REMOVAL OF SELECTED ORGANIC POLLUTANTS IN ANAEROBIC WASTEWATER TREATMENT PROCESS

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ABSTRACT

Aromatic compounds belong to one of most widely distributed classes of organic substances used in industry, services and households and, therefore they are relatively often measured in municipal wastewater. Their efficient biological degradation is essential to reduce the subsequent disposal of such substances into the waters of the receiver, or release into the atmosphere. The mineralization of organic compounds by facultative or obligate anaerobic bacteria can have importance for next state of treatment, because most of organic contaminants are toxic for microorganisms of conventional activate sludge process. The aim of the study presented was assessment of the removal of specific organic contaminations BTX (benzene, toluene and xylenes) found in wastewater in municipal WWTP during anaerobic treatment process. The experiments have been performed in the laboratory anaerobic reactor of the 5 L volume. The concentrations of BTX in wastewater were in the range of 0.8 and 5.3 mg/L as the sum of the compounds. After the anaerobic treatment process the concentrations of BTX decreased on average: 45 % for benzene, 59 % for toluene, 65 % for o-xylene and 76 % for p-xylene.

KEYWORDS

organic pollutants, aromatic hydrocarbons, BTX, anaerobic biodegradation, , anaerobic wastewater treatment

INTRODUCTION

The literature review and our own research results presents a quite common occurrence of various aromatics such as benzene and phenol derivatives in municipal wastewater. This applies to both aliphatic and chloro-or nitro-derivatives of these substances, as well as many other hydrocarbons, of which a large number of compounds is harmful to aquatic organisms, including humans. This kind of substances may be discharged with industrial wastewater, either with small manufacturing or service facilities and municipal wastewater. The presence of some hydrocarbons are detected in the effluent and also at different stages of their treatment processes. It means that some of aromatic compounds are not completely degraded in the wastewater treatment process and they can be discharged into the recipient and/or released into the atmosphere, (Albin and Holdern, 198; Meltzer et al. 1992; Hsieh et al. 1993; Bell et al. 1993; Melcer 1994; Suschka et al. 1996; Escalas et al. 2003; Kaleta, 2007).

Volatile mono aromatic hydrocarbons such as benzene, toluene and xylenes (collectively known as BTX) are key industrial solvents and are frequently required in industrial operations. These compounds are released into the environment during manufacture, transportation, usage and disposal. The liquid as well as gaseous states of these compounds in the environmental pose a significant threat to human health and the environment due to their toxic and carcinogenic properties. Mono aromatic hydrocarbons are hydrophobic volatile organic compounds, with maximum solubility in the range of

1740-1800, 515-540 and 178-220 mg/L for benzene, toluene and xylene respectively (Byrns, 2001; Maliyekkal et al., 2004).

Most of the liquid batch studies that have been carried out focused primarily on the degradation of individual substrates with a microbial population acclimatized to the substrates. A few studies have assessed the possibility of degradation a variety of compounds by one particular strain of microbes that was previously acclimatized for a single substrate. Although, each of BTX compounds can be mineralised by microorganisms, in mixture they cannot be successfully mineralised simultaneously. Moreover, even when mixed bacterial cultures are used, the presence of one compound may affect the degradation of another (Colombo et al., 2004).

Conventional aerobic biological wastewater treatment processes are commonly used to treat wastewater streams containing benzene, toluene, o-, m-, and p-xylene (BTX). However, these processes are not often out of compliance with the current national Emissions Standards for hazardous Air Pollutants regulations, due to stripping of the volatile compounds from the aqueous phase into the atmosphere.

Anaerobic (without oxygen) and anoxic (oxygen deficient) treatments are similar to aerobic treatment, but use microorganisms that do not require the addition of oxygen. These microorganisms use compounds other than oxygen to catalyze the oxidation of biodegradable organics and other contaminants, resulting in innocuous by-products. Compared to aerobic wastewater treatment processes, the most significant advantages of anaerobic processes are: significant excess sludge mass reduction, while many complex organic substances - aromatic compounds - are partially or even fully biodegradable and minimize the volatile substances emissions to the atmosphere. The basic principles responsible for BTX removal during anoxic and anaerobic activated sludge treatment are sorption and biological transformation.

