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CHEMICAL VARIABILITY IN CLAYS AND POTTERY FROM A TRADITIONAL COOKING POT PRODUCTION VILLAGE: TESTING ASSUMPTIONS IN PERERUELA

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Chemical variability in clays and pottery from a traditional cooking pot production village: Testing assumptions in Pereruela

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ABSTRACT

This paper explores analytically the contemporary pottery-making community of Pereruela (Northwest Spain) that produces cooking pots from a mixture of red clay and kaolin. Analyses by different techniques (XRF, NAA, XRD, SEM and petrography) showed an extremely high variability for cooking ware pottery produced in a single production centre, by the same technology using local clays. The main source of chemical variation is related to the use of different red clays and the presence of non-normally distributed inclusions of monazite. These two factors induce a high chemical variability, not only in output of a single production centre, but even in the paste of a single pot, to an extent where chemical compositions from one 'workshop', or even one 'pot', could be classified as having different provenances. The implications in the chemical characterisation and in provenance studies of archaeological ceramics are addressed.

KEYWORDS: ETHNOARCHAEOMETRY, CERAMICS, COOKING POTS, PROVENANCE, SPAIN, MONAZITE, NON-NORMAL DISTRIBUTION

INTRODUCTION

The analytical study of living pottery-making communities has increased in the last decades as a way to understand all stages of pottery production and to test methods and assumptions that form the basis of the analytical approach to archaeological ceramics. Among other aims, ethnoarchaeological cases have been particularly addressed to test some basic assumptions in provenance studies (i.e. Arnold *et al*.1991, Arnold *et al*. 2000) or as a basis for technological inference (Picon 1995). Indeed the possibility of documenting all stages and parameters -such as raw materials or paste recipes used by the potters- involved in pottery production converts archaeometric studies of contemporary pottery-making communities into 'cases with known solutions'. This is an ideal situation to confront analytical results and interpretations, and therefore methods and assumptions used in provenance studies. The application of archaeometric approaches to ethnoarchaeological cases constitutes an area of study which we suggest calling *ethnoarchaeometry*, as the term ethnoarchaeology has been used for ethnographic based studies carried out with an archaeological scope.

It is common practice in pottery provenance studies by chemical analysis to create 'control groups'. These groups are created to provide chemical profiles for the pottery produced in particular areas of study, and are used for comparisons with other pottery under investigation. Formation of these 'control groups' is done by the analysis of pottery from kiln sites, or by analysing pottery from consumption centres to form groups that are then attributed, on archaeological grounds, as presumably local to a certain area. In many cases, both approaches are assisted by the analysis of clays coming from the assumed production area.

Several assumptions underlie these approaches. Firstly, it is postulated that variability within one clay source is smaller than variability between different sources. A consequence of this is the common working hypothesis that pottery produced in a particular production site, by potters using the same technology and local clays, should exhibit chemical homogeneity and for this reason should be used as a reference group. Another basic assumption that has to do with the analysis itself is that the sample taken from each pot is, once homogenised, representative of the bulk chemical composition of the pot, provided that the amount sampled for analysis is over the minimum requirements. Usually, these are over those set by Brommund *et al*. (1976) in relation to maximum inclusion size and frequency. Finally, in terms of statistical analysis of the

analytical elemental data, most of the approaches and statistical techniques applied in archaeometric pottery studies assume that the elemental concentrations, either raw or logarithmic, follow a multivariate normal distribution.

In order to test some of the above assumptions, an analytical programme for the study of the contemporary pottery-making community of Pereruela was carried out including pots, paste and clays from the potters of Pereruela.

THE POTTERY-MAKING COMMUNITY OF PERERUELA

The village of *Pereruela de Sayago* lies around 15 km to the Southwest of the city of *Zamora* in Northwest Spain in an area of some geological complexity. Granitic rocks and a Pre-Ordovician metamorphic series, completed by minor Tertiary materials and Quaternary sediments dominate the village and its surroundings (Figure 1) (IGME 1980). The village lies in an area of low-grade gneiss with quartz, alkaline-feldspar and plagioclase, with muscovite and biotite as subordinates. This gneiss is similar in composition to the granites that appear in areas nearby. Series of schist and gneiss lie west of the village. This metamorphic outcrop is an alternating succession of schist and gneiss. The schist is generally mica-schist and the gneiss is differentiated rather by texture than by composition. An important megacristals granitic formation lies south of the village and it is characterised by a coarse granite composed of large alkaline-feldspar crystals, plagioclase, biotite, muscovite and several accessory minerals. Red clays have been formed *in situ* deriving from both the granitic and metamorphic rocks. Finally, the kaolin used for pottery making comes from the Tertiary outcrops of Lehm located at the outskirts of the village. This formation develops *in situ* on the granitic basement and presents quartz, highly altered feldspars and micas, with a general white to light grey colour.

In the 1950's, Cortés recorded the main characteristics of the pottery-making tradition from an ethnographic point of view (Cortés 1954). Since then Pereruela has been cited in several publications (i.e. Sempere 1982, Vossen *et al*. 1975, Cau 1998) and it has been used as example of household industry (Peacock 1982). Pottery production was a seasonal activity complementary to an economic system based on agriculture. The raw materials used were red clay (*bermejo*), collected in different locations, mixed with white clay/kaolin (*tierra*), extracted from a communal outcrop, in a proportion of 1:1. The pots were formed by women using the coiling technique with the aid of a turntable (*rueda*).

The production focussed on cooking pots, especially casseroles (*cazuelas*), kilns (*hornos*) and metallurgical crucibles (*crisoles*). In this sense, kaolin provided the necessary refractory properties to the pot, while the red clays were responsible for the workability of the paste. In this way all potters produced pastes with the same technological characteristics. In the cases where the work was carried out as a couple (*en pareja*) the men were in charge of collecting the raw materials, glazing, firing and trading the pots.

Nowadays, little has changed in relation to the use of raw materials, and potters still mix red clay extracted from several locations near the village and kaolin from the communal outcrop. Other aspects of pottery production have changed. Thus, although some women still work only seasonally, several workshops work all year on a full-time basis. The wheel has been introduced for reasons of comfort but this seems to have little effect on the forming process that is still carried out following the coiling technique. The typology has diversified and new forms have been introduced, although *cazuelas* and *hornos* are still the most distinctive products. Male potters are increasingly involved in the forming process and we interviewed at least two of them working on a full-time basis. Also firing processes have changed and many potters now use gas or fuel kilns instead of the traditional kilns. Finally the production has been intensified, trading systems have changed and trading networks have expanded.

