

# The circulation of South American precious metals in Brazil at the end of the 17th century

M.F. Guerra\*

*Centre de Recherche et de Restauration des Musées de France, UMR 171  
CNRS, Palais du Louvre - Porte des Lions, 14 quai François Mitterrand, 75001 Paris, France*

Received 30 September 2001; received in revised form 17 December 2002

## Abstract

The lack of important silver and gold sources in Brazil before the finds in the region of Minas Gerais at the end of the 17th century is known. Information concerning the first gold finds reached the Portuguese capital in 1695 but it was only in 1697 that the first ship arrived at Lisbon harbour with a gold load identified in the documents as ‘Peruvian’ but supposed to be Brazilian.

The first Brazilian mint was itinerant, and the first silver and gold coins were struck in Bahia from 1694 to 1698. In order to try to find out which metals were struck before 1698 in Brazil, we analyzed gold and silver coins issued by the first Brazilian mint, Bahia, and coins issued in Mexico and in South America—Peru, Bolivia, Colombia, Chile—as well as Brazilian gold ingots from different mining regions. Fingerprinting was done using several analytical techniques: 12 MeV proton activation analysis, moderated neutrons activation analysis and inductively coupled plasma mass spectrometry with laser ablation.

We could show that in general the platinum group elements discriminate gold and that elements such as gold, indium and tin discriminate silver. The comparison of the Brazilian coins and ingots with the coins issued elsewhere in South America and Mexico showed that the gold and silver coins circulating in Brazil between 1694 and 1698 were mostly made from a mixture of metals from the main Latin American sources.

© 2004 Elsevier Ltd. All rights reserved.

*Keywords:* Gold; Silver; South America; Fingerprinting; Coins; Ingots; Brazil

## 1. Gold and silver in Brazil

No important precious ore sources were known in Brazil before the find of gold at the end of the 17th century in the state of Minas Gerais. The precious metals used for the fabrication of the first Brazilian coinages could have come from numerous regions of the New World.

The Brazilian mint was created on the 8th March 1694 in Bahia by a royal decree and successively closed and transferred to Rio de Janeiro in 1698, to Pernambuco in 1700 and back to Rio de Janeiro in 1702. This itinerary is supposed to be correlated to the exhaustion of the local metal stocks. After the discovery of the Brazilian gold both Rio and Bahia mints started to issue

again in 1714, followed in 1724 by Vila Rica (Ouro Preto) and several other mints.

Little is known about the metals circulating in Brazil in the 17th century. According to Noya Pinto [26], in 1674 based on the finds in Bolivia and Mexico, King Peter II asked the inhabitants of São Paulo to search for precious metals in their lands, especially silver. In 1680 gold was found in Curitiba and Paranagua (in southern Brazil, see Fig. 1) and in June 1695 the Lisbon Court was informed about the first discovery of gold in Minas Gerais. The first ship carrying gold reached Lisbon only in 1697, the last operating year of the first Bahia mint before transfer to Rio. The records identified that gold as ‘Peruvian’, but Noya Pinto [26] (pp. 137 and 228) affirms that it was Brazilian.

Many authors agree with the fact that silver arrived in Brazil certainly through Rio de La Plata from Peru (from the Potosi mines, ancient Peru includes recent Bolivia and Peru) and also from Mexico. The

\* Corresponding author. Tel.: +33-1-40-20-24-58; fax: +33-1-47-03-32-46.

E-mail address: [maria.guerra@culture.fr](mailto:maria.guerra@culture.fr).



Fig. 1. Map of South America with the location of the mints and mines.

documents show that in 1651 the Brazilian governor wished to open a mint in Brazil in order to melt the Peruvian silver coins circulating in large amounts in that Portuguese territory [35] (p. 152).

The Potosi mines were found in 1545 but produced a large quantity of metal only after the introduction in 1570–1572 of the mercury amalgamation technique [2,5]. As we can see in Fig. 2, from Braudel [3], these mines continued to produce a large amount of silver until the middle of the 17th century, but the Mexican mines outweighed that quantity in the beginning of the 18th century.

In order to try to identify the metals struck before the gold finds of Minas Gerais, we analyzed coins issued from 1694 to 1698 in the first Brazilian mint, Bahia, as well as coins issued in Mexico, Peru, Bolivia, Colombia and Chile.

## 2. Analytical techniques and analyzed objects

Some of the South American metals have known characteristic trace elements. It is known that Colombian gold has numerous platinum inclusions, as

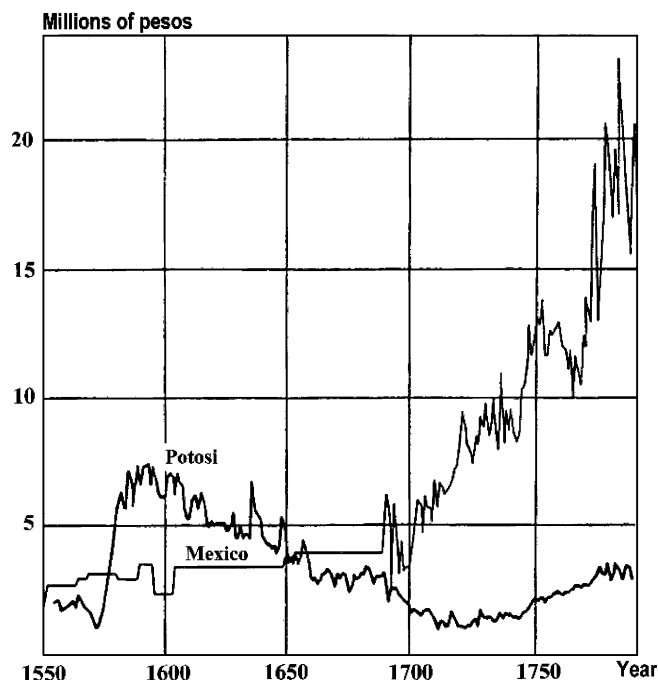


Fig. 2. The production curves of the silver mines of Potosi and Mexico from 1550 to 1800 [3].

demonstrated by the objects from the archaeological site of Las Tolitas (for example Refs. [33,34]). In the 1980s Guerra [14] and Guerra et al. [15] showed that Potosian silver is characterized by indium, while Morrisson et al. [25], following the work on Potosian silver and considering gold nuggets from Minas Gerais, Sabará and Rio de Janeiro analyzed by De Launay in 1913 [7], confirmed that the gold from Brazil is characterized by palladium.

However, for all the other precious metals exploited in the New World no characteristic elements were defined. In order to understand the circulation of the precious metals in Brazil, we tried to characterize the most important silver and gold metals circulating in Latin America by the determination of the trace element concentrations in a selection of coins issued in Mexico, Peru, Bolivia, Guatemala, Colombia and Chile. The results obtained for the composition of those silver and gold coins were compared with the composition of a group of Brazilian coins struck in the first Brazilian mint (1694–1698), Bahia, as well as with a group of 15 gold ingots fabricated in different Brazilian mints and expected to have a composition closer to the original ore.<sup>1</sup>

<sup>1</sup> The gold dust and nuggets were directly taken by the gold prospector to the mints where the ingots were made after the application of the 1/5 royal tax to the total weight of gold. Prober [28] (pp. 11 and 12) writes that the gold was melted and that the impurities were eliminated by addition of 'solimão', the popular name of 'sublimado corrosivo', which is the commercial name of mercury chloride. On the ingots the mint name, the craftsman's name, the reference, the date of issue, and so on, were struck and a certificate containing all that information was delivered to the owner.

Five Portuguese silver coins over-struck to circulate in Brazil were also analyzed.

