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Mini Review

Pharmaceutical Residues in the Aquatic Environment: Recent Studies in Estonia

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Introduction

Ecotoxicological impacts of pharmaceuticals in the environment were first paid attention to in the 1990-ies, when a massive death of the bird species Gyps africanus or white-backed vulture occurred in Pakistan and India. This was found to be caused by accumulation of diclofenac in their kidneys. These residues originated from veterinary treatment of free-range livestock that the birds had been eating Oaks et al. [1].

There are more than 3000 different active ingredients in use for both veterinary and human medicine, including analgesics, contraceptives, antibiotics, neuroactive compounds, beta blockers, lipid regulators and their metabolites. Carlsson [2] who has assessed the ecotoxicological effects of 27 different pharmaceuticals, has assessed, that the most harmful for the environment are diclofenac, ethinylestradiol, ibuprofen, metoprolol, norethisterone, oestriol and oxazepam. Li [3] added sulfamethoxazole into this list. There are various pathways for the medicine residues to fall into the environment. Part of the consumed medicaments are excreted by humans, discharged into wastewater treatment plants, accumulated in the sludge and, if the latter is used in greening or even agriculture, they may accumulate in plants, infiltrate into soil and ground water.

Table 1: Methods used to dispose unused pharmaceuticals Peake, et al. [13] cross-reference in the MSc thesis by Maiu Tiismus.

Year of Study	Country	Total No of Samples	Rubbish bin	Sink	Toilet	Other
2012	Nigeria	188	96	6	17	13
	Malaysia/liquid	885	25	31	19	14
	Malaysia/solid	885	65	42	17,7	11,5
	Malaysia/semi-solid	885	78	5,8	0.79	20.5
	Serbia/urban	104	85.6	0.68	8.7	5.8
	Serbia/rural	94	74.5	0	6.4	19.2
2013	Ireland/Malta/liquid	1130	57	28 sink and toilet		0
	Ireland/Malta/solid	1130	68	40 sink and toilet		0
	India	236	94	32	12	31
	Tanzania	409	66.6	0	19.7	0
	USA	444	59	0	31	0
2014	Malaysia/antibiotics	250	78.8	0	5	48
	Ireland/antibiotics	202	51	29	14	6

According to Oppel et al. [4] and Monteiro & Boxall [5] active sludge binds over 99% of salicylic acid, and 85…99.7% of caffeine, 65-77% of ketoprofen, and less than 40% of carbamazepine and celiprolol. Another pathway is by the incorrect handling of pharmaceuticals by population, flushing those in toilets and sinks or throwing those into garbage with other household waste from where the next destination is landfill and landfill leachate. A recent MSc study by Maiu Tiismus [6] in Tallinn University of Technology on the mindset and awareness of people, shows that the most common way of disposal of unused and expired medications in Estonia is still the garbage bin, although it is possible to take them to either pharmacy or local hazardous waste reception station free of charge. Discarded unused medicine should be mixed with municipal waste in a ratio 1:1000 to avoid halogen evaporation and incinerated in high temperature (>1200oC). Tiismus was awarded the degree of MSc in June 2019 (Table 1).

Estonia (1.3 million inhabitants) has collected statistical data on drugs (type and amount of medicines sold, sales value of pharmacies and wholesalers) for more than 20 years. An excel document on pharmaceuticals used in 2018 in defined daily dose (DDD) of medication needed for 1000 inhabitants per day is available on the webpage of the Estonian State Agency of Medicines (SAM). In 2017 the Secretariat of the Baltic Sea Convention (HELCOM) mentioned pharmaceutical pollution in the sea in their status report of the sea Zandaryaa et al. [7] and in spring 2019 the data of the first systematic screening of pharmaceutical residues in the Baltic Sea were published Delfi [8].

Measurable concentrations of diclofenac, ibuprofen and sulfamethoxazole were found in certain points of the Baltic Sea, especially near the eastern shore of Sweden. In 2010 the effluents of various wastewater treatment plants and their receiving waters in Sweden were studied by a Swedish researcher Ericson. Diclofenac was found ranging from 0.2to7.1 mg/l and ibuprofen between 0.1 and 0.2 mg/l. In Germany, 131 different pharmaceuticals have been detected in surface and ground water sources Weber et al [9] (Table 2).

Table 2: Best sold pharmaceuticals in Estonia, Finland, Germany, and Sweden together and for Estonia and Sweden separately: Fent et al. [15,16] cross-reference in the PhD thesis by Erki Lember [12].

Pharmaceutical	4 Countries, kg/year	Estonia kg/y, 2014	Sweden, kg /y, 2014
Paracetamol	529,035	16,950	338,007
Sulfamethoxazole	-	470	1,700
Metformin	346,888	18,800	13,500
Macrogol	273,687	980	54,400
Ibuprofen	180,208	15,100	
Mesalazine	41,966	685	17,100
Diclofenac	12,062	593	8,800

Paracetamol, diclofenac and ibuprofen are common analgesics, macrogol helps in case of constipation, mezalazine is used as an anti-inflammatory pharmaceutical and metformin treats diabetes. European Union has initiated and funded a thorough research of the impact of the release of pharmaceuticals and veterinary medicines on the state of the environment of the Baltic Sea. The watch list includes diclofenac, 17 alpha ethinylestradiol (EE2) 17 beta estradiol (E2), estrone (E1) and macrolide with the maximum permitted detection limits 10 ng/l, 0.035 ng/l, 0.4 ng/L and 90 ng/l respectively [10].

Worldwide, ca 30 000 kg of synthetic contraceptives E1, E2 and E3 are released into environment annually. In addition, 83 000 kg of cattle estrogens (natural hormones) is believed to be released in EU and USA Adeel et al. [11] affecting the living organisms. PhD student of Tallinn University of Technology, Erki Lember [12] analyzed the consumption rates of diclofenac and ibuprofen by the inhabitants of 8 seaside cities in Estonia from 2006 to 2014 and found a decrease of 19.9% for diclofenac consumption and an increase of 14,1% for ibuprofen. The European Medicines Agency had declared that diclofenac could cause heart attack or stroke and ibuprofen is as good analgesic as DFC. Lember modelled the fate of these pharmaceuticals in human metabolism as well as the removal of the residues of the partially modified and unmodified medicines in the activated sludge process of wastewater treatment plants (WWTPs). In the effluents of the WWTP, the theoretical model showed an average decrease from 1 to 0.8 mg/l g for diclofenac and increase from 11.4 to 13.4 mg/ for ibuprofen. Only a small part of the residues is decomposed in the biological treatment process or stays in the sediment. Such small concentrations are not harmful for the environment, but one should also investigate the accumulative impacts of different substances acting together.

Lember also measured the adsorption capacity (adsorbent dose and residence time) of powdered activated carbon (PAC) for removing three substances: diclofenac (DFC), sulfamethoxazole (SMX) and levofloxacin (LFX) from the effluents. 77% of the LFX could be removed in 5 minutes and 94% in 60 minutes, while only 68% of SMX was absorbed for 1 hour. An adsorption isotherm was compiled for future dimensioning of treatment processes. While the test was done in controlled conditions (using ultrapure ELGA water), some supplementary experiments were done using actual wastewater, measuring the decrease of total organic compounds with the TOC sum parameter. PAC process had the adsorption capacity per 1 g of PAC reaching 12096 mg/g, compared with APAC process (PAC was dosed into aeration tank) where the relevant value was 4060 mg/g. These results were first published in April 2017 in the 10th International Conference "Environmental Engineering" held in Lithuania, Vilnius Gediminas Technical University and in the fall 2018 Erki Lember was awarded the PhD degree.

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