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HAŽARDOUS MATERIALS

Journal of Hazardous Materials

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

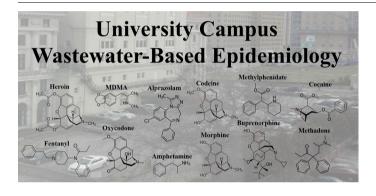
Tracking narcotics consumption at a Southwestern U.S. university campus by wastewater-based epidemiology



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GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords: Sewage-based epidemiology Urban metabolism metrology College campus Fentanyl Heroin

ABSTRACT

Wastewater-based epidemiology (WBE) was applied to estimate the consumption of twelve narcotics within a Southwestern U.S. university campus. Seven consecutive 24-hour composite raw wastewater samples (n = 80) were obtained once per month from sampling locations capturing > 95% of campus-generated wastewater. Samples were analyzed for indicators of consumption of morphine, codeine, oxycodone, heroin, fentanyl, methadone, buprenorphine, amphetamine, methylphenidate, alprazolam, cocaine, and MDMA using LC–MS/MS. Eleven indicator compounds (oxycodone, codeine, norcodeine, 6-acetylmorphine, EDDP, amphetamine, alprazolam, alpha-hydroxyalprazolam, cocaine, benzoylecgonine, and MDMA) occurred at 100% detection frequency across the study, followed by morphine-3-glucuronide (98%), noroxycodone (95%), methylphenidate (90%), heroin (7%), norfentanyl (7%), and fentanyl (5%). Estimates of average narcotics consumption ranked as follows in units of mg/day/1000 persons: heroin (474 ± 32), cocaine (80 ± 6), alprazolam (256 ± 12), methylphenidate (236 ± 28), methadone (72 ± 8), oxycodone (80 ± 6), alprazolam (60 ± 2), MDMA (88 ± 35), codeine (50 ± 4), and morphine (18 ± 3). This campus-based WBE study yielded baseline data on 12 narcotics for a U.S. campus and demonstrated for the first time the feasibility of detecting the fentanyl metabolite norfentanyl in this setting.

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https://doi.org/10.1016/j.jhazmat.2018.07.073

Received 17 April 2018; Received in revised form 17 July 2018; Accepted 18 July 2018 Available online 24 July 2018 0304-3894/ © 2018 The Author(s). Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/BY-NC-ND/4.0/).

1. Introduction

College-aged adults (ages 18-22) throughout the United States have historically been associated with the highest percentage of drug abuse rates of all U.S. age groups, with 24-28% of respondents in a 2016 survey admitting to illicit drug use within the past 30 days [1]. While 48% of high school respondents to the same survey reported trying at least one illicit drug in their lifetime, prevalence of drug use has been shown to be higher for those aged 18-29 years [1]. Americans between the ages of 15–24 have seen some of the lowest rates of overdose death (4-10 deaths per 100,000 persons) across all age categories from 2000 to 2016, but the subsequent age group (25-34 years) has been identified with the highest number of drug overdose deaths in 2016 (35 +deaths per 100,000 persons) [2]. This observation may be partially explained through drug-related associative learning, where drugseeking habits are sustained later in life, while the subjective effects that initially encouraged the drug use diminish [3]. Continued neurological development in the early 20 s [4] coupled with changes in brain chemistry due to drug use [5] may have a marked effect on this demographic group.

Addressing substance abuse within college-aged adults should be viewed as a principal task - but understanding the extent of misuse within this age category is met with significant difficulty. Current data analysis involves a combination of population surveys, crime statistics, medical records and narcotic seizure data [6], but these analyses provide data on previous years and may not adequately capture the current state of drug use. The costly and cumbersome procedures may also inject into studies unwanted bias through misrepresentation in self-reporting surveys [6]. First proposed in 2001 [7], wastewater-based epidemiology (WBE) has been shown to constitute a viable alternative to current methods of collecting data on narcotics use and abuse [8–12]. This tool has been applied worldwide to obtain narcotics abuse statistics in near-real time for varying population sizes [13-16]. The concept of tracking the consumption and fate of chemicals through diagnostics rooted in the analysis of urban process streams has been further expanded under the umbrella and moniker of urban metabolism metrology (UMM) [17], which examines multiple environmental matrices to estimated health statistics for a population or area of interest. Analysis of time- and flow-weighted samples of composited wastewater may provide unique wastewater-based epidemiological insights into consumption statistics, with flow-weighted estimates providing a more statistically favorable result, and theoretically could be obtained for a broad spectrum of chemical products consumed and excreted by a population [18]. Sampling for the purpose of wastewater epidemiological analyses generally focuses on the headworks of wastewater treatment plants (WWTP) [13,15,19] but the technique also has been applied to obtain equivalent information for smaller population sizes, such as college campuses [20-22] or prisons [14,23].

