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Chemical tracers of Lusitanian amphorae kilns from the Tagus estuary (Portugal)

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ABSTRACT

In this paper, the characterization of Roman amphorae from the Porto dos Cacos (PC) and Quinta do Rouxinol (QR) workshops, in the Tagus estuary, dating to a period between the 1st and 5th century AD was carried out on the basis of instrumental neutron activation analysis data on 260 amphorae fragments, together with mineralogical compositional studies obtained by X-ray diffraction.

Special attention was devoted to the study of the Dressel 14, Almagro 50/51c and Lusitana 3/9 amphorae in an attempt to establish whether or not it is possible to establish any correlation between the composition and typology, and between and within the production centres studied.

A description of the geochemical patterns associated with each production centre was first carried out separately, followed by a discussion comparing the two centres situated in the same sedimentary basin, identifying diagnostic chemical tracers for each one; also, in certain cases, a relative correlation with the typology was achieved. Both the definition of reference groups and the attribution of amphorae to their workshop origin relied on the use of chemometric techniques for data structure analysis, coupled with geochemical data analysis, especially regarding trace element data and its geochemical behaviour and distribution according to the geological environment of the region.

This approach complements and reinforces the conclusions drawn from typological and archaeological analyses. Considering the two kiln sites studied, we may talk of the production of two types of Roman amphorae in the lower Tagus, with the establishment of compositional groups defined according to their corresponding chemical signatures. The products of the PC workshop are characterized by high concentrations of Co, As and U, and low concentrations of Fe, Zn, Sb, Rb, K; QR workshop amphorae show instead higher concentrations of Fe, Sb, and also of Rb and Zn, and lower amounts of U.

These results make it possible to understand the crucial importance of the lower course of the Tagus River within the framework of the analysis of the economy of the estuary at the time. This, in turn, may lead to a better understanding of production and trade within Lusitania and also with other Roman provinces.

1. Introduction

The most abundant artefacts found in any Roman-period archaeological site, and particularly in workshops, are fragments of pottery. Thus, the study of amphorae occupies a distinguished place in Roman ceramics research. Amphorae were the larger two-handled pottery containers of the Greek and Roman epochs, used for the storage and transportation of liquids (i.e. wine, olive oil), as well as other foodstuffs (i.e. fish sauce products, *garum*). Generally, amphorae provide greater insight into socio-economic relations, since they were principally used to transport food products. Transport amphorae were ordinary containers of commodities, so today they are rightfully considered to be important historical documents, representing the most tangible evidence of ancient trade that remains today. The knowledge of the chemical tracer(s) of each workshop may provide insight into ancient transport, trade routes, and manufacturing practices, and also contributes to revealing improvements or decline in technology, as

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well as in raw materials exploitation strategies, such as changes in the exploration of clay pits.

We conducted an interdisciplinary study on Roman amphorae manufactured during the period between the 1st and the 5th century AD at two of the best known prominent workshops among the lower Tagus potteries, the Porto dos Cacos – PC (Alcochete) and the Quinta do Rouxinol – QR (Seixal) potteries. The two production centres were located on the left bank of the Tagus River (Fig. 1). Because of their geographical position, and according to the types of amphorae found at the locations, these workshops were most probably associated chiefly with the preparation of *garum* and fish salted products, in the surrounding areas, namely Lisbon, Troia and Setúbal which were important centres of exportation of fish preserves. Moreover, previous work comparing some amphorae from these production centres with amphorae found at the industrial trading site of Correeiros (located in Lisbon) points to the use of QR amphorae at this fish salting industry (Dias and Prudêncio, 2007; Raposo et al., 2005).

The close connection between these ceramics production centres and areas related to the production and handling of fish-based products is obvious. The exploitation of marine resources in this basin is to be expected, and archaeological remains point to an intense occupation, with fish-salting facilities close to the river mouth and small peripheral units further away, and with pottery centres located upstream, upon the banks of the main river and its tributaries (Fabião, 2004, 2008; Raposo et al., 2005).

In this work, a larger number of samples from each production centre were studied; together with the previously analysed samples, they constitute a large dataset used to establish differences and similarities between the chemical compositions of amphorae from the QR and PC Roman workshops. Therefore, the purpose of this paper is to study (i) whether all amphorae were manufactured with the same type of raw materials from the Tagus basin (ii) whether the presence of certain chemical elements in the ceramic pastes enables the differentiation and chemical tracing of each lower Tagus production centre,. This kind of approach will make available important geochemical patterns associated with the Roman production centres of the Tagus basin, to be used in further studies of provenance and in comparisons with consumption centres, thus providing significant information about the origin and development of industrial complexes and networks in Roman times.

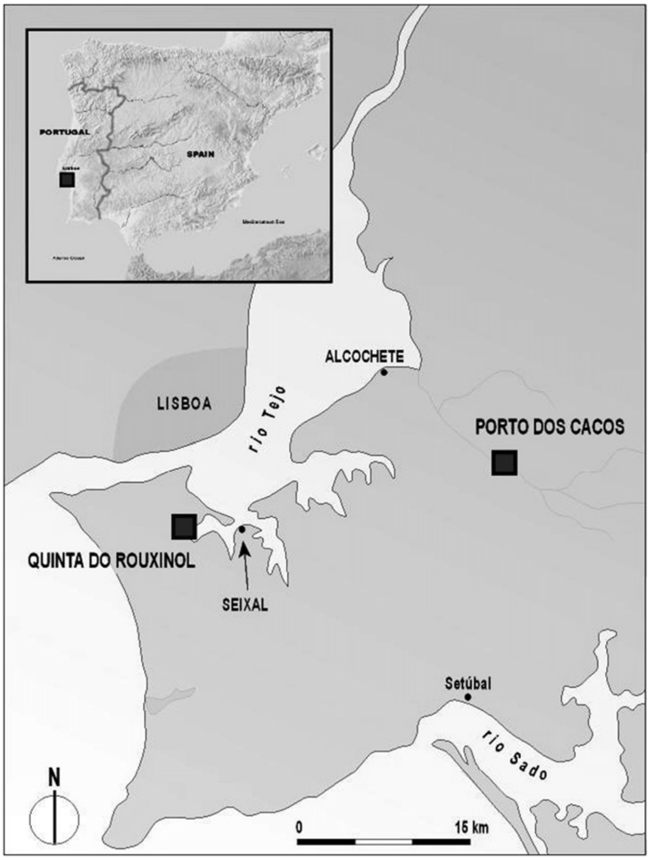

Fig. 1. Location of the Porto dos Cacos and Quinta do Rouxinol workshops.

Archaeometric studies of amphorae from the PC and QR Tagus production centres in comparison with a Sado basin production centre (Herdade do Pinheiro – HP) have already been performed, including a chemical characterization by instrumental neutron activation analysis (INAA) (Cabral and Gouveia, 1984; Cabral et al., 1993–1994, 1996, 2000, 2002; Prudêncio et al., 2003; Dias and Prudêncio, 2007). Some of these works emphasize the difficulty in distinguishing amphorae produced with similar raw materials from the Tertiary Tagus-Sado basin. Other works, including a petrographic identification of ceramic pastes from both basins (Mayet et al., 1996), also pointed to the difficulty in differentiating productions from the two estuaries (Sado and Tagus), due to the fact that they belong to a common Tertiary basin. Nevertheless, the application of adequate statistical treatment to a wide range of chemical analysis results has provided information that disclosed the relations among these production centres by classifying the samples into compositional groups. According to this data, subtle differences were found between them (Prudêncio et al., 2003; Raposo et al., 2005; Dias and Prudêncio, 2007) and within them as well (Dias et al., 2001). In addition, an assay on ceramic raw materials from the lower Tagus kilns has been carried out (Dias et al., 2003) by selecting some clay-supplying deposits from the surrounding area of each site (PC and QR). Chemical and mineralogical data obtained in the framework of that study emphasized the difficulty in establishing source materials of the same geological environment, especially estuarine ones, due to natural homogenization processes inherent to the transportation and deposition of those sediments. Even so, the results pointed to the use of two Mio-Pliocenic clays for amphora production at each site, and a certain similarity was also found between the iron content of QR amphorae and clays near that site.

More recently (Prudêncio et al., 2009), geochemical signatures of five Roman amphora production centres from the lower Sado have been established by the application of multivariate statistical analysis to chemical data obtained by INAA. Reference groups were identified, together with chemical indicators. Some of these sites are part of the same Tertiary Tagus-Sado basin.

Our intention in the present work is to complement the already existing information for the lower Sado basin, and also to provide chemical tracers for the most important Roman amphora production centres of the lower Tagus basin (Porto dos Cacos and Quinta do Rouxinol) identified so far, based on INAA. The methodology used emphasizes the encounter and complementarity between formal studies and chemical characterizations of pastes through the application of multivariate statistical methods necessary in order to quantify the similarities and differences between specimens and groups of specimens. Compositional groups are defined in accordance with the corresponding chemical signatures.

2. The Roman amphora production centres from the lower Tagus basin

The Porto dos Cacos (PC) archaeological site has been partially excavated in the late 1980s and two separate sets of kilns have been identified (Raposo, 1990; Raposo and Duarte, 1996). The first set featured a small kiln (kiln 1), which only maintained the lower part of its combustion chamber and the *praefurnium*. The second set included two other kilns found in close connection, and a possible third one, indicated by the geophysical survey. The second kiln (kiln 2) was completely excavated, showing a larger circular plant structure, with a furnace presenting a floor paved with small sandstone bricks and fragments of tiles. In addition, parts of the *suspensurae* and the grill they supported, together with parts of the vaulted baking chamber, could also be observed. The structure of kiln 3 was only partially excavated (Raposo and Duarte, 1996).

A significant amount of pottery was found at this site, such as domestic pottery, but especially amphorae, mostly used as containers for fish-based products, like the Class 20/21 = Dressel 14, Class 22 = Almagro 50 and Class 23 = Almagro 51c, and probably also for wine, like the Lusitana 3 and Lusitana 9. The entire set points to the abandonment of the PC production centre either at the end of the 4th century or during the first decades of the 5th century AD. It also indicates that Class 23 = Almagro 51c and Class 22 = Almagro 50 amphorae were produced in kilns 1 and 2, the latter also producing the Lusitana 9 form. No identification of the kilns with the early production stages, represented by the Class 20/21 = Dressel 14 and Lusitana 3 forms, was possible, as these were only found in the dumping areas so far (Raposo et al., 1995). Among the sherds belonging to the latter form, it is possible to observe the largest and most significant series of potter markings known so far in Portugal – the cognomen *Germanus* is the most abundantly documented (Guerra, 1996). An interesting but enigmatic feature was found among the infrastructures that supported the use of the kilns, comprising a conspicuous row of 46 Class 20/21 = Dressel 14 amphorae which are carefully placed vertically, defining an area whose function is still indeterminate.

The Quinta do Rouxinol (QR) production centre has also been excavated in the late 1980s (Duarte, 1990; Duarte and Raposo, 1996) and the excavations have revealed two kilns and the possible existence of a third one. The kilns are pear-shaped, and their only remains are the lower parts of the combustion chamber, the *praefurnium* and the bases of the *suspensurae* used to support the grills. Archaeological evidence points to a period between the middle of the 2nd century AD and at least until the end of the 4th century AD when these kilns were functional, producing amphorae of Class 22 = Almagro 50, Class 23 = Almagro 51c and Lusitana 9 form, as well as domestic pottery.

3. Experimental

Amphorae fragments from both sites (PC and QR), representative of the various typologies identified, have been chosen for chemical analysis. A total of 260 samples have been analysed distributed according to the typology for each site as described in Table 1. In the case of the PC workshop, most of the analysed sherds are samples of rims, and only 12 samples are fragments of bottom parts of the amphorae, six of A51c and six others of L3 typology.

The preparation for ceramic chemical analysis begins with the removal of soil or other surface materials from the sample. The procedure involves burring away the surface (inner and outer) with a tungsten carbide gouge, removing surface coatings and other impurities that may be attributed to contamination by weathering or other post-depositional processes. To the same purpose, the ceramic fragment is placed in deionised water for 24 hours, and afterwards for 30 more minutes in boiling clean deionised water. Next, ceramic fragments are dried in the oven at 80 °C, for a week. The interior paste from the ceramic fragment is then ground to a fine powder in an agate mortar, and homogenised. Aliquots of approximately 1 g each of powdered ceramics and reference materials are dried in an oven at 110 °C for 24 hours and stored in a desiccator. Once dried, 200–300 mg of powdered sample are weighed into clean polyethylene vials. Individual samples are prepared for long irradiations together with reference standard samples (GSD-9 and GSS-1) from the Institute of Geophysical and Geochemical Prospecting (IGGE). Reference values are obtained from the data tabulated by Govindaraju (1994).

One long irradiation and two gamma counts are performed. Samples and standards are bundled together in batches of 20 samples and 4 standards, and irradiated in the Portuguese Research Reactor pool for seven hours at a thermal flux of 3.34×10^{12} n cm^{-2} s^{-1}; $\varphi_{epi}/\varphi_{th} = 1.4\%$; $\varphi_{th}/\varphi_{fast} = 12.1$. The bundles are rotated continuously during irradiation, to ensure that all samples receive the same neutron exposure. Iron (Fe) flux monitors are irradiated with the samples to allow corrections due to variation in the neutron flux. Two gamma-ray spectrometers are used: (1) one consisting of a 150 cm^3 coaxial Ge detector connected through a Canberra 2020 amplifier to an Accuspec B (Canberra) multichannel analyser. This system has a FWHM of 1.9 keV at 1.33 MeV; and (2) the other, consisting of a low energy photon detector (LEPD) connected through a Canberra 2020 amplifier to an Accuspec B (Canberra) multichannel analyser. This system has a FWHM of 300 eV at 5.9 keV and of 550 eV at 122 keV. After a four-day decay, the samples are counted for 30 minutes in the high energy detector and 60 minutes in the low energy detector to measure medium-lived elements, including Na, K, Ga, As, Br, La, W, Sm and U. Following an additional four-week decay, the samples are counted again for 2 hours and 30 minutes in the low and high energy detectors, yielding the measurement of the following long-lived elements: Fe, Sc, Cr, Co, Zn, Rb, Sb, Cs, Ba, Zr, Ce, Nd, Eu, Tb, Yb, Lu, Hf, Ta and Th. Corrections were carried out for spectral interference from uranium fission products in the determination of barium, rare earth elements (REE) and zirconium (Gouveia et al., 1987; Martinho et al., 1991). Relative precision and accuracy are, in general, within 5%, and occasionally within 10%.

The mineralogical composition was obtained by X-ray diffraction (XRD), using a Philips X'Pert Pro diffractometer, with a PW 3050/6x goniometer, Cu Kα radiation, and fixed divergence slit, operating at 45 kV and 40 mA. The powdered samples were prepared as non-oriented aggregates and used to obtain the diffraction patterns. Scans were run from 3 to 70° 2θ, using a step size of 0.02° 2θ and a scan step time of 1.20 s. To estimate quantities, we measured the diagnostic reflection areas, considering the full width at half maximum (FWHM) of the main minerals and then weighted by empirical factors or calculated parameters

Table 1
Number of samples analysed for the Porto dos Cacos and Quinta do Rouxinol workshops according to their typology.

	Class 22=Almagro 50	Class 23=Almagro 51c	Lusitana 9	Class 20/21=Dressel 14	Lusitana 3	[A51c/L3]?	Total
Quinta do Rouxinol	49	49	32[a]	–	–	–	130
Porto dos Cacos	3	11	–	38	56[b]	22	130

[a] Six of these Lusitana 9 samples are sub-samples from the same amphora sherd.

[b] Among these Lusitana 3 samples, eight samples bear potters' marks (RVSTICI, TMM, CLARIAMI, AIVNIT (or TINVIA retro), three Germanus (CERF (Germanus), GERMAN (retro) and GERMA(ni)) and one non-identified.

