

PIXE PROVENANCING OF OBSIDIAN ARTEFACTS FROM PALEOLITHIC SITES IN KOREA

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Keywords: Korea, obsidian, PIXE, INAA, Paektusan

ABSTRACT

The trace element composition (based on Fe, Rb, Sr, Zr) of 50 obsidian artefacts from the Hopyung, Samri and Shinbuk Paleolithic sites were measured by the external beam PIXE method using the 3 MV Tandatron Accelerator Facility at the Seoul National University AMS Laboratory. About 85% of these obsidians originated from the Paektusan volcano (Korea); obsidian of Japanese origin was found only at Shinbuk. INAA analyses on 6 obsidian samples from the Hahwageri and Junghnugri sites are in good agreement with our PIXE results and those of the previous report by Popov *et al.* (2005).

INTRODUCTION

The elemental composition of obsidian depends on its specific geological origin. This property is important for archaeological research because the chemical composition of obsidian can help trace possible exchange and trade routes by linking artefacts found on sites with their geological sources. Consequently, the source identification by elemental analyses of obsidian excavated in various archaeological sites provides important clues to population migrations and cultural exchanges of prehistoric times. A recent survey recorded 90 archaeological sites in Korea that have produced obsidian artefacts (Kim 2002). The general belief is that they were derived from two major routes: one in the northern part of the Korean Peninsula, mostly involving obsidian from the Paektusan volcano, and the other from Japan through Sakhalin. However, few analytic data have been reported so far.

For this study, we collected obsidian from several Upper Paleolithic sites in Korea (Figure 1). These have radiocarbon dates in the range of 15,000-25,000 yr BP. Since the prime concern of Upper Paleolithic studies is the obsidian microlithic industry, the provenance of obsidian may be valuable for tracing the origin and spread of microlithic tools. PIXE measurements were carried out on obsidian artefacts using the Seoul National University's 3 MV Tandatron AMS facility (Kim *et al.* 2002). These

results were compared with a second analytical method involving instrumental neutron activation analysis.

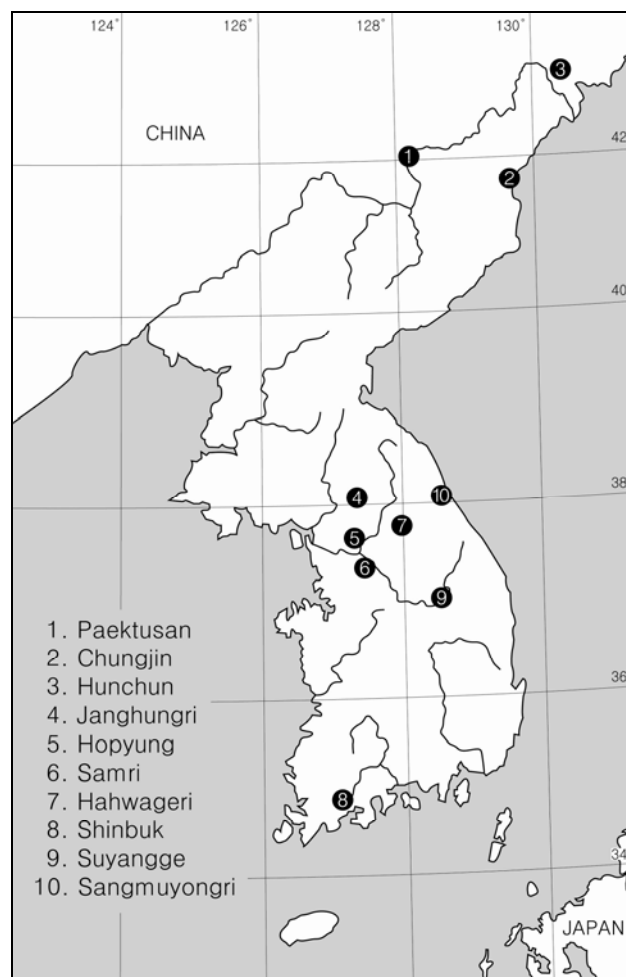


Figure 1. Location of the Paektusan obsidian source and the Paleolithic sites discussed in this paper.

STATUS OF SOURCE STUDIES

The most important aspect of provenancing archaeological obsidian is to have an excellent knowledge of the dis-

tribution and chemical composition of the geological sources. We therefore begin with a summary of the present status of studies on the Paektusan and Japanese obsidian sources because these are the most relevant to Korean archaeology.

