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### **Research Paper**

# Use of full-scale hybrid horizontal tubular photobioreactors to process agricultural runoff



María Jesús García-Galán <sup>a,\*</sup>, Raquel Gutiérrez <sup>a</sup>, Enrica Uggetti <sup>a</sup>, Víctor Matamoros <sup>b</sup>, Joan García <sup>a</sup>, Ivet Ferrer <sup>a</sup>

<sup>a</sup> GEMMA — Environmental Engineering and Microbiology Research Group, Department of Hydraulic, Maritime and Environmental Engineering, Universitat Politècnica de Catalunya BarcelonaTech, c/Jordi Girona 1-3, Building D1, E-08034, Barcelona, Spain

<sup>b</sup> Department of Environmental Chemistry, IDAEA-CSIC, c/Jordi Girona, 18-26, E-08034, Barcelona, Spain

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Diffuse pollution in rural areas due to agricultural runoff is a widespread and difficult problem to address due to the large areas affected. Drainage channels receive polluted water, but its introduction into conventional treatment network is often unfeasible. Within this context, microalgae-based treatment systems could be used as alternative treatment plants. A new design of semi-closed (hybrid) tubular horizontal photobioreactor (HTH-PBR) with low energy requirements has been evaluated for microalgae cultivation at full-scale (8.5 m<sup>3</sup>), using agricultural runoff as feedstock. This novel system was tested in batch and continuous mode over 4 and 135 d. Considering a full-scale application in an agricultural context, a batch test was carried out to evaluate the performance of the system. An increase of 22% in the biomass concentration in 4 d was registered, and all nutrients were consumed during the first two days. In the continuous experiment carried out over winter (December–April), productivity was between 2 and 14 g g [TSS]  $m^{-3}$  d, but values up to 76.4 g [TSS]  $m^{-3}$  d were reached at the end of the study in spring, despite the low nutrients concentration in the feedstock. Elimination of emerging contaminants was also evaluated, obtaining the highest removals for the synthetic musk fragrances tonalide and galaxolide (73% and 68%), and the anti-inflammatory drug diclofenac (61%).

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#### 1. Introduction

Aquatic ecosystems constantly receive a broad range of organic micropollutants of anthropogenic origin, from both point sources such as wastewater treatment plant (WWTP) effluents (urban, rural or industrial) (García-Galán, Petrovic, Rodríguez-Mozaz, & Barceló, 2016; Gros, Rodríguez-Mozaz, &

\* Corresponding author. Fax: +34 934017357.

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Barceló, 2012; Verlicchi, Galletti, Petrovic, & Barceló, 2010) and non-point sources such as urban or agricultural runoff waters after strong rain events (Dolliver & Gupta, 2008a,b; Sabourin et al., 2009; Topp et al., 2008). The equilibrium of river and stream ecosystems if jeopardised if their capacity to attenuate and neutralise these inputs (dilution, biodegradation, etc) is overcome. Groundwater systems are also

E-mail address: chus.garcia@upc.edu (M.J. García-Galán).

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COD	Chamical arrigan damand (tatal)
	Chemical oxygen demand (total)
CODs	Chemical oxygen demand (soluble)
D	Tube diameter (m)
DO	Dissolved oxygen
DOM	Dissolved organic matter
EOCs	Emerging organic contaminants
HRAPs	0 0 1
HRT	Hydraulic retention time
HTH-PBR Hybrid tubular horizontal photobioreactor	
$\rm NH_4^+-N$	Ammoniacal nitrogen
$NO_3^N$	Nitrates
OC	Organic carbon
PBR	Photobioreactor
PEs	Population equivalents
PhACs	Pharmaceutical active compounds
$PO_4^{-3}-P$	Phosphates
PLC	Programmable logic controller
R <sub>e</sub>	Reynolds number
TS	Total solids
TSS	Total suspended solids
UPC	Universitat Politècnica de Catalunya
VS	Volatile solids
WWTP	Waste water treatment plant
ρ	Density of the liquor (m s <sup>-1</sup> )
υ	Velocity of the liquor (m $s^{-1}$ )
μ	Dynamic viscosity of the liquor (kg m <sup><math>-1</math></sup> s <sup><math>-1</math></sup> )
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Nomenclature

indirectly affected if polluted rivers feed aquifers by infiltration or via artificial surface recharge or via reclaimed water (Díaz-Cruz & Barceló, 2008).