MATERIALS AND METHODS

Sampling

For the analytical programme of this study pottery samples of unused pots were collected from four potters (Table 1): Redondo (labelled RED, n=13), Pastor (PAS, n=5), Ramos, (RAM, n=12) and Riesgo (RIE, n=16). All potters are active in cooking pot production and trade, except Redondo who recently retired but still had pottery in her warehouse. Red clays (RCL, n= 8) were collected either at the workshops of the potters or from the clay beds used by them. All kaolin samples (KAO, n=4) came from the communal outcrop located very close to the village (50 m from the last houses). Finally, one clay paste (PST, n=1) already prepared for potting by Riesgo was also included.

For reasons explained in the discussion, one pot coming from Redondo (RED001) was divided into forty sections and analysed separately. This casserole, which had the shape

of a flat pan 34 cm in diameter and 13 cm high, was divided into 40 sections along the rim. Each section contained part of the rim and the corresponding piece of the body. The section weights were 10 cycles of 10, 15, 20 and 25 g in order to provide the maximum randomness in sampling. Also eleven pots from Redondo (RED049 - RED059) were analysed taking three different samples. These pots were smashed and three sherds were chosen randomly. Furthermore, multiple sampling of three red clays was carried out by crushing the original clay ball and randomly selecting several pieces to be analysed. Four samples were taken from RCL035 and five samples from each of RCL043 and RCL047. Moreover, RCL043 was fractionated into four different grain sizes: coarse sand >500 μm, medium and fine sand 500-70 μm, silt 70-20 μm and clay <5μm. About 450 g of this red clay was first gently crushed in a wooden mortar and then the first two fractions were taken by wet sieving, while the other two by levigation. The fractions were left to dry in air. Finally, they were powdered and homogenised in an agate mortar and kept for analysis.

Samples from potsherds were taken after mechanical cleaning with a diamond drill-bit, followed by cutting a piece of at least 15 g, which was homogenised in a tungsten carbide mill, and kept for analysis. Red clays, kaolin and the prepared paste weighed at least 1 Kg each. Portions of about 15 g were taken and powdered and homogenised in a tungsten carbide mill.

All samples were analysed by neutron activation analysis (NAA) and X-ray fluorescence (XRF) for major, minor and trace elements. A representative selection of pots from the four potters (8 individuals, including a multiple sampling of individual RED001) was also examined by X-ray diffraction analysis (XRD). The clay fractions (from RCL043) were analysed by NAA only. Moreover, thirteen samples from pots, red clays, kaolin and prepared paste were examined by thin section analysis under a polarising microscope. Finally, five samples from a single pot were examined under the scanning electron microscope attached to an energy dispersive X-ray analyser using the back-scattered electrons detector (SEM-EDX-BS).

Methods

For NAA, a portion of specimen was left overnight to dry at 120^{o}C and approximately 150 mg from each was weighed and heat sealed in polyethylene vials. The same procedure was followed for the reference materials used, an International Atomic

Energy Agency SOIL-7 and an in-house Lefkandi Brick standard. Individuals and standards were irradiated in batches of ten (8 individuals and two standards) at the swimming pool reactor of NCSR 'Demokritos' at a thermal neutron flux of $3x10^{13}$ $n.cm^{-2}.s^{-1}$. Eight days after irradiation, the individuals and standards were measured for Sm, Lu, U, Yb, As, Sb, Ca, Na, La and 20 days after irradiation for Ce, Th, Cr, Hf, Zr, Cs, Tb, Sc, Rb, Fe, Ta, Co and Eu.

XRF was performed using a Phillips PW 2400 spectrometer with a Rh excitation source. Portions of the specimens were dried at 100°C for 24 h. Major and minor elements were determined by preparing duplicates of glassy pills using 0.3 g of powdered specimen in an alkaline fusion with lithium tetraborate at 1/20 dilution. Trace elements and Na_2O were determined by powdered pills made from 5 g of specimen mixed with Elvacite agglutinant placed over boric acid in an aluminium capsule and pressed for 60 s at 200 KN. The quantification of the concentrations was obtained using a calibration line performed with 56 International Geological Standards. The elements determined comprised Fe_2O_3 (as total Fe), Al_2O_3, MnO, P_2O_5, TiO_2, MgO, CaO, Na_2O, K_2O, SiO_2, Ba, Rb, Mo, Th, Nb, Pb, Zr, Y, Sr, Sn, Ce, Co, Ga, V, Zn, W, Cu, Ni and Cr. The loss on ignition (LOI) was determined by firing 0.3 g of dried specimen, at 950°C for 3 h.

The complete chemical data-set was produced by joining the NAA and XRF data. For the common elements Na_2O, CaO, Fe_2O_3, Rb and Zr concentrations determined by XRF were selected, while for Ce, Th and Cr only NAA results were taken. Several elements were discarded: As and Sb due to the high variability, Tb, Mo and Sn due to the low counting statistics, and Co, Ta and W because of the possible contaminations from the tungsten carbide cell of the mill.

XRD measurements were performed using a Siemens D-500 diffractometer working with the Cu Kα radiation (λ=1.5406 Å), and graphite monochromator in the diffracted beam, at 1.2 kW (40 kV, 30 mA). Spectra were taken from 4 to 70°2Θ, at 1°2Θ/min (step size=0.05°2Θ; time=3 s). The evaluation of crystalline phases was carried out using the DIFFRACT/AT program by Siemens, which includes the Joint Committee of Powder Diffraction Standards (JCPDS) data bank.

Petrographic analysis was carried out by thin-section using a polarising microscope Leica Laborlux12 POL S working between X25 and X400 magnification. For the red

clays, kaolin and the prepared paste, briquettes were formed and fired at 300°C prior to thin sectioning. All the samples were impregnated with Epoxin resin and mounted using Loctite UV glue, thin-sectioned using a Struers Discoplan TS and finished by hand to 30 μm thickness.

Finally, polished sections of five samples from a single pot from Redondo (RED001) were examined by SEM-EDX-BS, using a Stereoscan S120 (Cambridge Instruments) working at 20 KeV for the characterisation of heavy minerals which are highlighted over the ceramic matrix, usually composed by minerals whose mean atomic number lies around 11 (Jones 1987).

RESULTS AND DISCUSSION

Chemical variability

In order to evaluate the chemical compositions, elemental concentrations were transformed to logratios using Sc as divisor (Aitchison 1986; Buxeda 1999). Sc was selected because it was the element with lowest variability within the dataset, and therefore most likely this variability to represent natural variation.

