Applied Radiation and Isotopes 135 (2018) 212–218

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Exploratory analysis for the identification of false banknotes using portable X-ray Fluorescence spectrometer

M.A. Zamalloa Jara[a,*], C. Luízar Obregón[b], C. Araujo Del Castillo[a]

[a] Academic Department of Physics – Faculty of Sciences – Universidad Nacional de San Antonio Abad del Cusco (UNSAAC), Av. de la Cultura, 733, Pabellón C-361, Cusco, Peru

[b] Academic Department of Chemistry – Faculty of Sciences – UNSAAC, Av. de la Cultura, 733, Pabellón LQ-201, Cusco, Peru

HIGHLIGHTS

- Falsification of currency banknotes is a quite frequent illegal activity.
- pXRF can also be used to identify illegal banknotes.
- pXRF has identified the presence of toxic elements in the false banknotes.

ARTICLE INFO

Keywords:
pXRF
Paper currency
Falsification
Security features
Fake

ABSTRACT

The aim of this study was to verify if a portable X-ray fluorescence (pXRF) spectrometer can recognize the security features in banknotes that are reproducible by counterfeiters. Peruvian Nuevo Sol banknotes were studied: 4 genuine and 3 fake ones, in 11 points of analysis for each one, at all 77 data set. The correlation analysis of spectra among original notes was 1.0, and there was no correlation with fake banknotes. pXRF prove that two security features were reproducible for counterfeiters.

1. Introduction

Falsification of currency banknotes is a frequent illegal activity. The falsification of Dollars (CNBC, 2016; Reuters NBC News, 2016; Rusanov et al., 2009), Euros (Cabitza, 2012; Europol, 2012) and other currencies has forced governments to elaborate ever more sophisticated security features (Dwan, 2002), as for example, the development of micro-printing, the use of fluorescent inks, plastic paper and the use of substances reactive against UV and IR, among others. In Peru, the latest false banknotes are very difficult to identify for most people (Franklin, 2016) because falsifiers can reproduce many of the security features established by Peru's Central Reserve Bank (BCRP) as are fluorescent images under UV light and chalcographic marks for touch recognition (BCRP, 2016). This problem affects low-income populations and tourists who are not familiar with the national money. Tourism is one of the most important financial income sources in Cusco, Peru.

Peruvian counterfeiters have managed to reproduce high quality forgeries of paper currency, one being the highest value banknote, the 200 Nuevos Soles bill, said to be the 'best fake'. Counterfeited banknotes present a texture very similar to that of a genuine banknote. They contain metallic threads with reflections and colorful fluorescent designs very similar to those expected to see in a UV detector for a genuine bill. Furthermore, the current development of printing technology allows the falsifiers to obtain a higher image quality, with details that are very similar to those present on genuine banknotes.

These and other aspects make the identification of fake banknotes difficult in Peru, and highlight the necessity of developing security marks that cannot easily be reproduced, and which, in turn, are easy to identify by common people. This problem also creates the need to develop new techniques that differentiate genuine banknotes from false ones.

In literature, the use of techniques to recognize fraudulent banknotes has been reported. For example, there is the use of image analysis for texture identification (Hassanpour and Farahabadi, 2009; Cao et al., 2012), intelligent recognition of paper currency systems based on image analysis (García-Lamont et al., 2012; Sargano et al., 2014), near infrared spectroscopy to characterize the cotton paper currency (Dale and Klatt, 1989), Raman spectroscopy to identify the organic nature of inks in banknotes (Jelovica Badovinac et al., 2010), and among others (Roy et al., 2015). The X-ray fluorescence spectroscopy (XRF) is a non-

* Corresponding author.
E-mail addresses: marco.zamalloa@unsaac.edu.pe (M.A. Zamalloa Jara), celina.luizar@unsaac.edu.pe (C. Luízar Obregón).

https://doi.org/10.1016/j.apradiso.2018.01.043
Received 12 January 2017; Received in revised form 22 December 2017; Accepted 31 January 2018
Available online 02 February 2018
0969-8043/

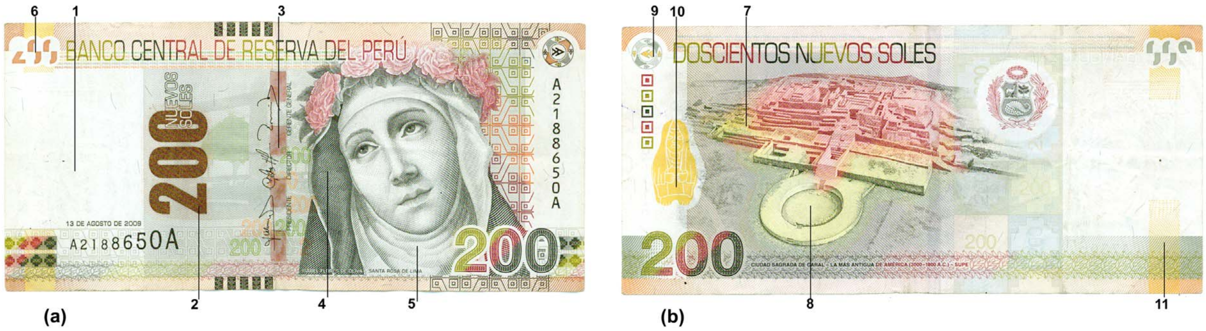

Fig. 1. pXRF analysis points for the two hundred Nuevos Soles banknotes, (a) front side, and (b) reverse side.

Table 1
Average intensity in beam 3 (Count / s) of the elements present in the original banknotes for each analysis point.

