

Adsorption of Diclofenac, Sulfamethoxazole and Levofloxacin with Powdered Activated Carbon

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Abstract. The presence of pharmaceutical residues in the receiving waterbodies of wastewater treatment plants (WWTP) and in the environment has become a global concern. We can now say for certain that, having metabolised in our bodies, partially modified or unmodified pharmaceuticals will reach WWTP. However, WWTP are not designed for the removal of such compounds. Only a small fraction of pharmaceuticals decompose during biological treatment or are adsorbed in sediment. Therefore, it is essential to find a treatment process that is capable of removing pharmaceutical residues. The aim of the present study was to research the removal of three pharmaceuticals found in the environment, namely diclofenac (DCF), sulfamethoxazole (SMX) and levofloxacin (LFX), through the use of powdered activated carbon (PAC). To this end, adsorption tests were conducted where the adsorption capacity was estimated according to the adsorbent dose and the residence time of the process. LFX had the highest adsorption rate: the removal effectiveness was 77% in a residence time of 5 minutes and in 60 minutes a stable indicator was achieved whereby 94% of LFX had become adsorbed. The worst adsorption property was observed for SMX, as 68% of SMX was adsorbed in a residence time of 60 minutes. According to the conducted tests, the Freundlich adsorption isotherms and constants characterising the adsorption were found where the DCF K was 23.8, the SMX K was 34.3 and the LFX K was 106.1. This test demonstrated that the pharmaceuticals selected for the experiment could easily be subjected to adsorption processes and could be removed by means of PAC.

Keywords: adsorption of pharmaceuticals, removal of pharmaceutical residues, diclofenac removal, sulfamethoxazole removal.

Conference topic: Water engineering.

Introduction

The presence of pharmaceuticals in the environment has become an important problem worldwide. In Germany, 131 different pharmaceuticals in sea and ground water have been found (Weber *et al.* 2014). Therefore, hundreds of scientific workgroups are working to understand the pathways of different human and veterinary drugs back into our food cycle (Miège *et al.* 2009; Li 2014; Białk-Bielińska *et al.* 2016) and to eliminate the long term effects of different pharmaceuticals on ecology (Fatta-Kassinos 2010; Klatte *et al.* 2016).

In examining different studies, one problem when searching for specific pharmaceuticals in the aquatic environment is that some pharmaceuticals are biologically degradable and form new chemical bonds (Langenhoff *et al.* 2013; Ericson *et al.* 2010). It makes analyses difficult because we simply don't know what chemical substance we are looking for or what ecotoxic effect this new chemical substance can have.

Emission of pharmaceuticals

The first step in managing the occurrence of pharmaceuticals in the environment is to understand the path of pharmaceuticals in the environment and back into the food chain. The amounts and kinds of medicines depend on the population's structure. Aging populations use more antibiotics and cardiovascular agents, while younger people use birth control pills. For different pharmaceuticals, the pathway can be different but a basic scheme is shown in Fig. 1 (Adamcza *et al.* 2012; Klatte *et al.* 2016; Weber *et al.* 2014).

In Fig. 1, the pathway of human and veterinary drugs is described. When we look the population box, we see two possibilities: the direct consumer who takes pharmaceuticals at home, including widely used medicines such as non-steroidal anti-inflammatory drugs (NSAID) and birth control pills. However, in hospitals, more effective and higher dosages of pharmaceuticals are used and are therefore described separately (Klatte *et al.* 2016). A third pathway is drugs that are used in veterinary medicine. Different antibiotics and NSAID are used to facilitate intensive animal farming (Oaks *et al.* 2004; Schwaiger *et al.* 2004). Some pharmaceuticals are not biologically degradable and pass through WWTP unchanged or are adsorbed to raw and activated sludge (ter Laak *et al.* 2010; Mohapatra *et al.* 2016). When the sludge is used as a fertiliser in agriculture, the pharmaceuticals can accumulate in plants (Lillenberg 2011; Nei *et al.* 2014). A similar cycle occurs with dung and fish faeces.

