Cocaine consumption can be evaluated through analysis of waste and surface water.

1. Introduction

Abuse of illicit drugs is a major problem in our contemporary society. The consumption of drugs leads not only to a high mortality and morbidity, but also to many socio-economic problems (economic damage, criminality, social insolation, a.s.o.). The European Monitoring Centre for Drugs and Drug Addiction (EMCDDA) estimates that even 4% of the European adults (approximately 12 million people) has used cocaine (COC) during their lifetime, and even 4% of the European adults (approximately 12 million people) has used cocaine (COC) during their lifetime, and estimates that even 4% of the European adults (approximately 12 million people) has used cocaine (COC) during their lifetime, and even 4% of the European adults (approximately 12 million people) has used cocaine (COC) during their lifetime. The consumption of drugs leads not only to a high mortality and morbidity, but also to many socio-economic problems (economic damage, criminality, social insolation, a.s.o.).

Cocaine is still administered as topical anesthetic compound in intranasal, ophthalmologic and bronchoscopic procedures (Dart, 2004). Illegally, cocaine is used by intranasal (“snorting”), intravenous or smoking routes (Cone, 1995). It has three important actions on the autonomic nervous system over-stimulation, risk of heart attack, pulmonary complications and altered serotonin levels (Lakoski et al., 1991). In humans, COC is rapidly hydrolyzed to benzoylecgonine (BE) and ecgonine methyl ester (EME) and some minor metabolites (Dart, 2004). Only a small fraction of COC is excreted in urine as parent compound, while the largest amount is excreted as BE, the most important metabolite, and EME (Ambre et al., 1991, 1994). In urine, COC can be detected for approximately 8 h, while BE and EME can be positively identified for more than 96 h (Hamilton et al., 1977; Jufer et al., 2000).

The European Monitoring Centre for Drugs and Drug Addiction (EMCDDA, 2007) observed, after a stabilizing trend in that 3 million European adults have used cocaine in 2007 (EMCDDA, 2007), probably as a consequence of their consumption of other drugs such as ecstasy and cannabis has been observed (Jufer et al., 2000). Such general indicators can give only raw estimations of drug abuse, but accurate and local measurements are not possible with this approach (Zuccato et al., 2005). Also in Belgium, limited information is available about the cocaine abuse (Lamkaddem and Roelands, 2007).

Cocaine abuse, a growing social problem, is currently estimated from population surveys, consumer interviews and crime statistics. A new approach based on the analysis of cocaine (COC) and metabolites, benzoylecgonine (BE) and ecgonine methyl ester (EME), in water samples was applied to 28 rivers and 37 waste water treatment plants in Belgium using solid-phase extraction and liquid chromatography coupled to tandem mass spectrometry. While EME was undetectable, COC and BE were detectable with concentrations ranging from <1 to 753 ng/L and <1 to 2258 ng/L, respectively. BE concentrations were used to calculate the local amount of abused cocaine. The highest values (up to 1.8 g/day cocaine per 1000 inhabitants) were found in large cities and during weekends. The estimation of cocaine abuse through water analysis can be executed on regular basis without cooperation of patients. It also gives clear geographical information, while prevention campaigns can easily be implemented and evaluated.

Cocaine and metabolites in waste and surface water across Belgium

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Cocaine
Waste water
Surface water
Cocaine equivalents
Cocaine consumption
LC–MS

COC can be detected for approximately 8 h, while BE and EME can be positively identified for more than 96 h (Hamilton et al., 1977; Jufer et al., 2000).
Recently, a new, direct and objective way of monitoring drug consumption has been proposed, based on the measurement of urinary excreted drugs and their metabolites in waste and surface water (Bones et al., 2007; Castiglioni et al., 2006; Gheorghe et al., 2008; Huerta-Fontela et al., 2007; Hummel et al., 2006; Kasprzyk-Hordern et al., 2008a,b; Zuccato et al., 2005, 2008). While these studies are particularly based on measurements by liquid chromatography coupled to tandem mass spectrometry (LC–MS/MS), they differ on the sample preparation approaches and the groups of investigated drugs of abuse.

