



Occurrence, removal, mass loading and environmental risk assessment of emerging organic contaminants in leachates, groundwaters and wastewaters

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ABSTRACT

A comprehensive study is presented, on the occurrence, removal, mass loading, and environmental risk assessment of 19 multi-class emerging organic contaminants in three Municipal Solid Waste landfills and three Wastewater Treatment Plants located at northeast Poland. The target compounds: methylparaben, ethylparaben, propylparaben, buthylparaben, benzophenone, benzophenone-2, benzophenone-3, 3-(4-methylbenzylidene) camphor, N,N-diethyltoluamide, 4-*n*-octylphenol, 4-*n*-nonylphenol, triclosan, bisphenol A, diclofenac, clotrimazole, carbamazepine, estrone, 17 β -estradiol, diethylstilbestrol were in a wide range of concentration, from below limit of detection to 116 micrograms per liter in leachates and to 18 micrograms per liter in groundwaters and wastewaters. Benzophenone and bisphenol A were found in all analyzed samples. The highest total content of target chemical classes were registered for pharmaceuticals and phenols, 44% and 41%, respectively. Benzophenone-2 and diethylstilbestrol were not detected in all examined samples. In this study, mass loading of target analytes to aquatic ecosystem was calculated. Finally, environmental risk assessment has been presented as the risk quotient for effluent wastewaters and groundwaters on three trophic levels. Diclofenac was found to be the most critical compound in respect of environmental risk.

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1. Introduction

Emerging organic contaminants (EOCs) are compounds now being found in environment that were previously not detectable, or thought to be significant. EOCs includes hundreds of organic contaminants belonging to diverse compound classes and usually detected in natural and polluted waters in concentrations at the level between ng/L and μ g/L. Their accepted concentration limits for drinking water and wastewater effluent have not been established. The EOCs comprise a variety of compounds - since these being in the use for a long time up to newly introduced. Among them are compounds which recently have become routinely detected thanks to improved techniques of extraction and detection (gas chromatography-tandem mass spectrometry, GC-MS/MS and liquid chromatography-tandem mass-spectrometry, LC-MS/MS) as well as those for which their harmful eco-toxicological effects have been recently discovered [1,2]. Some of the EOCs have potential to influence the hormonal balance of the organism, so they are perceived as endocrine disrupting compounds (EDCs). According to definition given by the Endocrine Society the EDC is: “an exogenous

chemical, or mixture of chemicals, that interferes with any aspect of hormone action” [3]. Although there may be hundreds or more environmental chemicals with EDC activity, several classes are most commonly studied and its effect on organisms is well- documented. These are mainly pesticides, industrial chemicals, including phthalates, bisphenol A, and surfactants, steroids, heavy metals. Other groups of compounds belonging to the EOCs are pharmaceuticals and personal care products (PPCPs). The annual global production of PPCPs is currently estimated at around 20 million tones and still growing due to the development of civilization, including the greater use of medicines for humans and animals and the growing demand for personal hygiene. The main PPCPs detected in natural environment include nonsteroidal anti-inflammatory drugs, hormones, antibiotics, lipid regulators, beta-blockers, anticonvulsants, preservatives, disinfectants, sunscreen agents and repellents [4,5].

Although EOCs are usually unstable they can be very harmful because of their continuous release into environment together with municipal and industrial sewage, which are their main source. Wastewater treatment plants (WWTPs) are not efficient in eliminating EOCs thus many of this compounds may pass through wastewater treatment processes. Discharge guidelines do not exist for most EOCs so precautions and monitoring actions have not been established in most

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WWTPs. The reported concentrations of EOCs in influent and effluent wastewaters reveal significant temporal and spatial variations depended on the rate of production, population size, type of waste received, excretion rate, water consumption per person and day, the size and efficiency of WWTPs as well as persistence of the individual compound [6,7]. Other important factors are climatic conditions like rainfall, temperature and level of sunlight [8,9]. Most EOCs occurred in WWTP influent in the concentration range between 0.1 and 10 µg/L, while some pharmaceutical compounds as well as nonylphenol, triclosan and certain industrial chemicals exhibit concentrations reaching mg/L [7]. The high levels of concentration could be explained by its high consumption or excretion rate (pharmaceuticals). The concentration of EOCs in effluent ranged from 1 ng/L to 1 µg/L, which were one to two orders of magnitude lower than those in influent [9]. Wastewater treatment plants employ primary (removing of suspended solids), secondary (dispersion, dilution, partition, biodegradation and abiotic transformation) and optional tertiary (advanced physicochemical processes used for production of high quality water, which are rarely used due to their high cost) processes. Biodegradation and sorption are two major mechanisms of EOCs removal in conventional WWTPs [10,11].

While wastewater has been studied in detail as the major source EOCs for the environment, relatively little research has been devoted to municipal solid waste (MSW) landfill leachate as a potential source of these compounds. Alkylphenols, bisphenol A and phthalates were determined in landfill leachates from Gothenburg region in Sweden [12]. Presence of alkylphenols, bisphenol A and other phenols in these polluted waters was confirmed during studies conducted in Japan [13]. Fifteen pharmaceuticals were detected in leachates from four MSW landfills in Taiwan [14]. Eight organic UV filters and stabilizers were determined in landfill leachate in Norway [15]. The presence of different trace organic pollutants was also investigated for leachates from municipal landfills located in the United States and Norway [16–19]. Pollutants present in a leachate can penetrate into the soil due to the insufficient degree of isolation between the storage basin and the ground. It was also observed that many landfill leachate affect the groundwater in the vicinity of MSW [20].