Anaerobic biodegradation of BTX

The documentation of anaerobic degradation of BTX is relatively recent (within the last two decades) and is constantly generating new insights. This activity is widespread, have been reported under nitrate-, iron-, manganese- and sulfate-reducing conditions as well as methanogenic conditions. Studies of microbial degradation of monoaromatic hydrocarbons have resulted in the identification and isolation a number of different anaerobic bacterial strains capable of degradation one or more monoaromatic hydrocarbons. Such cultures have initiated the elucidation of degradative pathways, intermediates, and genes encoding key enzymes. At the same time, pathways and metabolites have been inferred from complementary studies of mixed populations using analytical chemical methods to demonstrate relevance of laboratory results in situ. It is now clear that some key enzymatic steps in anaerobic hydrocarbon biodegradation involve novel biochemistry and versatile microbiota (Foth, 2008).

Evidence for the anaerobic degradation of benzene, the most stable aromatic hydrocarbon, has been provided by studies with enriched populations in or from sediments containing different electron acceptors. The mechanism of the initial reaction of benzene degradation in the absence of molecular oxygen is unknown (Widdel and Rabus, 2001). The occurrence and rate of biodegradation appears to be more site-specific for benzene than other BTX components and is subject to inhibition by those co-contaminants degradation, which is usually slow, incomplete and conditioned by long lag times (Foth, 2008).

Of these compounds, the anaerobic biodegradation of toluene is probably the most comprehensively understood. Toluene is biodegradable with nitrate, Fe(III), humic substances, sulfate and carbon dioxide as terminal electron acceptors (Grbic-Galic and Vogel, 1987; Edwards and Grbic-

Galic, 1994; Hunkeler et al. 1998; Hutchins, 1991; Chakraborty and Coates, 2005; Edvard et al. 1992; Hutchins et al. 1998; Coates et al. 1999; Beller et al. 1996; Foth, 2008). More recently, it has been demonstrated that toluene can also be assimilated anaerobically as a carbon source by anooxygenic phototrophs. Many organisms capable of anaerobic toluene degradation were alternatively capable of anaerobic degradation of other monoaromatic compounds such as benzene or xylenes. Although all three xylene isomers appear to be biodegraded via the fumurate addition pathway analogous to toluene, they have different susceptibilities to anaerobic biodegradation. Several organisms have been described that can completely mineralize m- and o- xylene coupled to the reduction of nitrate or sulfate (Foth, 2008).

Anaerobic degradation of p-xylene has been reported by an undefined nitrate-reducing enrichment culture, to date there is no organisms available in pure culture that can anaerobically mineralize this compound (Chakraboty et al., 2005).

Anaerobic biodegradation of individual BTX components often consisted of three distinct phases: (1) a lag period with little or no biodegradation; (2) a rapid degradation period; and (3) an asymptotic period where contaminant concentrations remained essentially constant (Borden et al., 1997). By the authors, this pattern of biodegradation cannot be accurately described with a simple first-order decay function. In contrast to the behaviour of the individual compounds, the biodegradation of total BTX appears to more closely approximate by first-order decay function. However, most often, biodegradation of organic compounds in sewage treatment systems is based on the Monod equation. The degree of compound removal depends on its concentration and activity of microbial biomass. This dependence is described by the equation of the first order with respect to substrate concentration (Rittmann, 1985; Janosz-Rajczyk, 2008). Due to the low concentration of trace substances, the degradation occurs mostly as a first order reaction, including sorption on suspended solids yields.

Sorption on activated sludge

Sorption on wastewater solids is an important mechanism for removal of organic compounds in biological wastewater treatment systems. Removal by sorption may occur either by removal of influent particulate matter or by excess sludge withdraw. In the case of organic micropollutants sorption, a distinction is made between: absorption (hydrophobic interreactions of the aliphatic and aromatic group of compounds with lipophilic cell membranes of miocroorganisms and fat fractions of the sludge) and adsorption (electrostatic interreactions of positively charged groups of chemicals with the negatively charged surfaces of microorganisms) (Siegrist et al., 2005). Due to the fact that sorption depends on various molecural characteristics of the micropollutants as well as of the sorbing particulate matter, there is generally no valid surrogate parameter, quantitatively describing this removal mechanism. For example, the partitioning coefficient for octanol-water $K_{OW,}$ or for organic carbon K_{OC}. They may correlate with the removal for rather lipophilic substances with similar molecular structure, but not if different group of compounds are compared. Therefore, a method characterizing the sorption coefficient of each compound for primary as well as for secondary sludge is needed. It is assumed that sorbed and dissolved fractions are in equilibrium due to the long detection time (several hours) within the reactor (Joss et al., 2008).

