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Removal of aromatic hydrocarbons (BTX) in anoxic and anaerobic wastewater treatment processes

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ABSTRACT

Benzene, toluene and xylenes (BTX) are a group of toxic and volatile aromatic compounds prevalent in many industrial wastewaters and they are often determined in municipal wastewater. Usually, BTX-containing wastewater is treated by conventional aerobic wastewater treatment process. However, the removal mechanisms are controversial since these compounds tend to be readily stripped from the aqueous phase to the atmosphere. On the other hand, their efficient removal is essential to reduce the subsequent disposal of such substances into the waters of the receiver. Batch tests with activated sludge process under anoxic and anaerobic conditions have been performed for assessment of the removal of specific organic contaminations BTX (benzene, toluene, and *o*- and *p*-xylene). The investigations were carried out in the laboratory anoxic reactors of 5 L volume. The content of activated sludge (TSS) was 5 g/L and concentrations of BTX in treated wastewater were in the range 0.25–2.37 mg/L of each of the compounds. The batch tests have showed enhanced BTX removal abilities under anoxic wastewater treatment. The rates of BTX removal in average: 0.214 mg/L d for benzene, 0.251 for toluene, 0.131 for *o*-xylene and 0.133 for *p*-xylene have been estimated. For anaerobic investigations the reactors of 2.5 L volume were used. The BTX removal under wastewater anaerobic digestion ranged from 28 to 63%.

Keywords: Volatile organic compounds; Aromatic hydrocarbons; Anoxic wastewater treatment; Psychrophilic anaerobic digestion; BTX biodegradation

1. Introduction

Our research results and the literature dates present a quite common occurrence of monoaromatic aromatics (benzene, toluene and xylenes—BTX) in municipal wastewater. Benzene and its homologues are widely used in different productions as bulk chemicals for industrial use, as solvents and starting materials for the manufacture of pesticides, plastics and synthetic fibers. 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 was 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 [1,3,11,19,23,25,34]. BTX are important

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contaminants, which give rise to cumulative hazardous effects in the environment. The compounds constituting an important group on the list of basic pollutants of waters, worked out by American Agency of Environment Protection (US EPA) and European Union. From the environment pollution point of view, aromatic hydrocarbons, especially benzene and his aliphatic homologues are considered as very harmful. It is connected with their toxic properties, even at very low concentrations [26].

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, previously acclimatized for a single substrate. Although, each of BTX compounds can be mineralized by microorganisms, in mixture they cannot be successfully mineralized simultaneously. Moreover, even if mixed bacterial cultures are used, the presence of one compound may affect the degradation of another [7].

Many microorganisms are capable of anoxic and anaerobic BTX biodegradation. Numerous laboratory and field studies have documented that aromatic compounds can be degraded under nitrate reducing [12,14,16–18], sulfate deducing [2,4,5,9,24,28], iron and manganese reducing [6,35], and methanogenic conditions [9,10,36–38].

In wastewater treatment processes, nitrate serves as an electron acceptor and thus results in anaerobic biodegradation of monoaromatic hydrocarbons, associated with the processes of nitrate reduction and denitrification. Anaerobic digestion is used mainly for industrial wastewater treatment. Application of the system for municipal sewage treatment is so far very limited, the predominant reason given for this is that municipal sewage are too weak (to BOD or COD) to maintain high biomass (in the form of granules—suspended solids or fixed film) content in reactor. There are, however, some successful examples in pilot and full scale, which were stated for psychrophilic digestion (<20˚C). The process has recently been proven feasible for the treatment of a range of industrial wastewater representing a technological breakthrough for environmental management [13,32,33]. Therefore, psychrophilic anaerobic treatment can be an attractive option to conventional anaerobic digestion for wastewater that are discharged at moderate to low temperature and pointed at removal of specific contaminants, such as BTX.

