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This study presents new geochemical data on dolerites from the Plussulien and Beulin Neolithic quarries in the Massif Armoricain (France) and for dolerite samples from their surrounding local regions. Using the major and trace element data obtained on these rocks, the two quarries and regional dolerite occurrences appear to be clearly distinct. Consequently, a set of chemical criteria is proposed to identify the dolerite axes produced uniquely at the Beulin quarry and to discriminate between the axes produced in the surrounding local region of Beulin, but outside the quarries, and those produced in the local region of Plussulien. These results could also be used, for example, to identify the existence of undiscovered quarries in the Beulin region. This work may provide the breakthrough needed to understand stone axe exchanges during the Neolithic times in north-western France and beyond.

KEYWORDS: DOLERITE, STONE AXE, GEOCHEMISTRY, NEOLITHIC

INTRODUCTION

The social organization of Neolithic societies may be explored, for example, through the knowledge of Neolithic axe exchange and of the localization of their source of production. The petrological and geochemical studies of these axes and their raw materials are without any doubt the necessary approaches to unravel such exchanges in Neolithic societies (Cogné and Giot 1952; Vuaillat *et al.* 1995). Most of the Neolithic axes from Brittany (France) are made of crystalline rocks and only a very few of them are made of flint (~4%). The considerable petrographic diversity observed for the stone axes made of crystalline rocks, led Cogné and Giot (1952) to classify these axes into three petrographic types. The first one, which represents ~8% of known Neolithic stone axes, corresponds to eclogite rocks composed mainly of sodic pyroxene (i.e., jadeite). This is by far the most studied petrographic type

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among all stone axes (Pétrequin and Jeunesse 1995; Ricq de Bouard 1996; Pétrequin *et al.* 2012). Most of the eclogite axes were found in deposits under big dolmens or in Carnac-style tumuli in the Gulf of Morbihan area (southern Brittany). The analyses of their diffuse reflectance (spectroradiometry) suggest a link with the Mont Viso outcrops in the Italian Alps (Errera *et al.* 2012). However, eclogites can also be found in the Brière river area of the Loire-Atlantique department (south-eastern Brittany). This is a much closer source than the Italian outcrops and their possible exploitation by Neolithic peoples cannot be excluded (Mens 1997; Tsobgou Ahoupe 2007). The second petrographic type, named 'fibrolite', represents 22% of the inventory of Neolithic stone axes of Brittany (Cogné and Giot 1952). The use of spectroscopic methods (i.e., spectroradiometry, and Raman and infrared spectroscopy) on stone axes from this group has provided significant information about their sources of production, suggesting only a few axe factory sites in the Massif Armoricain (Guiavarc'h 2009; Pailler 2009, 2012; Guiavarc'h and Querré 2012). The results emerging from these works on 'fibrolite' and eclogite axes have contributed to the development of the theory of a social inequality system during the Neolithic in some parts of France and Europe (Pétrequin *et al.* 1998, 2009). Finally, the third and last petrographic type, which designates 'greenstones'—embracing, among others, dolerites, diorites and amphibolites—represents ~66% of the Neolithic stone axes stored in the museums of Brittany (Cogné and Giot 1952). Most of these axes are made of metamorphic dolerites (metadolerites) and can be further subdivided into three petrographic types, A, B and C (see the review in Vuaillat *et al.* 1995). Type A dolerite, which is the type relevant to our study, has a fine-grained doleritic texture; it is composed of strongly altered plagioclase (saussuritized) and augite (uralitized), with elongated crystals of ilmenite having leucoxene rims (Cogné and Giot 1952). Le Roux and Giot (1965) proposed that the petrographic characteristics of type A dolerites match those from a dolerite outcrop directly quarried at the Plussulien-Roc'h Pol Neolithic site located in Brittany (Fig. 1), therefore identifying the source of production of type A dolerite axes. The discovery of this type A dolerite quarry in the Massif Armoricain has provided valuable information on the diffusion of Breton stone axes along the Loire valley (Le Roux 1999) and elsewhere. Plussulien was the only known Neolithic quarry until the recent discovery of the Beulin quarry (Mayenne, France), 250 km to the east (Kerdivel *et al.* 2011; Kerdivel 2012a).

The discovery of Neolithic dolerite axes in the Massif Central (Vuaillat *et al.* 1995) led to the question as to their origin of production. Dolerite rocks are indeed ubiquitous in both the Massif Armoricain and the Massif Central and the possibility of local production in the latter cannot be excluded. In fact, the comparison of the chemical compositions of 20 stone axes discovered in the Massif Central with those of dolerites outcropping in both massifs unambiguously indicates the existence of a local production area in the Massif Central that remains to be discovered (Vuaillat *et al.* 1995).

In this study, we characterize the chemical compositions of dolerites from the point sources of the Plussulien and Beulin Neolithic quarries and from their surrounding local regions. We use this information to describe the means by which axes from the two sites and their local regions may be distinguished from one another.

QUARRY EVIDENCE FROM THE PLUSSULIEN AND BEULIN NEOLITHIC SITES

Archaeological field surveys around Plussulien in Brittany (Fig. 1 (a); Le Roux 1999, 2011) reveal dolerite artefacts to be widespread over an area of ~1800 m^2 lying between the villages

