THE IMPACT OF AROMATIC HYDROCARBONS (BTX'S) ON SEWAGE QUALITY TREATED BY THE BIOLOGICAL METHOD

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Abstract In two medium size municipal sewage treatment plants designed and operated according to the enhanced biological nutrients removal process the presence of toluene, benzene, p-xylenes and o-xylenes have been investigated. Attention was given to the process of VFA production in witch as it was confirmed generation of primarily toluene takes place. Extremely high concentrations of toluene in the order of 35 000 μg/l are possible to be generated. Laboratory investigations on biological two step treatment - anaerobic - aerobic, in which BTX's were added in the range of 250 to 750 μg/l have been carried out. Such high concentrations of BTX's do have a negative effect on COD removal and the nitrification process efficiency. It was also stated that the presence of BTX's can be favourable for phosphorous removal.

Keywords: biological treatment; dephosphatation; nitrification; volatile fatty acids; BTX.

INTRODUCTION

Volatile Fatty Acids (VFA) production is often the crucial step for effective biological nutrients removal. The Enhanced Biological Nutrients Removal Processes (EBNRP) are working according to many different schemes, and the required VFA are produced in different places an procedures, from accidental production in primary settlers or thickeners, to specifically designed hydrolyses tanks. A primary or secondary sludge retention time for effective hydrolyse, i.e. production of optimal amounts of VFA is assumed to be 2 to 8 days (Oleszkiewicz and Barnard, 1997). Concentrations in the range of 3500 to 4000 mg/l of VFA expresses as acetic acid can be achieved. Almost no attention is given to other hydrolyses products of sludge organic substrate. It was found that in parallel to VFA production toluene formation occurred (Mrowiec et al., 2005).

Toluene and other aromatic hydrocarbons, as benzene and xylenes (BTX) are compounds that often occur in raw municipal effluents in the range from at detectable levels to 115 μg/l (Bell et al., 1988; Namkung and Riittman, 1987). By that time however, the presence of the aromatic hydrocarbons at much higher concentration was assigned only to external sources. The possibility of toluene bio-synthesis under anaerobic sludge digestion condition in the acidic phase was later proven (Suschka et. al., 1996; Mrowiec et al., 2005).

Further investigations have demonstrated that in the process of VFA production in several EBNRP, in average toluene was present at the level of 240 μg/l, p-xsylene 8 μg/l and o-xylene10 μg/l.

The paper elucidate further the possible quantity of BTX's to be produced in the process of VFA raising and the possible impact on sewage treatment effectiveness, especially for COD removal and nitrification.

EXPERIMENTAL METHODS

Substrate

Real municipal sewage and synthetic wastewater was used in order to show the connection to reality in two selected medium size municipal treatment plants, and perform correct laboratory experiments. Both treatment plants were designed for enhanced biological nitrogen and phosphorous removal. The treatment plants are using supernatant from primary digested sludge in order to enrich the sewage with easily biodegradable substrates (VFA) for the activated sludge microorganisms.

Laboratory apparatus and procedure

Primary sludge and excess sludge was retained for several day in anaerobic conditions in order to asses the amount VFA and toluene production in the laboratory conditions.

The investigations of biological treatment processes have been performed with the use of real activated sludge and raw municipal sewage or synthetic wastewater. Twelve series of biological treatment processes were carried out in laboratory reactors. Anaerobic and aerobic conditions were applied for experiments. The model of the reactor is shown in Fig. 1. In each step of treatment the hydraulic retention time was about 20 hour. The active volumes of the reactors were 5 litters. Two parallel tanks were working. To the first reactor benzene, toluene, p-xylene and o-xylene (BTX) dissolved in methanol were added. The content of the anaerobic reactors were continuously mixed by slow mixers. After sedimentation for 30 minutes, samples for analyses have been taken. The aerobic reactors were supplied by pressure air. Daily measurements of temperature, chemical oxygen demand (COD), pH, oxidation-reduction potential (ORP), dissolved oxygen, phosphorus (P_{Total}, $P\text{-}PO_4^{3-}$), nitrogen (TKN, $N\text{-}NH_4^+$, $N\text{-}NO_3^-$) and BTX concentration have been done. Determinations were done according to the Standard Methods (1995).

Results and discussion

Sludge samples were taken from a primary settling tank (plant A) and from a sludge hydrolyses tank (plant B), and next kept in laboratory under anaerobic conditions for 10 days.

