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A map of the Monte Arci (Sardinia Island, Western Mediterranean) obsidian primary to secondary sources. Implications for Neolithic provenance studies

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Abstract

Four types of obsidians from the Monte Arci volcanic complex (Sardinia) were used by Neolithic men in the North Tyrrhenian area of the western Mediterranean. A map of their occurrences from primary sources (mother rocks) to distant secondary deposits in the surrounding plains is presented. Some 1457 specimens were macroscopically characterized and in addition ~15% of them fingerprinted from their elemental compositions as determined by electron microprobe, neutron activation analysis or ion beam analysis. The results show that secondary sources, up to now largely neglected in provenance studies of 'archaeological' obsidians will have from now to be taken into account. ***To cite this article: C. Lugliè et al., C. R. Palevol 5 (2006).***

Résumé

Une cartographie des sources primaires et secondaires de l'obsidienne du Monte Arci (Sardaigne, Méditerranée occidentale). Implications pour les études de provenance au Néolithique. Quatre types d'obsidiennes du complexe volcanique du Monte Arci (Sardaigne) ont été utilisées par les néolithiques dans le secteur nord-tyrrhénien de la Méditerranée occidentale. Une carte de leurs gisements naturels, des sources primaires (roches mères) jusqu'à des gîtes secondaires parfois relativement éloignés, dans les plaines qui entourent le massif, est présentée. Plus de mille (1457) spécimens ont été caractérisés optiquement, dont ~15% également par leur composition élémentaire, déterminée par microsonde électronique, activation neutronique ou analyse sous faisceau d'ions. Les résultats montrent qu'il faudra dorénavant, dans les études de provenance d'obsidiennes archéologiques, prendre en compte les sources secondaires, jusqu'ici largement négligées. ***Pour citer cet article : C. Lugliè et al., C. R. Palevol 5 (2006).***

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Introduction

L'obsidienne, présente dans de nombreux sites néolithiques de Méditerranée occidentale, provient de gîtes situés dans les îles Lipari, Palmarola, Pantelleria ou de Sardaigne (voir par exemple [2,27]) (Fig. 1). L'obsidienne sarde, produite au sein du complexe volcanique du Monte Arci, dans le Centre-Ouest de l'île, n'a largement diffusé que dans la partie nord de la mer Tyrrhénienne et sur ses bordures continentales. L'obsidienne du Monte Arci [3] n'affleure pas seulement au sein de ses roches mères rhyolitiques (« sources primaires ») ou à leur voisinage immédiat, par suite de leur désagrégation (« sources sub-primaires »), mais encore à l'état de blocs et de galets, dans les sédiments des plaines environnantes (« sources secondaires ») [1,25]. Neuf types de compositions élémentaires y ont été reconnus, ne différant parfois que par les teneurs en quelques éléments traces [30], correspondant à autant de sources primaires. Toutefois, les obsidiennes de certains d'entre eux se trouvant mélangées dès les gîtes sub-primaires, il suffit, pour les études de provenance d'obsidiennes néolithiques, de ne reconnaître que trois, SA, SB et SC [9], voire quatre, SA, SB1, SB2 et SC [8], de ces types.

L'importance des gîtes secondaires d'obsidienne, peu considérés jusqu'alors, a été récemment soulignée [7,12,13]. Nous avons donc entrepris leur relevé, couplé à l'analyse d'une sélection d'échantillons [11,18,19,24, 26], dont nous présentons ici la première carte géochimique.

