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Removal of Emerging Contaminants from Municipal Wastewater Using a Compartmentalized Anaerobic Baffled Reactor: Effect of Organic Loading Rate

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THIS STUDY deals with the occurrence and removal efficiency of polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) as selected persistent organic pollutants (POPs)in municipal wastewater via a compartmentalized anaerobic baffled reactor (CABR). The treatment processes and removal efficiencies using CABR were assessed with application of three hydraulic retention times (8, 16 and 24 hrs). The detected concentration values for PAHs in raw wastewater were varied from 345 to 1960.1 with an average value of 1003.5 µg/l. While those for PCBs were 2.3 to 18.7 with an average value of 8.7µg/l.The removal efficiencies for PAHsachieved 52, 81 and 86%, whereas for PCBs they recorded 41, 67 and 77% at HRT of 8, 16 and 24 hrs, respectively. POPs contributor to surface water bodies in Egypt and better reduction efficiencies are achievable, the present study highlights the possibility of utilizing CABR for restoring water quality for reuse or safely discharging on the streams.

Keywords: Compartmentalized Anaerobic Baffled Reactor, Treatment, Wastewater, OLR, PAHs, PCBs.

Introduction

The explosive increase of the Egyptian population and industrial activities during the last decades lead to a considerable increase in the variability and complexity of wastewaters, that contain a large amount of toxic persistent organic pollutants (POPS). These compounds combine with domestic wastewater and go through sewerage system. Many of these POPs are very toxic even at low concentration levels with the effect of accumulation through the conventional biological wastewater treatment systems. Environmental occurrence of (POPs) is a worldwide rather than a regional problem, due to the formation of chlorinated compounds that used heavily in the tropical regions and certainly have bad impact on ecosystem. Chlorinated biphenyls are belonging to POPs group that primarily used in many industrial activities [1,2]. POPs can be derived as a component of agricultural drainage, urban runoff including wet and dry deposition from the atmosphere and via the contribution of industrial discharges into the sewerage system. Polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) are among the most toxic contaminates derived from POPs. These pollutants have been reported to cause a number of hazardous effects including carcinogenic, immunologic, tetratogenic and reproductive problems in organisms. Moreover, some congeners have shown some endocrine disrupting effects [2-5]. The fate of these contaminants in wastewater treatment plants (WWTPs) will be governed by process design, physico-chemical properties and operating conditions at the treatment system [6]. WWTPs are widely recognized as one of the most important sources of contaminants to the surface water and aquatic environment. Therefore, there is

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a growing need for controlling their effluents [7,8]. Among the developed modern high-rate anaerobic treatment systems, the compartmentalized anaerobic baffled reactor (CABR) showing promising for domestic wastewater treatment. The CABR was described as a series of up-flow anaerobic sludge bed blankets (UASBs). This design consisted of a number of vertical baffles to allow wastewater to flow up and down through a number of compartments containing mixed anaerobic bacteria for treatment [9,10]. One of the most important advantages of CABR that it has a simple design and maintains a high void volume without the need for further filter media and has no special sludge separation systems. The anaerobic baffled reactor is easy to build and easy to operate. Moreover, it has good solids retention, and requires low maintenance attentions. Four- and five chamber CABRs showed a slightly more efficient in converting solids and biogas compared with the two- and three-chamber CABRs. It has been proved that hydraulic and organic shock loads have little effect on the efficiency of treatment. The anaerobic baffled reactor avoids the problem of reactor clogging and sludge bed expansion like other biological treatment systems [1,10,11]. The most significant advantage of the CABR over all biological wastewater treatment systems is its ability to separate acidogenesis and methanogenesis longitudinal way down to the reactor. This character allows the different anaerobes to grow under the most favorable conditions, and consequently the reactor can behave as a two-phase system without the associated high cost-control Problems. So, the main theme of this work is to develop an efficient treatment system that can be implemented for domestic wastewater containing persistent organic contaminants [10,11].

Materials and Methods

A treatability study was carried out to assess the treatment efficiency of municipal wastewater using a compartmentalized anaerobic baffled reactor (CABR). To achieve the aim of this study, CABR simulating laboratory scale model was designed and manufactured. The treatment system was continuously fed with municipal wastewater from the public sewage network through a connection from the sewerage system. The influent raw wastewater was settled down in a settling tank for suspended solids minimization then fed to CABR treatment system that operated outdoors at ambient temperature. Sampling was carried out during winter season on basis of three times per week (n=48).