Mayor intensities			**Middle intensities**				
Point	Ca	Ti	Cl	Zn	Cu	Fe	K
1	31360 ± 3860	44375 ± 926	4079 ± 560	3800 ± 152	2087 ± 111	1941 ± 261	1433 ± 558
2	31728 ± 5170	38686 ± 689	3950 ± 295	3845 ± 218	2247 ± 61	2086 ± 238	1345 ± 425
3	42480 ± 7253	43742 ± 1590	6072 ± 427	3651 ± 196	2124 ± 151	2199 ± 278	1117 ± 258
4	51857 ± 8260	38234 ± 1491	3745 ± 273	3633 ± 93	2928 ± 107	2149 ± 239	1494 ± 442
5	37424 ± 5596	39448 ± 1363	4437 ± 237	3734 ± 127	2397 ± 106	1987 ± 216	1174 ± 383
6	36408 ± 4866	39399 ± 1579	4971 ± 518	3665 ± 49	2021 ± 35	1724 ± 191	1102 ± 306
7	42836 ± 8005	37371 ± 568	4147 ± 356	3669 ± 154	2303 ± 99	1915 ± 304	1158 ± 411
8	37394 ± 6720	38130 ± 1119	4502 ± 226	3741 ± 153	2301 ± 91	2007 ± 301	1209 ± 426
9	36009 ± 4332	37907 ± 1298	4784 ± 246	3646 ± 69	2116 ± 59	2034 ± 473	1291 ± 278
10	35404 ± 4922	39064 ± 543	4577 ± 408	3704 ± 144	2085 ± 84	2057 ± 409	1304 ± 566
11	35552 ± 5738	37578 ± 2021	4429 ± 510	3514 ± 172	2240 ± 120	1978 ± 162	1312 ± 462

	Lower intensities				
Point	**Sr**	**Mn**	**Zr**	**Cr**	**S**
1	702 ± 25	362 ± 8	206 ± 8	142 ± 18	120 ± 13
2	722 ± 26	400 ± 33	215 ± 3	139 ± 9	157 ± 29
3	681 ± 21	419 ± 68	216 ± 11	136 ± 22	147 ± 14
4	716 ± 35	594 ± 173	224 ± 2	140 ± 13	188 ± 8
5	719 ± 33	431 ± 72	225 ± 1	147 ± 5	145 ± 7
6	686 ± 35	346 ± 16	202 ± 5	127 ± 12	127 ± 13
7	687 ± 16	442 ± 83	203 ± 4	137 ± 7	145 ± 10
8	717 ± 41	395 ± 39	205 ± 5	131 ± 16	141 ± 9
9	736 ± 16	379 ± 16	199 ± 15	133 ± 10	136 ± 9
10	695 ± 12	398 ± 60	214 ± 3	132 ± 16	130 ± 20
11	661 ± 31	415 ± 62	199 ± 20	132 ± 7	125 ± 17

destructive technique that allows simultaneous identification of several elements, in short time (Cesareo et al., 2007). XRF was used for the identification of lead in American Dollar banknotes, as a drying agent, before the 90 s (Nir-El, 1994), and recently, to compare dyes used in stamps and one Lira banknotes, in Croatia. Appoloni and Melquiades (Appoloni and Melquiades, 2014) also showed that portable the X-ray fluorescence spectroscopy (pXRF) is a useful non-invasive technique, in the study of paper currency. In this sense, the present study intends to demonstrate that pXRF can also be used to identify fake Nuevos Soles banknotes and recognize the security features in banknotes that are reproducible by counterfeiters.

2. Material and methods

The analyzed samples were collected in the city of Cusco, and correspond to the 200 Nuevos Soles banknotes in circulation (BCRP, 2015). It is the highest denominator banknote in circulation, for which reason also makes it the most lucrative to falsify. Three of them were counterfeited (**I**, **III** and **V**), and four were genuine (**II**, **IV**, **VI** and **VII**). Each bill was taken randomly and was already in circulation in the economy. The origins of the fake bills are unknown.

A visual evaluation, under normal illumination, of the original banknotes was performed and, based on their color diversity 11 points were selected for pXRF analysis: six on the front side (from 1 to 6), and five on the reverse side (from 7 to 11) as shown in Fig. 1. For each point there were two kinds of data: first, the spectra with 2048 values (channels), and second, the element concentration in ppm.

For the pXRF analysis a handheld Premium DELTA Olympus portable fluorescence spectrometer was used, with a broad area SDD silicon drift detector, Rh 4 W X-Ray tube and, 200 μA maximum intensity. The analysis was performed in *Soil* mode. The equipment shot at the sample three times (three beams) consecutively at exactly the same point while maintaining equal geometry. The experimental conditions were acquisition time, tube voltage, and tube current, as follows, beam-1: 15 s, 40 KeV, 67 μA; beam-2: 20 s, 40 KeV, 34 μA and, beam-3: 30 s, 15 KeV, 78 μA respectively.

Table 2
Elemental composition (ppm) found in the false banknotes.