In the present study, a previously optimized and validated method based on solid-phase extraction (SPE) and hydrophilic interaction (HILIC) LC–MS/MS (Gheorghe et al., 2008) was applied to quantify COC, BE and EME in waste water from 37 waste water treatment plants (WWTPs) and in surface water from 28 rivers and brooks across Belgium. To the best of the authors’ knowledge, this is the largest monitoring study of cocaine and its metabolites in waste and surface waters up to date. The concentrations of COC and metabolites were then converted into cocaine equivalents, using the flow rate of the water stream and a formula that takes into account the molecular masses and the excretion pattern of cocaine and metabolites. The final aim was to detect regions with a high consumption of cocaine. This approach will ultimately lead to valuable information needed to tackle the growing problem of cocaine abuse in Belgium and in Europe.

2. Materials and methods

2.1. Sampling sites and sample collection

2.1.1. Sampling sites

WWTPs (n = 37) were chosen based on the amount of persons they serve and on their geographical location, so that the overall picture of cocaine use in Belgium could be made (Table 1, Fig. 1 and Fig. SI-1). Small WWTPs, which serve less than 10,000 persons, were excluded from this study. The chosen WWTPs cover about 3,700,000 inhabitants (~40% of the total Belgian population). Sampling sites on 28 rivers and brooks (n = 43) were also chosen so that they cover the whole Belgian hydrographic system (Table 2 and Fig. SI-2). Two rivers (the Demer and the Dijle) were sampled during the same day at four different downstream sites to evaluate differences in concentrations of COC and BE. One river, the Zenne, was sampled upstream and downstream of the urban region of Brussels.

2.1.2. Sample collection

Influent water samples (24-h flow dependent composite samples, 1.5 L) were collected from WWTPs across Belgium. For each WWTP, two samples were taken, one on Sunday and one on Wednesday to evaluate differences in cocaine abuse during the week. From the WWTP of Mouscron (4 Nov 07) we received a 99-h composite sample, from Turnai (20 Jan 08) a 48-h composite sample and from Waver (18 Nov 07) a 72-h composite sample. Surface water samples (grap samples, 2.5 L) were collected at 43 sampling points from 28 rivers and brooks across Belgium. All samples were collected during periods with low or no rainfall to prevent excessive dilution of analytes. Only for river Ijzer, the sampling was conducted after a heavy rainfall. For each sample, pH and temperature at the time of sampling were determined. A tap water sample spiked with 200 ng/L COC and 800 ng/L BE was also analyzed by the two laboratories. The relative standard deviation (RSD) of the mean concentrations of COC and BE analyzed by the two laboratories was in all cases lower than the recommended cut-off value of 20% (Reed et al., 2002). This is for the first time that an interlaboratory test is reported for the analysis of COC and metabolites in waste and surface water samples were made by the LC: (i) the use of a C8 UPLC column and (ii) the use of an UPLC–MS/MS instrument. A tap water sample spiked with 200 ng/L COC and 800 ng/L BE was also analyzed by the two laboratories. Table SI-2 summarizes the results for the interlaboratory test. The relative standard deviation (RSD) of the mean concentrations of COC and BE analyzed by the two laboratories was in all cases lower than the recommended cut-off value of 20% (Reed et al., 2002). This is for the first time that an interlaboratory test is reported for the analysis of COC and metabolites in waste water.

A quality control was executed during the analyses. With each batch of seven samples, a tap water sample spiked with 206 ng/L COC and 841 ng/L BE was analyzed and the recovery was calculated. Results are visualised in Figs. SI-3 and SI-4. All quality controls are within the control limits of 3 × SD, as suggested by Masson (2007).