As a first approach, a data set consisting of one sample from each pot (in total 46), red clays (16 samples, including the multiple sampling for RCL043 and RCL047), kaolinite (4 samples) and the prepared paste (1 sample) were evaluated. Principal Component Analysis on the covariance matrix was performed, with no rotation of axis, using the BMDP4M routine (Dixon 1983). The plot for the two first principal components, which account for 68.44% of the total variance, is shown in Figure 2. It is readily apparent in this plot that there is a considerable spread, considering that all clays and pots come from the same production site. In statistical terms the spread is a reflection of the high total variation (vt=4.3665) (Table 2), given by the Variation Matrix of this dataset (Buxeda and Kilikoglou in press). However, there are several patterns in the grouping of the samples that are included in the plot of Figure 2. The pottery samples are divided into three groups. The first contains the samples taken from Redondo and Pastor, the second the samples from Riesgo and the third the samples from Ramos. The average values along with the standard deviations in percent for each of these groups are given in Table 3, where significant differences in the absolute values can be observed (especially for La, Ce, Cs, Fe_2O_3, K_2O, Ba and Zr among others). The picture for the

clays is quite similar. The red clays are mainly divided into two groups, some remain single (Figure 2), the kaolin samples scatter all over the plot and the prepared paste from Riesgo is located among the pots of this potter. At the same time, none of the clays cluster firmly with the pottery samples analysed. Some red clays as well as Redondo and Pastor pots are attracted to PC2+, which is strongly influenced by the relative concentrations of REE, Th and Y and also some major and minor elements (MgO, Fe_2O_3 and K_2O). On the opposite side, Ramos and Riesgo pots and the prepared paste are attracted to PC1+. Furthermore, both groups of clays are separated by differences in CaO, Cs and Rb relative concentrations. A second group of red clays is clearly attracted to PC1- showing clear compositional differences. Finally, the kaolin are scattered but mainly attracted by PC2-.

By removing raw materials and the prepared paste from the previous data-set and performing again the calculation of the Variation Matrix on the data-set containing only the pottery samples, the total variation drops significantly to a value of vt=2.2106 (Table 2). This value is now half of the initial total variation. This decrease is due to the great chemical differences among pots, red clays and kaolin. In fact, this is clearly reflecting the mixing practice of the potters, which lead to a chemical composition for the final products very far from the composition of the original raw materials. Despite this decrease in total variation, chemical variability is still too high for an assumed monogenic population, for which values as low as 0.2 could be expected (Buxeda and Kilikoglou, in press). Moreover, any multivariate data treatment will still show a clear data structure in three different groups for the pots. If this was an archaeological case study, the existence of the three chemical groups, could be certainly interpreted as three different reference groups (Table 3). In the present case the groups cannot be linked to differences in proportion of mixed clays or typological/functional issues, as those are constant for all potters, but neither to different provenance in terms of production centre. Therefore, the existence of these three groups can be attributed to the diversity of red clay sources related to differences in the raw materials procurement, given that kaolin is collected by all potters from the same communal outcrop. Indeed, red clays are collected in different clay pits in the vicinity of the village, either in fields of their own property or in others with the permission of the landowners. Thus, exploitation of red clays is an activity scattered all around the village both in metamorphic and igneous terrains.

Petrographic and mineralogical variability

At a next step thin sections of the same samples were examined qualitatively under the petrographic microscope. In this case, all pots coming from different potters seemed petrographically very similar and the tendency was to gather them as a unique group, accepting some degree of variation. In thin-section, the pottery from Pereruela is characterised mainly by a groundmass that is brown-reddish in plain polarised light (PPL) to reddish in cross polars (XP), normally optically inactive, with high packing and poorly sorted non-plastics ranging from silt to coarse sand and sporadically very coarse sand. The main aplastics include quartz, highly altered alkaline-feldspar, abundant micas and fragments of metamorphic and granitic rocks, less abundant plagioclase, opaques and heavy minerals. This similarity in the final products is due to the mixing of red clays with kaolin. The addition of kaolin, which contains large amounts of rock fragments, alkaline-feldspar, plagioclase, micas and other mineral phases, blurs in the finished products the differences that can be observed in the red clays. Indeed the addition of kaolin to form the paste has a 'homogenising' effect that tends to obscure the differences existing in the red clays. Riesgo's red clay (RCL035) presents a relatively high amount of muscovite and metamorphic contribution, while Redondo's red-clay (RCL043) is relatively poor in muscovite, but richer in alkaline-feldspar, biotite and granite inclusions. This seems to be reflecting the differentiation in the red clays collected from the area dominated by metamorphic formations, close to the village, and the red clay sources located further East-Southeast deriving from the granite terrain.

In summary, the pottery analysed chemically belongs to three groups, one containing the Redondo – Pastor samples, the second Ramos and the third Riesgo, with most profound differences between the pairs Redondo – Pastor and Ramos – Riesgo. Petrographically they all look relatively homogeneous and the two pairs can only be separated after studying the raw materials. Nevertheless, chemical variability in Redondo - Pastor's group can also be considered as too high, especially in the rare earth elements (REE), Th and Y (Table 3). In order to explore this variability, two more arbitrary samples from all Redondo's pots (except RED039) were analysed, indicated as A (the original), B and C. Total variation for this data set drops significantly (vt=0.4748), being still high for what we could expect *a priori* for a monogenic population (Table 2). The cluster analysis dendrogram, performed using the mean square euclidean distance and centroid agglomerative method with Clustan (Wishart

1987), shows two main features (Figure 3). First, there is a complicated structure with several groups as well as several individuals placed at the left far side of the dendrogram, as outliers. Second, different samples from the same pot, at least for individuals RED001, RED053, RED056 and RED058, are clustered in different groups. This indicates that the raw materials used by Redondo introduce some variability affecting not only the homogeneity of Redondo's pottery production as a whole, but also the variability within a single pot. This single pot variation has been further explored by analysing up to 40 samples from one single pot, which was the casserole RED001. Labels for the resulting 40 samples are now assigned following the cycle and weight system explained in the *Sampling* paragraph. The results show again a high chemical variability, as reflected by the total variation value (vt=0.5982) (Table 2), especially considering that it accounts for variability within a single pot. Theoretically, by taking 40 samples from the same pot, which had never been used or buried, and with samples ranging from 10 to 25 g of homogenised powder, the only expected variability should be the sum of the analytical, plus the natural which exists in homogeneous samples following a multivariate normal distribution. As has been already pointed out, the main source of variability is related to REE, Th and Y which is demonstrated in Figure 4, where elemental concentrations of La and Th (representing the REE, Th and Y contents) have been plotted for the 40 samples, together with elemental concentrations of Sc and Hf (representing the stable elements). As can be seen, La and Th concentrations vary in a totally random way, not affected by the sampling area of the pot or the weight of the sample. When these elements (Sm, Lu, Yb, La, Ce, Th, Eu and Y) were removed and the variation matrix was re-calculated, the total variability dropped drastically to vt=0.0654 (Table 2). If we calculate the mean value for the cells from this Variation Matrix, and we use this value as an estimation of a possible contribution to the chemical variability for the removed elements, the total variation for all the elements in those 40 individuals would be vt=0.0948 (Table 2). This final value is now low enough to represent the expected result coming from multiple analyses of the same pot.

