Identification of the place of origin producing earthenware pottery based on chemical analysis of volcanic glass contained in ceramic paste

― Example for Usulután-type pottery in Chalchuapa, El Salvador, Central America ―

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1. Introduction

For the investigation to identify the origin of pottery production, mineralogical and chemical analysis techniques have been used in general. Mineralogical component is demonstrated by identifying and counting rock-forming minerals in ceramic paste under polarized microscope and calculating their ratio from the counts. X-ray diffraction analysis provides a good approximation of the original clay mineral composition that forms ceramic paste. Florescent X-ray analysis of the chemical elements of pottery can detect the original elements of the clay body regardless of whether the firing temperature is low or high, because it is relatively unaffected by the heat of firing.

These analyses are very helpful to identify the origin of the pottery production if the place of production is known because the earth of the place can be compared with the paste of earthenware by these analyses. However, if the place of production is not known and the property of earth candidate is not so unique, these analyses just point out similarity between the earthenware and the candidate earth materials, may not specify the place or area where the earth was produced as the material of the earthenware.

Moreover, in the case of earthenware, the ceramic is baked in lower temperature, so that the internal structure of the earthenware paste would not be homogenous, nor that sand-size material would not be melted or mixed together with clay material in earthenware paste. Although florescent X-ray analysis can detect “average” chemical composition and is appropriate for analysis of homogenous ceramic by melting, it may not be proper in cases for the ceramic of earthenware, which is heterogenous in many cases, and often added exogenous sandy grains to the main clayey material in various proportion.

In volcanic area, volcanic ash is one of general materials commonly used to produce pottery as sandy material for adjusting the hardness of the clay to make potteries, and then the ceramic paste commonly contains volcanic ash which yield in the area. Volcanic ash shows unique properties by each eruption, especially in the chemical composition of volcanic glass contained in volcanic ash. The areal distribution of a volcanic ash is restricted with in a range from the volcano of the origin. The unique chemical property and restricted areal distribution are quite valuable to identify the pottery production area and to consider the transport of pottery or the propagation of ceramic style from the place of production.

For this study, it is essential that the database or the catalogue of pyroclastic layers called “tephras” yielding in the area are previously obtained and that their areal distributions and the chemical properties of volcanic ash contained in them are also clarified.

Northern Central America is one of the area suitable for the study, because many volcanoes range along the pacific coast and the stratigraphy and the

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distribution of volcanic ash in the late Pleistocene has been demonstrated by volcanological and tephrochronological studies for more than fifty years. Especially in El Salvador, chemical composition of volcanic glass from each tephra was previously analyzed quantitatively with a wave-length-dispersive electron microprobe analyzer (WDS).

In the area from southern Mexico to Costa Rica, a remarkable Pre-columbian pottery style, called "Usulután" (photo 1), has been discovered. It has been believed to be derived from El Salvador because it was first discovered in eastern El Salvador. It was demonstrated to have been produced since the late Preclassic period until the early Classic period (B.C. 400 to A.D. 600). The spatially broad distribution and the temporally long continuity of the pottery style are very helpful to understand the background on production and transportation of potteries at that time and to consider the social relationship among the distant regions in the past. Therefore, identification of the origin or the place of production of Usulután-style pottery, which is valuable for the mesoamerican archaeology as described above, is possible in El Salvador by the comparison of ceramic paste with tephra in the analyses of chemical composition of volcanic glass contained in them.

This study aims at identifying the production area of two ceramic pieces of the Usulután-style pottery discovered in Chalchuapa, western El Salvador, by the chemical analysis of volcanic glass contained in the pottery paste using WDS, as the first step of the study.

Photo 1 A typical piece of the Usulután-style pottery (photograph by M. Murano)

2. Previous studies

1) Tephras in Northern Central America

A volcanic chain extends along the pacific coast of Central America from the Tacaná Volcano, Guatemala, to Irazú Volcano, Costa Rica, through southern Guatemala and El Salvador, on the E-W tensional field between the Motagua-Polochic transform fault and marine trench on the subduction zone of the Cocos Plate. In the area, many silicic centers which broadly dispersed pyroclastic material called "tephra" are known. Well-known five large calderas, called Atitlán, Amatitlán, Ayarza, Coatepeque and Ilopango are situated in the northern part of Central America (Fig. 1). A volcanic area, where various volcanoes are concentrated, is located on the eastern El Salvador, and is called Berlín-Pacayal volcanic area.

The largest eruption in the late Pleistocene occurred at Atitlán Caldera, southern Guatemala in 84 ka (Drexler *et al.*, 1980; Rose *et al.* (1981); Rose, *et al.*, 1999). Dispersed pumice and fine ash called Los Chocoyos, or H tephra (Koch and McLean, 1975; Hahn, *et al.*, 1979; Rose, *et al.*, 1979) broadly covers Central America and southern Mexico and the sea floors in the Pacific Ocean and Carribean sea (Fig. 2). At the Coatepeaque Caldera in the western El Salvador, huge silicic eruption occurred four times from 77 ka to 51 ka, and the products associated with the eruptions are called Bellavista, Arce, Congo and Conacaste from the lower to the upper (CEL, 1992). While the Arce tephra consists of three pumice-fall units, the others contain the pumice-flow component in addition to the pumice-fall units. Four tephras from Ilopango Caldera are called TB4, TB3, TB2 and TBJ, from the lower to the upper (CEL, 1992; Hernández, 2004). The TB4 tephra is fall-out pumice, the TB3 and the TB2 tephra are ash-fall deposit, and the TBJ tephra consists of multiple units of pumice fall, ash fall, ignimbrite and co-ignimbrite ash produced by phreatomagmatic and magmatic eruption (Hart and McIntyre, 1987; Hernández, 2004). Three tephras except the TB3 tephra are known to cover the western El Salvador (Kitamura, 2016). The eruptive ages of the TB4 and the TBJ tephra were determined as 36 ka and the 4^{th} to 6^{th} century, respectively (Kutterolf, *et al.*, 2008; Dull, 2001; Dull, *et al.*,2010; Kitamura, 2010; Smith, *et al.*, 2019). Eleven pumice-fall deposits are known in the Berlín-Pacayal

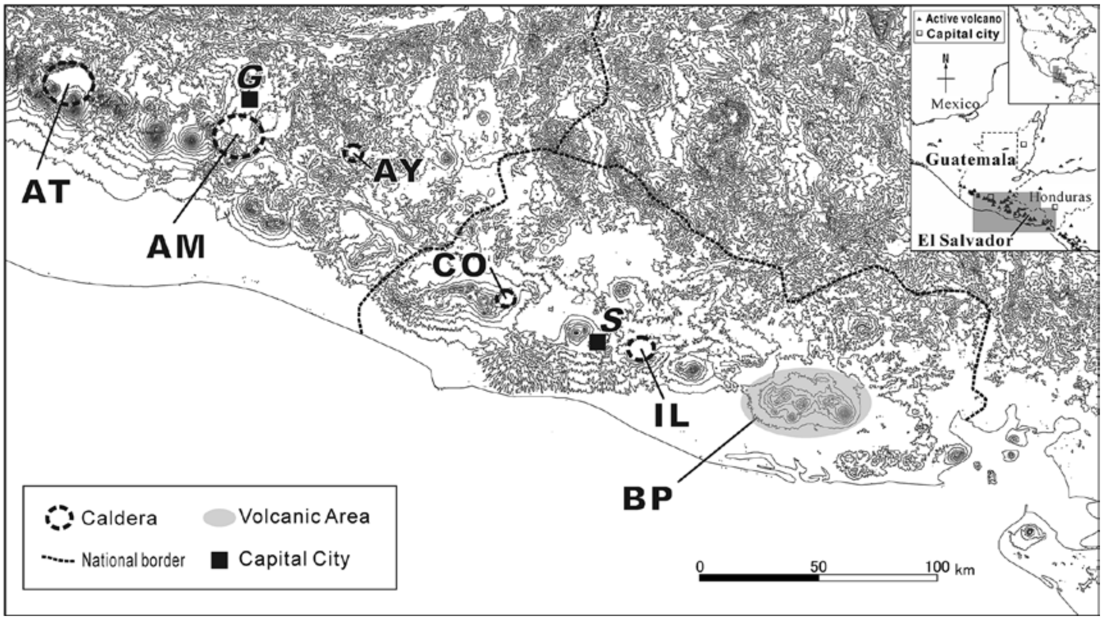

Fig. 1 Five Calderas in southern Guatemala and El Salvador, northern Central America

