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Estimation of amphetamine and methamphetamine uses in Beijing through sewage-based analysis

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HIGHLIGHTS

- First sewage-based epidemiology of methamphetamine and amphetamine used in Beijing
- Amphetamine present at low concentrations and from metabolism of methamphetamine
- Globally medium-level methamphetamine loads observed in Beijing
- Methamphetamine loads were higher at the urban center.
- Methamphetamine loads were greater in summer than in early winter.

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ABSTRACT

Sewage epidemiology has been proven as an effective approach to estimate the use of illicit drugs by a population. In this study, sewage analysis was applied to examine the patterns of amphetamine (AMP) and methamphetamine (METH) uses in the urban area of Beijing. Influent and effluent samples were collected from all the thirteen sewage treatment plants (STPs) in the urban area during two sampling campaigns. METH concentrations in influents were found to range from several tens to several hundred ng · L^{-1}, whereas AMP concentrations ranged from several to several tens ng · L^{-1}. The concentration ratios between AMP and METH in influents at most STPs were close to the rate of AMP excretion following METH ingestion, indicating that AMP in sewage in Beijing was predominately from the metabolism of METH. Much higher METH loads were observed in the center part of the urban area in Beijing, indicating a strong correlation between METH use and economic level and entertainment activities. Seasonal variation in METH loads was significant, with greater use in summer than in winter. Significant difference in METH loads between weekdays and weekend days were observed in winter but not in summer. No clear trend in diurnal variation of METH use was observed. Nearly complete removal of METH occurred at the STPS in Beijing. Apparent removal rates of AMP were lower than those of METH, likely due to degradation of METH into AMP during the wastewater treatment processes. In summary, this study represents the first application of sewage epidemiology to the entire urban population of a metropolitan in mainland China and provided an overview of METH and AMP uses in the city.

1. Introduction

A growing body of literature has reported that rapid increase in overall illicit drug consumption and extensive geographic expansion of drug use had occurred in the past quarter century in China (Cui, 2011; Liu et al., 2006; Lu et al., 2008; Qin, 2007; Wang et al., 2007; Xu et al., 2009; Yang, 2010). The drug use in China displays several distinct features: (1) the number of drug users has increased dramatically in recent years—the number of registered drug users increased from 1.2 million in 2007 to 1.81 million at the end of 2011 (the actual number is conjectured to be much higher) (Devaney et al., 2006; Yong-an, 2012); (2) several emerging synthetic illicit drugs such as amphetamine-like stimulants and ketamine are increasingly prevalent (Cui, 2011; Ha et al., 2007; Liu and Guan, 2006; Lv, 2011; Qin, 2007, 2011; Yang, 2010; Zhao and Wang, 2007), whereas the use of traditional illicit drugs (e.g., heroin) remains at high levels (Security NcBotmoP, 2006, 2011); (3) young drug users (between 15 and 35 years old) take an increasing percentage of the overall number of drug users (Han, 2007; Security NcBotmoP, 2011); (4) illicit drugs are commonly consumed at

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entertainment venues such as pubs, bars and nightclubs (Li and Nie, 2009; Lin, 2010; Liu, 2011; Shi, 2009; Zhao, 2011).

To better control the drug use in China, it is critical to precisely estimate the drug consumption, the pattern of drug use, and the number of drug users in the country. Currently, drug use monitoring in China relies on the passive registration (for the number of drug users), census system (mainly for the drug use pattern), and reports on drug seizures. These approaches yield estimates that are far from accurate. According to Chinese legislations on illicit drugs, the users would be arrested by law enforcement once they were found using drugs. Once arrested, they are not treated as common patients. Instead, they are detested and typically abandoned by the society. Thus, the users would do their best to conceal their use of drugs, resulting in a huge discrepancy between the registered and actual number of drug users. Drug census is typically performed within relatively small areas in China (e.g., in addiction treatment centers) and among specific populations (e.g., AIDs patients). As such, it is unlikely that the census results can reflect country-level patterns. Drug seizures are largely subject to randomness, providing limited information that can reflect the overall drug use trend at best.

In recent years, sewage epidemiology has emerged as a more reliable method to estimate drug consumption by a particular population. The approach starts with collecting wastewater samples (typically influents) from STPs and measuring the concentrations of the residues of drugs or its metabolites in the samples. The drug loads of the communities served by the sampled STPs, are then back calculated by taking account of the flow rates of STPs, populations of the communities, as well as the excretion and metabolism rates of the drugs (Khan and Nicell, 2012).