The effect of monazite

The study of polished sections of sample RED001 by SEM-EDX-BSE, reveals the presence of inclusions of monazite (mean atomic number 37.3). These inclusions are typically of small size, with diameters around 1 to 10 μm, but in several cases, inclusions of higher sizes, up to around 100 μm, are also found. An example of a relatively large monazite grain can be seen in the photomicrograph of Figure 5. Grains

of this size can also be recognised in thin section optical microscopy (Figure 6). Monazite is a light REE, Th, Y phosphate and it is found as an accessory mineral in small amounts in granitic igneous rocks, as it is the case in Pereruela. Therefore, we can attribute the light REE high values observed in the samples analysed to this mineral inclusion. It seems that the largest grains of positively identified monazite are restricted to the red clays from the granite terrain and, therefore, to the pots from potters using those clays (Redondo and Pastor). A careful examination of raw materials used by Riesgo and pottery from both Riesgo and Ramos, reveals that the red clays used by them (metamorphic) and the kaolins do not present either high REE, Th, Y concentrations nor significant fluctuations, and therefore the small inclusions present do not create noticeable variations in concentrations. On the contrary, granitic red clays used by Redondo and Pastor exhibit not only high concentrations of these elements, but also great fluctuations. This contrast is demonstrated in the plot of Figure 7, where the concentrations of two "variable" (La and Th) and two "stable" (Sc and Hf) elements are plotted. It can be seen that while the kaolins (KAO37, KAO38, KAO40) and red clays RCL35, RCL41, RCL44 exhibit stable concentrations of La, Th, Sc and Hf, red clays RCL48, RCL46, RCL47 exhibit much higher and variable concentrations for La and Th. A more detailed study of red clay RCL43, used by Redondo, in terms of different grain size fractions, shows that the highest amounts of light REE, and Th are found in the fraction below 70 μm. More specifically, the fraction between 70 and 20 μm exhibits a peak (Figure 7), which clearly supports the significant influence of the presence of the large monazite grains in the clays. In the same plot a high value of Hf is also observed in the latter fraction. This is attributed to the zircon grains which are also of this size (Figure 5) in which Hf exists as an impurity substituting Zr. This was confirmed by the NAA Zr values which were the highest (414 ppm) in the 70 - 20 μm fraction, while it was 63 ppm, 164 ppm and 203 ppm in the coarse sand, medium sand and clay fractions respectively.

As mentioned above, the casserole RED001, analysed 40 times, gave the most extreme concentrations, the highest and lowest of which are shown in Table 4. If we assume that the difference between both concentrations is solely due to the effect of monazite, we can calculate the composition of this mineral from the difference in these compositions, assuming that it is constant. An empirical formula of the mineral is given in the same table, after taking into account the La, Ce, Sm, Eu, Y, Th concentrations of the affected sample. Taking its specific weight equal to 5, and working on the basis of spherical inclusions of 70 μm diameter, the weight of one inclusion should be about 0.9 μg.

According to this, the differences between the concentrations in Table 4 could be explained by the presence of around 420 grains of that size. If we further assume that density of pottery is 2.5, we would obtain that 420 grains-of-monazite/g-of-pottery represent around 7 inclusions per cm^2. In such a case, the 10 – 25 g of sample is much higher than the minimum amount required for a representative sample, according to Bromund *et al*. (1976).

In this work it has been demonstrated that even the same red clay can produce different compositional patterns because of the great dependence of light REE, Th and Y concentrations on the presence of one single mineral, monazite. The existence of this kind of problem linked to accessory minerals is not new and has been cited before (i.e. Allen *et al*. 1989: 49), but the present case at Pereruela demonstrates the drastic effect it can cause. Here, within the granitic red clays, monazite grains exhibit a wide range of sizes up to around 100 μm. The larger grains, at least, are not normally distributed, and local concentrations of such grains produce a significant increase in absolute concentrations of the trace elements involved in the composition of this mineral phase. However, a minor element as phosphorus should not be affected in such a case. The amount of phosphorus provided by these 420 inclusions discussed above would be around 50 ppm, which is a negligible amount in the bulk composition of pottery where concentration of phosphorus is at least 25 times this value (Table 3).

SUMMARY AND CONCLUSIONS

The results of this ethnoarchaeometric study have implications at many levels of archaeological pottery provenance research and the assumptions that this is based on. Firstly, it has been demonstrated once more that, as far as chemical analysis is concerned, there might be no relation between raw materials and final products (Kilikoglou *et al*. 1988, Cogswell *et al*. 1996). In the case of Pereruela this was exaggerated because of the significant compositional differences between the red clays and the kaolin, which were mixed to produce the paste. This fact demonstrates, to an extent, the effect of human behaviour on paste preparation and points to the only valid axiom in provenance studies, that ceramic groups should be formed not on raw clay materials, but on a paste basis. The relationship between pottery and paste is close, since the former has only been affected by firing and possible alteration and contamination processes during use and burial. By contrast, the relationship between paste and raw materials is less straightforward because paste can vary from the simple clayey

sediment, used without any further preparation, to a very complex mixture of different raw materials.

Moreover, the results have clearly demonstrated how difficult it is sometimes to establish one single reference group, even if the pottery analysed comes from the same production centre (Whitbread *et al*. 1997), belongs to the same typology and was produced within a short chronological period with the same technology. In Pereruela, the cause of this difficulty was the geochemical variability of the red clays in the immediate vicinity of the village. The human parameter here could have been the actual selection of the red clay deposit by a potter, but this was not conscious and was not controlled by any quality or technological criteria. In that sense the observed differences are random and the assumption of the existence of one single reference group for pottery produced in a particular production centre, by potters using the same production technology and local clays is by no means valid here. Indeed, the assumed chemical homogeneity is in fact dependent on the geological environment and the resources exploitation strategies employed in each production centre during a particular chronological period.

