**. There were
247 no supporting data available regarding the concentrations of PG and TMA in the raw greywater.

248

249 **3.2 Performance of GROW System under different operational conditions**

250 The performance of GROW system was evaluated in two different phases, phase 1 was further
251 subdivided into four different sub-phases viz. a viz., start-up phase (Phase1.1), seasonal variation

252 (Phase 1.2), flow rate variations (Phase1.3) and organic load variations (Phase1.4). The first 4
253 weeks of Phase 1.1.covered the start-up stages of the GROW system. During this phase, the
254 system was fed with greywater at the flow rate of 70 L/day. During this period, the plants and
255 microbes were allowed to acclimatize to the newer environment. In Phase1.2, the performance of
256 the GROW system at various seasons and temperature, i.e., summer, monsoon, pre-monsoon and
257 post monsoon, were evaluated for the designed flow rate of 70 L/ day. In phase1.3, different
258 hydraulic loading rates were employed, i.e., 62 L/day, 82 L/day, 100 L/day and 120 L/day.
259 Finally in Phase1.4, the performance of the system was evaluated for various organic loading
260 rates (25.5 g COD/ cubic. meter/ day, 25.9 g COD/ cubic. meter/ day and 26.8 g COD/ cubic.
261 meter/ day) by adding sucrose as an external carbon source at a flow rate of 100 L/day. In Phase
262 2, the GROW system was operated at constant flow rate of 100 L/day to evaluate the effect of
263 different substrate materials and plant species on the performance of GROW system. The short
264 term equilibrium was attained within 2 months from the date of plantation in the GROW system
265 and performing well after 3 years of continuous operation. If the GROW system is properly
266 maintained, the system can work for another 2-3 years.

267

268 **3.2.1 Organics**

269 During the study period, the influent BOD and COD varied from 68-120 mg/L and 216-320
270 mg/L, respectively as shown in **Table 2**. However, the variation of influent quality did not affect
271 the outlet biochemical oxygen demand (BOD) and chemical oxygen demand (COD)
272 concentrations during the monitoring period. It was consistently below 10 mg/L for BOD and 20
273 mg/L for COD, which is below the USEPA standard limits for secondary reuse. It was also found
274 that the GROW system showed a better removal efficiency during the summer season compared

275 to other seasons as shown in Fig 3(a)-(b). As stated by Vymazal, 2002; Akratos and Tsihrintzis,
276 2007 that the organic pollutants are removed mostly by microbial degradation and also by
277 adsorption to a certain extent. Hence, at an elevated temperature, the activities of aerobic and
278 anaerobic microbes are enhanced, resulting in higher organic pollutant degradation during
279 summer season. During phase 1.3, it was found that the BOD and COD concentration in the
280 effluent were comparable at the flow rates of 62, 82 and 100 L/day and were increased as the
281 flow rate increased to 120 L/day. This indicates that 100 L/day can be considered as the optimal
282 flow rate for the maximum pollutant removal. As the flow rate increased, the retention time
283 (HRT) decreased, resulting in lesser removal of organic pollutant. Similar results were reported
284 by Akratos and Tsihrintzix, 2007. In phase 1.4, the effluent BOD and COD concentrations were
285 changed slightly. As the OLR increased from 25.5 to 26.8 g COD/ cubic. meter/ day, the effluent
286 COD and BOD concentrations were increased. It was observed that at 26.8 g COD/ cubic. meter/
287 day, the COD values were 16-24 mg/L, while it was <16 mg/L during the other two OLRs.
288 These results were in accordance with the results reported by Lin et al., 2002 and Saeed and Sun,
289 2012. This indicates that 26.8 g COD/cubic. meter/day OLR exceeded the degradation capacity
290 of the wetland system (Dalahmeh et al., 2014). The variance of means of the effluent quality at
291 various monitoring periods were found to be statistically significant ($p < 0.05$) [Supplementary
292 Table S1]. In phase 2, the COD and BOD removal efficiency was found to be 88% and 84%,
293 respectively [Supplementary Fig. S1]. The reason is that the organic pollutants are mostly
294 removed by microbial degradation and by adsorption (Vymazal, 2002).

295

296 3.2.2 Suspended solids

297 The inlet and outlet suspended solids concentrations and percentage removal during the
298 monitoring period are shown in Fig. 4. The suspended solids in the constructed wetlands are
299 removed from the wastewater by physical processes such as filtration and sedimentation
300 (Haghshenas-Adarmanabadi et al., 2016). According to Masi and Martinuzzi (2007), the solids
301 removals by the constructed wetlands are in the range of 72-84% in the Mediterranean countries,
302 and 65-91% in the tropical regions of developing countries (Singh et al., 2014). In the present
303 study about 85-90% (< 20 mg/L) removal of solids particles from inlet to outlet tank was
304 achieved. The reason for higher removal efficiency may be due to the baffled CW configuration
305 that prolonged the water flow path and enhanced the filtration process which favored the removal
306 of suspended solids.

307

308 It was also observed from Fig. 4 that during phase 1.1, the removal of suspended solids was less
309 (<80%). The microbes and plants started growing at this stage and they were not completely
310 matured. This may be the reason for the low performance. As the time progressed, in phase 1.2,
311 the removal of solids improved and remained almost at a constant level (88-95%). During phase
312 1.3, (i.e., change of flow rate), it was observed that as the flow rate increased, the removal of
313 solids concentration decreased due to lesser hydraulic retention time. Similar results were
314 reported by other researchers also (Akaratos and Tsihrintzis, 2007). As the organic load were
315 increases from 25.5 to 26.8 g COD/cubic. meter/ day, the solids concentration in treated water
316 also increased. The above obtained results were in good agreement with Dominguez et al., 2012
317 that the increased organic loading rate resulted in increased biomass growth which in turn
318 increased the suspended solids concentration in the effluent. However, the overall removal of
319 solids was comparatively lesser compared to other constructed wetlands like horizontal, vertical
320 and hybrid flow systems (Ramprasad and Philip, 2016). The statistical analysis (paired 't' test)

321 were conducted to evaluate the performance of GROW system for the removal of solids during
322 various monitoring periods (start-up, seasonal, hydraulic loading rate and organic loading rate)
323 and paired 't' test showed that the treated effluent quality during all the monitoring periods are
324 statistically significant at 95% confidence interval ($p < 0.05$; $p = 0.039$) [Supplementary Table
325 S1]. The removal of suspended solids was highly affected by lowering the HRT and increasing
326 the OLR.

