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Applicability of Anaerobic Digestion Model No. 1 (ADM1) for a specific industrial wastewater: Opium alkaloid effluents

Recep Kaan Dereli, Mustafa Evren Ersahin*, Hale Ozgun, Izzet Ozturk, Ali Fuat Aydin

Department of Environmental Engineering, Civil Engineering Faculty, Istanbul Technical University, 34469 Maslak, Istanbul, Turkey

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ABSTRACT

Opium alkaloid processing industries are quite limited throughout the world since cultivation and processing of opium are not practiced in most of the developed countries. However, wastewater generated from this industry is known to be highly polluted with considerable environmental impacts. High rate anaerobic digestion processes are effective in the treatment of opium alkaloid wastewaters that are characterized with relatively low pH and high soluble COD content. Experimental results obtained from the anaerobic treatability studies conducted with lab-scale upflow anaerobic sludge bed reactor (UASB) were simulated by the IWA Anaerobic Digestion Model No. 1 (ADM1). A model based influent characterization which provided consistent results was performed. The validation results indicated that the calibrated ADM1 was able to predict the experimental results of effluent COD, pH, methane and biogas flows with reasonable accuracy. This study demonstrates a useful approach for the implementation of the ADM1 for the treatment of a specific industrial wastewater.

1. Introduction

One of the oldest medicinal plants in recorded history is the opium poppy, *Papaver somniferum* L. (Papaveraceae) which has an annual herb with an erect stem, having a white, red, or purplish flower depending on the cultivar. The use of the plant as a soothing agent in medicinal purposes resulted in its spreading out through different cultures including ancient Sumeria, Rome, China and Europe for centuries. Presently, poppy straw is principally produced in India, Australia, France, Spain, Ukraine, Yugoslavia, and Turkey. Opium which has an alkaloid content of approximately 10–20% contains more than 40 individual alkaloids such as morphine, codeine, thebaine and narcotine [1].

Effluent from opium processing industries is known as high strength wastewater with a relatively low pH. Wastewater from opium processing industries has high COD concentration mainly of soluble and biodegradable character, with an initial inert COD content of less than 5%. The soluble COD content of wastewater consists of mainly acetic acid [2,3].

Information on characteristics, treatment and disposal of effluents from the opium alkaloids factories are quite limited since cultivation and processing of opium are not commonly practiced in the world. Hence, only a few studies have been reported for the treatment of opium alkaloid industry wastewaters such as chemical coagulation and biological treatment [2,4]; physico-chemical treatment [5]; anaerobic treatment [3], post-treatment of aerobically treated opium alkaloid effluents with lime and ozone [6,7]; activated sludge and catalytic wet air oxidation [8,9], post-treatment by fenton oxidation [10] and membrane treatment as a post-treatment alternative [11].

Fundamental models of various complexities describing anaerobic digestion process have been developed during the last four decades. Early models were very simple and assumed a rate limiting step [12,13]. However, the increasing knowledge on anaerobic digestion and the interactions of the multiple functional species involved in it require more complex models to be developed [14,15]. Furthermore, the recent developments in the computer technology stimulated the development of dynamic process models and they allowed to simulate the impact of the changing environmental conditions on complex biological treatment systems. Anaerobic Digestion Model No. 1 (ADM1), developed by the IWA Task Group for Mathematical Modeling on Anaerobic Digestion, is one of the most sophisticated and complex model involving 19 biochemical processes and two types of physiochemical processes [16].

Although ADM1 was principally developed for anaerobic digestion of wastewater sludges, its generic structure allows modeling of anaerobic processes for industrial wastewater treatment [17–20]. Rajinikanth et al. [21] applied the ADM1 and a modified version of ADM1 to the results obtained from a lab-scale hybrid upflow anaerobic sludge-filter bed reactor treating wine distillery vinasse wastewater. They concluded that the modified ADM1 was able to simulate well the dynamic behavior of the main variables in the liquid and gas phases. ADM1 was also successfully implemented for

* Corresponding author. Tel.: +90 212 285 3776; fax: +90 212 285 6545.
E-mail address: ersahin@itu.edu.tr (M.E. Ersahin).