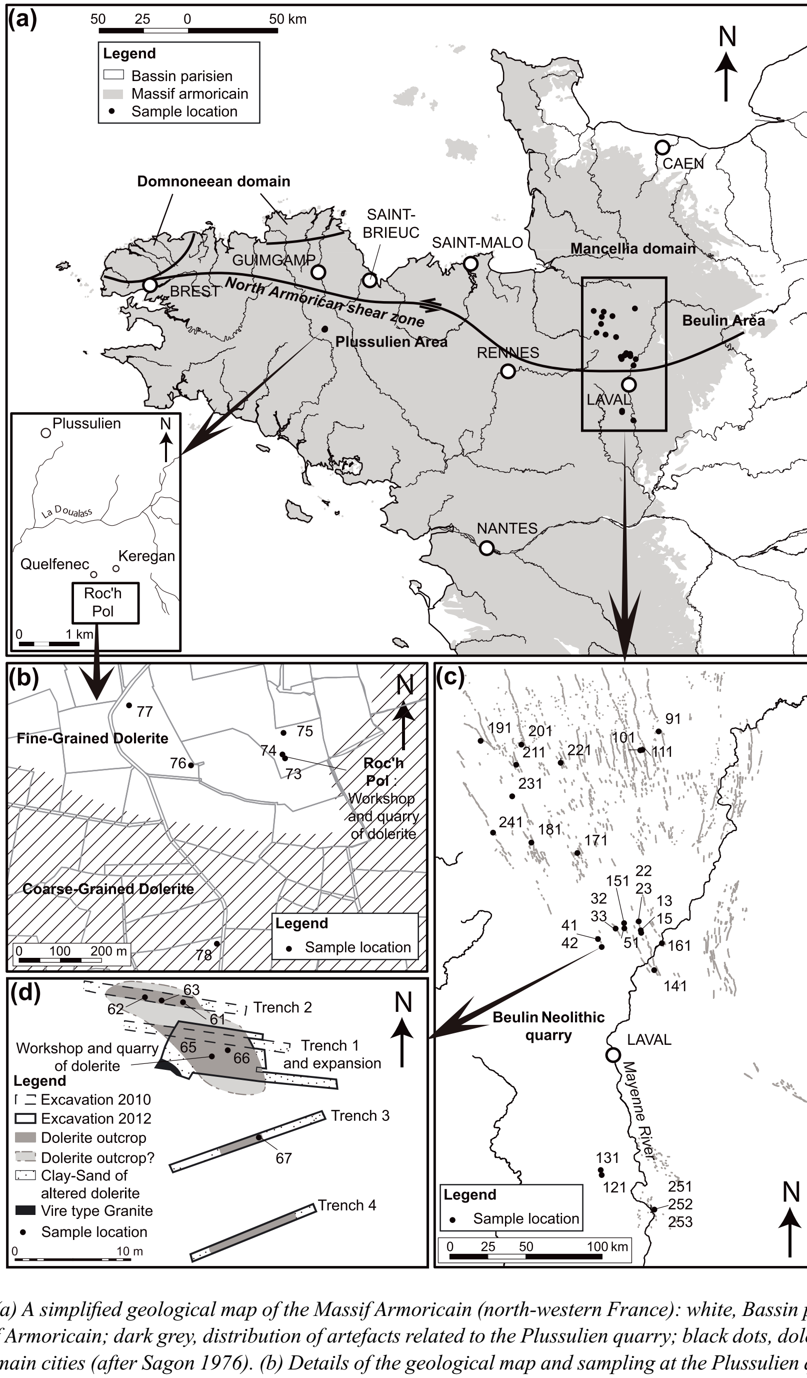

Figure 1 *(a) A simplified geological map of the Massif Armoricain (north-western France): white, Bassin parisien; light grey, Massif Armoricain; dark grey, distribution of artefacts related to the Plussulien quarry; black dots, dolerite samples; white dots, main cities (after Sagon 1976). (b) Details of the geological map and sampling at the Plussulien quarry (Roc'h Pol): black dots, sample locations with sample numbers; hatched zone, coarse-grained dolerite; white, fine-grained dolerite; other grey lines are fields and roads. (c) Details of the sampling of the Beulin area: grey lines, local map of the Mancellia dyke swarm. (d) Details of the geological sampling at the Beulin quarry: dark grey, observed outcrops of dolerite in the excavations; light grey, extrapolation of the dolerite outcrop that may define the orientation of the dykes observed in (c).*

of Quelfenec and Keregan and the Daoulas River to the north (Fig. 1 (a)). The distribution of artefacts near the villages of Quelfenec and Keregan displays four high-density clusters scattered over an area of 500 m^2 (Le Roux 1999, 2011). Le Roux and Giot (1965) first discovered this area near Plussulien (Côtes d'Armor) and identified the now well-known Roc'h Pol quarry (Fig. 1 (b)). In the following, we will refer to this site as the Plussulien quarry. The geomorphological situation of the archaeological site is a significant ridge formed by dolerite rocks that occasionally occur as 3–4 m wide outcrops. The excavations of Le Roux (1999, 2011) focused on two different outcrops that are very close to each other (~18 m apart). The most important outcrop is named Roc'h Pol (Fig. 1 (b)). The second one, located to the east of Roc'h Pol, is a small outcrop characterized by abundant veins of quartz, which certainly degrade the mechanical properties of the rocks there (Le Roux 1999, 2011). The Roc'h Pol outcrop displays abundant evidence of quarrying, such as fine-grained dolerite rocks with sharp edges, negatives flakes, and trimming and polishing marks (Le Roux 1979, 1999, 2011). Three different excavations have been conducted at the Roc'h Pol site over an area of ~250 m^2. The N1/O1 excavation (Le Roux 1979) provides the reference stratigraphic profile, reaching a depth of 4 m. The deepest soil consists of altered rocks, clays and sands resulting from the alteration of the dolerite; these form earth heaps visible in profile as a consequence of Neolithic quarrying. This first period of dolerite extraction has been radiocarbon-dated to around 4000 BC (Le Roux 1999). Two blocks of dolerite are also associated with this stratigraphic level and are interpreted as anvils set up by Neolithic people for trimming stages of the axe-making process. The second period of dolerite extraction is the direct quarrying of the Roc'h Pol outcrop. The technique of extraction takes advantage of the natural rock fracturing, which varies in spacing and orientation, leading to extraction of dolerite blocks of different sizes. During this period of extraction, dated between 3800 and 3200 BC, the dolerite wastes progressively bury both anvils (Le Roux 1999, 2011). The third stratigraphic level displays evidence of the use of fire-setting methods that seems to have been well known at the time, although it was only intermittently used prior to 3200 BC (Le Roux 1999). The extraction at this stratigraphic level was less intensive and limited to the prominent visible ridges of the outcrop. Dolerite axes produced at the Plussulien quarry have been recognized in the South of England as well as in the Rhone valley (Le Roux 1979; Vuaillat *et al.* 1995; Thirault 2004; Pétrequin *et al.* 2012). Geochemical data on axes and corresponding quarried material would certainly help to support the reality of such an important geographical diffusion of axes (Vuaillat *et al.* 1995).

The recent discovery of the Beulin Neolithic quarry, near Saint-Germain-le-Guillaume in Mayenne (France; Fig. 1 (a)), differs from the Plussulien quarry in its lack of massive dolerite outcrops and it was only the presence of clusters of dolerite flakes over an area of ~115 m^2 that led to its discovery (Kerdivel *et al.* 2011; Kerdivel 2012a). The field morphology at the Beulin site shows a scatter of small mounds corresponding to the presence of near-surface dolerite. The Beulin quarry was excavated over an area of ~50 m^2 (Figs 1 (c) and 1 (d)) revealing a quarry organized like an open-pit mine, with several quarrying sites (Kerdivel *et al.* 2011; Kerdivel 2012a). The Neolithic people preferentially quarried dolerite characterized by widely spaced fractures—mostly joints—as indicated by the use of fire-setting methods. Notably, the organization of dolerite blocks in the excavation suggests the extraction of blocks weighing ~150 kg. Flakes with a thick bulb and a great width were used to knap axes at this site. Three radiocarbon ages (Beta-287649, Lyon-8821 and Lyon-8820) on *Malus* (apple tree) and *Quercus* (oak) charcoal yielded ages between 4200 and 3600 BC, which indicate concurrent production of axes at the Beulin and Plussulien sites.

THE SCIENTIFIC APPROACH, THE GEOLOGICAL SETTINGS AND SAMPLING

The scientific approach

The use of chemical compositions of rocks may be used to pinpoint the origin of production of stone axes (e.g., Vuaillat *et al.* 1995). However, this approach requires a good knowledge of the regional chemical variation of outcropping dolerites—and, of course, of the chemical analyses of the stone axes themselves. In this study, we intend to set out a method by which to characterize the geochemical compositions of dolerites quarried by Neolithic people at the Beulin and Plussulien sites in the Massif Armoricain. In addition, we spread the net wider to characterize the local regional geochemical variation of outcropping dolerites around both quarries, to provide a geochemical catalogue of the dolerites that might have been available to the Neolithic people. Combining the use of this geochemical catalogue with the chemical analyses of the dolerite axes allows identification and distinction of the axes produced at the Beulin and Plussulien sites, or at least the definition of their region of production.

The geological setting and samples

First, a brief geological overview is presented of the Châteaulin basin sills and Mancellian dyke swarms where the Plussulien and Beulin quarries were, respectively, opened during the Neolithic. The Mancellian domain in the northern Massif Armoricain extends from Caen at its eastern end to Guingamp at its western end, and is bounded to the south by the North Armorican Shear Zone (NASZ; Fig. 1 (a)). A vast network of dyke swarms cross-cuts the Mancellian domain in a north to south direction. All the geological observations suggest that these dykes were injected during a brief period between the Devonian and Carboniferous (Le Gall 1999). Palaeomagnetic and K–Ar geochronology have given, for example, an age of 330 ± 10 Ma for the St Brieuc doleritic dykes (Perroud *et al.* 1986).

