Science of the Total Environment 496 (2014) 576–584

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Bioavailability and fate of sediment-associated trenbolone and estradiol in aquatic systems

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HIGHLIGHTS

- Steroid fate was measured in sediment and water by *in vivo* and *in vitro* methods.
- Estradiol was not bioavailable due to rapid degradation and reduced bioactivity.
- Trenbolone in sediment induced changes in whole organism endocrine function.

ARTICLE INFO

Article history:
Received 16 June 2014
Received in revised form 11 July 2014
Accepted 11 July 2014
Available online 8 August 2014

Editor: D. Barcelo

Keywords:
Hormone
Bioavailability
Sediment
Aquatic toxicology
Sorption
Fathead minnow

ABSTRACT

Endocrine disrupting effects in aquatic organisms have been observed in systems influenced by steroid hormones. Associating endocrine disruption with aqueous concentrations of steroids alone may overlook the influence of source–sink dynamics in sediments on steroid hormone bioavailability. The objective of this study was to determine the fate of 17β-estradiol and 17β-trenbolone in two field sediments and to evaluate the corresponding bioavailability of the compounds to the fathead minnow (*Pimephales promelas*). Steroid fate was evaluated using analytical chemistry and verified by assessing the biological activity using yeast based *in vitro* assays. Effective bioavailability of the steroids was inferred from changes in hepatic vitellogenin expression (increased expression in males exposed to 17β-estradiol, and reduced expression in females exposed to 17β-trenbolone). In experiments conducted with 17β-estradiol, no induction of hepatic vitellogenin mRNA expression was observed in male fish exposed to sediment-associated 17β-estradiol. In contrast, female minnows exposed to sediment-associated 17β-trenbolone experienced significant reductions in hepatic vitellogenin compared to negative controls. In both systems, the parent compounds were shown to degrade rapidly to the more persistent metabolites, estrone and trendione, both of which were found predominantly associated with the sediments. Results from the yeast estrogen screen indicate a reduction in biological activity as biotransformation of 17β-estradiol occurs; results from the yeast anti-estrogen screen were inconclusive and unable to substantiate 17β-trenbolone fate in aquatic systems. Collectively, these data support the contention that steroid hormones associated with the sediment can become bioavailable to fish, and that sediment characteristics influence the observed bioavailability of these compounds.

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1. Introduction

The biological effects of aquatic organism exposure to exogenous hormones include abnormal expression of secondary sex characteristics (Seki et al., 2006) and abnormal gonadal development resulting in intersex fish (Tetreault et al., 2011) that can ultimately lead to the collapse of wild fish populations (Kidd et al., 2007). Significant research has focused on the occurrence and biologic effect of steroidal compounds originating in domestic wastewater (Bradley et al., 2009; Lei et al., 2009; Tetreault et al., 2011; Zhou et al., 2012), and in these systems, steroid hormones have been described as being 'pseudopersistent' in the water column (Sumpter and Johnson, 2008) due to the continuous point source discharge of

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http://dx.doi.org/10.1016/j.scitotenv.2014.07.040
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effluent. More recently, agricultural production systems have also been cited as a significant source of steroidogenic compounds to aquatic systems as both estrogens and androgens have been detected in runoff from areas of intense agriculture including row crops (Gall et al., 2011; Sellin et al., 2009), animal grazing areas (Kolodziej and Sedlak, 2007) and animal feeding operations (Bartelt-Hunt et al., 2011, 2012; Kolok et al., 2007; Scott Mansell et al., 2011) as well as in surface water and sediments in proximity to animal production facilities (Bartelt-Hunt et al., 2012; Jeffries et al., 2011; Scott Mansell et al., 2011; Sellin et al., 2009, 2010).

Unlike municipal wastewater effluents, which occur as point sources, discharges from agricultural systems are typically non-point source and correlated with temporal events such as manure application, natural precipitation, and/or irrigation events (Bartelt-Hunt et al., 2012; Gall et al., 2011; Scott Mansell et al., 2011). Although there is limited information regarding the temporal variability of steroids originating from non-point source discharges, it is anticipated that after these loading events, hydrophobic steroid hormones are likely to be predominantly associated with aquatic sediment (Casey et al., 2003; Kim et al., 2007; Yu et al., 2004).

There is evidence from field studies that sediment-derived steroids remain bioavailable in the environment. Sellin et al. (2010) found that female fish exposed to river sediments from agriculturally-impacted surface water expressed decreased levels of vitellogenin (Vtg) and estrogen receptor alpha (ERα), although a specific compound responsible for the observed effects could not be identified. A subsequent laboratory exposure study using field-contaminated sediments from the same location confirmed that the result from Sellin et al. (2010) was due to an anti-estrogen and sediment extracts were found to contain several steroids including 17β-trenbolone (17β-Tb), α-estradiol, α-zearalenol, estrone (E1), testosterone, progesterone, and 4-androstenedione (Jeffries et al., 2011). Bioavailability of estrogens in field sediment was evaluated by Duong et al. (2009). In this study, field sediments from a river impacted by sewage and stormwater were found to elicit inappropriate expression of Vtg in male fish, and this response was attributed to the occurrence of 17 β-estradiol (17β-E2), E1 and ethynylestradiol (EE2) in the sediment.