Agricultural fields are probably the main source of diffuse pollution to both surface and groundwater systems in rural areas. Inorganic fertilisers, pesticides and farm yard manure, which is regarded as a very valuable fertiliser containing high amounts of nitrogen (N), phosphorus (P), potassium (K), organic carbon (OC), etc, are regularly applied to fields. These materials are usually highly soluble in order to easily reach roots. Thus agricultural runoff can contain a wide variety of contaminants including pesticides, heavy metals and nutrients. High levels of nitrates from fertilisers, together with some phosphate, have been detected for decades in groundwater as it eventually reaches surface waters such as rivers or lakes. Their presence leads to algae blooms that consume all the oxygen available and limit the sunlight reaching the benthic photosynthetic organisms, being toxic to freshwater life.

Intensive cattle farming is another relevant non-point pollution source, since a wide variety of veterinary pharmaceuticals (PhACs), mainly antibiotics, are used in the prevention and treatment of microbial infections. Depending on the drug, livestock can excrete high amounts of the administered dose as metabolites and also as the unchanged parent substance (Dolliver & Gupta, 2008a,b; Tong, Li, Wang, & Zhu, 2009; Wei, Ge, Huang, Chen, & Wang, 2011). The extensive use of manure from medicated animals in crop fields has been considered by different authors as one of the major routes by which veterinary antibiotics enter the environment. Once applied to top soil, these excreted residues can both percolate and reach groundwater bodies or reach surface waters during storm events (Dolliver & Gupta, 2008a,b; Kwon, 2011; Stoob, Singer, Mueller, Schwarzenbach, & Stamm, 2007; Watanabe, Bergamaschi, Loftin, Meyer, & Harter, 2010). Biosolids are applied as soil amendment and are an option favoured by governments for sludge management. This is because it results in the recycling of nutrients and it also improves soil structure (Edwards et al., 2009; Sabourin et al., 2009; Topp et al., 2008). Several studies have focused in the incomplete removal of organic and the emerging contaminants in WWTPs which are still present in effluents discharged into rivers, or, in the case of the more lipophilic compounds, found in digested sewage sludge. For instance musk, UV blockers, flame retardants, and bisphenol A (or triclosan) are frequently detected in biosolids (Clarke & Smith, 2011; Gago-Ferrero, Díaz-Cruz, & Barceló, 2011; Plagellat et al., 2006).

Drainage ditches or open irrigation channels can receive an important part of this agricultural runoff which finally discharges into rivers. Since these channels can also receive wastewater (treated or raw), particularly in underdeveloped countries, this can lead to the recycling of nutrients in crop fields (Christou, Karaolia, Hapeshi, Michael, & Fatta-Kassinos, 2017; Lees, Fitzsimons, Snape, Tappin, & Comber, 2016). However, when these channels discharge directly into rivers, potential contamination may be dispersed and affect a large number of non-target species. It is too costly to direct these drainage channels from agricultural fields into main collectors and towards WWTPs. Therefore, local and alternative treatments need be considered.

Recently great interest has been shown in wastewater treatment systems based on microalgae due to the ability of microalgae to remove nutrients, heavy metals and bacteria (Abdel-Raouf, Al-Homaidan, & Ibraheem, 2012; Muñoz & Guieysse, 2006). Microalgae can grow in low quality water, such as wastewaters, because wastewater still contain high amounts of nutrients, in particular N and P, which are essential for microalgae production (Pawar, 2016). Apart from the availability of light and water, C, N and P are essential for microalgal growth. The molar ratio of these three elements is determined by the Redfield ratio, that established that a proportion of 106:16:1 C:N:P is needed for the optimal growth of microalgae (Redfield, 1958). Microalgae carbon is derived from CO<sub>2</sub> (bacterial respiration and/or atmospheric exchange), nitrogen mainly from  $NH_4^+$ -N and phosphorus from  $PO_4^{-3}$ -P. In terms of the wastewaters likely to be found in rural areas, industrial wastewater effluents from the dairy or food industries, usually contain much higher concentrations of C, N & P than municipal wastewater (Pawar, 2016), making them more appropriate feedstocks for large scale algae cultivation. The use of rural wastewater as feedstock for microalgae biomass growth leads to a dual benefit; microalgae have proved to be highly efficient in removing these nutrients, producing clean water as a by-product, and at the same time the production of algal biomass increases; this biomass can be further processed and converted to bioenergy. Furthermore, it has been demonstrated that, under specific growing conditions, different added-value products can also be recovered from microalgae, such as glycogen or bioplastics (Arias,

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