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Table 2
Elemental compositions of Roman amphorae from the Porto dos Cacos workshop (μg/g unless specified otherwise).

Sample	Type	Na_2O%	K_2O%	Fe_2O_3T%	Sc	Cr	Co	Zn	As	Rb	Sb	Cs	Ba	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	Hf	Ta	Th	U
PC-1	L3	0.68	3.44	3.85	13.4	65.2	7.53	78.0	5.10	184	0.58	12.6	504	41.1	81.3	36.3	6.97	1.26	0.95	3.08	0.43	6.49	1.66	14.2	5.08
PC-3	L3	0.77	3.72	4.68	15.3	71.2	9.44	84.7	6.13	196	0.52	15.1	572	47.7	95.7	41.8	8.38	1.37	1.04	3.48	0.46	7.01	1.88	18.0	6.04
PC-5	L3	0.44	3.29	4.58	13.1	63.4	14.5	88.2	14.5	181	0.43	13.2	542	53.7	91.0	50.5	10.9	2.02	1.52	3.97	0.57	6.84	1.81	17.0	6.26
PC-9	L3	0.43	3.28	4.62	12.7	60.4	16.1	83.7	19.3	184	0.56	13.4	536	53.5	84.6	49.8	10.3	1.87	1.38	3.78	0.55	6.69	1.85	16.4	7.45
PC-12	L3	0.41	3.28	4.27	11.3	54.9	11.9	75.5	16.7	169	0.56	11.6	484	49.2	78.9	45.8	9.74	1.62	1.19	3.79	0.45	7.12	1.88	6.90	7.06
PC-14	L3	0.71	3.46	5.01	12.6	59.9	8.69	90.6	6.70	176	0.53	12.3	456	41.1	76.7	36.2	7.17	1.13	0.81	3.26	0.49	6.75	1.77	16.2	4.46
PC-15	L3	0.74	3.37	4.43	12.0	53.4	10.2	76.9	4.63	170	0.74	11.0	433	50.8	99.8	45.2	8.65	1.33	1.05	3.65	0.55	8.19	1.78	8.60	4.40
PC-16	L3	0.42	3.33	4.55	12.6	61.3	23.3	87.9	19.1	172	0.57	13.4	514	55.2	87.1	0.70	10.4	1.95	1.33	3.68	0.63	6.63	1.75	16.4	6.89
PC-20	L3	0.51	3.25	4.96	12.9	71.0	12.7	78.4	16.6	180	0.51	13.5	516	47.3	77.8	42.5	8.65	1.51	1.15	3.78	0.53	7.27	1.87	17.6	6.91
PC-21	L3	0.55	3.28	4.47	12.8	66.8	10.5	67.1	17.9	184	0.47	13.4	476	42.3	84.6	40.5	8.22	1.45	1.20	3.29	0.53	6.69	1.89	17.1	6.15
PC-22	L3	0.38	3.20	4.69	14.3	68.7	17.8	92.1	26.7	189	0.55	15.4	676	32.7	55.6	34.1	7.09	1.38	1.02	3.23	0.53	5.79	1.92	15.5	6.37
PC-23	L3	0.50	3.30	4.33	12.6	64.9	12.1	60.2	19.0	189	0.53	13.2	622	46.4	73.0	42.5	8.79	1.61	1.24	3.63	0.54	7.02	1.94	17.2	6.01
PC-24	L3	0.51	3.25	4.47	12.8	72.7	12.9	67.0	20.7	185	0.49	12.9	572	47.2	76.9	43.4	9.13	1.69	1.23	3.70	0.56	7.40	1.76	17.1	6.82
PC-26	L3	0.43	3.34	4.43	12.7	61.0	13.6	74.3	20.2	186	0.58	13.1	490	48.9	75.0	46.30	9.36	1.70	1.32	3.67	0.54	7.19	1.79	17.4	6.92
PC-29	L3	0.39	3.20	3.99	12.1	57.7	11.7	68.9	16.1	159	0.45	11.3	572	39.9	57.4	34.2	8.20	1.36	1.01	2.85	0.51	5.63	1.54	14.5	6.75
PC-31	L3	0.80	3.40	5.02	12.7	56.0	8.74	87.9	5.86	187	0.67	12.0	493	41.1	78.1	36.9	7.45	1.23	0.97	2.75	0.50	7.82	1.79	17.2	4.41
PC-32	L3	0.42	3.24	4.53	12.6	66.3	19.7	78.7	20.0	182	0.53	12.9	560	50.1	75.9	48.1	9.98	1.93	1.34	3.31	0.61	6.69	1.88	16.0	8.26
PC-34	L3	0.51	3.06	5.86	13.2	60.2	6.97	72.3	10.6	199	0.78	14.1	467	68.0	84.6	47.3	8.61	1.26	0.97	3.22	0.52	8.52	1.84	19.8	5.13
PC-37	L3	0.53	3.36	4.95	14.1	86.1	17.7	86.1	22.2	197	0.43	15.3	577	49.8	93.3	48.6	10.6	2.01	1.41	3.44	0.68	6.61	1.88	16.4	8.00
PC-38	L3	0.73	3.16	4.20	13.1	67.5	11.6	74.9	12.1	178	0.63	12.5	495	46.2	90.1	41.8	8.58	1.48	1.15	3.11	0.56	7.70	1.85	17.1	4.32
PC-39	L3	0.62	3.25	4.52	13.7	65.6	12.4	75.6	18.2	183	0.49	14.0	498	43.2	86.0	41.8	8.90	1.62	1.17	3.21	0.53	6.94	1.89	16.4	6.56
PC-41	L3	0.56	3.24	4.61	13.2	65.5	12.0	72.8	18.4	184	0.44	14.1	606	42.3	80.5	41.6	8.40	1.53	1.10	3.34	0.49	6.77	1.82	16.1	6.98
PC-47	L3	0.54	3.38	3.38	10.8	55.5	10.0	48.5	13.9	137	0.39	9.31	593	29.9	53.5	21.9	5.50	0.77	0.57	2.05	0.33	5.30	1.49	13.3	4.92
PC-48	L3	0.37	3.41	4.60	13.6	66.1	13.4	79.5	21.6	179	0.52	14.1	1030	34.1	55.9	29.2	5.95	1.05	0.90	3.06	0.42	6.39	1.98	16.7	6.56
PC-49	L3	0.36	3.40	4.81	12.7	65.5	17.5	78.9	20.6	191	0.60	12.9	554	43.1	65.2	37.8	7.89	1.54	1.18	3.11	0.51	5.93	1.84	16.1	5.84
PC-50	L3	0.79	3.62	4.68	15.2	75.6	9.57	84.2	5.86	197	0.67	14.6	534	48.6	90.6	40.6	8.41	1.33	1.01	3.17	0.52	6.99	1.99	17.9	5.48
PC-52	L3	0.42	3.20	4.55	13.3	65.8	14.4	80.6	12.7	190	0.53	13.8	485	51.4	88.6	51.0	11.2	2.02	1.49	3.95	0.60	6.99	1.95	16.8	8.57
PC-53	L3	0.33	3.18	4.98	14.0	72.1	16.4	85.8	25.0	191	0.36	13.8	540	55.5	83.0	52.8	11.5	2.11	1.56	4.24	0.66	7.30	2.09	18.3	7.81
PC-54	L3	0.35	3.09	4.83	13.1	65.2	17.8	84.5	18.9	187	0.56	12.8	529	54.5	83.2	50.3	10.8	1.85	1.48	4.01	0.62	7.73	1.86	18.4	7.78
PC-56	L3	0.63	3.13	3.74	12.3	58.1	6.53	60.6	9.16	169	0.44	11.4	573	33.6	68.0	31.0	6.22	0.99	0.85	2.64	0.42	6.57	1.51	15.0	4.59
PC-57	L3	0.46	3.15	4.57	12.1	57.7	12.1	68.1	17.0	184	0.46	12.3	552	41.2	71.8	37.2	7.88	1.26	1.04	3.25	0.50	7.55	1.69	17.5	7.05
PC-58	L3	0.82	3.26	4.25	11.6	55.0	11.7	62.5	6.29	183	0.67	12.0	608	44.7	90.3	42.9	8.70	1.38	1.19	3.67	0.56	7.96	1.81	17.6	3.82
PC-59	L3	0.42	3.36	4.63	13.2	63.6	16.3	81.1	21.4	201	0.50	14.1	485	48.8	75.2	47.2	10.0	1.83	1.41	3.96	0.62	7.03	1.89	17.2	8.05
PC-62	L3	0.34	3.12	5.11	14.4	66.8	17.8	79.3	21.7	202	0.55	14.8	494	58.1	83.7	56.1	12.5	2.27	1.72	4.56	0.69	7.24	1.96	18.0	8.93
PC-63	L3	0.38	3.15	4.47	13.3	63.5	16.4	77.0	18.6	196	0.52	13.7	510	48.2	76.5	50.9	11.1	2.00	1.61	4.06	0.62	6.87	1.82	17.1	8.01
PC-64	L3	0.34	3.29	5.24	14.6	68.6	20.8	78.8	22.4	208	0.60	14.9	515	59.0	85.2	55.2	12.9	2.31	1.69	4.49	0.70	7.56	1.91	18.8	9.14
PC-65	L3	0.34	3.30	5.10	14.4	71.2	17.9	78.3	22.7	209	0.51	14.7	522	59.5	91.3	57.9	11.9	2.26	1.66	4.76	0.68	7.23	1.96	18.4	8.39
PC-66	L3	0.41	3.38	5.05	13.7	70.6	20.2	77.9	21.1	220	0.65	14.8	512	53.8	118	50.7	10.5	2.04	1.42	4.22	0.63	7.03	1.80	16.8	7.69
PC-67	L3	0.39	3.54	5.10	14.2	68.9	15.3	74.4	22.5	209	0.60	14.8	488	58.4	86.8	55.8	11.4	2.11	1.62	4.40	0.69	7.41	2.06	18.6	7.39
PC-68	L3	0.43	3.10	4.41	12.4	59.0	16.8	70.5	15.6	187	0.49	12.7	492	49.7	90.9	46.5	10.2	1.73	1.32	3.41	0.55	7.52	1.77	17.6	7.25
PC-69	L3	0.50	3.33	4.74	13.3	62.4	14.7	76.9	17.8	196	0.59	13.9	455	51.1	82.0	45.4	10.2	1.76	1.31	3.84	0.57	7.17	1.78	17.0	7.63
PC-70	L3	0.25	2.10	4.91	13.9	65.7	15.4	80.5	14.5	199	0.65	14.5	516	37.7	84.8	53.4	7.23	2.09	1.53	4.27	0.64	7.55	2.19	18.6	4.91
PC-118	L3	0.42	3.25	4.89	13.5	66.0	15.9	81.9	19.1	183	0.56	13.4	399	53.1	97.1	52.7	10.5	1.85	1.39	4.10	0.59	7.83	2.03	18.4	7.47
PC-119	L3	0.40	3.21	4.67	13.3	63.6	14.4	85.6	18.4	187	0.61	12.8	392	55.1	95.3	56.4	10.3	2.03	1.48	4.28	0.65	8.19	1.96	18.7	6.29
PC-120	L3	0.67	3.29	4.50	13.9	67.3	9.24	78.1	7.60	176	0.54	12.4	465	50.0	94.4	44.2	7.74	1.31	1.05	3.28	0.51	7.71	1.79	17.8	5.25
PC-121	L3	0.43	3.17	4.65	12.9	61.3	14.2	73.2	16.8	180	0.49	12.5	409	52.0	84.2	49.0	8.88	1.75	1.32	4.05	0.59	8.54	1.86	18.7	5.74
PC-122	L3	0.45	3.36	4.73	12.8	60.7	15.0	78.9	15.4	183	0.53	12.8	442	54.9	94.2	52.7	10.1	1.78	1.31	4.10	0.60	8.55	1.81	19.7	6.59
PC-123	L3	0.45	3.41	4.87	14.2	65.2	17.1	187	20.7	202	0.53	14.7	471	52.8	76.4	46.7	10.4	1.96	1.52	4.53	0.65	7.44	2.07	17.9	7.46
PC-124	L3	0.47	3.31	4.86	13.8	64.0	16.1	84.2	15.5	197	0.49	14.2	414	50.3	76.5	50.1	10.0	1.95	1.46	4.16	0.60	6.75	1.78	16.0	6.17
PC-125	L3	0.48	3.13	4.77	12.7	59.6	10.2	60.2	20.8	184	0.56	13.4	447	44.9	77.8	42.4	8.52	1.35	1.08	3.61	0.52	8.05	1.84	18.3	6.37
PC-126	L3	0.53	3.41	4.74	13.3	64.9	15.3	85.5	15.5	186	0.48	13.3	402	47.4	72.5	45.8	9.38	1.75	1.28	3.77	0.61	6.99	1.82	16.2	6.17
PC-127	L3	0.80	3.46	3.92	11.6	54.5	9.43	74.7	4.29	171	0.60	10.7	496	41.6	82.0	39.9	7.73	1.24	1.02	3.27	0.48	6.90	1.70	16.4	3.81
PC-128	L3	0.38	3.26	5.23	12.6	67.6	16.2	82.5	34.3	188	0.59	12.6	589	43.6	74.6	42.7	8.63	1.73	1.23	3.64	0.53	6.14	1.78	15.3	8.18
PC-129	L3	0.37	3.03	4.67	12.9	60.3	12.5	78.3	16.4	181	0.48	12.7	421	55.2	92.5	52.4	10.5	1.82	1.41	4.30	0.65	8.18	1.72	18.70	6.29
PC-130	L3	0.79	3.41	4.04	11.8	53.5	13.6	73.8	4.45	180	0.62	11.5	438	46.0	84.3	40.3	7.76	1.28	1.04	3.39	0.56	7.73	1.81	16.5	3.73
PC-131	L3	0.45	3.15	4.29	11.7	58.1	13.8	70.8	17.8	174	0.42	11.7	542	41.6	63.4	35.6	7.50	1.30	0.96	3.23	0.46	6.80	1.69	15.8	4.94
PC-11	A51c	0.68	3.08	4.78	11.8	59.4	7.63	74.3	7.38	178	0.66	11.6	445	40.4	78.5	36.1	6.99	1.08	0.96	3.15	0.50	7.44	1.76	17.2	4.14
PC-17	A51c	0.59	3.01	4.33	10.7	51.5	54.4	95.2	10.4	156	0.62	10.5	517	40.6	75.6	34.1	6.65	0.88	0.83	2.67	0.43	8.30	1.61	19.4	4.31
PC-19	A51c	1.01	5.21	4.80	11.5	51.0	8.92	80.1	14.2	167	0.53	10.9	565	59.7	68.8	31.3	10.7	1.01	0.87	2.61	0.42	6.69	1.59	16.1	5.88
PC-28	A51c	0.65	3.28	5.16	13.0	57.2	10.1	83.1	8.04	190	0.69	12.6	437	42.8	82.0	39.8	7.75	1.29	1.06	3.26	0.50	6.93	1.77	16.9	3.94
PC-44	A51c	0.66	3.52	5.21	13.0	59.0	9.19	76.3	8.19	188	0.58	12.7	518	43.1	79.4	37.1	7.19	1.16	0.93	2.99	0.49	7.06	1.73	17.5	4.56
PC-112	A51c	0.78	3.29	4.95	14.6	66.7	11.9	79.5	13.4	190	0.75	14.3	441	48.1	93.3	45.6	9.16	1.50	1.24	3.59	0.54	7.28	1.97	18.0	4.77
PC-113	A51c	0.62	2.87	4.91	13.6	62.7	8.13	63.0	9.92	178	0.66	13.7	361	52.1	100	47.1	9.10	1.50	1.13	3.34	0.49	7.23	1.73	17.3	3.76
PC-114	A51c	0.64	3.37	5.02	12.0	52.4	9.14	77.9	9.25	188	0.69	11.7	378	45.4	91.0	43.1	8.26	1.17	1.06	3.38	0.52	8.36	1.70	20.0	4.46
PC-115	A51c	0.78	3.39	5.32	13.5	58.5	8.96	79.0	7.07	190	0.60	12.9	401	47.4	90.8	41.4	8.38	1.26	1.05	3.45	0.53	8.25	1.92	18.6	4.61
PC-116	A51c	0.68	3.38	4.23	13.8	63.2	8.60	82.4	6.79	178	0.45	12.6	455	47.4	91.7	40.7	7.86	1.31	1.02	3.34	0.53	6.72	1.65	16.4	5.29
PC-117	A51c	0.75	3.24	5.52	13.8	59.6	10.7	93.4	8.50	184	0.65	13.1	392	47.5	92.6	42.7	8.44	1.30	1.10	3.56	0.57	7.16	2.27	18.0	4.47
PC-74	D14	0.68	3.13	4.96	15.6	75.7	9.07	76.0	19.8	200	0.63	16.4	542	43.5	81.4	39.10	7.89	1.37	1.11	3.18	0.47	6.62	2.10	17.7	6.66
PC-75	D14	0.55	3.03	4.62	12.6	67.5	6.69	63.0	22.0	172	0.61	13.3	621	40.7	81.9	36.8	7.50	1.18	0.97	3.27	0.46	7.90	1.89	18.3	6.50
PC-76	D14	0.57	3.16	4.97	13.7	66.3	7.66	71.0	25.0	182	0.56	14.6	608	44.3	85.4	40.0	7.97	1.26	0.99	3.33	0.51	8.37	1.96	19.4	6.14
PC-77	D14	0.49	3.06	4.60	13.5	60.5	7.99	63.9	13.5	189	0.55	14.1	466	43.0	83.9	40.3	8.33	1.30	1.08	3.14	0.50	7.32	1.80	17.7	5.62
PC-78	D14	0.56	2.84	4.79	13.9	65.5	7.03	65.1	16.1	183	0.61	14.0	569	38.9	79.3	37.3	8.03	1.21	0.97	3.19	0.49	7.24	1.86	18.6	6.24
PC-79	D14	0.50	2.78	4.26	12.1	58.1	6.88	65.1	13.1	162	n.d.	12.3	383	44.8	82.8	39.7	7.78	1.20	0.90	3.00	0.48	7.54	1.59	17.5	5.03

