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Efficiency of nitrification and organics removal from municipal landfill leachate in the rotating biological contactor (RBC)

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ABSTRACT

The usability of moving beds both to ammonia oxidation and removal of refractory organic compounds from municipal landfill leachate was investigated. Organics and ammonia concentrations in the leachate were 1154 mg COD/L and 834 mg N-NH_4/L, respectively, whereas BOD_5/COD – 0.08. The humic substances expressed by UV_{254} and UV_{280} indexes were 10.04 and 8.0, respectively. The investigations have revealed that at ammonium load of 1.92 g N-NH_4/m^2·d (experiment 1), complete and stable nitrification (99%) was obtained and the single stage RBC may be sufficient in practical application, while at load 3.58 g N-NH_4/m^2·d (experiment 2), there was a necessity of two-stage system working (nitrification efficiency was 70% at 1st stage). At ammonium load of 4.79 g N-NH_4/m^2·d (experiment 3), the nitrification effectiveness was 59.7% in the 1st stage, but in whole system — 74.4 %. Simultaneously with nitrification, organics removal was observed. The efficiency of COD removal changed from 42.9% (experiment 1) to 32.6% (experiment 3). The efficiency of humic substances removal, based on UV_{254} and UV_{280} indexes, exceeded 30% and 20%, respectively. High concentrations of extracellular polymeric substances EPS (90.9-68.8 mg/g d.w.) in both stages of RBC and low biodegradability of organics in leachate (BOD_5/COD) allow to consider biosorption as a main process responsible for refractory compounds removal.

Keywords: Landfill leachate; Nitrification; Organics; Rotating biological contactor (RBC); EPS

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1. Introduction

Landfill leachate has been considered as problematical wastewater from the treatment point of view, because its quality and quantity changes within time at the same landfill. Leachate produced in young landfills is generally characterized by the presence of substantial amounts of volatile acids, as a result of the acid phase of fermentation. Typical old landfills produce leachate categorized as stabilized with slightly basic pH, relatively low chemical oxygen demand (COD) and the presence of refractory substances with high molecular weight compounds and low biodegradability (i.e. humic substances). Fan et al. [1] extracted and analyzed humic substances, humic acids (HA), fulvic acids (FA) and non-humic substances (NHS) from leachates originated from three different landfills and showed that percentage of HA was 11–28% and FA 26–30% of all humic substances, while the rest posed NHS. From other researches it results that humic acids might reach even 60% [2]. Moreover, leachate is reach in ammonium. Landfilled municipal solid waste (MSW) contain significant amounts of organic nitrogen in a non-

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degradable form as well as readily soluble nitrogen. As a result of anaerobic digestion of MSW, around 50% of the nitrogen undergoes solubilization [3]. According to Ehrig [4] the release of soluble nitrogen from municipal solid waste into leachate is slow and continues over a long period. Leachate from older landfills are rich in ammonia nitrogen due to hydrolysis and fermentation of the nitrogenous fractions of biodegradable substrates [5].

From literature data it results that wastewater with high ammonium content are difficult to effective treatment in suspended-growth systems (activated sludge) due to inhibitory effect on nitrifying microorganisms [6]. Moreover, in activated sludge, the ammonium oxidation proceeds slowly because of slow nitrifying bacteria growth. Therefore, it is necessary to provide high residence time of nitrifying cells (sludge age). Thereby, in order to obtain nitrification there are employed combined methods, such as activated sludge processes with biomass carriers. So far, nitrification was studied in reactors with activated sludge contained plastic carrier material as small cubes of macroporous cellulose or tubes made of polyethylene with an addition of ammonium chloride [7]. Commonly used appliances to nitrification are moving-bed biofilm process, based on the use of small, free-floating polymeric (polyurethane) elements, while biomass is being grown and attached as biofilm on the surface of these porous carriers [8]. Recently, many authors employed zeolite to improve nitrification in activated sludge systems. From literature view, it is concluded that two different mechanisms could cause enhancement of nitrification in activated sludge with these carriers. Added minerals adsorb some compounds working as nitrification inhibitors and nitrification may be improved as a consequence of decreased inhibition [9], and enable nitrifiers to colonize the attached biofilm, improving nitrification [10]. Kargi and Pamukoglu [11] compared two different adsorbents, PAC (powdered activated carbon) and zeolite, alone and with that adsorbent added to biological treatment of pre-treated landfill leachate. The percent of ammonium removal only by adsorption was 28% and 16% for clinoptilolite and PAC, respectively. Adsorbents added to biological system caused increase in ammonium removal to 40% (clinoptilolite) and 30% (PAC).

