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Multiple origins of Bondi Cave and Ortvale Klde (NW Georgia) obsidians and human mobility in Transcaucasia during the Middle and Upper Palaeolithic

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ABSTRACT

Using PIXE four types of elemental compositions were found among obsidian artefacts from the Bondi Cave and Ortvale Klde, Middle to Upper Palaeolithic sites in NW Georgia. One of those types corresponds to obsidians from the Chikiani source, whose compositions were determined with a very good agreement by PIXE and ICP-AES/MS. The composition of Chikiani obsidians is remarkably constant despite K—Ar and ³⁹Ar/⁴⁰Ar extrusion ages from ca 2.4 and 2.8 Ma. The compositions of two other groups of obsidian artefacts are similar to source materials from eastern Anatolia and Armenia, in particular Ikisdere, Sarikamis, Gutansar, and Hatis. Obsidian is only a minority component in the lithic assemblages at the Bondi Cave and Ortvale Klde. Both Neanderthal and Modern Human populations used obsidian in particular from Chikiani. Considering that the shortest walking distance to this nearest source is at minimum of about 180 km, and to other potential sources of more than 350 km it is suggested that this material reached these two sites mostly, if not exclusively, by a series of 'down the line' exchanges.

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1. Introduction

Obsidian was one of the preferred raw materials of prehistoric lithic industries, used from at least 1.8 Ma (Piperno et al., 2009). Along the 'obsidian belt' that extends from western Mediterranean to Transcaucasia (Blackman et al., 1998, figure 1; Poidevin, 1998, figure 1; Yellin, 1995), this raw material's use is documented regularly in archaeological contexts from 9 to 13 ka in western and eastern Mediterranean respectively (Carter, 2009; Lugliè, 2009) and

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since 14 ka in the Near and Middle East (Cauvin and Chataigner, 1998). In the latter region there is an even early occurrence, as represented by the Middle Palaeolithic site of Shanidar in northern Iraq, dated at 28 ka (Chataigner, 1998; Renfrew et al., 1966). Further to the north in southern Transcaucasia, where the archaeological record started 1.8 Ma ago, the oldest obsidian tools are found in undated open air Acheulean sites and in small quantities in Middle Palaeolithic (MP) sites located in the Lesser Caucasus range (Adler, 2002; Pleurdeau et al., 2007). Over the past 50 years, provenance studies in the Mediterranean basin have focused primarily on the use of obsidian during the Epipalaeolithic to post-Chalcolithic periods, with little investigation of the very early material.

In northern Transcaucasia, many archaeological sites attest the continuous presence of humans from the Middle Pleistocene. The archaeological record displays various types of MP Neanderthal settlements, especially in the near-mountainous environments of the Greater Caucasus. The extinction of Neanderthal occurred over a relatively short period of time when modern human, coming from the south, colonized the area (Adler et al., 2008; Bar-Yosef et al., 2006). Obsidian, while sometimes found in MP horizons, was more

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frequently used during the Upper Palaeolithic (UP). As no obsidian source is known in northern Transcaucasia, the material always has to represent a non-local import. The presence of cores in UP levels attest to some of this obsidian being worked on-site.

In this contribution we present the first Palaeolithic obsidian provenance study in northern Transcaucasia, for the two sites of Bondi Cave and Ortvale Klde, excavated since 2006 by a Georgian-French team. Both sites, whose stratigraphies include MP and UP horizons, are located in the Imereti region of NW Georgia. In both sites, the obsidian *chaînes opératoires* are incomplete, which suggests a procurement of at least part-manufactured artefacts. The goals of this work were to determine the provenance of these obsidians, of which the two nearest sources lie at linear distances of more than 100 km (Fig. 1) and to initiate a comparison between the behaviour of Neanderthal and *Homo sapiens* with regard to the procurement and use of this resource.

2. Archaeological background and sampling

2.1. Bondi Cave

The Bondi Cave is located north of the town of Chiatura at an altitude of 477 m asl. The cave, 30 m above the Tabagrebi River valley, opens towards the south (Fig. 2). Twelve squares were dug below the cave porch during the 2007 to 2010 field seasons. The excavation revealed a sedimentary sequence more than 2.8 m thick into which eight very distinct archaeological horizons are interbedded between several levels corresponding to major collapses of the cave roof (Table 1). The bottom of the excavation abuts on a level of dismantled limestone blocks, but the true limestone bedrock has yet to be reached (Moncel et al., 2007, 2008).

Bones and lithic artefacts are present in all archaeological layers. They are especially abundant in the UP (levels VI to I) horizons, deposited less than 30 ka ago (Table 1). Some ornaments with herringbone designs were found in layer III, one human tooth, assigned to *Homo* sp. in layer V and a small pierced disc of cockle-shell 3 mm wide and 1 mm thick (Tushabramishvili et al., 2012). The lithic industry of layers VII and VIII, dated between 40 and 43 ka have MP affinities. The Bondi Cave fauna is typical of that found in most Georgian sites of the region, and in particular to those attributed to the MP and/or the Early UP (Ortvale Klde, Dzudzuana) (Adler et al., 2006; Bar-Oz and Adler, 2005; Bar-Oz et al., 2002, 2004, 2008; Bar-Yosef et al., 2006; Meshveliani et al., 2004; Tushabramishvili et al., 2007). The UP lithic assemblage, almost exclusively made of various local flints (98.8%), is mainly composed of blades and bladelets. Laminar cores testify in situ production. Obsidian was only found in levels II, IV and V, where they represent $\approx 1\%$ of the lithic material. Sixty-one obsidian artefacts were found during 2007–2008. They are mainly comprised of unretouched pieces (50), including blades, bladelets and small flakes. Only 11 tools were found, with scrapers, burins, burin spalls, retouched blades. No cores were found in this recently excavated material; however the small sizes of some flakes suggest a certain amount of punctual resharpening. The technological characteristics of the obsidian are identical to those of the flint. Four pieces (bladelets and flakes) were obtained for a nondestructive sourcing study; they come from levels II (n = 2), IV (n = 1)and V (n = 1) (Fig. 3).