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Table 2 (*continued*)

Sample	Type	Na_2O%	K_2O%	Fe_2O_3T%	Sc	Cr	Co	Zn	As	Rb	Sb	Cs	Ba	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	Hf	Ta	Th	U
PC-80	D14	0.51	3.19	4.82	13.6	63.0	8.35	62.9	18.3	187	0.55	13.8	465	40.0	77.5	35.3	7.14	1.14	0.89	2.93	0.44	6.91	1.89	17.7	5.81
PC-81	D14	0.54	3.09	4.79	13.2	63.2	7.17	67.1	21.5	191	0.53	13.6	650	31.6	63.9	31.1	6.39	1.06	0.84	2.88	0.46	6.65	1.76	16.7	5.75
PC-82	D14	0.59	3.35	4.55	13.1	64.4	7.79	59.4	17.2	189	0.52	13.7	481	35.4	68.9	34.1	6.93	1.20	0.91	2.82	0.41	5.87	1.79	15.1	6.41
PC-83	D14	0.71	3.65	4.88	13.9	69.0	12.1	75.4	12.8	208	0.73	13.6	523	41.9	90.8	42.0	8.38	1.44	1.17	3.66	0.54	6.32	1.99	17.5	4.39
PC-84	D14	0.49	3.33	4.86	13.9	72.8	9.80	67.5	28.2	203	0.61	14.8	541	36.7	72.5	35.7	7.22	1.30	0.98	3.13	0.47	5.89	1.88	15.8	6.96
PC-85	D14	0.59	3.33	5.05	15.0	73.9	8.63	67.5	23.8	200	0.57	15.3	530	36.8	72.4	35.3	7.25	1.31	0.88	3.23	0.47	6.63	2.00	16.7	6.99
PC-86	D14	0.62	3.27	4.48	13.1	63.3	11.5	129	19.8	167	0.53	11.4	935	33.0	71.4	32.0	6.61	1.06	0.72	3.39	0.46	5.51	1.70	15.2	4.73
PC-87	D14	0.67	3.45	4.81	13.7	67.8	9.75	73.8	15.7	202	0.66	13.3	520	39.2	79.9	38.8	7.87	1.36	1.01	3.05	0.52	6.18	1.79	16.8	6.22
PC-88	D14	0.61	3.35	4.37	12.8	58.9	9.67	76.6	19.6	172	0.61	13.5	525	38.8	74.7	37.3	7.77	1.39	0.98	3.14	0.42	6.03	1.77	16.3	6.73
PC-89	D14	0.60	3.48	4.83	13.3	65.0	9.43	69.8	22.0	195	0.62	12.9	589	40.5	79.1	38.7	7.96	1.40	0.97	3.20	0.56	6.30	1.86	17.2	6.20
PC-90	D14	0.53	3.50	4.70	13.6	67.7	9.21	66.6	21.4	199	0.57	13.9	626	35.4	69.2	33.4	6.79	1.22	0.94	3.14	0.58	6.01	1.89	16.2	6.64
PC-91	D14	0.73	3.49	3.98	11.6	56.1	8.12	59.3	14.3	160	0.58	11.2	454	40.1	75.5	36.4	7.65	1.16	0.99	3.10	0.48	6.75	1.84	15.2	4.88
PC-92	D14	0.62	3.59	4.16	12.1	56.3	7.83	58.4	15.9	160	0.48	11.9	552	32.3	63.0	29.4	6.00	1.06	0.86	2.89	0.41	5.66	1.74	13.8	4.24
PC-93	D14	0.61	3.41	4.78	13.9	66.0	7.32	64.9	15.9	182	0.63	14.0	505	47.0	81.3	38.1	8.01	1.22	0.93	3.13	0.50	7.74	2.05	17.9	5.79
PC-94	D14	0.50	2.99	4.26	11.5	53.7	7.15	60.1	14.9	154	0.45	11.8	411	49.5	86.5	40.5	8.14	1.19	0.97	2.92	0.48	7.59	1.68	17.7	5.66
PC-95	D14	0.65	3.10	5.89	12.3	62.1	8.22	62.7	23.9	164	0.63	12.3	612	33.8	64.8	29.4	6.24	0.94	0.86	2.81	0.43	6.55	1.89	16.0	6.48
PC-96	D14	0.57	3.09	4.46	12.4	60.3	7.84	65.3	17.1	167	0.58	13.0	596	44.0	80.6	39.4	8.17	1.23	0.97	3.05	0.48	7.19	1.80	17.1	5.43
PC-97	D14	0.59	3.37	4.55	12.9	59.0	7.53	64.8	17.5	184	0.42	13.8	454	48.8	87.5	40.9	8.15	1.23	0.97	3.24	0.49	7.90	1.79	18.6	6.30
PC-98	D14	0.56	3.18	4.71	13.5	67.5	14.2	77.5	18.1	186	0.52	13.70	501	48.0	95.3	48.0	10.4	1.79	1.29	4.00	0.57	7.07	1.94	17.9	6.88
PC-99	D14	0.52	3.25	5.37	14.5	67.2	9.78	75.9	20.8	201	0.53	15.4	506	48.5	93.3	44.1	8.72	1.43	1.14	3.24	0.49	7.40	2.03	18.8	6.37
PC-100	D14	0.67	3.97	4.91	14.9	75.8	15.2	83.5	22.8	194	0.59	15.3	510	57.1	99.0	52.4	12.1	2.11	1.56	4.35	0.65	6.87	1.98	16.9	7.44
PC-101	D14	0.71	3.49	4.53	13.3	69.4	15.8	77.7	21.3	188	0.47	13.8	523	54.1	79.7	46.0	10.5	1.89	1.26	3.79	0.61	6.31	1.92	15.7	6.43
PC-102	D14	0.68	3.72	4.28	12.8	60.2	16.6	72.5	20.8	182	0.47	13.3	473	51.2	85.5	43.7	9.70	1.70	1.23	3.50	0.53	6.01	1.81	14.7	7.01
PC-103	D14	0.63	3.87	4.78	14.0	72.7	8.29	70.9	20.2	183	0.59	14.1	525	41.0	70.0	33.4	7.60	1.30	1.04	3.24	0.52	5.68	1.88	15.3	6.73
PC-104	D14	0.47	2.93	4.01	11.6	52.8	6.92	55.1	18.5	160	0.44	12.6	416	30.4	71.5	34.0	7.26	1.15	0.86	2.65	0.47	6.27	1.66	14.7	5.54
PC-105	D14	0.61	3.51	4.61	13.2	61.0	6.44	63.0	24.7	165	0.51	13.3	564	36.7	61.4	30.6	6.56	1.06	0.85	2.64	0.42	6.03	1.78	15.0	4.99
PC-106	D14	0.65	3.41	4.89	14.6	69.0	13.6	81.0	18.3	198	0.58	14.8	567	50.4	90.7	41.1	11.2	1.96	1.43	3.37	0.62	7.38	2.16	18.80	6.66
PC-107	D14	0.58	3.26	5.11	14.5	69.7	9.96	70.1	19.0	197	0.51	15.5	469	46.2	94.6	42.2	8.75	1.42	1.13	3.28	0.53	7.64	1.90	18.7	6.72
PC-108	D14	0.60	3.23	5.07	13.9	64.1	7.39	62.0	22.3	191	0.41	14.7	580	41.4	82.9	38.7	7.64	1.25	1.01	3.21	0.41	7.30	1.87	18.8	5.84
PC-109	D14	0.52	3.02	4.69	13.5	66.0	6.60	60.6	26.4	181	0.55	14.0	618	40.2	77.0	36.5	7.52	1.25	0.97	3.14	0.49	7.79	1.99	19.4	6.25
PC-110	D14	0.74	3.84	4.73	14.5	72.2	8.40	72.4	18.4	187	0.50	14.9	496	46.2	77.0	36.0	8.15	1.35	1.05	3.00	0.51	6.30	1.88	16.3	5.89
PC-111	D14	0.59	3.28	5.45	15.7	78.3	10.5	85.4	21.0	203	0.54	16.0	514	47.0	95.0	46.4	9.58	1.70	1.19	3.51	0.57	7.67	2.12	19.6	7.96
PC-71	A50	0.52	3.05	4.75	12.7	58.9	8.21	61.7	16.1	173	0.55	13.3	523	52.4	93.5	46.2	8.99	1.31	0.99	2.82	0.50	7.17	1.79	17.0	5.86
PC-72	A50	0.66	3.13	4.80	14.1	69.7	11.3	73.7	16.8	183	0.76	13.2	468	51.2	94.7	45.9	9.23	1.50	1.19	3.47	0.58	7.82	1.94	18.7	4.43
PC-73	A50	0.57	3.12	4.54	12.7	60.9	7.87	67.3	9.81	175	0.71	13.1	465	50.6	87.8	42.20	7.90	1.24	0.94	3.28	0.48	8.09	1.90	17.7	4.34
PC-2	A51c/L3?	0.51	2.81	4.07	10.5	50.0	5.37	52.8	9.41	164	0.67	10.8	406	48.2	89.6	42.2	7.90	1.03	0.96	3.23	0.50	10.5	1.93	22.6	5.29
PC-4	A51c/L3?	0.69	3.61	4.42	14.9	68.9	10.0	85.2	9.29	187	0.55	14.6	508	39.2	75.6	35.8	7.35	1.33	0.93	3.16	0.48	5.85	1.76	15.1	5.11
PC-6	A51c/L3?	0.57	3.10	4.64	12.3	57.2	8.26	65.4	13.5	181	0.77	12.2	423	52.9	82.6	42.1	7.80	1.20	0.99	3.41	0.51	8.82	1.76	18.7	4.31
PC-7	A51c/L3?	0.66	3.12	4.99	12.6	55.8	8.32	78.2	7.44	185	0.69	12.4	455	43.6	83.7	39.0	7.34	1.18	0.93	3.15	0.49	7.53	1.71	17.9	4.46
PC-8	A51c/L3?	0.38	3.12	4.76	13.5	69.3	7.31	76.5	18.0	176	0.75	12.5	536	40.3	69.5	32.6	6.86	1.18	0.78	3.02	0.44	7.10	1.89	16.5	5.38
PC-10	A51c/L3?	0.72	3.34	4.99	12.3	58.8	8.95	80.4	6.23	182	0.65	12.7	471	37.8	74.8	35.2	7.01	1.13	0.93	3.19	0.47	7.43	1.87	16.6	4.03
PC-13	A51c/L3?	0.68	3.13	4.15	12.3	61.6	8.15	67.7	11.5	173	0.73	11.6	467	42.9	79.1	38.6	7.28	1.16	0.92	3.29	0.51	7.78	1.77	18.2	4.08
PC-18	A51c/L3?	0.53	3.09	4.76	12.3	57.8	8.79	59.5	5.88	188	0.51	12.9	566	54.2	101	46.6	8.76	1.32	1.12	3.07	0.53	8.58	1.74	20.0	4.49
PC-25	A51c/L3?	0.60	3.07	4.83	13.7	71.2	9.71	71.0	11.6	182	0.68	12.3	465	49.3	96.2	48.2	9.07	1.53	1.18	3.59	0.55	7.80	1.95	18.9	4.37
PC-27	A51c/L3?	0.68	3.32	4.97	13.0	65.0	8.55	64.2	17.0	182	0.71	11.8	514	38.9	72.3	36.7	7.30	1.25	0.95	3.02	0.49	6.86	1.83	16.9	4.85
PC-30	A51c/L3?	0.52	3.10	3.96	11.9	57.6	10.4	67.8	13.9	171	0.44	12.4	471	41.8	67.1	36.9	7.84	1.40	1.01	2.98	0.49	6.05	1.58	14.5	6.77
PC-33	A51c/L3?	0.65	2.94	4.64	13.3	64.4	6.65	58.4	12.5	162	0.65	12.4	648	40.6	78.5	37.1	7.39	1.18	1.98	3.16	0.49	7.93	1.80	18.1	4.98
PC-35	A51c/L3?	0.57	2.81	4.14	11.0	52.1	5.85	54.1	10.3	153	0.58	10.6	520	42.0	74.6	37.4	7.35	1.12	0.88	2.92	0.47	7.03	1.46	16.8	4.03
PC-36	A51c/L3?	0.57	3.20	4.93	14.4	67.8	10.5	73.5	10.5	199	0.78	14.1	521	58.7	112	55.4	11.1	1.86	1.37	3.74	0.62	7.84	1.91	18.6	5.15
PC-40	A51c/L3?	0.45	2.70	2.61	6.0	44.0	2.95	44.0	10.1	91.6	0.39	4.97	250	36.0	62.0	30.1	5.73	0.46	0.90	2.50	0.35	5.67	0.83	14.5	3.16
PC-43	A51c/L3?	0.78	3.34	4.23	13.9	70.8	8.65	78.0	5.73	192	0.60	13.4	476	44.3	85.8	38.6	7.80	1.29	1.01	3.23	0.52	7.17	1.88	17.1	6.25
PC-45	A51c/L3?	0.49	3.59	3.79	10.7	55.3	14.8	69.8	17.4	168	0.50	10.1	557	41.3	85.5	37.2	7.22	1.22	1.01	3.10	0.47	7.14	1.78	17.1	5.36
PC-46	A51c/L3?	0.72	3.82	4.10	14.3	70.1	8.24	89.7	4.70	194	0.46	13.6	535	52.4	90.7	40.9	7.89	1.36	0.89	2.92	0.48	5.87	1.73	15.5	5.45
PC-51	A51c/L3?	0.66	3.19	5.17	12.5	55.3	8.88	84.7	8.26	190	0.72	12.6	415	40.9	78.5	37.0	8.10	1.24	0.95	2.96	0.48	6.42	1.75	16.0	4.19
PC-55	A51c/L3?	0.76	3.16	4.86	12.1	55.3	7.92	74.0	7.91	184	0.62	12.0	418	44.0	84.4	39.8	7.78	1.18	0.98	2.96	0.53	7.74	1.75	17.9	4.75
PC-60	A51c/L3?	0.63	3.31	4.65	14.7	70.4	11.6	64.3	11.7	179	0.49	14.7	700	26.6	55.0	24.0	4.94	0.76	0.71	2.44	0.39	5.81	1.67	15.6	4.48
PC-61	A51c/L3?	0.69	3.15	4.51	11.4	52.3	7.81	63.5	11.2	173	0.50	12.4	552	34.1	67.6	31.5	6.19	0.97	0.88	2.71	0.43	6.93	1.69	16.2	4.28

(Schultz, 1964; Biscaye, 1965; Martín-Pozas, 1968). The diagnostic peaks used and the corresponding reflection powers (in parenthesis) were the following: quartz – 3.35 Å (2), alkali feldspar – 3.25 Å (1), plagioclase – 3.20 Å (1), phyllosilicates – 4.46 (0.1), anatase – 3.52 Å (1), hematite – 2.70 (1.3) and mullite – 3.38 Å (1). Given the uncertainties intrinsic to this semi-quantitative method, the results obtained should only be taken as rough estimates of mineral percentages.

