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Wastewater-based assessment of regional and temporal consumption patterns of illicit drugs and therapeutic opioids in Croatia



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Drug consumption patterns in Croatia were studied using sewage epidemiology
- The most prevalent illicit drugs were cannabis, cocaine and heroin
- Drug consumption shows pronounced regional and seasonal differences
- Seasonal differences were observed between coastal and continental cities
- The results were compared with epidemiological data on treated addicts



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ABSTRACT

A comprehensive study of spatial and temporal consumption patterns of the selected illicit drugs (heroin, cocaine, amphetamine, MDMA, methamphetamine, cannabis) and therapeutic opioids (codeine, methadone) has been performed in six Croatian cities by applying wastewater-based epidemiology. The investigated cities (Bjelovar, Vinkovci, Varazdin, Karlovac, Zadar and Zagreb) varied widely in the population size (27,000-688,000 inhabitants) as well as in the number of registered drug consumers included in compulsory and voluntary medical treatment and rehabilitation programs (30-513 persons/100,000 inhabitants of age 15-64). The most consumed illicit drugs were cannabis (10–70 doses/day/1000 inhabitants), heroin (<0.2–10 doses/day/ 1000 inhabitants) and cocaine (0.2-8.7 doses/day/1000 inhabitants), while the consumption of amphetaminetype drugs was much lower (<0.01-4.4 doses/day/1000 inhabitants). Enhanced consumption of illegal drugs was generally associated with larger urban centers (Zagreb and Zadar) however comparatively high consumption rate of cocaine, MDMA and methadone was determined in some smaller cities as well. The overall average dose number of 3 major illegal stimulants (cocaine, MDMA, amphetamine) was rather similar to the number of corresponding heroin doses, which is in disagreement with a comparatively much higher proportion of heroin users in the total number of registered drug users in Croatia. Furthermore, the illicit drug consumption pattern in the large continental city (Zagreb) was characterized by a significant enhancement of the consumption of all stimulants during the weekend, which could not be confirmed neither for the coastal city of Zadar nor for the remaining small continental cities. On the other hand, the city of Zadar exhibited a significant increase of stimulant drug usage during summer vacation period, as a result of pronounced seasonal changes of the population composition and lifestyle in coastal tourist centers. The obtained results represent a valuable complementary data source for the optimisation and implementation of strategies to combat drug abuse in Croatia. © 2016 Published by Elsevier B.V.

1. Introduction

Wastewater-based epidemiology has been increasingly used as an additional source of information on the consumption of illicit drugs (e.g. Bijlsma et al., 2014; Irvine et al., 2011; Kankaanpää et al., 2014; Khan et al., 2014.; Nefau et al., 2013; Ort et al., 2014; Östman et al., 2014; Postigo et al., 2010; Terzic et al., 2010; Thomas et al., 2012; Vuori et al., 2014; Zuccato et al., 2008). The main advantage of this innovative approach is its objectivity, which is based on highly accurate chemical measurements of selected drug biomarkers in wastewater, and its suitability for near-real-time tracking of the changes in drug consumption patterns within the selected communities. Wastewater analvsis can provide information about spatial and temporal variations of illicit drugs use as well as on the impact of special events such as national holidays or music festivals on drug consumption patterns. Consumption patterns were investigated at different scales: as detailed studies in one selected municipality (e.g. Karolak et al., 2010; Terzic et al., 2010), as a part of national surveys (e.g. Banta-Green et al., 2009; Kankaanpää et al., 2014; Metcalfe et al., 2010; Nefau et al., 2013; van Nuijs et al., 2009; Vuori et al., 2014) or in the international context (e.g. Ort et al., 2014; Thomas et al., 2012; Zuccato et al., 2008). The recently performed international studies (Ort et al., 2014; Thomas et al., 2012) indicated rather pronounced geographical differences in illicit drug consumption patterns across Europe. The highest cocaine use was determined in western and central Europe, while the highest consumption of methamphetamine was determined in northern and eastern Europe. Furthermore, significant differences in drug consumption patterns were also demonstrated within the same country (e.g. Kankaanpää et al., 2014; Lai et al., 2013a; Nefau et al., 2013; Thomas et al., 2012; van Nuijs et al., 2009), indicating typically higher illicit drug abuse in highly urbanized metropolitan areas. Wastewater analyses can also be used to track temporal variability in illicit drug consumption. Zuccato et al. (2011) demonstrated a significant drop of cocaine use at the beginning of economic crisis in Italy. Moreover, a number of studies demonstrated a pronounced increase in consumption of stimulants during the weekend (Bijlsma et al., 2009; Huerta-Fontela et al., 2008; Karolak et al., 2010; Lai et al., 2011; Prichard et al., 2012; Reid et al., 2011; Terzic et al., 2010; Thomas et al., 2012; van Nuijs et al., 2009; Zuccato et al., 2008) as well as during music festivals (Bijlsma et al., 2014; Lai et al., 2013b; Mackulak et al., 2014) and sport events (Gerrity et al., 2011). Lai et al. (2013a) demonstrated that the consumption of several illicit drugs, including MDMA, cocaine and methamphetamine on the Australian vacation island significantly increased during the national peak holiday season, however the number of studies on seasonal variability of drug consumption is still rather low.