Another effective technology for nitrification are biofilm systems. According to literature data, in comparison to suspended growth system, biofilm systems allow to obtain nitrification due to long biomass retention time, more resistant to shock loadings, no problems of sludge bulking and higher sludge thickening properties. Moreover, fixed cultures are less strongly affected by changes in environmental conditions (temperature, pH, nutrient concentrations, metabolic products) than suspended cultures. Thereby, in order to obtain nitrification, there are employed systems with attached growth of microorganisms such as trickling filters or rotating biological contactors [12,13]. Apart from ammonium, leachates contain macromolecular compounds, mainly humic substances. They are resistant to biodegradation and most often physico-chemical methods are used for their removal, including sorption on activated carbon [14,15]. However, sorption of some organic substances from wastewater (e.g. phenantrene, benzene, toluene, m-xylene) to the extracellular polymeric substances (EPS) matrix has been confirmed [16,17]. Moreover, Esparza-Soto, Westerhoff [18] showed that biosorption of humic substances onto activated sludge microorganisms surface is effective method for their removal from river water.

It is known that microorganisms produce EPS in all biological treatment systems (activated sludge, granular sludge, biofilms). However, Park et al. [19] showed that during synthetic wastewater treatment in RBC, amount of EPS extracted from RBC disk was 2-fold higher in comparison with EPS amount in seeding sludge originated from sewage treatment plant.

Potentially high amount of EPS in RBC system and their sorption capacity have decided on the choice of RBC as an alternative in leachate treatment. The aim of this study was to determine efficiency of nitrification and removal of refractory organic compounds from municipal landfill leachate using rotating biological contactor (RBC).

2. Materials and methods

2.1. Leachate feed

Leachate was collected from municipal landfill located in Wysieka (Poland), exploited over 12 years. In the landfill, there are deposited municipal waste apart from fluid waste, faecal matter, hazardous substances, radioactive and toxic waste. The leachate was delivered to the laboratory twice a month and stored at 4°C. Landfill leachate composition is shown in Table 1.

2.2. Process configuration and system design

The experiments were conducted using rotating biological contactor (RBC) working in two-stage system. The RBC was inoculated with an aerobic sludge from wastewater treatment plant. Total discs area on each step was 0.68 m^2 and 45% of the total surface area of each disc was submerged in the leachate. Rotational speed of RBC discs was 7 rpm. Leachate flow rate in all experiments was 10 L/d (HRT was kept at 24 h) and temperature was maintained at 20°C.

The investigations were divided in three experiments differing at ammonium load. In order to obtain various loads, leachate was diluted with tap water. Ammonium concentrations in diluted leachate were 130.4 mg $N\text{-}NH_4$/L (experiment 1), 243.5 mg $N\text{-}NH_4$/L (experiment 2) and 334.0 mg $N\text{-}NH_4$/L (experiment 3). Organic concentrations (expressed as COD, UV_{254} and UV_{280}) were 196 mg/L, 1.76 and 1.41 (experiment 1), 358 mg/L, 3.42 and 2.76 (experi-

Table 1
Composition of raw landfill leachate

Leachate constituent	Mean value
pH	8.6
COD, mg/L	1154
BOD_5, mg/L	92
BOD_5/COD	0.08
k*, d^{-1}	0.12
UV_{254}**	10.04
UV_{280}***	8.0
Total nitrogen, mg/L	879
Ammonia nitrogen, mg/L	834
Organic nitrogen, mg/L	35
Total phosphorus, mg/L	57
Total dissolved solids, mg/L	7328
Volatile dissolved solids, mg/L	1234

* Biochemical oxygen demand rate constants
** Index of the humic substances concentration, caused by the presence of conjugated double bonds and representing aromatic and unsaturated organic compounds
*** Index representing aromaticity of organics

ment 2) and 478, 4.82 and 3.91 (experiment 3), respectively. Total experiment time amounted to 197 days, therein: 31 days (experiment 1), 46 days (experiment 2) and 120 days (experiment 3).