The results of the Pereruela study further demonstrate that even one single clay can give significant variation in chemical concentrations of particular elements, if the inclusions primarily responsible for such elemental concentrations are not normally distributed. Therefore, the assumption of normal multivariate distribution, which is the basis for the majority of the statistical procedures employed, becomes questionable.

Of course, all the above observations became clear because we were dealing with a problem ‘with known solution’. In practical terms, however, even if these assumptions are used as a starting point in archaeometric research projects, they should be proved before the final conclusions were drawn. In the case where only archaeological material is involved, it is not possible to know the contribution of the geological variability and the actual distribution of inclusions before the execution of the analytical programme. Moreover, these limitations impose serious restrictions when one is dealing with study cases at consumption centres. In such a case, the interpretation of the several groups of the Pereruela pottery - one for Riesgo, one for Ramos, and one for Redondo and Pastor, which would be really divided into further groups according to light REE, Th and Y concentrations – would not have been immediately interpreted as pottery with common provenance. Because the study was conducted at a production centre, where the whole

pottery production procedure was known, the chemical variability of sherds, which could not fit in the initial assumptions, was further investigated. This was achieved by detailed analyses using a combination of several chemical and mineralogical techniques which, realistically speaking, are rarely applied on a routine basis, due to cost, time and effort. As with the present case of Pereruela, a similar approach could have been followed in the case of an archaeological kiln site when facing a unique type of pottery with an unexpected high variability, which again would enable questioning of the original assumptions. However, at an ordinary consumption site it would be very unlikely that the chemical variability would be investigated in the way it was done for Pereruela or other kiln sites, and the interpretation would be most probably guided by the initial assumptions. An important consequence of this is that, unless provenance studies are routinely conducted on an intensive and well designed analytical basis, the *control groups* formed at kiln sites are much more valid than the ones formed at consumption sites.

Although there are many issues arising from the ethnoarchaeometric work at the pottery-making community of Pereruela, we have focussed the present paper only on the problem of geological variability and distribution of inclusions. Some other issues are related to major elements and petrography, and they will be reported separately. We believe that testing methods and assumptions is a way to strengthen the analytical approach to provenance of archaeological ceramics and a way to build a more solid theoretical framework for the discipline.

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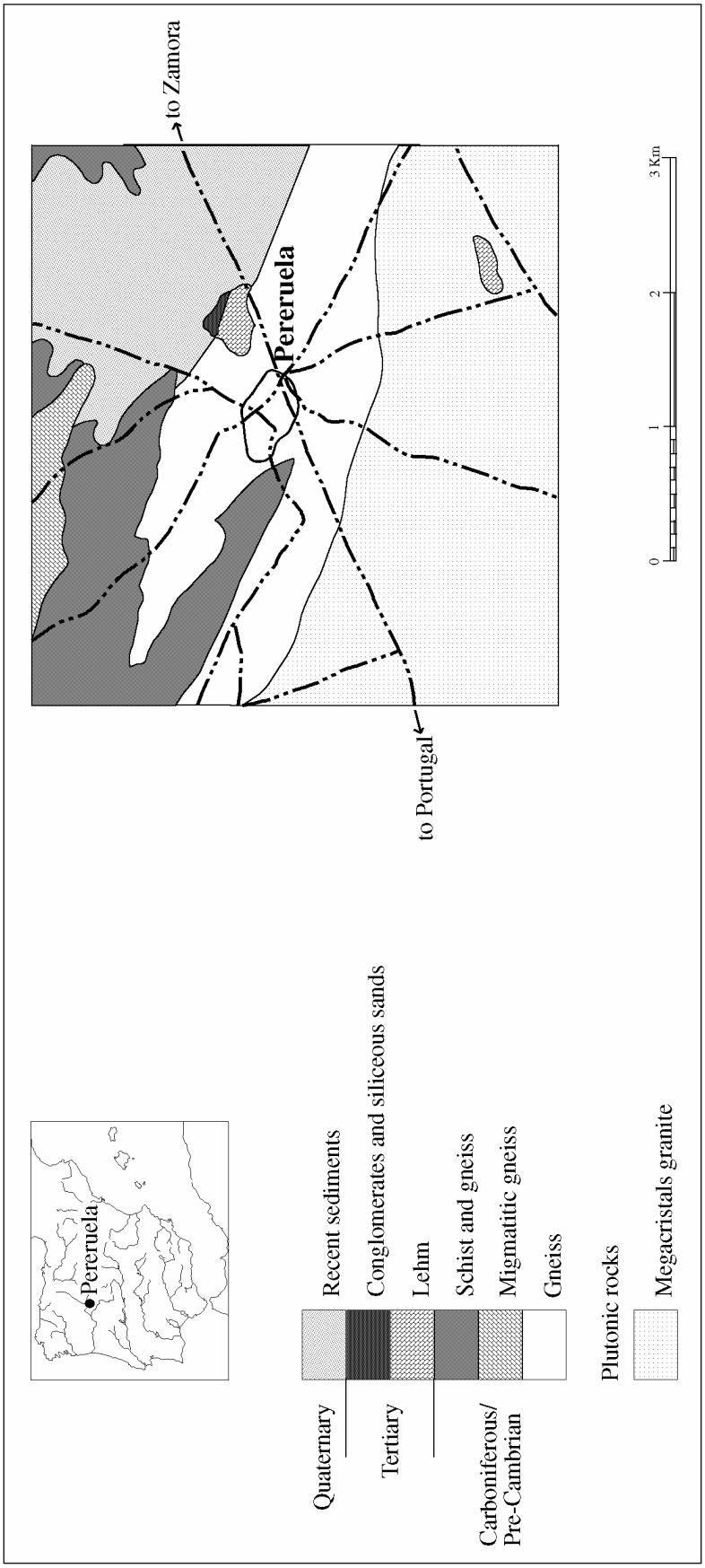

Figure 1. Location of Pereruela in the Iberian Peninsula and geological map of the area under study.