2.2. Ortvale Klde

The Ortvale Klde site, near to the city of Chiatura, is composed of two ('north' and 'south') chambers open to the East; it is located at an altitude of 530 m asl, some 35 m above the Cherula river (Fig. 4). The first excavations in this cave were undertaken by David Tushabramishvili from 1973 to 1992. Subsequent work, by a Georgian team (1992–1995), then by a Georgian-American team (Adler, 2002; Adler et al., 2006; Adler and Tushabramishvili, 2004) mainly focused on the MP/UP transition in the south chamber. The same team showed that the subsistence behaviours of Neanderthals and Modern Humans were largely identical (Adler et al., 2006; Bar-Oz and Adler, 2005; Bar-Oz et al., 2002). Several dates, by ¹⁴C, thermoluminescence and electron spin resonance documented the stratigraphic depositional history, fixing the MP/UP transition at 34 ± 1 ka (Adler et al., 2008).

The north chamber, 15 m wide, was first explored in the 1970s by two test pits dug into the slope outside the cave; they revealed MP



Fig. 1. Simplified map showing the localization of Bondi cave and Ortvale klde, and some obsidian sources.

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Fig. 2. Synthetic stratigraphical section (E-W, transversal) at Bondi Cave, with location of obsidian sampling.

horizons (Tushabramishvili, 1994; Tushabramishvili et al., 1999). In 2006, we opened four new test pits (Moncel et al., 2007). Three of those, similarly dug on the slope outside the porch, again reached MP levels while the fourth one, located below the porch, unearthed UP levels (Table 2). In the absence of radiometric dating, any correlation between the cultural levels of the two chambers would be hazardous. The animal species most represented are *Capra caucasica* and *Bison priscus*. The lithic industry was principally knapped from local flints of a very good quality. It includes mainly unilateral and bilateral unifacial points, close to the Djrujula MP tradition (Meignen and Tushabramishvili, 2006; Pleurdeau et al., 2007). In the UP levels, blade-bladelet cores and retouched tools are frequent.

In the 2006 excavations nineteen obsidian artefacts were found, of which nine were associated with the MP horizons and ten with the UP strata. This material comprised flakes, sometimes of very small size (largest dimension <15 mm) and bladelets (Fig. 3). Seven of these pieces were selected for a provenance study, five from the MP level and two from the UP level.

3. Regional obsidian sources

The immediate vicinity of the Bondi Cave and Ortvale Klde is devoid of obsidian sources. The nearest ones, the Baksan and Chikiani sources, lie at linear distances of about 120 km to the NNW

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Stratigraphical data on lithic artefacts at Bondi Cave (excavations 2006–2007) and obsidian sampling.	

Layer	Affinity	Number of	lithic artefacts		14 C AMS ages, ka (\pm	2σ) ^b	Samples
		Flint	Obsidian ^a	Total	Ages BP	Ages cal BP	
Ι	UP	332	0	332			
II	UP	1452	18 (2)	1470			LII-B3, LII-C3
III	UP	70	0	70	$14{,}330\pm90$	17,830-17,190	
					$14{,}050\pm90$	17,490-16,970	
IV	UP	2363	26(1)	2389	$19,360 \pm 120$	23,500-22,980	LIV-B4
					$\textbf{20,080} \pm \textbf{170}$	24,530-23,530	
					$\textbf{26,020} \pm \textbf{170}$	31,540-30,340	
V	UP	2426	17(1)	2443	$10{,}920\pm40$	12,960-12,720	LV-C4
					$18{,}010\pm140$	22,080-21,200	
					$21,550 \pm 120$	26,110-25,070	
					$\textbf{24,620} \pm \textbf{300}$	30,220-28,580	
VI	UP	65	0	65	$31,\!270 \pm 683$	27,270-23,870	
VII	MP	249	0	249	$35,070 \pm 340$	41,930-38,370	
					$\textbf{38,750} \pm \textbf{480}$	43,660-41,980	
VIII	MP	29	0				

UP and MP, Upper and Middle Palaeolithic.

^a Between parenthesis, number of obsidians sourced in this work.

^b Samples prepared and analysed at Centre de Datation par le Radiocarbone (Lyon, France) and at the Beta Analytic AMS facility (Miami, USA). Calibrated ages obtained from the Greenland-Hulu timescale (Weninger and Jöris, 2008) with the CalPal Radiocarbon Calibration program.

and SSW respectively (Fig. 1). The Baksan sources, on the northern flank of the Greater Caucasus, lie on the southern slope of the Ebrus volcano, in the Terek river basin. Obsidian is found as boulders in pyroclastic and in secondary contexts (river beds and terraces), close to the villages of Zayukovo in the Baksan valley and in the Chegem valley. Their primary source might be as far as 30 km away in the area of the modern town Tyrnyauz (V. Lebedev, pers. comm.) but unfortunately the eruption centre(s) are unknown. The available elemental analyses suggest a single homogeneous composition and possible discriminations from Caucasian obsidians on the basis of their Rb, Cs and As contents (Blackman et al., 1998; Keller et al., 1996). One fission track analysis of a piece of Baksan obsidian attributed an age of 2.2 \pm 0.2 Ma (Komarov et al., 1972). The obsidian from Baksan found in the Mezmaiskava Cave of northwestern Caucasus show that the Baksan source was used since at least the MP (Golovanova et al., 2010). The access to this source from the southern flank of the Greater Caucasus, where the Bondi Cave and Ortvale Klde are located is difficult, as the crest lines are at



Fig. 3. Obsidian artefacts: (a, b), Bondi Cave level IV (c, d) Ortvale Klde 175–195 cm deep.

an altitude higher than 3500 m and culminate locally at more than 5000 m.