The Plussulien Neolithic quarry was opened on a dolerite sill intruding cleaved shales of Lower Carboniferous age in the Châteaulin basin (Chauris and Guigues 1969) (Fig. 1). These sill rocks display doleritic textures varying from fine to coarse grained, with interstitial pyroxene (augite) and Fe–Ti oxide (ilmenite) embedded in a network of plagioclase laths. The dolerite outcrops at the Neolithic site are characterized by a fine-grained texture and strong hydrothermal alteration (for more petrological details, see the supplementary online material). During our field campaigns, we sampled dolerites 73 and 74 (Fig. 1 (b)) at the Plussulien site, the same as excavated by Le Roux (1999), and dolerites 71 and 72, which correspond to archaeological artefact samples (a nucleus and a flake, respectively) from the same area as Figure 1 (b). Our samples from the local region of Plussulien (four samples) were collected in an area restricted to a few km^2 around the Plussulien site and they belong to the same sill unit. The extent and number of sills (two or three) in this basin are severely restricted when compared to the vast geographical coverage of the Mancellia dyke swarm. There is, additionally, no evidence that the Plussulien sill and Mancellian Guingamp dyke swarm are related to the same magmatic event.

The Beulin Neolithic quarry was opened on a dyke that belongs to the south-eastern part of the Mancellian dyke swarm. The quarried rock is a dolerite with a medium-grained doleritic texture, showing the same mineralogical composition as dolerites from the Plussulien site (plagioclase, augite and ilmenite) but with less pronounced hydrothermal alteration (see the supplementary online information). Seven dolerites were collected at, or in close proximity to, the Beulin site (Fig. 1 (d)). Dolerites 65 and 66 come from a zone of axe production (trench 1), dolerites 61, 62 and 63 were sampled in the continuity of the production zone (trench 2) and dolerite 67 was sampled in close proximity to the production zone (trench 3). Dolerite 64 is an artefact collected on the quarry site. We have also collected a total of 27 rocks from the dyke swarm occurring in the Beulin local region over an area of ~120 km^2 (Figs 1 (c) and 1 (d)). All these local regional rocks are dolerites except for samples 33 and 161, which are more dioritic, with higher abundances of green amphiboles and plagioclase and less abundant pyroxene (See Petrographic descriptions). All sample locations are reported in Table 1.

ANALYTICAL METHODS

Major and trace elements were analysed at the laboratory of LPG-Nantes in Nantes, France, and samples were prepared as follows: the freshest parts of dolerites were first crushed into chips of ~2 mm diameter using a steel jaw crusher. The chips were then rinsed multiple times in ultrasonic baths of milli-Q water, dried and finely powdered in an agate grinder. Major elements were analysed by ICP–OES (iCAP-6300, Thermo), following the procedure described by Cotten *et al.* (1995). Samples were dissolved in closed Teflon PFA beakers using 0.5 mL of HNO_3 65% and 40 drops of HF 40% for 125 mg of rock powder. After 12 h at 110 °C, the acid mixture and dissolved sample were transferred to a 100 mL bottle that contained 5 g of our internal standard solution of 80 ppm Ge and Co, and diluted up to 50 g with a boric acid solution at 20 g L^{-1}. This final solution for analysis was then left for a minimum of 48 h prior to analysis. The geostandards materials BE-N, BHVO-2, BIR-1, JB-2, MO-5 and W-2 were used for external calibration, using the recommended values from Govindaraju (1994) for all standards except for BHVO-2 (Jochum *et al.* 2005). To ensure our reproducibility over time, during each analytical session we have analysed the geostandard WS-E and sample 61 from our sample collection (for the results, see the supplementary online material). Loss on ignition was determined after heating the sample powder at 900 °C for 3 h. All the major element and loss on ignition results are reported in Table 1. Trace elements were analysed by ICP–MS (820MS, VARIAN). Samples were dissolved in closed Teflon PFA beakers using 0.5 mL milli-Q water, 0.5 mL of double-distilled HNO_3 65% and 14 drops of double-distilled HF 40% for 50 mg of rock powder. After 12 h at 120 °C, the dissolved samples were evaporated at 90 °C, then re-dissolved in 1 mL of double-distilled HNO_3 65% at 120 °C and then evaporated to dryness at 90 °C. The final solutions for analysis were obtained by dissolution of sample residues in 250 g of 1 N HNO_3 with the following internal standards: 2 ppb for Ge and 0.8 ppb for Rh, In, Tm and Bi. The geostandards materials BE-N, BHVO-2, BIR-1, JB-2 were used for external calibration. All the trace element results are reported in Table 2. In the same way as for major elements, the geostandard material W-2 and sample ID-61 were analysed throughout our studies to ensure the good reproducibility of our analyses (for the results, see the supplementary online material). All the trace element results are reported in Table 2.

RESULTS

Major elements

Dolerites from the Neolithic quarries in Figure 2 show distinct and quite homogeneous compositions in their SiO_2 and MgO contents, except for dolerite 67 from the Beulin quarry, which displays lower SiO_2 and higher MgO contents compared to other dolerites from the same quarry. Dolerite 67 was sampled in trench 3, which is adjacent to trenches 1 and 2 that relate to the Neolithic quarry *sensu stricto* (Fig. 1 (d)). The major element composition of dolerite artefact

Table 1 *The localization and major element compositions of the dolerite samples analysed in this study, with results for geochemical reference material WS-E (SARM, CRPG, Nancy, France)*

Sample		*Longitude X*	*Latitude Y*	*Wt%*											*Sum*
				SiO_2	*TiO_2*	*Al_2O_3*	*Fe_2O_{3t}*	*MnO*	*MgO*	*CaO*	*Na_2O*	*K_2O*	*P_2O_5*	*L.O.I.*	
13	Beulin area	422520	6798373	47.88	4.49	12.74	15.27	0.20	5.01	8.24	2.90	0.832	0.389	1.23	99.18
15		422511	6798509	48.44	4.15	13.24	15.15	0.20	4.47	7.90	3.17	0.923	0.538	1.51	99.69
22		422357	6799481	47.01	4.74	11.85	17.12	0.22	4.99	8.28	2.58	0.720	0.395	1.52	99.43
23		422357	6799481	48.48	3.78	11.61	18.10	0.31	3.88	7.64	3.13	0.809	1.103	0.93	99.77
32		419322	6798833	51.06	2.63	12.52	16.46	0.20	3.18	7.06	2.61	1.748	0.776	1.32	99.56
33		419322	6798833	55.27	1.91	13.38	12.89	0.16	2.83	5.93	3.31	2.192	0.444	1.59	99.91
41		417363	6797359	46.37	4.05	13.21	15.65	0.23	5.06	8.67	2.94	0.697	0.759	2.32	99.95
42		417355	6797446	46.91	4.09	13.48	15.53	0.25	4.97	9.23	2.55	0.872	0.864	1.64	100.39
51		420332	6799089	46.90	3.70	13.37	13.61	0.19	6.43	9.87	2.63	0.364	0.357	2.18	99.59
64	Beulin artefact	–	–	51.59	2.55	14.02	11.01	0.15	5.63	9.31	2.02	1.116	0.242	2.21	99.85
62	Beulin quarry	417541	6796515	51.25	2.55	13.99	11.34	0.15	5.60	8.99	2.32	0.958	0.229	2.44	99.82
63		417541	6796515	51.65	2.51	14.00	10.90	0.15	5.52	9.15	2.19	1.265	0.239	2.09	99.67
65		417541	6796515	51.28	2.51	14.28	10.99	0.15	5.77	8.11	3.27	1.399	0.238	2.38	100.37
66		417541	6796515	51.00	2.61	14.01	11.46	0.15	5.84	8.22	3.22	1.074	0.263	2.3	100.14
67		417541	6796515	49.82	2.80	14.09	11.99	0.17	6.43	9.13	3.01	0.808	0.290	2.11	100.66
71	Plussulien	–	–	49.64	2.65	13.71	12.14	0.16	6.23	10.93	2.20	0.482	0.279	1.17	99.60
72	artefacts	–	–	49.69	2.53	13.80	12.17	0.16	6.36	10.90	2.25	0.522	0.257	1.26	99.89
73	Plussulien	251193	6812322	48.52	2.01	15.24	10.48	0.14	7.17	11.61	2.03	0.433	0.206	1.84	99.67
74	quarry	251201	6812314	48.50	1.96	15.20	10.06	0.13	7.50	12.07	2.59	0.342	0.208	1.84	100.41
75	Plussulien	251212	6812381	49.30	2.00	11.61	11.88	0.17	9.13	12.30	1.48	0.576	0.213	1.37	100.03
76	area	250980	6812308	49.20	2.70	13.85	12.31	0.15	6.37	11.35	2.41	0.329	0.288	1.67	100.63
77		250830	6812454	49.61	2.81	13.89	11.64	0.16	5.94	11.02	2.21	0.117	0.289	2.26	99.95
78		251035	6811868	49.63	3.21	14.31	14.31	0.18	4.57	7.29	3.76	0.232	0.293	1.84	99.63
91	Beulin area	425287	6824746	47.98	4.69	12.50	15.87	0.20	5.06	8.38	2.43	0.834	0.400	1.31	99.66
101		423014	6822252	47.97	4.52	12.26	16.05	0.20	5.01	8.10	2.58	0.752	0.436	1.51	99.39
111		423027	6822254	45.74	5.21	11.20	18.76	0.26	5.28	8.53	2.25	0.506	0.686	1.07	99.49
121		417802	6766862	46.73	4.28	13.05	14.99	0.19	5.44	8.79	3.31	0.923	0.456	1.53	99.69