According to expectations VFA have been released. The maximum obtained concentration of the sum of acetic, propionic and butyric acids, in case A was about 1500 mg/l, and in case B above 2500 mg/l in the process of hydrolysis (Fig. 2). The maximum values have been obtained respectively after 5 and 7 days. The supernatant from digested sludge for 7 days, were used in two series of laboratory experiments supplementing the sewage with easily biodegradable substrates (VFA).

Figure 1. Laboratory biological sewage treatment reactors.

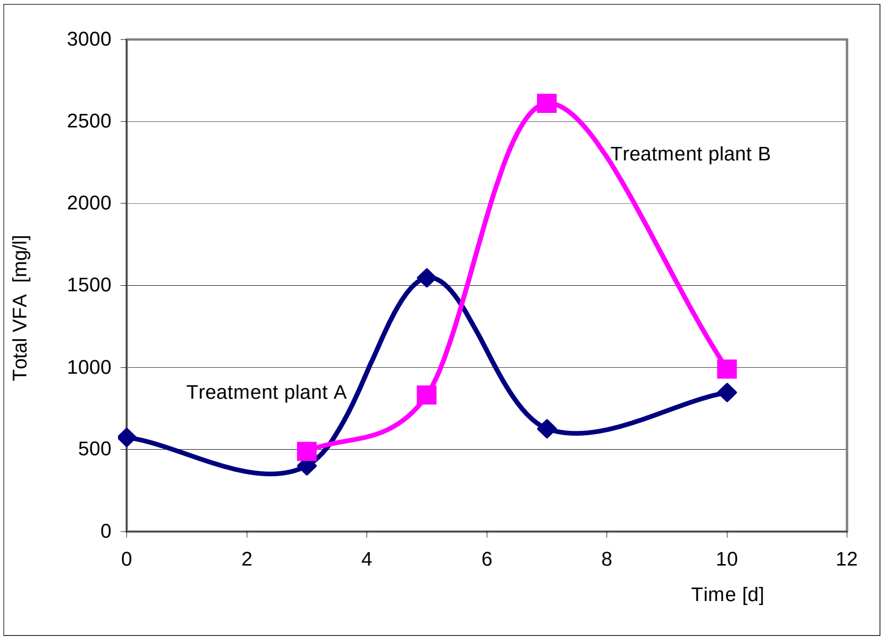

Figure 2. Release of VFA under anaerobic conditions.

In parallel to the release of VFA toluene was bio-synthesized (Fig. 3). The concentration of toluene increased to 32 930 μg/l and 37 524 μg/l respectively for plant A and B. Such high concentrations have not been measured directly at the treatment pants. The highest concentration measured in plant A was only 288 μg/l of toluene in the sewage before biological treatment process.

In order to be closer to real conditions the effects on biological treatment have been investigated with sewage taken from a municipal treatment plant and synthetic sewage spiked with toluene in the range of 250 to 750 µg/l. Although benzene, p-xylene and o- xylene was measured at treatment plants at distinctively lower concentrations (the maximum value was for benzene equal to 29 µg/l), they have been added in the similar to toluene range of concentration. The concept was to see better the possible effects of the BTX presence.