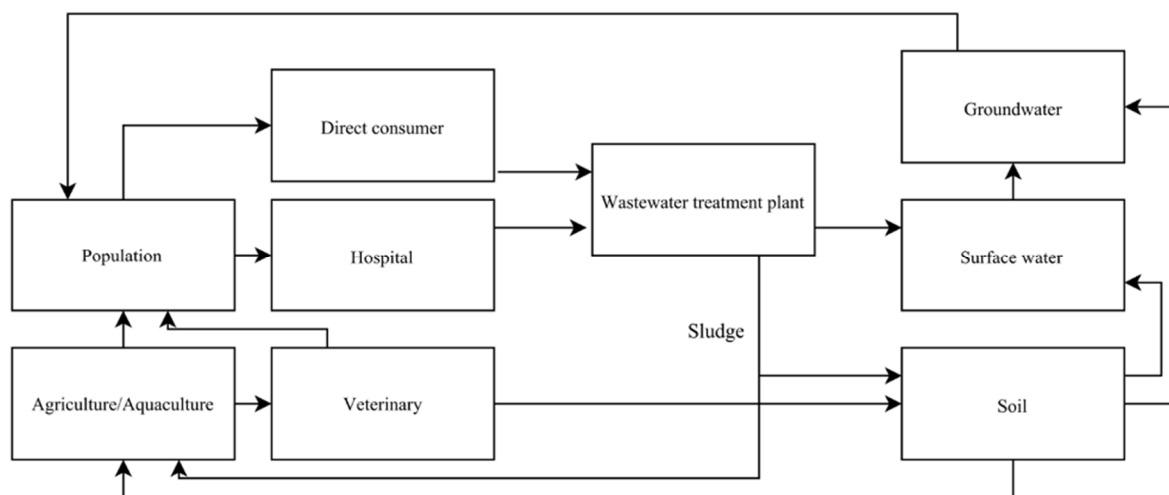


Fig. 1. Emission pathways of human and veterinary pharmaceuticals.

Risks for organisms and humans

The most widely known proven effect of pharmaceuticals on organisms is the major population collapse of white-backed vultures (*Gyps africanus*) in Pakistan and India in the early 1990s. The birds consumed the carcasses of cattle that had been treated regularly with NSAID diclofenac. For vultures, the concentration was high enough to cause kidney failure (Klatte *et al.* 2016; Oaks *et al.* 2004).

Various risk assessment methods have been developed in order to determine the most harmful drugs. In his research, Carlsson (2006) assessed the ecotoxicity risks of 27 different pharmaceuticals. Based on his study, the most dangerous drugs for the environment include diclofenac, ethinylestradiol, ibuprofen, metoprolol, norethisterone, oestriol and oxazepam (Carlsson *et al.* 2006; Lonappan *et al.* 2016). Li (2014) confirmed this data and added the antibiotic sulfamethoxazole to the list of ecotoxic pharmaceuticals (Li 2014; Sangion, Gramatica 2016).

For humans, the main concerns are bioaccumulation in our food, resistance to antibiotics and the toxicity of pharmaceutical mixtures and their effect on the human organism (Skórczewski *et al.* 2014; Fent *et al.* 2006; Klatte *et al.* 2016). Different studies show that the ecotoxic effect concentration of pharmaceutical mixtures are several times lower compared to pure active substances (Fent *et al.* 2006).

Removal of pharmaceuticals

In Fig. 1, we see two possible points for the removal of pharmaceutical residues: separately treating the effluent of hospitals or treating all water in a WWTP. Most European countries are examining ways to develop technology to remove pharmaceuticals at the WWTP stage because pharmaceutical residues are not the only problem (Nazari *et al.* 2016; Giannakis *et al.* 2015; Cruz-Morató *et al.* 2014). Many promising technologies to remove organic pollutions and heavy metals in the same treatment stage have already been determined.

In the advanced oxidation ozone, hydrogen peroxide or UV (UV+TiO₂) is used to degrade pharmaceutical residues from wastewater. However, it is disturbing that we don't in fact know the degradation products and the possible derivatives that the residues can form (Giannakis *et al.* 2015; Alvarez-Corena *et al.* 2016).