Caractérisation de l'obsidienne

L'obsidienne du Monte Arci a été caractérisée visuellement et instrumentalement. La reconnaissance visuelle des types SA, SB1, SB2 et SC a été établie à partir de critères discutés précédemment [15] et renforcée, dans les cas les plus délicats, par des déterminations de composition élémentaire. Pour ce faire, de telles déterminations ont été réalisées sur des échantillons de sources primaires, pour établir un référentiel. Les analyses « ponctuelles » réalisées sur sections polies à la microsonde électronique (10 éléments majeurs et mineurs [11]) et par émission de rayons X sous faisceau d'ions – PIXE – (15 éléments majeurs à traces [10]) distinguent sans ambiguïté les types SA, SB1, SB2 et SC d'obsidiennes. En revanche, les analyses par activation neutronique (33 éléments majeurs à traces) sur des aliquotes de 250 mg de poudres obtenues par broyage ne reconnaissent que trois ensembles, SA, SB (SB1 + SB2) et SC [18,19].

Échantillonnage et résultats

Un secteur d'environ 400 km^2, centré sur le Monte Arci, a été prospecté par l'un d'entre nous (C.L.) et 1430 échantillons d'obsidienne de localisation précisément obtenue par repérage GPS, collectés. Alors que les sources primaires (d'altitudes comprises entre 120 et 720 m) et sub-primaires sont toutes situées à l'intérieur du massif, les sources secondaires sont essentiellement distribuées dans les plaines environnantes plio-quaternaires. Les obsidiennes y affleurent en surface de dépôts d'origine colluviale à alluviale, souvent remaniés, ou sur les berges des rios Mannu et Mogoro. Des déterminations de composition élémentaire ont été effectuées sur 217 échantillons, dont 190 positionnés par GPS. Elles ont concerné principalement les obsidiennes de type SB1, SB2 et SC, dont les gîtes sont plus étendus que ceux des obsidiennes SA. En ce qui concerne les obsidiennes qualifiées de SB par l'analyse en activation neutronique, leur subdivision en SB1 et SB2, pour celles des sources secondaires, a été rendue possible par leurs apparences visuelles très caractéristiques [15]. Le résultat global des caractérisations visuelles et instrumentales est présenté dans la Fig. 2 sous la forme d'une carte géochimique. On y observe que seules les obsidiennes de type SB2 et SC débordent largement du massif. Vers le sud, le champ d'épandage de ces dernières est nettement séparé de celui, très réduit du fait de la géomorphologie locale, des obsidiennes SA. À l'ouest, il ne semble pas rejoindre celui des obsidiennes SB2. Les obsidiennes SB2 et SC, bien que présentant en sources secondaires des cortex très comparables, diffèrent nettement par leurs caractéristiques visuelles (inclusions minérales, transparence, etc.). Il en est de même entre obsidiennes SA et SC, les premières marquées, en outre, par une absence de cortex.

Par ailleurs, chaque type de composition élémentaire est très homogène (Le Bourdonnec et al., en préparation). Si l'on considère que l'échantillonnage a concerné des obsidiennes mises au jour aussi bien récemment par l'érosion (dans les sources primaires) qu'à différents moments (depuis la formation de ces obsidiennes, il s'est passé plus de 3 Ma), cela signifie que les roches mères présentaient une composition très homogène, une information importante pour les études de provenance des obsidiennes néolithiques.

Conséquences pour les études de provenance et conclusion

Bien que la présence d'obsidienne dans les plaines proches du Monte Arci ait été signalée depuis longtemps (voir par exemple [16,25]), aucune étude n'avait été consacrée spécifiquement à ces « sources » potentielles. Nous montrons ici que, pour les types SB2 et SC, elles sont en réalité très étendues. De plus, les obsidiennes qu'elles renferment présentent des dimensions pluricentimétriques, les rendant aptes à la production d'artefacts. Enfin, la « densité » (nombre de blocs ou de galets d'obsidienne par mètre carré) est apparue, en bien des lieux, comme suffisante pour qu'elles puissent avoir contribué significativement comme sources de matière première. Cependant, l'accessibilité de ces sources pour l'homme ancien, en raison du couvert végétal, pourrait avoir été moindre qu'aujourd'hui. Il n'en reste pas moins, pour les obsidiennes SC, que les rives du système rio Mannu–rio Mogoro devaient offrir de larges possibilités d'échantillonnage d'obsidiennes.