Compartmentalized anaerobic baffled reactor (CABR): The CABR treatment system was manufactured from a Perspex material. It contained a series of vertical baffles that divided the reactor into five identical compartments. Anaerobic flocculent sludge from a sewage treatment plant in Cairo was inoculated in the reactor. The content of the flocculent sludge was maintained at around 15 g VSS/l. The reactor total liquid volume of the reactor was about 15l. The dimensions of the reactor were as follows; 62 cm length, 15 cm width, and 17 cm heights. The CABR schematic diagram is presented in Fig. 1. The CABR was carried out at different hydraulic retention times (HRT), and consequently different organic loading rates (OLR), in order to reach the optimum operating conditions of the CABR treatment system. The operating conditions of the CABR can be shown in Table 1.

Chemicals, extraction and clean up: Reagents, solvents and chemicals as n-hexane, dichloromethane, ethanol and acetonitrile were purchased from Sigma and Alliance Bio, USA. Pesticide residue (PR) grade and standards were supplied from Supelco. Inc. One liter wastewater samples were extracted twice using dichloromethane via liquid-liquid extraction technique [12]. For PAHs, the collected samples (1-L) were liquid-liquid extracted using dichloromethane and then fractionated and cleaned up through 20 cm chromatographic column which filling with 12 cm of alumina over 6 cm of silica gel and 2 cm of anhydrous Na_2SO_4 on the top. Firstly, the aliphatic fraction was eluted with 50 ml of n-hexane, and then the aromatic fraction was eluted next with 30 ml of 10% dichloromethane followed by 20 ml of 20% dichloromethane. The collected fractions were concentrated to 1 ml prior to quantification by GC-FID coupled with capillary column HP-1A ultra1 methyl siloxane (30m x 0.2 mm ID, 0.33 um film thickness). For PCBs, 1-L of wastewater sample were liquid-liquid extracted using n-hexane and

TABLE 1. Operating conditions of the anaerobic baffled reactor

HRT (h)	8	16	24
HLR (m3/m3/day)	3	1.5	1
OLR (kg COD/m3/day)	1.9	1.1	0.8

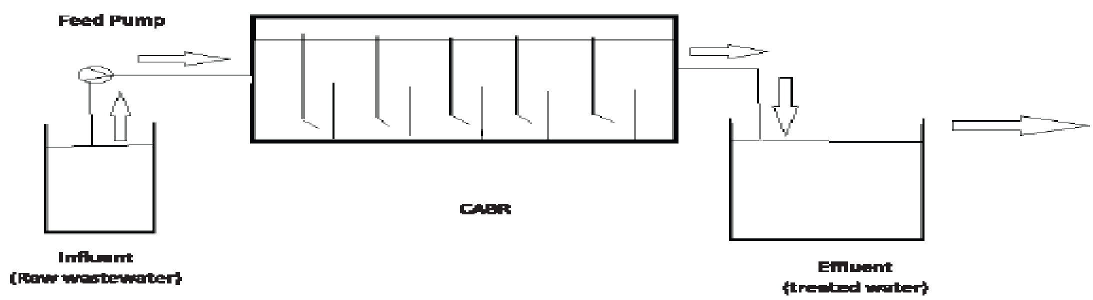

Fig. 1. Schematic diagram of CABR.

cleaned-up through silica gel chromatographic column. The extracted samples were cleaned up and fractionated through pouring about 20 g of activated florisil (0.54%) to column. The column was eluted with 60 mL of 30 % ethylene chloride in n-hexane to obtain the first fraction. However, the second fraction was obtained throughout the gradient elution of dichloromethane in hexane with 30 mL of 30 % and 50 ml of 50%, respectively. Then, fractions were concentrated using rotary evaporator until reach the volume of 1–2 mL. Finally, the fractioned samples were injected to gas chromatograph with mass spectra (GC-Ms) [4,13,14].