Paektusan sources

The Paektusan is the only known obsidian source on the Korean Peninsula (Figure 1). Paektusan obsidians were extensively studied by Kuzmin *et al.* (2002a; 2002b). More recently, Popov *et al.* (2005) have identified three different chemical groups of Paektusan obsidian by analysing geological specimens collected on field trips to Paektusan, combined with archaeological obsidian from southern Primorye in Far East Russia: Paektusan volcano-1 (PNK1); Paektusan volcano-2 (PNK2) and Paektusan volcano-3 (PNK3). As discussed below, we have also used PIXE to analyse four obsidian pieces related to the Paektusan source. These include two pieces claimed to be brought from an Early Iron age site in Hunchun, China and two from a Neolithic site in Chungjin, a coastal city of North Korea, 100 km south east of Paektusan (M.Y. Hong, pers. comm.).

Japanese sources

The following sources of obsidian from the Hokkaido area have been previously studied by Kuzmin *et al.* (2002a): Shiradaki-A; Shiradaki-B; and Oketo volcano. We procured six obsidian samples from the following Japanese sources: Koshidake from Kyushu (2); Shiradaki from Hokkaido (2); and Hariojima (2). Our PIXE analyses of the Shiradaki obsidians are in good agreement with those of Kuzmin *et al.* (2002a) (Table 1).

DESCRIPTIONS OF ARCHAEOLOGICAL SITES

In this section we give a brief description of the Paleolithic sites from which obsidian artefacts were collected for the PIXE and NAA analyses. Their locations are shown on Figure 1.

Junghnugri

The Junghnugri site was found in the early 1980s, but it was excavated only in 1999 in advance of road construction. Stone artefacts were found in fluvial deposits lying on basalt bedrock. Two C14 determinations, 24,200±600 BP (SNU00-380) and 24,400±600 BP (SNU00-381), were obtained from charcoal samples collected from the uppermost clay layer containing a microlithic industry with typical microblades and wedge-shaped microcores (Choi *et al.* 2001). If this association between the dates and the assemblage is accurate, the Junghnugri site represents the earliest microlithic industry on the Korean Peninsula. The presence of the Aira volcanic tephra from Japan (AT) which was found in this site corroborates the radiocarbon measurements (Okuno *et al.* 1997).

Hopyung

The Hopyung site is situated on the lower slope of a mountain range east of Seoul, in the city of Namyangju.

The Upper Paleolithic and Mesolithic industries were recovered from the third layer which was formed by colluvial processes. In total, five radiocarbon dates were obtained on charcoal collected from the uppermost part of this layer (Hong *et al.* 2002). Except for one value, 22,200±600 BP (SNU02-327), four other C14 dates are relatively consistent and cluster around c. 17,000 BP.

More than 1500 stone artefacts were collected from the two cultural horizons in the third layer, mainly from the lower horizon. Among them were typical microblades and wedge-shaped microcores, end scrapers, burins, borers, notches, etc. Many obsidian microlithic artefacts were found in this horizon. The C14 dates from this site along with data obtained for the Junghnugri site provide evidence that the beginning of microblade technology in Korea could be dated earlier than c. 20,000 BP. Another radiocarbon date, 33,200±1900 BP (SNU02-323), was obtained from the fifth layer associated with artefacts typical of an Upper Palaeolithic assemblage and lacking microblades.

Samri

The Samri site is situated near the city of Kwangju, c.50 km south east of Seoul. The site was formed in the low slope area (c.80 m a.s.l.) at the foot of a hill (c.600 m a.s.l.). It was excavated in 2000 by the Institute of Kijon Cultural Property (Han *et al.* 2003). Three cultural layers yielded c. 4000 stone artefacts. Obsidian was found in the first cultural layer. Although no radiocarbon date is available for this layer, it is estimated to be similar to Hopyung at c. 16,000-22,000 BP.

Hahwageri III

The Hahwageri III site is located on a terrace of the North Han River in Hongcheon county, in Gangwon province which is in the central part of the Korean peninsula. It was excavated in 2001-2 by the Kangwon National University and The Institute of Gangwon Archaeology (Choi 2003). Radiocarbon dates from the first and second cultural layers were 13,390±60 BP (SNU02-214) and 40,600±1500 BP (SNU02-212) respectively. The first cultural layer yielded microliths of obsidian and quartz crystal, along with microblade cores, arrowheads, cores, anvils, hammerstones and pecking tools (Choi and Yu 2005). Notable was the large number of obsidian artefacts identified to the Paektusan source by INAA (Table 2).