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simulating mesophilic and thermophilic anaerobic co-digestion of olive mill wastewater with olive mill solid waste in semi continuous tubular reactors under different operating conditions [22,23]. Chen et al. [24] successfully applied ADM1 for the simulation of a two stage treatment system consisting of a completely mixed reactor and an UASB reactor treating medicine production effluents.

Treatment of industrial wastewaters such as opium processing effluents will benefit from direct implementation of mathematical models for control, operation and optimization of full-scale treatment plants and assisting the transfer of modeling studies to industry. The present work was undertaken to apply ADM1 for simulating and analyzing the experimental results obtained from the anaerobic treatment of opium alkaloid wastewaters in a lab-scale upflow anaerobic sludge bed reactor (UASBR) at mesophilic (35 °C) conditions. Thus, the results obtained from this study present an example for application of a structured model for the treatment of a specific industrial wastewater.

2. Materials and methods

2.1. Experimental methods

The analysis of various parameters such as chemical oxygen demand (COD), biochemical oxygen demand (BOD), total kjeldahl nitrogen (TKN), total phosphorus, total suspended solids (TSS), total volatile solids (TVS) and total solids (TS) were performed according to Standard Methods [25]. pH was measured with Thermo Orion 720A pH meter. Volatile fatty acids (VFAs) were measured by liquid chromatography (HPLC 1100 equipped with Mbondapak C18 column). An appropriate mobile phase (H_2SO_4 solution at pH 3) was determined for VFA analysis and applied to all samples. Biogas flow was measured with Ritter wet type gas meter. Biogas composition was determined with Orsat type gas analyzer (Borucam 1-310-100) by measuring the CO_2 fraction. Soluble and particulate inert COD of the wastewater under anaerobic conditions were determined according to Germirli et al. [26] by using two anaerobic reactors operated for raw and filtered wastewater at 35 °C.

2.2. Wastewater characterization

The wastewater used in the study was generated from an opium processing industry located in Afyon Karahisar Province in Turkey. The wastewater characterization of opium processing industry was provided from the study of Aydin et al. [3] in Table 1. The process effluent characterized by its high strength and biodegradability had a relatively low pH. Soluble COD content and acetic acid related COD of the wastewater were 90% and 33%, respectively. The soluble and particulate inert COD fractions of the wastewater were experimentally determined as 1.64% and 2.42% of influent COD, respectively [3].

Table 1
Characterization of opium processing industry wastewater [3].

Parameter	Unit	Values
Total COD	mg/L	18,300–42,500
Soluble COD	mg/L	17,050–39,470
TOC	mg/L	7335–14,000
BOD_5	mg/L	4250–22,215
pH	–	4.9–6.3
Total alkalinity	mg $CaCO_3$/L	315–4450
TS	mg/L	27,235–29,750
TSS	mg/L	565–2295
TVS	mg/L	320–1775
TKN	mg/L	550–841
NH_3–N	mg/L	73–141
Total phosphorus	mg/L	3.1–15.0
Acetic acid	mg/L	3730–13,630