Contour was created using 7.5-arc-second DEM data from GMTED2000 (Danielson &Gesch, 2011), and GSHHG vector data (Ver. 2.3.6; August 19, 2016) distributed by National Oceanic and Atmospheric Administration (NOAA), U. S. Department of Commerce, was used for drawing coastlines. The contour interval is 200 m. AT: Atitlán, AM: Amatitlán, AY: Ayarza, CO: Coatepeque, IL: Ilopango, BP:Berlín-Pacayal, G: Guatemala City, S: San Salvador City

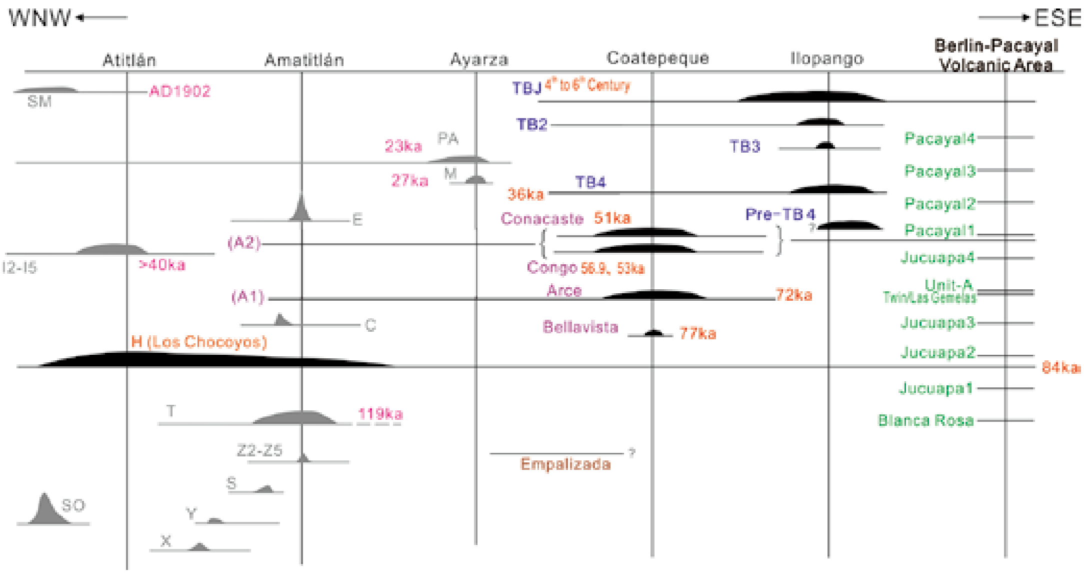

Fig. 2 Stratigraphic relationship of tephras in southern Guatemala and El Salvador

(Deformed from Rose *et al*., 1999)

Approximate extent of pyroclastic air-fall deposit is shown by horizontal solid line and the extent of pyroclastic-flow deposit is shown by filled-pattern. Faint color shows pyroclastic deposit which is not discussed in this paper.

volcanic area as shown in figure 2. Eruptions producing the Blanca Rosa tephra, Twin, Pacayal 1 and Pacayal 4 are assumed to be relatively larger, because they has been discovered in boring core at the sea floor of the Pacific Ocean (Kutterolf, *et al*. 2008). Although their eruptive ages are not determined yet, according to the stratigraphic relationship, the Jucuapa 1 tephra and the Blanca Rosa tephra are older than the Los Chocoyos tephra, and the Pacayal 1 tephra is younger than the Congo tephra or the Conacaste tephra (Kitamura, 2018).

The chemical composition of volcanic glass of the tephras mentioned above were previously illustrated by the analyses using a wave-length-dispersive electron microprobe analyzer (WDS), as shown in tables from 1-a to 1-e (Kitamura, 2006; Kitamura, 2016; Kitamura, 2017; Kitamura, 2018; Kitamura, 2019), and most of tephras are uniquely identifiable based on the chemical properties. Tephras from large calderas occupy their unique range in the FeO-CaO and the FeO-K_2O diagrams (Fig. 3-a).

The Los Chocoyos tephra from Atitlán Caldera in Guatemala shows unique properties, and in FeO-CaO and FeO-K_2O diagrams its dots are concentrated in the independent range from any other tephras discussed in this article, showing a trend that the CaO is rather

Table 1-a Chemical composition of volcanic glass of the Los Chocoyos tephra from Atitlán Caldera in Southern Guatemala
The numbers with parentheses show the quantity of analyzed volcanic glass shards.

tephra	sampling site	deposit	Sample No.		SiO_2 (%)	TiO_2 (%)	Al_2O_3 (%)	FeO (%)	MnO (%)	MgO (%)	CaO (%)	K_2O (%)	Na_2O (%)	Total (%)	Publication
Los Chocoyos (H tephra)	southern Guatemala	Pumice fall deposit	1	Average	77.8	0.1	12.4	0.5	0.1	0.1	0.6	4.5	3.9	100.0	Kitamura (2018)
				St. Dev.	0.4	0.1	0.2	0.1	0.0	0.0	0.1	0.2	0.4	(10)	
		pyroclastic-flow deposit	2	Average	77.8	0.1	12.5	0.5	0.1	0.1	0.6	4.5	3.9	100.0	Kitamura (2018)
				St. Dev.	0.3	0.1	0.2	0.1	0.1	0.0	0.1	0.3	0.2	(30)	
			3	Average	77.6	0.1	12.5	0.5	0.1	0.1	0.6	4.5	3.9	100.0	Kitamura (2018)
				St. Dev.	0.4	0.1	0.2	0.1	0.1	0.0	0.1	0.2	0.2	(42)	
			4	Average	77.7	0.1	12.4	0.6	0.1	0.1	0.6	4.5	4.0	100.0	Kitamura (2018)
				St. Dev.	0.2	0.1	0.2	0.1	0.1	0.0	0.1	0.2	0.2	(40)	
		co-ignimbrite ash	5	Average	77.8	0.1	12.2	0.5	0.1	0.1	0.6	4.6	4.0	100.0	Kitamura (2018)
				St. Dev.	0.3	0.1	0.2	0.1	0.1	0.0	0.1	0.2	0.2	(29)	
			6	Average	77.8	0.1	12.2	0.5	0.1	0.1	0.6	4.5	4.0	100.0	Kitamura (2018)
				St. Dev.	0.3	0.0	0.2	0.1	0.1	0.0	0.1	0.2	0.2	(30)	

Table 1-b Chemical composition of volcanic glass of the tephras from Coatepeque Caldera in Western El Salvador