327

328 3.2.3 Nutrients

329 Nutrients (nitrogen and phosphorous) presence in wastewater is one of the major factors that
330 causes eutrophication, deplete the dissolved oxygen level and can be toxic to the ecosystem.

331 Generally, the removal of nutrients is by ammonification, nitrification, denitrification, plant
332 uptake, volatilization and biomass assimilation (Vymazal, 2002; Akrotos and Tsihrintzis, 2007).

333 In the present study, the concentrations of nitrates, ammonia and phosphates present in the
334 influent were low as compared to earlier reports (Gilboa and Friedler, 2008; Antonopoulou et al.,
335 2013). The nitrogen compounds removal was around 88 to 99% during summer season and was
336 found to be lesser during other seasons (Fig. 5-). Microbial reactions such as organic nitrogen
337 decomposition, nitrification and de-nitrification are favored at higher temperature resulting in
338 greater removal efficiency. Similar to nitrogen compounds, phosphate removal was also favored
339 at high temperatures. During the summer season, the removal of total phosphate was maximum
340 at 92%, which was less during other seasons (Fig. 6). It was reported in earlier studies that the
341 main mechanism involved in phosphate removal were sorption and plant uptake (Vymazal,
342 2002). Sorption of phosphate is an endothermic reaction (Jin et al., 2005), which means that low
343 temperatures decrease the sorption capacity of the bed (Rustige et al., 2003). The results obtained

344 by GROW systems were compared with the other three wetlands (horizontal vertical and hybrid)
345 studied by Ramprasad and Philip, 2016. The hybrid wetland performed better than GROW and
346 other two systems. Many reports suggested that the nutrients removal in constructed wetlands
347 was predominantly due to de-nitrification activity (Vymazal, 2002). During phase 1.3, enhanced
348 activity of de-nitrifiers was observed due to high HRT which resulted in higher removal rate. In
349 phase 1.4, as the OLR increased from 25.5 to 26.8 g COD/ cubic. meter/ day, the removal rates
350 of nutrients increased (Fig. 5-6). The probable reason might be that, increased organic load
351 triggered an increased growth of anoxic microbes near the root nodules, providing a favorable
352 condition for de-nitrification (Dalahmeh et al., 2014). The statistical analysis confirms that the
353 effect of different operating conditions on the nutrients content was recognized as highly
354 significant ($T = 4.367$; $p = 0.005$).

355

356 In phase 2, the removal of nutrients and phosphate were in the range of 82-88% and 65-74%,
357 respectively [Supplementary Fig S1]. The GROW system was capable of removing the nutrients
358 from the greywater below the reusable standard level. It was observed that the nitrate - nitrogen
359 in the treated wastewater were in the range of 1.2-3.5 mg/L and 0.8-1.4 mg/L for total
360 phosphates. (Fig. 5 - 6)

361

362 3.2.4 Fecal Coliforms

363 The fecal coliform concentration in the inlet was relatively low (50-120 CFU/100 mL) compared
364 to the earlier reported values (Antonopoulou et al., 2013). The major reason for the lesser
365 coliform contamination may be due to the age group of inhabitants and avoidance of kitchen
366 wastewater. Most of the inmates were in the age group of 19-25 years. The fecal coliform

367 removal in constructed wetland is attributed to physical process such as sedimentation, filtration
368 and natural die-off. The removal rates of the fecal coliform were in the range of 70-85% during
369 the start-up stage, and gradually increased and reached around 98% during phase 1.2. It is clear
370 from Fig 7 that during the summer seasons the FC removal was more than the other seasons. It is
371 also evident that with increase in hydraulic retention time, during phase 1.3, the removal rates
372 also increased from 94% to 98% (Fig 7). Akaratos and Tsihrintzis, 2007 have previously
373 reported that as the HRT increased the coliform have higher contact time in the system to get
374 removed or degraded resulting in higher removal efficiency. As the OLR increased, the effluent
375 coliform concentration also increased from 4 CFU/ 100 mL to 12 CFU/ 100 mL, due to increased
376 biomass growth by utilizing the readily available carbon source. . Similar results were reported
377 by other researchers also (Dalahmeh et al., 2014). Statistically significant difference ($p < 0.05$;
378 $T = 5.860$) in the average fecal coliform content occurred in different operational phases. During
379 Phase 2, the fecal coliform removal was in the range of 88-90%. The coliform removal
380 efficiency of phase 2 was comparatively lower than one obtained during phase 1. The reason for
381 lesser removal is due to the high pore size available in gravel medium compared to sand gravel
382 mix. In spite of higher removal efficiency, the coliform counts did not comply with the USEPA
383 standard limits for reuse. Therefore the treated water should be provided with little dose of
384 disinfectant before reused.

385

386 **3.2.5 Emerging contaminants**

387 **3.2.5.1 Sodium do-decyl sulphate**

388 Sodium do-decyl sulphate (SDS) is the most commonly used surfactant, and the removal
389 efficiency of this pollutant in GROW system varied between 85-96% (Fig. 8) SDS is considered

390 to be highly hydrophobic in nature having a log K_{ow} of 3.6 (Hansch et al., 1996). Generally, the
391 compounds that are hydrophobic (with high K_{ow}) values are removed from the system by
392 adsorption, hydrolysis and microbial degradation/ biosorption (Lv et al., 2016; Ramprasad and
393 Philip, 2016). The SDS in the treated water was in the range of 2.8-4.2 mg/L (60-80%) during
394 the start-up phase, and was improved in the following phases to 82-96%. It was also found that
395 the removal of SDS was affected by seasons. The seasonal variability was mainly attributed to
396 two main physical conditions, namely solar radiation and water temperature. Low temperatures
397 decreased the bio-degradation rates and low solar irradiation decreased the phyto degradation
398 rates (Simonich et al., 2002). In phase 1.3, the removal efficiency of SDS increased from 88% to
399 96% with decrease in flow rate and increased hydraulic retention enhanced the biodegradation of
400 adsorption of SDS. This is in good agreement with the results reported by Langford et al., 2005.
401 In phase 1.4, as the OLR increased from 25.5 g COD/ cubic. meter/ day to 26.8 g COD/ cubic.
402 meter/ day, the rate of SDS removal decreased from 92% to 85%. The presence of readily
403 available carbon source (sucrose) reduced the biodegradability of SDS as reported by Nyberg et
404 al., 1992. The statistical analysis confirms that, although the difference in the effluent SDS
405 concentrations is statistically significant ($p < 0.05$), it is relatively small. The SDS removal
406 efficiency after the change of plant species and filling media was reduced to 85-88%, due to the
407 availability of less adsorption space as the media sizes were larger.

