

Non-Destructive energy dispersive X-ray fluorescence sourcing of glassy andesite and basalt artifacts in Upper Paleolithic southwestern Japan: A preliminary assessment

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ABSTRACT

X-ray fluorescence (XRF) analysis for lithic raw material provenance, based on comprehensive geological knowledge, offers indispensable insights into ancient human behavior. Nevertheless, the provenance study of non-obsidian materials is still a challenging field. Within the Setouchi region of southwestern Japan, the sourcing of lithic tools manufactured from glassy andesite and basalt (GAB) has been constrained due to skepticism surrounding the acquisition of compositional data from chemically weathered surfaces. GAB artifacts in Japan have historically been submitted to destructive analysis involving removing surface or sample homogenization, as the skepticism persists despite GAB constituting a substantial portion of lithic raw materials throughout the entirety of ancient occupation in southwestern Japan. This study attempts to overcome this impediment by adopting energy dispersive X-ray fluorescence (EDXRF) analysis using a calibration and methodology originally developed for obsidian. Given the understanding of the tectonic processes underpinning GAB formation, our preliminary assessment successfully discriminates nine distinct geochemical sources among four geologic source groups. Examination of archaeological samples from the region's largest Paleolithic site reveals that lithic raw materials primarily originate from sources approximately 73 km to the east and, to a lesser extent, 98 km to the west, which indicate the potential foraging radius and/or exchange networks of the site occupants. Although there is room for statistical enhancement in sourcing methodology and the implementation of the whole-rock chemical composition of geological GAB by NAA, the future accumulation of GAB sourcing data in this region stands to significantly augment our knowledge pertaining to foragers' exploitation radii, settlement structures, subsistence strategies, and social interactions.

1. Introduction

The sourcing of lithic raw material provides crucial insights into human behavior and social interaction. Globally, geochemical sourcing of obsidian, which is an effective material for producing a variety of lithic tools with sharp cutting edges, has been extensively pursued. These studies have revealed significant data on human mobility routes, the distance of mobility, resource exploitation, migration, and trade and exchange networks (Renfrew et al., 1965; Shackley 1998; Kuzmin and Glascock, 2007; Kuzmin, 2011; Freund, 2013; Ferguson, 2012; Ferguson et al., 2014). This trend is equally evident in Japan (Suzuki, 1969; Warashina, 1972; Warashina and Higashimura, 1973, 1975; Warashina et al., 1977, 1978, 1988; Mochizuki et al., 1994; Ikeya, 2015; Izuho and

Hirose, 2010; Izuho and Ferguson, 2016; Izuho et al., 2014, 2017; Suda, 2012; Suda et al., 2018).

By contrast, the advancement of provenance studies on lithic raw materials other than obsidian, has faced several challenges in Japan. Investigations into the sourcing of artifacts made of glassy andesite or basalt (GAB) have been limited, despite these being widely distributed volcanic rocks throughout Japan, with their glassy variants being especially valuable for lithic tool production. In the Setouchi region of southwestern Japan, where obsidian and other siliceous sedimentary rocks such as "hard" (siliceous) shale or chert are less abundant, GABs have been heavily utilized. Lithic tools made of GAB constitute a large proportion of each lithic assemblage from the Upper Paleolithic (ca. 39000–16000 BP) to the Yayoi period (ca. 3000–1700 BP) in this region.

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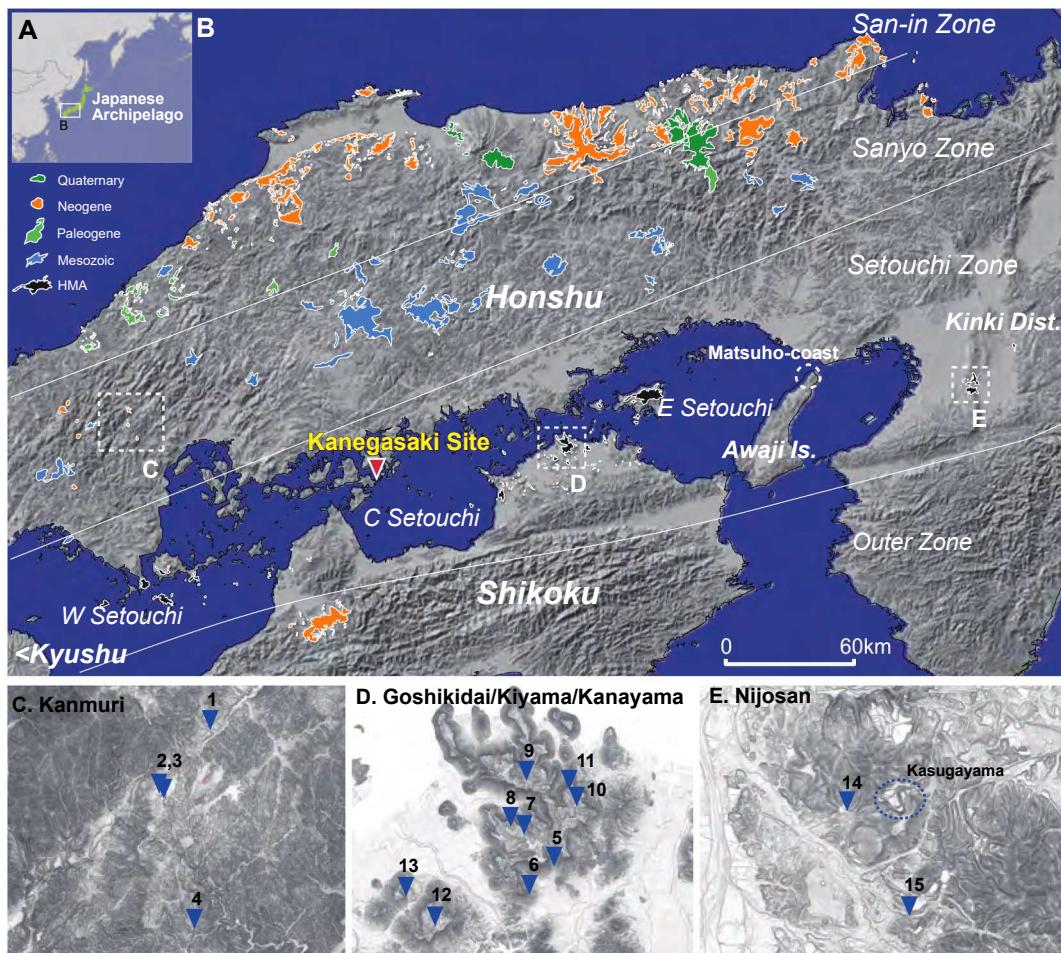


Fig. 1. Distribution of andesite and basalt from the Mesozoic to Quaternary in Setouchi and surrounding regions. Note that sea floor of the present Seto Inland Sea was exposed due to lowered sea-level during the Upper Paleolithic. C-E are three major GAB source groups. Numbers in Map C-E indicate the sampling stops for each source (refer also to Table 1). Maps B-E are adapted and modified from the hillshade and slope map provided by the Geospatial Information Authority of Japan (<https://maps.gsi.go.jp/development/ichiran.html>). Geologic information is sourced from [The Geological Survey of Japan, 2016](#). The classification of tectonic zones follows the criteria outlined by [Kimura et al. \(2003\)](#), while the Oki Zone is omitted in this figure.

