

TECHNICAL NOTE**CRIMINALISTICS**

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Occurrence of Contamination by Controlled Substances in Euro Banknotes from the Spanish Archipelago of the Canary Islands

ABSTRACT: The social problems of drug abuse are a matter of increasing global problem. Nowadays, international agencies need fresh methods to monitor trends of the use of illicit drugs. In this sense, small amounts of drugs are transferred to banknotes and they could be detected and quantified. An analytical procedure based upon extraction with organic solvent, liquid chromatography separation, and mass spectrometric detection allowed the identification of 21 drugs and metabolites in 120 used Euro banknotes collected in the Canary Islands (Spain). Most of the banknotes analyzed showed detectable drug residues (92.5%). Cocaine was the most frequently detected drug, present in approximately 90% of the samples. In addition, 75%, 35%, and 15% of the banknotes showed residues of amphetamine derivatives, opiates, and benzodiazepines, respectively. An average of three drug residues per banknote was detected. In summary, the presence of drug residues in banknotes could be useful as tracer for drugs prevalence.

KEYWORDS: forensic science, drugs of abuse, controlled substances, banknotes, tourist region, Canary Islands

Spain is one of the countries that shows high rates of consumption of certain drugs of abuse. Although cannabis is still the most commonly abused illicit drug by Spanish society, available official statistics also show that cocaine has overtaken amphetamine derivatives and opiates to become the second most commonly abused illicit drug in the country (1). In fact, Spain, as reported by the United Nations Office on Drugs and Crime (2), is the second country in the world in terms of cocaine abuse. However, the above-mentioned statistics are thought to greatly underestimate the actual levels of illicit drug usage within the community, because they do not include the usage data of recreational users, mainly in tourist areas, where foreign citizens are not normally enrolled in the surveys conducted to assess the consumption of controlled substances (3). Because of these circumstances, higher rates of abuse of recreational drugs than those discovered by statistics are expected in tourist areas, such as the Canary Islands archipelago.

Direct measurements of abuse drugs prevalence are unavailable (4). Because of the fact that the most misused drugs in a region are also the most detected ones in the environment, it has been proposed the measurement of abuse drugs and their byproducts in environmental samples as a suitable tool for this purpose. Thus, in past years, measurement of drugs in surface and waste waters has been allowed (5). Other proposed ways to monitor drug prevalence are the number of hospital recoveries or clinical treatments, and the amount of drugs seized by police forces. Because of the fact that banknotes are routinely involved in drug deals and, even in drug

use, they seem to be an appropriate sample to evaluate the prevalence of abuse drugs in a region.

The presence of drugs of abuse on Euro banknotes has been well established (6,7), and various techniques to detect or quantify the contamination of banknotes have been described previously in literature (6–12). Although banknotes are likely to become contaminated during drug deals, the major source of contamination is considered to be the use of banknotes by drug users, who roll them up to snort the illicit substances. As the paper currency is counted in banks mainly using machines, these may also become contaminated when coming in contact with heavily contaminated banknotes; these then serve to transfer small amounts of drugs to subsequent ones. In this manner, banknotes become contaminated by any type of drug that is used in a given region or country.

As cocaine is the most commonly used illicit drug in Western countries and is consumed mainly by snorting through rolled-up paper currency, most studies have focused on the presence of cocaine in banknotes. However, multidrug ingestion appears to be common among drug abusers in Spain (1,2). Consequently, studies analyzing the presence of other abuse drugs in banknotes are essential.

To assess the potential utility of the analysis of drug residues in banknotes as an indirect tool to evaluate the prevalence of consumption of abuse drugs in a region, 120 Euro banknotes were collected from the Canary Islands, a Spanish tourist region, and analyzed by liquid chromatography with tandem mass spectrometry (LC-MS/MS) to determine the presence of 21 abuse drugs and metabolites.