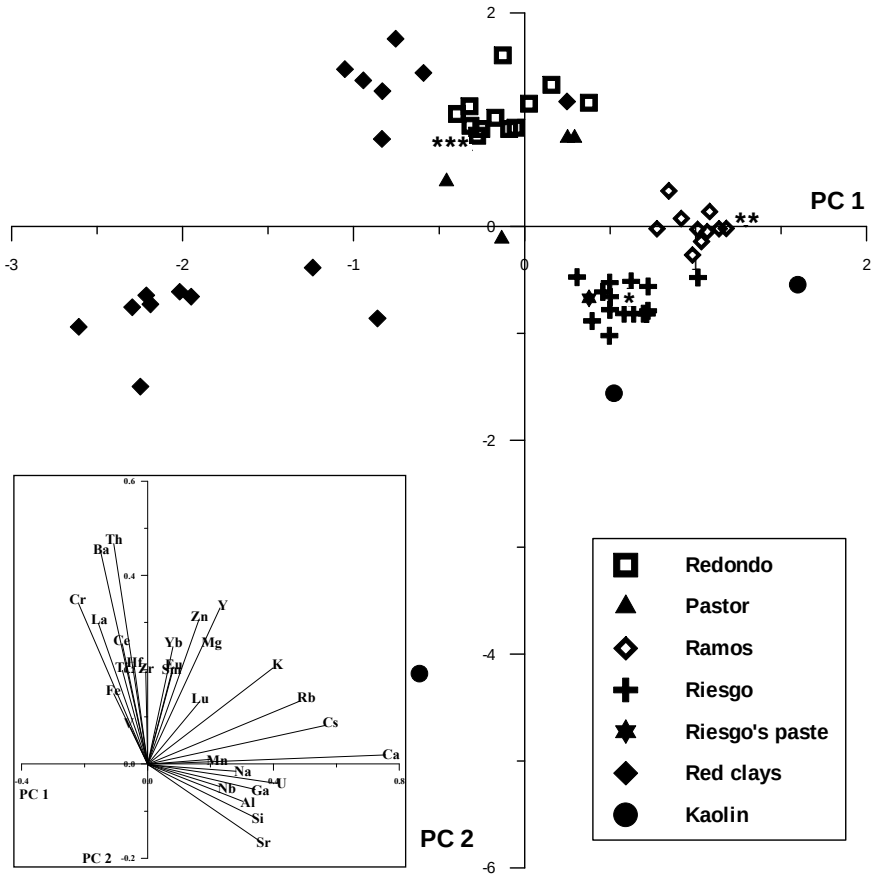

Figure 2. Bivariate plot with the first two principal components (PC1 and PC 2) using the pots, raw materials and paste. Overlappings of different materials are indicated as: * = 1 pot from Ramos and 1 pot from Riesgo; ** = 1 pot from Ramos and 1 kaolin; *** = 1 pot from Pastor and 1 pot from Redondo. On the left bottom corner, plot of the variables, labelled as elements, according to the loadings in the two first principal components.

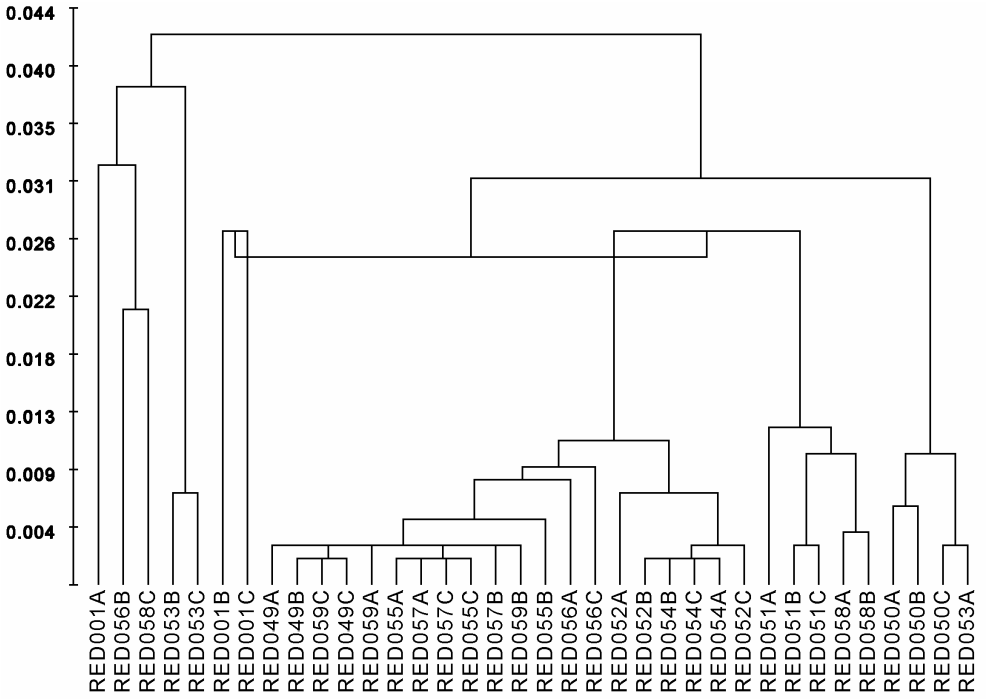

Figure 3. Dendrogram resulting from cluster analysis performed on 3 samples (labelled as A, B and C) from each of the 12 pots from Redondo.

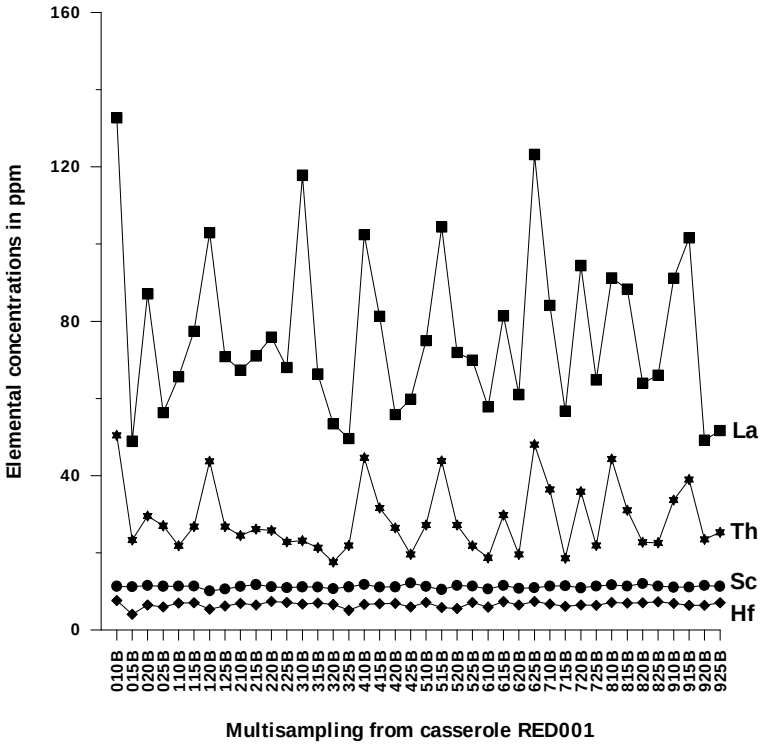

Figure 4. Elemental concentrations of La, Th, Sc and Hf for 40 different samples from Redondo's casserole RED001.