For the evaluation of data, and considering that the compositional data generated by INAA is considerable, multivariate statistical methods were applied, using the Statistica program (StatSoft, 2008) and the JMP7 program (SAS). Hierarchical and non-hierarchical cluster analyses (joining-tree and k-means methods) were used to identify possible groups and outliers, after which different techniques were employed for group refinement and classification. The amalgamation rule employed in the joining-tree clustering was the unweighted pair-group average, also referred to as UPGMA (unweighted pair-group method using arithmetic averages), as well as Ward's method. In the present case study, the Euclidean mean, representing the measure of the actual geometric distance between objects in space, was used as the similarity coefficient to identify outliers. The Pearson correlation coefficient was used to evaluate the correlation between chemical parameters and samples. Principal component analysis

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Table 3
Elemental compositions of Roman amphorae from the Quinta do Rouxinol workshop (μg/g unless specified otherwise).

Sample	Type	Na_2O%	K_2O%	Fe_2O_3T%	Sc	Cr	Co	Zn	As	Rb	Sb	Cs	Ba	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	Hf	Ta	Th	U
QR-2	A51c	0.35	3.06	6.62	12.7	61.3	6.66	69.5	9.12	187	0.95	13.4	436	33.5	63.8	30.3	5.76	0.97	0.82	2.70	0.41	6.70	1.80	15.2	1.78
QR-3	A51c	0.97	3.37	5.58	13.7	62.5	8.28	77.4	2.32	188	0.72	8.50	151	40.8	83.8	39.2	7.34	1.34	1.03	3.28	0.48	6.87	1.85	15.7	2.85
QR-4	A51c	0.46	3.18	6.58	13.4	65.5	6.08	67.6	5.44	203	0.73	15.4	468	34.9	67.0	32.4	5.94	1.07	0.82	2.61	0.40	5.32	1.75	15.9	4.23
QR-5	A51c	0.52	3.69	6.39	14.2	74.8	9.08	85.2	3.27	222	0.90	15.9	453	44.5	89.2	42.7	8.19	1.39	0.99	3.26	0.50	7.77	1.95	18.4	4.09
QR-6	A51c	0.63	4.05	6.67	14.5	72.7	8.54	94.7	7.56	234	0.96	15.1	549	40.5	79.1	38.7	7.51	1.38	0.93	3.24	0.47	6.27	1.91	16.3	3.76
QR-7	A51c	0.37	3.38	6.64	15.2	70.5	7.97	82.5	7.61	227	0.95	16.3	444	45.5	87.3	46.3	8.96	1.71	1.25	3.92	0.55	5.84	1.89	16.5	3.31
QR-8	A51c	0.43	3.32	5.17	12.9	61.4	7.78	75.2	11.9	192	0.83	12.4	423	37.8	77.4	37.1	7.82	1.38	1.04	2.8	0.49	6.12	1.62	14.9	2.77
QR-9	A51c	0.62	3.36	6.24	13.7	66.0	10.2	88.6	9.51	197	0.82	14.2	539	46.0	91.5	44.7	8.93	1.45	1.17	3.33	0.48	7.65	1.90	16.7	3.06
QR-10	A51c	0.47	3.30	4.87	12.4	59.5	6.80	70.2	11.3	185	0.66	12.5	380	36.9	71.9	34.9	6.85	1.12	0.88	2.73	0.41	6.23	1.69	15.3	3.05
QR-11	A51c	0.61	3.27	5.05	12.3	59.0	6.96	75.7	8.30	200	0.70	12.4	414	46.0	89.9	42.7	8.51	1.35	1.08	3.14	0.5	7.52	1.92	18.0	3.70
QR-12	A51c	0.46	3.59	5.62	14.8	67.0	9.62	96.6	4.93	216	0.86	14.7	449	51.5	100	51.1	10.4	1.72	1.39	4.07	0.58	8.07	2.17	18.1	3.53
QR-13	A51c	0.64	4.54	6.46	15.3	68.4	8.24	83.4	15.3	213	0.95	15.8	467	49.7	89.2	45.3	8.89	1.59	1.17	3.97	0.56	7.95	2.16	19.0	2.52
QR-14	A51c	0.41	3.60	5.99	14.4	62.7	7.66	88.5	6.08	222	0.91	15.7	431	47.1	91.1	45.7	9.14	1.54	1.18	3.99	0.61	8.35	2.03	18.2	2.60
QR-15	A51c	0.47	3.85	5.92	13.5	60.9	8.04	91.4	4.77	226	0.93	14.5	441	48.0	86.6	45.9	8.16	1.46	1.13	3.58	0.57	7.88	2.01	18.4	2.98
QR-16	A51c	0.71	4.16	6.83	14.5	67.1	10.3	97.4	9.43	218	0.92	14.2	527	52.0	91.5	49.7	9.50	1.71	1.26	4.06	0.61	8.40	2.14	18.8	3.62
QR-17	A51c	0.80	3.33	4.68	10.5	49.0	5.53	53.8	10.6	166	0.65	10.0	605	45.3	82.3	41.5	8.05	1.34	1.03	2.96	0.43	6.97	1.72	16.9	3.13
QR-18	A51c	0.71	3.59	4.58	12.1	59.2	7.13	78.7	5.70	188	0.70	12.9	397	48.8	91.4	43.7	8.79	1.41	1.11	3.21	0.50	7.71	1.89	17.7	3.19
QR-19	A51c	0.43	3.84	7.11	14.4	68.4	8.05	90.7	10.4	227	0.98	15.3	397	39.5	72.4	35.9	6.45	1.11	0.89	2.95	0.43	6.49	1.90	16.7	3.62
QR-20	A51c	0.50	3.53	5.15	12.4	57.7	6.66	70.1	11.5	175	0.88	12.0	367	43.8	78.4	38.6	7.14	1.18	0.92	2.99	0.40	6.94	1.88	16.8	3.80
QR-21	A51c	0.50	4.09	6.41	13.3	57.0	7.93	88.8	7.65	210	0.85	14.1	394	45.6	76.8	37.9	7.93	1.33	1.00	3.19	0.48	5.89	1.77	14.8	3.81
QR-22	A51c	0.47	3.95	6.46	14.5	64.2	7.44	86.1	6.97	209	1.03	14.4	425	49.2	86.2	42.8	9.09	1.54	1.20	3.93	0.57	7.27	2.01	17.8	4.02
QR-23	A51c	0.55	3.64	4.68	11.9	55.8	6.52	73.0	10.5	183	0.66	12.3	345	42.3	74.2	37.4	7.68	1.36	1.05	3.06	0.47	6.32	1.68	15.0	3.71
QR-25	A51c	0.78	3.26	5.21	10.8	52.5	7.73	68.0	6.72	169	0.73	10.4	346	44.9	80.1	40.2	7.95	1.39	1.00	2.97	0.45	6.67	1.63	15.7	2.68
QR-26	A51c	0.73	3.07	4.84	9.97	48.6	7.17	66.7	6.25	155	0.75	9.72	319	41.4	73.7	36.7	7.43	1.31	0.93	2.82	0.44	6.07	1.49	14.2	2.41
QR-27	A51c	0.47	3.70	5.69	13.1	59.7	9.08	89.0	8.31	208	0.78	13.5	389	43.3	80.0	40.3	8.26	1.45	1.10	3.51	0.49	6.58	1.79	16.0	3.34
QR-28	A51c	0.60	3.81	5.92	14.1	60.3	10.1	98.0	4.80	219	0.84	14.3	394	60.3	108	58.0	11.8	2.02	1.52	4.51	0.65	8.18	1.98	19.0	3.76
QR-29	A51c	0.49	3.61	4.09	11.5	54.1	7.05	76.0	4.99	174	0.59	11.6	336	41.3	74.4	37.2	7.61	1.14	0.93	3.01	0.47	5.39	1.59	13.5	3.18
QR-30	A51c	0.39	3.46	4.87	11.3	54.6	9.53	88.0	7.34	186	0.60	11.5	352	39.3	79.6	40.9	7.87	1.41	1.12	2.78	0.47	5.31	1.68	13.5	3.14
QR-31	A51c	0.64	3.63	5.91	12.4	55.5	14.0	95.8	4.13	225	0.78	13.2	432	51.9	104	57.5	11.9	2.23	1.49	3.78	0.57	6.71	1.94	16.1	2.60
QR-32	A51c	0.40	3.59	5.91	14.4	64.5	9.45	77.9	8.38	208	0.89	14.3	500	41.9	80.9	38.8	7.87	1.42	1.04	3.45	0.55	7.14	2.03	18.0	3.53
QR-33	A51c	0.44	3.30	5.39	12.2	57.6	5.81	68.4	11.3	176	0.70	11.3	568	37.8	68.3	34.7	6.86	1.21	0.91	2.87	0.44	5.89	1.66	14.7	3.05
QR-34	A51c	0.63	3.64	5.65	12.2	58.5	9.82	80.5	7.76	184	0.82	11.4	415	46.5	86.7	44.5	8.77	1.53	1.16	3.58	0.53	6.42	1.80	16.4	2.79
QR-35	A51c	0.31	2.74	4.29	11.8	58.1	5.36	56.4	6.08	166	0.85	12.3	352	45.9	82.4	38.8	7.21	1.20	0.94	3.02	0.41	6.06	2.02	17.4	3.66
QR-36	A51c	0.44	3.59	6.08	13.5	60.9	8.23	84.1	7.30	221	0.95	14.8	400	43.4	84.3	41.0	7.82	1.33	1.08	3.14	0.51	7.68	1.91	17.8	3.96
QR-37	A51c	0.49	3.50	6.40	13.4	62.1	9.16	93.1	6.72	223	0.83	13.7	392	43.0	85.0	42.2	8.38	1.41	1.11	3.25	0.50	6.87	1.85	16.7	3.26
QR-38	A51c	0.58	3.17	4.87	11.0	52.0	6.85	71.2	4.31	191	0.65	11.3	347	38.9	77.9	39.3	7.31	1.22	0.97	2.72	0.40	7.09	1.69	16.0	3.23
QR-39	A51c	0.47	3.50	5.97	13.4	60.3	8.07	90.0	9.69	214	0.89	13.7	421	41.8	81.3	40.2	7.83	1.37	1.00	3.21	0.48	6.64	1.72	16.6	3.58
QR-40	A51c	0.45	3.34	5.59	12.9	57.4	8.30	79.6	7.80	205	0.84	13.4	598	41.7	83.7	43.3	8.76	1.50	1.16	3.44	0.55	6.95	1.79	16.7	3.09
QR-169	A51c	0.44	3.10	5.37	12.4	54.4	8.22	85.4	6.33	197	0.75	12.3	362	51.8	98.9	58.4	11.4	1.91	1.36	3.83	0.59	7.17	1.81	17.8	3.19
QR-170	A51c	0.36	3.64	7.59	14.9	68.6	10.0	97.8	9.76	230	0.88	14.8	433	38.4	75.5	37.6	6.64	1.39	1.03	3.26	0.50	5.69	1.77	14.3	2.69
QR-171	A51c	0.40	3.67	6.83	13.6	62.8	7.51	84.4	11.8	225	1.00	14.9	375	38.0	68.6	33.1	6.71	1.04	0.83	2.82	0.45	5.90	2.66	15.9	3.27
QR-172	A51c	0.41	3.47	6.37	12.6	58.8	7.44	77.9	9.24	207	0.89	13.4	377	36.5	72.6	35.6	7.23	1.14	0.97	3.07	0.44	6.19	1.75	16.4	3.67
QR-173	A51c	0.47	3.27	5.37	11.9	52.0	7.04	68.5	5.99	194	0.70	13.6	362	44.1	84.7	39.4	7.82	1.14	0.97	3.19	0.48	8.63	1.86	18.9	4.06
QR-174	A51c	0.51	3.44	5.54	12.0	55.5	8.53	70.1	7.43	203	0.68	14.0	351	45.5	98.1	41.1	8.16	1.16	1.01	3.16	0.52	8.96	2.04	19.0	4.88
QR-175	A51c	0.37	3.44	6.95	13.8	63.4	6.78	78.3	10.8	217	0.81	13.7	564	33.9	66.0	34.9	6.84	1.13	0.88	2.97	0.47	5.82	1.76	15.9	3.14
QR-176	A51c	0.51	3.49	5.00	11.1	51.1	7.29	71.3	5.97	183	0.63	11.6	357	42.8	82.9	45.8	9.01	1.29	0.99	3.08	0.45	5.98	1.68	14.2	3.37
QR-177	A51c	0.67	3.37	5.38	12.1	56.3	7.13	73.6	7.35	189	0.76	11.6	435	43.7	82.4	42.4	8.29	1.35	1.09	3.28	0.46	6.75	1.77	16.7	2.81
QR-178	A51c	0.51	3.20	5.26	11.4	50.1	5.23	56.7	7.57	182	0.77	12.6	526	41.1	79.2	38.2	7.92	1.09	0.99	3.27	0.48	8.58	2.00	19.5	3.88
QR-179	A51c	0.42	3.47	5.63	12.9	60.9	7.75	85.8	9.04	206	0.72	13.4	345	41.6	81.0	44.8	8.11	1.43	1.04	3.28	0.46	6.12	1.76	15.9	2.79
QR-41	A50	0.81	4.46	6.99	14.9	68.3	9.26	91.7	8.96	243	0.96	15.2	625	40.1	79.0	36.2	7.14	1.28	1.01	3.05	0.46	6.46	1.88	15.6	2.59
QR-42	A50	0.62	3.64	6.19	14.5	65.1	8.44	87.8	8.28	242	0.93	15.5	457	43.6	84.7	39.6	7.87	1.30	1.02	3.14	0.52	7.35	1.87	17.2	2.41
QR-43	A50	0.64	3.57	6.86	15.2	69.4	8.20	84.7	6.26	233	1.08	16.1	439	47.0	94.0	48.3	9.21	1.58	1.27	3.61	0.57	7.15	1.87	19.1	3.57
QR-44	A50	0.36	3.33	4.67	11.5	59.5	6.24	62.9	7.89	180	0.68	12.2	389	35.4	72.3	35.9	6.94	1.25	0.86	2.87	0.38	5.27	1.48	13.7	2.69
QR-45	A50	0.39	3.12	6.02	13.5	63.2	7.27	78.1	5.72	186	0.77	12.7	475	39.9	80.3	39.8	7.57	1.33	1.01	3.31	0.48	6.83	1.92	17.4	3.51
QR-46	A50	0.41	3.39	6.86	14.5	64.6	9.81	59.1	17.5	187	1.10	13.6	554	38.4	77.9	38.3	8.00	1.30	1.07	3.32	0.49	7.21	1.98	17.6	3.32
QR-47	A50	0.60	3.49	5.87	13.7	62.4	10.1	95.1	6.61	221	0.81	14.0	414	50.5	103	53.5	10.6	1.84	1.35	3.89	0.56	7.22	1.96	18.4	2.90
QR-48	A50	0.67	3.26	4.26	11.2	54.5	8.49	79.7	6.18	181	0.77	10.6	410	46.2	89.2	44.8	9.11	1.56	1.19	3.37	0.49	7.51	1.93	16.0	2.86
QR-49	A50	0.69	3.02	4.59	11.5	63.3	6.81	67.3	6.94	179	0.85	11.8	417	41.5	83.3	39.7	7.42	1.15	0.96	2.85	0.45	8.26	1.81	19.3	3.88
QR-50	A50	0.41	3.54	7.60	16.8	76.5	9.62	99.1	6.95	218	0.96	15.2	450	48.8	95.5	47.3	9.60	1.76	1.29	4.06	0.58	6.93	2.47	19.3	3.73
QR-51	A50	0.59	3.83	7.02	16.6	75.3	11.5	94.6	8.07	253	1.01	16.3	492	47.1	93.9	47.0	9.32	1.70	1.27	4.07	0.55	7.13	2.12	18.9	4.01
QR-53	A50	0.27	3.39	6.01	13.0	63.7	7.33	84.3	11.6	182	0.87	13.1	322	33.7	59.1	28.5	5.27	1.01	0.76	2.66	0.40	5.45	1.55	14.0	2.44
QR-54	A50	0.49	4.17	6.78	14.4	66.5	9.08	103	11.2	221	1.05	15.3	391	44.0	77.9	39.4	7.28	1.24	0.99	3.05	0.48	6.04	1.84	16.0	3.19
QR-55	A50	0.51	3.81	6.38	13.0	59.7	7.65	92.3	8.83	207	0.93	13.8	397	38.5	69.1	29.0	6.79	1.16	0.89	2.90	0.45	5.32	1.67	14.3	3.33
QR-56	A50	0.57	4.12	6.11	14.4	64.8	8.22	93.5	8.09	229	0.94	15.2	430	47.4	82.1	39.9	7.54	1.22	1.03	3.31	0.50	7.29	1.97	18.5	3.41
QR-57	A50	0.49	4.13	4.61	14.5	65.0	4.07	65.3	10.3	203	0.65	15.2	514	45.3	58.4	32.1	8.18	1.85	1.17	3.52	0.52	5.06	1.65	15.2	4.09
QR-58	A50	0.51	3.76	6.17	13.1	58.8	7.74	84.0	7.76	218	0.84	13.5	414	40.4	78.6	39.0	8.03	1.37	1.15	3.57	0.53	6.65	1.81	16.6	3.71
QR-59	A50	0.49	4.02	7.52	15.3	72.9	8.74	98.0	9.65	244	0.93	16.0	433	43.1	77.5	36.9	7.65	1.35	1.05	3.46	0.50	6.32	1.99	16.9	3.90
QR-60	A50	0.50	3.63	5.37	13.3	61.7	7.76	91.0	7.31	193	0.66	12.6	419	44.2	78.8	40.0	8.05	1.37	0.98	3.28	0.50	6.27	1.83	15.7	3.33
QR-61	A50	0.54	3.82	6.05	14.2	61.0	9.53	94.5	7.07	214	0.79	14.0	422	62.1	110	61.8	12.7	2.22	1.58	4.58	0.63	8.12	2.02	19.3	3.88
QR-62	A50	0.55	4.00	6.12	14.0	60.9	9.46	92.5	5.95	220	0.93	14.9	386	62.9	112	60.9	12.0	2.08	1.56	4.73	0.67	8.90	2.14	20.5	4.24
QR-63	A50	0.61	3.53	5.17	11.7	54.4	7.64	74.0	3.76	202	0.73	12.4	385	38.8	79.8	40.1	7.58	1.39	0.99	3.01	0.51	6.33	1.72	15.5	3.01
QR-64	A50	0.42	3.28	6.98	14.7	66.9	11.3	107	8.70	198	0.84	12.9	405	51.5	103	55.8	10.8	1.94	1.41	4.04	0.66	8.15	2.19	19.3	3.57
QR-65	A50	0.33	3.26	6.23	13.1	63.7	7.80	85.8	9.63	194	0.87	13.1	383	37.1	70.6	34.0	6.59	1.19	0.93	3.03	0.46	5.79	1.76	14.8	3.18
QR-66	A50	0.37	3.75	7.60	15.9	79.3	8.40	99.4	13.1	215	1.11	15.1	472	42.3	78.7	38.6	7.28	1.34	1.06	3.44	0.53	7.13	2.05	18.2	4.15