Material and Methods

Study Area

The Spanish archipelago of the Canary Islands is located in the Atlantic Ocean, about 100 km away from the nearest point of the

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North African coast (southwest of Morocco). Geographically, the Islands are part of the African continent; yet, from a historical, economic, political, and sociocultural point of view, the Canaries are completely European. The archipelago consists of seven major islands, with a resident population of almost 2 million inhabitants. The economy of the Canary Islands archipelago is mainly based on tourism, these islands being one of the favorite tourist destinations for European citizens (approximately 9 million visitors annually). As in other tourist areas, in the Canary Islands also, there exists a high rate of consumption of recreational drugs (1,3,13).

Sampling Design

Samples, consisting of 120 used Euro banknotes analyzed in this study (30 each of 50-, 20-, 10-, and 5-€ banknotes), were randomly collected in the main tourist area of the archipelago, Playa de Maspalomas (located south of the Gran Canaria island), from the automated teller machines of four different branches of banks over a 5-day period in April 2009. Because automated teller machines use banknotes received from regional counting centers, these banknotes are more likely to be representative of the paper currency in the archipelago (10). After collection, the banknotes were placed in plastic storage bags until analysis.

Reagents

Standards of cocaine, cocaethylene, heroin, methadone, morphine, 6-acetylmorphine, oxymorphone, hydromorphone, hydrocodone, codeine, amphetamine, methamphetamine, *n*-ethyl amphetamine, methylenedioxyamphetamine (MDMA), 3,4-methylenedioxyamphetamine (MDA), *N*-ethyl-MDA (MDEA), 3,4-(methylenedioxyphenyl)-2-butanamine hydrochloride (BDB), *N*-methyl-1-(3,4-methylenedioxyphenyl)-2-butanamine hydrochloride (MBDB), *d*-lysergic acid diethylamide (LSD), diazepam, and alprazolam were from Cerilliant (Round Rock, TX). Ammonium formate, Formic acid (99%), and all LC-grade solvents were from Sigma-Aldrich (Manheim, Germany). Deionized water was generated from Elix 5UV and Milli-Q Gradient A10 water-purification systems (Millipore Corp., Billerica, MA).

Sample Preparation and Analysis

The Euro banknotes were subjected to the nondestructive extraction method described by Jenkins (8), and the extracts were analyzed by LC-MS/MS.

Equipment

Chromatographic analyses were carried out using an Accela ultra high-performance LC system, and data were acquired using a Quantum Access triple-stage quadrupole tandem mass spectrometer (both apparatuses were from Thermo Fisher Scientific Inc., San Jose, CA).

Preparation of Samples

Briefly, Euro banknotes were individually placed inside 15-mL, conical, glass centrifuge tubes, and 5 mL of acetonitrile was added. Each tube was capped and agitated by vortexing for 6 × 30-sec periods over 2 h. After 2 h, the banknotes were removed, and the solvent was filtered through 0.45- μ m polyethylene terephthalate filters (Chromafil PET-45/25; Macherey-Nagel, Düren, Germany). To improve the limit of detection of the method, the filtered

solvent was evaporated under a gentle nitrogen stream to near-dryness. The dried extract was then reconstituted in 0.5 mL of initial mobile phase and used for chromatographic analysis.

Chromatographic Conditions

Liquid chromatography separation was performed on a Hypersil GOLD PFP (perfluorophenyl) 5- μ m column of dimensions 50 × 2.1 mm (Thermo Fisher Scientific Inc.). The mobile phases were: (A) water, 0.1% formic acid, and 10 mM ammonium formate as the aqueous phase, and (B) acetonitrile and 0.1% formic acid as the organic phase. The flow rate was 200 μ L/min, and the injection volume was 10 μ L for all experiments. The separation run was completed in 30 min at 30°C in a gradient program, as follows: 0–5 min: 95% A; 5–20 min: 95% A → 5% A; 20–25 min: 5% A; 25–30 min: 5% A → 95% A; 5–20 min: 95% A → 5% B; 20–25 min: 5% A; and 25–30 min: 5% A → 95% B.

