Science of the Total Environment 524–525 (2015) 440–446

Contents lists available at ScienceDirect

Science of the Total Environment

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

The first application of wastewater-based drug epidemiology in five South Korean cities

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HIGHLIGHTS

- This is the first study on illicit drug use in Korea through sewage analysis.
- Only four compounds among 17 drugs and metabolites were detected.
- Methamphetamine was widely used but its estimated consumption rate was low in Korea.
- The characteristic spatial distribution of methamphetamine use was observed.

ARTICLE INFO

Article history:
Received 6 January 2015
Received in revised form 30 March 2015
Accepted 13 April 2015
Available online 29 April 2015

Editor: Adrian Covaci

Keywords:
Wastewater analysis
Sewage epidemiology
South Korea
Illicit drug
Methamphetamine
Consumption rate

ABSTRACT

Illicit drug consumption in five cities in South Korea was estimated by analyzing 17 drug residues in untreated wastewater samples collected during the Christmas and New Year period of 2012-13. Only methamphetamine, amphetamine, and codeine were detected at concentrations of tens of nanograms per liter or even lower concentrations in more than 90% of the samples. Other illicit drug residues (including cocaine, methadone, and benzoylecgonine) that have been detected frequently in wastewater from other countries were not found in this study. Methamphetamine was found to be the most widely used illicit drug in South Korea, and the estimated average consumption rate was 22 mg d^{-1} (1000 people)$^{-1}$. This rate is, for example, 2-5 times lower than the estimated average consumption rates in Hong Kong and other parts of China and 4-80 times lower than the estimated average consumption rates in cities in Western countries. It should be noted that the wastewater samples analyzed in this study were collected during a holiday season, when daily consumption of illicit drugs is often higher than on an average day. The methamphetamine usage rates were calculated for different cities in South Korea, and the usage rates in smaller cities was higher (2-4 times) than the average.

1. Introduction

Trafficking and abuse of illicit drugs are considered to be social problems around the world (UNODC, 2012). Therefore, understanding the extent of illicit drug consumption is essential for law enforcement to devise appropriate strategies aimed at controlling the use of these drugs. Currently, many studies have used wastewater-based drug epidemiology (WBDE), i.e. analysis of drugs and their metabolites or drug biomarker in wastewater samples from the target community, as an alternative approach to socio-epidemiological survey to estimate drug use in a community (Zuccato et al., 2008). Most of these studies have been conducted in European countries (Ort et al., 2014; Thomas et al., 2012) with others in Australia and the United States (e.g. Lai et al., 2011; Burgard et al., 2013). To date, only four studies have used WBDE to measure illicit drug consumption in Asian cities (China, Hong Kong, and Taiwan) (Khan et al., 2014; Lai et al., 2013b; Lin et al., 2010; Li et al., 2014). These studies have indicated that the use of methamphetamine (MA) has been relatively high whereas that of cocaine (COC) and cannabis has been low in Asia. Furthermore, the high use of ketamine has been found in Hong Kong and Taiwan. The estimated high use of MA in this region is consistent with the fact that Southeast and East Asia are the main sources of MA production and trafficking (Dargan and Wood, 2012; McKetin et al., 2008). It has been reported that 63% of the annually seized MA worldwide is produced in the Asia-Pacific region (McKetin et al., 2008).

The Republic of Korea (South Korea) has been called 'the country free from the needle' because of the low levels of narcotics abuse (INCB, 2003). This may lead the Korean society to pay less attention to illicit drug use and drug-related social problems than other countries. There is a paucity in data related to illicit drug use in South Korea and the limited available information on the use of illicit drugs in South

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http://dx.doi.org/10.1016/j.scitotenv.2015.04.065
0048-9697/

Korea come from the estimates made through socio-epidemiological surveys and from indirect drug usage data from the conviction rate for drug-related crimes (Dargan and Wood, 2012; SPO, 2012). These data sources suggest that more than 80% of the criminal activities related to illicit drugs are associated with methamphetamine and other amphetamine type substances (ATSs) (SPO, 2012). However, this information was presented with the common shortfall of social survey data, (i.e. no spatial distribution, aggregation of data collected over the year). It is difficult to assess drug usage patterns specific to local populations as demonstrated in other studies (e.g. Lai et al., 2013a) and, therefore, to develop appropriate strategies for such communities.

The WBDE approach may be used to overcome these disadvantages as it can provide objective estimation of the overall consumption of certain illicit drugs in the population serviced by a wastewater treatment plant. Therefore, in this study we aimed to use WBDE for the first time to estimate illicit drug consumption in different communities in the south of the South Korea and assess the spatial variability in drug use in this region. To the best of our knowledge, no data on illicit drug residues in wastewater from South Korea have previously been published.

2. Materials and methods

2.1. Sample collection

Raw influent wastewater samples were collected at the inlet (before the primary clarifier) of 15 wastewater treatment plants (WWTPs) located in five different cities in southeastern South Korea (Fig. S1). Ten of the WWTPs are in Busan, the second largest metropolitan city in South Korea (with 3.7 million inhabitants), two WWTPs are in Ulsan, the largest industrial city in South Korea, and three WWTPs are in small cities in the region (Changwon, Kimhae, and Milyang). Detailed information on the WWTPs that the samples were collected from is presented in the supplementary information (Table S1).