tephra	sampling site	deposit	Sample No.		SiO_2 (%)	TiO_2 (%)	Al_2O_3 (%)	FeO (%)	MnO (%)	MgO (%)	CaO (%)	K_2O (%)	Na_2O (%)	Total (%)	Publication
Bellavista	western El Salvador	pumice fall	7	Average	77.3	0.1	12.2	1.1	0.1	0.0	0.5	5.3	3.5	100.0	Kitamura (2018)
				St. Dev.	0.3	0.1	0.3	0.2	0.1	0.0	0.1	0.2	0.2	(29)	
		pyroclastic flow	8	Average	77.1	0.0	12.2	1.2	0.1	0.0	0.5	5.4	3.5	100.0	Kitamura (2018)
				St. Dev.	0.4	0.0	0.3	0.2	0.1	0.0	0.1	0.2	0.2	(30)	
Arce	southern Guatemala	vitric ash	9	Average	73.6	0.1	13.8	1.7	0.1	0.1	0.9	5.3	4.4	100.0	unpublished
				St. Dev.	2.4	0.1	1.5	0.5	0.1	0.1	0.3	1.0	0.5	(28)	
			10	Average	74.3	0.0	13.7	1.3	0.1	0.0	0.7	5.6	4.3	100.0	Kitamura (2006)
				St. Dev.	2.4	0.0	1.5	0.3	0.1	0.0	0.2	0.9	0.4	(26)	
			11	Average	73.4	0.1	14.6	1.2	0.1	0.1	0.8	5.7	4.2	100.0	unpublished
				St. Dev.	2.9	0.1	1.9	0.4	0.1	0.1	0.2	0.7	0.5	(28)	
	western El Salvador	pumice fall (lower unit)	12	Average	76.6	0.1	12.6	1.2	0.1	0.0	0.6	4.7	4.1	100.0	Kitamura (2017)
				St. Dev.	1.0	0.1	0.5	0.2	0.0	0.0	0.2	0.2	0.3	(39)	
		pumice fall (middle unit)	13	Average	74.9	0.1	13.6	1.5	0.1	0.1	0.8	4.7	4.3	100.0	Kitamura (2017)
				St. Dev.	1.5	0.1	0.8	0.3	0.1	0.1	0.2	0.2	0.3	(40)	
		pumice fall (upper)	14	Average	74.6	0.1	13.8	1.2	0.1	0.0	0.9	5.0	4.3	100.0	Kitamura (2017)
				St. Dev.	3.0	0.0	2.2	0.3	0.0	0.0	0.9	0.8	0.8	(39)	
		pumice fall	15	Average	74.2	0.1	13.5	1.4	0.1	0.0	0.8	5.7	4.3	100.0	added supplementing data to Kitamura (2006)
				St. Dev.	3.0	0.1	2.0	0.3	0.1	0.0	0.2	1.1	0.7	(30)	
	central El Salvador	pumice fall	16	Average	72.7	0.1	14.3	1.8	0.1	0.1	1.0	5.3	4.6	100.0	Kitamura (2017)
				St. Dev.	1.5	0.1	0.8	0.3	0.1	0.1	0.2	0.2	0.3	(35)	
Congo	western El Salvador	pumice fall	17	Average	74.0	0.1	13.7	1.8	0.1	0.2	1.1	4.7	4.2	100.0	Kitamura (2006)
				St. Dev.	0.7	0.1	0.5	0.2	0.1	0.1	0.2	0.4	0.4	(26)	
		pyroclastic flow	18	Average	73.3	0.1	14.4	1.7	0.1	0.1	1.1	5.1	4.2	100.0	Kitamura (2018)
				St. Dev.	0.4	0.1	0.2	0.2	0.1	0.0	0.1	0.2	0.3	(13)	
Conacaste	western El Salvador	pumice fall	19	Average	74.0	0.1	13.8	1.8	0.1	0.2	1.1	4.6	4.3	100.0	Kitamura (2006)
				St. Dev.	1.2	0.1	0.7	0.2	0.1	0.1	0.2	0.2	0.4	(23)	
		pyroclastic flow	20	Average	73.5	0.2	14.1	1.9	0.1	0.2	1.2	4.9	3.9	100.0	Kitamura (2018)
				St. Dev.	0.4	0.1	0.3	0.2	0.1	0.0	0.1	0.2	0.2	(16)	

Table 1-c Chemical composition of volcanic glass of the tephras from Ilopango Caldera in Central El Salvador

tephra	sampling site	deposit	Sample No.		SiO_2 (%)	TiO_2 (%)	Al_2O_3 (%)	FeO (%)	MnO (%)	MgO (%)	CaO (%)	K_2O (%)	Na_2O (%)	Total (%)	Publication
TB4	central El Salvador	pumice fall	21	Average	77.1	0.2	12.8	1.1	0.1	0.2	1.2	2.8	4.6	100.0	unpublished
				St. Dev.	0.6	0.1	0.3	0.1	0.1	0.1	0.1	0.1	0.2	(32)	
		pumice fall	22	Average	77.3	0.1	12.8	1.0	0.1	0.2	1.1	2.7	4.6	100.0	unpublished
				St. Dev.	0.6	0.1	0.4	0.1	0.1	0.0	0.2	0.1	0.2	(34)	
TB3	central El Salvador	pumice fall	23	Average	76.9	0.2	12.8	1.2	0.1	0.2	1.4	3.0	4.2	100.0	Kitamura (2016)
				St. Dev.	0.5	0.1	0.3	0.2	0.0	0.1	0.2	0.1	0.2	(28)	
		pumice fall	24	Average	77.2	0.2	12.8	1.1	0.1	0.2	1.3	3.0	4.2	100.0	Kitamura (2016)
				St. Dev.	0.5	0.1	0.2	0.2	0.1	0.1	0.1	0.1	0.3	(27)	
TB2	central El Salvador	pumice fall	25	Average	76.3	0.3	13.0	1.4	0.1	0.3	1.5	2.9	4.1	100.0	Kitamura (2016)
				St. Dev.	1.2	0.1	0.6	0.3	0.1	0.1	0.3	0.3	0.6	(26)	
		pumice fall	26	Average	76.4	0.2	13.0	1.3	0.1	0.2	1.4	2.9	4.4	100.0	Kitamura (2016)
				St. Dev.	0.7	0.1	0.4	0.2	0.1	0.1	0.2	0.2	0.2	(20)	
	western El Salvador	vitric ash	27	Average	76.5	0.2	13.0	1.3	0.1	0.3	1.4	2.9	4.4	100.0	Kitamura (2016)
				Std. dev.	0.9	0.1	0.7	0.2	0.0	0.1	0.3	0.4	0.2	(38)	
TBJ	central El Salvador	co-ignimbrite ash	28	Average	77.2	0.2	12.9	1.2	0.1	0.2	1.2	3.1	3.9	100.0	Kitamura (2016)
				St. Dev.	1.5	0.1	1.2	0.2	0.1	0.1	0.4	0.3	0.5	(29)	
		pyroclastic flow	29	Average	77.0	0.2	12.9	1.1	0.1	0.2	1.2	3.0	4.3	100.0	Kitamura (2016)
				St. Dev.	1.4	0.1	1.0	0.2	0.1	0.1	0.4	0.3	0.4	(21)	
		ash fall	30	Average	77.4	0.2	12.8	1.2	0.1	0.2	1.2	3.0	4.0	100.0	Kitamura (2016)
				St. Dev.	0.3	0.1	0.3	0.1	0.1	0.1	0.1	0.1	0.3	(27)	

Table 1-d Chemical composition of volcanic glass of the tephras from Berlin-Pacayal Volcanic Area in Eastern El Salvador