Insufficient progress of GAB sourcing is largely attributed to skepticism about acquisition of chemical compositional data from GABs. Unlike obsidian, surfaces of GAB show considerable visible weather changes even in buried contexts, but whether these changes are physical, chemical, both, or some other process is currently unknown. The potential for the chemically mutable nature of GAB artifact surfaces has been argued to reduce the precision of compositional data obtained through XRF analysis because some elements measured by EDXRF are primarily surface measurements ([Jones et al., 1997](#); [Warashina, 1972](#); [Warashina and Higashimura, 1973, 1975](#); [Warashina et al., 1977, 1978](#)). GAB artifacts in Japan have historically been submitted to destructive analysis involving sample homogenization (e.g., [Warashina et al., 1988](#); [Fujine, 2002](#)), impeding the accumulation of GAB sourcing data.

However, the potential impact of surface condition on the accuracy of compositional data acquired by non-destructive analysis needs to be tested rather than assumed. This problem could arise from a limited understanding of X-ray physics, geology, the calibration process, and insufficient testing of homogeneous and well-characterized reference materials for developing a valid calibration curve ([Ferguson, 2012](#); [Liritzis and Zacharias, 2011](#)). This hurdle can be overcome by adopting EDXRF with an established obsidian calibration ([Glascock and Ferguson, 2012](#)). Furthermore, archaeologists have not adequately addressed regional differences in the geochemical composition of GABs within the intensive study of regional igneous rock petrology. As a result, we have little knowledge of why and how the geochemical diversity occurred in

the formation of the arc-trench system of the Japanese Archipelago. In addition to the efforts of archaeologists to describe individual geological occurrences and its secondary deposits ([Takehiro, 2013](#); [Mitsuishi et al., 2021](#); [Okino and Okino, 2017](#); [Hofu Kokogaku Kenkyukai 2022](#); [Niwa, 2009](#)), a high degree of certainty in GAB source discrimination should be achieved based on the basic geologic knowledge of tectonic settings behind each volcanic rock ([Shackley, 1998](#); [Izuhō and Hirose, 2010](#)).

This paper aims to evaluate the potential to differentiate geochemical compositional data of each GAB source through non-destructive EDXRF ([Ferguson, 2012](#); [Izuhō et al., 2017](#)). First, we will review the formation processes of GABs in recent tectonics studies through a literature survey. Subsequently, we will test the feasibility of non-destructive source discrimination of GABs in the Setouchi region by EDXRF. Lastly, we will apply EDXRF source discrimination to an Upper Paleolithic site in the region. If this preliminary application proves successful for sourcing GABs, further methodological development with whole-rock compositional data and its comprehensive application could significantly contribute to our understanding of the ancient history of southwestern Japan and beyond.

2. Formation of GABs and the sourcing

2.1. Tectonics and volcanic activity

The current Japanese archipelago, situated at the western Pacific rim



Fig. 2. A distant view of Mt. Kanmuri from the northeast (Kanmuri area). Photo by K. Morisaki. The Tonbara source is located on the eastern foothill, while other sources are situated on the nearby volcano, several kilometers south from Mt. Kanmuri.

Table 1
GAB geologic sources in Setouchi and surrounding regions.

Source Group	Source Name	Source Type	Location	Geographic Coordinate	
				Latitude (N)	Longitude (E)
Kanmuri	Tonbara	primary	Fig.1C-1	34.454635	132.096319
	Kanmuri-kogen	primary	Fig.1C-2	34.430507	132.077930
	Kanmuriyama-ski-slope	primary	Fig.1C-3	34.434248	132.076867
	Hariyama	primary	Fig.1C-4	34.387672	132.091575
Goshikidai/Kiyama/Kanayama	Kokubudai-minami	primary	Fig.1D-5	34.318900	133.946898
	Renkoji	primary	Fig.1D-6	34.300777	133.936390
	Enshujo-kita	primary	Fig.1D-7	34.329272	133.933475
	Siraminedera	primary	Fig.1D-8	34.332956	133.927429
	Kitamine	primary	Fig.1D-9	34.352214	133.935422
	Aomine	primary	Fig.1D-10	34.348929	133.955942
	Aomine-kita	primary	Fig.1D-11	34.342227	133.957924
	Kiyama	primary	Fig.1D-12	34.291291	133.889738
	Kanayama-higashi	primary	Fig.1D-13	34.305369	133.875870
Awaji	Matsuho-coast	secondary	Fig.1B	34.605967	135.006127
Nijosan	Kasugayama*	primary	Fig.1E-14*	34.538085	135.650765
	Ishimakuri	primary	Fig.1E-15	34.521031	135.655670

*The primary geological occurrence of the Kasugayama source is reported to be at the summit of Kasugayama mountain (Fig.1-E) (Sato et al., 2009). Due to current access restriction to the land, we collected angular cobble samples from the western slope of the mountain.

(Fig. 1-A), evolved mainly from the Cambrian to the Quaternary, initially occurring on the continental margin (Taira et al., 2016; Taira, 2001; Wakita, 2013; Auer et al. 2019). However, most of the basement rocks were formed from younger Jurassic to Paleogene accretion of the Pacific Ocean floor to the Asian side of the continent (Taira et al., 1989; Taira, 2001). The growth of crust (turbidites and hemipelagic sediments) from the side of the Asia towards the ocean is well-documented and was accompanied by the emplacement of granitic rocks and extrusive rocks throughout Honshu, Shikoku, and Kyushu, spanning from the Mesozoic to the early Miocene (Taira, 2001; Imaoka et al., 2011; Sato et al., 2016). The volcanic succession persisted until the Late Paleogene (Imaoka et al., 2011). Extensional tectonics led to the opening of the Sea

of Japan in the early Miocene, within two extensional domains in the present-day Northeast Japan to Sakhalin and Southwest Japan, thereby forming the current backbone of the NE Japan arc, SW Japan arc, and the back-arc region of Japan (Jolivet et al., 1994; Yamaji and Yoshida, 1998).

In southwestern Japan, the clockwise rotation of the southwestern Japan arc, coupled with the opening events of the Sea of Japan, ultimately led to the subduction of the Philippine Sea Plate along the arc. This, in turn, initiated fore-arc volcanism since the middle Miocene. The magmatic province in this area is divided into distinct zones based on the classification proposed by Kimura et al. (2003). These zones include the Outer zone on the trench side, the Setouchi zone, the Sanyo zone, the



Fig. 3. Photo from the north of Kanegasaki site. Photo by K. Morisaki. The Kanegasaki site faces a narrow channel that has yielded a substantial number of large mammal fossils. This sea floor was exposed during the lowered sea-level of the Upper Paleolithic Period.

San-in zone, and the Oki zone, which are arranged from south to north (Fig. 1-B). During this period of volcanism, the initial activity was marked by tholeiitic magma eruptions in the Oki and San-in zones. Although volcanism became active throughout southwestern Japan during the opening of the Sea of Japan, the Setouchi region warrants special attention. Between 17 and 12 million years ago, the Setouchi zone experienced a substantial series of subalkali high-magnesium andesite (HMA) eruptions. These eruptions formed three major clusters: in the Nijosan area on the eastern edge of Setouchi region, another around the Goshikidai and surrounding areas near the boundary between eastern and central Setouchi, and the last in the Geiyo area of western Setouchi (Fig. 1-B). The extent of this volcanic activity spanned westward to Kyushu and eastward to the Kii region, extending beyond the confines of the Setouchi region. Similar lavas are unknown in northeastern Japan. This series of volcanoes is collectively referred to as the “Setouchi volcanic belt” (Tatsumi, 2001,2006). The origin of these volcanoes is attributed to the melt formation of the leading edge of the Philippine Sea Plate, with variations in chemical compositions resulting from the intricate interaction of slab melting and the overlying crust and sediments (Tatsumi, 2001,2006; Kimura et al., 2003,2005; Auer et al., 2019).