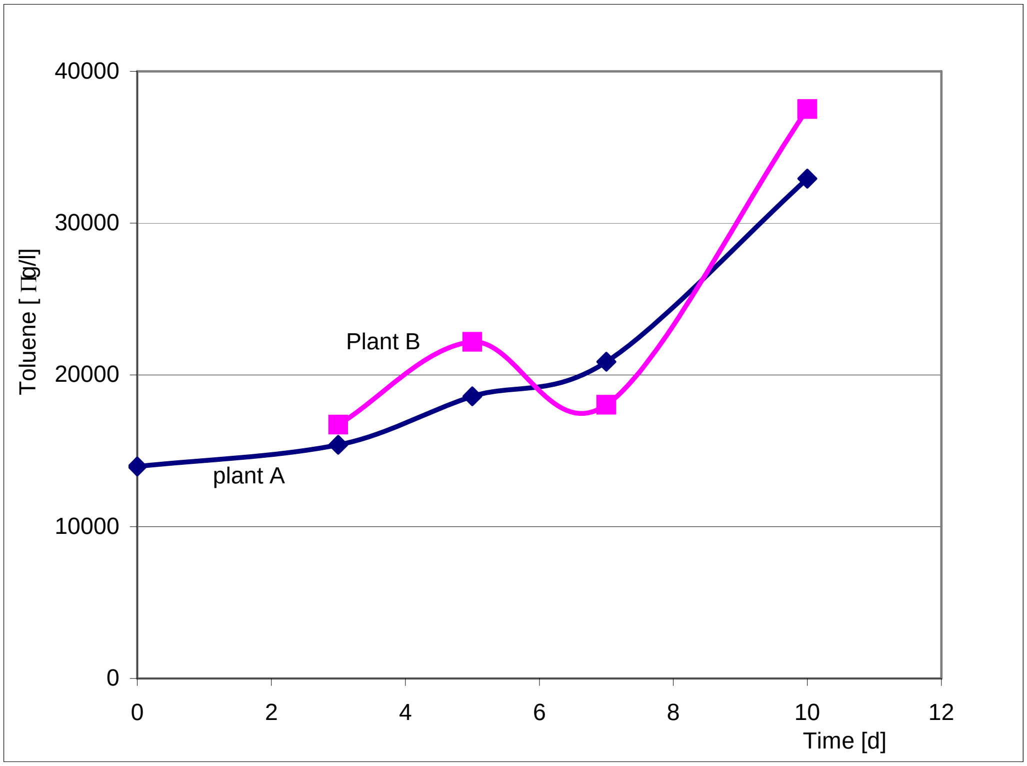

Figure 3. Toluene bio-synthesis in sludge in the anaerobic conditions (laboratory experiments).

The main aim of the laboratory investigations has been evaluation of BTX presence impact on COD and phosphorus removal as well as on the nitrification process efficiency.

The different values of COD in raw sewage have been investigated. For series I to VI the COD vales of the synthetic sewage were in the range from 115 to 490 mgO_2/l. For series VII to X, real municipal sewage was used with COD values ranging from 136 to 205 mgO_2/l. In addition two series XI an XII have been performed with the addition of digested sludge supernatant. In these two series the mixture had COD values respectively 1217 and 1157 mgO_2/l.

Comparing the effectiveness of COD removal for sewage (synthetic or natural) with and without the addition of various concentrations of BTX, a negative effect of their presence was found. The effects of COD removal for municipal sewage or synthetic sewage, without the addition of BTX's were in the range between 70 to 95 %, while with the presence of BTX's the COD removal effects decreased distinctly to the range of 35 to 75 %.

The negative effect of the presence of BTX in supplied sewage can be attributed to the very high increase of COD in the sewage after the first anaerobic stage of treatment. That can be clearly seen in Fig. 4 and Fig. 5, showing variation of COD in the different steps of treatment.

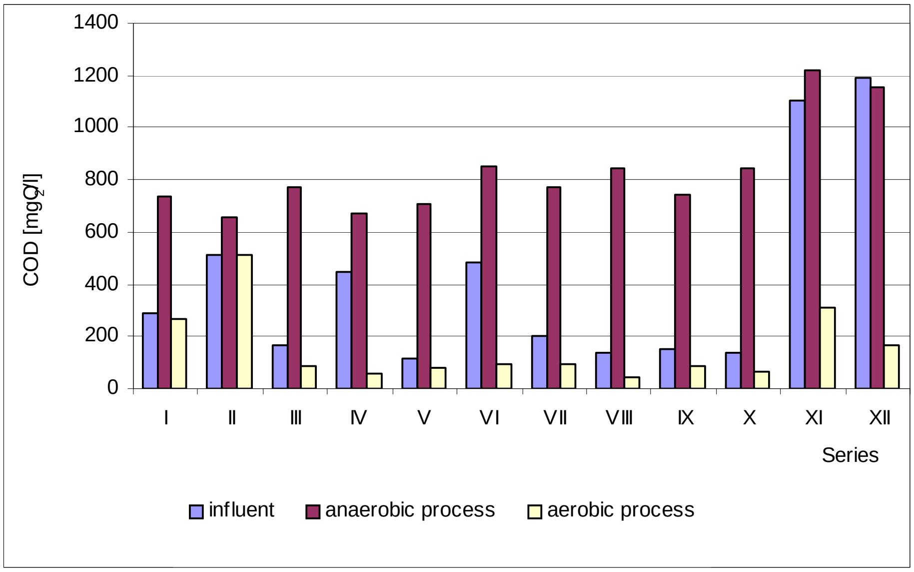

Figure 4. COD values in sewage with BTX addition at the various stages of treatment.