tephra	sampling site	deposit	sample No.		SiO_2 (%)	TiO_2 (%)	Al_2O_3 (%)	FeO (%)	MnO (%)	MgO (%)	CaO (%)	K_2O (%)	Na_2O (%)	Total (%)	Publication
Blanca Rosa	eastern El Salvador	pumice fall	31	Average	69.2	0.5	15.3	3.3	0.2	0.7	2.4	2.9	5.5	100.0	Kitamura (2019)
				St. Dev.	0.8	0.1	0.3	0.3	0.1	0.1	0.3	0.2	0.3	(28)	
		ash fall (Upper unit)	32	Average	69.4	0.4	15.4	3.1	0.1	0.7	2.6	3.0	5.2	100.0	Kitamura (2019)
				St. Dev.	0.8	0.1	0.7	0.3	0.1	0.1	0.4	0.2	0.3	(34)	
Jucuapa 1	eastern El Salvador	pumice fall	33	Average	66.7	0.8	15.1	5.1	0.1	1.2	3.9	2.5	4.7	100.0	Kitamura (2019)
				St. Dev.	0.8	0.1	0.7	0.5	0.1	0.3	0.4	0.2	0.4	(33)	
Jucuapa 2	eastern El Salvador	pumice fall	34	Average	65.6	0.6	16.5	4.5	0.1	1.2	4.4	1.9	5.2	100.0	Kitamura (2019)
				St. Dev.	1.8	0.1	2.0	0.9	0.1	0.3	1.1	0.3	0.3	(37)	
Jucuapa 3	eastern El Salvador	pumice fall	35	Average	65.7	0.7	15.8	5.0	0.1	1.3	4.3	2.0	5.1	100.0	Kitamura (2019)
				St. Dev.	1.0	0.1	0.8	0.9	0.1	0.3	0.4	0.2	0.3	(35)	
Twins-Las Gemelas	eastern El Salvador	pumice fall (lower unit)	36	Average	69.2	0.6	14.9	3.7	0.1	0.9	3.2	2.6	4.8	100.0	Kitamura (2019)
				St. Dev.	0.4	0.1	0.2	0.3	0.1	0.1	0.1	0.1	0.3	(24)	
Jucuapa 4	eastern El Salvador	pumice fall	37	Average	69.2	0.5	15.3	3.3	0.1	0.7	2.6	3.0	5.3	100.0	Kitamura (2019)
				St. Dev.	0.5	0.1	0.4	0.3	0.1	0.1	0.2	0.2	0.2	(30)	
Pacayal 1	eastern El Salvador	pumice fall	38	Average	74.1	0.4	13.5	2.0	0.1	0.4	1.9	3.1	4.5	100.0	Kitamura (2019)
				St. Dev.	0.3	0.1	0.2	0.1	0.1	0.1	0.1	0.1	0.2	(31)	
Pacayal 2/ volcan	eastern El Salvador	pumice fall	39	Average	65.7	0.8	15.5	5.3	0.2	1.3	4.1	2.1	5.0	100.0	Kitamura (2019)
				St. Dev.	0.7	0.1	0.8	0.6	0.1	0.2	0.4	0.2	0.3	(33)	
		pumice fall	40	Average	66.8	0.8	15.0	5.1	0.1	1.2	4.0	2.1	4.9	100.0	Kitamura (2019)
				St. Dev.	0.8	0.2	1.2	0.8	0.1	0.4	0.4	0.3	0.2	(33)	
Pacayal 3	eastern El Salvador	pumice fall (lower unit)	41	Average	67.5	0.7	14.9	4.8	0.2	1.2	3.6	2.2	5.0	100.0	Kitamura (2019)
				St. Dev.	0.4	0.1	0.2	0.2	0.1	0.1	0.2	0.1	0.2	(33)	
		pumice fall (upper unit)	42	Average	68.0	0.7	14.6	4.8	0.2	1.2	3.5	2.2	4.9	100.0	Kitamura (2019)
				St. Dev.	0.7	0.1	0.6	0.3	0.1	0.2	0.4	0.2	0.3	(34)	
Pacayal 4	eastern El Salvador	pumice fall	43	Average	67.2	0.7	15.5	4.6	0.2	1.2	3.7	1.9	5.0	100.0	Kitamura (2019)
				St. Dev.	0.8	0.1	0.7	0.6	0.1	0.5	0.4	0.2	0.3	(31)	

Table 1-e Chemical composition of volcanic glass of the tephras from unknown silicic centers in El Salvador

tephra	sampling site	deposit	Sample No.		SiO_2 (%)	TiO_2 (%)	Al_2O_3 (%)	FeO (%)	MnO (%)	MgO (%)	CaO (%)	K_2O (%)	Na_2O (%)	Total (%)	Publication
Empalizada	western El Salvador	pumice fall	44	Average	70.5	0.3	15.0	2.9	0.1	0.4	1.8	3.5	5.5	100.0	unpublished
				St. Dev.	2.0	0.1	0.8	0.7	0.1	0.1	0.4	0.3	0.4	(32)	
pre-TB	central El Salvador	pyroclastic flow	45	Average	75.3	0.2	13.4	1.6	0.1	0.3	1.7	2.9	4.5	100.0	unpublished
				St. Dev.	0.3	0.1	0.2	0.1	0.1	0.1	0.1	0.1	0.3	(29)	
		pyroclastic flow	46	Average	75.1	0.2	13.5	1.6	0.1	0.3	1.6	3.0	4.6	100.0	unpublished
				St. Dev.	0.3	0.1	0.2	0.2	0.1	0.1	0.1	0.1	0.2	(32)	

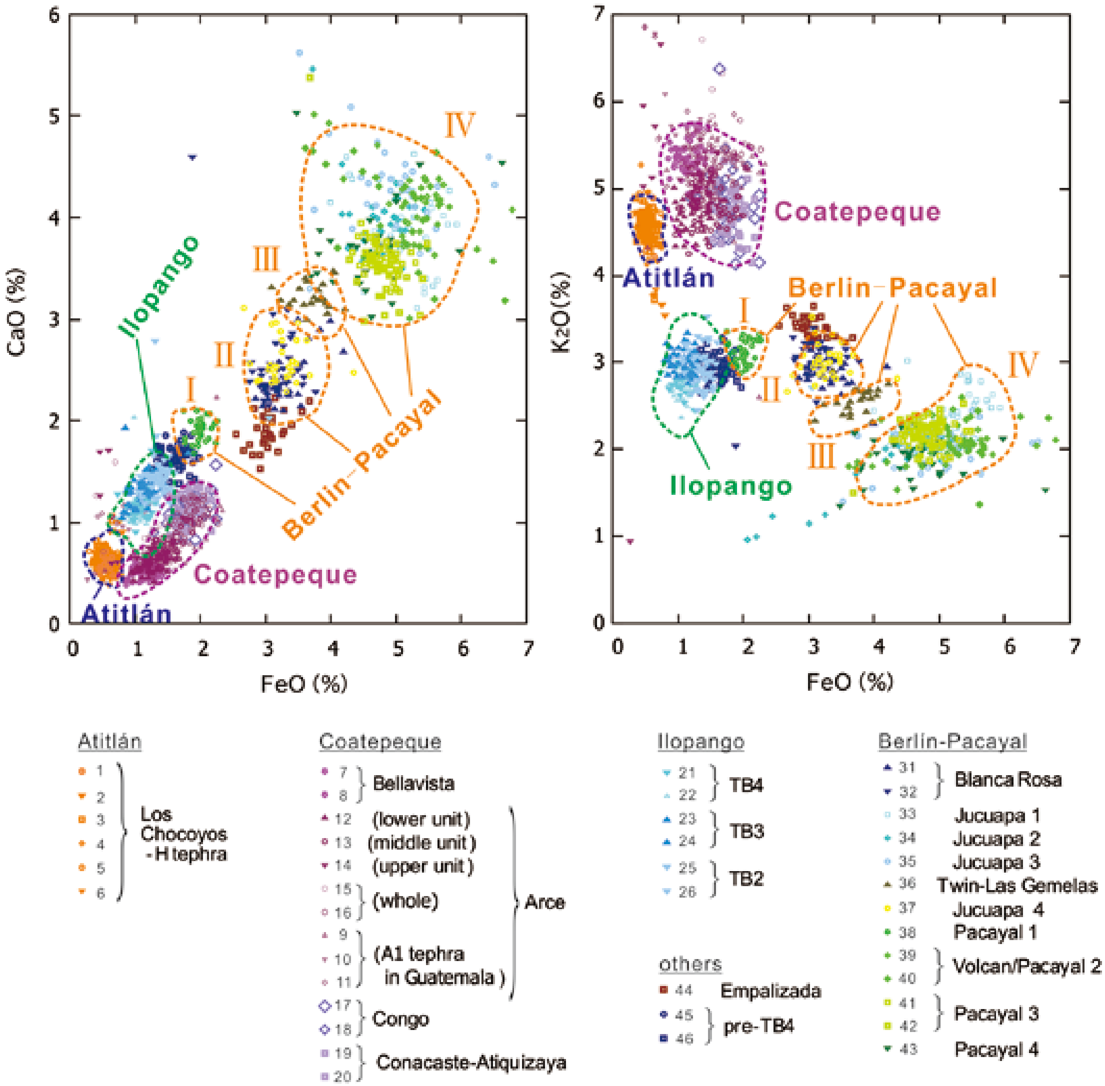

Fig.3-a Chemical composition of volcanic glass contained in tephras derived from five calderas and Berlin-Pacayal volcanic area shown in FeO-CaO and FeO-K_2O diagrams（Kitamura, 2018）.