(*Continues*)

Table 1 *(Continued)*

Sample		*Longitude X*	*Latitude Y*	*Wt%* SiO_2	TiO_2	Al_2O_3	Fe_2O_{3t}	*MnO*	*MgO*	*CaO*	Na_2O	K_2O	P_2O_5	*L.O.I.*	*Sum*
131		417722	6767411	61.99	0.80	16.20	5.31	0.08	2.15	4.36	3.69	1.838	0.170	2.02	98.61
141		424549	6793170	47.17	4.25	13.08	16.45	0.24	4.80	7.89	2.61	0.716	0.422	2.67	100.30
151		420308	6799383	47.88	3.13	13.73	12.18	0.15	5.75	10.11	2.25	0.446	0.350	2.42	98.41
161		425554	6796568	55.52	1.92	12.76	13.76	0.25	1.99	5.87	3.26	1.655	0.846	1.46	99.30
171		414610	6808832	49.18	3.07	13.61	13.34	0.18	5.27	8.15	3.07	1.210	0.486	2.06	99.63
181		408565	6810257	47.30	3.67	13.62	13.75	0.18	5.89	9.77	2.73	0.631	0.424	2.03	99.99
191		402020	6823551	49.76	3.28	13.74	13.18	0.17	6.15	9.70	2.54	0.717	0.316	1.75	101.31
201		407349	6823120	48.40	3.28	13.56	13.59	0.19	5.01	8.61	3.27	0.551	0.576	1.84	98.87
211		406591	6820370	47.62	3.60	13.24	13.63	0.16	5.57	8.93	2.45	0.787	0.407	2.06	98.47
221		412498	6820673	47.43	3.73	13.50	14.04	0.19	6.28	9.57	2.50	0.610	0.380	2.05	100.28
231		416084	6816283	46.31	4.27	13.06	14.92	0.19	5.43	9.46	2.77	0.646	0.499	1.7	99.24
241		403537	6811231	46.10	4.09	13.98	14.09	0.17	6.20	9.40	2.51	0.581	0.317	1.9	99.34
251		424300	6762195	47.17	2.55	16.76	11.93	0.16	6.22	9.64	2.68	0.565	0.200	2.06	99.94
252		424279	6762185	48.04	4.33	13.42	14.46	0.19	4.92	7.80	3.36	0.897	0.522	1.66	99.61
253		424272	6762185	47.31	4.55	13.54	15.14	0.19	5.20	8.08	3.05	0.505	0.471	2.18	100.22
61	Beulin quarry	417541	6796515	51.67	2.53	13.86	11.12	0.16	5.69	8.17	2.96	1.029	0.227	2.43	99.85
Replicate				51.40	2.60	13.15	10.66	0.15	5.34	7.83	2.70	1.048	0.230		97.54
Replicate				51.77	2.64	13.80	11.21	0.15	5.68	8.05	2.92	1.033	0.236		99.94
Replicate				51.67	2.58	13.75	11.15	0.16	5.68	8.08	2.91	1.018	0.222		99.65
Average				51.63	2.59	13.64	11.03	0.16	5.60	8.03	2.87	1.032	0.229	2.43	99.24
SD				0.16	0.04	0.33	0.25	0.00	0.17	0.15	0.12	0.012	0.006		
%RSD				0.31	1.74	2.44	2.29	1.35	3.11	1.81	4.09	1.207	2.577		
WS-E				50.78	2.42	13.86	13.17	0.17	5.47	9.06	2.55	1.022	0.306	1.3	100.11
WS-E				51.02	2.47	14.12	13.02	0.17	5.52	9.05	2.41	1.089	0.298		100.47
WS-E				51.04	2.49	13.62	13.05	0.16	5.51	8.90	2.42	0.987	0.311		99.80
Average				50.95	2.46	13.87	13.08	0.17	5.50	9.00	2.46	1.033	0.305	1.3	100.13
SD				0.15	0.04	0.25	0.08	0.01	0.02	0.09	0.08	0.052	0.006		
%RSD				0.29	1.57	1.77	0.58	3.23	0.45	1.01	3.25	5.032	2.127		
WSE (Georem)				50.70	2.40	13.78	13.15	0.17	5.55	8.95	2.47	1.00	0.30	1.30	99.77

64 displays a similar composition to those of the dolerite samples from trenches 1 and 2. Dolerites from the Plussulien quarry display on average lower SiO_2 and higher MgO contents when compared to the dolerites from the Beulin quarry. The bounding lines in Figure 2 illustrate the compositional fields for dolerite outcrops sampled around the Beulin and Plussulien quarries. For the purpose of clarity, these dolerites will be referred to as 'local regional' dolerites. The inverse relationship found in the Beulin local regional dolerites in Figure 2 suggests that these rocks are related to each other by a fractional crystallization process. Interestingly, the Beulin quarry dolerites plot outside the local regional Beulin compositional field and show, for a given MgO content, an SiO_2 excess of ~4 wt%. The inspection of other major element diagrams (not shown) indicates that the Beulin quarry dolerites are also characterized, for a given MgO content, by slightly lower $Fe_2O_{3\ total}$ and TiO_2 and slightly higher K_2O contents, compared to the Beulin local regional dolerites.

As mentioned earlier, the geographical coverage of our sampling around the Plussulien area is far less advanced and widespread compared to that of our sample collection from the local region of Beulin. This is partly related to the occurrence of a restricted number of sills in the Plussulien region when compared to the Mancellian dyke swarm in the Beulin region. For this reason, we cannot be sure that the Plussulien compositional field defined in Figure 2 is fully representative of the chemical variations displayed by other dolerites potentially related to this magmatic event (i.e., the Mancellian Guingamp dyke swarm). Bearing this in mind, we do not observe any differences in chemical composition between the Plussulien quarry dolerites and those sampled in the same local region. Finally, we note in Figure 2 that the Plussulien local regional dolerites are clearly distinguishable from the Beulin local regional and quarry dolerites.