Instrumentation: GC-FID was calibrated with PAHs standard of EPA 610 PAH Mixture, containing naphthalene (Nap), acenaphthalene (Ace), fluorine (Fle), anthracene (Ant), phenanthrene (Phe), fluoranthene (Frt), pyrene (Pyr), benz [a] anthacene (BaA), chrysene (Chr), benzo [b] fluoranthene (BbF), benzo [k] fluoranthene (BkF), benzo [a] pyrene (BaP), dibenz [a,h] anthracene (DBA), benzo [ghi] perylene (BghiPe) and indo [1,2,3-cd] pyrene (IDP). The collected fractions of PCBs were quantified through the injection in gas chromatograph with mass spectra (GC-Ms, Agilent, Folsom, CA) operated in a splitless mode. The florisil column capillary of 30 m length × 0.25 mm internal diameter × 0.25 µm film thickness, Agilent). The initial temperature of the column was initially set at 180 °C for 2 min, then raised to 220 °C for 1 min, finally raised to 280 °C for 30 min. Carrier and make-up gas was nitrogen (99.999 %) was used at flow rate of 4 mL/min. GC system was calibrated with PCBs standard contained 10 PCBs congener (PCB) congeners (C28, C44, C52, C70, C101, C105, C118, C138, C153 and C180). The PCBs and PAHs limit of detection (LOD) were determined by the signal to noise ratio (S/N). The obtained recoveries of the analyzed samples were ranged between 52 and 86 and from 41 to 77 % for PAHs and PCBs, respectively.

Results and Discussion

The anaerobic baffled reactor compartments efficiency: In the CABR, a series of vertical baffles forces the wastewater to flow up and down them as it passes from inlet to outlet. This configuration has been shown to result in a high degree of organic pollutant removal. The main advantage of using a CABR comes from its compartmentalized structure. One of the CABR mechanisms is to remove solids by settling as sludge. The amount of solids accumulated in each ig.1compartment during this study at the different HRT indicated that the sludge accumulation is directly proportional to the increase of OLR and decrease of the HRT. Compartment-wise, the sludge accumulation was found to be highest in the first compartment and least in the fifth compartment. The CABR operating conditions of OLR are 1.9, 1.1 and 0.8 (kg COD/m^3/day) and for HLR are 3, 1.5 and 1.0 (m^3/m^3/day) at HRT of 8, 16 and 24 hrs, respectively, as shown in Table 1.

PAHs concentrations and removal efficiency: The concentration levels of poly cyclic aromatic hydrocarbon (PAHs) in raw and treated wastewater at various detention times applied on the treatment system of compartmentalized anaerobic baffled reactor (CABR) are presented in Table 2. Performance of CABR in this study was investigated using three organic loading rates, ranging between 0.67 and 2.1 kg COD/m^3/day. The results of monitoring the performance of the CABR at HRT of 8, 16, and 24 hrs indicated a higher efficiency of PAHs removal at 24 hrs and go down in a descending manner until reached the minimum at 8 hrs as represented in Fig. 2. The total concentration of PAHs in raw sample is ranged from 345 to 1960.1 µg/l with mean concentration 1003.5 µg/l, these values were much higher than those obtained by [1,8,15,16]. Since, we have no wastewater separation and consequently the effect of petrogenic and pyrogenic contaminations. The most abundant PAHs compounds in raw samples are LMW-compounds represented in 2 and 3 ring PAHs followed by the HMW-compounds represented in 4 and 5 ring PAHs with mean concentration value of 241.4 and 762.1 µg/l, respectively. This is most likely due to volatilization for the LMW PAHs (i.e., ≤3 rings) and strong sorption for the HMW species (i.e., 4–6 rings). The most abundant PAHs in the influent were flouranthene and naphthalene (464.9 – 142.3 ng/L), which accounted for more than 61% of ∑PAHs (1003.5 ng/L). The overall removal values for 24, 16 and 8 hrs, were 86, 81 and 52% respectively. Moreover, the removal efficiencies for HMW-PAHs were higher than those for LMW-PAHs at different retention times as shown in Fig. 3. The high levels of these compounds may be attributed to their use as precursor and a key starting material for various industrial productions [5,8,17]. The removal percent (R %) of organic pollutants was calculated using the general following formula:

