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Wastewater analysis reveals regional variability in exposure to abused drugs and opioids in Finland

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HIGHLIGHTS

- First quantitative results for tetrahydrocannabinol-9-carboxylic acid (a cannabis metabolite) in Finnish wastewater
- Confirms low level of cocaine and heroin use in Finland
- Shows a great difference between the metropolitan area, university cities and small rural towns
- Confirms the finding of the greater use of party drugs (ecstasy and cocaine) during weekends compared to weekdays

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ABSTRACT

Abused drug concentrations were determined in nine Finnish wastewater treatment plants (WWTPs), representing the metropolitan area, university cities and rural towns. In an eight-day study period in August 2012, 24-hour composite influent wastewater samples were collected. Biological markers and census-based information were used to estimate the size of the population served. The analytical method included solid phase extraction, liquid chromatographic separation, tandem mass spectrometric identification, and quantification using isotope-labeled internal standards. The study covered amphetamines, cannabis and cocaine. The levels of some opioids used in treatment and their metabolites were also determined. Amphetamine was the most prevalent drug of abuse, the median loads varying between the cities from 4.16 to 29.6 mg/1000 inhabitants/d. In three western cities methamphetamine was detected in even higher amounts, ranging from 0.87 to 47.5 mg/1000 inhabitants/d. Ecstasy (MDMA) and cocaine (as benzoylecgonine, BE) were found in higher concentrations during weekends compared to weekdays, the difference being statistically significant. The concentration of tetrahydrocannabinol-9-carboxylic acid (THCA) was below the limit of quantification in the two rural towns, while in the other cities the load varied between 3.77 and 20.7 mg/1000 inhabitants/d. The average variation in BE load was 0.05–6.82 and that of MDMA 0–20.6 mg/1000 inhabitants/d. While the metropolitan area showed the highest loads of abused drugs, the substances were continuously detected at all WWTPs included in the study. The median concentration of codeine ranged from 164 to 325 mg/1000 inhabitants/d and that of morphine from 18.8 to 31.5 mg/1000 inhabitants/d. The methadone load was below the level of detection in two towns, and at the other locations were 1.22–9.46 mg/1000 inhabitants/d. The first metabolite of heroin, 6-monoacetylmorphine (6-MAM), was not detected at all. Although the method has limitations, wastewater analysis gives additional information for assessing the degree of drug abuse and range of drugs abused in a society.

1. Introduction

Untreated wastewater can be considered as a huge urine pool and hence wastewater analysis can be utilized to monitor the degree of drug abuse in a society (Daughton, 2001). The original idea was willingly accepted since the problems in assessing drug abuse with conventional methods such as using results of population surveys, recording amounts of confiscated drugs or monitoring drug-related criminality, diseases and deaths, are generally recognized (Wiessing et al., 2008). On the other hand, accurate and updated information on the profile and extent of drug abuse is vital to society, e.g. in decision making, allocating funds and implementing preventive measures. Analysis of wastewater can, at best, provide independent, low-cost, reliable and almost real-time information. Drug abuse is a vast problem all over the world; according to a recent estimate about 3.4–6.6% of the

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world population aged between 15 and 64 are illicit drug users, out of which 10–13% are problem drug users (United Nations Office on Drugs and Crime, 2012). It is therefore not surprising that wastewater epidemiological methods have been employed in Europe, the US and Australia (van Nuijs et al., 2011a; Banta-Green et al., 2009; Lai et al., 2011).

In addition to identifying the spectrum of drugs used in an area and measuring their concentrations in wastewater, additional information can be obtained if the flow rate into the wastewater treatment plant (WWTP) and the corresponding population served are known. Moreover, if the metabolism and excretion rate of a given drug is also known, it is possible to calculate an estimate of the amount of a drug consumed and, utilizing average doses, even the number of doses abused in a given society in a given time period. The method described is based on determination of the so-called drug target residues (DTRs), which give information on individual drug consumption (Zuccato et al., 2008).