MS/MS Parameters

The mass spectrometer and the electrospray ionization source worked with the following parameters: skimmer offset (4 V), sheath gas [8 arbitrary unit (au)], auxiliary gas (8 au), and capillary temperature (275°C), operating in positive mode with ion sweep cone gas (4 au), vaporizer temperature (200°C), and spray voltage (5000 V).

The target analytes were directly introduced into the ion source, and the collision energies of the MS/MS transitions were optimized for each of them. Table 1 lists the selective reaction monitoring (SRM) transitions used for the identification and quantification of all compounds. The peak width was 0.7 Da both in the collision cells Q1 and Q3, and the argon pressure in Q2 was set to 0.002 mbar. For the LC-MS/MS analysis, all SRM transitions were acquired in only one segment. Quantitation of analytes was conducted using external calibration.

Linearity was assessed from prepared five point calibration curves in the region of 0.005–5 mg/L equating to 0.1–100 ng/banknote. Repeatability was determined by performing six replicate injections of a 0.10 mg/L mixed standard solution while reproducibility was examined using six individually prepared 0.5 mg/L mixed analyte solutions. The limits of detection (LOD) and quantitation (LOQ) were defined as signals corresponding to the values that give a signal-to-noise ratio of 3 and 10, respectively. These values for each analyte are also listed in Table 1. Linearity was demonstrated in all cases with $r^2 > 0.99$. The calculated levels of precision were also deemed acceptable: in general, repeatability was <5% relative standard deviation (RSD), while reproducibility was in the range of 5–10% RSD. The method was also determined to be suitably sensitive with detection and quantitation limits in the pg-ng/banknote range.

Statistical Analysis

Standard descriptive statistics (mean values and standard deviations; medians and percentiles 25 and 75; and frequencies) were calculated for the levels of drugs in the banknotes. In the current sample, the distribution of drugs in the banknotes deviates significantly from normality. Hence, nonparametric tests were applied. To compare the levels of illicit drugs (or the groups of drugs) between banknotes, the Mann-Whitney test was used. Spearman's correlation coefficients were used to calculate the associations among drug levels. To compare the number of residues of drugs found in any Euro banknote of the four types of analyzed denominations

TABLE 1—Analytical features of the drug determination by liquid chromatography with tandem mass spectrometry.

Analyte	Precursor Ion (m/z)	Product Ion (m/z) Used for Quantitation	Collision Energy (V)	Product Ion (m/z) Used for Confirmation	Collision Energy (V)	Limits of Detection (pg/Banknote)	Limits of Quantitation (pg/Banknote)
Cocaine	304.3	182.1	17	82.3	27	3	10
Cocaethylene	318.4	196.2	19	82.3	29	5	18
Heroin	370.4	268.2	28	165.1	45	3	10
Methadone	310.4	265.3	15	135.1	20	4	14
Morphine	286.3	201.2	26	165.2	36	8	28
6-Acetylmorphine	328.4	165.1	36	211.1	26	4	14
Oxymorphone	302.3	284.3	19	227.2	28	10	24
Hydromorphone	286.2	185.1	30	157.2	38	3	10
Hydrocodone	300.3	199.1	30	171.1	36	3	10
Codeine	300.3	165.1	39	215.2	25	5	18
Amphetamine	136.3	91.3	17	65.4	35	60	210
Metamphetamine	150.3	91.3	17	65.4	38	40	140
<i>n</i> -Ethyl amphetamine	208.3	135.1	16	77.3	38	50	175
Methylenedioxyamphetamine (MDMA)	194.2	135.1	20	133.2	19	3	10
3,4-methylenedioxyamphetamine (MDA)	180.2	135.1	17	105.2	21	3	10
<i>n</i> -Ethyl-MDA (MDEA)	208.3	163.2	12	135.1	22	3	10
3,4-(Methylenedioxyphenyl)-2-butanamine hydrochloride (BDB)	235.3	135.1	22	77.3	48	15	55
<i>n</i> -Methyl-1-(3,4-methylenedioxyphenyl)-2-butanamine hydrochloride (MBDB)	208.3	135.1	18	77.3	37	15	55
d-Lysergic acid diethylamide (LSD)	324.4	223.2	23	207.2	43	8	28
Diazepam	285.4	193.1	30	154.1	25	3	10
Alprazolam	309.3	281.2	26	205.1	39	8	28

(5-, 10-, 20-, and 50-€ banknotes), Student's *t*-tests were used. A value of $p < 0.05$ (two-tailed) was considered to be statistically significant. Database management and statistical analysis were carried out with the software PASW v 17.0 (SPSS Inc., Chicago, IL).