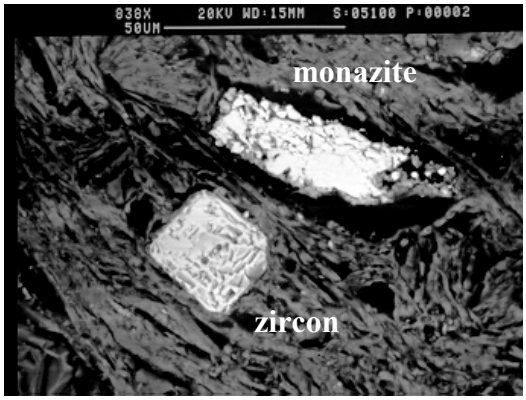

Figure 5. SEM-BS microphotograph of large inclusions of monazite and zircon in Redondo's RED001 casserole at 838X. Bar = 50 μm.

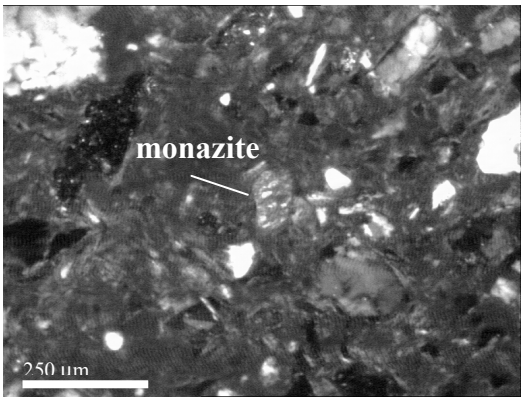

Figure 6. Photomicrograph from thin section analysis in cross polars on red clay RCL043 showing a large crystal of monazite. Bar = 250 µm.

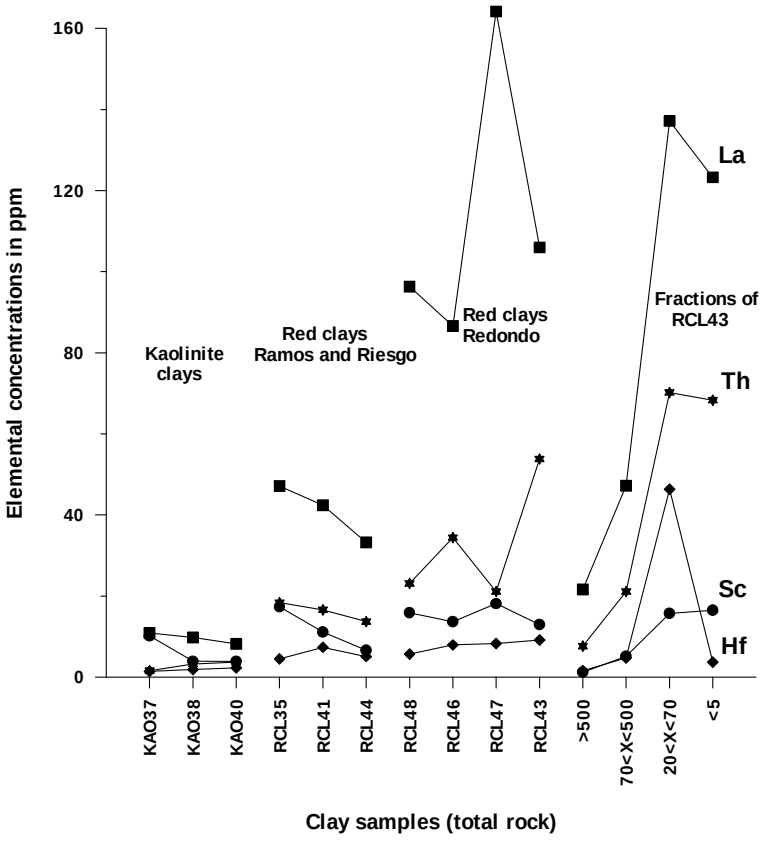

Figure 7. Elemental concentrations of La, Th, Sc and Hf for kaolin, red clays and fractions from red clay RCL043.

Table 1. Individuals included in the study of the production centre of Pereruela, with indication of number of samples from each individual and analytical technique. * = sample used for the analysis of different fractions.

Sample	XRF	XRD	Pet	NAA	SEM	Typology	Workshop
RED001	40	10	10	40	5	small casserole	A. Redondo
RAM002	1	1		1		small casserole	E. Ramos
RAM003	1		1	1		frying dish	E. Ramos
RAM004	1	1		1		chestnuts roaster	E. Ramos
RAM005	1	1		1		elongated casserole	E. Ramos
RAM006	1			1		elongated casserole	E. Ramos
RAM007	1		1	1		frying dish	E. Ramos
RAM008	1			1		casserole	E. Ramos
RAM009	1			1		casserole	E. Ramos
RAM010	1			1		casserole	E. Ramos
RAM011	1			1		elongated casserole	E. Ramos
RAM012	1			1		base	E. Ramos
RAM013	1			1		small casserole	E. Ramos
PAS014	1	1	1	1		base	F. Pastor
PAS015	1	1		1		lid	F. Pastor
PAS016	1			1		casserole?	F. Pastor
PAS017	1			1		casserole?	F. Pastor
PAS018	1		1	1		frying dish?	F. Pastor
RIE019	1			1		small high casserole	R. Riesgo
RIE020	1			1		kiln	R. Riesgo
RIE021	1		1	1		casserole	R. Riesgo
RIE022	1	1		1		casserole	R. Riesgo
RIE023	1			1		kiln	R. Riesgo
RIE024	1	1		1		casserole	R. Riesgo
RIE025	1			1		casserole?	R. Riesgo
RIE026	1			1		small casserole	R. Riesgo
RIE027	1			1		frying dish	R. Riesgo
RIE028	1		1	1		frying dish	R. Riesgo
RIE029	1			1		casserole	R. Riesgo
RIE030	1			1		casserole	R. Riesgo
RIE031	1			1		casserole	R. Riesgo
RIE032	1			1		casserole	R. Riesgo
RIE033	1			1		small casserole	R. Riesgo
RIE034	1			1		small high casserole	R. Riesgo
RCL035	1		2	4		Riesgo red clay	R. Riesgo
PST036	1		1	1		prepared paste	R. Riesgo
KAO037	1			1		kaolin	clay mine
KAO038	1		1	1		kaolin	clay mine
RED039	1			1		small casserole	A. Redondo
KAO040	1		1	1		kaolin	stored clay
RCL041	1			1		red clay	clay mine
KAO042	1			1		kaolin	clay mine
RCL043	5		1	5*		Redondo red clay	clay mine
RCL044	1			1		red clay	fields
RCL045	1			1		red clay	road trench
RCL046	1			1		red clay	L. Pérez
RCL047	5			5		red clay	L. Pérez
RCL048	1			1		red clay	F. Pastor
RED049	3			3		casserole	A. Redondo
RED050	3			3		casserole	A. Redondo
RED051	3			3		casserole	A. Redondo
RED052	3			3		casserole	A. Redondo
RED053	3			3		casserole	A. Redondo
RED054	3			3		casserole	A. Redondo
RED055	3			3		casserole	A. Redondo
RED056	3			3		casserole	A. Redondo
RED057	3			3		casserole	A. Redondo
RED058	3		1	3		casserole	A. Redondo
RED059	3			3		casserole	A. Redondo