Epidemiologic methods based on the analysis of wastewater suffer from several shortcomings, which have been discussed in detail (Zuccato et al., 2008; van Nuijs et al., 2011a; Baker and Kasprzyk-Hordern, 2011a; Mathiu et al., 2011; Lai et al., 2011; Castiglioni et al., 2013). The ethical issue of wastewater analysis has also come under scrutiny (Hall et al., 2012). Despite the limitations of the method, earlier studies have shown that the class and volume of abused drugs change geographically and also with time within a country (Zuccato et al., 2008; Gerrity et al., 2011; Irvine et al., 2011; Reid et al., 2011; Bijlsma et al., 2012; Lai et al., 2012; Thomas et al., 2012). Thus the results derived from a single community do not necessarily reflect the prevalent status in the larger area or in the country as a whole.

Not many wastewater epidemiological studies have been conducted in Finland to date. Since our preliminary study (Vuori et al., 2010) only one other study has been published (Thomas et al., 2012). This compared illicit drug use in 19 European cities. Two Finnish cities, Helsinki and Turku, were included. Both these studies confirmed the absence of heroin and the low consumption of cocaine in Finland, which was already known from mortality statistics (Vuori et al., 2012; EMCDDA, 2013). Both studies also showed that the abuse of party drugs (MDMA and cocaine) is most prevalent at weekends and much lower on weekdays. However, the latter study found an exceptionally high mass flow of methamphetamine in Helsinki and Turku compared to other European cities. Earlier studies were unable to estimate the consumption of cannabis because reliable quantitative results were lacking. In order to get a better picture of the status of drug abuse in Finland the present wastewater epidemiological study was extended to cover nine Finnish towns and cities.

2. Materials and methods

2.1. Sample collection

The locations of the nine towns and cities included in the study and their populations are shown in Fig. 1. Helsinki and Espoo compose the metropolitan area; Turku, Tampere, Jyväskylä, Oulu and Swedish-speaking Vaasa are smaller university cities while Savonlinna and Seinäjoki are rural towns. The WWTPs included in the study typically serve not only the city itself but also adjacent communities. The overall census-based population of the study covers more than two million inhabitants, which correspond about 38% of the total population of Finland.

The 24-hour composite influent wastewater samples were collected during eight days starting Sunday 12th August 2012 (Helsinki, Tampere, Turku and Savonlinna) or Monday 13th August 2012 (Espoo, Jyväskylä, Oulu, Seinäjoki and Vaasa). The sampling period was chosen so that the summer holiday season was almost over and the rainy autumn period had not yet begun. The samples were collected in two 500 mL polyethylene bottles provided by the study group; one bottle for drug analysis

Fig. 1. The nine Finnish towns and cities included in the study and their census-based populations.

and the other for estimating the population size. Otherwise the WWTP's own equipment was used. The WWTPs recorded the flow rate daily and the concentrations of nitrogen (N), phosphorus (P) and chemical oxygen demand (COD) according to each WWTP's individual practice and routine. The results were used as controls for our own corresponding laboratory results. Six out of nine WWTPs used flow-proportional sampling mode, while in Oulu and Savonlinna the mode was time-proportional. In Turku flow-proportional sampling mode was used during the first six days and time-proportional mode during the final two days. In flow-proportional sampling a subsample was taken after a given flow of incoming wastewater which varied according to the size of WWTP ranging from 66 m^3 (Vaasa) to 5000 m^3 (Helsinki). In time-proportional sampling the subsample was taken in Oulu and Savonlinna after every 30 min and in Turku after every 15 min. At all but one of the WWTPs the complete samples were frozen without delay and sent to the laboratory in cool boxes according to the instructions given. In Turku during the weekdays before freezing there was a two hour delay during which the samples were kept in a refrigerator. Additionally, during the weekend there were no staff present at the WWTP of Turku and hence the weekend samples were frozen on Monday morning and until that the samples stayed at approximately 15 °C.

2.2. Population size estimation

In order to allow a comparison of drug use between different cities and with previous studies the results are expressed per 1000