In anoxic and anaerobic conditions 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 [39]. 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 be more closely approximated 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 [21,30]. Due to the low concentration of trace substances, the degradation occurs mostly as a first-order reaction, including sorption on suspended solids yields. Assuming biodegradation and sorption of organic pollutants the rate of their removal (r_r) in the anoxic activated sludge process can be determined according to first-order reaction kinetic presented by Siegrist et al. [31]:

$$r_{\mathrm{r}} = k_{\mathrm{deg}} \cdot X_{\mathrm{SS}} \cdot S_{\mathrm{dis}} = k_{\mathrm{deg}} \cdot X_{\mathrm{SS}} \cdot \frac{S_{\mathrm{tot}}}{1 + K_{\mathrm{d}} \cdot X_{\mathrm{SS}}} = \frac{k_{\mathrm{deg}}}{1 + K_{\mathrm{d}} \cdot X_{\mathrm{SS}}} \cdot X_{\mathrm{SS}} \cdot S_{\mathrm{tot}} = k'_{\mathrm{deg}} \cdot X_{\mathrm{SS}} \cdot S_{\mathrm{tot}} \quad (1)$$

with

k_{deg}—degradation rate constant (L/gX_{SS}·d),
X_{SS}—concentration of activated sludge in the tank (g/L),
S_{dis}—dissolved concentration (μg/L),
K_{d}—sorption constant (L/gX_{SS}),
S_{sorb}—concentration sorbed onto activated sludge, per unit reactor volume (μg/L),
$S_{\mathrm{tot}} = S_{\mathrm{sorb}} + S_{\mathrm{dis}} = S_{\mathrm{dis}}\,(1 + K_{\mathrm{d}} \cdot X_{\mathrm{SS}})$—sum of dissolved and sorbed concentration.

This report describes a series of laboratory tests of anoxic and anaerobic wastewater treatment processes that were conducted to estimate the efficiency of BTX removal. Our main objective was to find out if there was a difference in BTX removal in anoxic activated sludge process and psychrophilic anaerobic digestion.

2. Materials and methods

The laboratory investigations of the anoxic 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 .The treatment plant was designed and operated for enhanced biological nitrogen and phosphorous

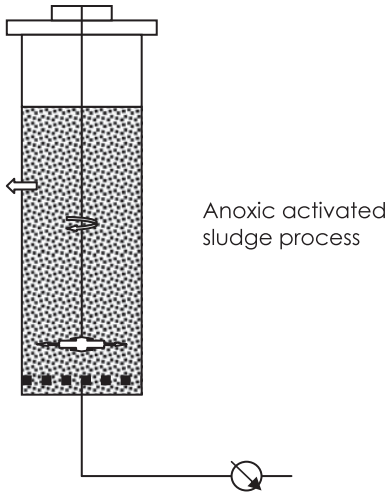

Fig. 1. Scheme of the anoxic laboratory reactor.

removal. Fifteen batch series were carried out in the laboratory anoxic reactor, 5 L in volume (Fig. 1). The content of activated sludge (TSS) was maintained at about 5 g/L (VSS 3.5 g/L) and concentrations of BTX in treated wastewater were in the range 0.25–and 2.37 mg/L of each of the compounds. The real concentration of BTX was measured at the beginning of consecutive experimental runs. The hydraulic retention time (HRT) was 18 h. The contents of the reactors during anoxic treatment process were continuously mixed by the application of slow mixing devices.

The investigations of the anaerobic treatment process have been performed with the use of anaerobic sludge acclimatized to metanogenic conditions and municipal wastewater taken from a municipal treatment plant in Tychy (Table 1). The treatment plant was designed and operated for enhanced biological nitrogen and phosphorous removal, too. Seven batch series were carried out in the psychrophilic anaerobic reactor, 2.5 L in volume. The HRT amounted to HRT 72 h and HRT 144 h. The content of activated sludge (TSS) was about 0.8 g/L (VSS 0.6 g/L) and concentrations of BTX in treated wastewater were in the range 0.6–1.1 mg/L of each of the compounds for HRT 72 h and 1.1 and 1.8 mg/L of each of the compounds for HRT 144 h. The contents of the anaerobic reactors were mixed twice a day.

