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Spatial distribution of pharmaceuticals in conventional wastewater treatment plant with Sludge Treatment Reed Beds technology

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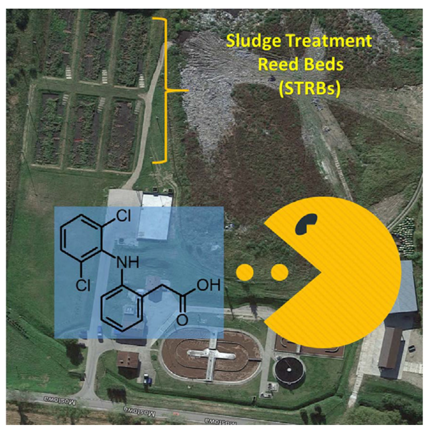
HIGHLIGHTS

- Analyzed WWTP was very effective in removal of basic pollutants.
- Pharmaceuticals distribution in WWTP +STRB differs between season and chemical type.
- Ibuprofen, naproxen and paracetamol were eliminated by conventional WWTP.
- Often higher level of diclofenac was observed in effluents compared to influents.
- Diclofenac and its metabolites were successfully removed by the STRB.

GRAPHICAL ABSTRACT



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ABSTRACT

Pharmaceutical residues are an emerging environmental problem. It is strongly confirmed that pharmaceuticals are present in soils and environmental waters (surface, marine and even groundwater), and that wastewater treatment plant (WWTP) effluents are the main source of pharmaceuticals in the watershed. The aim of this study was to recognize the spatial distribution and seasonal changes of selected pharmaceuticals in conventional WWTP with Sludge Treatment Reed Beds (STRBs) technology used for dewatering and stabilization of sewage sludge, because these systems have never been studied in terms of pharmaceuticals distribution or removal potential.

The research was conducted in conventional WWTP in Gniewino, where raw wastewater was treated using mechanical, biological and chemical removal of the organic matter and nutrients, and sewage sludge was treated with STRB. Determinations of pharmaceuticals (non-steroidal anti-inflammatory drugs - ibuprofen, paracetamol, flurbiprofen, naproxen, diclofenac and its metabolites) and basic parameters were carried out in samples of influent and effluent from WWTP and in the liquid phase of surplus activated sludge (SAS) as well as reject water from STRB.

The potential of removal varied among target pharmaceuticals. Ibuprofen and naproxen were completely removed by the standard applied technology of the Gniewino WWTP. Diclofenac and its metabolites were the chemicals with the lowest removal potential in wastewater and the highest detection frequency. These pharmaceuticals were also detected in the liquid phase of SAS as well as in reject water. However, removal potential when using STRB was higher than 94% (mostly higher than 99%), independent of the season. Indeed, the STRB

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technology is not only efficient in sludge dewatering and nutrient removal (primary purpose), but also elimination of polar pollutants. Nevertheless, removal in STRB did not mean that pharmaceuticals were totally eliminated because these compounds could be "trapped and stored" in beds (by the process of sorption) or transformed into other products. This study is a starting point for further exploration of STRB technology for elimination of emerging pollutants.

1. Introduction

Over the last decade, the scientific community has been focused on the presence of pharmaceuticals in the environment. Pharmaceuticals are a large group of bioactive chemical compounds used in veterinary medicine, animal farms and in medicine. They represent a diverse group of water pollutants that are not systematically monitored and can cause negative effects in the environment (Farré et al., 2008). The research indicates that pharmaceuticals are present in surface waters, sea water and even in groundwater (Borecka et al., 2015; Boxall et al., 2012; Caban et al., 2015; Farré et al., 2008; Watkinson et al., 2009), though the highest concentration of pharmaceuticals can be found in raw wastewater (inflow to municipal wastewater treatment plants - WWTPs) (Biel-Maeso et al., 2018; Sim et al., 2011; Verlicchi et al., 2012). Jarosova et al. (2012) undertook the investigation of the presence of pharmaceuticals in seven headwaters flowing through relatively unpolluted areas of the Czech Republic, a small country with a relatively low density of population (Jarosova et al., 2012). It was found that the WWTPs are the most significant source of pharmaceuticals in water bodies. Other sources were practically negligible. This was confirmed by many other research projects (Arlos et al., 2014; Zorita et al., 2009). It was also found that the distribution of the contaminations, including pharmaceuticals, was highly dependent on the treatment process and effluent quality (Arlos et al., 2014).