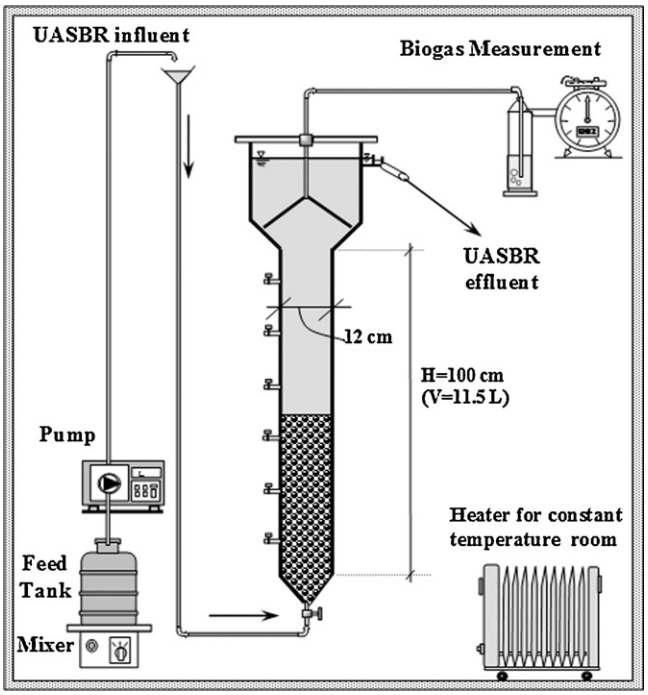

Fig. 1. Lab-scale UASBR set-up.

2.3. Lab-scale UASBR

An UASBR made of polyvinyl chloride (PVC) with a height of 1 m and an inner diameter of 12 cm was used for lab-scale studies (Fig. 1). The effective volume of the reactor was 11.5 L and it was operated at mesophilic conditions (35 ± 2 °C). The reactor was continuously fed by a peristaltic pump and the produced biogas was measured by a wet gas meter. The lab-scale UASBR was inoculated with granular sludge taken from a full-scale UASBR treating brewery industry wastewater. The TVS content of the seed sludge was 88%.

C:N:P ratio of the feed was adjusted to 300:5:1 by the addition of phosphoric acid. Trace elements were added into the feed periodically to supply the requirement of micronutrients necessary for the biomass growth. Trace elements Fe, Ni, Co, K, Zn and Mo were added into the feed at rates of 1.0, 0.2, 0.1, 100.0, 0.1 and 0.5 mg/L reactor-day, respectively [27].

The operational period of UASBR was divided into 15 phases due to the changes in hydraulic retention time (HRT) and organic loading rate (OLR). Table 2 presents the operating conditions of each phase. Lab-scale UASBR was operated at OLRs ranging from 3.40 to 10.0 kg COD/m^3 day and at HRTs changing between 1.62 and 1.15 days. More information about the reactor performance and operation can be found elsewhere [3].

2.4. Model implementation

ADM1 was implemented in Aquasim 2.1b which is a computer program for mathematical modeling and simulation of aquatic systems [28]. ADM1 requires a detailed characterization of COD in a specific form identifying the concentrations of soluble and particulate carbohydrates, protein, lipids, as well as individual VFAs. Since analysis of all these individual variables are generally not practical, at least not on a regular basis, reasonable approximations can be made depending on the available characterization of the raw material and wastewater (COD, TOC, TKN, NH_4–N) in the industry [20,29]. The studies about the constituents of the raw material "opium" presented that it is composed of approximately 5–20%

Table 2
Operating conditions of the experimental study [3].

Period number	Monitoring period (days)	Influent COD (mg/L)	Effluent COD (mg/L)	Hydraulic retention time θ (days)	Organic loading rate OLR (kg COD/m^3 day)	COD removal efficiency E (%)
C1[a]	40	5500	700	1.62	3.40	87.2
C2	32	6000	700	1.62	3.70	88.3
C3	29	8100	1130	1.62	5.00	86.0
C4	17	12,150	1950	1.62	7.50	84.0
C5	25	14,500	2610	1.62	9.00	82.0
V1[a]	15	5950	695	1.52	3.91	88.3
V2	17	7200	965	1.52	4.75	86.6
V3	13	10,600	1820	1.52	7.00	82.8
V4	13	12,500	2275	1.52	8.22	81.8
V5	15	15,200	2920	1.52	10.00	80.8
V6	16	13,200	2770	1.32	10.00	79.0
V7	13	6900	1310	1.15	6.00	81.0
V8	18	9800	2050	1.15	8.50	79.0
V9	40	11,500	2650	1.15	10.00	77.0
V10	17	11,700	1860	1.62	7.22	84.1

[a] C and V stand for calibration and verification periods, respectively.