After treatment, the BTX were determined in the samples with 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. Cleared sewage samples (after sludge sedimentation of 30 min) were analysed for temperature, pH, COD, ORP, phosphorus ($P–PO_4^{3-}$) and nitrogen ($N–NH_4^+$). All analyses were carried out according to Standard Methods [8].

Table 1
General characteristic of substrates used in the experiments

Parameters and units	Municipal wastewater	
	Anoxic process	Anaerobic process
pH	6.9–7.4	6.9–7.2
ORP (mV)	–323 to 178	–230 to –373
COD (mgO_2/L)	134–502	420–865
$N–NH_4^+$ (mg/L)	10–48	63–91
$P–PO_4^{3-}$ (mg/L)	2–11	22–33

3. Results and discussion

3.1. Anoxic activated sludge process

For raw sewage the BTX concentrations have been measured and the highest values were observed for toluene, up to 290 μg/L. The concentrations of benzene, *o*-xylene and *p*-xylene reached the range between 8 and 30 μg/L. In laboratory experiments, theoretical concentrations of BTX in wastewater before anoxic treatment processes were in the range between 0.25 and 1.25 mg/L of each compound (total BTX concentration 1.0–5.0 mg/L).

The total concentration of the aromatic hydrocarbons used for anoxic treatment investigations amounted from 0.8 to 5.3 mg/L. Lower than theoretical concentration has resulted from inaccuracy of doses 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 anoxic treatment process, the concentrations of BTX (the sum of aromatic hydrocarbons) in cleared wastewater decreased on average by 59%, while 44% decrease was observed in the samples with activated sludge. The average concentrations of BTX in the investigated wastewater before and after anoxic treatment process are shown in Fig. 2. The difference in concentrations between the treated wastewater with activated sludge and without sludge, indicates that approximately 26% of BTX was sorbed on the activated sludge particles. The average decrease of individual aromatic hydrocarbons was 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.

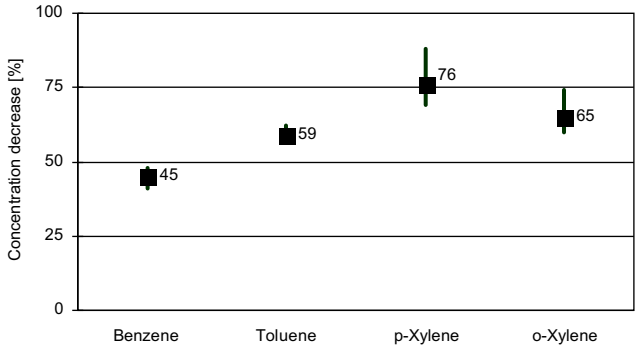

Fig. 2. Decrease of BTX concentration after anoxic treatment process.

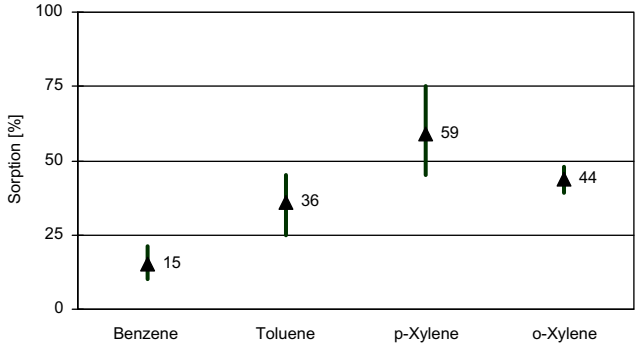

Fig. 3. Sorption of BTX on activated sludge particles.

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 as well as in the parallel samples of wastewater before sedimentation. The largest values of sorbed compounds were noticed 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 xylene isomers [40].