low and K_2O is rather high and the FeO content is uniquely low. Dots of four tephras as the Bellavista, the Arce, the Congo and the Conacaste, which originate from Coatepeque Caldera located near Chaluchuapa in western El Salvador, are concentrated in a range rather low CaO and high K_2O, and the FeO content obviously higher than that of the Los Chocoyos. Of the four tephras, Bellavista is plotted in a range with lower FeO, lower CaO, and higher K_2O, by which it can be distinguished from the Congo and the Conacaste with relatively higher content of FeO and higher content of CaO and lower content of K_2O, while the chemical properties of the Congo and the Conacate are so similar that it is difficult to distinguish the one from the other. The chemical component of the Arce is plotted in a broad range overlapping the ranges of other three tephras, and has also dispersal component. The K_2O content of the TB4, the TB3, the TB2 and the TBJ tephras from Ilopango Caldera in Central El Salvador, is obviously lower than that of the tephras derived from Atitlán Caldera and Coatepeque Caldera, while dots of the four tephras are concentrated in a range of higher CaO. 11 tephras in Berlín-Pacayal Volcanic area were chemically classified to four groups (CEL, 1995; Kitamura, 2019). All of them are lower in the K_2O content than the tephras form Atitlán Caldera and the Coatepeque Caldera, and they show a trend as higher FeO and higher CaO than other tephras. Two tephras from unknown origins called the Empalizada and pre-TB tephras are added in this study. In the FeO-CaO diagram, the pre-TB tephra occupies a range neighbor to the range of the tephras from Ilopango, while the chemistry of the Empalizada tephra is plotted in a unique range.

2) Usulután-style pottery

Usulután-style pottery is one of the Pre-Columbian ceramics distributed broadly in southern Mexico and Central America. It is characterized by its orange color, design of parallel, geometric or wavy lines that can be observe on the surface of earthenware (photo 1) . Its name is given in reference to the Department of Usulután, where the specimens of its style were identified at the first time and its origin has been considered to be in El Salvador. This style appeared during the late Preclassic period and survived until the early Classic period (B.C. 400 to A.D. 600). Although examples of this ceramic style can be found at the archaeological sites in Mexico, Guatemala, Honduras, part of Nicaragua, even in Costa Rica, the investigation at present records the largest deposits of this ceramic in the actual territory of El Salvador. As a result, it became to be considered that the influence of decorative technique expanded in all Central America during the Preclassic and the Classic periods and El Salvador was one of the most important origins. Such the temporally long continuity and spatially broad distribution are very helpful to obtain background information on production and transportation of this ceramic and to consider the social relationship among the distant regions (Murano, 2017).

3. Method of Analysis

1) Ceramic sample collection

The archaeological site of Chalchuapa is located at Chalchuapa City, the Department of Santa Ana, western El Salvador. It comprises 11 zones, called Tazumal, Casa Blanca, Laguna Seca, Las Victorias, Peñate, Laguna Cuzcachapa, El Trapiche, Pampe, Los Gavilanes, Nuevo Tazumal and La Cuchilla.

Several potteries and abundant earthenware pieces were excavated by the archaeological investigation in 2003 to 2005 at the eastern entrance of Chalchuapa City which belongs to La Cuchilla zone. Many earthenware pieces of them were identified as Usulután style.

From the Usulután-style earthenware pieces collected in the investigation, typical some pieces were sampled and brought to Japan by legal legitimate procedure for several analyses, and two pieces were served for this study (photo 2).

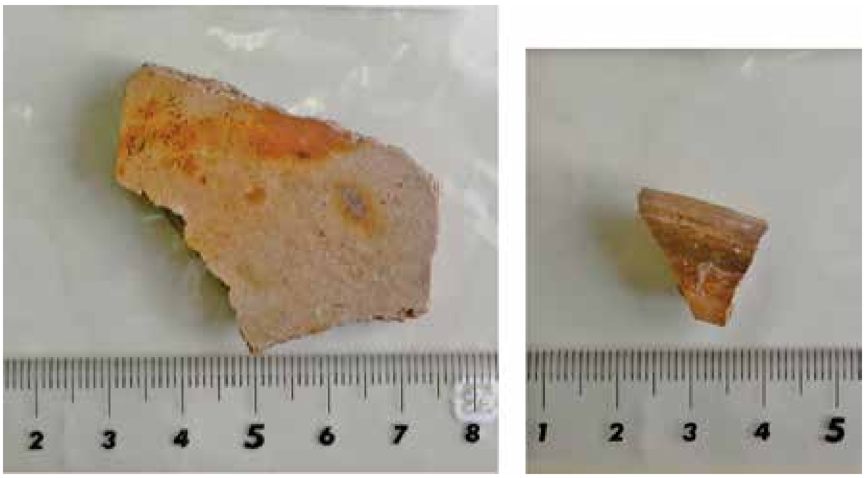

Photo 2 Two samples of the Usulután-style earthenware served for the analysis in this study

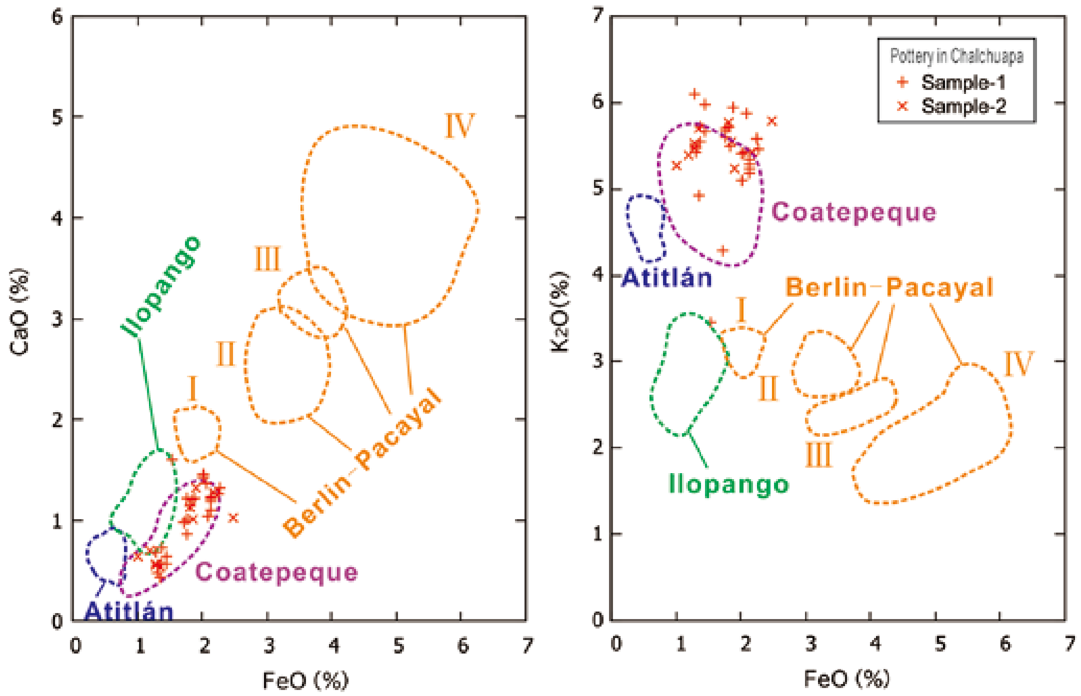

Fig. 3-b Chemical composition of volcanic glass contained in earthenware paste and comparison with tephras.

2) Chemical analysis of volcanic glass and identification of original tephra

The thin section was made from the ceramic piece by cutting earthenware along the vertical direction to the surface, polishing one side of the cross section, supporting and mounting the polished side on slide glass by epoxy resin, and polishing the cut section of the other side in mirror finish. After that, it was served for polarizing microscopic observation and chemical analysis by a wave-length-dispersive electron microprobe analyzer (WDS). For the latter analysis, the randomly selected glass shards found in the thin section were analyzed quantitatively with WDS (JEOL JXA-8800RL) in the Department of Earth and Environmental Science, Hirosaki University. Beam currents of 3×10^{-9} A and beam diameters of 10 micrometers were used at an accelerating voltage of 15 kV. Oxide percentages were renormalized to 100% for data comparison. These analytical device and analytical condition are the same as that in previous analysis for volcanic glass contained in tephras mentioned above.

