Forensic Science International 210 (2011) 69–73

Contents lists available at ScienceDirect

Forensic Science International

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

Population drug use in Australia: A wastewater analysis

Rodney J. Irvine [a], Chris Kostakis [b], Peter D. Felgate [b], Emily J. Jaehne [a], Chang Chen [a,*], Jason M. White [c]

[a] Discipline of Pharmacology, School of Medical Sciences, University of Adelaide, Adelaide, SA 5005, Australia
[b] Forensic Science South Australia, 21 Divett Place, Adelaide, SA 5000, Australia
[c] School of Pharmacy and Medical Sciences, University of South Australia, Adelaide, SA 5005, Australia

ARTICLE INFO

Article history:
Received 21 November 2010
Received in revised form 15 January 2011
Accepted 30 January 2011
Available online 9 March 2011

Keywords:
Geographical drug monitoring
Wastewater analysis
Cocaine
Benzoylecgonine
Methamphetamine
MDMA

ABSTRACT

Accurate information on drug use in communities is essential if health, social and economic harms associated with illicit drug use are to be addressed efficiently. In most countries population drug use is estimated indirectly via surveys, medical presentations and police and custom seizures. All of these methods have at least some problems due to bias, small samples and/or long time delays between collecting the information and analysing the results. Recently the direct quantification of drug residues in wastewater has shown promise as a means of monitoring drug use in defined geographical areas. In this study we measured 3,4-methylenedioxymethamphetamine (MDMA), methamphetamine and benzoylecgonine in sewage inflows in metropolitan and regional areas of Australia and compared these data with published European data. Cocaine use was small compared to European cities ($p < 0.001$) but was compensated for by much greater consumption of methamphetamine ($p < 0.001$) and MDMA ($p < 0.05$). MDMA was more popular in regional areas ($p < 0.05$) whereas methamphetamine and cocaine were mainly consumed in the city ($p < 0.05$). Greater than 5-fold increases in MDMA use were detected on weekends ($p < 0.001$). This approach has the potential to improve our understanding of drug use in populations and should be further developed to improve prevention and treatment programs.

1. Introduction

Accurate information on drug use in communities is essential if health, social and economic harms associated with illicit drug use are to be addressed efficiently. In most countries population drug use is estimated indirectly via surveys, medical presentations and police and customs seizures. All of these methods have at least some problems due to bias, small samples and/or long time delays between collecting the information and analysing the results. Surveys are also very costly, limiting their use, and are unable to provide sufficient resolution in small regional population areas.

There are several important consequences of these limitations in current methods. One such consequence is that direct measurement of changes in drug use as a result of a public health campaigns is difficult. A relatively inexpensive method that could provide near real-time measures would be needed for such evaluation. A second consequence is that international comparisons between countries based on self-reported drug use (e.g. World Drug Report 2009) are limited. Differences in questions, survey methods, etc. limit comparability. International comparisons have important ramifications for the implementation and development of global strategies to combat illicit drug use, and a more accurate method of comparison would be of value.

Recently, the measurement of illicit drugs in wastewater as a means of direct and quick assessment of drug use in a community has been explored in a number of countries [1]. The advantages of developing this technology to improve information on illicit drug use have been recognised [2,3]. The first reported study was conducted in Italy and sampled from the River Po as well as four wastewater treatment plants servicing medium-sized Italian cities [4]. Data showed that benzoylecgonine, the major human metabolite of cocaine, was present in the samples. Subsequent studies by this group and others have extended these findings to a number of different geographical locations in Europe and North America [1,5]. The markers for a number of additional illicit drugs including methamphetamine, heroin and cannabis have also now been assessed [6–8]. Calculations of *per* capita drug consumption have then been made based on populations served by the wastewater treatment plant, daily volumes of wastewater produced in the areas, and excretion rate of each drug. Although there are a number of technical issues outstanding for drugs with unstable metabolites, it is clear that this approach provides an identifiable method to objectively quantitate illicit drug use on a continual basis.

