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ORGANIC COMPOUNDS IN THE ENVIRONMENT

Occurrence of Antibiotics in an Agricultural Watershed in South-Central Idaho

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Abstract

The polar organic compound integrative sampler (POCIS) is a tool that has been effectively used to passively sample organic pollutants over long periods in aquatic environments. In this study, POCIS were used to investigate the spatial and temporal occurrence of 21 antibiotics in irrigation return flows and upstream sites of an intensively managed agricultural watershed in south-central Idaho. The antibiotic metabolite, erythromycin-H_2O, and the antibiotics monensin, oxytetracycline, sulfadimethoxine, sulfamethazine, sulfamethoxazole, trimethoprim, and tylosin were detected at frequencies ranging from 3.1 to 62.5%, with monensin having the highest rate of detection. The fact that monensin was the most frequently detected compound indicates that it is entering return flows in runoff from fields that had received livestock manure or wastewater. Antibiotics (except oxytetracycline, sulfamethazine, and tylosin) were also detected at an upstream site that consisted of diverted Snake River water and is the source of irrigation water for the watershed. Therefore, even cropped soils that are not treated with manure are still receiving low-level antibiotics during irrigation events. This study provides the first set of evidence that surface waters within this agricultural watershed contain antibiotic residues associated with veterinary and human uses.

Core Ideas

- Passive samplers were used to sequester antibiotics in surface waters.
- Eight of 21 antibiotics were detected in an intensively managed agricultural watershed.
- Monensin was the most frequently detected antibiotic (62.5% of samples).
- Antibiotics detected were associated with both veterinary and human uses.

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An estimated 100 to 200 Gg of antibiotic drugs are produced annually worldwide for the prevention and treatment of bacterial diseases in humans, animals, and crops (Franklin et al., 2016). In food-producing animals, the vast majority of medically important antibiotics are used as feed additives to enhance growth (FDA, 2014). Studies have shown that as much as 20 to 80% of antibiotics administered to animals can be excreted as the parent compound or metabolites in urine and feces (Arikan et al., 2007; Elmund et al., 1971; Winckler and Grafe, 2001). Because animal manures are regularly applied to agricultural soils to increase fertility, the antibiotic residues can be transported in runoff (Joy et al., 2013; Soni et al., 2015) and make their way to surface waters (Bartelt-Hunt et al., 2009; Dolliver and Gupta, 2008a; Kay et al., 2004). Additionally, veterinary antibiotics have been reported in runoff from unprotected manure stockpiles (Dolliver and Gupta, 2008b) and in groundwater underlying and downgradient from confined animal feeding operations (Bartelt-Hunt et al., 2011; Batt et al., 2006b). Antibiotics can also reach waterways from accidental or intentional spills of animal waste (Brands, 2014; Haack et al., 2015).

In addition to antibiotics from animal agriculture, antibiotics associated with clinical uses can reach the environment from municipal wastewater treatment plant effluent discharges (Batt et al., 2006a; Michael et al., 2013) and land application of municipal biosolids (Lapen et al., 2008). Regardless of source, antibiotic contamination is recognized as being widespread in water resources. During a nationwide survey of pharmaceutical compounds, 14 of 22 antibiotics were detected in up to 27% of 139 streams across 30 states (Kolpin et al., 2002). The occurrence of antibiotics at low concentrations may pose a health risk to humans and animals due to the proliferation of antibiotic resistance in the environment (Kemper, 2008). Subinhibitory antibiotic concentrations have been shown to act as signaling molecules between microorganisms and contribute to the evolution of antibiotic resistance by enriching for preexisting resistant bacteria and accelerating horizontal gene transfer, recombination, and mutagenesis (Andersson and Hughes, 2014; Davies and Davies, 2010; Knapp et al., 2008). Due to the potential

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Abbreviations: CD, Cedar Draw; DC, Deep Creek; HC, Hansen Coulee; IC, I Coulee; MC, Mud Creek; MD, MD, Milner Dam; NC, N Coulee; POCIS, polar organic compound integrative sampler; RC, Rock Creek; RCP, Rock Creek Poleline; TFC, Twin Falls Coulee; USR, Upper Snake Rock.

emergence and spread of antibiotic-resistant pathogens and the associated public health risks, there is great interest in determining the occurrence of antibiotics in waters affected by agricultural production and wastewater treatment plant effluents (Burkholder et al., 2007; Chee-Sanford et al., 2009; Jones-Lepp et al., 2012; Michael et al., 2013).