4. Results

The result of chemical analysis for the two samples is shown in table 2-a and 2-b, and it is also plotted in FeO-CaO, FeO-K_2O diagrams and Harker diagram with the plots of the candidate tephras (figs. 3-b, 4-a and 4-b).

In the FeO-CaO diagram, almost all the chemical plots of volcanic glass shards contained in the Usulután pottery paste are distributed within and around the range of tephras from Coatepeque Caldera (Fig. 3-b) except one exceptional dot (No.26 in table 2-a) located in the range of tephras from Ilopango Caldera. In the FeO-K_2O diagram, the chemical plots of the earthenware samples seem to be shifted to higher value by approximately 1%, although they are mostly concentrated in and around the range of tephras from Coatepeque Caldera (Fig. 3-b). The exceptional dot is also located in the higher margin of the range of tephra from Ilopango Caldera.

In Harker diagram, the proportion of SiO_2 of the volcanic glass shards from the ceramic paste shows a

Table 2-a Chemical composition of volcanic glass grain contained in the Usulután ceramic sample-1

Analysis No.	SiO_2 %	TiO_2 %	Al_2O_3 %	FeO %	MnO %	MgO %	CaO %	K_2O %	Na_2O %	Total %
1	71.09	0.03	15.44	1.87	0.17	0.17	1.22	5.93	4.06	100.00
2	71.16	0.12	14.88	2.01	0.09	0.12	1.43	5.41	4.79	100.00
3	71.41	0.10	15.27	1.82	0.10	0.15	1.20	5.49	4.47	100.00
4	71.50	0.15	15.30	2.14	0.15	0.11	1.19	5.29	4.18	100.00
5	71.51	0.00	15.21	2.13	0.11	0.13	1.23	5.24	4.45	100.00
6	71.53	0.07	15.03	2.12	0.12	0.11	1.10	5.18	4.74	100.00
7	71.57	0.11	15.24	2.14	0.12	0.12	1.23	5.34	4.13	100.00
8	71.60	0.01	15.31	2.07	0.00	0.17	1.04	5.87	3.93	100.00
9	71.74	0.19	14.70	2.06	0.24	0.18	1.36	5.42	4.11	100.00
10	71.80	0.11	15.07	2.26	0.23	0.19	1.32	5.45	3.56	100.00
11	71.94	0.08	15.01	2.24	0.03	0.10	1.27	5.58	3.75	100.00
12	71.95	0.09	14.85	1.77	0.18	0.20	1.22	5.59	4.14	100.00
13	72.08	0.14	14.62	1.81	0.05	0.07	1.14	5.72	4.37	100.00
14	72.16	0.03	14.75	2.02	0.15	0.16	1.46	5.10	4.18	100.00
15	73.34	0.05	13.94	1.77	0.15	0.16	0.87	5.70	4.02	100.00
16	74.61	0.03	13.59	1.76	0.10	0.07	1.02	5.67	3.15	100.00
17	75.24	0.07	13.24	1.30	0.14	0.00	0.55	5.47	3.98	100.00
18	75.29	0.13	12.88	1.44	0.10	0.04	0.64	5.66	3.82	100.00
19	75.32	0.07	12.89	1.43	0.14	0.02	0.57	5.98	3.59	100.00
20	75.82	0.12	13.04	1.37	0.28	0.00	0.57	5.71	3.09	100.00
21	76.26	0.12	12.41	1.37	0.09	0.00	0.73	5.55	3.46	100.00
22	76.30	0.03	13.50	1.71	0.00	0.19	0.98	4.29	3.00	100.00
23	76.34	0.00	12.66	1.29	0.04	0.02	0.69	6.09	2.87	100.00
24	76.56	0.03	12.46	1.36	0.19	0.04	0.43	4.92	4.01	100.00
25	76.59	0.14	13.03	1.54	0.10	0.25	1.61	3.45	3.28	100.00
26	77.19	0.03	12.07	1.30	0.13	0.00	0.48	5.43	3.38	100.00
average	73.53	0.08	14.09	1.77	0.12	0.11	1.02	5.41	3.87	100.00
std. dev.	2.21	0.05	1.13	0.33	0.07	0.07	0.34	0.54	0.52	(26)

Table 2-b Chemical composition of volcanic glass grain contained in the Usulután ceramic sample-2

Analysis No.	SiO_2 %	TiO_2 %	Al_2O_3 %	FeO %	MnO %	MgO %	CaO %	K_2O %	Na_2O %	Total %
1	70.85	0.11	16.99	1.89	0.17	0.17	1.32	5.24	3.26	100.00
2	71.41	0.00	15.49	2.18	0.11	0.17	1.27	5.43	3.94	100.00
3	71.50	0.08	15.77	2.48	0.00	0.24	1.02	5.79	3.13	100.00
4	72.43	0.03	14.82	1.80	0.16	0.10	1.13	5.77	3.75	100.00
5	72.72	0.13	14.46	1.85	0.26	0.08	1.01	5.54	3.95	100.00
6	75.94	0.00	12.47	1.35	0.08	0.00	0.54	5.70	3.92	100.00
7	76.14	0.03	12.42	1.27	0.00	0.04	0.55	5.47	4.09	100.00
8	76.39	0.01	12.19	1.28	0.10	0.06	0.57	5.54	3.86	100.00
9	76.61	0.08	12.56	1.19	0.01	0.01	0.69	5.40	3.45	100.00
10	76.71	0.05	12.57	1.00	0.00	0.03	0.64	5.27	3.72	100.00
average	74.07	0.05	13.97	1.63	0.09	0.09	0.88	5.51	3.71	100.00
std. dev.	2.47	0.05	1.74	0.48	0.09	0.08	0.31	0.19	0.32	(10)