Shinbuk

The Shinbuk site is located in Jangheung county, 50 km south of the city of Kwangju in the southwestern part of the Korean peninsula. It was excavated by the Chosun University Museum in 2003 and 2004. The excavation, covering over 130,000 m², yielded c. 30,000 artefacts of Upper Paleolithic origin (Lee 2004). The representative tool types are microblades, burins, end scrapers, side scrapers, knives, tanged points, and bifacial thick points. Polished stone artefacts, such as ground adzes, whetstones, pestles, and grinding slabs, were also found. The six C14 AMS dates are in the range of 18,500-25,500 BP:

18,500±300 BP (SNU03-912); 25,500±1000 BP (SNU03-914); 25,420±190 BP (SNU03-914); 20,960±80 BP (SNU03-568); 21,760±190 BP (SNU03-913); 18,540±270 BP (SNU03-915). It should be noted that a number of obsidian tools, quartz crystals and the AT tephra have also been unearthed at this site.

Only one cultural layer has been identified in the stratigraphy of the Shinbuk Paleolithic site. It is placed just below the topsoil horizon at the depth of c. 20 cm. The cultural layer, a pale to dark brown colored clay, is c. 1 m thick and is underlain by a sand layer with sub-angular rocks and then the granitic gneiss bedrock regolith. A very high density of artefacts was recovered: 32,000 artefacts in an excavated area of 20,000 m². Out of all these only 26 obsidian artefacts were recovered, which is markedly less than in the case of nearby Neolithic sites.

GEOCHEMICAL ANALYSES

PIXE

An external beam PIXE system was constructed on the 15 deg. exit port of the multipurpose beam line of the Seoul National University AMS Tandem accelerator (Kim *et al.* 2000). Proton beams of 2 MeV energy were extracted through a 10 µm thick Kapton window and were used to bombard obsidian samples. The samples were accurately positioned using crossed laser beams. X-rays were detected using a Si(Li) detector.

PIXE is an attractive characterization method for obsidian samples since it is a nondestructive technique. However, this procedure can lead to two errors, because it is focused only on the surface of the artefact. The first error is associated with potential surface weathering. A weathered surface might have an altered chemical composition of certain elements because of the effects of ion exchange mechanism with surrounding environments such as moisture, acidity, etc. The second potential source of error is the geometry for X-ray absorption, since curved surfaces of obsidian can cause angular uncertainties for the beam impinging on the target, thereby bringing about additional errors in measurements (also cf. Summerhayes *et al.* 1998: 140).

Low energy X-rays from elements such as Si, Cl, K, and Ca will be more sensitive to this angular effect. For this reason, we have confined our measurements to heavier elements such as Fe, Rb, Sr, and Zr by using a low energy X-ray absorber. As shown in Table 3, the ppm values for these heavier elements are not affected greatly by the shape of the surface and so should produce consistent results.

We measured 50 obsidian pieces: 30 waste flakes from the Hopyung site, 10 waste flakes from the Samri site, and 10 flakes from Shinbuk site. Our reference samples included 4 obsidian samples from the Paektusan source and 6 from Japanese sources (Table 1).

INAA

Secondly, we also analysed 3 obsidian flakes from the Hahwageri site and 3 from the Janghungri site using the Korea Multipurpose Research Reactor (KMRR) at the

Korea Atomic Energy Institute. The obsidian samples analysed by Kuzmin *et al.* (2002b; cf. Choi and Yu 2005) at the Research Reactor Center of the University of Missouri, were split into two and one set was analysed in Korea. As shown in Table 4, there is excellent agreement between these two sets of measurements. (Note that this table lists only those elements which are relevant to our PIXE measurements.)

Previous INAA analyses on 29 obsidian specimens from the Suyangge and Sangmuyongri Paleolithic sites were not assignable to sources (Cho *et al.* 2005; Lee *et al.* 2004:37-50). However, a comparison between their results and our data on obsidian sources (Table 4) enables us to definitely assign these to the Paektusan source (Table 2).