Sample was collected time proportionally using an auto-sampler which was set to imbibe 150 mL of wastewater per hour. Sampling was taken from 7 am to 7 am the next day and combined to make a 24-hour composite sample from each STP during the Christmas and New Year period in 2012–13 (26 December 2012 to 1 January 2013). The sampling method was kept consistent across all the studied sites to ensure the comparability of the data. However, it should be noted that the one-day sampling per STP with relatively low frequency sampling of the auto-sampler may result in large uncertainty for samples of small STPs.

After collection, the composite sample was mixed well and an aliquot of 500 mL was transferred into a polyethylene bottle pre-rinsed with methanol (MeOH) and was acidified to pH 2 using 2 M hydrochloric acid then stored at −20 °C. All collected samples were then shipped by air with dry ice to the University of Queensland, where the samples were received frozen and were stored at −20 °C until analysis.

2.2. Materials and chemical analysis

The analytical standards (COC, COC-d3, benzoylecgonine (BE), BE-d3, amphetamine (AMP), AMP-d6, MA, MA-d9, 3,4-methylenedioxymethamphetamine (MDMA), MDMA-d5, 3,4-methylenedioxyethamphetamine (MDEA), MDEA-d5, 3,4-methylenedioxyamphetamine (MDA), MDA-d5, codeine (COD), COD-d3, morphine, morphine-d3, Δ9-tetrahydrocannabinol (THC), THC-d3, 11-nor-9-carboxy-Δ9-tetrahydrocannabinol (THC-COOH), THC-COOH-d3, methadone, methadone-d3, 2-ethylidene-1,5-dimethyl-3,3-diphenylpyrrolidine (EDDP), EDDP-d3, ketamine, nor-ketamine, mephedrone, and methylone) were purchased from Cerilliant (Round Rock, TX, USA). Seventeen illicit drugs and metabolites in the samples were extracted using solid-phase extraction (SPE) as reported previously (Lai et al., 2011). Briefly, the sample (200 mL) was filtered (0.45 μm pore), and spiked with the deuterated standards (between 10 and 100 ng of each standard), and then loaded onto an Oasis MCX cartridge (Waters, Milford, MA, USA) which has been pre-conditioned with MeOH and Milli-Q water. After drying, the cartridges were eluted into two fractions with MeOH and 2% NH_4OH in MeOH, respectively. Both fractions were concentrated using a gentle stream of high purity nitrogen and the residues were reconstituted with MeOH and an aqueous solution of 5% acetonitrile and 0.1% formic acid, respectively. The target drug residues in the samples were analyzed using liquid-chromatography (Shimadzu Prominence; Shimadzu, Kyoto, Japan) coupled to tandem mass spectrometry (ABSciex, QTRAP®5500, Ontario, Canada) (see Lai et al., 2011, 2013b for the detailed analytical conditions). While the analytical conditions of this study remained the same as that of our previous study (Lai et al., 2011), another mass spectrometry model has been used in this study to analyze the samples with the limits of detection (0.03–7.5 ng/L) and quantification (0.10–25 ng/L) as shown in Table S2. Quality assurance and control measures were taken during the analyses, and these included analyzing duplicate samples and Mili-Q and wastewater matrix samples spiked with the native chemicals (10–100 ng). The matrix spike recovery of the target drug residues was determined by subtracting the analyte concentrations measured in the spiked samples from that measured in the respective unspiked samples and then compared to the expected spiked amounts. The recoveries of the illicit drug residues were 96–116% in Milli-Q water and 94–125% in wastewater samples (Table S2). Blank samples (Milli-Q and wastewater) were included to quantify any contamination of the samples that occurred during the analytical procedure. None of the target drug residues were quantified in the blank samples.

2.3. Back estimation of drug consumption

We used a common equation proposed by Zuccato et al. (2008) to estimate the consumption of illicit drugs in our target communities as shown in Eq. (1) below:

$$\text{Daily drug consumption rate}\left(\frac{\frac{\text{mg}}{\text{day}}}{1000\ \text{people}}\right) = \frac{C_i \cdot F \cdot \frac{R_i}{E_i}}{p} \quad (1)$$

where C_i is the concentration of drug residue i (parent drug or metabolite) in the raw wastewater sample (ng/L), F is the total daily flow (L/d) during the sampling period. P is the number of people in the WWTP catchment area, R_i is the ratio of the molar mass of the parent drug to the molar mass of the residue, and E_i is the average excretion rate for drug residue i. In this study, the population serviced by each WWTP (P) and total daily flow (F) was provided by the WWTP personnel.

3. Results and discussion

3.1. Daily illicit drug loads

Out of the 17 targeted drugs and metabolites analyzed, only three compounds (AMP, MA, and COD) were detected in most of the wastewater samples (Table 1). MDA was only detected in only one sample from WWTP (K-3).

MA was found in all of the samples, with the daily loads ranged from <1.12 to 29.2 mg d^{-1} (1000 people)$^{-1}$. The daily loads of AMP were lower than those of MA ranging from below the limit of detection (N.D.) to 7.18 mg d^{-1} (1000 people)$^{-1}$. The daily load of MDA from the only detected occasion was 5.55 mg d^{-1} (1000 people)$^{-1}$.

AMP and MA were more frequently detected than any of the other drugs and metabolites in the wastewater samples that were analyzed. AMP and MA were also the main drugs that were found in wastewater samples from Hong Kong and other parts of China (Khan et al., 2014; Lai et al., 2013b). The results of our study agreed with other