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Table 3 (*continued*)

Sample	Type	Na_2O%	K_2O%	Fe_2O_3T%	Sc	Cr	Co	Zn	As	Rb	Sb	Cs	Ba	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	Hf	Ta	Th	U
QR-67	A50	0.48	4.16	6.31	13.4	58.5	8.14	87.5	8.57	222	0.97	14.1	444	43.9	82.6	41.8	8.30	1.51	1.21	3.60	0.56	6.16	1.76	15.7	3.90
QR-68	A50	0.55	3.73	6.70	14.4	65.7	9.21	95.9	10.8	219	0.92	14.5	447	42.6	79.1	39.7	7.79	1.39	1.10	3.33	0.53	5.69	1.79	15.5	3.70
QR-69	A50	0.50	3.56	7.07	14.5	64.9	8.27	92.8	8.63	242	0.93	15.1	391	40.4	78.4	38.8	7.64	1.34	1.06	3.27	0.48	6.19	1.97	16.2	3.58
QR-70	A50	0.66	3.14	5.02	11.6	52.9	7.32	71.4	7.80	202	0.82	12.6	375	39.8	79.4	40.1	7.73	1.36	0.99	3.13	0.46	6.58	1.72	16.2	2.98
QR-71	A50	0.51	3.83	7.20	14.8	67.2	8.36	99.2	10.4	248	0.97	15.9	432	41.9	78.9	38.1	7.81	1.39	1.10	3.29	0.47	6.16	1.83	16.6	3.65
QR-72	A50	0.32	2.66	5.63	12.6	57.6	4.87	60.9	12.0	165	0.92	12.6	421	27.2	52.3	25.9	5.19	0.93	0.72	2.48	0.38	5.68	1.65	14.5	2.96
QR-152	A50	0.31	3.48	7.05	14.9	74.7	8.47	92.4	13.6	204	1.14	14.7	378	36.5	68.5	31.9	6.17	1.08	0.84	2.87	0.46	6.23	1.89	16.7	2.86
QR-153	A50	0.52	3.85	6.59	14.7	67.5	9.56	103	6.74	240	1.05	15.1	450	41.9	84.5	44.3	8.55	1.41	1.12	3.17	0.50	6.39	1.88	16.5	3.35
QR-154	A50	0.35	3.71	7.07	15.7	74.2	9.35	103	11.5	229	1.01	16.0	449	41.0	81.8	38.8	7.39	1.42	1.09	3.44	0.54	6.31	2.03	16.2	3.13
QR-155	A50	0.45	3.50	5.99	13.3	60.3	8.14	84.2	10.2	207	0.80	13.5	422	41.2	80.9	41.0	7.70	1.41	1.06	3.31	0.48	6.65	1.82	16.7	3.18
QR-156	A50	0.52	3.78	5.98	13.2	59.3	8.49	88.4	10.2	216	0.91	14.2	403	42.5	80.7	41.5	8.03	1.32	1.05	3.28	0.50	6.62	1.87	16.3	3.71
QR-157	A50	0.57	3.87	7.17	16.2	69.1	11.6	109	7.51	239	1.04	17.2	398	54.7	108	56.6	11.8	2.17	1.56	4.83	0.74	7.95	2.25	19.8	3.16
QR-158	A50	0.43	3.44	5.60	14.0	60.9	7.46	77.6	10.1	196	0.79	14.3	392	40.4	78.7	40.4	7.81	1.31	1.02	3.22	0.49	6.64	1.94	16.7	3.41
QR-159	A50	0.62	3.38	5.56	13.2	59.6	8.09	83.5	7.90	200	0.83	14.0	375	41.5	83.7	43.6	8.67	1.39	1.16	3.37	0.47	6.26	1.90	15.9	3.38
QR-160	A50	0.41	3.22	6.59	14.6	62.7	8.19	77.9	8.11	196	0.97	15.0	435	44.5	87.0	44.9	8.38	1.45	1.06	3.68	0.62	7.65	1.92	18.2	3.35
QR-161	A50	0.63	3.81	6.33	14.9	65.1	8.33	94.3	6.50	223	0.79	15.1	483	47.4	89.4	45.3	8.20	1.41	1.19	3.69	0.54	8.06	2.01	18.1	3.60
QR-162	A50	0.64	3.60	6.19	13.4	59.2	11.6	87.1	9.83	216	0.90	13.8	400	47.3	91.3	49.6	9.90	1.69	1.39	3.74	0.52	6.49	1.82	16.1	4.30
QR-163	A50	0.44	3.61	6.16	13.6	56.8	8.56	95.7	5.86	222	0.94	14.5	449	47.6	92.7	48.2	9.11	1.46	1.27	4.02	0.57	8.82	2.27	19.1	3.91
QR-164	A50	0.43	3.30	4.78	11.9	54.1	7.51	71.2	7.45	174	0.70	11.8	386	35.9	75.1	36.8	5.87	1.25	0.97	2.98	0.48	5.77	1.52	13.9	2.55
QR-165	A50	0.51	3.75	6.69	14.5	64.8	8.93	91.1	10.9	216	0.92	15.1	408	39.9	75.6	36.0	7.07	1.22	1.02	3.04	0.49	6.29	1.92	16.1	3.38
QR-166	A50	0.55	3.74	6.96	15.0	66.7	9.62	105	13.0	225	1.00	15.2	435	44.0	84.0	41.5	8.06	1.44	1.17	3.50	0.47	6.16	1.95	16.3	3.99
QR-167	A50	0.50	3.54	5.88	14.1	59.7	9.42	93.0	6.24	200	0.80	13.7	421	58.4	112	60.1	11.8	2.17	1.56	4.50	0.63	8.32	1.87	19.4	3.55
QR-168	A50	0.70	3.48	4.90	12.0	54.3	7.35	73.5	7.50	213	0.69	12.6	381	44.8	84.6	42.0	8.67	1.46	1.09	3.24	0.50	6.57	1.65	16.4	2.98
QR-180	A50	0.44	3.02	5.43	12.6	55.2	8.65	184	5.46	175	0.84	12.4	382	52.2	103	58.0	11.4	2.00	1.35	3.93	0.58	7.37	1.88	17.6	3.19
QR-24	L9	0.56	4.26	6.44	13.4	60.3	7.36	84.6	8.67	208	0.94	14.0	491	50.7	86.4	43.8	9.16	1.54	1.18	3.53	0.54	7.19	1.85	17.8	4.35
QR-73	L9	0.43	4.41	7.78	15.5	73.3	9.76	105	13.1	244	1.00	6.10	444	44.2	73.9	37.5	7.11	1.26	0.95	3.22	0.49	5.66	1.89	15.7	3.45
QR-74	L9	0.49	3.73	6.18	12.5	59.4	8.68	98.3	7.52	211	0.96	12.9	405	40.7	78.2	40.1	7.68	1.37	1.08	3.34	0.51	6.37	1.71	15.1	3.69
QR-75	L9	0.46	4.28	7.83	14.6	67.1	9.28	104	14.7	225	0.99	14.7	559	42.8	77.3	39.3	7.67	1.40	1.13	3.50	0.55	6.34	1.90	16.5	3.56
QR-76	L9	0.61	4.28	5.81	12.6	56.7	8.73	92.7	5.86	219	0.91	13.7	416	45.8	85.0	42.1	8.35	1.42	1.08	3.26	0.491	6.77	1.71	15.9	3.61
QR-77	L9	0.58	4.59	7.15	14.4	66.6	9.74	104	9.10	242	1.08	15.3	524	49.1	90.6	45.5	9.17	1.63	1.33	3.88	0.58	7.03	1.91	17.3	4.20
QR-78	L9	0.67	3.49	4.93	11.2	51.0	9.87	82.8	5.55	181	0.72	11.3	340	41.3	79.6	42.8	8.24	1.47	1.06	3.09	0.50	5.78	1.55	14.2	2.72
QR-79	L9	0.50	4.22	6.58	13.1	58.1	8.87	93.0	9.11	213	0.99	13.4	413	46.9	82.4	40.3	8.57	1.44	1.14	3.60	0.54	6.27	1.84	15.2	3.81
QR-80	L9	0.39	3.72	5.75	12.3	53.5	6.60	77.2	7.86	191	0.79	12.9	374	40.5	75.6	38.0	7.55	1.27	1.10	3.06	0.49	5.75	1.64	14.5	3.77
QR-81	L9	0.45	3.71	6.79	14.0	62.9	7.38	79.1	9.38	207	0.96	15.0	382	41.5	35.1	16.8	6.87	1.18	0.80	2.89	0.45	6.00	1.81	15.9	3.09
QR-82	L9	0.55	4.32	6.77	13.6	62.5	9.42	97.7	8.83	230	0.46	14.1	449	48.3	84.6	42.0	8.67	1.50	1.21	3.85	0.56	6.87	1.88	16.5	4.23
QR-83	L9	0.40	3.97	7.16	13.4	59.8	7.68	86.5	14.2	193	0.18	12.7	716	40.3	69.1	34.1	7.30	1.27	0.95	3.18	0.51	5.80	1.68	14.5	3.29
QR-84	L9	0.30	3.30	7.09	14.5	66.8	8.39	89.7	7.97	229	0.96	15.6	438	37.3	75.9	35.4	6.59	1.13	0.87	2.97	0.52	6.81	2.08	17.4	3.15
QR-85	L9	0.42	3.76	7.72	15.1	68.8	10.5	101	10.6	247	0.92	15.5	418	39.1	77.8	38.9	7.35	1.38	1.03	3.17	0.55	5.68	1.88	15.4	3.43
QR-86	L9	0.46	3.22	5.52	11.8	54.8	11.1	74.5	8.04	183	0.80	12.0	425	36.2	71.6	35.2	6.87	1.14	0.95	3.16	0.52	6.63	1.71	15.4	3.29
QR-87	L9	0.41	3.68	8.02	15.6	72.2	10.5	108	10.9	255	0.97	16.0	445	37.4	78.6	40.5	7.67	1.47	1.04	3.38	0.55	5.97	1.96	15.5	3.57
QR-88	L9	0.66	2.83	6.15	11.6	60.6	5.60	65.7	11.4	164	0.80	11.3	624	35.0	69.5	33.9	6.34	0.97	0.78	2.63	0.41	7.44	1.66	17.2	3.32
QR-89	L9	0.36	3.41	7.84	14.7	71.2	9.71	92.4	10.0	227	0.91	13.9	528	36.2	73.9	35.9	6.84	1.33	0.99	3.16	0.48	5.98	1.80	15.9	2.82
QR-90	L9	0.31	3.31	7.52	15.2	75.0	9.52	87.6	16.2	214	1.17	15.1	838	35.6	70.5	33.5	6.69	1.20	0.93	3.19	0.51	6.48	1.98	17.3	3.96
QR-91	L9	0.33	2.97	7.00	14.5	71.0	6.33	80.7	11.5	201	1.00	13.1	561	38.1	69.4	35.6	7.34	1.26	1.02	3.30	0.51	6.21	1.87	16.9	3.60
QR-92	L9	0.51	3.85	6.54	13.2	60.3	9.22	94.6	7.68	229	1.02	13.9	419	42.6	81.7	41.0	8.23	1.50	1.20	3.44	0.50	6.75	1.81	16.2	3.45
QR-141	L9	0.33	2.76	4.64	10.8	49.6	5.36	59.9	8.84	156	0.74	9.53	444	27.8	53.7	26.5	5.50	0.96	0.79	2.57	0.40	5.53	1.43	13.5	2.36
QR-142	L9	0.45	3.43	7.45	14.4	68.9	10.0	101	9.27	228	0.91	13.8	422	39.9	76.7	39.2	7.66	1.36	1.01	3.22	0.49	5.98	1.79	15.8	2.95
QR-143	L9	0.48	3.31	6.10	12.6	56.5	6.63	77.0	9.86	202	0.83	13.0	421	36.1	71.2	34.5	7.00	1.24	0.95	3.14	0.52	6.46	1.79	15.9	3.65
QR-144	L9	0.47	3.14	5.84	11.7	53.3	7.85	83.7	6.96	200	0.87	12.4	360	37.6	73.5	37.4	7.21	1.28	1.07	3.12	0.50	5.98	1.62	14.8	3.18
QR-145	L9	0.41	3.27	7.33	13.9	65.6	9.81	94.8	9.56	216	0.91	13.3	369	41.2	101	45.7	7.74	1.31	1.06	3.33	0.50	5.66	1.78	14.9	3.22
QR-146	L9	0.61	3.84	6.10	13.0	60.3	9.44	93.6	5.81	223	0.83	13.8	418	42.6	87.5	44.8	8.24	1.49	1.12	3.35	0.51	7.08	1.88	16.5	3.72
QR-147	L9	0.40	3.65	8.24	15.3	74.1	11.0	108	12.2	234	1.08	14.8	486	42.6	81.3	41.6	7.76	1.45	1.12	3.33	0.50	6.09	1.84	16.4	3.20
QR-148	L9	0.45	3.53	6.29	12.8	56.4	8.63	87.8	8.04	214	1.01	13.4	395	39.1	77.7	41.1	8.03	1.41	1.06	3.25	0.48	5.98	1.71	14.9	3.80
QR-149	L9	0.38	3.35	7.24	13.5	57.5	7.87	94.8	13.8	220	1.01	15.1	594	41.1	76.5	39.0	7.97	1.30	1.02	3.20	0.49	5.85	1.80	16.5	3.60
QR-150	L9	0.28	2.33	3.53	8.24	39.3	5.03	48.9	4.90	126	0.47	8.25	257	26.1	52.9	26.3	4.90	0.95	0.68	2.01	0.31	4.29	1.23	10.4	2.22
QR-151	L9	0.55	2.80	4.65	10.8	50.3	7.01	80.2	5.33	167	0.76	10.3	320	36.9	73.5	36.5	6.62	1.19	0.88	2.92	0.40	6.52	1.60	15.0	2.38