Setouchi HMAs, sampled from various locations, exhibit different isotopic trends. It is noteworthy that HMAs from Kinki, northeastern Shikoku, and Kyushu display systematic compositional differences. Even within the northeastern Shikoku region, the isotopic compositions and trace elements are not homogeneous (Tatsumi, 2006; Shinjo et al., 2016). Also, HMAs show a relative Nb depletion compared to those from non-subduction zones (Seno and Matsuura, 2000; Tatsumi, 2006; Nagao et al., 2008).

During the late Neogene (12–4 million years ago), the volcanic activity in the Sanyo, San-in, and Oki zones was characterized by small-scale monogenetic volcanoes that extruded alkali basalt or basaltic andesite (Kimura et al., 2003; Kimura et al., 2005). Differing from the HMA, these lavas possibly stemmed from the upwelling of the asthenosphere. Post 2 million years ago, volcanic activities in the San-in and Oki zones began producing monogenetic alkali basalt, subalkali intermediate rocks, and Adakitic dacites. Some of this activity continues today (Kimura et al., 2005).

2.2. Chemical sourcing of GABs

Glassy andesite around Sanuki district, northeastern Shikoku (Fig. 1-B,D), was first recognized by E. Naumann (1885), and later named ‘Sanukite’ (Weischenk, 1891). Petrologically, sanukite is a glassy variety of bronzite andesite, which has black appearance and breaks with

conchoidal fracture. The similar term ‘Sanukitoid’ was defined later (Koto, 1916), meaning bronzite andesite distributed throughout the Setouchi region. Some Japanese archaeologists casually use the term ‘sanukite’ to indicate glassy andesite, basalt, and similar volcanic rocks in and around the Setouchi region, whereas this terminology is clearly incorrect. To avoid misunderstanding, this paper uses the term GAB to indicates all glassy andesite and basalt suitable for stone tool manufacture.

Chemical sourcing of GABs in and around Setouchi region had been attempted since 1970s. Most of the works have been done by Dr. Warashina and his colleagues, applying EDXRF after the removal of weathered surface (Warashina, 1972; Warashina et al., 1977,1978; Warashina and Higashimura, 1973,1975). His identification approach on GABs was first to measure 12 elements (Al, Si, K, Ca, Ti, Mn, Fe, Rb, Sr, Y, Zr, and Nb) and then discriminate statistically the sources using Hotelling T^2 analysis based on ratios of elements (K/Ca, Ti/Ca, Mn/Sr, Fe/Sr, Rb/Sr, Y/Sr, Zr/Sr, and Nb/Sr). He identified a total 19 chemical groups of geological samples of GAB representing 13 geological sources in more than 700 GAB geologic samples analyzed (Warashina, 2006). These results have been published in numerous reports in rescue excavation reports.

His works expanded our knowledge of the GAB sources frequently exploited in the past. The majority of GABs used for Paleolithic tool production originated from two distinct distributional clusters of HMAs: the Nijosan area, the Goshikidai/Kiyama/Kanayama area, Awaji area (Fig. 1-B,D,E), and another source linked to a monogenetic volcano from the late Miocene situated in the Kanmuri area of the western Sanyo zone (Fig. 1-C).

Past attempts on GAB sourcing were, however, destructive, included only small numbers of samples, and unfortunately not based on the geological background. Given the knowledge on the regional difference in chemical compositions of HMAs and other GABs described by tectonic geology, we could target specific elements capable of differentiating source areas.

3. Materials and methods

3.1. Method

Although EDXRF’s sensitivity, precision, and limited range of detectable elements can sometimes create challenges in differentiating chemically similar sources of volcanic rocks, the GAB sources producing highly glassy rocks ideal for lithic tool production are limited, as previously noted. This limited distribution of flakable stone reduces the potential number of sources.

Table 2

General breakdown of the lithic assemblage by raw materials in the Kanegasaki site.

Toolkit	GAB	Rhyolite	Quartz	Chert	TOTAL
Backed point	74				74
Backed piece	14		1		15
Basal-retouched point	2				2
Tri-facial point	13	1			14
Unifacial point	1				1
Side scraper	18	1			19
End scraper	1	1			2
Drill	3		1		4
Truncation		1			1
Truncated piece	1				1
Notched piece	1	2			3
Biface	1				1
Chipped arrowhead	7				7
Wedge	3				3
Trapezoid	1				1
Microblade core		10			10
Spall	2	1			3
Core	50	14		1	65
Blade	6				6
Flake	932	90	9	2	1033
Retouched blade	2				2
Retouched flake	34	3			37
Pebble	3	1		1	5
TOTAL	1169	125	11	4	1309

XRF analysis was performed using a Bruker Tracer III-SD XRF. This handheld spectrometer features a rhodium-based X-ray tube, operated at 40 kV, and a thermoelectrically cooled silicon detector. Each sample is analyzed for 180 seconds using a protocol outlined in earlier studies (Ferguson et al., 2014; Izuho et al., 2014; Izuho and Ferguson, 2016). For calibration, we utilized a set of 37 thoroughly characterized volcanic rock sources based on measurements using a variety of analytical

techniques (Glascoc and Ferguson, 2012). Matrix-specific calibrations are preferred for accurate determination of elemental concentrations (Ferguson, 2012), but no specific GAB calibration exists and the established obsidian calibration from MURR provides internally-consistent ppm data for materials similar to obsidian and other rhyolites.

The analyzed elements include Mn, Fe, Zn, Ga, Th, Rb, Sr, Y, Zr, and Nb. However, only Rb, Sr, Y, Zr, and Nb data were reliably discernible for source discrimination. As previously highlighted, trace elements including Rb, Sr, Zr are not homogenous throughout the Setouchi region while Y concentrations are basically low. Nb appears crucial for distinguishing Setouchi and non-Setouchi sources.

The statistical interpretation of the compositional data obtained from archaeological sample analysis has been extensively discussed in other works (e.g., Baxter and Buck, 2000; Bieber et al., 1976; Bishop and Neff, 1989; Glascoc, 1992; Harbottle, 1976; Neff, 2000) and is briefly summarized in this study. The assignment of lithic artifacts to specific geologic sources primarily follows the method proposed by Ferguson et al. (2014) and further applied on Hokkaido (Izuho et al., 2014). Statistical analysis was performed using base-10 logarithms of concentrations, as logarithmic concentrations tend to yield more normal distribution for many trace elements compared to raw ppm data. The principal objective of the data analysis is to identify distinct chemical differences between geologic sources areas and then directly compare the artifact chemical data to the established geologic source groups. Decisions on assigning a specimen to a particular compositional group derived from the cumulative probability that the specimen's measured concentrations could originate from that group.

In processing quantitative compositional data, there are still limitations to these statistical approaches. For source discrimination, a line of statistically formal and robust procedures of provenience study pointing out the shortcomings of past provenance studies has been proposed (López-García and Argote, 2023). This issue will be addressed in future work along with the thorough acquisition of whole rock compositional