RESULTS AND DISCUSSION

Table 2 summarizes the results of provenancing obsidian from Paleolithic sites in the Korean peninsula. Of the 75 obsidian pieces studied in the present work, 64 are of Paektusan origin, four are from Japan and seven are of unknown origin. Considering only artefacts whose source has been identified, the Paleolithic sites in the Korean peninsula produced only Paektusan obsidian, with the sole exception of Shinbuk where a few pieces of Japanese obsidian were also found. To show the capability of the PIXE technique, in Figure 2 we present the results in terms of a two dimensional plot of Zr/Fe vs Rb/Fe for the artefacts from the Hopyung, Samri, and Shinbuk sites. In Figure 2 there are four clearly separated groups. In counter-clockwise order from the top left these represent an origin from Paektusan 2, Japan, Paektusan 1, and unknown (two cases on the far right side).

The Shinbuk results are very interesting in two respects. The first is that finding three pieces of Paektusan obsidian in this site, which is located in the southern end of Korean peninsula, proves very long distance movement of c. 800 km. Paektusan obsidian found in the Russian Far East Primorye region has been transported about the same distance (Kuzmin *et al.* 2002b).

The second is that Shinbuk is the only Paleolithic site in the Korean peninsula known to date which has produced obsidian of Japan origin. These four pieces demonstrate contact between the Korean peninsula and the Japanese archipelago as early as c. 20,000 BP. Previous suggestions about cultural exchange between the Japanese archipelago and the Korean peninsula have been based on artefact studies. For example, tanged points found in Suyangge were also found at a Paleolithic site in Kyushu, Japan. Matsufuji (1997) considered that the Upper Paleolithic culture in Kyushu was propagated through the Suyangge site. However, a study of Koshidake obsidian showed that it was only in the late Jomon period that this obsidian reached the Korean peninsula (Obata 2003). Thus our finding of Japanese obsidian in Shinbuk will shed a new light on this matter of cultural exchanges as early as c. 20,000 BP.

One possible problem is that the Shinbuk Palaeolithic site produced polished stone tools as well as microlithic

Table 1. PIXE results.

A. Hopyung and Samri

	Fe (%)	Rb (ppm)	Sr (ppm)	Zr (ppm)	Source assignment
Hopyung 1	3.6	277	0	1265	PNK2
Hopyung 2	3.6	293	0	1365	PNK2
Hopyung 3	4.2	349	0	1579	PNK 2
Hopyung 4	4.1	288	0	1451	PNK 2
Hopyung 5	4.1	310	0	1452	PNK 2
Hopyung 6	3.4	378	0	1465	PNK 2
Hopyung 7	1.47	270	10	293	PNK 1
Hopyung 8	1.39	273	0	266	PNK 1
Hopyung 9	1.50	274	0	325	PNK 1
Hopyung 10	1.40	248	0	263	PNK 1
Hopyung 11	1.52	267	39	253	PNK 1
Hopyung 12	1.49	266	49	277	PNK 1
Hopyung 13	1.44	230	43	257	PNK 1
Hopyung 14	1.50	238	32	309	PNK 1
Hopyung 15	2.32	1026	0	1145	Unknown
Hopyung 16	2.30	961	0	920	Unknown
Hopyung 17	3.10	333	0	1254	PNK 2
Hopyung 18	1.45	201	12	241	PNK 1
Hopyung 19	1.43	235	0	211	PNK 1
Hopyung 20	1.45	226	19	217	PNK 1
Samri 1	1.33	310	10	214	PNK 1
Samri 2	1.42	260	12	220	PNK 1
Samri 3	1.40	235	6	256	PNK 1
Samri 4	1.43	265	24	225	PNK 1
Samri 5	1.45	235	10	250	PNK 1
Samri 6	1.51	227	13	199	PNK 1
Samri 7	1.46	290	0	223	PNK 1
Samri 8	1.45	278	7	226	PNK 1
Samri 9	1.47	218	15	217	PNK 1
Samri 10	1.39	225	13	260	PNK 1
O-33 (Source)	1.39	267	15	247	ChungJin, North Korea
O-34 (Source)	1.45	244	18	216	ChungJin, North Korea
O-35 (Source)	1.50	269	8	190	Early Iron age / Hunchun
O-36 (Source)	1.46	273	0	211	Early Iron age / Hunchun

B. Shinbuk Obsidians

	Fe (%)	Rb (ppm)	Sr (ppm)	Zr (ppm)	Source assignment
SB-1	4.1	243	0	1270	PNK 2
SB-2	4.0	253	0	1238	PNK 2
SB-8	4.4	291	0	1383	PNK 2
SB-3	1.65	166	40	173	Ushinodake, Iimori, Ohmura
SB-5	1.9	254	150	158	unknown
SB-7	2.7	248	36	151	probably Japan
SB-9	2.5	214	34	207	probably Japan
SB-10	1.2	190	55	68	Hariojima
SB-4	1.5	153	80	92	Koshidake
SB-6	1.2	193	59	92	Koshidake

C. Japanese source obsidians

	Fe (%)	Rb (ppm)	Sr (ppm)	Zr (ppm)
Koshidake 1	1.78	186	52	82
Koshidake 2	1.41	199	75	73
Shiradaki 1	1.0	124	38	61
Shiradaki 2	1.2	164	47	66
Hariojima 1	1.2	150	58	63
Hariojima 2	1.36	197	43	88

Table 2. Summary of source identifications.