The second possibility for treat drugs from water is membrane technology, often in combination with a bioreactor (aerobic membrane bioreactor- AMBR). While this doesn't degrade the pharmaceuticals, it can be used to concentrate the pollution after which you can use other technologies to remove the residues such as advanced oxidation or adsorption with activated carbon (Priya, Philip 2015; Arya *et al.* 2016).

The most promising technology is adsorption with activated carbon because it removes different types of organic pollutions and heavy metals (Wu *et al.* 2015; Karnib *et al.* 2014; Larous, Meniai 2016; Raki *et al.* 2015). There are three ways to add an adsorption step to an existing WWTP:

- Filtration through a granular activated carbon filter bed after secondary clarification. This removes pharmaceuticals from the effluent water, but adsorbed pharmaceuticals in the sludge are not eliminated (Chang *et al.* 2015; Lima *et al.* 2004; Rattier *et al.* 2012; Meinel *et al.* 2015).
- Adding PAC to the aeration tank. The adsorbed pharmaceuticals and heavy metals are removed with the waste activated sludge (WAS) (Zietzschmann *et al.* 2016; Chang *et al.* 2015). Because the concentration of harmful substances would be higher than normal in the sludge, the sludge wouldn't be immediately applicable for use in agriculture. One way is to use a struvite reactor for the dewatered sludge water and to subsequently burn the sludge in order to recover the phosphorous from the ash (Rattier *et al.* 2012; Meinel *et al.* 2015).

- Using PAC after clarifiers as a separate treatment step. This method requires more significant investment, because we need a mixing tank, a clarifier for PAC removal and sand filtration for final treatment (see Fig. 2) (Weimar 2014).

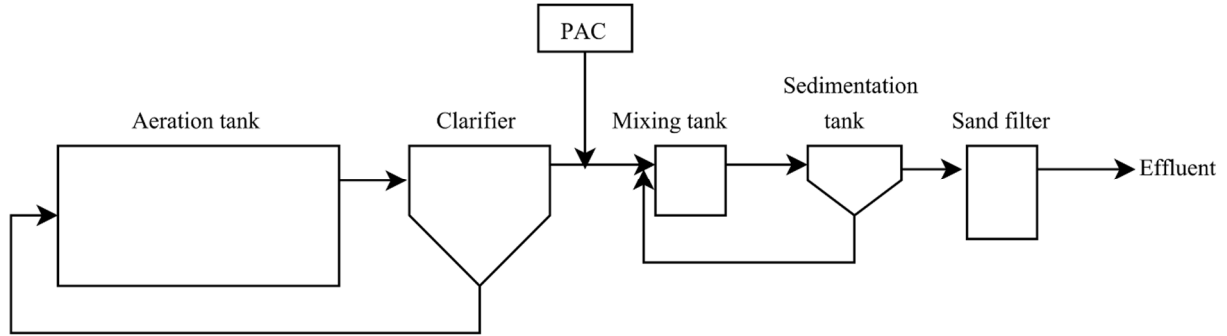


Fig. 2. Possible technical solution for PAC dosing in a conventional municipal WWTP (Weimar 2014).

The aim of this research was to study the removal potential of three common pharmaceuticals with PAC: non-steroidal anti-inflammatory drug diclofenac (DCF) and two antibiotics, sulfamethoxazole (SMX) and levofloxacin (LFX). To this end, different adsorption batch experiments were determined, and these provide information for process engineers on determining activated carbon treatment steps.

Materials and methods

For batch experiments, different solutions of DCF ($C_{14}H_{10}Cl_2NNaO_2$), SMX ($C_{10}H_{11}N_3O_5S$) and LFX ($C_{20}H_{24}O_2$) were prepared with ultrapure (ELGA) water and a known concentration of the pharmaceuticals under investigation (pH of all solutions was 7.6). All the pharmaceuticals were purchased from Sigma-Aldrich and are of lab quality. The concentration was measured as a TOC (total organic carbon). 100 ml of the solutions and 0.05 g of PAC (coal filter material silcarbon powder, Lach-Ner, M 12.01 g/mol) were mixed (stirring speed 150 rpm, at room temperature of 21.5 °C) and a sample was taken in intervals of 5, 10, 30, 60 and 120 minutes (when a stable value was achieved) in order to determine the adsorption capacity in relation to time. The same experiment was then repeated to discover the removal effectiveness and removal capacity, though different concentrations of PAC were added.