408

409 ***3.2.5.2 Propylene Glycol and Tri Methyl amine***

410 The propylene glycol (PG) and tri-methyl amine (TMA) are commonly used in personal care
411 products likes soap and shampoos. PG and TMA are highly water soluble, have low log K_{ow}
412 value and are also easily biodegradable. It was reported by Avila et al., 2014, that the compounds

413 that are highly water soluble is predominantly taken up by plants / phyto-degraded and
414 biodegraded. In phase 1.1, the PG and TMA removal efficiency was in the range of 40-60%
415 (Fig. 9 and 10) due to the low density of plants and microbes in the system. During phase 1.2,
416 the, the removal efficiency was more during summer than in winter or monsoon. In phase 1.3, as
417 the flow rate increased, the removal efficiency decreased (96% to 80%), due to less retention
418 time. Also, when the OLR increased (25.5 to 26.8 g COD/ cubic. meter/ day) the removal
419 efficiency decreased from 94% to 86%, as the system exceeded the biodegradation capacity. Due
420 to addition of external carbon source, the degradation rates of organic pollutants were hindered.
421 Sucrose is a readily biodegradable compound than PG and TMA. Therefore, microbial consortia
422 would have utilized more sucrose as a carbon source than the target pollutant. As a result, lesser
423 biodegradation was observed for target pollutants with increase in OLR. Similar trend was
424 reported by other researchers also (Nyberg et al., 1992). During phase 2, the removal efficiency
425 did not change much from phase 1, as the mechanism for PG and TMA removal was mostly
426 plant uptake and biodegradation.

427

428 **4. Conclusion**

429 This study confirmed that shallow horizontal subsurface flow GROW system with 8 varieties of
430 native plant species can effectively improve quality of greywater in tropical countries. The
431 performance of the GROW system was monitored over a significant period of time at various
432 operating conditions. The removal efficiency obtained for various parameters were; biochemical
433 oxygen demand (BOD) 90.8%, chemical oxygen demand (COD) 92.5%, total suspended solids
434 (TSS) 91.6%, nitrate-nitrogen ($\text{NO}_3 - \text{N}$) 83.6%, total phosphate (TP) 87.9%, total nitrogen (TN)
435 91.7%, fecal coliform (FC) 91.4%, sodium do-decyl sulphate (SDS) 85.7%, propylene glycol

436 (PG) 93.4% and trimethyl amine (TMA) 88.9%. It was found that the removal rate was high
437 during summer season compared to other seasons. Also the removal efficiency was more at
438 higher HRT. The promising results from this study may increase the applicability of GROW
439 systems as a robust, cost-effective and reliable green roof systems in India and other tropical
440 countries.

441

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445

446 **References:**

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Removal of chemical and microbial contaminants from greywater using a novel constructed wetland GROW

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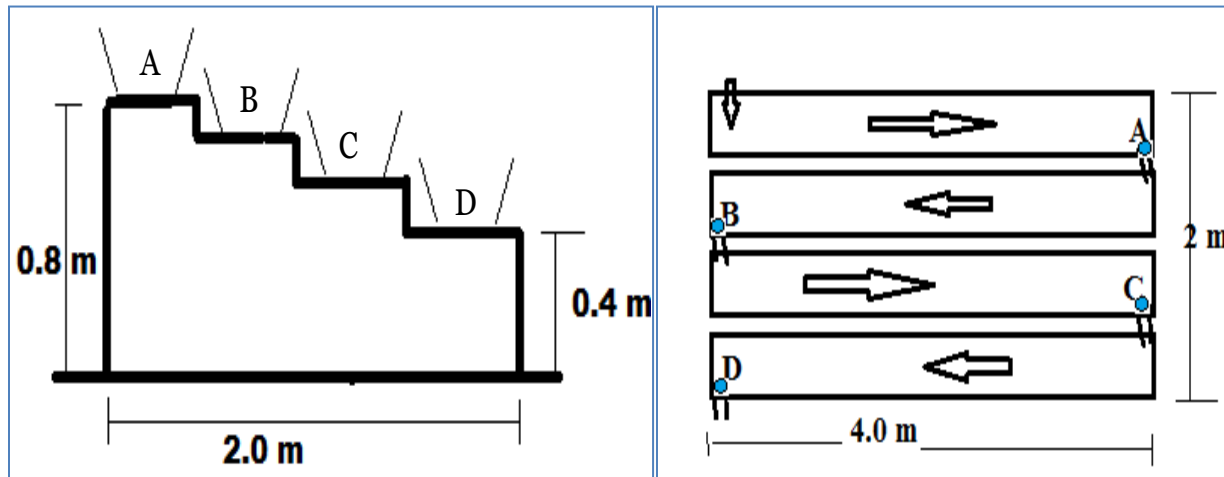