Passive sampling technologies are becoming a commonly accepted method for monitoring of organic contaminants and provide a cost-effective means for capturing low levels (Söderström et al., 2009). One such device is the polar organic compound integrative sampler (POCIS), which was designed to sequester polar compounds from water (Alvarez et al., 2005) and is particularly useful when concentrations are below detection limits in grab samples (Jaimes-Correa et al., 2015; Morin et al., 2012). Polar organic compound integrative samplers have been successfully used in a number of studies to determine the occurrence and quantity of antibiotics and other pharmaceutical compounds in surface waters, with comparable results to that of grab samples (Jaimes-Correa et al., 2015; Li et al., 2010).

Irrigation return flows, like runoff or drainage from nonirrigated watersheds, can transport sediment, nutrients, pathogens, and organic compounds from agricultural fields to surface waters (Bjorneberg et al., 2015). In this survey study, we evaluated the spatial and temporal occurrence of 21 antibiotics in surface waters of an intensively managed agricultural watershed in south-central Idaho. To sequester dissolved antibiotics, POCISs were deployed simultaneously during the irrigation season at eight return flow sites, where their flow ultimately discharges into the Snake River, and at two upstream sites. The antibiotics investigated in this study were selected to be representative of those that are likely used within the watershed. Many of the targeted antibiotics are used in both veterinary and human medicine and are considered medically important by the World Health Organization (WHO, 2016).

Materials and Methods

Watershed Description and Sampling Sites

The Upper Snake Rock (USR) watershed is 820 km^2 and is located along the south side of the Snake River in south-central Idaho (Fig. 1). This region has a semiarid climate and consists of cool wet winters and hot dry summers, with a mean annual temperature of 8.7°C and precipitation of 284 mm. The watershed is intensively managed, with a high percentage of the land in irrigated row crops (85%) and animal production. There are approximately 64,000 lactating dairy cows, 28,500 beef cattle, 80 pigs, 900 goats, and 12,300 sheep (NASS, 2014). In the USR watershed there are also 19 active aquaculture facilities raising mainly trout (Craig Thomas, personal communication, 2017). The manure solids (stacked and composted) and wastewater from the manure management systems, especially for dairy production, are applied to the surrounding cropland in the fall or spring.

About 65% of irrigated cropland is under furrow irrigation in the USR watershed, with the remainder under pressurized sprinkler irrigation. There are no rainfed crops in southern Idaho, and rainfall during the irrigation season (April through October) is negligible; thus, water in the return flows is predominantly from irrigation runoff. The rainfall total during the study period was 97 mm, whereas the amount of water applied through irrigation was about 1100 mm. All of the irrigation water is obtained from