(PCA) (non-supervised) and discriminate analysis (supervised) transformation provided a new basis for viewing the entire data distribution, allowing a better grasp of its structure. Bivariate and trivariate scatter plots were also used for a better scanning of the correlations between variables, also contributing to the detection of obvious groups and of outliers. For this statistical approach, the absolute concentrations of elements, as well as the normalized contents relative to the Sc content ([X]sample/[Sc]sample), were used as variables. This normalization procedure was done prior to any further statistical approach. Comparative studies of ceramics produced with similar raw materials, as are the ones from the Tagus estuary, must pass through a first step of normalization, especially because the temper is mainly composed of quartz, diluting chemical differences; normalization being a procedure allowing to compensate for the influence of natural processes on the measured variability of the concentrations of elements (Dias and Prudêncio, 2008). As scandium is a conservative element, it is most resistant and tends to survive in superficial environments during weathering; as such, it will not be leached and carried away easily during ancient ceramic burial. Another important argument for the choice of this element is that its concentration is obtained with good precision and accuracy by INAA.

4. Results and discussion

A description of the geochemical patterns of each production centre, taken separately, will be presented first, followed by a discussion comparing the two centres, belonging to the same

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Table 4
Average concentration values and corresponding standard deviations, minimum and maximum values of each chemical element for the PC groups (μg/g unless specified otherwise).

	GROUP 1					GROUP 2					GROUP 3				
	n	$\bar{x}$	Min.	Max.	*σ*	*n*	$\bar{x}$	Min.	Max.	*σ*	*n*	$\bar{x}$	Min.	Max.	*σ*
Na_2O, %	38	0.434	0.250	0.670	0.0785	24	0.686	0.510	0.820	0.0887	61	0.596	0.370	0.790	0.0979
K_2O, %	37	3.29	3.03	3.36	0.1617	24	3.25	2.81	3.520	0.1741	60	3.27	2.78	3.84	0.2279
Fe_2O_3T, %	39	4.71	4.27	4.91	0.2665	24	4.76	3.92	5.86	0.5120	56	4.65	3.96	5.37	0.3018
Sc	38	13.2	11.7	14.9	0.7110	24	12.3	11.0	13.8	0.6821	60	13.6	11.5	15.7	0.9596
Cr	39	64.7	54.9	75.8	4.806	24	56.7	52.1	61.6	2.8582	61	65.5	52.8	78.3	5.564
Co	38	15.3	10.5	20.2	2.139	21	8.71	6.97	10.7	0.9097	60	9.10	6.44	13.6	1.832
Zn	39	78.5	67.0	88.2	5.715	24	74.0	54.1	93.4	10.18	60	70.2	48.5	92.1	9.013
As	39	18.8	12.7	25.1	2.785	24	8.23	4.29	14.3	2.674	61	16.5	4.70	28.2	5.771
Rb	39	188	168	209	9.933	24	180	153	199	10.26	60	183	154	208	13.10
Sb	40	0.526	0.360	0.65	0.0638	24	0.647	0.500	0.780	0.0774	61	0.565	0.390	0.780	0.0931
Cs	39	13.5	11.6	15.3	0.9134	24	12.2	10.6	14.1	0.8319	60	13.6	11.3	16.4	1.179
Ba	40	503	392	612	54.68	23	459	378	566	48.24	59	529	361	700	70.71
La	40	50.5	37.7	59.5	5.425	23	44.1	34.1	54.2	4.802	61	42.1	26.6	58.7	6.610
Ce	39	83.1	63.3	99.0	8.621	24	83.3	67.6	101	7.993	61	79.6	53.5	112	12.62
Nd	40	48.0	35.6	57.9	5.629	24	39.8	31.5	47.3	3.812	59	38.0	24-0	48.2	5.040
Sm	40	9.97	7.22	12.9	1.385	24	7.76	6.19	8.76	0.6464	58	7.74	5.50	9.58	0.8992
Eu	40	1.83	1.22	2.31	0.2616	23	1.22	1.08	1.38	0.0768	56	1.29	0.940	1.62	0.1446
Tb	40	1.36	0.960	1.72	0.1807	24	0.988	0.810	1.19	0.0853	57	0.983	0.710	1.24	0.1286
Yb	40	3.87	3.10	4.76	0.4131	24	3.21	2.71	3.67	0.2524	58	3.15	2.64	3.66	0.2363
Lu	40	0.590	0.450	0.700	0.0648	24	0.506	0.430	0.570	0.0336	58	0.488	0.390	0.580	0.0459
Hf	38	7.07	5.93	8.19	0.5156	24	7.59	6.42	8.82	0.6667	61	6.84	5.30	8.37	0.7840
Ta	39	1.86	1.69	2.09	0.0958	22	1.78	1.69	1.92	0.0632	58	1.84	1.54	2.12	0.1257
Th	40	17.3	14.7	19.7	1.111	24	17.6	15.2	20.0	1.279	61	16.9	13.3	19.6	1.526
U	40	7.05	4.91	9.14	1.027	23	4.29	3.73	4.88	0.3032	61	5.77	3.76	7.96	0.8808

sedimentary basin, in order to establish the chemical tracers for each one and differentiate better between the two.

The chemical data obtained by INAA for the paste composition of amphorae from PC and QR is presented in Tables 2 and 3, respectively.

Samples will be classified into groups of similar elemental composition by exploratory methods (hierarchical cluster analysis, k-means and PCA). The average concentration values and corresponding standard deviations, and the minimum and maximum values of each chemical element for each group are presented in Table 4 for the Porto dos Cacos workshop, in Table 5 for the Quinta do Rouxinol workshop, and in Table 6 for the two workshops analysed comparatively (PC and QR).

The estimated mineralogical composition of selected sherds from the PC and QR workshops is presented in Table 7.

4.1. Porto dos Cacos workshop

Cluster analysis is a rapid and efficient technique for evaluating relationships among large numbers of samples between which distance measures have been calculated. The examination of the dendrogram resulting from the cluster analysis of PC amphorae by

Table 5
Average concentration values and corresponding standard deviations, minimum and maximum values of each chemical element for the QR groups (μg/g unless specified otherwise).

	GROUP 1					GROUP 2				
	n	$\bar{x}$	Min.	Max.	*σ*	*n*	$\bar{x}$	Min.	Max.	*σ*
Na_2O, %	27	0.461	0.306	0.661	0.0903	102	0.507	0.268	0.81	0.1183
K_2O, %	27	3.67	2.76	4.59	0.5019	99	3.54	2.74	4.17	0.2958
Fe_2O_3T, %	27	6.82	4.64	8.24	0.8707	103	5.92	3.53	7.60	0.8855
Sc	27	13.6	10.8	15.6	1.290	102	13.4	9.97	16.8	1.4372
Cr	27	63.1	49.6	75.0	6.977	101	61.3	48.6	76.5	6.319
Co	27	8.74	5.36	11.1	1.553	101	8.18	4.87	11.6	1.384
Zn	27	90.7	59.9	108	12.29	102	83.5	48.9	109	12.60
As	27	10.0	5.81	16.2	2.651	101	8.08	2.32	13.6	2.260
Rb	26	217	164	255	20.19	102	206	155	222	21.62
Sb	25	0.944	0.740	1.17	0.0988	103	0.845	0.47	1.14	0.1288
Cs	26	13.9	11.3	16.1	1.213	102	13.7	8.50	17.2	1.592
Ba	25	456	360	624	71.05	96	414	319	549	51.86
La	26	41.2	35.0	50.7	4.330	97	43.0	33.5	54.7	4.539
Ce	24	77.7	69.1	90.6	6.186	96	81.9	58.4	104	9.085
Nd	26	38.8	26.5	45.7	4.378	94	40.4	26.3	55.8	5.129
Sm	27	7.59	5.50	9.17	0.836	95	7.95	5.19	10.8	1.111
Eu	25	1.37	1.14	1.63	0.1221	93	1.35	0.930	1.84	0.1807
Tb	27	1.04	0.780	1.33	0.1286	97	1.06	0.680	1.41	0.1470
Yb	23	3.27	2.89	3.6	0.1556	97	3.29	2.48	4.07	0.3791
Lu	25	0.513	0.450	0.580	0.0298	100	0.501	0.380	0.660	0.0602
Hf	27	6.30	5.53	7.39	0.5171	103	6.76	4.29	8.96	0.9586
Ta	26	1.81	1.62	1.98	0.0927	100	1.85	1.48	2.27	0.1690
Th	27	15.9	13.5	17.8	0.9761	102	16.7	13.5	20.5	1.638
U	26	3.54	2.82	4.35	0.3786	101	3.34	2.22	4.30	0.4888

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Table 6
Average concentration values and corresponding standard deviations, minimum and maximum values of each chemical element for the PC and QR workshops (μg/g unless specified otherwise).

	Porto dos Cacos					Quinta do Rouxinol				
	n	$\bar{x}$	Min.	Max.	σ	n	$\bar{x}$	Min.	Max.	σ
Na_2O, %	129	0.612	0.250	0.820	0.0865	129	0.497	0.268	0.810	0.1142
K_2O, %	123	3.23	2.78	3.72	0.2145	127	3.56	2.66	4.46	0.3576
Fe_2O_3T, %	125	4.69	3.79	5.52	0.3758	130	6.11	3.53	8.24	0.9519
Sc	129	13.2	10.5	15.7	1.110	129	13.4	9.97	16.8	1.405
Cr	128	63.4	50.0	78.3	6.553	129	61.8	48.6	79.3	6.637
Co	128	9.07	2.95	20.8	2.410	128	8.30	4.87	11.6	1.434
Zn	128	69.9	44.0	95.2	9.399	128	85.3	53.8	109	12.47
As	130	15.3	4.29	34.3	5.798	128	8.49	2.32	15.3	2.453
Rb	126	183	154	209	12.50	129	208	155	255	22.16
Sb	130	0.585	0.360	0.780	0.0960	127	0.868	0.590	1.17	0.1248
Cs	128	13.3	10.1	16.4	1.272	128	13.7	9.53	17.2	1.560
Ba	126	504	361	676	68.44	115	415	319	539	46.81
La	129	43.3	26.6	59.7	6.584	123	42.6	33.5	54.7	4.542
Ce	129	81.5	53.5	112	10.95	119	81.2	59.1	103	8.424
Nd	129	39.0	24.0	57.9	5.532	117	40.1	28.5	53.5	4.491
Sm	128	7.95	4.94	12.1	1.234	117	7.83	5.50	10.4	0.8783
Eu	130	1.29	0.460	2.31	0.2527	120	1.35	0.930	1.76	0.1797
Tb	129	1.01	0.570	1.72	0.1522	124	1.06	0.720	1.41	0.1450
Yb	127	3.18	2.44	4.53	0.3189	124	3.28	2.48	4.07	0.3598
Lu	129	0.497	0.350	0.700	0.0549	125	0.499	0.380	0.630	0.0534
Hf	129	7.00	5.30	8.82	0.8074	129	6.64	4.29	8.90	0.8823
Ta	123	1.83	1.54	2.12	0.1164	127	1.84	1.48	2.27	0.1606
Th	129	17.1	13.3	20.0	1.457	129	16.5	13.5	20.5	1.555
U	130	5.47	3.16	9.14	1.044	128	3.37	2.22	4.35	0.4808

Ward's hierarchical clustering method and Euclidean distances, combined with box-and-whisker plot graphs, emphasize the fact that five ceramic samples remain chemically distinct and cannot be attributed to one of the groups. Among these samples, there are two A51c amphorae (PC 17 and PC 19), two A51c/L3? amphorae (PC 2 and PC 40), and one L3 (PC 123) amphora.

These five samples present outliers and extreme values in the chemical distribution, whose range was defined according to the 'classic' box-and-whisker plot. Considering these criteria: (i) PC 17 presents upper extreme values of Co, and upper outlier values of Zn, Hf and Th; (ii) PC 19 presents upper extreme values of K, and upper outlier values of Na, Fe and La; (iii) PC 2 presents upper extreme values of Th; (iv) PC 40 presents upper extreme values of K, Cr, Ce, upper outlier values of Fe, La, Nd, Yb, Lu, Hf, and lower outlier values of Cs; (v) PC 123 presents upper extreme values of Zn.

