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Effect of biochar on fate and transport of manure-borne estrogens in sandy soil

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ABSTRACT

The feasibility of using two types of biochars to reduce steroid hormone pollution from poultry and swine manure application on agricultural land was evaluated. The sorption affinity and desorption resistance of softwood and hardwood biochars were also determined for two estrogen hormones, 17β-estradiol (E2) and its primary metabolite estrone (E1). The softwood and hardwood biochars demonstrated high retention capacity for both estrogens. The effective distribution coefficient (K_d^{eff}) of soil-softwood-derived biochar (SBS_{450}) was significantly higher than soil-hardwood-derived biochar (SBH_{750}), indicating the stronger sorption affinity of SBS_{450} for estrogens. To validate the laboratory results, a field lysimeter experiment was conducted to study the fate and transport of E2 and E1 in soil and leachate in the presence of 1% softwood-biochar (BS_{450}) in topsoil and to compare it with soil without any amendments. The spatio-temporal distribution of both estrogens was monitored at four depths over a 46-day period. The lysimeters, in which the surface layer of soil was amended with biochar, retained significantly higher concentrations of both estrogen hormones. Although they leached through the soil and were detected in leachates, collected at 1.0 m depth, the concentrations were significantly lower in the leachate collected from biochar-amended lysimeters. The result confirmed the efficacy of biochar amendment as a remediation technique to alleviate the manure-borne hormonal pollution of groundwater.

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Introduction

Different types of antibiotics, growth promoters and hormones are administered in concentrated animal and poultry feeding operations; a large fraction of these in the parent or variant forms is excreted. Both poultry and swine operations generate large quantities of organic wastes (animal manure and biosolids). These polluted organic wastes are routinely disposed in agricultural land as organic soil-amendments. This waste management technique has become a major gateway for environmental exposure to a new class of toxic and high-risk organic contaminants with endocrine disrupting properties, known as emerging contaminants. These micro pollutants are poorly documented in terms of their toxicological and biological risks for environmental health and safety issues (Schriks et al., 2010). Among the emerging contaminants of greatest concern are the natural sex hormones (*i.e.*, 17β-estradiol, estrone, testosterone and progesterone) due to their high chronic toxicity and long-term health issues (*e.g.*, carcinogenicity, mutagenicity or teratogenicity) at concentrations as low as ng/L (Choi et al., 2004). The feminization

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of male fish or the masculinization of female fish (Soto et al., 2004; Vajda et al., 2011), the reproductive biology alteration of wild fathead minnows (*Pimephales promelas*) and rainbow trout at aquatic E2 and E1 concentrations as low as 1–10 ng/L and 25–50 ng/L, respectively (Fang et al., 2003; Hao et al., 2013) represent several biological hormone disruption incidences in aquatic media.

Animal manures are major sources of bioactive levels of natural estrogens including 17β-estradiol (E2) and estrone (E1). In spite of reported short half-lives and fast dissipation under aerobic conditions (Colucci et al., 2001; Fan et al., 2007; Ying and Kookana, 2005) and the high sorption affinity of estrgoens in topsoil (Abdullah and Wu, 2009; Lee et al., 2003; Neale et al., 2009), several studies (Jacobsen et al., 2005; Shore and Shemesh, 2003; Soto et al., 2004) have reported hormonal contamination of water resources by surface runoff from agricultural fields receiving animal manure. Finlay-Moore et al. (2000) observed concentrations of E2 up to 100–2500 ng/L in runoff from agricultural land after the application of poultry manure. Similar studies (Duncan et al., 2015; Kjær et al., 2007; Lee et al., 2003) have reported frequent detection of estrogens in the vadose zone beneath a manure-fertilized soil and in the leachate. Given the limited understanding of the environmental pathways and ecotoxicology of high-toxicity-at-low-concentrations of manure-borne steroid hormones in soil and water media, there is inadequate knowledge to address the remediation of manure-borne steroid hormones in the environmental matrix. Consequently, there is a pressing need to conduct detailed studies with the objective of developing feasible remediation techniques to reduce the environmental and biological effects of hormonal pollution.