The Chikiani obsidian source described by Blackman et al. (1998) and Badalyan et al. (2004) corresponds to a trachy-rhyolitic dome (2400 m high) located northeast of the Paravani lake (2081 m asl). Obsidian appears as segregations inside the ≈ 1 m thick rhyolitic flows which outcrop around the dome. They display very good quality black homogeneous obsidian, which can also turn into brown, red or green variants. The black variety is usually translucent with no visible spherulites to the naked eyes. There are numerous obsidian boulders on the dome slopes, which are then broken down into pebbles that are carried downslope and downstream by the Paravani River to the Krami River which constitutes the main secondary sources. The Chikiani obsidian presently appears to have a homogeneous composition, though there are some variations in several trace element contents, as Y, Sr, Zr, and Ba (Blackman et al., 1998; Keller and Seifried, 1990; Lebedev et al., 2008). The obsidian relates to extrusions of Middle Pliocene date as demonstrated by two fission ages of 2.34 \pm 0.10 and 2.63 \pm 0.10 Ma (Chataigner et al., 2003) and a K/Ar age of 2.70 ± 0.15 Ma (Lebedev et al., 2008). This area is of relatively easy access from the lake Tsalka area to the north and from the south along the Paravani River (Fig. 5), but is today covered by snow during at least six months per year. The Chikiani obsidian was used since the Lower Palaeolithic as evidenced by the bifaces found in the vicinity of the sources (Kikodze and Koridze, 1978) and was the main raw material for stone tool production from the Mesolithic to the Early Iron Age in the Kura basin (Badalyan et al., 2004).

The next known sources are all further to the south, roughly along a grossly oriented NW-SE direction, from Kars in NE Turkey to the Kel'Bedzar volcano of the Azerbaijani Syunik plateau (Fig. 1). Of those, only the Ashotsk source (also known as Eni-Ël or Kechut) in the northwest of Armenia and the Kars sources in northeastern Turkey are less than 180 km (linear distance) from the Bondi Cave and Ortvale Klde. Their elemental compositions are still poorly documented. Several samples of Ashotsk were analysed by EDXRF (Keller et al., 1996) and NAA (Blackman et al., 1998), while in the Kars area, where six obsidian sources are known, only four samples have been analysed by ICP (Delerue, 2007; Gratuze, in Poidevin, 1998; p. 187) and seven other by SEM-EDS (Delerue, 2007). There is evidence of long-term obsidian-producing volcanic activity in these two areas. Thus while two Ashotsk obsidians present concordant fission track ages of ca 1.0–1.1 Ma (Oddone et al., 2000) another one has

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Fig. 4. Simplified profile (E-W) of the North room of Ortvale Klde, with location of obsidian sampling.

a significantly older K–Ar date of 2.6 \pm 0.2 Ma (Komarov et al., 1972). The time span of obsidian production may have even be larger in the Kars area, with the K–Ar ages of three obsidian samples ranging from ca. 1.9 to 6.9 Ma (Innocenti et al., 1982), while another two fission track ages are around 4.0–4.1 Ma, followed by a much younger one at 3.0 \pm 0.42 Ma (Bigazzi et al., 1993).

Because the Chikiani sources are the closest and the easiest to be reached by foot from the two sites investigated, we determined the elemental composition of eight obsidians from this area (Fig. 5). Five samples were collected among the boulders bordering a railway line at a few tens of metres from each other and analysed by particle induced X-ray emission (PIXE). Three others, coming from different rhyolitic flows, were chemically characterized by inductively coupled plasma (ICP) and dated by K–Ar and 40 Ar/³⁹Ar.

Table 2

Stratigraphical data on lithic artefacts at Ortvale KIde (excavations 2006) and obsidian sampling.

Test pits	Layer	Affinity	Numb	er of lithic ai	tefacts	Samples
	depth cm		Flint	Obsidian	Total	
Cave	70-100	UP	89	1	90	
	100-130	UP	207	2	209	
	130-150	UP	597	7 (2) ^a	604	D28I, D28II ^b
Porch 1	150-175	MP	24	1	25	
	175-195	MP	71	5(2)	76	D28dI, D28dII ^b
	195-210	MP	10		10	
	210-300	MP	9		9	
	300-370	Sterile	0		0	
Porch 2	370-380	MP	80		80	
	380-390	MP	330		330	
	390-400	MP	356		356	
	400-410	MP	563		563	
	410-420	MP	289		289	
	420-430	MP	289	2(2)	291	C26aI, C26aII
	430-445	MP	495	1(1)	496	C26d
	445-460	MP	162		162	
	460-470	MP	96		96	
	470-480	MP	24		24	
	480-490	MP	65		65	
Porch 3	506		5		5	
	520		14		14	
	540		13		13	

UP and MP, Upper and Middle Palaeolithic.

^a Between parenthesis, number of samples sourced

^b The subscripts I and II were added by the Bordeaux laboratory.

4. Experimental procedures

4.1. Elemental compositions

The sourcing of the Bondi and Ortvale Klde Caves obsidians was tentatively determined from their elemental compositions. In order to fulfil a local authority requirement of non-destructive analysis. all the artefacts compositions were determined by PIXE. Five of those were analysed at Centre d'Etude Nucléaire de Bordeaux (CENBG, Gradignan, France) using the nuclear microprobe line of the AIFIRA facility, where the samples are exposed to the particle beam inside a chamber maintained in the accelerator high vacuum (Llabador et al., 1990). The remaining six artefacts and five geological samples were treated at Centre de Recherche et de Restauration des Musées de France (C2RMF, Paris) where the external particle beam of AGLAE is particularly adapted to the analysis of large samples, as shown for obsidians in other circumstances (Calligaro et al., 2005, figure 1; Le Bourdonnec et al., 2011, figure 4). Due to their 'large' sizes, the geological samples and artefact C26d had to be treated at AGLAE. In both PIXE systems, which were shown to provide concordant element contents (Le Bourdonnec et al., 2005) we used and a 3 MeV proton beam. Analytical procedures and data treatments are detailed elsewhere (Bellot-Gurlet et al., 2008; Lugliè et al., 2007; Poupeau et al., 2010).

Three samples, OBS1, 5 and 6, of the Chikiani volcano samples were analysed by ICP at European Institute for Marine Studies (Plouzané, France). For each sample, a master solution was first obtained by an acid (HF + HNO₃) digestion of about 100 mg of powdered sample and evaporated. For the determination of major elements Na to Fe by ICP-AES (atomic energy spectroscopy), a fraction of this dry residue was then dissolved in an H₃BO₃ solution and analysed in an atomic absorption spectrometer Horiba Jobin Yvon Ultima 2 (Cotten et al., 1995). PMS and ACE international standards and the BELC internal standard were used as external and internal controls respectively. A second fraction of the master dry residue was successively dissolved in two acid solutions (HNO₃ and HCl) for trace elements analysis by ICP-MS (mass spectrometry). An aliquot diluted in a third acid (HNO₃) solution, was spiked by Tm (Barrat et al., 1996, 2000) before its introduction inside a high resolution Thermo Finnigan Element II ICP mass-spectrometer. The international standards BCR2 and BIR1 were used as external controls and BHVO-2 used for internal control.

