Available online at www.sciencedirect.com

ScienceDirect

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

Comparison of export dynamics of nutrients and animal-borne estrogens from a tile-drained Midwestern agroecosystem

Heather E. Gall [a,b,*], Stephen A. Sassman [c], Byron Jenkinson [d], Linda S. Lee [c], Chad T. Jafvert [b,e]

[a] Pennsylvania State University, Department of Agricultural and Biological Engineering, 232 Agricultural Engineering Building, University Park, PA 16802, USA
[b] Purdue University, Lyles School of Civil Engineering, 550 Stadium Mall Drive, West Lafayette, IN 47907, USA
[c] Purdue University, Department of Agronomy, 915 West State Street, West Lafayette, IN 47907, USA
[d] Jenkinson Environmental, LLC, West Lafayette, IN 47907, USA
[e] Purdue University, Division of Environmental and Ecological Engineering, 550 Stadium Mall Drive, West Lafayette, IN 47907, USA

ARTICLE INFO

Article history:
Received 1 June 2014
Received in revised form
18 August 2014
Accepted 26 August 2014
Available online 6 September 2014

Keywords:
Emerging contaminants
Water quality
Manure
Effluent irrigation

ABSTRACT

Concentrated animal feeding operations (CAFOs) are known to be a source of nutrients and hormones found in surface water bodies around the world. While the fate and transport of nutrients have been studied for decades, much less research has been conducted on the fate and transport of hormones. To facilitate a comparison of nutrient and hormone export dynamics from farm fields, nitrate + nitrite (N), dissolved reactive phosphorus (DRP), 17α- and 17β-estradiol (E2), estrone (E1), and estriol (E3) were monitored in a tile drain and receiving ditch for one year on a working farm in north central Indiana. Repeated animal waste applications led to high frequency detection of hormones (>50% in tile drain; >90% in the ditch) and nutrients (>70% for DRP; 100% for N). Hydrologic variability was found to be a dominant factor controlling export of N, DRP, and E1 to the drain and ditch. Of the estrogens, the temporal trend in E1 export was most similar to that of DRP. Differences in temporal export between P and the other estrogens likely were due to differences in the biogeochemical processes that affect their fate and transport within the agroecosystem. During short periods when the flowrate exceeded the 80th percentile for the year, over 70% of the total mass export of DRP and E1 occurred for the year in both the tile drain and ditch, demonstrating the importance of high-flow events. Therefore, best management practices must be effective during large flow events to substantially reduce transport to downstream locations.

* *Corresponding author*. Penn State University, Department of Agricultural and Biological Engineering, 232 Agricultural Engineering Building, University Park, PA 16802, USA. Tel.: +1 814 863 1817.
E-mail address: heg12@psu.edu (H.E. Gall).
http://dx.doi.org/10.1016/j.watres.2014.08.041
0043-1354/

1. Introduction

The environmental impacts associated with land application of animal wastes have traditionally focused on nutrients; however, livestock waste has been identified as a source of hormones found in surface water bodies around the world (Chen et al., 2010; Kjær et al., 2007; Kolok and Sellin, 2008; Matthiessen et al., 2006; Nichols et al., 1997; Soto et al., 2004). The natural and anthropogenic processes influencing the availability of constituents for export from agricultural fields to downstream locations have important implications for water quality. While these processes for nutrients have been studied for decades, much less research has been conducted on the fate and transport of hormones. To date, no studies have been reported that fully compare the dynamics of exported loads (i.e., masses) of nutrients and hormones to receiving streams in an agroecosystem.

Hormone transport from agricultural fields has been investigated in several previous studies. In Delaware, Dutta et al. (2010) sampled surface runoff from fields treated with poultry litter during 10 storm events occurring up to 4 months after the litter was applied. Throughout the study period, significant correlations between rainfall depth and estrogen loads were found. In Denmark, Kjær et al. (2007) sampled tile drainage for one year following subsurface injection of swine slurry. They detected hormones in the tile drains for up to 11 months after the slurry had been injected. Our previous research on hormone concentrations (Gall et al., 2011) and loads (Gall et al., 2014) discharged from a tile-drained agroecosystem also found persistent hormone export, with export occurring up to 4 months following multiple applications of animal wastes. Overall, the results of these studies and others suggest that preferential flow (Gall et al., 2014, 2011; Kjær et al., 2007; Steiner et al., 2010) and surface runoff (Dutta et al., 2010; Finlay-Moore et al., 2000; Haggard et al., 2005; Jenkins et al., 2006) are important transport pathways for hormones.