$$K_d = \frac{S_{sorb}}{X_{SS} \cdot S_{diss}}$$

with K_d – sorption constant [L/gX_{SS}]
S_{sorb} – concentration sorbed onto activated sludge, per unit reactor volume [µg/L]
X_{SS} – concentration of activated sludge in the tank [gX_{SS}/L]
S_{dis} – dissolved concentration [µg/L]

Assuming biodegradation and sorption of organic pollutants the rate of their removal (r_r) in the anaerobic activated sludge process can be determined according to first order reaction kinetic presented by Siegrist et al. (2005):

$$r_r = k_{deg} \cdot X_{SS} \cdot S_{dis} = k_{deg} \cdot X_{SS} \cdot \frac{S_{tot}}{1 + K_d \cdot X_{SS}} = \frac{k_{deg}}{1 + K_d \cdot X_{SS}} \cdot X_{SS} \cdot S_{tot} = k'_{deg} \cdot X_{SS} \cdot S_{tot}$$

with k_{deg} – degradation rate constant [L/gX_{SS}·d]
$S_{tot} = S_{sorb} + S_{dis} = S_{dis}(1 + K_d \cdot X_{SS})$ – sum of dissolved and sorbed concentration

Laboratory investigations have been carried out in order to estimate the efficiency of BTX removal in the anaerobic sludge activated process. In addition, the importance of sorption participation in the aromatic hydrocarbons (BTX) removal from wastewater in the anaerobic treatment process was assessed.

MATERIALS AND METHODS

The laboratory investigations of the anaerobic treatment process have been performed with the use of both real activated sludge and municipal wastewater, taken from a municipal treatment plant in Bielsko-Biala and synthetic wastewater. The treatment plant was designed and operated for enhanced biological nitrogen and phosphorous removal. Twenty batch series were carried out in the laboratory reactor, (Fig.1).

Synthetic wastewater and mechanical treated municipal wastewater were used in 10 series of experiments for each type of investigated material. The synthetic wastewater have been prepared according to the composition given by Wu et al. (1984). The general characteristic of the wastewater used in batch tests is presented in Table 1.

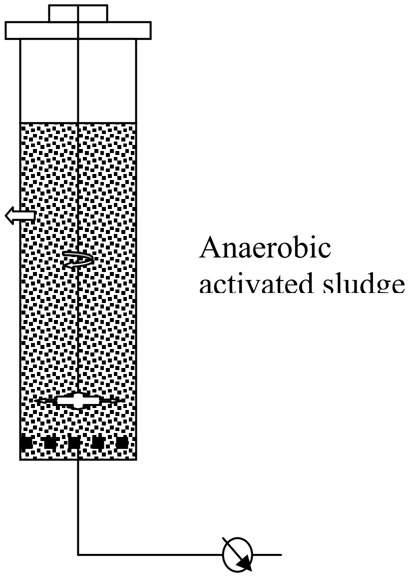

Figure 1. Scheme of the laboratory anaerobic reactor.

Table 1. General characteristic of substrates used in the experiments.

