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### Short Communication

# Influence of the seasonal variation of environmental conditions on biogas upgrading in an outdoors pilot scale high rate algal pond

David Marín<sup>a,b</sup>, Esther Posadas<sup>a</sup>, Patricia Cano<sup>a</sup>, Víctor Pérez<sup>a</sup>, Raquel Lebrero<sup>a</sup>, Raúl Muñoz<sup>a,\*</sup>

<sup>a</sup> Department of Chemical Engineering and Environmental Technology, School of Industrial Engineerings, Valladolid University, Dr. Mergelina, s/n, 47011 Valladolid, Spain
<sup>b</sup> Universidad Pedagógica Nacional Francisco Morazán, Boulevard Centroamérica, Tegucigalpa, Honduras

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## ABSTRACT

The influence of the daily and seasonal variations of environmental conditions on the quality of the upgraded biogas was evaluated in an outdoors pilot scale high rate algal pond (HRAP) interconnected to an external absorption column (AC) via a conical settler. The high alkalinity in the cultivation broth resulted in a constant biomethane composition during the day regardless of the monitored month, while the high algal-bacterial activity during spring and summer boosted a superior biomethane quality.  $CO_2$  concentrations in the upgraded biogas ranged from 0.1% in May to 11.6% in December, while a complete H<sub>2</sub>S removal was always achieved regardless of the month. A limited N<sub>2</sub> and O<sub>2</sub> stripping from the scrubbing cultivation broth was recorded in the upgraded biogas at a recycling liquid/biogas ratio in the AC of 1. Finally, CH<sub>4</sub> concentration in the upgraded biogas ranged from 85.6% in December to 99.6% in August.

#### 1. Introduction

Biogas from the anaerobic digestion of wastewaters and organic waste constitutes a renewable source of energy to generate electricity or heat (Muñoz et al., 2015). However, the use of biogas as a substitute of natural gas or fuel in transportation requires an effective purification to levels set by national regulations. For instance, biogas injection into natural gas grids typically requires concentrations of  $CH_4 \ge 95\%$ ,  $CO_2 \le 2\%$ ,  $O_2 \le 0.3\%$  and trace levels of  $H_2S$  (Muñoz et al., 2015; Toledo-Cervantes et al., 2017b).

Algal-bacterial processes have emerged as a platform technology capable of simultaneously removing  $CO_2$  and  $H_2S$  in a single stage, and constitute a cost-effective and environmentally friendly alternative to conventional biogas upgrading technologies (Bahr et al., 2014; Muñoz et al., 2015). Biogas upgrading in algal-bacterial photobioreactors is based on the oxidation of  $H_2S$  to  $SO_4^{2-}$  by sulfur oxidizing bacteria promoted by the high dissolved oxygen (DO) concentrations in the scrubbing cultivation broth, and on the photosynthetic fixation of the absorbed  $CO_2$  by microalgae. The economic and environmental sustainability of this biotechnology can be boosted via digestate supplementation as a nutrient and water source, which will support an effective recovery of nutrients in the form of algal-bacterial biomass (Posadas et al., 2017; Toledo-Cervantes et al., 2016).

Biogas upgrading coupled to digestate treatment has been typically evaluated indoors in high rate algal ponds (HRAPs) interconnected to biogas absorption columns (AC) under artificial illumination (Alcántara et al., 2015; Bahr et al., 2014; Meier et al., 2015; Posadas et al., 2016, 2015; Serejo et al., 2015; Toledo-cervantes et al., 2017a; Toledo-Cervantes et al., 2017b, 2016). The optimization of this process has reached promising results in terms of biomethane quality (CH<sub>4</sub> concentrations of 96.2  $\pm$  0.7%), nutrient removal (total nitrogen (TN)removal efficiencies (REs) of 98.0  $\pm$  1.0% and P-PO<sub>4</sub><sup>-3</sup>- REs of 100  $\pm$  0.5%) and biomass productivities (15.0 g m<sup>-2</sup> d<sup>-1</sup>) (Toledo-Cervantes et al., 2017b). Comparable results were also obtained by Posadas et al. (2017) in a similar biogas upgrading photobioreactor configuration operated outdoors during summer in Spain, when solar irradiation, temperature and the number of sun hours were most favorable to support algal-bacterial activity. In this context, a systematic year-round evaluation of the influence of the daily and seasonal variations of environmental conditions on biogas upgrading and nutrient recovery from digestate is needed to validate this technology under outdoor conditions.

This study investigated for the first time the year-round performance of biogas upgrading in an outdoors pilot HRAP interconnected to an external AC by monthly monitoring the daily variations of biogas quality and cultivation broth parameters under continental climate conditions.

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<sup>\*</sup> Corresponding author. E-mail address: mutora@iq.uva.es (R. Muñoz).