3. Results and discussion

3.1. Assumptions to calculate cocaine equivalents

To evaluate the cocaine abuse in the selected regions, it is necessary to transform the concentrations [in ng/L] into total cocaine equivalents (in g/day) and, if possible, to further report these cocaine equivalents per 1000 inhabitants. For these calculations, different parameters which play an important role have to be known: (i) the flow rate of the water to transform concentrations of COC and BE into loads of COC and BE (expressed in g/day); (ii) the relative amount of a cocaine dose excreted as COC and BE to make a back calculation from loads COC and BE to a total amount of abused COC (in g/day); (iii) the molecular masses of the different compounds/metabolites; (iv) the stability (temperature, pH and time of presence in water) of COC and metabolites in aqueous matrices; and (v) the amount of inhabitants that is served by a certain WWTP.

In the literature two approaches to calculate cocaine equivalents can be found. Zuccato et al. (2005) proposed a formula based...
excluding on BE loads. They assumed that about 45% of a cocaine dose is excreted as BE and only a small fraction as COC. Bones et al. (2007) calculated the cocaine equivalents based exclusively on COC loads and on the assumption that approximately 10% of a cocaine dose is excreted as COC. This group assumed also that COC is stable in waste and surface water and that the source of the COC found in waste and surface water comes from human excretion rather than from dumping of large quantities. The latter approach seems to be less reliable than the one proposed by Zuccato et al. (2005), since COC degraded rapidly in water, while BE is more stable (Gheorghe et al., 2008).

The above-described approaches (Bones et al., 2007; Zuccato et al., 2005) are based on several assumptions and they probably underestimate the real use of cocaine. The literature is inconsistent regarding the urinary excretion of COC. The relative amount of a COC dose excreted as COC and BE differs strongly between various experimental setups. Between 1 and 15% of a cocaine dose is excreted as COC, while between 15 and 55% of a cocaine dose is excreted as BE (Ambre et al., 1984, 1986; Cone et al., 1998).

In the future, it is necessary to develop a new mathematical model that takes every process (stability in water, urinary excretion, adsorption on solid particles) into account for a more exact estimation of the amount of cocaine abused in a certain region.

The calculation of the cocaine equivalents in Tables 1 and 2 is performed by applying the formula by Zuccato et al. (2005).

3.2. Cocaine and metabolites in surface water across Belgium

Table 2 and Fig. SI-2 summarize the concentrations of COC and BE and their corresponding COC equivalents in surface water across Belgium. In all samples, EMЕ was found below LOQ (20 ng/L), probably because of its low stability in water (Gheorghe et al., 2008) and their high LOQ compared to BE and COC, and therefore was not included in further calculations. COC and BE were present in surface water with a maximum of 115 and 520 ng/L, respectively. For surface water, no further association between the COC equivalents and the amount of inhabitants living on the river’s basin was made, since the latter cannot be estimated with accurate precision.

Generally, COC and BE levels were above LOQ in most surface water samples from Flanders (North of Belgium). Only for some smaller rivers, such as the Binchebeek and the Dommel, COC or BE was below LOQ. The absence of COC and BE in the Ijzer, a rather large river, is probably because of the massive rainfall during the sampling period (Table 2), which resulted in a high dilution of COC and BE in the surface water. Contrarily, we had a lower number of positive samples in surface water from Wallonia (South of Belgium), most probably due to the lower population density in Wallonia compared to Flanders.

The Dijle, a river that rises in Wallonia, was sampled at four different sites: Limelette, Florival, Korbeek and Wilsele (from upstream to downstream). An increasing trend in the COC equivalents was observed when going downstream the Dijle. This
observation agrees with earlier reports in other rivers (Zuccato et al., 2008) and could be explained by the continuous contamination of the rivers by waste water resulting from domestic use. Most of the domestic water runs through WWTPs, with variable removal efficiencies of COC and metabolites largely depending on the applied treatment technique (Castiglioni et al., 2006). Therefore, the effluent of the WWTPs may still contain COC and BE. Moreover, direct discharges of untreated waste water in the rivers are possible, since a large percentage of the population is not connected to a WWTP. The small drop in COC equivalents between Korbeek and Wilsele is probably a consequence of the fact that the Dijle does not receive waste water in this area and that, in the absence of additional source, the degradation of COC and BE occurs in water (Gheorghe et al., 2008). A similar trend in the COC equivalents was seen for the Demer, with the following sampling sites ranked downstream: Hasselt, Kermt, Halen and Aarschot.