2.3. Analytical methods

The raw leachate was analyzed for pH (pH-meter HI 8818), chemical oxygen demand (COD), according to the Standard Methods [20], biochemical oxygen demand, using OxiTop made by WTW company according to DIN EN 1899-1/EN 1899-2 official EPA method [21], UV optical density at λ = 254 nm (as the index of the humic substances concentration, caused by the presence of conjugated double bonds and representing aromatic and unsaturated organic compounds) [22] and at λ = 280 nm (representing aromaticity of organics) [1, 23]. UV optical densisty was analyzed after filtration through 0.45-µm Millipore membranes to remove suspended and particulate matter that could interfere with UV absorbance measurements. Kjeldahl nitrogen, ammonia-N, nitrite-N, nitrate-N, total phosphorus, total dissolved solids and volatile dissolved solids were determined according to the Standard Methods, as well. Measurements in the effluent included Kjeldahl nitrogen, ammonium-N, nitrite-N and nitrate-N. In treated leachate the following parameters were analyzed: COD, UV optical density at λ = 254 nm and λ = 280 nm, ammonium-N, nitrite-N and nitrate-N.

Biofilm dry mass (mg/cm^2) and EPS content in the biofilm microorganisms were done according to Park et al. [19] and Liu, Fang (2002) [24], respectively.

Table 2
Biofilm characteristics at the 1st and 2nd stage of the RBC

	RBC stage	
	1st	2nd
Wet weight, mg/cm^2	80.6	12.9
Dry weight, mg/cm^2	4.11	5.68
VSS, mg/cm^2	2.77	3.46
EPS, mg/g	90.9	68.6

3. Results and discussion

3.1. Biofilm characteristics

During all experiments, biofilm from the first stage had grayish colour and a gelatinous structure with white zones. Biofilm from the second stage was thinner and had a brown-like colour. Wet weight at the 1st RBC stage was six times higher than in the second one and amounted to 80.6 mg/cm^2 and 12.9 mg/cm^2, respectively. However, dry weight of the biofilm from the second RBC stage was 1.4-fold higher than from the first, that indicates high biofilm hydratation in the 1st RBC stage (Table 2).

EPS concentration was upper in the 1st stage of RBC, that probably is connected with microorganisms type growing in the following stages. Often, in the 1st stage filamentous bacteria grow and as Al-Halbouni et al. [25] showed seasonal variations in EPS concentrations from 17 mg/g dry matter in summer to 51 mg/g dry matter in winter, was correlated with an increased occurrence of filamentous bacteria in the colder season. The EPS concentration in own research was relatively high, but comparable with the content in RBC given by other authors. Park et al. [19] showed that during synthetic wastewater treatment in RBC, amount of EPS extracted from RBC disk was 2-fold higher (83–92 mg/g cell) in comparison with EPS amount in seeding sludge originated from sewage treatment plant (47 mg/g cell).

3.2. Organics in leachate — characteristics and removal

In analyzed leachate, organics concentration expressed as COD was relatively low — 1154 mg/L. According to Fan et al. [1] and Bila et al. [26] concentration of organic compounds (COD) in landfill leachate with similar operating time ranged between 3000–3500 mg/L. Similarly, Calace et al. [27] demonstrated that organics concentration (as COD) in landfill leachate changed from 2400 to 9100 mg/L, while the values obtained in own investigations were clearly lower.

UV_{254} and UV_{280} as the index of humic substances concentration were 10.04 and 8.0, respectively. Rivas et al. [28] reported that in municipal landfill leachate in Italy, the index of humic substances measured on the basis of absorbance at 254 nm was above 40, 4-fold higher than index estimated in present study. Fan et al. [1] using the value of UV_{280} as an indicator for the aromaticity of organics from three different samples of landfill leachate, showed that UV_{280} for leachate from closed landfill, landfill of municipal solid waste (MSW) with bottom ashes from MSW incinerators and landfill of MSW only, were about 1.63, 2.28 and 2.26, respectively.

It is known that high concentration of refractory organics, i.e. humic substances in old leachates might increase the difficulty of landfill leachate treatment, since they could not be easily biodegraded and oxidized. Therefore, removal efficiency of such organics in biological systems is very low. For example, Welander et al. [29] showed low COD removal (about 20%) during landfill leachate treatment from old site. In our study, although organics in leachate were characterized by low biodegradability confirmed by low BOD_5/COD ratio (0.08) and low constant rate of oxygen uptake k (0.12 d^{-1}), they were removed from leachate (Table 3).