The purpose of this study was to better understand the occurrence of the endocrine disrupting compounds, pharmaceuticals and personal care products in environmental matrices: influent and effluent wastewaters, MSW landfill leachates and leachate-affected groundwater from north-east Poland. Consequently, we measured the concentrations of 19 EOCs from different groups: antimicrobial preservatives: methylparaben (MP), ethylparaben (EP), propylparaben (PP), buthylparaben (BP), sunscreen agents: benzophenone (BPh), benzophenone-2 (BPh2), benzophenone-3 (BPh3), 3-(4-methylbenzylidene) camphor (4MBC), insect repellent: N,N-diethyltoluamide (DEET), antiseptic: triclosan (TRC), industrial chemical: bisphenol A (BPA), surfactants: 4-*n*-octylphenol (4OP), 4-*n*-nonylphenol (4NP), hormones: estrone (E1), 17β-estradiol (E2), diethylstilbestrol (DSB), non-steroidal anti-inflammatory drug: diclofenac (DIC), anticonvulsant: carbamazepine (CAR) and antifungal agent: clotrimazole (CLO) in influents and effluents from three WWTPs, landfill leachates from three MSW landfill sites and groundwater from two MSW landfill sites. Other specific objectives of the present study were to estimate the removal of the examined EOCs during wastewater treatment process and designated mass loadings of target analytes in the effluents. Finally, a risk analysis is done to preliminarily characterize the environmental risk of the target EOCs into Polish aquatic environment (effluent wastewaters and groundwaters).

2. Materials and methods

2.1. Reagents and solvents

MP, EP, PP, BP, BPh, BPh2, BPh3, 4MBC, DEET, 4*n*OP, 4*n*NP, TRC, BPA, DIC, CLO, CAR, E1, E2, DSB and hexane were obtained from Sigma-

Aldrich (Germany). Methanol, chloroform, acetone, anhydrous disodium hydrogen phosphate (V), anhydrous magnesium sulfate (VI), sodium bicarbonate, potassium bicarbonate, anhydrous potassium carbonate, sodium nitrate (V), sodium hydroxide, hydrochloric acid (35% to 38%), acetic acid (99%), anhydrous calcium chloride and urea were purchased from POCH (Poland). Acetic anhydride, ammonium bicarbonate (IV), magnesium chloride, potassium hydrogen phosphate (V) were provided by Chempur (Poland). Stock solutions of each analyte (at 1 mg/mL of each) were prepared separately in methanol and stored at –18 °C not longer than one month. Working solutions were prepared by diluting the stock standard solution in methanol and stored at –18 °C not longer than for two weeks. Deionized water was obtained using a purification system (Milli-Q RG, Millipore, USA) and was stored in glass bottles.

2.2. Leachates, groundwaters and wastewaters

Samples were collected into glass bottles and transported to the laboratory. Upon arrival the samples were filtered through a membrane filter with 0.45 µm pore size and acidified with concentrated hydrochloric acid to pH = 2. Later those samples were stored at –18 °C. Samples were collected between November 2011 and June 2014.

Samples were obtained from landfills of non-hazardous and inert waste having various characteristics (different in size, kind of insulation, the method of collecting leachate and age). Landfill leachates were obtained from three Municipal Solid Waste (MSW) landfill sites (A, B, C) and groundwater samples were collected from two MSW landfills sites (A, B), all located in north-eastern Poland. In MSW C piezometers are located at a depth of 35 m and we were physically not able to collect groundwater samples due to those conditions. Table 1 shows the characteristics of examined MSW landfills. Leachate was stored in open lagoons in the MSW landfill A and in wells in landfills B and C. The examined landfills transported leachate to wastewater treatment plants using tanker trucks. Additionally, a part of the leachate from landfill B is recycled by spraying it onto the landfill cap. Groundwater samples were collected from piezometers located under landfill sites. Target compounds were investigated in monitoring wells upstream and downstream of MSW landfill A. In the case of MSW landfill B samples were only taken from monitoring wells located upstream of this landfill [21].

Average daily wastewater samples were obtained from WWTPs in three cities in Poland. In these plants the treatment process includes both mechanical and biological purification with the use of activated sludge. Table 1 shows the average daily capacity and purification method of wastewater of studied WWTPs.

2.3. Analytical method and validation

In this study, the authors applied a ultrasound-assisted emulsification microextraction (USAEME) procedure [22] for the simultaneous isolation of eighteen EOCs. As the described procedure was developed for eight compounds, its current use for multiclass chemicals has required its re-optimization. All tests performed during the optimization process were carried out using distilled water containing 100 µg/L of each target compound.

Procedure of USAEME with in situ derivatization using acetic anhydride requires an addition of buffer salt. A series of experiments were performed using different quantities of sodium hydrogen phosphate (0.1 to 0.2 g) per 5 mL of sample. The highest peak areas were obtained using 0.15 g of buffering salt. Therefore, 0.15 g sodium hydrogen phosphate was used in subsequent experiments. The influence of the volume of acetic anhydride on relative peak areas was studied in regions ranging from 100 µL to 200 µL. Results indicated that the volume of acetic anhydride equal to 150 µL is optimal.

For the simultaneous USAEME and derivatization of target analytes aliquots of 5-mL of examined liquid sample were placed into 10-mL glass centrifuge test-tubes containing a mass of 0.15 g sodium hydrogen

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