Disregarding these five outliers for the further classification and grouping of samples with similar chemical composition, the data analysis points to a certain stratification of the results according to the typology (Figs. 2 and 3). This indicates that, considering the different amphorae shapes, at the PC site most of the L3 amphorae samples are distinct from the others due to their higher contents of REE, especially the heavy ones, Co and U, and lower values of Na. On the other hand, the majority of the D14 amphorae present higher values of U than the A51c and A51c/L3? amphora types. With regards to the A50 samples, the only conclusion to be drawn is that they do not fit with the L3 amphorae samples, but, due to the reduced number of samples (and also of finds), it is risky to establish a chemical tendency. Nevertheless, these samples are not outliers, and they appear to have been manufactured with the same raw materials used for the remaining PC workshop set.

This tendency of showing only a limited diagnostic chemical compositions for pottery produced at a particular workshop, is a sign of the use of similar raw materials for extended periods of time, and typologically different vessels. Nevertheless, for the PC amphora workshop, three main compositional groups may be defined after applying multivariate statistical tools, one comprising L3 amphorae (Group 1), the other consisting mainly of D14 (together with a few A51c and A51c/L3?) (Group 2), and a third, smaller one (Group 3), including A51c amphorae, together with L3 and A51c/L3? (Fig. 3).

Concerning the doubts about the allocation of the types A51c or L3 (A51c/L3?), we can consider the possibility of the existence of two groups of L3 with distinct composition, which may indicate a variation in the raw materials used over time. Therefore, there is only one group composed exclusively of L3 amphorae (Group 1), and another (Group 3) that includes samples of L3, A51c, and all the A51c/L3? samples whose typology cannot be established with precision (see Fig. 3). Thus, the typological classification of the fragments of amphorae cannot be made according to geochemical criteria, but merely by formal ones. This is because although most of the L3 amphorae have a composition distinct from the other samples, L3 amphorae are also present in another group, comprising the unidentified samples and A51c/L3?, not excluding the possibility of these belonging to the L3 typology. If one considers the analysed samples obtained from the bottoms of amphorae (six L3 and six A51c), and considering the L3 group, one is an outlier, four L3 bottom samples are included in the L3 group (Group 1), and the other in the D14 group (Group 2). Regarding the A51c bottom samples, three are included in Group 2 and the other three in Group 3 (A51c, L3 and A51c/L3?).

In terms of the samples that display potter markings, only one sample (TMM) is included in the D14 group, two samples (AIVNIT (or TINVIA retro); GERMA(ni)) are included in Group 3, and the other samples (RUSTICI; CLARIAMI; GERMAN (retro); CERF (Germanus); and non-identified) are part of Group 1.

This geochemical pattern of amphora pastes reflects, on one hand, the natural inhomogeneity of the raw material, intrinsic to the sedimentary basin, and on the other hand the recourse to diverse clay pits along the sedimentary basin, understandable considering the long period of operation of the workshop.

4.2. *Quinta do Rouxinol workshop*

Cluster analysis was also used in the initial inspection of the source data pertaining to Quinta do Rouxinol amphorae. The resulting dendrogram, using Ward's hierarchical clustering method

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Table 7
Estimated mineralogical composition for selected sherds from the PC and QR workshops (%).

	Type	Sample	Qtz	Phy	Kfs	Pl	Ant	Hem	Mul
			3.34	4.48	3.25	3.20	3.52	2.69	3.39
PORTO DOS CACOS	L3	PC63	79	16	4	–	1	–	–
		PC64	62	24	11	2	–	1	–
		PC65	55	32	11	1	1	–	–
		PC66	45	45	8	1	1	–	–
		PC67	40	43	15	2	–	–	–
		PC68	38	54	5	2	1	–	–
		PC69	52	34	11	1	1	1	–
		PC119	59	31	8	–	–	2	–
		PC122	53	38	7	–	–	2	–
		PC124	87	–	13	–	–	–	–
		PC126	88	–	6	4	–	2	–
		PC129	83	–	15	–	–	2	–
		PC130	45	42	3	3	–	1	6
		PC131	43	49	5	3	–	–	–
	A51c	PC112	68	–	5	–	–	7	20
		PC114	60	23	13	2	–	2	–
		PC115	80	–	14	4	–	2	–
		PC117	33	–	65	–	–	2	–
	D14	PC91	58	30	8	4	–	–	–
		PC92	43	44	11	1	1	–	–
		PC93	50	22	6	21	–	1	–
		PC94	86	–	12	1	–	1	–
		PC95	49	43	7	1	–	–	–
		PC96	58	28	10	3	1	–	–
		PC97	48	39	11	tr	1	tr	–
		PC98	50	41	6	2	–	1	–
		PC99	85	–	14	–	–	1	–
		PC100	50	39	9	–	1	1	–
		PC101	67	20	8	4	–	1	–
		PC102	86	–	13	–	–	1	–
		PC103	46	40	11	2	–	1	–
		PC104	51	32	6	10	tr	tr	–
		PC105	44	48	5	2	1	–	–
		PC106	68	22	9	–	–	1	–
		PC107	85	–	13	–	–	2	–
		PC108	50	35	11	3	–	1	–
		PC109	42	48	6	3	1	–	–
		PC110	63	21	12	3	–	1	–
		PC111	48	41	7	2	1	1	–
QUINTA DO ROUXINOL	A51c	QR2	61	28	8	1	2	1	–
		QR3	74	15	9	1	1	–	–
		QR4	83	–	15	–	–	2	–
		QR5	64	23	11	2	–	–	–
		QR6	93	–	7	–	–	–	–
		QR7	64	27	7	–	–	2	–
		QR8	72	14	13	–	1	–	–
		QR9	55	28	9	7	1	–	–
		QR10	75	10	14	–	–	1	–
		QR11	81	–	15	4	–	1	–
		QR12	88	–	11	–	–	1	–
		QR13	42	51	4	1	1	1	–
		QR14	61	31	7	–	1	–	–
		QR174	85	–	10	–	–	5	–
		QR175	37	56	3	–	3	1	–
		QR177	49	40	6	3	–	2	–
		QR179	91	–	8	–	–	1	–
	A50	QR41	88	–	12	–	–	–	–
		QR42	70	20	8	1	–	1	–
		QR43	64	22	9	3	2	–	–
		QR44	90	–	10	–	–	–	–
		QR45	64	27	6	2	1	–	–
		QR46	57	31	9	2	1	–	–
		QR47	79	11	10	–	–	–	–
		QR48	88	–	12	–	–	–	–
		QR49	67	19	10	4	–	–	–
		QR50	45	46	6	Trace	2	Trace	–
		QR51	62	27	6	2	2	1	–
		QR153	87	–	10	–	–	3	–
		QR161	33	56	7	2	–	2	–
		QR167	42	50	5	2	–	1	–

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Table 7 (*continued*)

Type	Sample	Qtz	Phy	Kfs	Pl	Ant	Hem	Mul
		3.34	4.48	3.25	3.20	3.52	2.69	3.39
	QR168	82	–	8	4	–	6	–
	QR180	87	–	10	–	–	3	–
L9	QR141	35	55	8	1	–	1	–
	QR144	84	–	13	–	–	3	–
	QR145	43	51	6	–	–	–	–
	QR146	86	–	13	–	–	–	–
	QR147	57	36	6	–	–	1	–
	QR149	30	58	7	4	–	1	–
	QR150	93	–	7	–	–	–	–
	QR151	49	46	4	–	–	1	–

Qtz – quartz; Phy – phyllosilicates; Kfs – K-feldspar; Pl – plagioclase; Cal – calcite; Ant – anatase; Hem – hematite; Mul – mullite

and Euclidean distances, clearly shows the order and levels of clustering, as well as the distances between individual samples. It is also important to emphasize that the five sub-samples analysed from the same L9 amphorae sherd present a very high level of resemblance, of identical degree to the rest of the group, which is a very good indicator of the high-quality clustering of these compositional groups. Cluster analysis also defines the existence of three chemically distinct samples from the QR workshop. Other statistical approaches (box-and-whisker plot graphs, PCA) point to the same distinct samples, representing one sample of each analysed typology: (i) QR3 (A51c) presents upper extreme values of Ba, lower extreme values of Cs and upper outlier values of Na; (ii) QR83 (L9) presents upper extreme values of Sb, lower extreme value of Ba and upper outlier values of Nd, Sm and Eu; (iii) QR180 (A50) presents upper extreme values of Zn.

Initially, on the basis of typological evidence, we tried to identify eventual chemical differences according to this criterion, but all of the specimens appeared to be compositionally quite similar, and had proven almost indistinguishable in earlier studies (Cabral et al., 1993–1994; Cabral et al., 2002). Even considering some ‘overlap’ in the chemical classification of QR samples, because a clear separation is not to be expected in the statistical analysis of chemical data, further studies (Dias et al., 2001) emphasize the possibility of slightly differentiating L9 amphorae from those belonging to the Almagro 51c typology, especially according to the iron enrichment and the higher chemical homogeneity. On the other hand, A51c amphorae present a wider spread of data from a compositional point of view. In fact, none of the bivariate plots examined so far convincingly distinguished QR typologies from each other, but we observed that, with the exception of a few L9 samples, the other L9 fragments were indeed distinguishable, as shown in Figs. 4 and 5, especially due to higher contents of Fe, as well as lower contents of REE, Ta, Hf, Th and Na.

From an archaeological point of view, this is an acceptable hypothesis, identifying a more heterogeneous chemical composition for A51c samples, especially as compared to L9. This was probably due to a longer period of production, allowing for a greater need to use diverse outcrop exploitations of clay raw materials.

4.3. *Porto dos Cacos and Quinta do Rouxinol workshops: a path to their differentiation*

The goal of the statistical analysis employed in this work is to isolate and refine a reference group for pottery production at each production centre (PC and QR) for further comparison to consumption centres, and also other kilns, in order to allow a better differentiation between them and to establish a geochemical signature for each workshop.

Several statistical approaches were carried out to this purpose, using both absolute and normalized values, coupled with diverse analyses of geochemical data, namely trace element data, in accordance with their geochemical behaviour and distribution by geo-environment.

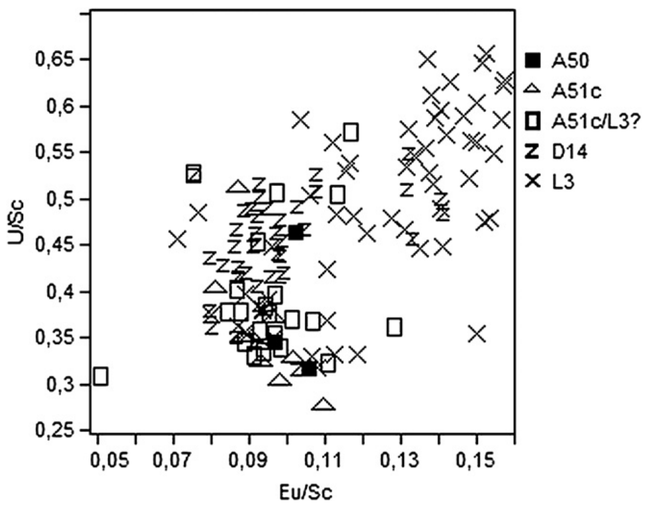

Fig. 2. Bivariate plot of Uranium versus Europium (both normalized to Sc) for the Roman amphorae from the Porto dos Cacos workshop, according to the typological classification.

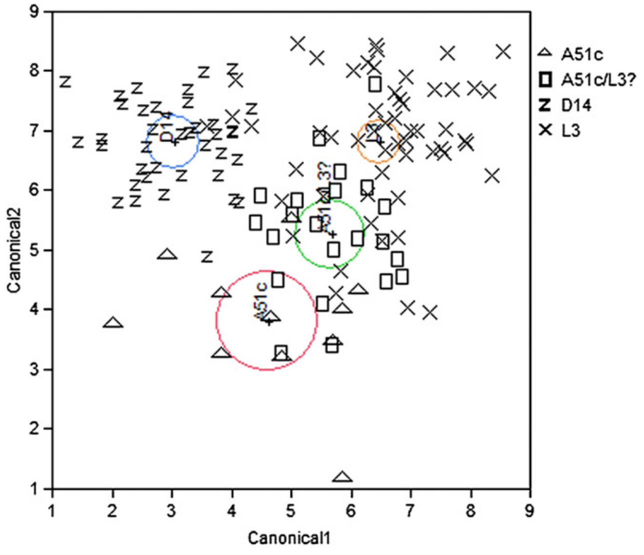

Fig. 3. Biplot showing amphorae samples from Porto dos Cacos on canonical roots 1 and 2 (chemical data normalized to Sc). Normal 50% contours are visible, containing roughly 50% of the points for that group.

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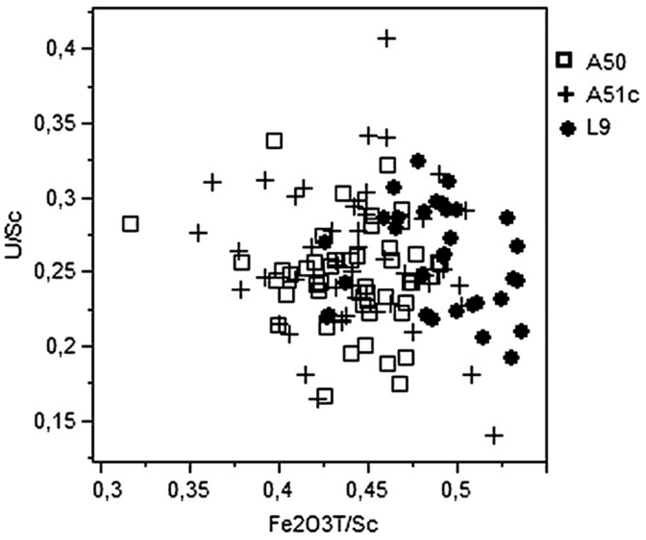

Fig. 4. Bivariate plot of Uranium versus Iron (both normalized to Sc) for the Roman amphorae from the Quinta do Rouxinol workshop, according to the typological classification.

Considering all the amphorae sherds analysed for each workshop, the resulting dendrogram using Ward's hierarchical clustering method and Euclidean distances suggests the existence of five outliers from the PC workshop (PC2, PC17, PC19, PC40, PC123) and two from the QR workshop (QR3, QR180), displaying the features mentioned above, with the exception of QR83, which also presented the lowest parting degree.

After disregarding these seven outlier samples, a new statistical approach has been taken in order to better define compositional groups. A first approach, also by cluster analysis applied to chemical elements normalized to Sc as variables, using Ward's amalgamation rule and the Pearson correlation coefficient, suggests the existence of two clusters (Fig. 6), both comprising samples from both the PC and QR workshops. Nevertheless, they are well separated, each sub-group including samples from only one workshop. The pottery

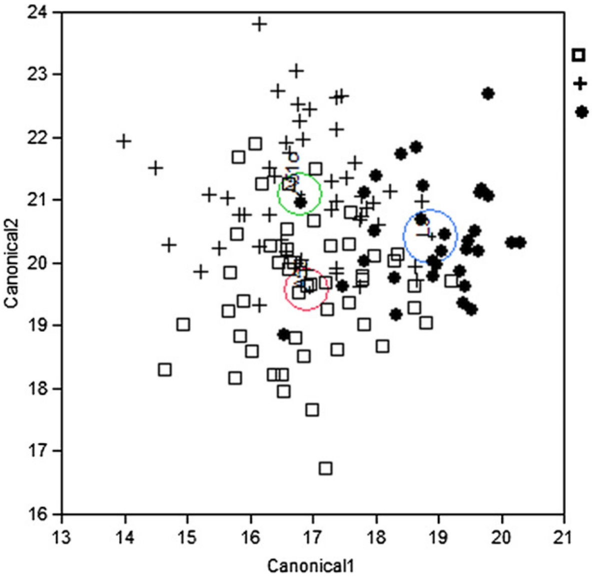

Fig. 5. Biplot showing amphorae samples from Quinta do Rouxinol on canonical roots 1 and 2 (chemical data normalized to Sc). Normal 50% contours are visible, containing roughly 50% of the points for that group.