The determination of trace elements at very low levels is not an easy task as classical techniques have in general too high detection limits to measure the characteristic elements of the metals. In order to determine as many trace elements as possible in a quantitative way, we used 12 MeV proton activation for the analysis of almost all the coins and ingots and inductively coupled plasma mass spectrometry (ICP-MS) with a UV laser for the analysis of some specimens. Some silver coins were also analyzed by thermal fast neutron activation analysis.

The proton activation technique using a cyclotron was first developed by P. Meyers in 1969 [23]. The proton energy used for this work was first reported by Barrandon et al. in 1976 [1] but it was only in 1983 that Poirier [27] published the methods of analysis and quantitative evaluation as well as the set-up. A 12 MeV proton beam with an intensity of 2  $\mu\text{A}$  is extracted from the CERI-CNRS cyclotron through a 25  $\mu\text{m}$  Ti window. At this energy only (p,n) reactions are produced in silver and gold avoiding interference. In order to perform  $\gamma$ -ray spectrometry after irradiation, for gold a 3 mm lead sheet interposed between the sample and the detector absorbs the low energy  $\gamma$  rays emitted by the two isomers of Hg197, while for silver a 2–3 day cooling time allows the decay of Cd107 (6.5 h half-life) after which we can measure the other elements. Quantitative calculations are performed by using the flux monitoring technique and the Ricci and Hann [30,31] mean  $\sigma$  method for the proton range in the sample; concentrations are normalized to 100 wt%. The range of proton in gold is about 240  $\mu\text{m}$ , most of the information coming from the first 50  $\mu\text{m}$ , and the analyzed region is of about 0.8  $\text{cm}^2$ . With this technique we can measure routinely (in 3–4 weeks) 12 elements with detection limits reaching the ppm level [18].

LA-ICP-MS analysis was performed with a Fisons PlasmaQuad PQXS device combined with a 2 mJ pulsed VG Nd:YAG laser working on Q-switched mode and at UV wavelength with a shot frequency of 6 Hz. Peak jumping acquisition mode was used with 10.24 ms dwell time and 60 s acquisition time after 10 s of pre-ablation. Craters vary from 40 to 70  $\mu\text{m}$  in diameter depending on the matrix and are about 130  $\mu\text{m}$  deep. Quantitative calculations are made by using the internal standard technique, the ions formed either by silver or by gold with argon in the plasma being used as internal standards [10]. Considering the concentration equivalent to  $3\sigma$  of three replicate analyses of high-purity standards, detection limits are typically between the ppm and the ppb level. About 40 elements are measured in 15 min with a reproducibility of about 5% (estimated on 30 consecutive measurements). Contrary to activation techniques, quantitative LA-ICP-MS requires standards with

compositions as close as possible to that of the sample. As no commercial standards are available for trace elements determination we use gold and silver samples ‘certified’ by activation analysis [10].<sup>2</sup>

Some of the silver coins were analyzed by neutron activation analysis developed to measure in a totally non-destructive way low contents of gold and indium. A 30  $\mu\text{A}$  current of 20 MeV HH+ issued from a cyclotron impinges on a thick Be target to produce by nuclear reactions a neutron flux moderated in a high-purity graphite block (moderated neutrons activation analysis (TFNAA)) [13,14]. After passing through 15 cm of graphite the estimated thermal neutron flux is  $2.7 \times 10^8$  n/cm<sup>2</sup> s. The homogeneous irradiation volume is 80 cm<sup>3</sup> which is big enough to irradiate several coins at the same time or small objects. With the two silver isotopes Ag108 and Ag110 produced by (n, $\gamma$ ) reactions having very short half-lives, of respectively 2.37 min and 24.6 s, and isotopes Ag108<sup>m</sup> and Ag110<sup>m</sup> having a very low sensitivity (respectively  $6 \times 10^{-5}$  and 0.0034 if we consider the 511 keV line of Cu64 as 1)  $\gamma$ -ray spectroscopy can be performed immediately after irradiation. Quantitative evaluation is made by using the internal standard technique with the introduction of the appropriate correction for neutrons self-absorption. The detection limits are strongly dependent on the element but attain 0.01 ppm for indium and 0.1 ppm for gold. However, if activation analysis has usually a 5% accuracy, TFNAA with the correction for self-absorption may sometimes exceed 10%.

### 3. Discussion

#### 3.1. The gold coinages

Gold is found in nature either in primary deposits alloyed with silver in quartz, in the lattice of arsenopyrite and pyrite or as tellurides. However, secondary deposits, placers, have always been the main source of gold [24]. Native ores may reach 98% of gold [8,12], the rest being mostly up to 40% silver, up to 1% copper and up to 4% iron [7,29].

Depending on the geology of the ore deposit, different elements enter the composition of native gold, and others can be panned together with it. It is known that many South American gold objects have very particular characteristic elements, such as platinum in Colombia and palladium-platinum in Brazil. Therefore, the most important elements to characterize gold seem to be platinum and palladium, those of the platinum

<sup>2</sup> The difficulty of this techniques is quantification: the laser pulse must be linear, the metal should not melt during ablation and the sample should not be lost during transportation to plasma. These issues are discussed in Gondonneau et al. [11].

group elements (PGE) that are the most soluble in gold [38]. However, we must be aware of the probability of precipitation of PGE in gold as exposed by Meeks and Tite [22] and more recently by Craddock [4].

The first Bahia mint struck in the 17th century while the other coins and the ingots were made during the 18th and 19th centuries. We assume that trace elements are not time-dependent.

In Fig. 3 we plot the platinum and palladium concentrations in ppm divided by the gold content expressed in percent (in addition to coins of Table 1, four coins from Peru, five from Colombia and two from Brazil-Bahia published by Morrisson et al. [25] and Guerra et al. [17] are also considered). This ratio of trace element concentration to gold content avoids fluctuations due only to changes of quality. We can observe that Colombian gold (mainly Popayan) has the highest concentration of platinum, while the highest concentrations of palladium were found in Brazilian ingots.<sup>3</sup> The high platinum content, about 52%, measured in the coin struck in Popayan in 1776 is explained by the fact that most of the analyzed volume was occupied by an inclusion which could not be avoided. This coin is not considered in the graphic.

The fact that in Ouro Preto ('black gold') native gold is associated with oxygen-bearing compounds of Pt-Pd-Fe [20] explains the high contents of palladium (2%) and also of platinum (900 ppm) found in some of the Brazilian ingots. This means that platinum can be present in Brazilian gold at higher concentrations than in some Colombian coins. This reinforces the fact that both platinum and palladium contents must be measured. We must also note that the ingots are supposed to be made with little purification, the gold being separated from the impurities by addition of mercury [28] (see also footnote 1). However, this might not be exact if we consider first the variation of elements such as palladium<sup>3</sup> and the absence of mercury.

With the exception of one coin, Peruvian gold has lower contents of platinum and palladium. Chilean and some Mexican coins fit with the group of Peru as well as the only Bolivian coin analyzed. The two Mexican coins in the group of Colombia could have been made with Colombian gold.