Site	No. of samples	PNK1	PNK2	Japan	Unknown	Method (lab)
Hahwageri	6	1	5			INAA (Missouri) ¹
Suyangge	8	6	2			INAA (KAERI) ²
Sangmuyongri	21	19			2	INAA (KAERI) ³
Hopyung	20	11	7		2	PIXE
Samri	10	10				PIXE
Shinbuk	10		3	4	3	PIXE
Total	75	47	17	4	7	

1. Choi and Yu 2005; 2. Lee *et al.* 2004; 3. Cho *et al.* 2005

Table 3. Measurement sensitivity for heavier elements on angular variations using PIXE.

Angle(°)	Obsidian standard sample (SRM278)			Hopyung #1		
	37	45	53	37	45	53
Fe/(%)	1.91	2.04	3.88	4.24	4.40	3.57
Rb/ppm	141	128	128	355	340	281
Sr/ppm	48	64	58			
Zr/ppm	290	290	244	1565	1651	1272

Table 4. Comparison of INAA results from two different laboratories. Measurements in ppm.

	K	Mn	Fe	Zn	Rb	Sr	Zr	Source
University of Missouri Research Reactor Center								
Hahwageri3	42596	1043	29742	237	284	0	1293	Paektusan volcano-2 (PNK2)
Hahwageri3	42586	1029	30929	253	293	0	1227	Paektusan volcano-2 (PNK2)
Hahwageri3	39608	948	29517	247	289	0	1272	Paektusan volcano-2 (PNK2)
Janghungri	47785	528	24547	227	317	0	1183	Outlier - probably from Paektusan volcano
Janghungri	41977	324	11108	112	252	0	267	Paektusan volcano-1 (PNK1)
Janghungri	38639	969	29458	252	282	0	1243	Paektusan volcano-2 (PNK2)
Korea Multipurpose Research Reactor (KMRR)								
Hahwageri	38971	1009	32008	256			1057	Paektusan volcano-2 (PNK2)
Hahwageri	36682	1005	33209	254			1023	Paektusan volcano-2 (PNK2)
Hahwageri	43603	10554	33150	258			952	Paektusan volcano-2 (PNK2)
Janghungri	33958	541	25898	213			908	Outlier
Janghungri	44557	326	12243	121			244	Paektusan volcano-1 (PNK1)
Janghungri	51340	1007	32514	248			938	Paektusan volcano-2 (PNK2)

artefacts. It is useful to note, however, that polished stone tools have also been found in the Jiepyun Paleolithic site (N. 35°15', E. 128°05')(Bae and Kim 2003), where the cultural layer was dated by AMS at 19,480±540 BP (SNU02-335) and 20,480±800 BP (SNU02-336). Although all charcoal dates at Shinbuk were in the range of c. 20,000 yrs, it may be worthwhile to do more careful studies such as direct dating of obsidian itself by obsidian hydration before hypotheses such as land-bridge formation across the Tsushima Strait or overseas travel during Palaeolithic times are contemplated.

The present analyses show that among the three kinds of Paektusan obsidian (PNK1, PNK2 and PNK3) only PNK1 and PNK2 were used for tool making by Paleolithic people in the Korean peninsula. It is known that the

PNK1 material is the best for tool making and that PNK3 is not suitable (Popov *et al.* 2005). Perhaps this explains why PNK1 is the most common type of Paektusan obsidian in the archaeological assemblages we analysed. Previously, researchers have failed to identify this Paektusan obsidian in archaeological sites in the Korean Peninsula because the geological outcrop has not been located. One plausible explanation may be the catastrophic eruption of Paektusan in c. 1000 AD. This huge volcanic event, which produced approximately 25 km³ of magma (Horn and Schmincke 2000), much larger than the c. 7 km³ for the Pinatubo volcano in 1991 AD, may have largely or completely covered over outcrops of PNK1 obsidian so they are no longer accessible.