Parameters and units		Synthetic wastewater	Municipal wastewater
pH		5,8 – 7,3	6,9 – 7,4
ORP	[mV]	-134 - 220	-323 - 178
COD	[mgO$_2$/L]	115 – 730	134 – 502
N-NH$_4^+$	[mg/L]	16 - 64	10 – 48
N-NO$_3^-$	[mg/L]	4 - 24	0 – 20
N$_K$	[mg/L]		12 - 85
P-PO$_4^{3-}$	[mg/L]	8 – 27	2 – 11
P$_{Tot}$	[mg/L]		5 – 13

The anaerobic process was carried out at a HRT of 20 hours. The active volume of the reactors was 5 liters. The activated sludge MLSS concentration was maintained at a level close to 5 g/L. A mixture of benzene, toluene, p-xylene and o-xylene (BTX) was added to the reactor. Theoretical doses of each hydrocarbons added, were in the range from 0.25 to 1.25 mg/L. The real concentrations of BTX and COD were measured at the beginning of consecutive experimental runs. The content of the reactor was continuously gently mixed. Examination of treated wastewater quality, after sedimentation for 30 minutes, included determination of; temperature, pH, COD, ORP, dissolved oxygen, phosphorus (P-PO$_4^{3-}$) and nitrogen (TKN, N-NH$_4^+$, N-NO$_3^-$). All analyses were carried out according to the Standard Methods. The BTX concentrations were determined in cleared wastewater and in the sample with activated sludge using a gas chromatography (GC) equipped with a "purge and trap system" and thermal desorption. The GC had a capillary column HP-5 Crosslinked 5% ME Siloxane (length 30 m, internal diameter 0.32 mm, film thickness 0.25 µm) and a FID detector.

RESULTS AND DISCUSSION

The measured BTX concentrations in real raw municipal wastewater varied in a wide range. The highest concentrations were observed for toluene 290 µg/L, for benzene 30 µg/L, for o-xylene 10 µg/L and for p-xylene 8 µg/L. In the laboratory investigations, theoretical concentrations of BTX in wastewater before treatment process were in the range from 0.25 to 1.25 mg/L of each compound (total 1.0 to 5.0 mg/L). But total real concentration of the aromatic hydrocarbons in investigated wastewater was from about 0.8 to 5.3 mg/L. Lower than theoretical concentration has probably resulted from inaccuracy of dose added and the possible losses when BTX have been introduced to the wastewater. Higher concentration was a result of BTX presence in real municipal wastewater. After the anaerobic treatment, the concentrations of BTX (as the sum of aromatic hydrocarbons) decreased on average by 59 % in case of cleared wastewater and 44 % in the samples with activated sludge. The average concentrations of BTX in the investigated wastewater before and after anaerobic treatment process are shown in Fig. 2. The concentrations difference in the treated wastewater with activated sludge and without sludge, indicate that approximately 26 % of BTX was sorbed on the activated sludge particles. The average decrease of individual aromatic hydrocarbons were determined at the level: of 45 % for benzene, 59 % for toluene, 65 % for o-xylene and 76 % for p-xylene. The decrease of BTX concentration after the treatment process is shown in Fig.3. As mentioned above, a part of BTX was sorbed on activated sludge particles and therefore, concentrations of BTX were determined in the samples without activated sludge and in the parallel samples in the wastewater before sedimentation. The largest values of sorbed compounds were observed for p-xylene, the average value was 59 % and about 15 % for benzene (Fig. 4). Probably, it was the result of different compounds solubility in the wastewater. The BTX solubility in water is the highest in the case of benzene and the lowest in the instance of a xylene isomers (Maliyekkal et al., 2004).