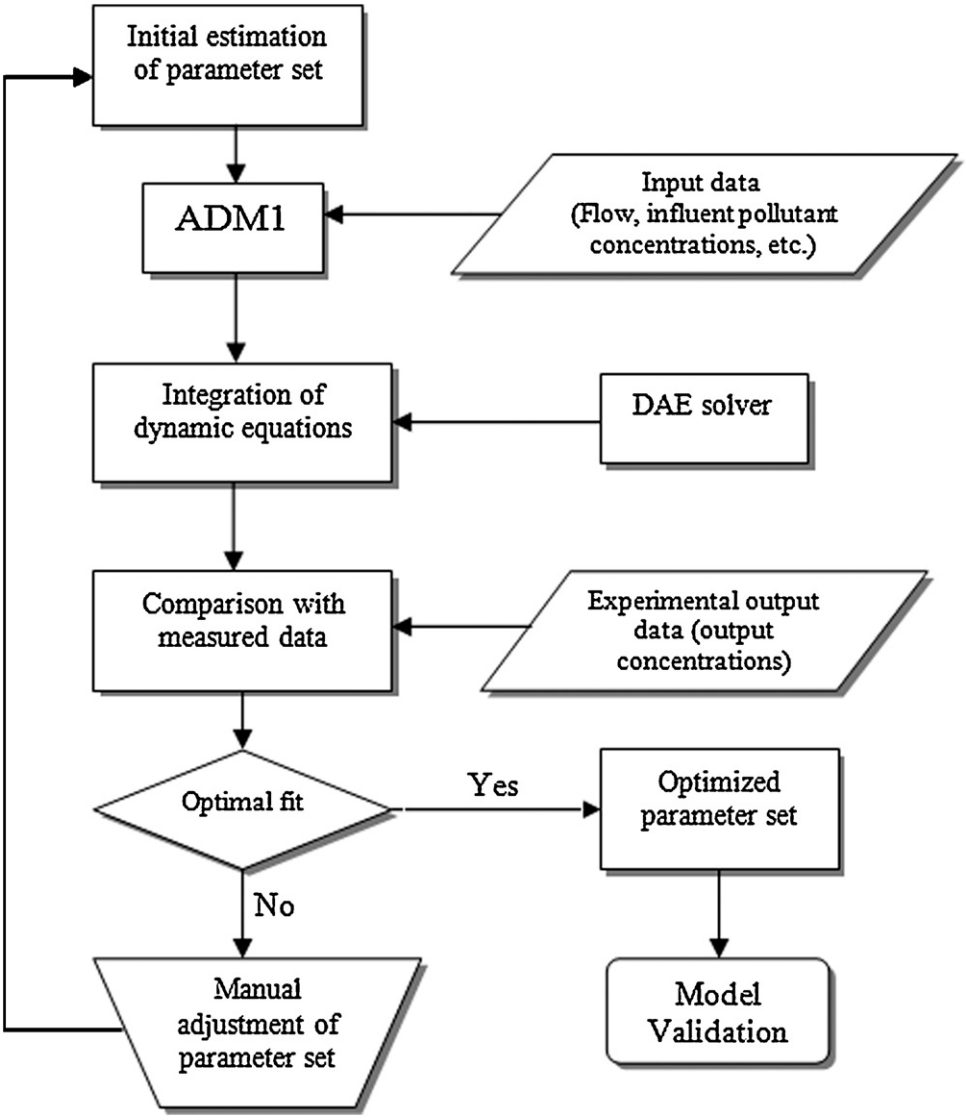

Fig. 2. Calibration algorithm used in the study.

water, about 20% various sugars, and several simple organic acids such as fumaric acid, lactic acid, oxaloacetic acid, and meconic acid [1]. An approximation of the influent wastewater characterization was made by using an algorithm offered by Kleerebezem and Van Loosdrecht [29]. The algorithm provides an initial estimate of COD fractions according to ADM1 by using minimum number of measured parameters such as total COD, total organic carbon, organic nitrogen and VFA alkalinity. The algorithm does not make an estimation of the inert fractions. Therefore, the inert fractions were determined by experimental methods. The model based COD fractionation of the wastewater from the opium processing industry was provided in Table 3.