Results

Most of the analyzed banknotes showed detectable levels of drug residues. Indeed, only one 5 € banknote did not show any drug contamination. An average presence of three drug residues per banknote (range: 0–7) was detected. Banknotes of higher denomination (10-, 20-, and 50-€) also showed statistically significant higher number of drug residues than the low-denomination banknotes (5 €). Thus, 5-€ denomination banknotes showed a mean level of 1.9 drug residues per banknote, whereas 10-, 20-, and 50-€ banknotes showed mean levels of 3.5 ($p = 0.048$), 3.6 ($p = 0.028$), and 3.4 ($p = 0.049$) residues per banknote, respectively.

As shown in Table 2, cocaine was the most frequently detected drug. More than 87% of banknotes showed cocaine levels above the LOQ. Additionally, many other samples ($n = 37$) were also positive for the cocaine metabolite, cocaethylene. Thus, considering the sum of the amounts of cocaine and cocaethylene (Σ cocaine), the current results showed that 92.5% of the analyzed banknotes had measurable levels of cocaine derivatives.

Regarding opiates, although morphine and oxymorphone were not detected in any banknote analyzed, methadone, heroin, hydromorphone, and hydrocodone were detected in 12, 9, 3, and 6 Euro banknotes, respectively. 6-Acetylmorphine was also detected in 13 banknotes in which no heroin residues were present. In addition, considering the sum of all the opiates measured as Σ opiates, the results showed that 42 banknotes (35%) had measurable residues of these drugs (Table 2).

Other illicit drugs that were detected in a significant number of banknotes were amphetamine and methamphetamine (32.5% and 45% of the analyzed banknotes, respectively). Furthermore, although

a number of “designer drugs,” such as BDB or MBDB, were not detected in any sample, other “designer drugs,” such as MDMA, MDA, and MDEA, were detected in a small percentage of samples (12.5%, 12.5%, and 7.5%, respectively). It should be highlighted that 75% of the banknotes showed detectable levels of some type of amphetamine–methamphetamine derivative (Σ amphetamines; Table 2).

The hallucinogen LSD was not detected in any sample.

In this work, the authors have also included the evaluation of prescription drugs frequently misused by drug consumers, such as benzodiazepines. Our findings showed that residues of diazepam and alprazolam were detected in a number of banknotes. In fact, several types of benzodiazepines (Σ benzodiazepines) were measured in 15% of the analyzed banknotes (Table 2).

Our findings indicated that most of the banknotes showed contamination by illicit drugs at trace levels, with the exception of cocaine. Thus, cocaine residues showed median levels nearly equal to 190 ng/banknote, whereas the remaining drugs detected showed median levels of 0.00 ng/banknote. Nevertheless, it should be highlighted that Σ methamphetamine residues were quantified at levels of 1.41 ng/banknote in the 75th percentile, and that Σ amphetamines reached levels of 2.93 ng/banknote in the same percentile (Table 2).

In any case, in spite of the fact that the sample with the highest value of cocaine residue (15023.09 ng/banknote) was a 5-€ banknote, 30% of the 5-€ banknotes did not contain detectable values of cocaine (data not shown). On the contrary, 100% the higher-denomination banknotes (50-, 20-, and 10-€) had measurable residues of cocaine (data not shown). Statistically significant differences were found between the cocaine levels quantified in 50-, 20-, and 10-€ banknotes and the cocaine levels present in 5-€ banknotes ($p = 0.003$, $p = 0.002$, and $p = 0.004$, respectively).