There have been more limited investigations of steroid-associated bioavailability in laboratory settings where steroid movement between sorbed and aqueous phases can be carefully monitored, and the focus of previous studies has been predominantly on estrogen bioavailability. Rempel et al. (2008) observed increases in Vtg expression in male hornyhead turbot exposed in laboratory aquaria to a single field sediment spiked with 17β-E2 at a concentration of 46.5 ng/g dry weight, however exposure to sediment with lower initial 17β-E2 concentrations did not result in significant changes in Vtg expression. The occurrence of sediment particles <1 μm has been demonstrated to increase the inappropriate expression of Vtg in fish exposed to an aqueous solution of 100 ng/L 17β-E2, indicating that 17β-E2 sorption to sediment can enhance 17β-E2 bioavailability (Duong et al., 2009, 2010), however, the specific mechanisms responsible for this observed effect have not been demonstrated. It is clear that sediment-associated steroids can remain bioavailable, however, to date, there is limited information regarding the route of exposure of sediment-associated steroids and how processes such as steroid sorption, desorption and biotransformation influence the bioavailability of sediment-associated steroids. In addition, there is limited information on whether estrogens and androgens will exhibit similar behavior as to date, field and laboratory studies of sediment-associated steroids have focused almost exclusively on estrogens. It is important to note that biological effects caused by steroids in the environment may be due to exposure to a complex mixture as parent compounds degrade or transform. The importance of combining chemical and biological methods has been highlighted by Blasco and Pico (2009) and the current study is one of the first to combine both chemical and biological methods for environmental assessment.

In this study, we investigated the effect of sediment-associated 17β-E2 and 17β-Tb on Vtg expression in an environmental sentinel species, the fathead minnow. The objective of this study was to conduct an integrated evaluation of the fate of steroidogenic compounds in sediment and water to understand their corresponding biologic activity and bioavailability. *In vitro* biologic activity of field sediments spiked with a known concentration of steroid was determined using yeast estrogen screen (YES) and yeast anti-estrogen screen (YAES) assays while bioavailability was evaluated using fathead minnows to determine if fish exposed to the same contaminated sediments were susceptible to changes in hepatic gene expression of Vtg. Two natural sediments with different physical properties and organic carbon contents were used to evaluate the effects of sediment type on both the bioavailability and fate of sediment associated steroids. Results of biological analysis were compared with analytical chemistry data obtained from controlled microcosms and periodic grab samples of sediment and water from exposure aquaria to correlate contaminant fate with in vitro biologic activity and bioavailability in the target organism.

2. Materials and methods

2.1. Sediment collection and storage

River bottom sediments were collected from two sites in Nebraska. Sediment classified as silty loam was collected from Plum Creek in Seward, Nebraska and sandy sediment was collected from the Elkhorn River near Winslow, Nebraska. At each site, sediment was collected from the top 10 cm of the streambed and kept frozen at −20 °C until use. Both of these sites were selected as steroid hormone contamination had not previously been detected at these sites (Sellin et al., 2009), and to evaluate bioavailability as a function of diversity in sediment properties. The sediments used in this study differed in total organic carbon (TOC) content and particle size (Table S10). The silty loam contained 1.44% TOC and 24% clay content with the dominant clay mineral being smectite. The sandy sediment has a TOC content of 0.13% and was 92% sand. The pH of both sediments was 7.4.

2.2. Fish exposure experiments

Laboratory experiments were conducted in 45-L glass aquaria using sexually mature fathead minnows (*Pimephales promelas*) from an on-site culture. Fish were divided into negative control (i.e. unexposed to steroids) and sediment-associated steroid exposed groups. Negative control experiments consisting of fish exposed to sediment mixed with the solvent only and fish exposed to the field sediment without hormone addition were conducted to ensure that any observed change in gene expression could not be attributed to pre-existing contaminants in the sediments or to solvents used in hormone stock solutions. In the sediment-associated steroid experimental tank, a layer of sediment approximately 6 cm deep was pre-equilibrated with the hormone of interest and the hormone was not replenished in the sediment tank during the experiment. A total of 4 experimental exposure aquaria were used: one for each hormone (17β-E2 and 17β-Tb) evaluated in each of the two sediments (sand and silty loam) as shown in the Supporting information Fig. S1.

To prepare hormone-contaminated sediment, 17β-E2 or 17β-Tb was dissolved in methanol, then added to DI water to give a final solution concentration of 100 μg/mL (17β-Tb) or 10 μg/mL (17β-E2). The hormone solution was added to each 45 L aquaria containing approximately 6 cm of gravity-drained sediment. Each hormone solution and sediment was combined in separate aquaria and mixed thoroughly every few hours over a 12-hour period to ensure a homogeneous substrate. The final methanol concentration was kept to a minimum (<0.1 μL/L) to avoid solvent effects in the fish. The target steroid concentration in both sediments was 1.1 17β-E2 ng/g dry sediment. Target

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