Concentrations of organic compounds expressed as COD in treated leachate changed from 112 mg/L to 322 mg/L, meaning that the removal efficiency changed from 42.9% (experiment 1) to 32.6% (experiment 3), while about 60–80% of COD was removed on the 1st stage of the RBC. On the basis of UV_{254} and UV_{280} indexes, the efficiency of humic substances removal exceeded 30% and 20%, respectively. Assuming that organics present in the leachate were refractory (low BOD_5/COD ratio), it might be supposed that main mechanism of their removal was biosorption. The possibility of sorption of humic substances is confirmed by Esparza-Soto, Westerhoff [18], who demonstrated removal of humic substances from river water as biosorption onto activated sludge microorganisms surface. According to the authors the main mechanisms responsible for biosorption were hydrophobic and cationic bridging effects between humic substances and activated sludge EPS. Moreover, the authors showed that the humic acid fraction was removed more efficiently than fulvic acid fraction because the humic acid was more hydrophobic. According to Jorand et al. [30] the higher removal of HA may be explained as the result of hydrophobic interactions between HA and biomass surface because of their incapability of interacting electrostatically with water.

In biological systems, removal of organics can occur by two mechanisms acting singly or in combination: sorption into biomass structures such as a biofilm and biodegradation. Biodegradation is limited to the easily biodegradable organics, however sorption may play significant role in the removal both biodegradable and refractory organics. As sorption sites can serve cell walls, cell membranes, cell cytoplasm and extracellular polymeric substances (EPS) synthesized by attached bacteria which have been shown to complex inorganic cations as well as sorbed organic molecules [31, after Characklis and Marshall, 1990]. These sites display different sorption properties, preferences and capacites. Spath et al. [16] showed that during BTX sorption, more than 60% of the total contens was found in the EPS, whereas the rest can be sorbed at the cell wall, the cytoplasmic membrane, or in the cytoplasm. However, in case of cadmium about 80% of total metal content was found in the cellular fraction and only a small amount was bound in the EPS. Because EPS is a significant component of biofilm, the chemical nature of EPS affects interactions between contaminants and biofilm. Carlson and Silverstein [31] showed that sorption of natural organic matter (NOM) molecules was inversely proportional to molecular size of organics. Moreover, significantly less removal was observed for anionic sorbates, compared with uncharged molecules.

In many cases organic matter contains biodegradable and non-biodegradable compounds, so it is difficult to study only sorption in the biofilm. However, Carlson and Silverstein [32] found that in a bench scale packed-bed reactors 18–22% of NOM was removed from the water by biosorption onto living biofilm in a packed-bed reactor, with negligible biodegradation, compared with 0–6% removal in an abiotic control reactor.

3.3. Nitrification

In this study, the nitrification efficiency and the main product of nitrification, depending on ammonium load in rotating biological contactor (RBC) working in two-stage RBC system, was estimated. Nitrogen concentrations in the effluent in dependence on ammonium load are listed in Table 4, however nitrification efficiency in Fig. 1.

It was shown that at ammonium load of 1.92 g

Table 3
Organics in treated leachate

Experiment No.	Organics in the effluent					
	1st RBC stage			2nd RBC stage (treated leachate)		
	COD (mg/L)	UV_{254}	UV_{280}	COD (mg/L)	UV_{254}	UV_{280}
Experiment 1	139	1.26	1.08	112	1.14	1.01
Experiment 2	252	2.61	2.24	219	2.26	2.13
Experiment 3	387	3.84	3.29	322	3.02	2.97

Table 4
Nitrogen concentration in treated leachate

Ammonium load ($g/m^2 \cdot d$)	Nitrogen concentration in the effluent (mg/L)					
	1st RBC stage			2nd RBC stage (treated leachate)		
	$N\text{-}NH_4$	$N\text{-}NO_2$	$N\text{-}NO_3$	$N\text{-}NH_4$	$N\text{-}NO_2$	$N\text{-}NO_3$
1.92	0.34	0.36	122	0.09	0.07	122.9
3.58	74.3	32.5	128.4	0.12	0.16	221.7
4.79	131.3	1.27	182.7	83.4	0.12	231.4