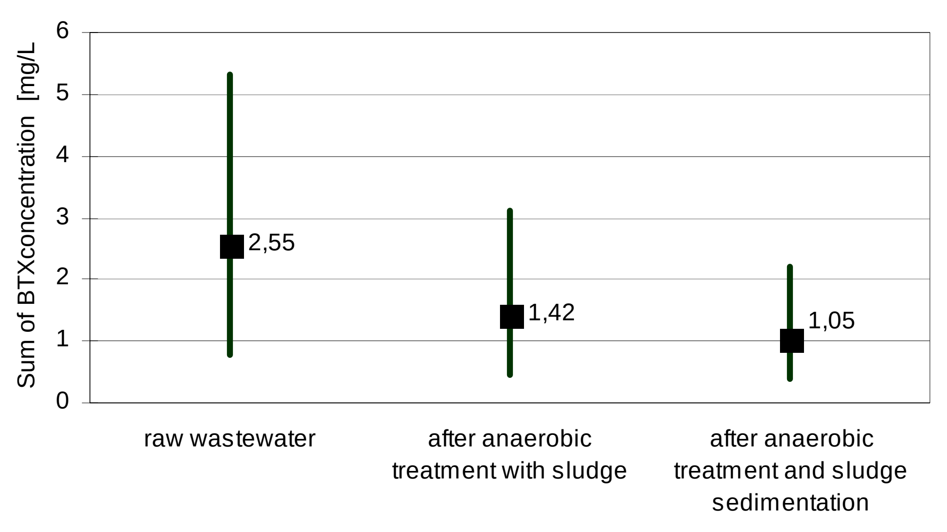

Figure 2. Decrease of BTX sum concentration after anaerobic treatment process in the cleared and with activated sludge samples.

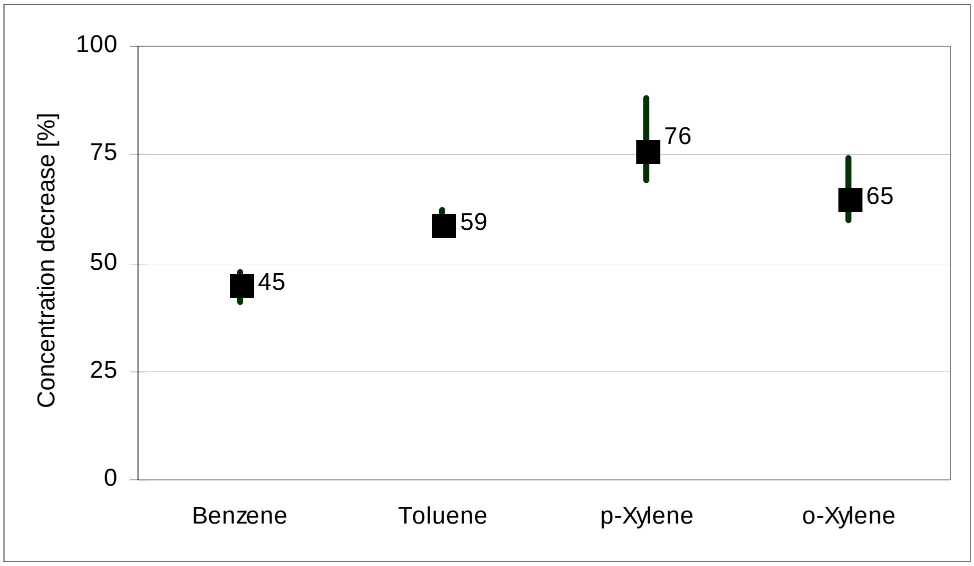

Figure 3. Average decrease of individual BTX concentration after anaerobic treatment process.

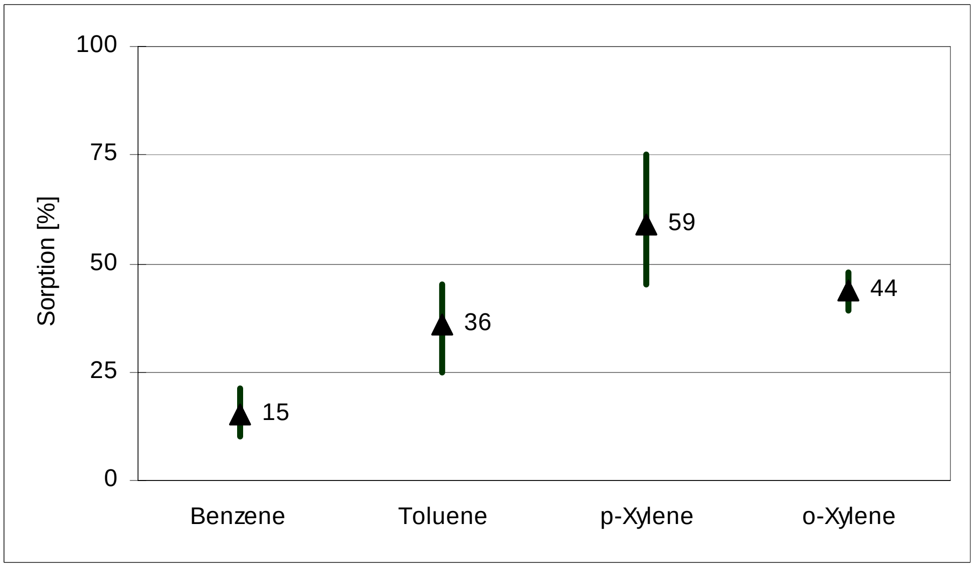

Figure 4. Sorption of BTX to activated sludge particles in the anaerobic treatment process.