Fig. 1 Schematics of GROW system

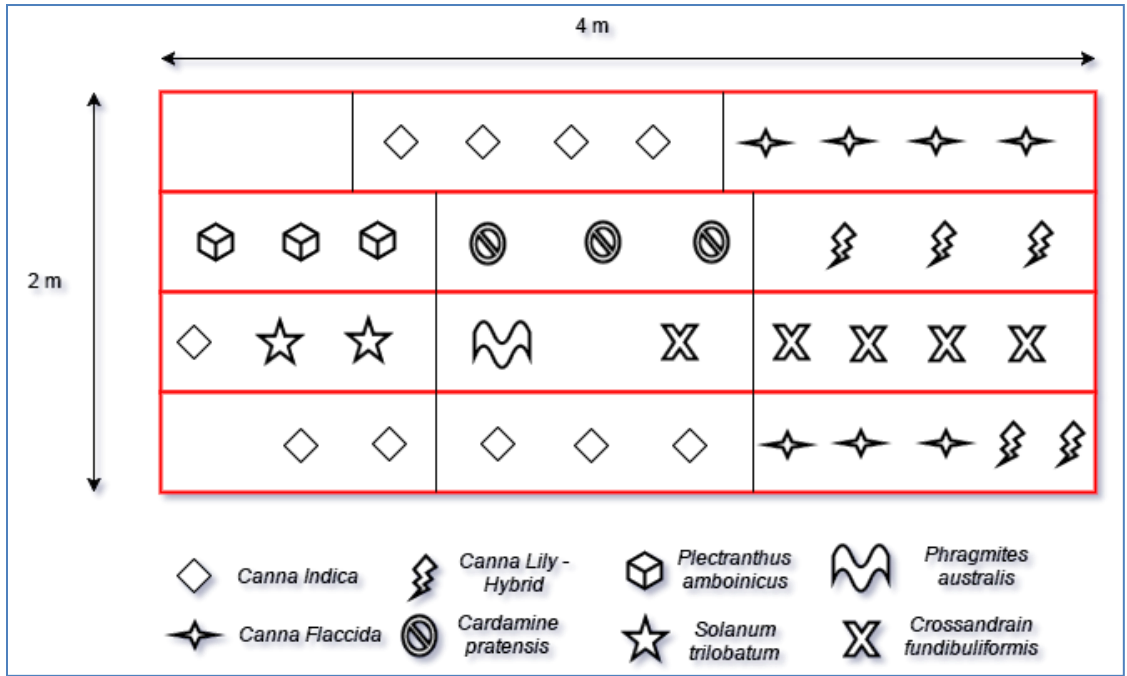


Fig. 2 Planting Plan of Each trough in GROW system

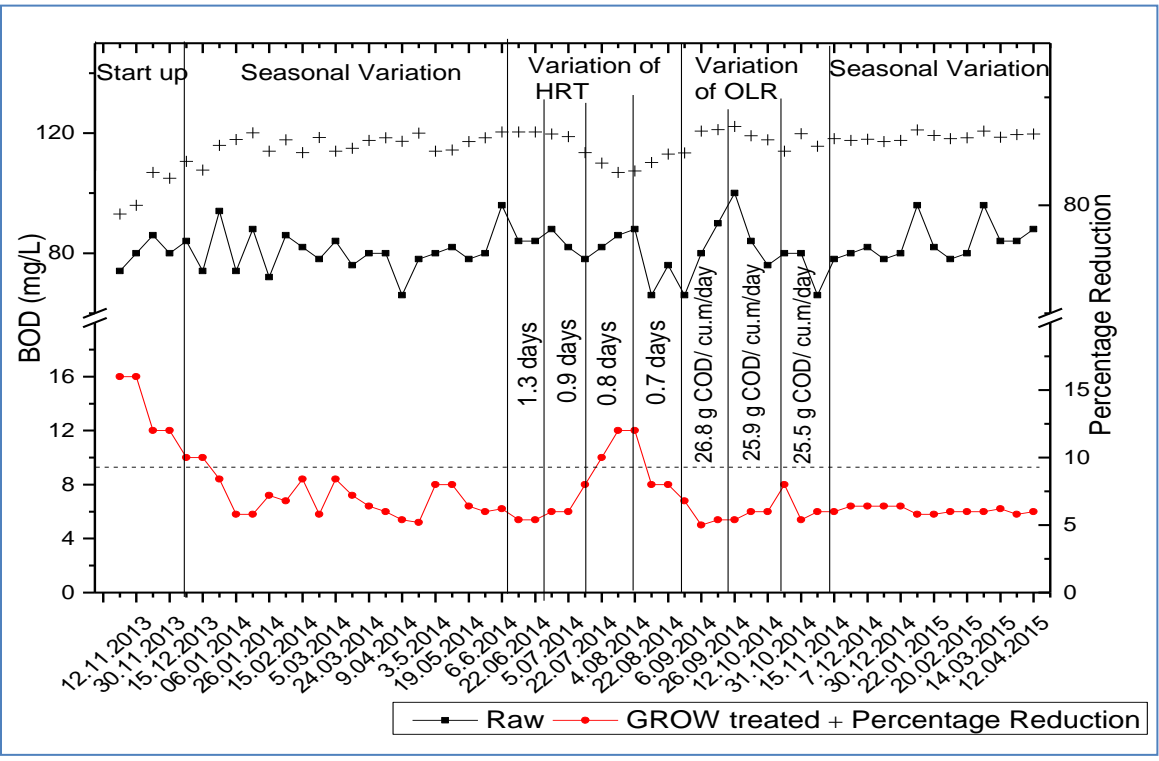


Fig. 3(a) Performance of GROW systems with respect to BOD removal during various operational conditions