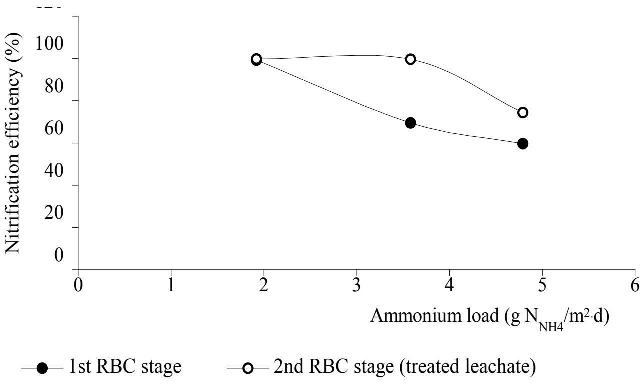

Fig. 1. Nitrification efficiency dependence on RBC ammonium load.

$N\text{-}NH_4/m^2 \cdot d$, the nitrate were the predominant product of nitrification and concentrations of nitrite and ammonium were below 0.2 mg/L (Table 3). Completely nitrification (> 99%) was obtained in the 1st stage of RBC (Fig. 1).

From these results it can be concluded that in case of old landfill leachate at load of 1.92 g $N\text{-}NH_4/m^2 \cdot d$, to reach complete and stable nitrification, the single stage RBC may be sufficient in practical application.

In the case of wastewater with high biodegradable organic concentration, in the initial stages of the RBC, organics can affect nitrification because of heterotrophic bacteria that are in competition to nitrifiers displacing them within the bioreactor [33]. Therefore, the maximum nitrification rate occurs after organics removal, i.e. in the latter stage of the RBC system. In our study, due to low concentration of biodegradable organic substances in raw landfill leachate, their influence on nitrification efficiency was negligible.

The average ammonium removal rate in RBC in all experiments was calculated on the material balance for rotating biological contactor:

$$Q_0 \cdot C_0 - r_{A_{t-total}} \cdot A_{t_{total}} = Q_e \cdot C_e \tag{1}$$

where Q_0 is landfill leachate flow rate (L/d), C_0 — ammonium concentration in raw leachate (mg/L), $r_{A_{t\text{-}total}}$ – the average ammonium removal rate in RBC ($g/m^2 \cdot d$), $A_{t\text{-}total}$ — total discs area (m^2), Q_e — landfill leachate flow rate (L/d) ($Q_0 = Q_e$), C_e — ammonium concentration in treated leachate (mg/L).

The calculated value of ammonium removal rate in experiment 1 amounted 1.91 g $N\text{-}NH_4/m^2 \cdot d$.

In experiment 2, the ammonium load of disc area was increased nearly 2-fold, in comparison with experiment 1, to 3.58 g $N\text{-}NH_4/m^2 \cdot d$. Nevertheless, complete nitrification (99.9%) was observed in two-stage RBC system, therein 70% process effectiveness was obtained in the 1st stage (Fig. 1). In treated leachate, in the 1st stage nitrate and ammonium were dominant forms of nitrogen (average concentration of 128.4 mg $N\text{-}NO_3/L$ and 74.3 mg $N\text{-}NH_4/L$, respectively) (Table 3). The leachate contained also high amount of nitrite with average concentration in the effluent of 32.5 mg $N\text{-}NO_2/L$. The concentration of the individual nitrogen forms in treated leachate was 0.12 mg $N\text{-}NH_4/L$, 0.16 mg NNO_2/L and 221.7 mg $N\text{-}NO_3/L$, respectively. The rate of ammonium removal amounted 2.49 g $N\text{-}NH_4/m^2 \cdot d$ (the 1st stage) and 1.09 g $N\text{-}NH_4/m^2 \cdot d$ (the 2nd stage). Summarizing, it should be noted that at ammonium load of 3.58 g $N\text{-}NH_4/m^2 \cdot d$, there was a necessity of two-stage system working to complete nitrification and obtaining stable products in form of nitrate.

In our study, most of ammonium was removed in the 1st stage of RBC. The results are in contradictory to these obtained by Tawfik et al. [34] during municipal wastewater treatment. The authors showed that in the 1st stage of two-stage RBC system only small amount of ammonium was eliminated and nitrification mainly proceeded in the 2nd stage. However, the investigations were conducted on municipal wastewater at high COD load in the 1st stage (21.3 g $COD/m^2 \cdot d$) and 3-fold lower in the 2nd stage, what may suggest that nitrification occurs after organics removal.