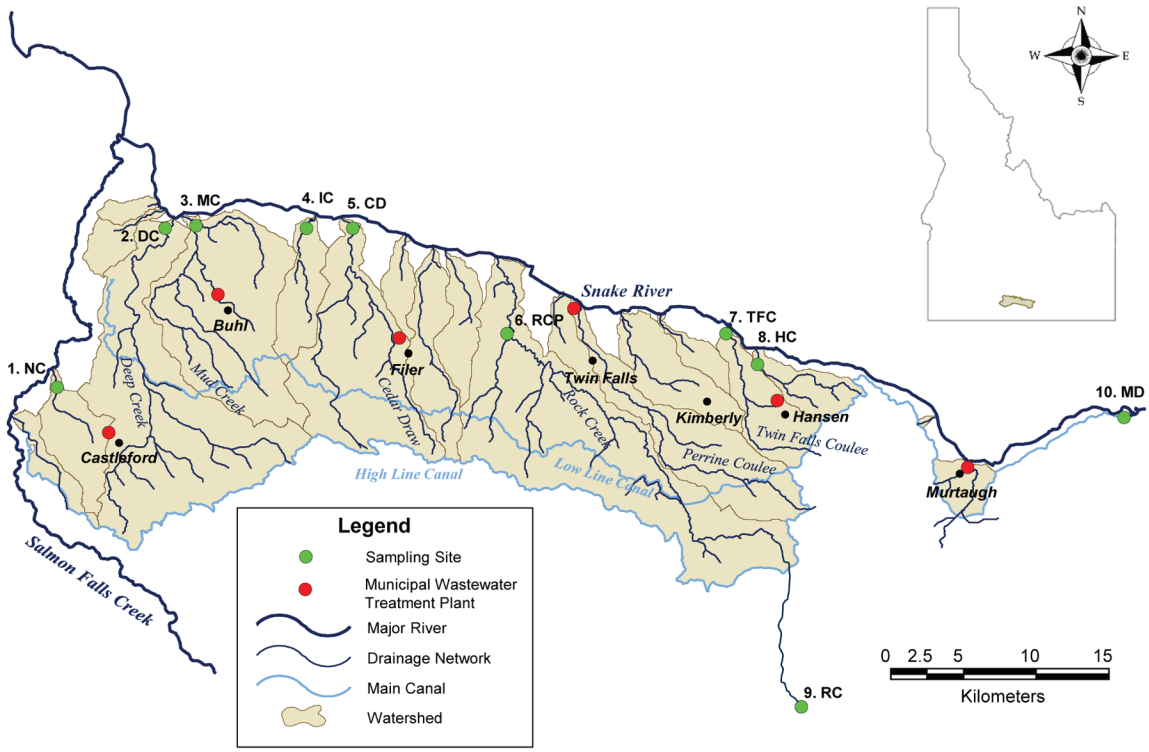

Fig. 1. The sampling sites within the Upper Snake Rock watershed in south-central Idaho. The POCIS were deployed at (1) N Coulee (NC), (2) Deep Creek (DC), (3) Mud Creek (MC), (4) I Coulee (IC), (5) Cedar Draw (CD), (6) Rock Creek Poleline (RCP), (7) Twin Falls Coulee (TFC), (8) Hansen Coulee (HC), (9) Milner Dam (MD), and (10) Rock Creek (RC). The Snake River flows to the west. The N Coulee and the other return streams (nos. 2–9) flow in a northerly direction into Salmon Falls Creek and Snake River, respectively.

the Snake River and is supplied in canals, ephemeral streams, and coulees as it is delivered to fields (Bjorneberg et al., 2008). Many streams flow only during the irrigation season, whereas others flow all year due to subsurface drain tiles and tunnels located sporadically throughout the watershed. Rock Creek is the only stream that flows into the watershed, and the upstream section is ephemeral, typically flowing only until early summer from snowmelt in the local mountains. Applying irrigation water to furrow-irrigated fields typically results in 20 to 50% of the water running off (Bjorneberg et al., 2015). The runoff water is reapplied to other fields, but a portion flows from the watershed to the Snake River through a return flow system.

The locations of the POCIS deployment sites in the watershed are presented in Fig. 1. The return flow sites from west to east were N Coulee (NC), Deep Creek (DC), Mud Creek (MC), I Coulee (IC), Cedar Draw (CD), Rock Creek Poleline (RCP), Twin Falls Coulee (TFC), and Hansen Coulee (HC). Upstream sites were Rock Creek (RC), which is 21 km south of Hansen, ID, and the start of the main irrigation canal near Milner Dam (MD). These latter two sites were designated as upstream sites (not background) because they are not pristine sites, and we expected to detect some antibiotics but at lower levels than in the irrigation return flows. Flow rates and water quality data at each irrigation return site can be found in Supplemental Tables S1 and S2, respectively.

POCIS and Field Deployment

Polar organic compound integrative samplers, holders, and canisters were obtained from Environmental Sampling Technologies. Each POCIS was composed of two sheets of microporous polyethersulfone membrane encasing a solid-phase sorbent (Oasis HLB, Waters Corp.). The membrane support rings, POCIS holders, and canisters were made of stainless steel. Triplicate POCISs were deployed at each site in canisters that were secured in the water by a metal cable and stake. The POCISs were deployed four times at each site during April to October 2015, with an exposure period of 2 wk (Table 1). Upon retrieval of a canister, the POCISs were placed inside a cooler with ice packs. At the laboratory, the POCISs were immediately processed by detaching them from the holder and rinsing with reverse osmosis–purified water to remove sediment and biofouling. The POCISs were dried using laboratory wipes, individually wrapped in aluminum foil, and placed inside a sealable plastic bag for storage at −20°C until processing by solvent extraction.