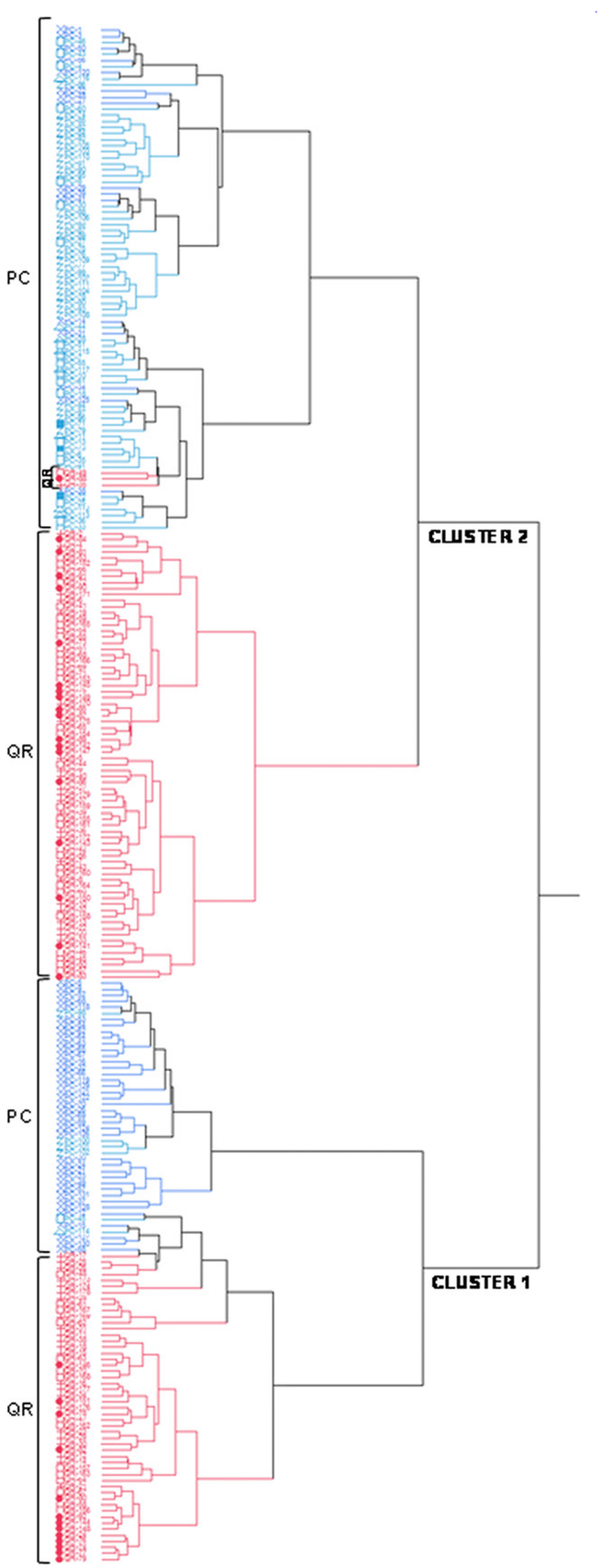

Fig. 6. Classification of amphorae from Porto dos Cacos and Quinta do Rouxinol considering typological taxonomy. The hierarchical cluster analysis was applied to chemical elements (normalized to Sc) using Ward's amalgamation rule and the Pearson correlation coefficient.

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from the two sites indicates clear intra- and inter-site interactions. There appears to be an intra-site distribution, as the presence of sub-groups within the ceramics from these two workshops implies the use of more than one clay source, and, at the same time, the existence of clusters comprising samples from both sites indicates the natural geochemical heterogeneity of this estuary basin. It is important to emphasize that some correlation with the typology has been established, again more evident for PC amphorae, as cluster 1 comprises mainly PC samples of L3 shape (Fig. 6). The other sub-group of cluster 1 comprises 50 QR samples of various typologies, and all the remaining samples are included in cluster 2, but with a clear distinction between PC and QR workshops (only samples QR 49 – A50, QR 88 – L9 and QR 35 – A51c are mixed within the PC sub-group of cluster 2; the other sub-group of cluster 2 only contains QR samples). It is important to underline the difficulty of assigning with certainty PC samples to the PC workshop and QR samples to the QR workshop by using this statistical approach, since, as mentioned above, some PC samples group with QR samples.

Principal component analysis (PCA) is one of the most frequently used techniques for exploring underlying relationships in multivariate data. Canonical discriminant analysis (CA) consists of finding a linear combination which gives the maximum ratio of the variability between groups applicable to the variability within groups. A PCA of the standardized chemical data identified the seven chemical outliers already suggested by cluster analysis. The seven outliers were subsequently omitted from all further analyses, to allow formal comparisons between different plots. Thus, we have repeated the PCA (Fig. 7) and CA (Fig. 8) analyses after this omission, and results are labelled according to whether the samples belong to the PC workshop or the QR workshop, also considering the typological classification. Following a principal components analysis that selected four components accounting for 70.79% of the original variation, two well-defined groups were mapped into the components space, each belonging to a specific workshop, especially considering factors 1, 2 and 3 (accounting for 64.80% of the original variation). In terms of the first principal component of the loading plots, lanthanides, actinides and Hf, Zr, Ta display large positive loadings, while Sb and Fe have a negative weight, and Rb, Zn, K, Cs, Ba, Cr, As have a very low positive weight. On the other hand, the second component clearly separates a set of elements, with large negative loadings, from the one including Cr, Co, As, Ba and U, and from another with positive loadings comprising K, Fe, Rb, Cs, Zn, Sb and Ta. The corresponding score plots show two groups (the outlier samples were omitted), each one corresponding to one of the workshops. The PC workshop is characterized by high concentrations of Co, As and U and low concentrations of the elements presenting large positive loadings on the second component (Fe, Zn, Sb, Rb, K); the other group, comprising QR workshop amphorae, shows instead large concentrations of Fe and Sb, and also of Rb and Zn. None of the two workshops shows a particular composition with respect to rare earth elements. Two robust clusters of the major sites are also clearly visible in the CA analysis (Fig. 8), corresponding to the samples from the PC and QR workshops.

Bivariate plots (after normalization to Sc) were also sometimes sufficient to reveal patterns in the chemical data, especially if they include iron and uranium (Fig. 9), as well as antimony and zinc,

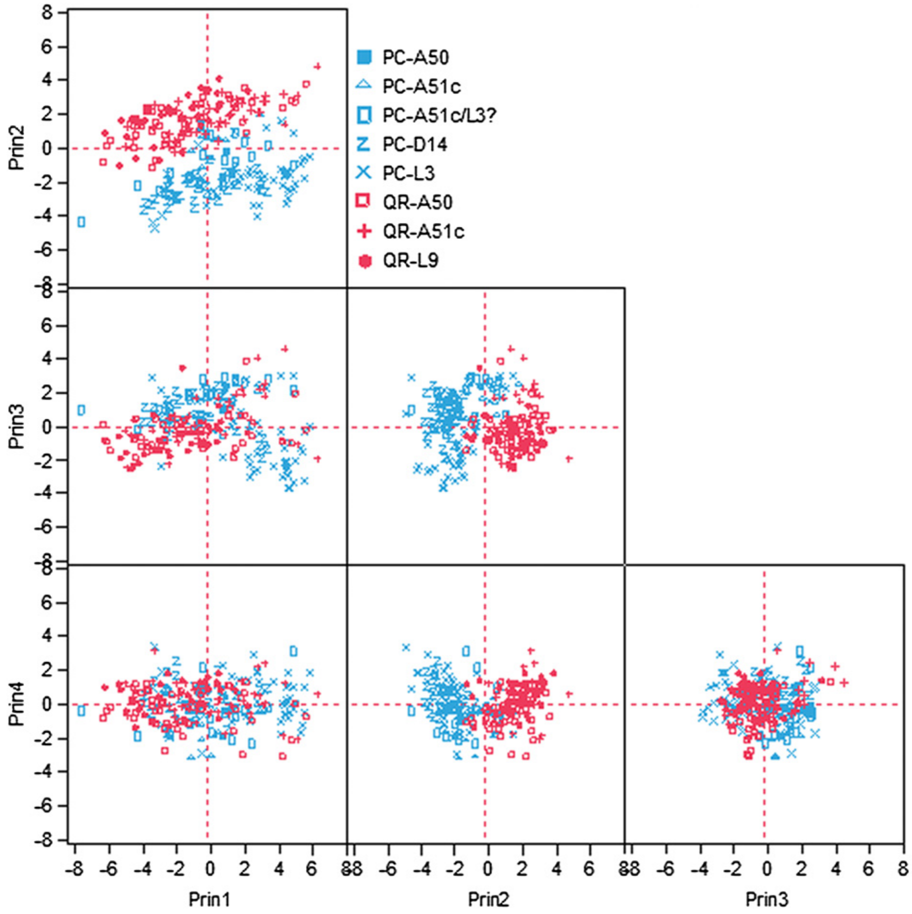

Fig. 7. A two-dimensional component plot based on PCA of the PC and QR chemical data (normalized to Sc), omitting seven outliers (five from PC and two from QR).

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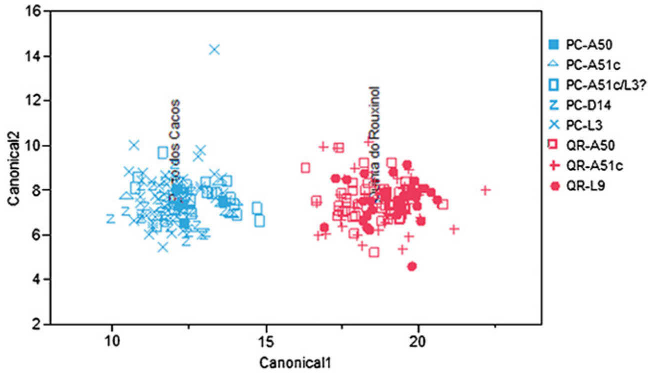

Fig. 8. A two-dimensional canonical plot based on CA of the PC and QR chemical data (normalized to Sc), omitting seven outliers (five from PC; two from QR).

antimony and uranium, arsenic and uranium, iron and zinc. According to these patterns, certain location-specific reference sets were defined, and the members of each set were ceramic specimens from each workshop.

In terms of the mineralogical composition obtained by XRD, quartz is the dominant mineral phase in almost all samples, particularly in QR analysed sherds (Table 3). The XRD analyses revealed, in addition to a great quantity of quartz, lesser amounts of mica (the only phyllosilicate detected) and K-feldspar, and finally plagioclase, hematite and anatase in trace amounts. The presence of mullite was only detected in two samples from PC kilns. XRD provided the key to the reconstruction of the firing temperatures that varied around 900 °C, and were higher than 1000 °C only in two cases. The range of the firing temperatures was determined according to the stability of mica components and the newly formed mineral phase of mullite, which is encountered when aluminous clay minerals are heated to temperatures of 1000 °C and above. Thus, the XRD data indicated relatively high firing temperatures, with some variability due to a few low-fired fragments and a very limited number of samples fired at temperatures higher than 1000 °C.

As a result, as expected considering the geological context of the Tagus estuary, the mineralogical composition obtained by XRD does not differentiate the two workshops, reflecting once again the natural inhomogeneity of the raw material, intrinsic to the sedimentary basin, with inputs belonging to diverse geological contexts.

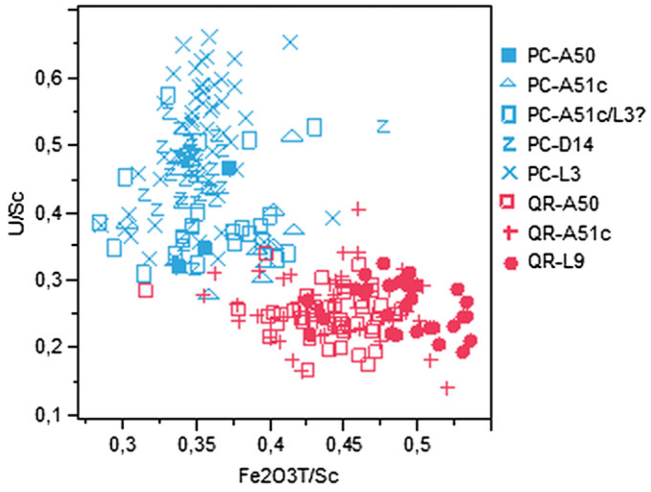

Fig. 9. Bivariate plot of Uranium versus Iron (both normalized to Sc) for amphorae from the two workshops (PC and QR).

The chemical variability within the PC workshop points to raw materials presenting a general enrichment of Co, As and U. Cobalt concentrations usually reflecting the abundance of mafic minerals are common in very fine-grained mudrocks, arsenic is usually concentrated in clays, hydrous Fe and Mn oxides, sulphides and phosphates, and uranium is closely related to redox conditions, representing the U enrichment attributed to oxidation-reduction processes. Thus, the high contents of these three elements may be related to the mixture of raw materials with brackish clays, as already noticed for the Zambujalinho workshop in the Sado basin (Prudêncio et al., 2009), where the high contents of U were associated with the organic matter present in the sediments of that estuarine environment.

In the QR workshop, the paste of the amphorae is particularly enriched in Fe and Sb (and also Rb and Zn). Fine-grained argillaceous and organic-rich sediments are typically enriched in antimony relative to their parent lithologies, reflecting the tendency for the element to become absorbed by hydrous oxides, organic residues and clay minerals in favourable environments (Ure and Berrow, 1982). The abundance of iron in sedimentary environments is determined by various factors, such as origin, being generally enriched in mafic rocks relative to felsic. Secondary hydrous oxides represent the dominant Fe phase, and there is a tendency for these hydrous Fe phases to form surface oxide coatings, reflecting a direct relationship between the total Fe content and the specific surface areas of particles (Ure and Berrow, 1982), consequently resulting in clays generally enriched in this element.

5. Conclusions

By providing geochemical reference groups for the PC and QR Roman production centres, with fine differentiation between them, the obtained results contribute significantly to our understanding of the nature of amphora production on the lower Tagus. This is especially important because the base raw materials at both sites are of a similar type, and the fine discrimination relies mainly on trace elements compositional differences revealed by INAA and related geochemical /chemometric approaches.

From a technological perspective, this work allowed us to estimate the firing temperatures of the amphorae, confirming the relatively high temperatures reached by these kiln types. The presence/absence and abundance of high-temperature phases confirm the indications pertaining to the temperatures reached inside the kiln, and the presence of mica has been shown to indicate firing temperatures lower than 900 °C. Another important technological aspect is the evidence of the mixture of raw materials with brackish clays in the PC workshop, as pointed out by the high contents of Co, As and U; also, in the QR workshop, the enrichment in Fe and Sb presented by the paste of amphorae seems to indicate the use of fine-grained argillaceous and organic-rich sediments.

This study of lower Tagus Roman ceramic production centres that sought to establish the geochemical fingerprints of each centre will be useful in further works of provenance and sourcing studies (comparison with consumption centres and with other regional /trans-regional workshops), contributing to the ascertainment of merchandise distribution patterns in Roman trade and their impact in Lusitania.

Once a framework for reference has been established, significant aspects related to pottery production and to the regional and imperial economy (in terms of production, distribution and consumption) will become clearer, thus confirming the potential of

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this type of study for understanding the social and economic history of Antiquity.

Ultimately, we may also revise our perception of the model of this type of kiln workshops, clarifying their function not only as simple production centres, but as veritable poles of regional economic development. This will also make possible clarifying the relationships and the levels of interdependence between pottery centres and the fish salting and transforming units related to them at a regional scale.

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