Traditionally, information on population drug use in Australia is mainly obtained from a project named National Drug Strategy

* Corresponding author. Tel.: +61 8 8303 5188; fax: +61 8 8224 0685.
E-mail address: chang.chen@adelaide.edu.au (C. Chen).

0379-0738/$ – see front matter © 2011 Elsevier Ireland Ltd. All rights reserved.
doi:10.1016/j.forsciint.2011.01.037

Household Survey (NDSHS), which is carried out by the government once every 3 years [9]. The reports derived from the survey are very informative, but limited data on weekly fluctuation and geographic difference of the drug use is provided, also up-to-date information is unavailable. It is also reported that the use pattern of illicit stimulants in Oceania differs from Europe, with methamphetamine and 3,4-methylenedioxymethamphetamine (MDMA) more popular in Australia and cocaine users equally distributed in these two continents [10]. However, these differences are based on survey, seizure and anecdotal evidence, and we hypothesised that wastewater analysis data of Australia and Europe would provide a more objective comparison.

Hence, in this study we applied this novel approach to wastewater samples collected in the State of South Australia from a number of metropolitan and regional wastewater treatment plants on midweek and weekend days, confined our analysis to the stimulant drugs (methamphetamine, MDMA and cocaine), and then compared our results with previously published data from Europe.

2. Materials and methods

2.1. Sample collection

From April 2009 to October 2009, 1.2-L samples were taken from sewage inlet pipes of metropolitan and regional wastewater treatment plants immediately after sewage has passed through screens during which large solids were removed. The metropolitan samples were obtained from three independent plants servicing the Adelaide greater metropolitan area using auto-samplers which collected 24-h composite samples flow-dependently. Regional samples were grabbed from 10 regional plants throughout the State of South Australia and ranged in the populations they serviced from 370 to 23,300 (Table 1). No more than one sample was collected from one plant in 1 day. The samples were stored frozen until analysis.

2.2. Drug analysis

Samples were thawed to room temperature and mixed by inverting several times, and then filtered under vacuum using glass microfiber filters GF/A 1.6 μm (Whatman, Kent, U.K.). 200 μL of deuterated internal standards of MDMA, methamphetamine and benzoylecgonine were added to 300 mL of duplicate samples to give resultant concentrations of 33.3, 33.3 and 166.7 ng/L, respectively. Acetic acid (2.5%) was added to lower the pH of the samples to 4.5–5. The acidified samples were loaded onto pre-conditioned mixed-mode solid phase extraction (SPE) cartridges (UCT™ XRDAH; 500 mg/6 mL). Cartridges were successively washed with 6 mL of pH 5.7 acetate buffer, 2 mL of 0.1 M acetic acid and 6 mL of methanol. Analytes were eluted with a mixture of 96% dichloromethane:i-propanol (80:20)/4% ammonia and evaporated to dryness. The dry residue was reconstituted with 20 μL of methanol and then mixed with 180 μL of 0.1% formic acid. A set of diluted extracts was prepared by transferring 40 μL of the original extract to new vials and diluting each with 160 μL of 0.1% formic acid. Both sets were analysed by liquid chromatography/tandem mass spectrometry (LC/MS/MS).

