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Investigating dynamic sources of pharmaceuticals: Demographic and seasonal use are more important than down-the-drain disposal in wastewater effluent in a University City setting

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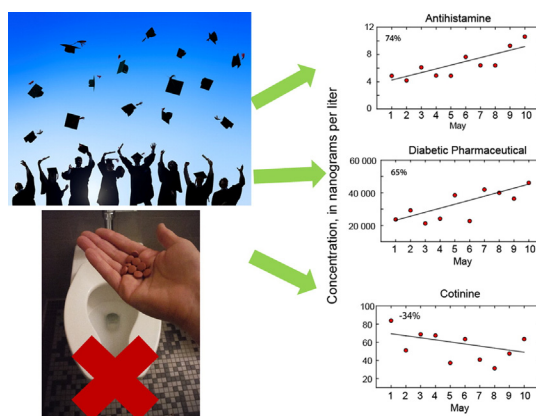
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HIGHLIGHTS

- Flushing unused medication not a common practice among University students
- Pharmaceutical concentration data in effluent supports survey data that flushing is not a common disposal practice
- Increased concentrations of several pharmaceuticals reflect a demographic shift toward an older population
- Increased concentrations of antihistamines may reflect increased use of pharmaceuticals to treat allergies

GRAPHICAL ABSTRACT



Credits: iStock.com/Rawpixel Ltd. (top left), C. Vatovec (bottom left).

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ABSTRACT

Pharmaceutical pollution in surface waters poses risks to human and ecosystem health. Wastewater treatment facilities are primary sources of pharmaceutical pollutants, but little is known about the factors that affect drugs entering the wastewater stream. This paper investigates the effects of student pharmaceutical use and disposal behaviors and an annual demographic shift on pharmaceutical pollution in a university town. We sampled wastewater effluent during a ten-day annual spring student move-out period at the University of Vermont. We then interpreted these data in light of survey results that investigated pharmaceutical purchasing, use, and disposal practices among the university student population. Surveys indicated that the majority of student respondents purchased pharmaceuticals in the previous year. Many students reported having leftover drugs, though only a small portion disposed of them, mainly in the trash.

We detected 51 pharmaceuticals in 80% or more of the wastewater effluent samples collected over the ten-day sampling period. Several increased in concentration after students left the area. Concentrations of caffeine and nicotine decreased weakly. Drug disposal among this university student population does not appear to be a major source of pharmaceuticals in wastewater. Increases in pharmaceutical concentration after the students

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left campus can be tied to an increase in the seasonal use of allergy medications directly related to pollen, as well as a demographic shift to a year-round older population, which supports national data that older people use larger volumes and different types of pharmaceuticals than the younger student population.

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1. Introduction

Pharmaceuticals are increasingly acknowledged as chemicals of emerging concern (CEC) (Daughton and Ternes, 1999; Vasquez et al., 2014) with known negative impacts on aquatic species (Brooks, 2014; Crane et al., 2006) and ecosystems (Celiz et al., 2009), and potential impacts on human health (Bruce et al., 2010; Cunningham et al., 2009; Kumar et al., 2010). Wastewater treatment facilities (WWTF) are major sources of human pharmaceuticals in the environment (Liu and Wong, 2013; Boxall et al., 2012; Azuma et al., 2015; Jain et al., 2013; Ort et al., 2010; Venkatesan and Halden, 2014); they were not designed to remove these chemicals, and as a result can release these compounds into surface waters (Verlicchi et al., 2012). WWTFs receive pharmaceutical compounds from consumers via three primary routes: excretion from the human body, bathing that washes off topically-applied medications, and disposal of unwanted drugs down-the-drain (Daughton and Ternes, 1999; Daughton and Ruhoy, 2008). Recent research in environmental risk assessment has called for integrated approaches to assessing pharmaceutical disposal practices in relation to wastewater (Petrie et al., 2016). The intent of this research was to determine what effect an annual university student spring move-out had on pharmaceutical concentrations in wastewater effluent, and in particular whether we could discern a change in pharmaceutical concentrations due to drug disposal as students departed a small university city in May 2014. As we will describe, our findings from both wastewater effluent sampling and a behavioral survey suggest something quite to the contrary.