Concentrations of COC and BE found in Belgian surface waters are in most cases comparable with concentrations measured in Italy (Zuccato et al., 2005, 2008), Spain (Huerta-Fontela et al., 2007) and the UK (Kasprzyk-Hordern et al., 2008a,b; Zuccato et al., 2005) (Table 3). Since COC is transformed readily in BE in the human body, a higher concentration of BE than COC in surface water could be expected. This was confirmed by our results and results from the other studies, in which the logical pattern of higher BE concentrations than COC concentrations is observed. However, our results differ from those of Bones et al. (2007), who have detected COC in surface water without being able to measure BE in most cases. Therefore, this is a remarkable (if not questionable) finding which has been partially explained by Bones et al. through the use of a different sample preparation technique.

The mean ± SD ratio COC/BE for the surface water samples was 0.18 ± 0.11 and no outliers were observed. This value agrees well with values reported for urinary excretion of BE and COC (mean 8% COC, 35% BE; COC/BE = 0.23) (Ambre et al., 1984, 1988; Cone et al., 1998). The relatively high RSD (60%) is a consequence of the very broad ranges of urinary excretion of COC and BE reported in the literature. Our findings are comparable with, but yet higher than the ratio measured by Zuccato et al. (2005) in the Po river (0.05), probably attributed to different temperatures of river Po and the rivers sampled in the present study, resulting in a different degradation pattern of COC and BE.

The concentrations of BE and COC in the river Zenne at Vilvoorde (downstream of Brussels) are well above the levels observed in other Belgian rivers (this study) and in other European rivers (Table 3). The river Zenne still receives directly a large amount of industrial and domestic untreated waste water from Brussels, which explains the high concentrations of COC and BE observed downstream of Brussels. This hypothesis is confirmed by the low levels of COC and BE measured in the surface water collected on the same day from river Zenne in Lot (upstream of Brussels) (Table 2), which suggests that indeed the urban agglomeration of Brussels is responsible for the dramatic increase in concentrations of COC and BE in the water of river Zenne.

### 3.3. Cocaine and metabolites in waste water across Belgium

COC and BE were quantifiable in all waste water samples collected from WWTPs spread across Belgium (Table 1 and Fig. 1 and Fig. SI-1). Similar to surface water, EME was below LOQ (20 ng/L) in all WWTP samples. It is worth mentioning that EME was neither reported nor measured in WWTP samples until now. The concentration ranges of COC and BE in the influent WWTP water samples were 10–753 ng/L and 33–2258 ng/L, respectively.
Every WWTP was sampled twice, on Wednesday and on Sunday, with the purpose to investigate differences in the abuse of cocaine throughout the week. Cocaine abuse was significantly higher (p = 0.018, paired samples t-test) during the weekend compared with the week (the average difference was 0.08 g/day per 1000 inhabitants). For some WWTPs, there were no clear differences between the COC equivalents calculated between the two days and this could be explained by a wide variety of factors. For instance, for Mouscron WWTP samples, we could not find a proper explanation, but WWTP samples from Oostende and Heist are susceptible to large variation during the 24-h sampling period. Due to expected cocaine intake, it means that the cocaine abuse on Friday is detected in the samples collected on Saturday. After intake, COC can be measured through the weekend (Hamilton et al., 1977; Jufer et al., 2000). Therefore, samples collected on Friday would still contain traces of BE originating from consumption throughout the weekend compared with the week (the average difference was 0.08 g/day per 1000 inhabitants).