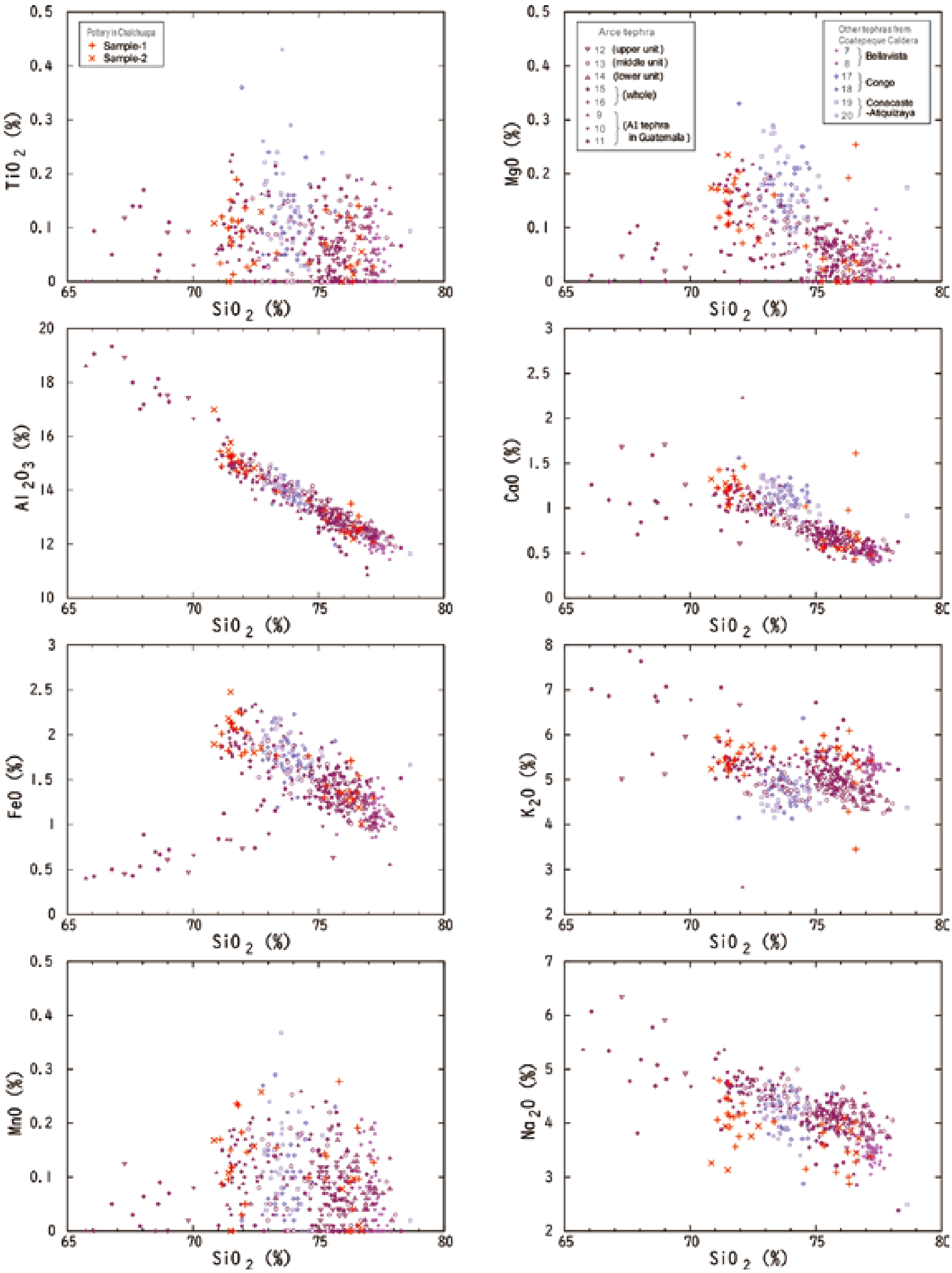

Fig. 4-a Chemical composition of volcanic glass contained in earthenware paste shown in Harker diagram and comparison with tephras from Coatepeque Caldera.

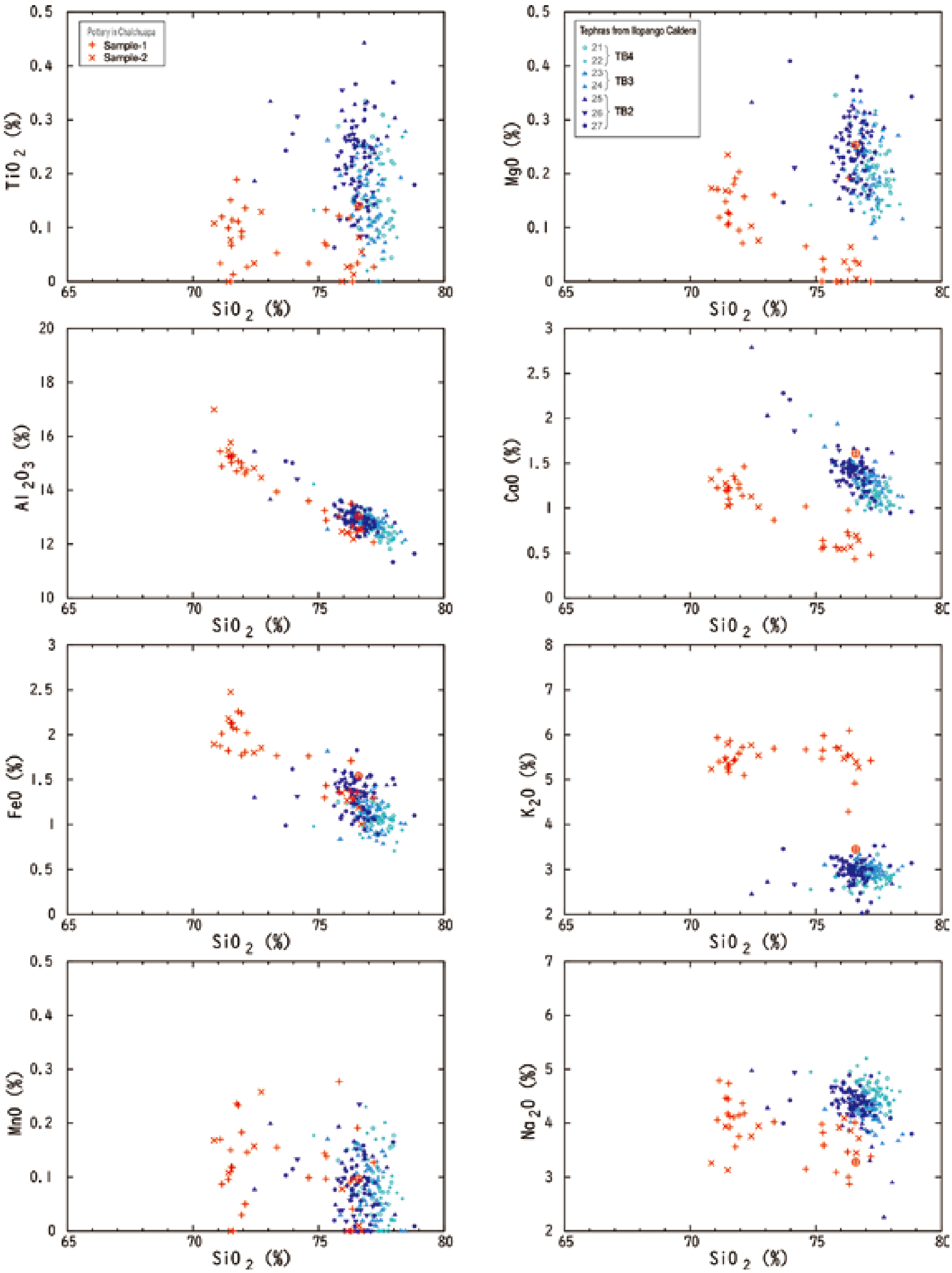

Fig. 4-b Chemical composition of volcanic glass contained in earthenware paste shown in Harker diagram and comparison with tephras from Ilopango Caldera.

relatively broad extent (Fig. 4-a). It is broader than the range of the Bellavista, while it is less consistent with the range of the Congo and the Conacaste, especially in the SiO_2-CaO diagram, in spite of partially overlapping. On the other hand, it is in good agreement with the range of the Arce, so that there is not inconsistent if almost all analyzed grains of volcanic glass are considered to be derived from the Arce tephra. While it is known that the Arce tephra comprise three fall units and their chemical properties are slightly different from one another, the chemical property of the volcanic glass contained in the earthenware samples seems in the best agreement with the that of the middle unit of the Arce tephra, because it does not have dispersal component as the upper unit, nor concentrated in narrow range of higher SiO_2 as the lower unit (Fig. 4-a).

In the FeO-K_2O diagram (Fig. 3-a), potassium content of samples seems to be shifted to the higher value by approximately 1%, as mentioned above. In Harker diagram, the sodium content seems to be shifted to the lower value, on the contrary to the potassium (Fig. 4-a). Such the chemical change in the potassium and sodium content appear in general at the ceramic production, the chemical shifts in volcanic glass shards in this study are considered to originate the alteration by heat when the ceramic was baked.

In the Harker diagram, the exceptional plot, mentioned above, is situated inside the ranges of the TB3, the TB2 tephras (Fig. 4-b), in spite that it is not consistent within the range of the TB4 tephra. Because most of the Usulután pottery is considered to have been produced during a period preceded to the TBJ eruption, the Usulután-style earthenware would not contain the volcanic glass from the TBJ tephra. The TB2 tephra is known to be distributed in the western El Salvador (Kitamura, 2016), so that it is possible that the exceptional volcanic glass may be derived from the TB2 tephra.

In the SiO_2-K_2O and the SiO_2-Na_2O diagrams, the exceptional plot is not located within the range of the TB2. However, it can be considered to be also consistent if the potassium value was shifted to the higher and the sodium value was shifted to the lower by approximately 1 %, under the effect of heat when the ceramic was baked.