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Fig. 5. Map of the Chikiani-Paravani area showing the obsidian sampling stations.

4.2. Potassium-argon and ⁴⁰Ar/³⁹Ar methods

The extrusion ages of three Chikiani obsidians were determined at the Argon Laboratory of *Laboratoire des Sciences du Climat et de l'Environnement* (Gif-sur-Yvette, France). Obsidian flows OBS1, 5 and 6 ages were determined using an unspiked K–Ar procedure (Charbit et al., 1998). Argon was extracted by the radio frequency heating of a 0.5–1.1 g sample. Periodic cross-calibrations of zeroage standards constraints the mass-discrimination to within $\pm 0.5\%$ on the ⁴⁰Ar/³⁶Ar ratios (Scaillet and Guillou, 2004). Replicate argon analyses on two aliquots were performed for each sample. The K contents were obtained by atomic absorption spectrometry.

⁴⁰Ar/³⁹Ar ages were obtained on samples OBS1 and OBS6. About 20 mm-sized fragments of each sample were irradiated in the OSIRIS (CEA, Saclay, France) nuclear reactor. The neutron fluence was monitored using the Alder Creek Sanidine ACR-2 (Nomade et al., 2005). The J value for each sample was determined from laser fusion analyses of three ACR-2 crystals. After irradiation, 10 glass pieces (250-350 µm) of each sample were loaded into single well and incrementally heated (six to seven steps) using a defocused CO₂ laser. Argon isotopes were measured using a VG5400 mass-spectrometer equipped with a single multiplier (Balzers[©] SEV 217 SEN) operating ion pulse mode. Detailed analytical procedures are presented elsewhere (Nomade et al., 2010). Full analytical dataset for each sample is provided in the Supplementary materials Table S1. Plateau ages and inverse isochron regressions were calculated using ArArCalc (Koppers, 2002).

5. Results

5.1. Chikiani obsidians

5.1.1. Elemental compositions

The element contents determined by PIXE or ICP-AES/MS in Chikiani obsidians are reported in Table 3. The results obtained by these three approaches are remarkably similar. This suggests that they are devoid of analytical biases between the techniques used and that the Chikiani obsidians present 'a single homogeneous composition', as previously observed by Blackman et al. (1998) from their NAA analyses of eleven samples from various outcrops. The other 30 elements determined for the three samples analysed by ICP-MS do not show either significant variations (Table 4).

Chikiani obsidians chemical compositions were obtained in several laboratories (Table 5). In spite of sampling effects and in the absence of inter-laboratory calibrations, the results are essentially concordant, with rare exceptions. The data from three laboratories/ techniques are in very good agreement, namely EDXRF (Keller et al., 1996), NAA (Blackman et al., 1998), PIXE, ICP-AES and ICP-MS (this work). The EDXRF analysis of a single sample from a fourth laboratory agrees with the preceding ones but for Na, Al, Fe, Co and U (Lebedev et al., 2008). A good agreement was also found for the major element data obtained by electron microprobe (EMP) (Frahm, 2010). However the results obtained by EMP on trace elements diverge significantly from the other techniques. The EMP Ga contents, about three times higher by EMP than obtained otherwise, were subsequently shown to result from Ga and Na interferences in the X-ray spectrum

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PIAE and ICP-A															
Sample	Na ₂ O	Al_2O_3	SiO ₂	K ₂ O	CaO	TiO ₂	MnO	Fe ₂ O ₃	Zn	Ga	Rb	Sr	Y	Zr	Nb
PIXE, run AGL	AE-AJ														
Chikiani 1	3.8	13.5	76.5	4.4	0.66	0.096	0.056	0.70	38	15	123	92	13	79	25
Chikiani 2	3.8	13.5	76.4	4.5	0.66	0.104	0.060	0.75	39	15	130	95	16	85	15
Paravani 1	3.8	13.5	76.5	4.5	0.65	0.091	0.057	0.67	41	14	130	94	17	77	20
Paravani 2	4.0	13.6	76.3	4.3	0.64	0.102	0.056	0.73	39	15	132	101	15	89	20
Paravani 3	3.9	13.7	76.4	4.4	0.62	0.095	0.058	0.69	40	14	129	90	11	75	22
ICP-AES/MS															
OBS1	3.90	13.90	76.4	4.7	0.66	0.11	0.06	0.79	42	14.2	124	80	15.3	74	19.5
OBS5	4.07	13.30	76.4	4.7	0.59	0.10	0.06	0.73	40	14.1	128	71	15.4	77	19.6
OBS6	4.00	13.18	76.2	4.7	0.62	0.10	0.06	0.75	43	14.7	128	77	15.4	86	24.7

 Table 3

 PIXE and ICP-AES/MS elemental compositions of Chikiani obsidians. Data for ICP-AES/MS limited to the elements also determined by PIXE.

Contents in oxides are in weight % and contents in elements in ppm. In bold characters, key element contents for the distinction of geochemical groups.

(Frahm, pers. comm.). The Zn and Ce are contents are characterized by a much larger standard error/dispersion and a higher mean value than obtained by other techniques, which suggests that the EMP signal/background ratio might be near to detection value and affects accuracy. The same tendencies might also affect, although in a lesser way, the Zr data. Thus the survey of Table 5 confirms the impression of the uniqueness of Chikiani obsidians composition.