Recently, the *in-situ* application of carbon-rich organic amendments, derived from biomass and biological residues, has been deployed as a financially-feasible approach to alleviate the natural pollutant transfer from contaminated to the aquatic environment (Alizadeh et al., 2016; Khorram et al., 2016; Wang et al., 2017). Biochar is the byproduct of the thermo-chemical conversion of biomass and biological residues in the absence of oxygen (pyrolysis). The amorphous structure of biochar is comprised of nano-scale condensed aromatic rings with a crystalline structure with the presence of both polar and non-polar surface sites. Thus, biochar offers a strong sorption affinity for inorganic contaminants (Beesley et al., 2011; Karami et al., 2011) and hydrophobic organic contaminants due to its high specific surface area and resistance to bio-decomposition.

The effect of biochar on sorption and dissipation of different types of pesticides (Zheng et al., 2010), herbicides and heteroaromatic amines (Khorram et al., 2017; Xiao and Pignatello, 2015), aminocyclopyrachlor, bentazone, fungicide pyraclostrobin (Cabrera et al., 2014) and endocrine disrupting compounds (Jung et al., 2013; Sarmah et al., 2010), have been determined by batch equilibration studies; however, there is a paucity of knowledge regarding the environmental remediation behavior of biochar under field conditions. Therefore, spatio-temporal investigations of the transport of hormones in field lysimeters, filled with soil, with and without biochar amended surface soil were carried out. The overall objective of this study was to evaluate the feasibility of the incorporation of two types of biochars (BS_{450}: soft-wood biochar produced at 450°C and BH_{750}: hard-wood derived biochar produced at 750°C) as a novel approach for reducing leaching of manure-borne estrogens, 17β-estradiol (E2) and its primary metabolite estrone (E1), from agricultural land. The specific objectives were (1) to determine the effect of both biochars on sorption–desorption and dissipation of poultry and swine manure borne E2 and E1 in sandy soil using a laboratory batch equilibrium technique and (2) to elucidate the fate and transport of manure-borne estrogens in soil alone, and in biochar-amended surface layer matrices, under field condition.

1. Materials and methods

1.1. Analytical chemicals

The pure analytical standards (≥99%) for E2 and E1 were purchased from Sigma Aldrich (St. Louis, MO, USA). The physiochemical and structural properties of both estrogens are summarized in Table S1 in supplementary information. High performance liquid chromatography (HPLC)-grade acetonitrile was purchased from Fisher Scientific. Double-deionized water (Milli-Q, Millipore, Molsheim, France) was used in the preparation of standard solutions and mobile phase solutions.

1.2. Soil and biochar characteristics

A sandy soil, belonging to the Sainte-Amable complex (ElSayed and Prasher, 2014) was used in both the sorption–desorption equilibrium study and in the field-scale evaluation of remediation capacity of biochar. The soil was collected from the lysimeters, located at the Macdonald campus of McGill University, Sainte-Anne-de-Bellevue, Quebec. The physical and chemical properties of the soil are given in Table 1. Softwood-derived biochar (BS_{450}) and hardwood-derived biochar (BH_{750}) were provided by BlueLeaf Inc., Drummondville, Quebec, Canada. BS_{450} was produced by slow-pyrolysis of the softwood fraction of main feedstock, bark of spruce trees, at 450°C for 2.5 hr. The processing of the hardwood segment of stock under fast pyrolysis at 750°C for a few minutes yielded the BH_{750}. The physical, chemical and elemental characterization of each biochar was provided by BlueLeaf Inc. in collaboration with the soil control lab of Control Laboratories Inc., Watsonville, California, USA. The characterization of each type of biochar is presented in supplementary information (Table S2).

1.3. Batch sorption–desorption experiments

The effect of both biochars on the sorption–desorption behavior of estrogens (E2, E1) was evaluated through a soil equilibrium technique using a modified protocol by Lee et al. (2003). Three different treatments, (1) soil (S), (2) soil and soft-wood biochar (SBS_{450}) and (3) soil and hard-wood biochar (SBH_{750}), were used in each batch. Each soil-biochar treatment contained 1% (W/W) of biochar. Triplicates of each treatment (2 g) were equilibrated with an aqueous solution of estrogens (20 mL) at initial spiking concentrations of 0.01, 0.05, 0.1, 0.5, 1 and 5 mg/L considered to be the reported estrogen content in swine and poultry manure (Bevacqua et al., 2011). All treatment samples were covered with aluminum foil to prevent any possible photo-degradation. The samples were placed on a reciprocating shaker for 24 hr at

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