Chemicals

Antibiotics, internal standards, and surrogates were purchased from Sigma-Aldrich. Isotopically labeled internal standards were purchased from Cambridge Isotopes Laboratories, Inc. Detailed physical and chemical information about the antibiotics can be found in Supplemental Tables S3 and S4. Antibiotics were grouped based on chemical class, compatibility with solvents, and eluting conditions from POCIS sorbent. High purity–grade solvents used for POCIS extraction and liquid chromatography were purchased from Fisher Scientific.

Table 1. POCIS exposure periods in 2015.

Month	Deployment	Collection	Exposure period
			d
Apr.	10 Apr.	23 Apr.	13
June	4 June	18 June	14
Aug.	5 Aug.	19 Aug.	14
Sept./Oct.	24 Sept.	8 Oct.	14

POCIS Extraction and Analysis

Prior to solvent extraction, each POCIS device was disembled, and the contents were quantitatively transferred to glass chromatography columns by rinsing with 5 to 10 mL of methanol. Recovery tests from separate portions of the HLB sorbent were performed to determine the optimal recovery composition for each compound group. In brief, Group 1 analytes were eluted from the sorbent by slowly passing 50 mL of an 80:20 mixture of dichloromethane/acetone. The sorbent was then extracted a second time using 50 mL of methanol (Group 2). Each extract was spiked with 50 ng of internal standard and surrogate compound and then concentrated separately using a RapidVap N_2 Evaporation System (Labconco). Concentrated Group 1 extracts were then analyzed by liquid chromatography tandem mass spectrometry. After Group 1 analyses were completed, these extracts were combined with the Group 2 extracts, and the combined extracts were analyzed to ensure complete recovery of the Group 2 analytes. Surrogate compounds provided information on matrix effects relative to the internal standards but were not used to correct for multiple reaction monitoring suppression or enhancement of individual compounds. The $^{13}C_6$–isotope labeled internal standard for sulfamethazine provided correction for this antibiotic by isotope dilution; however, concentrations of other compounds are uncorrected for recovery. Complete details on the analysis by liquid chromatography tandem mass spectrometry can be found in Supplemental Table S5.

Results and Discussion

Of the 21 antibiotics targeted for quantification in the POCIS extracts, only eight compounds were detected at least once (tylosin) to as much as 23 times (monensin) during the sampling campaign (Table 2). The detection frequencies in the return flows for monensin (62.5%), oxytetracycline (6.3%), sulfadimethoxine (18.8%), sulfamethazine (37.5%), sulfamethoxazole (12.5%), trimethoprim (9.4%), and tylosin (3.1%) were markedly similar to those reported by Lissemore et al. (2006) in the Grand River watershed (Ontario, Canada) but lower than in other agricultural watershed studies (Jaimes-Correa et al., 2015; Zhang et al., 2012). Erythromycin-H_2O (anhydroerythromycin) exhibited a detection frequency of 15.6% in this study, with no reportable values from other agricultural watershed studies.

At the upstream site RC, erythromycin-H_2O and monensin were detected once each, which was surprising because this location is relatively isolated. Although the land area upstream from this sampling point is national forest, one potential source of the antibiotics is open-range cattle that occasionally inhabit the area. Airborne transport of antibiotics has also been reported in dust from animal feeding operations (McEachran et al., 2015); thus, deposition of antibiotics is another possibility. At the other upstream site (MD), the occurrence of antibiotics was expected because the canal water at this site is diverted Snake River water. The likely sources of the antibiotics (i.e., erythromycin-H_2O, monensin, sulfadimethoxine, sulfamethoxazole,

and trimethoprim) are agricultural irrigation return flows and municipal wastewater effluents that discharge into the river at many locations upstream of this sampling site. Agricultural watersheds and wastewater treatment plant effluents have been identified as sources of antibiotics as well as a variety of other persistent pharmaceuticals (Arikan et al., 2008; Batt et al., 2006a; Jaimes-Correa et al., 2015).