La confrontation entre ces données et les données archéologiques sardes atteste l'utilisation de sources secondaires du Monte Arci, pour les sites de la « zone d'approvisionnement directe » (*supply zone* des auteurs) au Néolithique ancien et moyen, comme le montrent les cortex résiduels de certains artefacts façonnés à partir d'obsidiennes SB2 ou SC. Il s'agissait alors d'obtenir une industrie au caractère microlithique très marqué, obtenue à partir d'éclats détachés par percussion. À la fin du Néolithique moyen et au cours du Néolithique tardif (fin du V^{e}–IV^{e} millénaire avant notre ère), un changement dans les stratégies de réduction de l'obsidienne s'opère, plus orienté vers la production de lames. On observe aussi une augmentation importante de la production, basée sur l'exploitation dominante de sources primaires à sub-primaires. De vastes ateliers de taille apparaissent à proximité des roches mères des obsidiennes de type SA et SC. C'est aussi l'époque où une plus grande diversification de leur circulation dans la zone tyrrhénienne s'instaure. Les sources secondaires ne semblent plus employées qu'occasionnellement pour un usage local.

Il est devenu d'usage de relever les proportions relatives d'obsidiennes de type SA, SB1, SB2 et SC dans les séries néolithiques. Les résultats présentés ici montrent que l'appartenance à un type géochimique ne permet pas de préciser l'origine géographique d'une obsidienne taillée. C'est la combinaison caractérisation par la composition élémentaire/observation visuelle, avec notamment la présence ou non de cortex résiduel, qui pourra éventuellement permettre de remonter au type de source utilisée. Il est ainsi clair que les proportions relatives des types d'obsidienne dans un site ou un niveau culturel n'auront pas la même signification au Néolithique ancien et moyen, d'une part, et au Néolithique tardif, d'autre part.

Enfin, si les caractéristiques optiques suffisent, en pratique, à déterminer le type d'une obsidienne prise dans un gîte naturel associé au Monte Arci, il n'en est plus de même pour les obsidiennes taillées, parce qu'il peut s'agir, soit de petits objets à l'allure pétrographique apparente peu caractéristique, soit d'objets plus volumineux, mais opaques, et qu'il est peu judicieux, voire inadmissible, de fragmenter afin d'obtenir de meilleures conditions d'observation. Ainsi, des examens menés à la fois visuellement et instrumentalement ont montré que la part de succès dans l'attribution d'un type à une obsidienne sarde pouvait n'atteindre que 65–69% [28,29], voire au mieux 85% [15]. La première esquisse de banque de données sur les compositions élémentaires des obsidiennes du Monte Arci, constituée ici pour réaliser une cartographie de leurs gîtes, a déjà puissamment contribué à renforcer la justesse d'études de provenance portant sur de grands nombres d'obsidiennes dans des sites néolithiques sardes [14,15].

1. Introduction

Obsidian artefacts are often present in Neolithic sites of the Tyrrhenian Islands (western Mediterranean) and of surrounding continental regions, from eastern Algeria to Italy and southern France. Provenance studies based on fission track dating and/or elemental analyses have shown that the raw materials of this industry were exclusively coming from the four Italian islands of Lipari, Palmarola, Pantelleria and Sardinia (see, e.g., [2, 27]) (Fig. 1). Whereas obsidians from the first three islands are found, albeit by times in quite small numbers, in about all this area, the diffusion of Sardinian obsidians seem to have been restricted to nearby Tyr-