Table 4 shows the concentrations of cocaine (COC) and benzoylecgonine (BE) in the waste water of several WWTPs in the Antwerp area. The highest BE concentrations were found in Antwerpen-Zuid and Deurne WWTPs compared to Antwerpen-Noord WWTP (Table 1). This is most probably due to two reasons: (i) these WWTPs receive waste water from the centre of Antwerp and (ii) a different social standard, more reported criminality and drug traffic in the suburbs whose waste water ends up in these WWTPs (Fig. SI-5). As expected, higher COC equivalents were calculated in the waste water samples from Antwerpen-Zuid and Deurne WWTPs compared to Antwerpen-Noord WWTP. For Brussel-Noord WWTP, four 24-h composite samples were collected within the same week, on Friday, Saturday, Sunday and the following Wednesday, to evaluate possible daily variations in the cocaine equivalents (Table 4). As expected, we observed a clear peak in the COC equivalents during the weekend. Since it takes approximately 24 h for water to run from the domestic site to a WWTP (personal communication, Alain Vandelannoote, Aquafin), it means that the cocaine abuse on Friday is detected in the samples collected on Saturday. After intake, COC can be measured in urine up to 6 h, while BE is excreted over a larger period (up to 96 h) (Hamilton et al., 1977; Jufer et al., 2000). Therefore, samples from Sunday would still contain traces of BE originating from cocaine abuse on Friday. This explains the increasing BE loads from the centre of Antwerp and from surrounding suburbs, while Antwerpen-Noord WWTP receives only waste water from the northern suburbs (Fig. SI-5).
throughout the weekend and the peak in COC loads observed on Saturday.

The concentrations and ratios of COC and BE observed in the present study are comparable to the results from Italian (Castiglioni et al., 2006; Zuccato et al., 2005, 2008), Spanish (Huerta-Fontela et al., 2007), and UK WWTPs (Kasprzyk-Hordern et al., 2008a,b; Zuccato et al., 2005) (Table 3). As hypothesized from the urinary excretion pattern, higher BE concentrations than COC concentrations were found in waste water. However, Bones et al. (2007) measured higher levels of COC than BE in Irish waste water, which conflicts with the normal expected pattern resulting from the metabolism of cocaine.

The mean \(\text{COC/BE} = 0.28\) and no outliers were observed. This ratio is comparable with the mean value (0.23) resulting from averaging the percentages of urinary excretion of BE and COC (Ambre et al., 1984, 1988; Cone et al., 1998). The calculated RSD (26%) is a consequence of the broad ranges of urinary excretion of COC and BE reported in the literature and of the differences in the stability of COC and BE in aquatic systems (Gheorghe et al., 2008). Yet, the RSD for waste water is lower than for surface water, indicating that for surface water, more factors, such as weather conditions, time of residence and turbidity, are responsible for the observed variations. The ratio COC/BE in waste water was higher than in surface water. The longer presence of COC and BE in surface water and the instability of COC in water are

Table 3
Concentrations of BE and COC of other studies

<table>
<thead>
<tr>
<th>Place</th>
<th>Type</th>
<th>Conc. BE (ng/L)</th>
<th>Conc. COC (ng/L)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Olona, IT</td>
<td>Surface</td>
<td>183</td>
<td>44</td>
<td>Zuccato et al. (2008)</td>
</tr>
<tr>
<td>Lambro, IT</td>
<td>Surface</td>
<td>50</td>
<td>15</td>
<td>Zuccato et al. (2008)</td>
</tr>
<tr>
<td>Po, IT</td>
<td>Surface</td>
<td>4</td>
<td>1</td>
<td>Zuccato et al. (2008)</td>
</tr>
<tr>
<td>Arno, IT</td>
<td>Surface</td>
<td>22</td>
<td>2</td>
<td>Zuccato et al. (2008)</td>
</tr>
<tr>
<td>Thames, UK</td>
<td>Surface</td>
<td>13</td>
<td>4</td>
<td>Zuccato et al. (2008)</td>
</tr>
<tr>
<td>Po, IT</td>
<td>Surface</td>
<td>25</td>
<td>1</td>
<td>Zuccato et al. (2005)</td>
</tr>
<tr>
<td>Taff, UK</td>
<td>Surface</td>
<td>78</td>
<td>2</td>
<td>Kaspzyk-Hordern et al. (2008a)</td>
</tr>
<tr>
<td>Ely, UK</td>
<td>Surface</td>
<td>23</td>
<td>&lt;0.3</td>
<td>Kaspzyk-Hordern et al. (2008b)</td>
</tr>
<tr>
<td>Broadmeadow, RI</td>
<td>Surface</td>
<td>nd</td>
<td>25</td>
<td>Huerta-Fontela et al. (2007)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Place</th>
<th>Type</th>
<th>Conc. BE (ng/L)</th>
<th>Conc. COC (ng/L)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Llobregat, ES</td>
<td>Surface</td>
<td>77</td>
<td>6</td>
<td>Present study</td>
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</tbody>
</table>