Table 2. Total variation values for different data-sets of Pereruela material.

n	Individuals considered	Total variation	Comments
67	Pots, red clays, kaolin and prepared paste	4.3665	All 59 individuals, including multiple sampling for RCL043 and RCL047
46	All pots from the four potters	2.2106	
36	3 samples from all Redondo's pots but 1	0.4748	
40	40 samples from Redondo's pot RED001	0.5982	
40	40 samples from Redondo's pot RED001	0.0654	Removing Sm, Lu, Yb, La, Ce, Th, Eu, Y
40	40 samples from Redondo's pot RED001	0.0948	Calculating the mean value of the previous VM for the removed elements

Table 3. Mean values (m), standard deviations in % (s%) and minimum and maximum (min-max) values for the three main pottery groups established at Pereruela. All mean, minimum and maximum values are in ppm, except for major and minors oxides which are expressed in %.

	Redondo & Pastor (n=18)			**Riesgo (n=16)**			**Ramos (n=12)**		
	m	s %	min-max	m	s %	min-max	m	s %	min-max
Sm	12.06	25.46	7.87-20.69	6.42	10.90	4.78-7.50	7.77	8.49	5.99-8.62
Lu	0.30	10.00	0.24-0.36	0.19	15.79	0.16-0.27	0.28	14.29	0.21-0.34
U	4.0	17.50	3.0-6.1	5.3	13.21	4.5-6.8	5.4	14.81	3.9-6.6
Yb	2.45	15.10	1.77-3.50	1.10	9.09	0.92-1.30	1.75	14.86	1.28-2.18
La	75.6	27.64	41.1-133.9	29.3	6.82	25.5-32.1	30.6	7.52	27.4-35.1
Ce	145.6	23.14	88.9-240.9	70.0	7.43	60.3-78.6	77.2	7.77	67.1-87.2
Th	24.1	33.19	10.7-50.9	7.8	8.97	6.6-9.0	8.1	17.28	6.6-11.9
Cr	44.6	25.34	19.9-56.4	16.4	15.24	11.0-19.7	20.6	12.62	17.0-24.3
Hf	6.19	15.51	3.71-7.69	3.04	13.81	2.31-3.69	2.82	16.31	2.23-3.66
Cs	20.9	11.00	18.5-27.0	13.0	7.85	11.5-15.0	30.4	23.35	12.3-41.1
Sc	10.63	7.52	8.64-11.75	6.96	7.04	6.00-8.15	7.09	6.77	6.44-8.11
Eu	2.08	24.52	0.74-3.15	1.03	20.39	0.41-1.32	1.38	16.67	0.82-1.57
Fe_2O_3 %	4.25	14.59	2.84-5.00	2.57	10.51	2.12-3.01	2.58	13.56	2.19-3.37
Al_2O_3 %	20.33	2.66	19.43-21.01	21.03	3.09	19.31-22.05	20.08	1.34	19.51-20.39
MnO %	0.02	20.00	0.02-0.03	0.01	40.00	0.01-0.02	0.02	15.00	0.01-0.02
P_2O_5 %	0.20	26.02	0.13-0.32	0.15	12.10	0.13-0.20	0.16	8.73	0.13-0.18
TiO_2 %	0.61	9.84	0.50-0.73	0.31	9.68	0.25-0.35	0.31	12.90	0.23-0.39
MgO %	0.99	14.14	0.76-1.19	0.39	5.13	0.35-0.42	0.65	15.38	0.37-0.77
CaO %	1.22	73.77	0.39-4.05	1.81	27.07	0.76-2.49	1.53	26.80	0.88-2.21
Na_2O %	0.18	27.78	0.12-0.27	0.13	15.38	0.10-0.17	0.19	15.79	0.13-0.23
K_2O %	4.62	9.96	3.60-5.37	2.24	6.25	2.03-2.54	2.80	8.93	2.19-3.16
SiO_2 %	67.57	2.15	64.55-70.18	71.40	1.32	69.55-73.98	71.71	1.09	70.39-72.85
Ba	962	15.90	788-1421	217	22.58	86-302	268	20.15	180-335
Rb	215	7.44	189-247	138	8.70	107-154	177	9.04	133-194
Nb	16	6.25	14-17	14	7.14	12-16	14	7.14	13-15
Zr	176	13.07	115-214	94	8.51	75-107	90	11.10	73-106
Y	29	17.24	18-38	12	16.67	8-15	20	15.00	11-23
Sr	209	25.84	158-336	247	16.60	177-341	280	12.86	223-327
Ga	26	7.69	23-30	29	17.24	21-43	27	7.41	24-30
V	63	14.29	44-75	42	7.14	35-50	44	6.82	39-50
Zn	82	10.98	61-95	30	3.33	28-33	47	14.89	28-59

Sample	Ce	La	Y	Th	Sm	Eu	Total
320B	112	53.4	27	18	8.30	1.74	
120B	239	102.9	42	43.7	14.64	3.57	
difference	127	49.5	15	25.7	6.34	1.83	225.37
atomic weight	140.12	138.91	88.906	232.038	150.35	151.96	

Experimental formula of monazite	$(Ce_{0.56}\ La_{0.22}\ Sm_{0.03}\ Eu_{0.01}\ Y_{0.11}\ Th_{0.01})PO_4$

Table 4. Above: elemental concentrations (in ppm) for samples 320B and 120B, from Redondo's casserole RED001, with their absolute differences. Below: atomic weight of the considered elements and formula derived from sample compositions.