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Long-term monitoring of drug consumption patterns in a large-sized European city using wastewater-based epidemiology: Comparison of two sampling schemes for the assessment of multiannual trends

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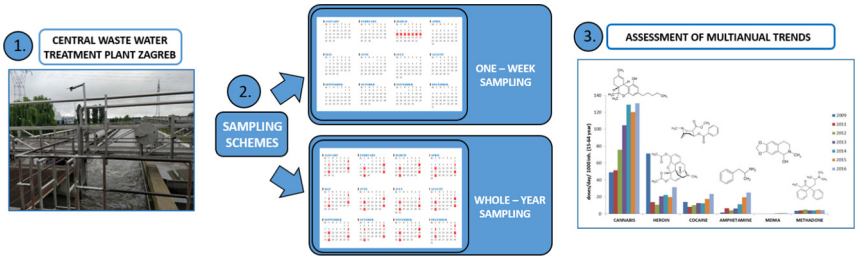
HIGHLIGHTS

- Drug consumption patterns were studied using wastewater-based epidemiology.
- The 8-year study was performed in a large European city.
- Comparison of one-week and whole-year sampling strategies was made.
- Significant multiannual drug consumption changes were determined.
- A comparison with epidemiological data was performed.

GRAPHICAL ABSTRACT



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ABSTRACT

A comprehensive study aimed at monitoring of temporal variability of illicit drugs (heroin, cocaine, amphetamine, MDMA, methamphetamine and cannabis) and therapeutic opiate methadone in a large-sized European city using wastewater-based epidemiology (WBE) was conducted in the city of Zagreb, Croatia, during an 8-year period (2009–2016). The study addressed the impact of different sampling schemes on the assessment of temporal drug consumption patterns, in particular multiannual consumption trends and documented the possible errors associated with the one-week sampling scheme. The highest drug consumption prevalence was determined for cannabis (from 59 ± 18 to 156 ± 37 doses/day/1000 inhabitants 15–64 years), followed by heroin (from 11 ± 10 to 71 ± 19 doses/day/1000 inhabitants 15–64 years), cocaine (from 8.3 ± 0.9 to 23 ± 4.0 doses/day/1000 inhabitants 15–64 years) and amphetamine (from 1.3 ± 0.9 to 21 ± 6.1 doses/day/1000 inhabitants 15–64 years) whereas the consumption of MDMA was comparatively lower (from 0.18 ± 0.08 to 2.7 doses ±0.7 doses/day/1000 inhabitants 15–64 years). The drug consumption patterns were characterized by clearly enhanced weekend and Christmas season consumption of stimulating drugs (cocaine, MDMA and amphetamine) and somewhat lower summer consumption of almost all drugs. Pronounced multiannual consumption trends were determined for most of the illicit drugs. The investigated 8-year period was characterized by a marked increase of the consumption of pure cocaine (1.6-fold), THC (2.7-fold), amphetamine (16-fold) and MDMA (15-fold) and a concomitant decrease (2.3-fold) of the consumption of pure heroin. The heroin consumption decrease was associated with an increase of methadone consumption (1.4-fold), which can be linked to its use in the heroin substitution therapy. The estimated number of average methadone doses consumed in the city of Zagreb was in a good agreement with the prescription data on treated opioid addicts in Croatia.

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1. Introduction

Abuse of illicit drugs has become a major global problem with numerous negative consequences including increase in crime rate, negative impacts on public health, economic damage as well as costs of treatment of drug addicts (EMCDDA, 2009). Consequently, knowing the extent and patterns of drug abuse is very important for planning timely and effective actions to mitigate these problems. The official data about illicit drug consumption usually include the information about the amount and purity of seized drugs, number of treated drug addicts and general population survey data, whose frequency in different countries may be rather different. In recent years, wastewater-based epidemiology (WBE) has been used as a complementary approach for the estimation of drug consumption across the world (e.g. Bijlsma et al., 2016; Bones et al., 2007; Huerta-Fontela et al., 2008; Khan et al., 2014; Kankaanpää et al., 2014; Kasprzyk-Hordern et al., 2009; Irvine et al., 2011; Lai et al., 2013a, 2016; Metcalfe et al., 2010; Postigo et al., 2010; Terzic et al., 2010; van Nuijs et al., 2009; Zuccato et al., 2008).

The main advantages of the WBE approach are objectivity and suitability for near-real-time monitoring. In order to improve and expand the WBE approach, several publications addressed the problem of uncertainties associated with sample collection (Ort et al., 2010), sample stability (McCall et al., 2016; van Nuijs et al., 2012; Senta et al., 2014) as well as back-calculation of drug consumption (Castiglioni et al., 2013; Gracia-Lor et al., 2016; Lai et al., 2011). A number of studies have already demonstrated the potential of WBE to provide information about the spatial (Been et al., 2016; Bijlsma et al., 2016; Kankaanpää et al., 2016; Nefau et al., 2013) and temporal (Bade et al., 2018; Been et al., 2016; Lai et al., 2016; Mastroianni et al., 2017; Tscharke et al., 2016) drug consumption patterns, including large international comparative studies (Ort et al., 2014a; Thomas et al., 2012), which showed a pronounced regional and temporal variability of drug abuse across the Europe. In several studies, the potential of this approach as a complementary tool to support epidemiological and seizure data (Baz-Lomba et al., 2017; Been et al., 2016; Zuccato et al., 2016) was demonstrated. The WBE approach was also successfully applied to study the differences in drug consumption patterns between the large and small cities (Krizman et al., 2016; van Nuijs et al., 2009), with a clear indication that large cities represent communities with significantly enhanced drug consumption and, consequently, are very suitable for the investigation of the drug consumption patterns.