5.1.2. Ages

The three K/Ar ages obtained range from 2.41 \pm 0.05 to 2.83 \pm 0.06 Ma (Table 6). Samples OBS1 and OBS6. collected 100 m apart on the northern side of the Chikiani dome are very close in ages, whereas OBS5, from the southern side of the dome is about 400 ka younger. The two plateaus obtained for OBS1 and OBS6 yield concordant ages spectra with 100% of the defining gas included (Fig. 6). The ⁴⁰Ar/³⁶Ar low intercepts values found for the corresponding isochrons are atmospheric, indicating that neither sample was affected by excess argon or kinetic isotope fractionation (Morgan et al., 2009). The $^{40}\mathrm{Ar}/^{39}\mathrm{Ar}$ ages derived from the plateaus are of 2.84 \pm 0.04 Ma and 2.73 \pm 0.05 Ma for OBS1 and OBS6 respectively. The ages obtained using the corresponding isochrons are identical to the plateau ages and also consistent with their K-Ar ages. The weighed mean ages calculated for OBS1 and OBS6 from their K-Ar and ⁴⁰Ar/³⁹Ar ages are of 2.84 \pm 0.03 Ma and 2.76 \pm 0.04 Ma respectively. Considered altogether, the set of numerical ages obtained on Chikiani obsidians seems to have recorded two major eruptive phases around 2.4 and 2.8 Ma.

5.2. Bondi Cave and Ortvale Klde obsidians sourcing

Four compositional types are represented among the 11 artefacts analysed (Table 7). The distinction between these types relies

 Table 4

 Other elements determined by ICP-MS for the Chikiani obsidians.

		-					
Element	OBS1	OBS5	OBS6	Element	OBS1	OBS5	OBS6
MgO	0.09	0.06	0.08	Nd	15.52	14.34	15.08
P_2O_5	0.03	0.02	0.02	Sm	2.94	2.84	2.90
Li	48.1	42.5	54.6	Eu	0.59	0.57	0.49
Be	2.62	2.63	2.55	Gd	2.32	2.28	2.25
Sc	2.04	2.03	2.11	Tb	0.39	0.39	0.38
V	1.20	0.74	1.24	Dy	2.32	2.39	2.34
Cr	1.2	0.4	3.6	Но	0.47	0.48	0.48
Со	0.2	0.1	0.2	Er	1.36	1.39	1.35
Ni	0.8	0.3	2.4	Yb	1.41	1.44	1.41
Cu	0.9	0.7	0.7	Lu	0.21	0.21	0.21
Cs	5.42	5.67	5.67	Hf	2.35	2.63	2.90
Ba	727	638	682	Ta	1.43	1.20	1.40
La	24.61	21.37	23.16	Pb	18.50	18.95	20.07
Ce	46.30	41.28	42.35	Th	14.16	14.30	15.08
Pr	4.89	4.44	4.79	U	5.22	5.57	5.41

essentially upon their Al, Ca, Ti, Fe, Ti, Zn, Sr and Zr contents. One group of four artefacts display a Chikiani-Paravani type of composition. It includes two pieces (LIV-B4 and LV-C4) from the Bondi Cave UP levels IV and V, and two others (C26aI and C26d) from two successive layers (420–430 and 430–445 cm) of the Ortvale Klde MP test pit 'porch 3' (Table 2). The seven other artefacts fall into the other three compositional types (Fig. 7). Obsidian of the type 1 composition have the highest Sr contents (>150 ppm) of all artefacts; they can also be distinguished by their Ca and Ti contents.

Table 5

Comparisons of elemental compositions obtained in different laboratories for the Chikiani obsidians.

Element	PIXE	ICP-AES/ MS	NAA	EDXRF-1	EDXRF-2	EMP
Na ₂ O	3.9	3.99	$\textbf{4.18} \pm \textbf{0.04}$	4.02	3.52	3.97 ± 0.050
Al_2O_3	13.6	13.46		13.49	12.49	13.5 ± 0.071
MgO		0.077		0.16	0.17	0.1 ± 0.01
SiO ₂	76.4	76.35		75.73	77.03	75.8 ± 0.205
K ₂ O	4.4	4.69	4.54 ± 0.26	4.66	4.52	$\textbf{4.78} \pm \textbf{0.07}$
CaO	0.65	0.623		0.78	0.75	0.690 ± 0.06
TiO ₂	0.098	0.103		0.13	0.14	0.11 ± 0.010
MnO	0.057	0.06		0.06	0.06	0.06 ± 0
Fe ₂ O ₃	0.71	0.76	0.82 ± 0.01	0.95	1.28	
Со		0.2	0.27 ± 0.01	0.61	2	
Zn	40	41.7	$\textbf{37.4} \pm \textbf{0.97}$	40.2	34	71 ± 19
Ga	15	14.3			15	47 ± 4
Rb	129	127	139 ± 6.3	116	124	
Sr	94	75.9		109	90	
Y	15	15.4		16	21	
Zr	81	79	114 ± 19	106	101	85 ± 13
Nb	20	21.3		16	20	56 ± 16
Cs		5.6	4.16 ± 0.09	4.38		
Ba		682	906 ± 103	921	441	836 ± 92
La		23	$\textbf{33.2} \pm \textbf{0.5}$	33.3		
Ce		43.3	$\textbf{52.3} \pm \textbf{0.6}$	59.8		82 ± 21
Nd		15	18 ± 2.2	17.2		
Sm		2.9	$\textbf{2.79} \pm \textbf{0.07}$	3.49		
Eu		0.5	0.56 ± 0.01	0.71		
Tb		0.4	0.39 ± 0.02	0.37		
Yb		1.4	1.59 ± 0.06	1.46		
Lu		0.2	0.14 ± 0.02	0.28		
Hf		2.6	$\textbf{3.38} \pm \textbf{0.04}$	3.61		
Ta		1.3	1.39 ± 0.03	1.39		
Th		14.5	15.8 ± 0.17	16.2	5	
U		5.4	5.94 ± 1.2	5.3	7	

Contents in oxides are in weight % and contents in elements in ppm.

PIXE: this work; average on 5 samples.

ICP-AES/MS: this work; average on 3 samples.

NAA, Blackman et al. (1998): average on 11 samples; standard errors calculated from the uncertainties given in per cent.

EDXRF-1, Keller et al. (1996): in *italic*, measurements on one sample; other elements, average on 4 samples.

EDXRF-2, Lebedev et al. (2008): measurement on a single sample.

EMP: Frahm (2010), average on 13 samples; element contents on a single sample first averaged when several measurements were made. On each sample, from one to seven measurements were made.

Contents in oxides are in weight % and contents in elements in ppm.