$$R\ \% = ((C_{inf} - C_{eff})*100)/C_{inf}$$

Where C_{inf} is the concentration of pollutant in influent and C_{eff} is the concentration of pollutant effluent of individual treatment stage or the whole treatment process respectively. The treatment removal efficiency (% R) of CABR at different hydraulic retention times (HRT) revealed that the highest removal efficiency was achieved at HRT of 24 followed by 16 then 8 hrs for both LMW and HMW PAH compounds. as shown in Fig. 4. During the biological treatment of CABR, wastewater was fully mixed within contained sludge and then allow for the anoxic-anaerobic-aerobic (Inverted A^2/O) process. Removal of PAHs was mostly attributed to the microorganisms' biological activities. The reduction mechanisms include adsorption-precipitation, biodegradation and volatilization [14]. PAHs removal by volatilization mechanism was less than 2% in conventional activated sludge process, while the biodegradation effect may also be minor in the absence of microbes selected for PAHs degradation. The observed overall removal in CABR is comparable to those reported in earlier studies[10,11].

PCBs concentrations and removal efficiency: The measured concentration of PCBs in wastewater samples is presented in Table 3; 12 PCB congeners were detected in raw samples.

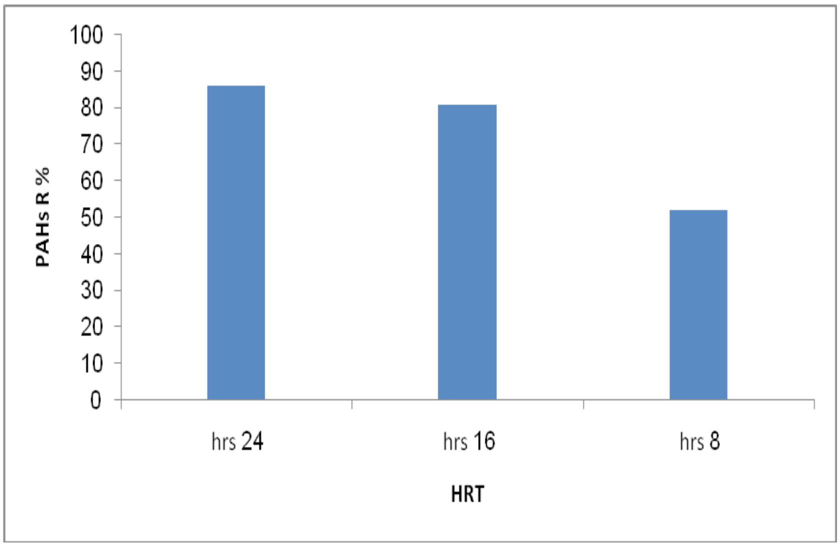

Fig. 2. The removal efficiency of PAHs via CABR at different HRT.

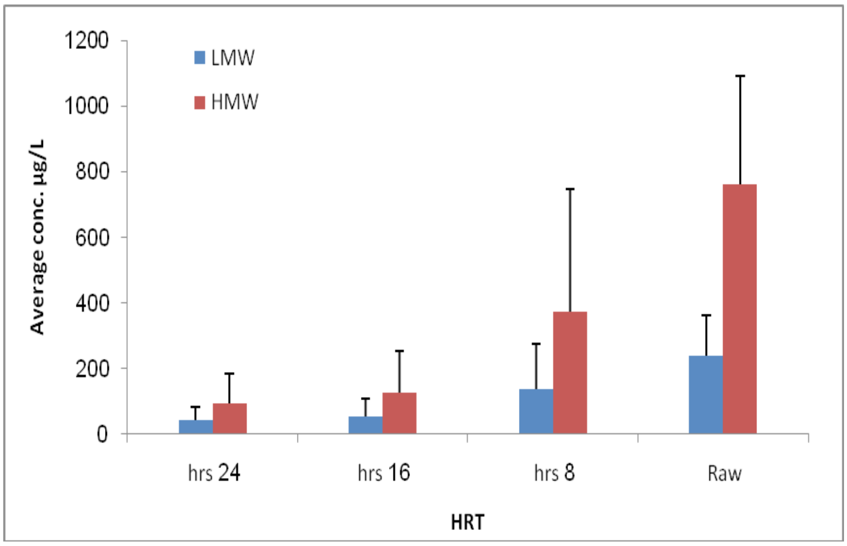

Fig. 3. Average concentration values of raw and treated LMW and HMW PAHs via CABRat different HRT.