Trace elements

The incompatible trace element patterns of dolerites, normalized to the primitive mantle values (McDonough and Sun 1995), are presented in Figures 3 (a) and 3 (b). In such diagrams, the trace elements are ordered from the most incompatible element (left) to the least incompatible element (right). The incompatibility of an element refers to its preferential partitioning into magmas relative to the solid phases at equilibrium. The trace element patterns of the Beulin local regional dolerites (grey patterns in Fig. 3 (a)) reveal, relative to primitive mantle values, an overall increasing enrichment from Lu to Nb–Ta, then a decrease of enrichment from Nb–Ta to Ba, and finally a noticeable increase of the Rb content relative to the Ba content at the most incompatible end. The high field strength element (HFSE) pairs Nb–Ta and Zr–Hf also display positive anomalies relative to their neighbouring elements U–La and Sm–Gd, respectively. Such overall geochemical features are commonly diagnostic of intra-plate (hotspot) magmatism. Interestingly, dolerites from the Beulin quarry (black patterns in Fig. 3 (a)) do not display the positive Nb–Ta anomaly observed for dolerites from the local region of Beulin, but instead display a well-marked positive Th–U anomaly relative to neighbouring elements Nb and Ba. Dolerite 67 from trench 3 does not display the positive Th–U anomaly observed for the quarried dolerite and therefore conforms to the trace element description of the Beulin local regional dolerites. Finally, we note that the trace element composition of dolerite artefact 64 cannot be distinguished from that of the other dolerites from the quarry.

The trace element patterns of the Plussulien local regional dolerites (grey patterns in Fig. 3 (b)) also display the geochemical characteristics of intra-plate volcanism. This result agrees well with previous studies of the Mancellian dyke swarm (Lahaye *et al.* 1995; Vuaillat *et al.* 1995; Caroff

Table 2 *The trace element compositions of dolerite samples, with results for geochemical reference material WS-E (SARM, CRPG, Nancy, France)*

Sample	*Localization*	*Li*	*Be*	*Sc*	*V*	*Cr*	*Co*	*Ni*	*Cu*	*Ga*	*Rb*	*Sr*
61	Beulin quarry	33.3	1.55	30.0	276	194	32	33	18	20.5	32.7	318
62	Beulin quarry	33.2	1.63	29.7	277	194	32	34	19	21.8	28.3	306
63		32.6	1.73	29.1	264	190	36	35	22	21.8	42.4	297
65		29.7	1.48	28.5	259	188	33	34	21	19.9	40.2	252
66		30.0	1.44	29.3	272	196	35	36	26	21.0	30.1	294
67		39.6	1.29	32.3	298	216	38	37	29	20.4	28.5	264
64	Beulin artefact	35.8	1.59	30.3	270	208	34	36	24	20.9	36.7	264
13	Local region	16.9	1.71	32.6	422	38	40	31	44	23.6	24.0	329
15	of Beulin	18.7	1.75	29.6	381	26	40	26	41	24.5	22.5	345
22		20.0	1.71	32.3	436	28	41	27	46	24.9	19.4	312
23		17.9	2.35	30.8	156	1	27	2	12	27.2	20.1	344
32		32.5	2.55	32.5	168	29	30	8	11	25.5	81.3	243
33		21.7	3.26	25.8	146	54	21	6	7	26.7	61.4	210
41		30.2	1.82	29.4	278	69	36	31	31	23.5	23.1	410
42		32.7	1.74	28.4	277	65	37	33	38	23.9	20.0	371
51		40.9	1.47	30.7	341	138	43	62	78	21.9	24.7	357
91		22.2	1.69	32.5	441	51	43	42	37	23.9	22.6	297
101		23.6	1.76	31.2	413	35	40	31	27	23.8	20.7	271
111		16.3	1.63	36.0	483	39	49	33	48	24.2	9.9	267
121		19.7	1.57	31.1	369	67	43	44	65	23.1	18.8	407
131		48.4	1.72	14.9	110	28	14	14	15	19.2	79.4	204
141		43.0	1.45	31.8	378	23	38	27	45	22.4	19.3	303
151		35.6	1.64	28.2	284	167	40	66	60	21.5	14.8	359
171		24.8	1.81	25.9	270	76	38	47	52	22.2	33.5	307
181		22.5	1.50	30.5	324	97	42	62	66	22.5	19.3	344
191		26.0	1.34	30.6	309	156	42	75	76	21.0	24.2	311
201		18.8	1.83	26.7	289	60	36	40	49	24.3	13.3	379
211		20.9	1.64	29.6	339	86	42	60	78	23.0	25.7	373
221		30.6	1.47	31.7	338	114	46	72	76	21.5	24.6	307
231		25.8	1.69	30.7	370	60	43	49	66	23.3	27.1	353
241		30.7	1.26	35.1	410	66	51	31	45	21.9	21.4	334
251		26.7	0.79	25.5	249	187	43	102	54	19.7	13.8	405
252		18.8	1.72	31.0	389	70	38	50	44	25.2	14.9	385
253		20.1	1.57	33.0	428	77	42	54	59	24.8	11.6	338
73	Plussulien quarry	34.8	0.91	31.1	251	372	41	109	78	18.9	10.7	303
74		34.1	0.84	31.6	255	398	39	116	34	18.9	9.2	299
71	Plussulien artefacts	34.0	1.33	33.3	294	221	40	80	93	21.1	9.6	304
72		30.1	1.31	32.5	289	229	40	82	97	20.8	12.2	288
75	Local region of	30.2	0.90	41.4	318	360	56	133	108	17.3	16.1	252
76	Plussulien	31.9	1.10	32.9	292	228	41	81	140	20.1	9.4	329
77		36.3	1.14	33.3	301	218	37	77	35	20.8	31.3	582
78		40.4	1.73	33.2	285	87	37	11	27	22.3	6.9	723

et al. 1996). However, the Plussulien quarry dolerites display slightly negative Zr–Hf anomalies (relative to Sm and Gd) that clearly separate them from the marked positive anomalies observed for the Beulin quarry and its local regional dolerites (Fig. 3 (a)). This observation is reflected by

Table 2 *(Continued)*

Sample	*Y*	*Zr*	*Nb*	*Cs*	*Ba*	*La*	*Ce*	*Pr*	*Nd*	*Sm*	*Eu*	*Gd*
61	31.9	202	20.7	1.35	158	17.6	40.3	5.44	23.8	5.96	1.92	6.49
62	32.6	213	21.5	1.17	145	18.3	41.9	5.62	24.5	6.15	1.86	6.77
63	33.2	213	20.7	1.26	231	18.5	42.8	5.74	25.1	6.32	1.96	6.95
65	31.0	198	19.1	1.50	168	17.1	39.9	5.31	23.1	5.86	1.80	6.43
66	34.0	209	19.9	0.96	152	17.9	41.7	5.64	25.1	6.35	1.95	6.67
67	32.8	201	19.4	1.94	130	15.7	37.0	5.18	24.0	6.14	2.07	6.55
64	32.4	211	20.6	1.23	165	17.9	41.1	5.56	24.4	6.11	1.86	6.77
13	36.7	234	30.0	2.26	134	18.4	44.9	6.39	29.2	7.57	2.58	8.20
15	43.8	247	32.8	2.28	154	23.6	58.1	8.27	38.2	9.55	2.99	10.05
22	38.8	282	32.5	2.06	136	18.2	45.2	6.64	30.7	8.02	2.82	8.66
23	62.2	497	47.8	2.57	175	32.9	81.4	11.74	55.3	14.26	5.46	15.25
32	52.8	335	34.7	10.41	306	36.3	81.4	10.95	47.8	11.64	3.62	12.27
33	58.4	438	36.2	3.26	406	38.7	88.0	11.79	50.1	12.03	3.62	12.64
41	41.7	309	34.8	1.75	138	23.9	58.8	8.33	38.8	9.72	3.55	10.42
42	50.5	344	36.6	3.37	180	28.7	70.4	10.16	47.3	11.71	4.12	11.63
51	35.9	242	34.8	6.47	70	20.9	50.4	7.03	31.7	7.92	2.54	8.45
91	42.6	297	32.0	1.81	172	22.1	52.5	7.35	33.5	8.48	2.81	9.24
101	44.2	304	31.5	1.87	131	21.4	51.5	7.21	33.0	8.46	2.87	8.87
111	49.3	272	41.9	1.59	115	23.7	58.0	8.48	40.1	10.42	3.34	11.39
121	41.1	293	33.7	1.33	168	22.3	54.2	7.64	34.9	8.79	2.87	8.84
131	29.8	126	6.0	2.60	344	16.1	35.4	4.49	18.2	4.51	1.07	4.97
141	37.1	300	28.2	3.12	143	19.7	48.0	6.77	31.4	7.95	2.65	8.62
151	36.3	214	29.3	1.99	63	20.7	49.1	6.73	30.0	7.47	2.24	7.70
171	41.6	306	31.4	1.36	186	26.1	62.2	8.62	38.0	9.39	2.86	9.81
181	37.7	223	29.6	1.73	103	19.9	48.6	6.95	32.1	8.14	2.65	8.32
191	34.2	210	23.6	4.46	116	17.3	41.1	5.71	26.2	6.70	2.14	7.04
201	46.2	265	36.0	1.58	98	25.7	63.4	8.99	41.6	10.33	3.23	10.26
211	38.4	231	29.5	2.25	116	21.6	53.1	7.36	34.8	8.42	2.68	8.36
221	36.3	251	27.5	2.16	161	20.0	48.4	6.80	30.4	7.76	2.42	8.26
231	40.8	291	33.3	1.68	100	23.2	56.4	7.94	36.4	9.06	2.92	9.30
241	31.8	211	23.2	2.51	116	16.3	39.6	5.58	25.4	6.48	2.10	7.07
251	23.3	123	15.4	2.82	69	10.6	24.5	3.37	15.9	4.14	1.55	4.88
252	43.4	285	32.1	1.09	158	23.3	56.4	8.03	36.8	9.40	3.17	10.14
253	40.2	271	30.1	0.53	91	21.5	51.7	7.35	33.8	8.63	2.86	9.43
73	23.3	88	14.0	0.37	63	12.0	28.3	3.89	17.3	4.49	1.57	5.06
74	24.0	83	13.6	0.29	53	11.5	26.7	3.81	17.6	4.45	1.52	4.85
71	31.2	155	21.4	0.57	111	15.6	37.4	5.28	23.7	6.01	1.93	6.72
72	30.3	159	21.4	0.52	101	15.1	36.2	5.03	22.7	5.81	1.90	6.48
75	26.1	109	15.3	0.94	118	12.5	30.2	4.19	19.1	4.94	1.60	5.68
76	31.1	118	20.2	0.49	42	15.8	37.4	5.16	23.8	5.98	1.84	6.38
77	32.1	128	21.7	0.17	20	16.5	38.6	5.47	24.9	6.30	2.11	6.67
78	41.7	139	22.3	1.16	97	23.6	53.6	7.15	31.1	7.58	2.24	8.33

