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Estimating heroin abuse in major Chinese cities through wastewater-based epidemiology

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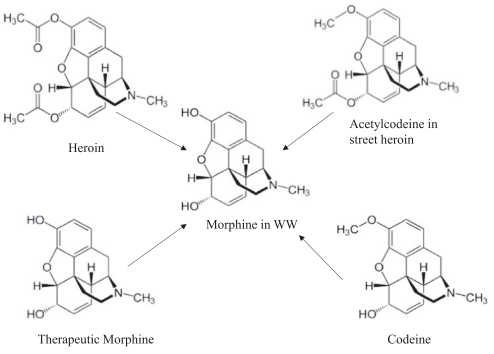
HIGHLIGHTS

- First large scale wastewater-based investigation on heroin abuse in major Chinese cities
- Morphine and codeine in wastewater were predominantly from street heroin.
- Codeine abuse was evident in cities in Guangdong province.
- Heroin consumption was estimated based on morphine loads in wastewater.
- Heroin consumption in Northwest and Southwest was much higher than in other regions.

GRAPHICAL ABSTRACT



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ABSTRACT

Heroin consumption in major cities across China was estimated for the first time via wastewater-based epidemiology. Influent and effluent wastewater samples were collected from 49 wastewater treatment plants (WWTPs) in 24 major cities that cover all the geographic regions of the country. Concentrations of morphine, 6-acetylmorphine, and codeine were measured. Near complete removal of morphine by wastewater treatment processes was observed, whereas removal rates of codeine were slightly lower. Morphine loads were much higher than codeine loads at most WWTPs in China, a trend opposite to that in many European countries. In addition, morphine and codeine loads were strongly correlated at most WWTPs, indicating morphine and codeine in wastewater were predominantly from the same source, street heroin. At WWTPs in Guangzhou and Shenzhen, codeine loads were considerably higher than morphine loads, consistent with previous reports of codeine abuse (e.g., as cough syrup) among middle and high school students in Guangdong province. Heroin consumption was derived based on morphine loads and taking into account therapeutic use of morphine and codeine, as well as contribution of codeine and acetylcodeine in street heroin. Highest heroin consumption was observed in northwestern and southwestern China. The average heroin consumption of the sampled cities was 64.6 ± 78.7 mg/1000 inh/d. The nation-wide average heroin consumption was much lower than that of methamphetamine, consistent with seizure data and numbers of registered heroin and methamphetamine users in China.

1. Introduction

Heroin had long been the primary drug of abuse in China since the reemergence of the drug problem in the country in early 1980s (Office of China National Narcotic Control Committee, 2016a). For

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example, registered heroin abusers (0.745 million) accounted for 82.7% of the total number of registered drug abusers (0.901 million) in China in 2000 (Office of China National Narcotic Control Committee, 2001). Although seizure of heroin was exceeded by that of methamphetamine about ten years ago (Office of China National Narcotic Control Committee, 2006), heroin abuse remains at high level (Office of China National Narcotic Control Committee, 2016b). Heroin seizure increased gradually from 5.79 t in 2007 to 9.3 t in 2014 (Office of China National Narcotic Control Committee, 2008, 2015b). However, exact heroin abuse, its spatial distribution, as well as its temporal variation were largely unknown in the country.

About a decade ago, Zuccato et al. (2005, 2008) estimated illicit drug abuse through wastewater-based epidemiology (WBE) for the first time. This methodology involves collecting wastewater samples (typically influents) from wastewater treatment plants (WWTPs) and measuring the concentrations of the drug target residues in the samples. The drug consumption of the communities served by the WWTPs is then back-calculated by taking in account flow rates of WWTPs, populations of the communities, as well as the excretion rates of the drugs (Zuccato et al., 2008). This approach uses objective measures to estimate drug use, generates results in near real time, and allows comparison of drug use between different communities and different time periods.

In the past decade, wastewater-based epidemiology has been applied in many countries, mainly in Europe (van Nuijs et al., 2009, 2011a; Subedi and Kannan, 2014; Jiang et al., 2015; Senta et al., 2015; Mackulak et al., 2015, 2016; Karolak et al., 2010; Repice et al., 2013; Andres-Costa et al., 2014; Bijlsma et al., 2014; Damien et al., 2014; de Castro et al., 2014; Guerra et al., 2014; Kankaanpaa et al., 2014; McCall et al., 2016). In particular, this approach has been used to estimate heroin use in Spain (Boleda et al., 2009; Postigo et al., 2010), Belgium (van Nuijs et al., 2011b), Italy (Zuccato et al., 2008, 2011, 2016), Switzerland (Zuccato et al., 2008; Been et al., 2015), UK (Zuccato et al., 2008), Canada (Yargeau et al., 2014), and Croatia (Terzic et al., 2010). In these studies, heroin consumption was back-calculated based on concentrations of either of its two metabolites, morphine or 6-acetylmorphine in wastewater. Morphine (in free and conjugate forms) is the major yet non-exclusive metabolite, whereas 6-acetylmorphine is the exclusive yet minor metabolite of heroin (Baselt, 2004; D'Ascenzo et al., 2003). Among the countries examined, heroin consumption ranged from a few tens to a few hundreds of milligrams per 1000 inhabitants per day.