First Group- Same elements as in the original banknotes

Point		Ca	Cl	Ti	K	S	Fe	Mn	Zn	Sr	Cr	Cu	Zr
1	Bill-I	146456 ± 621	44980 ± 292	27758 ± 123	2308 ± 43	1349 ± 196	167 ± 8	77 ± 3	61 ± 2	65 ± 2	23 ± 2	14 ± 2	6 ± 1
1	Bill-III	183336 ± 802	21838 ± 205	62073 ± 271	1752 ± 44	1064 ± 214	112 ± 7	143 ± 3	41 ± 2	74 ± 2	25 ± 3	14 ± 2	4 ± 1
1	Bill-V	158041 ± 685	34009 ± 256	54517 ± 236	3736 ± 54	1099 ± 205	119 ± 7	124 ± 3	61 ± 2	67 ± 2	26 ± 2	18 ± 2	ND
2	Bill-I	94194 ± 414	160500 ± 775	34911 ± 152	6712 ± 68	1693 ± 224	286 ± 9	56 ± 2	45 ± 2	66 ± 2	11 ± 2	13 ± 2	5 ± 1
2	Bill-III	157636 ± 677	89960 ± 490	32818 ± 145	789 ± 34	2237 ± 228	93 ± 6	116 ± 3	46 ± 2	78 ± 2	341 ± 4	9 ± 2	ND
2	Bill-V	142999 ± 610	82227 ± 452	30035 ± 132	2192 ± 42	3018 ± 226	135 ± 7	106 ± 3	67 ± 2	70 ± 2	663 ± 6	13 ± 2	ND
3	Bill-I	120324 ± 515	45916 ± 295	28032 ± 123	1919 ± 40	811 ± 181	155 ± 8	96 ± 3	62 ± 2	61 ± 2	22 ± 2	9 ± 2	8 ± 1
3	Bill-III	123737 ± 519	15968 ± 158	34552 ± 147	1504 ± 37	1290 ± 175	134 ± 7	127 ± 3	54 ± 2	69 ± 2	26 ± 2	14 ± 2	ND
3	Bill-V	142296 ± 584	24609 ± 195	29037 ± 123	1831 ± 38	1678 ± 185	144 ± 7	113 ± 3	49 ± 2	67 ± 2	32 ± 2	16 ± 2	ND
4	Bill-I	143139 ± 595	36055 ± 247	21837 ± 97	1545 ± 37	895 ± 181	193 ± 8	84 ± 3	47 ± 2	67 ± 2	18 ± 2	16 ± 2	5 ± 1
4	Bill-III	172966 ± 733	23229 ± 198	33387 ± 146	1081 ± 36	931 ± 195	134 ± 7	192 ± 3	45 ± 2	76 ± 2	21 ± 2	14 ± 2	ND
4	Bill-V	165602 ± 705	40082 ± 275	32318 ± 142	2690 ± 47	1360 ± 204	139 ± 7	104 ± 3	57 ± 2	68 ± 2	19 ± 2	14 ± 2	ND
5	Bill-I	90335 ± 379	211751 ± 934	15583 ± 70	794 ± 28	1946 ± 224	153 ± 7	79 ± 2	40 ± 2	69 ± 2	ND	11 ± 2	8 ± 1
5	Bill-III	130943 ± 548	46741 ± 294	26724 ± 116	1079 ± 33	8274 ± 258	214 ± 9	366 ± 4	45 ± 2	73 ± 2	14 ± 2	10 ± 2	ND
5	Bill-V	137001 ± 571	110642 ± 554	26737 ± 116	1664 ± 37	1529 ± 212	125 ± 7	109 ± 3	69 ± 2	64 ± 2	8 ± 2	7 ± 2	ND
6	Bill-I	150489 ± 624	36013 ± 248	25666 ± 112	1737 ± 38	1181 ± 188	171 ± 8	83 ± 3	75 ± 2	69 ± 2	24 ± 2	9 ± 2	6 ± 1
6	Bill-III	187094 ± 795	22891 ± 200	43078 ± 187	1838 ± 42	1216 ± 207	104 ± 7	122 ± 3	48 ± 2	78 ± 2	28 ± 2	9 ± 2	ND
6	Bill-V	187009 ± 831	31813 ± 252	45986 ± 207	1893 ± 45	1715 ± 226	118 ± 7	128 ± 3	64 ± 3	70 ± 2	30 ± 2	8 ± 2	4 ± 1
7	Bill-I	170084 ± 733	54492 ± 342	34407 ± 153	2623 ± 47	1999 ± 220	145 ± 7	81 ± 3	55 ± 2	65 ± 2	44 ± 2	10 ± 2	8 ± 1
7	Bill-III	193236 ± 828	31531 ± 241	37296 ± 165	2155 ± 45	1388 ± 214	124 ± 7	120 ± 3	56 ± 2	76 ± 2	28 ± 2	18 ± 2	ND
7	Bill-V	170802 ± 727	41331 ± 281	35323 ± 155	3580 ± 52	1795 ± 212	146 ± 7	108 ± 3	64 ± 2	68 ± 2	35 ± 2	23 ± 2	ND
8	Bill-I	153338 ± 660	68476 ± 399	25700 ± 116	1122 ± 36	1444 ± 210	131 ± 7	75 ± 3	41 ± 2	68 ± 2	15 ± 2	12 ± 2	7 ± 1
8	Bill-III	162566 ± 686	41575 ± 278	32817 ± 143	1034 ± 35	5433 ± 244	177 ± 8	261 ± 4	53 ± 2	76 ± 2	17 ± 2	9 ± 2	ND
8	Bill-V	160215 ± 670	63862 ± 370	32058 ± 139	3512 ± 50	1665 ± 210	135 ± 7	99 ± 3	66 ± 2	67 ± 2	15 ± 2	14 ± 2	ND
9	Bill-I	155915 ± 659	53264 ± 328	29936 ± 131	1478 ± 38	1225 ± 200	147 ± 7	76 ± 3	47 ± 2	60 ± 2	20 ± 2	14 ± 2	6 ± 1
9	Bill-III	195459 ± 826	34570 ± 252	38133 ± 166	1977 ± 43	1580 ± 216	112 ± 7	109 ± 3	52 ± 2	76 ± 2	21 ± 2	7 ± 2	ND
9	Bill-V	169972 ± 715	35833 ± 254	34325 ± 149	2467 ± 45	930 ± 198	120 ± 7	103 ± 3	48 ± 2	68 ± 2	24 ± 2	14 ± 2	ND
10	Bill-I	167759 ± 716	50949 ± 323	28627 ± 128	1876 ± 41	1379 ± 208	159 ± 8	78 ± 3	56 ± 2	71 ± 2	24 ± 2	7 ± 2	9 ± 1
10	Bill-III	166097 ± 715	60685 ± 368	33807 ± 150	1898 ± 42	1897 ± 219	120 ± 7	125 ± 3	53 ± 2	77 ± 2	21 ± 2	13 ± 2	ND
10	Bill-V	134688 ± 570	107414 ± 549	31592 ± 137	2891 ± 46	1843 ± 217	118 ± 7	106 ± 3	53 ± 2	64 ± 2	27 ± 2	19 ± 2	ND
11	Bill-I	185394 ± 772	18089 ± 169	27564 ± 121	1571 ± 39	1055 ± 195	165 ± 7	91 ± 3	98 ± 3	77 ± 2	25 ± 2	12 ± 2	9 ± 1
11	Bill-III	198976 ± 845	24679 ± 208	40205 ± 175	1578 ± 41	1599 ± 215	98 ± 7	120 ± 3	56 ± 2	78 ± 2	29 ± 2	7 ± 2	ND
11	Bill-V	163518 ± 702	40428 ± 280	42117 ± 184	5205 ± 62	1813 ± 213	143 ± 7	124 ± 3	61 ± 2	71 ± 2	36 ± 2	30 ± 2	ND