Regarding temporal variability, a significant emphasis of existing studies was on short-term consumption variability, especially regarding so-called recreational stimulating drugs. A number of WBE studies performed in different countries confirmed an enhanced consumption of stimulating illicit drugs during the weekend (e.g. Krizman et al., 2016; Terzic et al., 2010; Thomas et al., 2012), large sport events (Gerrity et al., 2011), music festivals (Bijlsma et al., 2014; Jiang et al., 2015; Lai et al., 2013b; Mackuľak et al., 2014) and the peak of tourist season in the vacation areas (Krizman et al., 2016; Lai et al., 2013c). In contrast, only few reports addressed the issue of multiannual changes in drug consumption patterns within the selected population (e.g. Kankaanpää et al., 2016; Mastroianni et al., 2017; Ort et al., 2014a; Tscharke et al., 2016; Zuccato et al., 2016). Most of the published multiannual studies were based on the comparison of one-week wastewater sampling campaigns in a given time-period (Kankaanpää et al., 2016; Mastroianni et al., 2017; Ort et al., 2014a; Zuccato et al., 2016.). In such cases, possible week-to-week variability during the particular year was not taken into account, which might increase the uncertainties related to the annual consumption estimates. In order to get a more accurate estimate, representative of average annual drug consumption, a recent study by Ort et al. (2014b) recommended the use of stratified annual sampling to minimize the errors associated with day-to-day variability. The importance of sampling scheme for the assessment of consumption was also discussed in Humphries et al. (2016).

In this study we investigated the multiannual trends in the consumption of 6 illicit drugs (cannabis, cocaine, heroin, MDMA, amphetamine, methamphetamine) and one therapeutic opioid (methadone) in the city of Zagreb in the period 2009–2016, by applying two different sampling schemes (one-week sampling scheme and a whole-year sampling scheme). The city of Zagreb is the capital and the largest Croatian city, representing almost 20% of Croatia's population. Furthermore, an initial WBE study conducted in Zagreb (Terzic et al., 2010) indicated specific drug consumption patterns which were different from those reported for most of the other European cities, in particular regarding comparatively higher prevalence of heroin consumption and lower prevalence of cocaine and amphetamine drug consumption.

The specific goals of the present study included: a) long-term study of the weekday-related drug consumption patterns; b) impact of the holiday season on drug consumption patterns; c) seasonal changes in drug consumption patterns; d) testing different sampling schemes for the assessment of multiannual trends; e) tracking the multiannual changes of the drug consumption over a period of 8 years and comparison with the available epidemiological data.

2. Materials and methods

2.1. Selection of target compounds

The selection of target compounds was based on the available data on drug consumption patterns in Croatia (Glavak Tkalic et al., 2013) and in the city of Zagreb (Krizman et al., 2016; Terzic et al., 2010). Selected analytes included morphine (MOR), morphine 3 glucuronide (M3G) and 6-acetylmorphine (6-AM) as principal heroin-derived substances as well as benzoylecgonine (BE), amphetamine (AMP), methamphetamine (MAMP), 3,4 methylendioximethamphetamine (MDMA), 11 nor 9 carboxy tetrahydrocannabinol (THC-COOH) and 2 ethylidene 1,5 dimethyl 3,3 diphenylpyrrolidine (EDDP) as principal biomarkers of cocaine, amphetamine, methamphetamine, MDMA, cannabis and methadone consumption, respectively.

2.2. Chemicals and materials

Standard solutions of all target analytes (1 g/L) and their deuterated analogues (0.1 g/L) were purchased from Lipomed AG (Switzerland). Mixed standard solutions of the analytes and their deuterated analogues, used as surrogate standards, were prepared in methanol (MeOH) at concentrations of 10 mg/L and 2 mg/L, respectively, and kept in the dark at −20 °C. Aqueous ammonia solution (NH_3, 25%) and LC-MS grade MeOH were purchased from Merck AG (Darmstadt, Germany). Acetic acid (CH_3COOH), also LC-MS grade, formic acid (HCOOH) and phosphoric acid (H_3PO_4) were purchased from Fluka (Switzerland). Milli-Q water was obtained by purifying with an Elix-Mili-Q-system (Millipore, Bedford, USA). Oasis MCX cartridges (150 mg/6 mL) were purchased from Waters (Milford, MA, SAD) whereas Strata NH_2 (200 mg/3 mL) cartridges as well as HPLC columns used for the chromatographic separation (Synergi Polar; 4 μm, 150 mm × 3 mm, Kinetex PFP; 2.6 μm, 100 mm × 2.1 mm) were purchased from Phenomenex (Torrance, California, USA). Glass-fiber filters (GF/C) were purchased from Whatman (USA).

2.3. Wastewater sampling and analysis

The 24-h composite samples (from 8 a.m. of the previous day to 8 a.m. of the sample collection day) of untreated wastewater were collected at the inlet of the central WWTP of the city of Zagreb in the period 2009–2016, except in 2010. All collected samples were time-proportional, with the sampling time interval of 15 min. A total number of 282 samples, having an average pH of 7.6 ± 0.2, was collected. Depending on the specific research goals, different sampling schemes were applied to cover both short-time and long-term variability: one-week sample scheme, a whole-year sampling-scheme and Christmas season sampling scheme.