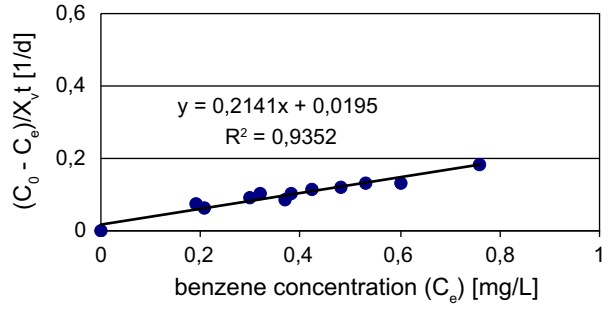

Fig. 4. The removal rate of benzene from wastewater under anoxic conditions (constant k').

The obtained dates showed that the sorption was important for the removal of BTX from wastewater during anoxic process. The results also shows that the benzene was the least sorbed on activated sludge particles. Under these conditions, the highest removal efficiency 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 [41]. 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 [42]. Also, the process can be important for efficiency of biological wastewater treatment processes, both for the removal of aromatic hydrocarbons and other contaminants present in wastewater.

Removal of BTX in the anoxic activated sludge process was analyzed based on specific substrate utilization rate. An abbreviated form of Monod equation, reduced to a first-order reaction was used in describing the kinetics of biodegradation of hydrocarbons. First-order kinetics are much more popular than Monod kinetics, because of its sensible approach at low substrate concentrations. In the investigations, removal rate constants (k') were determined, based on the concentration of test substances in the effluent after wastewater treatment process in anoxic conditions. BTX removal rate constants were determined by the graphical method as the slope straight line in the system: substrate concentration in the effluent (C_e [mg/L]) and substrate removal rate ($(C_0 - C_e)/X_{SS} \cdot t[1/d]$). The values of first-order removal constant k' considering biodegradation and sorption of BTX on the activated sludge particles have been determined for each analyzed hydrocarbon. The average values of 0.2141 mg/L·d for benzene, 0.2517 for toluene, 0.131 for *o*-xylene, and 0.133 for *p*-xylene have been estimated. A graphical display of the BTX biodegradation rate constants (k') is shown below in Figs. 4–7.

Removal of BTX in the process of biological treatment is a function of biodegradation rate constant and concentration of active biomass (X_V) in the reactor [20]. Complete removal of volatile substances occurs when biodegradation rate constant reaches a value of approximately 100 L/g·d and biodegradation is the dominant process. Biodegradation rate constant values at the level are achieved under aerobic conditions in which biological degradation with oxy-

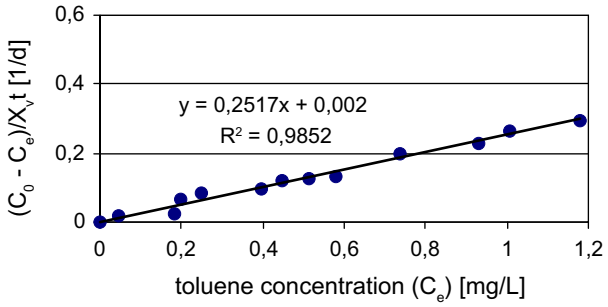

Fig. 5. The removal rate of toluene from wastewater under anoxic conditions (constant *k´*).

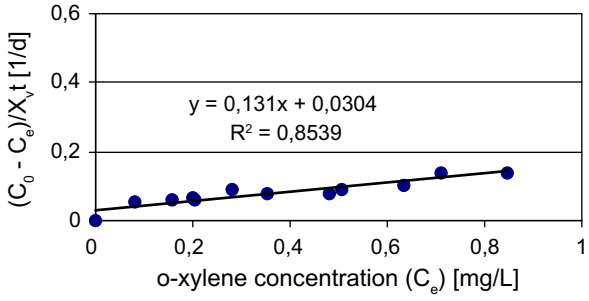

Fig. 6. The removal rate of o-xylene from wastewater under anoxic conditions (constant *k´*).