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Table 6

K-Ar ages of three obsidian flows from the Chikiani dome. Ages calculations based on the decay and abundances constants from Steiger and Jäger (1977).

Sample	K, Weight %	Weight molten, g	⁴⁰ Ar			Mean age value, Ma $\pm 2\sigma$
			%	10^{-11} moles/g	Weighed mean, $\pm 1\sigma$	
OBS1	3.769 ± 0.038	1.02050	54.979	1.858	1.849 ± 0.007	2.83 ± 0.06
		1.01107	50.381	1.840		
OBS5	$\textbf{3.877} \pm \textbf{0.039}$	0.93816	12.037	1.609	1.681 ± 0.006	2.41 ± 0.05
		0.41252	14.109	1.627		
OBS6	$\textbf{3.81} \pm \textbf{0.038}$	0.58800	42.554	1.815	1.848 ± 0.007	$\textbf{2.79} \pm \textbf{0.06}$
		0.43004	19.514	1.885		

This group includes three UP and one MP artefact. Among the UP artefacts, two (LII-B3 and LII-C3) come from the Bondi Cave UP laver II and one (D28dII) from the 130–150 cm UP layer. The MP artefact (C26aII) was collected in the 420-430 cm depth layer of the Ortvale Klde site. The type 2 obsidian is defined by their low Sr (<10 ppm) but also their high Zn and Zr and low Ca and Ti contents. The two artefacts of this group (D28I and D28II) were found in the 130–150 cm UP level of Ortvale Klde. Finally, the one type 3 artefact differs from the type 2 obsidian only by a significantly higher Ca and by marginally higher Zn and Sr contents (Fig. 7). We believe that the origin of types 1 to 3 obsidian has to be searched for among the faraway sources of eastern Anatolia and Armenia. Up to now, almost no obsidians from these areas were characterized by PIXE (Poupeau et al., 2010). Therefore, any provenance attribution of archaeological obsidians of types 1–3 has to be based on data from existing literature. In general this may be somewhat risky, due to the inherent biases between techniques and differences in laboratory calibrations/procedures (Poidevin, 1998: p. 152; Hancock and Carter, 2010). However such pitfalls can be minimized by a careful choice of reference data. In our case, the element contents reported in Table 5 for the Chikiani-Paravani source suggests that we can rely, for a number of elements, on the data obtained in four other laboratories. One can also rely on the electron microprobe (EMP) and ICP-MS data of the Poidevin laboratory (Poidevin, 1998, referenced as 'Cauvin, inédit' in a footnote of his Annexe II Table, p. 169; Gallet, 2001), as shown from the concordant results obtained

on various sources (compare with Poupeau et al., 2010) and by an unpublished inter-laboratory comparison on aliquots of several eastern Anatolia samples (Delerue, 2007 vs. Gallet, 2001). Finally, it must be stated that the agreement between the PIXE and ICP data obtained by our group is in general very good, as shown previously (Bellot-Gurlet et al., 2005; Poupeau et al., 2010) and confirmed in Table 5. It must also be kept in mind that a provenance study in the regions of interest, eastern Anatolia, Caucasia and Azerbaijan is hampered by a limited corpus of data. What about EDXRF (Keller) and PIXE agreement (as with Poupeau et al., 2010).

In spite of these limitations, some inferences can still be drawn about the provenance of the Bondi Cave and Ortvale Klde obsidian. Apart from the Chikiani source, the three 'nearest' source-areas to consider are those of Baksan, Ashotsk and Kars. The former two do not account for our data. The Baksan source is easily distinguished from types 1 to 3 obsidian on the basis of its Sr and Rb Zr contents (Fig. 8), while the distinction of artefacts from the Ashotsk source relies principally on their Rb, Zn, and Zr contents (Fig. 9). Testing for a Kars provenance is a challenge, as several obsidian outcrops are dispersed in an area of more than 1000 km². More than ten years ago, Poidevin (1998) complained about the limited number of characterization data from this source area. Since then, almost no other data have been produced. At Bordeaux, we analysed two samples of the Kars-Digor source by ICP-MS and for the major elements by EMP (Delerue, 2007), both of which are incompatible with our artefacts data.





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Table 7					
PIXE data on	Bondi	Cave and	Ortvale	Klde a	artefacts.

Sample reference	PIXE run	Na ₂ O	Al ₂ O ₃	SiO ₂	K ₂ O	CaO	TiO ₂	MnO	Fe ₂ O ₃	Zn	Ga	Rb	Sr	Y	Zr	Nb	Туре
Upper Palaeolithic	artefacts							_									
Bondi Cave, Samplir	ng of 2007																
LII-B3	AGLAE-AH	4.2	14.0	75.1	4.2	0.87	0.143	0.051	1.07	33	15	101	315	13	159	24	1
LII-C3	AGLAE-AH	3.8	13.7	75.5	4.3	0.92	0.160	0.047	1.07	49	16	112	176	18	140	21	1
LIV-B4	AGLAE-AH	3.8	13.5	76.5	4.5	0.64	0.096	0.060	0.68	39	14	123	84	12	69	17	Chikiani
LV-C4	AGLAE-AH	3.6	13.7	76.1	4.4	0.72	0.112	0.052	0.81	38	14	125	117	17	97	21	Chikiani
Ortvale Klde																	
D28I ^a	CENBG31	4.3	12.8	76.9	4.4	0.47	0.078	0.074	0.99	73	19	147	5	50	211	39	2
D28dI	CENBG31	4.0	13.3	75.3	4.5	1.05	0.109	0.098	1.12	108	23	144	18	49	190	39	3
D28II	CENBG31	4.4	12.9	76.5	4.4	0.47	0.084	0.082	1.12	80	23	152	8	51	234	48	2
D28dII	CENBG31	3.6	13.4	75.9	4.6	0.84	0.124	0.074	1.06	50	20	139	148	14	132	44	1
Middle Palaeolithic	artefacts																
Ortvale Klde																	
C26aII	CENBG31	3.7	13.5	76.0	4.7	0.73	0.109	0.073	0.98	58	19	170	151	13	128	38	1
Klde C26d	AGLAE-AH	3.5	13.9	76.1	4.4	0.69	0.111	0.060	0.80	40	15	121	103	17	80	21	Chikiani
Klde C26aI ^a	CENBG31	3.4	13.4	76.6	4.9	0.70	0.107	0.052	0.70	38	15	132	89	16	86	29	Chikiani

Contents in oxides are in weight % and contents in elements in ppm.