Furthermore, numerous statistically significant associations were found between the drug residues measured. Thus, there was a positive association between the residues of cocaine and cocaethylene

TABLE 2—Drug determination in Euro bank notes by liquid chromatography with tandem mass spectrometry.

	Mean (SD)	Median (p25–p75)	D (%)	ND (%)	Minimum/Maximum
Cocaine	955.22 (2671.78)	188.48 (25.75–473.29)	105 (87.5)	15 (12.5)	0.00/15,023.09
Cocaethylene	0.31 (1.32)	0.00 (0.00–0.00)	37 (22.5)	93 (77.5)	0.00/8.10
∑Cocaine	955.53 (2673.10)	188.48 (25.75–473.42)	111 (92.5)	9 (7.5)	0.00/15,041.02
Methadone	0.26 (1.16)	0.00 (0.00–0.00)	12 (10)	108 (90)	0.0/7.2
Heroin	3.85 (23.79)	0.00 (0.00–0.00)	9 (7.5)	111 (92.5)	0.00/150.60
6-Acetylmorphine	0.08 (0.25)	0.00 (0.00–0.00)	13 (11.0)	107 (89)	0.00/1.10
Hydromorphone	0.01 (0.34)	0.00 (0.00–0.00)	3 (2.5)	117 (97.5)	0.00/0.20
Hydrocodone	0.01 (0.25)	0.00 (0.00–0.00)	6 (5)	114 (95)	0.00/0.10
Codeine	0.02 (0.1)	0.00 (0.00–0.00)	6 (5)	114 (95)	0.00/0.50
∑Opiates	4.22 (25.08)	0.00 (0.00–0.00)	42 (35)	108 (65)	0.00/158.88
Amphetamine	1.38 (3.18)	0.00 (0.00–1.01)	39 (32.5)	81 (67.5)	0.00/13.90
Metaamphetamine	0.91 (1.46)	0.00 (0.00–1.41)	54 (45)	54 (55)	0.00/5.50
MDMA	0.01 (0.06)	0.00 (0.00–0.00)	15 (12.5)	105 (87.5)	0.00/0.20
MDA	0.02 (0.06)	0.00 (0.00–0.00)	15 (12.5)	105 (87.5)	0.00/0.90
MDEA	0.01 (0.01)	0.00 (0.00–0.00)	9 (7.5)	111 (92.5)	0.00/0.10
∑Amphetamine-derivatives	2.40 (3.91)	0.55 (0.16–2.93)	90 (75)	30 (25)	0.00/18.56
Diazepam	0.01 (0.78)	0.00 (0.00–0.00)	9 (7.5)	108 (90)	0.00/1.40
Alprazolam	0.17 (0.78)	0.00 (0.00–0.00)	12 (10)	108 (90)	0.00/4.80
∑Benzodiazepines	0.22 (0.84)	0.00 (0.00–0.00)	18 (15)	102 (85)	0.00/5.04

D, detectable; ND, nondetectable.

($r = 0.501$; $p = 0.001$). Interestingly, positive associations between cocaine and amphetamine ($r = 0.261$; $p = 0.038$) and between cocaine and 6-acetylmorphine ($r = 0.352$; $p = 0.026$) were also evident. In addition, a positive association was found between ∑cocaine and ∑amphetamines ($r = 0.352$; $p = 0.026$), and between ∑cocaine and ∑benzodiazepines ($r = 0.421$; $p = 0.007$).

Opiates were also detected at trace levels, although it should be highlighted that in one 5-€ banknote, 150.6 ng of heroin was determined.

With reference to methamphetamine, although only 45% of the banknotes showed residues of this drug, it was mainly detected in the highest-denomination banknotes (50-, 20-, and 10-€). Approximately 70% of the 20-€ banknotes were positive for this drug, and the highest level of contamination by methamphetamine (5.50 ng/banknote) was found in a 20-€ banknote. Similar to the results obtained for cocaine, methamphetamine levels in 50-, 20-, and 10-€ banknotes were higher than methamphetamine levels shown by 5-€ banknotes ($p = 0.036$, $p = 0.004$, and $p = 0.036$, respectively).