At ammonium load of 4.79 g $N\text{-}NH_4/m^2 \cdot d$ (experiment 3) the nitrification effectiveness was 59.7% in the 1st stage, but in whole system – 74.4% (Fig. 1). The rates of ammonium removal were 2.86 g $N\text{-}NH_4/m^2 \cdot d$ and 0.70 g $N\text{-}NH_4/m^2 \cdot d$ in the 1st and 2nd stages of RBC system. After the 1st stage, the leachate contained mainly ammonium (average 131.3 mg $N\text{-}NH_4/L$) and nitrate — 182.7 mg $N\text{-}NO_3/L$ (Table 2). The concentration of individual nitrogen forms in treated leachate was 83.4 mg NNH_4/L, 0.12 mg NNO_2/L and 231.4 mg NNO_3/L, respectively (Table 3). It means that at ammonium load exceeding 4.79 g $N\text{-}NH_4/m^2{\cdot}d$, two-stage system turned out insufficient to complete nitrification.

From literature data it results that nitrification failure can be caused by high ammonium load, short HRT or low oxygen concentration. In aerobic RBC, the rotation of the media supports oxygen transfer which promotes nitrification. The higher rotation speed, the higher oxygen transfer capacity of RBC in terms of the overall oxygen transfer coefficient [35]. Usually, with the increase of rotation speed, increases the dissolved oxygen concentration available to microorganisms. As a result they are able to degrade the substrate at a higher rate. Investigations of Israni et al. [36] and Najafpour et al. [37] have proved that in aerobic RBC an increase of the rotational speed caused higher COD removal. On the other hand, if rotational speed is too high, the microorganisms will be stripped off the discs, deteriorating the effluent quality and lowering the biodegradation rate in the reactor. Radwan, Ramanujam [38] investigated the influence of COD/$N\text{-}NH_4$ ratio on organics removal and nitrification from synthetic wastewater in a four-stage RBC. The COD and ammonium concentrations were about 100–1500 mg/L and 20–185 mg/L, respectively. Hydraulic load was 0.03 $m^3/m^2{\cdot}d$ and ammonium loadings were in range of 0.66–5.5 g $N\text{-}NH_4/m^2{\cdot}d$. The authors showed that at ammonium load up to 1.85 g $N\text{-}NH_4/m^2{\cdot}d$, the effluent ammonium nitrogen was less than 5.0 mg/L (at rotation speed of 6 and 12 rpm), however, percentage of COD removal was approximately 97%. At load higher than 1.85 g $N\text{-}NH_4/m^2{\cdot}d$, increasing the rotational speed from 6 to 12 rpm caused improved of nitrification rate. Rostron et al. [39] in the reactor with immobilised biomass, observed complete nitrification at HRT of 2.2–8 d. After HRT shortening to 1.5 d build-up of nitrite took place. According to the authors, the lower cell yield of Nitrobacter in comparison to *Nitrosomonas* (0.042 mg cells/mg N and 0.142 mg cells/mg N, respectively) indicated that a lack of *Nitrobacter* was probably the reason for the build-up of the nitrite. When HRT was further decreased to 1 d, the concentration of ammonium increased, indicating washout of *Nitrosomonas*.

4. Conclusions

RBC working in two-stage system is effective technology that can be applied as the 1st stage of leachate treatment from stabilized municipal landfills. Complete nitrification and nitrate as stable reaction product were obtained at ammonium loads from 1.92 g $N\text{-}NH_4/m^2{\cdot}d$ to 3.6 g $N\text{-}NH_4/m^2{\cdot}d$. Simultaneously with nitrification, organics removal was observed. Taking into consideration low biodegradability of organic substances (BOD_5/COD ratio was 0.08), and high content of extracellular polymeric substances EPS (68.6–90.9 mg/g) produced by biofilm microorganisms, it was assumed that main process responsible for removal of refractory compounds was biosorption. As the process is an initial step to remove high molecular weight organic compounds, future research should involve studies on different parameters controlling biosorption in order to gain a deeper understanding the role of EPS in mechanism of refractory compounds removal.

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