Second Group- Elements (ppm) identified just once or twice but in different false banknotes

	Bill-I		Bill-III			Bill V	
Point	**As**	**Pb**	**As**	**Y**	**Pb**	**As**	**Pb**
1	ND	ND	ND	ND	ND	ND	ND
2	ND	5 ± 1	6 ± 1	ND	16 ± 1	5 ± 1	14 ± 1
3	ND	ND	ND	ND	9 ± 1	ND	13 ± 1
4	ND	5 ± 1	ND	567 ± 10	7 ± 1	ND	8 ± 1
5	ND	6 ± 1	ND	1157 ± 18	5 ± 1	ND	ND
6	ND	4 ± 1	ND	ND	6 ± 1	ND	7 ± 1
7	6 ± 1	12 ± 1	ND	6 ± 1	9 ± 1	ND	10 ± 1
8	ND	4 ± 1	ND	650 ± 11	4 ± 1	ND	4 ± 1
9	ND	4 ± 1	ND	ND	6 ± 1	ND	7 ± 1
10	ND	5 ± 1	ND	ND	9 ± 1	4 ± 1	11 ± 1
11	ND	ND	3 ± 1	ND	8 ± 1	ND	12 ± 1

Before the experiment, the equipment was checked to make sure it was working correctly using a 316 Stainless Steel Calibration Check Reference Coin. For all the experiments a stand workstation was used. The X-ray gun was placed under the workstation and faced upwards, then the bill was placed on the shooting window. Through a mini CCD camera (model DP-600-CC) the irradiated area could be observed, that corresponded to a circular diameter of 8 mm. The spectra and the element concentration were obtained using the Innov-X Delta software.

The 11 points analyzed on each bill correspond to 77 sets of data. In the case of the spectral analysis, 77 spectra were considered, each containing 2048 data, corresponding to 2048 channels (1–2048), and the respective number of counts per channel. Each channel is associated with an energy value (0–40 KeV). This involves the manipulation of 157696 values. In addition, the Soil mode of the pXRF spectrometer provided three spectra for each point, since three beams were used, making the initial total amount of data about 473,088. However, the preliminary analysis showed that the elements present could be studied with the third beam (0–40 KeV), which significantly decreased the amount of data used.

During the experiment, the bills were mixed in a way that they could not be recognized as originals or false. A set of XRF chemical emissions were considered to characterize and identify the original bill, therefore the genuine bills should have equal or similar emissions, because in theory they came from the same manufacturer and so should show equal correlations (*cor (x,y) = 1*) between spectral intensities for each energy.

Then, applying R-Studio v. 0.99.486 the correlation between all the spectra using the function "cor (x,y)" was calculated. That operates as a default Pearson's linear correlation, and measures the lineal relationship between two quantitative random variables and is independent

Table 3
Spectral correlation in the eleven points, between the original (II, IV, VI, VII) and false (I, III and, V) banknotes.

Banknotes Correlation	Analysis points										
	1	2	3	4	5	6	7	8	9	10	11
Bill I x II	0.97	0.81	0.99	1	0.67	0.97	1	0.98	0.99	0.98	0.93
Bill I x III	0.97	0.86	0.97	0.99	0.53	0.99	0.99	0.93	0.99	1	1
Bill I x IV	0.97	0.8	0.99	1	0.66	0.95	1	0.99	0.98	0.98	0.94
Bill I x V	0.98	0.86	0.98	1	0.87	0.99	1	1	0.99	0.92	0.98
Bill I x VI	0.96	0.75	0.99	0.99	0.64	0.98	1	0.99	0.99	0.98	0.96
Bill I x VII	0.95	0.77	0.99	1	0.69	0.96	0.99	0.99	0.98	0.97	0.93
Bill II x III	0.95	0.98	0.93	0.99	0.84	0.94	0.98	0.92	0.96	0.99	0.94
Bill II x IV	**1**	1	1	1	1	1	1	1	1	1	1
Bill II x V	0.98	0.98	0.94	1	0.95	0.95	0.99	0.99	0.97	0.95	0.98
Bill II x VI	0.99	1	1	1	1	1	1	1	1	1	1
Bill II x VII	1	1	1	1	1	1	1	1	1	1	1
Bill III x IV	0.95	0.97	0.95	0.99	0.83	0.93	0.98	0.93	0.95	0.99	0.96
Bill III x V	**1**	1	1	0.99	0.79	1	1	0.94	1	0.94	0.99
Bill III x VI	0.97	0.97	0.94	0.99	0.84	0.96	0.99	0.93	0.96	0.99	0.97
Bill III x VII	0.94	0.97	0.95	0.99	0.83	0.94	0.97	0.92	0.96	0.98	0.95
Bill IV x V	0.97	0.98	0.96	1	0.94	0.95	0.99	0.99	0.96	0.96	0.99
Bill IVx VI	0.99	1	1	1	1	1	1	1	1	1	1
Bill IV x VII	**1**	1	1	1	1	1	1	1	1	1	1
Bill Vx VI	0.98	0.97	0.95	1	0.93	0.97	1	0.99	0.97	0.94	0.99
Bill Vx VII	0.96	0.97	0.95	1	0.95	0.95	0.99	0.99	0.97	0.96	0.98
Bill VI x VII	0.99	1	1	1	1	1	1	0.99	1	1	1