The removal effectiveness $E\%$ was calculated using the following equation:

$$E\% = \frac{C_0 - C_n}{C_0} \cdot 100\%, \quad (1)$$

where: C_0 is the initial concentration of adsorbate, C_n final concentration of adsorbate and C_e equilibrium concentration of adsorbate.

Freundlich adsorption isotherm was used to describe the adsorption process. It shows us the relationship between the adsorbate amount, what is adsorbed per unit adsorbent and the adsorbate equilibrium concentration q_e (Sekar *et al.* 2004; Kumar, Kirthika 2009):

$$q_e = K \cdot C_e^{1/n}, \quad (2)$$

where: K is the Freundlich adsorption constant (mg/g) and n heterogeneity factor. K and n are the indicators denoting adsorption capability and intensity.

The maximum adsorption capacity $q_{e_{\max}}$ dependent on time was calculated using the following relationship:

$$q_{e_{\max}} = \left(\frac{1}{q} \right) \cdot m_s, \quad (3)$$

where: q is the amount of adsorbent (g), m_s weight of adsorbed adsorbate (mg) and $q_{e_{\max}}$ maximum adsorption capacity (mg/g).

Results and discussion

Removal effectiveness

In Fig. 3, the results of the adsorption studies are shown, where the adsorption effectiveness dependant on time and adsorbent dosage were investigated. The best results were achieved in LFX removal; within 5 minutes, 77% percent had already been removed, while the highest removal rate was 94% after 60 minutes detention time. The equilibrium concentration was reached in 60 minutes. 0.03 g/100 ml was determined to be the preferable PAC dosage for this LFX concentration, for which the adsorption effectiveness was 86%. A similar removal effectiveness of 93% was reported by L. Lima (2004) (Lima *et al.* 2004). In higher PAC concentrations, the removal capacity stays similar, which means that at the initial concentration researched by us the higher PAC dose would only increase adsorbent consumption without significantly increasing the treatment effectiveness. Rapid adsorption is an indicator that we need significantly shorter residence times for creating a treatment stage of LFX with specific activated carbon than for other pharmaceuticals. Therefore, the planning of the first stage of multistage filters may be dimensioned smaller. After a residence time of 120 minutes, the concentration of the substance to be removed even increased and the removal effectiveness dropped by 2%. Similar rises in concentrations and adsorbate equilibrium (Chang *et al.* 2015), suggesting that it is due to continuous changes in the adsorbent and adsorbate equilibrium (Chang *et al.* 2015).