higher Sm/Zr ratios for the Plussulien dolerites compared to those for Beulin (Fig. 3 (c)). Nonetheless, the Plussulien archaeological artefacts tend to show Sm/Zr ratios akin to those from the Beulin area dolerites (Fig. 3 (c)). This observation points to our still poor characterization of the local regional Plussulien dolerites.

Table 2 *(Continued)*

Sample	*Tb*	*Dy*	*Ho*	*Er*	*Yb*	*Lu*	*Hf*	*Ta*	*Pb*	*Th*	*U*
61	1.04	5.95	1.15	2.96	2.58	0.375	5.04	1.27	2.03	3.10	0.892
62	1.07	6.23	1.19	3.01	2.66	0.385	5.19	1.26	3.32	3.26	0.916
63	1.09	6.31	1.19	3.10	2.71	0.397	5.29	1.23	4.03	3.22	0.919
65	0.99	5.83	1.12	2.88	2.56	0.369	5.03	1.19	2.79	3.05	0.857
66	1.08	6.18	1.21	3.06	2.62	0.383	5.03	1.26	2.64	3.18	0.887
67	1.05	5.91	1.15	2.95	2.51	0.371	4.78	1.20	2.54	1.91	0.531
64	1.07	6.16	1.16	3.00	2.64	0.383	5.15	1.25	2.52	3.14	0.895
13	1.27	7.26	1.35	3.33	2.81	0.412	5.38	1.74	1.75	1.72	0.569
15	1.55	8.47	1.64	4.05	3.22	0.469	5.81	1.96	3.10	2.24	0.698
22	1.35	7.71	1.42	3.56	2.98	0.431	6.55	1.89	1.28	1.78	0.614
23	2.26	12.70	2.32	5.57	4.59	0.685	10.22	2.62	1.77	2.54	0.799
32	1.83	10.32	1.90	4.77	4.11	0.604	7.80	1.94	6.11	5.15	1.659
33	1.93	11.14	2.09	5.32	4.78	0.715	10.05	2.04	10.20	7.29	2.269
41	1.54	8.54	1.54	3.79	3.09	0.451	6.96	2.02	5.44	2.13	0.669
42	1.76	9.47	1.79	4.36	3.43	0.494	7.37	2.21	4.03	2.33	0.740
51	1.28	7.22	1.31	3.23	2.58	0.366	5.63	2.02	1.48	1.62	0.498
91	1.43	8.15	1.57	3.95	3.39	0.488	7.19	1.99	2.94	2.46	0.743
101	1.42	8.03	1.56	3.95	3.35	0.494	7.14	1.96	2.91	2.55	0.784
111	1.76	9.69	1.86	4.66	3.78	0.551	6.71	2.51	2.29	2.03	0.694
121	1.40	7.81	1.47	3.66	2.92	0.408	6.69	2.06	2.01	1.82	0.583
131	0.78	4.90	0.98	2.70	2.72	0.396	3.27	0.52	9.42	4.87	2.207
141	1.32	7.44	1.42	3.57	3.01	0.442	7.01	1.73	3.83	1.91	0.661
151	1.23	6.78	1.28	3.23	2.60	0.375	5.34	1.84	3.92	2.64	0.772
171	1.50	8.15	1.57	3.89	3.16	0.456	7.23	1.91	1.55	2.52	0.785
181	1.31	7.13	1.36	3.34	2.73	0.389	5.22	1.82	1.98	1.53	0.469
191	1.13	6.38	1.23	3.10	2.60	0.369	4.97	1.47	2.14	2.05	0.631
201	1.62	8.83	1.66	4.12	3.27	0.469	6.18	2.19	1.47	1.99	0.610
211	1.33	7.24	1.37	3.40	2.81	0.403	5.39	1.84	1.91	1.74	0.517
221	1.25	6.99	1.32	3.34	2.69	0.388	5.98	1.67	1.69	1.89	0.583
231	1.44	7.85	1.48	3.69	2.96	0.427	6.78	2.03	1.57	1.82	0.564
241	1.09	6.17	1.20	3.04	2.56	0.376	5.12	1.49	2.21	1.76	0.566
251	0.75	4.42	0.85	2.14	1.80	0.255	2.99	0.91	1.16	0.78	0.234
252	1.56	8.54	1.64	4.07	3.28	0.480	6.87	1.98	1.70	1.99	0.597
253	1.45	7.95	1.54	3.77	2.96	0.419	6.35	1.83	2.26	1.81	0.549
73	0.78	4.51	0.87	2.16	1.70	0.224	2.37	0.88	2.32	1.21	0.371
74	0.77	4.47	0.86	2.14	1.68	0.229	2.41	0.88	1.58	1.13	0.305
71	1.06	6.15	1.15	2.86	2.38	0.319	3.75	1.25	1.83	1.66	0.493
72	1.01	5.94	1.11	2.81	2.29	0.312	3.75	1.27	2.31	1.69	0.507
75	0.88	5.10	0.97	2.43	1.96	0.268	2.67	0.95	2.16	1.32	0.396
76	1.01	5.72	1.11	2.78	2.30	0.323	3.14	1.25	2.03	1.57	0.478
77	1.06	6.01	1.16	2.93	2.33	0.322	3.15	1.31	3.85	1.62	0.466
78	1.33	7.78	1.56	3.98	3.19	0.440	3.90	1.35	5.57	3.29	0.756