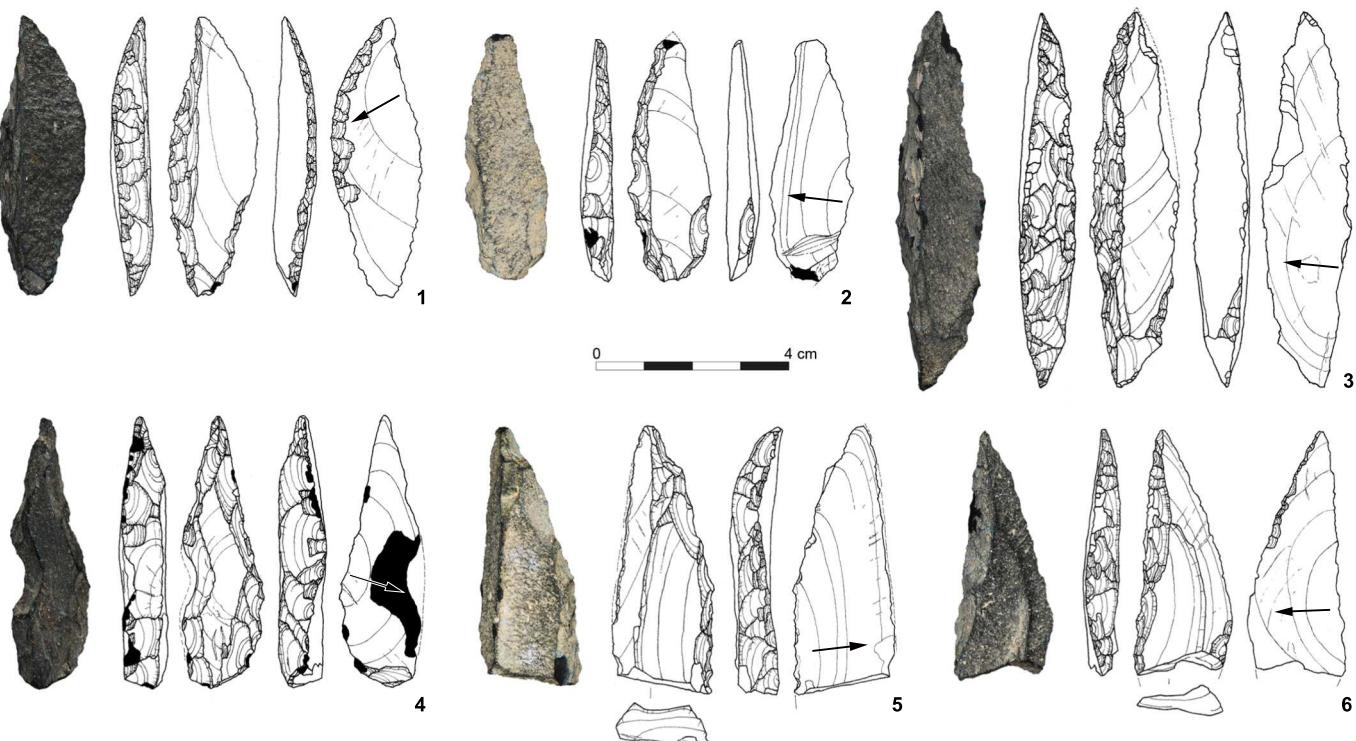


Fig. 4. Hunting weapons made of GAB from Kanegasaki site (Lithic illustration adapted from Museum of Ehime History and Culture, 2020. Photo by K. Morisaki with permission). 1-3,5,6: Backed points on wide flakes. 4: Tri-facial point. Most tools were made from wide flakes as indicated by arrows in the figure. The tools are the collection of the Museum of Ehime History and Culture.

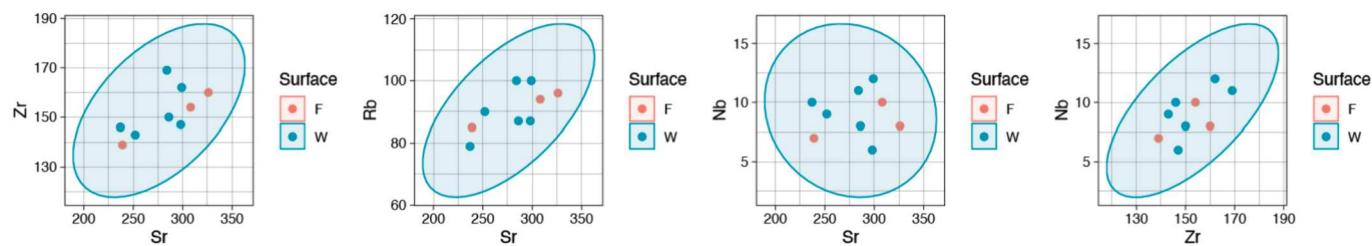


Fig. 5. Comparison of chemical composition of Rb, Sr, Zr, Nb measured on fresh (F) and weathered (W) surface of five Renkoji (Fig. 1D-6) sample. Created by R (R Core Team, 2022).



Fig. 6. Two GAB samples from Renkoji source of Goshikidai/Kiyama/Kanayama source group. GAB is black inside with grayish weathered surface (thinner than 1 mm).

data of GAB which is now underway. At this stage, this paper's purpose is to preliminarily compare the EDXRF data of geologic and weathered archaeological samples in non-destructive method based on the knowledge of the geological background which has never been provided by archaeologists in this region.

3.2. Materials

3.2.1. Geological samples

From our previous discussion on the geologic framework, it is evident that GAB sources predominantly exist in and around the Setouchi region, with concentrations in the Kanmuri, Goshikidai/Kiyama/Kanayama, Awaji, and Nijosan areas (Fig. 2). Table 1 lists the locations of these sampled sources, and Fig. 1-C, D, E visually present them, capturing virtually all known sources. The Kanmuri area, located in the Sanyo zone, encompasses four sub-sources. The other sources are situated in Setouchi zone. The Goshikidai/Kiyama/Kanayama area source group comprises nine sub-sources, and around the Awaji area, samples were specifically collected at Matsuhō-coast. The Nijosan area source group includes the Kasugayama and Ishimakuri sources. From each of these sources, up to 10 samples were collected to discern internal compositional variability. Except for the Matsuhō-coast area, all are identified as primary sources. A total of 167 geologic samples were analyzed by EDXRF (Supplementary Information: S1).

3.2.2. Archaeological samples

The focus here is the Kanegasaki Upper Paleolithic site located in Ehime Prefecture, southwestern Japan (34.211667 N, 133.137500 E; Fig. 1-B). The site is situated on the narrow hill-top at the eastern edge of Hakatajima Island (38 m.a.s.l.). During Last Glacial period when the Seto Inland Sea floor was exposed due to the sea-level lowering, the island was a terrestrial low mountain (Fig. 3). The site's location would have provided a vantage point overlooking the expansive western Setouchi basin. Although no excavations have been undertaken at the

site, Sachio Takahashi, a local archaeologist, provided a large surface collection ($n = 1,309$) from the site (partly reported in [Museum of Ehime History and Culture, 2020](#)). The majority of the collected assemblage is stored at the Imabari City Board of Education with the rest of collections stored at the Museum of Ehime History and Culture. While the collection contains a small number of microblade cores made of rhyolite, and Jomon and later period artifacts made of GAB, it predominantly consists of middle Upper Paleolithic artifacts (ca. 30–19 ka BP) represented by flake-based GAB lithic tools such as backed points, backed pieces, unique tri-facial points, side scrapers, other occasional tools, and debitage (Table 2, Fig. 4). This site stands as the region's largest Upper Paleolithic forager mobility and settlement patterns. Moreover, the locale presents an incomparable opportunity to examine Upper Paleolithic foragers' subsistence, given the vast number of fossil remains of *Palaeoloxodon naumannii* (Naumann's elephant) and *Sinomegaceroides yabei* (giant deer) recovered from the bottom of the Seto inland sea (e.g., [Takahashi, 2022](#), [Kitagawa et al., 2006](#)).

For this study, 678 GAB artifacts (larger than 2 cm in length and width) from the Kanegasaki site were analyzed by EDXRF. A small number of artifacts of phenocryst-rich andesite were excluded ($n = 10$). KM obtained the permission from Imabari City Board of Education in 2022 to borrow the collection and transport to the University of Tokyo for the selection of samples to be analyzed. Following the sample selection, both KM and MI conducted the EDXRF analysis at Tokyo Metropolitan University during 2022–2023. Smaller artifact samples kept at the Museum of Ehime History and Culture were analyzed on-site using the same instrumentation and procedure.