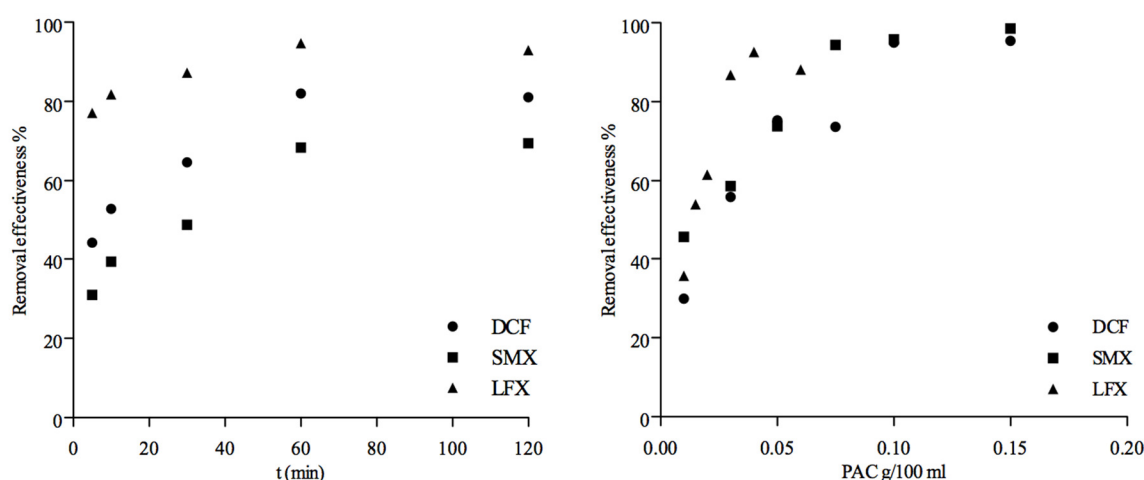


Fig. 3. Removal effectiveness depended on adsorption time (a) and adsorbent dosage (b)

In the case of DCF, the adsorption was significantly slower. 45% of DCF was removed in 5 minutes and, as with LFX, the equilibrium concentration was achieved in 60 minutes with 82% of the initial concentration removed. The removal efficiency of 70% for DCF was achieved by L. Lima (2004) in a similar study (Lima *et al.* 2004). The highest adsorption capacity was on PAC dosage of 0.1 g/100 ml, which is 70% higher than that of LFX. For DCF removal, we need longer contact time and higher PAC dosage than for LFX removal. The greatest difference can be seen in Fig. 3b. While diclofenac usually adsorbs better than SMX, with different PAC concentrations it can be observed that the removal effectiveness is lower than in other experiments where fixed amounts of PAC (0.05 g) are used. DCF only adsorbs better than SMX at 0.05 g.

SMX has the lowest adsorption capability. 31% of SMX was removed in the first five minutes. The equilibrium between adsorbate and adsorbent was reached in 60 minutes and the removal effectiveness was 68%. The same result was also achieved by L. Lima (2004) (Lima *et al.* 2004). The most effective SMX removal was attained with a PAC concentration of 0.075 g/100 ml. The poor adsorption of SMX was also confirmed by an earlier study, which found the adsorption of SMX was 76% worse than that of DCF (Nam *et al.* 2015).

Adsorption is influenced by many factors, such as porosity of the activated carbon, pH of the solution under study and temperature. In the case of the current test, pH dropped from 7.6 to 6.3 after PAC dosing (0.05 g). B. Bhadra (Bhadra *et al.* 2016) found that the adsorption capacity of diclofenac increases when the pH decreases. This is attributed to the surface charge of the activated carbon, which decreases when the pH falls and makes the adsorption more efficient (Bhadra *et al.* 2016). In addition, the adsorption is influenced by the solubility in water of the removable substance. The higher the solubility the worse the removal rate through adsorption. In our study, LFX turned out to be insoluble in water, DCF is poorly soluble (water solubility 2.37 mg/l) and SMX partly soluble (water solubility 610 mg/l). These study findings confirm the relationship between the water solubility and adsorption, where LFX was removed in higher amount than DCF and SMX (National Center for... 2016b, 2016a, 2016c).

Maximum adsorption capacity and Freundlich adsorption isotherm

In Fig. 4a, we see the adsorption capacity of DCF, SMX and LFX dependant on time. LFX has the highest adsorption capability as is also shown in Fig. 3. The equilibrium was achieved in 60 minutes, where 324 mg of LFX per 1 gram PAC was adsorbed. DCF has an adsorption capacity of 231 mg/g on the studied concentration, which is 29% lower than that of LFX. For SMX, the maximum adsorption potential in the same time was 185 mg/g, which is 43% less than that of LFX.