According to Verlicchi et al. (2013) the range of pharmaceutical concentrations in raw wastewater is from 10^{-3} to 10^{2} μg/l and even more, and common WWTPs are not able to efficiently remove all of them from liquid effluent as well as sludge. It was observed that removal efficiencies varied in a wide range for the different compounds, as well as for the same substance, due to the different chemical and physical characteristics and to operational conditions. Other research indicated that the total concentration of the individual pharmaceuticals (except carbamazepine and crotamiton) in the influent was efficiently removed by 80% during the biological treatment. It was also found that the total concentrations in the effluent from conventional activated sludge process were 1.5 times higher than that from biological nutrient removal process (Okuda et al., 2016). Many research confirms that ibuprofen is nearly completely removed from wastewater in conventional WWTPs (removal rates >90%) (Clara et al., 2005; Paxéus, 2004; Joss et al., 2005). The lower removal efficiency was found in case of naproxen (80%) and diclofenac (39%) (Clara et al., 2005). Other studies show the lower efficiency of pharmaceuticals removal in conventional activated sludge processes. According to Tiwari et al. (2017) the removal rates of ibuprofen and naproxen are common ranges between 75% and 85% and 50–60%, respectively. Diclofenac revealed low and varied removal rate ranging from 10 to 50%.

The processes occurring in Sludge Treatment Reed Beds (STRBs) are similar to those in constructed wetlands (CWs). According to Carvalho et al. (2016), CWs present similar or better removal of pharmaceuticals compared to conventional WWTP systems. The pharmaceuticals are removed mostly thanks to (i) degradation in a hydroponic medium vegetated by wetland plants, (ii) uptake by the wetland plants, and (iii) degradation in CW mesocosms.

Chen et al. investigated pharmaceuticals in wastewater from rural areas treated in CWs located in the Czech Republic (Chen et al., 2016). The removal efficiencies of pharmaceuticals and personal care products (PPCPs) in the rural CWs exhibited large variability with 11–100% for anti-inflammatories, 37–99% for β-blockers and 18–95% for diuretics. The statistical results revealed significant correlations between removal efficiencies of some PPCPs and removal efficiencies for organic matter, ammonia and phosphorus (Chen et al., 2016). Other research (Vymazal et al., 2017) of wastewater treated in CW indicated wide variation in removal efficiency among systems as well as among pharmaceuticals. The highest average removal was found for paracetamol (91%). Moderate removal was found for ibuprofen. Diclofenac removal was insufficient and did not exceed 50%. Matamoros et al. also confirmed that diclofenac was not effectively removal in CWs (Matamoros et al., 2009). Although efficiency of pharmaceuticals removal in CWs is rather well know, their removal in STRBs has not been studied.

STRB technology offers simultaneous dewatering and stabilization of sewage sludge taken from conventional WWTPs. These systems are used for treatment of sludge from very small single-family WWTPs (for a few persons) to big WWTP (for example Kolding STRB for 125,000 pe - personal equivalent), but mostly they are useful for medium-size WWTPs (Nielsen, 2003).

STRB technology is based on the same processes that occur in natural wetland ecosystems. STRBs are built as concrete constructions or as tight tanks placed in the ground. The whole system is divided into several beds planted with reeds. The long-term experiences indicate that in medium or big WWTPs the number of beds should be at least eight. STRB technology consists of periodical loading of sludge with low content of dry matter (0.5–1.5%) (Kołecka and Obarska-Pempkowiak, 2008; Nielsen, 2003). The time of loading typically takes about 3–7 days. After discharging of sludge onto a bed, time for its dewatering (so-called resting time) is needed, therefore sludge should be loaded onto another bed. The resting time is about 21–49 days (Brix, 2017). The sludge is stored in system for about 10–15 years. After this time it is removed from the system and can be used as fertilizer (Kołecka and Obarska-Pempkowiak, 2013; Nielsen, 2011).

STRBs are especially useful in rural areas and housing estates where economic considerations limit the use of expensive mechanical equipment. These systems can be established in any area and are simple to build and operate. Their low energy consumption is their main advantage. Additionally, they do not require addition of chemicals for improvement of dewatering capability (Kołecka et al., 2017).

Research shows that sludge dewatering efficiency in reed systems is comparable to that of mechanical equipment such as a filter press (content of dry matter can even reach up to 40%). It has also been proven that sludge after long-term treatment in STRBs is stabilized and has a chemical composition similar to that of humus. Additionally, it was proven that the obtained product is safe with regard to its microbiological characteristics (Nielsen, 2007).Unlike most other conventional methods, reject water from STRBs released from the sludge during dewatering is treated as it percolates through the bed (Brix, 2017; Nielsen, 2007).

The secondary function of STRBs could be the removal of hazardous pollutants, for example pharmaceuticals, which are classified as new emerging pollutants with a global awareness statute (Gavrilescu et al., 2014). It has been proposed that systems containing plants and soil can participate in elimination of pharmaceuticals and their metabolites.

The aim of this study was to recognize the spatial distribution and seasonal changes of selected pharmaceutical in conventional WWTP with STRB technology. The distribution as well as removal potential