In general, the installation of tile drains and subsequent changes from natural to agricultural land use cause an increase in discharge and nutrient export (Skaggs et al., 1994). However, as the drainage density of tile drains increases, phosphorus (P) and organic nitrogen loads generally decrease and nitrate (NO_3^-) loads generally increase compared to agricultural sites with lower drainage densities (Kladivko et al., 2004; Simard et al., 2000; Skaggs et al., 1994). This trend is due to differences in the affinity for soil and the transport pathways for phosphorus compared to nitrate. Surface runoff is reduced and the soil infiltration capacity is increased when tile drains are installed, thereby reducing the primary transport pathway for P and enabling P to be retained by the soil matrix (Simard et al., 2000). These changes cause an increase in the primary transport pathway for NO_3^- (Kladivko et al., 2004), which does not sorb to the soil matrix.

While the transport pathways for hormones and P in tile-drained systems are similar, the magnitude and rates of the biogeochemical processes affecting phosphorus (P) and hormones such as sorption/desorption and microbial degradation can differ greatly. Hormones can be biotransformed or mineralized (Carmosini and Lee, 2008). Under aerobic conditions, 17α-estradiol (17α-E2) and 17β-estradiol (17β-E2) biotransform to estrone (E1), with an apparent half-life of hours to days (Colucci et al., 2001; Combalbert and Hernandez-Raquet, 2010; Lee and Liu, 2002; Lee et al., 2007; Mashtare et al., 2013). Under anaerobic conditions, both 17β-E2 and 17α-E2 degrade predominantly to E1, but reversibly, and interconversion between isomers also occurs (Mashtare et al., 2013). Anaerobic biotransformation of 17α-E2 to estriol (E3) has also been observed (Xuan et al., 2008). The microbial processes affecting nutrients are quite different. Conversion of dissolved reactive phosphorus (DRP) into organic matter makes phosphorus less mobile (Mullins, 2009). During mineralization, organic phosphorus is converted to DRP, which is then more available for plant uptake or hydrologic export. The processes influencing nitrate levels include plant uptake, production from mineralization of organic matter and nitrification, and losses due to denitrification and immobilization.

Sorption of hormones occurs predominantly due to hydrophobic partitioning; however, hydrogen bonding and electron donor–acceptor interactions with soil organic matter likely also cause hormones to sorb to soils (Mashtare et al., 2011; Shareef et al., 2006; Sun et al., 2012; Yamamoto and Liljestrand, 2003). Qiao et al. (2011) identified hydrogen bonding as a more important sorption mechanism for 17β-E2 compared to 17α-E2. Sorption of orthophosphate is largely through electrostatic interactions to specific sites. The results of desorption studies have yielded mixed results, with hormones being observed to desorb from soil and sediments relatively quickly and easily (Hildebrand et al., 2006) and with continued extraction from soils that have been washed successively (Loffredo and Senesi, 2002), while others observed strong hysteretic desorption (Durán-Álvarez et al., 2014; Lai et al., 2000). Dissolved organic carbon (DOC) has been shown to play an important role in the sorption of hormones to soils, with greater DOC impacts to sorption occurring in soils amended with sludge or manure (Stumpe and Marschner, 2010; Qiao et al., 2011). P desorption exhibits strong hysteresis, with only a small amount of adsorbed P readily available for desorption (Pierzynski et al., 1994). However, rates of P release can become higher when soils approach P saturation (Torrent and Delgado, 2001).

Significant insight into the role of biogeochemical and hydrologic factors affecting hormone transport may be gained by comparing the export dynamics of hormones to nutrients, which have been studied extensively. Previous studies have shown that tile drained fields export hormones and nutrients, but were not conducted at a sufficiently high temporal resolution to quantify and compare hormone and nutrient export dynamics. The goal of this study was to compare the export dynamics of nutrients and estrogens from a tile-drained agricultural field using several techniques in order to assess the roles that hydrologic and biogeochemical factors play in controlling their fate and transport. These techniques include an examination of concentration–discharge (C–Q) relationships, load–discharge (L–Q) relationships, and a quantification of the importance of high-flow events to total annual eported loads. An improved understanding of the similarities and differences of how natural and anthropogenic drivers affect nutrients (e.g., N, P) and hormones (e.g., estrogens) will provide additional insight into whether best management