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Estimating daily and diurnal variations of illicit drug use in Hong Kong: A pilot study of using wastewater analysis in an Asian metropolitan city

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ABSTRACT

The measurement of illicit drug metabolites in raw wastewater is increasingly being adopted as an approach to objectively monitor population-level drug use, and is an effective complement to traditional epidemiological methods. As such, it has been widely applied in western countries. In this study, we utilised this approach to assess drug use patterns over nine days during April 2011 in Hong Kong. Raw wastewater samples were collected from the largest wastewater treatment plant serving a community of approximately 3.5 million people and analysed for excreted drug residues including cocaine, ketamine, methamphetamine, 3,4-methylenedioxymethamphetamine (MDMA) and key metabolites using liquid chromatography coupled with tandem mass spectrometry. The overall drug use pattern determined by wastewater analysis was consistent with that have seen amongst people coming into contact with services in relation to substance use; among our target drugs, ketamine (estimated consumption: 1400–1600 mg/day/1000 people) was the predominant drug followed by methamphetamine (180–200 mg/day/1000 people), cocaine (160–180 mg/day/1000 people) and MDMA (not detected). The levels of these drugs were relatively steady throughout the monitoring period. Analysing samples at higher temporal resolution provided data on diurnal variations of drug residue loads. Elevated ratios of cocaine to benzoylecgonine were identified unexpectedly in three samples during the evening and night, providing evidence for potential dumping events of cocaine. This study provides the first application of wastewater analysis to quantitatively evaluate daily drug use in an Asian metropolitan community. Our data reinforces the benefit of wastewater monitoring to health and law enforcement authorities for strategic planning and evaluation of drug intervention strategies.

1. Introduction

Illicit drug consumption is among the top 20 contributors to the global burden of disease and injury [1] and has a substantial negative economic impact [2]. As such, systematic surveillance of the extent of substance use and changes over time is important, particularly, to plan and to determine the success of law enforcement and health intervention strategies [3].

Hong Kong is one of the most densely populated cities in the world and its role as one of the key international financial centres draws a large number of international visitors each year. With such dynamic flow in people of different nationalities and high efficiencies in finance and transportation exchanges, Hong Kong is attractive for drug trafficking organisations [4]; for example, Hong Kong is found a key embarkation point for drugs to other Asian cities from China where illegal drug manufacturing appears often active [5].

The Narcotics Division of the Security Bureau in Hong Kong reports trends in substance use through its "Central Registry of Drug Abuse (CRDA)" reports. This report compiles data from law

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enforcement agencies (all arrests for substance use), drug rehabilitation and treatment centres, welfare and social work services (where substance use is suspected in clients) and hospitals (where withdrawal syndromes are present or individuals self-identify); and demographic and substance use information is collected [6]. The figures recorded in the CRDA are based on those drug consumers who have been identified with the agencies reporting in the system. While this dataset is the primary source for understanding drug use trends in Hong Kong, it is highly likely that many consumers may not be identified through this system. For example, for a population of seven million, just 12,400 consumers were identified for the 2010 CRDA report: <0.2% of the total population, which is extremely low by global standards (3.4–6.6% of adults) [3]. It is likely that infrequent consumers will not come into contact with the reporting agencies. The majority of consumers identified in CRDA were unemployed and had low education levels, suggesting that the consumers in other demographics are not well captured by the system. To obtain more comprehensive information about substance use, multiple methods can help overcome the limitations of individual datasets [7].

An alternative method to estimate drug use is the quantification of drug metabolite residues in raw wastewater sampled at inlets of wastewater treatment plants. The feasibility of this approach – in this paper subsequently referred as *wastewater analysis* – to back-estimate drug consumption has been widely demonstrated [e.g. 8–10]. The basic concept of the approach is that excreted drug residues are collectively delivered from toilet systems to wastewater treatment plants in a catchment. Thus, a raw wastewater sample represents a pool of the excreted drug residues within a population and allows tracing back per capita consumption rates in the catchment. Daily composite samples are commonly collected for understanding day-to-day changes in population's drug use; higher consumption is typically identified in weekends than weekdays [e.g. 11,12–16]. Analysing shorter time periods allows evaluating intra-daily variations in drug use [17,18]. Such diurnal monitoring to date is less common in the literature.

Despite the fact that this approach cannot reveal patterns of individual drug use such as dose or the presence of poly-drug use, the final estimates from wastewater analysis are objective and maintain the anonymity of individual consumers. Hence, it produces less ethical issues compared to traditional epidemiological methods such as self-reporting surveys [19]. Another benefit of wastewater analysis is that it provides information about the use of chemically specified substances, which is particularly relevant to tablets sold as 'ecstasy', which may vary substantially in purity and content over time without the knowledge of the consumers [20,21]. As such, wastewater analysis has been widely applied across different cities in western countries such as Australia, Canada, Europe and North America [e.g. 18,22–28] but to date has not been conducted in any Asian communities.

In this study, we applied wastewater analysis to estimate the extent of use of ketamine, cocaine, methamphetamine and MDMA over nine days in the major urban community of Hong Kong. The data was then compared with that from the existing CRDA drug reporting system. Additionally, we examined diurnal variations of drug residue loads in the community through analysis of two-hourly composite wastewater samples.

