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Review

Fate of estrogenic hormones in wastewater and sludge treatment: A review of properties and analytical detection techniques in sludge matrix

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ABSTRACT

Estrogenic hormones (estrone (E1), 17β-estradiol (E2), estriol (E3), 17α-ethinylestradiol (EE2)) are the major contributor to the total estrogenicity in waterways. Presence of these compounds in biosolids is also causing concern in terms of their use as soil amendment. In comparison with wastewater treatment, removal of estrogenic compounds in sewage sludge has received less attention. This paper presents a literature review regarding the source and occurrence of these pollutants in our environment. The removal pathways of estrogenic compounds in engineered systems, such as full-scale wastewater treatment plants (WWTPs), are also discussed. Review of the fate studies revealed that activated sludge system with nutrient removal shows very high (>90%) removal of estrogenic hormones in most of the cases. Although, aerobic digestion showed better attenuation of estrogenic compounds, anaerobic digestion increased the overall estrogenicity of biosolids. Finally, this paper highlights the challenges involved in analytical determination of these compounds in sewage sludge matrix.

Abbreviations: θ, Temperature co-efficient; AMO, Ammonium mono-oxygenase; AOB, Ammonia oxidizing bacteria; APCI, Atmospheric pressure chemical ionization; ASE, Accelerated solvent extraction; BOD, Biochemical oxygen demand; C, Total compound concentration; C_s, Concentration in solids; C_w, Concentration in water; CAS, Chemical abstract services; CEE, Conjugated Equine Estrogens; DTE, Dithioerythritol; E1, Estrone; E2, 17β-estradiol; E3, Estriol; EE2, 17α-ethinylestradiol; EDC, Endocrine disrupting compound; EDSP, Endocrine disruptor Screening Program; EDTA, Endocrine Disruptors Testing and Assessment; Eq, Equilin; Eqn, Equilenin; ER, Estrogen receptor; ESI, Electrospray ionization; GC, Gas chromatography; GLU, Glucuronide; GPC, Gel permeation chromatography; H, Henry's law constant; HRT, Hydraulic retention time; HRTh, Hormone replacement therapy; k, Reaction rate constant; K_d, Distribution coefficient; K_F, Freundlich parameter; K_{OM}, Distribution coefficient normalized to organic matter; K_{OC}, Distribution coefficient normalized to organic carbon content; K_{ow}, Octanol/water partition coefficient; LC, Liquid chromatography; LDTD, Laser diode thermal desorption; MAE, Microwave assisted extraction; MS, Mass spectrometry; MSTFA, N-(trimethylsilyl) trifluoroacetamide; NAS, Nitrifying activated sludge; OC, Percentage of organic carbon; OCED, Organization for Economic Cooperation and Development; pKa, Acid dissociation constant; PhATE, Pharmaceutical Assessment and Transport Evaluation; PPCP, Pharmaceutical and personal care product; S, Soluble compound concentration; SPE, Solid phase extraction; SPEED, Strategic Programs on Endocrine Disruptors; SRT, Solid retention time; SS, Suspended solids; SUL, Sulfate; t, Time; TMSI, Trimethylsilylimidazole; TSS, Total suspended solids concentration; USEPA, United States Environmental Protection Agency; VSS, Volatile suspended solids concentration; WWTP, Wastewater treatment plant; YES, Yeast estrogen screen.

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Contents

1. Introduction

Some natural and synthetic compounds are attracting attention due to their interference with the usual functioning of the endocrine system in humans and animals. Collectively these are called endocrine disrupting compounds (EDCs). When present in environment above a certain concentration (threshold limit value), these compounds can cause adverse health effects on wildlife (Hansen et al., 1998; Tyler et al., 2005). EDCs mainly consist of natural hormones, synthetic hormones and their metabolites; several non-steroidal, synthetic compounds that are used as plasticizer, flame retardants, surfactants, and pesticides; some pharmaceutical and personal care products (PPCPs) (Caliman and Gavrilescu, 2009). Most of the compounds that interact with hormone signaling are estrogenic in nature; only a few have androgenic or anti-androgenic potency (McLachlan et al., 2006). Among these different classes of endocrine disrupters, human and animal waste born hormones, often known as endogenous steroidal hormones, has been characterized by very high estrogenic potency. Compared to the exogenous endocrine disrupters, such as the organochlorine pesticides and industrial compounds, endogenous hormones has been found to be 10^2–10^7 times more potent (Legler et al., 1999, 2002a, 2002b).

Municipal wastewater is the main disposal pathway for the human waste born estrogenic compounds. In addition, synthetic estrogens widely used in oral contraceptives and hormone replacement therapy are ingested in humans and after excretion enters the wastewater stream. Optimizing the removal of micropollutants like estrogenic compounds is not a design criterion for conventional wastewater and sludge treatment plants. Upon wastewater treatment, stabilized biosolids may act as source of these micropollutants due to incomplete removal from solid and/or liquid phase of wastewater. Studying the fate of these chemicals throughout different unit treatment processes is important to determine the removal and incoming load of estrogens to environment. The goal of this paper is to provide a state of the art review of the current treatment techniques employed in wastewater and biosolids treatment in regards to their removal efficiency and mechanism of estrogenic hormones. Furthermore, the current practices and the challenges involved in analytical determination of these compounds in biosolids samples are highlighted.

2. Properties of hormones as endocrine disrupters

A number of natural and synthetic hormones from humans and animals act as endocrine disruptors as well as some estrogen mimicking compounds derived from plants. Considering their origins, these compounds can be grouped as following (Caliman and Gavrilescu, 2009; Burkhardt-Holm, 2010):

i) Natural estrogenic/androgenic hormones: E2, E1, testosterone etc.
ii) Synthetic hormones: EE2, diethylstilbestrol, 19-norethindrone etc.
iii) Phyto- and mycoestrogens: daidzein, genistein, zearalenone etc.