As the COD content of the wastewater entering to the UASB reactor is readily biodegradable and rapidly hydrolysable and also composed of acetic acid, amino acid and monosaccharide as presented in Table 3, it is not necessary to apply disintegration step in ADM modeling. Therefore, disintegration step was only used to fractionate the particulate COD which constituted the lower portion (7%) of the influent COD and to represent the lysis of biomass.

Most of the stochiometric coefficients and kinetic parameters of the original ADM1 model were used without any changes [16].

Table 3
Model based influent wastewater characterization.

Stochiometric parameters	Definition	Value (%)
Solubles		
$f_{Sac,CODt}$	Acetic acid fraction of total COD	33.00
$f_{Saa,CODt}$	Amino acid fraction of total COD	18.10
$f_{Ssu,CODt}$	Sugar fraction of total COD	29.62
$f_{Sfa,CODt}$	LCFA fraction of total COD	10.64
$f_{SI,CODt}$	Soluble inert fraction of total COD	1.64
Particulates		
$f_{XIi,CODt}$	Lipid fraction of total COD	1.21
$f_{Xch,CODt}$	Carbohydrate fraction of total COD	3.37
$f_{XI,CODt}$	Particulate inert fraction of total COD	2.42

Table 4
Initial and calibrated values of kinetic parameters.

Kinetic parameters	Names	Units	Initial values	Calibrated values
k_{dec}	Decay constant	1/day	0.02	0.05
$k_{m,ac}$	Maximum uptake rate for acetate utilizers	COD/COD day	8	4
$k_{m,aa}$	Maximum uptake rate for amino acid utilizers	COD/COD day	50	40
$k_{m,c4}$	Maximum uptake rate for valerate/butyrate utilizers	COD/COD day	20	13
$k_{m,fa}$	Maximum uptake rate for fatty acid utilizers	COD/COD day	6	4
$k_{m,h2}$	Maximum uptake rate for hydrogen utilizers	COD/COD day	35	25
$k_{m,su}$	Maximum uptake rate for sugar utilizers	COD/COD day	30	20
$K_{s,ac}$	Half saturation constant for acetate utilizers	kg COD/m^3	0.15	0.3
$K_{s,fa}$	Half saturation constant for fatty acid utilizers	kg COD/m^3	0.4	0.6
K_{l,NH_3}	Ammonia inhibition constant for acetate utilizers	M	0.0018	0.002