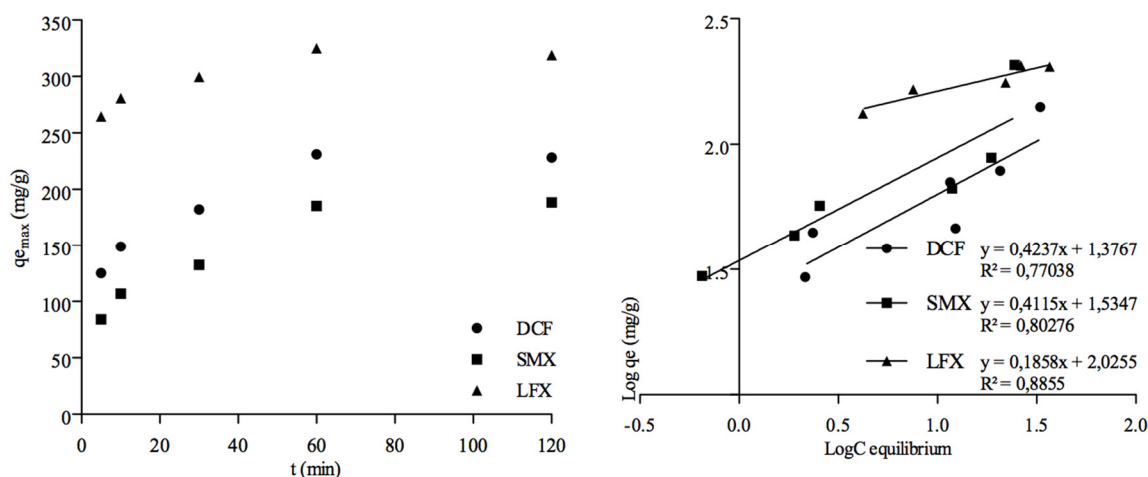


Fig. 4. The maximum adsorption capacity $q_{e_{max}}$ (mg/g) dependent on time (a) and Freundlich adsorption isotherms (b)

From Fig. 4b, K and n for DCF, SMX and LFX experiments was found. K was determined from the intersection of the graph with the y-axis and n is equal to the slope of the graph (Okeola, Odebunmi 2010). The higher K is the more effective adsorption process. With the n value, we can see the adsorption intensity and whether the adsorption process is integrated or not. When $1/n$ is higher than 1, it is uniform; the smaller the $1/n$ value, the greater the heterogeneity of the surface of PAC. If n is under 1, then the process is unfavourable for adsorption (Sekar *et al.* 2004; Desta 2013). In Table 1, the results according to Fig. 3b are shown.

Table 1. Freundlich parameters for DCF, SMX and LFX adsorption with PAC

Parameter	DCF	SMX	LFX
K	23.8	34.3	106.1
$1/n$	0.4237	0.4115	0.1858
n	2.36	2.43	5.38
q_e	34.2	53.8	138.5
R^2	0.77038	0.80276	0.8855

The results show that the adsorption processes of DCF, SMX and LFX align with the Freundlich adsorption isotherm. As our previous discussion already showed, LFX has the highest adsorption capability, as confirmed through the K, q_e and n values where it is higher than DCF and SMX. At the equilibrium concentration, the maximum adsorption capacities were 106.8, 34.3 and 23.8 for LFX, SMX and DCF, respectively. Though DCF was otherwise better adsorbable than SMX, the reason for the low q_e of DCF was explained above in Fig. 2. In reviewing other similar studies where different adsorbents were used, K was found to be 2.26 in one study and 44.9 in another, which demonstrated that adsorption processes largely depend on the adsorbent being used (Larous, Meniai 2016; Nam *et al.* 2015).

The data from Table 1 is helpful in designing an activated carbon process for DCF, SMX and LFX removal. With these results, we can calculate the required amount of PAC and detention time for the adsorption.

Conclusion

The removal of three pharmaceuticals found in the environment was surveyed in this article. It was found that the pharmaceuticals selected for the study could be effectively subjected to adsorption processes, and they could be removed in a WWTP by adding an additional treatment process. LFX has the best adsorption property and SMX the poorest.

It should be taken into account that in wastewater there are in addition to pharmaceutical residues other substances that can influence the adsorption. Thus, the next step is pilot-scale experiments in an operating WWTP. This is a prerequisite for planning and designing a full-scale PAC treatment step.

Disclosure statement

I don't have any competing financial, professional, or personal interests from other parties in connection with this research.

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