4. Result

4.1. Separation of XRF source groups of geologic samples

We initially tested the potential effects of surface deterioration on

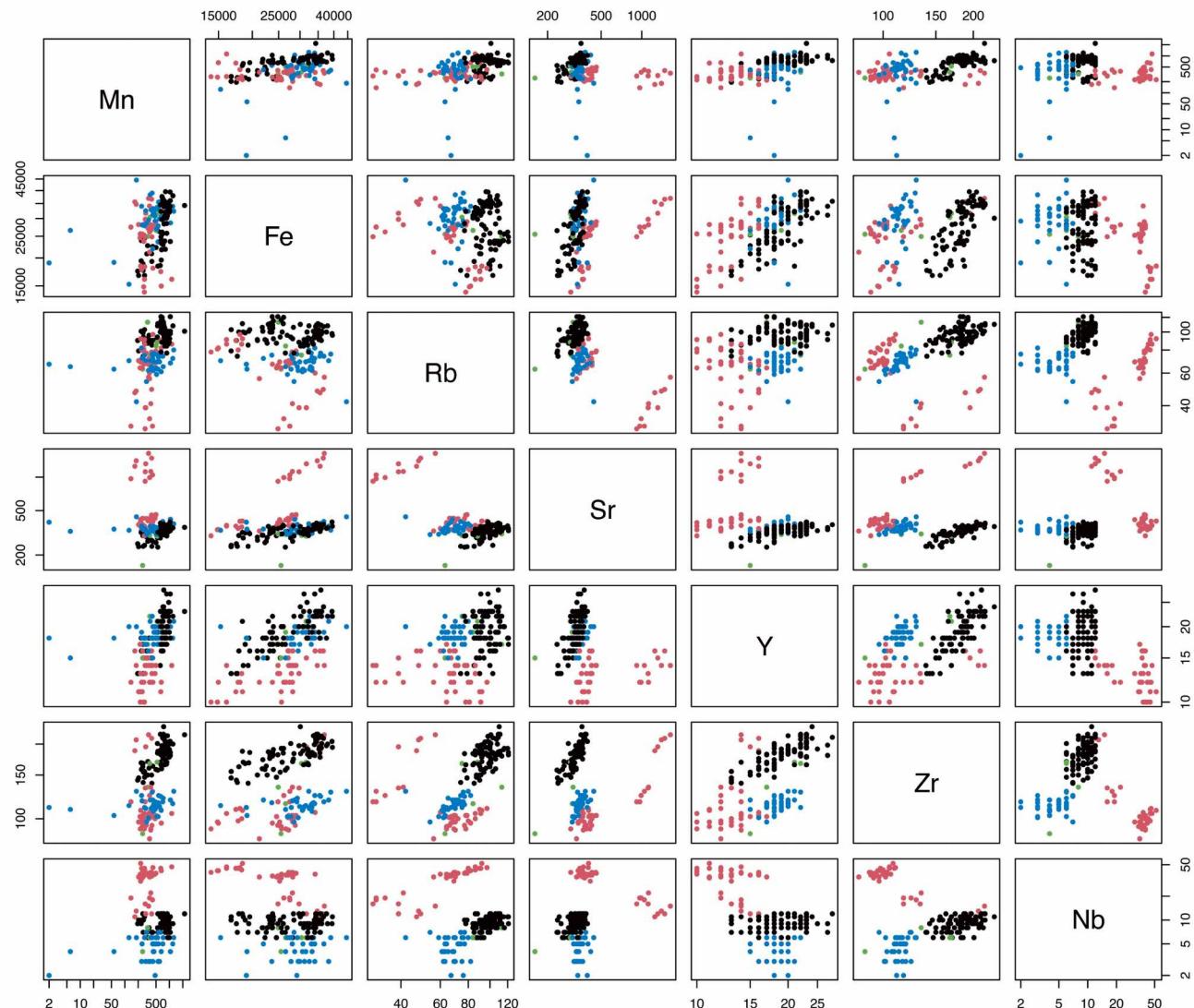


Fig. 7. Scatterplot matrix with logarithmic axes of seven elements (Mn, Fr, Rb, Sr, Y, Zr, Nb) measured on 167 geologic samples. Colored by the source groups in Table 1. Red: Kanmuri, Black: Goshikidai/Kiyama/Kanayama, Green: Awaji, Blue: Nijosan. Data source in Supplementary Information (S1). Created by R (R Core Team, 2022).

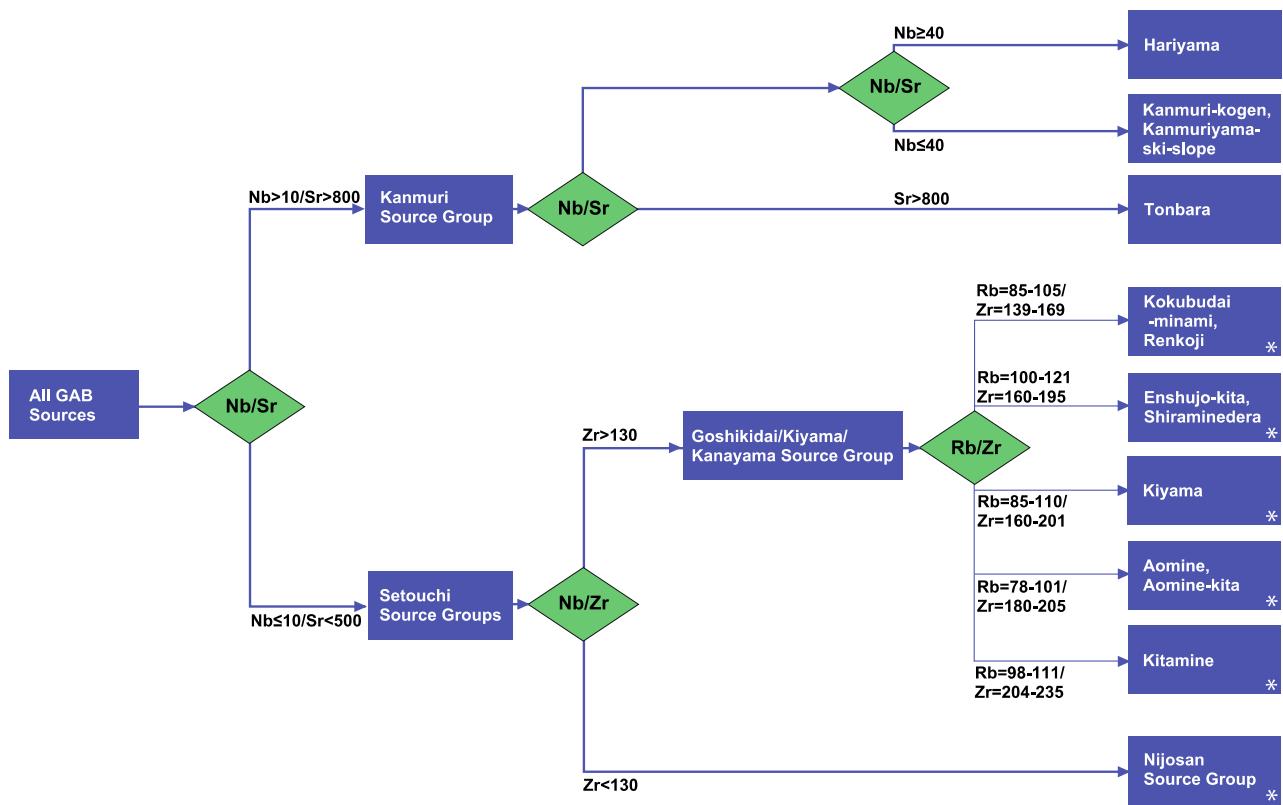


Fig. 8. A flowchart of the source discrimination procedure adopted in this paper. The threshold values are shown in ppm. Asterisked sources are identified in the secondary deposit of Matsuho-coast, possibly derived through alluvial action.

compositional data by analyzing the weathered natural rock surface, the weathered flat scar surface, and the fresh flat scars of two identical geologic samples (also see MIH16804, 16804b, 16804c, and MIH16808, 16808b, 16808c of S1). The results generated indicates that there is no significant difference between those surface conditions (Fig. 5 and 6). More detailed examination of the nature of the weathering and any potential impacts on compositional analysis are currently underway focusing on Ca and ratio of water content, but we are confident that the weathering has little, if any, impact of compositional data collected by EDXRF.