Assessing the BTX removal in the anaerobic wastewater treatment a simplified form of the Monod equation was used. The equation was transformed to the first order reaction with respect to the substrate concentration. Adopting this simplification the removal rates constants (k), based on the tested substances effluent concentrations have been determined. The values of the first-order removal constants (k) were determined for the sum of BTX and individual for each analyzed hydrocarbons. Because, the investigations were stated the significant impact of the sorption process on the BTX removal from wastewater have also been determined similarly removal rate constants k ', which are including the BTX sorption on the activated sludge particles, according to the relation defined by Siegrist et al. (2005). In addition, the BTX sorption constants (K_d) on the activated sludge suspension have been determined for individual compounds and mixtures of BTX in the anaerobic conditions (Table 2).

Table 2. The first-order rate constants of BTX removal (k and k') and the sorption constants (K_d) of BTX on activated sludge particles in the anaerobic wastewater treatment.

Compounds	Removal kinetic coefficient k [L/g·d]	Removal kinetic coefficient k' [L/g·d]	Sorption constant K_s [L/g]
Benzene	0,2508	0,1381	0,164
Toluene	0,3206	0,1862	0,144
o-Xylene	0,3465	0,0823	0,642
p-Xylene	0,4755	0,0692	1,174
BTX mixture	0,3550	0,1623	0,237

The compound, which has been removed in the least range was benzene. In the literature, benzene is considered as slow biodegradable in the absence of dissolved oxygen, whereas its aliphatic derivatives (toluene and xylenes) are regarded as relatively easily biodegradable under anaerobic conditions (Grbic-Galić and Vogel, 1987; Edwards and Grbitch-Galić, 1994; Hutchins, 1991, Evans et al., 1991; Edwards et al., 1992; Beller et al, 1992; Beller et al, 1996; Borden et al., 1997; Ma, 1999).

The obtained data also shows that the benzene was the least sorbed on activated sludge particles. Under these conditions, the highest removal efficience of BTX from wastewater was for p-xylene and the compound has had the highest degree of sorption on solids (sludge). Such a ranking (benzene < toluene < o-xylene < p-xylene) of analyzed BTX in terms of the size of their sorption on activated sludge suspension is consistent with the data presented by Zytner (1994), in which he defined the partition coefficient and rate of migration of these compunds in various liquid-solid systems. The determined average values of BTX sorption as the difference of hydrocarbons concentrations in the clarified wastewater and in the wastewater with activated sludge and the participation the process in hydrocarbons removal are about 10 times higher than those presented by Byrns (2001), Jean et al. (2002) and Parkerton (2001). It should be noted however that, as depicted Byrns (2001) and Jean et al. (2002), the determined values for benzene, toluene and xylenes are significantly different from each other in the range of values but have similar quantity dependences. The benzene sorption has the lowest degree, twice as much occurred in the case of toluene, and the highest value was noticed for xylenes. The same relationship have been stated in investigations conducted. In addition, the mentioned authors have not presented which of the xylene isomers have been analyzed. Sorption process is directly related to the chemical structure of a hydrocarbon and depends on the individual BTX concentration in a mixture, however, does not represent a linear relationship (Jean et al. 2002).

SUMMARY

The designated in experiments values of the constant rate of BTX removal in anaerobic wastewater treatment process significantly differ from literature data (they are lower than obtained for aerobic conditions). Analysis of the kinetics of the BTX removal and biodegradation in the anaerobic conditions of activated sludge process is a problem so far unprecedented in scientific reports, but important from the point of view of biological municipal wastewater treatment and BTX elimination. Sorption of these compounds on the particles of activated sludge and their partial biodegradation under anaerobic conditions can significantly help to reduce atmospheric emissions of the pollutants during aerobic wastewater treatment processes and increase the overall efficiency of their removal.

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