For the coins struck in Bahia we can notice that those issued by the first mint in the 17th century fit together (except the two included in the Colombian group) in-between the Colombian chemical group and most of the Peruvian coins, very close to the low platinum contents in Colombian coins. The two coins analyzed by Morrisson et al. [25] struck in the 18th

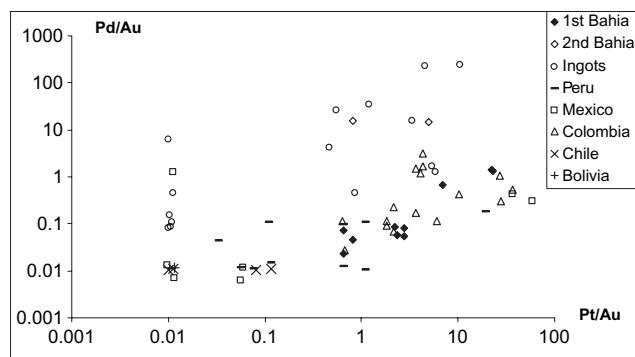


Fig. 3. Ratio of platinum and palladium concentrations in ppm to gold content in percent for the gold coins struck in Mexico, Chile, Colombia, Peru, Bolivia and Brazil (first Bahia and second Bahia mints) as well as for the Brazilian ingots.

century by the second Bahia mint fit with the Brazilian ingots.<sup>4</sup>

If we consider (a) the fact that two Brazilian coins struck by the first Bahia mint fit with the high platinum content Colombian gold, (b) that in the 17th century Colombia was the biggest gold producer in South America—as shown in Table 2 where the data of Rigway [32] for gold production between the first half of the 16th century and 1800 are summarized—and (c) that all the other coins struck in the first Bahia mint fit with the low platinum content Colombian gold, we can suggest that this gold reached Brazil in the 17th century and became part of the Brazilian stocks.

However, the gold struck in the first Bahia mint is not a pure Colombian gold. These coins seem to be close to Colombian gold by their palladium and platinum contents, but do not show the same contents of tin and antimony (no correlation could be found between these elements and copper or silver). The concentrations of tin and antimony shown in Fig. 4 indicate that the highest contents were found in Colombian gold. With tin increasing with antimony, we could suggest a correlation between these two elements. The coins from Peru and Bolivia also show lower contents of tin and antimony. These elements could be removed by refining, but the concentration of palladium would also decrease. We could suggest the use of a mixture of two golds, the second one with very low contents of tin and antimony.

Table 3 shows the concentrations of elements measured by LA-ICP-MS for a group of Mexican, Peruvian, Colombian and Chilean gold coins. Due to the scarcity of gold standards, the concentrations given in Table 3 are estimated by extrapolation based on the closest quantified element. Little new information is obtained

<sup>3</sup> Two ingots show low palladium and high platinum contents. They might have been made with either a refined gold or a gold coming from other South American regions. We must note that an ingot has a higher value than the same weight of gold.

<sup>4</sup> If we consider the high quantity of copper for the ingot made in Ouro Preto in 1818 we could say that it may be a fake. This explains the fact that the craftsman, whose name is indicated on the ingot, was unknown in that mint.

Table 1  
Composition of the gold coins and ingots obtained by PAA

		%			ppm													
		Au	Ag	Cu	Pb	Pt	Pd	Sn	Sb	As	Fe	Zn	Te	Ga	Hg	Cr	Ti	Ru
<b>Brazil</b>																		
<i>Coins</i>																		
Bahia	1695	92.3	6.7	1.0	25	60	7	7	5	6	275	10	2		385	2	2	7
Bahia	1696	91.9	6.7	1.3	40	75	4	25	8	10	300	85	2		515	4	2	8
Bahia	1696	92.2	6.3	1.4	30	60	2	15	4	8	410	305	1	1	40		1	15
Bahia	1697	91.3	7.4	0.9	30	2040	125	40	10	13	805	50	5	1	340	9	2	10
Bahia	1697	91.8	7.2	0.9	15	215	5	25	6	6	285	15	5	1	235	1	1	1
Bahia	1697	91.3	6.8	1.8	15	205	8	20	10	20	345	25	2		460	4	1	5
Bahia	1698	92.7	6.3	0.9	30	260	8	35	10	8	305	15	3		30	3	2	5
Bahia	1688	90.8	7.3	1.7	40	620	60	65	30	15	230	15	7	1	400	9	10	4
Bahia	1698	91.5	6.8	1.3	70	2100	120	105	40	25	400	50	9	1	360	10	1	15
Bahia	1698	89.3	9.2	1.3	180	250	5	115	35	15	600	60	4	1	375	8	1	10
<i>Ingots</i>																		
Cuyaba	1821	97.1	2.5	0.3			15	15		4	560	2						
Goias	1814	98.5	1.0	0.3			615				30							
Mato Grosso	1817	95.4	4.5	0.0	125		8	9		10	400	2						
Sabara	1807	86.7	11.0	0.1		910	2.1%			71	415	9						
	1809	91.8	5.6	0.4		415	2.1%		15	106	315							
	1814	92.3	7.6	0.1			10			3	355	2						
	1816	91.7	7.7	0.2		110	3160			3	180	60						
	1817	88.4	11.4	0.1			40			16	25	140						
Serro Frio	1809	97.0	2.9	0.0		565	125	3			20	1						
	1811	96.4	3.3	0.1		325	1465	9			260	4						
	1818	97.7	2.2	0.0		85	45	5		1	100	1						
Ouro Preto	1808	96.7	2.9	0.3		45	410				25							
	1810	99.0	0.9	0.1			8			1	95	1						
	1818	91.2	8.5	0.1		50	2345			20	55	85						
	1818	91.7	1.9	6.3		500	155		5	2	220	10						
Peru	1751	90.8	5.9	3.1	110	7	1	120	20	11	950	15						
	1753	90.5	7.0	2.4	35	100	1	115	20	15	605	40				4	4	10
	1758	91.2	8.0	0.6	100	60	1	250	45	44	720	15						
	1761	91.4	6.7	1.8	100	1	1	130	35	22		65						
	1766	87.5	10.4	2.0		5	1	100	15	3	1245	6					20	
	1809	87.2	7.0	5.6		10	1	55	55	47	1615	10					6	
	1817	85.2	4.2	10.3		1670	15	55	110	63	1465	5					3	
Mexico	1736	87.7	8.4	3.9		1	1	3	25	1	280	10			110		8	
	1745	89.3	5.7	4.9		1	1	3	5	1	130	3					7	
	1757	88.9	8.6	2.5		1	115	1	1	1	60	1	3				15	
	1786	87.6	5.6	6.7		5	1	2	20	1	70	3					2	
	1809	84.4	10.3	4.8		3100	35	25	30	17	1870	10	25				4	
	1816	84.3	4.2	11.5		5	1	8	20	0	100	10					2	
	1818	65.8	25.4	8.4	240	3885	20	45	35	27	190	50						
<b>Colombia</b>																		
Cartagena	séc.XVI	89.7	10.0	0.1	300	60	3	500	95	27	770	30						
	1793	86.6	12.7	0.4	185	355	100	670	100	21	1200	80	20			4		35
	1838	87.9	11.7	0.1	520	160	8	500	185	32	1500	20						
	1848	91.1	0.8	8.0		390	150	30	60	70		30	55		50		2	
Popayan	1776	38.2	5.5	4.4	825	52%	310	15	15	14	1300		8	1	15			
	1792	88.7	10.1	0.8	45	2400	95	165	40	17	1290	50	10			4	3	20
	1808	86.7	9.4	3.4	50	3170	45	50	35	9	1960	10	15		15		8	
Bogota	1767	92.2	7.6	0.1		935	40	265	50	4	770	5	5		10			
	1816	85.1	14.0	0.6	170	520	10	300	85	12	1430	20	4					
	1820	84.1	15.2	0.3		2355	25	430	40	18	1520	55	2					
	1833	88.1	11.3	0.3	110	190	6	1300	235	94	1300	30						
	1848	87.4	12.1	0.3	130	320	15	820	90	29	1000	30						
Chile	1750	89.7	8.5	1.8		1	1	80	15	7	645	25	10		150		2	
	1788	88.8	7.7	3.4	45			20	40	35	990	125	20			3	10	60
	1800	86.7	9.5	3.5		7	1	120	15	10	2050	20	10				3	
	1811	84.5	11.5	3.7	110	10	1	155	10	9	2400	115	40				3	
<b>Bolivia</b>																		
Potosi	1782	86.7	7.4	5.7	35			45	30	11	1560	45				15	3	35

Where data are missing this is because the elements were under the detection limits.