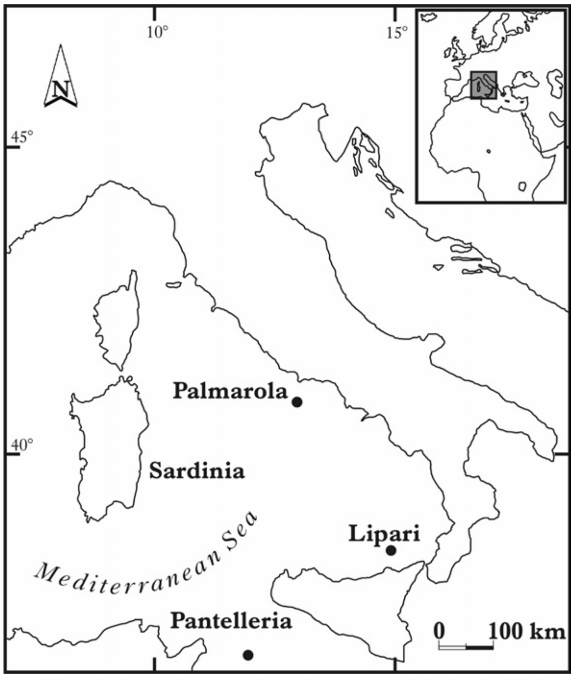

Fig. 1. Schematic map of western Mediterranean showing the obsidian source-islands.

Fig. 1. Carte schématique de la Méditerranée occidentale montrant les îles-sources d'obsidienne.

rhenian Islands and from northwestern Italy to southwestern France.

Sardinian obsidians were formed some 3.2 Ma ago in the Plio-Pleistocene Monte Arci volcanic complex dominating the coastal plain to the East of the Oristano Gulf [3]. As a consequence of weathering, erosion and natural transports, obsidian occurs not only in these 'primary sources', but also in their immediate proximity, in 'sub-primary' sources, as more or less angular down-warped blocks and cobbles [1]. They are also found in more distant 'secondary sources', as rounded pebbles along the river beds and in the plains sediments surrounding the southern half of the massif [25]. Nine elemental types of composition were identified among the Monte Arci obsidians, corresponding to as many primary sources [30]. However, as obsidians of several of these types, only discernible from some trace element contents, are found together from easily accessible sub-primary sources, for provenance studies only three, SA, SB, SC [9] to four obsidian types, resulting from the partition of SB into SB1 and SB2 types, have to be distinguished [8].

The importance of distant secondary sources in Neolithic man raw material procurement strategies in Sardinia was recently pointed out [7,12,13]. Hence the need, for Neolithic obsidian provenance studies, to know the geochemical fingerprinting of these sources [11,18,19, 24], their geographical extensions and their potential 'productivities'. We give here the status of an ongoing program aimed at a better knowledge of the Monte Arci-related obsidians (see also [26]) and the first detailed map of their field occurrences.

2. Obsidian characterization methods

All the obsidians sampled were first visually characterized. In most cases a type (SA, SB1, SB2 or SC) attribution could be obtained directly from visual examination with the naked eye and/or with the help of a stereomicroscope (100 ×). Our visual criteria for obsidian-type identification are discussed elsewhere [15]. Ambiguous cases were resolved by instrumental determination of the elemental composition, which in turn contributed to refine the visual criteria [15].

A majority of samples were analysed by instrumental neutron activation analysis (INAA); the elemental compositions were performed using the standard techniques adopted in the Radiochemistry Laboratory of the Department of General Chemistry of the University of Pavia [20,21]. The obsidian samples were ground to a fine powder in an agate mortar, of which aliquots of 250 mg were sealed in polyethylene vials and irradiated for 12 h in the Triga Mark II reactor of the University of Pavia with a nominal thermal flux of 1×10^{12} n cm^{-2} s^{-1}. The contents of elements Na, K, Ca, Sc, Cr, Fe, Ni, Co, Zn, Ga, As, Se, Rb, Sr, Zr, Sb, Cs, Ba, La, Ce, Nd, Sm, Eu, Gd, Tb, Ho, Tm, Yb, Lu, Hf, Ta, Th and U were determined by γ-ray spectrometry using a Ge HP detector connected to a multichannel pulse height analyser and to a personal computer. Counting was started after 3 h of decay time to determine radionuclides with half-lives less than one day and was repeated after three and 15 days to determine the other elements. Data reduction was carried out using a personal computer with software for spectral analysis. A geological standard from the National Institute of Standards and Technology (obsidian rock NIST-SRM 278) and a nitric solution of the analysed elements were irradiated together with the samples as reference standards. Accuracy was evaluated by comparing determined and certified abundances for the NIST-SRM 278 standard [20].