^a The subscripts I and II were given by the Bordeaux Lab.

Among the other eastern Anatolia potential sources, very good agreements were found between the elemental compositions of type 1 and 2 obsidian with those from Ikisdere (Weiss et al., 2002) and Sarikamis North (Gallet, 2001; Delerue, 2007) obsidians respectively (Table 8). The type 3 obsidian, which differs only marginally from type 2 obsidian, might come from another Sarikamis source. Type 1 obsidians also displays some (less marked) chemical affinity with obsidian from the Meydan Dağ, Mus and Pasinler sources (Keller and Seifried, 1990; Poidevin, 1998; Gratuze, 1999; Delerue, 2007; Poupeau et al., 2010) (Fig. 10). Currently, the only eastern Anatolian sources which can be firmly rejected as suppliers to the Bondi Cave and Ortvale Klde are, in addition to the quite different peralkaline obsidians of Bingöl (A) and Nemrut Dağ, those of Bingöl (B) calkalcaline, Erzincan, Suphan Dağ and West Erzurum, which can be discriminated primarily on the basis of their Ca, Ti, Fe, Rb, Sr, Zr and Nb contents (Keller and Seifried, 1990; Poidevin, 1998; Bellot-Gurlet, 1998; Gratuze, 1999; Poupeau et al., 2010).

The corpus of data for the ca. 20 obsidian source-areas of Armenian obsidians is even more limited than that for eastern Anatolia (Poidevin, 1998; Blackman et al., 1998). However, even from the small number of elements in common with the previous analyses and our PIXE data, it seems that most of these sources, with the possible exception of Gutansar and Hatis, are incompatible with the artefacts' data.

Thus if Ikisdere and Sarikamis North appear as good candidates for the origin of types 1 and 2 obsidian, we cannot categorically exclude other possibilities in northeastern Anatolia and central Armenia based on presently available data.

6. Archaeological considerations

It is not surprising to find Chikiani obsidian at Bondi and Ortvale Klde (Fig. 7), as the route to this source is relatively straightforward (low energy expenditure) when following the valleys of the hydrographic basins, specifically via the lower valley of the Kura River (Mtkvari) near Borjomi, then along the Likhi range (or Surami ridge) that connects the Greater and Lesser Caucasus (Fig. 10). This 'shortest' path to the Chikiani source, estimated to be about 170 km long, is not a difficult one even if it includes some ascent near to its end. However it must be kept in mind that especially during the cold interval between 15 and 40 ka, this source was probably only accessible for a very short period of time each year.



Fig. 7. 3D diagram of the Ca, Fe and Zr contents of obsidians from Bondi Cave, Ortvale Klde and the Chikiani source determined in this work by PIXE and ICP-AES/MS.



Fig. 8. 3D diagram of the Rb, Sr and Zr contents of obsidians from Bondi Cave, Ortvale Klde determined in this work by PIXE, and of the Baksan source (Keller et al., 1996).

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Conversely, the way to the Baksan sources is clearly more difficult, separated as they are from the Bondi Cave and Orvale Klde by the Greater Caucasus belt, which culminates at more than 5000 m at Mounts Dikh Tau and Elbruz. The rare passes which would allow human groups to cross the mountains are at altitudes of no less than \approx 3500 m. Nearly constant cold and dry winds blowing from the north made this chain a topographic and climatic barrier difficult to cross, especially during most of the Early UP. However, the similarity of Early UP assemblages North and South of the Great Caucasus suggests that modern humans did cross this barrier, possibly during favourable temperate periods; in contrast, there is no evidence for Neanderthals crossing these mountains. To reach the Baksan sources from the Bondi Cave and Ortvale Klde sites, it is necessary to use a pass at 3500 m between Mounts Dikh Tau and Elbruz involving a difference in elevation of nearly 3000 m over 50 km before descending through various rivers of the Inguri basin, skirting the western end of the coastal plain and reaching the Rioni valley near Kutaisi (Fig. 7). The length of this route, of \approx 270 km, and its topographic and climatic difficulties, might account for the absence of Baksan obsidians at Bondi Cave and Ortvale Klde. This absence might also be a reflection of the fact that we have only sourced a small number of artefacts (see Tables 1 and 2).

The Sarikamis and Ikisdere sources are farther from Bondi cave and Ortvale Klde than the Baksan and Chikiani sources. Nevertheless, the potential routes to these sources appear as practical as those to Chikiani irrespective of their distance (\approx 350 and \approx 400 km respectively). Sarikamis is not far from the high Kura Valley, easily reachable via the Lake Cildir. Then, following the high Kura Valley until Borjomi city, the easiest way to reach the two caves is similar to the route for Chikiani. The location of Ikizdere (\approx 35 km from the Black Sea coast) makes the way to reach Bondi and Ortvale Klde sites effortless in terms of ascent, following the coastline through Batumi city until the Rioni delta from where the two caves are not very far. The distance to the Meydan Dağ and Mus sources makes them unlikely candidates as the source of our two sites' obsidians.