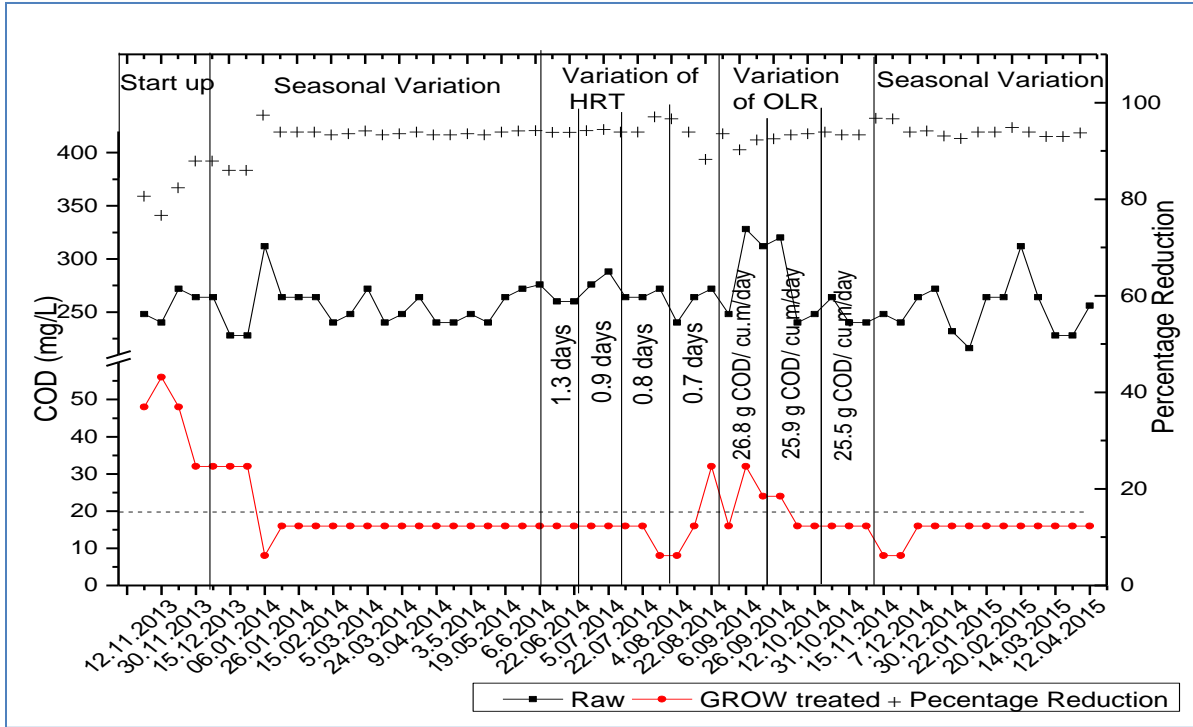


Fig. 3(b) Performance of GROW systems with respect to COD removal during various operational conditions

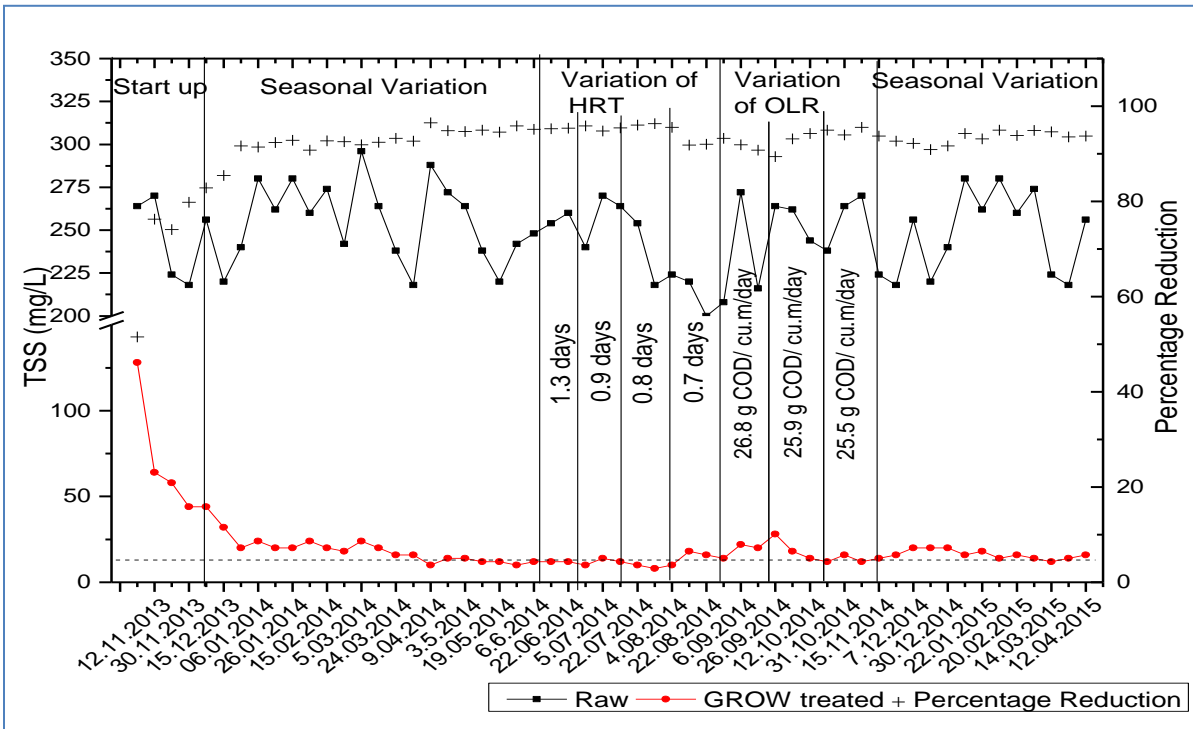


Fig. 4 Performance of GROW systems with respect to the total suspended solids removal during various operational conditions