Although the Ba content is in itself a good discriminator of the SA, SB and SC obsidian types of Hallam et al. [9], most elements do contribute to this separation, as shown by multivariate analysis, and more clearly even by the rare-earth elements contents and patterns [18,19]. NAA does not allow one to partition the SB obsidians into SB1 and SB2 types [18].

A second group of samples were analysed from polished sections with the SX50 (CAMECA) electron microprobe (EMP) of IFREMER (Brest, France). Analyses were performed on polished sections with a 20-nA and 15-keV defocused (5-μm diameter) electron beam. To insure internal consistency between analytical sessions, the Monte Arci SA type sample SA-66 was repeatedly measured. The element Na, Mg, Al, Si, P, K, C, Ti, Mn and Fe contents were computed with the PAP software [22] as 100% oxides. Concentration in Al, K, Ca and Ti allows one to easily separate the SA, SB1, SB2 and SC obsidian types. A discriminant analysis derived from the Na, Al, Si, K, Ca, Ti and Fe contents highlights this separation [11].

Finally, several obsidians were treated also in the form of polished sections by ion-beam analysis. Analyses by particle induced X-ray emission (PIXE) were carried out with the AGLAE facility (external beam) of the 'Centre de recherche et de restauration des musées de France' (C2RMF, Paris, France). As two Si(Li) detectors were available, light elements Na, Al, Si, K, Ca, Ti, Mn and Fe and heavy elements Zn, Ga, Rb, Sr, Y, Zr and Nb were determined simultaneously with a 3-MeV proton beam [4,5]. The elemental quantification of the detected elements was realized with the 2000 version of the GUPIX software [6,17]. The SA, SB1, SB2 and SC obsidian types can be singled out, among other elements, from their Ti, Zn, Sr, Y, Zr and Nb contents [10,23].

3. Sampling and results

An area of some 400 km^2 centred on the Monte Arci was systematically prospected and sampled by one of us (C.L.). The localisations of the 1430 obsidians collected during this survey were systematically GPS-recorded. Whereas primary sources, at altitudes between 120 and 720 m a.s.l. and sub-primary sources are all located in the Monte Arci massif s.s., the secondary sources are almost entirely situated in the Plio-Quaternary sediments of the surrounding plains. Obsidian pebbles often outcrop on the surface of these more or less reworked colluvial to alluvial deposits. The secondary source samples collected were taken either in such terrains or along river (Rio Mogoro, Rio Mannu) banks. Internal reference values for the SA, SB1, SB2 and SC types of elemental compositions were obtained from samples taken in primary to sub-primary sources. In all, 217 samples were analysed, of which 190 with GPS-recorded localisations. As can be seen in Table 1, special attention was given to SB and SC obsidians. This is due to the larger extension of their secondary sources as compared to that of SA obsidians (see below). For obsidians from secondary sources, the agreement between optical and instrumental obsidian type attributions was excellent. The two SB types, not singled from each other by NAA, were easily sorted from their visual characteristics (colour, transparency, mineral inclusions, etc [15]). The overall result of our Monte Arci obsidian characterizations is summarized in the form of the geochemical map presented in Fig. 2. The contours of the four greyed areas are those of the field-recorded distribution of obsidian occurrences. The points reported give the localisation of the samples whose geochemical type was determined strictly instrumentally (EMP, NAA, PIXE) or by using a combined visual/instrumental (NAA) characterization (for SB obsidians). The largest source area of a given type is by far that of SC obsidians. It extends from the primary sources east of the Monte Arci to the plains surrounding the eastern, southern and southwestern parts of the massif; while in primary sources the SC obsidians may be coated by a thin patina, in secondary sources, where they are more or less rounded, they present a well-developed cortex that testifies at different degrees to their more or less complex transportation/sedimentary history. In the South of Monte Arci, there is a gap between the SC spreading field and the SA obsidians. The latter are situated in a low topographic position and accordingly are distributed only in the immediate vicinity of their primary source. In any case, they are easily discriminated by the naked eye from that of the SC type. The SC obsidians are still convenient for knapping purposes 5 km from the Gulf