from the scale of measurement of the variables, which correspond to different spectra. Thus, a matrix with 2048 rows (energy channels) was constructed per 77 columns (counts or intensities) of the 11 analysis points for each of the 7 banknotes. Only beam-3 spectra counts were used, since they contained all the elements of interest. To find the number of correlations in the algorithm of the program in R, the equation **[n (n-1) / 2]** was used, where n is the number of banknotes (n = 7), obtaining 21 correlations for each point, making a total of 231 correlations (21 × 11 analysis points).

After observing the results, the correlations between the selected points were almost identical on the authentic bill, despite being well-used and already in-circulation. From this, we could conclude our experiments on the genuine notes, as they all showed the same result. The software Innov-X Delta reported the presence of the elements, the spectral peaks were matched to emission energy tables, after that the presence of the elements were evaluated and confirmed.

Furthermore, in order to validate the results, it is necessary to use matrix standards similar to paper money with a thin film geometry. However, we only had the SRM® 2710a Montana I Soil and the SRM® 2711a Montana II Soil standards that feature infinitely thickness geometry. These were placed in the stand workstation of the pXRF and analyzed under the same conditions as the bills. Then, the standards were used only with the intention of demonstrating the validity of the results reported by the equipment.

The PCA multivariate analysis used Pirouette 4.5 and the concentrations obtained from three beams of Innov-X Delta software, for the 15 identified elements. These were considered as variables, meanwhile the 11 points, in each one of the 7 banknotes, constituted a total of 77 samples. A mean centering pre-processing was performed, to mitigate the influence of the Ca and Ti variables magnitude, and 7 factors were considered as a maximum.

3. Results and discussion

Twelve (12) elements were identified using the genuine banknotes pXRF elemental analysis: Ca, Cl, Ti, Zn, Cu, Fe, K, Sr, Mn, Zr, Cr, and S (Table 1). These elements can be separated into three groups, the first, with mayor intensities as Calcium. The second group, with middle intensities: Ti, Zn, Cu, Fe, and K. The third group with lower intensities: Sr, Mn, Zr, Cr, and S. Zirconium was found only in two of four original bills.

Twelve (12) elements were identified using the genuine banknotes pXRF elemental analysis: Ca, Ti, Cl, Zn, Cu, Fe, K, Sr, Mn, Zr, Cr, and S (Table 1). These elements can be separated into three groups, the first, with mayor intensities: Ca and Ti. The second group, with middle intensities: Cl, Zn, Cu, Fe, and K. The third group with lower intensities: Sr, Mn, Zr, Cr, and S. Zirconium was found only in two of four original bills.

These results coincide with those reported by Appoloni and Melquiades (2014) who highlighted the presence of Fe, Ti, Ca, Sr and Zr in a R/.50.00 Brazilian Reales currency banknote, and in a USD $50.00 American Dollar banknote, they reported Fe, Ti, Ca and Zn and, finally, the presence of Ti, Ca and Y, in a €50.00 Euro banknote. The above-mentioned authors indicate that these banknotes coincide in the presence of titanium´s white pigment in paper currency.

The false banknotes show a different elemental composition (Table 2). 15 elements were identified and can be divided into three groups: The first is composed of the same elements as in the originals: Ca, Cl, Ti, K, S, Fe, Mn, Zn, Sr, Cr, Cu and Zr. The second group can be constituted by As, Y, Pb, and where As and Y show up once or twice. Meanwhile lead is present in more than 7 of the 11 analyzed points in all the fake bills.

It is logical to expect that the spectral correlation on each point should be 1.0 with all original banknotes. The pXRF analysis confirms the previous statement. Table 3 shows the spectral correlation of the elements identified by pXRF for the false banknotes (**I, III** and **V**) and the genuine ones (**II, IV, VI** and **VII**). For example, in the second analysis point (**2**), the correlation between the false banknote (**I**) and the genuine one (**II**) is 0.81, meanwhile the correlation, at the same point, between the original banknotes **II-IV, II-VI** and, **II-VII** is 1.

It was observed that any one of the correlations, between the genuine banknotes (Fig. 2a), is always equal to 1, with the exception of the correlations between the **IIxVI, IVxVI** and **VIxVII** banknotes, at point 1, which could be due to the banknote´s contamination and wear from being in circulation. This shows the fabrication reproducibility of the studied original banknotes, and the potential pXRF as an applicable