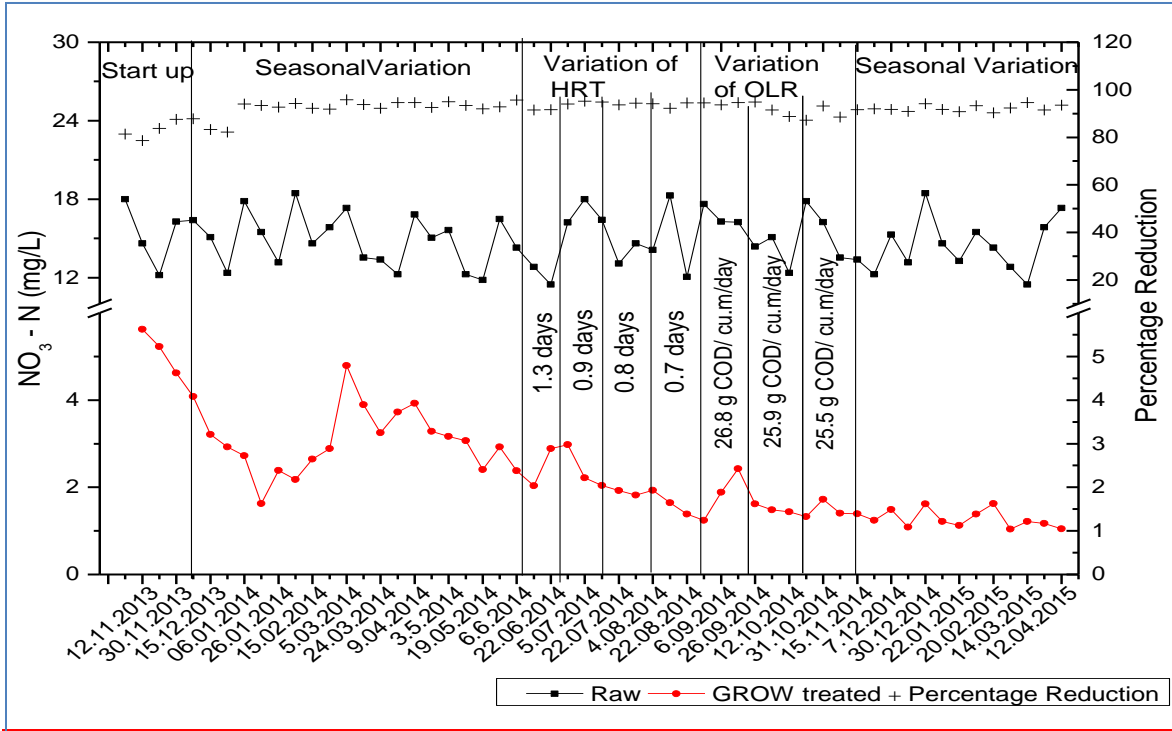


Fig. 5 Performance of GROW systems with respect to nitrate nitrogen removal during various operational conditions

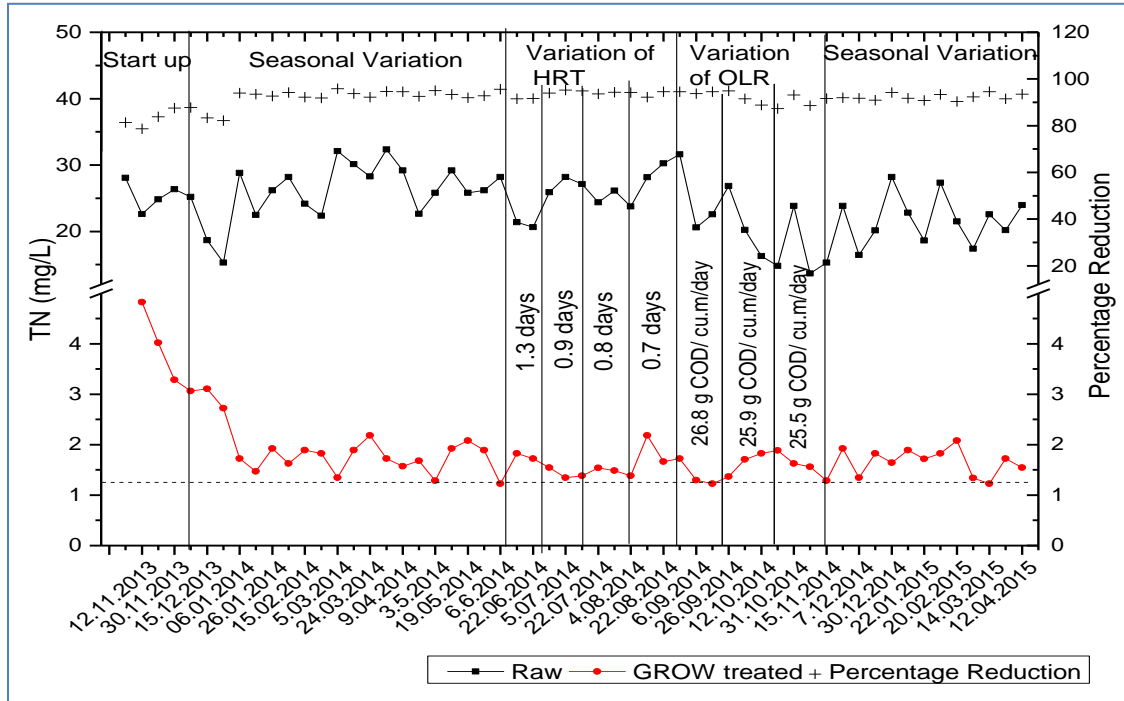


Fig. 65 Performance of GROW systems with respect to total nitrogen removal during various operational conditions

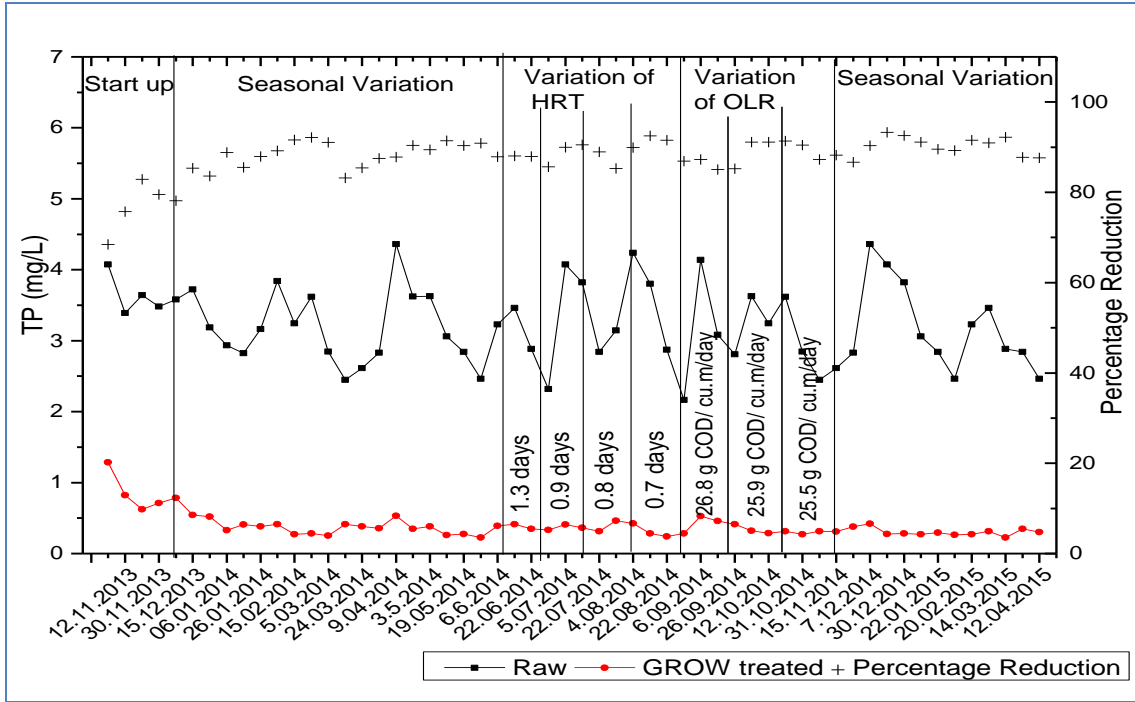


Fig. 7.6 Performance of GROW systems during with respect to total phosphate removal during various operational conditions