#### D. Marín et al

#### 2. Materials and methods

#### 2.1. Biogas and centrate

A synthetic biogas mixture composed of CO<sub>2</sub> (29.5%), H<sub>2</sub>S (0.5%) and CH<sub>4</sub> (70%) was used as a raw biogas in the present study (Abello Linde; Spain). Centrate was monthly obtained from the centrifuges dehydrating the anaerobically digested mixed sludge of Valladolid wastewater treatment plant (WWTP) and stored at 4 °C. The composition of centrate varied along the experimental period as a result of the seasonal operational variations of the WWTP: total organic carbon (TOC) = 16–523 mg L<sup>-1</sup>, inorganic carbon (IC) = 450–600 mg L<sup>-1</sup>, TN = 374–718 mg L<sup>-1</sup>,  $P-PO_4^{3-} = 26-135 mg L^{-1}$  and  $SO_4^{2-} = 0-38 mg L^{-1}$ . The IC concentration in the centrate was adjusted to 1999 ± 26 mg L<sup>-1</sup> via addition of NaHCO<sub>3</sub> and Na<sub>2</sub>CO<sub>3</sub> in order to maintain the required high alkalinity and pHs (≥9) in the cultivation broth to support an effective CO<sub>2</sub> and H<sub>2</sub>S absorption in the AC (Posadas et al., 2017).

#### 2.2. Experimental set-up

The experimental set-up, constructed according to Posadas et al. (2017), was located outdoors at the Department of Chemical Engineering and Environmental Technology of Valladolid University (41.39° N, 4.44° W). The pilot plant consisted of a 180 L HRAP with an illuminated area of  $1.20 \text{ m}^2$  (width = 82 cm; length = 170 cm; depth = 15 cm) and two water channels divided by a central wall and baffles in each side of the curvature. The internal recirculation velocity of the cultivation broth in the HRAP was  $\approx 20 \text{ cm s}^{-1}$ , which was supported by the continuous rotation of a 6-blade paddlewheel. The HRAP was interconnected to an external 2.5 L bubble AC (height = 165 cm; internal diameter = 4.4 cm) provided with a metallic biogas diffuser of 2 µm pore size located at the bottom of the column. The HRAP and the AC were interconnected via an external liquid recirculation of the algal-bacterial cultivation broth from an 8 L conical settler (Fig. 1). The efficiency of the settler in terms of biomass removal was almost complete.

#### 2.3. Operational conditions and sampling procedures

Process operation was carried out from November the 1st 2016 to October the 30st 2017. The HRAP was inoculated to an initial concentration of 210 mg TSS  $L^{-1}$  with a microalgae inoculum composed of *Leptolyngbya lagerheimii (54%), Chlorella vulgaris (28%), Parachlorella* 

kessleri (9%), Tetrademus obliquus (5%) and Chlorella minutissima (2%) from an indoor HRAP treating biogas and centrate at the Department of Chemical Engineering and Environmental Technology of Valladolid University (Spain). Five different operational stages (namely I, II, III, IV and V) were defined as a function of the temperature, photosynthetic active radiation (PAR), number of sun hours and biomass productivity imposed (Table 1). The synthetic biogas was sparged into the AC under co-current flow operation at 74.9 Ld<sup>-1</sup> under a recycling liquid to biogas ratio (L/G) of 1.0 according to Posadas et al. (2017), which resulted in gas and liquid retention time of 48 min and. The liquid velocity accounted for 2 m h<sup>-1</sup>. The HRAP was fed with IC-supplemented centrate as a nutrient source at a flow rate of 3.5 L d<sup>-1</sup>, which entailed a hydraulic retention time of 50 d. Tap water was supplied in order to compensate water evaporation losses and allow process operation without effluent (Table 1).

The pH, temperature and DO concentration in the cultivation broth of the HRAP, AC and settler, along with PAR, were monitored every thirty minutes during the daytime of one day every month where the environmental conditions were representative of the conditions in the entire month. Gas samples of 100  $\mu$ L from the upgraded biogas were drawn every hour to monitor the gas concentrations of CH<sub>4</sub>, CO<sub>2</sub>, H<sub>2</sub>S, O<sub>2</sub> and N<sub>2</sub>. Liquid samples of 100 mL from the cultivation broth of the HRAP, AC and settler were drawn every two hours to monitor the concentrations of dissolved TOC, IC, TN.

#### 2.4. Analytical procedures

PAR was measured using a LI-250A light meter (LI-COR Biosciences, Germany), while pH was determined with an Eutech Cyberscan pH 510 (Eutech instruments, The Netherlands). Temperature and DO were measured using an OXI 330i oximeter (WTW, Germany). Gas concentrations of CH<sub>4</sub>, CO<sub>2</sub>, H<sub>2</sub>S, O<sub>2</sub> and N<sub>2</sub> were determined using a Varian CP-3800 GC-TCD according to Posadas et al. (2015) (Palo Alto, USA). Dissolved TOC, IC and TN concentrations were measured using a Shimadzu TOC-VCSH analyzer (Japan) coupled with a TNM-1 chemiluminescence module.

#### 3. Results and discussion

#### 3.1. Biogas upgrading

#### 3.1.1. $CO_2$ biomethane concentration

Negligible variations in CO<sub>2</sub> concentration in the biomethane were

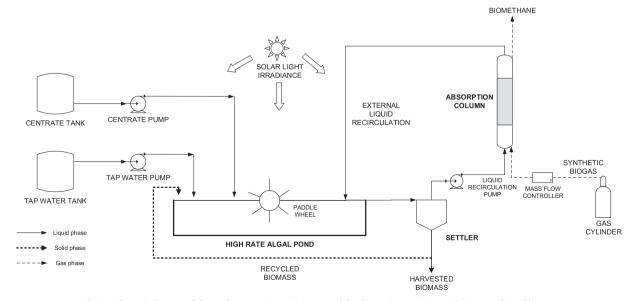


Fig. 1. Schematic diagram of the outdoors experimental set-up used for the continuous photosynthetic upgrading of biogas.

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