Table 1
Samples characterized by instrumental methods
Tableau 1
Échantillons caractérisés par méthodes instrumentales

Method	*n*	SA	SB1	SB2	SC	Reference
NAA	110	11	34[a,b]	18[a,b]	47	[19] and this work
EMP	67	12	10	7	38	[11] and this work
PIXE	13	1	0	1	11	This work
	27	9	6	7	5	[15,23][c,d]

n, number of samples analysed / nombre d'échantillons analysés.

[a] SB1 and SB2 types sorted visually from the type determined by NAA as SB (see text).

[b] Types SB1 et SB2 différenciés visuellement à partir du type déterminé par NAA comme SB (voir texte).

[c] Samples without GPS positioning, not reported in the map of Fig. 2.

[d] Échantillons non référencés par GPS, non reportés sur la carte de la Fig. 2.

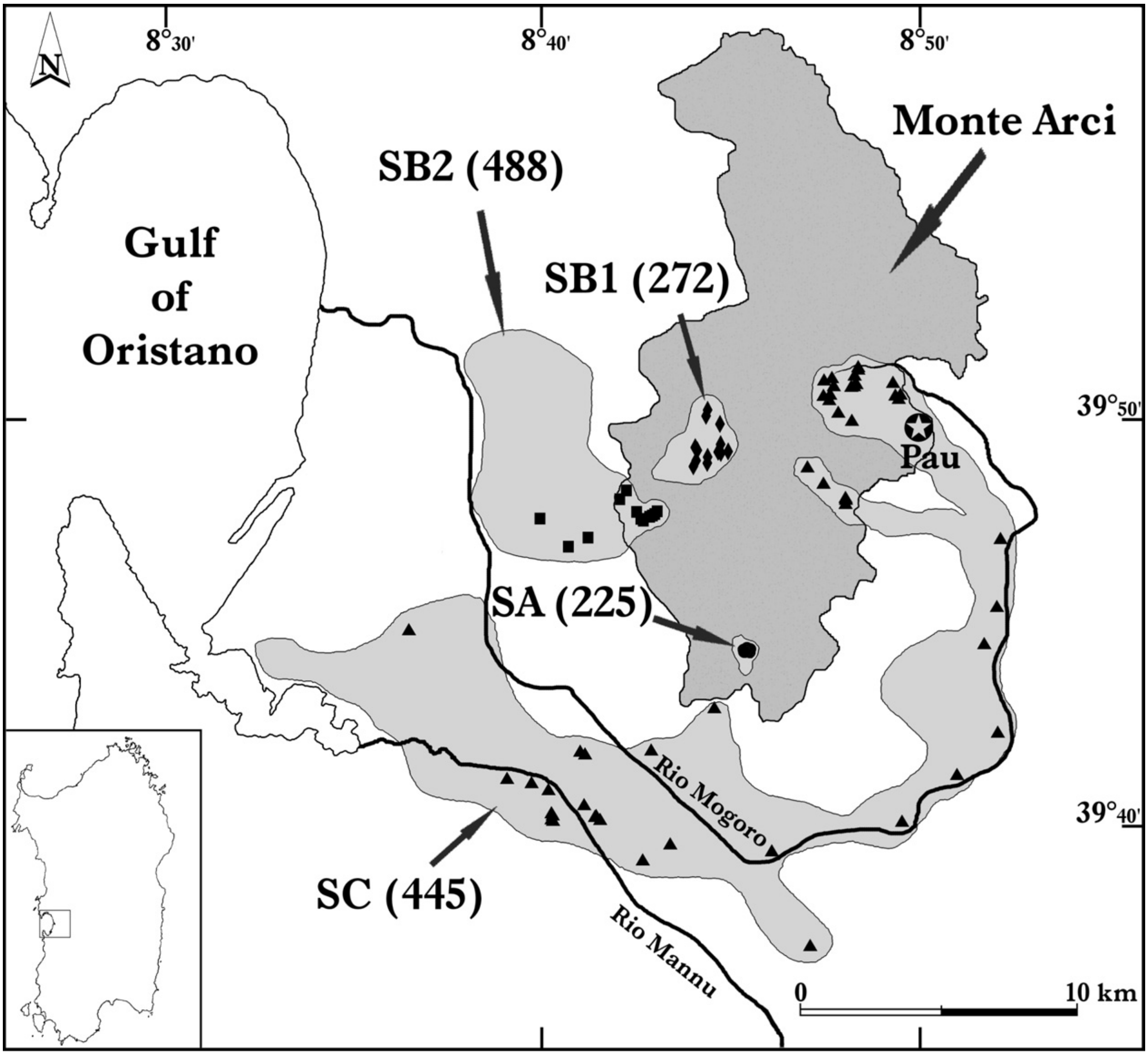

Fig. 2. Geochemical map showing the extension of SA, SB1, SB2 and SC obsidian sources. All primary and sub-primary sources are localised inside the Monte Arci massif. Between parentheses, the number of samples characterized.
Fig. 2. Carte géochimique montrant l'extension des sources d'obsidiennes de types SA, SB1, SB2 et SC. Les sources primaires et sub-primaires des obsidiennes sont toutes situées à l'intérieur du massif du Monte Arci. Entre parenthèses, le nombre d'échantillons caractérisés.

of Oristano shoreline. On the western side of the Monte Arci, although the SB1 and SB2 primary sources are localised not far from each other; the local morphology seems to have prevented their mixing in secondary sources. While SB1 obsidians are found only at proximity of its mother rock, SB2 obsidians are found up to several kilometres from its sources in the coastal plain, where however their field of occurrence does not seem to overlap that of the SC obsidians.

As discussed elsewhere, each one of the instrumentally determined type of elemental composition is very homogenous (Le Bourdonnec, et al. in preparation). Considering that obsidians from primary sources may have been denuded by erosion at different times since 3.2 Ma, this shows that the corresponding mother rock compositions are also quite homogeneous, an important point for further regional Neolithic obsidian provenance studies based on elemental composition.