Chromatographic separation was carried out using an Agilent 1200 series liquid chromatograph with a Phenomenex™ Luna PFP column (3 μm, 50 mm × 4.6 mm) connected to a PFP guard column (5 μm, 4 mm × 2.0 mm). The mobile phase consists of methanol (solvent A) and 0.1% formic acid (solvent B) with a flow rate of 0.5 mL/min. The gradient started with 95% B for 1 min. Then it was brought down to 5% B in the next 14 min and kept there for 1 min. Finally, the gradient was brought back to 95% B in 0.1 min and kept there for 2 min. Sample injection volume was 10 μL. Mass spectra were obtained using a 4000 Q-Trap™ (Applied Biosystems, Toronto, Canada) system equipped with an electrospray ionisation source. Mass spectrometric analysis was performed in positive mode via multiple-reaction monitoring (MRM). The optimum MS/MS parameters for the detection of our analytes were as follows: nitrogen was used as the nebulizer and auxiliary gas, the ion spray voltage (IS) was maintained at 4.0 kV and the source temperature (TEM) was 650 °C, the curtain gas (CUR), gas 1 (GS1), gas 2 (GS2) and collision gas (CAS) were set at 30, 70, 70 and 'medium', respectively. Three transitions were used for each analyte and the most responsive one was used for quantitation. The most responsive transition of each internal standard was also monitored for quantitation. Settings for compound-dependent parameters are summarised in Table 2.

Table 1
Concentration of MDMA, methamphetamine and benzoylecgonine in wastewater samples collected from metropolitan and regional treatment plants in South Australia.

Plant type	Plant name	Average flow rate (kL/d)	Population served	Sample type	Sampling day of week	No of samples	Concentration of residues (ng/L) (mean ± SEM or single value)		
							MDMA	Methamphetamine	Benzoylecgonine
Metropolitan	Bolivar	141243	820000	Composite	Sun	2[*,Δ]	208 ± 124	346 ± 75	55 ± 8
					Mon	2[*,Δ]	265 ± 138	4108 ± 1118	52 ± 8
					Tue	3[*,Δ]	39 ± 20	216 ± 35	27 ± 8
					Wed	4[*,#,Δ]	26 ± 8	224 ± 17	30 ± 7
					Thu	1[*,Δ]	63	287	31
					Fri	1[Δ]	72	338	40
					Sat	2[*,Δ]	85 ± 51	311 ± 103	50 ± 14
	Beach	27520	150000		Sun	2[*,Δ]	706 ± 264	640 ± 116	74 ± 1
					Mon	1[*,Δ]	321	445	35
					Tue	3[*,Δ]	76 ± 26	388 ± 116	31 ± 14
					Wed	4[*,#,Δ]	62 ± 21	367 ± 45	17 ± 8
					Fri	1[Δ]	192	527	30
					Sat	2[*,Δ]	330 ± 81	553 ± 12	58 ± 1
	Glenelg	52550	200000		Sun	1[*,Δ]	326	357	118
					Mon	4[*,Δ]	209 ± 83	368 ± 67	64 ± 13
					Tue	3[*,Δ]	66 ± 12	277 ± 18	38 ± 4
					Wed	3[*,#,Δ]	51 ± 23	368 ± 58	47 ± 5
					Thu	1[*,Δ]	114	524	67
					Fri	1[Δ]	136	505	69
					Sat	1[*,Δ]	392	586	103
Regional	Angaston	364	1900	Grab	Tue	2[#]	20 ± 19	69 ± 30	ND
	Finger Point	5226	23300		Tue	2[#]	183 ± 16	160 ± 31	ND
	Mt. Burr	121	370		Tue	2[#]	5 ± 3	12 ± 0	12 ± 0
	Nangwarry	134	480		Tue	1[#]	ND	305	ND
	Naracoote	900	4780		Tue	1[#]	ND	2	ND
	Pt. Augusta East	1323	5000		Tue	1[#]	167	218	ND
	Pt. Augusta West	627	3500		Tue	1[#]	200	507	ND
	Pt. Lincoln	2842	12660		Tue	4[#]	268 ± 50	105 ± 36	14 ± 4
	Pt. Pirie	3414	13260		Tue	2[#]	31 ± 20	85 ± 47	ND
	Whyalla	4139	21270		Tue	7[#]	134 ± 27	282 ± 77	ND

ND: not detected.
[Δ] Samples used for international comparisons.
[*] Samples used for comparisons of midweek days and weekend days.
[#] Samples used for comparisons of metropolitan and regional areas.