5. Consideration

By the analysis of the study, most of the volcanic glass contained in the ceramic paste from the two pieces of the Usulután pottery was identified to originate from the Arce tephra, while exceptional one grain would be derived from the TB2 tephra. These data suggest that the Usulután pottery discovered by the excavation in La Cuchilla, Chalchuapa, was not moved for so long distance, because the volcanic glass contained in the ceramic paste was correlated to the tephras that are generally found out in and near Chalchuapa. At least, the two pieces of the pottery were possible to be produced in and around the area between Coatepeque Caldera and Ilopango Caldera, and it is unlikely that they are derived from the eastern El Salvador where both of the Arce tephra and the TB2 tephra are not distributed.

The chemical composition of the volcanic glass contained in the earthenware paste shows a best accordance to the middle unit of the Arce tephra among the fall units, and the middle unit is found, in Chalchuapa and its surroundings, as well-sorted and coarse-sand-sized fine pumice, which is much finer than the upper and the lower units composed of pumice lappili. This fact suggests the possibility that naturally sorted sand-size material of the middle unit of the Arce tephra would be useful for adjusting the clay hardness of the earthenware paste and could be utilized arbitrarily and selectively for production of the Usulután pottery in the area around Chalchuapa.

6. Further investigation

This study is the first step of the investigation to identify the place of origin of the Usulután-style pottery and to provide the information on the transport of the pottery or the propagation of the style. In this study, only two pieces of Usulután pottery were analyzed, only one thin section was made for each analysis, and only ten grains were analyzed for the second piece, so that the quantity of the WDS analysis is not enough to prove the place of pottery production. Therefore, it is necessary to increase the amount of earthenware samples for analysis as well as to increase the amount of volcanic glass to be analyzed for each earthenware piece.

References

CEL (Comisión Ejecutiva Hidroeléctrica del Río Lempa) (1992) Desarrollo de los Recursos Geotermicos del Area Centro-Occidental de El Salvador. Prefactibilidad Geotérmica del Área de Coatepeque. Reconocimiento Geotérmico. Informe Final, Internal report.

CEL (Commisión Ejecutiva Hidroeléctrica del Río Lempa) (1995) Prestación de servicios de consultoria para desarrollar estudios geocientíficos complementarios en el campo geotérmico Berlín, Departamento de Usulután, El Salvador - Partida 4: Estudio geovulcanológico, y recursos geotérmicos del área Berlín-Chinameca, internal report, San Salvador.

Danielson, J. J., and Gesch, D. B. (2011) Global multi-resolution terrain elevation data 2010 (GMTED2010): U.S. Geological Survey Open-File Report 2011-1073, 26 p.

Drexler, J. W., W. I. Rose Jr., R. S. J. Sparks, and Ledbetter, M. T. (1980) The Los Chocoyos Ash, Guatemala: A major stratigraphic marker in Middle America and in the three ocean basins, Quat. Res., 13, 327-345.

Dull, R. A., Southon, J. R., and Sheets, P. (2001) Volcanism, ecology, and culture: A reassessment of the Volcan Ilopango TBJ eruption in the southern Maya realm. Latin American Antiquity, 12, 25-44.

Dull, R., Southon, J., Kutterolf, S., Freundt, A., Wahl, D., and Sheets, P. (2010) Did the IlopangoTBJ eruption cause the AD 536 event? Poster presentation in Ame. Geophys. Union conference, San Francisco.

Hahn, G. A., Rose, W. I., Meyers, T. (1979) Geochemical correlation of genetically related rhyolitic ash-flow and airfall ashes, central and western Guatemala and the Equatorial Pacific. In: Elston, W. and Chapin, C. (eds), Ash Flow Tuffs Geol. Soc. Am. Special Pap., 180, 100-114.

Hernández, Guevara E. W. (2004) Características geomecánicas y vulcanológicas de las tefras Tierra Blanca Joven, Caldera de Ilopango, El Salvador. Proyecto final de Master en Tecnologías Geológicas. Universidad Politécnica de El Salvador y Universidad Politécnica de Madrid.

Kitamura, S. (2006) A preliminary report of the tephrochronological study of the eruptive history of Coatepeque Caldera, El Salvador, Central America. Bull. Fac. Social Welfare, Hirosaki Gakuin Univ., 6, 8-13. (http://id.nii.ac.jp/1610/00000168/)

Kitamura, S. (2010) Two AMS radiocarbon dates for the TBJ tephra from Ilopango Caldera, El Salvador, Central America. Bull. Fac. Social Work, Hirosaki Gakuin Univ., 10, 24-28. (http://id.nii.ac.jp/1610/00000201/)

Kitamura, S. (2016) Tephrochronological data for correlation of distal air-fall tephra from Ilopango Caldera in the central highlands of El Salvador, Central America. Bull. Fac. Social Welfare, Hirosaki Gakuin Univ., 16, 21-34. (http://id.nii.ac.jp/1610/00000248/)

Kitamura, S. (2017) Temporal chemical variation of the Arce tephra from Coatepeque Caldera, El Salvador, Central America. Bull. Fac. Social Work, Hirosaki Gakuin Univ., 17, 21-30. (http://id.nii.ac.jp/1610/00000257/)

Kitamura, S. (2018) Wide-spread tephras as a time-marker and chronological framework of eruptive history in the Berlín-Pacayal volcanic area, eastern El Salvador, Central America. Bull. Fac. Social Work, Hirosaki Gakuin Univ., 18, 17-32. (http://id.nii.ac.jp/1610/00000500/)

Kitamura, S. (2019) Mineralogical and chemical properties of pumice in the Berlín-Chinameca volcanic area, eastern El Salvador, Central America. Bull. Fac. Social Work, Hirosaki Gakuin Univ., 19, 1-10. (http://id.nii.ac.jp/1610/00000515/)

Koch, A. J., McLean, H. (1975) Pleistocene tephra and ash-flow deposits in the volcanic highlands of Guatemala. Geol. Soc. Am. Bull., 86, 529-541.

Kutterolf, S., Freundt, A. and Peréz, W. (2008) Pacific offshore record of plinian arc volcanism in CentralAmerica: 1. Along-arc correlations. Geochem. Geophys. Geosyst, 9 (2) (doi:10.1029/2007 GC001631).

Murano, M. (2017) Una síntesis del estudio sobre la técnica para la elaboración de una cerámica de estilo Usulután (in Japanese and Spanish). In Nakamura, S. (ed.),"New Developments in Maya Archaeology: An Interdisciplinary Approach", Kanazawa cultural resource studies, 16, 87-137.

Rose, Jr., W. I., Grant N. K., Easter J. (1979) Geochemistry of the Los Chocoyos Ash, Guatemala. In: Elston, W. and Chapin, C. (eds), “Ash Flow Tuffs”, Geol. Soc. Am. Special Pap., 180, 87-99.

Rose, Jr. W. I., Hahn, G. A., Drexler, J. W., Malinconico, M. L., Peterson, P. S., Wunderman, R. L. (1981) Quaternary Tephra of Northern Central America. In Proceedings of the NATO Advanced Study Institute, “Tephra Studies as a Tool in Quaternary Research”, held in Laugarvatn and Reykjavík, Iceland, June 18–29, 1980 (eds. Self, S. and Sparks, R. S. J.), 193-211.

Rose, W. I., Conway, F. M., Pullinger, C. R., Deino, A., McIntosh, W. C. (1999) An improved age framework for late Quaternary silicic eruptions in northern Central America. Bull. Volcanol., 61, 106-120.

Smith, V. C., Costa, A., Aguirre-Díaz, G., Pedrazzi, D., Scifo, A., Plunkett, G., Poret, V., Tournigand, P-Y., Miles, D., Dee, M. W., McConnell, J. R., Sunyé-Puchol, I., Harris, P. D., Sigl, M., Pilcher, J. R., Chellman, N. and Gutiérrez, E. (2020) The magnitude and impact of the 431 CE Tierra Blanca Joven eruption of Ilopango, El Salvador. Proceedings of the National Academy of Sciences,117(42), 26061-26068. https://www.pnas.org/lookup/suppl/doi:10.1073/pnas.2003008117/-/DCSupplemental.