The MP and UP obsidian artefacts from the Bondi Cave and Ortvale cave were brought to the sites as unretouched and retouched debitage products. The two obsidian sources (Chikiani/Paravani) are represented in both sites, despite of their locations in different, albeit nearby, valleys. The presence of other obsidian sources in several of their MP and UP horizons reveals a diversity in obsidian procurement. There are no clear relationships between the specific raw material and the way in which it was flaked, or the type/form of the final product. The flaking systems are always partial thus indicating the movement of debitage products both in the MP and UP. In the other chamber of Ortvale Klde, Adler and Tushabramishvili (2004) observed that some obsidian cores were flaked inside the site, but only in UP levels. The management and the type of mobility of obsidian cannot as yet be described in detail in these sites. We can just observe here that the obsidian pieces arrived, as small and very thin (sometimes transparent) flakes of blades/bladelets and were rarely retouched. In Ortvale Klde MP levels, the tip of a broken point had been abandoned, its presence suggests a resharpening after breakage. Only rare evidence for the re-use of a raw material are found in the two assemblages. The Neanderthals and Modern Humans of Bondi Cave and Ortvale Klde partly used the same obsidian sources, testifying at the very least to a high level of human mobility or artefact exchange through the inter-Caucasus belts plains. The provenances of the artefacts' raw materials attest that both Neanderthals and Modern Humans exploited sources situated in Southern Transcaucasia and beyond. The durability of this behaviour, from MP to UP, might be related to common traditions and oral transmission. As there is evidence, from technological and typological analyses, of Modern Humans crossing the Greater Caucasus belt it is expected that Baksan obsidians, although not identified here, might be found in UP horizons south of this belt. At Bondi Cave and Ortvale Klde, obsidian represents only a small/tiny fraction of the lithic assemblages which are otherwise dominated by locally available good quality flints. This raises the question as to whether the obsidian might have had a non-utilitarian significance rather than a functional status and/or indicates that mobility of raw materials was limited to very good quality and precious stones. The distances involved between obsidian sources and sites documented in this work are of the same order as previously observed in Central Europe for obsidian (Dobosi, 2011) and for other types of extra-ordinary stones (for example rock crystal at Külna in the Czech Republic [Moncel and

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Samples	Method	п	Na ₂ O	Al_2O_3	SiO ₂	K_2O	CaO	TiO ₂	MnO	Fe_2O_3	Zn	Ga	Rb	Sr	Y	Zr	Nb	Reference
Type 1	PIXE	4	3.8	13.7	75.6	4.4	0.84	0.134	0.061	1.04	47	17	131	197	15	140	32	This work
artefacts			0.2	0.3	0.4	0.2	0.08	0.022	0.014	0.04	10	3	31	79	2	14	11	
Ikizdere A	?	27	4.02	13.87	75.09	4.90	0.89	0.16	0.05	1.05	_	_	131	147	15	163	-	Weiss et al. (2002)
Ikizdere B	?	5	3.99	13.76	75.10	4.95	0.87	0.16	0.05	1.02	-	-	137	138	15	155	-	Weiss et al. (2002)
Type 2 artefacts	PIXE	2	4.3	12.9	76.7	4.4	0.47	0.081	0.078	1.06	77	21	149	7	51	223	43	This work
Sarikamis	EMP/ICP-MS	5	4.6	12.6	75.8	4.76	0.45	0.09	0.09	1.06	_	_	169.2	6.4	35.4	151.4	32.8	Gallet (2001)
			0.1	0.27	0.1	0.54	0.01	0.01	< 0.01	0.16	_	_	11.7	2.5	0.3	28.8	2.1	
	EMP/ICP-MS	1	4.44	12.86	76.42	4.68	0.29	0.06	0.10	0.67	64.2	16.2	147	7.4	48.0	129.4	27.0	Delerue (2007)

n, number of samples analysed. When more than one sample is analysed, the element contents reported are mean values and eventually in *italic* their standard deviations. Weiss et al. indicated in their poster only mean values. Contents are given with same units than in previous table. EMP = electron microprobe, other acronyms as in text. The sample measured by Delerue is an aliquote of one Gallet sample.



Fig. 10. Possible obsidian routes from potential eastern Anatolian sources and the Bondi Cave and Ortvale Klde sites (see text).

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Neruda, 2000]) though even greater distances are attested for flint in areas where artefact quality raw materials are absent or rare (cf. Fernandes et al., 2008; Di Modica, 2010).

7. Conclusion

During the Palaeolithic, the use of obsidian by Neanderthals and Modern Humans in northern Transcaucasia is an established feature. This work shows that from at least 30 ka, during the Middle Palaeolithic, several obsidian sources were exploited at Bondi Cave and Ortvale Klde both by Middle (Neanderthals) and Upper (modern men) Palaeolithic groups.

The only source unambiguously identified so far is that of Chikiani, on the northern flank of the Lesser Caucasus. The K–Ar and ³⁹Ar/⁴⁰Ar ages obtained for several Chikiani obsidians are in the same range as obtained previously by K-Ar and fission tracks in other laboratories but these more reliable and precise and suggest that obsidians were emplaced in several eruptions which took place about 2.4 and 2.8 Ma ago. Their elemental characterization by PIXE, ICP-AES and ICP-MS suggest however a very homogeneous composition, in agreement with Keller et al. (1996) and Blackman et al. (1998). The other three types of obsidian are thought to most likely to be products of some of the many sources in eastern Anatolia, Armenia and Azerbaijan, perhaps Ikisdere and Sarikamis in northern Anatolia in particular. However, almost of these sources lack the precise mapping and geochemical characterization we have for the Chikiani source. Precise and unequivocal sourcing of these raw materials will be only be possible once these sources' ages, elemental composition, magnetic properties have been documented fully.

Concerning the modes of procurement, in the case of Chikiani, the closest identified obsidian source products attested at the Bondi Cave and Ortvale Klde, we are dealing with a minimum walk of 170 km; this might conceivably have been undertaken by the inhabitants themselves. In contrast, we are dealing with distances in excess of 350 km when we come to consider those sources that might represent our Types 1–3, being located in eastern Anatolia (e.g. Ikisdere and Sarikamis), and Armenia (e.g. Hatis and Gutansar), distances that suggest obsidian procurement likely involved some form of intermediary exchange. The obsidian brought to the Bondi Cave and Ortvale Klde are mostly in the form of small semi-finished products, with only rare evidence for on-site re-shaping and re-use. Neanderthals and later Modern Humans apparently employed the same behaviour with regard to how they worked their obsidian, which raises the question of an eventual transmission of this tradition. However it is not sure that the MP horizons are definitely associated with Neanderthals, or early Modern Humans. The history of obsidian use in this region, where it is attested since the early Palaeolithic (Tushabramishvili et al., 2007) is in its infancy, and calls for the beginning of large scale sourcing/ typo-technological studies. Future provenance studies of complete obsidians assemblages, notably at Bondi Cave and Ortvale Klde, would be an important step towards the understanding of prehistoric human behaviour given that our data suggests the existence of early exchange processes.

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Appendix. Supplementary material

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jas.2011.12.008.

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