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Pharmaceuticals in a temperate forest-water reuse system

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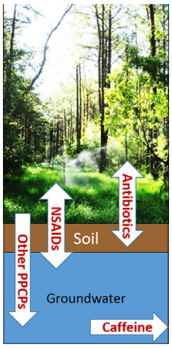
HIGHLIGHTS

- Forest-water reuse systems sustainably treat and manage wastewaters through land-application to natural forests.
- Fate and effects of wastewater contaminants remain uninvestigated in these systems.
- 33 pharmaceuticals were quantified in wastewater, groundwater, surface water, and soil.
- Land application of municipal wastewater contributes pharmaceuticals to the forest-water reuse system.
- Pharmaceutical concentrations observed do not pose a significant risk to human health.

GRAPHICAL ABSTRACT



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ABSTRACT

Forest-water reuse systems infiltrate municipal, industrial, and agricultural wastewaters through forest soils to shallow aquifers that ultimately discharge to surface waters. Their ability to mitigate regulated nutrients, metals, and organic chemicals is well known, but the fate of non-regulated chemicals in these systems is largely unstudied. This study quantified 33 pharmaceuticals and personal care products (PPCPs) in soils, groundwaters, and surface waters in a 2000-hectare forest that receives ~1200 mm/year of secondary-treated, municipal wastewater in addition to natural rainfall (~1300 mm/year). This forest-water reuse system does contribute PPCPs to soils, groundwater, and surface waters. PPCPs were more abundant in soils versus underlying groundwater by an order of magnitude (5–10 ng/g summed PPCPs in soil and 50–100 ng/L in groundwater) and the more hydrophobic chemicals were predominant in soil over water. PPCP concentrations in surface waters were greater at the onset of significant storm events and during low-rainfall periods when total summed PPCPs were >80 ng/L, higher than the annual average. With few exceptions, the margins of exposure for PPCPs in groundwater and surface waters were several orders of magnitude above values indicative of human health risk.

1. Introduction

Climate change and population pressures are expected to increase the scarcity and diminish the quality of water resources (Ingram et al., 2013). While significant attention has been paid to more arid regions of the world, temperate climates are also experiencing water availability shortages and competition globally (Ingram et al., 2013). As such, managing water security through water re-use will become increasingly important as urbanization and human population growth continues. One strategy to improve water quality and regulate water availability is the use of forest-water reuse systems (Braatz and Kandiah, 1996; Isosaari et al., 2010). Globally, forests are crucial to water resource capacity for

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30% of the major cities in the world (Dudley and Stolton, 2003). Worldwide, forest water-reuse systems treat municipal wastewater through land application of wastewater to managed tree plantations as well as to natural forest stands (Nichols, 2016).

Forest water-reuse systems are slow-rate land application systems that treat municipal, industrial, and agricultural wastewaters via soil infiltration and groundwater recharge prior to surface water discharge (Crites, 1984; Pound and Crites, 1973). Slow-rate irrigation is the most ubiquitous and oldest wastewater land application technology and can be designed for different natural and managed landscapes such as crop lands, golf courses, and forest ecosystems (Crites, 1984; McKim et al., 1982). Relative to other land application technologies, slow-rate infiltration systems are designed to treat the greatest amount of wastewater on a given amount of land by using irrigation rates in excess of crop needs (Crites et al., 2014). In temperate regions, forest slow-rate infiltration systems enable year-round wastewater treatment, nutrient storage, and intermittent soil saturation without known adverse impacts on forest integrity and water quality. When compared to traditional wastewater treatment plants (WWTP) of similar wastewater volume treatment capacity, these systems are more cost-effective and energy efficient. Forest-water reuse systems provide ecosystem services such as provisioning wood products, biodiversity, and carbon storage that conventional wastewater treatment systems cannot provide (Shifflett et al., 2014). Under proper management and design, these systems effectively mitigate regulated nutrients, metals, and organics to acceptable water quality criteria levels for groundwater and surface waters (Crites, 1984; Crites et al., 2014; Hutchins et al., 1985), which is another important ecosystem regulating service. However, very little is known about the fate and effects of non-regulated wastewater contaminants in these water-reuse systems and studies are needed to understand the extent to which these systems regulate water quality.