Table 2  
Summarized gold production of the Latin American countries concerned by this work from the 16th century to 1800 (from Ref. [32])

	Period	Quantity (fine ounces)	World total (%)
Mexico	1521–1600	771,618	3.36
	1601–1700	1,231,373	4.27
	1701–1800	2,932,148	4.79
Bolivia	1545–1600	1,800,442	7.84
	1601–1700	3,323,677	11.59
	1701–1800	2,314,853	3.78
Brazil	1681–1700	482,261	1.67
	1701–1800	27,006,623	44.12
Chile	1545–1600	1,543,236	6.72
	1601–1700	1,125,276	3.90
	1701–1800	2,764,964	4.52
Colombia	1537–1600	4,115,295	17.91
	1601–1700	11,252,760	39.01
	1701–1800	15,110,849	24.69
Peru	1533–1600	745,898	3.25
	1601–1700	1,607,535	5.57
	1701–1800	1,768,290	2.89

by such a technique. As expected, considering its high PGE contents, Colombian gold has higher concentrations of rhodium and iridium. We can also notice in Table 3 that Mexican gold has lower gallium and cadmium contents. Elements close to nickel exhibit dispersed values due to the composition of the ICP-MS extraction cones.

It is difficult to compare the concentrations obtained by LA-ICP-MS and proton activation analysis (PAA). Elements such as germanium, rhenium, osmium, bismuth, thorium, iridium, etc. are not measured by PAA. In Gondonneau et al. [11], 400 gold coins analyzed by both techniques showed that zinc, platinum, tin, palladium, and particularly lead and arsenic may present

dispersed concentrations. This dispersion is connected to laser ablation<sup>2</sup> as the concentrations obtained by ICP-MS on about 25 ancient gold samples dissolved in the British Museum research laboratory [11] showed very good agreement with PAA.

Our data suggest that the coins issued by the first Bahia mint were made with a mixture containing Colombian gold, but we could not show where the other gold comes from, and in fact several countries of Table 2 could have supplied the metal. In Fig. 3 we notice that some of the Peruvian coins are close to the Brazilian coins and also in Fig. 4 in terms of tin and antimony. However, no chemical group is clearly defined for Peru and its production was rather low in the 17th century (Table 2).

### 3.2. The silver coinages

Most authors agree with the fact that the silver struck in South America came through Rio de la Plata from ancient Peru—more precisely Potosi in present Bolivia—and from Mexico. Fig. 2 shows that between 1694 and 1698 the Potosian production had already decreased while the Mexican mines started to produce a large quantity of silver which in the 18th century was about three times the highest production of Potosi.

The chemical characterization of silver from the region of Potosi (ruby silver type) [37] was done by Guerra several years ago [14] using TFNA. In Fig. 5 we can see that the quantification of a single element, indium, expressed in ppm divided by the silver content expressed in weight percent was sufficient to determine the date of arrival of Potosian silver in Spain. The same type of study was done for several French and Italian mints in order to explain the inflation observed in Europe in the 16th century [14–16,19].

Fig. 6 shows that the concentrations of gold and indium separate Mexican from Potosian silver. A few

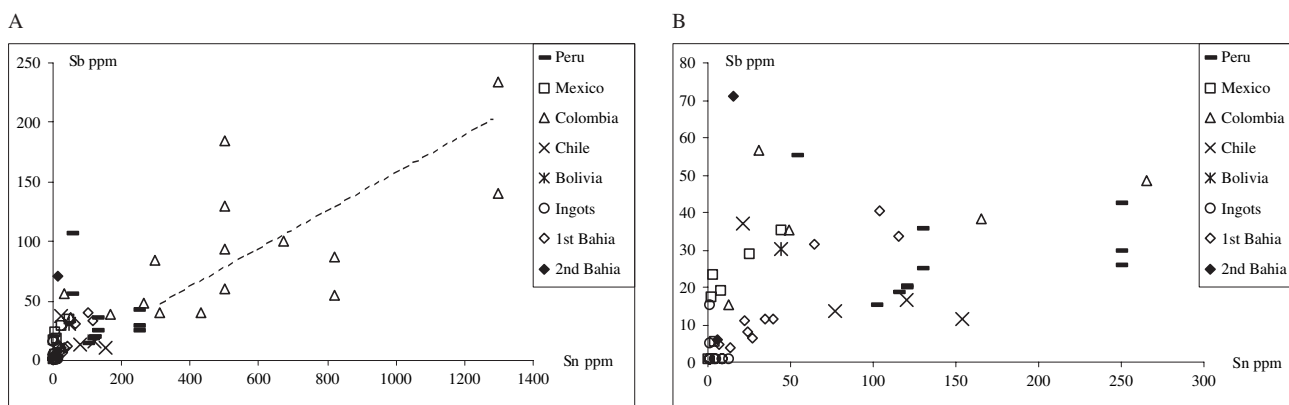


Fig. 4. Tin and antimony concentrations for all the analyzed gold coins and ingots. (A) All samples; (B) expanded zone of lower contents. For Brazil the ingots, the first Bahia mint and the second Bahia mint are represented separately.

Table 3  
Concentrations in ppm obtained for some trace elements measured for a group of gold coins analyzed by LA-ICP-MS

	Date	Co	Ni	Ga	Ge	Ru	Rh	Cd	In	Re	Os	Ir	Tl	Bi	Th	U	
Peru	1751	2	11	1	0.4	0.1		0.1	0.04	1	0.1	0.2	0.1	6	0.01		
	1758	3	8	1	1	0.05	0.4	0.4	0.1	0.2		1	0.1	6	0.01	0.01	
	1761	3	10	0.5	20	0.1		0.2	0.04		0.04	0.2	0.1	2	0.01	0.003	
	1766	1	0.4	1	3	0.1	0.02	0.1			0.1	0.2	0.1	0.4			
	1809	1	17	1	17	0.3		0.3	0.03	2	4	0.3	0.1	0.2			
	1817	1	35	1	20	1	5	0.1	0.01	1	23	22	0.1	1		0.01	
Mexico	1736	0.2	4	1	13	0.1		0.01	0.03		1	0.003	0.2	14	0.01	0.003	
	1745	0.3	3	0.3	24	0.2		0.04	0.01		0.1	0.01	0.03	1	0.1	0.01	
	1757		0.2	0.1		0.1	2	0.02	0.01		0.1	0.004	0.1	1		0.002	
	1786	0.1	1	0.2	8	0.3		0.1	0.02		0.3	0.1	0.1	3		0.01	
	1809	30	20	2		1	8	1	0.1			13		14			
	1816	0.2	3	0.4		0.4		0.04		2	0.1	0.1	0.1	5		0.001	
	1818	1	23	0.1		0.4	9	0.4	0.01			10		26		0.02	
	Colombia	Cartagena	1500	1	5	1	0.3			1			0.5	0.2	9	0.003	0.01
1838			2	17	2	0.2	0.2	2	0.1	1	0.3	0.2	2	0.1	6	0.005	0.01
1848			2	130	3	3	0.5	2	0.3	0.03	0.3	3	9	0.1	2	0.03	0.004
Popayan		1808	8	13	3	7	3	15	0.4		0.1	52	47	0.2	13		0.02
		Bogota	1767	1	7	1	9	0.2	5	1		0.1	5	0.1	7		0.01
Bogota		1816	1	10	1	8	1	3	0.2		1	0.1	21	0.04	9		0.0002
		1820	1	10	1	3	1	14	0.3		0.2	0.1	19	0.1	6		0.003
		1833	2	11	2	1	0.2	2	1	0.5	0.1		2	0.1	12		0.001
		1843	2	8	2	1	0.2	3	1	1	0.1	0.1	3	0.01	17	0.01	0.01
		Chile	1750	1	1	1		0.1		0.2			0.1	0.004	0.1	16	0.03
	1800	10	10	2	8	0.1		0.1		1	0.2	0.1	0.1	4		0.1	
	1811	6	10	1	15	0.1		0.4		2	2	0.1		26	0.01		

Where data are missing this is because the elements were under the detection limits.

coins struck in Lima are not made of pure Potosian silver, except the only coin struck in the 17th century. However, these two elements are not adequate to characterize the Mexican silver. In fact, if we consider the coins struck in Spain before Philip II we observe gold

and indium contents equivalent to those of the Mexican coins [14].

