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Seasonal occurrence of antibiotics and a beta agonist in an agriculturally-intensive watershed

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ABSTRACT

We evaluated the occurrence of 12 veterinary antibiotics and a beta agonist over spatial and temporal scales in Shell Creek, an intensively agricultural watershed in Nebraska, using Polar Organic Chemical Integrative Samplers (POCIS). Twelve pharmaceuticals were detected with concentrations ranging from 0.0003 ng/L to 68 ng/L. The antibiotics measured at the highest time-weighted average concentrations were lincomycin (68 ng/L) and monensin (49 ng/L), and both compounds were detected at increased concentrations in summer months. Analysis of variance indicates that mean concentrations of detected pharmaceuticals have no significant ($p > 0.01$) spatial variation. However, significant temporal differences ($p < 0.01$) were observed. This study demonstrates the utility of passive samplers such as POCIS for monitoring ambient levels of pharmaceuticals in surface waters.

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1. Introduction

The occurrence of veterinary pharmaceuticals, including antibiotics and beta agonists, in surface water is of significant concern due to their potential human health and ecological impacts. These compounds can enter surface water through illicit or accidental wastewater discharge, or in storm runoff from agricultural fields fertilized with manure from animals that were administered pharmaceuticals (Davis et al., 2006; Kim et al., 2010; Joy et al., 2013). Although there is significant evidence showing that pharmaceuticals can be transported to surface water in runoff from land-applied manure, to date, a limited number of studies have evaluated the fate and persistence of antibiotics in surface waters in agricultural watersheds.

Antibiotics are commonly prescribed to humans and livestock for prevention and treatment of disease. Beta-adrenergic agonists stimulate skeletal muscle growth of livestock species (Beermann, 2002). Ractopamine, sold as a feed additive, is used to promote weight gain (Centner et al., 2014) and is administered to swine (Alder et al., 2010) and cattle (Koontz et al., 2010). The Center for Disease Control (CDC) has estimated that 50 million pounds (~2.26 million kg) of antibiotics are produced in the United States annually, with approximately 40 percent used in agriculture (Nawaz et al., 2001). Antibiotics administered to animals are excreted in manure, and up to 90% of certain antibiotics may be excreted unmetabolized (Jjemba, 2006). Antibiotic concentrations in manure have been reported to range from trace levels to hundreds of mg/kg, with typical concentrations between 1 and 10 mg/kg (Joy et al., 2013; Ji et al., 2012; Zhao et al., 2010; Martínez-Carballo et al., 2007; Kumar et al., 2005).

The occurrence of antibiotics in the environment may pose a human health risk as they may be linked to proliferation of antibiotic resistance (Jjemba, 2006; Hughes and Andersson, 2012; Truszczyn'ski and Pejsak, 2013). Some antibiotics are structurally similar to pesticides (e.g. fluconazole vs. azaconazole; sulfanilamide vs. asulam) and may also impose a risk to non-target organisms (Swanton et al., 2011). For instance, lomefloxacin, sulfamethoxazole, and chlortetracycline are toxic for *Lemna gibba* at effective concentrations of 38, 37 and 114 μg/ml, respectively (Brain et al., 2004).

Potential adverse effects of antibiotics on microorganisms, aquatic life and human health have motivated the need to evaluate their occurrence in the environment, especially in water receiving municipal wastewater treatment plant effluents (Alvarez et al., 2005; Moldovan, 2006; Ying et al., 2009; Watkinson et al., 2009). In contrast, fewer studies have analyzed the occurrence, temporal, and spatial variability of antibiotics in water bodies within predominantly agricultural watersheds (Arikan et al., 2008;

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Kurwadkar et al., 2013; Zhang et al., 2012). Antibiotics including tetracyclines and sulfonamides (Arikan et al., 2008; Zhang et al., 2012; Lissemore et al., 2006) and monensin (Kurwadkar et al., 2013) have been detected in watersheds influenced by agricultural inputs, and some evidence suggests that there are temporal trends in antibiotic concentration (Arikan et al., 2008; Zhang et al., 2012) with higher concentrations occurring during winter or low-flow conditions. Typical concentrations of antibiotics measured in agricultural watersheds range from hundreds of ng/L (Lissemore et al., 2006) to tens of μg/L (Kurwadkar et al., 2013).

Traditionally, grab sampling methods are used to evaluate the occurrence of pharmaceuticals in aquatic environments. The efficacy of grab samples can be limited when evaluating low concentrations of pollutants, especially pollutants that dissipate quickly or enter the water body in discrete temporal events. An alternative is to use passive samplers such as the Polar Organic Chemical Integrative Sampler (POCIS) (Alvarez et al., 2005). POCIS are useful for evaluation of water-soluble compounds like veterinary pharmaceuticals where concentrations may be below detection limits in grab samples. POCIS are efficient due to the ease of deployment, and do not require collecting or processing multiple water samples (Alvarez et al., 2005). POCIS have been used to provide TWA concentration of pharmaceuticals at a location for periods of weeks to months (Alvarez et al., 2004; Matthiessen et al., 2006; Bartelt-Hunt et al., 2009).