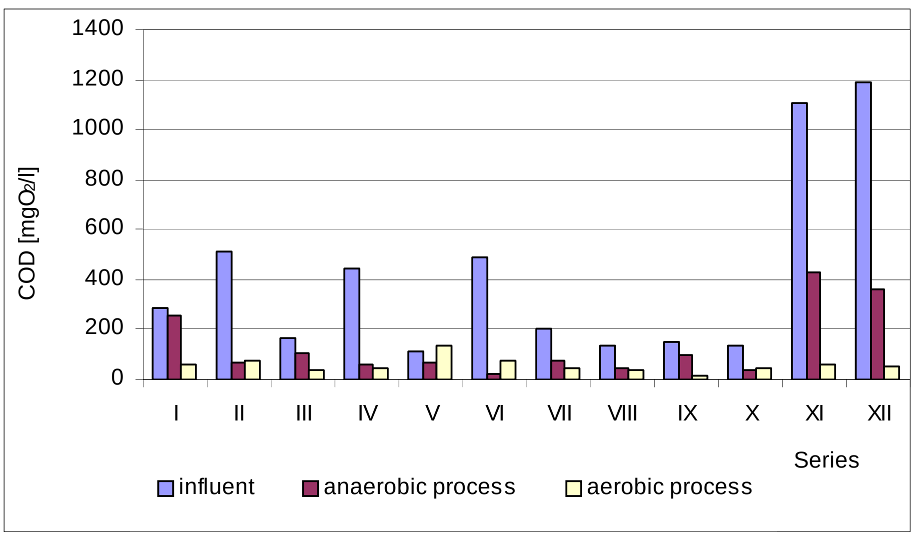

Figure 5. COD values in sewage without BTX addition at the various treatment stages (blank).

In contrary, and surprisingly to some extend, the presence of BTX's in the influent has contributed to higher effects of phosphorous removal. The remaining concentration of total phosphorous and phosphates after the aerobic stage of treatment was definitively lower in the case of sewage to which BTX's were added. Fig. 6 shows that especially with higher phosphates concentrations in the influent the differences are distinct.

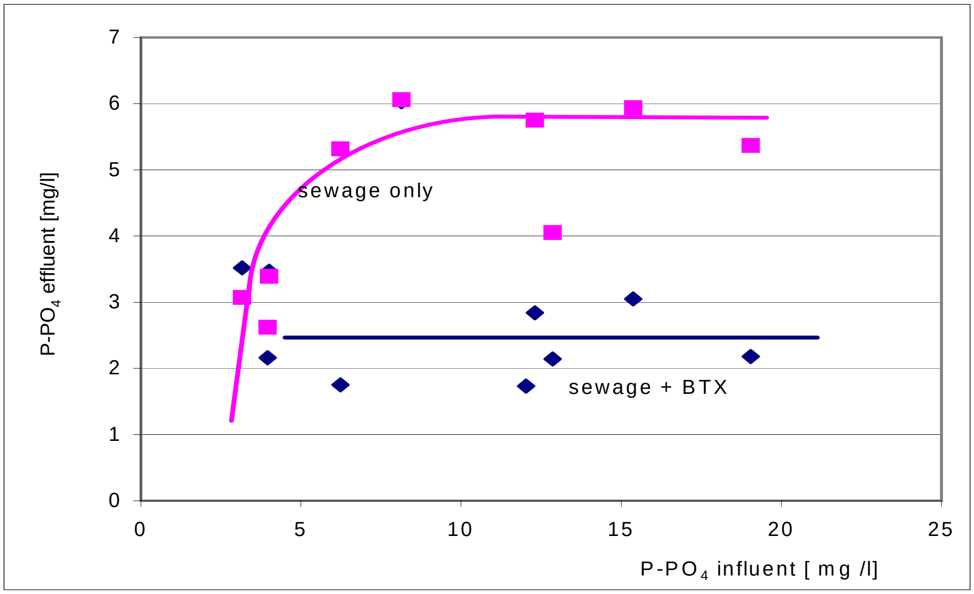

Figure 6. Remaining phosphates after two steps: anaerobic- aerobic treatment

Probable the high values of COD after the first anaerobic step of biological treatment process, can be assumed to be easily biodegradable and be a favorable substrate for phosphorous accumulating bacteria. Definitively that subject has to be further investigated and clarified. The correlation between the concentration of P_{Tot} in the effluent and BTX's concentration is showing clearly the positive effect of their presence. The correlation is shown in Fig. 7. The observed correlation requires to be better explained on the base of further investigations.