A new set of South American and Mexican coins together with some coins already studied by TFNAA were analyzed by PAA. A few coins were also analyzed

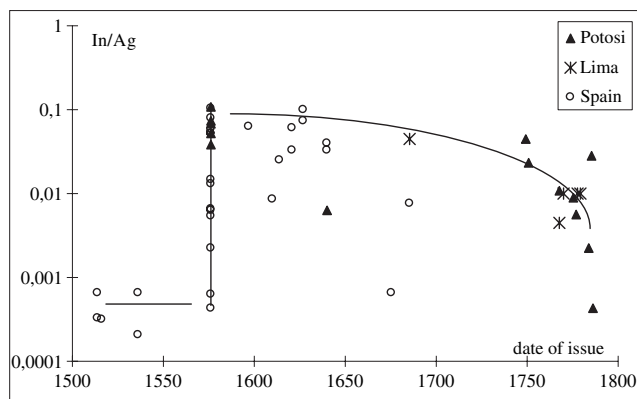


Fig. 5. Ratio of indium concentrations in ppm to silver content in percent measured by TFNAA for the silver coins struck in Potosi, Lima and Spain as a function of the date of issue. Potosian silver arrived in Spain under Philip II.

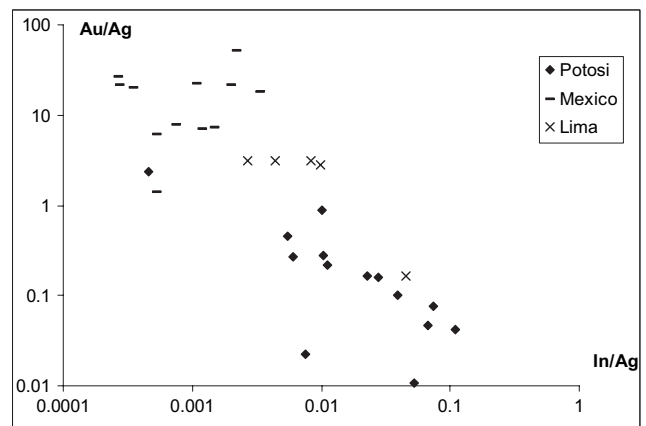


Fig. 6. Ratio of indium and gold concentrations in ppm to silver contents in percent measured by TFNAA for the coins struck in Potosi, Mexico and Lima. The coins struck in Lima in the 18th century are not of the 'Potosian type'.

Table 4  
Composition of the silver coins obtained by PAA and TFNAA

	Date	%			ppm								
		Ag	Cu	Pb	Au	As	Sn	Sb	Zn	Hg	Ti	Fe	In
<b>Afonso VI</b>													
	1656–1683												
PII3		89.4	10.5	0.11	7	310	1495	290	55	185	2	159	ND
PII4		92.5	7.0	0.40	440	45	120	180			2	84	ND
PII6		95.2	4.7	0.13	4	260	60	100	25		2	73	ND
PII10		92.6	7.0	0.35	270	55	140	160	95	140	5	89	ND
PII11		91.8	7.9	0.27	4	845	210	415	70		15	968	ND
<b>Bahia</b>													
PII2	1695	93.7	6.0	0.23	415	330	360	270	35	40	5	99	ND
PII17	1695	91.6	8.1	0.20	385	205	155	200	20	40	1	39	ND
PII20	1695	94.3	5.4	0.22	420	225	290	215	25	90	1	33	ND
PII18	1695	92.8	6.9	0.22	290	360	195	230		200	1	58	ND
PII20	1695	94.8	5.0	0.21	340	235	265	260	20		1	100	ND
PII22	1695	91.4	8.3	0.23	460	460	280	210	25		1	37	ND
PII15	1695	93.2	6.5	0.21	320	345	290	230			1	327	ND
PII8	1695	93.1	6.6	0.20	340	345	215	170	40	135	1	71	ND
PII5	1696	92.6	6.9	0.40	370	115	275	230	45	65	5	87	ND
PII16	1696	92.2	7.6	0.22	370	360	310	240	20	30	1	104	ND
PII7	1696	95.2	4.6	0.21	250	215	245	225	10		2	96	ND
PII14	1696	93.7	6.0	0.22	300	380	340	235	15		1	113	ND
PII19	1697	94.6	5.2	0.18	420	210	180	180			2	34	ND
PII9	1698	94.8	5.0	0.14	220	160	275	105	100	95	1	547	ND
PII1	1698	91.9	7.8	0.16	210	515	210	270	45		1	50	ND
PII12	?	93.3	6.5	0.18	235	440	350	225	20		1	54	ND
PII13	?	92.4	7.3	0.20	280	510	330	240	30	80	2	220	ND
<b>Guatemala</b>													
G10	1808	92.2	7.7	0.06	870	25	15	40	10		1	62	ND
<b>Potosi</b>													
225	1556–1598	90.8	9	0.17	4	300	850	240	20	145	7	170	6.2
231	1556–1598	90.7	9.2	0.13	7	625	630	185	95	60	9	510	6.6
2348	1556–1598	92.5	7.4	0.10	9	75	2530	185	175	350	20	880	3.7
b1	1556–1598	91.9	7.8	ND	1	2280	ND	160	ND	ND	ND	ND	4.9
2340	1556–1598	93.2	6.4	ND	4	90	ND	440	ND	ND	ND	ND	10.2
767	1621–1665	94.1	5.6	0.64	25	2290	410	535	280	150	7	160	0.6
b2	1749	90.8	9.2	ND	15	720	ND	280	ND	ND	ND	ND	2.1
b3	1751	89.2	10.7	ND	2	155	ND	45	ND	ND	ND	ND	0.7
501	1768	91.2	8.6	ND	20	1995	ND	130	ND	ND	ND	ND	1.0
502	1775	89.7	10.2	ND	25	440	ND	225	ND	ND	ND	ND	0.9
498	1777	88.6	11.4	ND	40	295	ND	100	ND	ND	ND	ND	0.5
499	1784	91	8.8	ND	80	1170	ND	70	ND	ND	ND	ND	0.9
255	1785	92.9	6.8	ND	15	1990	ND	620	ND	ND	ND	ND	2.6
257	1786	93.4	6.3	ND	220	188	ND	395	ND	ND	ND	ND	0.04
<b>Lima</b>													
342a	1685	91.3	5.6	ND	15	2075	ND	330	ND	ND	ND	ND	4.1
500	1768	90.4	9.6	ND	285	100	ND	110	ND	ND	ND	ND	0.8
503	1770	91	8.9	ND	285	260	ND	135	ND	ND	ND	ND	0.4
497	1778	89.3	10.6	ND	275	145	ND	95	ND	ND	ND	ND	0.2
504	1778	89.4	10.5	ND	250	410	ND	320	ND	ND	ND	ND	0.9
L8	1788	91.6	8.3	0.02	100	990	910	40					ND
CZ4	1824	90.8	9.1	0.01	215	1335	425	10					ND
<b>Mexico</b>													
625	1516–1556	94.5	5.2	1.30	2055	85	160	115	10	160	4	70	0.2
627	1516–1556	94	5.5	0.92	4870	160	30	5	55	35	6	59	0.2
629	1516–1556	94.3	5.6	0.76	580	85		230	80	20	6	78	0.1
630	1516–1556	94.1	5.8	0.96	725	30	25	70	15		8	142	0.1
631	1516–1556	93.6	6.4	0.38	675	55		20	20	470	3	100	0.1
632	1516–1556	93.8	6.1	0.15	130	60	40	125	80	140	20	97	0.1