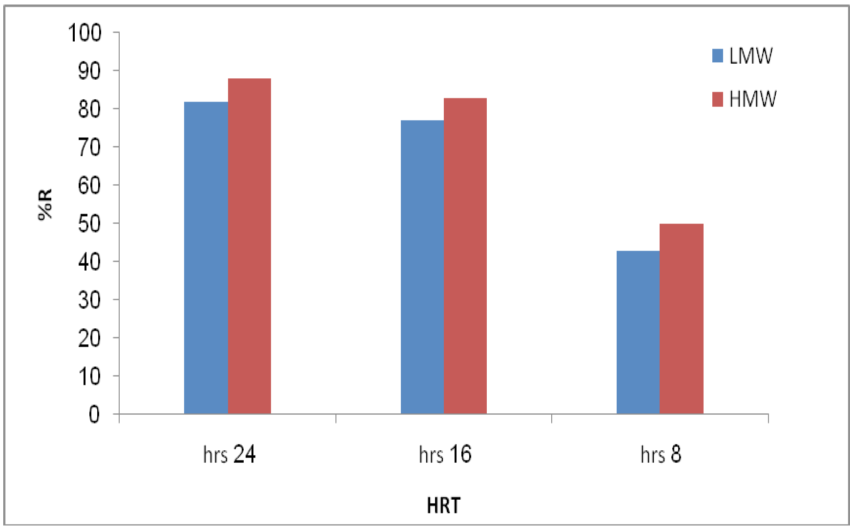

Fig. 4. CABR removal efficiencies of LMW and HMW PAHs at different HRT.

The average concentration levels of PCB congeners in raw wastewater samples are ranged from 2.3 to 18.7 µg/l with average concentration of 8.7 µg/l. These values are higher than those measured by other authors [5, 7, 8, and 9]. The most abundant concentrations of the individual PCBs in raw samples are C-52, C-18 followed by C-44 with values of 1.7, 1.5 and 1.0 µg/l, respectively. Whereas, the PCBs total average concentration values resulted from the treatment system (CABR) at different HRT are 5.1, 2.2 and 1.9 µg/l for 8, 16 and 24 hrs, respectively. These values are higher than those previously reported and the literature values of ΣPCBs in treated wastewaters vary largely depending on influent concentration [3,13,18]. The occurrence of PCBs are mostly similar to that in raw samples, whereas the removal efficiencies for PCBs congeners in the treatment system (CABR) at different HRTs are varied from 41, 67 and 77% for 12, 16 and 24 hrs respectively, as shown in Fig. 5. The removal efficiency of PCBs in CABR treatment system was increased following the order of 8 < 16 < 24 hrs, because PCBs undergo degradation by biological treatment, This removal efficiency of PCBs was in good agreement with literature values [13,18].

TABLE 2. Concentration levels of PAHs in raw and treated wastewater using ABR at different HRT