Contaminants of emerging concern, specifically pharmaceuticals and personal care products (PPCPs), are non-regulated contaminants that have been routinely reported in the environment (Barnes et al., 2008; Boyd et al., 2003; Kim et al., 2007; Kolpin et al., 2002). These contaminants can elicit reproductive (Bringolf et al., 2010; Panter et al., 1998), behavioral (Weinberger Ii and Klaper, 2014), and developmental (Olmstead and LeBlanc, 2000) effects on aquatic organisms. While the environmental input of PPCPs has been documented from many sources (i.e. animal waste, landfill leachate, etc), municipal wastewater release to surface waters represents the largest source as most wastewater treatment technologies do not completely remove them prior to discharge to surface water bodies (Metcalfe et al., 2010; Papageorgiou et al., 2016; Ternes et al., 2004). Various studies have reported concentrations of PPCPs in surface waters (de Jongh et al., 2012) and groundwaters (Meffe and de Bustamante, 2014) at levels far below acceptable daily intake doses for humans, but human health risks may exist when pharmaceuticals remain in water sources through drinking water intake. Exposure to unregulated pharmaceuticals is often calculated relative to acceptable intake values in adult humans to assess risk (Simazaki et al., 2015; Webb et al., 2003), and margins of exposure are rarely exceeded in drinking water (de Jongh et al., 2012). To date, PPCPs in forest-water reuse systems have yet to be evaluated relative to human health risks.

This study reports on the monthly occurrence, concentration, and human health risk of 33 PPCPs in applied wastewater, soils, groundwaters, and surface waters with seasonal changes for a full year. This temperate forest-water reuse system has been irrigated with secondary-treated, municipal wastewater for two decades. Previous research has documented that this system effectively manages regulated nutrients for groundwater and surface water protection (Shifflett et al., 2014). A recent hydrological study of this system found that municipal wastewater comprised 50–76% of groundwater and 23% of surface water in irrigated catchments (Birch et al., 2016). Additionally, select PPCPs were identified in forest groundwater and surface water leaving the irrigated subwatershed during a preliminary investigation to ascertain if PPCPs were present (McEachran et al., 2016).

This work expands on the previous preliminary investigation by extending the sampling period to one year and addressing the seasonal differences in PPCP concentrations to better assess ecosystem and water regulating services for this system given the biological functioning of both built (reservoir lagoons) and natural (forest and soils) components. Preliminary research also did not address PPCP presence and concentrations in soils, an important compartment for PPCP mitigation and transport to groundwater. The occurrence and concentrations of PPCPs in soils and waters were evaluated using physiochemical parameters (log Kow, Koc), and margins of exposure (MOE) of PPCPs in groundwater were calculated to evaluate human health exposure potential via drinking water, which has not been previously assessed in forest-water reuse systems. Communities in this area utilize groundwater for drinking water resources, and quantifying PPCP human health risks is warranted to ensure that forest-water reuse systems are designed and implemented as an environmentally safe and sustainable water reuse technology.

2. Materials and methods

2.1. Study area and sampling

The majority of the study area of interest has been previously described (Birch et al., 2016; McEachran et al., 2016; Shifflett et al., 2014). Briefly, the study site is a 2000 ha, wastewater land-application facility which services a municipality of 70,000 defined by a large military presence and young (average age 22.8 years) population (United States Census Bureau, 2015). After a 7–14-day residence time in open reservoirs, secondary-treated wastewater is land-applied onto 930 ha of forested land in the 2000 ha watershed using one meter irrigation risers with weekly application rates of 25–75 mm. The 930-hectare land-application area receives approximately 1200 mm of wastewater and 1300 mm of precipitation annually (Birch et al., 2016; Shifflett et al., 2014) (see Supplementary material Fig. S2). Regulatory permits require that wastewater is applied only when conditions support infiltration of wastewater into soils; wastewater pooling and run-off at the soil surface is not allowed (Pound and Crites, 1973) and was not observed throughout the sampling period, even during periods of extreme rainfall when application still occurred under permit to reduce reservoir water levels.

Wastewater, groundwaters, and surface waters were collected over 12 consecutive months from August 2014 to July 2015 for analysis of PPCPs. Prior data were included from August 2014 to January 2015 (McEachran et al., 2016) and are identified in the tables and figures below. Wastewater was collected from a central distribution spigot within the irrigation network and reflects wastewater after secondary treatment and storage in open reservoirs. Surface water samples were collected from Southwest Creek upstream of the land-application facility (Upstream site), at a small tributary within the site (TA SW sites), and at the subwatershed outlet on Southwest Creek (Outlet site) shown in Supplementary Material Fig. S1. Groundwater samples were collected along two separate gradients in two separate catchments, Transect A (TA 1–2, Well 3) and Transect B (TB 1–2), and from a reference groundwater well in a forested area that is not irrigated with wastewater (Reference). Surface and groundwater sampling procedures followed USGS field manual protocols (Wilde et al., 1999) and have been previously described (McEachran et al., 2016). One-liter amber glass sampling bottles were cleaned, solvent rinsed with acetone, and baked at 300 °C for 24 h before use. All water samples were, transported to the laboratory on ice and stored at 4 °C until extraction, within 7 days of sampling. A separate surface water sampling event took place during a 24-hour storm event in July 2015. The stage of the watershed outlet surface water was continuously monitored during

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