2. Materials and methods

2.1. Wastewater sampling

Samples were collected at the inlet of the largest wastewater treatment plant (WWTP) in Hong Kong. It serves approximately 3.5 million people, which is about half of the local population living in the mainly urban catchment. The WWTP is fed by two main inlet pipes (channels A and B) receiving wastewater from seven preliminary treatment works (PTWs). These PTWs physically remove coarse particles and sediments (screening and de-gritting) and continuously pump the wastewater to the WWTP under study. The average overall hydraulic residence time of wastewater collected and pumped into the WWTP through channel A is approximately three hours and four hours for channel B. The sewer layout and hydraulic properties provide considerable mixing of wastewater and attenuation of short-term concentration variations, facilitating the collection of representative samples. Samples were collected throughout the working week in 2011 on April 14th, 17th–21st and 24th–28th, representing the weekdays Sunday through Thursday. Unfortunately, samples from Fridays and Saturdays are missing since the WWTP does not conduct routine sampling on weekends and does not allow access for non-staff.

Hourly raw wastewater composite samples were collected at both inlet channels applying a time-proportional sampling mode, 250 mL every 15 min. With a few exceptions, intra-hour flow coefficient variations (CV) were relatively small: 3.6–53% (channel A) and 1.2–29% (channel B) (Fig. S1 and Table S1). Individual hourly samples were flow-proportionally mixed onsite in the laboratory of the WWTP to obtain representative daily composite samples for both channels. Additionally, to assess diurnal variations, the hourly samples from channel B were mixed flow-proportionally to two-hour composite samples on April 24th–28th. Milli-Q water samples were prepared and put aside during the sample composition process as field blanks for quality control. Samples were preserved at pH 2 using 2 M hydrochloride acid and frozen until analysis. The method of preservation has been widely applied and demonstrated to stabilise target analytes in wastewater during storage [29–31].

2.2. Materials and chemical analysis

Reference materials, sample preparation and analytical measurement applied in this study have been previously reported [32]. Briefly, cocaine, benzoylecgonine, amphetamine, methamphetamine, 3,4-methylenedioxymethamphetamine (MDMA), ketamine and norketamine, together with their corresponding deuterated analogues, were purchased from Cerilliant (USA) (purities ≥99%). Methanol and acetonitrile (LC grade) were purchased from Merck (Germany) while hydrochloric acid (37%), formic acid and acetic acid were purchased from Sigma–Aldrich (Australia).

Samples were filtered (0.45 μm, RC, Phenomenex) before spiking with deuterated standards (i.e. internal standards, 1–10 ng/mL) and then analysed together with seven calibration standards (0.05, 0.1, 0.5, 1, 5, 10, 50 ng/mL) using liquid chromatograph (Shimadzu Nexera UHPLC system, Kyoto, Japan) coupled with tandem mass-spectrometry (AB SCIEX QTRAP® 5500, ON, Canada) (LC–MS/MS). Targeted analytes were chromatographically separated using a Luna C18 column, 3 μm, 150 mm × 2 mm, (Phenomenex, Torrance, CA). Scheduled multiple reaction monitoring with positive electrospray ionisation were operated to identify and quantify the masses of analytes (see Lai et al. [32] for details of analytical conditions).

For quality assurance and control of the analysis, duplicate samples and wastewater matrix spiked with native chemicals (1 ng/mL) were arranged for analysis. Also, blank samples of Milli-Q water were included to check for contamination in every batch of sample preparation and analysis. Milli-Q water samples were spiked with native chemicals as procedural recovery checks. The coefficient variation (CV) of duplicate samples was on average <6% ($n = 5$). No target chemicals were quantified in the blank samples and field blank samples (Table S2). Procedural and matrix spike recoveries were on average in a range of 97–110% (CV: 4–16%; $n = 3$) and 89–104% (CV: 8–27%; $n = 5$), respectively, and inter-day analytical variability (2 days; $n = 8$) was in a CV range of 2–9% (Table S2). Average recoveries of individual internal standards in the samples were in a range of 62–120% (CV: 5–15%; $n = 77$) (Table S3).

2.3. Targeted drug residues

Seven drug residues, including parent drugs and/or its major metabolites, were targeted. These are methamphetamine, amphetamine, cocaine, benzoylecgonine, ketamine, norketamine and MDMA. The drugs have been reported to be commonly consumed in Hong Kong [6] and are regulated under Schedule 1 of the Dangerous Drugs Ordinance in the Laws of Hong Kong [33], meaning that use is illegal without authorised licenses.

2.4. Back calculation of drug consumption

The back calculation method was based on the model previously proposed [11] and has been commonly applied in the literature [e.g. 23,26,28]. A mass load of a given chemical was estimated by multiplying concentrations by the wastewater flow. The figure is then multiplied with a correction factor which comprised of the average urinary excretion rate and molecular mass ratio of a parent drug to its metabolite. The correction factor of 3.14 (1.1/0.35) was used to back estimate cocaine consumption. This was derived from the average excretion rate of cocaine to benzoylecgonine (35%, covering administration routes of smoking, snorting and injection) [34–36] and the molecular weight ratio of cocaine to benzoylecgonine (1.1). Similarly, the average excretion of methamphetamine itself (33%, covering administration routes of oral, smoking, snorting and injection) [37,38] was used to calculate the correction factor of 4.06 (1.0/0.33) for back estimating methamphetamine consumption. The correction