Monensin is an ionophore exclusively used in the cattle and poultry industries to prevent coccidiosis and to increase weight gain and milk production efficiency. By weight, it can be one of the most significant antibiotics used on a dairy farm (Watanabe et al., 2008). In this study, monensin was the most frequently detected antibiotic, and it was present at all eight irrigation return sites (Table 2). The mean mass of monensin recovered ranged from 0.15 to 2.3 ng POCIS^{-1}. Monensin was always detected in

Table 2. Mean mass of pharmaceuticals recovered from POCIS that were deployed in the Upper Snake Rock watershed

Site†	Month	Erythromycin-H_2O	Monensin	Oxytetracycline	Sulfadimethoxine	Sulfamethazine	Sulfamethoxazole	Trimethoprim	Tylosin
		ng POCIS^{-1}							
1. NC	Apr.	–‡	1.3 ± 0.12	–	–	–	§	§	–
	June	–	0.38 ± 0.14	–	1.2 ± 0.95¶	–	§	–	–
	Aug.	§	0.31 ± 0.03¶	–	§	–	–	–	–
	Sept./Oct.	0.51 ± 0.05#	–	–	–	–	0.51 ± 0.57¶	–	–
2. DC	Apr.	–	1.4 ± 0.25	–	1.1 ± 0.06	–	1.6 ± 0.35	–	–
	June	–	0.38 ± 0.05	–	–	3.3 ± 1.3¶	–	–	–
	Aug.	–	§	–	0.12 ± 0.02	0.19 ± 0.09¶	–	–	–
	Sept./Oct.	0.20 ± 0.07	0.32 ± 0.09	80.1 ± 16.1	§	–	0.67 ± 0.14§	–	0.06 ± 0.004¶
3. MC	Apr.	–	0.63 ± 0.19	23.5 ± 16.0	–	4.7 ± 0.86¶	–	–	–
	June	–	0.70 ± 0.21	–	§	19.7 ± 6.5	–	–	–
	Aug.	–	–	–	–	0.39 ± 0.03¶	–	–	–
	Sept./Oct.	0.23 ± 0.06	0.15 ± 0.005	–	0.66 ± 0.19	3.4 ± 0.83	–	–	–
4. IC	Apr.	–	2.3 ± 0.50	–	–	–	–	–	–
	June	–	0.38 ± 0.12	–	–	31.4 ± 37.7¶	–	–	–
	Aug.	§	–	–	–	–	–	–	–
	Sept./Oct.	0.20 ± 0.19	–	–	–	–	–	–	–
5. CD	Apr.	–	0.71 ± 0.08	–	–	–	–	1.2 ± 0.41	–
	June	–	0.29 ± 0.13	–	–	8.1 ± 4.1	–	–	–
	Aug.	–	–	–	0.48 ± 0.19	0.22 ± 0.09	–	17.7 ± 3.5	–
	Sept./Oct.	–	–	–	§	§	–	–	–
6. RCP	Apr.	–	1.5 ± 0.30	–	–	–	§	8.2 ± 0.80¶	–
	June	–	0.49 ± 0.12	–	–	6.8 ± 1.3	§	–	–
	Aug.	§	–	§	0.30 ± 0.10¶	–	–	–	–
	Sept./Oct.	0.22 ± 0.15	0.21 ± 0.04	–	–	–	1.2 ± 0.26	–	–
7. TFC	Apr.	–	1.0 ± 0.26	–	–	–	§	–	–
	June	–	0.79 ± 0.14	–	–	19.2 ± 6.2	§	–	–
	Aug.	§	§	–	–	–	–	–	–
	Sept./Oct.	§	–	–	–	–	–	–	–
8. HC	Apr.	–	0.58 ± 0.31¶	–	–	–	§	–	–
	June	–	0.69 ± 0.05	–	§	21.2 ± 6.9	§	–	–
	Aug.	–	§	–	–	–	–	–	–
	Sept./Oct.	§	–	–	–	–	–	–	–
9. RC††	Apr.	–	0.08 ± 0.03	–	§	–	§	–	–
	June	–	–	–	–	–	–	–	–
	Aug.	–	–	–	–	–	–	–	–
	Sept./Oct.	0.28 ± 0.13	–	–	§	–	–	–	–
10. MD††	Apr.	–	0.23 ± 0.09	–	4.2 ± 4.5¶	–	1.2 ± 0.39	3.0 ± 0.80	–
	June	–	0.14 ± 0.04	–	§	–	§	–	–
	Aug.	–	–	–	–	–	–	–	–
	Sept./Oct.	0.19 ± 0.06¶	–	–	–	–	0.36 ± 0.03	–	–

† CD, Cedar Draw; DC, Deep Creek; HC, Hansen Coulee; IC, I Coulee; MC, Mud Creek; MD, Milner Dam; NC, N Coulee; RC, Rock Creek; RCP, Rock Creek Poleline; TFC, Twin Falls Coulee.