In this study, we evaluated the occurrence of 12 veterinary antibiotics and a beta agonist in the Shell Creek watershed, Nebraska (Fig. 1). POCIS devices were deployed at eight stations simultaneously between September 2008 and October 2009. Our purpose was to assess the temporal variation in occurrence of dissolved pharmaceuticals in an intensively agricultural watershed.

2. Materials and methods

2.1. Watershed description

The Shell Creek watershed drains approximately 1200 km^2 in east-central Nebraska and is located primarily within Boone, Madison, Platte and Colfax counties of Nebraska. The towns of Schuyler, Platte Center, Lindsay and Newman Grove are located within the watershed and the city of Columbus is directly to the south. Shell Creek has three major tributaries in the watershed: Elm Creek, Loseke Creek and Taylor Creek (Fig. 1).

The watershed is a heavily agricultural region with a high percentage of the land in irrigated row crops (46%). The counties comprising the watershed include approximately 1550 farms with over 1,050,000 head of swine, cattle, and poultry (USDA-NASS, 2012). Land cover types include cultivated (78.2%), herbaceous (14.6%) and forest (1.85%); while urban developed areas cover only 4.4% of the watershed (Fig. 2). In this region, antibiotics are commonly administered prophylactically for disease prevention as well as for disease treatment. Animals in the region are typically raised in confined animal feeding operations (CAFOs) where animals are confined to production barns (swine and poultry) or feedlots (cattle). Manure from animal production facilities is typically collected and land applied to agricultural fields as a soil conditioner and fertilizer. The five towns in the watershed combined include a population of 1675 people (U.S. Census Bureau, 2010). The daily discharge at the USGS gage station (Fig. 1) averages 1.40 ± 5.13 m^3/s between 1947 and 2014 (U.S. Geological Survey, 2012), and mean precipitation at the rain station (Fig. 1) is 2.18 ± 7.31 mm/day (3076.16 ± 1317.94 mm/year) between 2005 and 2010 (Menne et al., 2012).

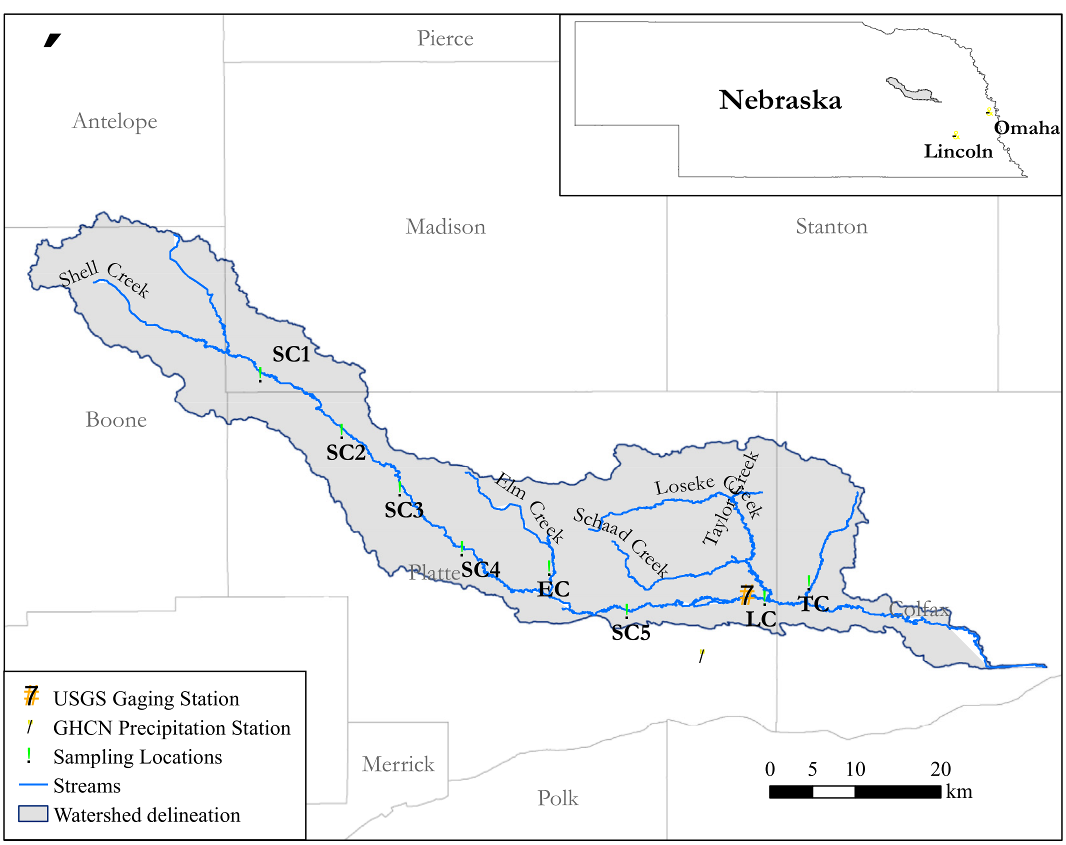

Fig. 1. Shell Creek watershed and monitoring stations.

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