Although direct disposal of pharmaceuticals to wastewater through flushing of unused medications has been suggested as a means of pharmaceuticals in wastewater (Braund et al., 2009; Tong et al., 2011; Vellinga et al., 2014), studies have generally used population surveys in lieu of actual measurement of pharmaceuticals in wastewater to estimate the importance of direct disposal. However, scholars have noted that there is little to no data to show the importance of flushing of medications in impacting observed concentrations of pharmaceuticals in wastewater samples (Daughton, 2016; Petrie et al., 2016). Thus, the occasional direct disposal of unused medications by consumers is sufficiently small in comparison to normal excretion patterns that it is not expected that the effects of these inputs would not be observable in wastewater samples. Studies that have shown an impact of direct disposal have attributed increases in observed wastewater concentrations to disposal of fluoxetine from a pharmacy (Petrie et al., 2016) or to direct disposal of MDMA (3,4-methylenedioxymethamphetamine) by an illegal production facility in response to a raid by law enforcement personnel (Emke et al., 2014).

The limitations of most studies of direct disposal of pharmaceuticals to the wastewater system by consumers, then, is that survey data is not often linked to wastewater measurement. Even if survey data and wastewater sampling were coordinated, the temporally random nature of pharmaceutical disposal by patients would result in it being very difficult to discriminate between normal excretion and direct disposal. Indeed, the few studies of the effect of direct disposal have depended on coincident sampling during law enforcement activities or patterns of racemic mixtures of pharmaceuticals that strongly indicate but do not directly show disposal by a pharmacy.

1.1. Leftover medications

Current estimates suggest that 49% of Americans have used prescription medications within the past 30 days (CDC., 2015), and 82% of adults

have used non-prescription, or over-the-counter (OTC), drugs (IMS., 2013). A large volume of these medications go unused, as evidenced by the >350 tons of prescriptions drugs collected at the most recent National Drug Take-Back Day (NDTB) in September 2015 (DEA., 2015). Surveys have indicated that the majority of Americans dispose of unwanted pharmaceuticals down the drain (31% to 54%) or in the trash (54% to 59%) (Kuspis and Krenzelo, 1996; Tong et al., 2011; Wiczorkiewicz et al., 2013). In order to reduce pharmaceutical contamination of aquatic environments, multiple government agencies and non-profit organizations have developed public education campaigns to end the practice of flushing unused medications down-the-drain (Environmental Protection Agency E, 2016; Product Stewardship Institute, 2014). However, a quantitative link between flushing as a source of pharmaceuticals being discharged to the environment has not yet been established. Understanding how the population within a WWTF catchment area uses and disposes of pharmaceuticals, in relation to pharmaceuticals observed in wastewater (Venkatesan and Halden, 2014) is an important factor in understanding the efficacy of interventions designed to minimize the volume of these compounds that enter surface waters (Castensson and Ekedahl, 2010).

1.2. Short-term factors affecting pharmaceuticals in wastewater

Recently, researchers have begun assessing various short-term social and cultural factors that affect the presence and concentrations of pharmaceutically-active compounds in wastewater and surface waters. Two of the primary factors that have been studied to date are events that increase the recreational use of illicit drugs within a sewershed, and seasonal health changes that lead to increased use of certain medications (Kasprzyk-Hordern and Baker, 2012; Singer et al., 2014; Jiang et al., 2014; Gerrity et al., 2011).

The recreational use of illicit drugs has been shown to cause significant fluctuations in wastewater pharmaceutical concentrations over a short period of time. One study examining the effect of a Taiwanese youth festival (600,000 music fans) on CEC in wastewater and surface water reported that the large influx of concert-goers had a significant impact on the concentrations of several compounds including a suite of illicit drugs (Jiang et al., 2014). Similarly, a study on CEC in wastewater during the National Football League Super Bowl found a significant increase in illicit drugs, including cocaine during the event as compared to baseline weekends, indicating the impact that a large cultural sporting event can have on CEC pollution (Gerrity et al., 2011). Furthermore, the size and demographics of a population center can influence the volume of illicit drugs in wastewater (Yargeau et al., 2014).

In comparison, studies investigating the effect of seasonal health-induced changes in pharmaceutical use on wastewater effluent show that a range of prescription and OTC medications change based upon the needs of the human population. The drugs ephedrine and pseudoephedrine have been found in higher concentration in the winter months as compared to the summer, presumably because these drugs are commonly found in OTC cold medications (Kasprzyk-Hordern and Baker, 2012). Several studies have focused on increased concentrations of antibiotics (Singer et al., 2014) and antivirals (Azuma et al., 2015; Leknes et al., 2012) in wastewater due to influenza pandemics. Such studies are of interest in understanding the ecological effects of pharmaceutical pollution because these changes can result in increased antibiotic resistance or antiviral resistance in bird populations (Ghosh et al., 2010; Jain et al., 2013; Orozovic et al., 2011; Soderstrom et al., 2009).

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