‡ Below detection limit.

§ Analyte above detection limit in one of three POCIS but not reported.

¶ Mean ± SD of duplicate POCIS.

Mean ± SD of triplicate POCIS unless stated otherwise.

†† Upstream site; not irrigation return flow.

April and June at all sites but was detected only once in August (at NC) and three times in September/October (at RCP, MC, and DC). In addition, the general trend at the sites was that the mass of monensin recovered from the POCIS was greatest in April and lower during each subsequent sampling period (Fig. 2). Monensin was also detected at MD in April and June, and, because it is the source of irrigation water in the watershed, it is logical that monensin was detected at all return flow sites during the same period. The mass recovered was higher at the return flow sites than at MD, indicating that field runoff transported additional monensin to the irrigation return flows. Because the irrigation water becomes available in early April, the highest concentration of monensin in field runoff and return flows can be expected at this time. The flow rates in the irrigation returns were also generally at their lowest level in April (Supplemental Table S1); thus, this could be an additional contributing factor.

Steers fed oral doses of radiolabeled monensin were found to excrete about 50% as unmetabolized monensin, with none excreted in urine (Donoho et al., 1978). Like the excretion of other antibiotics by animals, monensin is readily disseminated on farm and to environments located near agricultural operations. As a result, monensin has been detected in wastewater lagoons, shallow groundwaters, river sediments, and surface waters (Bartelt-Hunt et al., 2011; Hafner et al., 2016; Kim and Carlson, 2006; Lissemore et al., 2006). In a rainfall simulation study conducted by Davis et al. (2006), monensin was reported to have the greatest runoff potential among a mixture of antibiotics (including tetracycline, chlortetracycline, sulfathiazole, sulfamethazine, erythromycin, and tylosin) that was applied to the soil surface. However, only 0.08% of the applied monensin was lost in surface runoff, with Dolliver and Gupta (2008a) also reporting low mass losses of <0.05 to 2% in runoff and <0.005% in leachate.

Sulfamethazine (also called sulfadimidine) is approved for use in beef cattle, swine, and poultry to treat a variety of bacterial diseases and was designated a highly important antibiotic for human medicine (WHO, 2016). It was the second most frequently occurring antibiotic in the irrigation return flows and was detected at least once at all sites except NC (Table 2). The mean mass of sulfamethazine recovered ranged from 0.22 to 31.4 ng POCIS^{-1}. Sulfamethazine was generally not detected in the return flows in April, but it was detected in June and August. Sulfamethazine detection in MC was the exception to this trend because it was detected during all four sampling periods. The MC sampling site was immediately downstream of the confluence of two return flows, both of which contain waters from a relatively large network of ditches (Fig. 1). This area, however, does not contain extensive land under crop production due to shallow soils overlying basalt and undulating topography. Although a few dairies and fish farms can be found in this immediate area, sulfamethazine is not approved for use in lactating dairy cows (NMPF, 2016) and aquaculture (FDA, 2017). Potential sources are manure deposition from beef cattle (stockers) on pasture and a wastewater treatment plant that discharges treated effluent into this area of the USR watershed.

Other sulfonamides detected in the irrigation return flows were sulfadimethoxine (six occurrences) and sulfamethoxazole (four occurrences). Both antibiotics were detected at the RCP, DC, and NC sampling sites, whereas sulfadimethoxine

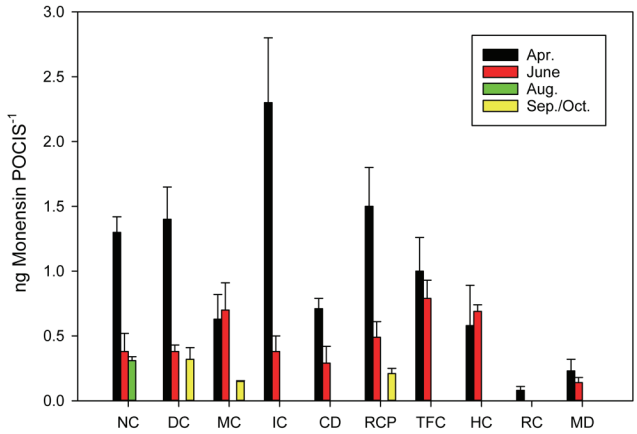

Fig. 2. Mass of monensin extracted from POCIS that were deployed at the sampling sites within the Upper Snake Rock watershed in south-central Idaho. CD, Cedar Draw; DC, Deep Creek; HC, Hansen Coulee; IC, I Coulee; MC, Mud Creek; MD, Milner Dam; NC, N Coulee; RC, Rock Creek; RCP, Rock Creek Poleline; TFC, Twin Falls Coulee. Columns represent means ± SD (*n* = 3).