Fig. 7 is a scatterplot matrix of seven elements. Here we include Mn and Fe as well as Rb, Sr, Y, Zr, Nb. Some pairs of Sr/Nb, Zr/Nb, Sr/Zr shows tighter cluster of plots, although Mn, Fe and Y making unclear cluster seems less useful for source discrimination.

As expected from the geological literature, we found it possible to separate the currently known major source groups and some sources according to the following procedure (Fig. 8).

First, the individual sources of the Kanmuri source group are clearly distinguished from the other Setouchi source groups in a plot of Nb vs. Sr based on the scarcity of Nb (≤ 10 ppm) and lower Sr (≤ 500 ppm) in Setouchi source groups (Fig. 9: Upper). Focusing on the sub-sources of the Kanmuri source group (see Fig. 1-C), Hariyama source ($Nb \geq 40$ ppm) and Kanmuri-kogen/Kanmuriyama-ski-slope source ($Nb \leq 40$ ppm) are also basically distinguishable in this plot along with consideration of positive correlation with Sr value. Also, high Sr (> 800 ppm) concentrations in samples from the Tonbara source create clear chemical separation.

Secondly, the concentration of Zr is very useful for the separation of Setouchi source groups between the Goshikidai/Kiyama/Kanayama source group ($Zr > 130$ ppm) (Fig. 1-D) and the Nijosan source group ($Zr < 130$ ppm) (Fig. 1-E) (Fig. 9: Lower). As seen in this plot, no difference is observed between the two sources in the Nijosan group, while

sources in Goshikidai/Kiyama/Kanayama source group make several concentrations slightly separate from each other.

Fig. 9 also shows that samples from the Matsuho-coast encompass the compositional variability of the Goshikidai/Kiyama/Kanayama area and the Nijosan area, illustrating the previous supposition that GAB from Matsuho-coast represent secondary deposits transported by alluvial actions from several areas of central and eastern Setouchi during the Quaternary.

Furthermore, the third plot of Rb vs. Zr (Fig. 10) can be more effective in distinguishing the sources in the Goshikidai/Kiyama/Kanayama source group into five groups: Kokubudai-minami/Renkoji sources, Enshujo-kita/Shiraminedera sources, Kiyama source, Aomine/Aomine-kita sources, and the Kitamine source, although they are partially overlapping (see Fig. 8 for the range of Rb and Zr concentrations in each source) and the discrimination of the Kanayama-higashi source is not clear (Fig. 10). It should be noted that these five source groups are distributed apart from each other (Fig. 1-D), implying systematic difference of chemical composition of magma sources at larger geographic/geologic scales. Clear separation of sources in Goshikidai/Kiyama/Kanayama source group should need future improvement of methodology including statistical analysis.

4.2. Sourcing of the GAB artifacts from the Kanegasaki site

As shown in the scatterplot plot of Nb vs. Sr (Fig. 11: Upper), the artifact samples are grouped into three distinct clusters: A, B and C. Cluster A consists of 529 samples, Cluster B consist of 116 samples, while the minor cluster C consists of only 4 samples. 29 pieces do not consistently plot in or near the confidence ellipses of any of the geologic sources and may represent minor or distant GAB sources not included in our current study or possibly unexplained chemical variability.

The artifacts in cluster B plot within or near the confidence ellipses of

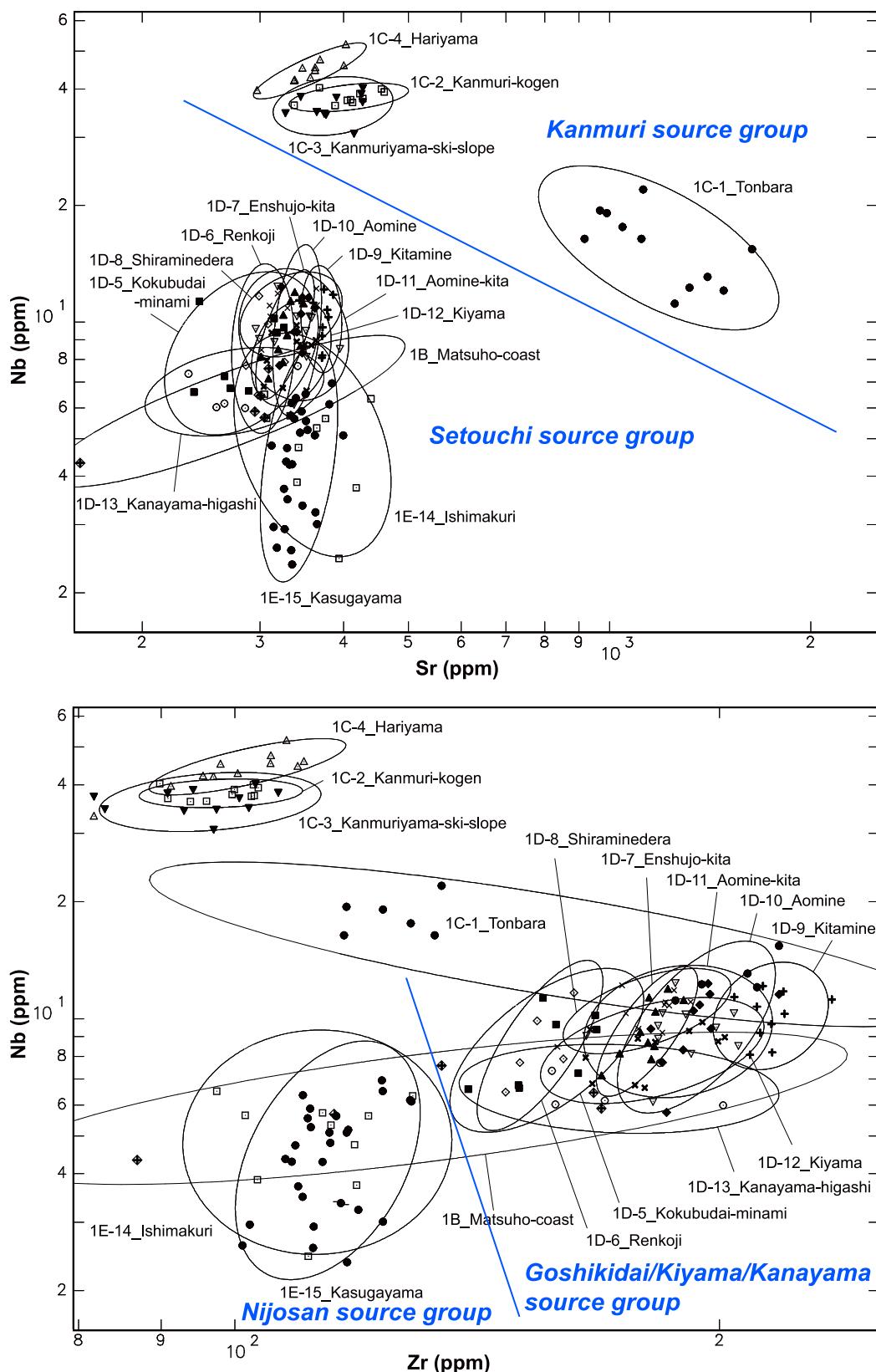


Fig. 9. Scatter plots of Nb vs. Sr (Upper) and Nb vs. Zr (Lower) concentrations from EDXRF for all geologic sources analyzed in this paper. Ellipses represent 90% confidence intervals of membership in each geologic source group. Source names are consistent with Fig. 1 and Table 1.