In the case of amphetamine, the situation was similar. The highest-denomination notes were more frequently contaminated by this drug (40% for 50-€ notes, 60% for 20-€ notes, and 30% for 10-€ notes), whereas no banknote of 5-€ showed detectable residues of amphetamine. Indeed, a 50-€ banknote had 13.90 ng of amphetamine. In addition, statistically significant differences were found between the amphetamine levels in 50- and 20-€ banknotes and the levels in 5-€ banknotes ($p = 0.031$ and $p = 0.005$, respectively). As cited previously, a positive association between amphetamine and cocaine was evident ($r = 0.261$; $p = 0.003$), also in addition to a positive one between ∑cocaine and ∑amphetamines ($r = 0.352$; $p = 0.026$).

Interestingly, numerous banknotes showed detectable residues of benzodiazepines. Thus, nine banknotes showed residues of diazepam, and 12 showed measurable residues of alprazolam. Although benzodiazepines were measured at trace levels, it should be highlighted that 4.80 ng of alprazolam were found in a 20-€ banknote.

Discussion

The presence of residues of several drugs of abuse in Euro banknotes has been well documented (6,7). Banknotes are likely to be

contaminated when they come in contact with significant quantities of drugs, especially when handled by drug users (10). These contaminated banknotes are then distributed throughout the banking system. Thus, counting machines can pass detectable amounts of drugs from heavily contaminated banknotes to those subsequently counted (14). However, it is not clear whether only contamination within the counting machines can explain the widespread occurrence of low levels of illicit drugs on banknotes (14). Although to a lesser degree, the presence of trace levels of illicit drugs on banknotes may well be because of the transfer of drugs by handling. Because automated teller machines use banknotes received from regional counting centers (10), the existence of drug residues on banknotes from automated teller machines could help to identify the regional prevalence of abuse drugs. From this point of view, the extent of drug contamination in banknotes could possibly reflect trends in illicit drug usage in a region.

Our findings agree with previously published studies (6–12) in that cocaine is one of the most extensively abused illicit drugs in the world, and that most of the paper currency in general circulation is contaminated with cocaine (8). As previously cited, Spain has had the highest abuse rates for cocaine in Europe for the last decade and rates higher than U.S.A. in recent years (2). Furthermore, in Europe, the U.K. and Germany (countries that provide the highest number of tourists to the Canary Islands each year) also show high prevalence rates of cocaine (2). Thus, in agreement with these data, 82.5% of the Euro banknotes from the Canary Islands analyzed in this work showed detectable residues of cocaine, and this percentage increased to 92.5%, if we also include the cocaine derivative cocaethylene present in the banknotes.

Other authors have established a cut-off point of 100 ng of cocaine per banknote as the background level of the drug on banknotes to distinguish the banknotes in general circulation from those that have been directly in contact with the drug (used both in drug deals or to snort) (10). The assumption of this cut-off point would imply that of the 120 Spanish Euro banknotes analyzed in this work, 75 were actually used for snorting and/or drug dealing (62.5%). It could be possible that, as a consequence of the increasing rates of cocaine usage reported in the preceding decades in Spain, the real cut-off point should be even higher. In any case, applying the value of the median obtained for cocaine in this work (188.48 ng/banknote) as the cut-off point, 50% of the herein-analyzed Spanish Euro

banknotes showed values higher than the median, pointing to the possibility that they had been in close contact with the drug (involved in drug deals or even snorting). Although it appears that the 5-€ banknote with the highest value of cocaine (15,023.09 ng/banknote) was used to snort the drug, our findings indicate that the banknotes more frequently involved in drug deals or for snorting are the higher-denomination ones (10-, 20-, and 50-€ banknotes).