4. Consequences for provenance studies and conclusion

The presence of scattered obsidians in the Campidano Plain, south of Monte Arci, is known since long (see, e.g., [16,25]). However, the importance of this and of the other plains surrounding the massif as potential obsidian secondary sources had not been taken into consideration. The map in Fig. 2 suggests that for SB2 and especially SC obsidians, these plains may have had a significant role in procurement strategies, especially as they present pebbles of convenient sizes for making them valuable raw materials. Thus SB2 obsidians lay on the ground surface in the form of rounded to well-rounded pebbles of the order of 1.5 kg at 1.5 km from their primary source, down to 0.5 kg at 2.5 km. At greater distances SB2 obsidians can be found in smaller subprismatic, from subangular to subrounded blocks. Small cm-sized samples are still workable 6 km from the primary source. The observed frequency of SB2 secondary deposits is actually strongly biased (lowered) by modern land reclamations. Nevertheless, good concentrations of excellent pebbles can still be easily found in the plain, some 4 km from their mother rocks. The SC obsidian secondary deposits, still workable 5 km from the Gulf of Oristano shoreline, increase notably in density (up to about one piece per square meter) at a distance of ~15 km inland. The SA and SB1 'secondary deposits' are more concentrated near to the primary sources and therefore are less suspect of having influenced Neolithic sourcing strategies. Testifying of the short distances of secondary sources from the primary ones, their obsidian cobble surfaces are not much weathered, which makes them often not easily distinguishable from post-depositional patinas on artefacts.