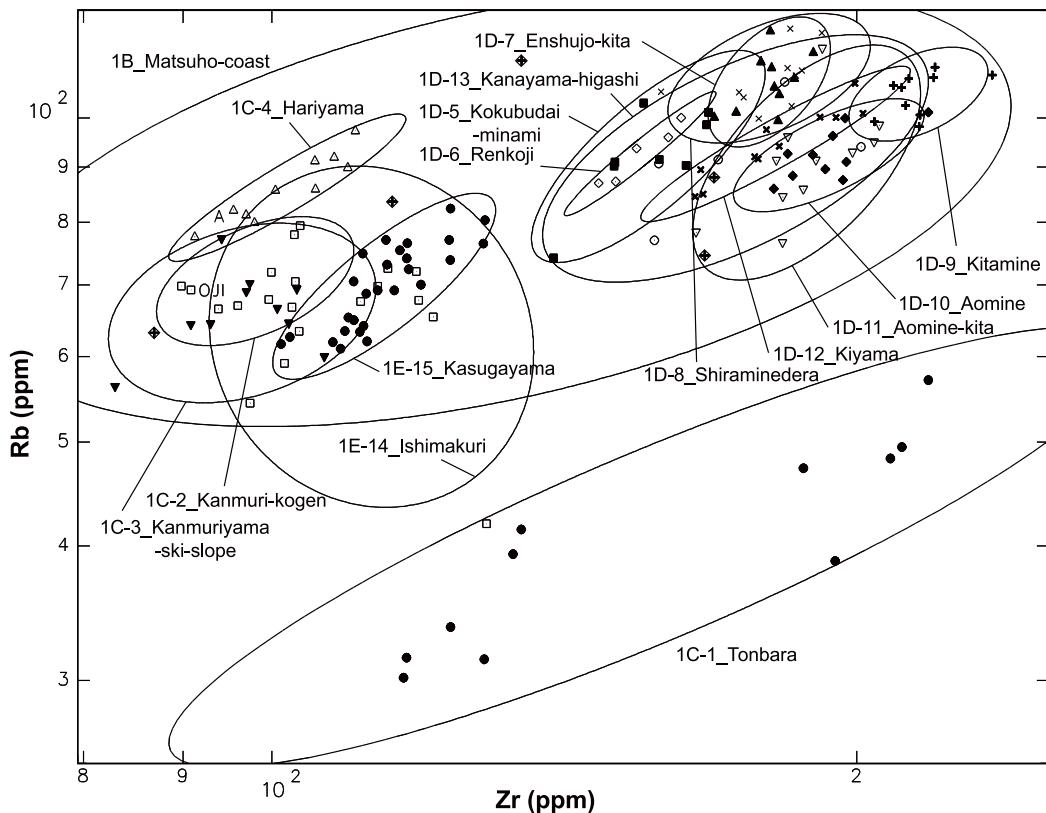


Fig. 10. Scatter plot of Rb vs. Zr (Upper) concentrations from EDXRF for all geologic sources analyzed in this paper. Ellipses represent 90% confidence intervals of membership in each geologic source group. Source names are consistent with Fig. 1 and Table 1.

the Kanmuri source group. Most of them fall within the Hariyama source, while some are within the Kanmuri-kogen/Kanmuriyama-ski-slope source. The smaller cluster C, with four artifacts, falls within the ellipse of the Tonbara source. The second plot of Nb vs. Zr (Fig. 11: Lower) reveals that the major cluster A is plotted not in the Nijosan source groups but around the ellipses of the Goshikidai/Kiyama/Kanayama source group.

Based on the procedure for the separation of geologic sources, a plot of Rb vs. Zr is more effective to breakdown Goshikidai/Kiyama/Kanayama source group into sub-sources (Fig. 12). It shows that they form two elongated clusters a1 and a2, which respectively fall around the Enshujo-kita/Shiraminedera sources and the Kiyama source. Considering this pattern, most of them would not be assigned to the other sources.

Table 3 concisely summarizes the assemblage composition in number and the result of geologic sourcing. Detailed chemical compositional data is available in the [Supplementary Information \(S2\)](#).

5. Discussion and conclusion

Given the geological knowledge, trace elements that are widely useful for obsidian sourcing are shown to be applicable also to GAB sourcing with non-destructive EDXRF. Specifically, Nb concentrations are quite effective for discriminating monogenetic GAB outside the Setouchi region from HMAs of the Setouchi volcanic belt. Furthermore, GAB sources from the Setouchi volcanic belt can be distinguished using Zr, Sr, and Rb concentrations, while at the same time we realized that the more robust discrimination of the Goshikidai/Kiyama/Kanayama source group should need future improvement of methodology including statistical analysis.

The GAB source of lithic tools from the Kanegasaki site is

demonstrated to consist mostly of the Goshikidai/Kiyama/Kanayama source group, and partly of Kanmuri source group. Moreover, our preliminary presumption suggests that the most used sub-sources are the Enshujo-kita/Shiraminedera sources and Kiyama source, constituting the western part of the source group. The Kanegasaki site is located far from both Goshikidai/Kiyama/Kanayama area and Kanmuri area, while lithics made of GAB sources other than the Goshikidai/Kiyama/Kanayama group are limited to a minor proportion.

Given the compositional data, one possible explanation is that the occupants of the Kanegasaki site were foragers primarily inhabiting the flat river basin stretching eastward from the site. In the meantime, the lithic assemblage of the GAB from the Kanmuri source group is characterized by almost the same tool types and toolkit composition. This implies that the foraging radius of the occupants of Kanegasaki site encompassed the Kanmuri area to the west. While at the same time, it is equally possible that another local group mainly inhabiting the area stretching westward from the site including the Kanmuri also occupied the site. Furthermore, considering the inter-social network, the possibility remains to be considered that GAB artifacts of the Kanmuri source group were acquired through exchange networks. All of these may combine to explain the source patterns observed. Our limited understanding of raw material procurement and transportation cannot determine which possibility is more likely because of the current lack knowledge of the lithic reduction strategy and submerged landscape. Therefore, future studies will also focus on clarifying the lithic reduction stage of each site based on the toolkit composition and on reconstructing sea bottom topography of the present Seto Inland Sea by using geographic information system, and these will allow us to further refine the procurement model as the sourced artifact database expands.