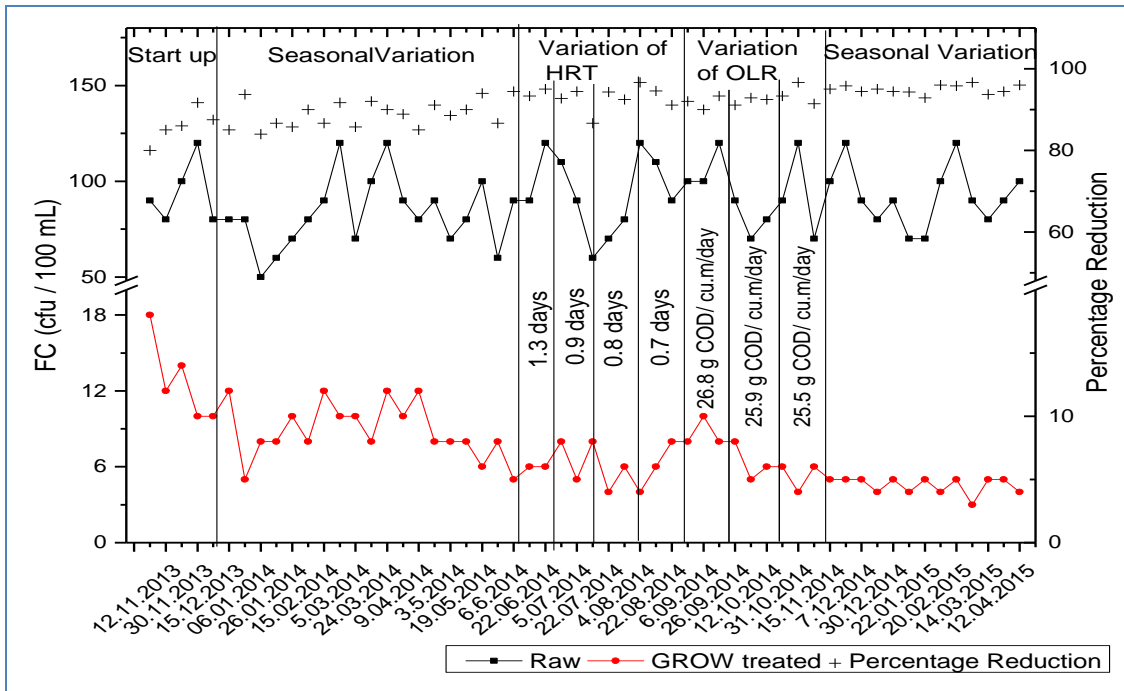


Fig. 7.8 Performance of GROW with respect to the fecal contamination removal during various operational conditions

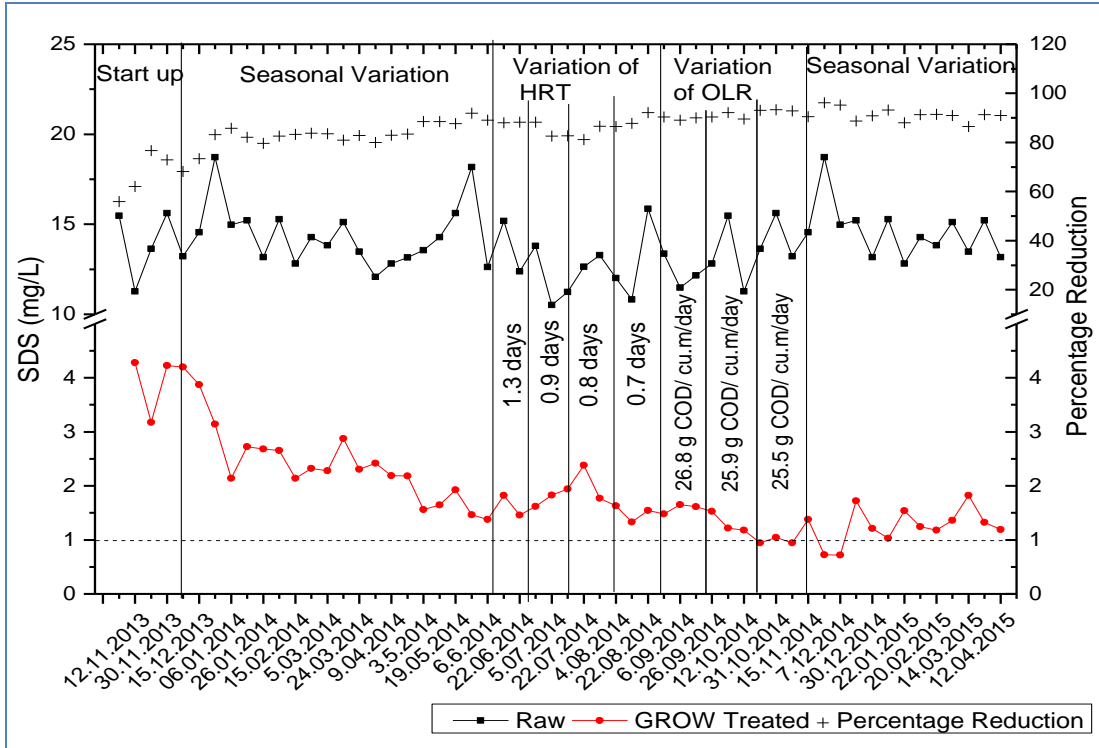


Fig. 89 Performance of GROW systems with respect to sodium do-decyl sulphate removal during various operational conditions