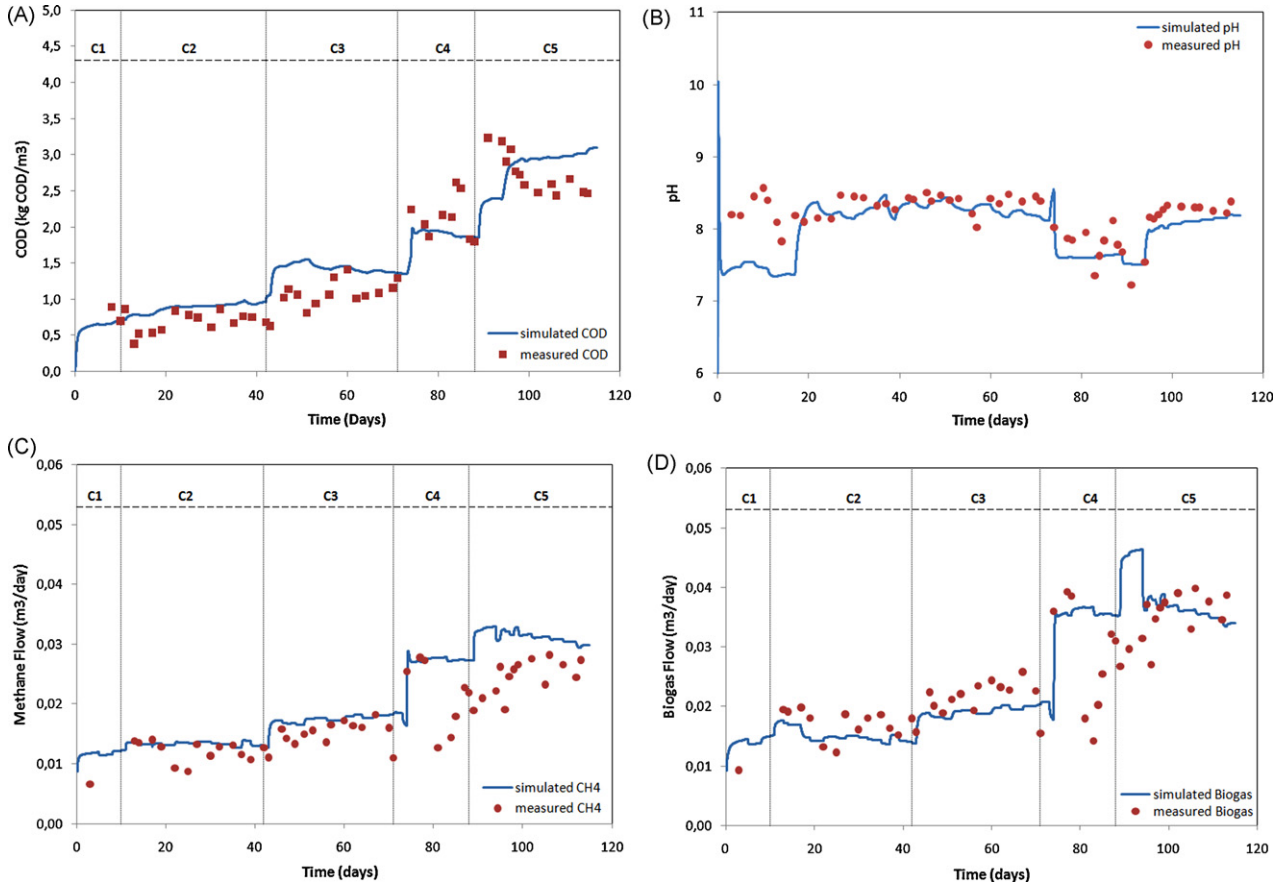

Fig. 3. (A) Effluent COD, (B) pH, (C) methane flow, and (D) biogas flow levels in comparison with experimental data after model calibration.