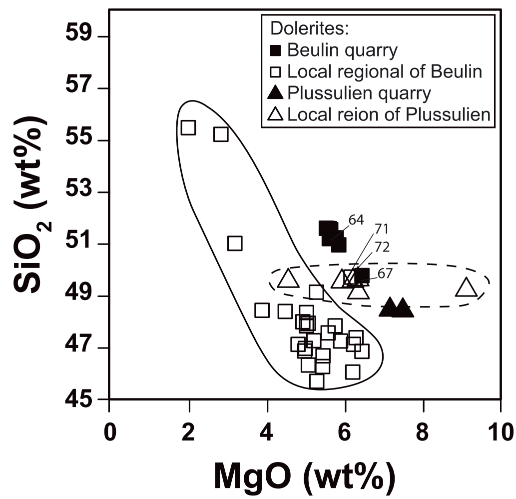

Figure 2 *SiO_2 versus MgO contents for dolerite rocks analysed in this study: ■, dolerite from the Beulin Neolithic quarry; ▲ , dolerite from the Plussulien quarry; □, dolerite from the Beulin local region; Δ, dolerite from the Plussulien local region; ———, compositional field of Beulin local regional dolerites; – – –, compositional field of Plussulien local regional dolerites.*

DISCUSSION

The geochemical signature of the dolerites from the Beulin and Plussulien quarries

We have demonstrated that the Beulin quarry dolerite is characterized by distinct major and trace element compositions compared to the Plussulien quarry dolerite. This is clearly illustrated in Figures 3 (c) and 3 (d), where the chemical compositions of dolerites from the two quarries barely overlap. These differences are remarkable and beyond the analytical errors, which are smaller than the drawn symbols in Figures 3 (c) and 3 (d). From this observation, we have drawn up a list of chemical criteria (Table 3) that can easily be used to discriminate between the rocks that originate from these two quarries. However, these chemical criteria do not have any meaning without knowledge of the chemical variations displayed by the corresponding local regional dolerites in both areas. The question of as yet undiscovered Neolithic quarries of dolerite with similar chemical compositions to those of dolerites from the Beulin or Plussulien quarries certainly needs to be addressed. As an example, in Figure 3, dolerites from the Plussulien local region display similar chemical compositions to dolerites from the Plussulien quarry. This implies that dolerites from the Plussulien quarry cannot be specifically distinguished from other local regional dolerites on the basis of their chemical compositions. As mentioned earlier, our sample collection from the Plussulien region is restricted to only a few samples that, furthermore, are located in close proximity to the quarry. This illustrates the need for a detailed geochemical study of the Châteaulin basin sills and the Guingamp dyke swarm to fully understand the regional chemical variability of dolerites in the local region of the Plussulien site. At a greater geographical scale, a comparison of the Beulin and Plussulien local regional dolerites reveals different and distinct major and trace element characteristics (Figs 2 and 3). Such differences can certainly be used to discriminate between the axes produced in these two regions by using the geochemical criteria listed in Table 3.

In contrast to the Plussulien dolerites, dolerites from the Beulin quarry display distinct and different geochemical features when compared to its local regional dolerites. They show, for

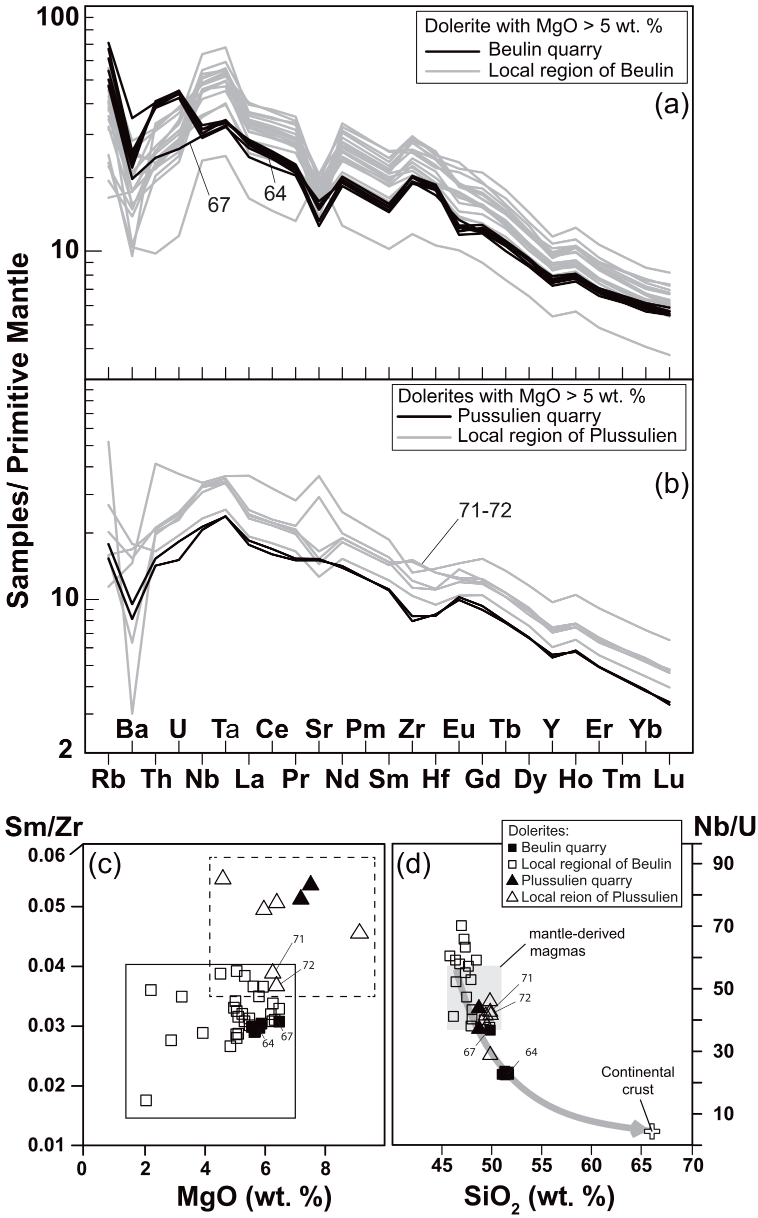

Figure 3 *(a,b) Trace element patterns for dolerites with MgO contents higher than 5 wt% MgO. (a) Dolerite from the Beulin local region (grey) and the Beulin Neolithic quarry (black). Note that sample 67 shows a trace element pattern that is not matched by any dolerite from the Beulin quarry. (b) Dolerites from the Plusssulien local region (grey) and Plussulien Neolithic quarry (black). Note the differences in the trace element patterns for the Th, U, Nb and Ta contents when comparing dolerites from the Beulin quarry with those of the Beulin local region. Note also the differences in the Sm, Zr, Hf, and Eu contents when comparing dolerites from the local regions of Beulin and Plussulien. (c) The Sm/Zr ratio versus the MgO content of the dolerite rocks: ———, compositional field of Beulin local regional dolerites; – – –, compositional field of Plussulien local regional dolerites. (d) The Nb/U ratio versus the SiO$_2$ content of the dolerite rocks: the grey curve is the calculated mixing curve between the chemical composition of the upper continental crust and the chemical composition of the rock (sample 121) derived from a magma characterized by a mantle-like U/Nb ratio.*

Table 3 *Analytical criteria to discriminate between Neolithic axes produced using Massif Armoricain dolerites. Note that averages have been calculated for samples with MgO > 5 wt%, and that averages for local regions do not include data from the quarries*