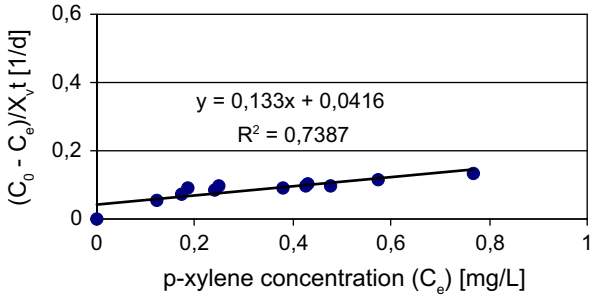

Fig. 7. The removal rate of p-ksylene from wastewater under anoxic conditions (constant *k´*).

gen as an electron acceptor is relatively easy and fast, practically for BTX. Hsieh [20] showed that the increase in the concentration of active biomass raises the overall efficiency of removal of volatile substances from wastewater, and also a significant increase in efficiency of biological treatment and decreases the stripping of the BTX into the atmosphere.

3.2. Psychrophilic wastewater digestion

Anaerobic psychrophilic digestion was carried out as a preliminary process for wastewater treatment. The wastewater used in the experiments were characterized by the presence of BTX concentrations ranging from 0.6 to 1.1 mg/L for the rector of HRT 72 h and from 0.6 to 1.8 for the reactor of HRT 144 h. In the conditions applied, the BTX removal was achieved in a more differentiated range than in anoxic conditions. However, somewhat surprising was the greater removal of BTX achieved in the case of wastewater in the reactor with a shorter retention time (HRT 72 h). The average removal of BTX in psychrophilic digestion in the reactor with HRT 72 h was 58% for benzene, 63% for toluene, 48 for the *p*-xylene, and 44% for *o*-xylene. Slightly lower values have been obtained for the wastewater anaerobically treated in the reactor with HRT 144 h, which amounted to 54% for benzene, toluene 50%, 31% for *p*-xylene, and 28% for *o*-xylene.

In the anaerobic conditions the different values of BTX removal were obtained than in anoxic conditions. Higher degrees of removal were obtained for benzene and toluene and lower for xylenes. Such a relationship could result from the other side with a much lower concentration of active biomass (about six times lower concentration in anaerobic conditions than in anoxic conditions). For the conditions of anaerobic wastewater treatment, the sorption on sludge particles was not determined, due to the low concentration of biomass and implementation of treatment in characteristic conditions for the wastewater digestion. Therefore, lower concentration of anaerobic sludge probably limited the xylenes removal from wastewater by sorption to sludge. The results of the BTX removal in the psychrophilic fermentation are also different from the data presented in the literature. In the literature, the compound which is removed in the least range is benzene. The compound 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 [9,15].

However, as demonstrated by Jo et al. [22] and Rene et al. [29], increased concentration of individual hydrocarbons in the treated wastewater stimulated the efficiency of BTX removal. Assessing individual substances separately on the efficiency of BTX removal has effected their individual toxic properties, the mechanism of decomposition and the concentrations of other hydrocarbons. Similarly to studies conducted in terms of anaerobic wastewater containing toluene, for the retention time of 24 h, Enright et al. [10] obtained the removal of toluene in the range 55–99%. These were the results comparable to those obtained

in the studies of this work. For toluene, as mentioned, the removal fluctuated between 50% and 63%. Given that, these are average values and the obtained removal of toluene was below 50%, it is believed that this could be due to the presence of other hydrocarbons in the effluent from the group of BTX, especially benzene, which inhibited the efficiency of removal of other substances present in the mixture [22,27,29]

4. Summary

The batch tests have showed enhanced BTX removal abilities under anoxic and anaerobic wastewater treatment. Analysis of the removal and biodegradation of BTX in anoxic conditions with high activated sludge concentration is an issue so far unprecedented in scientific reports, but important for the realization of biological wastewater treatment and elimination of substances as BTX. Sorption of these substances on activated sludge particles and their partial biodegradation under anoxic and anaerobic conditions can make a significant contribution to reducing atmospheric emissions of these substances in aerobic wastewater treatment processes as well as increase the overall efficiency of their removal from wastewater. The results of this study indicated the feasibility of anoxic and anaerobic conditions for the treatment of BTX-contaminated wastewater.

Acknowledgement

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