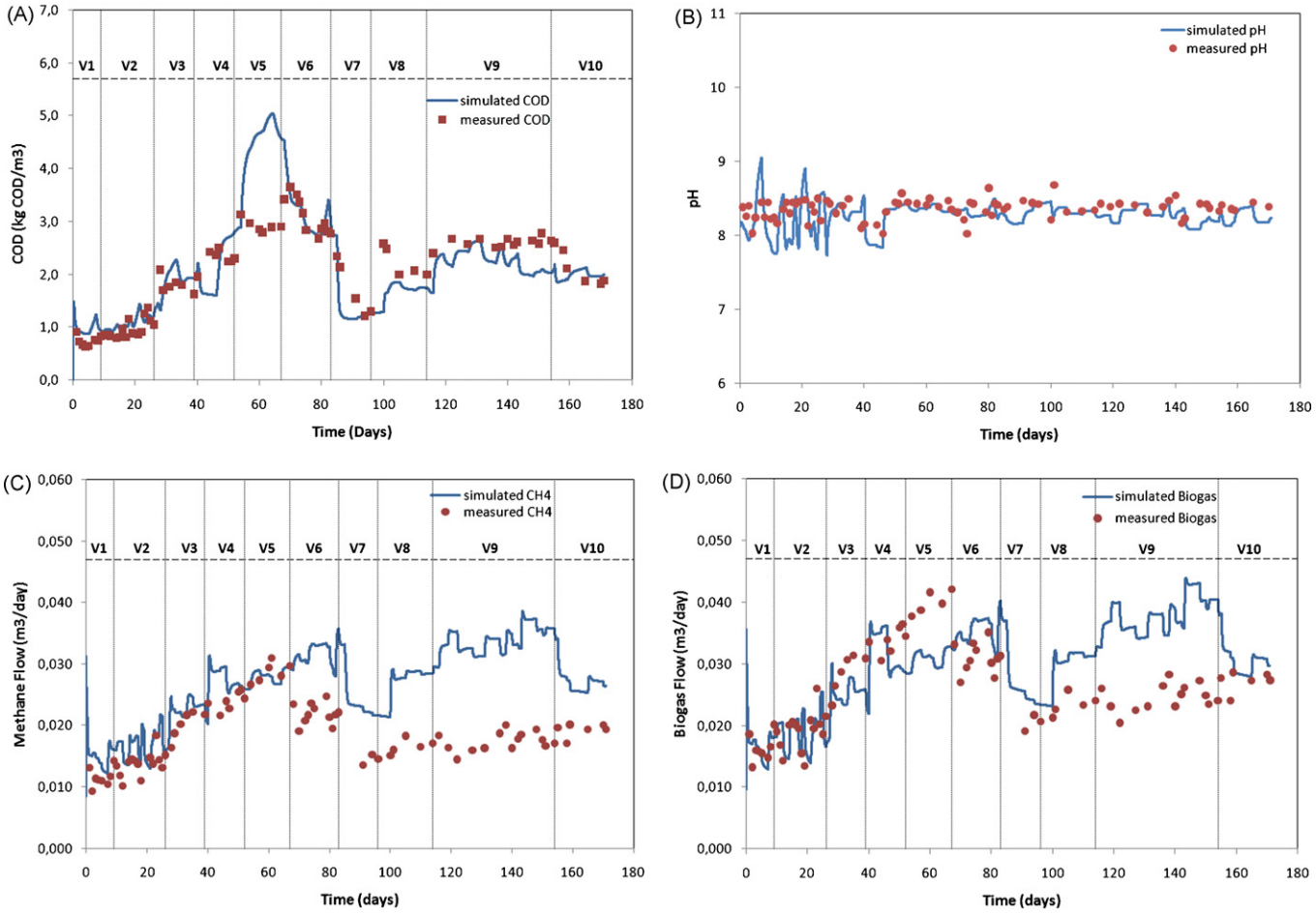

Fig. 4. Validation of model outputs with experimental data: (A) effluent COD, (B) pH, (C) methane flow, and (D) biogas flow.

The other kinetic parameters, modified by the calibration of the model simulations with the experimental results, were provided in Table 4.

3. Results and discussion

3.1. Model calibration

The model was calibrated by using 113 days dynamic data covering C1–C5 periods. During these periods, the volumetric loading rate of the UASBR was gradually increased from 3.4 to 9.0 kg COD/m^3 day at a constant HRT of 1.62 days. An iterative method was applied for the calibration of the most sensitive parameters to fit the model outputs to the experimental results (effluent COD, pH, biogas and methane flows). The used iterative procedure can be seen in Fig. 2. The estimated parameter values providing the best fitting between model outputs and experimental results were given in Table 4. The influent wastewater characterization makes the kinetic coefficients of the biomass types that degrade acetate, amino acid, monosaccharide and fatty acids more important in comparison to the rest of the parameters. Therefore, the most sensitive parameters that have significant effects on the ADM1 model outputs were identified as $k_{m,ac}$, K_{I,NH_3}, k_{dis}, k_{dec} [30], $k_{m,pr}$, $k_{m,c4}$, k_La [31]. Other parameters, except from Table 4, have been used without any modification as suggested by Batstone et al. [16].