Ring No.	PAH (µg/l)	Raw Min	Raw Max	Raw Average	8 hrs Min	8 hrs Max	8 hrs Average	16 hrs Min	16 hrs Max	16 hrs Average	24 hrs Min	24 hrs Max	24 hrs Average
2-ring	**Nap**	44.8	297.6	142.3 ± 77	34.2	189.6	89.3± 43.2	14.1	61.8	33.2 ± 17.1	12.3	41.2	26.4 ± 12.2
3-ring	**Ace**	18.9	87.1	22 ± 10.3	6.1	78.4	13.2 ± 6.6	2.3	11.2	5.3 ± 3.1	ND	7.9	4.0 ± 1.8
	Fle	7.2	50.4	18.2 ± 7.6	2.3	28.7	9.2 ± 4.2	2	8.6	4.3 ± 2.7	1.1	6.8	3.2 ± 1.5
	Phe	18.9	79.3	38.1 ± 16.7	5	44.2	17.1 ± 8.3	3.3	16.9	9.2 ± 4.3	ND	11.2	6.4 ± 3.5
	Ant	10.7	54.2	20.8 ± 9.4	1.3	16.1	8.3 ± 4.0	ND	6.1	2.8 ± 1.5	ND	2.2	2.1 ± 1.1
	Tot. (2,3 ring)	100.5	568.6	**241.4 ±121.7**	48.9	357	**137.1 ±66.8**	21.7	104.6	**54.8 ±29.3**	13.4	69.3	**42.1 ±20.4**
	%R (2,3 ring)						**43**			**77**			**82**
	Frt	112.3	798.4	464.9 ± 193.4	102.4	415.2	219.4 ± 109.8	24.5	103.4	76.1 ± 38.5	16.1	79.9	52.3 ± 25.7
4-ring	**Pyr**	68.3	231.2	129.2 ± 62.9	24.6	102.2	69.8 ± 35.2	15.2	55.7	29.8 ± 13.3	11.2	30.2	18.2 ± 10.1
	BaA	44.8	203.9	111.7 ± 55.8	21.3	95.2	60.8 ± 31.9	10.2	34.2	18.9 ± 11.0	8.1	25.3	14.7 ± 7.6
	Chr	17.6	100.7	38.2 ± 18.9	8.2	36.4	17.6 ± 9.3	3.7	15.9	8.4 ± 3.8	ND	12.4	5.8 ± 2.4
	BbF	N.D	29.4	9.6 ± 4.5	N.D	9.4	4.3 ± 2.2	ND	5.2	1.9 ± 1.0	ND	4.8	1.5 ± 0.9
5-ring	**BkF**	ND	ND	ND	N.D	ND	ND	ND	ND	ND	ND	ND	ND
	BaP	N.D	18.6	5.1± 2.2	N.D	9.4	2.1 ± 1.3	ND	3.1	0.8 ± 0.3	ND	31.2	0.6 ± 0.3
	DBA	1.5	9.9	3.4 ± 1.4	N.D	53.5	1.5 ± 1.0	ND	1.5	0.4 ± 0.2	ND	1.2	0.3 ± 0.2
	Tot. (4,5 ring)	244.5	1392.1	**762.1 ±331.2**	156.5	721.3	**374.8 ±188.5**	53.6	219	**126.2 ±67.6**	35.4	185	**93.2 ±47.2**
	%R (4,5 ring)						**50**			**83**			**88**
6-ring	**BghiPe**	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	IDP	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	Total (average)	345	1960.1	**1003.5 ± 460.1**	205.4	1078.3	**483.3 ± 257**	75.3	323.6	**191.1 ± 96.8**	48.8	254.3	**135.5 ± 67.3**
	Total % R						**52**			**81**			**86**

TABLE 3. Concentration levels of PCB congeners in raw and treated wastewater using ABR at different HRT

PCBs (ng/l)	Raw			8 hrs			16 hrs			24 hrs		
	Min	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**	**Min**	**Max**	**Average**
C-18	928.1	2466.2	1541.2 ±722.1	310.9	1391	964.2 ±341.2	207.1	1021.3	559.4 ±272.3	155.8	479.6	383.5 ±129.4
C-31	182.5	1766.4	1024.2 ±439.4	100.3	876.3	698.3 ±224.5	83.2	607.2	416.8 ±200.3	59.1	293.2	217.9 ±97.3
C-28	ND	1500.3	512.4 ±266.3	ND	604.2	311.7±137.9	ND	322.4	188.2 ±81.2	ND	159.1	92.3 ±40.2
C-52	199.2	2439.2	1723.9 ±611.3	98.2	1870.4	1024.6 ±420.7	ND	664.5	419.1 ±177.3	ND	568.2	415.8 ±185.2
C-44	211.3	2921.1	1035.3 ±289.3	102.3	698.5	491.3 ±163.6	86.4	358.8	289.3 ±127.2	53.1	297.3	211 ±81.8
C-101	ND	642.4	211.2 ±98.7	ND	278.9	115.5 ±49.1	ND	133	77.2 ±31.4	ND	111.2	56.2 ±21.6
C-149	321	2413.2	725.9 ±322.6	201.6	578.7	401.3 ±178.5	136.9	461.1	310.3 ±113.9	101	276.4	199.4 ±82.7
C-118	ND	265.7	131.2 ±55.4	ND	122.3	60.9 ±27.6	ND	69.3	41.2 ±16.9	ND	ND	ND
C-153	107.4	1129.5	652.3 ±247.9	89.7	512.1	386.1 ±168.9	66.3	354.6	244.5 ±97.8	ND	223.1	139.4 ±71.2
C-138	324.9	2151.4	819.4 ±321.2	188.9	569.8	432.6 ±204.6	131.4	355.3	293.7 ±124	113.9	321	187.3 ±84.1
C-180	ND	732.4	279.1±112.8	ND	219.7	144.3 ±72	ND	108.4	79.8 ±34.8	ND	88.9	23.5 ±12.9
C-194	ND	319.8	81.3 ±31.3	ND	104	52.5 ±23.1	ND	ND	ND	ND	ND	ND
Total	2274.4	18747.6	8736.4 ±2671.3	1091.9	7825.9	5083.3 ±1897	711.3	4455.9	2191.5 ±947.2	482.9	2818	1926.3 ±677.2
Total % R						0.41			0.66			0.77