There are three major specific aims to study patterns of drug abuse in Croatia. As a transition country, Croatia is characterized by fast socio-economic changes in the last two decades, some of which could be linked with the changing patterns in drug abuse. Specifically, Croatia is situated on a so-called Balkan route of drug trafficking, making it vulnerable to enhanced availability of illicit drugs originating from Middle East. After the end of the war conflicts in the former Yugoslavia, this route has become increasingly actual. Finally, Croatia's fast growing tourism during the last couple of years has become an important issue when addressing different routes of drug trafficking. Traditionally, sources of information on illicit drug consumption in Croatia have included primarily the data on the number of treated drug addicts and police drug seizures, while general population surveys as well as wastewater-based epidemiology have been used only recently. The general population survey conducted in 2011 indicated that the lifetime prevalence rates of almost all investigated illicit drugs in Croatia were below the European average (Glavak Tkalic et al., 2013), which is in a good agreement with the results of the two recent wastewaterbased Europe-wide studies (Ort et al., 2014; Thomas et al., 2012). On the other hand, the same study indicated that the lifetime prevalence of heroin consumption in Croatia in 2011 (0.4%) was at the European average (0.41%). Moreover, the wastewater-based epidemiology study conducted in the city of Zagreb in 2009 indicated that the consumption prevalence of heroin was even higher than in some other European cities (Terzic et al., 2010). Currently available wastewater-based epidemiology data for Croatia are limited only to the city of Zagreb while the data on regional differences in illicit drug consumption are still missing. The aim of this study was therefore to investigate regional variability of the consumption patterns of illicit drugs (cocaine, heroin, amphetamine, MDMA, methamphetamine and cannabis) and two therapeutic opioids (methadone, codeine) in Croatia. The specific goals of the study included: a) a preliminary comparison of drug consumption patterns in six selected Croatian cities with regard to the city population size and geographic position; b) study of weekday-related consumption patterns; c) study of changes in drug consumption patterns associated with summer tourist season in two selected cities and d) comparison of wastewater-based consumption estimates with the available epidemiological data.

2. Material and methods

2.1. Selection of target compounds

The study included analyses of 6-acetylmorphine (6-AM), morphine (MOR) and 3- β -D-morphine glucuronide (MG) as principal heroinderived substances, cocaine (COC) and its main metabolite benzoylecgonine amphetamine (AMP). 3.4-(BE). methylenedioxymetamphetamine (MDMA, ecstasy), methamphetamine (MAMP) as well as two urinary metabolite of cannabis, 11-nor-9-carboxy- Δ^9 -tetrahydrocannabinol (THC-COOH) and 11-hydroxy- Δ^9 tetrahydrocannabinol (THC-OH). Methadone (MTHD) and its metabolite, 2-ethylidene-1.5-dimethyl-3.3-diphenylpyrrolidine (EDDP) as well as codeine (COD) were monitored as representatives of therapeutic opioids. The selection of target compounds was made based on the data on drug seizures in Croatia and general population survey performed in Croatia in 2011 (Glavak Tkalic et al., 2013).

2.2. Chemicals and materials

Standard solutions of all target analytes (1 g L^{-1}) and their deuterated analogs (0.1 g L^{-1}) were purchased from Lipomed AG (Switzerland). Mixed standard solutions of the analytes and their deuterated analogs (used as surrogate standards) were prepared in methanol (MeOH) at concentrations of 10 mg L⁻¹ and 2 mg L⁻¹, respectively, and kept in the dark at -20 °C. Aqueous ammonia solution (NH₃, 25%) and LC-MS grade MeOH were purchased from Merck AG (Darmstadt, Germany). Acetic acid (CH₃COOH), also LC-MS grade, formic acid (HCOOH) and phosphoric acid (H₃PO₄) were purchased from Fluka (Switzerland). MQ water was obtained by purifying in Elix-Mili-Q-system (Millipore, Bedford, USA). Oasis MCX cartridges (150 mg/6 mL) were produced by Waters (Milford, MA, SAD) while Strata NH₂ (200 mg / 3 mL) cartridges as well as HPLC columns used for the chromatographic separation (Synergi Polar; 4 µm, 150 mm × 3 mm and Kinetex PFP; 2.6 µm, 100 mm × 2.1 mm) were manufactured by Phenomenex (Torrance,

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