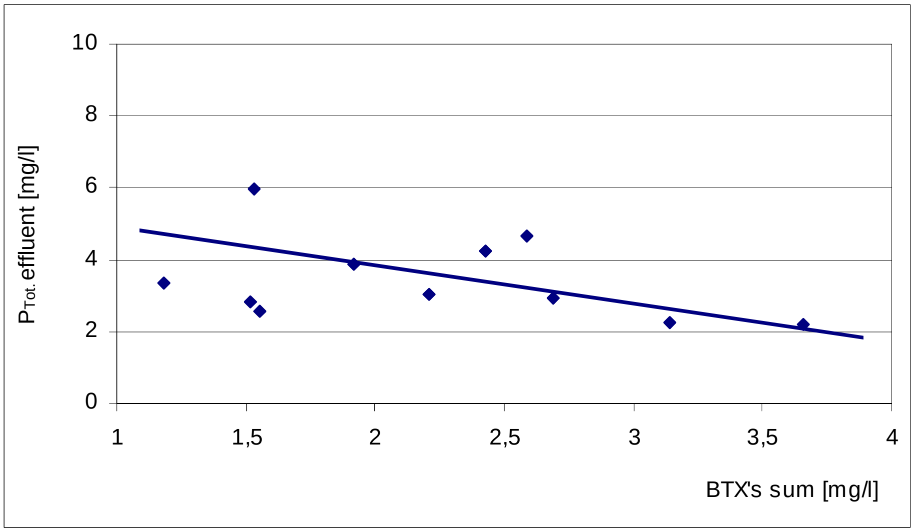

Figure 7. Correlation between P_{Tot} concentration in treated sewage and BTX concentration in raw sewage, based on laboratory experiments.

The laboratory investigations have been performed in the anaerobic (I-step) and next in the aerobic (II- step of biological treatment) conditions. Therefore only nitrification process can be analysed in the case of nitrogen transformations. After the anaerobic treatment, the concentration of ammonia nitrogen was measured as the effect of amonification process and after the aerobic conditions concentrations of ammonia nitrogen and nitrate nitrogen were determined as the final products. Only for five series the Kjeldahl nitrogen was measured for raw and treated sewage. With COD increase in raw sewage an increase of ammonia nitrogen concentrations were observed. It was conformable to assumption, especially for synthetic sewage and in the case of supernatant addition. The correlation between COD and ammonia nitrogen concentrations in raw sewage is shown below in Fig. 8.

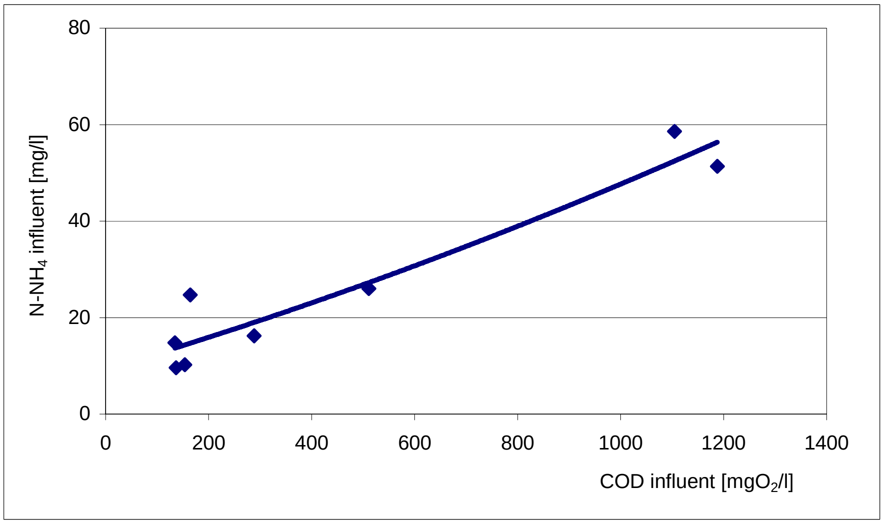

Figure 8. Correlation between COD and ammonia nitrogen concentrations in raw sewage (laboratory investigations).