(continued on next page)



Table 4 (continued)

	Date	%			ppm								
		Ag	Cu	Pb	Au	As	Sn	Sb	Zn	Hg	Ti	Fe	In
200	1516–1556	93.7	6.1	ND	2500	35	ND	175	ND	ND	ND	ND	0.0
201	1516–1556	94.2	5.6	ND	1890	40	ND	110	ND	ND	ND	ND	0.0
201	1516–1556	94.1	5.7	ND	2015	45	ND	185	ND	ND	ND	ND	0.0
227	1556–1598	93	6.9	ND	640	30	ND	10	ND	ND	ND	ND	0.1
228	1556–1598	92.7	7.1	ND	1670	10	ND	20	ND	ND	ND	ND	0.3
2346	1556–1598	93	6.8	0.92	2080	2		4	15	40	8	117	0.1
M3	1758	92.5	7.3	0.12	560	4		4	60		2	61	ND
M7	1556–1598	93.5	5.2	1.07	2000	3		20	40	70	2	71	ND
Z9	1821	91.6	7.6	0.55	2155	3		25		110	2	17	ND

Major elements, arsenic and antimony were measured by both techniques; tin, lead, zinc, mercury, titanium and iron were measured by PAA; indium was measured by TFNAA. Where data are missing this is because the elements were under the detection limits. 'ND' indicates that the element was not determined.

by LA-ICP-MS (see Table 4). In Fig. 7 we observe the ratio of antimony to silver as a function of the ratio of gold to silver—antimony and gold in ppm and silver in percent. Potosian silver, with high antimony and low gold contents, forms one chemical group while the Mexican coins form a different chemical group with high contents of gold and low contents of antimony.

The highest contents of antimony found in the Mexican coins correspond to the issues of the first half of the 16th century, before the introduction of the amalgamation technique in South America. Even if lead seems to be constant and mercury does not increase, we must mention that cupellation was replaced by amalgamation in 1570–1572 in Potosi (introduced by Pedro Fernando Velasco during the visit of the Spanish Vice-Roy Francisco de Toledo) and in 1555 in Mexico (introduced by Bartolomeo de Medina). Several remains of the use of amalgamation can still be found in South America [6,36]. As antimony is not correlated with copper, the change on the trace elements composition could come from the silver (antimony is expected in the case of dark ruby silver also known as pyrargyrite). We

suggest the use of a different silver ore<sup>5</sup> before the increase of the Mexican mines production.

The Brazilian group in Fig. 7 is very homogeneous and lies between the Mexican and the Potosian chemical groups together with a group of coins from Lima and two of the five Portuguese coins with Brazilian mintmarks, struck in the 17th century under Alphonse VI. The three other Portuguese coins struck under Alphonse VI as well as one from Lima are in the group of Potosi.<sup>6</sup> If we consider the concentration of tin (measured only by PAA) in Fig. 8 we find the same chemical separation as for antimony. Only coin 257 issued in Potosi has an antimony concentration close to that of the Brazilian coins. The same situation is observed for the gold concentration in Fig. 6. This coin could have been issued more recently (the date could not be read on the coin) when Potosi mines were almost exhausted.

The large amounts of Peruvian silver coins circulating in Brazil in the 17th century are evidenced by a demand of the Brazilian governor who in 1651 asked for the opening of one mint in Brazil in order to melt those coins [35]. We also know that Potosian silver reached several European countries and that it travelled through Rio de la Plata to Brazil. We can suggest that this metal became part of the Brazilian stocks. However, Bahia did not strike a pure Potosian silver.

Contrary to gold, silver was not massively exploited in many American regions. Mexico and Peru (ancient Peru includes also present Bolivia) were the most important Latin American silver producers [36]. No silver mines were known in Brazil and Portugal, and as previously mentioned in 1674 King Peter II asked the inhabitants of São Paulo to look for silver in their lands. By the end of the 17th century the Mexican silver mines

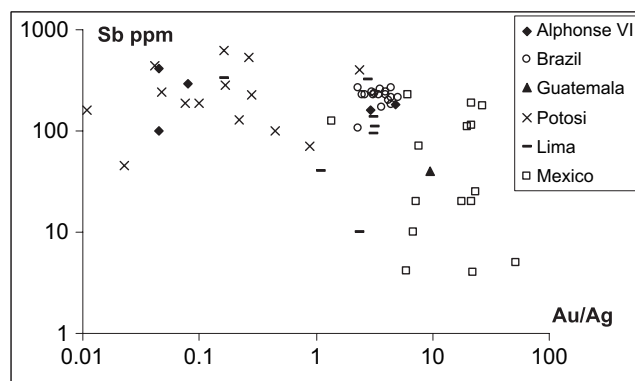


Fig. 7. Antimony concentrations as a function of the ratio of gold content in ppm to silver concentration in percent for all the silver coins.

<sup>5</sup> We could suggest the use of light ruby silver (proustite) instead of dark ruby silver, but the first ore is more common than the latter.

<sup>6</sup> Considering the results obtained for Spain, France and Italy, it is expected that Potosian silver will be found in the 17th century Portuguese issues.

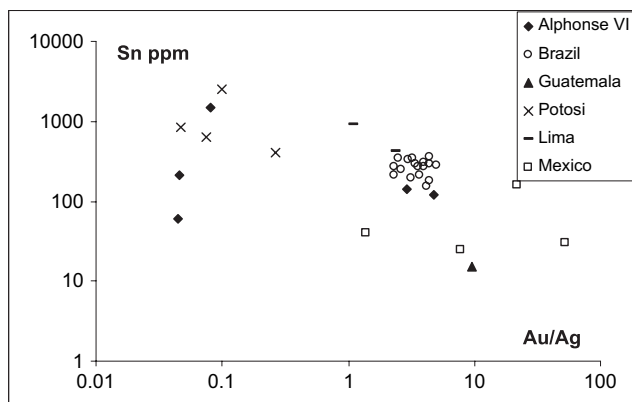


Fig. 8. Tin concentrations measured by PAA as a function of the ratio of gold content in ppm to silver concentration in percent for some of the silver coins.

were producing about 4–5 million pesos and Potosi about 2–3 million pesos (Fig. 2). However, the Bahia mint struck from 1694 to 1698, when it was transferred to Rio de Janeiro after exhaustion of the stocks. This means that the silver and gold supplies already existed in those states and that the coinages could have been issued with the metals supplied by the most important sources of the 16th and 17th centuries. It is possible that Mexico also provided Brazil. The use of a mixture of Mexican and Potosian silver would explain our analytical results.

The only Guatemalan coin analyzed in this work matches the composition of the Mexican coins in Figs. 7 and 8. This coin, issued in 1808, could have been made with Mexican silver, Mexico being the most important South American producer in the beginning of the 19th century.

In Fig. 9 the arsenic content in ppm is plotted as a function of the copper content in percent. We can observe that these elements are correlated for Brazil and may be anti-correlated for Mexico. We could suggest the importation of copper from different sources. We know that the Portuguese mines of Alentejo produced a copper characterized by high concentrations of arsenic [21]. However, Portugal was already unable to cover the copper needs in India and this metal was lacking in the Lisbon mint [9] (p. 49).