	Neolithic quarries		*Plussulien region*	*Beulin region*
	Plussulien (Roc'h Pol)	*Beulin*		
Sm/Zr	0.05	0.0295 ± 0.0005	0.04 ± 0.01	0.033 ± 0.003
Nb/La	1.18	1.14 ± 0.03	1.3 ± 0.1	1.5 ± 0.1
Nb/U	41	22.8 ± 0.5	47 ± 3	52 ± 10
Th/Nb	0.08	0.155 ± 0.004	0.079 ± 0.004	0.06 ± 0.01
Ce/Pb	15	15 ± 3	16 ± 4	25 ± 9
SiO_2 (wt%)	48.51	51.4 ± 0.3	49.5 ± 0.2	47 ± 1
MgO (wt%)	7.33	5.6 ± 0.1	6.8 ± 1.3	5.6 ± 0.5
TiO_2 (wt%)	1.98	2.55 ± 0.04	2.5 ± 0.3	3.9 ± 0.7

example, a relatively high SiO_2 content for a given MgO content (Fig. 2) and a particularly low Nb/U ratio of ~23 (Fig. 3 (d)). Such geochemical characteristics for highly incompatible trace element ratios are at odds with the expected values for mantle-derived magmas, which would be characterized by an Nb/U of 55 ± 10 (Hofmann *et al.* 1986). Note that most dolerites analysed in this study, including the Beulin local regional dolerites, plot within the compositional field labelled 'mantle-derived magmas' in Figure 3 (d). The combination of a low Nb/U ratio and a high SiO_2 content strongly suggests that the magma from which the Beulin quarry dolerite originates has assimilated significant amounts of continental crust during its emplacement. Rocks from the continental crust are indeed characterized by a low Nb/U ratio of ~4.5, and high silica contents of ~67 wt% (Hawkesworth and Kemp 2006), which strongly contrast with the expected mantle-derived magma values mentioned just above (see the cross in Fig. 3 (d)). Such a crustal contamination process in the context of intra-continental plate magmatism is quite common and has been widely documented (Patchett 1980; Fodor *et al.* 1985; Caroff *et al.* 1996). We have calculated the mixing relationship expected between a pristine magma composition with Nb/U = 55 ± 10 and SiO_2 = 48 ± 2 wt% (e.g., dolerite 121) and the average composition of the continental crust. The result is illustrated in Figure 3 (d) by the thick grey line connecting the end-member compositions labelled 'continental crust' and 'mantle-derived magmas'. The good agreement between our mixing model and the data (Fig. 3 (d)) suggests that all the Plussulien and Beulin dolerites have experienced significant crustal contamination. More importantly, this model shows that dolerites from the Beulin quarry are the most affected by this process, with up to 25 wt% assimilation of continental crust by the magmas *en route* to the surface. This extensive contamination has therefore imprinted a specific geochemical signature on dolerites from the Beulin quarry relating to both major and trace elements.

ARCHAEOLOGICAL PERSPECTIVES

The geological fingerprint of Neolithic axes

Fifty years after the discovery of the type A dolerite quarry by Le Roux and Giot (1965), Kerdivel *et al.* (2011; see also Kerdivel 2012a) discovered the Beulin site, a new dolerite axe source in the eastern part of the Massif Armoricain. This finding opens up new perspectives for the study of the social and territorial organization of Neolithic populations in the north-west of France through Neolithic axe exchange. Le Roux (1999) suggested a noticeable decrease of interest in type A dolerite material (i.e., from Plussulien) in the east of the Massif Armoricain. Although this decline has not been dated, it might be connected to the discoveries of new Neolithic dolerite quarries at that time (Cogné and Giot 1957; Le Roux 1999; Kerdivel 2012b). The opening of the Beulin quarry might be one of these new resources competing with the Plussilien quarry (Kerdivel *et al.* 2011). Given the unique geochemical signature of the Beulin quarry (Table 3), we propose that it is now possible to identify the axes produced at the Beulin quarry and provide new constraints to understand how the discovery of new doleritic resources might impact the social structures during Neolithic.

Neolithic knowledge of raw materials

There is no doubt that Neolithic people had a very good knowledge of the raw materials in their environment. This knowledge was probably transmitted from generation to generation as an increasing heritage, with its origins going back as far as Mesolithic times. Tsobgou and Dabard (2010) have shown, for example, that in the Massif Armoricain during the Late Mesolithic, specific *phtanite* (radiolarian chert) was used inland as a substitute for flint, which was the main usage near the coast. This is certainly related to the higher abundances of *phtanite* relative to flint rocks inland and to the mechanical properties of *phtanite*, which bears some similarities to flint—they are both siliceous. Fábregas Valcarce and Rodríguez Rellán (2008) and Rodríguez Rellán *et al.* (2011) have also shown, in the western Iberian Peninsula, that schists were used during prehistoric and Bronze Age times due to their suitability for the making of good arrow-heads. The Neolithic people in the area of Sablé-sur-Sarthe favoured flint from a specific source —that is, the Vion quarry (Sarthe, France)—rather than from other random flint outcrops in the region (Georges and Kerdivel 2012; Georges and Lenormand 2012). In another example of Neolithic discrimination, the type A dolerites outcropping at the Plussulien site display two different types of texture: fine-grained and coarse-grained (Fig. 1 (b)); however, the Plussulien quarry was opened exclusively to exploit the fine-grained variety. Tsobgou (2007) has shown that the mechanical properties of coarse-grained dolerites are less suitable than those of finer-grained dolerites, which tends to indicate preferential choice by the Neolithic people based on experience. In the same way, the presence of dolerite axes competing with flint axes, which are less hard than dolerites (Tsobgou 2007), outside the Massif Armoricain may indicate educated choices by Neolithic people that are certainly related to cultural distinctions that remain to be elucidated.

We have shown that dolerites from the Beulin quarry display, for a given MgO content, a significantly higher SiO_2 content compared to other dolerites from the Beulin area. We think that the Neolithic quarrying of this specific high-silica dolerite is not fortuitous and might be correlated with its better knapping capabilities, probably approaching the mechanical properties of flint owing to its high SiO_2 content. As an experiment to challenge our hypothesis, we found out that sculptor J. Huard, who lives in Mayenne, close to the Neolithic Beulin quarry, uses this specific dolerite as a raw material because of its mechanical properties. Interestingly, the technique of identification developed by J. Huard when looking for a raw material is based on the specific sound produced by a shock or blow between two such blocks of dolerite. This demonstrates that basic skills of field prospection can be developed to allow the identification of this rock without the need for modern analytical facilities, and that this specific dolerite is particularly well suited for sculpture. We suggest, therefore, that a Neolithic population in the area did not choose this dolerite dyke for its axe source at random. This study provides new evidence for the knowledge of rocks held by Neolithic peoples.

CONCLUSION

In this paper, we have characterized the major and trace element compositions of dolerites from the Beulin and Plussulien Neolithic quarries and from their respective local regions. The data shows that dolerites from the Beulin quarry display a unique geochemical signature among all the dolerites analysed in the study. This chemical singularity, which can be recognized in both major and trace elements, arises from the particularly extensive assimilation of continental crust by the magma at the origin of the Beulin quarry dolerite. This geochemical singularity of the Beulin quarry dolerite can be used to fingerprint the chemical composition of Neolithic axes that originate from this specific production site. We provide a set of geochemical parameters that are usable in such identifications. We have also shown that the local regional dolerites from Plussulien differ in both major and trace elements from those from Beulin. This result can be used to argue in favour of new sites of axe production in the local regions of Plussulien and Beulin beyond the context of known dolerite quarries. Unfortunately, the dolerites from the Plussulien quarry do not display any unique chemical signature when compared to the local regional dolerites. Among the chemical specificities of the Beulin quarry dolerites, we note the particularly elevated SiO_2 content for a relatively high MgO content. We propose that such high-silica dolerite might have specific physical properties that enhance its knapping capabilities and that these were recognized by Neolithic populations. The results of this study also open up new perspectives on the study of Neolithic stone axe exchange beyond the boundaries of the Massif Armoricain.

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SUPPORTING INFORMATION

Additional Supporting Information may be found in the online version of this article at the publisher's web-site:

Table A1. Major elements replicate analyses of Dolerite 61 and reference material WS-E.
Table A2. Trace elements replicate analyses for dolerite 61 and referenced material W-2. Petrographic descriptions.