Wastewater-based epidemiology has not been applied in mainland China until quite recently. Khan et al. (2014) applied the approach for the first time to four megacities (Beijing, Shanghai, Guangzhou, and Shenzhen). In total nine wastewater treatment plants were sampled in that study to examine the use of heroin, methamphetamine, ketamine, and other seven illicit drugs in the cities. Li et al. (2014) examined methamphetamine loads in wastewater of the urban districts of Beijing. Du et al. (2015) carried out a nation-wide reconnaissance of methamphetamine and ketamine abuse. More recently, Gao et al. (2017) used this approach to examine use of new psychoactive substances (synthetic cathinones and piperazines). However, large-scale survey of heroin use has not been performed in the country.

The objective of this work was to examine, for the first time, heroin use in major cities across China. Influent wastewater samples were collected from 49 WWTPs of 24 provincial capitals or equivalent cities that cover all the geographic regions of the country. Concentrations of morphine, 6-acetylmorphine, and codeine (a pharmaceutical that is metabolized into morphine and an impurity in street heroin) in the wastewater samples were analyzed. Heroin consumption was derived based on morphine loads in wastewater that were subtracted by contribution of therapeutic morphine and codeine as well as contribution of codeine from heroin. A clear geographic pattern of heroin use in China was then demonstrated.

2. Materials and method

2.1. Reagents and chemicals

Morphine (MOR), codeine (COD), 6-acetylmorphine (6-AM), and their deuterated analogs used as internal standards (MOR-d_3, COD-d_6, 6-AM-d_6) were purchased from Cerilliant (Round Rock, TX, USA). Formic acid and ammonium formate (HPLC grade) were purchased from CNW Technologies GmbH (Düsseldorf, Germany). HPLC grade of acetonitrile (AcN) and methanol (MeOH) was obtained from Fisher Scientific (Waltham, MA, USA). Hydrochloric acid (AR) and ammonium hydroxide (AR) were purchased from Beijing Chemical Works (Beijing, China). Ultrapure water was prepared using a Milli-Q ultrapure system (Millipore, MA, USA). Oasis MCX SPE cartridges (60 mg, 3 mL) were purchased from Waters Corporation (Milford, MA, USA).

2.2. Wastewater sampling

Wastewater samples were collected from 24 cities in China (Fig. 1, Table S1). Beijing (BJ) and Shanghai (SH) are municipalities directly administrated by the central government. Harbin (HRB), Xi'an (XA), Lanzhou (LZ), Yinchuan (YC), Taiyuan (TY), Shijiazhuang (SJZ), Wuhan (WH), Jinan (JN), Nanjing (NJ), Hangzhou (HZ), Nanning (NN), Guangzhou (GZ), Chengdu (CD), Kunming (KM) and Guiyang (GY) are provincial capitals. Dalian (DL), Luoyang (LY), Qingdao (QD), Suzhou (SuZ), Xichang (XC), Xiamen (XM), and Shenzhen (SZ) are equivalent to provincial capitals in terms of economy and population. The sampled cities are distributed in all the eight geographical regions of China: Northeast (HRB and DL); North (BJ, SJZ and TY); Northwest (LZ, XA and YC); Central China (LY and WH), East (JN, QD, NJ, SuZ and SH); Southeast (XM and HZ), Southwest (CD, XC, KM and GY), South (NN, GZ and SZ). The total population of sampled cities was 217.5 million, about 16% of the entire population of the nation.

Wastewater samples were collected from 49 WWTPs in the 24 cities. In most cities, two or more WWTPs were selected for wastewater sample collection. Only one WWTP was sampled in LZ and JN. Influent samples were collected in all 49 WWTPs, whereas effluent samples were collected in 19 selected WWTPs (Fig. 2) in 9 cities (Table S1). Most sampled WWTPs treat domestic wastewater from the urban districts of the cities. The population served by the WWTPs from which influent samples were collected totals 43.9 million, representing about 20% of the total population of the cities and 3.2% of the entire population of the country. The WWTPs are named as SZ-1 (first WWTP of Shenzhen), BJ-5 (fifth WWTP of Beijing), etc.

The sampling campaign was conducted during two stages. The wastewater in DL, TY, QD, SuZ, NN, GZ, SZ, XC, CD and HRB-2 were collected between May and September of 2015. Samples from other cities were collected from early July to early October of 2014. Sampling over a period of one year may involve seasonal changes, which may affect the spatial trend of heroin use. However, sampling across a country as vast as China was too huge an undertaking to be completed within a short sampling period. Each WWTP was sampled at least two days (one weekday and one weekend day) except LZ-1, KM-1 and HZ-2 (sampled for one day) (Table S1). Most of samples were collected by collecting 24-h time-proportional composite samples with several types of auto-samplers, such as FC-9624 (GRASP Science & Technology Co., LTD, Beijing), ISCO 3000, 3700, 4700, 6712 (Teledyne Technologies Inc., Lincoln, NE, USA), Sigma-SD900 (HACH Inc., USA). Time-proportional sampling is not an ideal approach to collect samples (Ort et al., 2010). However, flow- and volume-proportional samplers were not available at the WWTPs. In addition, time-proportional sampling was adopted in many regional and national scale WBE studies (e.g., Kankaanpaa et al., 2014; Mackulak et al., 2014; Du et al., 2015).

During collection, each auto-sampler was programmed to imbibe a specific volume (100 mL) of influent (or effluent) for each hour of sampling. A composite sample was obtained by mixing the samples