In order to look for new trace elements a small group of coins was analyzed by LA-ICP-MS (Table 5). The concentrations given in Table 5 are estimated by extrapolation based on the closest element for which we have a standard.<sup>7</sup> No complementary information on the silver provenance was obtained by this technique. We can observe that only bismuth seems to show

significant differences: coins from Lima have very low contents of bismuth while Mexican coins (together with two coins from Brazil) show high contents of this element.

#### 4. Conclusions

The analysis of a group of silver and gold coins issued from the 16th to the 19th century by different Latin American mints showed several trends.

As expected, countries owning important gold and silver mines strike purely local metals, including Bolivia, Peru, Colombia, Mexico and Brazil. Otherwise it seems that a mixture of metals is minted. If some elements were identified as characteristics of the most known South American golds and silvers we could show with this work that monetary metals have the following characteristics: Colombian gold with high platinum contents also has high concentrations of tin and antimony; Brazilian gold with high palladium contents also has high concentrations of platinum; Bolivian silver with high indium contents also has low concentrations of gold and high concentrations of antimony and tin; and Mexican silver has high gold and low antimony and tin contents.

Based on these chemical groups we tried to identify the silver and gold struck from 1694 to 1698 in the first Brazilian mint, Bahia. For this purpose we analyzed coins from the cited countries as well as gold ingots from Brazil and gold coins from Chile. We could show that Peruvian and Chilean gold present low palladium and platinum contents while Mexican coins present quite dispersed concentrations for trace elements. The arrival of Colombian and Brazilian gold in Mexico could explain this dispersion.

If the second Bahia mint issued as expected gold coins with compositions matching those of the Brazilian ingots, gold and silver struck in the first Bahia mint seem to be a mixture of Latin American metals. In the case of gold the high production of the Colombian mines in the 17th century, and the fact that two Brazilian coins clearly fit with the high platinum content Colombian coins and the others with the low platinum content Colombian coins seem to indicate that Colombian gold reached the Brazilian stocks. We must mention that a few Peruvian coins also match the group of the Brazilian coins.

However, in contrast to Brazilian coins, Colombian coins have high tin and antimony contents. This could suggest the use of a gold mixture in Bahia. We could find some Peruvian specimens in the same situation.

In the case of silver the high productions of the Potosian mines and the documents showing the large amounts of Peruvian coins circulating in the 17th century in Brazil suggest that this metal reached the

<sup>7</sup> PAA and LA-ICP-MS have a better agreement for silver than for gold. However, we must be aware of the lack of trace element standards. These results were published in 2003 by Gondonneau, Guerra and Cowell [11].

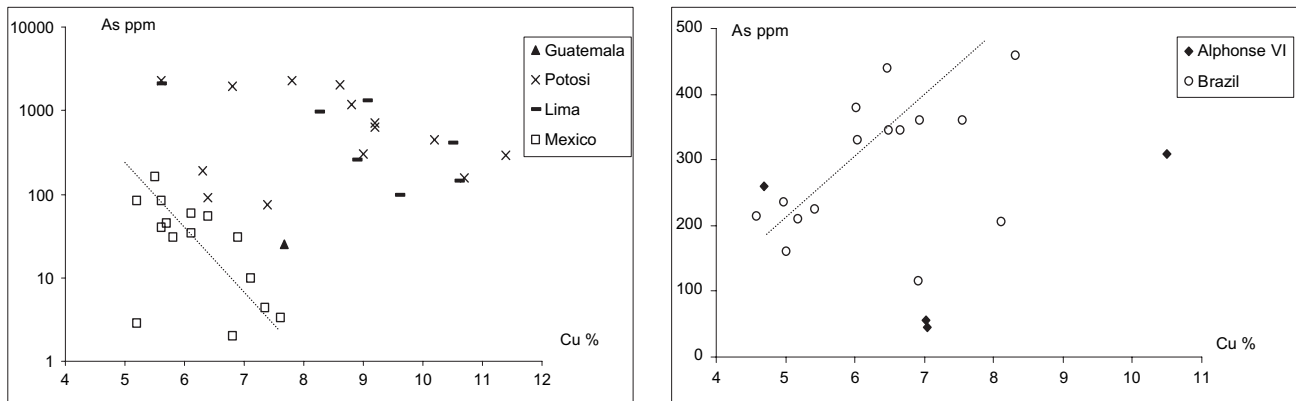


Fig. 9. Arsenic concentrations in ppm as a function of the copper content in percent for all the silver coins, with two different scales.

Brazilian stocks. However, the Bahia silver does not have the exact characteristics of the Potosian silver. The gold, tin and antimony contents place the Brazilian coins together with some issues from Lima in a chemical group situated between Potosi and Mexico. We must mention that the other coins from Lima match either the Mexican or the Potosian group.

Potosi and Mexico produced equivalent amounts of silver by the end of the 17th century. We could suggest that a mixture of Mexican and Potosian silver was used to strike the silver coins of the first Bahia mint.

The LA-ICP-MS analysis performed on a small group of gold and silver coins did not show new characteristic trace elements.

Table 5

Concentrations in ppm obtained for some trace elements measured for a group of silver coins analyzed by LA-ICP-MS

	Date	ppm							
		Ru	Rh	Pd	Cd	In	Ir	Pt	Bi
<b>Afonso VI</b>									
PII3	1656–1667		0.8	0.5	1	9	0.3	0.01	0.01
PII4			0.9	0.6	1	11	0.2	0.01	0.01
PII6		0.3	0.7	0.3	1		0.2	0.24	0.19
PII10		0.1	0.4	0.3	1	2	0.1	0.03	0.03
PII11		0.1	0.4	0.3	1	3	0.05	0.02	0.03
<b>Bahia</b>									
PII2	1695	0.1	0.6	0.2	35	7	0.8	0.04	0.05
PII5	1696	0.3	0.5	0.2	4	9	0.6	0.17	0.22
PII8	1695	0.3	0.4	0.2	3	7	0.3	0.06	0.07
PII7	1696		0.4	0.2	0.4			0.02	0.05
PII1	1698		0.7	0.3	1	6		0.04	0.06
PII9	1698		0.5	0.2	3	6		0.02	0.06
PII12	?		0.4	0.2	6	19	0.9	0.14	0.22
PII13	?	0.1	0.9	0.2		10			0.04
<b>Guatemala</b>									
G10	1808		0.7	0.3	3	2		0.04	0.02
<b>Lima</b>									
L8	1788	0.1	0.7	0.3	2			0.01	0.0002
CZ4	1824	0.2	0.6	0.2	11			0.02	0.0045
<b>Mexico</b>									
M7	Ph II		0.7	0.2	1	2		0.02	0.14
M3	1758		0.7	0.2	1			0.03	0.34
Z9	1821		0.4	0.3	22	2		0.01	0.28

Where data are missing this is because the elements were under the detection limits.

## Acknowledgements

The author is grateful to C.M. Costa and F.A. Magro from the Associação Numismática de Portugal for providing most of the analyzed coins and the Brazilian ingots as well as for all the help with the historical development, to Alexandra Gondonneau for the support given during the analysis, to Michael Cowell from the British Museum for the ICP-MS samples, to the Bibliothèque Nationale de France and to the CERI and IRAMAT CNRS laboratories.

## References

- [1] J.-N. Barrandon, J.-L. Debrun, M. Hours, Application des Réactions Nucléaires Induites par des Protons de Moyenne Énergie à la Caractérisation des Ors Anciens, *Accademia Nazionale dei Lincei*, Rome, 1976, pp. 77–85.
- [2] D. Brading, H.E. Cross, Colonial silver mining: Mexico and Peru, *Hispanic American Historical Review* 52 (4) (1972) 545–579.
- [3] F. Braudel, *La Méditerranée et le Monde Méditerranéen à l'Époque de Philippe II*, A. Colin, Paris, 1966.
- [4] P.T. Craddock, Historical survey of gold refining. Parts 1 and 2, in: A. Ramage, P.T. Craddock (Eds.), *Kings Croesus' Gold*, The Trustees of the British Museum, British Museum Press, London, 2000, pp. 27–71.
- [5] H.E. Cross, South American bullion production and export 1550–1750, in: J.F. Richards (Ed.), *Precious Metals in the Later Medieval and Early Modern Worlds*, Carolina Academic Press, Durham, NC, 1983, pp. 397–423.
- [6] E. Cumenge, E. Fuchs, *L'Or: Ses Propriétés, ses Gisements et son Extraction*, A. Lahure, Paris, 1900.