The nitrification processes was limited in the analysed samples to which BTX's were added. The nitrate nitrogen concentrations in sewage after aeration stage in all samples were from 0 to 8 mg/l. In the case of higher ammonia nitrogen concentrations in raw sewage (series XI and XII) higher concentrations of ammonia nitrogen in the effluent were observed. It is shown in Fig. 9. For lower ammonia concentrations the nitrification process was more pronounced, and the dominating form of nitrogen were nitrates. In the case of samples without BTX's only for very high ammonia nitrogen concentrations (over 100 mg $N\text{-}NH_4^+/l$) in raw sewage the ammonia nitrogen concentrations in treated sewage were on the same level as nitrates (Fig. 10).

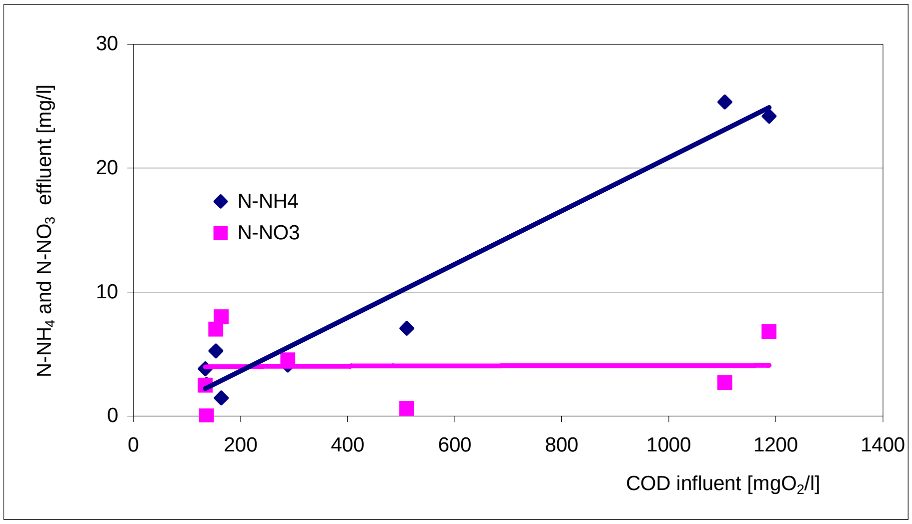

Figure 9. Dependence of ammonia and nitrate nitrogen concentrations in the treated sewage from COD values in raw sewage with BTX's.

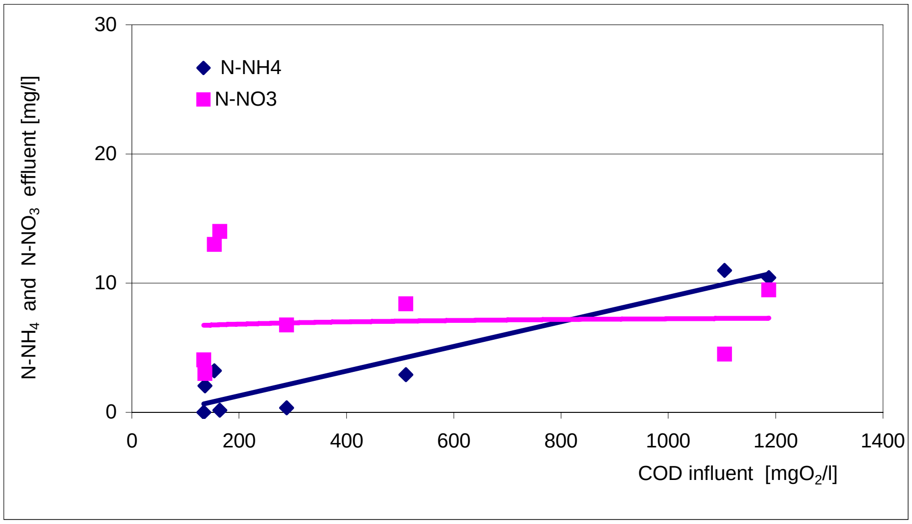

Figure 10. Dependence of ammonia and nitrate nitrogen concentrations in the treated sewage from COD values in raw sewage without BTX's .

The presence of BTX in sewage treated biologically, can have a negative effect and limit the nitrification process. For comparison the quantity of ammonia nitrogen oxygenated under aerobic conditions in the sewage with and without BTX's is given in Fig. 11. For high ammonia nitrogen concentrations in raw sewage the differences have been greater, and the highest concentrations of ammonia nitrogen in the treated sewage have been measured.