Wastewater-based epidemiology has seen limited application in the United States [24]. To the authors' knowledge four studies have applied WBE technology at sampling points local to U.S. college campuses to obtain drug use statistics. Two studies were primarily interested in quantifying non-medical attention deficit hyperactivity disorder (ADHD) prescription drug use [21,22], whereas two other ones screened for a wider suite of illicit and prescription drugs including: amphetamines, opioids, cocainics, cannabinoids, and lysergics [20,25]. None of these studies screened for the potent synthetic opioid fentanyl, despite its known association with still increasing overdose rates and fatalities from drug abuse [26]. These studies typically focus on a single university and thus are limited in their generalizability due to the social, economic, and circumstantial factors that cause variation in drug use across different locations [27,28]. Therefore the main objectives of the present study were to: (i) apply a liquid chromatography tandem mass spectrometry method for the detection of 12 drugs of abuse, including some of their known metabolites in a university setting; (ii) obtain the first data on fentanyl occurrence in university campus wastewater where the contributing population is predominantly college-aged adults; and (iii) assess and quantify potential consumption of targeted prescriptions and illicit narcotics within the campus population.

2. Materials and methods

2.1. Study location and methods of wastewater sampling

Seven consecutive 24-hour flow-weighted wastewater samples were collected using automated samplers for a one-week period per month from August 2017 to December 2017. Sampling location 1 accounted for approximately 95% of the total campus-borne wastewater while sampling location 2 accounted for approximately 5% of the total flow. The sewershed contributing population for both locations ranged between approximately 15,000 to 60,000 persons depending on the day of sampling. Mean age (26.5 years) of the catchment population was estimated by comparing average age of the population from years with available age data (2004-2009). Approximately 53.6% of students were male and 46.4% female, with 81.8% pursuing undergraduate degrees and 18.2% pursuing graduate degrees. Undergraduate ethnicity demographics were as follows: white: 50.5%; Hispanic/Latino: 21.7%; international: 10.6%; Asian: 6.6%; African American: 4.3%; American Indian: 1.3%. Graduate ethnicity demographics were as follows: white: 45.5%; Hispanic/Latino: 10.9%; international: 30.8%; Asian: 4.9%; African American: 3.1%; American Indian: 1.2%. Population demographics were obtained from available public record of the participating university. The campus features a sewer system designed to separate municipal wastewater from stormwater inputs. Ambient temperatures throughout the study period ranged from 3.4 to 42.8 °C. The average sewage travel distance within the study catchment was estimated to be 2700 m. Sewage retention time in the catchment system was estimated to average about 50 min but could range between 10-110 min depending on travel distance and sewer flow conditions (Table S1). Sampling occurred one week per month during the fivemonth study period through a joint effort between the study researchers and municipality personnel. Samples were stored in polyethylene terephthalate (PET) bottles for transport and storage and immediately processed through solid phase extraction upon receipt by laboratory personnel. Remaining samples and concentrated sample extracts were stored at -20 °C until analysis.

2.2. Target analytes

Ten parent prescriptions and illegal narcotics and nine (9) metabolites were monitored in raw wastewater collected from two sampling locations on university campus accounting for a majority of campusborne wastewater (see supporting information). The investigated drugs were morphine's major metabolite morphine-3-glucuronide (M3G), codeine (COD), its major metabolite norcodeine (NCOD), oxycodone (OXY), its major metabolite noroxycodone (NOXY), fentanyl (FENT), its major metabolite norfentanyl (NFENT), heroin (HER), its minor but exclusive metabolite 6-acetylmorphine (6-AM), methadone's major metabolite 2-ethylidene-1,5-dimethyl-3,3-diphenylpyrrolidine (EDDP), buprenorphine (BUP), its metabolite norbuprenorphine (NBUP), amphetamine (AMP), methylphenidate (MPH), alprazolam (ALP), its metabolite alpha-hydroxyalprazolam (OH-ALP), cocaine (COC), its metabolite benzoylecgonine (BZE), and 3,4-methylenedioxymethamphetamine (MDMA). High purity (> 97%) standards of the target compounds originated from Sigma Aldrich (Milwaukee, WI) and were prepared by Cerilliant (Round Rock, TX, USA) as solutions in methanol or acetonitrile. Eighteen deuterated compounds, one for each of the parent opioid target compounds were also purchased from Cerilliant for use as internal standards (IS) for quantification (section S-1.2).

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