In spite of removal efficiency of treatment for PCBs seemed to be good, high concentration from banned PCBs still present in final effluent.

Conclusion

The occurrence of persisting pollutants as PAHs and PCBs were analyzed and demonstrated in municipal sewerage wastewater that receives many industrial and domestic wastes. A treatment system of compartmentalized anaerobic baffled reactor (CABR) is used for wastewater treatment and operated at different HRT and OLR as well. In case of PAHs, Fluoranthene and Anthrathene have the highest detected concentration levels among all detected PAHs with average total concentrations of 464.9 and 142.3 µg/l, respectively. The PAHs removal efficiency in treatment system (CABR) was varied from 52, 81 and 86% at HRT of 8, 16 and 24 hrs, respectively. Whereas, for PCBs the highest detected concentration levels are 1.7 and 1.5µg/l for C52 and C18, respectively. The PCBs removal efficiency in treatment system (CABR) was varied from 41, 67 and 77% at HRT of 8, 16 and 24 hrs, respectively. Despite of high reduction load of organic micro-pollutants assessed in present study using a decentralized wastewater treatment units as CABR, discharged wastewater resulted from insufficient treatment remain important sources of hazardous and toxic pollutants to aquatic environment thus it is necessary for controlling the effluent for further post treatment. The impact of discharged effluent to aquatic environment will have to be evaluated.

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إزالة الملوثات الخطره من مياه الصرف الصحي باستخدام المخمر اللاهوائي المجزأ: تأثير معدل الحمل العضوي

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تتناول هذه الدراسة حدوث وكفاءة إزالة الهيدروكربونات العطرية متعددة الحلقات (PAHs) وثنائي الفينيل متعدد الكلور (PCBs) كمواد عضوية خطره (POPs) في مياه الصرف الصحي وذلك بواسطة المفاعل اللاهوائي ذو الحواجز .(CABR) تم تقييم عمليات المعالجة وكفاءة الإزالة باستخدام CABR عن طريق تطبيق ثلاث مراحل من مدد المكث الهيدروليكية (٨ و ١٦ و ٢٤ ساعة). كانت قيم التركيز المكتشفة لل PAHs في مياه الصرف الصحي الخام تتراوح بين ٣٤٥ إلى ١٩٦٠,١ بمتوسط قيمة ١٠٠٣,٥ ميكروغرام / لتر. في حين تراوحت الازاله الخاصة بمركبات ثنائي الفينيل متعدد الكلور من ٢,٣ إلى ١٨,٧ بمتوسط قيمه ٨,٧ ميكروغرام / لتر. وقد حققت كفاءة الازالة لمركبات PAHs ٥٢ و٨١ و ٨٦٪ علي التوالي، في حين سجلت ٤١ و ٦٧ و ٧٧٪ لمركبات ال PCBs وذلك باستخدام مدد مكث HRT تراوحت مابين ٨ و ١٦ و ٢٤ ساعة، على التوالي. حيث ان هذه الملوثات العضوية موضع الدراسه تتواجد بشكل كبير وبتركيزات عاليه في المجاري المائيه بمصر لذا كان لزاما استخدام تقنيات فعاله لمعالجة مثل هذه الملوثات. لذلك فان اهمية هذه الدراسه تكمن في تسليط الضوء علي احد تقنيات معالجة المياه باستخدام المخمر اللاهوائي المجزأ CABR لاستعادة وتحسين جوده المياه العادمه بغرض إعادة استخدامها أو تصريفها بأمان على المسطحات المائيه.