It should be also highlighted that nearly 23% of the analyzed banknotes showed detectable levels of cocaethylene. As far as we know, this is the first time that this metabolite has been investigated in banknotes. Cocaethylene is a cocaine byproduct that can be formed if the cocaine cooks use ethanol to purify the cocaine base, to processing cocaine base to cocaine HCl via ethanolic HCl, or when cocaine HCl is dissolved in ethanol for smuggling purposes (15). But cocaethylene can also be a metabolite formed in the liver of users who have consumed cocaine and ethanol simultaneously. Nowadays, it seems evident that cocaine users frequently also are alcohol abusers (1,16), increasing the formation of cocaethylene and its potential transfer to bills.

Amphetamine, methamphetamine, and “designer drugs” were detected in a large number of banknotes. The Canary Islands Government statistics (13) indicate that there is a high ratio of prevalence of these drugs (among illicit drugs, only cannabis and cocaine are present in higher ratios than amphetamine and its derivatives) in the Canary Islands. Our findings agree with these data because most banknotes showed detectable residues of these drugs. The presence of high levels of amphetamine and/or methamphetamine in a number of Euro banknotes (13.90 and 11.70 ng of amphetamine in a 50- and a 20-€ banknote, respectively) appear to indicate that, in spite of the fact that these drugs are mainly consumed by oral ingestion, they were also consumed by smoking or snorting. As in the case of cocaine, our findings appear to indicate that the banknotes more frequently involved in drug deals or snorting are the highest-denomination ones (10-, 20-, and 50-€ banknotes).

In our study, 20% of the banknotes showed detectable residues of heroin or its main metabolite 6-acetylmorphine, indicating an illicit use of heroin, whereas only 10% of the Euro banknotes showed residues of methadone (opiate legally used in the methadone-maintenance programs). As previously cited in 2007, the Spanish Government statistics reported a very low prevalence rates of consumption of opiates (0.12%) in the Spanish population (15–64 years) (1). However, 35% of the analyzed banknotes showed the presence of residues of opiates. The possibility exists that the level of contamination of paper currency in the Canary archipelago by these drugs could be influenced by the fact that the Canary Islands receives hundreds of thousands of visitors from the U.K. every year (a country that shows high ratios of prevalence of opiates) (2).

In our opinion, the fact that 15% of the analyzed samples showed detectable residues of benzodiazepines is very interesting. This result agrees with the epidemiological studies indicating that the prevailing abuse rates of these drugs are very high in Spain (1).

Currently, the abuse of multiple illicit drugs among drug abusers is the most frequently encountered condition all over the world (1,17,18). Our findings showing a positive association between \sum cocaine and \sum benzodiazepines, between \sum cocaine and \sum amphetamines, and between cocaine and 6-acetylmorphine point to the possibility that multiple drug abuse is very common among recreational drug abusers, especially among amphetamine and cocaine abusers, in these islands. Additionally, the number of drug residues quantified increased as the values of the banknotes

increased. Such a result may well indicate that higher-value banknotes are more frequently involved in drug deals than lower-value banknotes (such as 5-€ banknotes).

There are some limitations to the main objective of this study. It is evident that any Euro banknote could be freely in circulation throughout the entire Euro zone. Such circumstances could make it difficult to obtain epidemiological conclusions about any specific region of the Euro zone through analyses of paper currency. However, this study is sufficiently large and has been conducted in a relatively isolated geographical region (the Canary archipelago), increasing the possibility that most of the analyzed banknotes originate from the regional counting centers (10). In addition, the analytical methods applied in this work allowed the quantification of a large number of drug residues and metabolites. As cited earlier, the most misused drugs in a region are also the most detected ones in any environmental sample, including banknotes. Thus, this type of study may well contribute to evaluate the prevalence of abuse drugs in a region. In this sense, the detection of an controlled substance, which is not frequently consumed by a population, in a high percentage of banknotes could indicate that the specific drug is an emerging menace or that it has remained unnoticed in epidemiological surveys.

In conclusion, the pattern of contamination of Euro banknotes in circulation in the Canary Islands confirms the date of prevalence of abuse drugs reported previously by epidemiological studies. It is possible that examination of paper currency from countries with a different drug abuse problem will demonstrate a varying distribution of contamination. Considering that, currently, multidrug analysis of banknotes is a fast and feasible methodology, our findings are significant enough to allow us to state that the analyses of drug residues in paper currency may be a useful tool to evaluate the prevalence of abuse drugs or to detect the presence of new emerging drugs in a region.