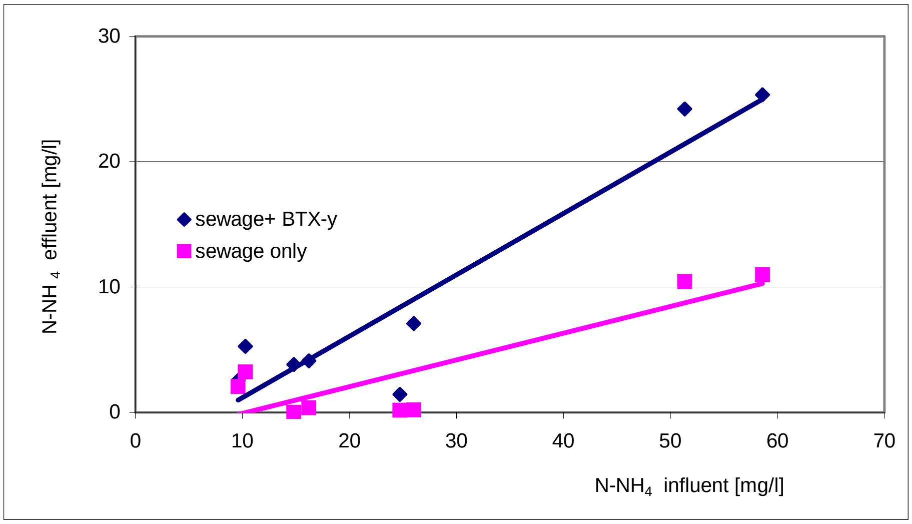

Figure 11. Comparison of ammonia nitrogen quantity oxygenated in the aerobic conditions with and without BTX's.

The differences in the nitrification process efficiency were distinctly shown for Kjeldahl nitrogen concentrations. The nitrification process has been adversely affected in all analysed samples (Fig. 12).

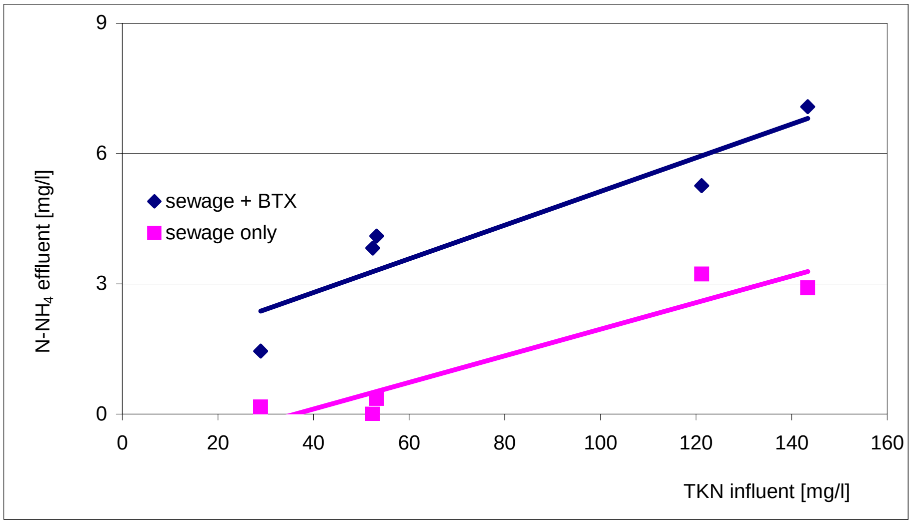

Figure 12. Effect of Kjeldahl nitrogen oxidation in the aerobic conditions in samples with and without BTX's.

CONCLUSIONS

The presence of BTX's (toluene, benzene, p-xylene and o-xylene) in municipal sewage has so far not raised sufficient attention.

Previous and present investigations have demonstrated the phenomenon of bio-synthesis, mainly of toluene under anaerobic conditions.

It was shown, that in the step of VFA production being a part of the EBNRP system, relatively high concentrations of toluene may also be produced and next discharged to the bioreactors with the sludge supernatant.

The presence of BTX's in treated sewage, arising in the hydrolysis step for VFA production can affect negatively COD removal and efficiency of nitrification process. It can result in higher ammonia nitrogen concentration in treated sewage.

However, due to the hydrolysis of organics in the anaerobic stage to simpler products being apparently a favourable substrate for phosphorous accumulating bacteria, better effects of phosphorous are possible.

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