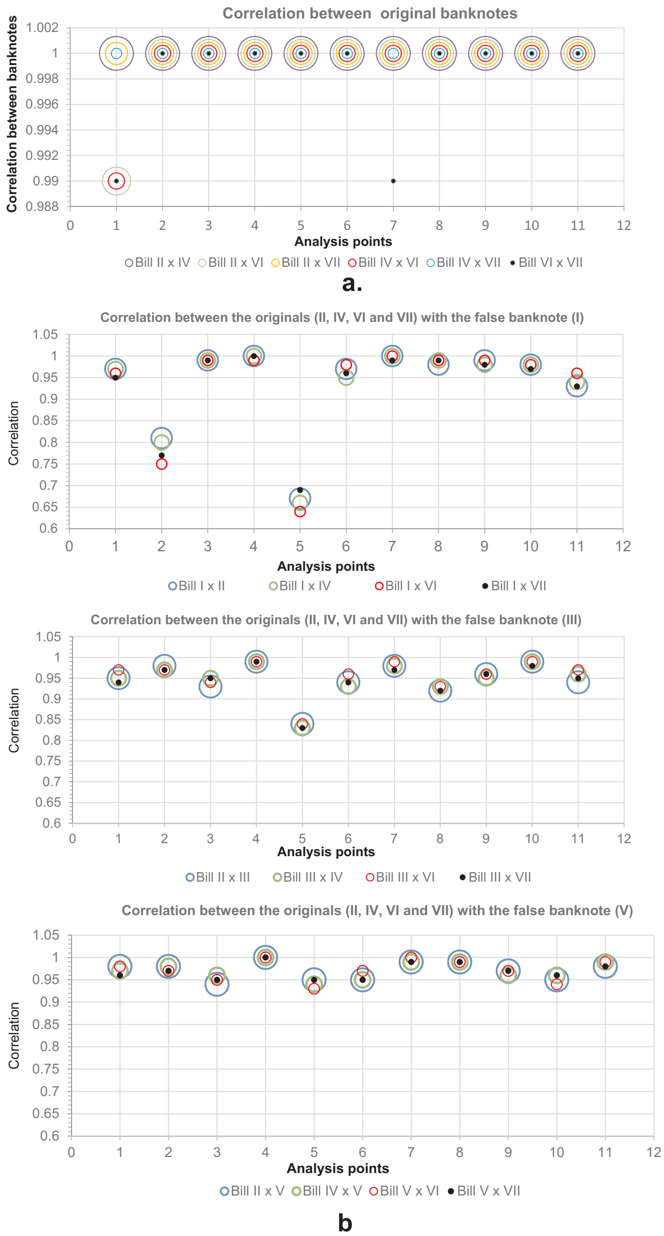

Fig. 2. a. Spectral correlation in each one of the eleven analysis points between the originals banknotes (II, IV, VI and VII). b. Spectral correlation in each one of the eleven analysis points between the originals (II, IV, VI and VII) banknotes and false ones I, III and V.

technique in the forensic analysis sustained in the elemental analysis.

In a similar way, it was observed that the points **4** and **7** present high original/false Pearson correlations, and even give a value equal to one between false banknotes (**I** and **V**), implicating that these points could be falsified with greater ease (Fig. 2b).

False notes generate different emissions than the authentic ones, which is shown with correlations between 0.65 and 0.8. Likewise, there are points of analysis in counterfeit notes with correlation in respect to the original, close to 1. Thus, we consider that these points can be reproduced by counterfeiters. From Fig. 2b, we can conclude that the best falsification corresponds to bill 5, which has correlations between 0.93 and 1 with the original bills.

The pXRF data show that point **5** is the most difficult one to reproduce (Fig. 2b). Therefore, one can state that pXRF is a technique that allows for orientation of the type of security feature to be developed in banknotes.

In Table 3, it is observed that the difference in correlation obtained from pXRF, between real banknotes is of a centesimal (0.01), considering the banknotes were not new, and that they were in circulation, these values might reflect a certain degree of contamination proceeding from manipulation for being in circulation. The use of new banknotes, in the study, would give correlations equal to 1, as results. Thus, it is possible to assert that pXRF is a technique that not only allows identifying false Soles banknotes, but, also, the development of new ones.

The result of the principal components analysis (100% of accumulated variance), based on the concentration obtained by pXRF, presents the separation of two groups: genuine and false. Although samples **b1-2** and **b1-5** (points 2 and 5 in banknote **I**, respectively) could look like outliers, their exclusion does not conduce to a better PCA result. The same happens with the exclusion of Ca, Ti and Cl.

The 3D graphic of factors (Fig. 3a) shows the separation of the two groups: Originals in red (**B2, B4, B6** and **B7**), and false in brown (**b1, b3** and **b5**). The loadings' graphic (Fig. 3b) shows, that the presence of Ca is important in the false banknotes' characterization, as well as that of Titanium, in the genuine banknotes. This fact could be related to the type of paper used and Titanium ink previously reported. The presence of Calcium (Fig. 4), in the genuine banknotes is notably lesser than in the false ones.

Banknote **I** has a different presence of Chlorine to that observed in other fake bills. Point 2 (**b1-2**) and point 5 (**b1-5**) are characterized by their content in this element (16.1% and 21.2%) respectively.

On the other hand, in point **5**, the false banknotes also show a different elemental composition to that of the genuine ones. That is to say that, although under UV light, these points could seem red to the human eye, and identical to the original. However, the pXRF emission shows that they correspond to different energies, thus identifying the falsification.

It is known that the trace elements distribution, in different samples, tends to relate to the sample´s origin. In this sense, Fig. 5 shows the variability of the minority elements profile, the false banknotes (**I, III** and **V**) composition is different to the genuine banknotes (**II, IV, VI** and **VII**).

In Table 4 we compare our results from the experiments using the specified values from Standard Reference Material SRM 2710a Montana I Soil and SRM 2711a Montana II Soil. The experiment values correspond to the average of three measurements made under the same condition for all the points analyzed on the 200 Nuevos Soles bill. The table shows that the experiment concentrations were very close to the certified values, which validates the results obtained by the pXRF.