Another question is that of the suitability to Neolithic man of these secondary obsidian resources. Few data are actually available for a detailed reconstruction of the landscape and vegetation of the Monte Arci area during these times: the most general and commonly accepted picture is that of a land entirely covered by forests and vegetation, which might have hidden the obsidian secondary sources in the plains, especially to the West and the South of the massif. Some pedostratigraphic data referring to the 6th millennium BC demonstrated the presence of a Mediterranean climate with biostatic conditions developed on sandsoils belonging to stage 2. It could thus be argued that, at least along riverbeds of the Rio Mannu–Rio Mogoro system, some superficial erosion might have brought to light obsidian-bearing alluvial deposits, offering to Early Neolithic man their first contacts with this raw material. Further agricultural advances might have enhanced the erosion and visibility of the ground, giving to Neolithic communities a more detailed outline and awareness of obsidian secondary deposits and of related outcrops as well.

Several lines of evidence, notably the eventual presence of cortex remnants on artefacts, show that Early to Middle Neolithic man of the so-called 'supply zone', up to 30 km far from Monte Arci, used these secondary sources. The shape and size of their cobbles were in effect convenient to produce a toolkit exhibiting a strongly microlithic character. This small-size industry is generally and almost exclusively obtained from flakes detached with direct percussion reduction techniques and counts for the largest part of the archaeological assemblages. At the end of Middle Neolithic and during the Late Neolithic (late 5th–4th millennium BC), one observes both a shift in the obsidian reduction strategies, which became more oriented towards blade production and a high increase in the production rate. The secondary deposits seem then to be only scarcely exploited, for local and expedient production essentially, whilst large obsidian workshops appear directly close to the lavas outcrops, chiefly of the SA and SC types. This new acquiring strategies and scales of production are reflected in the coeval increase and more diversified circulation of SC obsidian from these workshops to the whole northward Tyrrhenian area.

In the western Mediterranean region, it has become usual, since the late 1970s, to note the percent relative abundances of SA, SB (or SB1, SB2) and SC Monte Arci obsidians in provenance studies. The belonging of a group of archaeological obsidians to a given geochemical type was in practice equated to their common provenance of the same 'source'. The results presented here show that the relative abundances of SA, SB1, SB2 and SC obsidians in a Neolithic series cannot be interpreted in the same way in the Early/Middle Neolithic and in the Late Neolithic. It also shows the need to closely associate technological and 'sourcing' studies on the same archaeological pieces.

Finally, we would point out the usefulness of instrumental type determinations of archaeological artefacts. We observed here a one-to-one agreement between visual and instrumental determination of obsidian types SA, SB1, SB2 (or SB1+SB2 for NAA) and SC. This was possible because the only geological samples collected were those of a size large enough to be potentially used for the production of at least a microlithic industry. Moreover, these samples could be broken, to best reveal to the naked eye its mineral inclusions, fresh fractures brightness, transparency, etc. Neolithic artefacts may be too small or too thick to allow the observer a good determination. Combined visual and instrumental determinations have shown that, depending on the series, from about 15% [15] to 31–35% [28,29] of the visual determinations can be erroneous. Thus instrumental determinations will keep essential in any future 'provenance' study. The data bank accumulated for the making of the map presented here strongly contributed to recent provenance studies [14,15]. We expect from its increase and from the related enrichment of visual criteria in progress in our laboratories some refinements in future provenance studies.

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