was also detected at CD and MC sites. The mean mass of these sulfonamides ranged from 0.12 to 1.6 ng POCIS^{-1}. Sulfadimethoxine and sulfamethoxazole were also detected upstream at MD in April, with recovered masses of 4.2 and 1.2 ng POCIS^{-1}, respectively, which are generally equal to or higher than the sampling sites. These sulfonamides, however, were not detected at all return flow sites in April, indicating that transport of dissolved antibiotics is complicated. Batt et al. (2006b) found detectable levels of sulfadimethoxine (and sulfamethazine) in groundwater affected by agricultural sources in western Idaho. Among many other sulfonamides, sulfadimethoxine and sulfamethoxazole have been identified as highly important for human medicine (WHO, 2016). Sulfadimethoxine is also approved for use in nonlactating dairy cattle (i.e., bulls, calves, and replacement heifers), beef cattle, poultry (chickens, turkeys), and aquaculture. Sulfamethoxazole (often used in combination with trimethoprim) is approved for use in pets (dogs, cats) and horses.

Compared with the other antibiotics that were detected, erythromycin-H_2O, oxytetracycline, trimethoprim, and tylosin were generally detected less frequently in the irrigation return flows from April to October (Table 2). Oxytetracycline and trimethoprim, both of which are used in veterinary and human and medicine, are considered highly important antimicrobials (WHO, 2016). Tylosin is used exclusively in veterinary medicine but is considered critically important because it is structurally related to erythromycin and other macrolides (Marshall and Levy, 2011). Although erythromycin was not detected in POCIS extracts, its main degradation product, erythromycin-H_2O, was detected at five sampling sites (i.e., RCP, IC, MC, DC, and NC) during the September/October campaign. Erythromycin-H_2O was also detected in the other return flows (i.e., HC, TFC, and CD), but values were not reported because erythromycin-H_2O was detected in extracts from only one of three POCIS. Under acidic conditions, erythromycin is unstable and loses one water molecule to form erythromycin-H_2O (Göbel et al., 2004). However, the average pH of the irrigation return flows was near 8, and it did not drop below 7.4 during the sampling campaigns (Supplemental Table S2).

Alternatively, erythromycin could have been dehydrated by gastric acid after being ingested (Fan et al., 2009). In a composited sample of fresh dairy manure collected at a local composting facility in south-central Idaho, we detected erythromycin-H_2O (not erythromycin) at a parts per billion level (data not shown). Erythromycin-H_2O was detected at a similar level in composted manure at the same facility.

In conclusion, we demonstrated that POCISs can be used to effectively sequester antibiotics at low levels due to their ability to accumulate dissolved compounds over long periods of time. Out of 21 target compounds, seven antibiotics and one antibiotic metabolite were present in return flows during the irrigation season in the USR watershed, with detection frequencies ranging from 3.1 to 62.5%. Five of the antibiotics detected are associated with both veterinary and human uses, whereas monensin and tylosin are for veterinary use only. Except in the case of monensin, there was no relationship between antibiotic level in the POCIS and month during the irrigation season. Due to the abundant use of monensin in dairy and beef cattle production, its high detection frequency suggests that it is entering the irrigation returns in runoff from fields that had received livestock manure or wastewater. Manure could be the source of the other antibiotics detected, but wastewater treatment plant effluents, aquaculture effluents, and biosolids-treated soils are other likely sources. It should be noted that cropped soils in the watershed that are not treated with manure, wastewater, or biosolids still receive low-level antibiotics because the canal water at Milner Dam and return flows contained antibiotic residues, and both are used for irrigation. Ultimately, these antibiotic residues make their way to the Snake River via the irrigation return flows. The information from this study raises questions about antibiotic and land use practices in the USR watershed and can be a starting point for discussions between farmers, land managers, scientists, and regulatory authorities about how to manage antibiotics.

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