This approach, while still being improved, can provide more consistent and logical estimates on drug consumptions and use patterns as wastewater analysis uses objective measures that avoids possible bias associated with self-reporting (Zuccato and Castiglioni, 2011). In addition, it can generate results in near real time and allows comparison of drug uses between different communities and at different time periods. Sewage epidemiology has been extensively applied in North America (Bisceglia et al., 2010; Chiaia-Hernandez et al., 2011; Chiaia et al., 2008; Metcalfe et al., 2010; Postigo et al., 2008), Europe (Kasprzyk-Hordern and Baker, 2012; Postigo et al., 2008; van Nuijs et al., 2009, 2011; Zuccato et al., 2008, 2011), Australia (Irvine et al., 2011; Lai et al., 2011, 2013a), Hong Kong (Lai et al., 2013b) and Taiwan (Lin et al., 2010). Recently, Khan et al. used the sewage epidemiology approach, for the first time, to examine the use of ten illicit drugs in four megacities in China (Khan et al., 2014). Their results revealed that drug use patterns in China were very different from those observed in Europe and North America (Berset et al., 2010; Bisceglia et al., 2010; Bones et al., 2007; Huerta-Fontela et al., 2008b; Karolak et al., 2010; Metcalfe et al., 2010; Postigo et al., 2010; Zuccato and Castiglioni, 2009; Zuccato et al., 2008). Cocaine and ecstasy, the most popular drugs in Europe, were detected at very low concentrations, while AMP, METH and ketamine are detected most frequently and at relatively high concentrations. Particularly, highest AMP and METH concentrations were found in influents of STPs in Beijing, whereas other drugs (including MDMA, MDA, ketamine) were rarely detected in sewage of the same city. The overall nationwide use pattern revealed in their study is qualitatively consistent with UNODC reports (UNODC, 2012, 2013a,b). While Khan et al.'s work demonstrated overall geographic pattern of drug use in China, the study only collected sewage samples from a small number (<4) of STPs in only one sampling campaign in each city. Thus their results did not provide comprehensive information on drug use in a particular city (e.g., diurnal, daily, and seasonal pattern of drug use).

The overall objective of this study was to conduct a systematic sewage-based analysis to assess AMP and METH use in Beijing to estimate spatial and temporal use patterns. All thirteen STPs in the urban area were sampled, each during two campaigns, to examine spatial, seasonal and daily variations of concentrations, as well as removal efficiencies of the two drugs. Diurnal fluctuations were also investigated at selected STPs by collecting 24 × 1 h composite samples. Loads of METH were back calculated based on the concentrations, flow rates, and population data and compared to loads of different countries. This study provided comprehensive information on AMP and METH use in the Beijing urban area that is valuable for a more accurate monitoring of the drugs.

2. Materials and methods

2.1. Reagents and materials

AMP and METH as well as their deuterated analogs (AMP-d_8 and METH-d_8) used as internal standards (purity > 98%) were purchased from Cerilliant (Round Rock, TX, USA). Dilutions and working mixtures with concentrations ranging from 0.02 to 100 ng · μL^{-1} were further prepared in acetonitrile (AcN) (HPLC grade, Fisher Science). Ammonium hydroxide (NH_4OH) (AR) was purchased from Sinapharm Chemical Regent Co. Ltd. (Beijing, China). Ammonium acetate (HPLC grade) was obtained from CNW Technologies GmbH (Düsseldorf, Germany). Deionized water was prepared by a Milli-Q system (Millipore, MA, USA). Oasis MCX (60 mg, 3 mL) SPE cartridges were purchased from Waters Corporation (Milford, MA, USA). CNW™ SPE vacuum manifold (CNW Technologies GmbH) with twelve connections and a self-cleaning dry vacuum system were used for loading wastewater samples and for drying the cartridges.

2.2. Sampling

In total, thirteen domestic sewage treatment plants (STPs) (Gabobeidian (GBD), Qinghe (QH), Xiaohongmen (XHM), Beixiaohe (BXH), Jiuxianhe (JXH), Fangzhuang (FZ), Wujiacun (WJC), Lugouqiao (LGQ), Tongzhou Bishui (TZ), Xiaojiahe (XJH), Beiyuan (BY), Yongfeng (YF) and Wenquan (WQ)) located in the urban area of Beijing were sampled (see Table S1 for details of the sampled STPs and Fig. 1 for the locations of STPs). All these thirteen STPs are secondary types. Sampling was conducted for two periods—summer season (between 30th of June and 6th of August in 2013) and early winter season (between 14th of Nov. and 24th of Nov. in 2013) (see Table S1 for the specific dates on which each STP was sampled). Each STP was sampled for four days (one weekend day and three weekdays) by collecting 4 consecutive 24-h composite samples at both sewage inlets and effluent exits in the first period and for two days (one weekend day and one weekday) at inlets in the second period. Two models of autosamplers, FC-9624 (GRASP Science & Technology Co., Ltd., Beijing) and ISCO-6712 (Teledyne Isco, NE, USA), were used for sampling purposes. Each auto-sampler was programmed to imbibe 100 mL of influent for each hour of sampling. After collection for 24 h, samples collected every hour were mixed on spot to constitute 24-h composite samples. Diurnal variations of AMP and METH concentrations in sewage were examined at four selected STPs (GBD, XJH, LGQ and FZ) by collecting separate hourly samples. The sampling dates for GBD, XJH, LGQ and FZ STPs were from June 30 (Sun.) to July 1 (Mon.), from Aug. 5th (Sun.) to Aug. 6th (Mon.), from June 29 (Sat.) to June 30 (Sun.) and from Nov. 24 (Sun.) to Nov. 25 (Mon.), respectively. Samples were collected at an interval of 1 h from 8:00 AM of the first day to 7:00 AM of the next day at a particular plant. All samples were adjusted to pH 2 by using hydrochlorine acid once the sample was collected. Following collection, samples were stored at −20 °C until analysis.

2.3. Analysis

Samples were pretreated using solid-phase extraction (SPE) and analyzed by liquid-chromatography coupled to tandem mass spectrometry (LC–MS/MS). Sample pretreatment followed the procedures developed by van Nuijs et al. (2009, 2013), with minor modifications.