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<td>Surface</td>
<td>77</td>
<td>6</td>
<td>Present study</td>
</tr>
</tbody>
</table>

IT: Italy, UK: United Kingdom, RI: Republic of Ireland, ES: Spain, BE: Belgium, CH: Switzerland.

Table 4
Results for WWTP Brussels-Noord

<table>
<thead>
<tr>
<th>Day</th>
<th>Conc. BE (ng/L)</th>
<th>Conc. COC (ng/L)</th>
<th>Loads BE (g/day)</th>
<th>Loads COC (g/day)</th>
<th>COC eq (g/day)</th>
<th>COC eq/1000 inh. (g/day)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fri 14/12/2007</td>
<td>856</td>
<td>221</td>
<td>232.3</td>
<td>59.9</td>
<td>601.3</td>
<td>0.707</td>
</tr>
<tr>
<td>Sat 15/12/2007</td>
<td>1157</td>
<td>362</td>
<td>292.7</td>
<td>91.5</td>
<td>773.5</td>
<td>0.910</td>
</tr>
<tr>
<td>Sun 16/12/2007</td>
<td>1291</td>
<td>348</td>
<td>327.0</td>
<td>88.0</td>
<td>849.9</td>
<td>0.999</td>
</tr>
<tr>
<td>Wed 19/12/2007</td>
<td>909</td>
<td>274</td>
<td>222.7</td>
<td>66.8</td>
<td>585.6</td>
<td>0.689</td>
</tr>
</tbody>
</table>

BE = benzoylecgonine, COC = cocaine, COC eq = cocaine equivalents, inh = inhabitants.
possibly an explanation for this observed difference. Such difference between the COC/BE ratios in waste and surface water samples agrees with the difference observed by Zuccato et al. (2005).

4. Conclusions

The present study offers for the first time a deeper insight in the daily/weekly variation of COC and its major metabolite, BE, in a large number of waste and surface water samples collected throughout whole Belgium. The detection of COC and BE in all waste water samples indicates a widespread abuse and emphasizes that such monitoring is important to get a better view on the growing issue of cocaine. The present approach of estimating cocaine use could be executed on a regular basis for short term (monthly) to long term (yearly) monitoring and gives an immediate and clear picture of the geographical variation in cocaine abuse. This information will ultimately serve to setup prevention campaigns in the regions with high cocaine abuse, while the regular analysis of waste water can quickly evaluate the effect of these prevention campaigns. Future assessments of cocaine abuse through analysis of waste water should include more accurate mathematical models which need to include various factors such as urinary excretion profiles, stability of COC and metabolites in water and adsorption on solid particles.

Acknowledgements

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Appendix. Supplementary data

Results of waste water samples taken on Wednesdays are visualised in Fig. SI-1. Sampling locations and results of surface water are presented in Fig. SI-2. Details and results of the method validation and the interlaboratory test are described in Tables SI-1 and SI-2, respectively. Quality control charts for COC and BE are given in Figs. SI-3 and SI-4, respectively. Fig. SI-5 gives a geographical situation of the three sample WWTPs from Antwerp. Supplementary data associated with this article can be found in the online version at doi:10.1016/j.envpol.2008.07.020.

References