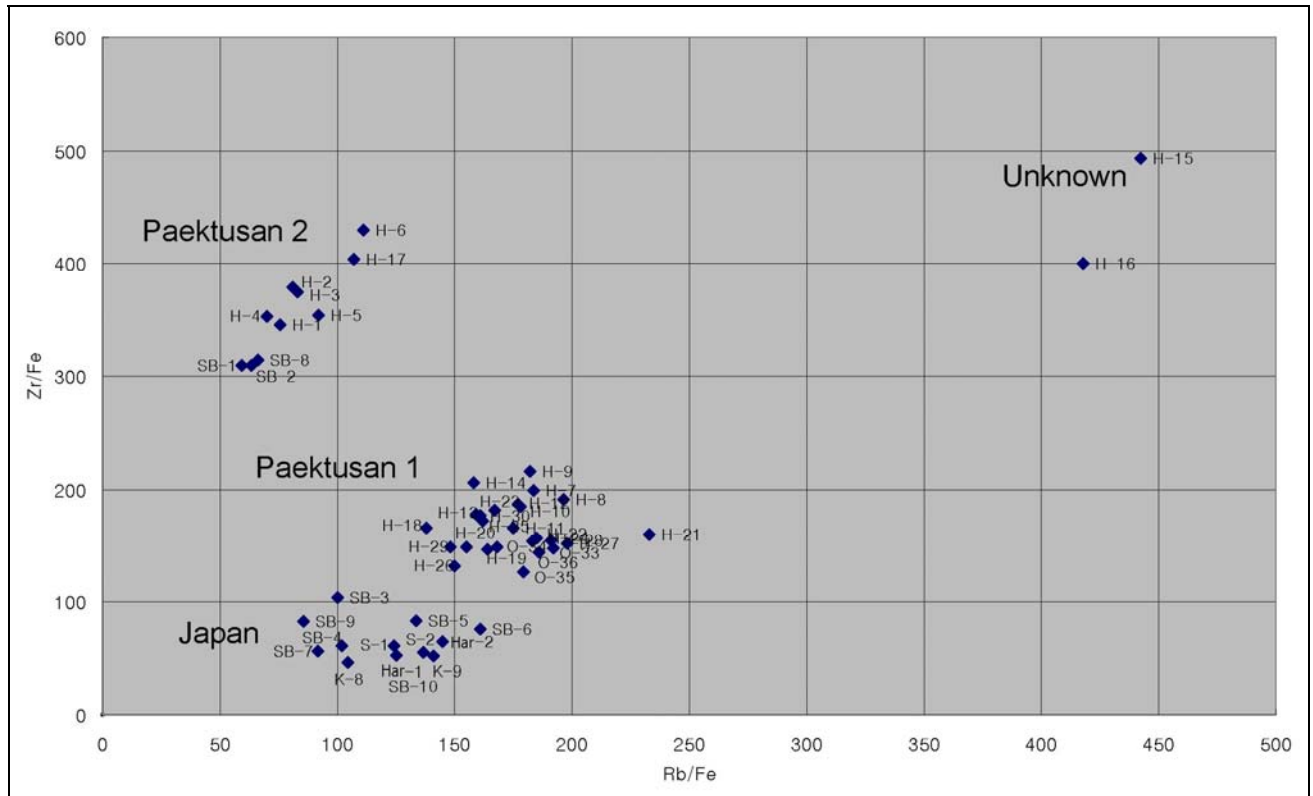


Figure 2. Two dimensional plot of Rb/Fe and Zr/Fe for obsidian artefacts from the Hopyung (H), Samri (S), and Shinbuk (SB) sites. Also plotted are source obsidians from Koshidake (K), Hariojima (Har) and Chungjin-Hunchun (O). In counter-clockwise order from the top left these represent an origin from Paektusan 2, Japan, Paektusan 1, and unknown (two cases on the far right side).

CONCLUSIONS

The external beam PIXE has been successful at identifying the origin of obsidian found on a series of Palaeolithic sites in the Korean peninsula, especially those from the Paektusan volcano. In contrast, sources of obsidian from the Japanese archipelago were not clearly differentiated and may require higher resolution measurements, using techniques such as LA-ICPMS, or HR-SIMS. Results of the characterization study show that obsidian was transported over long distances during the Paleolithic period. Future research should investigate the nature of cultural connections that are implied by the movement of obsidian at this early date.

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