Moreover, pXRF let us identified the presence of small quantities of some toxic elements in the false banknotes: Cr, Pb, and As (Table 5). Chrome was found in point 2 of all fake bills, in the false banknotes **Bill-III** (341 ppm) and in **Bill-V** (663 ppm), it exceed the limit for toys and desk utensils (60 ppm), allowed by Dirección General de Salud Ambiental-DIGESA in Peru (DIGESA, 2008). Pb and As are under the permissible limits.

4. Conclusions

The results from the semi quantitative elemental analysis, using pXRF, show that the chemical composition of the ink used in the false banknotes is different to that of original ones. The data analysis allows an insight into the pXRF´s potential, as a technique applicable to high quality fraudulent paper currency detection and also allows identifying

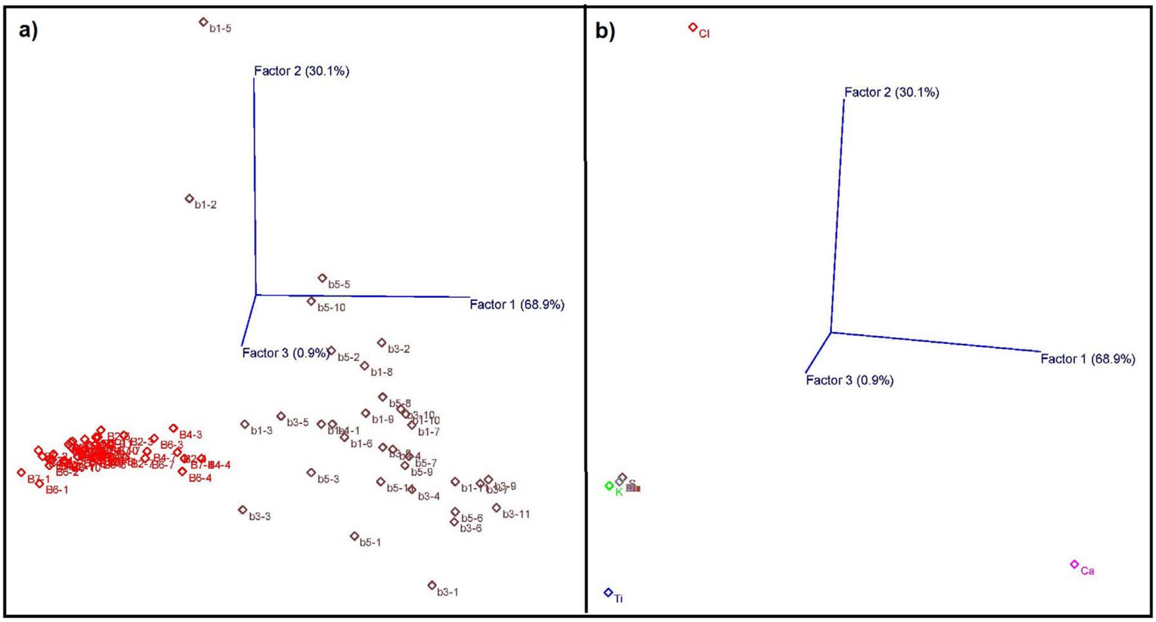

Fig. 3. Principal components analysis 3D graphic of (a) scores and (b) loadings. Original banknotes are in red (B2, B4, B6 and B7) and false banknotes in brown (b1, b3, b5) with their eleven sampling points.

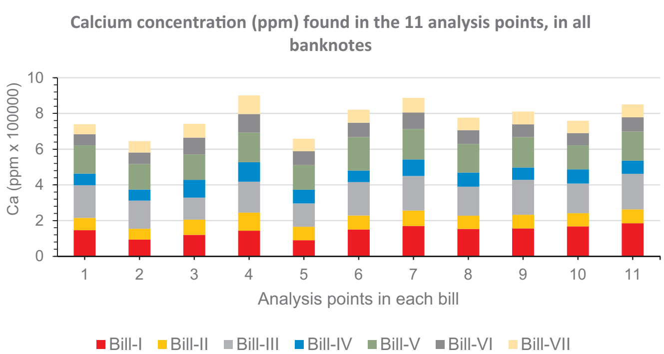

Fig. 4. Calcium concentration (ppm) found in the 11 analysis points, in all banknotes.

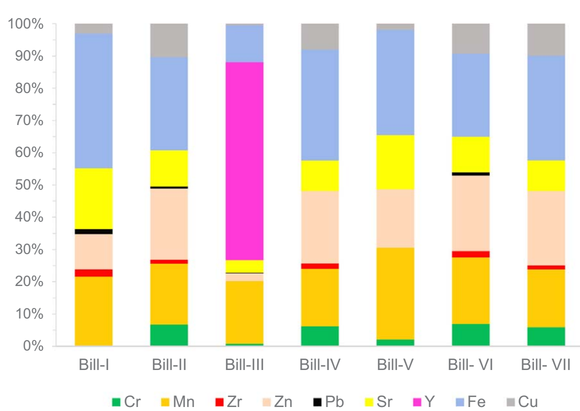

Fig. 5. Minority elements in point 5, in all banknotes.

which are the security marks that are easily and more faithfully reproduced by falsifiers. The use of principal components analysis, in the data processing, also allows an insight into the pXRF's applicability in this field.

Table 4
Values of elemental composition in SRM 2711a and SRM 2710a obtained by pXRF.