References

1. Observatorio Español sobre Drogas. Plan Nacional de Drogas. Gobierno de España, 2007, <http://www.pnsd.msc.es/Categoria2/publica/pdf/oed-2007.pdf> (accessed November 22, 2009).
2. United Nations Office on Drugs and Crime. World Drug Report 2009, <http://www.unodc.org/> (accessed January 20, 2010).
3. Bellis MA, Hughes K, Calafat A, Juan M, Schnitzer S. Relative contributions of holiday location and nationality to changes in recreational drug taking behaviour: a natural experiment in Balearic Islands. *Eur Addict Res* 2009;15:78–86.
4. Cecinato A, Balducci C, Nervegna G. Occurrence of cocaine in the air of the world's cities, an emerging problem? A new tool to investigate the social incidence of drugs? *Sci Total Environ* 2009;407:1683–90.
5. Zuccato E, Chiabrando C, Castiglioni S, Bagnati R, Fanelli R. Estimating community drug abuse by wastewater analysis. *Environ Health Perspect* 2009;116:1027–32.
6. Esteve-Turrillas FA, Armenta S, Moros J, Garrigues S, Pastor A, de la Guardia M. Validated, non-destructive and environmentally friendly determination of cocaine in euro bank notes. *J Chromatogr A* 2005; 1065:321–5.
7. Bones J, Macka M, Paull B. Evaluation of monolithic and sub 2 μ m particle packed columns for the determination of drug contamination on Irish euro banknotes. *Analyst* 2006;132:208–17.
8. Jenkins AJ. Drug contamination of US paper currency. *Forensic Sci Int* 2001;121:189–93.
9. Ebejer KA, Brereton RG, Carter JF, Ollerton SL. Rapid comparison of diacetylmorphine on banknotes by tandem mass spectrometry. *Rapid Commun Mass Spectrom* 2005;19:2137–43.
10. Ebejer KA, Lloyd GR, Brereton RG, Carter JF, Sleeman R. Factors influencing the contamination of UK banknotes with drugs of abuse. *Forensic Sci Int* 2007;171:165–70.
11. Ebejer KA, Winn J, Carter JF, Sleeman R, Parker J, Körber F. The difference between drug money and a “lifetime's savings.” *Forensic Sci Int* 2007;167:94–101.

12. Zuo Y, Zhang K, Jingping W, Rego C, Fritz J. An accurate and nondestructive GC method for determination of cocaine on US paper currency. *J Sep Sci* 2008;31:2444–50.
13. Dirección General de Atención a las drogodependencias. Consejería de Sanidad y Consumo. Gobierno de Canarias, 2007, <http://www2.gobiernodecanarias.org/sanidad/dgad/> (accessed December 15, 2009).
14. Carter JF, Sleeman R, Parry J. The distribution of controlled drugs on banknotes via counting machines. *Forensic Sci Int* 2003;132:106–12.
15. Casale JF. Cocaethylene as a component in illicit cocaine. *J Anal Toxicol* 2007;31:170–1.
16. Rubio G, Manzanares J, Jiménez M, Rodríguez-Jiménez R, Martínez I, Iribarren MM, et al. Use of cocaine by heavy drinkers increases vulnerability to developing alcohol dependence: a 4-year follow-up study. *J Clin Psychiatry* 2008;69:563–70.
17. Musshoff F, Trafkowski J, Lichterman D, Madea B. Comparison of urine results concerning co-consumption of illicit heroin and other drugs in heroin and methadone maintenance programs. *Int J Legal Med* 2010;124:499–503.
18. Black DL, Cawton B, Robert T, Moser F, Caplan YH, Cone EJ. Multiple drug ingestion by ecstasy abusers in the United States. *J Anal Toxicol* 2009;33:143–7.

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