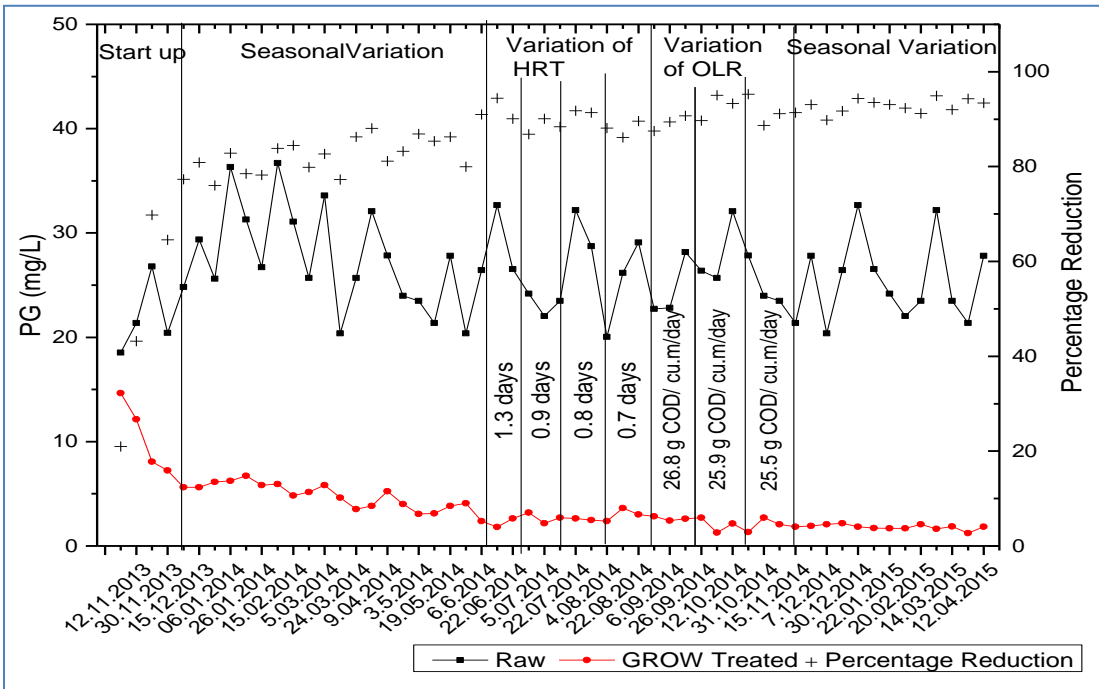


Fig. 910 Performance of GROW systems with respect to propylene glycol removal during various operational conditions

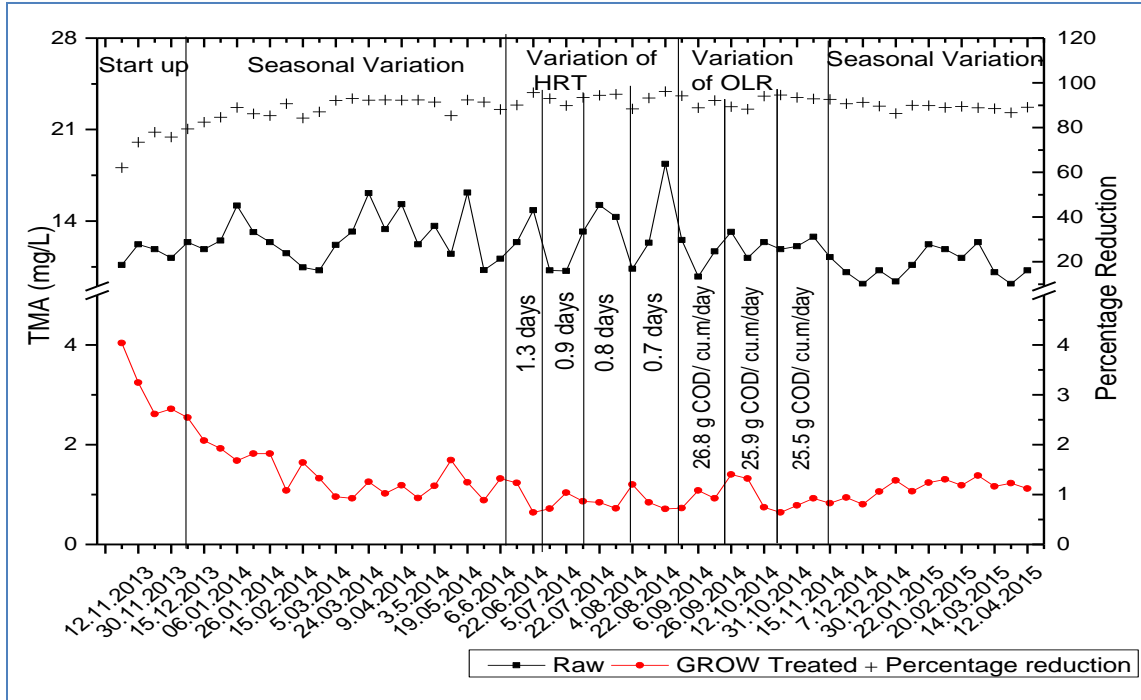


Fig. 11-10 Performance of GROW system with respect to tri-methyl amine removal during various operational conditions

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Table 1 Operating history for GROW constructed wetland

Months of operation	HRT (days)	HLR (L/ cu. m/ day)	OLR (g COD/ cu. m / day)
Start – up phase			
November to December, 2013	1.09	58.3	14.0
Performance Evaluation of GROW system			
Jan to June, 2014 and Nov – August 2015	1.09 – 1.22	58.3	14.0
Effect of Flow			
3 rd and 4 th week of July 2014	1.3-0.9	58.9	12.9
August 2014	0.7-0.8	53.1	14.9
Effect of additional organic loading			
September – October 2014	1.09	53.3	25.5 - 26.8

Table 2 Raw greywater characteristics

Parameters	Raw Greywater
pH	7.24 - 8.34
COD (mg/L)	216 - 320
BOD (mg/L)	68 - 120
TSS (mg/L)	240 - 280
TOC (mg/L)	23 -36.48
TN (mg/L)	17 - 28.82
NO ₃ – N (mg/L)	12.32 -17.84
TP (mg/L)	2.934 – 3.84
NH ₄ – N (mg/L)	10.28 -14.56
FC (CFU / 100 mL)	50 - 120
SDS (mg/L)	14.99 – 35.89
PG (mg/L)	11.58 - 46.59
TMA (mg/L)	8.67 - 15.54