- [7] L. De Launay, *Traité des Gîtes Minéraux et Métallifères*, Librairie Polytechnique Ch. Beranger, Paris/Liège, 1913.
- [8] E. Fuchs, L. De Launay, *Traité des Gîtes Minéraux et Métallifères*, Baudry et Cie, Paris, 1893.
- [9] V.M. Godinho, *Os Descobrimentos e a Economia Mundial*, vol. V, second ed., Editorial Presença, Lisbon, 1985.
- [10] A. Gondouneau, M.F. Guerra, The analysis of gold by ICP-MS with an UV laser. Application to the study of the gold currency of the Muslim West Africa, in: S. Young, A. Pollard, P. Budd, R.A. Ixer (Eds.), *Metals in Antiquity*, BAR International Series 792, Archaeopress, Oxford, 1999, pp. 262–270.
- [11] A. Gondouneau, M.F. Guerra, M.R. Cowell, Searching for the provenance of gold. The methodology of gold analysis by ICP-MS: first developments, in: *Proceedings of Archaeometry 2000*, CD-ROM, University of Mexico, 2001.
- [12] A. Gondouneau, M.F. Guerra, The circulation of precious metals in the Arabic Empire: the case of the Near and the Middle East, *Archaeometry* 44 (4) (2002) 473–599.
- [13] M.F. Guerra, J.-N. Barrandon, Thermal neutron activation analysis of archaeological artifacts using a cyclotron, in: *The Archaeometry Laboratory* (Ed.), *Proceedings of Archaeometry '88*, University of Toronto Press, Toronto, 1988, pp. 262–268.
- [14] M.F. Guerra, *Da caracterização dos arqueometais. Análise quantitativa por métodos nucleares e perinucleares*, PhD Thesis, New University of Lisbon, Portugal, 1990.
- [15] M.F. Guerra, J.-N. Barrandon, B. Collin, E. Le Roy Ladurie, C. Morrisson, The diffusion of the silver from Potosi in the 16th century Spanish coinage: a new analytical method, in: P. Budd, B. Chapman, C. Jackson, R.C. Janaway, B.S. Ottaway (Eds.), *Oxbow Monograph 9*, Oxbow Books, Oxford, 1989, pp. 28–35.
- [16] M.F. Guerra, J.-N. Barrandon, E. Le Roy Ladurie, C. Morrisson, B. Collin, The diffusion of the silver from Potosi in the 16th century European coinage, *Archaeometry '90*, Birkhäuser Verlag, Basel, 1991, pp. 11–18.
- [17] M.F. Guerra, A. Gondouneau, J.-N. Barrandon, South American precious metals and the European economy, A scientific adventure in the Discoveries time, *Nuclear Instruments and Methods B136–138* (1998) 875–879.
- [18] M.F. Guerra, J.-N. Barrandon, Ion beam activation analysis with a cyclotron, in: W.A. Oddy, M. Cowell (Eds.), *Metallurgy in Numismatics*, 4, Royal Numismatic Society, SP 10, London, 1998, pp. 15–34.
- [19] M.F. Guerra, The mines of Potosi: a silver Eldorado for the European economy, in: G. Demortier, A. Adriens (Eds.), *Ion Beam Study of Art and Archaeological Objects*, European Communities, Luxembourg, 2000, pp. 88–94.
- [20] J. Jedwab, J. Cassedane, Historical observations on oxygen-bearing compounds of platinum and palladium in Minas Gerais, Brazil, *The Canadian Mineralogist* 36 (1998) 887–893.
- [21] F.C. Magro, M.F. Guerra, J.-N. Barrandon, Les ceitis portugais (XVe–XVIe siècles): composition des alliages utilisés et problèmes numismatiques, *Revue de Numismatique VIe série*, Tome XXXVI, 1994, pp. 199–219.
- [22] N.D. Meeks, M.S. Tite, The analysis of platinum-group element inclusions in gold, *Journal of Archaeological Science* 7 (1980) 267–275.
- [23] P. Meyers, Non-destructive activation analysis of ancient coins using charged particles and fast neutrons, *Archaeometry* 11 (1969) 67–84.
- [24] W.E.L. Minter, Ancient placer gold deposits, in: R.P. Foster (Ed.), *Gold Metallogeny and Exploration*, Chapman and Hall, London, 1993, pp. 283–308.
- [25] Ch. Morrisson, J.-N. Barrandon, C. Morrisson, *Or du Brésil, Monnaie et Croissance en France au XVIIIe Siècle*, Cahiers Ernest-Babelon, 7, CNRS, Paris, 1999.
- [26] V. Noya Pinto, *O Ouro Brasileiro e o Comércio Anglo-Português*, second ed., vol. 371, Brasileira, Edição Nacional, São Paulo, 1979.
- [27] J. Poirier, *Contribution à l'analyse de l'or antique. Application aux monnayages du monde Méditerranéen du Ile au XVI siècles*, PhD Thesis, University of Orléans, France, 1983.
- [28] K. Prober, *Ouro em Pó e em Barras, Meio Circulante no Brasil (1754–1833)*, Paqueta Editora, Rio de Janeiro, 1990.
- [29] Ch.J. Raub, The metallurgy of gold and silver in prehistoric times, in: G. Morteani, P. Northover (Eds.), *Prehistoric Gold in Europe*, Kluwer Academic, Dordrecht, 1995, pp. 243–260.
- [30] E. Ricci, R.L. Hann, Theory and experiment in rapid, sensitive helium-3 activation analysis, *Analytical Chemistry* 37 (1965) 742–748.
- [31] E. Ricci, R.L. Hann, Sensitivities for activation analysis of 15 light elements with 18-MeV helium-3 particles, *Analytical Chemistry* 39 (1967) 794–797.
- [32] R.H. Rigway, Summarized data of gold production. United States Department of Commerce, Bureau of Mines, Economic Paper 6, Washington, 1929.
- [33] P.E. Romero, Platino en el Ecuador precolombino, *Boletín Museo del Oro No. 44–45*, Banco de la República, Santafé de Bogotá, 1998, pp. 159–180.
- [34] D.A. Scott, W. Bray, Pre-Hispanic platinum alloys: their composition and use in Ecuador and Colombia, in: D.A. Scott, P. Meyers (Eds.), *Archaeometry of Pre-Columbian Sites and Artifacts*, The J. Paul Getty Trust, Los Angeles, CA, 1994, pp. 285–322.
- [35] J.V. Serrão, *História de Portugal (1640–1750)*, vol. V, second ed., Verbo Editora, Lisbon, 1982.
- [36] E. Tandeter, L'Argent du Potosi dans l'Amérique Coloniale, *Recherches d'Histoire et de Sciences Sociales*, 76, Ecole des Hautes Etudes en Sciences Sociales Ed, Paris, 1997.
- [37] F.S. Turneure, The Bolivian tin-silver province, *Economic Geology* 66 (2) (1971) 215–225.
- [38] U. Zwicker, An investigation of platinum-group metals in ancient coinage, in: W.A. Oddy, M. Cowell (Eds.), *Metallurgy in Numismatics*, 4, Royal Numismatic Society, 30, London, 1995, pp. 171–201.