Decay constant (k_{dec}) was found to have a great impact on the simulation outputs. Batstone [32] indicated that higher decay rates were more valid for the modeling of anaerobic mixed and biofilm systems. Thus, the decay rate in this study was increased to the value of 0.05 day^{-1} which was found to be consistent. Since the wastewater had high acetic acid content, the model outputs such as effluent COD and methane flow were mostly affected from the kinetic parameters of acetate utilizers. Therefore, $k_{m,ac}$ and $K_{s,ac}$ values were changed to 4 COD/COD day and 0.3 kg COD/m^3, respectively in order to obtain the best fit. Although the ammonia concentrations in the influent were low compared to the other waste types such as slaughterhouse effluents or primary sludge, the model simulated a very significant unionized ammonia inhibiton for acetate utilizers due to the high operating pH of the reactor. This effect was reduced by increasing the value of K_{I,NH_3} to 0.002 M.

The model calibration results for effluent COD, pH, biogas and methane flow by using the optimized parameter set were presented in Fig. 3. Effluent COD, biogas and methane flows were predicted quite well for relatively low organic loading (3.40–5 kg COD/m^3 day: C1–C3 periods) conditions. The model predicted the adaptation of the system adequately during the periodical changes. However, the accuracy of the model prediction decreased with the increase in the organic loading rate (7.50–9 kg COD/m^3 day: C4–C5 periods). It was seen that the model simulated an overload situation for methane and biogas flows. It was difficult to further calibrate the model parameters to get better simulation results, and a complete replication of measured data by the model for all loading periods could not be obtained. This might arise from complication of applying ADM1 for a lab-scale reactor which has relatively lower tolerance to changes in operating conditions in comparison to full-scale systems. Besides, similar results can be found elsewhere [33]. Finally, ADM1 simulated pH variations with high accuracy for all periods.

3.2. Model validation

A validation study was performed to assess the quality and applicability of the optimized parameters and influent characterization values. The model outputs were compared with measured data obtained in V1–V10 period from the investigated UASBR without changing the previously optimized parameter set. During these periods, OLR and HRT of the UASBR were applied between 3.91–10.0 kg COD/m^3 day and 1.15–1.62 days, respectively. Varied HRT values were applied in the periods covering validation study unlike the periods of the calibration study.

Fig. 4 presents the comparison of model outputs and experimental data for effluent COD, pH, methane and biogas flows. As can be seen from the Fig. 4, COD and pH were reasonably good predicted by the model (except V5 period for COD). A good estimation of influent cation and anion concentrations resulted in the prediction of pH measurements with high accuracy. The mean absolute relative error for COD prediction was calculated as 20% which can be regarded as medium accuracy [17]. Although the model predicted an overload situation for COD in V5 period, the lab-scale reactor could maintain the high OLR values without losing its treatment efficiency probably due to the acclimation of biomass. Also methane flow was quite well predicted until the period of V6 and biogas flow was adequately simulated until the period of V8. Methane and biogas flows were over predicted in V6–V10 periods and V8–V10 periods, respectively.

4. Conclusions

The study revealed the applicability of ADM1 for opium processing effluents in the prediction of effluent characteristics under a wide range of operational conditions. The remarkable discrepancies between simulations and biogas and methane flows in the latter periods of the experimental study are attributed to a systematic error related to the gas flow measurement system rather than the weaknesses of ADM1 model in the prediction of biogas flow at overload situations as mentioned in other studies [20,23,34,35]. Moreover, adequate results provided for effluent COD values through the whole experimental study and consistency of it with biogas and methane flows during the former periods justified this argument.

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