Element	SRM 2711a Experimental (%)	Certified Values (%)
K	2.75 ± 0.02	2.53 ± 0.10
Ca	2.74 ± 0.02	2.42 ± 0.06
Ti	0.319 ± 0.030	0.317 ± 0.008
Fe	2.41 ± 0.02	2.82 ± 0.04
Element	**Experimental (ppm)**	**Certified Values (ppm)**
Cr	37 ± 3	52.3 ± 2.9
Mn	640 ± 6	675 ± 18
Cu	125 ± 4	140 ± 2
Zn	370 ± 6	414 ± 11
As	125 ± 8	107 ± 5
Sr	236 ± 5	242 ± 10
Y	35 ± 2	–
Zr	402 ± 8	–
Pb	1322 ± 11	–
	SRM 2710a	
Element	**Experimental (ppm)**	**Certified Values (ppm)**
Sr	262 ± 6	255 ± 7
Pb	5299 ± 37	5520 ± 30

Table 5
Values (ppm) of some toxic elements in the false banknotes (I, III and V).

	Cr		Pb		As	
Maximum value all studied banknotes		–	Bill-I:	12 ± 1	Bill-I:	6 ± 1
	Bill-III:	341 ± 4	Bill-III:	16 ± 1	Bill-III:	6 ± 1
	Bill-V:	663 ± 6	Bill-V:	14 ± 1	Bill-V:	5 ± 1
Maximum limit allowed by DIGESA in toys	60			90		25

References

Appoloni, C.R., Melquiades, F.L., 2014. Portable XRF and principal component analysis for bill characterization in forensic science. Appl. Radiat. Isot. 85, 92–95. http://dx.doi.org/10.1016/j.apradiso.2013.12.004.

BCRP, B., 2016. Elementos de seguridad de los Billetes [WWW Document]. URL ⟨http://www.bcrp.gob.pe/billetes-y-monedas/familia-de-billetes/elementos-de-seguridad-de-los-billetes.html⟩ (accessed 12 December 2016).

BCRP, B., 2015. seguridad-billetes-200.pdf [WWW Document]. URL ⟨http://www.bcrp.

gob.pe/docs/Billetes-Monedas/medidas-de-seguridad/seguridad-billetes-200.pdf〉 (accessed 12 December 2016).
Cabitza, M., 2012. Why fake dollars are big business in Peru. BBC News.
Cao, S., Nie, X., Cheng, Z., 2012. New auto identification technology on paper Currency using pseudo binocular stereo imaging.pdf. Comput. Sci. 39, 19–22.
Cesareo, R., Ferretti, M., Gigante, G.E., Guida, G., Moioli, P., Ridolfi, S., Roldán Garcia, C., 2007. The use of a European coinage alloy to compare the detection limits of mobile XRF systems. A feasibility study. X-Ray Spectrom. 36, 167–172. http://dx.doi.org/10.1002/xrs.960.
CNBC, 2016. Peru seizes $30 million counterfeit dollars in record bust [www document]. CNBC (URL). 〈http://www.cnbc.com/2016/11/18/peru-seizes-30-million-counterfeit-dollars-in-record-bust.html〉.
Dale, J.M., Klatt, L.N., 1989. Principal component analysis of diffuse near-infrared reflectance data from paper currency. Appl. Spectrosc. 43, 1399–1405.
DIGESA, R.M., 2008. República del Perú. Reglamento de la Ley N° 28376, Ley que prohíbe y sanciona la fabricación, importación, distribución y comercialización de juguetes y útiles de escritorio tóxicos o peligrosos, RM517-2008/MINSA.
Dwan, B., 2002. Counterfeit and Fraud. Comput. Fraud Secur. 2002, 11.
Europol, E., 2012. Four million counterfeit euros confiscated in Peru [WWW Document]. Europol (URL). 〈https://www.europol.europa.eu/newsroom/news/four-million-counterfeit-euros-confiscated-in-peru〉.
Franklin, J., 2016. "Counterfeiting is an art": Peruvian gang of master fabricators churns out $100 bills. Guardian.
García-Lamont, F., Cervantes, J., López, A., 2012. Recognition of Mexican banknotes via their color and texture features. Expert Syst. Appl. 39, 9651–9660. http://dx.doi.org/10.1016/j.eswa.2012.02.132.
Hassanpour, H., Farahabadi, P.M., 2009. Using Hidden Markov Models for paper currency recognition. Expert Syst. Appl. 36, 10105–10111. http://dx.doi.org/10.1016/j.eswa.2009.01.057.
Jelovica Badovinac, I., Orlić, N., Lofrumento, C., Dobrinić, J., Orlić, M., 2010. Spectral analysis of postage stamps and banknotes from the region of Rijeka in Croatia. Nucl. Instrum. Methods Phys. Res. Sect. Accel. Spectrometers Detect. Assoc. Equip. 619, 487–490. http://dx.doi.org/10.1016/j.nima.2009.10.174.
Nir-El, Y., 1994. Determination of lead in US dollar paper currency by X-ray fluorescence spectroscopy. Bull. Environ. Contam. Toxicol. 52, 787–793.
Reuters NBC News, 2016. U.S., Peru seize $30 million in counterfeit dollars, biggest bust ever [WWW Document]. NBC News. URL 〈http://www.nbcnews.com/news/world/u-s-peru-seize-30m-counterfeit-dollars-biggest-bust-ever-n685646〉 (accessed 14 December 2016).
Roy, A., Halder, B., Garain, U., Doermann, D.S., 2015. Machine-assisted authentication of paper currency: an experiment on Indian banknotes. Int. J. Doc. Anal. Recognit. IJDAR 18, 271–285. http://dx.doi.org/10.1007/s10032-015-0246-y.
Rusanov, V., Chakarova, K., Winkler, H., Trautwein, A., 2009. Mössbauer and X-ray fluorescence measurements of authentic and counterfeited banknote pigments. Dyes Pigments 81, 254–258. http://dx.doi.org/10.1016/j.dyepig.2008.07.020.
Sargano, A.B., Sarfraz, M., Haq, N., 2014. An intelligent system for paper currency recognition with robust features. J. Intell. Fuzzy Syst. 27, 1905–1913.