In sum, our regional case study preliminarily evidenced the effectiveness of non-destructive EDXRF geochemical sourcing for major GAB

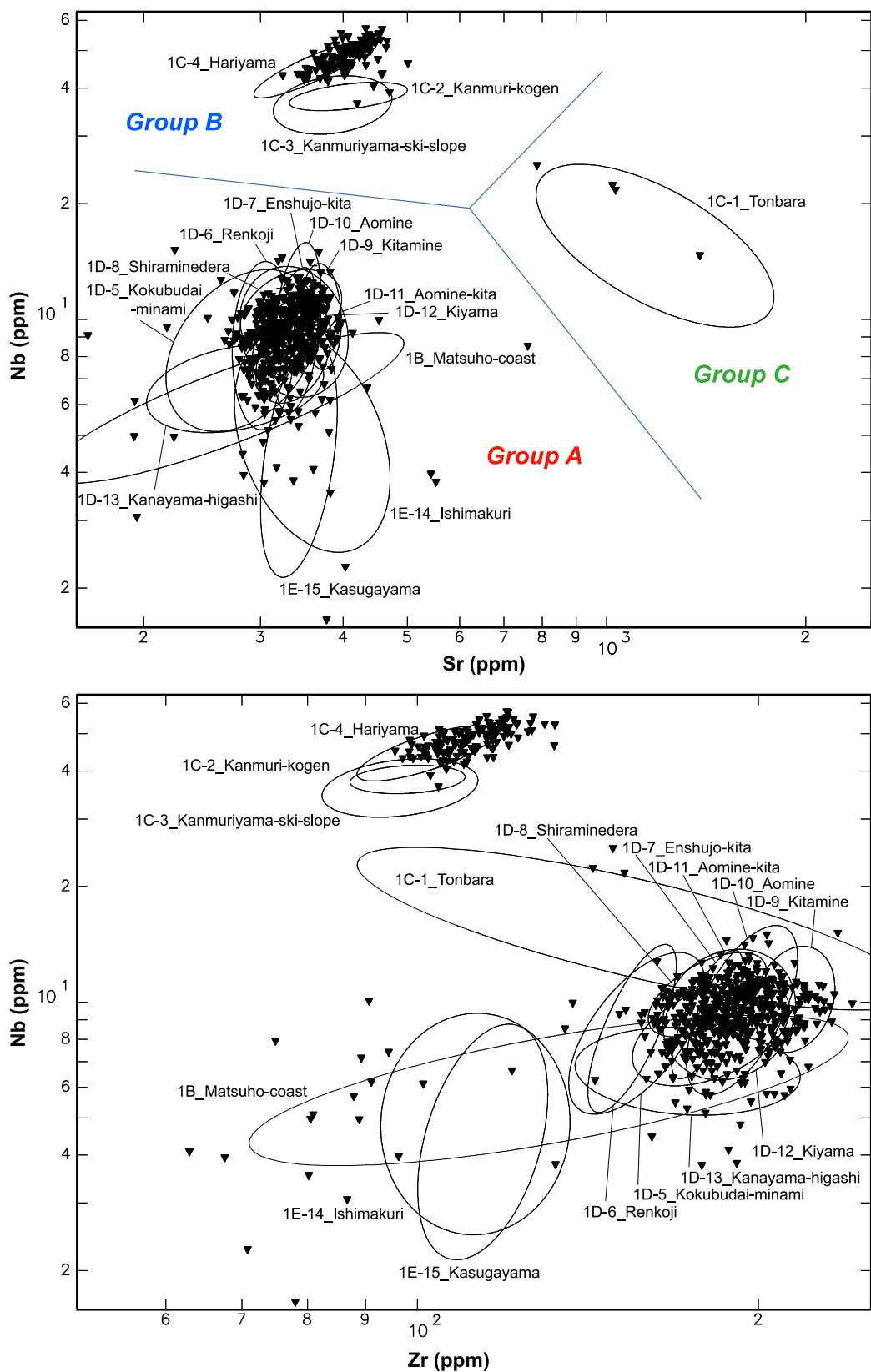


Fig. 11. Scatter plots of Nb vs. Sr (upper) and Nb vs. Zr (lower) concentrations from EDXRF for 678 archaeological samples. Ellipses represent 90% confidence intervals of membership in each geologic source group. Source names are consistent with Fig. 1 and Table 1.

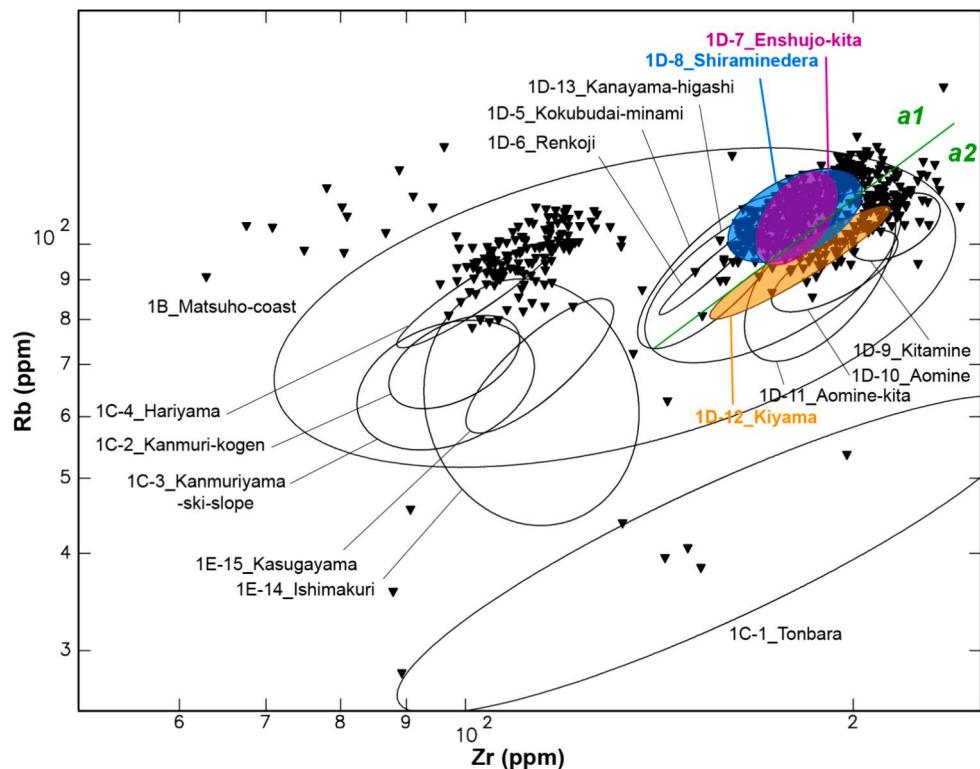


Fig. 12. Scatter plot of Rb vs. Zr concentrations from EDXRF for 678 archaeological samples. Ellipses represent 90% confidence intervals of membership in each geologic source group. Source names are consistent with Fig. 1 and Table 1.

Table 3

The concise summary of the source discrimination result for 678 GAB artifacts from the Kanegasaki site. Refer to Supplementary Information (S2) for details.

Toolkit	Goshikidai/ Kiyama/ Kanayama	Kanmuri		Unknown	TOTAL
		Kanmuri-kogen/ Kanmuriyama-ski-slope	Hariyama		
Backed point	56		15	1	73
Backed piece	10		2	2	14
Basal-retouched point	2				2
Tri-facial point	9		3	1	13
Unifacial point			1		1
Side scraper	15		2	1	18
End scraper			1		1
Drill	2		1		3
Truncation	1				1
Notched piece	1				1
Biface	1				1
Chipped Arrowhead	6			1	7
Wedge	3				3
Trapezoid	1				1
Spall	1			1	2
Core	41	1	5	1	49
Blade	4		2		6
Flake	347	4	73	2	445
Retouched blade	1		1		2
Retouched flake	28		5	1	34
Pebble				1	1
TOTAL	529	5	111	4	678

geologic sources and weathered artifacts in the Setouchi region. Although improvement of statistic methodology and the implementation of the whole-rock chemical composition of geological GAB by NAA is now ongoing, our result strongly suggests that future accumulation of geochemical data of GAB could drastically expand the knowledge on the forager's exploiting radius, the settlement pattern, the subsistence strategy, and the social network not only during the Upper Paleolithic but also throughout the later periods.

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CRediT authorship contribution statement

Kazuki Morisaki: Writing – review & editing, Writing – original

draft, Visualization, Project administration, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Masami Izuho:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation. **Jeffrey R. Ferguson:** Writing – original draft, Visualization, Software, Methodology.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jasrep.2024.104589>.

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